

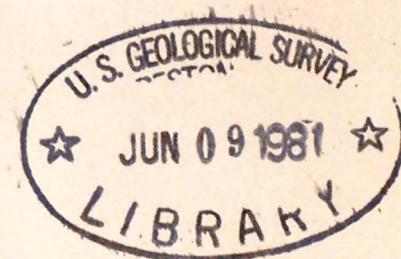
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# Genesis of Uranium- and Gold-Bearing Precambrian Quartz-Pebble Conglomerates

Proceedings of a Workshop,  
October 13-15, 1975, Golden, Colorado

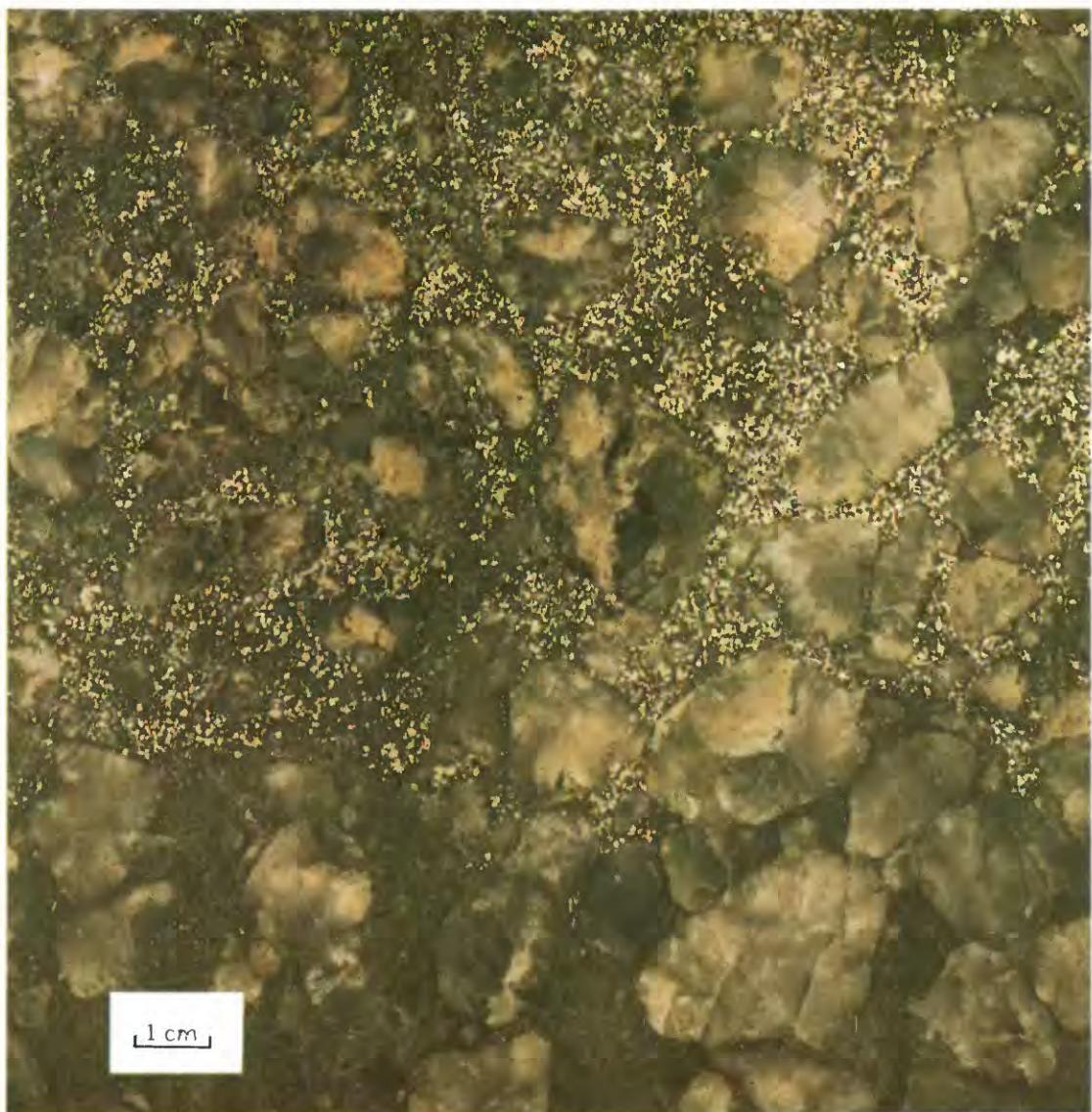
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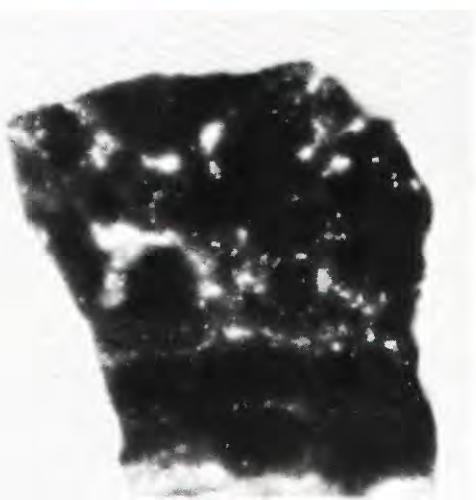
**GENESIS OF URANIUM- AND GOLD-BEARING  
PRECAMBRIAN QUARTZ-PEBBLE CONGLOMERATES**



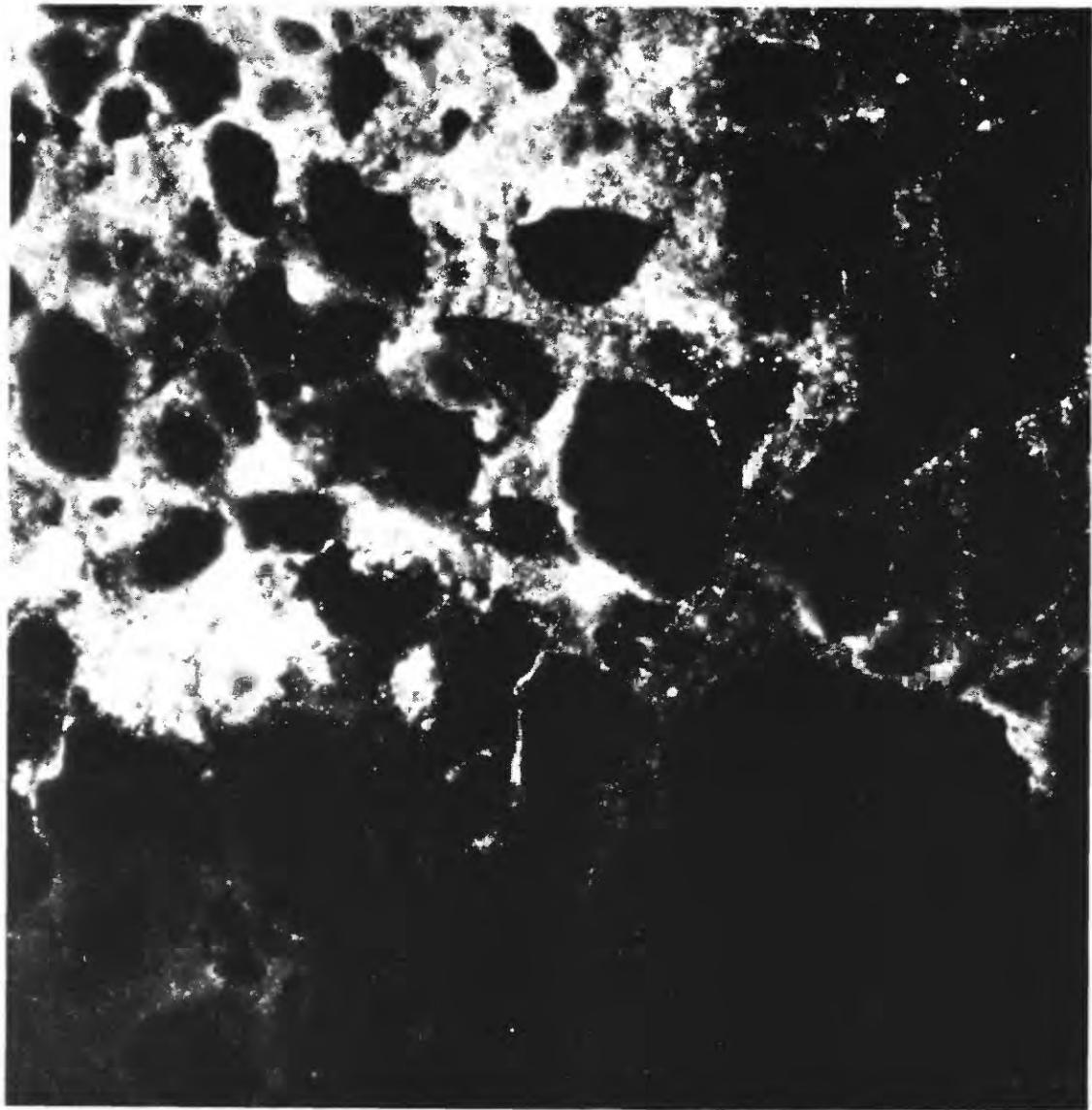
A1



B1



B2



A2

Polished specimens of quartz-pebble conglomerate uranium ore.  
Both actual size.

A. AB Reef, Denison Mine, Elliot Lake, Ontario. (Samples courtesy of V. Ruzicka, photography by David M. Watson, both Geological Survey of Canada.)

1. **Photograph, artificial light.** Note distribution of quartz pebbles; fine-grained pyritiferous matrix concentrated in upper two-thirds of photograph. The bottom of the photograph is the stratigraphic top of the specimen.
2. Radioluxograph\* of A1. Radioactivity is mostly confined to the pyritiferous matrix. Large radioactive areas result from clusters of minute detrital grains of radioactive minerals, mostly uraninite, not large detrital grains of radioactive minerals.

B. Carbon Leader Reef, about 1500 m depth, Blyvooruitzicht Gold Mine, Carletonville, Witwatersrand. (Sample courtesy of D. K. Hallbauer, Chamber of Mines of South Africa ; radioluxograph by David M. Watson, Geological Survey of Canada.)

1. Photograph, artificial light. The band of thucholite is the stratigraphic base of the specimen; note gold in it and along its upper margin. Compare quartz pebble size with A1. Note concentration of pyrite in matrix.
2. Radioluxograph\* of B1. Note that radioactivity is concentrated in the thucholite layer and in the pyritiferous matrix.

\* Dooley, J. R., Jr., 1958. The radioluxograph: a fast simple type of autoradiograph *in* Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy: v. 3, p. 550-553.



# Genesis of Uranium- and Gold-Bearing Precambrian Quartz-Pebble Conglomerates

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*Edited by* FRANK C. ARMSTRONG

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GEOLOGICAL SURVEY PROFESSIONAL PAPER 1161-A-BB



UNITED STATES DEPARTMENT OF THE INTERIOR

JAMES G. WATT, *Secretary*

GEOLOGICAL SURVEY

Dallas L. Peck, *Director*

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## INTRODUCTION

By FRANK C. ARMSTRONG

The shock of the Arab oil embargo in 1973 suddenly made everyone conscious of the dependency of modern industrial societies upon assured steady supplies of energy-source materials. Two responses to this new awareness were the acceleration of the search for alternate energy sources and the substitution of abundant materials for scarce materials; in addition, scarce materials were channeled into more beneficial uses and were reserved to uses for which no acceptable substitute was known. Immediately, of course, a high-priority target for change was the use of petroleum and natural gas in base-load central electricity-generating stations. The world had a developed expanding technology that appeared made-to-order as a heat source for central generating stations, namely, nuclear reactors. The promise of nuclear reactors created a strong demand for uranium, and consequently the price of  $U_3O_8$  rose. Increased price for  $U_3O_8$  opened geologic environments for exploration that theretofore had not been considered because they were not economically viable. Almost simultaneously, the oil-deficient industrial nations of the world began to look at their uranium resources. Because of the long lead times involved and because of the large capital expenditures necessary to construct nuclear reactors, attention focused on the adequacy of uranium resources to supply the anticipated new nuclear reactors for their entire useful lives of thirty to forty years. The world cumulative need for uranium to supply  $U_3O_8$  for the lifetime needs of the anticipated nuclear expansion proved to be very large indeed. Only a few countries with large uranium reserves and small nuclear-reactor programs had adequate supplies of known uranium resources. The rest of the western world would have to depend on imports or new discoveries. As the cost of generating electricity is relatively insensitive to the cost of  $U_3O_8$ , continued high prices for secure supplies of uranium seemed assured.

Such was the climate of the uranium market in late 1974. Exploration for deposits that previously were considered low-grade was undertaken, known low-grade deposits were re-examined, and consideration was given to resumption of production from low-grade mines where production had ceased, particularly in South Africa, where uranium had been produced almost exclusively as a byproduct of gold mining. The rising price of gold and the prospect of a favorable uranium market in late 1974 encouraged former South African uranium

producers to reactivate their long dormant extraction circuits and also encouraged others who had not previously recovered uranium to install new uranium-recovery plants. At that time about 35 percent of the known world uranium resources was contained in quartz-pebble conglomerates in South Africa and Canada.

In late 1974 the United States started to take a hard look at the adequacy of its uranium supplies in the light of the cumulative need for uranium to meet the forecast demands for electricity to be generated by nuclear reactors. At this same time a report on South African gold- and uranium-bearing quartz-pebble conglomerates that was in preparation by a U.S. Geological Survey scientist, W. Bradley Myers, came to the attention of the Director of the Geological Survey, Vincent E. McKelvey. Because no uranium-bearing quartz-pebble deposits were known in the United States, despite the occurrence of favorable geologic environments in the Lake Superior-Upper Michigan area and elsewhere, and because of the importance of this type of deposit in nearby Ontario and in South Africa, the Director suggested to the Chief Geologist that a workshop on uranium-bearing quartz-pebble conglomerates be sponsored by the U.S. Geological Survey to broaden understanding of the origin and distribution of deposits of this type as a basis for further exploration. The Chief Geologist asked me to organize such a workshop and to compile the proceedings and discussions as a Professional Paper.

The main burden of presentation at the workshop necessarily fell on scientists from South Africa and Canada; contributions were also to be made by others in the United States, Brazil, the United Kingdom, Germany, and Australia. Unfortunately, Australia could not be represented because the workshop was called on such short notice that their personnel had other commitments that took priority. Originally it was also planned to have the panel informally discuss the quartz-pebble conglomerates in Ghana, Finland, and Russia. As it turned out, however, except for the remarks made by Dr. Ruzicka about two Russian deposits, there was not time to discuss these other deposits.

Selection of the panelists and their subjects for the workshop would have been a formidable task had it not been for the help of friends and colleagues and their knowledge of up-to-date developments. The speakers on the Rand were selected with the advice of Dr. Desmond

A. Pretorius, Director, Economic Geology Research Unit, University of Witwatersrand, and Dr. Johann W. von Backström, Director, Geology, Atomic Energy Board of South Africa. The Canadian panelists were invited following the suggestions of Dr. Heward W. Little, who at that time was Head, Uranium Resource Appraisal Section, Economic Geology Subdivision, Geological Survey of Canada. Dr. John M. A. Forman, Mineral Resources Director, NUCLEBRAS, assisted in selecting the Brazilian panelist. Dr. S. H. U. Bowie, Director, Institute of Geological Sciences, London, suggested that Dr. Peter R. Simpson attend the workshop in his stead. Numerous colleagues assisted in selecting the other speakers.

To a student of Precambrian quartz-pebble conglomerates it must seem strange that such a workshop could be held without including Dr. Desmond A. Pretorius. It was originally intended to hold the workshop in the late spring of 1975, but it soon became apparent that many people, including Dr. Pretorius, could not attend at that time. Accordingly, the date was changed to October 1975 to coincide with a time when Dr. Pretorius would be in the United States on other matters. As it later developed, he had to cancel all plans of visiting the United States at that time. The workshop did benefit, however, from his advice in selecting the panelists.

Originally it was anticipated that the workshop would be an in-house study to be held in Reston, Va., but in the early stages of organization it was realized that all geologists in the Branch of Uranium and Thorium Resources, based in Denver, Colo., should attend, and that industry would also be interested in attending a meeting at which experts from around the world would be speaking. As many companies have uranium-exploration offices in Denver, moving the workshop to Denver would fulfill two needs: personnel of the Branch of Uranium and Thorium would be able to attend, and by opening the workshop to all, the Geological Survey would be fostering its continuing mutually beneficial dialogue with industry. Accordingly, the workshop was held at the excellent conference facilities at the Green Center, Colorado School of Mines, Golden, Colo., on October 13-15, 1975, and was attended by about 250 specialists from government, academe, and industry; about 125 companies connected with the uranium industry were represented. Following presentations by the speakers, discussion was among the speakers and the other invited participants. If time permitted, discussion was then extended to the general audience.

The workshop was designed to examine genetic concepts and their possible bearing on exploration. Whether or not it was successful in that respect is left to the reader to decide. Most papers presupposed a

familiarity with the subject matter; the reader who regrets the lack of an extensive review paper for the Rand is referred to D. A. Pretorius, 1974, The Nature of the Witwatersrand Gold-Uranium Deposits: University of Witwatersrand, Johannesburg, Economic Geology Research Unit, Information Circular No. 86, 50 p.

Before the workshop was held, it was decided that the papers for the Professional Paper would not be put through the rigorous editing "mill" of the U.S. Geological Survey. The Geological Survey did not think that much of its editorial system, for example, the Geologic Names Committee, could or should be used for authors almost all of whom were not Geological Survey employees and the majority of whom came from foreign lands and discussed geology in foreign lands. Accordingly, most of the editing has been for clarity and internal consistency. Subject matter, data, reasoning, and the like, were not changed from what the author submitted for publication. The reader will be interested to note the lack of agreement among the authors on many subjects. If nothing else, the workshop pointed out additional areas for investigation.

To preserve the international flavor of the meeting, slightly different English usage, spelling, and phrases, such as "on the Rand," have not been changed; similarly, to record the spontaneity of the meeting, the discussions have been transcribed as faithfully as possible. Some of the discussion was lost because of technical failures in tape recording, but it is estimated that more than 90 percent of the pertinent discussion was recovered and appears in the "Discussions."

All additions and comments by the editor are enclosed in square brackets ([ ]).

Usage in this report is as follows:

$$\begin{aligned}1 \text{ short ton} &= 1 \text{ ton} \text{ (abbreviated t)} \\1 \text{ metric ton} &= 1.1023 \text{ short ton} \\1 \text{ short ton} &= 0.9072 \text{ metric ton} \\1 \text{ metric ton uranium metal (U)} &= 1.3 \text{ t U}_3\text{O}_8 \\1 \text{ t U}_3\text{O}_8 &= 0.7693 \text{ metric ton U}\end{aligned}$$

A large part of the success and smooth running of the workshop must be credited to the U.S. Geological Survey's Center for Continuing Education in Denver, headed by the late Dr. Harley Barnes and ably assisted by Helen E. Eichler, Cynthia Rathbun, and David L. Macke. They made all arrangements for the meetings and housing and recorded the proceedings. Special credit must go to Carolyn Willis for phonetically transcribing the discussions. She spent many long hours trying to understand unfamiliar accents pronouncing unfamiliar place names and discussing esoteric subject matter. Thanks are also due Drs. von Backström and Little for presiding over the South African and Canadian parts of the workshop, respectively. We are most indebted to the panelists for their willingness to take

part in the workshop and to prepare their papers for publication, and we are greatly indebted to the companies, governments, and other organizations who made possible the participation of the panelists.

About one-third of the manuscripts were not received by the editor until 1978, and the revised draft of the final

one was not submitted until April 1979. Most papers submitted after the workshop differ somewhat from the authors' original presentations; in some instances this difference may make the discussion difficult to follow. As can be seen from dates of references cited, many authors have updated their papers past 1975.



**HISTORY**



# Investigation of Witwatersrand Uranium-Bearing Quartz-Pebble Conglomerates in 1944-45

*By* WESTON BOURRET

GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN  
QUARTZ-PEBBLE CONGLOMERATES

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## GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN QUARTZ-PEBBLE CONGLOMERATES

# INVESTIGATION OF WITWATERSRAND URANIUM-BEARING QUARTZ-PEBBLE CONGLOMERATES IN 1944-45

By WESTON BOURRET<sup>1</sup>

### ABSTRACT

The paper discusses the results - both short- and long-term - of a 1944 study undertaken by a team from the Manhattan Project (supported by the governments of Great Britain and Canada) to assess the potential for uranium occurrence in the Witwatersrand mining region of South Africa.

Details are given on the methods used by the Manhattan team and the results of the sampling study and survey that the team conducted in South Africa. The findings for 23 mines that were surveyed in the Witwatersrand are documented. These mines were the major producing mines in the Witwatersrand region.

The prior research that led the Manhattan Project administrators to concentrate efforts on the Witwatersrand is described. The history of uranium production in the Rand since the 1944 study is delineated.

Tables show the findings of the sampling study in terms of various major mines. A graph is included to show the growth of uranium production in the Witwatersrand from the initiation of the first operation in 1952 to 1975.

### INVESTIGATION OF WITWATERSRAND URANIUM-BEARING QUARTZ-PEBBLE CONGLOMERATES IN 1944-45<sup>2</sup>

The presence of uraninite on the Central Rand was first reported in a heavy-mineral concentrate recovered from City Deep mines in October 1923 by R. A. Cooper. It should be mentioned, however, that Fred Wartenweiler, consulting metallurgist for the Johannesburg Consolidated Investment Group, was indirectly responsible for this discovery, in that he initiated a change in Rand milling practice from conventional

amalgamation plates through the installation of corduroy riffle tables ahead of the cyanide circuit for the recovery of coarse gold and heavy minerals. This change made possible Cooper's discovery.

Because of the abundance of pyrite and a variety of other heavies characteristic of placer deposits, the volume of concentrate caught on the corduroy far exceeded the capacity of the amalgamation plant to recover a high percentage of free gold. As a consequence, a number of Wilfley-type tables were installed to recover a middling product from the corduroy tables with a specific gravity of about six or less.

Since Wartenweiler's decision to install corduroy riffles did, in fact, result in improved recovery of free gold, as well as osmiridium and placer heavies, his flow sheet was quickly adopted by a major segment of the Rand gold industry. As a natural consequence of reconcentrating the corduroy fraction over tables, curiosity as to their composition and mineral content led Fred Wartenweiler to ask his mill superintendent, R. A. Cooper, to examine and identify the chemical and mineral content of the heavy fraction. The result of this research is the now-famous paper of Cooper's that appeared in the Journal of the Chemical, Metallurgical and Mining Society, in October 1923, followed by a reply to discussion in May 1924. In his reply, Cooper reported on the radioactivity of a concentrate from the City Deep Mine. This concentrate was recovered over a period of about three months in the course of dressing the concentrates collected from corduroy tables, representing 300,000 tons<sup>3</sup> of mine-run ore. The total weight of the

<sup>1</sup> Consultant, Belvedere, Calif.; formerly Senior Geologist, Manhattan District Project, U.S. Corps of Engineers

<sup>2</sup> This summary records events initiated through the Manhattan Project in 1944 that led to sampling and preliminary evaluation of more than 20 producing Witwatersrand gold mines. The work was made possible through the joint effort of the Rand Gold Mining Industry, Manhattan Project engineers, and the South African Chamber of Mines. The complete results of the sampling study and survey are presented in the First Uranium Report, Witwatersrand, South Africa, published in 1973. This document is in the libraries of the South African Atomic Energy Board, the Chamber of Mines of South Africa, and numerous South African universities, in the U.S. Geological Survey libraries at Reston, Va., and Menlo Park, Calif., and in the Congressional Library, Washington, D.C.

<sup>3</sup> [Originally all record-keeping on the Rand was in English units and the short ton (2,000 lbs) was used. When the work was done on which this report is based, the English system was in use. Accordingly, unless otherwise specified, English units are used in this report. Record-keeping on the Rand is now in metric units; the change from English to metric units was made in the mid-1960's. In Figure 2 all uranium production is shown in metric units.]

final (reconcentrated) product was only 720 grams; it assayed as follows:

<i>Assay of radioactive fraction</i>	
Uranium Oxide ( $U_3O_8$ )	44.0 percent.
Radium	127 mg per ton
Osmiridium	28.35 grams

Then, Cooper went on to say (p. 266):

The streak on the concentrating table \*\*\* is brownish black, the uraninite itself being black. \*\*\* As uraninite is distinctly softer than pyrites it is, of course, possible that a considerable amount may have been lost by slimming. The quantity recovered certainly is only a homeopathic trace, but that does not detract from its technical interest.

The uranium exploration unit of the Manhattan District Project was established in 1942 for the purpose of discovering and evaluating uranium deposits believed to hold economic potential for production. The governments of Great Britain and Canada cooperated in this effort during and following World War II.

Consistent with this objective, a literature research unit was established in 1943 in New York and Washington and at the Massachusetts Institute of Technology, under the direction of Dr. Joseph Sinclair, to identify potential uranium occurrences outside the United States that appeared to justify follow-up examination by field geologists and mining engineers of the Manhattan Project staff.

Early in 1944, Margaret Cooper of Dr. Sinclair's group directed attention to R. A. Cooper's paper on the Witwatersrand "heavies" research. Even though only 720 g of final radioactive product were isolated from some 300,000 t of ore, the decision was made to send a field team under the direction of Weston Bourret to South Africa to expand on and look deeper into the work initiated by Wartenweiler and Cooper in 1923. Since Cooper's work provided results from only one sample from the City Deep Mine, the main objective of the Manhattan Project investigation, of course, was to examine and sample as many of the major operating mines along the East, Central, and West Rand as time permitted—specifically, those with gravity circuits incorporated in the flow scheme capable of recovering some uraninite.

Upon arrival in Johannesburg in May 1944, Bourret, assisted by Frank West, established contact with the Witwatersrand Chamber of Mines and consulting metallurgists of the major mining houses. Despite stringent wartime secrecy, the cooperation of the mining industry, South African government, and Chamber of Mines was essential, of course, to gaining access to the larger mines and getting the job done. Their superb cooperation is gratefully acknowledged, for without the help of so many metallurgists, engineers, and mine managers, successful negotiations and preparation for

South African uranium production by 1952 could have been delayed by several years. Although space limitations do not permit identifying every individual, the complete cooperation of consulting metallurgists and managers of the major mining houses is gratefully acknowledged, particularly the efforts of Fred Wartenweiler, T. K. Prentice, R. A. Cooper, and E. T. Pinkney.

In the course of the investigation, more than 20 Rand mining operations (representing more than 50 percent of total Rand gold production) were examined, and samples of concentrated products from gravity circuits, similar to those shown in figure 1, were taken for analysis and mineralogic study. In 1944 the most westerly operating mine on the Rand was Blyvooruitzicht, a new and high-grade mine that had just come into production. Unusually encouraging sample results from the Blyvooruitzicht table concentrates immediately focused attention on the importance of the Far West Rand and continuity of major uranium and gold mineralization to the west and south. It was at Blyvooruitzicht also that the presence of thucholite was first identified by Dr. D'Arcy George, chief mineralogist of the Manhattan Project, from high-carbon ores of the Carbon Leader Reef. This footwall carbon seam is subsequently referred to in the literature as an algal mat.

During the three and one-half month period spent on the Witwatersrand field investigation, the project team accomplished the following:

1. Visited more than 20 mining or milling operations and obtained bulk samples of uraniferous gravity-circuit mineral concentrates. These samples included Wilfley tables, batea, and amalgamation barrel fractions.
2. Examined and assayed more than 42 product samples.
3. Examined deep underground stoping operations at 16 Rand mines.
4. Observed methods of recovering uranium-gold concentrates and attempted estimates of slime losses from various-sized fractions.
5. Discussed broad aspects of Rand geology, mineralization, and distribution of gold with the consulting geologists and mine operators.
6. Discussed recovery (and losses) of heavy-mineral concentrates by means of corduroy and Wilfley tables with consulting metallurgists.
7. Separated and prepared mineral fractions utilizing a Haultain superpanner. Sized fractions were prepared from sieved samples and mineral determinations were made.

The field team was equipped with a Herbach and Rademan type 215 gamma counter. This instrument



FIGURE 1.—Corduroy table feed on deck of Wilfley table, Robinson Deep mine. *a*, Wide light-gray band: pyrite, quartz and zircon. *b*, Sinuous, narrow black band: principally chromite and niccolite. *c*, Narrow, somewhat sinuous light-gray streak: coarse iron. *d*, Wide black band: uraninite. *e*, Wide light- and medium-gray band: osmiridium with some gold.

proved adequate for approximating the  $U_3O_8$  content of concentrate samples. It was entirely inadequate, however, for testing ores containing only a few hundred parts per million. A number of samples were sent to the laboratories of the Manhattan Project in the United States for check assay. (This transfer was accomplished through diplomatic air pouch of the U.S. State Department.) The time interval of from two to three weeks in receiving results enabled confirmation of assays made in the field with the gamma ray counter.

The mines visited and sampled during the investigation are shown in the following list:

West	Central Rand	East
West Rand Consolidated	Durban-Roodepoort Deep	East Rand Proprietary
Randfontein Estates	Consolidated Main	Van Ryn Mines
Blyvooruitzicht	Crown Mines	New Modderfontein
Klerksdorp	Robinson Deep	Modderfontein Gold
Dominion Reefs	City Deep	East Modderfontein
	Nourse Mines	Government gold areas
	Geldenhuys Deep	East Geduld
	Simmer and Jack	Daggafontein
	Rose Deep	Vogelstruisbult

The results of analyses of bulk samples taken at these operations are set forth in tables 1 and 2.

East and West Rand sample results show that the  $U_3O_8$  content of the heavy-mineral fractions recovered from the reconcentrated corduroy riffle tables is generally less than 1.0 percent, and the higher grade

ores, such as those from the Carbon Leader and Main Reef, contain far more uranium than gold on a weight ratio basis. The actual uranium-to-gold ratio of different mines and different reefs varies over a wide range; however, the ratio appears to be between 15 and 20 parts uranium to one part gold-- and often even higher. Based on the 1944 production figures for gold bullion, indications are that the Witwatersrand has the capability of producing more than 500 t of gold annually. Therefore, it may be conservatively concluded (assuming the installation of uranium plants) that the potential for uranium should be about 15 to 20 times the gold figure, or 7,500 to 10,000 t of  $U_3O_8$  contained in concentrate per year.

Upon returning to the United States in early 1945, the author prepared a summary report describing sampling methods, losses, and conclusions for submission to the directors of the Manhattan Project. Late in 1945, Dr. D'Arcy George and Frank West, together with other members of the Manhattan Project staff in New York City, cooperated in the preparation of the final report. The findings and recommendations of that report forced Dr. George Bain, consultant to the Manhattan Project, and Dr. Charles F. Davidson, senior geologist for the British Atomic Energy Board, to visit South Africa to continue the investigation.

TABLE 1. - *Description and analysis of samples*

[June-October 1944]

Sample	Mine	Sample description	Reef	Tons milled per month	Percent $U_3O_8$ (chem.)	Pounds $U_3O_8$ per ton
6915	Randfontein Estates	Chiefly pyrite; concentrate from Wilfley tables following removal of osmiridium streak (see sample 6905).	Main and Johnston	340,000	0.04	0.80
6907	West Rand Consolidated	Daily composite concentrate, sized to 70 percent - 200 mesh; 82 percent $U_3O_8$ values in - 200 mesh fraction.	Main, Johnston, and Bird	240,000	.08	1.60
6931	Durban-Roodeport	Daily composite; osmiridium concentrate from corduroy riffles (see sample 6930).	Main	200,000	<sup>1</sup> .03	.60
6927	Consolidated Main Reef	Composite of weekly gravity concentrate; contains 3-4 oz Au per ton.	Main	210,000	.09	1.80
6935	Crown Mines "A" Mill	About equal amounts of quartz and pyrite	Main and South	105,000	.53	10.6
6936	Crown Mines "B" Mill	More quartz than in sample 6935	Main and South	75,000	.48	9.6
6937	Crown Mines "C" Mill	Weighted average of the three Crown mine samples is 9.0 lb $U_3O_8$ per ton; 75 percent quartz and 25 percent pyrite.	Main and South	110,000	.32	6.4
6932	Robinson Deep	Chiefly pyrite with visible uraninite	South and Main Leader	100,000	1.98	39.6
6912	City Deep	Before removal of osmiridium; pyritic concentrate with quartz.	South and Main Leader	95,000	.71	14.2
6920	Nourse Mines	Mostly pyrite and quartz	South and Main Leader	80,000	.85	17.0
6923	Geldenhuys	Quartz and pyrite; osmiridium concentrate not removed (see sample 6921).	Main and South Leader	65,000	.09	1.80
6913	Simmer and Jack	Contains osmiridium (see sample 6914 for uranium osmiridium concentrate).	South and Main Leader	150,000	1.30	26.0
6922	Rose Deep	Concentrate prior to amalgamation and removal of osmiridium.	Main Leader	70,000	<sup>1</sup> 1.08	21.6
6924	East Rand Proprietary	Chiefly quartz and pyrite (Cason Mill)	Main Leader	105,000	.12	2.4
6925	ERP-Angelo Mill	Osmiridium concentrate removed	Main Leader	80,000	.12	2.4
6916	Modderfontein New North Mill	Daily composite heavy-mineral concentrate before osmiridium cut; essentially pyrite and pyrrhotite.	Upper Leaders and Black	72,000	.11	2.20
6917	Modderfontein New South Mill	Daily composite heavy-mineral concentrate before osmiridium cut; essentially pyrite and pyrrhotite.	Upper Leaders and Black	90,000	.03	.60
6918	Modderfontein B	Daily concentrate composite sample	Main Leader	70,000	.02	.40
6919	Modderfontein New East Mill	Daily concentrate composite sample	Main Leader	130,000	.03	.60
6906	Government areas	Daily composite after removing osmiridium.	Leaders, Black, and Kimberley	210,000	.13	2.60
6934	Government areas	Composite concentrate representing six week milling run.	Upper Leaders and Kimberley	220,000	.10	2.0
6933	Vogelstruisbult (East Rand)	Composite concentrate	Main Leader	75,000	1.74	34.8
6926	Blyvooruitzicht (West Witwatersrand areas)	Weekly composite containing osmiridium, visible uraninite, and thucholite.	Carbon Leader	12,000	1.20	24.0

<sup>1</sup> Radiometric equivalent.

TABLE 2. - *Description and analysis of osmiridium and miscellaneous samples*

[June-October 1944]

Sample	Mine	Sample description	Reef	Percent $U_3O_8$ (chem.)	Pounds $U_3O_8$ per ton
<b>Osmiridium gravity concentrates</b>					
6905	Randfontein Estates	Gold-pyrite-osmiridium concentrate with chromite, zircon, rutile, and uraninite.	Main and Johnston	1.01	20.2
6914	Simmer and Jack	Wilfley table composite concentrate	Main and South	3.28	75.6
6921	Geldenhuys Deep	Composite pyrite concentrate from corduroy.	Main and South	.81	16.2
6929	Consolidated Main Reef	Composite from corduroy tables, with pyrite.	Leader Middle South	1.52	30.4
6930	Durban Roodeport	Recovered over corduroy, pyrite, mill iron, and quartz.	Main and Leader	.24	4.8
6938	Robinson Deep	Isolated uraninite streak cut from Wilfley table; about 75 percent uraninite, with osmiridium, gold, mill iron, and pyrite.	Leader Middle South	63.8	1,276.0
<b>Miscellaneous and hand specimens</b>					
6928	Consolidated Main Reef	Wilfley table tails after dressing corduroy concentrates; essentially quartz.	Leader Middle South	nil <sup>1</sup>	-----
6939	Daggafontein	Heavy-mineral accumulation from ballmill liners; gift of Dr. E. Mendelsohn; 30-40 percent visible uraninite; also chromite, rutile, and iron.	Kimberly	14.5	290.0
5977	Vogelstruisbult	Hand specimens of high-grade gold ore; high pyrite content.	Black	.335	6.70
5978	Do	Hand specimen of Kimberley Reef	Kimberley	.018	.36
5979	Randfontein Estates	Sample of Main Reef gold ore	Main	.05	.12

<sup>1</sup> Radiometric equivalent.

Following Drs. Bain and Davidson's conferences with the South African government, Dr. L. T. Nel of the South African Geological Survey was placed in charge of a team of geologists and metallurgists organized to expand the sampling of reefs reported to contain significant uranium. These locations included Blyvooruitzicht, Vogelstruisbult, Western Reefs, and East Daggafontein. Moreover, in 1946 a special uranium research committee was organized to recommend metallurgical research on the recovery of uranium by flotation, leaching, and alternative methods. This team consisted of T. K. Prentice, Fred Wartenweiler, and O. A. E. Jackson. About the same time, a research mineralogist, Dr. W. R. Liebenberg, was appointed to study and report on the heavy minerals contained in the Witwatersrand conglomerate ores. With the assistance of a consulting metallurgist Robert R. Porter, the first test pilot plant was erected at Blyvooruitzicht and Western Reefs in 1949 to work out a flow sheet based on sulfuric-acid leaching for the recovery of uranium.

In 1951 a small amount of yellow cake was produced by the pilot plant at the Blyvooruitzicht mine; commercial production on the Rand started in 1952. Since that time, South Africa is credited with the production of more than 82,000 metric tons of uranium oxide (from more than 20 mines) in the form of yellow cake concentrates valued at more than \$2 billion. The record of this production by years is set forth in figure 2.

According to the annual reports of the Chamber of Mines of South Africa, the average recovery from approximately 300 million metric tons of ore treated from

1952 through 1975 was 0.271 kg  $U_3O_8$  per metric ton, or 0.54 lb  $U_3O_8$  per short ton. Since discovery of the Rand in 1886, more than 3.3 billion metric tons of ore have been treated in cyanide mills, from which more than 32,000 metric tons of gold bullion have been recovered.

The tailings dumps and slime dams resulting from milling over 3 billion metric tons are intact around the north rim of the Witwatersrand Basin. Although the uranium content of these dumps is low, the uranium contained in them reaches an impressive figure, probably approaching 500 million pounds of  $U_3O_8$ . Possibly the day is not too far off when it will become economically feasible to re-treat some of the old tailings, assuming a price, say, of \$40 to \$50 per pound  $U_3O_8$ . In any event, the future of the Rand as one of the world's major uranium producers is secure.

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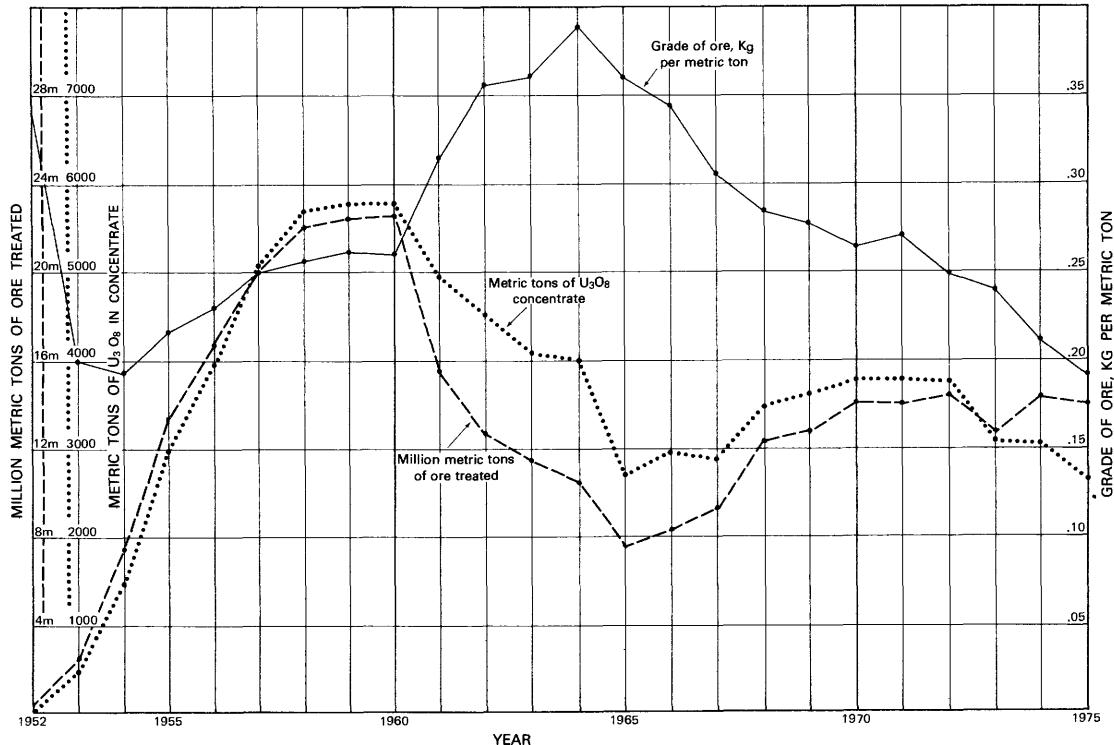


FIGURE 2.—Uranium production from South African gold mines.

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## DISCUSSION

*von Backström:* I must say that I really enjoyed this historical review. I just want to mention to the audience what the situation is at the present. We have fifty gold mines in production at the moment and of these there are seven that have extraction units for uranium. There are ten uranium plants. We think that with the demand building up and some of our extraction plants which are moth-balled being prepared for production, we could probably eventually double the number of mines that would be able to produce uranium. It is very difficult to say at this stage what the uranium production capability would be from the Witwatersrand mines because there are so many factors influencing production. I was very interested to hear the figure of 10,000 tons of  $U_3O_8$  per year as the production capability of the Rand. I think that level of production probably could be reached provided that all economic factors were favorable. At the moment we certainly are far away from that situation. I think it is generally known that labor costs have increased to a very great extent and will certainly affect production

capability. Another important item is, of course, the gold price, which has not been very consistent. This inconsistency makes the decision-makers uneasy about installing expensive extraction plants. Another unknown economic factor, of course, is inflation.

*Bourret:* Dr. von Backström, I apologize for not specifically including your name among those I mentioned. At that time, of course, you were not working on the Rand. Nevertheless, I should call attention to the fact that you have done a tremendous amount of work on the Rand since the days of the early investigations. Obviously, when I mention a figure of 10,000 tons, I simply cite that as an ultimate productive capability, assuming that the economic factors were reasonably favorable.

Also I should mention in passing that there are slime dams and dumps, as you well know, from which uranium has not been recovered. In a sense, uranium has been stock piled in these enormous dumps and they are available for reprocessing in the event that the price of uranium and the market were favorable. Recovery of uranium from these dumps is a project for the future. As in excess of one hundred million tons of ore is processed annually, and at least ninety-nine and a half percent of that material ends up in slime dams, the dumps contain a large uranium resource.

*Myers:* You spoke of making tests on some of the central Rand mines. Could you give a statement as to the amount of uraninite contained in crude ore?

*Bourret:* I would say that the ore averaged perhaps 0.3 pound of  $U_3O_8$  per short ton.

*Myers:* This would be a reasonable average for the central Rand?

*Bourret:* Yes. Another way of checking this is to go to the Annual Report of the Chamber of Mines, where the grade is given in kilograms per metric ton treated. Now if in some of these mines they had sorting belts, if they hand sorted out some of the waste rock or shale

or obviously barren material, then these figures would be a little on the high side.

[In 1975 the average recovery from ore processed by the seven uranium-producing mines on the Rand was 0.189 kg  $U_3O_8$  per metric ton, that is, 0.378 lbs.  $U_3O_8$  per short ton.]

[The following is quoted from Mining Survey (no. 74, April 1974, p. 28), a magazine published quarterly by the Chamber of Mines of South Africa, Johannesburg.]

#### URANIUM REPORT

Publication in the United States of the "First Uranium Report - Witwatersrand, South Africa" has thrown fresh light on the events that led to the establishment of South Africa's uranium industry.

The report is by Dr. Weston Bourret who was employed by the United States Government in 1944 to undertake an investigation of the uranium mineralisation on the Witwatersrand as a part of the effort mounted by the "Manhattan Project" to develop the atomic bomb. Dr. Bourret visited South Africa to carry out field work. The results of his work could not be published previously for security reasons.

The presence of uraninite or other uranium minerals in the gold-bearing conglomerates of the Witwatersrand had been first reported in 1923 in a technical paper by R. A. Cooper entitled "Mineral Constituents of Rand Concentrates". These findings attracted little attention at the time but were recalled when the Manhattan Project was under way.

Dr. Bourret concluded that the Witwatersrand was potentially one of the world's most important and lowest cost producers of uranium and he estimated possible output at between 5,000 and 7,000 tons of uranium oxide a year. Events were to prove him right: production in the late 1950s ran in excess of 6,000 tons a year.

Dr. Bourret recommended that "a detailed investigation be undertaken promptly with a view to recovering uranium and determining the overall uranium potential of this very important field."

As a result follow-up investigations were initiated by the Manhattan Project and the British Atomic Energy Commission. Distinguished contributions to the establishment of South Africa's great uranium industry were then made by Professor George Bain, Consulting Geologist of the United States Atomic Energy Commission and Dr. C. F. Davidson of the Geology Survey of Great Britain. Production began in 1952 and South Africa remains today one of the top world producers.



**GENERAL**



# The Precambrian Development of an Oxygenic Atmosphere

By J. WILLIAM SCHOPF

GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN  
QUARTZ-PEBBLE CONGLOMERATES

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GEOLOGICAL SURVEY PROFESSIONAL PAPER 1161-B





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## THE PRECAMBRIAN DEVELOPMENT OF AN OXYGENIC ATMOSPHERE

By J. WILLIAM SCHOPF<sup>1</sup>

## ABSTRACT

It has become rather widely accepted in recent years that (1) during the geologic past, the Earth's atmosphere evolved from an initial "anoxic" to a later "oxygenic" state; that (2) this change was a result of the cumulative effects of "green plant" (i.e., oxygen-producing) photosynthesis; and that (3) the transition occurred during Precambrian, and probably Early Proterozoic, time. Although evidence in support of these views appears to be both more circumstantial and more qualitative than has perhaps been generally appreciated, a broad range of biological (and paleobiological, biochemical, and organic-chemical) data seems to establish that such a transition did in fact occur during the Precambrian as a result of blue-green algal photosynthesis, with geological (and mineralogical) data suggesting a plausible date for the transition of about 2,000 million years ago. Nevertheless, it has become increasingly evident that the Earth's early atmosphere was not totally devoid of free oxygen, as had apparently been assumed by many workers. Diagenetic oxidation, to yield such sediments as banded iron-formation and red beds, may thus have occurred on the primitive Earth prior to the advent of oxygen-producing photosynthesizers; if so, the mere presence of such deposits should not be construed as necessarily evidencing the occurrence either of biological activity or of an environment sufficiently "oxygenic" to have been habitable by aerobic forms of life.

## INTRODUCTION

During the past quarter century—since the discovery in the early 1950's of the now-famous microfossil assemblage of the mid-Precambrian Gunflint Cherts (Tyler and Barghoorn, 1954)—and especially within the past decade, there has been a marked increase of interest throughout the world in the paleobiology of the Precambrian. Long-standing questions regarding the nature, antiquity, and even the existence of Precambrian life, questions dating from the mid-nineteenth century and the writings of Charles Darwin (1859), have at last begun to be answered. The question now is not whether evidence of Precambrian life exists, but rather, what does it say—what can it tell us of the timing and nature of early evolutionary advances and, especially, of the interactions between the evolving Precambrian biota and the developing early environment?

Probably foremost among all such biotic-environmental problems is the matter of Precambrian atmospheric evolution or, more precisely, the timing and nature of the development of an "oxygen-rich" atmosphere, one capable of supporting the type of aerobic metabolism on which we and most other living systems are dependent. In the present ecosystem, anaerobic organisms of course also exist, but collectively they are only a small fraction of the Earth's extant biomass. Although anaerobic metabolic pathways occur in living aerobic organisms, they tend to be of subsidiary importance only, operating for rather limited periods as a sort of "back-up system" to aerobic metabolism in times of special stress. Among the various tissues of higher mammals, for example, the probably most complex—the specialized nerve tissue that forms most of the human brain—inevitably dies if it is deprived of oxygen for more than about four minutes. Such dependence on free oxygen, while differing in degree from species to species, is exhibited by virtually all other megascopic organisms as well, plants included (since they, too, respire aerobically, using pathways that are essentially identical with those of higher animals). Indeed, among nonmicrobial forms of life, one is hard-pressed to find exceptions to this general rule of oxygen dependence; certain protists and other intestinal parasites no doubt qualify (although all such forms appear to be evolutionary derivatives of originally free-living, aerobic respirers), but the list is short (aquatic mammals, which can remain submerged for extended periods of time, are not included—special mechanisms have evolved that enable such mammals to maintain oxygen supplies to the brain at the expense of other, less oxygen-demanding, tissues). Free atmospheric oxygen plays a necessary and integral role in the extant ecosystem of the planet.

From the perspective of the present, therefore, it seems rather remarkable to realize that dependence on oxygen is by no means an inherent requirement for the existence of life on Earth—indeed, during the past decade it has become increasingly apparent that for an appreciable segment of geologic time, a segment encompassing a large portion of Precambrian Earth history and as much as half of the total history of life on Earth,

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the planet's atmosphere was virtually devoid of free oxygen and the Earth's biota was solely anaerobic; free oxygen was neither used by such life forms nor, probably, could its presence (in excess of some minimal level) have been tolerated. The anaerobes of the present ecosystem, the majority of which appear to be evolutionary derivatives of this ancient, pre-aerobic biota, hark back to this early anoxic environment, as do the anaerobic metabolic pathways that still function (albeit in some cases only under special circumstances) in extant plants and animals.

These views, interpretations of the early history of life that have become widely accepted in recent years, have important ramifications for an understanding of geological and biological processes on the Precambrian Earth. It is thus appropriate to inquire as to just how convincing the evidence really is regarding such a supposed major change in atmospheric composition—do available data require that such a change in atmospheric oxygen content actually occurred during the Precambrian? If so, what do they suggest regarding the timing of this event, the source of the atmospheric oxygen, the "morphology" of the transition (was it part of a long-term continuum, or did it occur as an abrupt step function?), and the impact of this rise in oxygen content on the Earth's biota and its environment? As will be evident from the following discussion, evidence bearing on these questions tends to be circumstantial rather than direct, and the answers generated, qualitative rather than quantitative. Still, while the various lines of relevant evidence (coming from rather disparate branches of the natural sciences) may individually seem less than fully compelling, they each are at least consistent with—and they commonly seem to point to—the occurrence of just such a transition, with several types of data suggesting that this event probably occurred during the mid-Precambrian, some 2,000 m.y. (million years) ago. Although additional and more telling data are no doubt needed, this most crucial problem in atmospheric evolution appears to be rapidly nearing solution.

### BIOCHEMICAL CONSIDERATIONS

Of the several lines of evidence that seem to bear on the question at hand, studies of the biochemical characteristics of modern organisms, coupled with an understanding of phylogenetic relationships and of the temporal distribution of appropriate types of fossils, are among the most powerful. While necessarily indirect—this line of evidence being based on the reasonable but unproved premise that the metabolic characteristics of fossil organisms can be accurately inferred from their morphology (by analogy with related modern organisms) or from their particular level of cellular or organismal complexity—this approach seems

generally quite sound. Just as morphological comparison with modern analogues can be used to infer that trilobites were mobile rather than sessile and that saber-tooth cats were carnivores rather than herbivores, biochemical comparison with modern taxa can be used to infer that both of these types of now-extinct heterotrophs were obligate aerobes, dependent upon the availability of free oxygen for various biosynthetic pathways and for aerobic metabolism. Indeed, such "argument by analogy," reinforced by an understanding of relevant evolutionary lineages, forms the basis of a vast majority of all paleoecologic and phylogenetic inference. One "knows" for rather similar reasons and with similar degrees of certainty that Paleozoic trilobites were both mobile and oxygen dependent.

Let us now apply this biochemical approach to the question of atmospheric evolution. Oxygen-requiring biosynthetic pathways (e.g., of tyrosine, hydroxyproline, collagen, steroids) and oxygen-dependent metabolism (namely, aerobic respiration) are characteristic of invertebrate metazoans; it thus seems evident that free atmospheric oxygen must have been readily available at least as long as such metazoans have been extant, by current estimates beginning some 700 m.y. ago (near the beginning of the "Ediacaran" or "Terminal Riphean" phase of the late Precambrian). Although it is perhaps not widely appreciated, this line of reasoning is not limited to the occurrence of fossil megascopic eukaryotes such as metazoans; indeed, it seems applicable, and with equal force, to the occurrence of microscopic eukaryotes as well, for even the most primitive of such organisms, unicellular microscopic algae (whether rhodophytes or, as according to some phylogenies, chlorophytes), require molecular oxygen for intracellular synthesis of such compounds as sterols and polyunsaturated fatty acids, for use in aerobic respiration, and possibly also for that most characteristic of eukaryotic traits, eukaryotic (i.e., mitotic) cell division (Amoore, 1961a, 1961b). As is true at the multicellular level of eukaryotic organization, unicellular eukaryotes are aerobic, oxygen-dependent "biological machines"; the occurrence of such cells in the fossil record would thus seem indicative of the presence of biologically usable concentrations of free atmospheric oxygen. Although there is some uncertainty regarding the time of origin of eukaryotes, the most recent, thorough compilation of data bearing on the subject suggests that the lineage had probably become established by about 1,400 m.y. ago (Schopf and Oehler, 1976); on this basis it can be inferred that appreciable quantities of atmospheric oxygen were present at least as early as the mid-Proterozoic.

Regardless of the time of origin of the eukaryotic cell, however—whether somewhat earlier or substantially

later than the date noted above (and both possibilities have been suggested in the recent literature)—it is apparent that eukaryotes were preceded by an entirely prokaryotic biota, an ecosystem composed solely of forms that, were they alive today, would be classed among the bacteria or the “cyanobacterial” blue-green algae. These prokaryotes provide additional evidence regarding the development of an oxygenic environment. Like their modern counterparts, fossil prokaryotes appear certain to have encompassed a broad range of physiological types, a spectrum including strict anaerobes, facultative anaerobes, microaerophiles, and obligate aerobes. Indeed, this striking versatility in oxygen relations is one (among many) of the factors that separate the prokaryotic and eukaryotic lineages and is one of the principal characteristics (again among many) that have led many workers to conclude that prokaryotes diversified during a time of changing oxygen content in the primitive environment. The data supporting this conclusion are many and varied, but perhaps the most instructive are those derived from biochemical studies of modern blue-green algae, prokaryotes of the kind that were apparently responsible for deposition of most types of Precambrian stromatolites (laminated, organo-sedimentary structures that are abundant and widespread in Precambrian carbonate facies younger than about 2,300 m.y. in age).

Although blue-green algae are oxygen-producing photoautotrophs and although they can cope with the intracellular oxygen thus produced (i.e., they possess oxygen-mediating enzyme systems) and can apparently use such oxygen both metabolically (namely, in aerobic respiration) and in what may be oxygen-requiring biosyntheses (i.e., in pathways leading to formation of phycobiliproteins and chlorophyll-*a*), they exhibit a number of biochemical characteristics that suggest that they probably evolved under changing conditions of ambient pO<sub>2</sub>. For example, many (but possibly not all) blue-green algae can use molecular oxygen to produce polyunsaturated fatty acids (via “oxidative desaturation”); the precursors of these fatty acids, however, are produced via a totally anaerobic pathway (similar to that which occurs in obligately anaerobic bacteria), and it has thus been inferred that the oxygen-requiring portion of this pathway was “added on” to a previously established anaerobic pathway during an early stage in blue-green-algae evolution (and certainly prior to the origin of eukaryotes, since they share this same oxygen-requiring biosynthetic system). Similarly, some (but not all) blue-green algae are capable of oxygen-requiring sterol synthesis, again involving a biosynthetic step (the cyclization of squalene) that was apparently “added on” to an earlier established anaerobic pathway that produces the necessary precur-

sor. Moreover, as is noted above, blue-green algae also use oxygen in aerobic respiration; and, as in apparently all aerobic (e.g., eukaryotic) organisms, the steps involved in this energy-yielding process (the so-called “citric acid” or “Krebs” cycle) have been superimposed upon—“added on” to—a shorter and completely anaerobic pathway, one that is less efficient and that yields far less cellular energy per molecule of substrate metabolized than its aerobic derivative. As a rule, although such oxygen-related “add-ons” are exhibited by eukaryotes, they may be present or absent in prokaryotes, such as blue-green algae; this rather striking difference in oxygen relations presumably reflects the fact that prokaryotes had become established prior to, and became diversified during, a major change in the oxygen content of the Earth’s environment.

In this regard, it is of significance to note that although blue-green algae are “good aerobes” (i.e., they flourish in many fully oxygenic settings on the present Earth), the group tends to be “less aerobic” than is typical of modern eukaryotes. Of those blue-greens for which relevant data are available, virtually all “tend to prefer” (i.e., grow most rapidly within, whether in the laboratory or the field) microaerophilic rather than fully aerobic environments—optimum growth rates are commonly observed in the presence of about 10 percent free oxygen, in contrast with the 20 percent of the present atmosphere. Moreover, although it has long been recognized that some blue-green algae are capable of living within and reproducing under totally anoxic conditions (both in the laboratory and in the field), it has recently been discovered that several species, among them the filamentous cyanophyte *Oscillatoria limnetica* (Oren and others, 1977), are capable of “switching on” or “turning off” their aerobic cellular machinery as a function of the environment in which they occur. Under anoxic conditions, these species can photosynthesize anaerobically (using H<sub>2</sub>S as a hydrogen source, like photobacteria, and thus not producing oxygen as a byproduct of photosynthesis), but if placed under oxygenic conditions, they can “switch on” their more normal aerobic apparatus (one using H<sub>2</sub>O as the hydrogen source, which thus results in the photosynthetic production of free oxygen).

During the past decade, it has become increasingly well established that the “heterocyst,” a particular cell type that occurs in several orders of filamentous blue-green algae, probably evolved in response to an increase in environmental oxygen content (Schopf, 1974, p. 29). These specialized, thick-walled cells have been shown to enclose the oxygen-sensitive nitrogenase enzyme system, a system that “fixes” atmospheric nitrogen by combining it with hydrogen and thus reduces it to the proper oxidation level for its incorporation into amino acids (and a system that is entirely lacking among

modern eukaryotes—in legumes, for example; nitrogen-fixation is carried out by prokaryotic symbionts rather than the eukaryotic host). In addition, such cells are known to lack "Photosystem II" (that portion of the photosynthetic apparatus involved in production of free oxygen) and to be capable of high levels of aerobic respiration. Under present-day oxygenic conditions, heterocysts apparently serve to protect the nitrogenase enzyme system from oxidation by ambient oxygen. They are thick-walled, so diffusion of oxygen into them is relatively slow; they lack Photosystem II, so oxygen is not generated within them; and they carry out active aerobic respiration, so the cell is scrubbed of any uncombined oxygen that may be present. Under anoxic conditions, however, many species of extant blue-green algae (including some that lack heterocysts) have been shown to be capable of fixing atmospheric  $N_2$ . Thus, it seems likely that the enzymes required for  $N_2$  fixation probably first appeared at a time during which the environment was essentially anoxic and that as oxygen concentrations increased (owing principally to the photosynthetic activities of the blue-green algae themselves!), specialized, thick-walled heterocysts developed as a means of providing (on a localized, microscopic scale) the anaerobic environment needed to protect the oxygen-labile nitrogenases and, thus, to ensure that  $N_2$  fixation could take place. Interestingly, *in vivo* and *in vitro* studies of nitrogenases from various sources (Stewart and Lex, 1970; Stewart and Pearson, 1970; Haystead and others, 1970) indicate that the enzyme-complex is partially deactivated (60 to 80 percent inhibition) by concentrations of ambient oxygen of about one percent and is completely deactivated at a  $pO_2$  of about 5 percent; read literally, this would seem to suggest that heterocysts may have first appeared with the attainment of this level of ambient oxygen content, and since heterocyst-like cells are first known from the fossil record in blue-green algalike filaments about 2,200 m.y. in age (Nagy, 1978), their presence may indicate that a  $pO_2$  of perhaps 1 to 5 percent may have been attained by mid-Precambrian time.

Finally, a word should be said regarding the evolutionary precursors of blue-green algae, the photosynthetic bacteria. Although several groups of such photobacteria occur in the extant biota, differentiated one from the another principally by the nature of their pigmentation (and thus their capability to absorb light energy in various portions of the spectrum), the important point is that, without exception, the process of photosynthesis in such microbes is obligately anaerobic. Such organisms are incapable of using water as a hydrogen source in their version of the photosynthetic reaction (rather, they commonly use  $H_2S$ , hydrogen gas, or hydrogen atoms derived from organic compounds); thus, oxygen is never produced as a byproduct of

bacterial photosynthesis and, indeed, the process cannot take place under aerobic or apparently even under microaerophilic conditions. On the face of it, this seems a paradox—photosynthesis is a light-requiring process and the photobacteria are benthonic, commonly mat-building, microorganisms; but although they require anaerobic conditions in which to carry out bacterial photosynthesis, the sediment-water interface, for which they are otherwise well adapted (and at which they are required to live if they are to absorb the maximum available solar energy), is an oxygenic environment. It would appear that to photosynthesize optimally, photobacteria must inhabit an environment that prohibits their photosynthesis from occurring! Whether stochastic or not, it seems literally incredible that evolution could be this capricious. In fact, the seeming paradox can be rather easily explained: If one assumes that a transition in atmospheric-oxygen content actually occurred during the Precambrian, then it would seem reasonable to suggest that primitive photobacteria (precursors of the present several strains) evolved prior to the appearance of oxygenic conditions, at a time when the sediment-water interface was still essentially anoxic (rather than being oxygenic, as it is at present). With the development of aerobic photoautotrophs (precursors of present-day blue-green algae), conditions at the interface began to change and, by the ecological Principle of Competitive Exclusion, oxygen-producers came to supplant photobacteria in the upper reaches of bacterial mat communities (since they produced a toxin, free oxygen, that during daylight hours served to inhibit bacterial photosynthesis); the photobacteria thus became adapted to lower, relatively anoxic reaches of the mat community, an environment that many such microorganisms inhabit to the present day.

In summary, diverse types of biochemical data seem to indicate the following:

1. Appreciable quantities of atmospheric oxygen were available prior to the development both of invertebrate metazoans (about 700 m.y. ago) and of unicellular eukaryotes (about 1,400 m.y. ago).
2. Prokaryotes, such as blue-green algae, evolved in response to (and, as is discussed below, were the oxygen-producers responsible for) a transition from essentially anoxic to oxygenic conditions during the Precambrian.
3. Earlier evolving, more primitive prokaryotes, such as the photobacteria, were adapted to an anoxic, rather than an oxygenic world.

If these evidences have been read correctly, the conclusion seems inescapable that a transition in atmospheric oxygen content must have occurred and have had a profound effect on the Earth's biota, relatively early in geologic time.

### RELEVANCE OF MODEL STUDIES OF THE ORIGIN OF LIFE

Since the early 1950's and the initial studies of Garrison and others (1951) and Miller (1953), literally hundreds of "early Earth experiments"—studies demonstrating the non-biologic formation of organic compounds under conditions simulating those believed to be present on the primitive Earth—have been carried out in scores of laboratories throughout the world. The vast majority of these have been gas-phase syntheses, experiments carried out both under highly reducing conditions (using a starting mixture of  $H_2$ ,  $CH_4$ ,  $NH_3$  and  $H_2O$  vapor; for example, Miller, 1953) and under less reducing conditions (using a starting mixture of  $CO$  or  $CO_2$  plus  $N_2$ ,  $H_2$  and  $H_2O$  vapor; for example, Abelson and Hoering, 1967) and by using a rather wide assortment of plausible energy sources (such as spark discharge, ultraviolet light, and ionizing radiation). Compounds so synthesized include amino acids, hydrocarbons, fatty acids, nitrogen-containing heterocyclics, sugars, and many other types of small molecules. The important point about these experiments is that in all such studies for which relevant data are available, such gas-phase syntheses have been reported to be inhibited by "small concentrations" or by even "traces" of free oxygen (see, for example, Abelson and Hoering, 1967).

Recently it has been demonstrated that a second type of abiotic synthesis, one involving the photocatalytic production of organic compounds on mineral surfaces (from  $CO$  or  $CH_2$  and traces of  $H_2$  vapor), is relatively unaffected by the presence of uncombined  $O_2$  (Hubbard and others, 1973). However, the yields of these apparently oxygen-insensitive, surface-dependent syntheses are quite low and, because of their inhibition by even small amounts of liquid water—the syntheses having been carried out under conditions designed to simulate those of the arid Martian regolith—they appear to be of "questionable importance on the primitive Earth" (Hubbard and others, 1975).

Thus, as currently understood, the nonbiologic synthesis of small organic compounds on the primitive Earth, the first step in the long chain of events that led ultimately to the formation of the earliest forms of life, seems to have required the near-absence of free oxygen. Of those synthetic reactions for which the mechanisms have been studied in detail, several require the presence of two specific, highly reactive intermediates: formaldehyde (e.g., an intermediate in the synthesis of sugars and of amino acids) and hydrogen cyanide (involved in the production of amino acids and of such nitrogen-containing heterocyclics as purines and pyrimidines). This latter compound is of special interest since, like carbon monoxide and hydrogen sulfide (starting materials commonly used in such syntheses),

hydrogen cyanide is lethal, even if present in only minute amounts, to many extant forms of life. From the perspective of the present, it thus seems puzzling (and, indeed, at first glance seems inexplicable) that living systems could have originated in a milieu that was in fact poisonous to the process of living. Such, of course, was not the case—both  $HCN$  and  $CO$  are lethal only to aerobic forms of life (since they react with, and deactivate, cytochromes involved in aerobic respiration), and  $H_2S$  is a compound both used as a reactant by (e.g., photobacteria) and produced as a metabolic product of (e.g., sulfate-reducing bacteria) numerous extant anaerobic microbes. Thus, the inferred involvement of these compounds in the origin of life—like the results of virtually all other experimental work that has been done in this field—is consistent with, and seems supportive of, the concept that the origin of life on Earth could have occurred only under essentially anoxic conditions.

The application of results of these "early Earth experiments" to an understanding of the primitive atmosphere is, of course, highly model dependent. Knowledge of the origin of living systems is far from complete, and even if the Oparin-Haldane model is accepted without reservation, virtually no data are available to indicate, quantitatively, the degree to which abiotic syntheses are inhibited (but perhaps not completely terminated) by various low concentrations of ambient oxygen. Still the overall model seems too sensible to be seriously questioned with the data on hand—the "early Earth experiments" seem to require essentially anoxic conditions; the abiotic organic matter of interstellar dust clouds and that occurring in carbonaceous meteorites seem certain to have been synthesized in the absence of free oxygen; and the morphologically and biochemically least complex, and thus presumably most primitive, of known living organisms—simple, heterotrophic or  $CO_2$ -using bacteria (namely, the clostridia and the methanogens; Broda, 1975)—are all obligate anaerobes. The conclusion seems difficult to escape: early in the Precambrian, at the time when life arose on Earth, the Earth's environment must have been virtually devoid of free atmospheric oxygen.

### GEOLOGIC AND PALEOBIOLOGIC CONSIDERATIONS

The foregoing discussion has dealt almost exclusively with biochemical and organic chemical evidence relating to the development of an oxygenic environment. These matters have been here treated in some detail, principally because they have been largely overlooked in much of the other geologic literature on the subject and because, taken together, they appear to me to constitute perhaps the single most telling line of evidence establishing that a transition in atmospheric oxygen content actually occurred in the geologic past.

The timing of this transition, however, is another question altogether, one that is best addressed by referral to the geologic-paleobiologic record. As is noted above, available paleobiologic data seem to indicate that the transition occurred earlier than about 1,400 m.y. ago—the suggested time of origin of the eukaryotic cell type—but how much earlier remains in question (and it should not be overlooked that some workers have suggested a much later time for the origin of eukaryotes than the date here preferred). To resolve this question, one must turn to the Precambrian rock record and a consideration of the temporal distribution and possible significance of various types of sedimentary, oxygen-sensitive mineral assemblages. Over the years, much has been written on this subject. Traditionally regarded as intertwined with the classic question of the origin of banded iron-formations, it has been a topic of debate and speculation for several decades, a discussion most recently revived in a flurry of stimulating papers by Preston Cloud (1976) and by Eric Dimroth and his associates (Dimroth and Kimberley, 1976). Indeed, the geologic literature is replete with discussions of this general problem area, and other than to expand and update ideas I have previously expressed (Schopf, 1975, 221–222), I have little new to contribute. Nevertheless, as requested by the Convener of this symposium, I will here dutifully summarize my current thoughts on the matter (but I do so only briefly, and rather reluctantly, for it seems to me that what is really needed are more data, not more verbiage).

Numerous workers have read the geologic record as indicating that a transition in atmospheric oxygen occurred roughly 2,000 m.y. ago. The principal lines of evidence usually cited are as follows:

1. "Easily oxidized" detrital pyrite and uraninite are known to occur together in placer deposits older, but not in those younger, than about 2,000 m.y. in age;
2. Red beds, interpreted as being indicative of subaerial oxidation and thus of an "oxygenic" atmosphere, are known to occur in sediments younger, but not in those significantly older, than this age;
3. The bulk of banded iron-formations of the world—deposits containing individual reserves of tens of billions of tons of oxidized iron—occur in strata approximately 2,000 m.y. in age but are extremely rare in appreciably younger terranes; the concept here is that these deposits might evidence the anoxic-oxygenic transition since their deposition is thought to require both a predominantly anoxic water column (necessary for dispersal of dissolved ferrous iron over basin-sized areas) and an at least marginally oxygenic atmosphere (with the oxygen diffusing into the upper reaches of the water col-

umn and there reacting with dissolved iron to cause precipitation of ferric and ferro-ferric oxides).

The first of these three types of evidence, that relating to detrital uraninites, is discussed in detail by David Grandstaff elsewhere in this volume (chap. C). He gives an excellent summary of his studies of the kinetics of oxidation and of deposition of the mineral species in question and of their apparent relevance to the problem at hand.

Although the second line of evidence, that relating to the distribution of red beds, is commonly cited in much of the recent literature on this problem, its significance now seems open to question since

1. the temporal distribution of these deposits is relatively poorly known (compared, for example, with the distribution of banded iron-formations; Goldich, 1973);
2. terrigenous deposits, of which continental red beds are one of many types, are generally very susceptible to geological recycling and are thus preserved only exceptionally in very ancient terranes (consequently, the present absence of red beds older than 2,000 m.y., even if firmly established, would not necessarily indicate that such deposits were originally lacking);
3. the Viking explorations of Mars have shown that red beds need not necessarily reflect the occurrence of an "oxygenic" atmosphere, at least as the term is normally used—the Martian atmosphere contains only trace amounts of free oxygen (about 0.13 percent, a value less than that required by terrestrial organisms for aerobic metabolism), all of which is apparently of inorganic, photolytic origin, an atmosphere that has nevertheless produced a vast "red bed" on the planet's surface; and
4. an increasing body of geologic data (e.g., Dimroth, 1975; Dimroth and Kimberley, 1976) seems to indicate that free oxygen (perhaps at a low  $pO_2$ , with the oxygen having been produced by photolysis of atmospheric water vapor) was available for subaqueous oxidation during the Archean—if so, subaerial oxidation to produce red beds might also have occurred.

Thus, it would appear that red beds need not be indicative of a fully oxygenic atmosphere, that the oxygen required for their formation need not be of biological origin, and that red beds might reasonably be expected to have been formed (and to have been subsequently recycled) prior to 2,000 m.y. ago.

Finally, as a corollary to the foregoing remarks regarding red beds, it seems likely that the presence of banded iron-formations need not reflect a transitional stage in atmospheric evolution. If red beds can be formed by reaction of iron with photolytically-produced ox-

ygen (as appears to have occurred on Mars), and if UV-induced photolysis of water vapor occurred in the Archean atmosphere (an occurrence dependent only on the presence of water, of solar energy, and of an exosphere temperature sufficient to insure loss of hydrogen from the gravitational field of the Earth—conditions that all seem certain to have been satisfied since very early in Earth history), then such oxygen might be expected to have reacted to produce banded iron-formation even at a low, nearly “anoxic” level of atmospheric  $pO_2$ . In contrast to continental red beds, the resulting subaqueously deposited iron-formation might be rather readily preserved in the rock record. It is conceivable, therefore, that although deposition of Archean banded iron-formations and of other oxidized facies of similar age (e.g., Dimroth and Kimberley, 1976) may have involved reaction with free oxygen, ambient oxygen concentrations could have been quite low. The mere presence of such units provides little indication of the actual  $pO_2$  and no evidence as to whether the  $pO_2$  had become stabilized (for example, at a low, photolytically-regulated level) or whether it might have been increasing over time (as would be required for such units to reflect a transitional stage in atmospheric evolution).

However, the very large volume of banded iron-formation that occurs throughout the World in strata aged about 2,000 m.y. (Goldich, 1973), and thus the consumption and burial of the huge amounts of oxygen that their deposition seems to reflect, requires the existence of an oxygen source far more efficient than that suggested above for the scattered oxidized facies known from Archean-age sediments. The key here is that measured or calculated rates of  $O_2$  production by inorganic photolysis (e.g., Brinkmann, 1969) are simply too low, by more than an order of magnitude, to account for the enormous amounts of oxygen that were entombed during this phase of mid-Precambrian iron-ore deposition (see Holland, 1973). Indeed, only one plausible mechanism capable of accounting for the required rate of oxygen production is known: aerobic (e.g., blue-green algal) photosynthesis, followed by burial (to prevent aerobic recycling) of the organic matter produced. That aerobic photosynthesizers were extant by this time seems evidenced (albeit indirectly) by the “evolutionary trees” generated from studies of biochemical evolution (for example, Schwartz and Dayhoff, 1978) and is firmly supported by the now well-known occurrence of blue-green-algalike microfossils in mid-Precambrian stromatolites (Schopf, 1974, 1975; Nagy, 1978). Moreover, the fact that such stromatolites are first known to have become abundant and widespread about 2,300 m.y. ago (Schopf, 1975), just prior to the onset of the major phase of banded iron-formation deposition, raises the intriguing possibility that this phase of diver-

sification and rapid spread of such structures may reflect the origin and early diversification of  $O_2$ -producing photosynthesizers. Thus, the mid-Precambrian banded iron-formations may simply be the geologic result of a major event in biologic history, the origin and early evolution of oxygen-producing and oxygen-utilizing microorganisms. And, as several workers have previously suggested (for example, Garrels and others, 1973; Cloud, 1974), the cessation of this major phase of iron-formation deposition, about 1,800 m.y. ago, may be a result of the essentially total consumption of accessible, easily oxidized ferrous substrates—the oceans had become “swept free” of unoxidized iron and this, together with the saturation of other oxygen sinks (e.g., the conversion of  $H_2S$  to sulfate), led to the establishment of a stable oxygenic environment.

## SUMMARY

Current concepts regarding the origin of life on Earth, concepts supported by numerous laboratory studies of the nonbiologic synthesis of organic compounds under simulated “early Earth conditions,” require that living systems originated at a time when the Earth’s atmosphere was virtually devoid of free oxygen. The present atmosphere, however, is highly oxygenic, and the present ecosystem is based largely on oxygen-requiring metabolism. At some point in the geologic past, therefore, the Earth’s atmosphere must have undergone a transition from an early, essentially anoxic stage to its present oxygenic state. Comparison of oxygen-related biosynthetic pathways, metabolic capabilities, and other relevant biochemical characteristics of invertebrate metazoans, primitive unicellular eukaryotes, and prokaryotic blue-green algae and bacteria demonstrates that there is a strong correlation between degree of organizational complexity and biochemical dependence on free oxygen. This correlation seems most logically interpreted as reflecting the evolution of these groups in response to a rise in atmospheric oxygen content during Precambrian time:

1. The most primitive forms of life, prokaryotes comparable in many respects to extant clostridial and methanogenic bacteria, were obligate anaerobes that developed during an early, essentially anoxic stage in atmospheric evolution.
2. The earliest photoautotrophs (cf. extant photobacteria), which similarly evolved prior to the development of oxygenic conditions, carried out anaerobic photosynthesis at the anoxic sediment-water interface; these microorganisms probably formed laminated, matlike communities, stratified as a function of the light-absorbing capabilities of various components of the biocoenose (and they

thus may have produced calcareous, bacteriogenic stromatolites).

3. The derivation from such photobacteria of oxygen-producing, aerobic photoautotrophs (cf. extant blue-green algae) resulted in evasion of free oxygen into the Precambrian environment and the beginning of a major change in the Earth's ecosystem. Although many contemporary anaerobes, especially those that were planktonic or those that were benthonic and nonmobile, would have been hard pressed to cope with the newly abundant, highly reactive, free oxygen (and the beginnings of oxygenic conditions may thus have resulted in an early, major episode of biotic extinction<sup>2</sup>), the totally prokaryotic biota diversified to give rise to facultative aerobes, microaerophiles, and obligate aerobes; biologically mediated, anaerobic-aerobic geochemical cycles of various elements (such as carbon, nitrogen, sulfur, and iron) became established; and photoautotroph-dominated, benthonic biological communities became stratified into an uppermost oxygenic zone (inhabited by aerobic photoautotrophs and other oxygen-tolerant prokaryotes) and an underlying anoxic zone (inhabited by facultative or obligate anaerobes, and including photobacteria in its upper portions). Thus, with the advent of aerobic photoautotrophs, photobacteria were supplanted at the sediment-water interface. Because of the absence of effective competition for photosynthetic space at this interface, the oxygen-producing, stromatolite-forming photoautotrophs spread rapidly to occupy virtually all usable, accessible niches; they thereby became the dominating element of the Earth's ecosystem and, through their photosynthetic activity (and the subsequent burial of the photosynthetically-produced organic matter) caused an irreversible rise in atmospheric oxygen content.

4. By the time of origin of unicellular eukaryotes, the Precambrian atmosphere had become oxygenic, stabilized at a  $pO_2$  similar to (but perhaps somewhat lower than) that of the present, and members of this newly evolving lineage were "wholly dependent" (for their metabolism, for important biosynthetic

capabilities and, possibly, for mitotic reproduction) on the availability of free  $O_2$ .

5. Like their unicellular progenitors, multicellular eukaryotes (e.g., invertebrate metazoans) are fundamentally aerobic, a characteristic reflecting their derivation from oxygen-dependent precursors and indicating that the Earth's atmosphere has been fully oxygenic since their origin during late Precambrian time.

This evolutionary sequence, apparently indicating that the transition in atmospheric oxygen content occurred earlier than about 1,400 m.y. ago (the suggested time of origin of the eukaryotic cell type), during the mid-Precambrian, is consistent with, and seems supported by, available mineralogic and geologic data. The occurrence of placer deposits containing easily oxidized pyrite and uraninite (the well-known uraniferous gold deposits of South Africa, Brazil, and southern Canada), in strata older, but not younger, than about 2,000 m.y., suggests that the Earth's atmosphere may have been essentially anoxic prior to this time (Grandstaff, this workshop, Chapter C). This is not to imply, however, that the atmosphere was absolutely devoid of uncombined oxygen prior to 2,000 m.y. ago. Even today, free oxygen is produced readily (albeit slowly) in upper reaches of the atmosphere by ultraviolet-induced photolysis of water vapor and the accompanying loss of hydrogen from the Earth's gravitational field. This mechanism would have been operable since the formation of the planet and the beginnings of its atmosphere, and it thus seems reasonable to suggest that this may have been the source of the free oxygen apparently required for diagenetic oxidation, and the production of such sediments as banded iron-formations, during the Archean. Studies of this inorganic process indicate, however, that it would have been insufficient to have produced the large amounts of oxygen sequestered in oxide-rich rocks formed during the major episode of banded iron-formation deposition about 2,000 m.y. ago; indeed, the amounts of oxygen required appear to be far too great to have been produced in the time involved by any known mechanism other than aerobic (e.g., blue-green algal) photosynthesis. It is thus perhaps significant that calcareous stromatolites apparently produced by blue-green algae are first known to become abundant and widespread in the geologic record in strata about 2,300 m.y. in age. Although this temporal relationship may be merely fortuitous, perhaps a result of rare preservation of older stromatolitic facies, this seemingly abrupt appearance of widespread stromatolites could reflect the origin and early diversification of aerobic photosynthesizers. If so, the few older stromatolites now known would appear to be most plausibly interpreted as being of photobacterial origin (produced by

<sup>2</sup> Recently, based on sequence data from six protein superfamilies (ferredoxin, 5S ribosomal RNA, c-type cytochromes, azurin-plastocyanin, flavodoxin, and rubredoxin) that occur in modern prokaryotes and eukaryotes, Schwartz and Dayhoff (1978) have constructed a composite "evolutionary tree," which they interpret as indicating that aerobic respiration evolved in prokaryotes prior to the advent of oxygen-producing photosynthesis. If their concept is correct, the oxygen utilized by these earliest aerobes would have to have been a product of ultraviolet-induced photolysis of water vapor; the oxygen content of the environment inhabited by these aerobes would apparently have to have been on the order of 0.2 percent or greater (the so-called "Pasteur Point," the approximate concentration of oxygen required by extant microbes for aerobic respiration); such organisms (which must have possessed oxygen-mediating enzyme systems), even if planktonic or if benthonic and nonmobile, would have been little affected in the episode of biotic extinction that may have accompanied the advent of aerobic photoautotrophy.

anaerobic photoautotrophs), and the apparent timelag between the origin of oxygen-producing photoautotrophs and the cessation of the major phase of banded iron-formation deposition (about 1,800 m.y. ago) could be interpreted as encompassing the time required for oxidation of previously unoxidized inorganic substrates and the buildup of a stable, oxygenic atmosphere.

It is important to note that several aspects of this transition are as yet poorly defined. For example, although available data seem to suggest that the transition occurred at "approximately" 2,000 m.y. ago (i.e., apparently between 2,200 and 1,800 m.y. ago), the data do not indicate whether the transition occurred slowly as a more-or-less unbroken continuum, during this period; whether it occurred in a single episode, as a relatively abrupt step-function of short duration; or whether the transition was sporadic, with atmospheric oxygen content fluctuating widely as a result of variable and essentially uncoupled rates of oxygen production (via photosynthesis and photolysis) and oxygen consumption (via oxidation of inorganic substrates and, assuming the presence of aerobic respiring microorganisms, via biological recycling of photosynthetically produced organic matter). Moreover, the ranges of atmospheric  $pO_2$  prior to and after the event are as yet ill defined (although biochemical data seem to suggest that  $pO_2$  levels on the order of 1 to 5 percent may have been attained during the mid-Precambrian), and direct evidence of several possible ramifications of the transition, such as a suggested episode of extinction of oxygen-intolerant anaerobes and the presumed inhibition of abiotic synthesis as a result of increased  $pO_2$ , has yet to be detected in the rock record. Thus, although it seems evident that such a transition in atmospheric oxygen content actually did occur during the Precambrian, and although available data suggest a "best guess" for its time of occurrence of about 2,000 m.y. ago, additional and more telling data will be needed before the timing, nature, and ecologic impact of this event can be fully elucidated.

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## DISCUSSION

*Irving Breger, U.S.G.S., Reston, Va.:* If you are assuming that photosynthetic organisms existed before 1.4 billion years ago, what protective mechanism do you assume against the lethal ultraviolet radiation?

*Schopf:* In the first place, there are very good data indicating the presence of planktonic organisms earlier than 1.4 billion years ago. There are good data for planktonic organisms back to about 2.2 or 2.3 billion. These organisms are flushed up into the top of the water column; if there were a high ultraviolet flux they would get "fried." They presumably weren't getting "fried," and one answer is that like modern organisms, these organisms must have had an effective DNA repair mechanisms for UV damage. Secondly, as I think we will hear later, there is evidence for some free oxygen as early as 3.76 billion years ago, that is the presence of oxidized iron minerals in the Isua (Greenland) iron-formation. Geologists maintain the iron minerals are primary sedimentary syngenetic. If that is the case, there was oxygen, and it is my hunch that there was always some small amount of oxygen in the Earth's atmosphere as a product of photodissociation of water vapor. This being the case, it seems to me probable that there was always some degree of an ozone shield. One other thing I might mention is a paper by Preston Cloud and Gerry Licari, dedicated to John Gruner, in the *American Journal of Science* about three years ago, on filamentous microorganisms presumed to be blue-green algae in the Pokegama Quartzite, in which Cloud and Licari show that the organisms occur in re-entrants in an erosion surface. These organisms are sitting there on an erosion surface not underneath the water, only a couple of centimeters down. The Pokegama underlies the Biwabik and the Gunflint and is probably around 2.2 billion years old. So again, it seems to me probable that UV flux was not a major problem at that point in Earth's history.

*Hallbauer:* You have talked about only very simple microorganisms so far, but you didn't mention the carbonaceous material in the Witwatersrand, where there are branched filaments and septate filaments and forms which could be called megascopic fossils. And I might mention here that E. S. Barghoorn of Harvard recognized these forms as megascopic fossils. Where could they fit in? Apparently there

are different mechanisms in the evolution of life which should fit the model.

*Schopf:* Unlike Professor Barghoorn, I have not seen the material studied by Dr. Hallbauer—my comments can be based only on what I have read of Dr. Hallbauer's work and on the photographs and photomicrographs he has published. As you know, he has interpreted these structures to be fossil lichens, and as such, he has interpreted them to contain a fungal, and thus eukaryotic, component. If his interpretation is correct, it would then follow that there must have been appreciable quantities of free oxygen available during the time of deposition of the Witwatersrand, since it would be difficult to envision fungi either originating or surviving under essentially anoxic conditions, and this would set the transition to an oxygenic environment appreciably earlier than the 2,000 m.y. ago that I have suggested as being most plausible. However, the photomicrographs published by Dr. Hallbauer lead me to suspect that the structures he has discovered have probably been misinterpreted—specifically, they have a branch pattern that appears to me to be quite unlike that known to occur in any modern fungi, and, in fact, in any modern thallophytes of which I am aware. I thus suspect that these structures are most likely to be nonbiologic and, thus, that they have no obvious bearing on the time of origin of oxygenic conditions. I hate to disagree, but it seems to me that it just doesn't fit in with everything else that is known in the Pre-Cambrian. On the other hand, that doesn't mean that it is wrong; it just means that it doesn't fit in and maybe you're looking at a different facies. If what you are saying is correct, that these are lichens, 2.5 billion years in age, and if as lichens they have a fungal component, and if those fungi had the same metabolic features as modern ones, then there must have been appreciable quantities of atmospheric oxygen, above 1½ percent of present atmospheric levels. This seems to me not to fit in with a lot of other data, but on the other hand, if they are fungi, it's of very considerable interest obviously.

*Kalliokoski, Michigan Technical University:* There are a series of things that are being called fossils—these are little clusters of pyrite grains. They are very common in the recent record, and they can be chased back into the Precambrian. The question is, do you consider these to be fossils?

*Schopf:* If I may, without meaning to be facetious, yes and no. Sedimentary pyrite in general is evidence of biologic activity in the sense that the hydrogen sulfide produced by sulfate-reducing bacteria can combine with iron and you can get pyrite

precipitated. On the other hand, Bob Sweeney has shown one can get similar structures without the intervention of biologic activity. It is well established that pyrite can infill cells, and not only of microscopic algae but of forams and a variety of other organisms; even the epidermal cells of leaves are sometimes infilled by pyrite. Here, apparently, the organic matter is used by bacteria for local production of  $H_2S$ , the hydrogen giving the reducing power which reduces sulfate in a marine environment and thus pyrite is precipitated within cells. In that case they are fossils, but I would be very hesitant to say that all frambooidal pyrite could be properly interpreted as Precambrian fossils or fossils generally.

*Skinner:* I don't think this is going to be answered by anybody else, Bill, so I hope you'll have a shot at it. If you do in fact have large amounts of abiologic carbon back in the early Precambrian, should you not see some progressive change in the carbon isotopic fractionation in this material?

*Schopf:* That's a good question to ask Manfred Schidlowski.

*Schidlowski:* Actually we had a look at the carbon isotope composition of sedimentary carbonates from the very start. Now we must say from almost the very start because we did not include the Greenland material. The delta  $C^{13}$  values of the carbonates were practically constant, always near zero per mil.

*Schopf:* I would say one other thing. Since Bill Ruby's paper in the early fifties, people have been doing mass balances on carbon and oxygen in the geologic record and using the relation between them as indicating that it is a plausible and consistent model to argue that all of the organic carbon that is sequestered in the Earth's crust could come from photosynthesis and therefore that all of the oxygen in the Earth's crust and in the Earth's atmosphere and oceans could also come from photosynthesis.

Mass balance calculations seem to be within an order of magnitude and seem to make good sense. It turns out also that if you look at those numbers, and I've just redone that whole business again, the new data give you a better hunch as to what the true numbers might be. These data also allow you to have half of the organic matter sequestered in the Earth's crust from other than photosynthetic processes. I think what the data say is that there is a big slop in the data. If you want to argue that it is all from green-plant photosynthesis, you can argue that way. On the other hand, if you want to point out that there is a big slop in the data, that they are not all that reliable, you're allowed at least up to half the organic matter in the Earth's crust from other than photosynthesis. And if there was a large amount of abiotic synthesis going on or there was a fixation by organisms that were photosynthetic but non-oxygen producing, the mass balance equations allow you to do that. They certainly do not rule it out, and it is as logically consistent with the data as the other hypothesis that says it all comes from oxygen-producing photosynthesis.

*James Trow, Michigan State University:* Two short questions. First, do you know the age when a lot of these carbon beds show up, and how far down in the stratigraphic section do those carbon-bearing beds occur? Second, do you have evidence of any native sulfur beds in these rocks?

*Schopf:* With regard to the first question, there certainly are graphitic schists that go way back into the Archean, the oldest ones of which I am aware, highly carbonaceous material, come from the Swaziland sequence. I don't know of comparably highly carbonaceous material from the Isua iron-formation region, although there is graphite and there is organic matter in those rocks which are reliably dated at older than 3.76 b.y. With regard to sulfates and sulfur deposits, I do not know the distribution of those in the Precambrian.



# Uraninite Oxidation and the Precambrian Atmosphere

*By* D. E. GRANDSTAFF

GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN  
QUARTZ-PEBBLE CONGLOMERATES

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GEOLOGICAL SURVEY PROFESSIONAL PAPER 1161-C





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# GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN QUARTZ-PEBBLE CONGLOMERATES

## URANINITE OXIDATION AND THE PRECAMBRIAN ATMOSPHERE

By D. E. GRANDSTAFF<sup>1</sup>

### ABSTRACT

Although uraninite is thermodynamically unstable at oxygen pressures greater than approximately  $10^{-21}$  atmospheres, calculations based on the dissolution kinetics suggest that uraninite from the Witwatersrand Sequence may have survived as a detrital mineral at oxygen pressures as high as  $10^{-2}$  to  $10^{-6}$  of the present atmospheric level (PAL). Therefore, the mid-Precambrian atmosphere may have contained small amounts of free oxygen, produced by aerobic photosynthesis and photodissociation of water vapor.

### INTRODUCTION

Detrital uraninite is extremely rare in modern sediments because it is rapidly destroyed by oxidation. A search of black sands for uraninite during World War II failed to reveal any uraninite in recent placers. In contrast, several Precambrian sequences contain uraninite that many geologists believe is detrital (Davidson, 1965). Of these, the Witwatersrand Triad of South Africa and the Elliot Lake Group-Huronian Supergroup of Canada are the best known and documented occurrences. Although C. F. Davidson (1960, 1965) and others have supported a hydrothermal origin for the Witwatersrand and Elliot Lake ores, a detrital-placer origin now seems well supported by the available evidence.

The occurrence of detrital uraninite has been cited as evidence that oxygen probably was much less abundant in or absent from the Precambrian atmosphere (Randardohr, 1958). Holland (1962) calculated that uraninite would be thermodynamically stable only at oxygen pressures less than  $10^{-21}$  atmosphere. However, photosynthesis by blue-green algae and the photodissociation of water vapor may have contributed significant amounts of oxygen to the Precambrian atmosphere (Berkner and Marshall, 1965; Brinkmann, 1969). Although oxygen may have been rapidly consumed by oxidation of reduced volcanic gases and iron and

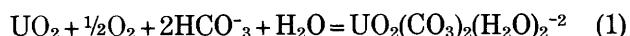
other reduced species in sediments, it seems likely that some free oxygen was present in the mid-Precambrian atmosphere.

It is well known that minerals may exist for appreciable periods out of equilibrium with their environments. Magnetite, for example, should be oxidized to hematite or some other ferric compound under Earth surface conditions. Magnetite survives as a common detrital mineral because its oxidation rate is quite slow under oxidizing surface conditions. Although the oxidation of uraninite is much faster, modern occurrences of detrital uraninite have been reported at Nation River, British Columbia (Steacy, 1953), Cornwall, England (Davidson and Cosgrove, 1955), Hogatza placer, Alaska (Staatz, this volume, chap. Y) and the Indus River, Pakistan (Zeschke, 1959, 1961). These reports indicate that some disequilibrium may prevail. Estimates of the amount of disequilibrium have not been made, however, because of lack of knowledge of the kinetics and rate of uraninite dissolution and the rate of geological processes (Rutten, 1971).

The purpose of this paper is to use experimental data on the kinetics of uraninite dissolution in order to make a more quantitative estimate of the upper limits of oxygen in the Precambrian atmosphere that would allow the survival of uraninite as a detrital mineral during periods of weathering, erosion, transportation, deposition, and burial and the formation of the uraniferous conglomerate of the Witwatersrand, Elliot Lake and other similar deposits.

### EXPERIMENTAL DATA

The oxidation of uraninite (represented by  $\text{UO}_2$ ) in water containing carbonate species at intermediate pH probably takes place according to the ideal formula



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Experimental data and discussion of the kinetics of uraninite dissolution have been reported in detail elsewhere (Grandstaff, 1973), and only a brief summary is given here. The dissolution of uraninite is represented by the following kinetic equation

$$R = \frac{-d(\text{uran})}{dt} = 10^{20.25} (SS) (RF)^{-1} (10^{-3.38-10.3} NOC) (2)$$

$$(a_{\Sigma\text{CO}_2})(\text{D.O.})(a_{\text{H}_+}) \exp(-7045/T) (\text{day}^{-1})$$

in which

$R$  = rate of the dissolution reaction, the diminution of uraninite as a function of time ( $-d(\text{uran})/dt$ )

$SS$  = geometric specific surface area ( $\text{cm}^2\text{g}^{-1}$ )

$NOC$  = nonuranium cation mole fraction

$\text{D.O.}$  = dissolved oxygen concentration (ppm)

$\Sigma\text{CO}_2$  = total dissolved carbonate concentration (Molar)

$$\Sigma\text{CO}_2 = [\text{H}_2\text{CO}_3] + [\text{HCO}_3^{-1}] + [\text{CO}_3^{2-}]$$

$T$  = absolute temperature ( $^{\circ}\text{K}$ )

$RF$  = organic retardation factor. Retardation factors of 5 and 35 have been found in a New Jersey river water and in coastal sea water.

#### FACTORS IN THE SURVIVAL OF PRECAMBRIAN URANINITE

It can be seen from the experimental data that the survival of uraninite as a detrital mineral is influenced by environmental and physiographic factors, such as pH, temperature, presence of organic species, rates and time required for erosion, transport, deposition, and burial; mineralogical factors, such as chemical composition and grain surface area; and atmospheric factors, such as oxygen and carbon dioxide partial pressures.

A schematic representation of the rate of dissolution of uraninite during the sedimentary cycle is shown in figure 1. Uraninite grains deep in the rock probably are not subject to significant attack by oxidation. Oxidation of organic matter, sulfides, and iron-bearing silicates, as well as of uraninite, depletes the ground water of oxygen. However, as erosion proceeds, the uraninite is brought closer to the surface, and the content of oxygen in the water and the rate of dissolution increase. At the surface the rate of dissolution is defined by atmospheric and environmental conditions. Dissolution of uraninite is quite rapid during periods of residence in acidic soil profiles. During transportation the rate of dissolution is somewhat slower due to the higher alkalinity and higher pH generally encountered in alluvial sediments and river waters. Finally, after deposition, the rate of dissolution decreases as the grain is buried and the concentration of oxygen in the ground water decreases.

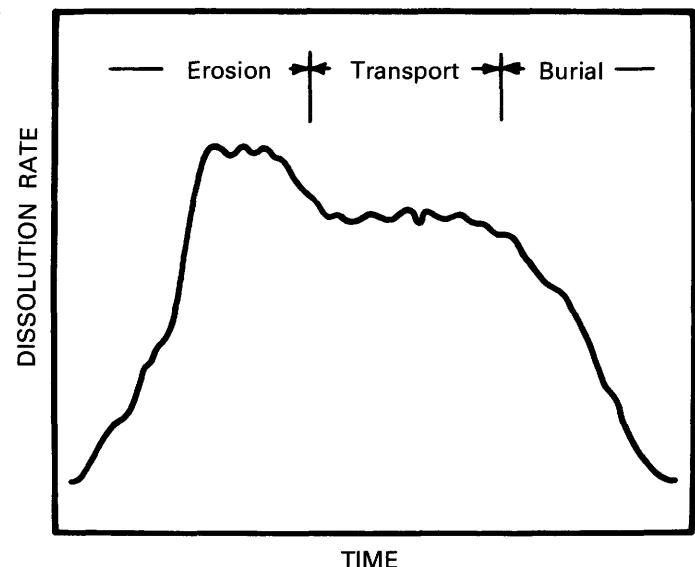


FIGURE 1.—Schematic representation of the relative rate of dissolution of uraninite during the sedimentary cycle.

The experimentally derived reaction equation (2) may be represented as

$$R = \frac{-d(\text{uran})}{dt} = R[f(SS, NOC, RF, \dots, t)] \quad (3)$$

in which the instantaneous reaction rate, the diminution of uraninite with time by chemical corrosion ( $-d(\text{uran})/dt$ ) is equal to a rate function  $R[f]$ , given by equation (2), and itself a function of time.

Rearranging the equation

$$-d(\text{uran}) = R[f]dt \quad (4)$$

and integrating

$$DF = \int R[f]dt \quad (5)$$

in which  $DF$  is the fraction of the original grain destroyed by oxidation. To simplify calculations, and in the absence of data on exact variations in environmental conditions during the Precambrian sedimentary cycle, the complex dissolution rate function shown in figure 1 was approximated by a summation. The elements of the summation represented the rate of dissolution during periods of weathering, transportation, and burial.

The rate equation may be written as a summation

$$DF = \sum_i R'_i t_i \quad (6)$$

in which  $R'_i$  is the average rate of dissolution in, and  $t_i$  is the duration of, the  $i$ th period. If the rate term is written as an explicit function of atmospheric oxygen, for example, the summation terms will have the form

$$P_{\text{O}_2} \sum_i R'_i t_i$$

If the oxygen concentration is constant during some geological process, it, and other such constant terms,

may be separated from the summation or integral to produce

$$DF = P_{O_2} \sum_i R'_i t_i \quad (7)$$

This equation may be rearranged to evaluate for  $P_{O_2}$ , for example

$$P_{O_2} = \frac{DF}{\sum_i R'_i t_i} \quad (8)$$

Calculation of the maximum concentration of oxygen in the Precambrian atmosphere that would have permitted the survival of detrital uraninite and the deposition of the uraniferous conglomerates requires that as many of the environmental, mineralogical, and atmospheric variables as possible should be fixed and limits placed on the others.

#### COMPOSITION

The composition of the uraninite is quite an important variable in its oxidation kinetics. Unfortunately, the composition and the rate of oxidation measured on the uraninite as it exists today are probably not representative of the composition and the rate of oxidation of the uraninite at the time of weathering, transport, and deposition, more than 2 b.y. (billion years) ago. Some estimate of the composition of the uraninite at the time of sedimentation must be made. Several analyses of uraninite from the Witwatersrand and Elliot Lake deposits are given in table 1.

TABLE 1.—Chemical compositions and NOC (nonuranium cation) values of some uraninite samples from the Witwatersrand and Elliot Lake

	Blyvooruitzicht (1)	Sub Nigel (1)	Dominion Reef (2)	Nordic (3)
UO <sub>2</sub>	—	—	17.10	22.7
UO <sub>3</sub>	—	—	46.19	42.6
U <sub>3</sub> O <sub>8</sub>	74.10	66.65	—	—
ThO <sub>2</sub>	2.70	1.63	6.52	6.1
PbO	16.70	19.44	14.85	22.6
REE	1.00	2.13	3.43	4.0
ZrO <sub>2</sub> + TiO <sub>2</sub>	1.40	1.04	.50	.0
SiO <sub>2</sub>	1.78	2.07	1.43	.0
MgO	.16	.30	.11	.0
CaO	.81	1.54	1.30	.7
Fe <sub>2</sub> O <sub>3</sub>	.84	1.81	1.12	.2
Al <sub>2</sub> O <sub>3</sub>	1.33	.99	.08	.0
S	—	—	1.26	.9
H <sub>2</sub> O <sup>+</sup>	—	—	2.50	—
SnO <sub>2</sub>	—	—	1.43	—
Other	—	—	.62	—
Total	100.82	97.60	98.44	99.8
NOC:				
Minimum	.13	.17	.21	.15
Maximum	.19	.23	.24	.17

References: (1) Liebenberg (1957), (2) Hiemstra (1968), (3) Grandstaff (1973).

Unfortunately, the uraninite analyses from Liebenberg (1957) and Hiemstra (1968) were made on uraninite concentrates by wet-chemical methods.

Cassiterite, monazite, leucoxene, hydrated iron oxides, quartz, and sericite were probably present as impurities (Liebenberg, 1957; Hiemstra, 1968). Therefore, the analytical results for SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, Fe<sub>2</sub>O<sub>3</sub>, SnO<sub>2</sub>, H<sub>2</sub>O<sup>+</sup>, S, ZrO<sub>2</sub>, and TiO<sub>2</sub> reported in these analyses were excluded as present primarily in impurities. The remaining elements were recalculated to 100 percent. Much of the lead was present as galena inclusions in the uraninite rather than in the uraninite lattice. However, it was included in the elements retained because the lead undoubtedly had its origin in the radioactive decay of uranium and thorium.

A first approximation of the composition of the uraninite can be made if the lead present in the chemical analyses is reconverted to uranium and thorium. The analysis calculated will approximate the composition of the uraninite at the time of crystallization if primary lead were negligible. Then, knowing or assuming an age for the uraninite at the time of erosion, the amount of lead created by radiogenic decay during that period may be calculated. This composition will approximate the composition of the uraninite at the time of erosion and deposition if there have been no major postdepositional changes in the composition of the uraninite other than that of radioactive decay of uranium and thorium.

The age of crystallization of uraninite from the Dominion Reef is approximately  $3,100 \pm 100$  m.y. (million years) (Nicolaysen and others, 1962). An extrusive in the Dominion Reef has been dated at  $2,800 \pm 60$  m.y. (Niekerk and Burger, 1969). Therefore, the age of the uraninite at the time of deposition of the Dominion Reef must have been approximately  $300 \pm 120$  m.y. The Witwatersrand System overlying the Dominion Reef System must be somewhat younger. In the absence of dates on the time of sedimentation of the Witwatersrand System, the age of this system can only be specified as between 2,800 m.y., the age of the underlying Dominion Reef System, and 2,300 m.y., the age of the overlying Ventersdorp System (Niekerk and Burger, 1966). Assuming the uraninite from the Witwatersrand System had an age of crystallization of 3,100 m.y., the uraninite probably had an age of  $550 \pm 250$  m.y. at the time of deposition of the Witwatersrand System.

If the age of crystallization of the Elliot Lake uraninite is approximately 2,500 m.y. and the age of the Matinenda Formation is greater than  $2,288 \pm 87$  m.y. (Roscoe, 1969), the age of the Elliot Lake uraninite at the time of deposition was approximately  $150 \pm 150$  m.y.

The results of the calculations are shown in table 1. Values of NOC for the samples lie between 0.13 and 0.24. Variation of NOC in each sample is a result of the uncertainty in the age of the uraninite at the time of erosion. Because of the number of operations and assumptions involved in the calculations of NOC at the time of

sedimentation and the possibility that the samples from which these analyses were made are heterogeneous (Grandstaff, 1975), these figures are only approximate.

The uraninite most sensitive to the Precambrian atmosphere, that is, the fastest reacting uraninite, the uraninite having the lowest *NOC* content, is that from Blyvooruitzicht. This uraninite has a probable range of *NOC* from 0.13 to 0.19. An average value of 0.16 has been used in calculations. Use of other permissible *NOC* values in the range 0.13 to 0.19 would affect the dissolution rate and the oxygen pressures calculated by about a factor of three at the extremes.

#### SIZE, SHAPE, AND ROUNDING

The source areas of the Elliot Lake and the Witwatersrand uraninite have not been located. In the absence of unweathered uraninite from the source areas for comparison, it is not possible to assess directly possible mineralogical changes in grain size, shape, and composition that might have occurred as a result of chemical corrosion or abrasion.

Liebenberg (1957) measured the size of uraninite grains from the Witwatersrand in polished sections. He found a very small range of size and determined that the average diameter of the uraninite grains in the Witwatersrand Sequence is about 75  $\mu$  and that in the Dominion Reef and Ventersdorp Contact Reef it is about 100  $\mu$ . Roscoe (1969) found that uraninite grains in Elliot Lake are about 100  $\mu$  in diameter. Since the measurements were made on polished sections, the actual dimensions of the uraninite grains must be somewhat larger. Although uraninite grains from pegmatites may be quite large, the small size variations of uraninite grains in the Precambrian deposits and, particularly, the absence of any larger grains in the Witwatersrand and Elliot Lake deposits suggest that the uraninite grains were never much larger than they are now. Furthermore, some of the grains are euhedral or subhedral; they retain what appear to be original crystal faces. Since physical attrition and chemical corrosion would tend to destroy the planar crystal faces, these euhedral or subhedral crystals must be close to their original size. This suggests that only relatively small variations in the mineralogical parameters occurred. Therefore, as a first approximation, these factors are assumed constant and are evaluated by examination of existing uraninite samples from the Witwatersrand and Elliot Lake.

Seventy-five micrometers have been chosen as a low-average particle dimension. The exact specific surface area cannot be calculated since this requires a precise knowledge of the particle shape of each grain. The particles as seen in polished section are intermediate in

shape between circles and squares, implying that the particle shape is between spheres and cubes, and a specific surface area of  $90 \text{ cm}^2 \text{g}^{-1}$  has been estimated on this basis.

An additional constraint is imposed by the degree of rounding and the survival of the uraninite grains. The degree of rounding of the grains varies markedly, even within single samples. Some grains are angular or subangular. The shape of these grains appears to have undergone very little modification by mechanical abrasion or chemical corrosion. Other grains are subrounded to rounded - often referred to as "muffin shaped" (Schidlowksi, 1966) - and their shape appears to have undergone extensive modification.

The literature on interpretation of the rounding of the uraninite is quite extensive (Hiemstra, 1968). Liebenberg (1957) believed that the grains had a water-worn appearance. Hiemstra (1968) found that uraninite from the Dominion Reef System had a mean roundness value in accordance with those of monazite and zircon grains in the same samples. However, Koen (1961) and Davidson (1958) believed that the uraninite had too high a degree of rounding to be consistent with a granitic or pegmatitic origin.

The descriptions of Hiemstra (1968) and Liebenberg (1957) suggest that angular and subangular grains are more common in the Dominion Reef than in the main Witwatersrand System. Hiemstra (personal commun., 1973) believed that uraninite from the Dominion Reef System had a higher degree of angularity than uraninite from the Witwatersrand or Ventersdorp Systems.

This apparent variation in rounding may be due to physical characteristics of the grains, normal distribution of mechanical processes, reworking, and variation of the period of exposure of the uraninite to oxidizing conditions. It seems possible, however, that some rounding is influenced by compositional variations.

Rounding of grains during weathering and erosion may occur by two processes, (1) mechanical abrasion and (2) chemical corrosion and dissolution. Both processes will be active, but one process may dominate. If the amount of material removed by dissolution is much less than that removed by mechanical abrasion, the shape of the grain will be consistent with mechanical abrasion. If, however, the amount of material removed by dissolution is of the same order as or greater than that removed by mechanical abrasion, the grain will be more highly rounded than is indicated by a purely mechanical model.

At the time of erosion the uraninite from the different ore reefs probably had quite different chemical compositions. This probably would have affected the rate of chemical corrosion. For example, if extreme values of *NOC* (table 1) are used, the average Blyvooruitzicht uraninite may have dissolved about fifteen times faster

than the average Dominion Reef uraninite. If the two uraninite samples were exposed to oxidizing conditions for approximately the same length of time, about fifteen times more material would be dissolved from the Blyvooruitzicht uraninite. If oxygen pressures were fairly high and the absolute velocity of the dissolution reaction were fast, this could have resulted in appreciable chemical corrosion and rounding of the Blyvooruitzicht uraninite. The Dominion Reef uraninite, however, having a slower reaction rate resulting in less chemical corrosion would have higher angularity.

Therefore, highly resistant grains, having high NOC content, may exhibit degrees of attrition and rounding consistent with purely mechanical processes. This appears to be the case in the Dominion Reef and perhaps the Elliot Lake uraninite grains. Less refractory grains or grains exposed to oxygenated environments for long periods of time would exhibit roundness values greater than those expected from purely mechanical attrition. This does not necessarily indicate that they were deposited colloidally, but that they have undergone a high degree of chemical corrosion in addition to physical attrition.

To estimate the composition of the Precambrian atmosphere, some estimate of the degree of chemical rounding should be made. Unfortunately, accurate estimates of the amount of material removed from the uraninite grains by chemical corrosion cannot be made because the initial size and shape of the grains and the proportion of physical attrition to chemical corrosion are not known. If, however, the particles are near their original dimensions, an estimate of the maximum degree of chemical corrosion may be made. The small grain-size variation and the presence of euhedral uraninite grains suggest that, in general, the grains were not reduced appreciably in size by corrosion or attrition. If a sphere is created from an originally cubic particle with the same dimension or diameter, nearly 50 percent of the original particle must be removed. This factor of 50-percent destruction of the original grain by chemical corrosion has been used in calculations. This amount of corrosion is probably a maximum, since few of the uraninite grains are spherical and since both physical and chemical attrition are involved.

#### PRECAMBRIAN ORGANISMS AND ORGANIC MATTER

Prokaryotic bacteria and blue-green algae were probably the only organisms present during the early and middle Precambrian (see, however, Hallbauer, this volume, chap. M). These organisms were undoubtedly

present in the oceans, and the remains of some of them may be preserved in marine and deltaic sediments of the Witwatersrand and Elliot Lake. It is not known, however, if the organisms were present on land.

Berkner and Marshall (1965) suggested that at low atmospheric oxygen partial pressures the flux of solar ultraviolet radiation may have been sufficiently intense to kill any organisms exposed on land. However, the "lethal level" of ultraviolet radiation proposed by Berkner and Marshall may be too low (Godward, 1962). If Precambrian organisms had higher tolerance to ultraviolet radiation than has been proposed, or if oxygen pressures were higher than proposed, it is possible that microorganisms such as algae or bacteria did form a ground cover during the Precambrian. Even if ultraviolet radiation was at lethal levels, some organisms may have lived in niches in rock or deep in soil zones (Fischer, 1965).

The presence of organic matter in water decreases the rate of uraninite dissolution (Grandstaff, 1973). The organic matter may bond to the surface of the uraninite, blocking oxidation sites. Chave (1965), Chave and Schamlz (1966), and Suess (1970) found that polar organic matter formed surface complexes on calcium carbonate grains and that these complexes decreased rates of nucleation, growth, and dissolution. Suess (1970) found that the surfaces on calcite and dolomite grains became saturated with absorbed organic matter even at very low concentrations of dissolved organic matter. Suess (1970) estimated that ocean water contains approximately 1,000 times as much organic matter as necessary to saturate the calcite surfaces. The distribution of organic matter between the surface of uraninite and water is not known. It seems likely, however, that Precambrian river and ocean water contained dissolved organic matter and that some organic matter was absorbed on uraninite surfaces. This would result in a decrease or retardation of the dissolution rate.

The type and concentration of organic matter in water probably influence the degree of retardation of the dissolution rate (Grandstaff, 1973), but the type and concentration of organic matter in Precambrian river and ocean water and the extent of possible retardation are unknown. It has been found (Grandstaff, 1973) that dissolution rates were decreased by a factor of 35 times in New Jersey coastal sea water, while the rate in water of the Molluca River in New Jersey was decreased by about 5 times. In the absence of knowledge of Precambrian retardation factors, the retardation factors of 5 and 35 times found in modern rivers and the ocean have been used as a first approximation.

#### ATMOSPHERIC CARBON DIOXIDE PRESSURE

It does not appear possible to specify the pressure of carbon dioxide in the Precambrian atmosphere with any certainty. The carbon dioxide pressure in the atmosphere appears to be controlled by equilibrium between magnesium carbonate and magnesium silicate systems, by rates of input of  $\text{CO}_2$  into the atmosphere by volcanic exhalation and biological respiration, and by removal from the atmosphere by photosynthesis, weathering, and precipitation (Holland, 1968).

Calculations suggest that the Precambrian carbon dioxide pressure may have been as much as 10 to 100 times PAL (present atmospheric level) (Holland, 1968), but Cawley, Burruss, and Holland (1969) have suggested that the carbon dioxide pressure in the pre-Silurian atmosphere may have been as low as 5 PAL. Therefore, limits of 5 and 100 PAL have been used in the present calculations.

#### TEMPERATURE

Temperature has been regarded as a factor in the survival of detrital uraninite. Since reactions are generally slower at lower temperatures, glacial or periglacial conditions have been cited as environments in which the survival of uraninite would be more probable. However, decrease in the temperature from  $25^\circ\text{C}$  to  $0^\circ\text{C}$  only decreases the reaction rate by a factor of three. Although this aids the survival of uraninite, it is probably not the major factor, since much greater reduction of the oxidation rate is probably required to permit the survival of uraninite to the extent observed in Precambrian sediments.

However, weathering in glacial or cold environments is almost entirely mechanical, resulting in minimal chemical attack on mineral grains. The liberation of uraninite from the parent rock by glaciers is probably responsible for the survival of the present Alaskan, Canadian, and Pakistani occurrences of detrital uraninite.

Tilloids have been described in association with the Witwatersrand (Wiebols, 1955) and Elliot Lake deposits (Casshyup, 1969). The relative chemical and mineralogical maturity of the Witwatersrand and Elliot Lake rocks suggests, however, that most of the sediments were probably not of glacial origin. Therefore, glacial action seems unable to account for the survival of the Precambrian uraninite.

If, however, a glacial origin for part of the Witwatersrand and Elliot Lake Sequences is accepted, then at least some of the radioactive quartz-pebble conglomerate may have been deposited in a moderate or cold climate, under periglacial or interglacial conditions. A

temperature of  $15^\circ\text{C}$  has been used in the calculations. Variation in the chosen temperature by  $10^\circ$  in either direction would produce a change in the dissolution rate by less than a factor of two.

#### WEATHERING AND DENUDATION

Actively eroded uplands, even in the tropics, yield fresh mineral grains of relatively unstable silicates and oxides mixed with products of chemical weathering (Van Houten, 1972). Such grains apparently are derived from scouring of bedrock by rivers, and the grains are mechanically released from the rock without undergoing significant chemical weathering.

Brock and Pretorius (1964) suggested that the source areas surrounding the Witwatersrand were undergoing uplift along basin margin faults. Frarey and Roscoe (1970) and Pienaar (1963) have suggested that the deposition of the conglomerates containing the uraninite in Elliot Lake occurred after uplift of the source area. Under such conditions rivers in the source area would be rejuvenated, moving old, mature detritus from the source area and downcutting into fresh rock. Under these conditions uraninite could be scoured from bedrock by rivers without significant chemical attack.

Glaciers may also deliver virtually unweathered grains to rivers, where they may be mixed with chemically weathered minerals. Although glacial derivation of the uraninite in Witwatersrand and Elliot Lake remains possible, it appears unlikely.

If uraninite was not scoured from bedrock by rivers or glaciers it must have been derived by way of soil profiles.

Estimates of denudation rates range from 0.5 cm/1,000 years near Hudson Bay (Holmes, 1965) to as much as 10 m/1,000 years in tectonically active areas (Gilluly, 1949). Ahnert (1970) found that the rate of denudation in large midlatitude basins is related to the mean elevation, and he calculated rates of summit denudation as high as 60 cm/1,000 years in the Alps. Rates of denudation as high as 100 cm/1,000 years have been calculated for the Himalayas (Leopold and others, 1964).

Soil profiles and leached zones vary widely in depth. Under tropical climates in tectonically inactive areas leached zones may reach 50 or 100 m in depth (Leopold and others, 1964). On slopes and in cold climates the soil zone may be only a thin film.

Figure 2 indicates probable residence times of grains in a soil profile under various conditions of denudation rate and soil-profile depth, assuming a steady-state soil profile. In mountainous areas after tectonic rejuvenation, rates of denudation would have been fairly high,

several tens of centimeters per thousand years. On mountainous slopes and in the absence of vascular plants to hold soil formed, soil and leached zones could have been fairly thin. Under these conditions, the diagram indicates that particles would probably reside in soil zones for periods on the order of 500 to 10,000 years.

#### TRANSPORT

The source areas of the uraninite in Elliot Lake and the Witwatersrand have never been discovered. Although extensive nonmarine deposits may occasionally be found hundreds of kilometers from their source areas (Stanley and Wayne, 1972), if a molasse model is used (Reinike, 1930), the source area of the sediments was probably within 100 km. Conglomerates in the Alps, Siwaliks, and the Andes are probably deposited within 100 km of their source areas. Brock and Pretorius (1964) have suggested that the source areas for the Witwatersrand lie immediately to the north, presently buried under the Bushveld, to the west or northwest of Klerksdorp, and to the south of the basin buried under the Karroo.

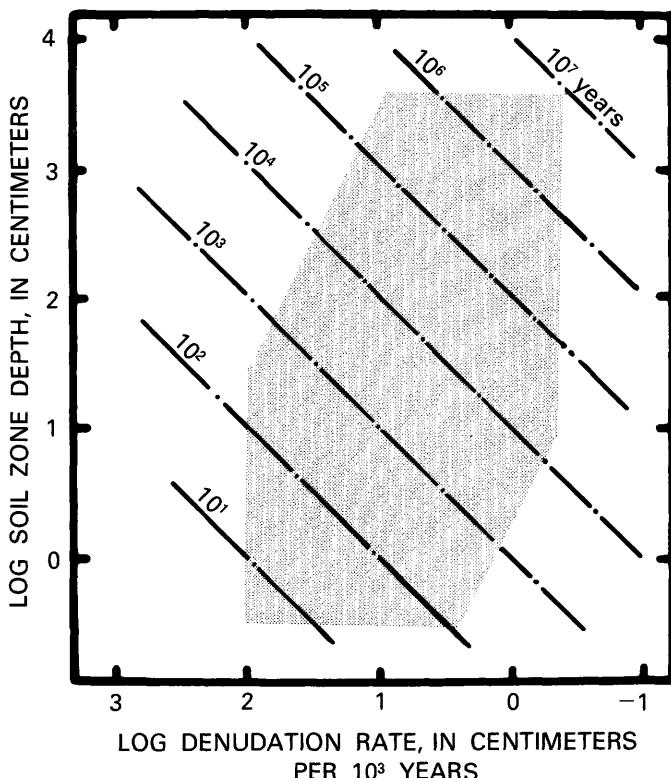


FIGURE 2.—Residence time of particles in a soil profile, assuming a steady-state soil profile. The range of most geological conditions is shown by the shaded area; residence times are given by diagonal lines.

Sediment grains in rivers may be carried at various velocities, depending on the grain size, the velocity of the flow, the bed configuration, and other factors (Leopold and others, 1964). Uraninite grains of the Witwatersrand and Elliot Lake have diameters on the order of 0.1 mm. These grains are hydraulically equivalent to quartz grains of 0.3 to 0.4 mm diameter (McIntyre, 1959). Therefore, they probably formed part of the bedload.

During transport down rivers, sediment grains may undergo deposition and re-entrainment by the river many times. In entrenched rivers, transport of grains may be fairly continuous, but in rivers having broad flood plains, a sediment grain may be abandoned in alluvium for considerable periods of time by shifts of the river channel. This increases the residence time of the particle in the river system.

For periods of transport, assuming steady state, an approximate velocity of bedload transport may be calculated by the equation

$$V = \frac{dM/dt}{\rho WD} \quad (9)$$

in which  $V$  is the velocity of transport,  $dM/dt$  is the discharge of bedload sediments,  $\rho$  is the density of the sediments, approximately  $2.6 \text{ g cm}^{-3}$  (Judson and Ritter, 1964),  $W$  is the width of the channel or portion of the channel, and  $D$  is the depth of sediment being transported.

It appears quite difficult to estimate accurately the depth of sediment being transported by the river. In some cases this depth may be estimated by using the height of sand waves or sand bars developed by grain movement.

A sand wave (also called a sand dune or bar) is a ridge that is formed on the bed of a river by movement of bed material and that is usually approximately normal to the direction of flow (Lane, 1947). Movement of the bedload takes place when grains are removed from the dune in regions of high flow and redeposited in regions of lower flow. The size of the dunes depends on the flow velocity and the sediment size (Morisawa, 1968). The full scale of variation in size within the range of conditions in rivers is not known. Dunes approximately 2 m high and 100 m wide have been found in the Columbia River (Jordan, 1962), and dunes as much as 10 m high and 150 m wide, in the Mississippi River (Carey and Keller, 1957). If the sand waves are approximated as triangular solids, the equivalent depth of sediment moved in the sand waves is one half of the wave height. Therefore, the depth of sediment moved by the Mississippi River in sand waves is as much as 5 m.

The suspended-sediment discharge of the Mississippi River is approximately  $3 \times 10^{14}$  g yr<sup>-1</sup> (Judson and Ritter, 1964). Fisk (1944) estimated that the bedload of the Mississippi River is approximately 7–10 percent of the suspended load. Therefore, the bedload discharge is about  $3 \times 10^{13}$  g yr<sup>-1</sup>. If the channel width of the Mississippi River is approximately 1 km (Carey and Keller, 1957) and the amount of sediment being moved is equivalent to 5 m depth, assuming steady state, the rate of transport calculated from equation (9) is approximately 2.3 km yr<sup>-1</sup>.

Assuming that the bedload is equal to 10 percent of the suspended load, data for the Columbia River (Judson and Ritter, 1964; Jordan, 1962) suggest that sediment bedload material is transported at a rate of approximately 1 km yr<sup>-1</sup>.

These figures are only approximate since probably not all of the width of the channel is active in transport of material at any one time and the depth of sediment transported may also be variable.

By examination of the sediment type in Elliot Lake and the Witwatersrand it may be possible to estimate the rate of bedload transport in these Precambrian rivers.

The diameter of pebbles in the central Witwatersrand generally ranges from 25 to 75 mm and may be as great as 150 mm (Pretorius, 1964). The size of pebbles in the Elliot Lake ranges from 25 to 100 mm (Pienaar, 1963). This indicates that, at least during floods, the rivers were competent to move cobbles of this size. The ability of a river to carry material of this size indicates that the river had a flow velocity of approximately 1–3 m s<sup>-1</sup> (Sundborg, 1956). From this flow rate and the empirical relationship derived by Colby (1961), such a river would have a bedload discharge of 0.3 mm sand-sized particles on the order of 2 to  $10 \times 10^{10}$  g m<sup>-1</sup> of channel width per year. If a depth of 5 m of sediment in transport is assumed, a transport rate of 1.5–7.5 km yr<sup>-1</sup> may be calculated from equation (9). This calculation should only be taken as indicating an order of magnitude, particularly since the calculation of the flow rate is influenced by flood conditions and the depth of sediment transported is assumed. However, the calculation indicates rates of transport of bedload material similar to those of present rivers.

These calculations indicate that during periods when the bedload is continuously entrained it may be transported at rates on the order of 1 km yr<sup>-1</sup>. Under these conditions, residence time of the grains in river systems 100 km in length would have been on the order of 100 years.

In alluvial rivers, however, with broad flood plains, sediment grains may be abandoned in alluvium for con-

siderable periods of time by shifts in the river channel. This increases the residence time of the particle in the river system. In order to estimate the average transport rate of particles in such cases, the entire width and depth of sediments in the alluvium must be considered.

The Mississippi River is used as an example. The alluvial valley of the Mississippi River is approximately 1,000 km long. The valley varies in width from a maximum of approximately 200 km at Helena, Ark., to a minimum of 40 km at Natchez, Miss. The depth of sediments in the valley is approximately 30–40 m (Thornbury, 1966). Using an average value of 120 km width, an average sediment depth of 35 m, a transport rate of approximately 30 m yr<sup>-1</sup> may be calculated from equation (9). Thus, the residence time of sediments in the Mississippi River valley is approximately 35,000 years.

Data from Everitt (1968) indicate a transport rate of approximately 16 m yr<sup>-1</sup> for sediments in the valley of the Little Missouri. The transport rate and residence time of particles will undoubtedly vary from one river system to another and within individual river systems.

It is not possible to make any calculations of the mean residence time of uraninite grains in the Precambrian rivers of the Witwatersrand and Elliot Lake since virtually no trace of the rivers remains. If the results for the Mississippi River or the Little Missouri River are taken as order of magnitude estimates, residence times of tens of thousands of years are suggested.

However, the degree of oxidation and dissolution may not be proportional to the total time spent in the alluvium. From figure 3 it can be seen that the pore water in sediments somewhat removed from Bugaboo Creek was highly undersaturated with respect to atmospheric oxygen. The degree of undersaturation, if any occurs, is dependent on the relationship of the sediments to the river and to the water table, the permeability, the content of organic matter, and the mineralogy. In impermeable sediment containing oxidizable organic matter or minerals, the pore water may become highly undersaturated with oxygen with respect to the atmosphere. If the oxygen pressure in the Precambrian atmosphere and waters was less than at present, completely anoxic conditions might have been more easily achieved at that time. In this case, the rate and the amount of oxidation may be negligible. However, if the grain is deposited above the water table, or in a zone of fluctuating water table, or in a relatively permeable sediment, the degree of undersaturation may be minor. In the first case, the period the particle is exposed to oxidation may be approximately equal to the time during which the particle is actually in sediments being moved by the river. In the second case, the particle is exposed to oxidation for the entire period it is in

transit in the river system. The actual case must lie somewhere between these two extremes.

Therefore, it seems virtually impossible to establish with any degree of certainty the period of time uraninite grains might have spent in oxidizing zones during transport. In the extreme cases in which a sediment particle was continuously entrained by the river or in a river surrounded by anoxic sediments, periods in oxidizing environments might have been as short as 100 years. At the other extreme, in which the particle remained in oxidizing zones during transit, periods of tens of thousands of years might have been reached.

#### DEPOSITION AND BURIAL

The exact mode, or modes, of deposition of the placers is still uncertain (Brock and Pretorius, 1964; Frarey and Roscoe, 1970). The two major models for deposition of the placers are those of terrestrial fluvial deposition and marine littoral deposition. The fluvial model appears to be well supported for the major uraniferous conglomerates in Elliot Lake (Pienaar, 1963; Frarey and Roscoe, 1970). However, no single model appears able to explain the deposits of the Witwatersrand. Brock and Pretorius (1964) have suggested that several mechanisms may have been responsible for deposition.

Pienaar (1963) suggested that the gravels constituting the main Elliot Lake uraniferous conglomerates were probably laid down during periodic increases in the current velocity of the streams. Under such conditions oligomictic gravels would be deposited on the bottoms of wide stream channels as gravel bars or lag gravels. He attributed the interlacing of lenses of varying grain size to periodic fluctuations in the current velocity, as well as to differences in the current velocity in different parts of the channel.

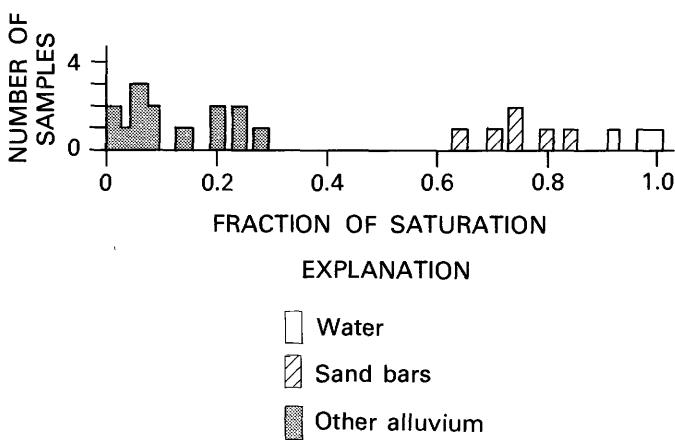


FIGURE 3.—Concentration of dissolved oxygen (as a fraction of saturation) in water and alluvial sediments of Bugaboo Creek, British Columbia, Canada.

The intimate intercalation of the uraninite-bearing conglomerates with generally uneconomic conglomerates may indicate that placers were produced by very local current and depositional variations, without significant diminution of the sedimentation rate.

This should have applied to the fluvial deposits of the Witwatersrand as well. However, conditions in marine littoral or submarine deposits might have been quite different. Tracer studies of movement of beach sand have been undertaken (Ingle, 1966), but periods of milling and concentration in beach deposits are largely unknown. It appears, however, that concentrations of heavy minerals may be formed within a few years.

Even if the uraninite were delivered to the basin of deposition virtually unweathered, it might still be destroyed if oxidizing conditions prevailed in the sediment (Walker, 1967). There has been long discussion of the origin of pyrite in both the Witwatersrand and the Elliot Lake deposits (Ramdohr, 1958; Liebenberg, 1957; Davidson, 1960, 1965; Schidlowski and Trurnit, 1966; Roscoe, 1969; and others). Part of the pyrite appears to be detrital. However, observations of pyrite overgrowths and frambooidal pyrite indicate that some of the pyrite is of postdepositional, epigenetic origin. Ramdohr (1958) suggested that pyritization occurred shortly after deposition. He suggested that some of the concretionary pyrite, which is probably secondary in origin, may have undergone some transportation. This would indicate either erosion from an older formation or, more probably, erosion from alluvial sediments caused by shifts in the position of the stream channel. This suggests that pyritization occurred close to the surface and that ground waters in some sediments surrounding the river or beach contained so little free oxygen that pyrite could be precipitated. Therefore, it may be postulated that the sedimentary environment became reducing near the surface. The fluvial or marine facies of the placers would probably have been aerated in the top few centimeters owing to the aeration of the river or ocean, but lagoonal facies of the placers might have been anoxic, even very near the surface (Saager, 1970).

Rates of deposition in molasse settings are fairly rapid. Rates are estimated at between 15 cm/1,000 years and 5 m/1,000 years (Fischer, 1969). Rates of deposition in the Siwalik Formation, one used by Reinike (1930) as a model for Witwatersrand sedimentation, may have been as high as 50–100 cm/1,000 years (Gansser, 1964). Therefore, grains may have been removed from strongly oxidizing environments within a few thousand years.

#### NUMERICAL CALCULATIONS

The experimentally derived reaction-rate equations (2 and 8) were solved for the maximum oxygen pressure

that would allow survival of uraninite as a detrital mineral.

For purposes of the calculations the following values or ranges of values were used:

Specific surface area (SS)	90 cm <sup>2</sup> g <sup>-1</sup>
Organic Retardation Factor (RF)	5-35
Nonuranium cation content (NOC)	0.16 (mole fraction)
Carbon dioxide pressure	5-100 PAL (0.0016-0.032 atm)
Temperature (T)	15°C
Alkalinity of river waters (A)	1×10 <sup>-3</sup> Molar
Residence time in soil profiles (T <sub>E</sub> )	500-10,000 years
Residence time in oxidizing environments during transit and burial (T <sub>r</sub> )	1,000-100,000 years
Fraction of original grain destroyed by 0.5 chemical corrosion (DF)	

The hydrogen ion concentration in solution was calculated assuming that all activity coefficients were unity, that carbonic acid was the only major source of hydrogen ion, and that the solution was open to the atmosphere. This may lead to an underestimation of the acidity of ground and river waters if oxidation of sulfides contributed significant amounts of sulfuric acid or the decay and metabolism of organisms contributed organic acids or excess carbon dioxide to the water. Since pyrite apparently survived as a detrital mineral, the rate of oxidation of sulfides must have been quite slow. This suggests that addition of sulfuric acid to ground and river water may have been minor.

The hydrogen ion concentration in erosional zones was calculated assuming negligible alkalinity by the equation

$$[\text{H}^+] = (\mathbf{K}_1 \mathbf{B} P_{\text{CO}_2})^{1/2} \quad (10)$$

in which  $\mathbf{B}$  is the Henry's law coefficient and  $\mathbf{K}_1$  is the first dissociation constant of carbonic acid ( $\mathbf{B}\mathbf{K}_1 = 1.6 \times 10^{-8} M \text{ atm}^{-1}$  at 15°C).

The hydrogen ion concentration in river and alluvial waters was calculated from the equation

$$[\text{H}^+] = \frac{\mathbf{B}\mathbf{K}_1 P_{\text{CO}_2}}{(A)} \quad (11)$$

in which (A) is the bicarbonate alkalinity

$$(A) \equiv (\text{HCO}_3^-) \equiv (\text{Na}^+) + (\text{K}^+) + 2(\text{Ca}^{+2}) + 2(\text{Mg}^{+2}) - (\text{Cl}^-) - 2(\text{SO}_4^{-2}) \quad (12)$$

(Broecker and Oversby, 1971).

The alkalinity of present average river water is approximately  $1 \times 10^{-3} M$  (Livingstone, 1966). The rate of chemical weathering appears to have been relatively constant over geologic time (Garrels and Mackenzie, 1971). If the runoff from continents during the mid-Precambrian was similar to the present amount, the alkalinity of Precambrian rivers may have been similar to that of present rivers. If this is correct, the average

pH of Precambrian rivers would not have been lower than about 6.3, even at carbon dioxide pressures as great as 100 times the present level.

Holland (1972) has shown that the pH of ocean water is unlikely to have been less than 7 since the early Precambrian, even under very high atmospheric carbon dioxide pressures.

Assuming carbonate equilibria largely control solution pH, it can be shown that at pH greater than 6.3 the dissolution rate of uraninite in systems open to the atmosphere is independent of alkalinity or hydrogen-ion concentrations (Grandstaff, 1973). Therefore, small variations in Precambrian river- or ocean-water chemistry may not be significant in influencing the rate of uraninite dissolution. (Other details of the calculations are given in Grandstaff, 1973.) Results of the rate calculations are shown in figures 4 and 5.

Figure 4 is constructed under the assumption that oxidation during weathering is minor and that chemical corrosion occurs only during transportation and burial. This would occur if the uraninite were derived by scouring from bedrock or glacial weathering. Although glacial erosion seems unlikely in the case of the Witwatersrand and Elliot Lake deposits, the figure suggests that uraninite of the Blyvooruitzicht type could survive for some time as a detrital mineral if it were derived from glacial weathering, even under present atmospheric conditions. This is supported by observations in Pakistan, Canada, and Alaska of detrital uraninite that is apparently derived from glacial sources. The chemical composition and resistance to chemical corrosion of the uraninite in these recent placers is not known. The persistence of uraninite in the modern environment may be due not only to cold temperatures and glacial weathering but also to high resistance to chemical corrosion.

Figure 5 shows the maximum atmospheric oxygen pressure allowing the survival of uraninite as a detrital mineral under various periods of exposure to oxidizing conditions during weathering, transportation, and burial.  $T_E$  is the residence time in soil profiles.  $T_r$  is the residence time in oxidizing zones during transport and burial. Because the rate of dissolution during weathering is more rapid than that during transport or burial, the maximum pressures allowed are largely influenced by residence periods in soil profiles except under conditions in which the time required for transport and burial is much longer than the weathering period ( $T_r \gg T_E$ ). The oxygen pressures allowing uraninite survival decrease with increasing periods of exposure. For the most likely conditions, oxygen pressures of  $10^{-2}$ – $10^{-6}$  PAL are indicated.

These calculated pressures are maximum pressures. It is likely that the actual atmospheric oxygen pressure was somewhat lower. Also, these calculations do not

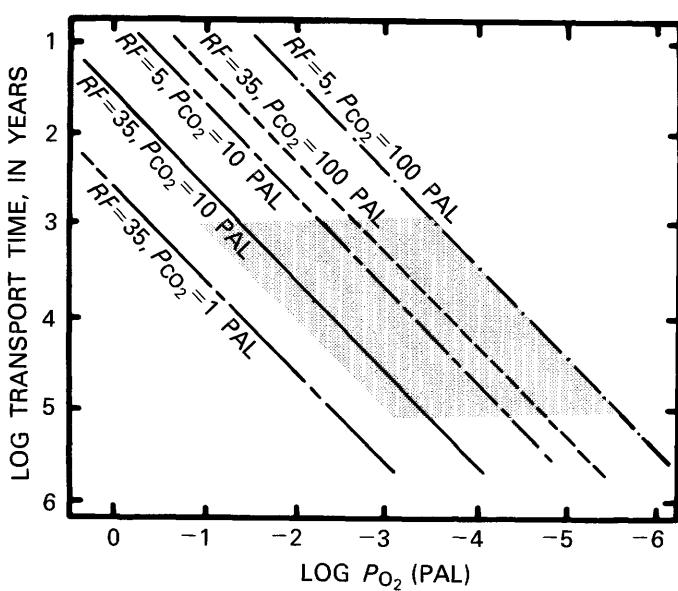


FIGURE 4.—Maximum oxygen pressures allowing survival of uraninite as a detrital mineral, assuming that significant dissolution occurs only during transport and burial. The most reasonable conditions are shown by the shaded area.  $RF$ , organic retardation factor; PAL, present atmospheric level;  $P_{CO_2}$ , partial pressure of carbon dioxide in atmosphere.

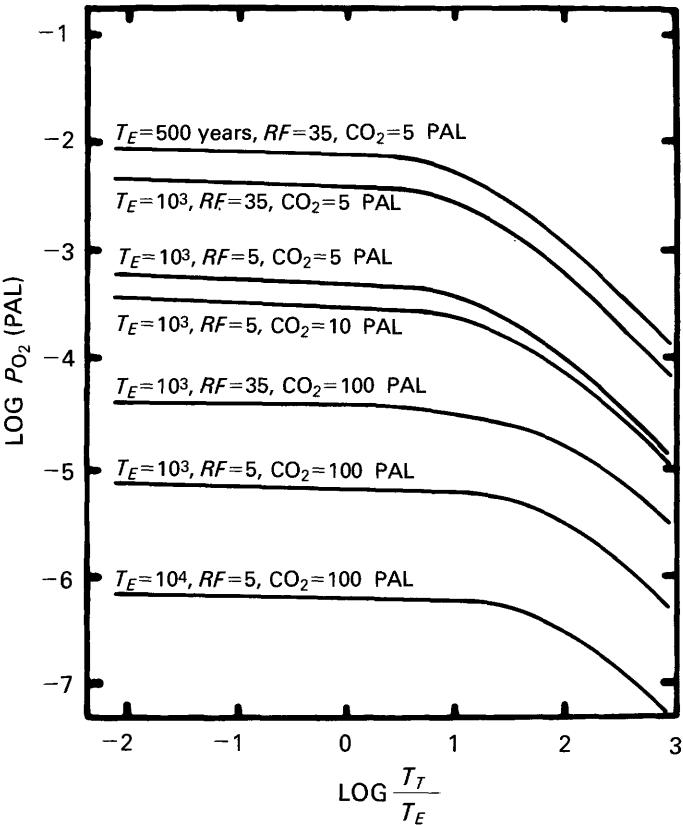


FIGURE 5.—Maximum oxygen pressures allowing the survival of uraninite as a detrital mineral during the sedimentary cycle.  $T_E$ , residence time in soil profiles;  $T_I$ , residence time in oxidizing zones during transport and burial;  $RF$ , organic retardation factor; PAL, present atmospheric level.

take into account other possible mechanisms by which uraninite might be attacked and dissolved, such as by uranium-oxidizing bacteria or by formation of oxygen or other oxidative species by hydrolysis of water by radiation. If uranium-oxidizing bacteria or other oxidizing agents were present in the ancient environment, oxygen pressures required for survival of uraninite may have been much lower.

#### IMPLICATIONS FOR THE PRECAMBRIAN ATMOSPHERE

Present data suggest that the earliest terrestrial atmosphere was composed of reduced species, hydrogen and methane with nitrogen and water vapor (Holland, 1962). Then, at some time before 3,500 m.y. ago, this atmosphere was converted into one containing nitrogen, argon, and carbon dioxide, possibly by the disappearance of free iron from the upper mantle and consequent volcanic exhalation of less-reduced species. Finally, at some more recent time, the photodissociation of water vapor with hydrogen escape and the aerobic photosynthesis of plants with burial of carbonaceous matter converted the atmosphere into one containing oxygen as a major component.

Four general models of oxygen increase have been proposed (see Fischer, 1972). They are shown schematically in figure 6 and are described as follows:

1. The oxygen content of the atmosphere has been progressively increasing from aerobic photosynthesis since the appearance and evolution of blue-green algae.
2. The atmospheric oxygen remained low (less than 0.001 atm) until the late Precambrian and early Phanerozoic time. Berkner and Marshall (1965, 1966) proposed that this concentration of oxygen was reached as a result of photodissociation of water vapor. They ascribed the appearance of metazoa at the beginning of the Cambrian to the rise of oxygen pressure to the Pasteur point (approximately 0.01 PAL). They believed that the appearance of fossil land biota at the beginning of the Devonian could be correlated with a rise in atmospheric oxygen pressure to approximately 0.1

PAL, a level providing an ozone layer capable of shielding the Earth's surface from deadly ultraviolet radiation.

3. The oxygen level remained low until about 2,000 m.y. ago and then rose toward its present concentration (Cloud, 1968). Cloud ascribed the disappearance of banded iron-formations and the appearance of red beds about 2,000 m.y. ago to a rise in atmospheric oxygen at that time.
4. The atmospheric oxygen increased rapidly near the beginning of the Precambrian to a level near that of the present time. This rapid rise was due either to photodissociation of water vapor (Brinkmann, 1969) or to the evolution of photosynthetic algae.

The present study indicates that the pressure of oxygen in the Precambrian atmosphere between 2 and 3 b.y. ago was probably less than approximately  $10^{-2}$  to  $10^{-6}$  PAL. This appears to rule out the progressive accumulation model (model 1) and the model of early high oxygen levels (model 4).

Because of the possible presence of photosynthetic organisms as early as 3 b.y. ago, this low level of oxygen implies that either the rate of photosynthetic oxygen production was quite low, possibly as a result of low nitrate nutrient concentrations (Holland, 1973), or the oxygen produced by photosynthesis or photodissociation was rapidly consumed by oxidation of reduced volcanic gases or reduced elements in sediments.

Brinkmann (1969) calculated that the content of oxygen in the Earth's atmosphere produced by photodissociation could have been as high as approximately 0.27 PAL, depending on the extent of oxidation of reduced species. However, it appears possible that the quantum

yield and the escape efficiencies used in the calculations may be too high (Venugopalan and Jones, 1968; Walker, 1974). Thus the oxygen pressure from photodissociation calculated by Brinkman (1969) appears too high.

Probable eucaryotic organisms are present in strata as old as 1.3 b.y. (Schopf, 1970). Eucaryotic organisms are fundamentally aerobic. This suggests that the concentration of oxygen in the atmosphere at that time was above the Pasteur point, approximately 0.01 PAL, unless the eucaryotic organisms were restricted to special oxygen oases (Fischer, 1965; Weyl, 1968). Certainly, by the time of origin of metazoa, before the beginning of the Cambrian, the level of atmospheric oxygen had reached 0.01 PAL. By the beginning of the Cambrian, the presence of carbonate skeletons and cartilaginous connective tissue indicates that the oxygen content of the atmosphere must have been more than 0.1 PAL (Towe, 1969).

This evidence suggests that model 3 is schematically correct. Low levels of oxygen, less than  $10^{-2}$  to  $10^{-6}$  PAL, were present in the early and middle Precambrian atmosphere. Higher oxygen pressures, greater than 0.01 PAL, were reached by 1.3 b.y. At the beginning of the Cambrian, the requirements of collagen (Towe, 1969) indicate that the oxygen pressure was greater than approximately 0.1 PAL.

#### ACKNOWLEDGMENTS

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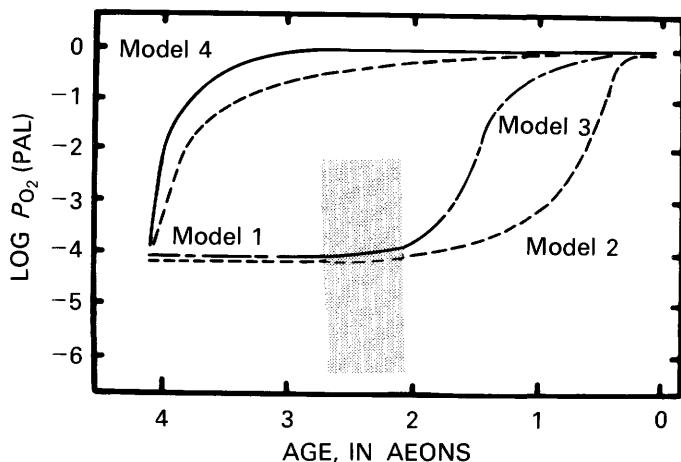


FIGURE 6.—Models for the evolution of oxygen in the Earth's atmosphere through geologic time. (See text for further description of models.) Shaded area shows most probable ranges of oxygen pressure and time for formation of quartz-pebble uranium deposits.

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## DISCUSSION

[Hallbauer?]: Is there anything known about why some of the recent rivers carry uraninite, for instance, the Indus River in India?

Grandstaff: There are several possibilities here. First of all, that the uraninite could be quite highly thorian or have a high content of rare earths, which would tend to stabilize it against oxidation. So these would oxidize

very, very slowly, much more slowly than the Blind River or Witwatersrand uraninite. Secondly, the uraninite appears to be derived glacially without any oxidation during the erosion process and in the case of the Indus is flushed rather rapidly down to the Hazara district, where it is currently being buried. I think this is about the same for other occurrences. Of course, in England the uraninite, pitchblende really, was derived from mine tailings, so didn't undergo any particular oxidation during its "erosion." I went up to Bugaboo Creek, British Columbia. As I indicated, that's one area in Canada which is supposed to have uraninite. I didn't find any uraninite, unfortunately; it may still be there, but if so, it is very rare.

Simpson: My question follows on from the previous speaker. The Indus River gravels today contain thorian uraninite with a thorium-to-uranium ratio of 1:4, very similar to the ratio in the Dominion Reef of 1:4 to 1:5. The question I want to ask is this. What comparative conclusions, if any, do you draw about the atmospheres for these two occurrences?

Grandstaff: As was pointed out, thorium is not the whole story; you need a complete analysis of the uraninite. Stacy, in some conversations I have had with him, suggested that the uraninite of the Bugaboos might be very cerian rich. His X-ray data suggested it has a very small unit cell, much smaller than  $\text{UO}_2$ . So I think you need a complete chemical analysis. Other than that, I'd suggest again that the very cold temperatures in the glacial environment tend to allow the uraninite to persist, at least for a time.

Simpson: Would you agree, then, that your data doesn't say much about the atmosphere in Dominion Reef times?

Grandstaff: Well, the Dominion Reef's uraninite is more resistant to corrosion, and I have suggested about 15 to 20 times, which would have allowed oxygen pressures of, say, a tenth to one hundred-thousandth of the present. Obviously, since it's not the most sensitive uraninite, it doesn't really play any part in saying what oxygen pressures in the Precambrian might have been.

Cannon: Have you considered what effect either ozone or monatomic oxygen might have on these rates? Both of these might be important species in a primitive atmosphere and should react more rapidly than  $\text{O}_2$  does.

Grandstaff: Yes, that's correct. However, if the oxygen pressure is less than 0.01 PAL or so, the ozone pressure would have been only a very, very small fraction of that. I didn't consider that it would be important because of its relatively low probable concentration, even though it is somewhat more reactive.

Roscoe: I wonder if the molasse model you have of erosion and deposition of the Huronian sands and gravels

would give perhaps a falsely short time of residence over what the actual situation may be, because I don't think the comparison to molasse is really valid here. You've got extremely coarse, fairly mature sediments throughout the geological column of up to 20,000 feet, interrupted only perhaps by glacial episodes where we have immature sediments. In other words, over an immense period of time, this time must have been long if you consider also that the clastics are very coarse, we have very coarse sediments, so what we are seeing is only the coarsest fraction of the granitic rocks that have been eroded and only the quartz out of that, only a few percent of the rocks piled up in this immense column. The sediments, therefore, must have had a very volumetrically large source area through a very appreciable length of time to have developed this whole sedimentary system. Moreover, the source area must have been soil mantled virtually continuously, and not cut by canyons, steep slopes or so on; the slopes would have to have been fairly moderate and I think the molasse type of deposition would be an exaggeration of what must have prevailed at that time.

*Grandstaff:* I agree there has been a large pass-through of finer grained sediments. The sediments are not, perhaps, as mature as one might think since there is a lot of evidence for feldspars and other easily weathered minerals. They are submature rather than completely mature sediments, so I would say that the molasse model is not too bad.

[*Schopf?*:] I notice that your data are inconsistent with the Brinkmann calculation. It would be instructive to me if you could discuss what's wrong with the Brinkmann calculation and why it doesn't fit your experimental data.

*Grandstaff:* Brinkmann looks at the reaction water vapor plus a photon yielding hydrogen plus oxygen, and he calculated that oxygen pressure could have been brought up, at a maximum, to about one quarter of our present oxygen pressure. Brinkmann makes two assumptions which may not be entirely correct. The first assumption is that the quantum yield of the above reaction is unity; that is, every photon causes the splitting of water and the creation of odd oxygen. The quantum yield in this reaction is not very well known. It appears to be very highly dependent on pressures, on other species, and varies between about one and one millionth. So, depending on quantum yield, oxygen pressures which could be created might vary between a quarter and a quarter of a millionth of the present. The second thing that Brinkmann suggests is that all this hydrogen escapes; that is, that the escape factor is very high. In our present atmosphere, at present exospheric temperatures of around 1,000 to 1,500 degrees Kelvin, the escape efficiency is only

about one thousandth to one hundredth; that is, only about one in a thousand to one in a hundred of the hydrogen ions actually leaves the atmosphere. In my opinion his escape efficiency is unreasonably high, and I think if you consider what the atmosphere might have been like during the mid-Precambrian, particularly with a lower solar constant, that is, lower luminosity, you will agree that the escape efficiency would have been quite low. Take Venus as an example. Exospheric temperatures are only on the order of 600 to 800 degrees Kelvin; that would give an efficiency of perhaps a hundred-thousandth. So I'd say that, rather than the quarter that he suggests,  $10^{-3}$  to  $10^{-6}$  is more reasonable for the maximum amount of oxygen which could be produced by photodissociation.

[*Minter?*]: You have taken samples from different environments and placed them in the Dominion Reef, which is a very old deposit. I do think that the evolution in the model that you are purporting to describe is not as general as you imply and that probably the oldest deposits were certainly deposited on very long low-gradient paleoslopes. Secondly, your samples from Blyvooruitzicht are samples from a reef which has certainly been recycled many times; that reef lies on an angular unconformity and is certainly not a primary reef.

*Grandstaff:* Again, it seems to me that there is very little necessary relation between the slope on which the material is resting and the source area; that is, if you do have uplift in the source area, you probably do indeed have virtual peneplanation at the beginning of this episode. I would say that peneplanation was followed by uplifting in the source area, downcutting by rivers, relatively rapid erosion, and flushing of this material into the reef. Obviously I have tried to indicate some range in permissible exposure times to allow for this very basic uncertainty in the rates. With regard to recycling the sediments, this too would tend to increase exposure periods. It is not really very clear to me how long and how necessary this recycling is that is commonly alluded to. How much recycling lengthens the exposure period I just don't know; recycling can happen very rapidly, particularly in oceanic milling and formation of black sands on beaches. Also, the uraninite and the host sediment do not necessarily have to come from the same area or have been exposed to the environment for the same period of time.

*Myers:* Could you explain your statement about organic matter slowing the rate of uraninite oxidation?

*Grandstaff:* I believe that the polar organic matter latches onto the positively and negatively charged surfaces and in this case essentially blocks the oxygen from approaching the oxidation site, so that essential-

ly the organic matter completely covers the surface and doesn't allow the oxygen to get in, slowing down the reaction rate. The same thing has been noted in calcite. You can acidify a solution containing calcite, and the reaction should go quite quickly, but it doesn't because of the organic coating on the grains.

*Myers:* Do I understand that you actually observed this slowing of the rate?

*Grandstaff:* Yes.

*Myers:* And you are attributing it to the organic matter?

*Grandstaff:* Yes.

*Skinner:* I would like first to ask you, if you only have carbon dioxide at one hundred times the present level, where was the rest of the carbon? It doesn't seem to be in carbonates or in sediments if you make mass balance for that period, despite Bill's [Schopf] 50 percent slop.

*Grandstaff:* I have to disagree with you there. All we are saying is that you have carbonates and you have a reaction between the carbonate, calcite and the magnesium silicate, some sort of chlorite and dolomite, and silicic acid giving you  $\text{CO}_2$  in solution, or in the atmosphere. Because of the absence of silicifying organisms during the Precambrian, we believe that the  $\text{CO}_2$  pressures must have been somewhat higher. This doesn't have really anything to do with the mass balance between carbonate and organic carbon.

*Skinner:* It requires you to keep adding  $\text{CO}_2$  from the mantle or somewhere for that kind of model, or else you have to have almost as much carbonate in the section as you now have.

*Grandstaff:* You certainly have carbonates, primary looking carbonates, all the way back to Onverwacht time.

*Skinner:* Do you have the same amount?

*Grandstaff:* Well, this is certainly a big question. There is a problem here, of course, in that the carbonates tend to be preferentially dissolved by later weathering. Garrels and Mackenzie have shown that the turnover time is only a few hundred million years for carbonates as compared to the billion years or so for silicates, so that it's not very clear how much our present exposures really reflect the true Precambrian carbonate record.

*Skinner:* Could I pursue not the abundance of carbonates but the question surrounding carbon dioxide a little further? Since yours and anybody else's model for the early Precambrian atmosphere has a higher

carbon dioxide content, presumably we were dealing with much higher average surface temperatures than we see today.

*Grandstaff:* No, not necessarily.

*Skinner:* At least five to ten degrees more. No?

*Grandstaff:* Okay. The thermal balance of the Earth is still very much up in the air, so to speak. Calculations assuming a lower solar constant suggest the atmospheric temperature was actually lower than at present. Rasool and Schneider find that the effect of  $\text{CO}_2$  pressure on temperature is more logarithmic than linear, and that doubling  $\text{CO}_2$  pressure causes an increase of roughly  $1^\circ\text{C}$ , the  $\text{CO}_2$  pressure must be increased ten times to get the next degree C, and so forth. This is one mechanism by which the Precambrian temperature could have been made higher. Opposing this is the fact that the solar constant at that time was lower, perhaps only about 75 percent of the present, and would cause temperatures to be lower. Also, the rate of orogeny and rate of volcanism at that time, as suggested by Richard Armstrong, may have been higher because of the higher radioactive decay. Intense volcanism would have erupted much material into the atmosphere, aerosols, which would have tended to depress the temperature. The effect of aerosols on atmospheric temperature seems to be much more pronounced than the effect of carbon dioxide. I think rather than fearing that the surface temperature would get too high, I think we have to fear that the surface temperature would get too low. This sort of thing has not been treated in a quantitative fashion.

*Skinner:* If the surface temperature gets too low, then you depress the dissolution rate of the phosphides and of course you know they are all gone. The surface temperature cannot remain too low for very long without destroying the only weathering processes you've got to get rid of phosphides.

*Grandstaff:* Well obviously, if you freeze the whole surface of the planet there is no life. But within the sort of permissible range of  $1^\circ$  up to  $30^\circ$ , using an activation energy for the hydrolysis of feldspar of 8.7 kilocalories per mole, as suggested by Helgeson, you get a variation in rate of weathering of only a factor of two. It is about the same for uraninite. It's hard to tell what we are seeing in the Precambrian in regard to the maturity of Precambrian sediments and the effectiveness of Precambrian weathering.

**SOUTH AFRICA**



# Uraniferous Quartz-Pebble Conglomerates in South Africa

By J. W. VON BACKSTRÖM

GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN  
QUARTZ-PEBBLE CONGLOMERATES

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## GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN QUARTZ-PEBBLE CONGLOMERATES

# URANIFEROUS QUARTZ-PEBBLE CONGLOMERATES IN SOUTH AFRICA

By J. W. VON BACKSTRÖM<sup>1</sup>

### ABSTRACT

The purpose of this paper is to give a short background statement summarising data on the Dominion Reef Group, the Witwatersrand Supergroup, and the Ventersdorp Contact Reef, with particular reference to the close relationship of gold and uranium with sedimentary features as well as the mineralisation, conditions of deposition, and the nature of the quartz-pebble conglomerates.

### URANIFEROUS QUARTZ-PEBBLE CONGLOMERATES IN SOUTH AFRICA

Important concentrations of uranium minerals occur in auriferous conglomerates present in four successive Precambrian stratigraphic units, namely, Dominion Reef, Witwatersrand, Ventersdorp, and Transvaal, of which the two first named are important because of their relatively large uranium content. These four units do not conform to one another; they cover some tens of thousands of square kilometres in the Provinces of the Transvaal and the Orange Free State.

An outcrop map of the above-mentioned formations is shown in figure 1, whilst figure 2 illustrates the suboutcrop when all the younger formations are removed.

The Dominion Reef Group comprises a basal group of conglomeratic, arkosic, and quartzitic rocks up to 100 m thick, covered by about 900 m of lava of andesitic or acidic composition. The group of sedimentary rocks forming the base is, from an economic point of view, the most important part of the Dominion Reef Group. It is composed largely of the disintegration products of the Basement Granite. The sediments consist of medium-grained to coarse-grained feldspathic, and somewhat micaceous, sandstone or quartzite, in which there are lenticular bodies of grit and conglomerate.

Two conglomerate bands along or close to the base of the arenaceous sediments have been worked or prospected for gold and uranium; these are known as the Upper and Lower Reefs. They are separated by barren conglomerate or by a layer of quartzite. The Lower Reef, which occurs at the base of the conglomerate zone, occupies depressions in the erosion surface of the Basement Granite. It is lenticular and has been the main gold

carrier in the area. The Upper Reef occurs some 18 m or more higher up in the succession of conglomeratic, arkosic, and quartzitic rock. The reef is a small pebble conglomerate passing laterally into a thin layer of grit containing scattered pebbles. It contains a relatively high concentration of detrital monazite and cassiterite. The reef, varying in thickness from 5 to 120 cm, is remarkably consistent and was nowhere absent over some 900 m of underground development on strike. Where it is thin, the concentration of uraninite may be very high, with sections 2–5 cm thick assaying as high as one percent uranium. This reef is the main carrier of uranium and, apart from the minerals already mentioned, carries minor amounts of detrital chromite, garnet, zircon, ilmenite, and columbite-tantalite.

In the Witwatersrand Supergroup, economically important uranium and gold-bearing conglomerate bands occur on flat planes of intraformational diastems, disconformities, and unconformities, where they are known as reefs or banks. Each of these conglomerate bands represents a deposit formed after a hiatus in the process of sedimentation. They obviously consist largely of reworked and resorted detritus derived from the erosion of underlying pebble and quartzite beds. Channels and hollows, filled with eroded detritus derived from the erosion of previously laid sediments and known as Hybrid or Footwall Reefs, are of common occurrence immediately below the surface of contemporaneous erosion, upon which the principal conglomerate ore bodies were laid down. Fluvial structures are often clearly revealed by the channel deposits, some of which are more than 100 m deep. Pay streaks in the Footwall Reefs are aligned parallel to the course of the winding channel, and the higher values of gold and uranium, where present, tend to be confined to the basal portion of the reefs.

The remarkably close relationship of the concentrations of the gold and uraninite with sedimentary features strongly suggests that the gold and uraninite were deposited at the same time as the pebbles and other detrital components of the conglomerate. The pebbles show variations in their packing index. They are usually not close packed, and sometimes only isolated pebbles are present in the matrix. In some areas pebbles

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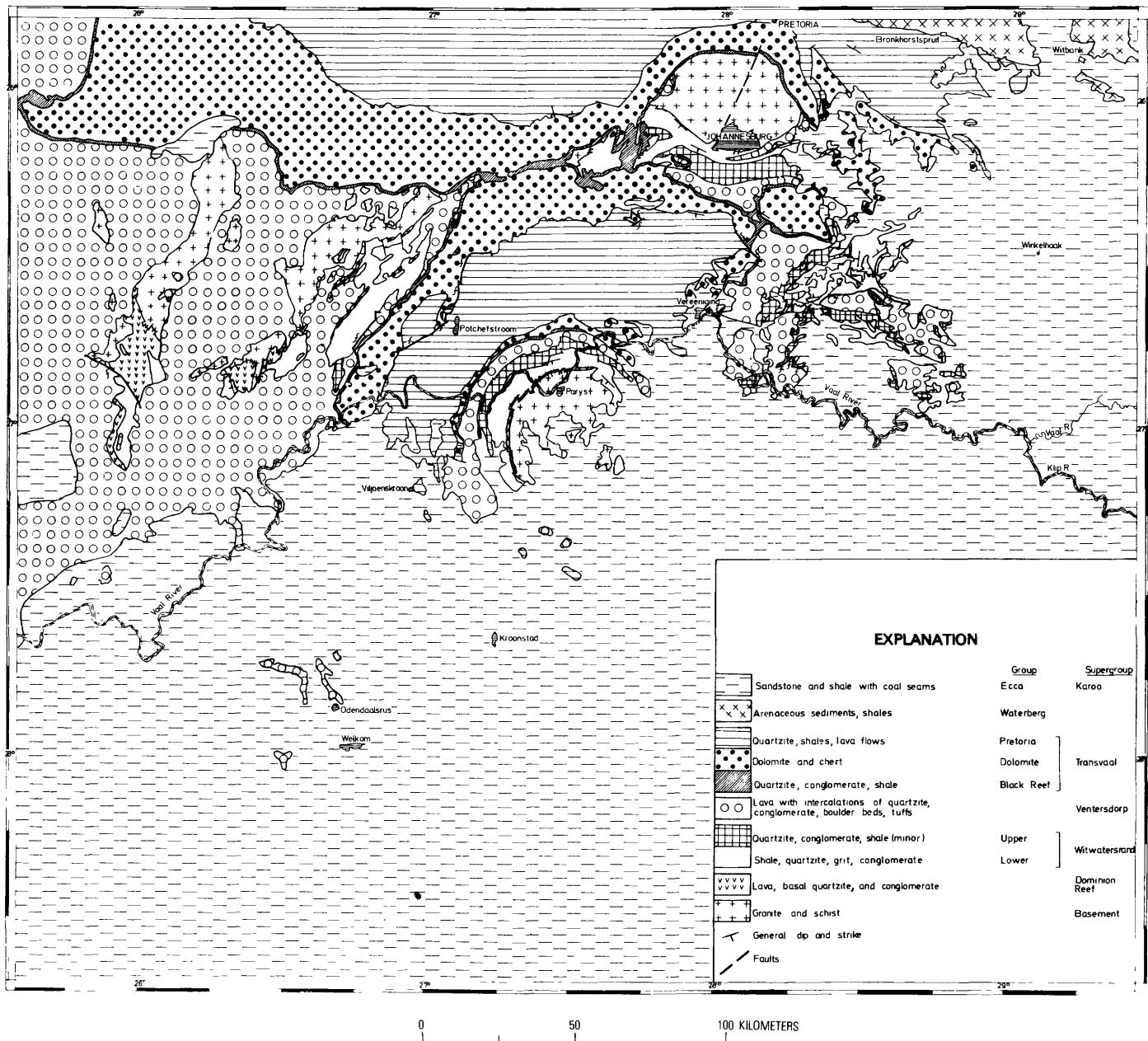


FIGURE 1.—The Witwatersrand basin, surface geology (from R. Borchers, 1964, Exploration of the Witwatersrand System and its extension, in Haughton, S. H., ed., The geology of some ore deposits in southern Africa: Geological Society of South Africa, v. 1, diagram I).

are well sorted, with the larger ones at or near the base, and are oriented with the longest axis parallel to the direction of strike of the beds in which they occur.

The pebbles in the ore-bearing conglomerates are composed mainly of vein quartz, with occasional pebbles of chert, quartzite, red jasper, quartz porphyry, and, more rarely, tourmalinized rocks, greenish slate, and schistose-type pebbles. The most abundant constituent of the matrix is quartz, which occurs together with phyllosilicates such as sericite and pyrophyllite and with

lesser quantities of chlorite, chloritoid, and tourmaline. Pyrite, in the form of rounded grains, crystals, and irregular patches, is a ubiquitous and often abundant constituent. Other sulphides that may be present are pyrrhotite, arsenopyrite, galena, chalcopyrite, sphalerite, marcasite, cobaltite, and linnaeite. Hydrocarbon is commonly found as granules and less often as thin layers or bands. Hydrocarbon containing minute particules of uraninite is also known as thucholite.

The conglomerates form only a minor part of the

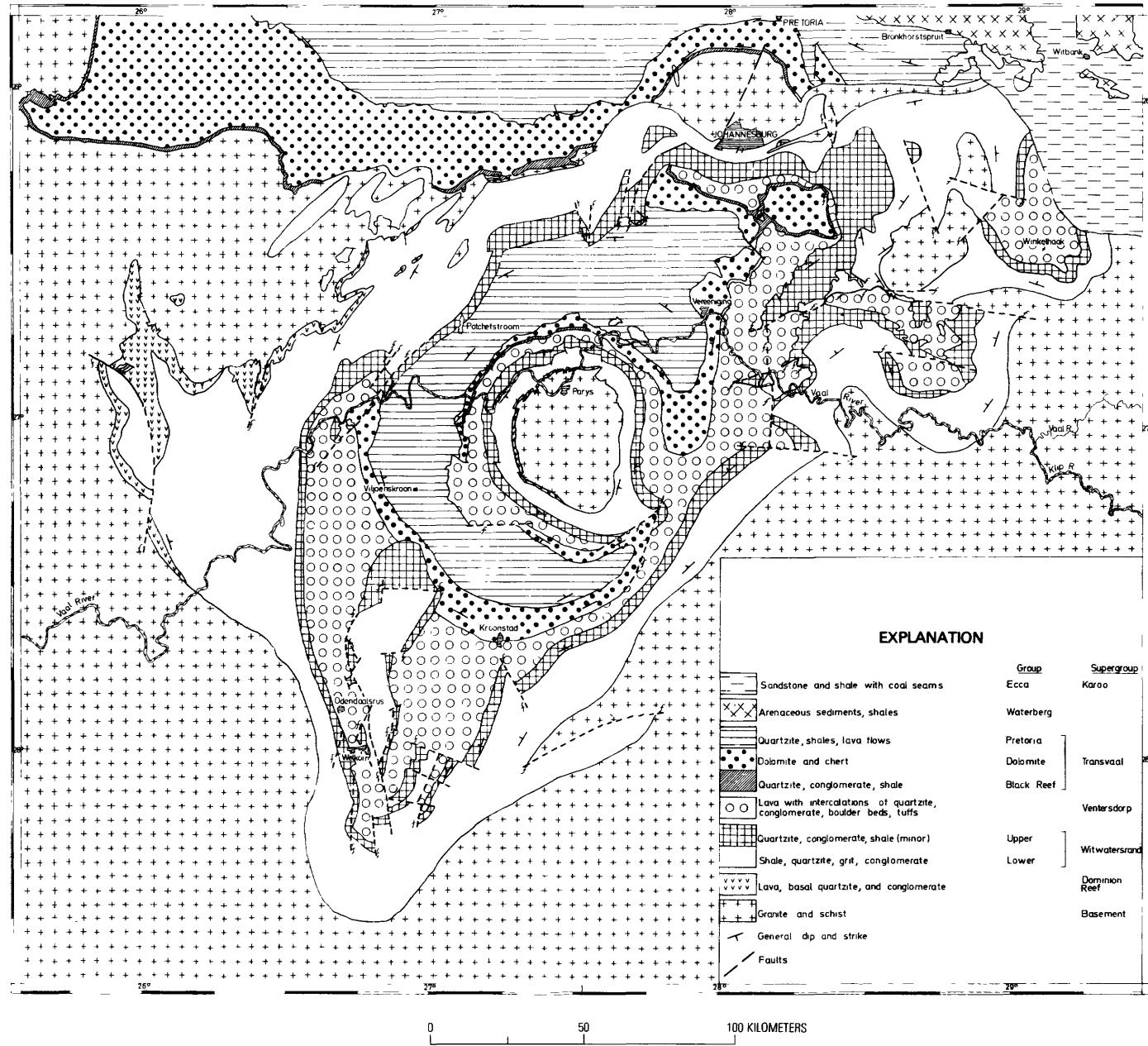
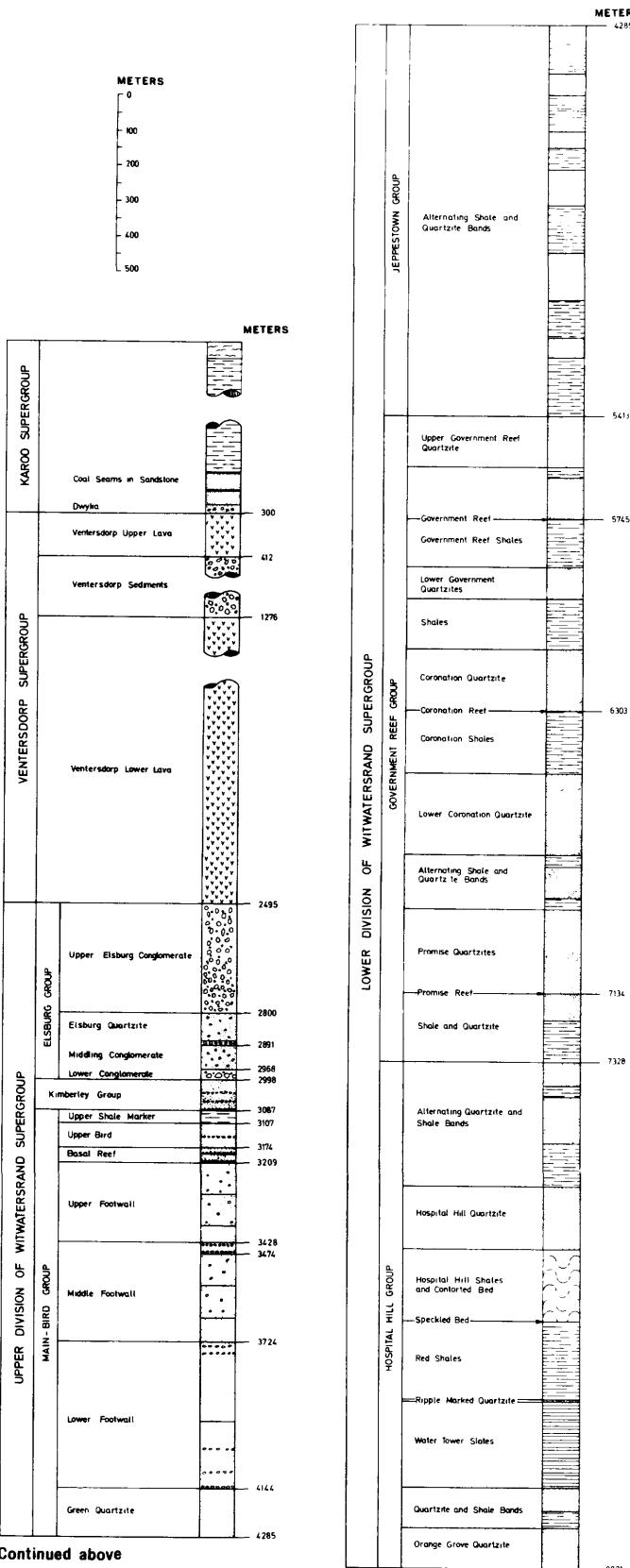


FIGURE 2.—The Witwatersrand basin, distribution of the Witwatersrand System (from R. Borchers, 1964, Exploration of the Witwatersrand System and its extensions in Haughton, S. H., ed., The geology of some ore deposits in southern Africa: Geological Society of South Africa, v. 1, diagram II).

whole Witwatersrand Supergroup (fig. 3) and vary in abundance in the different regions. In the Central Witwatersrand, conglomerates form 600 m of the 7,500 m of sediments (fig. 3) and are most abundant in the Upper Division of the Witwatersrand Supergroup (figs. 3, 4). Most of them contain both gold and uraninite, but not more than a dozen are rich and persistent enough to repay the cost of mining and extraction. These few are the conglomerates that have produced by far the largest output of gold and uranium in South Africa. They are

generally remarkably regular in their mode of occurrence, and individual conglomerate bands persist over wide areas. Beds of auriferous conglomerate also occur in the Lower Division of the Witwatersrand Supergroup, particularly in the Government Reef Member, but, with the exception of those west of Klerksdrop, they have not proved to be of economic importance.

From the bottom upwards, the principal uranium-bearing conglomerates in the Upper Division of the Wit-



Continued above

FIGURE 3.—Generalised geological section of the Upper Witwatersrand, showing cover of Venterdorp and Karoo Supergroups (Orange Free State), and of the Lower Witwatersrand (Central Rand).

watersrand Supergroup are the following: the Carbon Leader, belonging to the Main Reef Group of conglomerates; the White, Monarch, and one or two other conglomerate reefs of the Bird Reef Group in the West Rand, and their equivalents the Vaal Reef in the Klerksdorp Goldfield and the Basal and Leader Reefs in the Orange Free State Goldfield; and the May Reef, and its equivalents in the Kimberly Reef Group of conglomerates in the Far East Rand. Besides these, there are other gold-bearing reefs from which uranium is being, or may be, profitably extracted as a byproduct of gold production, for example, the Commonage Reef on the horizon of the Main Reef Group, some of the Western or Elsburg Reefs in the Klerksdorp Goldfield, and the A, B, and Rainbow Reefs in the Kimberley-Elsburg Groups in the Orange Free State Goldfield. The only uranium produced so far from the Lower Witwatersrand Division comes from the Afrikaner and Inner Basin Reefs in the Government Reef Group west of Klerksdorp (see fig. 4).

The uraninite and gold are more or less uniformly distributed over wide, flat areas occupied by the conglomerate sheets. As in the case of gold, uraninite can therefore be spoken of as evenly distributed in a general way, when the whole extent of the goldfields is considered, especially when comparison is made with the much greater variations in the distribution of values shown by nearly all lode or hydrothermal deposits. The richer gold and uraninite concentrations in an individual blanket sheet tend to collect in pay streaks, which are usually characterised by the presence of pebbles larger than those in the rest of the blanket and by a greater concentration of other ore particles, some of which are clearly detrital. The economically important uranium- and gold-bearing conglomerates, which form relatively thin, unbroken sheets of great length and breadth, eventually, with increasing distance from the original shoreline or source rocks, either break up into isolated patches or lenses of conglomerate containing notable amounts of very finely divided gold separated by lean or barren quartzite or shale, or they thin out into a single, rich line of heavy ore minerals and granules of hydrocarbon, along which isolated clusters of pebbles or scattered pebbles may occur. It is not unusual to find visible gold associated with these pebbles. Still farther away from the source rocks, the conglomerate lenses become smaller and smaller, and spaced farther and farther apart until, ultimately, the conglomerate has passed laterally into quartzite. Concomitantly, gold and uranium values decline until only traces or no values are left where quartzite occupies the horizons on which the conglomerate ore bodies occur near the source rocks.

Mineralisation is confined to the ore-bearing conglomerate and seldom crosses the stratification into the arenaceous wall rock, except where it has obviously

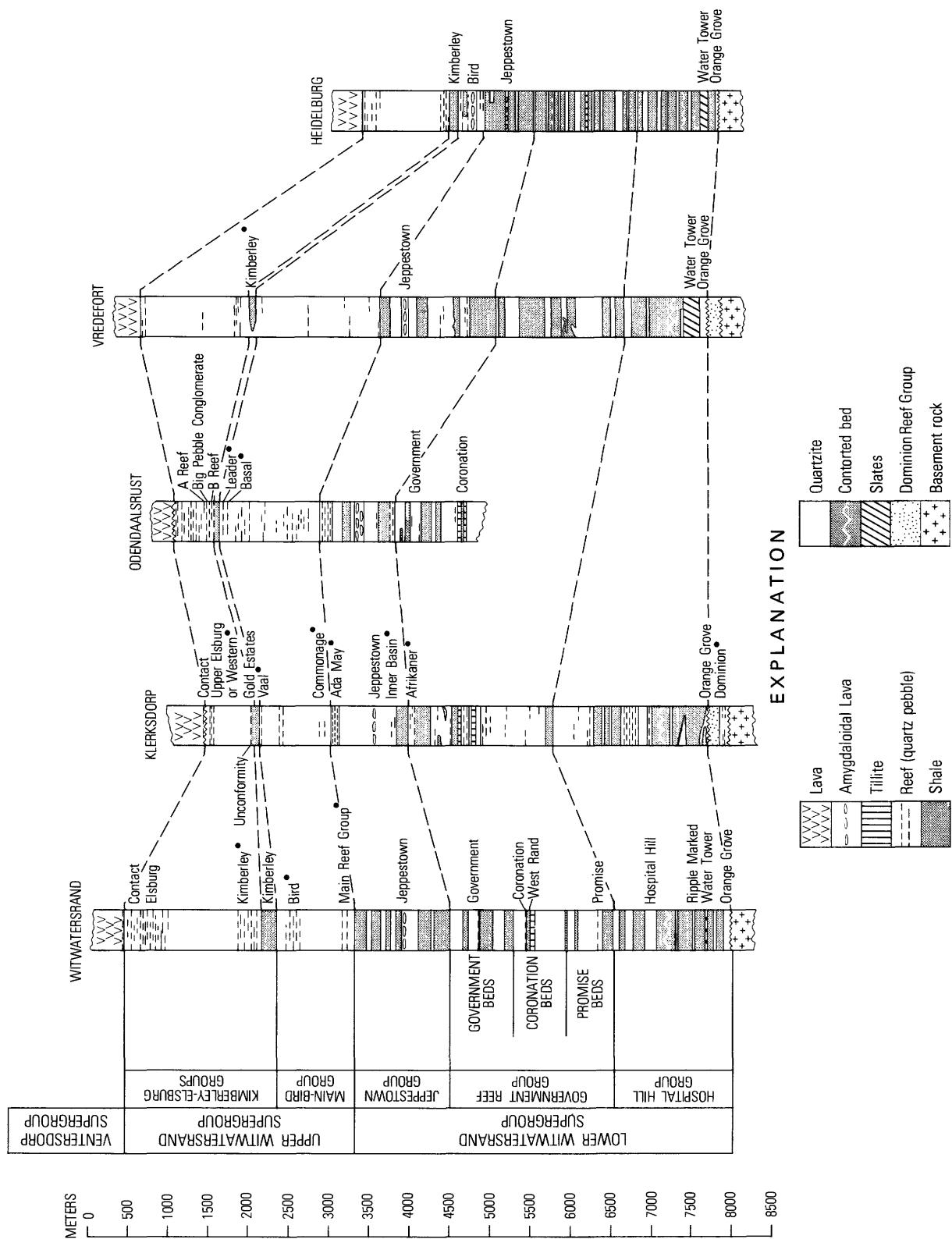


FIGURE 4.—The Witwatersrand Supergroup, comparative sections showing uranium-bearing horizons.

been derived from the conglomerate ore body itself. There is a close relation between the distribution of values and purely sedimentary features. Richer concentrations are commonest near the base of the conglomerate along well-defined shaly or schistose footwall partings and in conglomerate less than 30 cm thick. They also tend to be disposed in pay streaks that have many of the characteristics of those found in present-day placers.

The uraninite is finely disseminated as minute, discrete particles in the quartzose matrix that fills the interstices between the pebbles. Along with the fine particles of gold and grains of other heavy minerals, those of uraninite are very often more highly concentrated along the bottom of the conglomerate; in some cases, however, the higher concentrations of these heavy minerals are distributed parallel to the bedding and towards the middle or top portions of the conglomerate.

The Ventersdorp Contact Reef and the Black Reef occur at the base of the Precambrian Ventersdorp and Transvaal Groups, respectively. The Ventersdorp Group, predominantly a volcanic formation, overlies a surface of Witwatersrand beds that has been eroded down to varying depths. Gently inclined or flat-lying basal beds of the Transvaal Supergroup in turn rest with a pronounced unconformity on the tilted and eroded Witwatersrand and Ventersdorp rocks. Both the Ventersdorp Contact Reef and the Black Reef occur as lenticular bodies of conglomerate in depressions and channels in the Witwatersrand beds and are largely composed of the erosion products, including the heavy concentrates, of the immediately underlying Witwatersrand rocks. Many of the quartz, chert, and quartzite pebbles of these reefs came from the Witwatersrand conglomerates and already had their round shape when they were released as a result of the disintegration of these conglomerates.

The Ventersdorp Contact Reef appears to consist of partly eluvial and partly alluvial material; a small admixture of volcanic tuff accounts for its dark colour. The presence of uraninite and gold in the Ventersdorp Contact Reef and in the Black Reef is related to the distribution of the mineralisation in the bedrock from which most of their components have been derived. Both these reefs are pyritic, and they carry uraninite and gold in those localities where they rest upon or occur in the vicinity of suboutcrops of the uranium- and gold-bearing ore beds in the Upper Witwatersrand. Away from these ore beds, uraninite and gold have hitherto not been found in significant amounts in the Ventersdorp Contact and Black Reefs. The composition of these conglomerates resembles that of the Witwatersrand banket in many respects and also shows mineralogical changes as a result of low-grade regional metamorphism.

## DISCUSSION

*B. Nagy:* Just a very minor point of semantics. I wonder if you would object to using the words "organic matter" instead of "hydrocarbons," because in the Witwatersrand Vaal Reef, which we are studying, the carbonaceous matter is not hydrocarbons. Also, I wonder, again from a point of semantics because this is going to develop tomorrow, whether the term "thucholite" should really be used for this carbonaceous matter which actually may be determined to be a polymerlike organic material. Now I want to emphasize that by polymer I do not mean polymerization of methane, but I mean a polymer like a protein, macromolecules. Would it not perhaps be better not to use thucholite, because really nobody knows what thucholite is?

*von Backström:* Dr. Nagy, I take both your points. I speak as a regional geologist.

*Wilfried Meyer, Falconbridge Nickel Mines, Ltd., Canada:* Dr. von Backström, where in the stratigraphic succession would you place any change in the atmosphere from anoxic to oxygenic?

*von Backström:* I'm afraid I wouldn't think myself capable of answering that question. I would have to refer it to colleagues who presented the previous papers.

*Meyer:* Is there an answer from anybody else?

*Grandstaff:* I think we have pointed out already that the appearance of eukaryotic organisms, the disappearance of banded iron-formations, the appearance of red beds, all at around 1.5 to 2 billion years appears to indicate that the oxygen pressure did increase in that time interval.

*Meyer:* The point I'm trying to make is this: In Canada we've got this very rapid change from pyrite-bearing beds to hematite-bearing beds in the Huronian, and this change has often been used as evidence for change in the oxygen content of the atmosphere. If you take absolute early age dating from the Witwatersrand, the corresponding change must, in my opinion, come somewhere in the Transvaal Group, or even earlier than that, yet you have detrital uraninite in the basal conglomerate there and in some other. There seems to be an inconsistency.

*Schopf:* I'm aware of that work and I think it's very interesting. It has bothered me, however, that many geologists seem to be looking at this in a rather simplistic way. It seems to me that it is probably incorrect to think of the evolution of atmospheric oxygen as being a nice smooth curve. It is often drawn that way. But I would suppose that there must have been fluctuations in the period between 2.3 and 1.9 b.y., ups and downs, that will be reflected in what will be interpreted perhaps as conflicting evidence: oxygenic con-

ditions and then maybe reducing conditions, then oxygenic conditions, and so forth. It would seem to me, however, that looking at the dynamic processes involved, that's exactly what should be expected rather than a simple non-all situation. I suspect that it's really more complicated and that there was a period of time in which both obtained.

*Grandstaff:* Also, when you're speaking about oxidation of hematite, hematite is stable above oxygen pressures around 10 to the minus 70, so what is oxidizing for hematite still may not be very oxidizing for uraninite. There's quite a range in which the two can quite happily co-exist.

*Meyer:* Yes, but all this still does not explain this time change from pyrite to hematite, fluctuations or not, because somehow you don't repeat that change. At least the pyrite in detrital form is not repeated. Why not?

*Bourret:* May I suggest that in a very broad sense we are overlooking one important factor here and that is the tremendous changes in the weather cycle. Torrential rainfall in high ancestral mountains flushed large quantities of coarse debris into the basin followed by periods of quiescence during which fine sediments were deposited and even erosional gaps developed. With repeated tectonic and weather changes, the cycle was repeated over and over again. So, in a sense, we're prone to oversimplify the cycle of change that caused the working and reworking and deposition of coarse material.

*Paul Weis, U.S.G.S., Reston, Va.:* You mentioned that the Witwatersrand conglomerates are deposited on surfaces unconformably and that the unconformities represent considerable periods of erosion or at least nondeposition. Do you feel that the deposition of the conglomerates on these unconformities represents very long periods of time or relatively short periods of time of actual deposition?

*von Backström:* Well, I think both because there are some very major breaks. Although we considered the Witwatersrand Supergroup as one formation, it is actually a large number of formations with some very distinct angular unconformities. Also, when you take these various formations, again you find minor unconformities, in this case down to very short periods where you have channel fillings and this sort of thing.

*Irving Breger, U.S.G.S., Reston, Va.:* Dr. von Backström and the previous speaker and other members of the panel, can you tell me, since I don't know, is the uraninite always associated with organic matter or can you find it without organic matter?

*von Backström:* Is it always associated with organic matter? I don't think so.

*Schidlowski:* Definitely not; I'll come back to that in my talk tomorrow.

*Samuel Adams, Anaconda Co., Uranium Division, Salt Lake City, Utah:* This goes back to the paper on the rates of oxidation of pitchblende or uraninite. You mentioned, or some of the other speakers did, the influence of thorium on the rates of that dissolution, but it seems to me that perhaps we did not pay enough attention to the state of oxidation of the uraninite itself at the initiation of those reactions. For example, if I understand your data correctly, the degree of oxidation of the samples you used ranged from about 12 percent  $\text{UO}_3$  up to about 60 percent. The sample which showed the highest rate of dissolution was the sample with the smallest amount of  $\text{UO}_3$ , it was the least oxidized.

*Grandstaff:* That's not correct.

*Adams:* That isn't?

*Grandstaff:* No.

*Adams:* That was the 22.7 figure?

*Grandstaff:* If you look at the Witwatersrand and the Blind River uraninites, both being about 60 percent  $\text{UO}_3$ , they both oxidized very, very slowly. In fact, we looked at this, and there did not seem to be any very simple relation between  $\text{U}_2 - \text{UO}_3$  ratios and rates of oxidation; they both oxidized very, very slowly.

*Adams:* Right. The figures that I have for the percent oxidation of the uranium in the Dominion Reef and the Nordic sample are 46 and 42 percent, respectively. So those really still are not as oxidized as many of the uraninite samples. And what I wonder is whether or not such studies shouldn't look at two phenomena: first, the dissolution of uranium from those pitchblends and, secondly, the oxidation of uranium followed by the dissolution. So my question would be, have you looked at the dissolution of these samples in, say, oxygen-poor solutions to see what the solubility is, strictly as a function of, say, the carbonate content, which ought to be fairly predictable? The other thing that I wonder about is the distribution of the  $\text{UO}_2$  and the  $\text{UO}_3$  in the uraninite. In other words, it may not be uniformly distributed so that even these ratios may not tell us very much about the behavior of your samples until we know how the oxidation states are distributed within the grains; in other words, it may already be armored with  $\text{UO}_3$ .

*Grandstaff:* The samples I took were chemically homogeneous, at least to the microprobe. Obviously you cannot get at oxidation states with a probe; you have to do this by titration with ferric sulfate, or ferrous sulfate, but in X-ray, using diffraction, there was no line broadening or any suggestion that the uraninite might have been heterogeneous with respect to  $\text{UO}_2 - \text{UO}_3$ . Further, we took big uraninite crystals, took off the outside layer and any gangue minerals and anything that didn't look like uraninite. Then we crushed them so that the surfaces were all reasonably

fresh, fresh as one can make them, at any rate geologically. We looked at the dissolution reaction in water in those experiments. In a solution without carbonate a scale of schoepite or some hydrated form of  $\text{UO}_3$  forms on the surface of the fragment. Oxidation probably forms  $\text{UO}_3 \cdot 2\text{H}_2\text{O}$ , then carbonate attacks this armored surface and pulls off the hexavalent uranium exposing another fresh surface, so the two actually go on in steady state with respect to one another.

*Adams:* I would still submit that the heterogeneity of the samples wouldn't be demonstrated even by breaking

off the outsides, and it might not be surprising that the rates of dissolution would be very difficult to correlate with chemical parameters. Another comment I'd like to make just in passing is that commonly when we speak of uraninite and try to understand it either in the conglomerates or the veins, we are rather casual about precisely what we are talking about, in particular what is the oxidation state of the uranium? And if anyone would like to pursue that subject, I think it would add a great deal to at least nailing down the chemical parameters of the systems we are trying to understand.

Examples That Illustrate Sedimentological Aspects of  
the Proterozoic Placer Model on  
the Kaap-Vaal Craton, Witwatersrand, South Africa

By W. E. L. MINTER

GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN  
QUARTZ-PEBBLE CONGLOMERATES

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GEOLOGICAL SURVEY PROFESSIONAL PAPER 1161-E





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## GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN QUARTZ-PEBBLE CONGLOMERATES

# EXAMPLES THAT ILLUSTRATE SEDIMENTOLOGICAL ASPECTS OF THE PROTEROZOIC PLACER MODEL ON THE KAAP-VAAL CRATON, WITWATERSRAND, SOUTH AFRICA

By W. E. L. MINTER<sup>1</sup>

### ABSTRACT

In the fluvial-fan model of Proterozoic placers in South Africa, midfan and fanbase environments are illustrated by the Ventersdorp Contact Reef, proximal braided-belt environments by the B Reef, distal braided-belt environments by the Vaal Reef, and deltaiclike environments by sheets, ribbons, and pods at the extremities of the South and Basal Reefs. The nature of distribution of gold and uranium in these various environments has been evaluated and the knowledge successfully applied by the author to the valuation and practical mining of the reefs since 1965.

### INTRODUCTION

My contribution is essentially sedimentological. I have divided the contribution into five short papers so that they can be inserted at opportune intervals between other papers to remind us that these samples come from rocks and that we ought to view the results and shape our thinking in the light of the sedimentological framework from which the samples have been derived.

In the conceptual model of gold-bearing placers on the Witwatersrand, described by Pretorius (1966), alluvial fanhead, midfan, flanks and fanbase environments were included. Pretorius has more recently (1974) reworded "alluvial" to read "fluvial," but the fan terminology remains the same. During recent years a considerable amount of detailed sedimentological research on many of these "reefs" has been accomplished. In particular, results of research by Armstrong (1965), Knowles (1966), Sims (1969) and Minter (1972), presented in theses to the University of the Witwatersrand, have confirmed the applicability of Pretorius' model and have provided data with which to identify the sedimentary environments that prevailed. These principles have been extensively and successfully applied to the evaluation and exploitation of many different Witwatersrand reefs

(Minter, 1969, 1970), and the results are recorded in several confidential company reports prepared between 1965 and 1973.

### THE PRESENT MODEL

The preservation potential of placers in proximal environments along the upwarping edge of the intracratonic Witwatersrand basin was low sedimentologically because of transgressions and low structurally because of subsequent peripheral faulting. Conversely, the preservation potential of placers in distal environments was high. The distal extremities of placer deposits have not been accessible, however, because of technical difficulties associated with mining diminishing gold contents at increasing depths. The full sedimentary sequence in the model of a Witwatersrand reef has consequently been constructed from the economic parts of various reefs that have been geologically preserved and exploited.

In essence, the present model describes the dispersal of sediment from eroding auriferous provenance areas down dendritic systems of tributary channels into braided belts and finally into deltaic-shaped sand sheets, ribbons, and pods. Placer deposits were developed in all these environments, and the rock records display characteristics appropriate to the various positions down the paleoslope.

Witwatersrand reefs have been broadly classified into two different groups, the bankets and the carbon-seam reefs. The bankets were deposited in proximal environments and are composed of pebble-supported conglomerates with detrital heavy minerals dispersed throughout their matrix. The carbon-seam reefs were sandbodies containing a minor small-pebble population distributed as single-layered lags, as scattered pebbly

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quartzites, and, rarely, as pebble-supported conglomerates. Detrital heavy minerals were concentrated at the base of the unit; lesser amounts were concentrated on sedimentary partings within the reef unit. The carbon-seam reefs were deposited in distal environments. In both reef groups, the reef unit is defined by the maturity of its sand-sized sediment that is cleaner than that in the sediments either above or below the reef unit.

#### ALLUVIAL-FAN PLACERS

Preserved gold-bearing alluvial-fan deposits appear to be confined to the Ventersdorp Group, which was largely shed off reworked Witwatersrand horsts and deposited into structurally controlled yoked basins. The most extensive example is the Ventersdorp Contact Reef in the Far West Wits Goldfield near Carltonville. This deposit, described by Knowles (1966), is of the blanket type. At its proximal end, on West Driefontein, it is composed of a thick accumulation of very coarse, somewhat angular clasts (figs. 1, 2) with a mean size in excess of  $-4$  phi units. The largest clasts reported are up to 36 cm in diameter. At a distance of 2.5 km down the paleoslope from this position, the mean pebble size has decreased rapidly to less than  $-3$  phi units. The standard error of these mean size measures is 0.02 phi units.

Although the maturity of the reef indicates a wet alluvial-fan rather than a dry alluvial-fan environment, black shale interbedded in the conglomerate could be interpreted as playa-lake muds (fig. 3). Terracing or multilevel channelling is evident, but insufficient areally spread data are available to outline details of channel patterns.

As in all bankets, the detrital heavy-mineral concentration is closely related to packing density. Maximum concentrations occur in densely packed (pebble-supported) conglomerate, whereas pebbly quartzites and quartzites of the reef unit are poorly mineralized.

Similar but less extensive wet alluvial-fan deposits were shed off the Witpoortjie horst in the West Rand. At South Roodepoort (Minter, 1970), reworked material was shed southeast off the Roodepoort Fault scarp, which dips  $45^\circ$ . About 6 km of the midfan part of the deposit has been preserved. A very narrow fan was shed northwest off the Witpoortjie Fault scarp, which dips  $85^\circ$  (Minter, 1971, personal files).

#### FLUVIAL-FAN PLACERS

The Livingstone Reefs described by Steyn (1963) at West Rand Consolidated on the West Rand have a fluvial-fan geometry. As many as twelve gold-bearing

conglomerate layers are interbedded with quartzites to form a clastic cone of sediment, which is over 30 m thick at its proximal end and thins laterally and downslope, within the area of economic interest, to about 15 m. Over the exposed downslope distance of 2 km, the mean of the ten largest pebbles changes from 40 mm to 27 mm. The reefs that have been mined in this deposit are of the blanket type.

#### PROXIMAL BRAIDED-BELT PLACERS

The most commonly preserved Witwatersrand reefs were deposited in braided-belt environments. In their more proximal position they occur as channels averaging 50-100 cm deep with a channel index of as much as 500. The channels, as in the B Reef at Freddies in the Orange Free State Goldfield, were eroded into the underlying footwall sediments and are separated by minor interchannel topographic elevations. The chan-

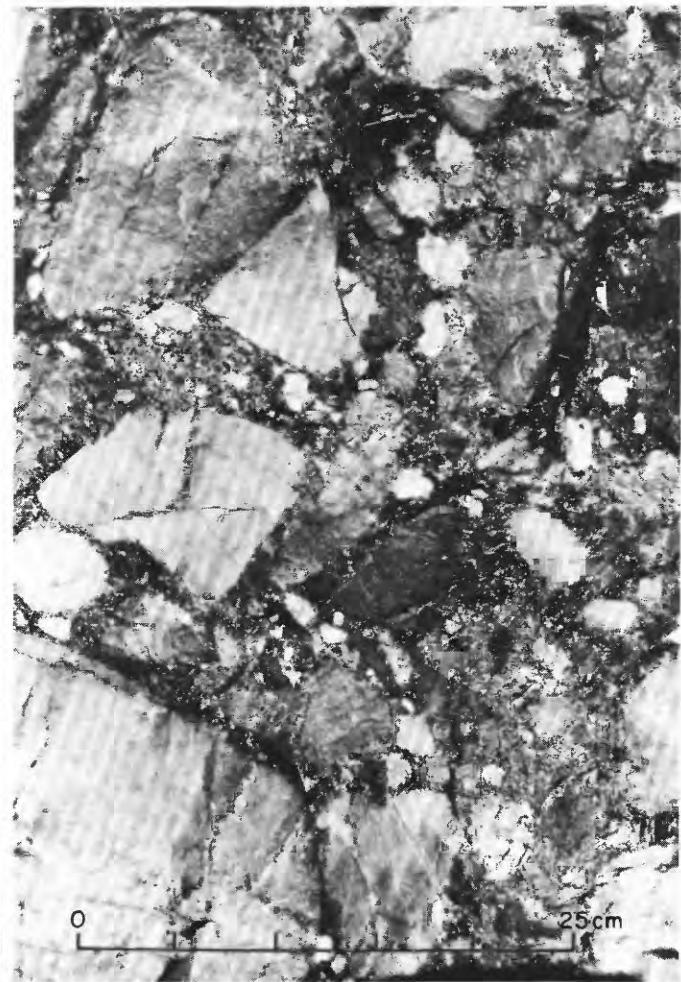


FIGURE 1.—Large subangular clasts of the Ventersdorp Contact Reef in a proximal, wet alluvial fan, position at West Driefontein gold mine near Carletonville on the Far West Witwatersrand.



FIGURE 2.—A thick layer of coarse, poorly sorted Venterdorp Contact Reef at West Driefontein, where the deposit is considered by Knowles (1966) to represent the base of an alluvial fan.

nels, defined by their fill of more mature sediment, have well-defined edges (fig. 4A). They are either filled with large pebble-supported conglomerate containing detrital heavy minerals throughout the matrix or with mature pebbly quartzite or quartzite, in which detrital heavy minerals are less well concentrated. The conglomeratic fills in some instances are gravel bars (fig. 4B). In shallower channels only thin lags occur (fig. 4C), overlaid by argillaceous quartzite. These lags may, however, contain very high concentrations of detrital heavy minerals (fig. 4D).

#### DISTAL BRAIDED-BELT PLACERS

Braided belts in a more distal position anastomose to form an apparent sheet of placer sediment, for example, the Vaal Reef (fig. 5; see Minter, 1972). The channelled bedforms are still evident (fig. 6), however, and indicate the central position of the channelways. Channels are as



FIGURE 3.—Large-pebble Venterdorp Contact conglomerate interbedded with laminated black shale, which in turn is channelled by prograding conglomerate and buried beneath amygdaloidal lava. Western Deep levels gold mine near Carletonville on the Far West Witwatersrand.

much as 100 cm deep but generally are less than 50 cm and have a channel index of as much as 3,000. The placer sediment in this more distal position is no longer a pebble-supported conglomerate reef (banket) but is a pebbly quartzite carbon-seam type of reef. The preservation of detrital heavy-mineral concentration was not dependent on physical protection by pebble layers (Minter and Toens, 1970) but, in the prevailing lower-energy environment, the heavy minerals were deposited on sedimentary partings in the reef, usually the bottom contact (fig. 7).

In the very distal positions, as in some South Reef exposures at West Rand Consolidated in the West Rand Goldfield and in the most easterly borehole exposures of the Basal Reef in the Orange Free State Goldfield, the reef unit becomes a thin sand layer with occasional grits or small pebbles (–2.5 phi units) and a thin detrital layer of heavy-mineral concentrates. The reef unit is patchily distributed in very distal environments, possibly taking the form of pods and ribbons in deltaic-shaped areas.

### CONCLUSIONS

Detailed sedimentological data from Ventersdorp and Witwatersrand reefs support a fluvio-deltaic conceptual model in which a fan-shaped geometry of distribution with fan-head, braided-belt, and deltaic-sheet environments occurs.

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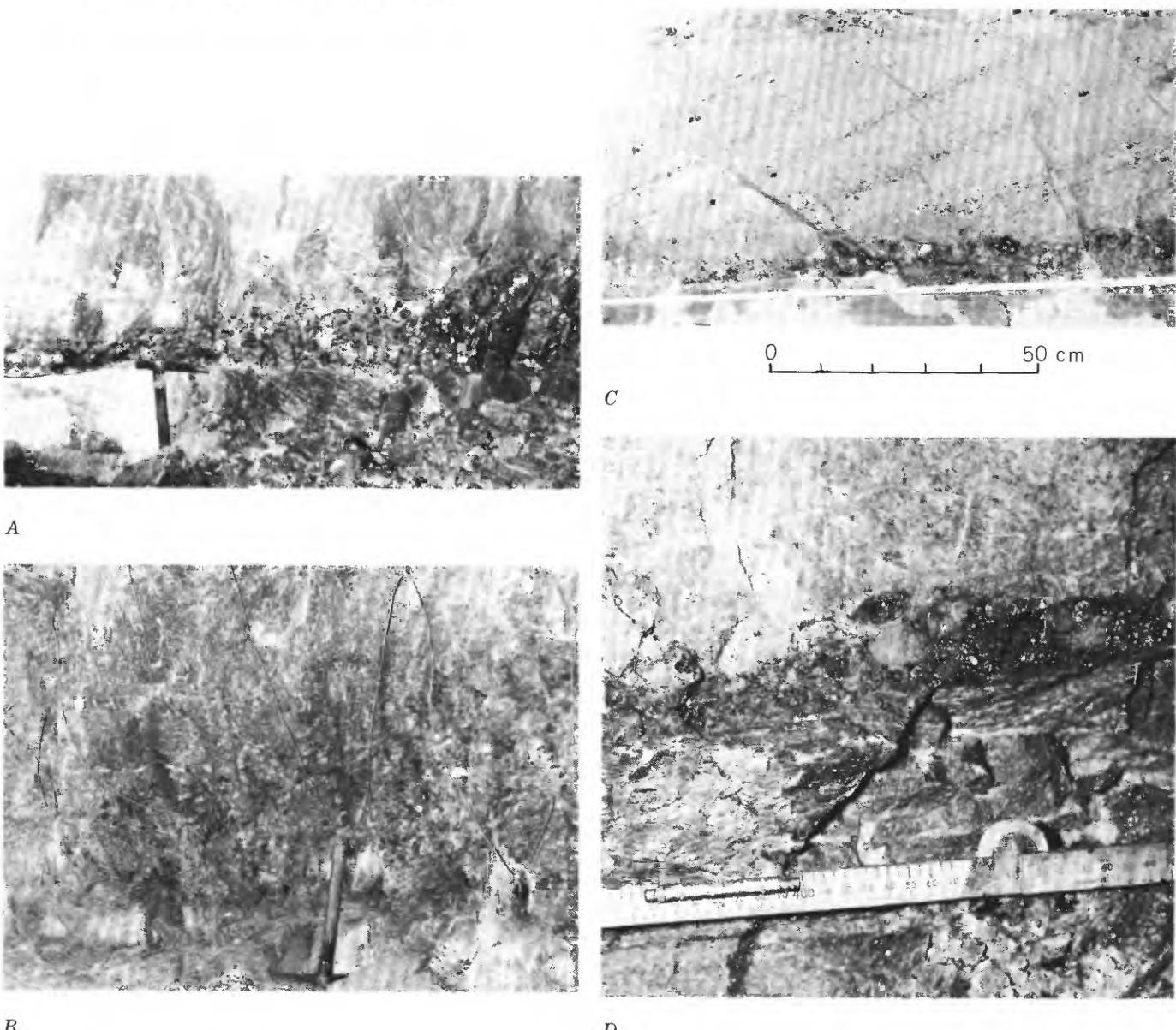


FIGURE 4.—Sections from the B Reef of a channel at Freddie's gold mine, Orange Free State Goldfield, as an example of a proximal braided placer deposit. A, The edge of a B Reef channel; the conglomeratic channel-fill thins out against the channel bank to the left of the hammer. B, The edge of a gravel bar in the B Reef; the gravel bar feathers out across the flat channel floor, which is evident at a position halfway down the hammer handle. C, A thin gravel lag of B Reef beneath crossbedded argillaceous quartzite. D, Closeup of a thin basal gravel lag of B Reef; this illustrates the dense concentration of coarse-grained nodular pyrite and other detrital heavy minerals.

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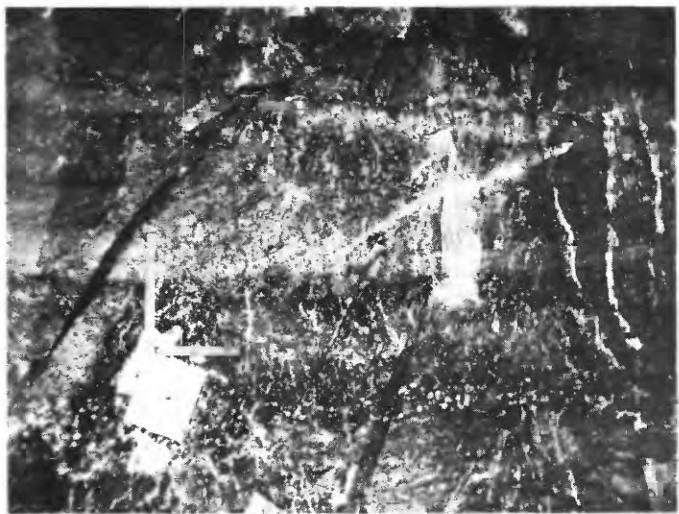


FIGURE 5.—Longitudinal view of three trough-crossbedded cosets in a paleochannel of the Vaal Reef placer, which was deposited in a distal braided-stream environment. Hartebeestfontein gold mine in Klerksdorp Goldfield. The horizontal arm of the rule, to the left of center in the photograph, is 10 cm in length.



FIGURE 6.—Transverse view of the edge of a paleochannel that has anastomosed with adjacent shallow channelways in the distal braided Vaal Reef placer, Stilfontein gold mine, Klerksdorp Goldfield.

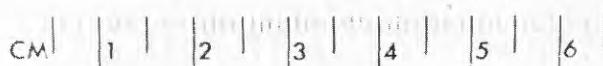


FIGURE 7.—A polished slab of Vaal Reef, which is a pebbly quartzarenite and is classified as a carbon-seam type of reef. The dense concentration of nodular pyrite that is evident is accompanied by other detrital minerals. The basal centimeter of reef, 2 cm above the base of the specimen, contains 750 ppm gold while the remainder of the reef contains 35 ppm gold.

## DISCUSSION

*Skinner:* Would it be possible for you to help me clarify my thinking and perhaps some of the others by drawing a cross section through one of these fans and perhaps putting what you think might be water depths on it?

*Minter:* Water depths? Well, we try to make calculations in connection with water depths using Allen's formula in which he bases the calculation on the actual scale of the trough crossbedding; one estimates that the water depth was about one-half to one meter.

*Skinner:* Throughout the entire fan?

*Minter:* Yes. Well, an extensive area of ten miles wide by ten miles downslope, the water depth probably is about a meter, with some areas actually being subaerial in the higher positions. In the proximal environments I think that the waters are primarily in the channel ways, but in the distal braided belts I think there was almost a continuous sheet of water with very little sediment sticking out. I would quote perhaps the Athabasca River, where you have extensive braided belts of sand almost covered by water and very shallow water conditions. Do you still want a diagram?

*Skinner:* No, no, that's all right. Then would most of the sedimentation and working be done during intermittent floods, or do you envisage a continuous water flow, as in the Athabasca model?

*Minter:* Continuous water flow. I have divided my contribution into a number of papers and I will elaborate on that later, but certainly one has a very unimodal transport direction. It's a fluvial deposit with constant reworking and the velocity is such that one has a pi crossbedding developed. It's a trough crossbedding, which is a fairly high velocity crossbedding which moves along in lunate sand dunes or linguoid sand dunes and continuously destroys its form and reworks the sediment, thereby producing a very concentrated heavy-mineral concentration at the base.

*B. Nagy:* I would like to ask a question regarding the Athabasca model. The Athabasca River has fluvial plains, some of which are basically sand and some of which are basically clay. All of them are rather heavily impregnated with organic matter. This does not seem to be the case here.

*Minter:* It is the form I am really referring to.

*B. Nagy:* Apparently the history is different. I just want

to clarify in my mind that in the Athabasca model the fine-grain sediments are the ones which contain indigenous life rather than developing some kind of carbonaceous matter later on. Is that what you have in mind?

*Minter:* Yes, possibly. I think of the environment really being a coarse sand with not very much fine material, certainly not in the basal placer lode. There is very little fine material; I think the fine silicate percentage is about 16 percent.

*Nagy:* I was talking about the clay-size fraction.

*Minter:* I use that [the Athabasca] as an example because we have mapped out channel ways and this is the configuration they have.

*Simpson:* You didn't describe where the carbon was in the carbon-seam reefs. Could you tell us?

*Minter:* I am going to do that later. Is that all right?

*Skinner:* If you look at the entire basin downslope from the fans do you find enough clastic material to account for all the rest of the detritus that would have been eroded from the source areas? Is all the rest of it there?

*Minter:* Yes, I think so.

*Skinner:* You don't have some prior sorting in other pathways for the material before it gets into the Wits Basin?

*Minter:* I think that the sand was perhaps held in valleys and flushed out. A lot of concentration perhaps took place in valleys in the drainage area. I don't think there's an outlet to the Wits Basin. I think that all the material is in there. Does that answer your question?

*Skinner:* Yes, that is what I was curious about.

*Minter:* These deposits have been described as very extensive deposits but in fact when one takes them apart, one finds that they are in effect coalescing deposits, and this often accounts for the difference in gold concentration laterally.

*Hallbauer:* What significance do you attribute to the occurrence of ventifacts in these deposits?

*Minter:* I have found ventifacts in the deposits but they aren't in situ. They are upside down, some have facets on both sides, they are very small pebbles, and all have obviously been recycled. I can show you an example later in my next paper where the sedimentation prior to the deposition of one of these reefs produced a deltaic flat upon which pebbles could, and I am sure would, have been faceted.

# The Dominion Reef Group, Western Transvaal, South Africa

By J. W. VON BACKSTRÖM

GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN  
QUARTZ-PEBBLE CONGLOMERATES

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GEOLOGICAL SURVEY PROFESSIONAL PAPER 1161-F





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# GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN QUARTZ-PEBBLE CONGLOMERATES

## THE DOMINION REEF GROUP, WESTERN TRANSVAAL, SOUTH AFRICA

By J. W. VON BACKSTRÖM<sup>1</sup>

### ABSTRACT

The Dominion Reef Group, disconformably overlain by the Witwatersrand Supergroup, outcrops in four separate areas in the Western Transvaal, Republic of South Africa. It consists essentially of a basal sedimentary formation as much as 90 m thick, which contains two important uranium-bearing quartz-pebble conglomerates known as the Upper Reef and the Lower Reef; an intermediate formation of altered andesitic lava flows as much as 650 m thick with some intercalated beds of tuff; and an upper formation of altered cherty rhyolites and dacites with a thickness of at least 1,500 m. The basal sedimentary formation of the Dominion Reef Group represents the earliest sedimentation in the region (>2,800 million years) predating that of the Lower Witwatersrand Group (>2,700 million years).

This paper is devoted mainly to a description of the auriferous "reefs," their depositional environment, pay streaks that are a feature of these "reefs," their mineralogy, and the highly mineralised, pebbly, silicified sandstone known as the Pay Band present near the top of the Upper Reef.

The relation of ore deposits to structures, and the possible source of the uranium, are discussed. Reasons are given why a hydrothermal origin for the presence of uranium in the "reefs" is thought to be untenable.

### PREAMBLE

The Dominion Reef Group finds its major development south of the village of Hartebeestfontein and around the village of Ottosdal in the Western Transvaal (fig. 1).

The Dominion Reef mine, situated 16 km south of Hartebeestfontein, started its life as a gold producer in 1888 but, owing to low gold grades, was subsequently closed down. It was reopened in 1936 as a gold producer, the bulk of production coming from the lower Reef, which initially yielded 5 g per metric ton; this yield stood at less than 2 g per metric ton when the mine closed down in 1953. A vigorous search for uranium was initiated in 1958, when sampling of the slimes indicated average values of more than 0.46 kg per metric ton, and the mine acquired a new lease of life in 1959, when uranium production commenced and the Upper Reef was mined ex-

tensively for uranium. When, because of overproduction, the mine was closed down in 1961, more than 1,500 short tons of  $U_3O_8$  and 13 million rands (R) worth of gold had been produced.

A recent press release (Rapport, 10 June 1975) gave the average uranium and gold values of different sections of the Dominion Reef deposit south of Hartebeestfontein as shown in table 1.

TABLE 1. - *Average uranium and gold values for different sections of the Dominion Reef deposit*

Area	Values per metric ton	
	Uranium (kg)	Gold (g)
Afrikander Lease	0.25	2.5
Rietkuil Section (part of Afrikander Lease)	1.25	.7
Rhenosterhoek	.50	1.5
KCG (Klerksdorp Consolidated Goldfields)	.89	4.5
Dominion Reef	.80	.4
Vlakfontein	.50	Trace
Ottosdal	.34	Trace
Lucaskraal (south of Ottosdal)	.15	Trace

Very limited drilling (13, 11, and 2 boreholes, respectively) was undertaken in the three areas named last on table 1; the results have been known since 1972 (von Backström, 1972).

Between 1966 and 1970, exploration by means of core drilling of the uraniferous conglomerates present in the basal sedimentary rocks of the Dominion Reef Group was undertaken. Then and during earlier exploration, 230 boreholes, with a total of 31,980 m, were completed by the mining industry. Boreholes range in depth from less than 100 m to more than 1,000 m. Large deposits of uranium of low to medium grade have been proved. Associated gold values are very low. There are in existence two developed mines, which have been closed down, but a feasibility study is under way to reopen them and other possible uranium producers.

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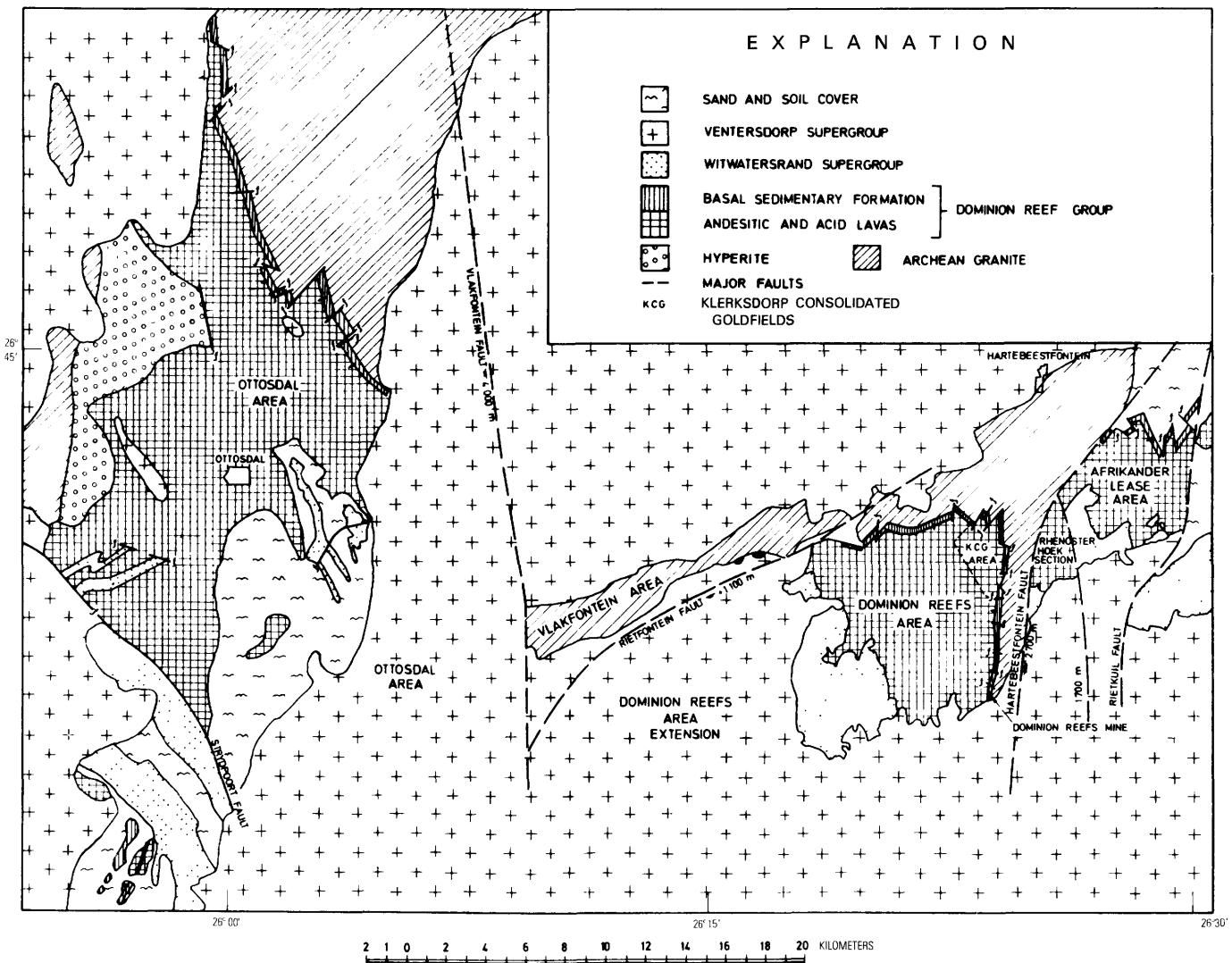


FIGURE 1.—Major geological features of study area.

### GENERAL GEOLOGICAL SETTING

South of Hartebeestfontein in the Western Transvaal, granite of the Archean Complex is unconformably overlain by quartzites and conglomerates, which together form the basal section of the predominantly volcanic sequence of the Dominion Reef Group. These rocks are in turn disconformably overlain by shales and quartzites of the Lower Witwatersrand Group. The lavas and volcano-sedimentary rocks of the Ventersdorp Supergroup cover the above-mentioned flat-lying or inclined older rocks, which are eroded to varying stratigraphic depths.

Although part of a single depositional unit, rocks of the Dominion Reef crop out in four separate areas, each of which can be considered a structural entity (fig. 1):

1. The Afrikander Lease area constitutes a fault block 7

km wide bounded by the large northeast-striking Rietkuil and Hartebeestfontein faults.

2. The Dominion Reef area forms a quasi-synclinal structure plunging to the southwest. The sediments crop out over a north-south strike distance of at least 10 km and continue westward an additional 15 km.
3. The Vlakfontein area is almost entirely covered by lavas of the Ventersdorp Supergroup, so that only two small outcrops of Dominion Reef rocks appear. The area is bounded by the Rietfontein and Vlakfontein faults. The latter fault has no surface manifestation; its presence was proved by drilling.
4. The basal sediments in the Ottosdal area, although broken by a number of cross faults, can be followed along the line of strike for 20 km north of the village before they are again covered by

Ventersdorp lavas. South of the village, the basal sediments crop out south of the Strydpoort fault.

### GEOLOGY

The Dominion Reef Group consists of three members:

1. a basal sedimentary formation as much as 90 m thick, consisting of conglomeratic, arkosic, and quartzitic rocks with subordinate shale bands;
2. an intermediate formation as much as 650 m thick, consisting of altered andesitic lava flows with intercalated beds of tuff; and
3. altered cherty rhyolites and dacites at least 1,500 m thick, with intercalated tuff bands (known as Wonderstone near Ottosdal) and schists and minor breccias at the top.

Only the basal sedimentary formation, which contains uranium and gold, is described further.

The basal formation is of economic importance because its conglomerate beds are auriferous; it has been exploited in the past for gold and uranium. Extensive resources of uranium of good grade were proved, but associated gold values are low and few developed mines exist. The economic viability of such mines would be dependent on a satisfactory and sustained uranium price based on long-term contracts.

The basal sedimentary formation of the Dominion Reef Group rests unconformably directly on the basement granite and represents the earliest sedimentation in the region ( $>2,800$  m.y. (million years)), predating that of the Lower Witwatersrand Group ( $>2,700$  m.y.). The thickness of the basal formation varies from 3 to 50 m south of Hartebeestfontein to 90 m north of Ottosdal (fig.1).

Many deep boreholes have proven that intercalated sediments, from 60 cm to 40 m thick, occur in the first 60 m of the overlying andesitic lava.

The basal sedimentary formation consists largely of the disintegration products of the basement granite. The sediments consist of medium-grained to coarse-grained feldspathic and somewhat micaceous sandstone or quartzite, in which there are lenticular bodies of grit and conglomerate. Evidently, the micaceous or sericitic material in the rocks was derived from the breakdown of the original detrital grains of feldspar. A conglomerate along or close to the base of the arenaceous sediments and another higher up in the succession, both of which have been worked or prospected for gold and uranium, are known as the Dominion Reefs. The thickest bodies of conglomerate occupy depressions in the old granite floor, whereas the conglomerates are absent on humps. Usually the lowest bed of conglomerate is separated from the underlying granite by a band of sheared granite or by a variable thickness of transition beds comprising one or more of the following rock types: arkose,

grit, and quartz-sericite schist. Where there is no basal conglomerate, feldspathic sandstone often grades imperceptibly into the granite below. In many prospecting boreholes the contact between sandstone and granite could be established only radiometrically.

Two main auriferous conglomerate zones known as "reefs," separated by barren conglomerate or quartzite, are present in the basal formation, which can be divided into (1) a lower quartzite and conglomerate member, from 0 to 40 m thick, containing the Lower Reef; and (2) an upper quartzite and conglomerate member, between 4 and 50 m thick, containing the Upper Reef.

The Lower Reef, a lenticular, well-developed large-pebble conglomerate, was the main gold carrier in the mined area. It occurs sporadically at the base of the lower member, where it occupies depressions in the erosion surface of the basement granite. It shows marked variation in texture where it rests on irregular portions of the pre-Dominion erosion surface and more uniformity where the basement is fairly even. On the whole, the conglomerate thickens and pinches in a manner reflecting the surface on which it was deposited, and deposition and preservation were controlled by depressions in the floor. The thickness ranges from 0 to more than 3 m, and pebbles vary in diameter from less than 5 to more than 10 cm. Near the base, the Lower Reef is often poorly packed and sorted and merges into a pebbly quartzite. The packing index improves at the top, where there is often the best concentration of heavy minerals, including uraninite. The quartzite is medium grained to coarse grained and sericitic, the sericitic being derived from the feldspar of original arkosic sediments.

There is no significant angular unconformity between the upper and lower members and, apart from channeling, there is little differential erosion of the lower member.

The upper member is developed throughout almost the entire area. South of Hartebeestfontein it ranges from 4 to 30 m thick; north of Ottosdal it is a maximum of 75 m thick. The Upper Reef, taken as the basal bed of the upper member, consists in fact of several conglomerate bands, which vary from poor to good in packing and sorting. The conglomerate bands may rest on any unit of the lower member. Pebbles range in size from 2 to 5 cm, are well rounded to subrounded, and consist almost exclusively of quartz. They may represent reworked and concentrated detrital material derived from the erosion of earlier conglomerate and arenaceous rocks. The Upper Reef normally ranges from 0.15 to 3 m thick; in some places it is as much as 5 m thick.

The conglomerates of both the Lower and Upper Reefs closely resemble the Witwatersrand banket and are likewise composed of pebbles of vein quartz set in a

dark-grey, highly siliceous, fine-grained matrix. The pebbles show variations in their packing index. They are usually not closely packed, and sometimes only isolated pebbles are present in the fine-grained matrix. The pebbles are often severely sheared and fissured and are also replaced along their edges and along fissures by the surrounding matrix materials. In some areas pebbles are well sorted, the larger ones being at or near the base, and are oriented with the largest axis parallel to the direction of strike. Some are split in half along their central axis parallel to the dip and are slightly displaced.

The matrix is sporadically mineralised by pyrite, and veins of pyrite have also been observed in fissures in the pebbles. In weathered conglomerate the matrix is stained by yellow-brown oxidation products of hydrated iron oxide and leucoxene.

Pay streaks similar to those found in alluvial deposits are a feature of these reefs, but unfortunately there is no general uniformity in grade, and the distribution of the uranium minerals is neither characteristic nor a significant feature everywhere. There appears to be a connection, however, between the distribution of detrital minerals like monazite and cassiterite and that of uraninite in the pay streaks. The pay streaks appear to have been formed by stream action and not by the introduction of lateral mineralising solutions. Features such as the alteration of wall rocks or uranium mineralisation crossing the stratification were not observed, and cross-cutting quartz, calcite, or other veins that might have acted as feeders are conspicuously absent.

It would appear that conditions during and after deposition of the sediments were such that they permitted the formation of a thin upper reef or, in places, an almost purely black sand well endowed with a full range of heavy alluvial material such as uraninite, monazite, cassiterite, zircon, and garnet. This reef is completely different in appearance from the main conglomerate body and is amazingly consistent, the average thickness being about 13 cm.

Although this reef always carries uraninite, a very wide range of values is encountered. The concentrations occur in well-defined bodies of considerable width, with very little internal low-grade ore. These so-called pay streaks vary in width from 130 to more than 300 m, and no high values have been encountered outside these shoots. Recognition of the above-mentioned feature is naturally of extreme importance where mining operations are concerned. As far as could be established, these streaks all follow a northeast trend and represent ancient shorelines or a channel system under deltaic conditions.

The width and consistency of the streaks seem to indicate continuity down to considerable depths and, in-

deed, the auriferous shoot in the Lower Reef, which has supplied Dominion Reef Mines with ore for 18 years, was mined to a vertical depth of 800 m. As mentioned above, the average thickness of the mineralised Upper Reef is approximately 13 cm, and there are well-defined partings above and below it. This allows the application of mining methods that, together with ore sorting, should permit a low-tonnage but high-grade mill feed.

An extraordinary concentration of heavy minerals occurs in an argillaceous, gritty, pebbly, silicified sandstone known as the Pay Band, near or at the top of the Upper Reef, from which it differs completely. It is generally less than 0.3 m thick, but can reach 1 m in thickness. It extends from the eastern part of the Afrikander Lease Area westward to the Hartebeestfontein fault along an east-west trending belt 300 to 900 m wide. The concentration of uraninite is usually three to four times that in the best zone of the Upper Reef conglomerate. Where the band is thin (20–50 mm), concentrations of uraninite may be as high as 1 percent; otherwise the Pay Band is virtually all detrital monazite and cassiterite, with minor amounts of detrital chromite, garnet, zircon, ilmenite, and columbotantalite.

## MINERALOGY

The mineralogy has been previously described in reports by Liebenberg (1955, 1956, 1958, 1960), Malan (1959), Ortlepp (1962), Taylor, Bowie, and Horne (1962), and especially Hiemstra (1968), who gave a detailed description of the following minerals:

1. Radioactive minerals: thorian uraninite, thorite, monazite, betafite, euxenite, leucoxene, and zircon
2. Sulphides: pyrite, marcasite, galena, chalcopyrite, covellite, sphalerite, cobaltite, linnaeite, arsenopyrite
3. Metal: gold
4. Silicate minerals: garnet, pyroxene, epidote, sphene, biotite, chlorite, sericite.

Uraninite, the principal radioactive mineral present, occurs as oval and roundish detrital grains similar in all respects to those in the Witwatersrand banket. According to Liebenberg (1955), the

\*\*\* galena is ubiquitous in the uraninite and much more common than in the banket of the Witwatersrand. The detrital shape of the majority of the grains has been retained, but some are reconstituted, and specks of uraninite are redistributed throughout the quartz-chlorite-sericite matrix immediately surrounding the uraninite. Minute specks of secondary uraninite also accompany the partly reconstituted grains of detrital uraninite. Contamination of uraninite by sulphides, other than galena, and by gold is rare. The grains of uraninite are slightly larger than those in the Witwatersrand banket and are also well graded.

Monazite is one of the most abundant heavy minerals in the Upper Reef, and it is preferentially associated with uraninite. In the Lower Reef the concentration is

much lower. In thin sections, the monazite is to be seen as small grains, many of them well rounded, of the order of 0.1 mm in diameter and always traversed by cracks. All samples tested contained some monazite; the ore contains about 12 percent monazite.

### RELATION OF ORE DEPOSITS TO STRUCTURES

The distribution of uraninite and gold in the conglomerate is intimately related to certain lithological and structural features of sedimentary origin.

Mineralisation is confined to the ore-bearing conglomerate and seldom ever crosses the stratification into the arenaceous wall rock, except where it has obviously been derived from the conglomerate ore body itself. There is a close relation between the distribution of values and purely sedimentary features. Richer concentrations are commonest near the base of the conglomerate along well-defined shaly or schistose footwall partings.

Both uraninite and gold can, in general, be said to be more or less uniformly and regularly distributed in the conglomerate reefs when the whole extent of the deposit is considered. However, when comparison is drawn with the much greater variation in the distribution of values shown by nearly all lode deposits, there are variations, often marked, in uranium content within the limits of each locality (fig. 1). The richer uraninite concentrations tend to collect in pay streaks, usually characterised by a closer packing of well-rounded and sorted pebbles composed mainly of quartz and by a greater concentration of other heavy-mineral particles including those of detrital origin. Except for local deviations, the pay streaks generally run roughly parallel to the original shoreline. Enrichment in gold and uraninite is usually more pronounced where the conglomerate reefs rest on intraformational disconformities. From the information available, this would seem to be the case particularly where these conglomerates occupy depressions or hollows in the floor of deposition, or where they lie against slight barriers across the floor, which acted as traps.

In some areas, high values are present in well-developed conglomerate filling depressions that are relatively shallow and have gentle slopes. Where the depth of a depression becomes excessive, and especially where its marginal slope suddenly steepens, conglomerate development in the centre of the depression is poor compared with that along its margins. The marginal portions of such depressions are characterised by well-developed and closely packed conglomerate generally containing high values of gold and uranium, whereas the central portion of the depressions is usually filled with relatively thin conglomerate bands separated by barren medium-grained and gritty quartzite.

The general distribution of payable concentrations of gold and uranium in the conglomerate is parallel to the original shoreline of the sedimentary basin as inferred from the spread of the coarser sediments. Richer concentrations in the conglomerate tend to be disposed in pay streaks, which have many of the characteristics of those found in modern placers, and a similarity in mode of deposition is strongly indicated. The distal part of an ore-bearing conglomerate sheet may break up into isolated lenticular bodies, which diminish in size and number until ultimately they are spaced too far apart for profitable mining. There is a decrease in pebble size and metal content outwards from the point of entry into the depositional basin.

In summary, uranium, which is intimately related to gold, occurs in bedded conglomerate and quartzite near the margin of a continental structural basin that is situated well within the boundaries of what is regarded as a uranium province. It is assumed that the uranium and the gold were deposited together as an integral part of the sedimentary series, but after the conglomerate was laid down, and that they were transported no very great distance, by water, from an original source (still unknown) to the north and west.

### SOURCE OF THE URANIUM

There are two obvious possible primary sources of the uranium: (1) older rocks from whose destruction were derived the materials that form the shale, quartzite, and conglomerate of the succession; and (2) fluids containing uranium that penetrated the sediments during or after their deposition and from which the oxide was precipitated.

The sedimentary rocks were deposited in a large intracratonic basin whose boundaries have not yet been precisely defined. If we consider the Dominion Reef Group, with its preponderance of lava over sediment, as part of the Witwatersrand Supergroup, and if we accept the correlations of both the partly exposed rocks of the Godwan beds of the Uitkyk Formation and the Godwan beds of the Derdepoort strip near the Transvaal-Botswana border with the lower beds of the supergroup, it is clear that this basin of deposition diminished considerably as time progressed.

The material that was transported into the basin must have been derived primarily from the exposed parts of the basement complex—from the sediments and metamorphosed sediments of the Kheis and Swaziland Supergroups (*sensu lato*) and from the magmatic rocks intruded into them—mainly granites and pegmatites, the so-called “Old Granite.” In such a basin, coarser and heavier sediments would tend to have been deposited close to the shores, the finer and lighter material in

deeper water. As the area covered by water shrank, the coarser equivalents of the Dominion Reef, Hospital Hill, Government Reef, and Jeppestown Groups would be exposed and could have contributed detritus to the overlying Main-Bird and Kimberly-Elsburg Groups. Moreover, there is abundant evidence in the areas that are being mined that fluctuations of water level within the basin and local tectonic movements led to considerable reworking of already deposited material.

If the uranium now present in the conglomerates were syngenetic, it could have been carried into the basin either in solid form or in solution, or both. But having arrived there, it had to remain—subject to the natural processes of decay and, if in solution, of groundwater movement.

The source rocks of the Witwatersrand Supergroup and the Dominion Reef Group are known to be mineralised, and their mineralisation is almost wholly pre-Witwatersrand in age. Part of the mineralisation is a uranium mineralisation. The question naturally arises, therefore, whether the quantity of uranium present in these beds, and particularly in the beds above the Jeppestown Formation, of which we have more detailed knowledge, is greater than could be accounted for on the assumption of a syngenetic origin.

Any attempt to answer such a question involves the consideration of a number of factors of which our knowledge is very scant. Among these factors are the total original extent of the sediments of the succession, the degree to which earlier beds in it were eroded and incorporated into later beds, the average uranium content of the source rocks, and the average uranium content of the existing remnants of the system.

Only for the last of these factors have we sufficient information available to permit a rough approximation to be made. This information is derivable from assays of core samples from comparatively few of the hundreds of boreholes that have been drilled in the search for auriferous reefs in the Upper Witwatersrand beds. Nel (1958) states that he obtained "information on uranium distribution in a vertical direction" from unpublished notes by S. H. Haughton, formerly in charge of the Geological Unit. Haughton reported that "physical assays of cores from certain boreholes logged in the Orange Free State and Klerksdorp Goldfields have been

made." His information, as cited by Nel, is summarised by the author in table 2.

The important, but at present unknown, factor concerning these cores is the actual average uranium content of the sections which carry less than 0.003 percent. Simpson's borehole logs of radioactivity (1951) show adequately that the radioactive content of quartzitic members of the succession varies from layer to layer and from hole to hole. A few random specimens have yielded, on assay, values of up to 0.001 percent  $U_3O_8$ , but it would be hazardous to accept this, or any other figure, as an average of all material carrying less than 0.003 percent in order to arrive at an average for the whole succession in a given area. A significant fact revealed by these figures is that the important concentrations of uranium are to be found only in certain narrow zones, which usually form less than 5 percent of the whole Upper Witwatersrand Group—the average usually being about 2 percent. The remainder is not barren and generally shows as "normal" on the radiometric logs, which is higher than the "normal" of the igneous material that intrudes the sediments. But there is not, in this 95 to 98 percent of the sediments, anything like the concentration found in the other 2 to 5 percent. What is true and significant, however, is that the average uranium content would appear to be much higher than that of the many billions of metric tons of granite in the world which, on average, contain 3.5 ppm uranium and which have been mooted as very low-grade potential uranium resource rocks.

With regard to the uranium content of the granite basement, the presence of the pre-Witwatersrand granitic rocks in an overturned sequence of strata at Vredefort afforded a unique profile covering a considerable vertical section of the upper and middle crust. Uranium determinations of the "normal" granite undertaken by Hart, Nicolaysen, and Gale (in press) averaged 2.45 ppm (range, 1.03–3.93 ppm) or 0.000245 percent. The outermost margins of the central core, however, showed much higher values, 20–30 ppm, or 0.002 and 0.003 percent uranium. The uranium concentration found in 2 to 5 percent of the sediments of the Witwatersrand Supergroup is thus from 3 to 70 times greater than that present in the underlying basement granite.

TABLE 2.—*Uranium values of upper Witwatersrand sedimentary rocks*

Area	Formation	Core (m)	Mineralized (percent)	Number of holes	Equivalent percent $U_3O_8$ <sup>1</sup>
Klerksdorp Townlands	Upper Witwatersrand, Gold Estates Group, Vaal Reef, Ada May Reefs, and Commonage Reefs.	4,943	5.29	8	0.110
Potchefstroom	Upper Witwatersrand	7,035	2.8	8	.080
Ventersburg OFS	Upper Witwatersrand, Footwall quartzite, and Basal Reef.	4,068	1.33	8	.170

<sup>1</sup> Cutoff. 0.003 percent.

## DEPOSITIONAL ENVIRONMENT

Since Witwatersrand gold was first discovered in 1886, there has been no unanimity amongst geologists as to the conditions under which Dominion Reef and Witwatersrand sedimentary rocks were formed, as is witnessed by the large number of papers propounding the many different theories. The reader is referred to the excellent recent review by Pretorius (1975) of this large volume of information, which he summarised as follows:

A review has been undertaken of the very large volume of information that has been published in the 89 years since gold was first discovered on the Witwatersrand.

The literature has been surveyed for opinions voiced and theories propounded for the nature of the depository in which the sediments were laid down and the environmental conditions under which the gold- and uranium-bearing reefs were formed. The review has not been directed towards examining the arguments for and against the various hypotheses that have been advanced concerning the source of the metals and the mineralising processes that emplaced them in the conglomerates, quartzites, and carbon seams. It is considered that about 90 contributions to the literature can be ranked as having added significantly to a better understanding of the depositional environment. It is apparent that, in the early period of mining operations, speculation favoured the conglomerates to have been formed on a beach. As limited amounts of quantitative data become available, preference was given to a deltaic and, later, a valley-flat environment. With the advent of detailed sedimentological studies, conclusions became far more objective, and systematic observation replaced subjective speculation. The result is that a fair degree of consensus now prevails on deposition having taken place along the interface between a fluvial system that brought the sediments and heavy minerals from an elevated source-area to the northwest and a lacustrine littoral system that reworked the material and redistributed the finer sediments along the shoreline. The goldfields were formed as fluvial fans that built up at several points along the northwestern periphery of the intermontane, intracratonic lake, or shallow-water inland sea. Each fan was the result of sediment accumulation at the mouth of a river, which discharged through a canyon and flowed across a relatively narrow piedmont plain, before entering the basin. The review has also covered ideas on the origin of the carbon that is present in the sediments, since it has now been shown to be an important environmental indicator. Evidence is strongly in favour of the material representing the remains of algal colonies that flourished, at certain times only, about the mouths of the rivers. There is a suggestion that the plantlike forms might even have been more akin to lichens. Other ideas are subscribed to by many geologists, and there can be little doubt that, when the centenary of the Witwatersrand goldfields is celebrated in 1986, disputes will still be raging as to how and where the auriferous sediments formed.

Hallbauer (1975), with the aid of the necessary scientific instrumentation developed in recent years, established that a well-differentiated plant life existed during Witwatersrand times, including bacteria, algae, fungi, and lichenlike plants.

Sharpe (1949) first put forward the idea that the carbon found in the basket might be the remains of some of the earliest forms of life.

Liebenberg (1955) emphasized that thucholite is very scarce in the Dominion Reef and could hardly have

played any significant role in the deposition of the uranium and gold in these rocks.

## EPILOGUE

With artists' licence, supporters of hydrothermal introduction paint impressive and interesting pictures of the amazing ability of mineralising solutions to move from intrusive igneous bodies deep into sedimentary rocks, without leaving any trail, until the site of deposition is reached, and then to select, in an uncanny manner, only certain beds in which to deposit their load of uranium. Yet, as far as the distribution of uraninite and gold in the Witwatersrand Supergroup and contiguous formations is concerned, there are many features which hydrothermalists do not seem to have been able to explain satisfactorily—features, for example, such as the absence of ore tracts between a hydrothermal source and the distant sites of ore deposits, the more or less uniform and consistent distribution over hundreds of square kilometres of gold and uraninite in the economic conglomerate reefs, the fact that the mineralisation does not cross from these conglomerates into quartzite partings and adjacent beds (including other conglomerates), the lack of mineral zoning and alteration of wall rocks—all of which features tend to refute hydrothermal lode deposits.

Notwithstanding detailed regional mapping, the vast amount of deep and extensive mining from the West Rand to the Orange Free State goldfields, and widespread prospecting (including the large number of prospecting boreholes, some of which are as much as 3,500 m deep), no intrusive bodies have been found which could have acted as a source of radioactive mineralisation solutions. But hydrothermalists confidently believe that there are such hidden or unknown sources deep down in, or elsewhere in other rocks beyond, the Witwatersrand Basin—no matter how far away. In passing from these unknown sources, the mineralising solutions have left no trace of their paths through and across the succession of four supergroups (the Dominion Reef, Witwatersrand, Ventersdorp, and Transvaal) with a total thickness of at least 18,000 m (perhaps even much more), assuming, as postulated by hydrothermalists, that the mineralisation solutions were introduced in post-Transvaal time. No channels along which these solutions could have been introduced have been found. Hydrothermalists therefore contend that all these formations were simply permeated by the mineralising solutions. In whichever way the solutions were introduced—whether along channels or by permeation—they completely disregarded favourable sites for deposition of their load of minerals, for example, permeable volcanic tuffs in the Witwatersrand and

Ventersdorp Supergroups, volcanic breccias in the latter, and breccias in the Dolomite Formation of the Transvaal Supergroup. Instead, the solutions selected the lenticular Black Reef and Ventersdorp Contact Reef, as well as certain preferred conglomerate beds in the Witwatersrand Supergroup, which form some 2 percent of the total number of conglomerates in the Upper Division of this Supergroup. The mineralising solutions also selectively mineralised the stratigraphic equivalents of the ore reefs in areas where the conglomerates are not continuous sheets but are lenticular patches that become smaller and smaller and are spaced many kilometres apart. The solutions even selectively mineralised the lenticular conglomerates at the base of the Dominion Reef Group.

The age of the mineralisation postulated by hydrothermalists, namely, post-Transvaal, is definitely suspect on geological grounds. In barren Ventersdorp conglomerate and volcanic breccia, well-mineralised boulders of Witwatersrand rocks have been found, showing that the gold and uraninite were already in fully consolidated and cemented conglomerate before Ventersdorp time. The principal economic conglomerate reefs in the Witwatersrand Supergroup were cut by streams during intraformational sedimentary breaks. Several exposures underground reveal islands or remnants of reef completely surrounded by impervious and barren argillaceous channel fillings. The tenor of the reef in the islands is the same as that of the broken reef on opposite sides of the channels. Boulders of reefs cut by transgressive stream channels have also been recorded in barren channel fillings. Such channel fillings, which consist of conglomerate or quartzite, carry higher or lower values than the principal reefs across which they cut. Such occurrences can reasonably be interpreted as indicating that the gold and uraninite were present in the conglomerates before they were cut by stream channels during intraformational breaks in sedimentation.

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# The Crossbedded Nature of Proterozoic Witwatersrand Placers in Distal Environments and a Paleocurrent Analysis of the Vaal Reef Placer

By W. E. L. MINTER

GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN  
QUARTZ-PEBBLE CONGLOMERATES

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GEOLOGICAL SURVEY PROFESSIONAL PAPER 1161-G





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## THE CROSSBEDDED NATURE OF PROTEROZOIC WITWATERSRAND PLACERS IN DISTAL ENVIRONMENTS AND A PALEOCURRENT ANALYSIS OF THE VAAL REEF PLACER

By W. E. L. MINTER<sup>1</sup>

### ABSTRACT

The Proterozoic placers in South Africa that were deposited in distal environments are referred to as carbon-seam reefs. They are pebbly quartz-arenite units with a characteristic pi-crossbedded internal geometry. Their external geometry, which is sheetlike, was produced by the braiding of shallow channels in a dendroidal belt. Low-variance, unimodal, paleocurrent data from the Vaal Reef are used to illustrate a transgressing, braided, fluvial environment followed by longshore drift in a deepening water environment. The total variance, evident in detrital heavy-mineral concentrations of carbon-seam placers, is related to the internal geometry, external geometry, and areal position within the deposit.

### INTRODUCTION

I have introduced the conceptual model (this volume, chap. E), and I think we are aware of the framework. In this paper I want to make you aware of the processes that actually took place and that produced heavy-mineral concentrates.

The term "reef" as used in the Witwatersrand context refers to a fossil placer deposit. During early mining activities the reefs being mined were mostly "bankets," pebble-supported conglomerates, that were identified by the pebbly layer. Most reefs being mined today are of the carbon-seam type, in which pebbles are of less significance. Nevertheless, the term "reef" has been applied to the pebble layer often evident on the basal contact of carbon-seam reefs. This layer is, however, only a part of the whole reef.

### CROSSBEDDED INTERNAL STRUCTURE

The carbon-seam type reef is a siliceous quartzite (quartz-arenite) unit that is overlain and generally underlain by quartzites that are more argillaceous. This is an important aspect of the identification of a reef unit

and is closely related to the nature of the internal geometry. Every carbon-seam reef examined by the author has been characterised by the presence of trough crossbedding, more explicitly defined as pi crossbedding (Allen, 1963).

Trough crossbedding is a structure that develops at a higher flow velocity than that required for planar crossbedding. Planar crossbedding represents the primary structure that develops at the lower velocity end of a range of crossbedded structures in which the linear edge of the lee side of the sand dune becomes increasingly curved with increasing velocity. Trough crossbedding is developed by a scour-and-fill mechanism, and most units owe their origin to the migration of trains of large-scale asymmetrical dunes with pronouncedly curved crests. The coarse sand deposited under this flow regime contains very little muddy material (figs. 1, 2).

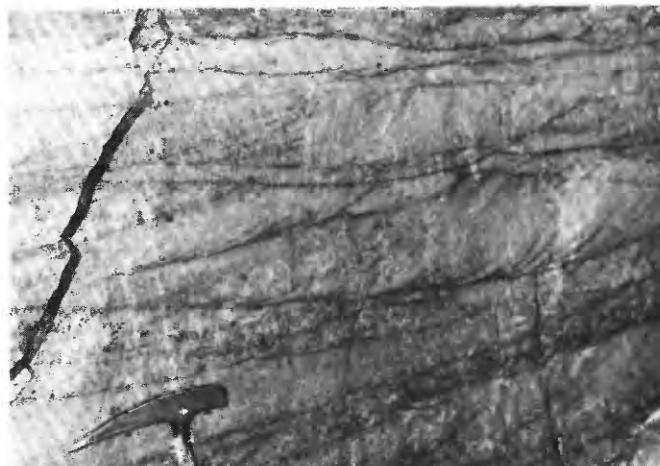


FIGURE 1. - A longitudinal view of trough crossbedded sets illustrating the external geometry of each set and the concentration of dark heavy detrital minerals on the toes of foresets, which merge along the trough-shaped base of each set. Footwall quartzite at Free State Saaiplaas in the Orange Free State Goldfield.

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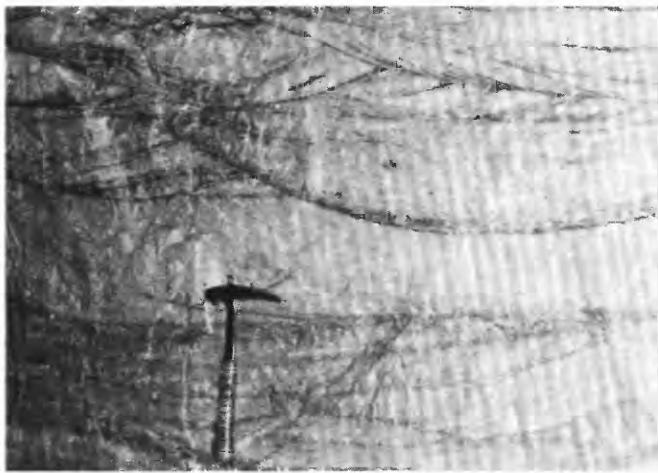


FIGURE 2.—A transverse view of trough crossbedding to illustrate the slight discordance of the curved foresets, which dip in both directions, with the trough-shaped base of the sets. Footwall quartzite at Free State Saaiplaas in the Orange Free State Goldfield.

It is evident from figure 3 that this mechanism also operated in proximal environments where coarse-grained sand containing large pebbles and coarse-grained heavy minerals was transported as 1-m-high sand dunes. However, these are rarely observed in proximal environments, probably because they are destroyed. Trough crossbedding is much more common in distal deposits like the Carbon Leader (fig. 4).

#### THE VAAL REEF

The Vaal Reef is an ancient placer that was deposited 2,500 million years ago over an area of 260 km<sup>2</sup> on the Kaap-Vaal Craton in the Western Transvaal (Minter,

1972, 1976). The placer, stratigraphically situated in the Main-Bird Series of the Witwatersrand System, lies upon an extensive, regular, angular unconformity with a paleoslope truncation angle of between 1:120 and 1:350 (fig. 5).

Mining, 2,000 m below surface, of the 1-m-thick sheet of sedimentary rock that contains the placer deposit has provided superb exposures and a unique opportunity to analyse Proterozoic paleocurrents in detail.

The placer sediment, which is a pi-crossbedded quartzarenite with a mean grain size of 2.54 phi units, was deposited in a dendroidal fluvial system of shallow, braided channels (fig. 6). Measurements of 484 foresets from longitudinal sections of the trough-shaped sets indicate a mean scale of 8 cm, a mean inclination of 18°, a vectoral mean of 144° with a variance of 4,721 (fig. 7). Analysis of 142 foresets from a local part of the deposit indicates a variance of 2,209, which implies that the increase in variance obtained from more widespread data is attributable to "between braided belt" variance. Areal distributions of moving-average vectoral-means conform closely with local directions of the braided channels.

Examination of a quartz-arenite unit beneath the angular unconformity, upon which the placer lies, indicates that it is a lobate body that lies on a disconformity. Analysis of 480 foreset measurements in this pi-crossbedded unit indicates a mean scale of 8 cm, a mean inclination of 23°, a vectoral mean of 150° with a variance of 2,484 (fig. 8). Analysis of 297 foresets from the same local part of the deposit selected before indicates a slightly reduced variance of 2,169. It is evident that very little variance can be attributed to "between braided belts" and that the unit is essentially a fluvio-deltaic fan.

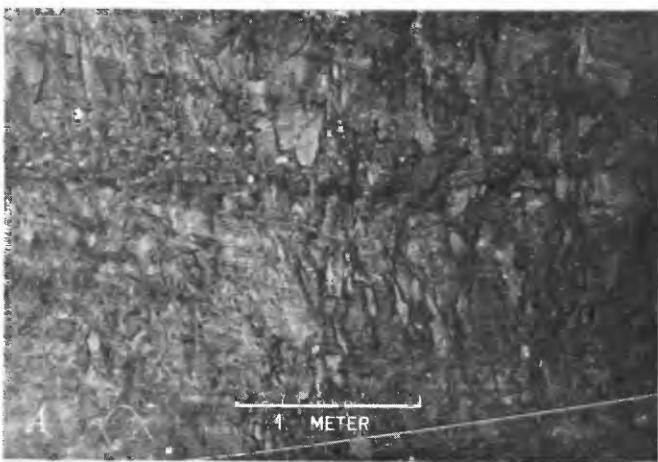


FIGURE 3.—Two oblique, transverse views of very large scale trough crossbedding in proximal Steyn Reef at Western Holdings Mine, Orange Free State Goldfield.



FIGURE 4.—Distal Carbon Leader Reef illustrating a longitudinal view of the lowest set of trough crossbedded quartzite with dark detrital heavy-mineral concentrations on foresets, bottomsets, and particularly at the base of the unit at a position halfway down the hammer handle. Blyvooruitzicht, Far West Wits Goldfield.

The sediment overlying the Vaal Reef placer is a pi-crossbedded argillaceous quartzite. Mining of the placer unit has exposed the base of this overlying unit, on which trough forms are evident. Analysis of 500 measurements of axes of these troughs indicates a mean scale of 25 cm and a vectoral mean of 065° with a variance of 269 (fig. 9). Analysis of 193 measurements from the same local part of the deposit selected before indicates a slightly reduced variance of 256. The small difference is attributed to a regional curve in the paleocurrent direction from 045° to 085° as the paleocurrent follows the strike of the angular unconformity, which describes an embayment.

Integration of these paleocurrent data from three superimposed quartzite layers reveals deposition of a fluvio-deltaic fan, transgressive truncation of the fluvio-deltaic fan and deposition of a Proterozoic gold and uranium placer by braided channel processes, burial of the placer beneath argillaceous quartzite, which was transported and deposited by longshore paleocurrents (fig. 10).

The very low variance of paleocurrent vectors measured from pi-crossbedded sets in this study ap-

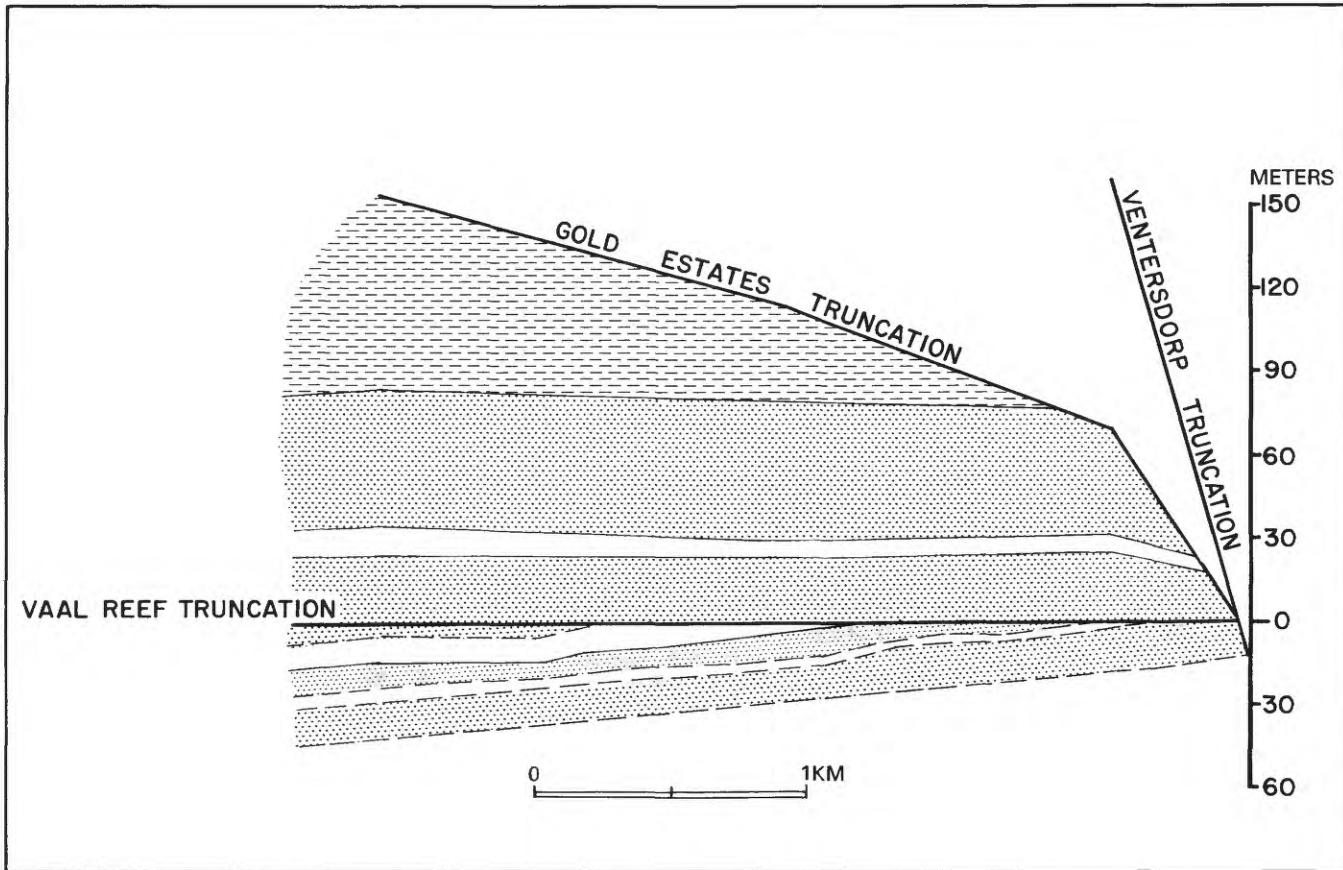


FIGURE 5.—A paleodip section of the strata in the vicinity of the Vaal Reef, illustrating the angular unconformity that the reef was deposited on.

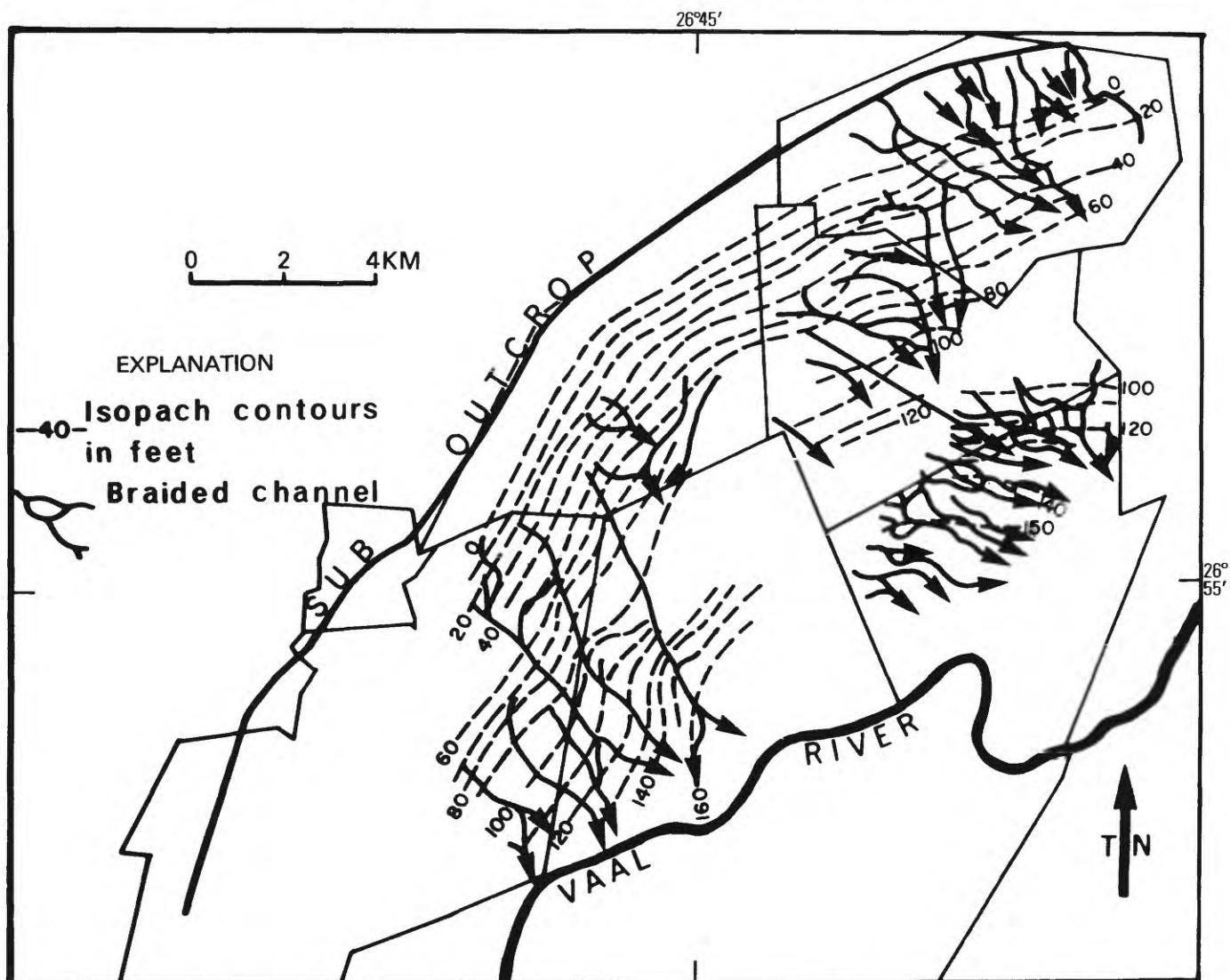


FIGURE 6.-- Isopach plan of the truncated MB5 quartzite, which underlies the Vaal Reef unconformably. The strike of isopachs reflects a synclinal axis. Final erosional etches on the paleosurface are evident as a braided channel pattern.

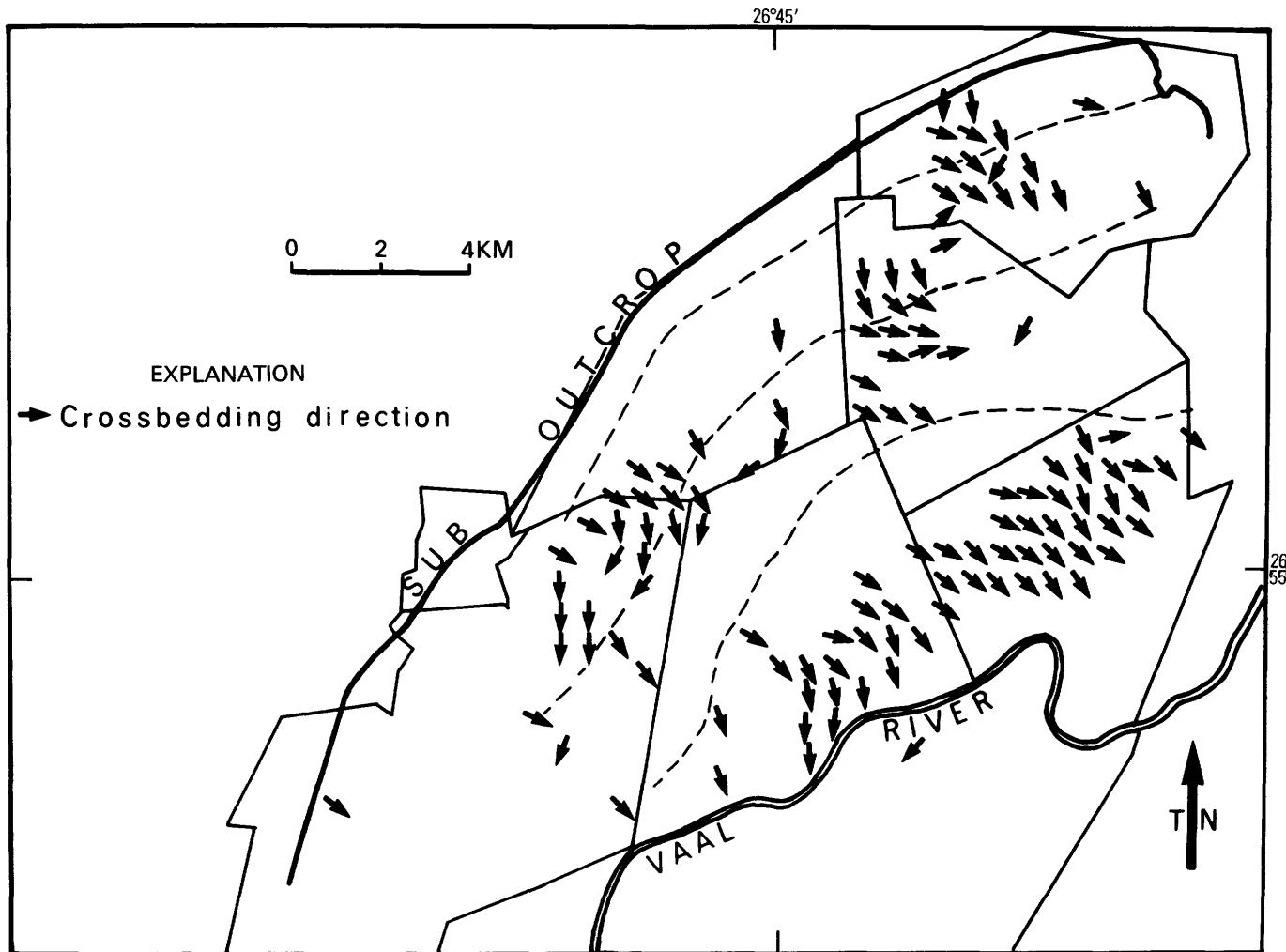


FIGURE 7.—Moving-average vectors measured from trough crossbedded Vaal Reef.

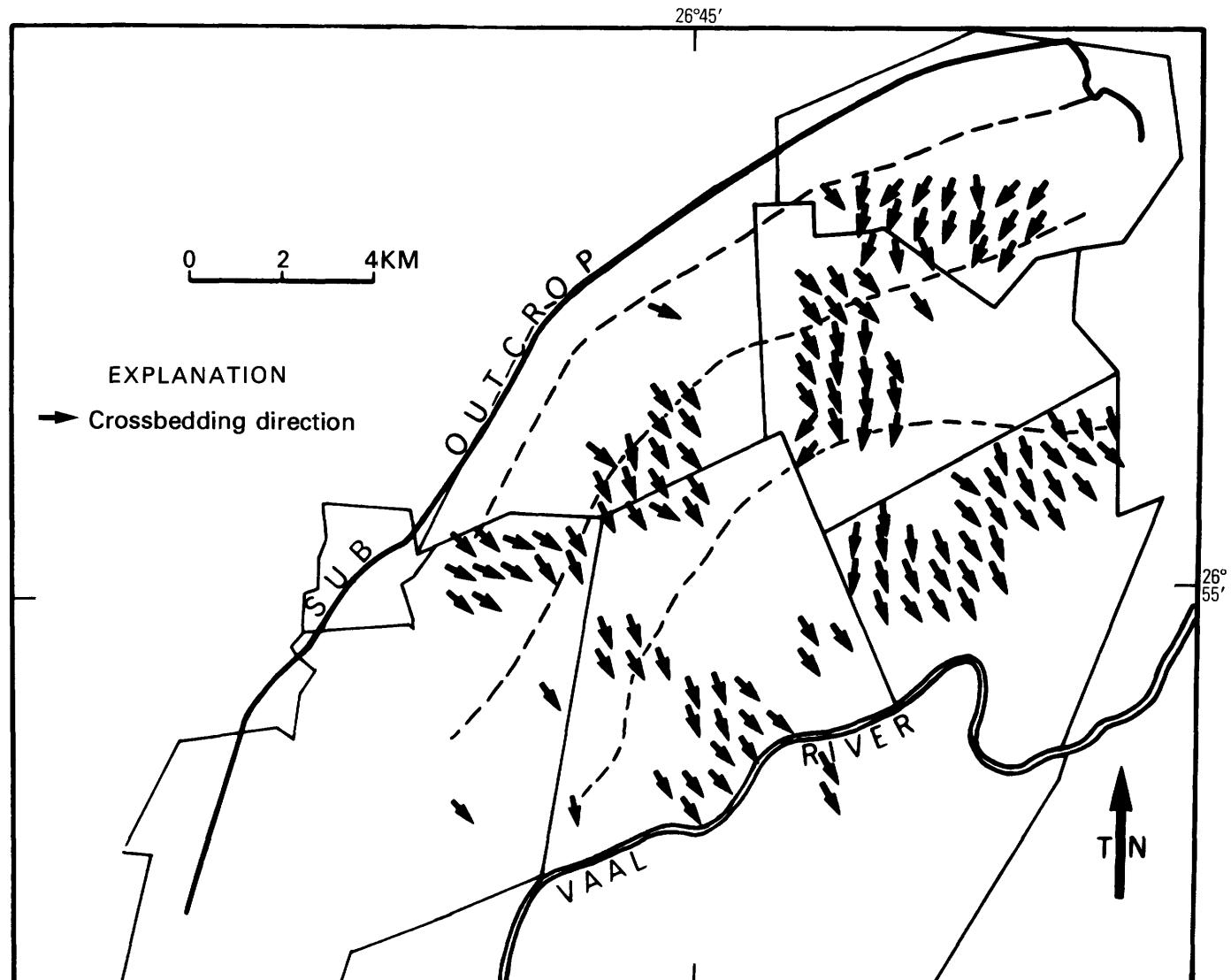


FIGURE 8.-- Moving-average vectors measured from trough crossbedded quartzite beneath the Vaal Reef.

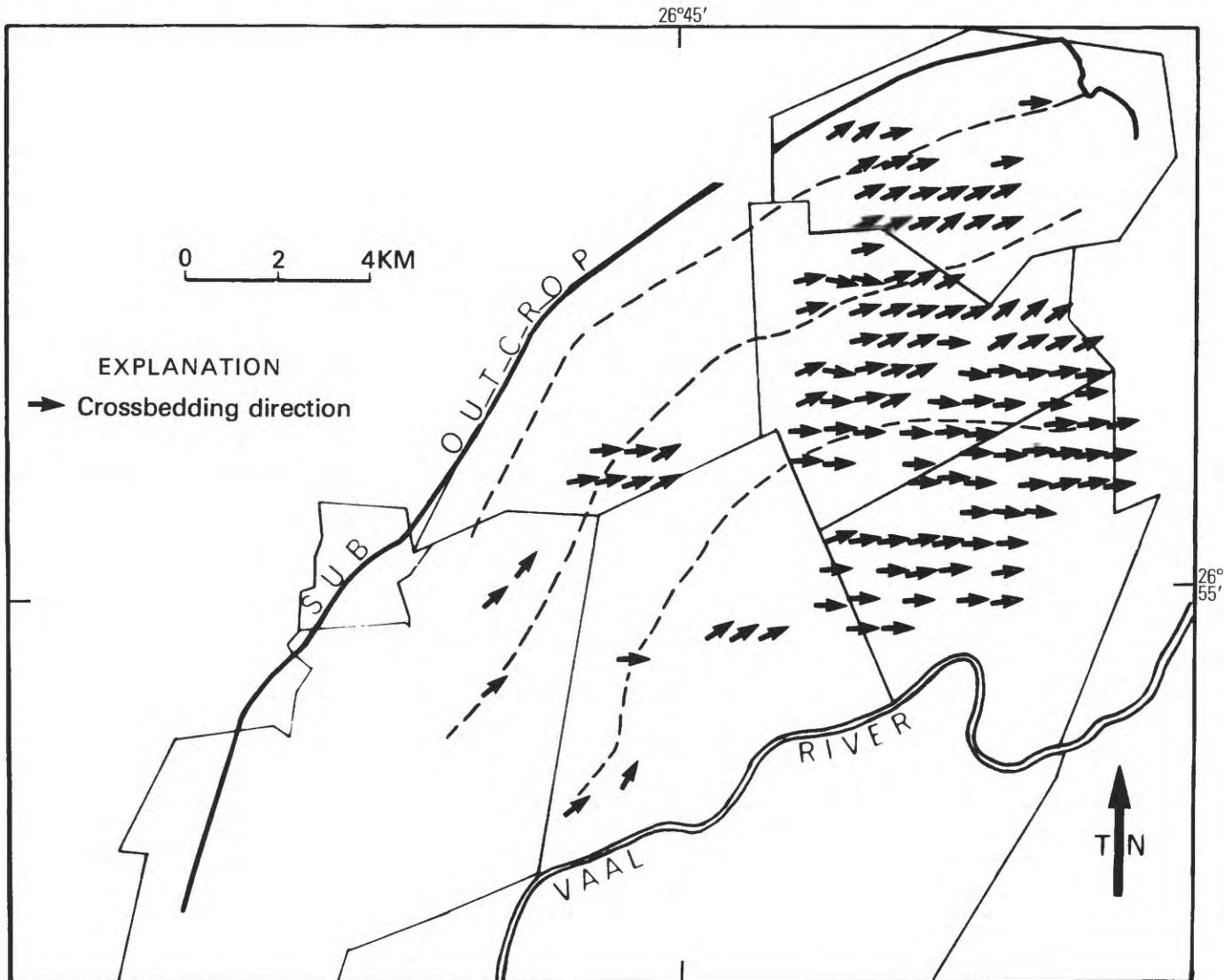


FIGURE 9.—Moving-average vectors measured from the axes of trough crossbedded units in argillaceous quartzite overlying the Vaal Reef.

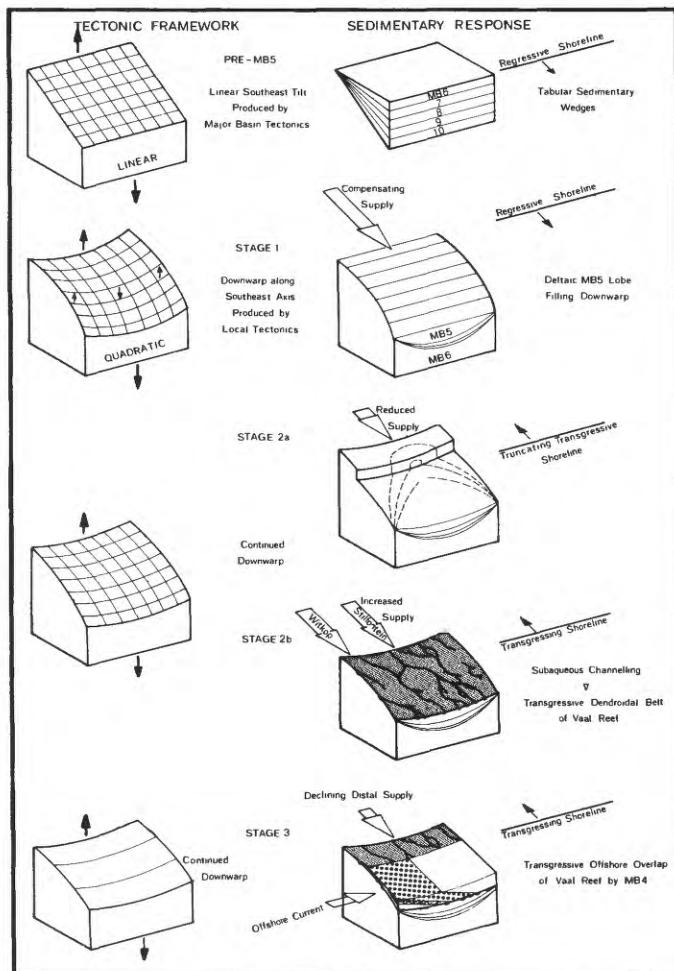


FIGURE 10.—A schematic model of the sequence of events that resulted in deposition and preservation of the Vaal Reef.

proaches ideal limits and is attributed to good stratigraphic control of the data, the small size of the area, and the excellent exposures.

#### SPATIAL DISTRIBUTION OF DETRITAL HEAVY MINERALS

The scour-and-fill mechanism that transported placer sediment effectively concentrated detrital heavy minerals on the toes of foresets and the trough-shaped bases of sets (fig. 11). When the rate of migration of these sets exceeded accumulation, the detrital heavy minerals became concentrated at the base of the bed in a heterogeneous arrangement. This is well illustrated by figure 11, in which the barely preserved remnants of many trough units are marked by dark heavy-mineral concentrates.

Rates of accumulation and migration fluctuate so that a sequence of predominant migration followed by accumulation and then migration again produces a basal

concentration in the reef unit and an upper layer where heavy minerals and small pebbles are concentrated (fig. 12). Although the reef unit is stratigraphically continuous from one position to another, the internal structures are not, because of their curved geometry and the associated heterogeneous distribution of detrital heavy minerals. The total concentration of detritals is related to the amount of migration and is reflected by the number of sets per unit of accumulation.

#### CONCLUSION

As a result of the heterogeneous spatial distribution of heavy minerals, the gold and uranium contents from ad-

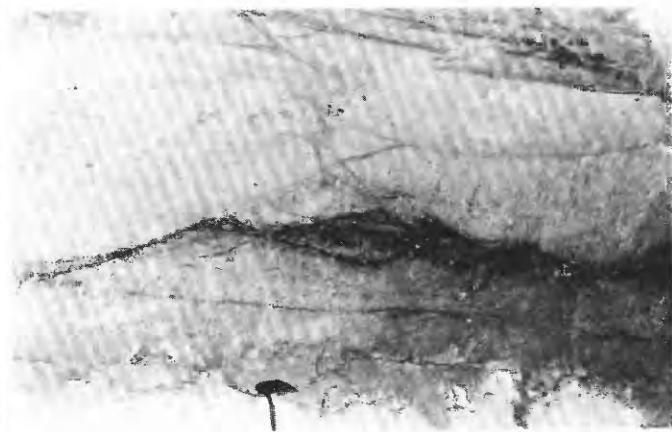


FIGURE 11.—A transverse view of trough crossbedded distal Steyn Reef in which the barely preserved remnants of many trough units are marked by dark heavy-mineral concentrates at the base of the reef unit. The overlying trough crossbedded sets that are better preserved accumulated more rapidly with consequently less reworking and lower concentration of detrital heavy minerals. Free State Saaiplaas, Orange Free State Goldfield.



FIGURE 12.—Distal Steyn Reef, in which is illustrated a sequence of predominant migration followed by accumulation and then migration again, which has produced a basal concentrate in the reef unit and an upper layer where heavy minerals and small pebbles are concentrated.

jacent samples display a standard deviation of as much as 300 percent, and a single sample is seldom representative. This variance of mineral content is a result of the crossbedded geometry of distal placer sediment and may be thought of as a first order of variance.

A second order of mineral concentration variance, which is displayed by stretches of different concentration ranges, is attributed to changes of hydraulic energy level and subtle paleosurface topography. These stretches, in the case of the Vaal Reef, are oriented parallel to the braided channel pattern evident on the Vaal Reef paleosurface.

A third order of mineral concentration variance occurs on an areal scale and is apparently related to the overall geographic distribution of the deposit.

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#### DISCUSSION FOLLOWING PAPERS BY DRS. VON BACKSTROM AND MINTER

*Bourret*: May I first of all compliment you on an outstanding paper. I've been in most of the mines you mentioned and I appreciate particularly what you have to say, and I think you've done a tremendous job.  
*Myers*: Larry [Minter], could you explain a little bit more about the difference between the pi crossbedding and the trough crossbedding?

*Minter*: It's the same thing.

*Myers*: All right.

*Minter*: It's just that John Allen has classified cross-bedding into 15 varieties, the reason being that there is a different mechanism for each variety. This is very important and particularly in this case because one needs a reworking process to create heavy-mineral concentrates and it's interesting to see that one has this process in the reef. It has never been emphasized before, but it occurs in all the Witwatersrand reefs. It also occurs in the Blind River deposits.

*Charles Miles*, *Protocol Corp., Denver, Colo.*: Dr. Minter, earlier we had a source of these gravels or placers sort of boxing the compass, and you have one in your first

paper that had to do with a horst shedding material on both sides. Do you attribute the gravels to localized sources like that?

*Minter*: In those Ventersdorp deposits in which we have alluvial fans, which I used in the example for the left part of the conceptual model, I think that the Witwatersrand has been reworked and that all the material in the Ventersdorp Contact Reef in those alluvial fans came from the horsts.

*Miles*: So that you had a sort of tectonic center.

*Minter*: That's a tectonic center and is applicable to the younger part of the Witwatersrand, in fact the Ventersdorp. We did not have that type of source, I think, in the older Witwatersrand.

*Miles*: That is, the horst contained gravels which on exposure were reworked and deposited in the Ventersdorp?

*Minter*: That's right.

*Miles*: Thank you.

*Minter*: Just one other reply to that question. I think Larry Schloss would call these younger deposits yoked deposits, whereas the others are intracratonic deposits in a very much gentler depression, as it were.

*Larry Milliken*, *Public Service, Denver, Colo.*: This perhaps should be directed to Dr. von Backström. You mentioned the presence of several refractory minerals that make up a fairly significant percentage of the heavy-mineral suite. Has any effort been made to extract uranium from these, or is all your uranium taken from uraninite?

*von Backström*: I wouldn't be able to answer this directly myself, but I wonder if Dr. Whiteside, who is here, could answer it. I know there was some uranium production from the Dominion Reef mine. Dr. Whiteside, do you know what they did in connection with any refractory minerals?

*Whiteside*: As far as I know, all that happened was that it messed up the mill circuit at the Western Reefs Mine and that we eventually gave up mining the Dominion Reef for that reason. Material from the Vaal Reef and material from the Dominion Reefs were antipathetic, rather.

*Milliken*: Thank you. Is any work being done at this time on the milling treatment of those refractories?

*Whiteside*: They are definitely working on the Dominion Reef, and they will put up a separate plant for it, so that would take care of the refractories. I think according to Dr. [S.H.U.] Bowie, the uraninite is not a true uraninite and he will concede that the uraninite in the Dominion Reef is detrital only because it is thorium-bearing uraninite.



# Primary Dispersal Patterns of Uraninite in the Proterozoic Vaal Reef Placer Deposit, Witwatersrand, South Africa

*By* W. E. L. MINTER

GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN  
QUARTZ-PEBBLE CONGLOMERATES

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## GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN QUARTZ-PEBBLE CONGLOMERATES

# PRIMARY DISPERSAL PATTERNS OF URANINITE IN THE PROTEROZOIC VAAL REEF PLACER DEPOSIT, WITWATERSRAND, SOUTH AFRICA

By W. E. L. MINTER <sup>1</sup>

### ABSTRACT

The Vaal Reef is a strata-bound uranium orebody. Uraninite, together with nodular pyrite and gold, is a detrital heavy-mineral component of the sediment. Consequently, the areal distribution patterns of uranium content match the braided, southeasterly dispersal pattern evident in the Vaal Reef. Deeper channelways contain more uraninite.

### INTRODUCTION

Well, I've really primed you, and now I would like to relate the uranium distribution in the Vaal Reef to the sedimentary environment that I have painted.

The Vaal Reef is a distal Proterozoic placer deposit that is being mined for its gold, uranium, and pyrite content in the Klerksdorp Goldfield (fig. 1), on seven mining-lease areas (fig. 2). Stratigraphically situated near the top of the Main Bird Series of the Upper Witwatersrand System (fig. 3), the Vaal Reef was a pebbly sand sheet, 1 to 150 cm thick, that covered approximately 260 km<sup>2</sup> (100 mi<sup>2</sup>). It is the sedimentary product of anastomosing braided river channels in a shallow fluvial environment.

The ancient southeasterly dispersal pattern indicated by primary crossbedding structures within the sand body is substantiated by meandering braided channel forms, evident at the base of the sand body, that eroded into underlying sediments (fig. 4).

Nodular pyrite (30,000 ppm (parts per million)), uraninite (500 ppm), and gold (15 ppm) are detrital heavy-mineral components of the Vaal Reef that were concentrated at the base of the sand body and on internal sedimentary partings. Fossil hydrocarbon, ranging from scattered fine-grained specks to a seam 5 cms thick, is generally evident on the basal contact and less commonly on sedimentary partings.

### URANIUM DISTRIBUTION

Uraninite is distributed vertically throughout the Vaal Reef, 20 percent of the total content being concentrated

near the base of the reef. Similar concentrations may also occur on internal sedimentary partings.

The positively skewed log-normal frequency distribution of uranium contained in total Vaal Reef samples was reduced by combining large numbers of samples with a moving-average technique. More than 900,000 samples from the area were used to produce a meaningful contour plan (fig. 5). The southeast channel fabric is well defined by the uranium distribution.

A contour plan prepared in a similar way using total-thickness data of the Vaal Reef (fig. 6) describes the external geometry of the sedimentary sheet. The southeasterly trend and braided nature of the reef are well defined. It is evident from a comparison of the uranium and the thickness contour plans that a sedimentary relationship exists - a thicker reef contains more uranium.

A stratigraphically higher pebbly layer known as the Zandpan Marker onlaps the Vaal Reef (fig. 7) from the west, and its uranium content enhances that of the Vaal Reef in the area of convergence (fig. 8).

A moving-average contour plan (fig. 9) of the basal hydrocarbon thickness, based on 499 samples, describes an area of thick hydrocarbon along the western side of the deposit that is a mile wide and extends for at least four miles down the paleoslope. This is interpreted as having been an area favourable for the growth and preservation of organic plant material (Minter, 1972). The uranium content in areas where thick hydrocarbon seams are prevalent is of the same order as in areas to the east where hydrocarbon is conspicuously sparse. The ratio between uranium and gold, calculated from the moving-average data used for figure 5 ranged from 7 to 19 over most of the area (fig. 10), and the distribution pattern does not appear to have significant relation to the carbon-seam isopach (fig. 9). These data do not support previous concepts that the carbon seam was produced from liquid hydrocarbon by irradiation from uranium nor that the uranium was precipitated from solution by carbon. Had either effect predominated, the

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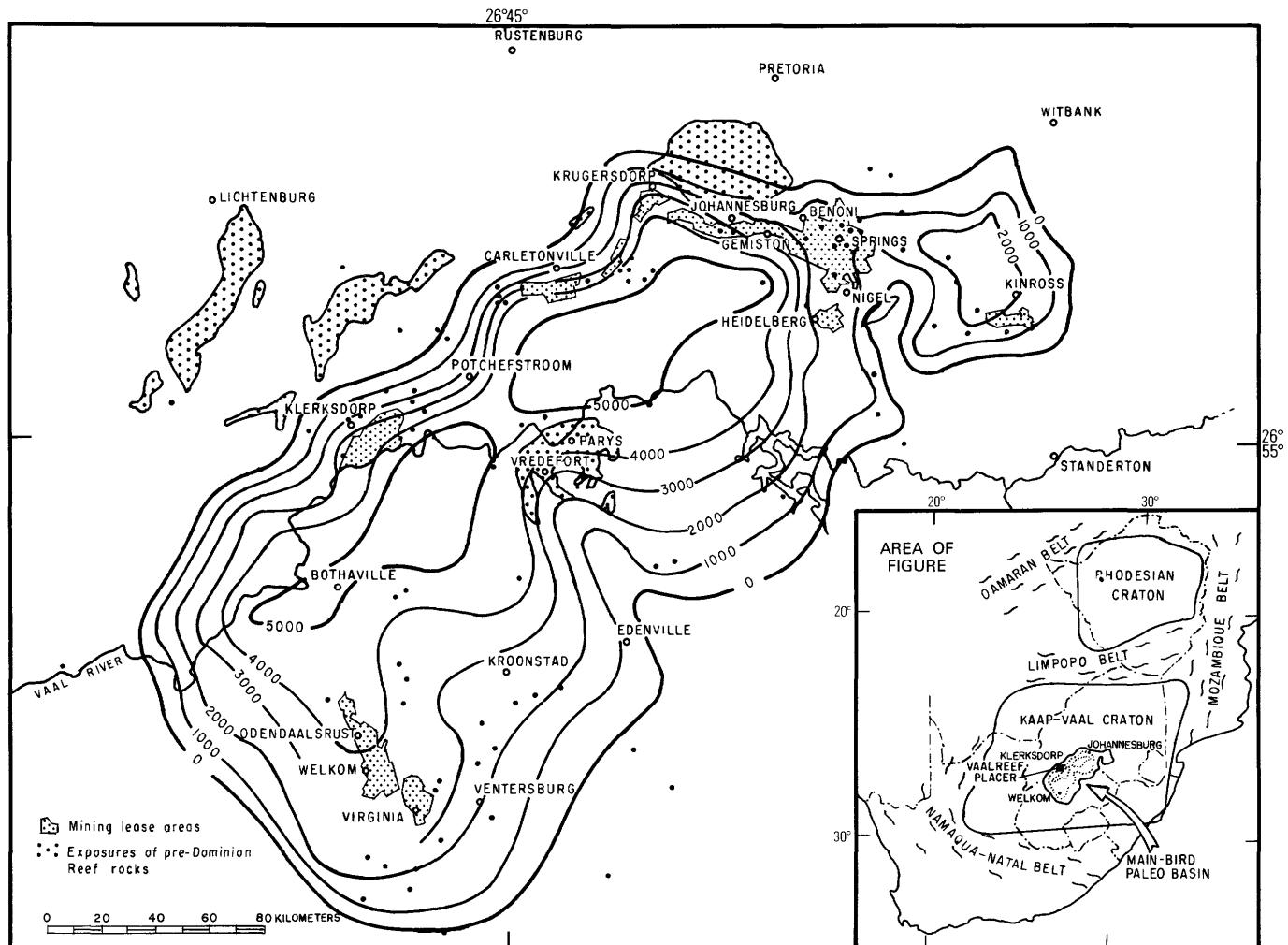


FIGURE 1. – Isopach map of the Main Bird Series (modified from Brock and Pretorius, 1964, pl. 2) showing the relative position of the Vaal Reef in the Witwatersrand basin. Isopach contours in feet, contour interval 1,000 ft.

uranium content would have been preferentially greater in thick carbon-seam areas.

### CONCLUSIONS

Uraninite is a detrital component of the Vaal Reef placer. The total uranium content is related to the thickness of the Vaal Reef, and the areal distribution patterns of uranium content and thickness are sympathetic.

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### DISCUSSION

[Hallbauer?]: [An incompletely recorded statement was made to the effect that uranium occurs in two forms in the Vaal Reef, as detrital uraninite and in the organic matter.]

Minter: That's another story which is coming later, but certainly there are at least three positions where one can find radioactive minerals in the reef. As I said, only 20 percent is on that bottom contact and a lot of that 20 percent is probably in the carbon, some of it is perhaps in the structure itself, some as detrital grains

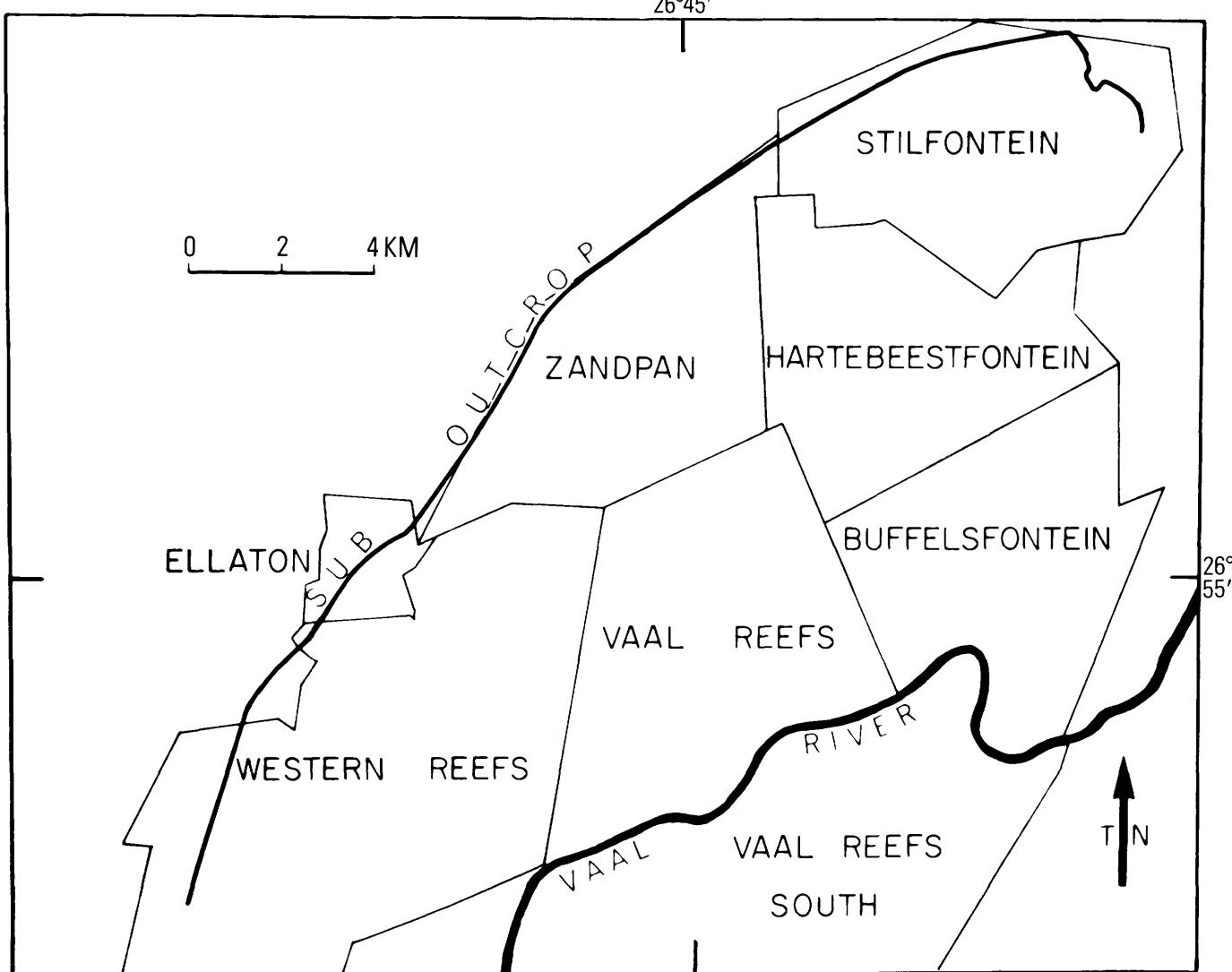


FIGURE 2.—Location plan of the seven mining lease areas where the Vaal Reef is being exploited.

within the carbon. Certainly I am implying that the 80 percent which is distributed through the reef and which isn't associated with carbon is presumably detrital.

*Bourret:* In all your sections you showed the mineralization of uranium pretty much terminating as you proceed to the southeast corner of the slides. Is this a structural feature or property lines?

*Minter:* Property lines. We've reached a depth of something like 7,000 feet and we are still going. But I can bring you up to date in a few years time. There's a very interesting point associated with the mineralization trends shown and that concerns application of sedimentology to mining. We have very broad trends and we can project them downdip, which also happens

to be down the paleodip, and consequently we can perhaps exercise grade control, predict grades, etc. With uranium it is quite easy because the concentration is relatively high. With gold it's more difficult, but that's certainly one of the economic applications we use.

*Bourret:* This does not mean, then, that this is the end of the economic mining depth?

*Minter:* No. That big black line at the bottom, which represents the Vaal River, separates Vaal Reefs from Vaal Reefs South, which is an extensive new mine lease area just to the south that presumably has a life of 20 years. We keep these maps up to date on a computer program; data goes in monthly and we use it.

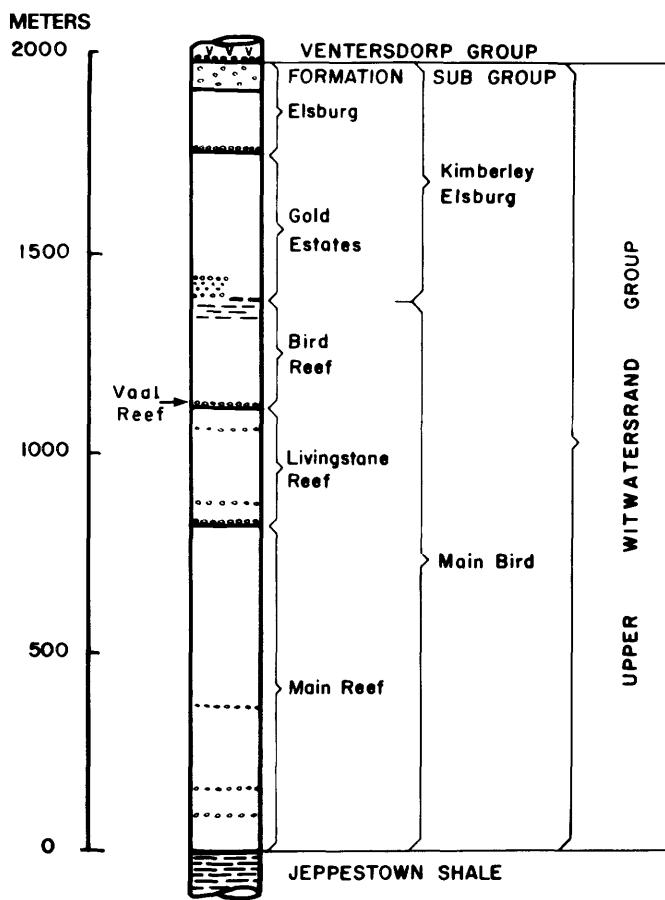


FIGURE 3. - Stratigraphic column of Upper Witwatersrand Group  
in the Klerksdorp area.

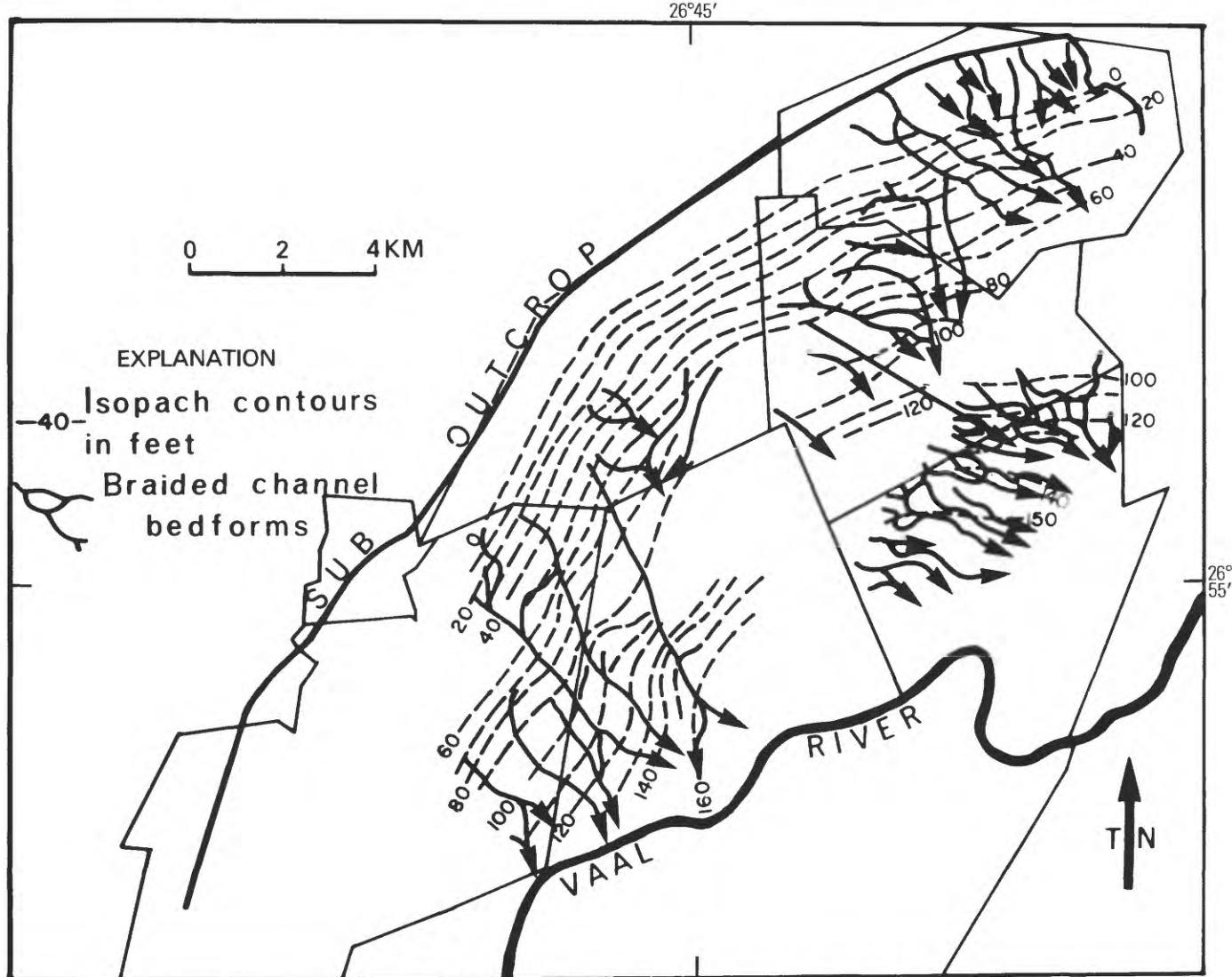


FIGURE 4. – Isopach plan of the MB5 footwall sediments that the Vaal Reef truncated. The braided channel pattern that is superimposed reflects the final erosional etch on the paleosurface and the Vaal Reef distributary system.

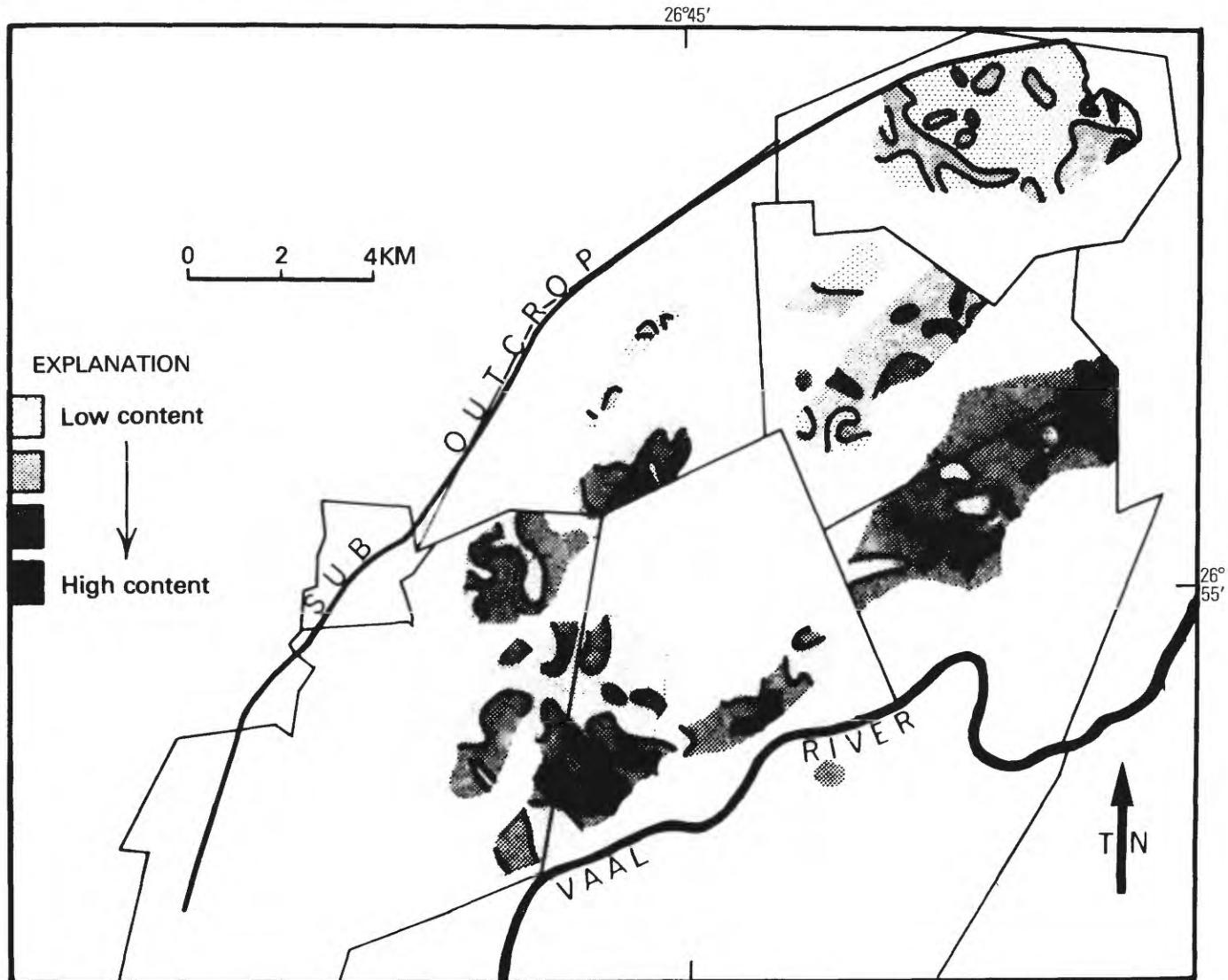


FIGURE 5. - Moving-average plan of uranium content prepared from more than 900,000 samples of Vaal Reef.

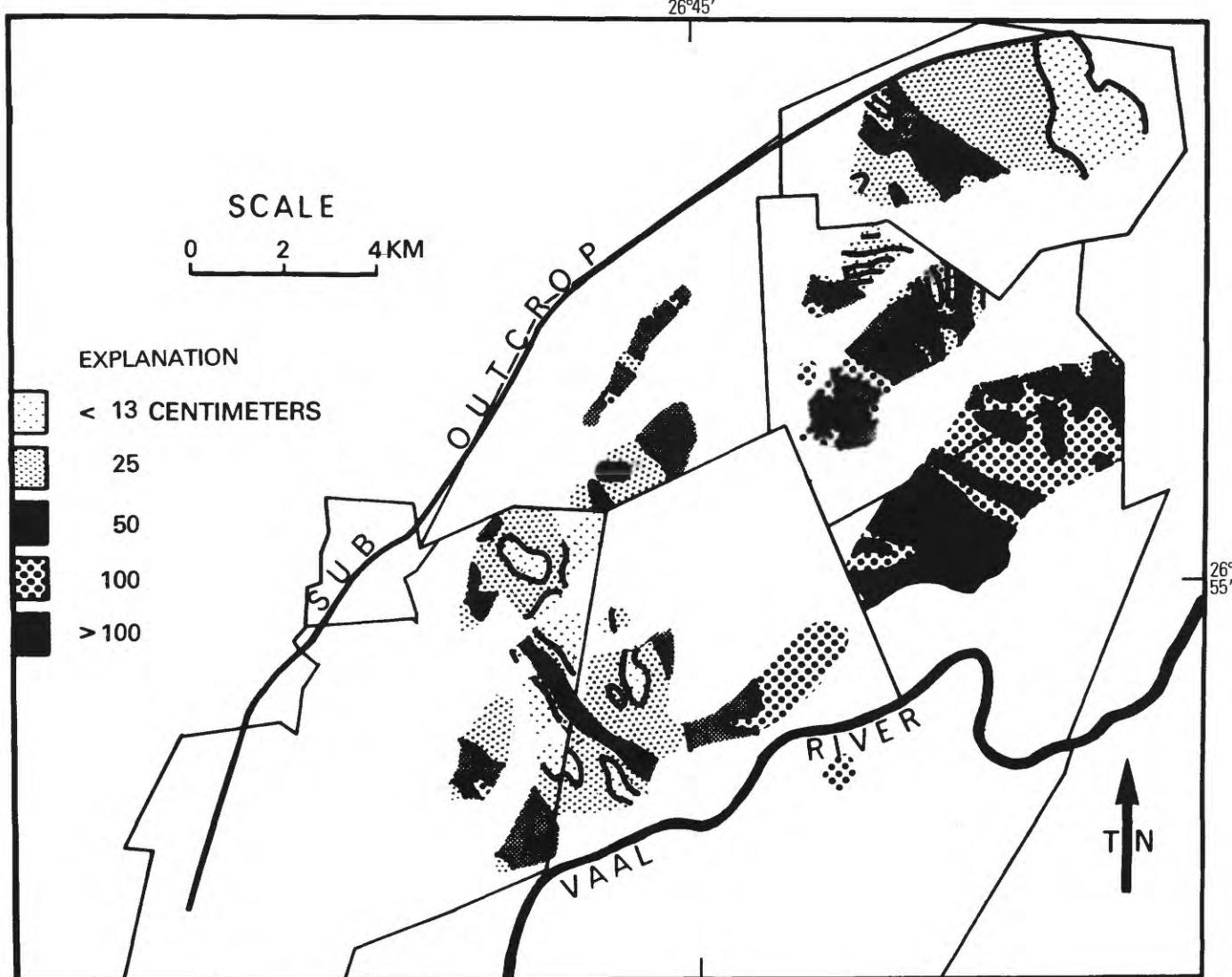


FIGURE 6.—Moving-average isopach plan of the Vaal Reef prepared from more than 900,000 data points.

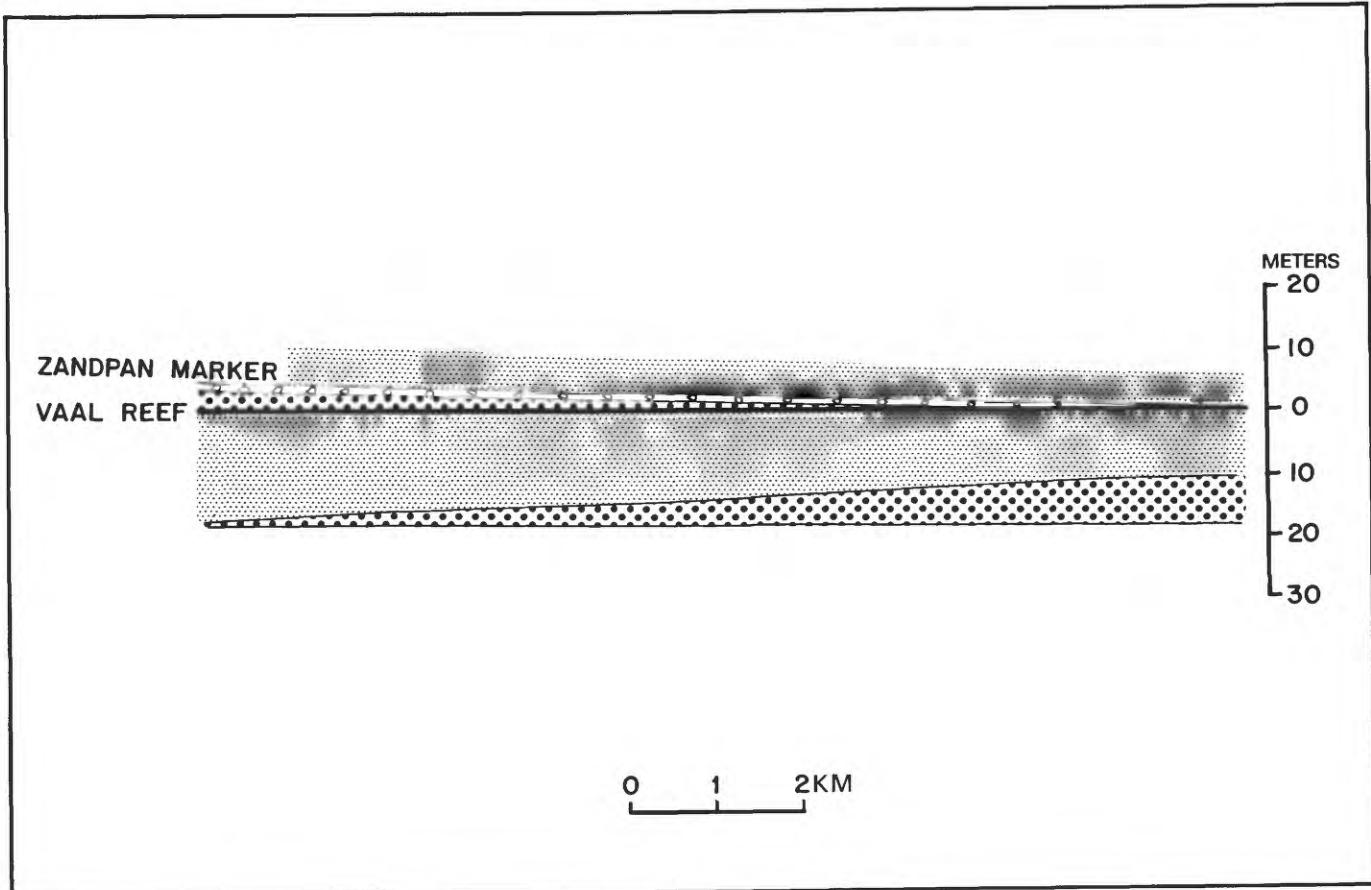


FIGURE 7.—Section along Vaal Reef paleostrike to illustrate the stratigraphic relation between the Zandpan Marker and the Vaal Reef.

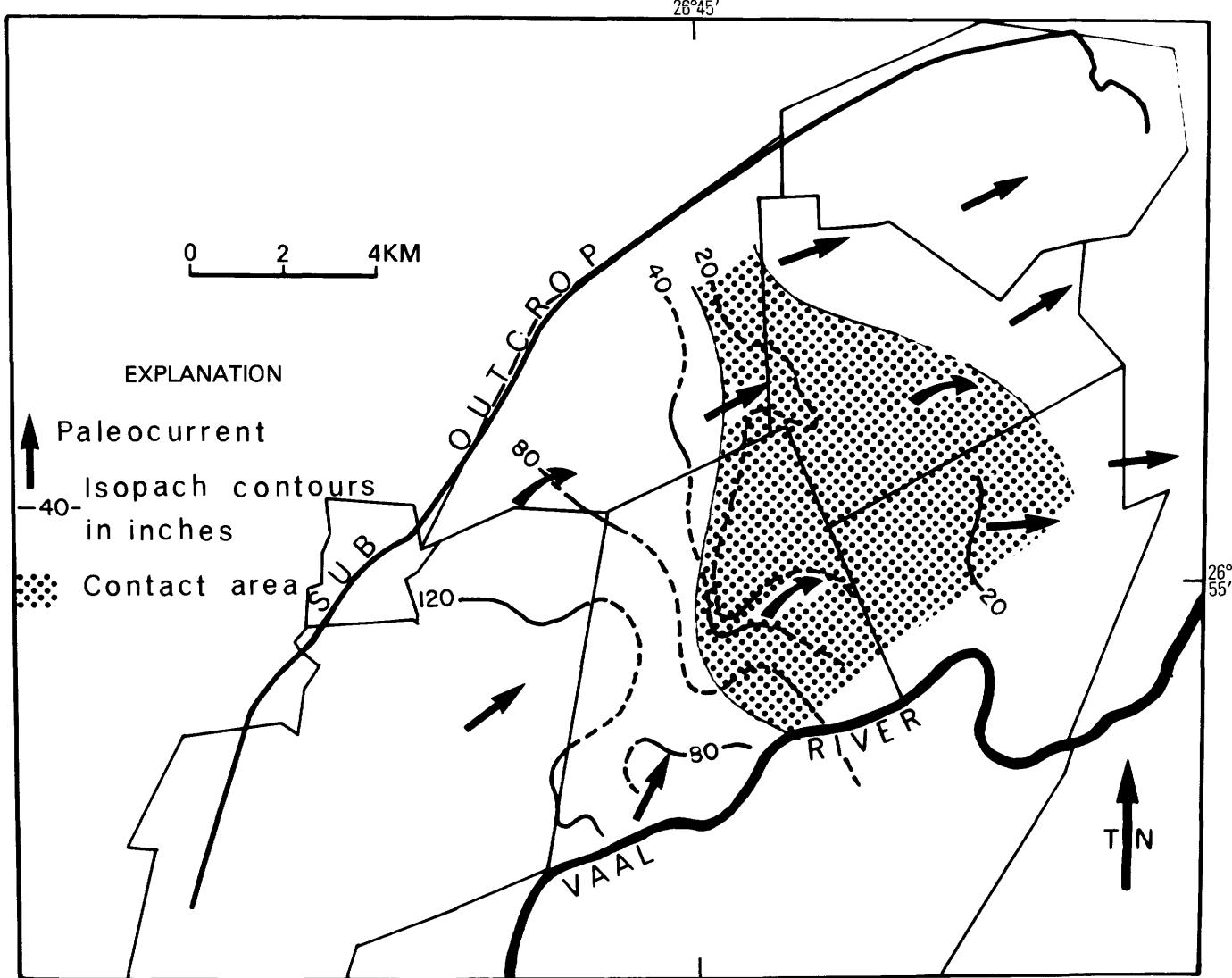


FIGURE 8. - Isopach and paleocurrent plan of the sediment between the Vaal Reef and the Zandpan Marker, illustrating the Zandpan Marker's onlap.

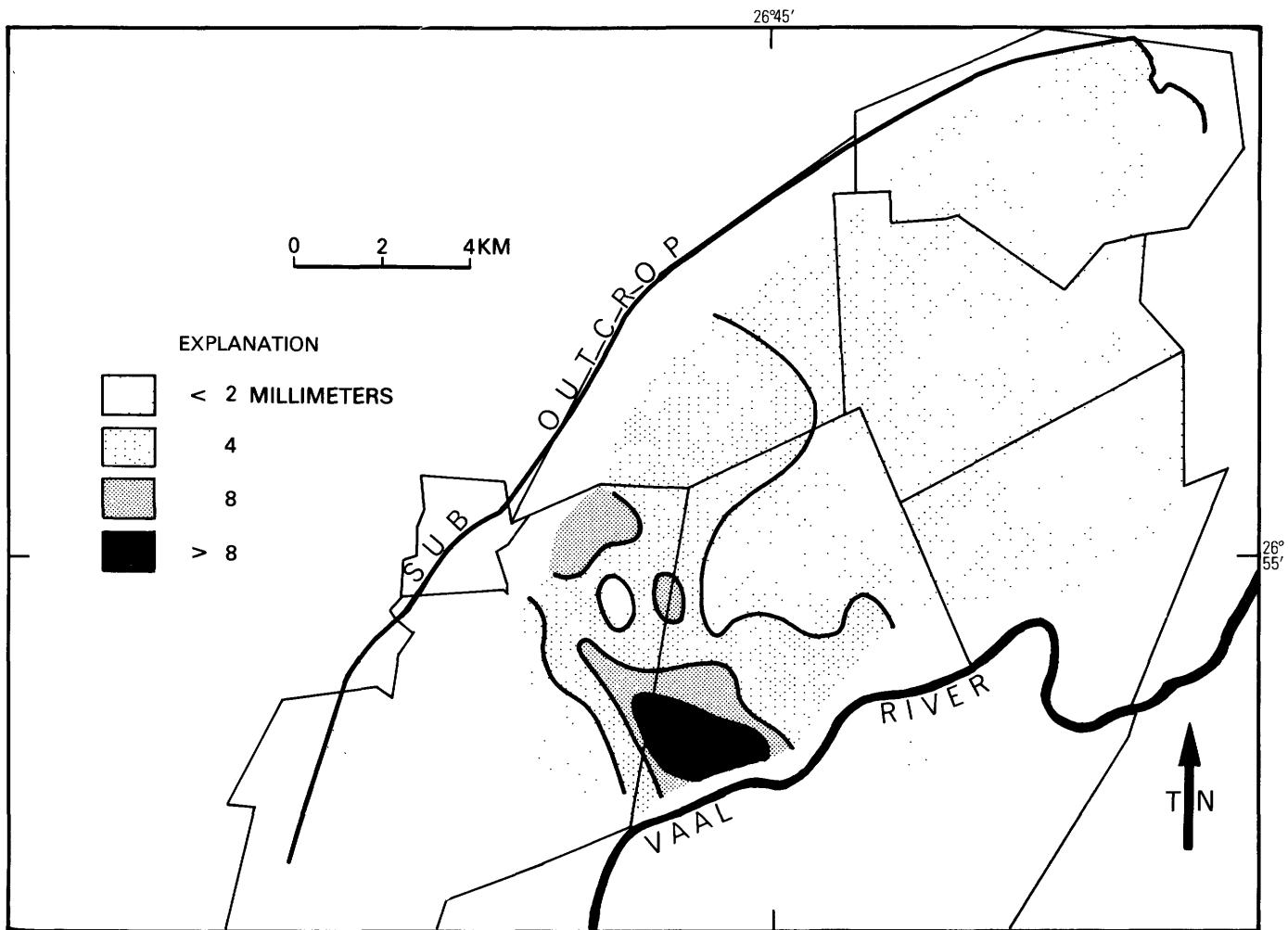


FIGURE 9.—Moving-average isopach plan of the basal carbon-seam measured at 499 locations.

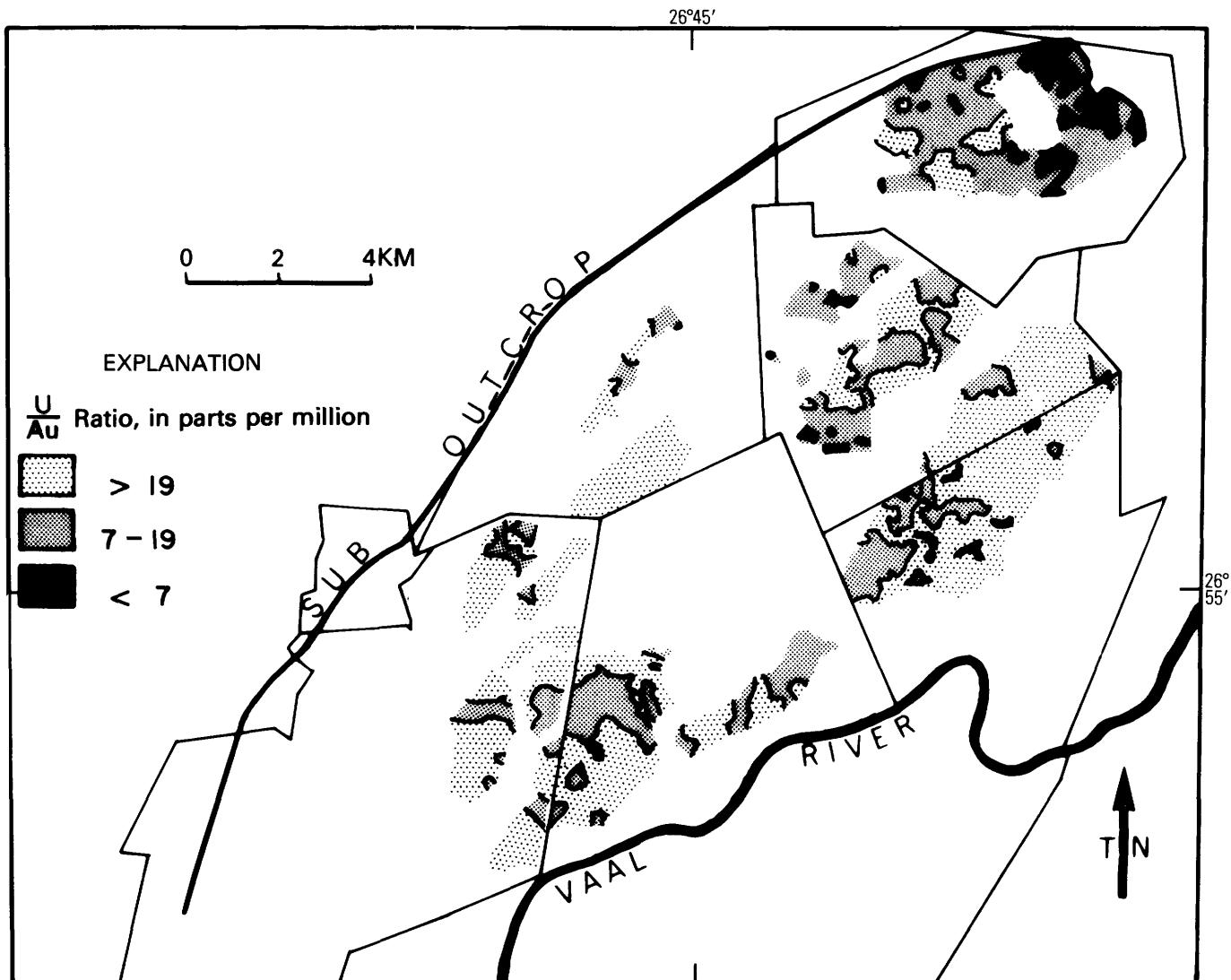


FIGURE 10. - Moving-average uranium-gold ratio plan calculated from moving averages of more than 900,000 samples of the Vaal Reef.



# Examples of Possible Movement of Gold in Solution in the Witwatersrand, Ventersdorp, and Transvaal Systems

*By* H. C. M. WHITESIDE

GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN  
QUARTZ-PEBBLE CONGLOMERATES

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## GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN QUARTZ-PEBBLE CONGLOMERATES

### EXAMPLES OF POSSIBLE MOVEMENT OF GOLD IN SOLUTION IN THE WITWATERSRAND, VENTERSDORP, AND TRANSVAAL SYSTEMS

By H. C. M. WHITESIDE<sup>1</sup>

#### ABSTRACT

Many South African geologists consider that gold especially, and probably uraninite, as well as other heavy minerals such as zircon, chromite, and diamond, are detrital in the Precambrian quartz-pebble conglomerates of South Africa. It is suggested here that solution of gold and its redeposition elsewhere may have played a part in its enrichment or impoverishment. Two examples of possible solution of gold are cited, and the lack of modern placers associated with auriferous Witwatersrand is briefly discussed.

#### INTRODUCTION

It is generally considered that gold, and probably uranium as well, has been concentrated by hydraulic processes in the conglomerates of the Dominion Reef, Witwatersrand, Ventersdorp, and Transvaal Systems. (It is proposed to retain the old divisions into systems and series as these are more familiar than the terms supergroup, group, and the like). Practically all South African geologists consider that gold and uranium, as well as other heavy minerals such as zircon and chromite, are detrital and deposited along with the pebbles and sand grains. The gold and uranium deposits of the Witwatersrand type would therefore be syngenetic.

This short paper is an attempt to show that South African geologists may have reacted too strongly against the hydrothermal theory, which postulated that gold was introduced into the conglomerates by mineralized hot waters emanating from a deep-seated igneous source. Using "hydrothermal" in its widest sense of "heated waters," it is suggested that warm ground waters may have taken gold and uranium into solution and modified their concentration and distribution. This does not mean that gold and uranium are not predominantly detrital, merely that there may have been movement of these metals in solution in the period between their deposition and the consolidation of the sediments.

Two examples are described that are suggestive of solution of gold and, in one of these cases, redeposition in a new environment. The lack of modern placers associated with erosion of the auriferous Precambrian conglomerates in and around the Witwatersrand basin is also briefly discussed.

#### HEAVY METALS IN THE BLACK REEF, EAST RAND

The Black Reef, a dark conglomerate whose colour is attributed to disseminated particles of carbon, is developed in places at the base of the Black Reef Series, the lowest member of the Transvaal System. It contains appreciable gold but not much uranium and has been mined in several localities encompassing the northern part of the Witwatersrand basin, usually close to or overlying the edge of that basin. The majority of the orebodies are restricted in size, and mining was on a small scale compared with those mines that worked the Witwatersrand or Ventersdorp conglomerates. From descriptions in the literature it would appear that most of the Black Reef deposits were infillings of scour channels, composed of coarse, fairly thick conglomerates extending for only a few hundred metres in width and 200–300 m down-dip, separated by barren areas in which the conglomerates are thin or even absent.

It has been stated by several geologists that the Black Reef has derived its mineral content through the erosion of the underlying rocks in the vicinity and subsequent concentration of the heavy minerals by sedimentary processes. This can be said to have been adequately demonstrated in some instances, particularly regarding the two largest known deposits on Government Gold Mining Areas and Randfontein Estates, where auriferous Witwatersrand conglomerates form the immediate footwall or are very close by. A rough analysis shows that the

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footwall of the Black Reef where it has been mined is as follows, in percent:

Ventersdorp:	
Lavas	47
Sediments and tuffs	13
Witwatersrand:	
Upper	20
Lower	7
Basement, essentially granite	13

This tabulation seems to show that auriferous Black Reef may be deposited some distance away from the original source of gold.

A peculiarity of the Black Reef is that it contains osmiridium in greater than normal concentration, compared with Witwatersrand conglomerates, in at least three mines. J. A. Papenfus (1964) has demonstrated that at Government Gold Mining Areas in the East Rand there is a relation between quantity of osmiridium recovered and the proximity of the Black Reef, which was being mined, to a particular conglomerate (U.K.9 c) in the Kimberley Series. Beds of the Kimberley Series constitute the immediate footwall of the Black Reef, and a large channel, filled with Black Reef conglomerates and quartzites, has been cut into the Kimberley for a distance of some 5 km.

It is reasonable to suppose that, in this distance, the osmiridium in the Black Reef was derived from the Kimberley Series, but it is difficult to account for the osmiridium in areas where the footwall is granite or Ventersdorp lavas and sediments. One explanation is that heavy metals such as gold and osmiridium can be transported in their metallic state farther than is at present thought possible, although the fact that concentrations of gold are found only near the edge of the Black Reef basin appears to contradict this. A second explanation is that there is a local but concealed source of these metals.

More than 25,000,000 metric tons of Black Reef ore was mined on Government Gold Mining Areas, in addition to very many millions of metric tons from the Main Reef Leader, the main ore horizon, and a fair tonnage from conglomerates in the Kimberley Series. Four metals and minerals were recovered—gold, silver, osmiridium, and pyrite—and statistics were kept of all recoveries. The following differences, compared with the Main Reef Leader and the Black Reef, emerge. Although it is not possible to quantify the following ratios because ore was derived from three sources, it is believed that the subsequent statements are correct because there were periods when either the Main Reef Leader or the Black Reef contributed more than 80 percent of all ore milled. It is not possible to assess the Kimberley Reef because it never contributed a high percentage to the mix, according to the available information.

1. The gold-to-osmiridium ratio is lower in the Black Reef than in the Main Reef Leader. There is not a great difference between the specific gravities of these two minerals, depending on their composition, so the ratio should not change appreciably.
2. The gold-to-silver ratio is lower in the Black Reef than in the Main Reef Leader. It is generally accepted that the silver content of detrital gold is less than that of vein gold, some of the silver being dissolved during transportation. If the Black Reef gold has been derived from older conglomerates the opposite should be the case—that is, the gold-to-silver ratio should be higher.
3. The pyrite content of the Black Reef is considerably higher than that of the Main Reef Leader. "Buck-shot" pyrite, rounded grains varying in size from 0.5 mm to more than 2 cm, is the form in which it generally occurs, and these grains are considered to be replacements of detrital ferruginous material or chert pebbles.

Summing up these facts, it is suggested that:

1. Normal sedimentary processes concentrated pebbles and heavy minerals such as gold, osmiridium, zircon, and chromite from previously existing sources such as the Witwatersrand succession, the Older Granite, and perhaps the Ventersdorp sediments.
2. Sulphur-bearing waters were introduced after deposition but before consolidation. The sulphur could be derived from organic matter or residual fluids from the Ventersdorp volcanicity. Iron-bearing minerals such as magnetite and hematite, as well as small banded ironstone pebbles, would be altered to pyrite nodules.
3. If the gold in the Black Reef contains more silver than normal it suggests that silver has been added or that gold has been leached. During deposition by sedimentary processes gold and osmiridium should have retained a ratio similar to that of the source rock, in this case, Witwatersrand conglomerates and quartzites. This is not so because the gold:osmiridium ratio is lower in the Black Reef than in nearly all Witwatersrand conglomerates. This must mean that either additional osmiridium was introduced or gold was reduced in quantity. These conclusions suggest that gold was leached penecontemporaneously with the deposition of the Black Reef.

#### GOLD IN AN AMYGDALAE IN VENTERSDORP LAVA

This note is merely to record that one grain of gold was found in an amygdale in lava a few inches, no more than 15 cm, above the contact of lava with the top of the

Ventersdorp Contact Reef. This occurrence was in core from a borehole drilled to the west of Western Deep Levels Mine for the purpose of proving continuity of the Ventersdorp Contact Reef.

As far as is known, this is the only recorded instance of visible gold in Ventersdorp lava. Gold in small quantities has been found in cores that have intersected sediments in the Ventersdorp System, but the gold is not visible and has probably been brought in as detrital grains. Occasionally, amygdales have been noted that contain pyrrhotite and chalcopyrite.

It is considered that the gold in the amygdale was taken into solution by heated waters from the Ventersdorp lavas circulating through the Ventersdorp Contact Reef, which presupposes that the conglomerate was not fully consolidated at the time.

This is no proof that gold has been carried in solution in large quantities or over great distances, but the observations cited above may support the modified placer hypothesis, which contends that the gold in the Witwatersrand conglomerates is detrital but has been modified by hot fluids and now often is indistinguishable from hydrothermal gold.

#### RECENT ALLUVIALS ASSOCIATED WITH THE WITWATERSRAND BASIN

It has often been noted by geologists working on the Witwatersrand that there is no large-scale development of auriferous alluvials related to present-day topography.

There are numerous examples of erosion of auriferous beds and redeposition of the various constituents, such as pebbles and heavy minerals, throughout the succession from the Witwatersrand to the Black Reef Series. The U.K.9 Kimberley Reef has been shown to contain more gold downstream from areas where the middle Kimberley has been truncated. The Ventersdorp Contact Reef clearly derives some of its gold from erosion of underlying auriferous conglomerates of the Upper Witwatersrand. On the West Wits Line, at West Driefontein, and Western Deep Levels Mines the Contact Reef is enriched in the vicinity of the Main, Bird, and Kimberley groups of conglomerates where the latter crop out below it. An example of concentration of heavy metals and minerals in the Black Reef has been described above.

It has been suggested that glaciation during Dwyka time dispersed gold and heavy minerals so that concentration into alluvials was impossible. This may be true for that particular period, but the Karoo System is Permo-Carboniferous in age and there must have been a considerable time interval after the Permo-Carboniferous during which the Karoo was stripped from the Central Witwatersrand area and the Upper

Witwatersrand rocks with their auriferous conglomerates were eroded, yet no gold placers are known to be related to that period of erosion.

On average, the conglomerates carried much more gold at outcrop than in depth, and much of it was native, suggesting secondary enrichment. Could it be that gold went into solution and was dispersed and that this is the reason why no alluvials were formed?

#### SELECTED REFERENCES

At the meeting held in Golden, Colo., in October 1975, several of the geologists who attended expressed the view that there was insufficient description of the general background of the Witwatersrand System and other Precambrian Systems that are hosts to auriferous and uraniferous quartz-pebble conglomerates.

The following papers are thought to cover the geology of the Witwatersrand both in general and, to a certain extent, in detail:

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Winter, H. de la R., 1964b, the geology of the Virginia section of the Orange Free State Goldfields, in Haughton, S. H., ed., The geology of some ore deposits in southern Africa: Geological Society of South Africa, v. 1, p. 507-548.

## DISCUSSION

[As an aside in his talk Dr. Whiteside gave the following approximate dates for the Rand: Black Reef, 2,000 m.y.(million years); Ventersdorp, 2,300 m.y.; Witwatersrand, 2,500 m.y.; Dominion Reef, 2,800 m.y.]

*B. Nagy:* When you talk about the transport of gold, do you mean gold in molecular solution, or do you mean gold in colloidal suspension?

*Whiteside:* I cannot answer. I am a field geologist and I don't know.

*B. Nagy:* We are working on the Vaal Reef area and we have considered various kinds of transport, placer gold transport, even solution with hydrogen cyanide, which doesn't seem to work. On the other hand, there are a number of possibilities. For example, hydrothermal gold can interact with various organic compounds. Small gold particles can be carried this way, but such transport is not on a molecular level. Also it is possible that at the site of deposition very small particles of gold can be partially mobilized by the organic compounds present, giving the impression of dissolution. What I am saying is that from our experience I cannot see molecular solution of gold. Despite the difficulties involved I can see, however, transport of gold in colloidal suspension or as very fine placer grains. Did you make the comment that in the Transvaal the carbon is graphite?

*Whiteside:* I said it is carbon, but some people have said it is graphite. I don't think it is.

*B. Nagy:* We published a paper in *Nature* which came out June 26, 1975, in which we described two cyclic ethers from a sample of the Transvaal. Well, for the chemists' sake, we found two ethyls, two propyls, three methyltetrahydrofuran, two propyltetrahydrofuran. In addition, we have found by ionization mass spectrometry at least five thousand different peaks of molecular ions of various compounds: oxygen, hydrogen, nitrogen, and hydrocarbon compounds ranging from very low molecular weight to very high molecular weight. We did not find any graphite whatsoever. We did find, however, a very complex organic chemistry which to us is indicative of a biota that had evolved a paleobiochemistry by Transvaal time.

*Whiteside:* Thank you.

*Simpson:* Would you regard uranium as possibly concentrated in carbon by a similar process to the one you describe for gold?

*Whiteside:* Yes.

*Skinner:* I would like to comment on gold transportation. In a very short, interesting paper a few years ago

in *Economic Geology*, Holgeson and Garrels, discuss transportation of gold and of silica in an ordinary brine; transportation is small but significant. It is not zero. If you have a brine and have gold present, that brine is going to be saturated with gold. It is impossible not to move some of it. If you had any kind of thermal gradients developed in these basins you must have moved some gold around. It seems quite conceivable to me that a perfectly normal, ordinary brine may indeed be the cause of what you describe, and I rather doubt that organic compounds have much to do with it.

*Whiteside:* Thank you.

*Feather:* Did you discover there were any secondary minerals associated with that gold inclusion within the amygdale? Were there any secondary sulfide or arsenide minerals present?

*Whiteside:* Quite honestly, I don't know. The mineralogists told me they were so scared of wiping the gold off when they wanted to do more polishing on it, that they were not able to determine very much except that it was gold. Sorry.

*Feather:* Do we still have the sample at the research laboratory?

*Whiteside:* Yes, that sample is in a core shed on Western Deep Levels. I must say, I would like to know a little bit more about that.

*Feather:* Yes, I would like to look up the sample and in fact also analyze for the silver content of the gold. That would be interesting, too.

*Whiteside:* Yes, it would be. I should think they could do that with a microprobe.

*Wilfried Meyers, Falconbridge Nickel Mines, Ltd., Canada:* I'd like to ask you more about the absence of magnetite throughout the Witwatersrand. This is something that apparently also holds true for the Blind River area and other pyritic conglomerates in Canada and other parts of the world. What happened to the magnetite? It must have been there originally with a vast granite source terrane. Where has it all gone to?

*Whiteside:* Well, as I say, I think it has been converted to sulfide.

*Myers:* Where did all the sulfur come from?

*Whiteside:* That's one of the problems.

*Meyers:* If you expect sulfur to move into every nook and cranny within that vast sedimentary pile to change every grain of magnetite or hematite that may have been there originally, you must have had an absolute, complete pervasion by solutions, or however that sulfur got in there. What I am trying to drive at is, if you can do that, you can also bring in uranium and gold in solutions and not by detrital means.

*Whiteside:* Well, the one thing I can assure you is that

there is no magnetite in the Witwatersrand. The only place it has ever been identified has been in pebbles, not in the matrix. Occasionally I believe that you do get small grains of magnetite in the iron-bearing rocks as pebbles. And another thing, incidentally, you get very little ilmenite, but you do get a lot of leucoxene, and that's almost spherical. Say, I'm talking out of turn. I am not a mineralogist. Sorry, but I don't know where the sulfur comes from, but there are, as I say, various stages roughly in the Witwatersrand where you could bring in sulfur. We have volcanicity in the Dominion Reef, in the Witwatersrand in two places,

and also an enormous amount in the Ventersdorp. Going further, you've got the black carbon of the dolomite, and then finally you've got the Bushveld Complex, which presumably could have brought in quite a lot of sulfur.

*Schidlowski:* Just a very quick comment on the sulfur budget. If you convert part of the pyrite into pyrrhotite you get a sulfur surplus which can be used for sulfidation processes. In fact, conversion of pyrite into pyrrhotite has obviously occurred during the pseudo-hydrothermal reconstitution of the reef; that has been observed in several places.



# Microprobe Analyses of Uranium and Thorium in Uraninite from the Witwatersrand, South Africa, and Blind River, Ontario, Canada

By D. E. GRANDSTAFF

GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN  
QUARTZ-PEBBLE CONGLOMERATES

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## MICROPROBE ANALYSES OF URANIUM AND THORIUM IN URANINITE FROM THE WITWATERSRAND, SOUTH AFRICA, AND BLIND RIVER, ONTARIO, CANADA

By D. E. GRANDSTAFF<sup>1</sup>

### ABSTRACT

Micropore analyses of uranium and thorium in uraninite grains from the Witwatersrand, South Africa, and Blind River, Ontario, reveal that although individual grains are fairly homogeneous, the assemblage of grains is quite heterogeneous. This heterogeneity appears to favor genetic concepts advocating a detrital, placer origin for the uraninite.

### INTRODUCTION

The bulk chemical compositions of uraninite concentrates from the Witwatersrand basin of South Africa and the Blind River-Elliot Lake area of Ontario, Canada, have been reported by several investigators (Liebenberg, 1955; Hiemstra, 1968; Roscoe, 1979). The results of these analyses reveal that the concentrates from the Blind River area are quite similar in regard to their uranium and thorium content. The  $UO_2/ThO_2$  ratio of the Blind River concentrates varies from approximately 8 to 12 and averages about 10 (Roscoe, 1979). In contrast, analyses of concentrates from the Witwatersrand basin reveal a wider range of composition. The  $UO_2/ThO_2$  ratio varies from approximately 10 to 40 as a result of the variation in  $ThO_2$  content, which ranges from 1.5 to 6.5 percent. However, very little has been reported on the composition of individual uraninite grains (Schidlowski, 1966) or variations within or between grains. This paper reports the results of analyses made by electron microprobe of uranium and thorium in individual uraninite grains.

### MATERIAL

Two samples were examined in this study, a sample of very high grade uranium ore from the Rio Algoma Nordic Mine, Elliot Lake, Ontario, Canada, and a lower grade ore sample from the Vaal Reef West mine, Klerksdorp area, Transvaal, South Africa, from the upper division of the Witwatersrand System.

The Canadian sample was unusually rich in uraninite, and a single polished surface exposed a large number of uraninite grains. All the grains analyzed from this sample appeared on this polished surface and lay within an approximately  $4 \text{ cm}^2$  area.

Because the ore grade of the Witwatersrand sample was somewhat lower than that of the Blind River sample, only a few uraninite grains were exposed on any single polished surface. Therefore, a small cube approximately  $64 \text{ cm}^3$  was crushed to between 80 and 200 mesh, approximately 60 to  $200 \mu$ . This is about the size range of many of the uraninite grains (Liebenberg, 1955). The uraninite grains were separated by a method involving two heavy liquids, bromoform and Clerici solution, and the Frantz isodynamic separator. A polished section grain mount was made of the separated grains. Microscopic examination revealed that in some cases the uraninite grains were still in contact with matrix material around a large portion of their perimeters. These grains were probably nearly whole and therefore were used in the examination for internal zoning.

### ANALYSES

The analyses were made using an ARL-EMX<sup>2</sup> microprobe. The beam diameter was approximately  $1 \mu$ , the sample current was  $0.05 \mu \text{A}$  and the accelerating voltage was 15 kV. The data obtained were corrected for drift (which was negligible), deadtime, and background. Artificial  $UO_2$  and  $ThO_2$  crystals were used as standards.

### RESULTS

Results of analyses for uranium and thorium are shown in figures 1-4.

Step scans for uranium and thorium were made across several grains. Representative results are shown in

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<sup>1</sup> Department of Geology, Temple University, Philadelphia, Pa. 19122.

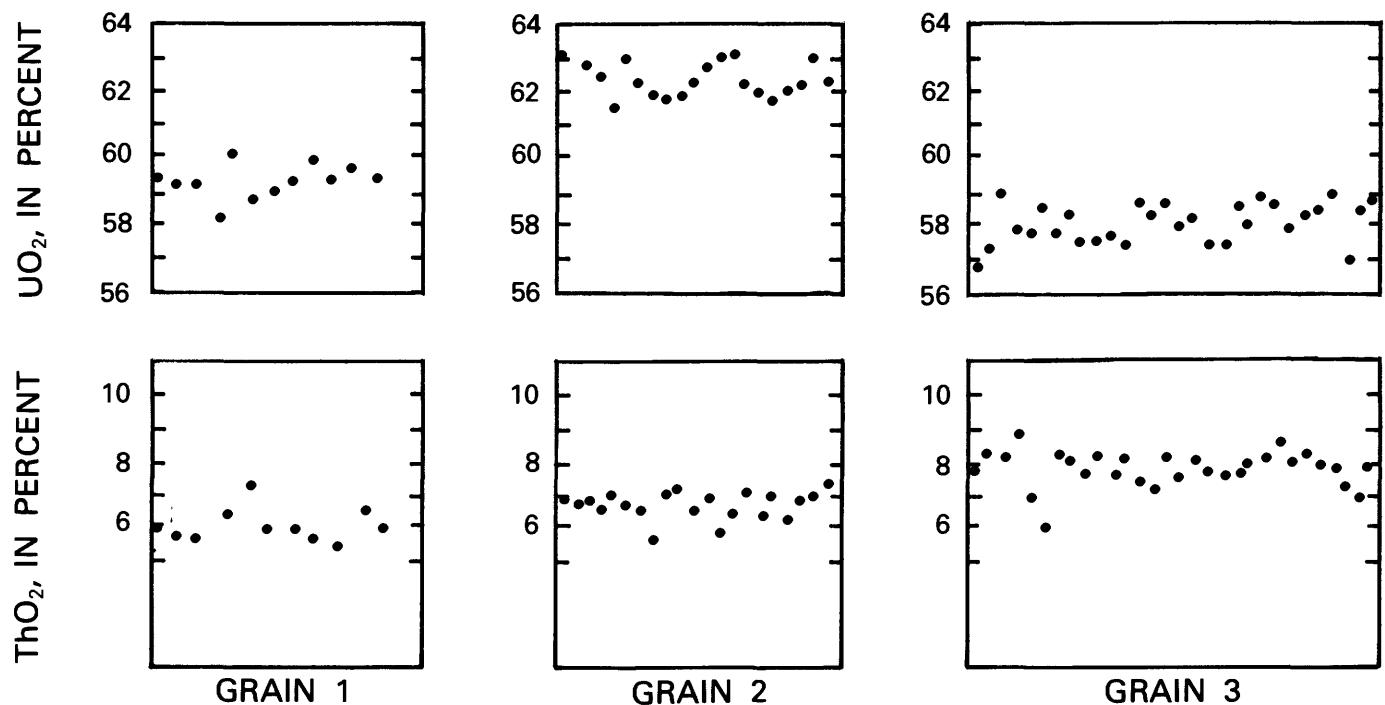


FIGURE 1.—Representative step scans across three uraninite grains. These step scans illustrate the lack of zoning within individual uraninite grains. Note, however, the difference in  $\text{UO}_2$  and  $\text{ThO}_2$  contents among the grains.

figure 1. The step scans did not reveal any major zoning within the grains examined. Other grains were examined for zoning by use of X-ray area displays for uranium and thorium. The X-ray area displays did not reveal zoning in the grains examined, although this method is less sensitive than is use of step scans.

Quantitative analyses of spots in the uraninite grains are shown in figure 2. Quantitative analyses were made in areas that were optically free of inclusions.

The results show that although individual grains are fairly homogeneous internally, the assemblage of grains is quite heterogeneous. Uranium concentration, calculated as  $\text{UO}_2$ , varies from 58 to 69 percent in the Blind River sample and from 55 to 77 percent in the Witwatersrand sample. Thorium, calculated as  $\text{ThO}_2$ , ranged from 4 to 9 percent in the Blind River sample and from 0.8 to 11 percent in the Witwatersrand sample. The uranium and thorium in both samples are approximately log normally distributed.

The  $\text{UO}_2/\text{ThO}_2$  ratio as a function of  $\text{ThO}_2$  concentration is shown in figures 3 and 4. The figures show a hyperbolic trend, which would be expected from variation of a minor or trace element (thorium) in a matrix of a major, relatively invariant element (uranium). The

trend is particularly clear in the Witwatersrand sample, owing to the wide variation in grain compositions, but is also apparent in the Blind River sample.

It is possible that some scatter of the data may be due to the presence of inclusions of galena or silicates in the region impinged upon by the electron beam or to the expulsion of differing amounts of radiogenic lead as galena by the different uraninite grains. However, this probably does not account for the major trend observed. If the inclusions or exsolution products contain little or no uranium or thorium, either mechanism should generate apparent uraninite compositions lying along a mixing line having a constant  $\text{UO}_2/\text{ThO}_2$  ratio equal to that of the original grain. Such a trend would lie at a high angle to the trend actually observed in figures 3 and 4 except at very low  $\text{UO}_2/\text{ThO}_2$  ratios. Therefore, these mechanisms are not considered important in causing the scatter of data shown in figure 2.

## DISCUSSION

The origin of the Blind River and Witwatersrand uraniferous conglomeratic deposits has been extensively discussed. A variety of stratigraphic, geochronological,

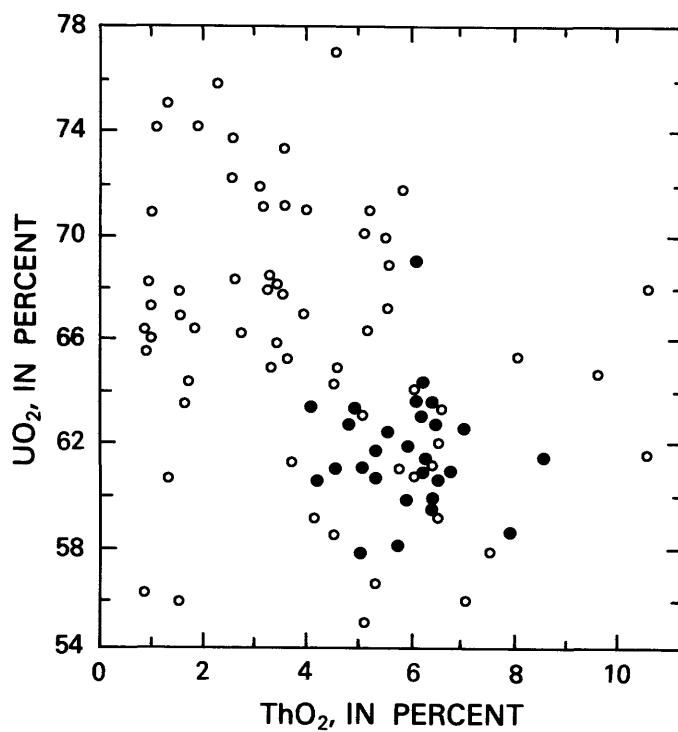


FIGURE 2.— $\text{UO}_2$  and  $\text{ThO}_2$  content of uraninite grains from the Rio Algoma Nordic Mine, Blind River, Ontario (closed circles), and from the Vaal Reef West Mine, Witwatersrand, South Africa (open circles).

geochemical, and sedimentological evidence has been brought to bear on the problem of origin. Most geologists appear to favor the placer or modified placer hypothesis, which states that the uraninite grains were deposited syngenetically in the sediments as placer origins.

The composition of the uraninite grains has major bearing on the problem of genesis. Most discussion has centered on the thorium content of the grains. Low-temperature, hydrothermally derived uraninite contains little, if any, thorium, while uraninite derived from pyrogenetic sources, from granites or pegmatites, may contain appreciable thorium (Frondel, 1958; Ramdohr, 1969). Therefore, the high thorium content of many of the grains examined in this study supports the hypothesis that they are derived from granitic or pegmatitic sources.

The great compositional variability of the grains from within a small rock volume produces additional problems for advocates of a hydrothermal origin of the uraninite grains. Such a great compositional variability would require that the grains grew in very specialized microenvironments and that the mineralizing fluid was heterogeneous over short distances. Such conditions are unlikely. Any movement of a heterogeneous mineralizing fluid through the rock might be expected to produce

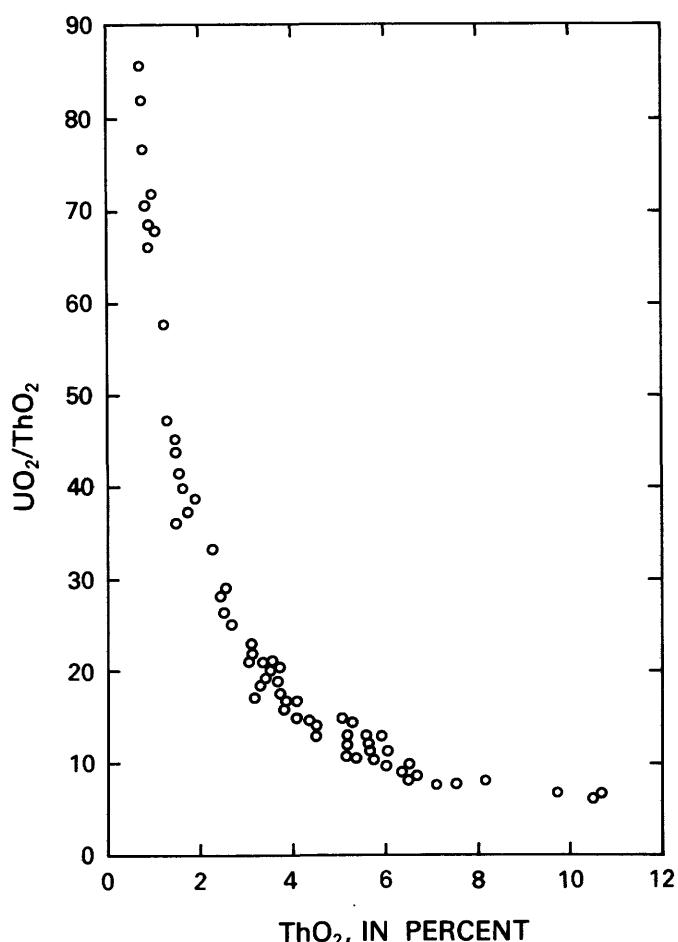


FIGURE 3.—Plot of  $\text{UO}_2/\text{ThO}_2$  against  $\text{ThO}_2$  in uraninite grains from the Vaal Reef West mine, Witwatersrand, South Africa.

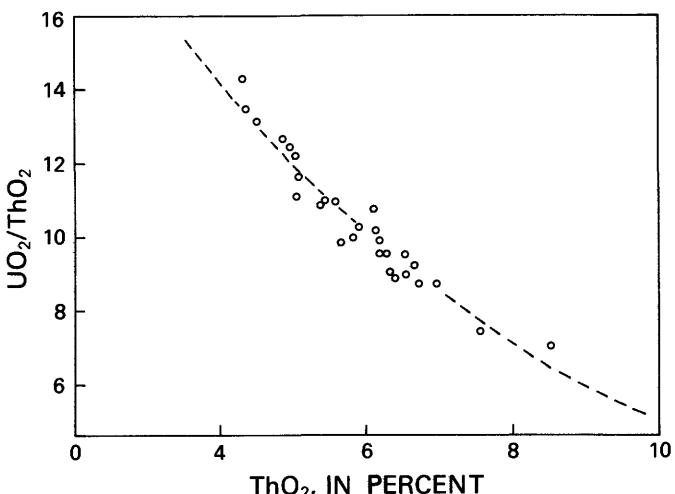


FIGURE 4.—Plot of  $\text{UO}_2/\text{ThO}_2$  against  $\text{ThO}_2$  in uraninite grains from the Rio Algoma Nordic Mine, Blind River, Ontario.

zoning in the growing grains. However, no zoning was observed in this study. Nor does it seem likely that a later thermal event could have erased any zoning. The Witwatersrand sediments are regionally metamorphosed only to the greenschist facies (Fuller, 1958), while the Blind River sediments are virtually unmetamorphosed (Roscoe, 1969). Therefore, the chemical variability appears more consistent with the hypothesis that pyrogenetic uraninite grains of varied compositions were gathered from different localities during erosion and this chemically heterogeneous mixture was transported and deposited as a mechanical mixture.

The difference in variability of the composition of the two samples may yield some evidence concerning their source areas. The uranite from the Blind River sample has much smaller variability than the Witwatersrand sample. This may indicate that the uraninite in the Blind River sample was derived from a single relatively homogeneous source area, whereas the uraninite in the Witwatersrand sample had multiple sources.

Such possibilities have already been suggested. Fuller (1960) and Hiemstra (1968) have suggested that the sediment may have been derived from different source areas. Viljoen and others (1970) have argued for a heterogeneous source for the Witwatersrand basin uraninite in order to account for the variation of thorium content between the Dominion Reef and Witwatersrand Series uraninites. Variations of  $UO_2$  and  $ThO_2$  in samples from many parts of the Witwatersrand basin should be investigated to study the possibility of multiple or heterogeneous sources for the uraninite.

#### ACKNOWLEDGEMENTS

I would like to thank Dr. E. S. A. Antrobus and the Anglo-American Mining Corp. and the Rio-Algom Mining Corp. for providing the samples, and Mr. C. Kulik for aid with the microprobe analysis. This work was undertaken as a part of a doctoral dissertation at Princeton University. The work was supported by NDEA grants under Title IV, and by grants to the Princeton University computer center.

I would also like to thank the Geological Society of South Africa for permission to have this article republished. It appeared first in the Transactions of the Geological Society of South Africa in 1975, v. 77, p. 291-294. The U.S. Geological Survey also thanks the Geological Society of South Africa for permission to republish this paper.

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#### DISCUSSION

*Skinner:* Perhaps this is not a fair question for you, David, but one of the South Africans may step in. If the Witwatersrand can be considered to be a large sample of the detrital minerals from the crust and if the crustal abundance figures for thorium and uranium are in fact correct, showing that thorium is four or five times more abundant than uranium, where is all the thorium?

*Grandstaff:* I don't think that, in fact, the Witwatersrand represents a major sample of the crust. If you plot up analyses of pegmatitic uraninites from the various parts of the world, they scatter quite widely in uranium-to-thorium ratio. I think we are really looking at an individual chance segregation of uranium over thorium.

*Skinner:* Then would it be a correct conclusion to draw that if there is another hidden pebble conglomerate somewhere the chances are it is a thorium rich one, not a uranium rich one?

*Grandstaff:* Could well be, yes.

*Robertson:* In comment to Dr. Skinner's question, perhaps one should point out that certainly in the Blind River ores monazite is a fairly significant component.

*Roscoe:* You could add to that that we do get conglomerates in the Blind River ores which have very high thorium-uranium ratios, 4 to 1, and they appear to complement the high uraninite ones. The concept is, of course, that this is hydraulic differentiation.

*Grandstaff:* Those high thorium concentrations that you pointed out are largely monazite-zircon concentrates rather than uraninite concentrates.

*Simpson:* Did you analyze any uraninite within carbon? Our analyses show that within a given sample, this type of uraninite is generally thorium poor and homogeneous.

*Grandstaff:* I analyzed only a couple of grains from carbon leaders, and they contained several percent thorium and were relatively homogeneous.

*Feather:* I would like to comment on your question, Peter [Simpson]. I have analyzed a great many uraninite grains, both the distinctly detrital grains, that is, grains not associated with carbon, and grains within carbon. And I have found as wide a variation of thorium content in uraninite grains included in carbon as I have found in uraninite grains that were not associated with the carbon.

*Grandstaff:* I analyzed one thucholite uraninite grain which had about 8 to 10 percent thorium. They do get that high.



Preliminary Notes Concerning the Uranium-Gold Ratio and the  
Gradient of Heavy-Mineral Size Distribution as Factors of  
Transport Distance down the Paleoslope of the  
Proterozoic Steyn Reef Placer Deposit,  
Orange Free State Goldfield, Witwatersrand, South Africa

*By* W. E. L. MINTER

GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN  
QUARTZ-PEBBLE CONGLOMERATES

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GEOLOGICAL SURVEY PROFESSIONAL PAPER 1161-K





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## GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN QUARTZ-PEBBLE CONGLOMERATES

# PRELIMINARY NOTES CONCERNING THE URANIUM-GOLD RATIO AND THE GRADIENT OF HEAVY-MINERAL SIZE DISTRIBUTION AS FACTORS OF TRANSPORT DISTANCE DOWN THE PALEOSLOPE OF THE PROTEROZOIC STEYN REEF PLACER DEPOSIT, ORANGE FREE STATE GOLDFIELD, WITWATERSRAND, SOUTH AFRICA

By W. E. L. MINTER<sup>1</sup>

### ABSTRACT

The size decrease of quartz pebbles, pyrite nodules, and zircon grains, evident from samples of Steyn Reef taken from various positions down a paleoslope indicated by crossbedding data, confirms their detrital origin. An increase in the ratio of uranium to gold, which appears to be related to their original size-frequency distribution, also indicates the paleoslope direction and effectively distinguishes between basket and carbon-seam reefs.

### INTRODUCTION

The hydraulic size equivalence between uraninite grains and the sand-sized fraction of South African Proterozoic placer deposits, described by Koen (1961), was substantiated by Viljoen (1963), who found that the equivalence also applied to pyrite and zircon grains and to a number of other minerals. These were consequently considered to have been detrital components of the placers. Furthermore, Viljoen demonstrated that there was a decreasing size gradient of these minerals down the paleoslope of the placers he sampled.

A gradient of this nature depends, however, on the hydraulics of the sedimentological environment in which the placer was deposited and on the part of the paleoslope that has been preserved. For instance, the exponential tail of grain-size decrease that may be expected in a distal position on a paleoslope plus the transgressive process involved during deposition of the Vaal Reef (Minter, 1972) resulted in a practically insignificant size gradient.

In the Orange Free State Goldfield, the Steyn Reef placer deposit, which is a regressive deposit, lies upon practically the same paleosurface as the Basal Reef

(Sims, 1969) and has a sequence from proximal, large-pebble conglomerate basket reef to distal, small-pebbly sand carbon-seam reef, preserved down a paleoslope distance that exceeds 15 km. This provides an opportunity to study size changes of detrital minerals over a wide range of hydraulic energy levels.

### PARTICLE-SIZE CHANGES IN THE STEYN REEF

The mean sizes of quartz pebbles, massive pyrite nodules, and zircon grains were obtained by measuring their apparent maximum axes, in phi units, from the smoothed surfaces, cut normal to bedding, of samples collected down 15 km of paleoslope. Over this distance, quartz pebbles grade from cobble to small-pebble size, pyrite nodules grade from small-pebble to pinhead size (fig. 1), and zircons grade from fine to very fine grained. The change in zircon sizes is less conspicuous than for either pebbles or pyrite (fig. 2), but it was noticeable that zircon grains become much more abundant in distal samples. However, all three parameters reflect the paleoslope that has been well defined by abundant cross-bedding data (Sims, 1969). Isopleths will be drawn when more data are available and after the area has been structurally reconstructed.

### MINERAL CONCENTRATION CHANGES IN THE STEYN REEF

Abundant sampling data obtained from mining of the Steyn Reef down the entire paleoslope indicate that the overall gold content decreases while the overall uranium content increases. This is illustrated in figure 2 by a plot of the ratio of uranium to gold, the concentration of both having been measured in parts per million. Isopleths be-

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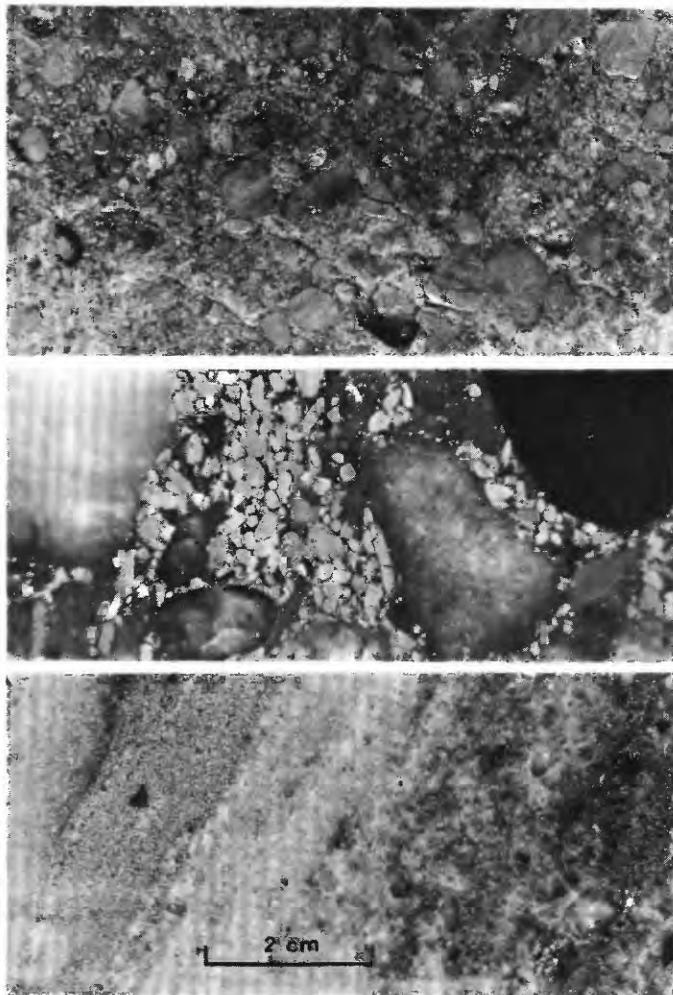


FIGURE 1.—Photographs of samples of Steyn Reef taken at 5-km intervals down a paleoslope that was indicated by crossbedding data in the Orange Free State Goldfield.

ing constructed substantiate the paleostrike obtained from crossbedding data.

High uranium-gold ratios appear to characterize the small-pebble distal environments of all Witwatersrand carbon-seam reefs, and low ratios, those of proximal, basket reefs (Minter, 1972). Zircon and uraninite are considered to have been derived from the same granitic provenance area, and, consequently, their low abundance in proximal deposits may be linked to an inherent sparsity of coarse grains in the original supply.

## CONCLUSIONS

Size gradients of pebbles, pyrite nodules, and zircon grains in the Steyn Reef placer indicate the dispersal direction of the prevailing paleocurrents and the detrital nature of the mineralization.

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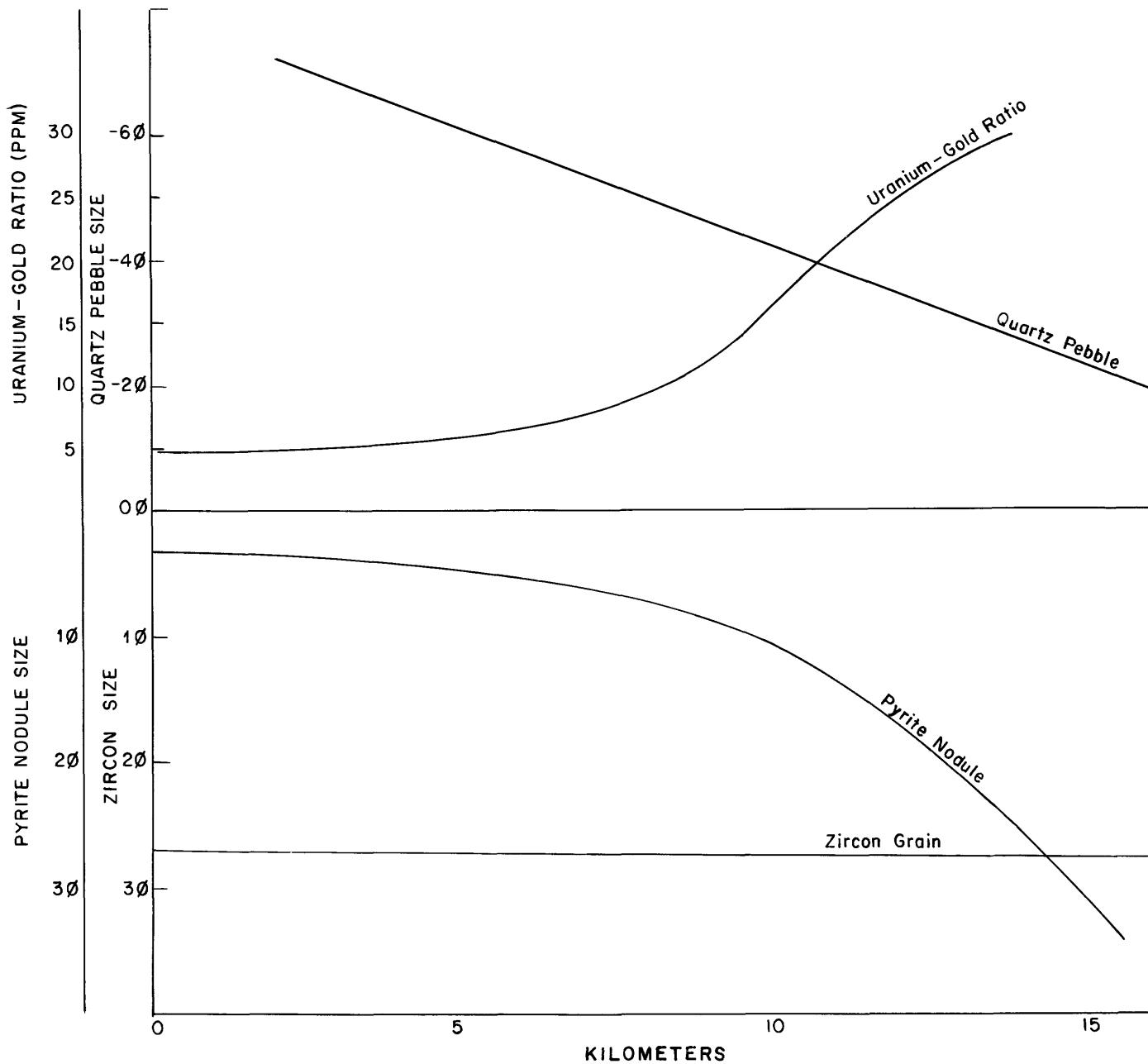


FIGURE 2.—A graphic plot that illustrates changes related to particle size and uranium-gold ratio that took place during transport of the Steyn Reef placer down 15 km of paleoslope.



# Geochemical Studies on the Origin of the Detrital Pyrites in the Conglomerates of the Witwatersrand Goldfields, South Africa

*By* RUDOLF SAAGER

GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN  
QUARTZ-PEBBLE CONGLOMERATES

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## GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN QUARTZ-PEBBLE CONGLOMERATES

# GEOCHEMICAL STUDIES ON THE ORIGIN OF THE DETRITAL PYRITES IN THE CONGLOMERATES OF THE WITWATERSRAND GOLDFIELDS, SOUTH AFRICA

By RUDOLF SAAGER<sup>1</sup>

### ABSTRACT

The principal source area of the Witwatersrand sediments, including heavy minerals such as gold, uraninite, and pyrite, lies to the west and north of the basin of deposition. Because this area is overlain by younger rocks, the actual identification of the source area is virtually impossible. On the basis of a hypothetical provenance model it has been suggested earlier that the primary source of the sediments was in the Archean granite-greenstone terrane of the Kaapvaal Craton.

For the present study, 57 pyrite samples from primary gold deposits of the greenstone terrane (Swaziland Sequence) and from various gold reefs of the Witwatersrand deposit have been investigated for their trace-element content (Co, Ni, Cu, Zn, Pb, Ag) in order to study the possible presence of similar geochemical trends within both groups of pyrite. The analytical data have been treated by statistical methods, and the results indicate that both the primary and the detrital Witwatersrand pyrites, with respect to the investigated elements, were drawn from the same population. They are both clearly different from the Witwatersrand pyrites formed in situ. Lead isotope investigations carried out on sulfides from primary gold deposits of the Swaziland Sequence and on detrital and authigenic pyrites from the Witwatersrand deposit showed similar isotopic compositions of the lead in detrital pyrite and in primary sulfides from the Swaziland Sequence. The results of the present geochemical studies thus constitute strong evidence that the Archean greenstone belts of the Kaapvaal Craton were the most probable source of the detrital pyrites and also of the gold in the sediments of the Witwatersrand Basin.

### INTRODUCTION AND GEOLOGICAL SETTING

For many placer deposits the question of the original source of the heavy minerals is an intriguing and often hotly debated subject of discussion. The solution of the problem is not only of great academic interest but in many instances has proved to be of considerable economic importance.

In the case of the early Precambrian fossil placers of the Witwatersrand Basin in South Africa, the problem of the provenance of the gold, uraninite, and other detrital conglomerate constituents has been overshadowed for a long time by the metallogenetic con-

troversy between the hydrothermalists and the placerists (see for example Becker, 1909; Mellor, 1916; Young, 1917; Graton, 1930; Liebenberg, 1955; Ramdohr, 1955; Davidson, 1957, 1960, 1961). The very common criterion, "Gold is where you find it," and the preoccupation of the mine geologists on the Rand with the daily problems of exploiting the precious metal also hindered the search for the original source of the placer minerals, in spite of its utmost importance to the case of the placerists.

A great number of sedimentological, geological, and mineralogical studies carried out during the last twenty years led to the general acceptance of the modified placer theory as a metallogenetic concept for the Witwatersrand deposit. The concept implies that, after their deposition, some of the detrital minerals, notably gold and the sulfides, were reconstituted and lost their waterworn shapes as a result of the low-grade regional metamorphic overprint of the embedding Witwatersrand and Dominion Reef sediments.

Today most investigators of the Witwatersrand deposit agree that the sediments and heavy minerals originated essentially from a source area situated to the north and west of the basin of deposition, with only minor amounts of sedimentary material being introduced from the southeastern edge of the Witwatersrand Basin (Pretorius, 1974). Some of the evidence for this conclusion is found in the conspicuous occurrence of all the producing goldfields along the northwestern arc of the Witwatersrand Basin, in the asymmetry of the gradients of the paleoslopes, which are considerably steeper on the northwestern side of the basin, and in the distinct decrease of conglomerate thickness and pebble size in a southerly to southeasterly direction. Pretorius (1966, 1974) convincingly demonstrated that each of the six major goldfields represents a fluvial fan or fan delta (fig. 1). Furthermore, he concluded that the fluvial system was short, the provenance area being close to the edge

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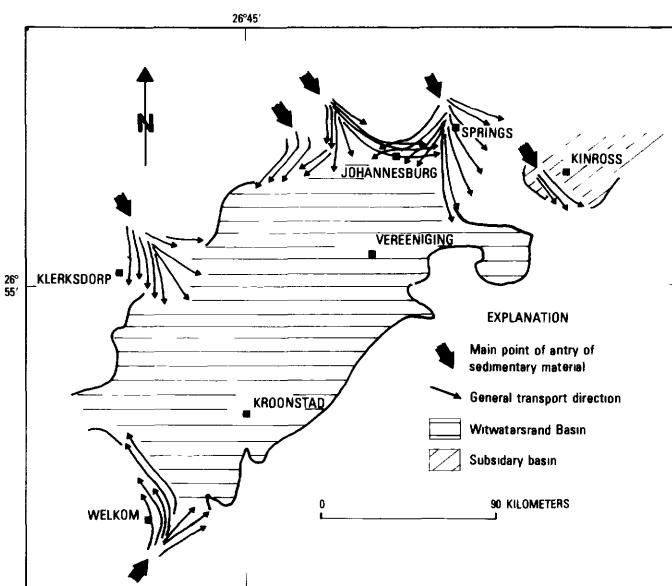


FIGURE 1.—Locations of main points of entry and general transport directions of sedimentary material into the Witwatersrand Basin (modified from Brock and Pretorius, 1964, pl. 6).

of the basin of deposition. The entry points of sedimentary material into the basin, corresponding to the apices of the fans, thus must have remained relatively fixed during a considerably long period of time—that is, from Dominion Reef till Upper Witwatersrand times.

To pinpoint the actual provenance area of the sediments and the placer minerals and thus to locate the primary gold and uranium-mineralized areas, however, is virtually impossible, as these areas are largely covered by younger Proterozoic rocks, such as the Ventersdorp lavas, the Transvaal sediments, and the Bushveld Igneous Complex. The few windows in which pre-Witwatersrand rocks are exposed, for example the Johannesburg Dome, Westerdam Dome, and Amalia Dome, show only rock types similar to those found in the Archean basement of the eastern Transvaal Lowveld but to no other pre-Witwatersrand sequences.

Viljoen and others (1970), after synthesizing recently published data on the Witwatersrand Basin and on the basement of the southern African Kaapvaal Craton, suggested that the primary source of the Witwatersrand gold and detrital sulfides should be sought in the Archean greenstone belts of the craton. As a hypothetical model source terrane, they proposed the classic area of the Barberton Mountain Land (fig. 2).

In the Kaapvaal Craton, the generally northeast-trending Archean greenstone assemblages are referred to as the Swaziland Sequence. The lowermost members of the Swaziland Sequence, which in the Barberton area are fully preserved, constitute the Onverwacht Group,

characterized by ultramafic, mafic, and acid volcanics (Viljoen and Viljoen, 1969; Anhaeusser, 1971). These rocks are overlain by the argillaceous sediments of the Fig Tree Group, which, in turn, is followed by the principally arenaceous sediments of the Moodies Group, forming the topmost members of the sequence. As with other greenstone belts in the Kaapvaal Craton, the Barberton Mountain Land is well mineralized and forms, next to the Witwatersrand Basin, the most important gold province in South Africa (fig. 2).

Most of the gold occurrences in the Barberton Mountain Land are located along a stratigraphic horizon in basic volcanics at the base of the Upper Onverwacht Group and along unconformities between the Upper Onverwacht Group and the sedimentary groups of the Swaziland Sequence. Viljoen and others (1969) showed that the gold content of the ultramafic and mafic magnesium-rich volcanics in the Barberton Mountain Land ranges from 0.005 to 0.01 ppm (parts per million). Gold contents of as much as 0.112 ppm have been observed where the rocks have been weathered and recycled. The gold content in the accompanying intrusive basement granites, on the other hand, is less than 0.005 ppm. From these geochemical trends and from various geological and mineralogical observations, Viljoen and others (1969) and Saager (1973a, b) concluded that the gold deposits in the greenstone belts are genetically related to the magnesium-rich mafic to ultramafic volcanics and not to the late volatile phases of the intruding granites. Descriptions of the various processes that led to the formation of the gold deposits in the greenstone belts of the Kaapvaal Craton are given by Viljoen and others (1969) and Saager (1973b).

The suitability of the Barberton Mountain Land as a model source terrane for the sedimentary filling of the Witwatersrand Basin was demonstrated by Viljoen and others (1970) owing to a remarkable correlation between the expected heavy minerals derived from successive erosional levels of such a terrane and the actual heavy minerals encountered in the various horizons of the Witwatersrand sediments. The results of their study are summarized in table 1. It demonstrates the relative abundance of uraninite, monazite, cassiterite, and garnet in the lower portions of the sedimentary succession, especially in the conglomerates of the Dominion Reef Sequence. Chromite and the platinoids show a distinct increase towards the topmost members of the sediments, and the gold, as well as the sulfides, reaches a maximum in the Main Bird Series of the Upper Witwatersrand Sequence. This heavy-mineral distribution indicates that, in the provenance area, the lower erosional level consisted mainly of mafic to ultramafic rocks, whereas the higher levels of erosion consisted more and more of granitic, pegmatite-rich rocks.

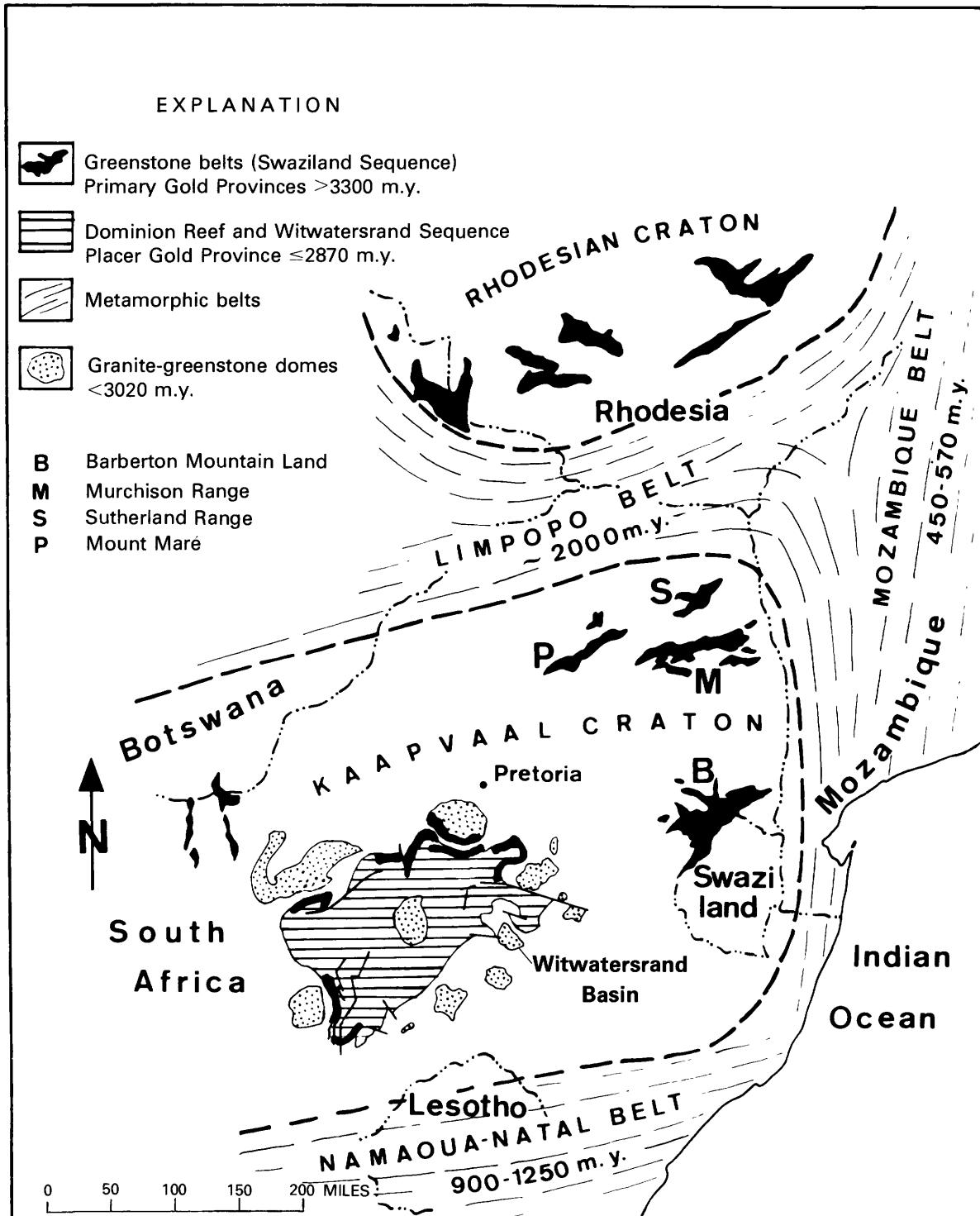


FIGURE 2.—Part of the southern African Shield, showing general geological setting of the Witwatersrand Basin and the major Archean greenstone belts of the Kaapvaal Craton. The locations of the major goldfields along the western and northern rim of the Witwatersrand Basin are marked with a heavy black line (modified from Viljoen and others, 1970, fig. 1).

TABLE 1.—*The relative abundance of some heavy minerals in the rudaceous sediments of the Witwatersrand Basin and their corresponding erosion units in the model source area of the Barberton Mountain Land*

(Modified from Viljoen and others (1970, p. 174). Relative abundance of heavy minerals (U, uraninite; G, garnet; M, monazite; C, cassiterite; Z, zircon; G, gold; S, sulfides; C, chromite, P, platinoids): †, not abundant; ††, fairly abundant; †††, very abundant)

Stratigraphic position in sediments of the Wit- watersrand Basin, in stratigraphic sequence	Detrital minerals								Main rock types in the model provenance ter- rane (Barberton Moun- tain Land), in reverse stratigraphic sequence
	U	G	M	C	Z	G	S	C	
Kimberley-Elsburg Series.	†				††	††	†††	††††	I. <i>Ultramafic Unit (Lower Onverwacht Group):</i> Ultramafic to mafic lavas. Minor felsic tuffs and porphyries. Low- volatile tonalitic granite.
Main-Bird Series	—	—	—	—	—	—	—	—	II. <i>Mafic to Felsic Unit (Upper Onverwacht Group):</i> Mafic, andesitic, and felsic lavas. Felsic to mafic pyro- clasts. Banded chert. Low-volatile tonalitic granite. Some high-volatile granite (root zone).
Lower Witwatersrand Sequence.	††			—	—	—	—	—	III. <i>Argillaceous Unit (Fig Tree Group):</i> Shales, greywackes, banded ironstone. Low-volatile tonalitic granite. High-volatile granite (root zone).
Dominion Reef Sequence.	—	—	—	—	—	—	—	—	IV. <i>Arenaceous Unit (Moodies Group):</i> Quartzites, conglomer- ates. High-volatile "hood" granite. Pegmatites.

The present geochemical investigation was undertaken to further test the source hypothesis of Viljoen and others (1970). The study was restricted to pyrite because it forms the most abundant ore mineral in the primary gold ores of the Archean greenstone belts and in the Witwatersrand conglomerates. It was carried out, first, by a trace-element investigation of pyrites from the two gold provinces and, second, by a comparison of the lead-isotope composition of Witwatersrand pyrites with that of pyrites and lead-bearing sulfides associated with various gold deposits in the Barberton Mountain Land.

## PYRITE MINERALOGY

### WITWATERSRAND BASIN

In the Witwatersrand ores pyrite generally constitutes more than 90 percent of the ore minerals present and appears to be the only macroscopically identifiable ore mineral (Saager, 1970). It generally occurs disseminated throughout the entire width of individual ore horizons but in many places is concentrated, together with other detrital ore minerals (such as gold,

uraninite, chromite), in false footwalls and in footwall contacts. Between 50 and 75 percent of the pyrites in the reef horizons are rounded (fig. 3A). These grains have been described by Becker (1897), who considered them to be of detrital origin.

Witwatersrand pyrites have been classified by Ramdohr (1955) and Saager (1970); the different pyrite structures are summarized in table 2. In general, pyrite can be differentiated into the three following groups:

1. Rounded allogenetic pyrites form the bulk of the rounded grains. They are exceptionally unweathered, are generally less than 0.2 mm in size, and acquired their roundness by abrasion during sedimentary transport (fig. 3A-D). Besides their waterworn shapes, occasional primary inclusions of silver-rich gold particles, possessing a higher reflectivity than the yellow (silver-depleted) reconstituted gold in the conglomerate matrix, provide further evidence for a detrital origin of these pyrite grains (see also Feather and Koen, 1975). Other primary inclusions in the rounded allogenetic pyrite grains—pyrrhotite, pentlandite, chalcopyrite, and, less often, sphalerite—are essentially the same as in the pyrite crystals of the primary gold mineralizations of the Archean greenstone belts.
2. Concretionary authigenic pyrites have diameters several times larger than those of rounded allogenetic pyrites and form the largest proportion of the "buckshot pyrites," which often are as large as hazelnuts. In spite of their generally loose structures and delicate forms, the concretions have distinctly rounded outlines and are very well preserved (figs. 3E-I). On account of their loose structures, long transportation distances seem to be improbable, and Ramdohr (1955) and Saager (1970) have suggested that the concretions were formed in situ. Skeletal-type growth structures, such as delicate axial crosses, have been found in practically all pyrite concretions and are characteristic features. Saager and Mihálik (1967) observed that the concretionary pyrite aggregates very often consist of an isotropic and an anisotropic variety, which differs markedly in colour, reflectivity, hardness, and trace content of nickel, cobalt, and arsenic. The pores in the marginal area of the concretionary pyrite grains sometimes contain numerous fillings of gold, chalcopyrite, and galena. This indicates that the concretions acted as traps for metamorphically mobilized ore minerals, which, after deposition of the concretionary grains, infiltrated the pores where they now form inclusions.

In rounded allogenetic pyrite grains, inclusions of galena are practically absent.

TABLE 2.—*Different pyrite structures in Witwatersrand reefs*

[Modified from Saager, 1970, p. 32. Printed with the permission of the Geological Society of South Africa]

Morphology	Structures	Allogenic	Authigenic
Rounded	Compact or porous	Detrital pyrite and black sands pyritized before their final deposition.	Black sands pyritized in situ.
	Pseudomorphic	Inhomogeneous black sands pyritized before their final deposition.	Inhomogeneous black sands pyritized in situ.
	Concretionary	—	Generally formed in situ; very rarely signs of short transportation.
	Nodules containing "mineralized bacteria."	—	Formed in situ; indicating presence of primitive life.
Idiomorphic to hypidiomorphic	Compact or porous	—	Formed in situ by reconstitution of ferrous sulfide; rarely hydrothermal along dikes.
	Compact or porous encrustations	—	Formed in situ by reconstitution during metamorphism; generally overgrow older detrital pyrites.
Xenomorphic	Veinlets and fracture filling	—	Formed in situ by reconstitution during metamorphism.

Rounded pyrite nodules containing microspherical structures of the "mineralized bacteria type" are closely related to the concretionary pyrites. The microspheres reach diameters of 10 to 15 m (micrometers) and, in rare cases, have been observed to be as large as 45 m. In most cases, the structures are irregularly distributed in the nodules (figs. 3G, H). Saager (1970) has suggested that these microspheres may represent organic morphologies. He concluded that the concretionary pyrites and rounded pyrite nodules, as products of precipitation, were formed in situ by the activity of anaerobic  $H_2S$ -generating, sulfate-reducing microorganisms, which preferably existed in small pocketlike areas in the Witwatersrand Basin.

3. Reconstituted authigenic pyrites form idiomorphic to hypidiomorphic grains or occur as late encrustations on detrital components of the conglomerate, particularly on pyrite (see figs. 3J, K). The pyrite encrustations lead directly to the common stringer-like pyrite veinlets that cut through detrital minerals and form fracture fillings in other minerals. This authigenic pyrite variety has been formed by recrystallization during the metamorphism of the Witwatersrand sediments (Saager, 1970).

In certain places dikes intrusive into the Witwatersrand rocks have influenced the content of authigenic minerals and have led to the formation of enriched masses of pyrite, galena, chalcopyrite, and sphalerite. Such occurrences, however, are local and account only for a very small proportion of the authigenic idiomorphic to hypidiomorphic

pyrites and base-metal sulfides in the Witwatersrand ores (Saager, 1968).

#### ARCHEAN GREENSTONE TERRANES

In the primary gold mineralizations of the Archean greenstone belts of the Kaapvaal Craton, pyrite is generally the most abundant ore mineral and often forms the most important carrier of gold. In the gold-vein deposits, generally two generations of pyrite have been observed (Saager, 1973b). The older generation forms large, usually idiomorphic grains measuring as much as 10 mm across. Inclusions of pyrrhotite and chalcopyrite are abundant; less common inclusions are gold, magnetite, rutile, and sphalerite. The younger pyrite generation generally exhibits pentagonal-dodecahedral outlines, grain sizes being less than 100  $\mu$ . It generally surrounds the pyrite of the first generation and rarely contains inclusions of other ore minerals.

A noticeable feature of many pyrites from the greenstone belts is the common occurrence of a distinct zonal structure caused by a variation in the trace-element content of nickel, cobalt, and arsenic (fig. 3L; see also Viljoen and others, 1969). Microprobe analyses of brownish-coloured, relatively soft, weakly anisotropic pyrite zones yielded nickel, cobalt, and arsenic contents of as much as 1.75 percent, 0.17 percent, and 0.24 percent, respectively (fig. 4). For more detailed descriptions of the mineralogy of the gold ores in the Archean greenstone belts, the reader is referred to de Villiers (1957), Schweigart and Liebenberg (1966), Viljoen and others (1969), Saager (1973b) and Liebenberg (1973).

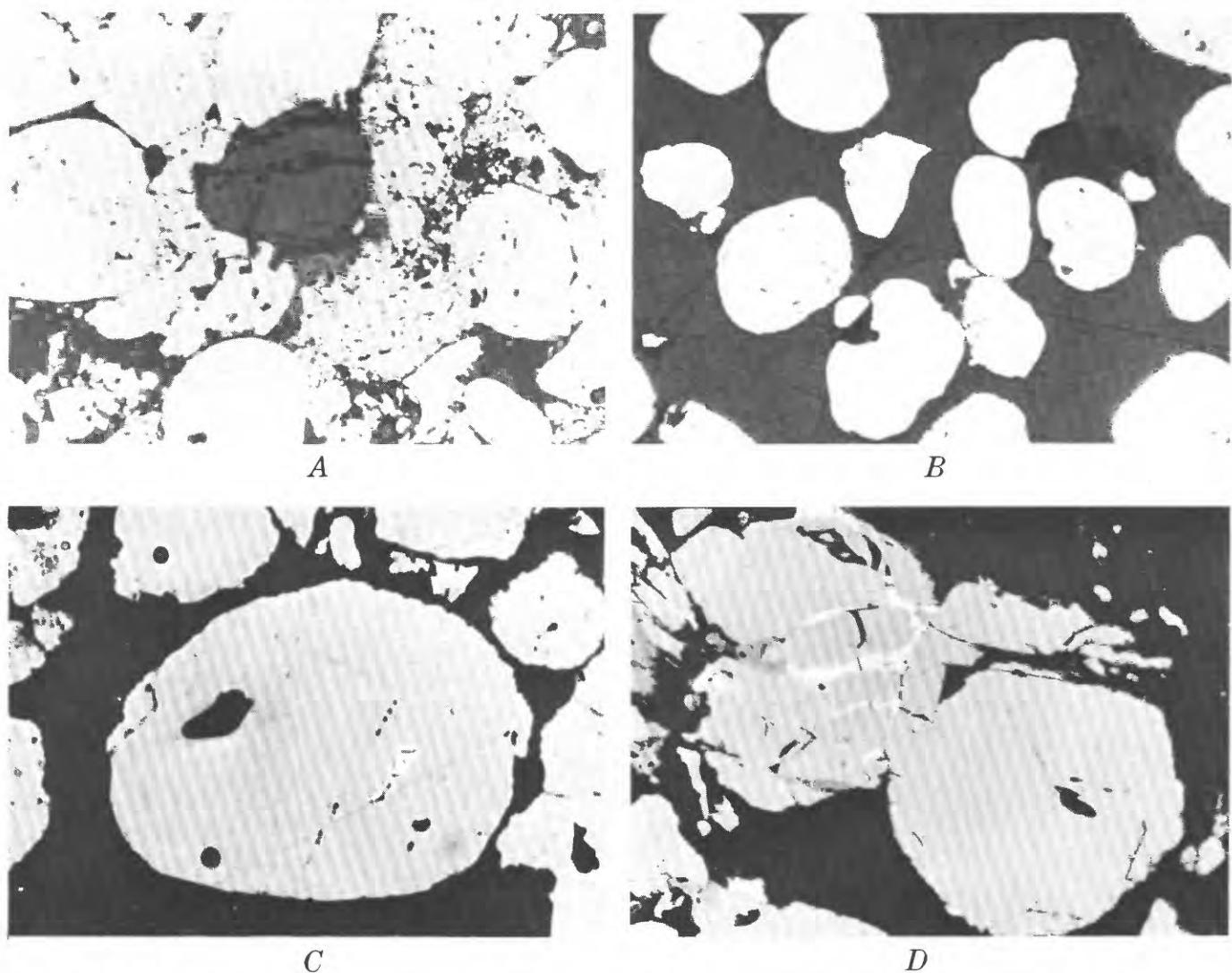


FIGURE 3.—Examples of different pyrite structures in the Witwatersrand reefs. *A*, Typical ore sample from the Basal Reef, showing abraded alloigenic pyrite and reconstituted authigenic pyrite, which molds around the rounded grains (white). Note the detrital chromite in the centre of the photograph (dark grey). Free State Geduld Mines; reflected light,  $\times 200$ . *B*, Concentrate of rounded alloigenic pyrites from the Leader Quartzite horizon. Welkom Gold Mines; reflected light,  $\times 50$ . *C*, Rounded alloigenic pyrite with "primary" inclusions of gangue (black) and gold (bright white, scratched). Free State Geduld Mines; reflected light, oil immersion,  $\times 500$ . *D*, Two rounded alloigenic pyrites in gangue (black) from the Basal Reef. Note the fracture fillings of reconstituted gold (bright white) and chalcopyrite (dark grey) in the shattered pyrite grain on the left side of the photograph. Free State Geduld Mines; reflected light, oil immersion,  $\times 500$ .

#### SAMPLE PREPARATION

Forty-four pyrite samples from 20 different primary gold deposits in the Barberton Mountain Land, the Murchison Range, and the Pietersburg Schistbelt, as well as 13 pyrite samples obtained from pay reefs of the Central Rand, Klerksdorp, and Orange Free State Goldfields, were used for the present investigation. Before concentration, a polished section was made of each ore sample, and the homogeneity and intergrowth relations of the pyrites were studied under the ore microscope.

Most of the ore specimens of primary gold deposits showed large individual pyrite grains as much as 12.5 mm in diameter. They were isolated from the sample with the use of a dentist's drill, or, after gently crushing and cleaning, the grains were concentrated by handpicking under the binocular microscope.

The concentration of rounded alloigenic Witwatersrand pyrites proved to be more complicated owing to the small grain sizes of the detrital pyrites ( $<0.5$  mm) and the omnipresence of various pyrite varieties. For these

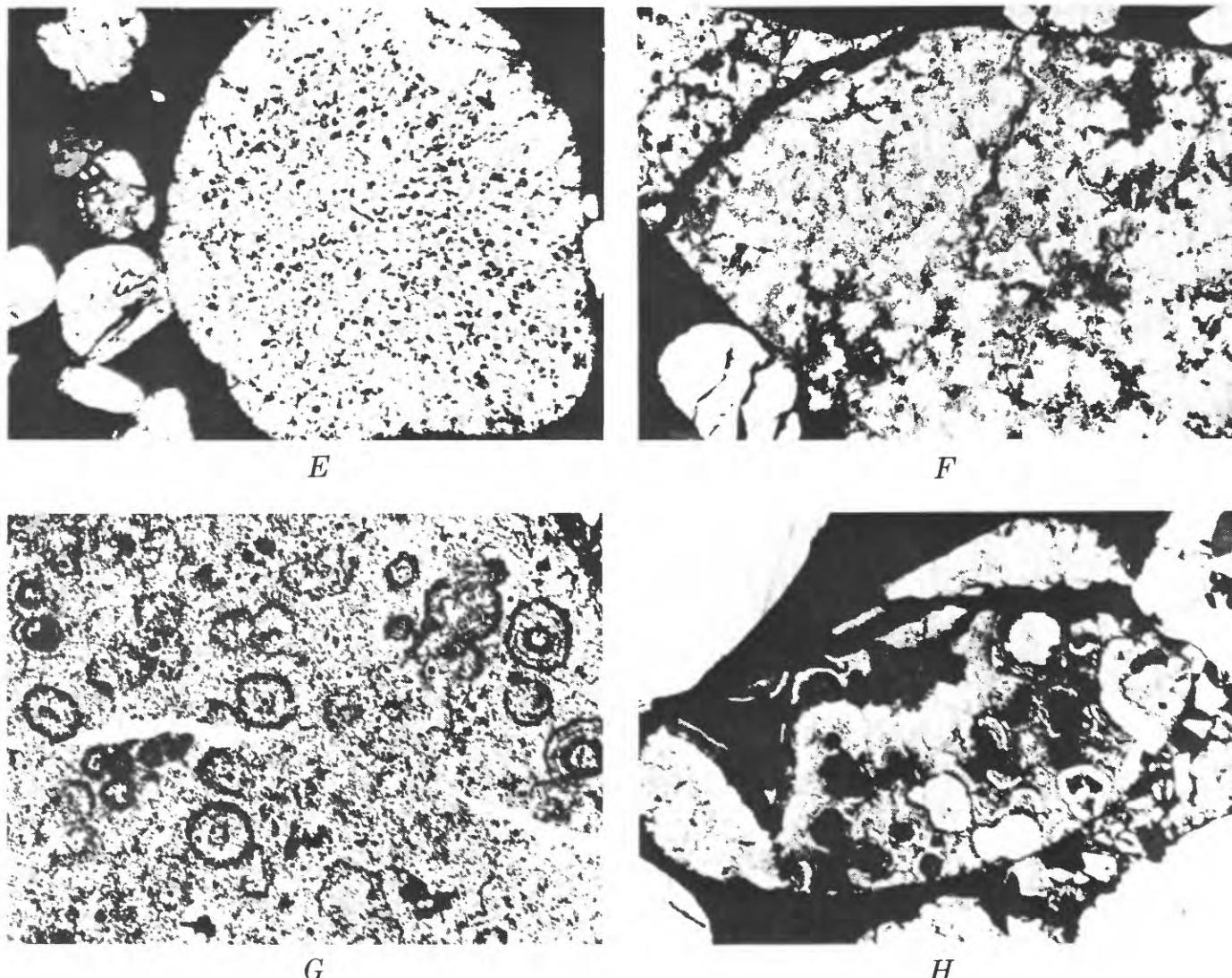


FIGURE 3.—Continued. *E*, Ore sample from the Basal Reef, showing rounded alloogenetic pyrites (partly cataclastic) and a large porous pyrite concretion. Free State Geduld Mines; reflected light,  $\times 60$ . *F*, A large, fractured, porous pyrite concretion. Free State Geduld Mines; reflected light,  $\times 100$ . *G*, "Mineralized bacteria" structures in a large authigenic pyrite concretion from the Basal Reef horizon. Free State Geduld Mines; reflected light, oil immersion,  $\times 500$ . *H*, Pyrite aggregate consisting of differently shaped microspherical structures. Note that along the edge of the pyrite aggregate some of the globules are distinctly abraded, which suggests some transport of the aggregate. Free State Geduld Mines, Basal Reef; reflected light,  $\times 500$ .

reasons, only those samples could be used that had an unusually high content of one of the various types of pyrites. Suitable samples were so scarce that, although more than 100 polished sections were investigated, only 5 were of samples that could be used for the concentration of detrital pyrites. The selected samples were crushed, sieved, and cleaned in a water stream. Following this, the concentrates were obtained by heavy-liquid separation and by handpicking the detrital grains under the binocular microscope according to their distinct morphological features.

The much larger grain sizes of the authigenic Witwatersrand pyrites, notably the concretionary buckshot pyrites, facilitated concentration and allowed a greater number of samples to be investigated.

Following these concentration and separation processes, all the pyrite samples were treated with 20 percent HCl and again cleaned ultrasonically in distilled water until free of visible turbidity. Polished sections were then made from about one-half of the concentrate, and their purity was examined under the ore microscope. From these examinations, it was estimated that the

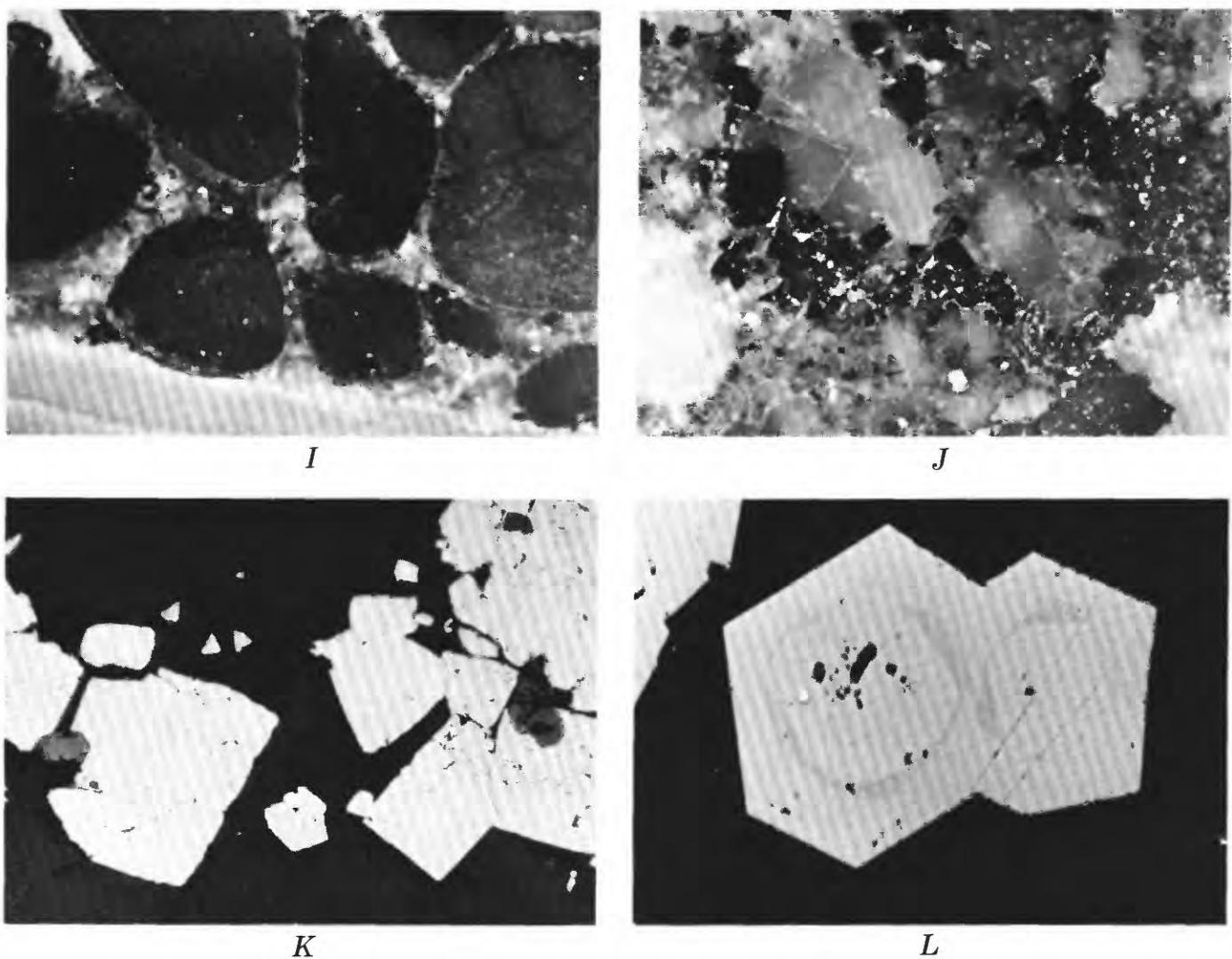


FIGURE 3.—Continued. *I*, Photograph of a polished slab, showing the footwall contact of the B-Reef horizon with underlying ore-free shales. The sample shows an unusually rich concentration of large concretionary buckshot pyrites. Free State Geduld Mines;  $\times 5$ . *J*, Photograph of a polished slab from the Basal Reef horizon. This sample contains predominantly idiomorphic authigenic pyrites in a quartz-pebble conglomerate. Free State Geduld Mines;  $\times 5$ . *K*, Microphotograph of the authigenic pyrites shown in figure 3 *J*. Note the small detrital chromite grains (dark grey), which are partly overgrown by the reconstituted pyrite formed in situ (white). Free State Geduld Mines; reflected light, oil immersion,  $\times 350$ . *L*, Zoned pyrite grains from a primary gold deposit in the Barberton Mountain Land. Sheba Queen Mine; reflected light, oil immersion,  $\times 500$ .

contamination of the pyrite concentrates by silicate minerals or other sulfides was less than 5 percent by volume and that, for the Witwatersrand samples, the separation of the authigenic from the allogenic pyrite variety was between 80 and 90 percent successful.

#### ANALYTICAL PROCEDURE

The copper, zinc, lead, cobalt, nickel, and silver contents were determined with a Perkin Elmer Atomic Absorption Spectrophotometer at the University of Heidelberg, and a number of control analyses were carried out

by an outside laboratory (Metallgesellschaft, Frankfurt) using photometric and polarographic methods. The limits of detection were 10 ppm for zinc and silver, 20 ppm for copper, cobalt, and nickel, and 50 ppm for lead. The precisions (two standard deviations) range between 10 and 30 percent.

The selection of the elements under investigation was based mainly on the following considerations:

1. Copper, zinc, and lead occur only in restricted amounts as substitutions in the lattice of most natural pyrites (Boyle, 1965; Radcliffe and Mc-

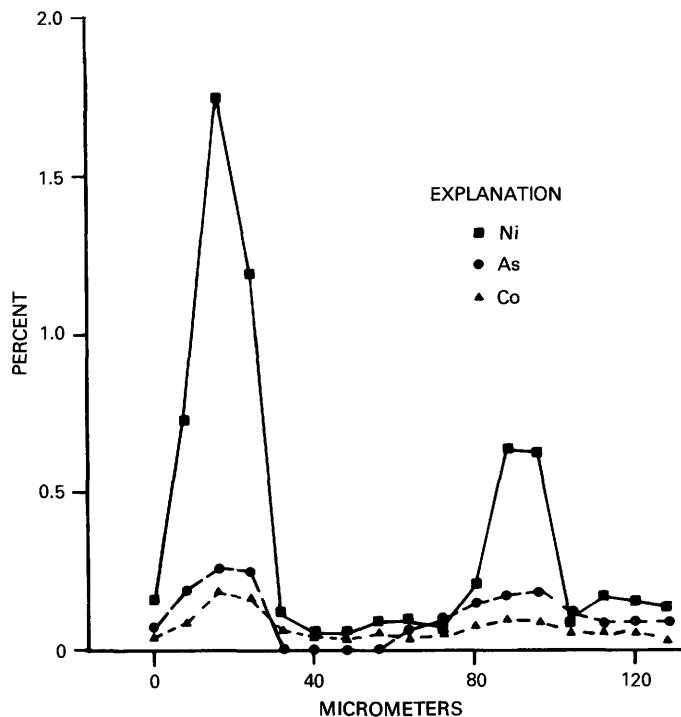


FIGURE 4.—Electron-microprobe step-scanning profiles across a zoned pyrite from the Eagles Nest gold mine in the Barberton Mountain Land.

Ween, 1969 and 1970; Scott and Barnes, 1972; and others). The observed trace contents of these elements thus indicate the extent to which the investigated pyrites were contaminated by microscopic to submicroscopic inclusions of base-metal sulfides—for example chalcopyrite, sphalerite, and galena, as the most common ones.

2. Nickel and cobalt, in contrast, enter the pyrite structure with ease (Kullerud and others, 1962; Ramdohr, 1969; and others), and since nickel-cobalt sulfides occur rarely in the investigated ore samples (minute pentlandite exsolutions in pyrrhotite), one

can assume that the nickel and cobalt values obtained indicate the overall availability of these two elements during the formation of the pyrites.

3. Silver occurs almost exclusively in solid solution with gold. Silver-bearing sulfides have been found only as a few grains in a galena aggregate from the Basal Reef in the Witwatersrand deposit (Saager, 1968). The silver values, therefore, serve as excellent indicators for the amount of gold present as inclusions in the pyrite. Primary gold inclusions in detrital Witwatersrand pyrites contain approximately 10 percent silver (Saager, 1969); and for the gold grains in primary ores of the Barberton Mountain Land, silver contents of 0.2-21.6 percent have been reported by Liebenberg (1973).

## DISCUSSION OF RESULTS

According to the mode of formation and the origin of the sampled pyrites, the geochemical data were divided into the following three groups (see also table 3):

Group 1 (G1), Authigenic Witwatersrand pyrites  
 Group 2 (G2), Allogenic Witwatersrand pyrites  
 Group 3 (G3), Greenstone-belt pyrites

The silver content of samples in Group 1 ranged from 0 to 390 ppm; in Group 2, from 0 to 10 ppm; and in Group 3, from 0 to 380 ppm. The silver content in most samples, however, lies below the limit of detection for the analytical method used (10 ppm), and consequently silver is not considered in the statistical analysis.

For cobalt, nickel, copper, lead, zinc, and each of the three groups of samples, the sample cumulative frequency distribution was constructed (fig. 5). With the aid of nonparametric Kolmogorov-Smirnov statistic the various cumulative frequency distributions were compared, and, for each element, the null hypothesis was tested that the three sample frequency distributions (the three postulated pyrite groups) were drawn from populations having the same frequency distribution.

TABLE 3.—Grouped results of trace-element analyses

[ $\bar{x}$ , mean;  $s$ , standard deviation;  $C$ , coefficient of variation]

Group		Element				
		Co	Ni	Cu	Zn	Pb
G1 (8 samples), authigenic Witwatersrand pyrites:						
$\bar{x}$	ppm	1,301	1,347	251	56	963
$s$	ppm	932	490	109	30	1,209
$C$		.71	.36	.43	.54	1.07
G2 (5 samples), allogenic Witwatersrand pyrites:						
$\bar{x}$	ppm	216	450	118	64	135
$s$	ppm	88	112	63	21	69
$C$		.41	.25	.53	.31	.51
G3 (44 samples), greenstone-belt pyrites:						
$\bar{x}$	ppm	198	514	341	151	123
$s$	ppm	195	532	456	379	73
$C$		.98	1.04	1.13	2.51	.59

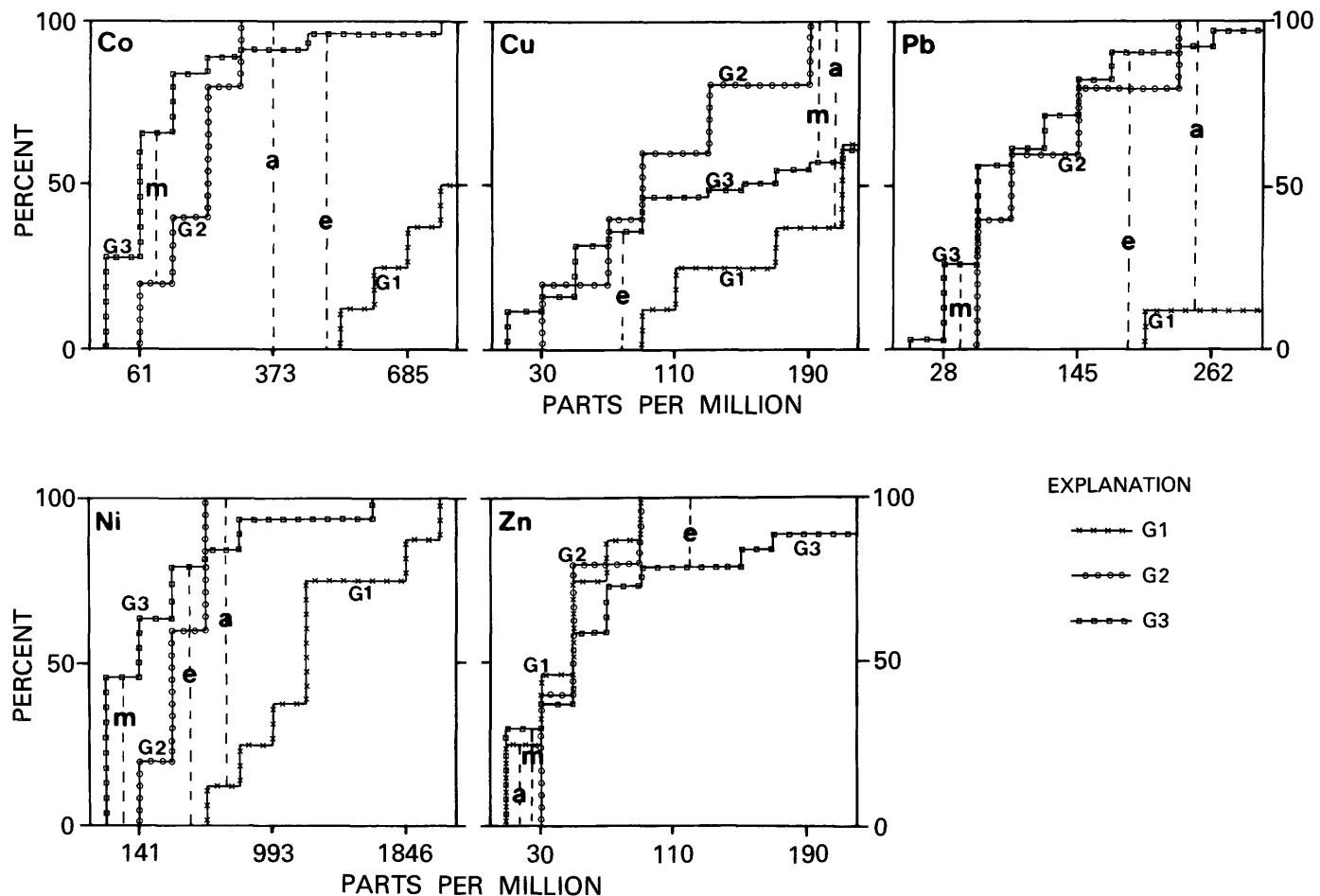


FIGURE 5.—Cumulative frequency distributions of trace-element values from the grouped pyrite samples, and graphical solutions of Kolmogorov-Smirnov statistic. G1, authigenic Witwatersrand pyrites; G2, allogenic Witwatersrand pyrites; G3, greenstone-belt pyrites (primary).  $d_n$ , maximum vertical deviations between the sample frequencies: a,  $d_{G1-G2}$ ; m,  $d_{G2-G3}$ ; e,  $d_{G3-G1}$ .

The Kolmogorov-Smirnov statistic (Smirnov, 1948; Dixon and Massey, 1951; Miller and Kahn, 1965) was used because no assumptions are needed about the normality of the distributions and because it is not subject to the very small sample limitation of chi-square. The test is a graphic one, and the maximum vertical deviation ( $d_n$ ) between the sample cumulative frequencies is directly measured on the graph (see fig. 5). The obtained  $d_n$  values are then compared with values extracted from tables, and the null hypothesis is either accepted or rejected at a certain level of significance.

The results of the test are shown in table 4. At a 0.05 level of significance the measured maximum deviations ( $d_n$ ) do not in all cases exceed the values required to say that the investigated sample groups were drawn from different populations. It follows that the proposed grouping of the samples is not always geochemically significant. This is particularly the case for the allogenic Witwatersrand pyrites and the greenstone-belt pyrites

(G2-G3). At a 95 percent level of probability—with respect to all five investigated elements—the samples were drawn from the same population. It thus can be assumed that both the allogenic Witwatersrand pyrites and the pyrites from mineralizations of the greenstone belts were formed in similar geological environments under similar chemical and physical conditions. These results support the source hypothesis of Viljoen and others (1969) for an origin of the detrital Witwatersrand pyrites from mineralizations in the Archean greenstone terranes of the Kaapvaal Craton.

With respect to the other pairs of pyrite groups (G1-G2, G1-G3)—that is, authigenic Witwatersrand pyrites and allogenic Witwatersrand pyrites—on the one side, and authigenic Witwatersrand pyrites and greenstone-belt pyrites on the other side, the results are not as distinct and uniform (fig. 5; table 4). For the elements cobalt, nickel, and lead the maximum deviations ( $d_n$ ) exceed the required values, and the postulated

TABLE 4.—*Results of the Kolmogorov-Smirnov statistics*  
 [Minimum deviation required at level: G1-G2,  $\text{sign.}_{0.05} = 0.700$ ; G2-G3,  $\text{sign.}_{0.05} = 0.580$ ;  
 G1-G3,  $\text{sign.}_{0.05} = 0.590$  (sign., significance)]

Element	Pyrite groups		
	G1-G2	G2-G3	G1-G3
Cobalt	$d_{G1-G2} = 1.000$ $H_o: G1 \neq G2$	$d_{G2-G3} = 0.475$ $H_o: G2 = G3$	$d_{G1-G3} = 0.955$ $H_o: G1 \neq G3$
Nickel	$d_{G1-G2} = .875$ $H_o: G1 \neq G2$	$d_{G2-G3} = .455$ $H_o: G2 = G3$	$d_{G1-G3} = .785$ $H_o: G1 \neq G3$
Copper	$d_{G1-G2} = .625$ $H_o: G1 = G2$	$d_{G2-G3} = .425$ $H_o: G2 = G3$	$d_{G1-G3} = .355$ $H_o: G1 = G3$
Zinc	$d_{G1-G2} = .250$ $H_o: G1 = G2$	$d_{G2-G3} = .290$ $H_o: G2 = G3$	$d_{G1-G3} = .210$ $H_o: G1 = G3$
Lead	$d_{G1-G2} = .875$ $H_o: G1 \neq G2$	$d_{G2-G3} = .270$ $H_o: G2 = G3$	$d_{G1-G3} = .920$ $H_o: G1 \neq G3$

sample groupings seem to be justified. The distribution of the elements copper and zinc, however, deviate very little, and, at a 0.05 level, it can be assumed that the samples were drawn from the same population. The proposed grouping, with respect to copper and zinc, seems to be invalid.

These contradictory results require further comment:

1. The copper and zinc trace contents of the pyrites, as explained, have been caused by inclusions of base metal sulfides and, thus, are an indication of the contamination of the pyrite by other sulfides. Inclusions of chalcopyrite and sphalerite have been observed irregularly distributed in the pyrites of all three postulated sample groups. They are primary inclusions, which formed as mechanical admixtures (Ramdohr, 1969) during pyrite crystallization, or are infiltrated secondary inclusions, which migrated into the marginal areas of porous pyrites during metamorphic periods (see also section on pyrite mineralogy). Both types of inclusions are not characteristic of a specific pyrite group, and this is reflected in the Kolmogorov-Smirnov test.
2. Cobalt and nickel in pyrite show a different crystal-chemical behaviour, occurring in solid solution. The trace contents of these two elements are therefore an indication of the availability of cobalt and nickel during pyrite formation. It follows that these two elements represent better indicators for the study of the metallogenic origin of the investigated pyrites.

Carstens (1941), Hegemann (1943), Cambel and Jarkovský (1967), and other workers have stated that sedimentary and hydrothermal pyrites possess different and characteristic cobalt and nickel trends. Sedimentary pyrites have low Co/Ni ratios and rather constant cobalt and nickel contents of about 10 ppm and 200 ppm, respectively. In contrast, hydrothermal pyrites have more erratic and considerably larger cobalt and nickel contents (several thousand parts per million) and Co/Ni ratios that are greater than 0.1 and may be as high as 830 (Berg and Friedensburg, 1944). Results of

work by Hawley and Nichol (1961) indicate that pyrites from magmatic ores commonly show higher cobalt and nickel contents than do pyrites from hydrothermal ores, and Wilson (1953), on the basis of relative availability, concluded that pyrite derived from early magmatic melts would have a higher nickel content, and probably a lower Co/Ni ratio, than would pyrite formed from late fluids.

Recent investigations (Tourtelot, 1964; Roscoe, 1965; Saager, 1965; Loftus-Hills, 1967; and others) suggest that the foregoing criteria are only valid in a generalized way and cannot be used to distinguish between different types of ore deposits. The studies indicate, however, that in many cases pyrites from specific occurrences or metallogenic provinces exhibit characteristic and persistent cobalt and nickel trends, a conclusion that agrees with the present study and that also explains why the elements cobalt and nickel yielded the best results in the Kolmogorov-Smirnov test.

3. The extremely high lead contents obtained (between 230 and 3,400 ppm) are significantly restricted to the authigenic Witwatersrand pyrites (see table 3) because the generally very porous pyrites quite commonly have metamorphically infiltrated galena inclusions in their marginal zones, and it can be assumed that the lead in these inclusions is of radiogenic origin (Saager, 1970; Burger and others, 1962).

The allochthonous Witwatersrand pyrites, which are very compact (Saager, 1970) and practically free of galena inclusions of infiltrated radiogenic lead, have lead contents of less than 240 ppm. Similar low lead contents were found in the pyrites from the greenstone belts, which have been formed in a geological environment characteristically devoid of lead-bearing minerals (Villiers, 1957; Saager and Koeppel, 1976). It is for these reasons that the lead values yielded the same statistical trend as displayed by the cobalt and nickel data, in spite of the fact that the crystal-chemical behaviour of lead is markedly different from that of nickel and cobalt.

## FACTOR ANALYSIS

To further investigate the trace-element data of the pyrites and to test the results of the graphic Kolmogorov-Smirnov statistic, a factor analysis of the data was carried out. Factor analysis is a branch of multivariate statistics that recently has found increasing application in various fields of the Earth sciences (Imbrie and van Andel, 1964; Nichol and others, 1969; Saager and Sinclair, 1974; and others). The primary aim of the analysis is to investigate the intercorrelations

within a set of data and to find the smallest noncorrelated set of basic vectors that accounts for most of the variance in the original set of data. Factor analysis, therefore, tries to simplify a complex set of data by expressing it in a minimal number of theoretical variables or factors without significant loss of information. The factors, which are linear combinations of the original variables, must be interpreted in terms of all available geological and other information. For a detailed mathematical elucidation of factor analysis and its problems the reader is referred to the appropriate texts by Spearman (1904), Kaiser (1958), and Harman (1960).

Q-mode factor analysis as employed in this study examines the variation from sample to sample, and its objective is to find groups of samples that are similar in terms of their total composition. The observed variations from sample to sample are not independent but are dependent upon underlying causes. If these causes are the same for two samples, then the values measured in these samples will be similar. The measure of similarity used in the Q-mode analysis is the cosine "theta" of the angle between two sample vectors in  $n$ -dimensional space. All the cosines obtained yield the cosine "theta" or similarity matrix. It is analysed mathematically, and the principal components or factors are extracted by simultaneous consideration of all the sample similarities. Once the minimum number of factors required has been established, the factors are rotated according to specific mathematical criteria—in the present study, the varimax rotation—to aid interpretation of geological data.

The Q-mode factor analysis was carried out at the Computing Center of the University of Heidelberg, using a modified FORTRAN program written by J. D. S. Wilson, Department of Geology, University of British Columbia. The results of the analysis showed that a 2-factor model accounts for 69.8 percent of the total variance in the original pyrite data. The composition of the two factors, or end members, can be roughly determined by scanning down the columns of the factor-score matrix (table 5).

TABLE 5.—Varimax factor score matrix

Element	Factor 1	Factor 2
Cobalt	-1.0994	-0.3326
Nickel	-1.5330	.1660
Copper	.2406	-1.7544
Zinc	.0034	-1.3042
Lead	-1.1863	-.2646

Factor 1 is essentially a cobalt-nickel-lead factor accounting for 37.2 percent of the available information, and factor 2 is a copper-zinc factor accounting for 32.6 percent. This grouping into two factors corresponds

closely with the trend found by the Kolmogorov-Smirnov statistic (it shows that the behaviour of the elements cobalt, nickel, and lead is independent from that of the elements copper and zinc).

The factor loadings of each pyrite sample are the projection of each factor axis on each sample vector or, more simply, the composition of each sample with respect to the extracted 2-factor model (table 6). The factor loadings, as determined from the Q-mode factor matrix, have been plotted on a scatter diagram (fig. 6). The axes of the diagram represent the two factors, and the units on the axes are given in factor loadings. They vary between -1.0 and +1.0. The sum of the squared factor loadings of each sample gives the communality. This value indicates, for each sample, what proportion of the variance has been explained by the factor model used. The communality has a value of 1.0 if the particular sample has been explained at 100 percent by the extracted factors (table 6).

TABLE 6.—Partial Q-mode varimax factor matrix

Sample	Communality	Factor 1	Factor 2
B 24	0.7580	-0.5682	-0.6594
SW 8	.9632	.3311	.9239
SW 1	.9294	-.8541	.4472
EN 4	.3685	-.4252	.4333
EN 85	.8799	.4970	.7955
ST 29d	.6562	-.0182	.8099
SG 29	.8044	-.3615	.8205
SB 191	.2739	.4096	.3257
528	.8199	.3324	.8423
NO 3	.6467	.0690	.8012
25	.8154	.4806	.7645
BR 9	.5274	.6046	.4025
3.3	.9028	-.9500	-.0197
3.4	.5997	-.7398	-.2288
BS 184	.7320	-.8163	-.2562
2 HH	.9046	-.9473	-.0845
SB 231	.8854	.9278	-.1566
H231	.7985	.7922	-.4133
H 401	.2412	.2774	-.4052
120	.8199	.3324	.8423
F III	.4721	-.6855	-.0472
Variance, in percent		37.1910	32.6009
Cumulative variance, in percent		37.1910	69.7919

The scatter diagram (fig. 6) demonstrates the heterogeneous distribution of all the investigated trace elements in the greenstone-belt pyrites (see also table 3). These pyrites originated from a large number of genetically different deposits that occurred in quite different geological environments. The generally low cobalt, nickel, and lead contents of these samples are reflected in the weak but distinct accumulation of data points in the positive half of factor 1.

The alloigenic Witwatersrand pyrites show a rather similar, irregular distribution on the diagram. Only one

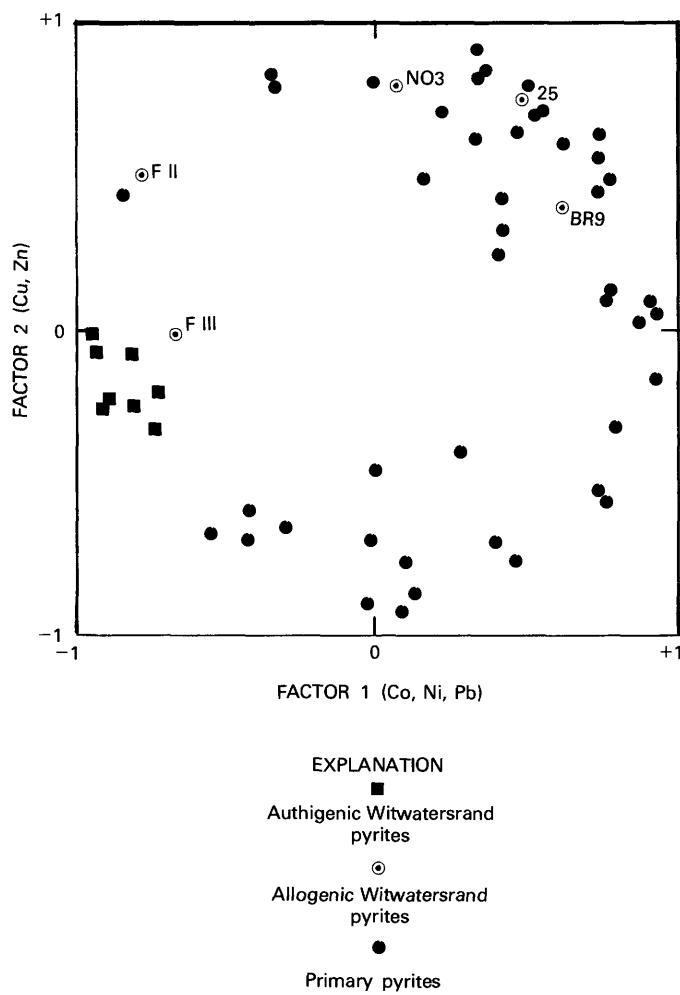


FIGURE 6.—Scatter diagram of the factor loading values.

sample, F III, exhibits a certain affinity to the data points of the authigenic Witwatersrand pyrites, possibly reflecting an imperfect separation of the two pyrite varieties during sample preparation.

The authigenic Witwatersrand pyrites, in contrast, are far more homogeneous and exhibit a distinct cluster (fig. 6). With respect to the system investigated, these samples possess a very high degree of similarity. As all authigenic Witwatersrand pyrites have comparatively high cobalt, nickel, and lead contents, they all plot close to the -1.0 value of factor 1. The high cobalt, nickel, and lead values can be regarded as an indication of the high availability of these elements in the Witwatersrand Basin where authigenic pyrite formed, possibly as late as the metamorphism of the sediments (Saager, 1970; Koeppel and Saager, 1974). Relatively high cobalt and nickel contents in the Witwatersrand sedimentary materials are in good agreement with the source model of Viljoen and others (1970), who postulated a granite-

greenstone source terrane that, in its lower members, is largely built up of mafic-to-ultramafic rocks. The high lead contents can be explained by the radioactive decay of the uranium- and thorium-bearing placer minerals in the Witwatersrand sediments.

Like the Kolmogorov-Smirnov test, the Q-mode factor analysis shows that the allogenic Witwatersrand pyrites and the greenstone-belt pyrites have geochemical similarities that clearly set them apart from the authigenic Witwatersrand pyrites. The investigation also indicates that pyrites formed in a sedimentary environment are far more homogeneous in trace-element contents than those originating from hydrothermal deposits. This observation agrees with the general trends of the trace-element contents in sedimentary and hydrothermal pyrites.

#### LEAD-ISOTOPE INVESTIGATION

As part of the same research project, various pyrite and galena samples from Witwatersrand reefs and from primary gold deposits in the major greenstone belts of the Kaapvaal Craton were investigated for their lead-isotope composition. The results of the study have been reported in detail in an earlier investigation (Koeppel and Saager, 1974). They bear additional important information on the provenance of the detrital pyrites in the Witwatersrand sediments and are therefore summarized below.

Uranium-thorium-lead geochronological dating of various sequences of the Witwatersrand Basin has been undertaken. It yielded for the Dominion Reef Sequence, at the base of the assemblage, a maximum sedimentary age of  $2,850 \pm 55$  m.y. (million years) (Allsopp, 1964) and for the Ventersdorp Sequence, which directly overlies the Witwatersrand Sequence, an age of  $2,300 \pm 100$  m.y. (Niekerk and Burger, 1966). Nicolaysen and others (1962), by means of uranium-lead investigations of rounded uraninite and monazite grains and of total conglomerate samples from the Dominion Reef sediments, showed that these minerals have a crystallization age of  $3,080 \pm 100$  m.y., which is consistent with the hypothesis of a detrital origin of the uraninite and monazite. The uranium minerals apparently were reconstituted and underwent a first complete lead loss 2,040 m.y. ago; a second lead loss occurred in recent times (Wetherill, 1956; Nicolaysen and others, 1962). Lead-isotope studies carried out on galena samples from various reefs of the Witwatersrand deposits by Burger and others (1962) yielded independent evidence for old detrital components approximately 3,000 m.y. old in the sedimentary filling of the basin. The investigation furthermore confirmed a period of lead loss 2,040 m.y. ago. This alteration period coincides with the emplacement of the Bushveld Granite at Houtbeck (Nicolaysen and others,

1958), representing a period of mild regional metamorphism of the Witwatersrand sediments (the solution stage of the modified placer theory).

The results of the lead-isotope study of Koeppel and Saager (1974) are condensed on a lead-lead diagram (fig. 7). On this diagram a hypothetical field is delineated in which all the data points of the authigenic Witwatersrand minerals should plot. The construction of the field necessitated the following assumptions:

1. The crystallization age of the uranium-bearing minerals in the sediments is 3,100 m.y.
2. Only two periods of lead loss occurred, the first one 2,040 m.y. ago and the second one in recent times.

The upper boundary of the field is then given by the secondary isochrone due to the first lead loss of the 3,100 m.y. old uranium-lead system. This isochrone has a slope of 0.3834 and corresponds to the "main trend" of Burger and others (1962). The lower boundary of the field, owing to the second lead loss, starts at the lower end of the upper boundary and has a slope of 0.124, which is the present-day  $^{207}\text{Pb}/^{206}\text{Pb}$  ratio of a uranium-lead system 2,040 m.y. old. It is important to note that

all investigated authigenic Witwatersrand minerals of Burger and others (1962) and of Koeppel and Saager (1974) did plot within the hypothetical field. Only two data points, by the latter authors, are given because most samples contain large amounts of radiogenic leads and therefore plot outside of the diagram (fig. 7) on the open side of the wedge-shaped field.

The field of data points of sulfides from primary gold ores in the greenstone belts was experimentally determined (Saager and Koeppel, 1976). It does not completely overlap the field of authigenic Witwatersrand minerals.

The results of sample 25 in this study are of special interest. No. 25 is a detrital-pyrite sample originating from the Basal Reef horizon of the Free State Geduld Mine in the Orange Free State Goldfield. It has a lead-isotope composition that on the diagram (fig. 7) plots outside the predicted field of authigenic minerals. Thus, the lead-isotope composition of this sample concurs with the rounded, waterworn shapes of the pyrites and, in fact, confirms their detrital origin from a source outside the Witwatersrand Basin.

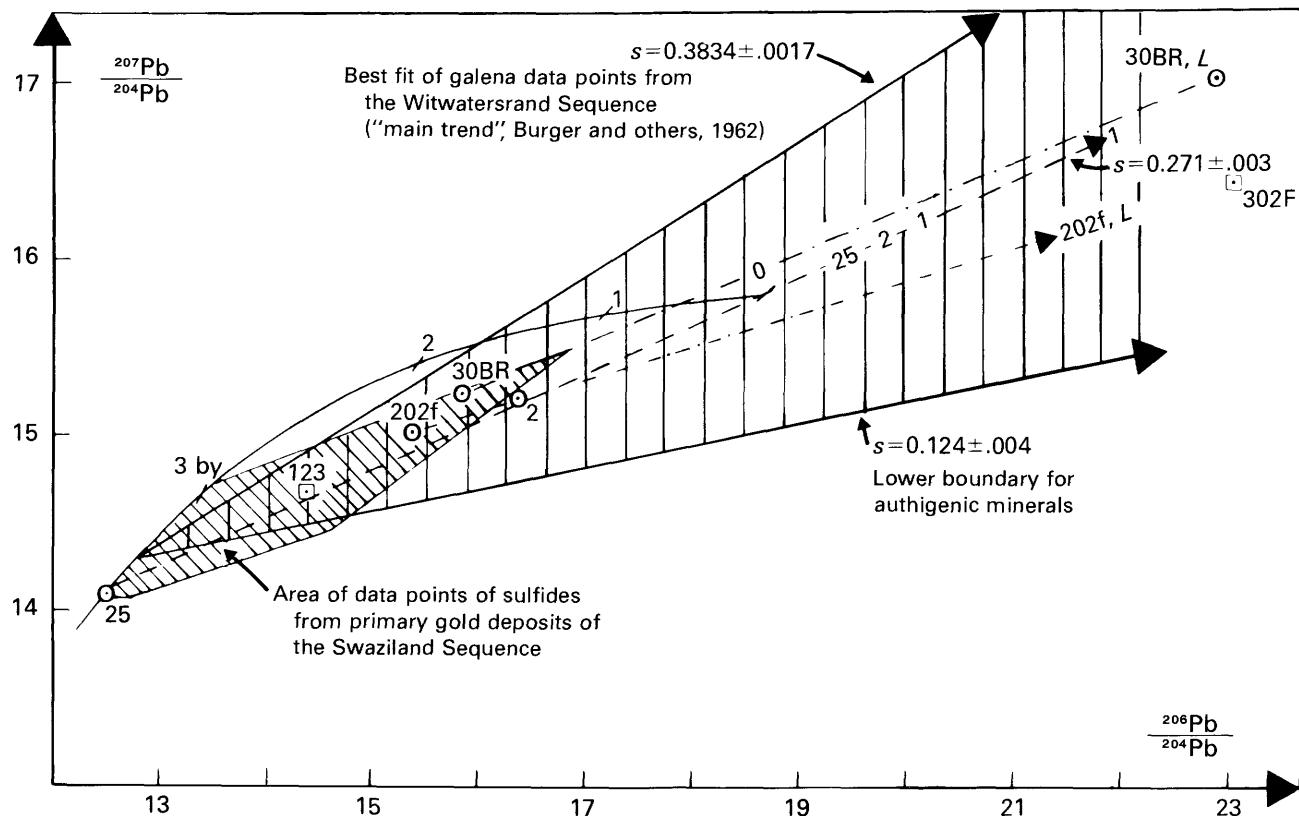


FIGURE 7. – Diagram showing  $^{207}\text{Pb}/^{204}\text{Pb}$  versus  $^{206}\text{Pb}/^{204}\text{Pb}$ . The field of authigenic Witwatersrand minerals is vertically hatched; the field of sulfides from primary mineralizations in the Archean greenstone belts is diagonally hatched. Circles denote allogenic pyrites; squares, authigenic pyrites from the Witwatersrand sediments.  $L$  denotes an HCl leach of the respective sample;  $s$ , slope of the line (modified from Koeppl and Saager, 1974, p. 327). The following parameters were used for calculating the growth curve:  $(^{206}\text{Pb}/^{204}\text{Pb})_0 = 9.56$ ;  $(^{207}\text{Pb}/^{204}\text{Pb})_0 = 10.42$ ;  $T_0 = 4.55$ ;  $^{238}\text{U}/^{204}\text{Pb} = 9.0$ ;  $\lambda_{238} = 1.537 \times 10^{-10} \text{ yr}^{-1}$ ;  $\lambda_{235} = 9.722 \times 10^{-10} \text{ yr}^{-1}$ .

The lead-isotope composition of sample 25 is identical with that observed in galena from the Rosetta Mine, a primary gold lode deposit located in Onverwacht Group rocks of the Barberton Mountain Land. A remarkable lead line was also obtained from the three detrital-pyrite samples 25, 2, and 1 (sample 1 lies outside of the graph), which were collected at the same locality in the Basal Reef. The samples contain all the same first-stage lead indicative of an origin from a lead source similar to that found in the Archean greenstone terrane. The tieline of the data points (fig. 7) has a slope of 0.271, which corresponds with the present-day  $^{207}\text{Pb}/^{206}\text{Pb}$  ratio of a uranium-lead system 3,360 m.y. old. Slopes yielding such ages were commonly observed in pyrite samples from greenstone belts by Saager and Koeppel (1976) and also in whole rock samples from the Onverwacht Group (Sinha, 1972). Significantly, the age of 3,360 m.y. is identical with the minimum age of the volcanic rocks of the Upper Onverwacht Group of the Barberton Mountain Land as reported by Niekerk and Burger (1969).

Samples 202f and 30 BR are also detrital pyrite samples from the Klerksdorp and the Orange Free State Goldfields, respectively. They exhibit lead-isotope compositions similar to those observed in sulfides from primary gold mineralizations in the greenstone belts, but also their data points fall into the predicted field of authigenic Witwatersrand minerals. However, it is conspicuous that the slopes of the tielines to the respective HCl leaches of the two allochthonous pyrite samples are steeper than analogous tielines of the authigenic Witwatersrand minerals. The hypothetical first-stage lead of these detrital pyrites resembles that of galenas from primary deposits in the greenstone belts (fig. 7; see also Ulrych and others, 1967).

## SUMMARY AND CONCLUSIONS

1. Compact, rounded pyrite grains from the Witwatersrand sediments and pyrites from primary gold deposits in greenstone terranes of the Swaziland Sequence exhibit identical heterogeneous trace-element contents. At a high significance level their cobalt, nickel, and lead values were drawn from populations having the same distribution. It follows that both pyrite varieties were formed under similar chemical and physical conditions in similar geological environments and that the compact, rounded Witwatersrand pyrites are in fact of detrital origin.
2. The large, often extremely porous Witwatersrand pyrites that were formed in the basin of deposition possess remarkably homogeneous trace-element values and conspicuously high lead, nickel, and cobalt contents. These trends clearly differentiate

this pyrite variety from the other pyrites investigated and suggest a different mode of formation, probably during sedimentation of Witwatersrand strata.

3. Although comparatively few pyrite samples have been analyzed for their lead-isotope composition, the results agree with the trends observed in the trace-element investigations. The isotope study provides evidence that the detrital Witwatersrand pyrites derive from a source area with a lead-isotope composition identical with that of the greenstone belts of the Swaziland Sequence. The observed lead-isotope compositions, furthermore, indicate that the detrital Witwatersrand pyrites were formed much earlier than the Witwatersrand sediments.
4. The provenance areas of the Witwatersrand sediments, as demonstrated by sedimentological and geological evidence, lay to the north and west and close to the rim of the basin. The provenance areas had far-reaching similarities and were of the same age as the Barberton Mountain Land – a typical Archean granite-greenstone terrane – used to explain the succession of the sediments and the heavy minerals in the Witwatersrand Basin. However, it must be noted that the Barberton Mountain Land, which lies some 300 km east of the Witwatersrand deposits, can only be regarded as a model for the source terrane and that the actual provenance areas lay closer to the basin but are now eroded away and (or) covered by younger strata.
5. Pyrite and gold are intimately associated in the primary and the fossil placer-gold province. It can be assumed that not only the detrital Witwatersrand pyrites but also the Witwatersrand gold have originated from pyritic and often gold-rich deposits in the greenstone belts of the Swaziland Sequence. The gold deposits are genetically related to ultramafic and mafic, often magnesium-rich, volcanics, which are exceptionally abundant in the Lower Onverwacht Group at the base of the Swaziland Sequence. It is possible that the larger portion of the detrital sulfides came from the acid volcanics of the Upper Onverwacht Group.
6. The detrital uranium- and thorium-bearing minerals in the Witwatersrand sediments have a crystallization age of 3,040–3,160 m.y. This agrees well with the U-Pb and Rb-Sr ages reported for some of the granites in the basement of the Kaapvaal Craton. Because of the high thorium content of many uraninites in the Witwatersrand sediments, it has been suggested that they are derived from a granitic or pegmatitic source. According to the provenance model such source rocks form the highest

erosional levels in a typical Archean granite-greenstone terrane. The model thus provides an explanation of why no primary uranium mineralization has yet been found in the granite-greenstone terranes of the Kaapvaal Craton.

7. Two geochemically and petrogenetically different rock assemblages provided the sources for the gold and uranium in the Witwatersrand sediments. Both rock assemblages occupied different erosional levels and, depending upon the actual prevailing denudation and drainage conditions, varying amounts of gold and uranium were deposited in the basin. The expected trend from uranium-rich horizons at the bottom of the sedimentary sequence to gold-rich horizons at its top is still observable. Reworking of the sediments, weathering of material, and various other sedimentary processes have led to considerable deviations from this overall trend.

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# Geochemistry and Morphology of Mineral Components from the Fossil Gold and Uranium Placers of the Witwatersrand

*By* D. K. HALLBAUER

GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN  
QUARTZ-PEBBLE CONGLOMERATES

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## GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN QUARTZ-PEBBLE CONGLOMERATES

# GEOCHEMISTRY AND MORPHOLOGY OF MINERAL COMPONENTS FROM THE FOSSIL GOLD AND URANIUM PLACERS OF THE WITWATERSRAND

By D. K. HALLBAUER <sup>1</sup>

### ABSTRACT

Using the hydrofluoric-acid leaching technique to obtain heavy-mineral grains intact from Witwatersrand conglomerates and the scanning electron microscope with microanalyzer attachment, the technique of morphological microscopy was applied to observe the three dimensions of the grains and to study their composition.

In combination with stereo X-ray radiography of rock slabs and the application of various geochemical techniques, this afforded new information on the processes that led to the formation of Precambrian placer deposits.

### INTRODUCTION

For close to 100 years the Witwatersrand deposits have been studied by mineralogists and geologists, helped by the ever-increasing sophistication of methods and techniques. For some time now these deposits have been widely recognized as fossil placers. It can therefore be assumed that the concentration of gold, uranium, and other heavy minerals took place in large palaeoecological systems in which the distribution of these minerals was determined by the palaeoenvironment. The combination of modern and established mineralogical techniques can increase substantially the retrieval of information on these environments that is contained in rock parameters and rock components. The recognition and understanding of the environment in which a fossil placer was formed and of the relation between the various morphological forms and components of the deposit present a potential approach for use to determine economic horizons even if the economic minerals are absent in a particular small sample.

In previous mineralogical studies of the various minerals in such ores the classical methods of microscopy, using thin sections and polished sections of rock, permitted observation of two dimensions of a mineral grain. The study of rocks and ores by these methods was developed to a very advanced stage by great mineralogists such as Paul Ramdohr (1975), and the

knowledge of geological processes was enhanced greatly by them. The development of the scanning electron microscope with its high resolution (100 Å and better) and increased depth of focus, however, has opened up the third dimension of mineral grains to examination by microscopy. The use of the scanning electron microscope in combination with hydrofluoric-acid digestion method (Neuerburg, 1975), which produces concentrates of undamaged grains of many ore minerals, has made it possible in the past few years to advance further the knowledge of geological processes that led to the formation of the Witwatersrand and other deposits.

### METHODS OF STUDY

In order to be able to observe and study the morphology and micromorphology of minerals it is essential to free the mineral grains from the matrix rock. Depending on the type of matrix, agents such as hydrochloric acid, sulphuric acid, or hydrofluoric acid can be used to dissolve the matrix and liberate the grains to be examined. Softer, porous rocks can often be split into their components by the crystallization pressure of common alum ( $KAl(SO_4)_2$ ). Repeated soaking of the rock in a solution of alum and slow drying causes the rock to disaggregate into its components. After having been washed, the mineral particles can be studied.

For the study of heavy minerals in Precambrian rocks of the Witwatersrand and similar deposits, the siliceous matrix and the quartz pebbles were decomposed in hydrofluoric acid. This process requires about six weeks of continuous acid treatment to liberate the heavy minerals from about 100–200 g of uncrushed rock. Many heavy minerals are insoluble in hydrofluoric acid and can be recovered in their natural shape (Neuerburg, 1975). Tests have shown so far that gold, pyrite, zircon, and chromite are not attacked or altered by hydrofluoric acid, but that uraninite, apatite, monazite, and magnetite are.

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Of the major silicates present in the Witwatersrand rocks, only pyrophyllite is not dissolved in hydrofluoric acid. Fine-grained pyrophyllite, if present in large amounts, can be a disadvantage, but repeated, careful washing of the sample can remove most of it. The inertness of pyrophyllite in hydrofluoric acid led, on the other hand, to the discovery of pyrophyllite pebbles containing fine-grained crystalline gold, providing an important contribution to knowledge of the origin of the Witwatersrand deposits.

After being cleaned in water and acetone, the mineral

grains are ready for examination. Heavy fractions can be obtained by heavy-liquid separation or by using the micropanner (Muller, 1959), and individual grains are selected for microscopy and mounted on an aluminium holder to be placed in the scanning electron microscope, a Leitz AMR 1000. The mounting medium may be conducting silver paint or colloidal graphite for larger grains or double-sided cellotape for small grains. Non-conducting specimens must be coated with carbon, gold, or palladium.

The scanning electron microscope is used widely in

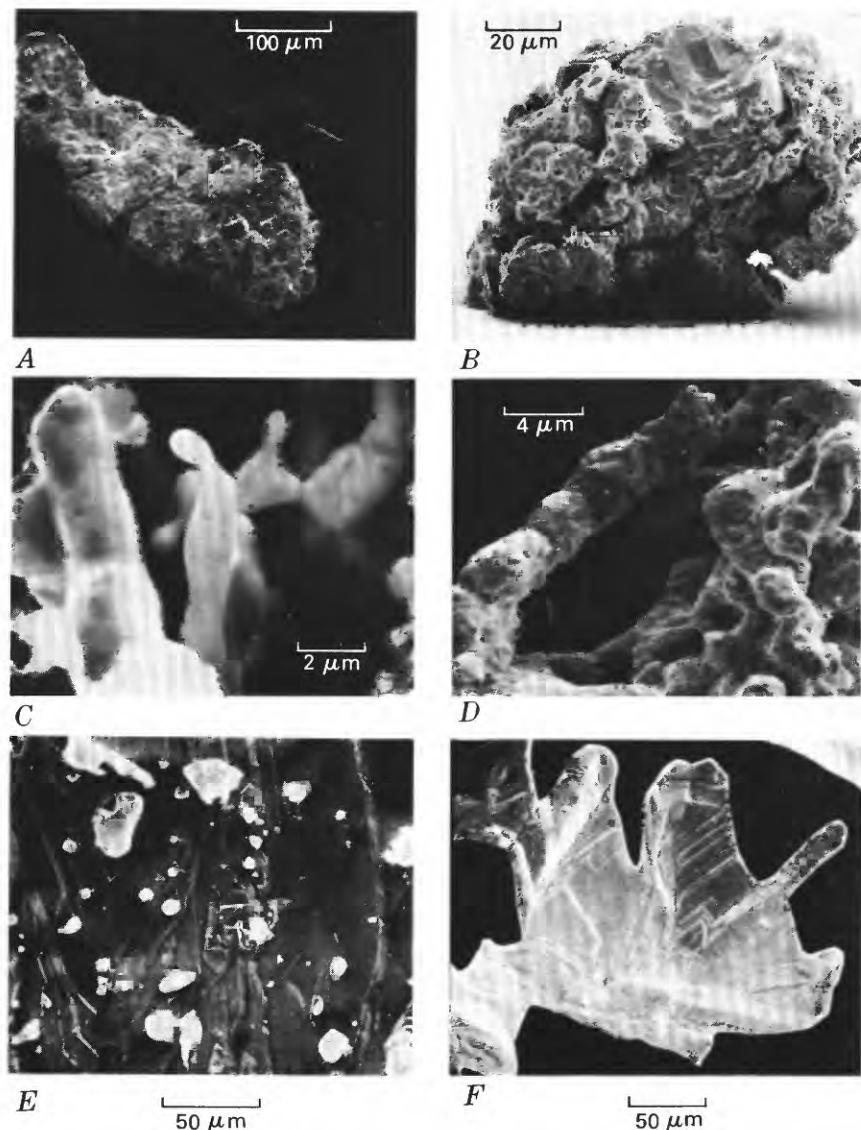


FIGURE 1. Scanning electron photomicrographs of the three major types of gold particles from Witwatersrand reefs. A, Detrital, flaky gold particle, indicating a medium to long transport distance (40–60 km). B, Slightly distorted, detrital gold particle with partially preserved crystalline texture, indicating a transport distance of up to 30 km. C, Filamentous biochemical gold from the interior of carbonaceous matter. D, Filamentous gold from the interior of carbonaceous matter, showing cell-like structures. E, Oriented growth of secondary gold on pyrite. F, Recrystallised gold from small fissures in quartzite.

biological, physical, and mineralogical research (Johan, 1974; Stoops, 1970; Metzger and others, 1977), as well as in other fields. For the type of investigation described here it was greatly advantageous to have an attached ORTEC microanalyser on the AMR 1000 to exploit the energy-dispersive X-ray spectra of the particles examined and to obtain a semiquantitative analysis of the samples and, in particular, to measure the fineness of gold particles.

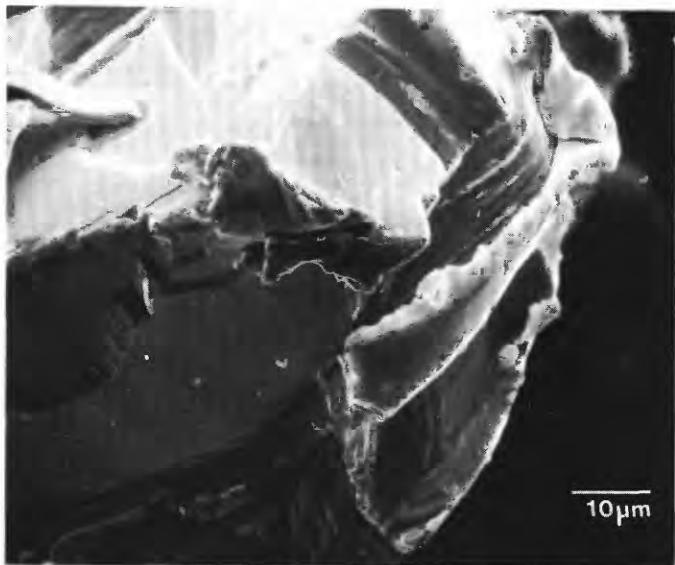


FIGURE 2.—Scanning electron photomicrograph of part of a crystalline gold particle from a Witwatersrand reef showing abrasion and distortion on protruding corners and edges, indicative of a short transport distance (up to 10 km).

### THE COMPONENTS OF WITWATERSRAND CONGLOMERATES

#### GOLD PARTICLES

Gold is present in the Witwatersrand reefs as discrete particles of various shapes and sizes. From studies of the morphology of the individual gold particles it has been possible to classify the particles into three major groups: detrital gold, biochemically concentrated gold, and recrystallised gold (fig. 1).

#### DETTRITAL GOLD

During transport of primary gold particles, their typical crystalline microtexture (fig. 2) is deformed slowly by repeated collisions with the bedload in the fluvial system until the crystalline texture is destroyed and replaced by a distorted and flaky microtexture (fig. 3). In a comparison with gold from recent alluvial gold deposits it was possible to show that the shapes of gold particles undergo characteristic changes during transport down the slope of a placer deposit.

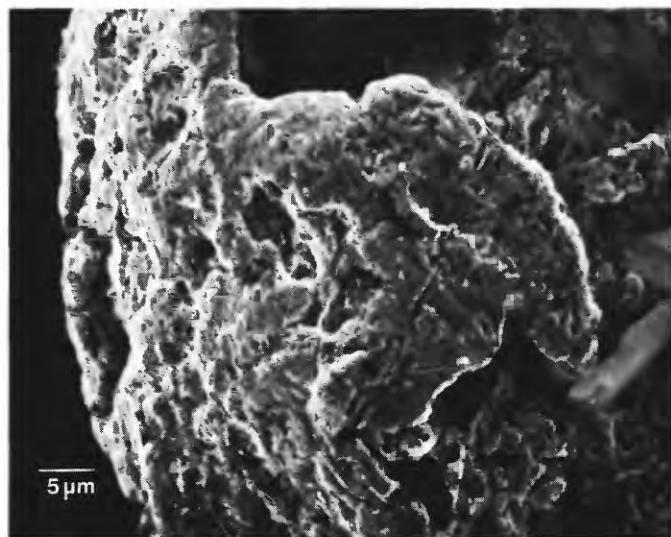


FIGURE 3.—Scanning electron photomicrograph of part of a gold particle from a Witwatersrand reef exhibiting a severely distorted surface typical for transport distances in excess of 30 km.

Thus the morphology of detrital gold should reflect the conditions of transport, distance of transport and, when observed on a lateral scale, the direction of transport (Hallbauer and Utter, 1977). From a large number of observations it was then concluded that the average transport distance for Witwatersrand gold was about 30 km.

All transitional shapes, ranging from crystalline to completely deformed, have been observed in gold particles recovered from Witwatersrand deposits. It should be possible, therefore, to reconstruct, from a few strategically selected small samples of gold-bearing conglomerate, the major outlines of a fossil placer, including the general transport direction.

In a study conducted along these lines, consideration was given to the abundance of gold particles with crystalline texture, and from this it was predicted that a separate entry point existed in the northern part of the Orange Free State goldfields (Hallbauer and Utter, 1977). This was confirmed by independent sedimentological investigations (Minter, 1977). Observations on some of the other heavy minerals accompanying the gold supported the findings of the gold-morphology studies.

It is of interest to note that gold particles recovered by the hydrofluoric-acid leaching method from samples originating from other fossil placers, such as the Dominion Reef, Klerksdorp, and the Blind River Conglomerate of the Elliot Lake District, Canada, showed typical detrital features indicating a transport mechanism similar to that identified for rocks of the Upper Witwatersrand Series.

The fineness— $F = (Au \times 1,000)/(Au + Ag)$ —of Witwatersrand gold was previously known to vary between

different localities and stratigraphic horizons (Viljoen, 1971; Saager, 1973; Schidlowski, 1968a). No definite trends, however, could be established, as the number of grains analysed was comparatively small. Analysis of a larger number of gold particles not only gave the statistical distribution within a given sample but provided reliable information on lateral trends within a deposit and differences between stratigraphic horizons.

Under recent transport conditions gold is rapidly depleted of its silver content, owing to the oxidising conditions in most modern fluvial environments. It can be shown (Hallbauer and Utter, 1977) that the gold-silver alloy is stable during fluvial transport in an oxygen-deficient atmosphere, a condition commonly assumed for the Precambrian.

The fineness of the gold in Witwatersrand deposits should consequently reflect the fineness of the primary deposits. It is therefore not surprising that many conglomerate horizons have a characteristic fineness that does not vary within one geological unit (Hallbauer, Utter, and Hirdes, 1978; Utter, 1977). Facies changes, reworking of older horizons, and other geological parameters can thus be "read off" the fineness of gold particles (Utter, 1977). Geochemical maps showing the distribution of gold fineness can thus be used successfully to supplement sedimentological work.

Geological observations in the past have shown that the Ventersdorp Contact Reef in the Klerksdorp Goldfield truncates a number of older horizons. Consequently, the fineness of gold particles from the Ventersdorp Contact Reef showed a spread of fineness values from 750 to 960 (fig. 4), which reflects the fineness of gold from the truncated horizons (Utter, 1977).

The bimodal distribution of fineness for the Vaal Reef (fig. 4) can be correlated to the sedimentological evidence that predicted two facies on the Vaal Reef horizon (Minter, 1976).

In contrast to these observations, the fineness of other conglomerate units showed a remarkable unimodal distribution, pointing to one source of gold for these units (fig. 4).

Grain-size determinations, which were carried out simultaneously with the fineness measurements, showed that no relationship exists between the size of gold particles and the fineness.

#### BIOCHEMICALLY CONCENTRATED GOLD

The second type of gold, the biochemically concentrated gold, is associated genetically with layers of carbonaceous matter. The association between high gold values and carbonaceous matter in some Witwatersrand reefs is well known. It has been possible in the past few years to show that the carbon seams are the fossilized

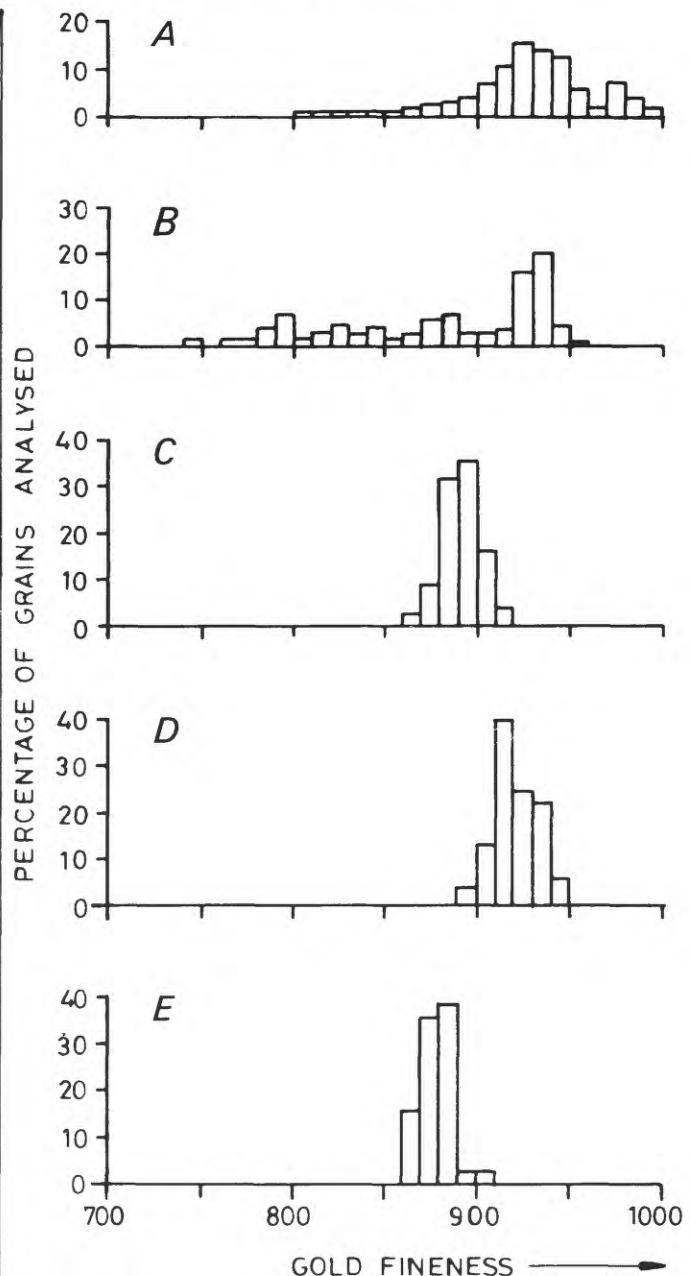


FIGURE 4.—Fineness of gold from various reefs in the Witwatersrand area. A, Vaal Reef, Klerksdorp Goldfield (Utter, 1978). B, Ventersdorp Contact Reef, Klerksdorp Goldfield (Utter, 1978). C, Basal Reef, St. Helena gold mine. D, Carbon Leader Reef, Blyvooruitzicht gold mine. E, Ventersdorp Contact Reef, East Driefontein gold mine.

remains of Precambrian plants (Hallbauer and van Warmelo, 1974; Hallbauer, 1975); the most common plant represented by columnar carbonaceous material was named *Thuchomyces lichenoides* (Hallbauer and others, 1977). Colonies of these plants formed mats or carpets, which trapped detrital gold by a corduroy-tablelike action (fig. 5); this gold was apparently then dissolved and redistributed either intracellularly or ex-



FIGURE 5.—Photograph of a carbonaceous layer, showing trapped detrital gold on the surface and filamentous gold within the carbonaceous matter, Basal Reef, St. Helena gold mine.

tracellularly by the organisms. From this process there originated the biochemical gold, which is present in the carbonaceous material as small filaments, agglutinated aggregates of filaments (fig. 1C, D) or irregularly formed, often perforated, particles.

#### RECRYSTALLISED GOLD

The recrystallised gold apparently was remobilised and transported for short distances of the order of a few millimeters within the consolidated sediment and has only limited, local significance. Transportation and mobilization could be attributed to local rises in the temperatures caused by dyke intrusions, overlying volcanic rock, or hydrothermal activity along faults and fracture planes.

Recrystallised gold generally occurs as crystalline aggregates in small quartz veins or in thin fractures and as orientated growth on secondary pyrite (fig. 1E, F).

#### DETritAL GOLD INTERGROWTHS

Of special interest in samples of Witwatersrand conglomerates are detrital particles consisting of intergrowths of gold and other minerals and of pyrite and other detrital minerals. From their detrital nature, these can be regarded as primary intergrowths, and they serve as indicators of the types of the provenance deposits. In addition, a large concentration of these particles can serve to indicate proximity to the entry point to the basin.

In samples of Basal Reef, primary intergrowths of sphalerite and gold and of sphalerite and chalcopyrite, as well as the gold contents of sphalerite (about 250 ppm (parts per million)) and of chalcopyrite (about 6,000 ppm), point to a strong genetic relation between gold, sphalerite, and chalcopyrite. Typical of detrital gold-sphalerite intergrowths (fig. 6C) are the etched surface of the sphalerite and the distorted and abraded edges of

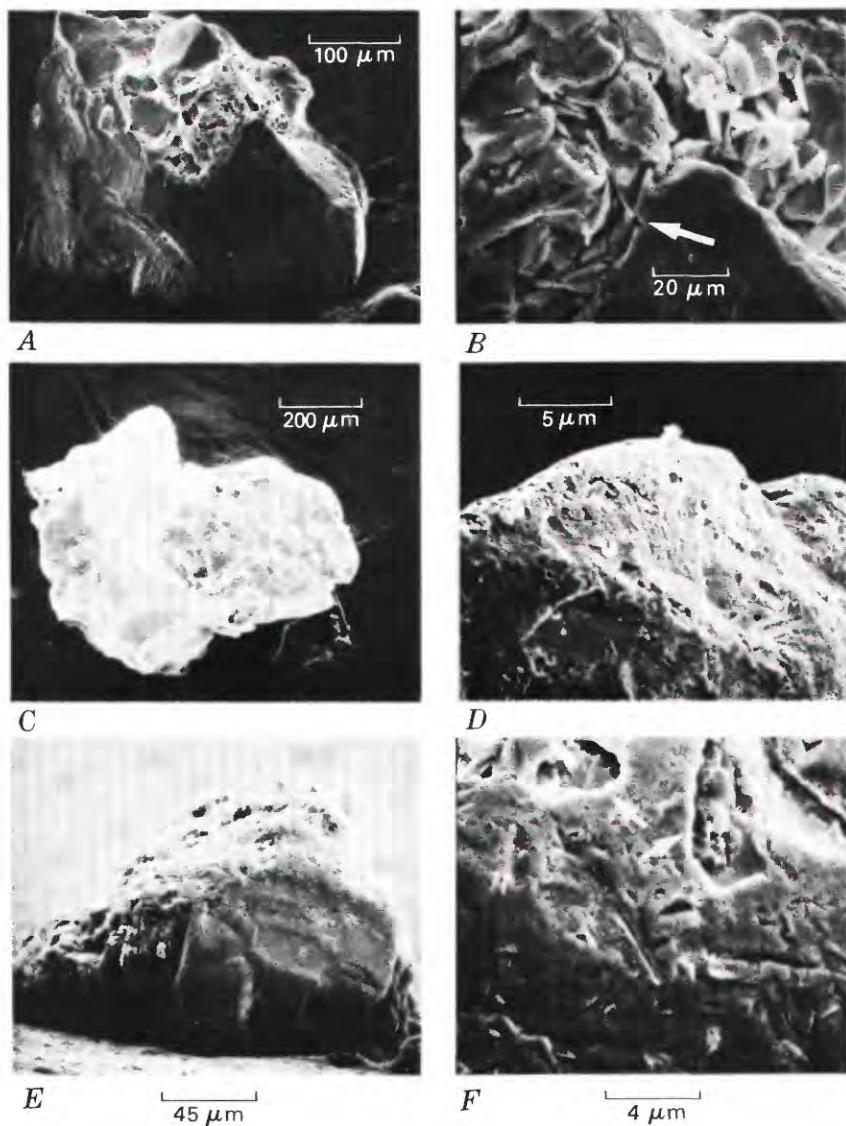


FIGURE 6.—Scanning electron photomicrographs of some intergrowths between gold and other primary minerals. *A*, Primary intergrowth of detrital pyrite and molybdenite from a sample of Ventersdorp Contact Reef, East Driefontein gold mine. *B*, Surface details of *A*, showing an aggregate of molybdenite flakes attached to the pyrite surface but partially displaced from the original position (arrow). *C*, Detrital particle of gold-sphalerite intergrowth from a sample of Basal Reef, St. Helena gold mine, Orange Free State, with gold on the left partly surrounding a piece of sphalerite on the right. *D*, Detail of *C*, showing the surface of the gold distorted along the edges during alluvial transport. *E*, Side view of a slightly rounded particle consisting of an intergrowth of gold and gersdorffite from a sample of *B* Reef, Lorraine gold mine, Orange Free State. *F*, Surface detail of *E*, showing the scratched and distorted gold surface with small gersdorffite inclusions (arrows).

the gold (Fig. 6*D*). Primary intergrowths of gold and cobaltite-gersdorffite were observed in samples of *B* Reef from the Lorraine gold mine (fig. 6*E, F*). In these samples, detrital gold particles with low overall distortion are a common feature; they are accompanied by pyrite and smaller amounts of molybdenite, tourmaline, and other heavy minerals, as well as various amounts of pyrophyllite in the form of small pebbles and fine-grained material.

In samples of Ventersdorp Contact Reef from the East Driefontein gold mine, primary intergrowths of pyrite and molybdenite (fig. 6*A, B*), gold and bravoite, and pyrophyllite pebbles and fine-grained gold were observed. As the detrital pyrite from this deposit contains small inclusions of gold (average gold content 250 ppm) that can be regarded as primary, a common origin can be assumed for gold, pyrite, molybdenite, bravoite, and pyrophyllite. Pyrophyllite is regarded as the

hydrothermal alteration product of potassium feldspar (Deer, Howie, and Zussman, 1962) and is a major constituent of the sericite-quartz pyrite zone of porphyry-type ore deposits (Hollister, 1975). This suggests, therefore, that some primary deposits of the Witwatersrand gold were perhaps similar to porphyry copper deposits or mineralised acid volcanics and not to a greenstone terrain as found in the Barberton Mountain Land (Viljoen and others, 1970). Concurrent geological considerations led to the formation of a similar model supporting the findings of the studies using scanning electron microscopy (Button, A., University of the Witwatersrand, Economic Geology Research Unit, personal commun., 1977).

A recent search in the vicinity of the Klerksdorp Goldfield succeeded in finding small occurrences of mineralised acid volcanics that are older than the Witwatersrand rocks and that contain minute quantities of free gold and are accompanied by pyrite, cobaltite and other minerals typical of Witwatersrand conglomerates.

#### CARBONACEOUS MATTER

Prominent in a number of conglomerate horizons in the Upper Witwatersrand and also locally in the Dominion Reef are layers of a coal-like material called "carbon" or "thucholite." A recent investigation into its organic geochemistry has shown that it is in fact a kerogen similar to bituminous coal and considered to be the result of polymerization of biochemicals from decayed primitive Precambrian micro-organisms (Zumberge and others, 1978). The majority of the pyrolysis breakdown products of this kerogen are alkyl-substituted aromatic hydrocarbons, low molecular weight aliphatic hydrocarbons, and some aromatic sulphur and aliphatic oxygen compounds. The evolution of the kerogen from the original biochemicals of primitive micro-organisms appears to have progressed through free radical reactions influenced by a constant radiation from the accompanying finely dispersed uraninite.

Although some of the structures of the carbon in the carbon seams of the Witwatersrand deposits had been observed in polished sections and in thin sections (Snyman, 1968), recognition of this carbon as the fossilized remains of Precambrian plants and the full reconstruction of the internal structures of these plants were made possible only by scanning electron microscopy, while the spatial arrangement relative to rock component was studied by X-ray radiography of slabs of conglomerate.

Most of the smaller seams of carbonaceous matter prominently display a columnar structure. Stereo X-ray radiographs in particular show that these small seams consist of a carpetlike arrangement of vertical columns

about 0.5 mm in diameter and as much as 10 mm long. Between the columns and on top of the layer are heavy minerals, including gold, pyrite, zircon, and others (fig. 7), which points to a mechanism of concentration similar to a corduroy-table.

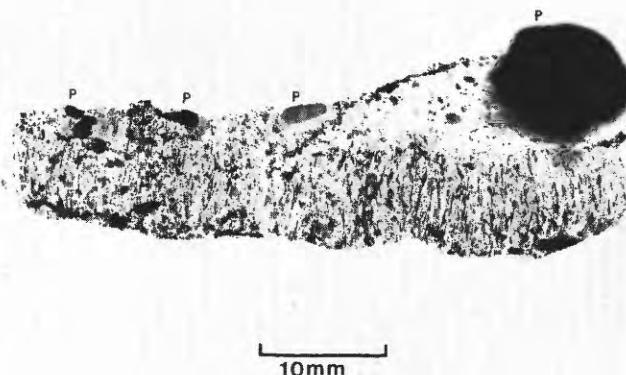


FIGURE 7.—X-ray radiograph of a carbonaceous layer with overlying quartzite showing small gold filaments (black) in the carbon and irregularly formed gold particles (black) in the upper portion of the layer and in the quartzite. Pyrite grains (P) can be distinguished from the gold by their lighter grey colour. Middle Elsburg Reef, Cook II gold mine.

Thin sections of such layers show finely laminated quartz as filling between the columns, while coarse-grained quartz and pebbles at the top form the transition to the overlying conglomerate (fig. 8A). The finely laminated quartz, which in places contains single filaments of an obvious biological nature (fig. 8B), can be interpreted as silicious sediment deposited during quiet periods.

Various sedimentological investigations and geological observations point to an environment comprising small stagnant pools, lagoons, wet sand, and clay on the lower slopes of wet alluvial fans or shallow water along the shore lines of an inland lake in which this Precambrian plant life flourished (Minter, 1976; Pretorius, 1975).

The more-or-less constant shape of the columnar form all over the Witwatersrand can be taken as an indication that it is the primary shape of a small plant or unit of a plant colony.

Some of the interior structure of individual columns is revealed in X-ray radiographs taken of thin slices on high-resolution plates. These radiographs when magnified show distinctly a vertical arrangement of filaments (fig. 9). Rather more pronounced is this on columns after low-temperature ashing (fig. 10). Even with a group of

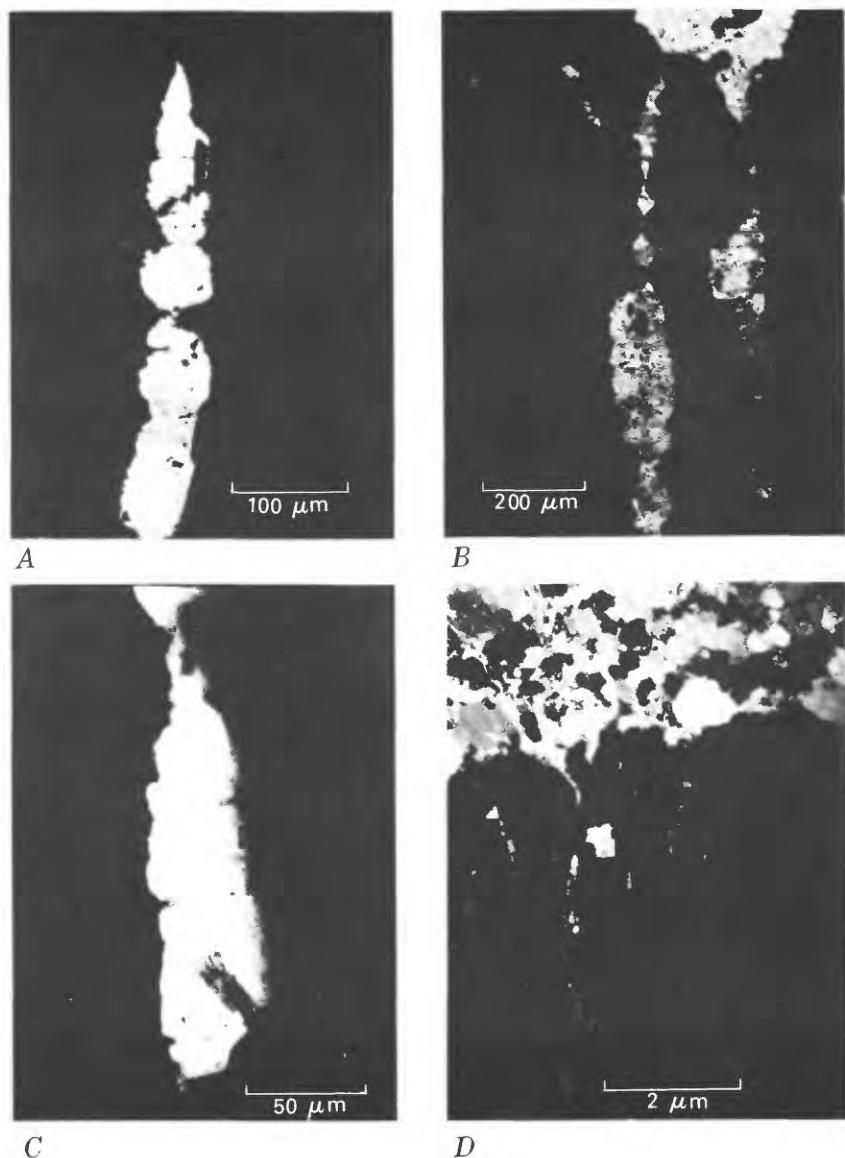


FIGURE 8.—Photomicrographs *A*–*D* showing thin sections of carbonaceous matter. The thin sections show the finely layered nature of quartz within the columnar carbon and grainy texture of the detrital quartz on top of the carbon (*D*), as well as a filament of a biological nature protruding from the carbonaceous matter (*C*), are shown. Photomicrograph *E* (a color reproduction of *C*) shows algal-like filaments of biological origin protruding from carbonaceous matter. Carbon Leader Reef, Blyvooruitzicht gold mine.



FIGURE 8.—(See facing page for explanation.)

Additional support for the biological nature of the carbonaceous or kerogen layers is obtained from gold filaments within the columns. While most of the filamentous structure in the columns consists of an almost constant mixture of uranium and lead, with small amounts of silicon, alumina, and iron, the number of gold filaments varies from sample to sample, and gold filaments are absent in some. Common to all gold contained within the kerogen is a shape composed of irregular, often perforated, particles and filaments (fig. 11A-D) with a form very closely resembling inorganic encrustations found on living micro-organisms (Hallbauer, 1975, 1978). It is therefore assumed that the Precambrian organisms in the Witwatersrand conglomerates collected gold directly by biochemical processes or indirectly from solutions and deposited this gold extracellularly on the living or perhaps on the decayed filaments.

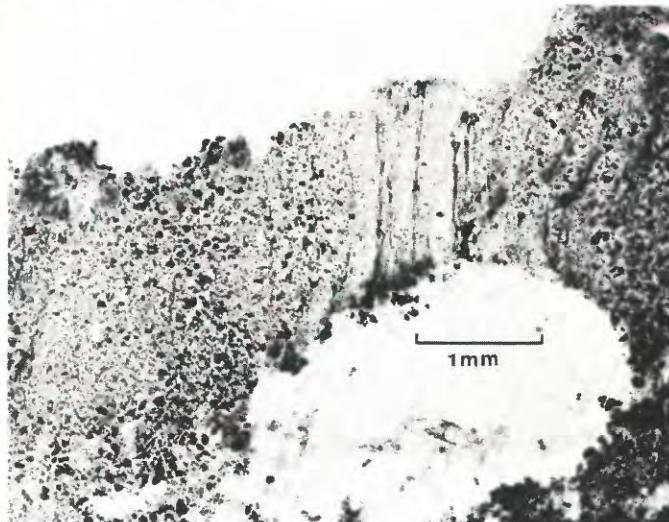


FIGURE 9.—Magnified X-ray radiograph of columnar carbonaceous matter, taken on a high-resolution plate and magnified to show the filamentous interior structure (white, rounded part is quartz). Carbon Leader Reef, Blyvooruitzicht gold mine.

columns, each individual is clearly defined by a thin, membrane-like structure surrounding it. Although most of the filaments when viewed in a light microscope do not show biological features, occasional filaments with typical cell structures were observed (Hallbauer, 1975). This is somewhat surprising when considering the age of these deposits (approx 2,700 m.y. (million years)). The low metamorphism (lower greenschist facies) and a relatively quiet tectonic history of the Witwatersrand rocks, however, might be some influencing factors in the preservation of these structures.

In addition to the biological structures mentioned, filaments resembling actinomycetes or related organisms have been observed in samples from the St. Helena gold mine (Hallbauer, 1975; Hallbauer, Jahns, and Beltman, 1977). They appear to be replacements of organisms by lead and zinc sulphide. Although these filaments are well preserved (fig. 12A, B) and readily recognized by microbiologists as bacterial or fungal filaments, they are a localized occurrence and appear to be of a parasitical nature. Their acceptance or nonacceptance does not influence the theory on the biological origin of the Witwatersrand carbonaceous matter or kerogen.

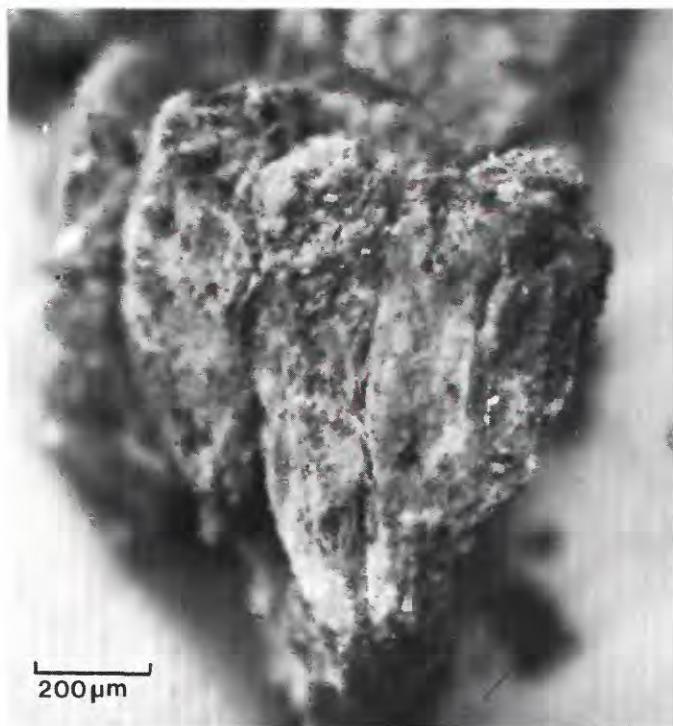


FIGURE 10.—Scanning electron photomicrograph showing structural details of columnar carbonaceous matter after oxidizing of kerogen at 500°C. (Oxidizing in a plasma furnace (International Plasma Corp.) at temperatures less than 100°C leads to the same type of structures.)

#### URANINITE

Uraninite was recognized early as a detrital component of the Witwatersrand conglomerates (Liebenberg, 1955; Ramdohr, 1955). Of special interest, however, is the association between uraninite and the carbonaceous matter. Uraninite can be found as small (<100  $\mu\text{m}$ ), irregular grains finely dispersed in carbonaceous matter and as rounded, detrital grains completely surrounded or in various degrees intergrown with the latter. Often the surrounding carbonaceous matter has the appearance of a felt of small filaments (Schidlowski, 1968b). Detailed observations on particles cleaned in hydrofluoric acid appear to indicate that microorganisms attacked detrital uraninite and slowly assimilated the mineral matter.

#### PYRITE

The detrital nature of most pyrites in the Witwatersrand conglomerates was recognized long ago from macroscopic observations and studies of polished sections (Ramdohr, 1955; Liebenberg, 1955), and as many as nine different types of pyrite identified by ore microscopy have been described in detail (Schidlowski, 1970; Saager, 1973). These various types can, however, be classified into three major groups: (1) allogenic

detrital pyrite; (2) pyrite formed under sedimentary conditions shortly before deposition; and (3) authigenic pyrite formed under various conditions in the solidified sediment.

Allogenic detrital pyrite is present in samples of Witwatersrand and similar conglomerates as massive, non-porous particles that exhibit all stages of rounding, the result of fluvial transport. Typical of the Witwatersrand pyrites are inclusions of various minerals—such as pyrrhotite, chalcopyrite, sphalerite, gold, and others (Utter, 1977)—that are similar to those found in primary deposits. The allogenic detrital pyrite is, therefore, most likely associated genetically with detrital gold.

Low chemical stability of pyrite in recent fluvial deposits has been taken to be an indication of an absence of oxygen in the Precambrian atmosphere (Cloud, 1976; Schidlowski and Junge, 1973). This is an assumption of considerable importance to studies of the evolution of life (Margulis and others, 1976). Although the presence of detrital pyrite in recent rivers has been reported repeatedly, little is known about periods of time for which pyrite can survive in a fluvial environment under the present oxygen partial pressure. Observations on recent detrital pyrite from some Eastern Transvaal, South Africa, rivers have revealed numerous orientated etched pits on the surface of the pyrite grains. A similar microsurface texture can be observed on some pyrite particles from Witwatersrand reefs and from the similar Blind River, Canada, conglomerates. If these textures still represent those of the original particle surfaces, it can be assumed that there was a low-level chemical attack on the surface during prolonged transport of the particle and, therefore, that a low oxygen partial pressure existed in the Precambrian atmosphere.

Laboratory experiments were conducted during which pyrite was tumbled in closed glass containers filled half with oxygen-free water and half with artificial atmospheres or normal air. After 30 days of continuous tumbling the  $\text{SO}_4^{2-}$  content of the water and the weight loss of the pyrite were determined. The results show that the pyrite lost 0.01 percent of its weight by tumbling in water under the present atmosphere. This weight loss was reduced to about 0.002 percent if the atmosphere had an oxygen content of 2 percent, or one tenth of the present (fig. 13). These results appear to indicate that a low-level chemical attack on pyrite is possible under conditions assumed to have existed during the Precambrian (Grandstaff, this volume, chap. C).

Within the group of sedimentary pyrite types, the pyrite "mud balls" are of particular importance as indicators of the nature of the palaeoenvironment. These porous, rounded pyrite particles consist of fine-grained pyrite aggregate and are known as "buckshot pyrites," a term often used to describe all rounded pyrite grains;

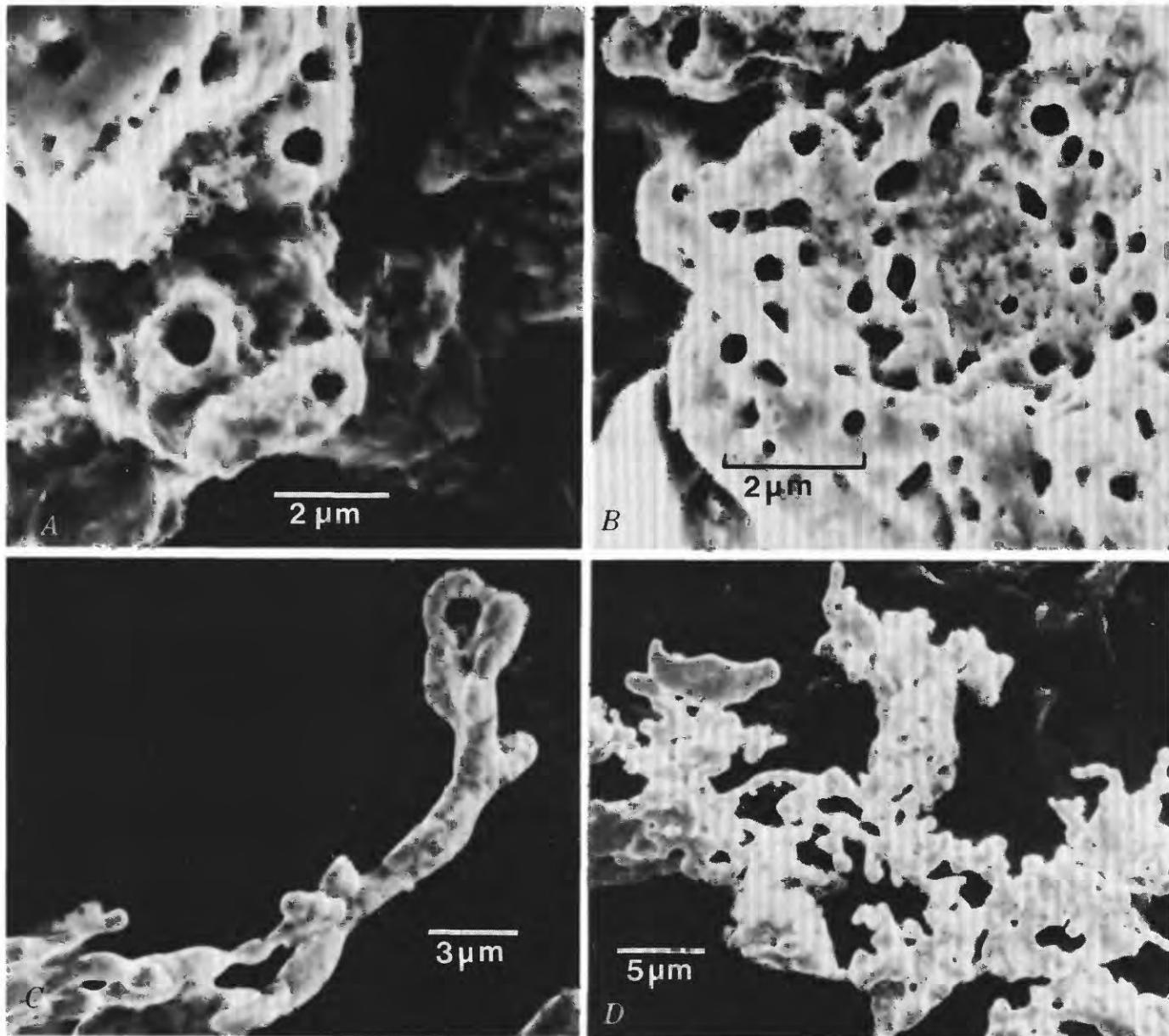


FIGURE 11.—Scanning electron photomicrographs showing examples of biochemically concentrated gold extracted from carbonaceous matter.

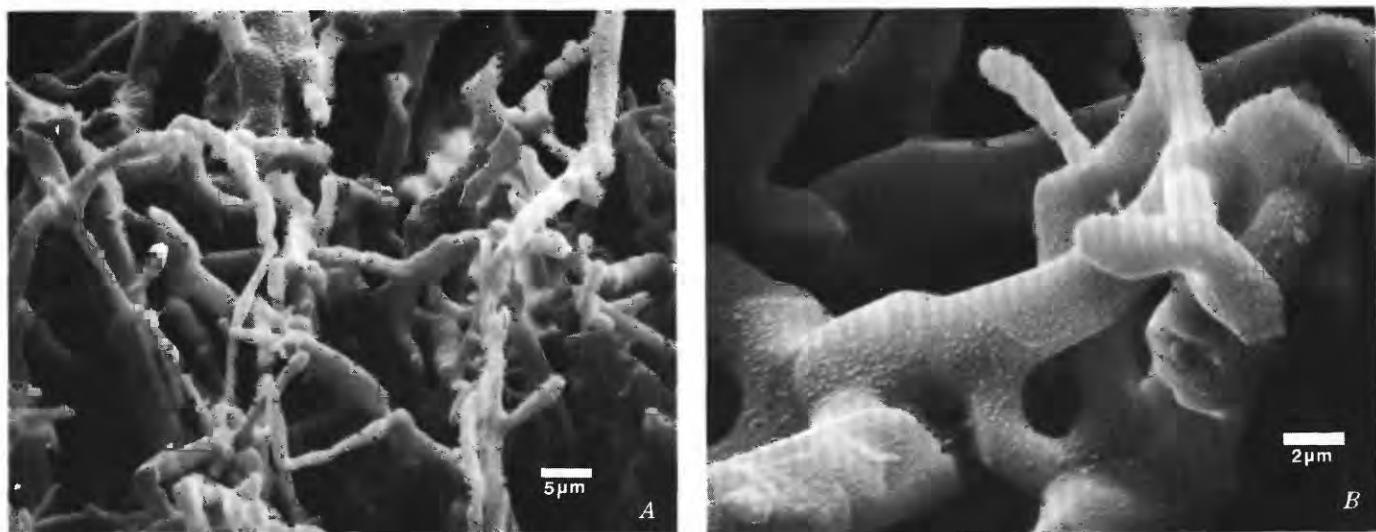


FIGURE 12.—Scanning electron photomicrographs showing filaments from carbonaceous matter where bacterial or fungal structures have been replaced and preserved by sphalerite and galena. Basal Reef, St. Helena gold mine.

they can occur in sizes ranging from 0.1 to 10 mm in diameter and are indicative of economic horizons in the Witwatersrand sequence. Polished sections of this type of pyrite occasionally show cracks and indentations, which previously were regarded as having been caused by solution under pressure (Schidlowski and Trurnit, 1966). However, three-dimensional examination of these cracks on the intact grain after treatment with hydrofluoric acid suggested that they were caused by shrinkage and (or) plastic deformation of soft material (fig. 14). From this it was deduced that pyritic mud was formed during Witwatersrand time and was deposited in pools and puddles of an ephemeral-stream system; flooding after a dry period would then produce mud pebbles and mud balls, as have been described for other fluvial environments (Ball, 1940; McKee and others, 1967). It was further deduced that during a subsequent dry period, shrinkage cracks as observed (Hallbauer, 1977) would form on the surfaces of the mud balls and overlying pebbles would cause the formation of indentations in, and plastic deformation of, the mud pebbles.

The occurrence of pyrite mud balls in conjunction with ventifacts in Witwatersrand conglomerates points to a palaeoenvironment characterised by desert conditions interrupted by frequent flooding of the area. Stereo X-ray radiographs of rock slabs showing mud flake structures in pyrite layers support this interpretation (Hallbauer, 1974).

Pyrite mud balls generally contain small fragments of most other detrital minerals, as is shown by the study of polished sections and trace-element analyses (Utter, 1977). Rounded pyrite particles with typical mud-ball

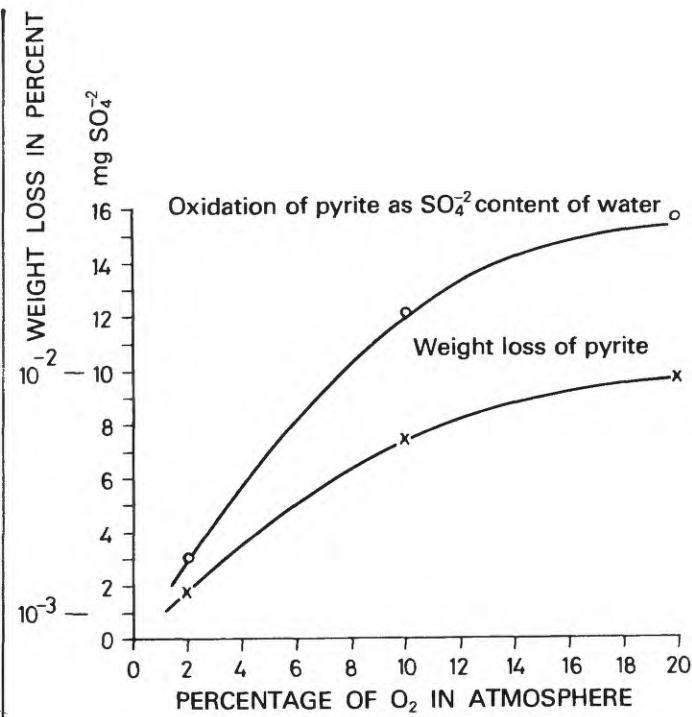


FIGURE 13.—Stability of pyrite in atmospheres with various oxygen contents.

structures have been observed in most Witwatersrand conglomerates, including those from the Vredefort area and the Dominion Reef.

Authigenic pyrite in fossil placers was formed in the rock during and after diagenesis and is easily recognized by its crystalline morphology. It has been observed that

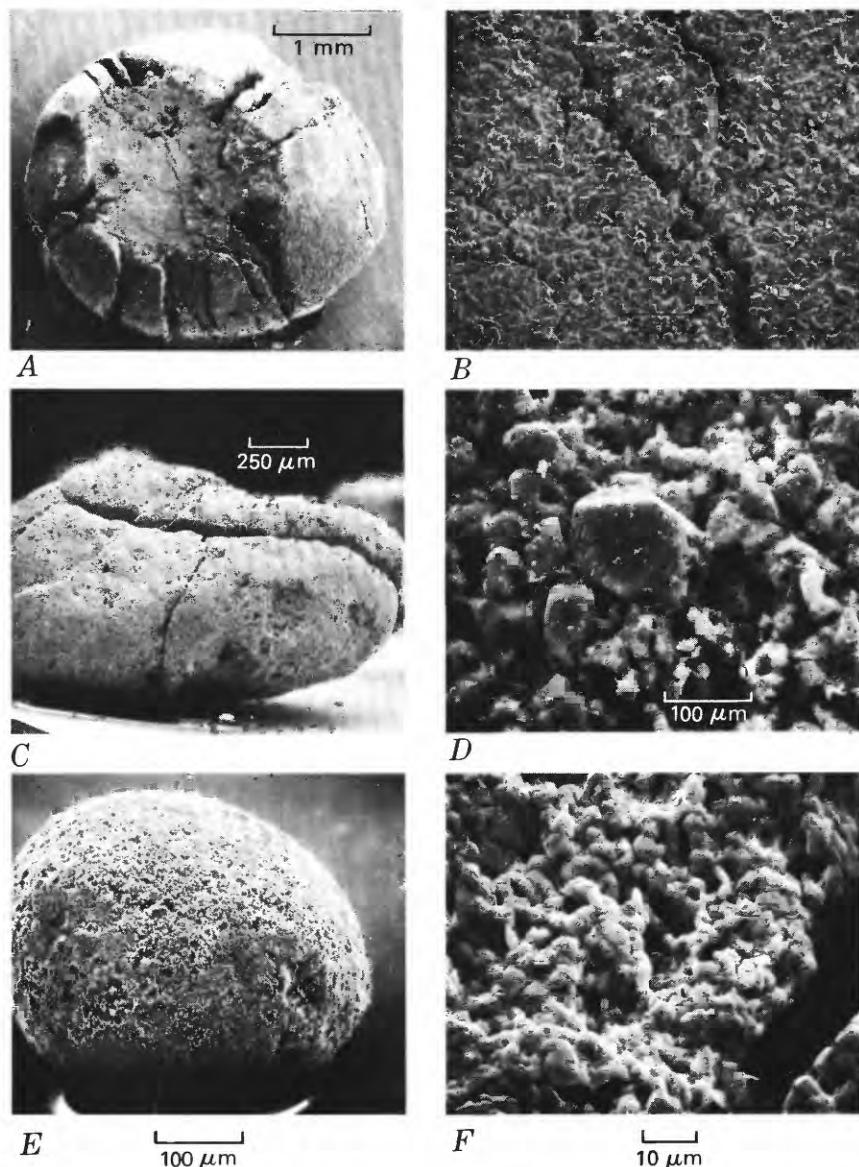


FIGURE 14.—Scanning electron photomicrographs of pyrite mud balls. *A*, Pyrite mud balls from an Elsburg Reef (EA 12), Lorraine gold mine, Orange Free State, with well-preserved indentation and system of cracks caused by plastic deformation by overlying pebbles. *B*, Portion of the surface showing the fine-grained texture and small cracks, as well as partial recrystallisation. *C*, Pyrite mud ball from the Vredefort area (Bird Series). *D*, Surface details of *C*, showing the grainy texture. *E*, Pyrite mud ball from the Dominion Reef showing the typical porous surface. *F*, Surface of *E*, showing the fine-grained texture of irregularly formed grains.

stratigraphic horizons in the Witwatersrand that contain large amounts of authigenic pyrite have, in most instances, subeconomic gold contents. This can be seen as a reflection of the original depositional environment, which could be described as a low-energy environment producing fine-grained arenaceous sediments with sulphide gels and mud, the precursors of the authigenic pyrite. Only limited amounts of very fine grained gold could be transported under such conditions.

The occasional occurrence of minute inclusions of gold in, as well as epitaxial growth of gold on, authigenic pyrite points to a local redistribution of gold under conditions of elevated temperatures, which could result from regional metamorphism or would exist in the vicinity of volcanic intrusions.

#### OTHER DETRITAL HEAVY MINERALS

Gold and pyrite normally are accompanied in the conglomerates by varying amounts of other detrital heavy minerals. Of economic importance in particular localities are the platinum-group minerals and uraninite; the lesser-known minerals are chromite, zircon, cobaltite, gersdorffite, arsenopyrite, rutile, molybdenite, graphite, tourmaline, and others.

The detrital cobalt-nickel arsenosulphides and the detrital sphalerite in Witwatersrand conglomerates are of special interest because they sometimes occur as primary intergrowths with gold (fig. 6). Chemically, they are even less stable than pyrite in a normal oxidizing fluvial environment; thus their presence is an additional indicator for a low oxygen content, if not an absence of oxygen in the palaeoenvironment of Witwatersrand time. To date, all geochemical work on the Witwatersrand and similar rocks elsewhere support this concept. Work on the distribution of rare-earth elements in particular showed that only from about 1,950 m.y. B.P. (before present) on did the oxygen partial pressure approach the present level (Fryer, 1977), and there appears to be no evidence for either a reducing Precambrian atmosphere (Shimizu, 1976) or a highly oxidizing atmosphere, as postulated by several authors (Kimberley and Dimroth, 1976).

Except for a few obviously recrystallised gersdorffite particles, most arsenosulphides encountered in HF-treated samples displayed a typical rounded, detrital morphology (fig. 15).

The onset of recrystallisation observed on some grains could easily be misleading when observed in polished sections. There appears to be a tendency for incipient recrystallisation to be more evident in the nickel-rich members of this group.

There is a wide range in the compositions of the iron-, cobalt-, and nickel-arsenosulphide particles in Wit-

watersrand conglomerates. Within a certain reef horizon, however, the composition often remains fairly constant. As shown in figure 16, only particles from the Basal Reef, St. Helena gold mine, vary greatly in their compositions, whereas samples examined from the other localities have well-defined compositions.

Gold from the Basal Reef at the St. Helena Mine occurs as two distinct varieties: coarse, crystalline, hardly deformed particles; and flaky, extremely deformed gold typical of a long transport distance. Seen together with the variation in arsenosulphide composition, this could be interpreted as indicating transport from two or more sources to its present position.

A short transport distance for part of the heavy-mineral assemblage at St. Helena is supported by the occurrence of detrital sphalerite (fig. 15) and gold-sphalerite intergrowths, as well as molybdenite and graphite. The excellent cleavage and low hardness of these minerals make them good indicators of low-energy environment and short distance of transport. A geochemical indication of a genetic relation between sphalerite and gold is given by the gold content of sphalerite grains, which averages about 200 ppm.

As in the case of pyrite, etch pits on the surface of detrital sphalerite and arsenosulphide grains point to the presence of small amounts of oxygen in the Precambrian atmosphere rather than to a complete lack of oxygen.

Although it is a small part of the detrital heavy-mineral assemblage, chromite is a common constituent of all Witwatersrand conglomerates. It normally occurs as well-rounded particles, often with a verrucose surface structure, which could be of primary origin. In a study of chromite composition in the Klerksdorp Goldfield (Uitter, 1977), no significant changes in the chemistry of detrital chromite throughout the Upper Witwatersrand could be observed, and the composition could best be compared with that of chromite from Archean greenstones.

The comparatively large amount of zinc observed in chromite particles from the Basal Reef appears to be a local phenomenon.

It has been claimed in the past that a major portion of the pyrite in Witwatersrand conglomerates was derived from pyritization of "black sands," that is, from magnetite and ilmenite (Liebenberg, 1955; Ramdohr, 1955). The evidence is based, however, on only a few scarce occurrences of transitional stages, and considering the well-documented detrital nature of most of the pyrite, ilmenite and magnetite can be considered to have been scarce in the primary deposits.

Well-rounded ilmenite particles without any sign of pyritization have recently been observed in samples of the Amazon Reef from the Vredefort area in the centre

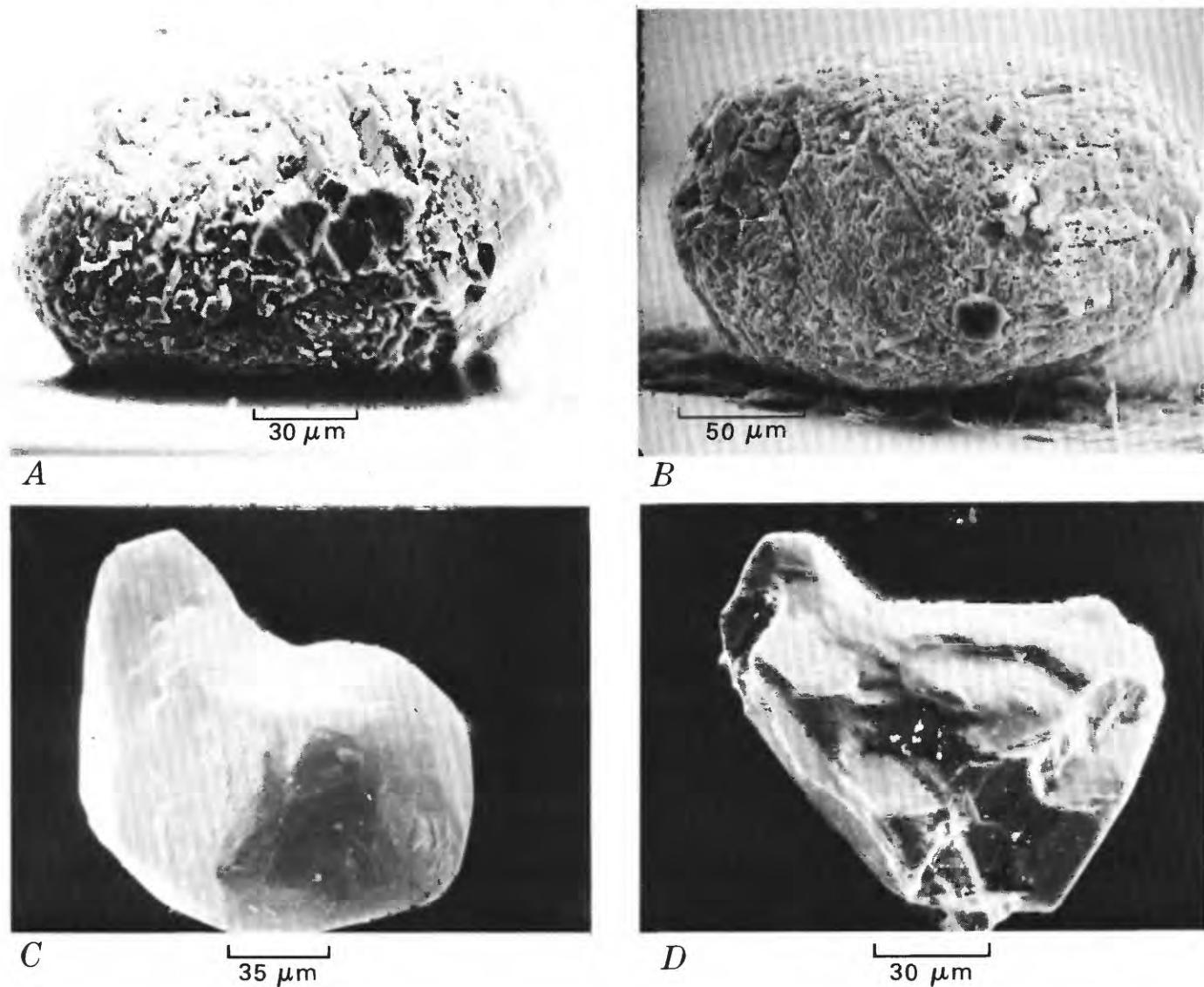


FIGURE 15.—Scanning electron photomicrographs of detrital cobaltite, gersdorffite, and sphalerite. A, Partly recrystallised detrital grain of gersdorffite from an Elsburg Reef (EA 12), Loraine gold mine, Orange Free State. B, Detrital particle of a nickel-bearing cobaltite from a sample of B Reef, Loraine gold mine, Orange Free State. C, Rounded, obviously detrital particle of sphalerite with a slightly etched surface from a sample of B Reef, Loraine gold mine, Orange Free State. D, Rounded, octahedral fragment of sphalerite from a sample of Carbon Leader Reef, Western Deep Levels gold mine, Carletonville.

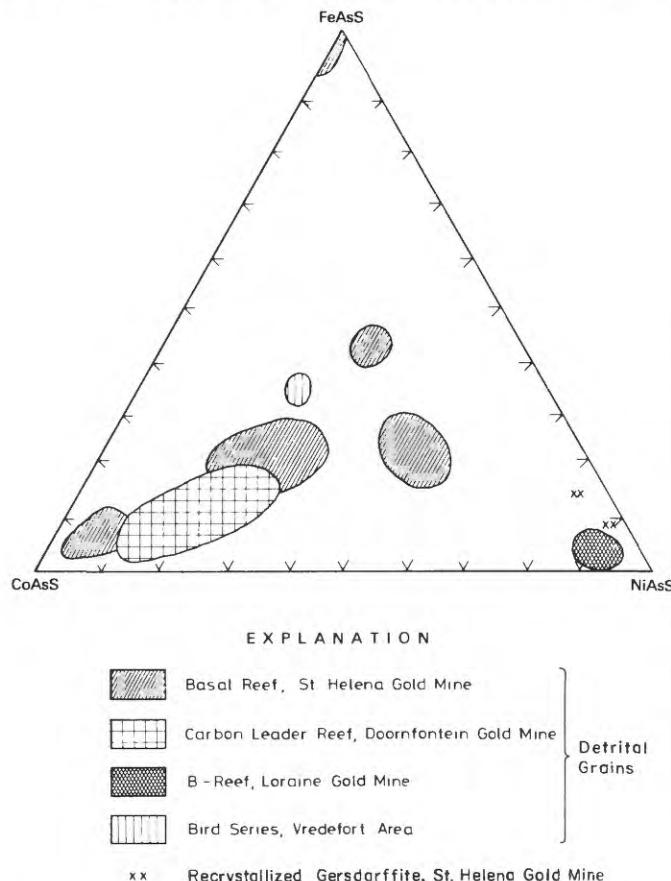


FIGURE 16.—Composition of arsenosulphides from various conglomerate horizons in the Witwatersrand sequence.

of the Witwatersrand. Although the ilmenite was accompanied by detrital pyrite, no intergrowths of the two minerals were observed.

Another prominent constituent of most Witwatersrand conglomerates is zircon. It occurs as rounded, single crystals less than 100  $\mu\text{m}$  across, and as fractured, partly rounded remnants of larger crystals. Experiments have shown that hydrofluoric-acid treatment at room temperature has no noticeable effect on zircons. A comparison between the trace-element composition of zircon concentrated from beach sand and the same treated with hydrofluoric acid has shown no difference between the two samples.

These samples and other concentrates of zircons from various reef horizons were analysed by instrumental neutron activation (Watterson, 1977). In all samples analysed the zirconium-hafnium ratio was found to range between 43 and 48, which is similar to values quoted in the literature for granites.

Zircons from one of the three horizons analysed, the Vaal Reef, were found to contain anomalous amounts of antimony and iron. Zircons from the Kimberley Reef,

Evander Goldfield, and the Kimberley Reef, East Rand Goldfield, had almost equal trace-element compositions as well as similar rare-earth patterns (fig. 17). The rare-earth pattern for the Vaal Reef zircons, in contrast, showed a less pronounced europium anomaly. Although the results from more samples are still being awaited, it appears that different source areas can be recognized and geological units delineated by the trace-element content of zircons in the heavy-mineral suite.

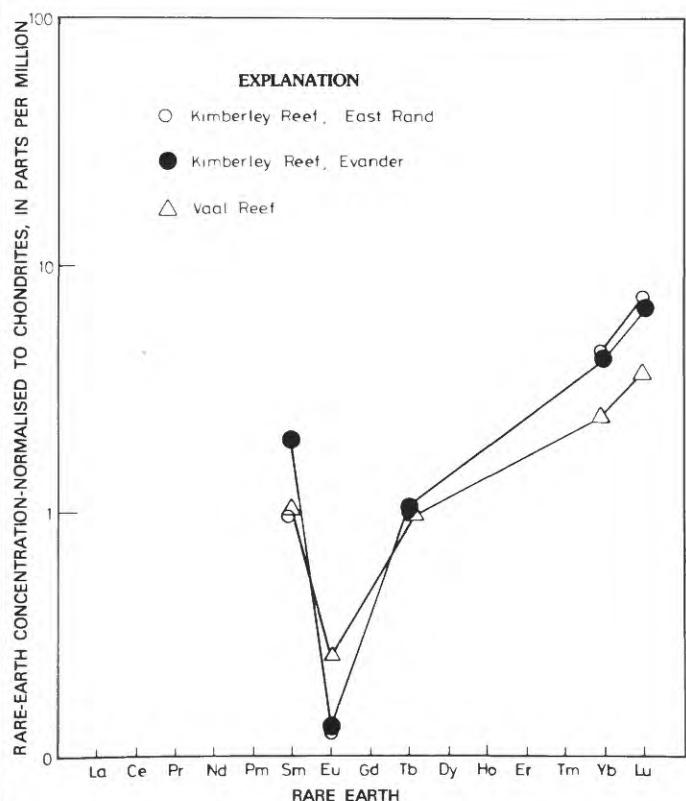


FIGURE 17.—Rare-earth contents of zircons from three conglomerates in the Upper Witwatersrand.

Although quartz is not part of the heavy-mineral suite, it is nevertheless the major component of Witwatersrand conglomerates. Recently a research programme has been initiated to study the trace-element content of quartz and especially the rare-earth-element pattern by instrumental neutron-activation analysis, as well as the daughter minerals in fluid inclusions. The results so far indicate that a classification of quartz pebbles can be made using the above-mentioned criteria, and that more information is to be gained on the origin of certain quartz pebbles and their relation to gold mineralisation.

Various other heavy minerals, including the platinoid minerals, tourmaline, and graphite (not actually a heavy mineral), have been observed but not specially investigated.

## CONCLUSIONS

In studies of the distribution of gold, uranium, and other heavy minerals in fossil placers the technique of morphological examination in the scanning electron microscope opens up the third dimension to microscopy. This supplements the classical two-dimensional microscopy as a tool for geological investigation and makes possible observations not previously attainable with earlier methods.

New investigative methods apply especially to the geochemical studies of mineral grains made with the microanalysing attachment of the scanning electron microscope and with instrumental neutron-activation analysis of single grains, as well as sample suites.

Fossilised organisms could consequently be recognized from the morphology of their carbonaceous remains, permitting their role in the concentration of gold and uranium to be explained.

Studies of the morphology of different types of gold, pyrite, and other minerals have permitted conclusions on the origin and distribution of economically important minerals, as well as on the palaeoenvironment of Precambrian placers that were not possible with the classical methods of microscopy.

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## DISCUSSION

[At the workshop Dr. Hallbauer presented three short papers, titled: (1) Stereo X-ray radiography and chromatographic painting techniques applied to the study of Witwatersrand conglomerates; (2) Morphology of gold, pyrite, zircon, and chromite in the Witwatersrand Series; (3) Genetic relation between organic material and gold in Witwatersrand conglomerates. The first one followed the presentation by Dr. Whiteside, the second one followed Dr. Saager's talk, and the third one followed Dr. Schidlowski's paper.

Dr. Hallbauer has combined the three talks into a single manuscript, chapter M of this report, which somewhat complicates orderly placement of the discussion following each talk. The headings to the discussions will aid the reader in following the different discussions.]

### DISCUSSION FOLLOWING PAPER 1 BY DR. HALLBAUER

*Simpson:* Your larger gold grains associated with the carbonaceous material don't look like detrital grains.

Do you consider it possible, therefore, for the gold to have recrystallised diagenetically on the carbonaceous material which your beautiful photographs, if I may say so, show so clearly as being relatively barren in gold?

*Hallbauer:* First of all, the large gold particles in the carbonaceous material don't look detrital in the photographs which I've shown, but they look detrital in the stereophotographs. And, apart from this, we have dissolved many samples of conglomerate in hydrofluoric acid and paid special attention to these large gold particles, and I will show a few of them later on and you can see quite clearly that they are detrital.

*Minter:* On your first radiograph, on the right-hand side on the foresets there were three or four gold particles. You said there was no gold in the quartzite between the pebble layers. We, on the contrary, have sliced specimens and find that in the reef unit, which is that mature layer of quartz arenite, between 8 and 18 percent of the total gold content is in the quartzite on foreset layers. That's quite a high percentage of the total gold content and this is where one would expect to find it as detrital particles.

*Hallbauer:* Yes, but on the other hand if you look at the comparatively thin conglomerate, most of the gold is either on the footwall contact or in the matrix surrounding the larger particles. In the photographs I've shown you it's very difficult to distinguish between pyrite and gold. You would have to look at the original X-ray photographs where you can quite easily see the difference, but you are quite right in so far as we've found examples of gold distributed on foresets in this reef and in the footwall rocks.

*Skinner:* Have you any specific reagents that you use for your staining techniques with uraninite? Have you developed any?

*Hallbauer:* We have done very little, but we've found that if you use nitric acid as a dissolving agent and use potassium hydrate (KOH), then the uraninite shows up as brown dots, that is, if you don't have too much iron. You have to mask the iron before you do that. So quite often, it might be three treatments, one after the other. And if you do it in quick succession, then the solution which is in the pores has no time to disperse; otherwise, you get blurred images of your grains. We found with the contact method that you can detect gold particles down to a diameter of 10 microns, but in that case you have to expose for a longer time. The small gold particles are surrounded by a small halo of precipitate, but under the ordinary stereomicroscope you can observe the small core within.

**DISCUSSION FOLLOWING PAPERS BY DRs. MINTER, SAAGER, AND HALLBAUER (PAPER 2)**

*von Backström:* Well, ladies and gentlemen, I think you'll agree with me that we have listened to three extremely interesting papers, and these are now open for discussion.

If you would permit me to kick the ball off, I must say I am a bit confused. I think we have seen some very strong evidence presented by Dr. Hallbauer that the distance of transport was very small, and this I believe is also shown by one of the previous slides that was projected that was taken from work by Professor Pretorius, and Dr. Saager also gave us some very interesting data showing that the provenance was quite far away. The Barberton area from which the gold could have been transported, I think, for those of you who don't know South Africa, is of the order of about 300 miles from the central Rand, and is probably about 450 miles from the Orange Free State. Now, I wonder what is the general view on this question today? Could you give us the answer?

*Saager:* Yes. I think you should not look at the Barberton Mountain Land as the actual source area. It is a model source area because we haven't got any greenstone belts which we can actually look at closer to the Witwatersrand Basin except the small greenstone remnants in the Johannesburg dome, but unfortunately we haven't found any sulfides in the greenstone remnants. The small remnants preserved are the very roots of the greenstone belts, and the rest has been completely eroded away. I don't think the actual source is the Barberton Mountain Land or any of the greenstone belts in the eastern Transvaal, but similar greenstone belts, now covered by younger rocks or eliminated by the Bushveld Complex, much closer to the basin.

*Grandstaff:* I have a question for Dr. Hallbauer. I am a little concerned about possible alteration of the grain surfaces during the HF treatment. Have you made any SEM photos on grains prior to and after the HF treatment to determine any possible alteration in the surface characteristics?

*Hallbauer:* Yes, we examined some grains, particularly zircon grains recovered from beach sands, before and after hydrofluoric acid treatment, and there was no sign of any etching. More detailed investigations of resistance to hydrofluoric acid treatment have been done by Neuerburg from the U.S. Geological Survey to develop this method of hydrofluoric acid treatment, but as far as we are concerned, we haven't found any signs of hydrofluoric acid attack, apart from acid attack on uraninite grains, of course. They preserve their round shape, but they are completely altered to

hexafluoride. We found no evidence of acid attack on pyrites.

*Whiteside:* May I make a couple of comments on the remarks that von Backström made just now. First of all, Barberton is over to the northeast and I believe everybody believes the transport direction is from the northwest or north; and secondly, there are remnants of greenstone belts in the Vreysrus [?] dome, and there was actually one small gold mine there.

*Little:* I'd like to ask Dr. Hallbauer if he has been able to make any correlation of the degree of transport with the paleogeography?

*Hallbauer:* [Hallbauer drew a sketch map of the Witwatersrand Basin and indicated that the gold particles that showed signs of short transport were collected near a source and gold particles that showed increased distortion of their surfaces were collected farther from a source. He said that that was the only information he had at that time, but that they are continuing to collect data.]

*Saager:* We've been looking at some lead-isotopic composition of nodular pyrites on which Dr. Hallbauer identified mud cracks. Now the interesting result of this investigation is somehow a puzzling result. It shows that this pyrite probably has formed as early as 2,000 million years ago, which would have been during the metamorphism of the sediments. Could you make some remarks on that? We don't really understand this age we have been getting.

*Hallbauer:* So far, our geochemical investigations have proceeded only as far as the gold content of these pyrites. We have found that the rounded crystalline pyrite from one particular reef has an average gold content of 10 ppm, which is more or less the amount that can be accommodated in the crystal lattice. The pyrite which could be typical for pyritic concretions has the lowest gold content, average of 1½ ppm. The pyrite which we interpret as pyritic mud balls has gold contents of something between 600 and 900 ppm, and we selected these pyrites very carefully to make sure that no visible gold was attached to them. First of all, they were selected manually under the stereomicroscope and then the same fraction was X-rayed on fine-grained film to make sure that no large gold particles were included within the pyrite. Even so, the gold content was very high. The other results concerning the other trace elements—cobalt, nickel and others—are not completed yet.

*Saager:* Wouldn't a high gold content be proof of a very late formation of this mud cake, during probably the solution stage of the modified placer theory?

*Hallbauer:* The possibility is that fine-grained gold was included when the mud was still soft and pliable, or the

gold was transported into the porous pyrite and was included in the conglomerate. We haven't yet made polished sections and we can't answer this.

[Some discussion was not recorded while recording tapes were changed.]

*Myers:* I think it's worth commenting on the fact that the osmiridium grains are recovered only from amalgam barrel residues from the mill and so you can get only one point per mine. Osmiridium occurs in parts per billion, so osmiridium grains are almost never seen in polished sections.

*Bourret:* In past years there was a substantial production of osmiridium; of course, that is not the case today. In view of the knowledge gained over the years, it occurred to me that this might be a successful source area to work on.

*Whiteside:* Can I just make a comment there? There was a time, I think, when most of the production of osmiridium came from the East Rand, and the ratio of osmiridium to gold fell as you went across the Central Rand until, I'm just quoting from memory, you found that on the West Rand or on the western side of the Central Rand, it was about one part in six thousand. The greatest production now comes from our latest field, the Evander Goldfield, which is not so young now. It lies to the east and is detached from the main Witwatersrand Basin, and only the Kimberly Reef is mined. It thus looks as though there is a gradual reduction of osmiridium from east to west.

*von Backström:* I observed when I worked in the gold mines that many of these reefs, especially at great depth, are sheared and compacted. As a matter of fact, I think you know that in reefs that have been mined there can be, unless there is very strong support, a plastic flow; the reefs start pushing out. Could some of this movement in the reef itself be responsible for the bending of gold pebbles?

*Hallbauer:* The structures you get on the gold particles are so small that there is hardly any chance of movement within the conglomerates. And apart from this, the structures are so very consistent with the structures on nuggets that the movement within the confined spaces of the rock should produce a totally different structure from the movement within loose pebbles.

*Schopf:* It would seem to me that these rocks have been compacted during lithification and I wonder whether a certain amount of the markings you interpret as abrasion could be introduced during the compaction of the sediment and therefore not necessarily be entirely introduced during transport?

*Hallbauer:* I am sure there are signs which could have been produced during the diagenesis and compaction of the sediments, but on the other hand, the surrounding quartz pebbles normally create a pressure-free

area within their matrix, and movement in that space should be confined to only microns. I admit there is a possibility that these things could be mixed with movement within the rock; it is very difficult to differentiate between these.

*Schopf:* Have you generated a set of criteria that will allow you to differentiate between markings produced at various times during transport, during sedimentation, during diagenesis, things of this sort?

*Hallbauer:* This is something we haven't done yet. There are examples, however, of gold particles which show only small signs of distortion and others which show signs of complete distortion. How to differentiate between them will be shown in further work.

*Simpson:* Dr. Hallbauer, I would like to ask you with which detrital gold particles are you comparing your material, which you have alluded to several times in your talk, because they don't compare at all to the type of detrital gold particle that you see deposited by glacial action in, say for example, the strath of Kildonan in Scotland?

*Hallbauer:* The gold particles we used came from rivers in the Barberton area, and we assumed transport distance from the last known outcrop of gold-bearing rock to the place where we took our samples, about 30 to 50 km. In addition, we had samples of gold particles from Blind River, but apart from that we haven't examined gold particles from other alluvial deposits yet.

*Feather:* I would like to ask Dr. Hallbauer, would you care to comment on what proportion of the original detrital gold was redistributed during subsequent metamorphic activity in the reefs?

*Hallbauer:* That's a bit difficult to answer. In a large number of samples we found signs of gold being redistributed mostly as oriented growths on secondary pyrites, but from rough judgment I would say that this is an insignificant amount, probably less than 20 percent.

*James B. Black, Atlas Minerals, Utah:* I have a couple of rather naive questions perhaps. To Dr. Saager: Could you refer me to the method by which you independently establish the age of a lead loss? Some document that I might be able to read that might detail your method?

*Saager:* There are two papers: One is by Berger, which appeared in 1962 in *Cosmochimica Acta*, and the other one is by Nicolaysen, which is in the same volume, also 1962.

*Black:* Thank you, that's fine. Then to Dr. Hallbauer: Have you observed similar indications of transportation on grains of uraninite?

*Hallbauer:* Unfortunately, most of the uraninite is attacked by hydrofluoric acid, and it would be extremely difficult to say anything specific about the surface structure of uraninite grains. Of course, you have to

take into account the signs of acid attack, and it might be better to say nothing at all. I think there is a certain amount of caution required in these statements.

*Black:* How about attempting to get a photomicrograph of a particle in place that has not been etched by hydrofluoric acid?

*Hallbauer:* That we haven't attempted. I would say that it is very difficult to find a grain of uraninite within the matrix which has not been broken and where the surface has been kept in such a state that you can really make statements.

*Black:* Thank you very much.

*Bourret:* I am sure that everyone is aware that there is an excellent paper by Liebenberg with dozens and dozens of photomicrographs of uraninite grains in reef ore, 50, 75, or 100 of them. They are all described and they show clearly rounded detrital grains.

*Hallbauer:* If I may quickly comment, these detrital grains from Liebenberg come from milled products. They have been through the mill, and surface structures are somewhat dubious.

*Myers:* If I recall the literature correctly, he [Liebenberg] specifically said that he dug some of these [rounded uraninite grains] out of reef samples.

*Feather:* May I comment on this. I have looked at a lot of uraninite grains recently. These are specifically exposed on fresh fractures in the reefs, so that there is not the problem that the grains might be rounded during the milling process. I would say that invariably the grains contain at least one or two distinct crystal faces with distinct corners and many grains are beautiful crystals. The evidence seems to suggest that they have recrystallised in the reefs and that therefore any original features have been completely destroyed. The previous idea that the grains were completely rounded I would contest, certainly from examining freshly exposed grains in the reefs. In my talk I'll show you some photographs.

*Kalliokoski, Michigan Technological University:* I would like to make a comment about the nature of gold. It seems that the gold in the Witwatersrand is in some respects anomalous from more recent gold. I was introduced to this subject by a gentleman at Stanford in the metallurgy department, a Colonel Crook, who became experienced in studying gold. He was involved in some law cases where the question was whether the gold was Recent or Tertiary, and he was able to demonstrate that Recent gold was quite irregular whereas Tertiary gold had recrystallised and had very definite crystal faces. A similar kind of recrystallised gold has been found in some of the placers in Surinam, and at a place in Venezuela called Icabaru you can find octahedra. My wife has one that she forbids me to cut

which is about the size of a pea that shows seven faces. There is a geologist, Dr. Cecelia Valetsia [sp.?], who has a gold crystal about three-quarters of an inch in diameter which has recrystallised in place. Why can you be so sure that they recrystallised in place? Because there are no hydraulic equivalents with the gold. There are no gold veins in the vicinity; therefore, the gold must have been transported a considerable distance. From this kind of evidence one can then conclude that the gold must have recrystallised. So, it is peculiar that the gold in the Witwatersrand does not show recrystallisation. Now, there is a possibility that the chemical conditions were different in those days. We know that uraninite was persistent; perhaps because of similar conditions gold refused to migrate and therefore didn't recrystallize. But there is also the other fact that gold is exceedingly soft. Accordingly, I think that one would have to take particular care in collecting gold from in place or even collecting gold from present-day placers to make sure that, if it has crystallised in place, it doesn't become scratched between the time it is dug out and the time that it is examined under the microscope.

*James Trow, Michigan State:* I have two questions. The first for Dr. Minter: Would you care to speculate concerning the greater proportion of uranium compared to gold in the Canadian occurrences compared to those in the Witwatersrand? Do you ascribe those differences to perhaps the Canadian occurrences being in a more distal portion of the fluvial system, or are the differences more related to provenance, with greater greenstone surface exposure near the Rand compared to what we have in Canada?

*Minter:* I think the reason is that one does have different provenance areas and different original amounts of gold and uranium so that the ratio change has to be based on each separate population. And, moreover, one finds that there is change going down the paleoslope of a single reef. We have done this with a number of reefs and find that there is a ratio change down the paleoslope but that one can't compare one reef directly with another because on some paleoslopes one can go from a coarse pyrite nodule to a fine pyrite nodule in only 4 km, whereas in the Steyn Reef it takes 22 km. Obviously the gradients are different, the sizes of the deposits are different, and I think that certainly provenance areas supply different proportions. The deposit at Denison [Elliot Lake, Canada] is not a distal deposit; it looks quite proximal from the sizes of pebbles, et cetera, so therefore the original uranium content must have been extremely high. I have wondered whether the uraninite grains are actually larger than those in the Witwatersrand, but I have no data.

*Trow:* The second question is to Dr. Hallbauer: Have you seen any octahedra of pyrite or any forms which might suggest that the pyrite is pseudomorphic after magnetite?

*Hallbauer:* No, I haven't observed any octahedra or any signs of alteration from magnetite to pyrite.

*Unidentified [Growick?]:* A remark on that. I think that Ramdohr and several other authors have shown inclusions of magnetite in pyrite pebbles, suggesting a replacement of magnetite by pyrite, but all these authors could not give a clear indication where this replacement actually would have taken place. Has it taken place in the basin or outside the basin? I do not know.

*Hans Adler, Energy Research and Development Administration:* I would like to ask this question of Dr. Minter. In regard to this 70/30 approximate ratio of detrital to authigenic pyrite, is there any observable change in this ratio going from the proximal to the distal portions of the reefs, and also is there an observable difference in this ratio between the carbon-bearing and the non-carbon-bearing reefs? Secondly, has an attempt been made to determine differences in the sulfur-isotope ratios of the authigenic and the detrital pyrites?

*Minter:* I haven't observed any difference in that wild estimate of the ratio going from proximal to distal deposits, and the only places where I have seen a big change have been in the vicinity of dikes which have

cooked up the reefs and remobilized the pyrite. In fact, in specimens of that nature one can't measure the pyrite nodules; they have been completely recrystallized. One other criterion in collecting samples was to make sure that one collected pyrite from samples taken some distance from any intrusives. I think I've seen comments made on the sulfur-isotope ratios in textbooks, and I think that somebody else is more qualified to comment on that.

*Schidlowski:* I think I'd better do that in the frame of my talk.

*Adler:* That's fine. Could you comment on the ratio of the two types of pyrites in the carbon and noncarbon reefs? Is there any observable difference there?

*Minter:* No, none.

*Adler:* Thank you very much.

*Weis, U.S. Geological Survey:* This is for Dr. Minter. You commented on the increase in the percentage of zircon down the paleoslope and the change in the gold-uranium ratio and the increase in uranium content down the paleoslope. Is there a change in the gold content, and if so, what is it?

*Minter:* There is a small change in the gold content. It decreases down the paleoslope slightly, but not nearly as much as the increase in the uranium content. We intend to measure the size of the zircon grains and also to analyse the zircon concentrations so that we hope eventually to tie this up completely.

Uraniferous Constituents of  
the Witwatersrand Conglomerates:  
Ore-Microscopic Observations and  
Implications for the Witwatersrand Metallogeny

By MANFRED SCHIDLOWSKI

GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN  
QUARTZ-PEBBLE CONGLOMERATES

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## GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN QUARTZ-PEBBLE CONGLOMERATES

### URANIFEROUS CONSTITUENTS OF THE WITWATERSRAND CONGLOMERATES: ORE-MICROSCOPIC OBSERVATIONS AND IMPLICATIONS FOR THE WITWATERSRAND METALLOGENY

By MANFRED SCHIDLowski<sup>1</sup>

#### ABSTRACT

A review is given of the available ore-microscopic evidence pertaining to the origin of the main uraniferous constituents of the banket. Uraninite occurs principally in the footwall portions of the reefs as rounded or muffin-shaped grains some 100 micrometers in diameter, which leave little doubt as to their waterworn, abraded nature. It can be demonstrated by comparison with detrital monazite grains from present-day beach placers that the muffin shape displayed by individual uraninite particles constitutes an unquestionable sedimentary abrasion form. Brannerite appears to be an authigenic constituent of the reef, most probably formed from uraninite and rutile as a result of postdepositional "pseudohydrothermal" reconstitution. Branneritization of primary detrital uraninite grains has often preserved the characteristic abrasion forms of the uraninite parents (so-called "uraninite ghosts"). As for carbonaceous matter or "thucholite," microscopic evidence decidedly favors a genetic concept involving a radiolytic polymerization of originally mobile hydrocarbons (most probably indigenous to the rock) by pre-existing uraninite constituents, with these latter subsequently subjected to progressive replacement by the ambient organics. Apart from a morphologic description of the principal "carbon" aggregates in the reef, a critical appraisal is given of a recently proposed different concept for the origin of the organic substances. Altogether, the mineragraphic evidence lends strong support to a modified placer theory previously advanced for the origin of the Witwatersrand metallogenic province as a whole.

#### INTRODUCTION

The rediscovery of uraninite in the Witwatersrand conglomerates, originally recorded by Cooper (1923), during the World War II hunt for fissionable material gave rise to a temporary revival in the 1950's of the longstanding Witwatersrand controversy between the "placerist" and "hydrothermalist" schools (Davidson, 1953, 1957; Liebenberg, 1955; Ramdohr, 1955, 1958a, b). During these discussions, the crucial importance of mineragraphic studies for the elucidation of the Witwatersrand metallogeny became apparent. Following the pioneering work by Ramdohr and Liebenberg, an impressive array of detailed ore-microscopic investigations has appeared during the last two decades, furnishing *prima facie* evidence of a detrital origin for the banket deposits (Koen, 1961; Schidlowski, 1966a, 1970a;

Schidlowski and Trurnit, 1966; Hiemstra, 1968; Saager, 1970). The following paper aims at briefly reviewing the basic observational and microchemical evidence accrued in these publications to provide a synopsis of our present knowledge of the principal radioactive constituents of the reef.

#### URANINITE

##### OCCURRENCE WITHIN THE CONGLOMERATES

Both microscopic inspection and autoradiographs of complete reef sections have invariably furnished evidence that the bulk of the uraninite is mainly concentrated in the lower portions of the conglomerates, along with gold and other heavy minerals. The richest assemblages of uraninite particles have always been observed at the reef footwalls, the individual grains sometimes perching like beads on a string immediately at the footwall contacts. (All figure 1 photomicrographs are of samples from such footwall seams.) Occurrences of uraninite in the upper reef portions are relatively rare and mostly confined to so-called "false footwalls" forming the base of a new depositional cycle within the same conglomerate horizon. Pending the accumulation of contradictory evidence, this would identify the uraninite fraction as a member of the original heavy suite of the conglomerates.

##### MICROSCOPIC CHARACTERISTICS

Both Ramdohr (1955) and Liebenberg (1955) have identified the prototype of the primary, unaltered uraninite grain within the Witwatersrand conglomerates, which was subsequently recognized in all further ore-microscopic surveys. In general, this prototype shows up as well-rounded particles some 80–150  $\mu\text{m}$  in diameter (fig. 1). The ore microscopist will be particularly struck by the ubiquitous occurrence of grains exhibiting a typical "muffin shape," characterized by one flat side, the opposite side forming a gentle bulge (fig. 1A, C, D, F, I, J).

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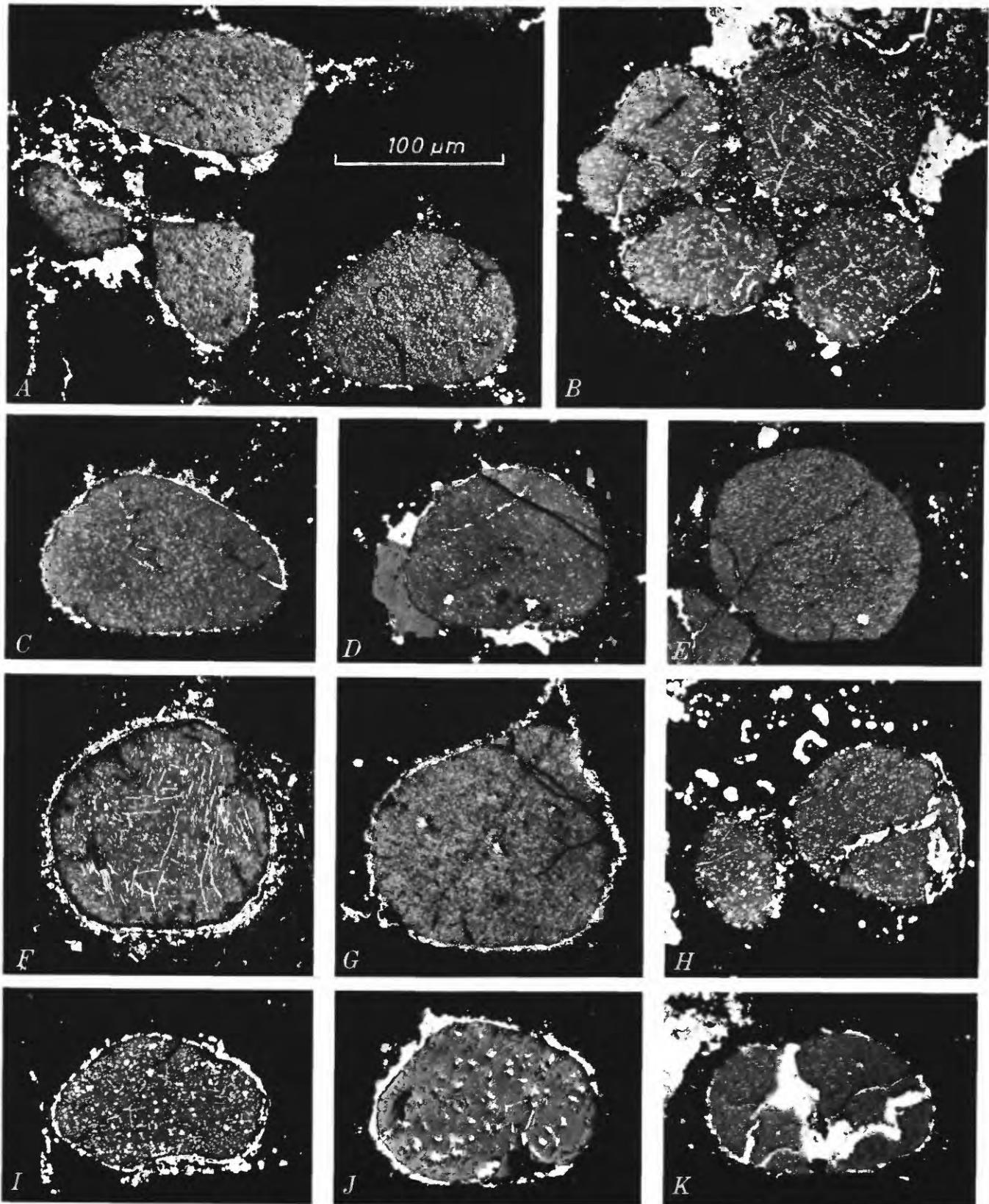


FIGURE 1.—Detrital grains of uraninite from various Witwatersrand conglomerates, mostly Basal and Elsburg Reefs, Orange Free State, many of them displaying excellent muffin shapes (A, C, D, J). Note dustlike inclusions of galena (white) in all grains, as well as concentrations of this mineral in cracks (B, C, H) and on octahedral cleavage planes (F). Parts of the galena have been “sweated out,” forming outgrowths (A, B, C, D, H) or aureoles (G, J) around individual uraninite particles. Single grains are veined by pyrrhotite (K) intergrown with galena or overgrown by authigenic pyrite (F, I) or sphalerite (D, left side). All photomicrographs taken in oil immersion.

In polished section, the gray groundmass of the grains is always speckled with myriads of dustlike inclusions of galena (PbS), the radiogenic lead having been obviously subjected to sulfurization during postdepositional reconstitution of the blanket. (With both detrital and authigenic sulfides present in the reef in large quantities, this was actually to be expected.) Radiogenic galena often appears to be mobilized, forming crack fillings within (fig. 1B, C, H, I, K) or aureoles around (fig. 1B, C, G, J) the individual uraninite grains, with small amounts of the galena also dissipating into the surrounding groundmass. Apart from galena, reconstituted sulfides,—mostly pyrite, pyrrhotite, and chalcopyrite (fig. 1K)—and also gold have been observed as crack fillings within uraninite particles, occasionally occupying octahedral cleavage planes of the host mineral (figs. 1F, 2B). Some grains are also transected by minute stringers of carbonaceous matter (fig. 2A, C).

Apart from the typically well-rounded uraninite particles described and their cataclastic fragments, hypidiomorphic grains have also been encountered (fig. 2). With the exception of single occurrences from which they have been reported in larger amounts—for example, the Dominion Reef (Hiemstra, 1968)—these grains are, however, relatively rare constituents of the blanket.

#### CHEMISTRY

X-ray diffraction data supplied by Liebenberg (1955) have furnished proof that the microscopically identified uraninite particles yield a powder diagram identical in all respects with that of artificially prepared  $UO_2$ . Chemical analyses of uraninite concentrates from two different reefs, the Main Reef and the Carbon Leader, have shown a  $ThO_2$  content of 1.6 and 2.7 percent, respectively (Liebenberg, 1955, p. 126). This general tenor was subsequently confirmed by a microchemical

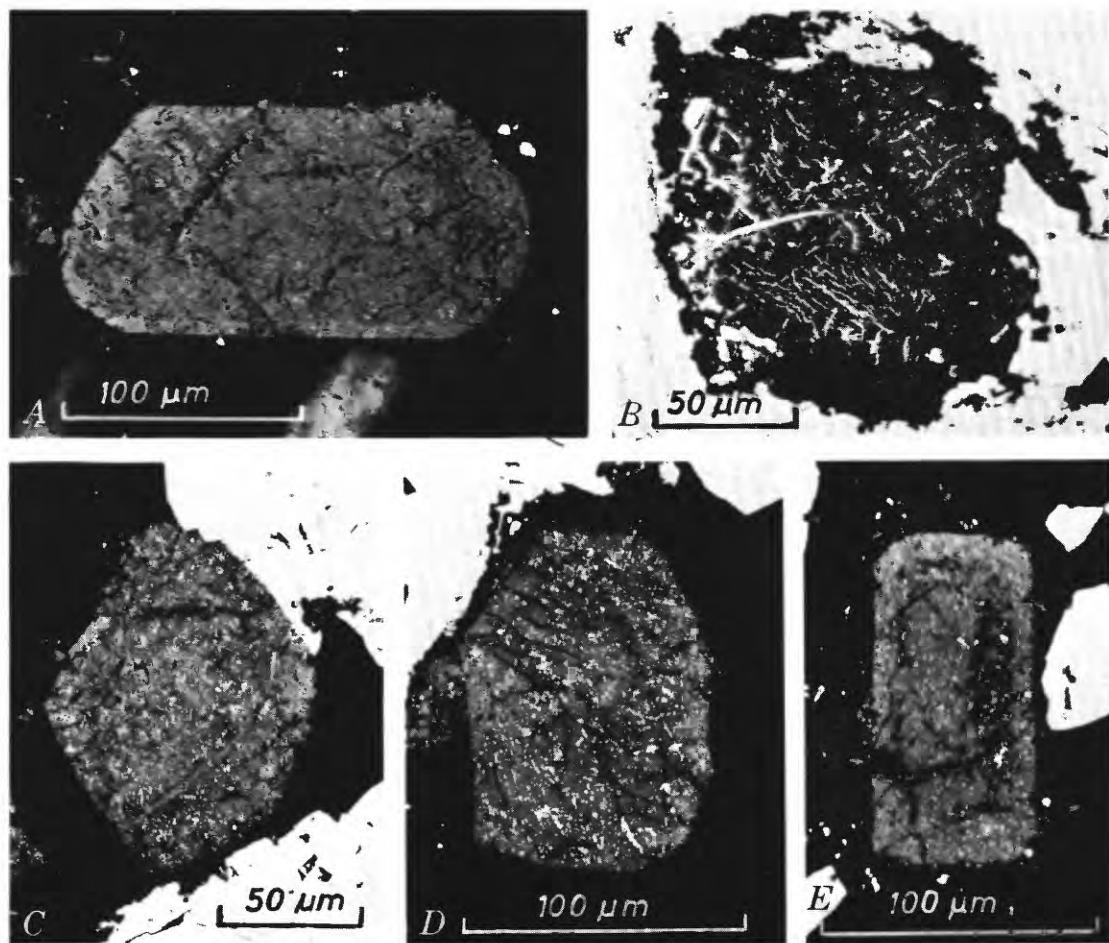


FIGURE 2.—Hypidiomorphic uraninite particles from various Orange Free State gold conglomerates. Some grains (A, C) contain minute stringers of "carbon" showing up in different shades of gray due to pronounced reflection pleochroism of the carbonaceous material. One particle (B) overgrown by authigenic pyrite is veined by gold, the veins following, in part, the octahedral cleavage pattern. Note dustlike inclusions of galena (white) in all grains. All micrographs taken in oil immersion.

analysis of an individual uraninite particle from the Basal Reef, Orange Free State (fig. 3; Schidlowksi, 1966a, p. 190. For the uraninite fraction of the Dominion Reef, Hiemstra (1968) has reported an average  $\text{ThO}_2$  content of about 6.5 percent. Recently, Grandstaff (1974) has submitted the results of a comprehensive microprobe investigation of uraninite from the Vaal Reef that showed the  $\text{ThO}_2$  tenor of individual grains to lie mostly in the range of 2 to 7 percent.

#### ORIGIN OF THE URANIUM MINERALIZATION

With all information presently available on the uraninite fraction of the blanket, there can be no doubt that this mineral is a detrital constituent of the rock. In particular, its distribution pattern within the conglomerates is typical for a member of a suite of heavy minerals, corresponding to that of gold and several other heavy minerals. Further, the relation of the economic mineralization in general to sedimentation features of the primary depositional environment long has been recognized (Reinecke, 1928; Sharpe, 1949).

Prima facie evidence of a detrital origin of the uranium mineralization is provided by the clearly water-worn, abraded nature of the individual uraninite grains.

The claim advanced by Davidson (1957) in the Witwatersrand controversy, that abrasion is unlikely to have occurred in the small size class to which the bulk of the uraninite fraction belongs, does not stand up to critical review. According to Correns (1960, p. 136), particles smaller than 0.1 mm in diameter may still be excellently rounded in certain sedimentary environments. Further, Pettijohn (1957, p. 63) regarded 0.1 mm as a possible lower limit for the rounding of sand grains. According to a graph by Pettijohn and Lundahl (1943), the quasi-linear relation between the sphericity of grains and their mean diameter still holds for the size class of about 100  $\mu\text{m}$  in the case of well-worked beach sands, whereas the respective function of a fluvial sand suffers a breakdown at about 170  $\mu\text{m}$  (fig. 4). However, with the degree of abrasion, particularly of the smaller size classes, largely dependent on the maturity of a sediment, low sphericity values in immature detritals have no significance at all for the issue under consideration.

Moreover, if ordinary sand grains some 100  $\mu\text{m}$  in diameter still fall within the subrounded category,  $\rho > 0.25$  (fig. 4), an abrasion of the considerably heavier uraninite particles (S.G. = 9.0–9.7) of the same size class should present no problems at all. Actually, reasonably well-rounded uraninite grains as small as some 60  $\mu\text{m}$  in

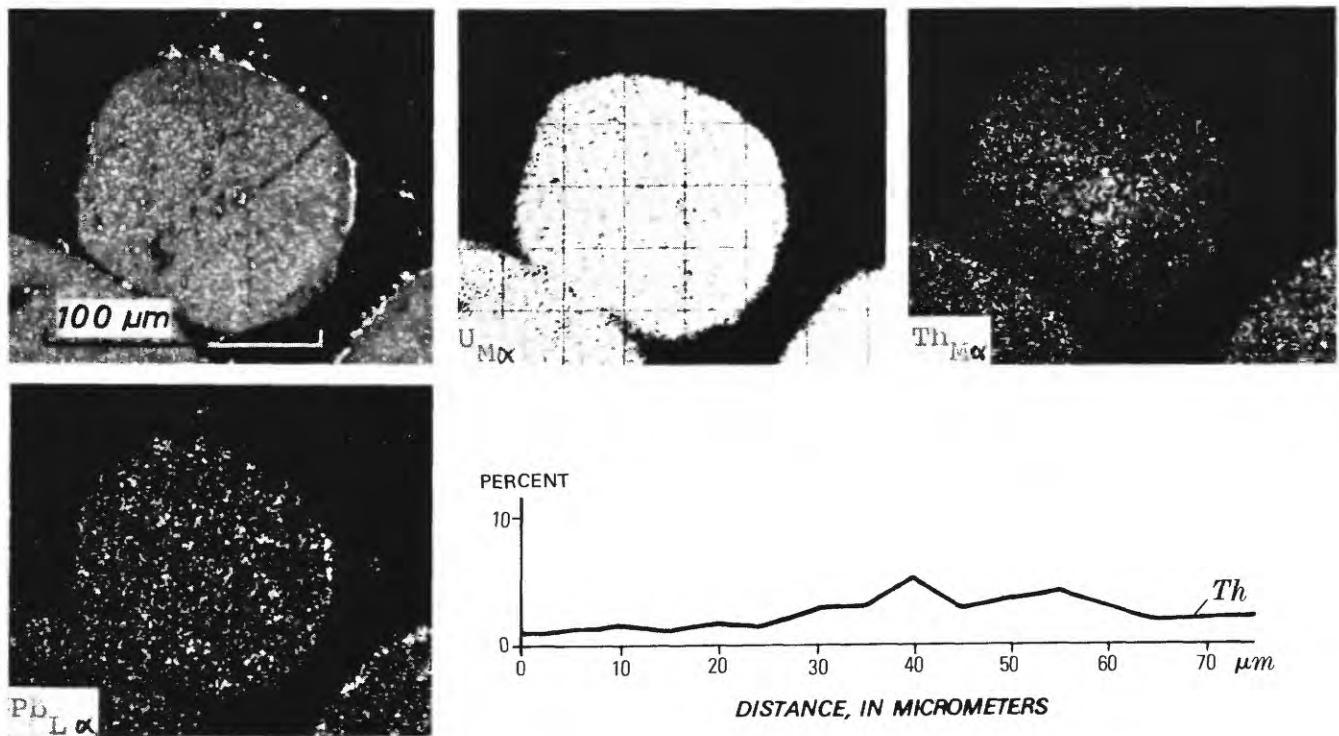


FIGURE 3.—Detrital grain of uraninite with minute specks and outgrowths of galena, and microprobe scanning pictures for uranium, thorium, and lead. Note higher thorium concentrations in central part of grain, shown also by quantitative step-scanning diagram traversing the particle over a distance of some 7  $\mu\text{m}$ . Average thorium is 2.42 percent; average  $\text{ThO}_2$  is 2.75 percent. From Basal Reef, Orange Free State; optical picture taken in oil immersion.

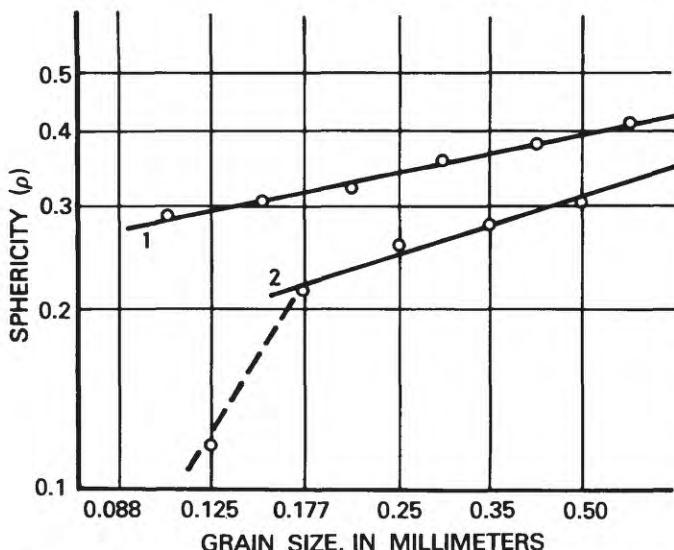


FIGURE 4.—Relation between roundness and average grain size in a Lake Erie beach sand (1) and a Mississippi River sand (2). Note the continuous decrease of sphericity values with decreasing grain size. In the case of well-worked beach sands (1), the size fraction of about 0.1 mm falls within the "subrounded" category  $\rho$  between 0.25 and 0.40 (from Pettijohn and Lundahl, 1943, fig. 6; published with permission of The Society of Economic Paleontologists and Mineralogists, *Journal of Sedimentary Petrology*).

diameter (fig. 1A, B) have been observed in the conglomerates. According to the experience of this author, this is about the smallest grain size to display abrasion phenomena. Uraninite particles of smaller size are either angular clastic debris or relict bodies of larger grains that had suffered partial replacement by carbonaceous matter (see fig. 13).

Among the various undoubted abrasion forms observed, the muffin shape of a large number of uraninite grains (fig. 1) is most conspicuous. This morphology is fairly widespread in practically all size classes of a large variety of clastic rocks and probably constitutes one of the most common abrasion forms known in pebble morphology. Major quartz pebbles of this type have also been described and illustrated from Witwatersrand conglomerates (Du Toit, 1954, p. 100).

Comparison of the uraninite "muffins" with the shapes of other ancient and modern detrital clasts (fig. 5, 6) is likely to lend unequivocal support to a placer origin of the uraninite fraction. The morphological analogies between the grain populations are actually so self-evident, that any further detailed discussion seems superfluous. It should be pointed out, however, that the excellently rounded monazite muffins from a recent Australian beach placer shown in figure 6 fall within a size range similar to the Witwatersrand uraninites. This observation should definitely dispose of Davidson's (1957) idea that grains of these dimensions are no longer susceptible

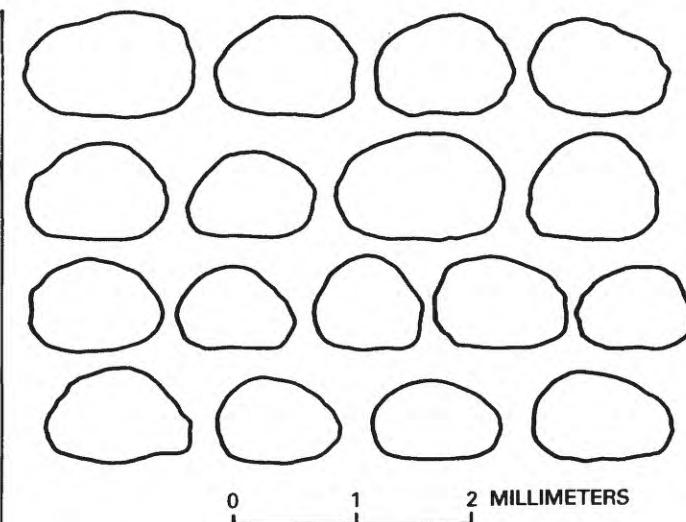


FIGURE 5.—Muffin-shaped quartz grains from "Ottawa Sand" facies of Ordovician St. Peter Sandstone, Ottawa, Illinois. Drawn from thin section of grain mount.

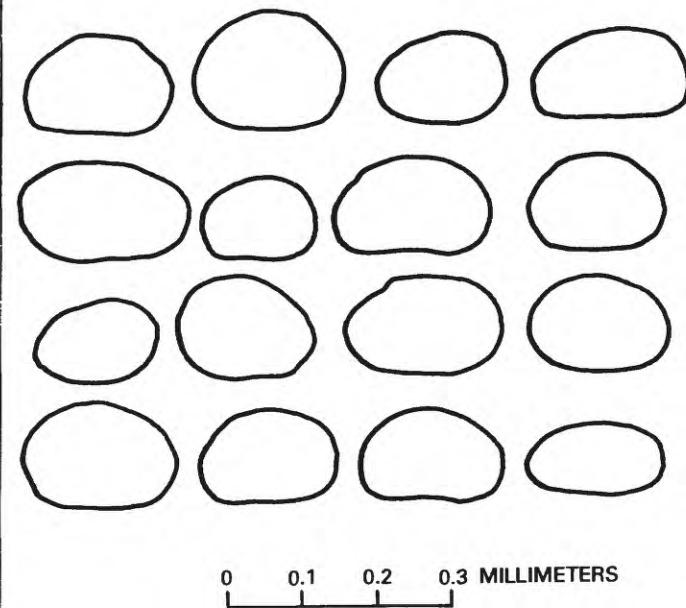


FIGURE 6.—Detrital monazite grains with typical muffin shapes from a recent West Australian beach placer, drawn from grain mount of a monazite concentrate.

to rounding. The obvious correspondence in grain morphology between the Witwatersrand uraninites and the heavy-mineral fraction of a modern marine beach placer is suggestive of genetically related primary sedimentary environments, thus vindicating previous interpretations of the economic conglomerates as principally ancient littoral placer deposits (Brock and Pretorius, 1964, p. 570 ff.). Incidentally, the ubiquitous detrital sulfide grains within the Witwatersrand conglomerates, as well as

other members of the heavy fraction—for example, chromite—exhibit similar morphological characteristics, with muffin-shaped individuals abundant (Schidlowski, 1970a).

Brief reference should also be made in this context to Davidson's (1955, 1957) statement that the rounded appearance of the uraninite particles of the blanket is that typical of pitchblende from hydrothermal veins. Instead of a lengthy discussion of this point, two micrographs of colloform, botryoidal pitchblende aggregates from a genuinely hydrothermal deposit are included in this paper for the convenience of the reader not familiar with textures of primary ore deposits (fig. 7). Comparison of these aggregates with the Witwatersrand uraninites reveals a large array of fundamental distinctions; in particular, the ovoid bodies do not display the faintest indication of a muffin shape. Moreover, clustering and intergrowth of the ovoids is very common, whereas the Witwatersrand uraninites are always individual, discrete particles. Rare mention of uraninite "clusters" in the blanket refers either to closely packed assemblages of detrital grains (Liebenberg, 1955, pl. 19, fig. 3) or to obviously reworked and redeposited fragments from older reefs rich in uraninite particles (Koen, 1961, pl. 2). Furthermore, hydrothermal and other low-temperature pitchblende contains very little,

if any, thorium, the  $\text{ThO}_2$  content always staying well below 0.1 percent (Frondel, 1958; Ramdohr, 1969).

The relatively high  $\text{ThO}_2$  and rare-earth-element content of the Witwatersrand uraninites (Liebenberg, 1955, p. 126) is, on the other hand, indicative of a high-temperature (pegmatic) parent material and thus decidedly compatible with the concept of a detrital origin. The wide scatter in the  $\text{ThO}_2$  tenor, between 0.5 and 11 percent with 2–7 percent being most common, recently reported by Grandstaff (1974) for individual  $\text{UO}_2$  particles of the same grain population from polished sections or hand specimens, provides additional proof of a heterogeneous sedimentary assemblage. The compositional variability of the grains would suggest a polygenetic source, with each of the contributing areas characterized by a definite thorium content of the uraninite as a function of different temperatures of formation of the parent pegmatite. As pointed out convincingly by Grandstaff, an *in-situ* origin of the uraninite particles would require that the mineralizing fluids were heterogeneous over extremely small distances, which (1) is unlikely and (2) should definitely produce zoning in the growing grains owing to the successive movement through the rock of fluids of different composition. Similar conclusions had been drawn by Ramdohr and others (1965) from semiquantitative microprobe scan-

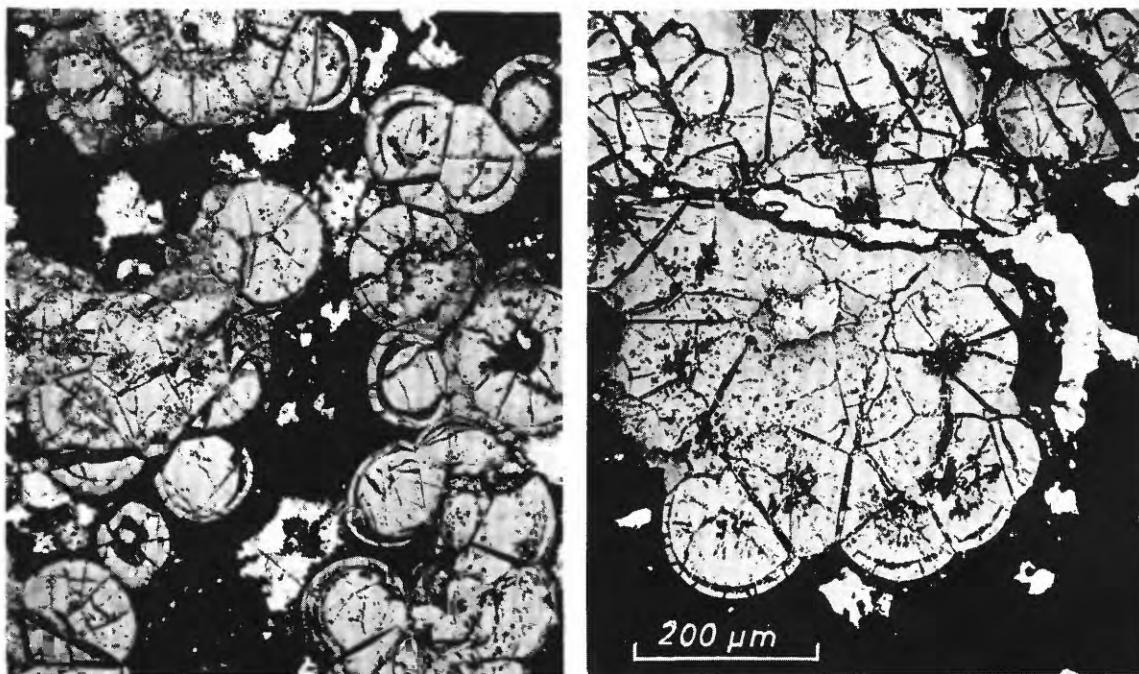


FIGURE 7.—Two typical aggregates of colloform pitchblende from a vein-type uranium deposit, Sudetes Mountains, central Europe. Note that ovoid bodies on left picture, with concentric shells and radial cracks formed during the aging process of the parent gels, differ basically from the detrital uraninite particles of the Witwatersrand. Right photomicrograph depicts large botryoidal pitchblende aggregate. Oil immersion.

ning diagrams. The variable lead content of individual uraninite grains noted during this investigation was interpreted as reflecting different ages of formation of the primary pegmatitic  $\text{UO}_2$  material.

### BRANNERITE

In contrast to the early recognition of brannerite<sup>2</sup> ( $\text{UTi}_2\text{O}_6$ ) as one of the principal uranium carriers in the Blind River conglomerates (Nuffield, 1954), the pioneering ore-microscopic studies of the Witwatersrand blanket lack any specific references to this material. However, in 1957 Ramdohr, by comparing with Blind River samples, was able to identify this phase in the Dominion Reef, expressing also the suspicion that the material of his so-called "uranium ghosts" previously described and at that time not yet identified might actually represent brannerite. Similarly, Davidson (1957) considered that the "uraniferous leucoxene" mentioned by Liebenberg (1955) might be brannerite. Finally, Ramdohr (1958b) was able to definitely identify this mineral in the Witwatersrand conglomerates.

### MICROSCOPIC AND CHEMICAL CHARACTERISTICS

The difficulties inherent in the identification of Witwatersrand brannerite are primarily due to (1) its occurrence as mostly tiny needle-shaped crystals at a micrometer scale, often intergrown in feltlike aggregates, and (2) to its metamict state. It is optically isotropic, and it does not yield the X-ray diffraction pattern of brannerite. The lines for anatase, rutile, and galena usually show up on the diffractometer charts.

However, as demonstrated by Nuffield (1954) for the Blind River material, a brannerite phase is obtained after momentary heating at red heat, a phenomenon corroborated for material from both the Dominion Reef (Taylor and others, 1962) and Witwatersrand reefs (Feather and Koen, 1975, p. 203).

In polished section, the mineral usually appears as felts of needle- or lath-shaped crystals (fig. 8A, B), grading sometimes into irregular patches of massive brannerite (fig. 8C). Both the needles and the compact variety are speckled with minute inclusions of galena; the larger ones sometimes display cubic outlines. The massive variety of brannerite is often characterized by lath-shaped inclusions of pyrrhotite (fig. 8C) recognized by Ramdohr (1958b) as being an aid in identifying brannerite in both Blind River and Witwatersrand samples. Minute dustlike inclusions of anatase have been observed and have been explained as the radioactive breakdown of uranium in the brannerite lattice, with the remaining  $\text{TiO}_2$  subsequently undergoing reconstitution to anatase. Close association of brannerite with patches of rutile is also common (fig. 8C). In places, massive brannerite forms overgrowths on detrital uraninite grains and has a markedly lower reflectivity (see fig 11A, B) than the uraninite. Pending the accumulation of confirmatory chemical evidence, it is suspected that the "secondary" uraninite described by Liebenberg (1955) is largely identical with the compact variety of brannerite.

Peculiar brannerite aggregates are represented by the so-called "uraninite ghosts" originally described by Ramdohr (1955). Subsequent work (Ramdohr, 1958b; Schidlowksi, 1966a) was able to show beyond doubt that these "ghosts" actually represented pseudomorphs of brannerite after detrital uraninite grains (fig. 9). As a

<sup>2</sup> [Brannerite is essentially a uranium titanate of variable composition that contains thorium and minor amounts of calcium, cerium, yttrium, niobium, and ferrous iron.]

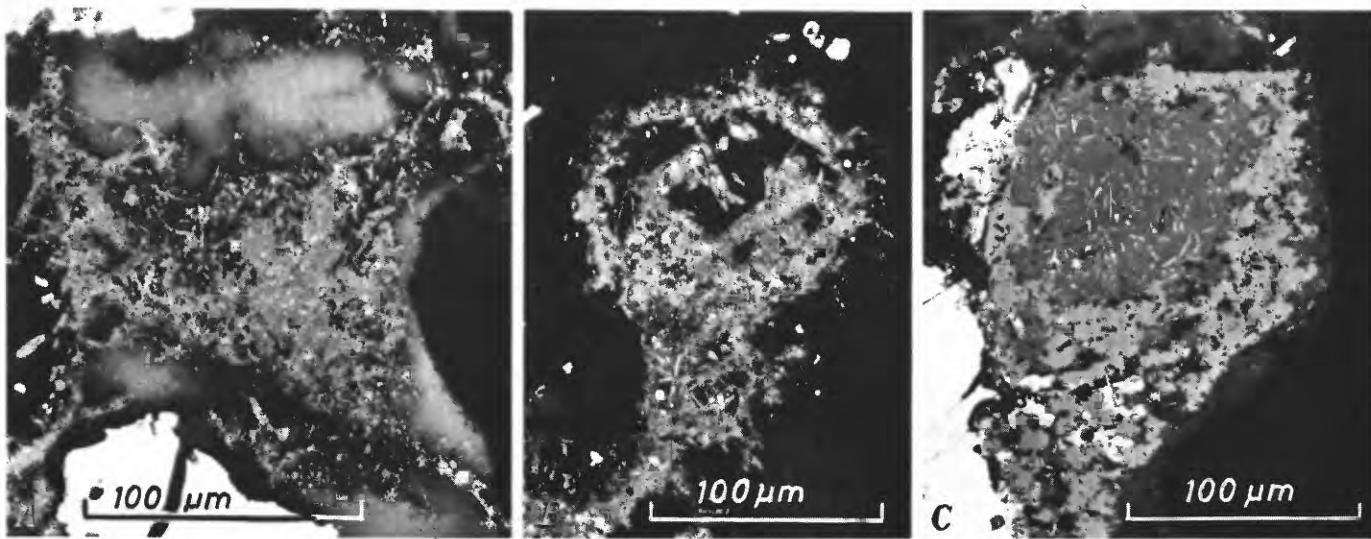


FIGURE 8.—Felts of needle- and lath-shaped brannerite crystals (A, B) and "compact" brannerite aggregate (C) from various Orange Free State reefs. The compact variety (C) contains tiny needles of pyrrhotite and is largely surrounded by rutile. Oil immersion.

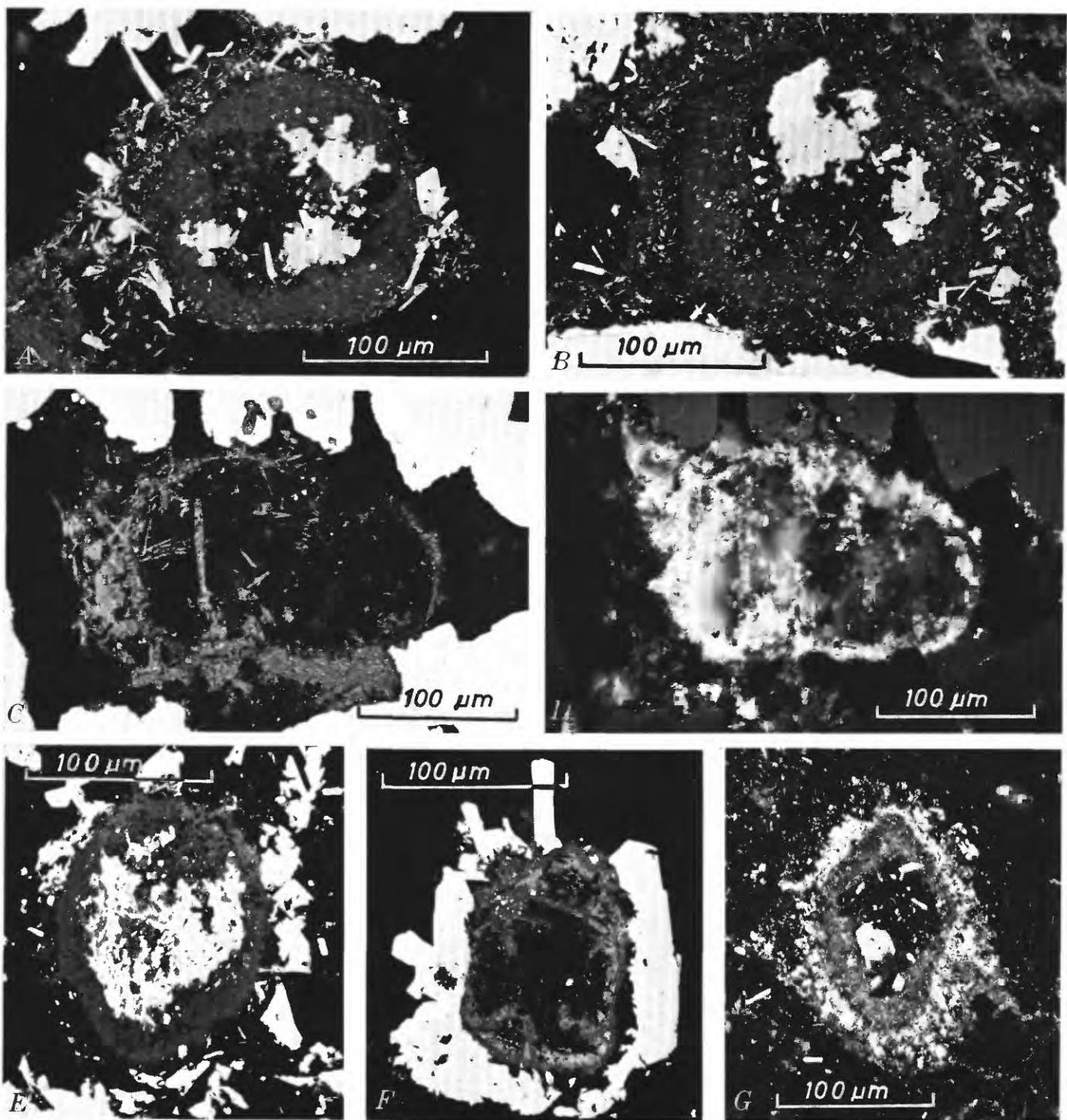


FIGURE 9.—Pseudomorphs of brannerite after grains of detrital uraninite, so-called “uraninite ghosts.” In A, C, and E the abraded condition of the uraninite parents has been excellently preserved. With crossed nicols (D), the muffin shape of C is accentuated by abundant internal reflections of the brannerite. Other “ghosts” derive from hypidiomorphic UO<sub>2</sub> parents (B, F, G). The central parts of the pseudomorphs are leached and the cavities partially invaded by authigenic chalcopyrite, pyrrhotite, and usually a little galena (A, B, E, G). Several grains are surrounded by aureoles of reconstituted sulfides, mostly lath-shaped pyrrhotite crystals (A, E, F) or pyrite (G). Note minute specks of galena in the brannerite matrix of all aggregates. All examples from Orange Free State reefs. Oil immersion.

rule, these "ghosts" excellently preserve the original forms of the parent  $\text{UO}_2$  particles, and many of them exhibit distinct muffin shapes (fig. 9A, C). The central portions of the "ghosts" are usually leached, with the resulting cavities filled, in part, by reconstituted chalcopyrite, pyrrhotite, and galena (fig. 9A, B, E, G). Often the grains are also surrounded by aureoles of authigenic sulfides, most conspicuous in figure 9E, G.

X-ray spectrographic analyses usually show the presence of titanium, uranium, thorium, and lead along with traces of iron and niobium. Microprobe scanning

pictures reflecting the distribution of the principal elements in two samples of Witwatersrand brannerite are presented in figures 10 and 11.

Among the diagnostically valuable optical characteristics of brannerite listed by Ramdohr (1958b, 1969) and by Ferris and Ruud (1971) is its lower reflectivity relative to uraninite (fig. 11) and to rutile (fig. 8C). While the compact variety of the mineral shows almost no internal reflections, the latter are abundant in felted nests of needle-shaped crystals or "cloudy" brannerite patches, particularly when observed in oil immersion

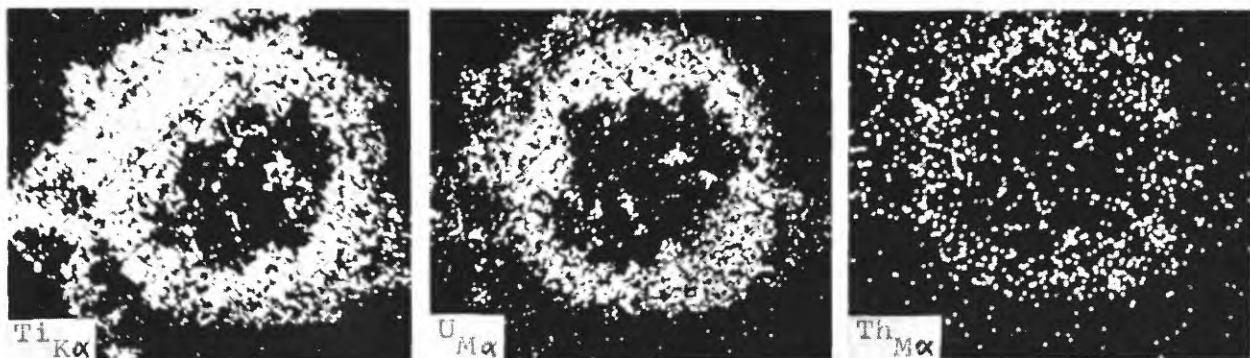


FIGURE 10.—Microprobe scanning pictures for titanium, uranium, and thorium obtained from the muffin-shaped "uraninite ghost" shown in figure 9A.

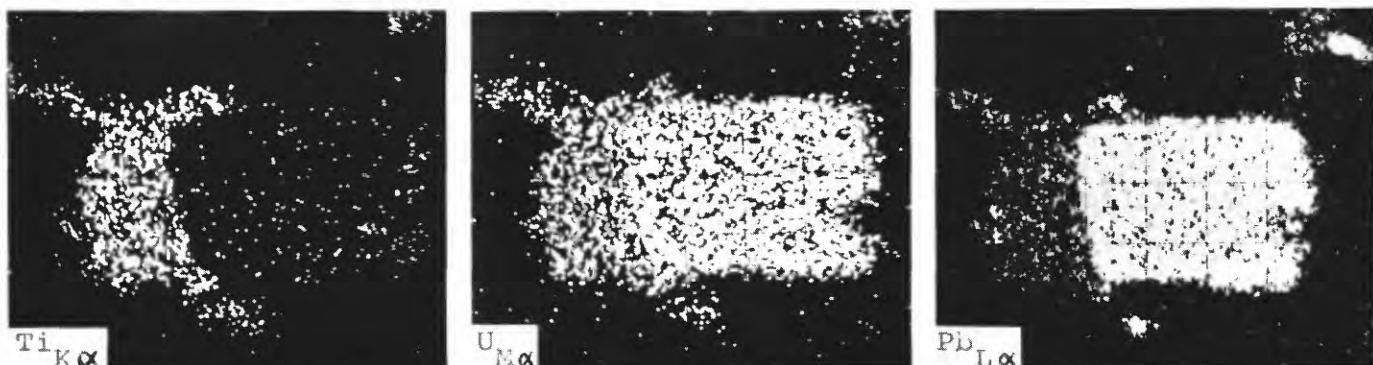
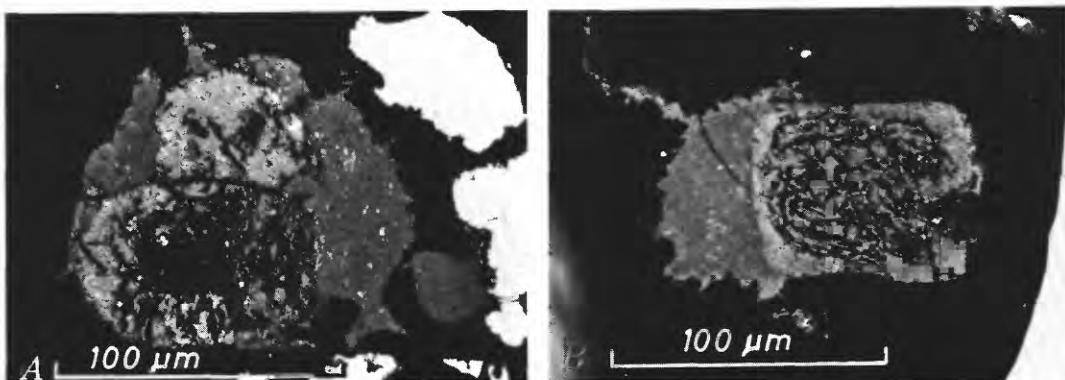


FIGURE 11.—Two grains of partially leached uraninite (A, B) with outgrowths of brannerite. Note distinctly lower reflectivity of brannerite as compared to uraninite. Below, microprobe scanning pictures of B for titanium, uranium, and lead. The brannerite overgrowth stands out prominently in the titanium scan. The rather low lead content of the brannerite indicates that this mineral is considerably younger than the neighboring uraninite. From Elsburg Reefs, Orange Free State. Optical pictures taken in oil immersion.

(fig. 9D). As a result of metamictization, the Witwatersrand species is practically isotropic, although an occasional low birefringence has been reported for other brannerite occurrences.

Radioactive blasting and corrosion phenomena in the blanket caused by radioactive decay of brannerite have been recorded by Ramdohr (1958b) and the writer (Schidlowksi, 1966c).

#### ORIGIN OF THE WITWATERSRAND BRANNERITE

According to the microscopic evidence summarized in figures 8, 9, and 11, there can be no doubt that brannerite is an authigenic constituent of the Witwatersrand conglomerates. With a syngenetic origin excluded, it must be a secondary mineral formed as a result of a large-scale reconstitution of primary detrital minerals during later processes of diagenesis and metamorphism.

Ramdohr (1957) was the first to assume that the brannerite from the Blind River deposits had originated from detrital uraninite and rutile according to his so-called "Pronto reaction,"  $UO_2 + 2TiO_2 \rightarrow UTi_2O_6$ , that was believed to have taken place under "pseudohydrothermal" conditions during the postdepositional history of the conglomerates. With uraninite and rutile present in the Witwatersrand reefs in large quantities, the same reaction should have occurred there also, probably enhanced by the low-grade (greenschist-facies) metamorphism to which the Witwatersrand region was subjected. Besides, it is well known that several thermal events, Ventersdorp volcanism, intrusion of the Bushveld Igneous Complex, and emplacement of Karroo dolerites, have left their imprint on the region as a whole and were responsible for large-scale reconstitution of the detrital sulfides with the formation in places of authigenic mackinawite (Schidlowksi and Ottemann, 1966). However, as the Blind River province is virtually unmetamorphosed (Roscoe, 1969), the "Pronto reaction" can be expected to proceed readily of its own accord in "normal" diagenetic environments. It has been shown that brannerite can be synthesized easily from  $UO_2$  and  $TiO_2$  in the laboratory (Patchett and Nuffield, 1960).

With partial mobilization in the blanket of both uranium and titanium, obvious replacement phenomena, as manifest in the formation of "uraninite ghosts," can be readily accounted for. In "ghosts"  $UO_2$  was largely stationary and titania was the mobile phase, whereas replacement of rutile by brannerite would require mobilization of uraninite constituents. The ubiquitous small brannerite specks interspersed in the conglomerate matrix have certainly formed there in situ as a result of a random encounter of uranium- and titanium-bearing mineralizing fluids. Details of the process of brannerite formation in the reef have been discussed elsewhere by the writer (Schidlowksi, 1966a).

From the microprobe scanning diagram for lead (fig. 11) of coexisting uraninite and brannerite it is obvious that the brannerite, because of its sparse lead content, is considerably younger than the detrital  $UO_2$  constituent of the reef, which conclusion is consistent with the assumed mode of brannerite formation. Possible consequences of this type of radiogenic lead distribution for the interpretation of the uranium-lead ages hitherto proposed for the Witwatersrand have been pointed out elsewhere (Ramdohr and others, 1965).

In discussing the origin of brannerite it should be noted that rutile, as a necessary prerequisite for the "Pronto reaction," is fairly widespread in the Witwatersrand conglomerates (Ramdohr, 1955; Schidlowksi, 1970a, p. 48 ff). Alteration of detrital ilmenite constituents to rutile, as microscopically evident in many excellent samples, should also have contributed extensively to the  $TiO_2$  budget of the blanket. According to Ferris and Ruud (1971), the brannerite of the Blind River province has principally formed from detrital ilmenite that was diagenetically altered to rutile and anatase in the presence of dissolved uranium.

#### CARBONACEOUS MATTER

##### MICROSCOPIC OBSERVATIONS AND THEIR GENETIC SIGNIFICANCE

The widespread occurrence of "carbon" constituents in the Witwatersrand conglomerates was well known by early workers (Young, 1917) and has received increased attention since the discovery of the pronounced radioactivity of the material. The application of reflected light microscopy to the study of "thucholite"<sup>3</sup> (Davidson and Bowie, 1951; Ramdohr, 1955; Liebenberg, 1955; Schidlowksi, 1966a, 1966b) has shown it to be a heterogeneous mixture of "carbon" and uraninite with varying amounts of practically all other minerals contained in the neighboring reef.

For a genetic interpretation of this peculiar carbon-uraninite association, ore-microscopic observations have turned out to be of prime importance. The initial stage in the formation of such aggregates is represented by uraninite particles transected by stringers, and coated by layers, of carbonaceous matter (fig. 2A, C; fig. 12A, B.). Progressive replacement by carbon of the primary  $UO_2$  phase as a result of gradual enlargement of both the carbon aureoles and the crack fillings is documented in figure 12C and D, in which the muffin shapes of the original detrital grains are still easily recognizable. In an advanced stage of replacement (fig. 13) the uraninite has almost wholly disappeared except for small angular

<sup>3</sup> The term "thucholite," coined from the chemical symbols of the main constituent elements (Th, U, C, H, and O) was originally introduced by Ellsworth (1928) for uraninite-bearing carbonaceous materials. In the Witwatersrand literature it is largely used as a synonym for carbonaceous matter.

relict bodies floating in the groundmass of nodular carbon aggregates. Specks of visible carbon thus originated have come to be known as "fly speck" carbon. Typical of the replacement process is a "drifting apart" of the resulting uraninite relicts, which, however, still fit each other excellently, like pieces of a puzzle (fig. 13D-G).

In a remarkable consensus, the principal ore-microscopic investigators have explained these finding in terms of an encounter of a pre-existing uraninite phase and mobile, preferably gaseous, hydrocarbons. These hydrocarbons, while migrating through the strata, were obviously polymerized by ionizing radiation

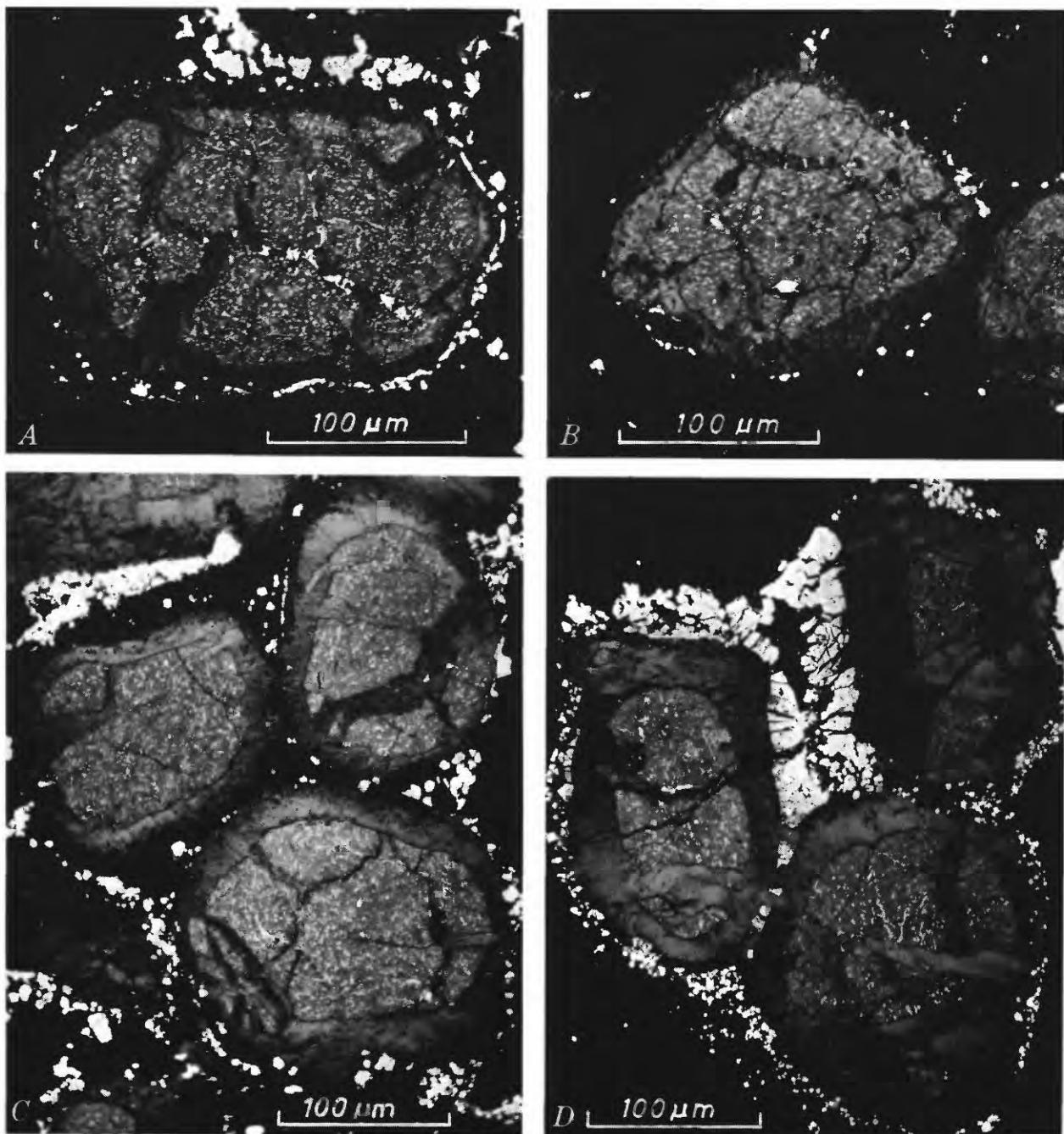


FIGURE 12.—Grains of uraninite coated and veined (A, B) or partially replaced (C, D) by carbonaceous matter. Because of its reflection pleochroism the organic material shows up in different shades of gray. In the initial stages of replacement the pebble shape of the primary uraninite grains is still excellently preserved (C, D). Remnants of uraninite floating in a "carbon" groundmass may be readily recognized by their dustlike inclusions of radiogenic galena (white). Note loosely knit aureole of authigenic pyrite around one of the major aggregates (D). Examples from various Orange Free State reefs. Oil immersion.

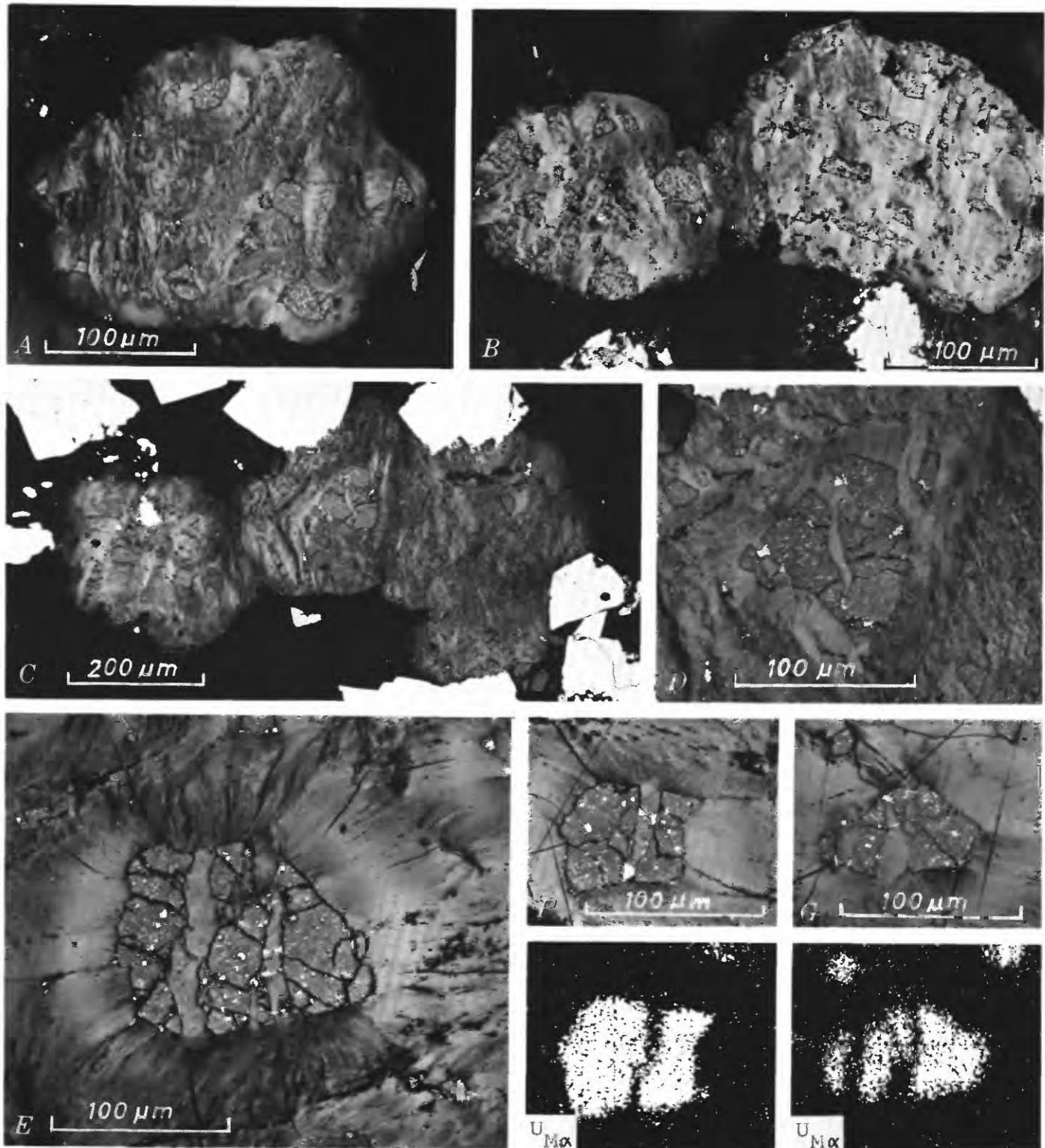


FIGURE 13.—Advanced stages of uraninite replacement by carbonaceous matter. The primary uraninite phase has largely disappeared except for small angular relict bodies. It is reasonable to assume that carbon nodules such as depicted in *A* derive from the replacement of single uraninite grains. Replacement of chains of  $\text{UO}_2$  particles would result in aggregates such as shown in *B* and *C*. Note characteristic "drifting apart" of individual uraninite particles during progressive replacement (*D*–*G*) with galena (white) locally accumulating in the interstices. An enlargement of a particle (*D*) in the center of (*C*) is shown. Element scanning for uranium carried out on samples shown in *F* and *G* shows that the "carbon" groundmass is virtually devoid of uranium. The uranium content of the material is thus confined to the interspersed uraninite particles. Examples taken from various Orange Free State conglomerates. All photomicrographs taken in oil immersion.

emanating from uranium-bearing constituents of the reef. With the process of polymerization entailing a transition into the solid state, the mobile organic substances were fixed in close proximity to the sources of radiation, thus accounting for the carbonaceous aureoles and crack fillings of many  $\text{UO}_2$  particles. The characteristic "drifting apart" of the uraninite fragments during replacement (fig. 13) may be conveniently explained by a continuous supply of mobile organic material to the interface between the hydrocarbons already solidified and the remaining pieces of uraninite.

This concept for the origin of the Witwatersrand thucholite is plausible because processes of radiolytic polymerization of simple, mostly unsaturated, hydrocarbons by  $\alpha$  and  $\gamma$ -radiation have been simulated experimentally by various workers (Lind and Bardwell, 1926; Lind, 1928; Colombo and others, 1964). Radiation effects on the carbonaceous material can also be seen as haloes around many uraninite inclusions (figs. 13E, 18A). More difficult to explain is the mechanism of uraninite replacement by carbon constituents as is seen under the microscope. Microprobe scanning pictures (figs. 13F, G, 14) show the carbonaceous material to be virtually devoid of uranium. Accordingly, the replaced  $\text{UO}_2$  constituents are obviously not resorbed or "digested" by the organic materials in some form such as organo-metallic compounds. This does not mean, however, that such compounds may not have been instrumental in a transitory phase in the breakdown and mobilization of uraninite. Pending a better understand-

ing of the chemical background of the replacement process, a final assessment of these matters must be postponed.

Another mechanism for bringing about a uranium carbon association that is not less important geochemically than the one described should be mentioned here. It is well known that hexavalent uranium of the soluble uranyl complex  $[\text{U}^{6+}\text{O}_2]^{2+}$ , which is often carried by weathering solutions, may be precipitated as  $\text{U}^{4+}\text{O}_2$  in the reducing environment of sedimentary organic matter (McKelvey and others, 1955, p. 472). This reaction, for the most part, is responsible for the conspicuous uranium concentrations in some coals, lignites, peats, and the like, which have been known for a long time (Berthoud, 1875; Breger and others, 1955). The economically important uranium province of the Colorado Plateau (Rosenzweig and others, 1954; Gruner, 1955) was formed mainly in this way. In this area, fossil wood is commonly replaced by pitchblende, the low-temperature,  $\text{ThO}_2$ -poor cryptocrystalline variety of  $\text{UO}_2$ . In the Colorado Plateau, organic materials concentrate uranium by providing a change in redox potential favorable for precipitation of the uranyl complex carried in water, whereas in the Witwatersrand deposits migrating hydrocarbons are concentrated around preexisting uranium-bearing minerals as a result of radiolytic polymerization. Although both processes produce a close association of uranium and carbon compounds, they achieve the association by completely different geochemical pathways.

Attempts to invoke a Colorado-type pedigree for the

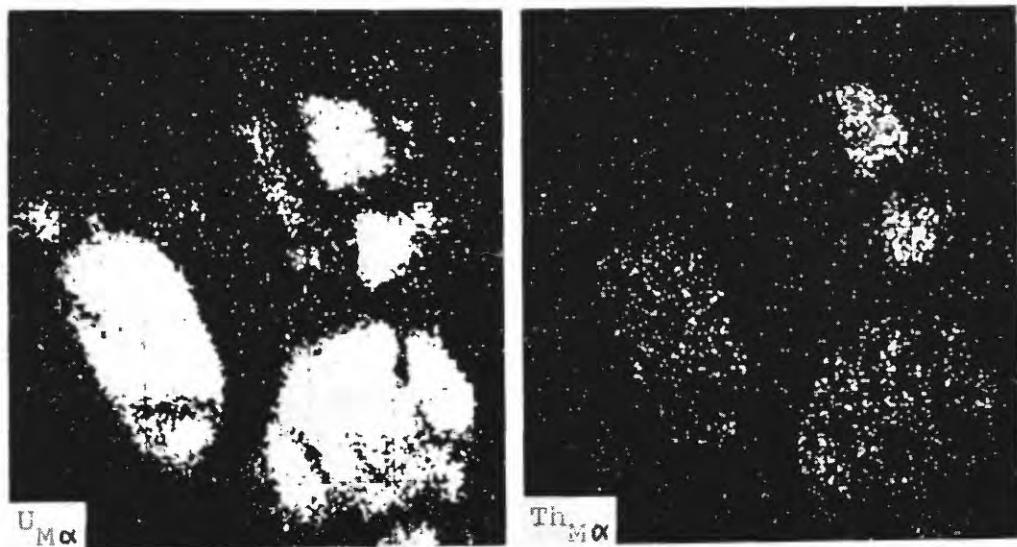


FIGURE 14.—Microprobe scannings for uranium and thorium of the carbon-uraninite aggregate depicted in figure 12D. It is evident that the distribution pattern of uranium almost coincides with the areas occupied by uraninite. Note the increased thorium content of the uraninite particles in upper right corner.

uraninite constituents of the Rand conglomerates (Sharpe, 1949; Snyman, 1969; Reimer, 1975), in part combined with the concept of subsequent reworking by wave action of the  $\text{UO}_2$ -precipitates (Koen, 1961), may safely be dismissed because of the high  $\text{ThO}_2$  content in Rand uraninite, which is not present in low-temperature uraninite. Besides, it is well known that a large part of the uraninite fraction of the blanket is not associated with carbon at all. A crucial prerequisite for the formation of Colorado-type deposits is, furthermore, an oxygenic weathering cycle to oxidize tetravalent uranium of the primary deposits to the soluble hexavalent form, which, in turn, is susceptible to subsequent precipitation by reducing agents. With all evidence presently available strongly favoring anoxigenic weathering on the ancient continents prior to some  $2 \times 10^9$  years B.P. (before present), this is a very unrealistic premise that, in the opinion of this writer, does not stand up to critical examination.

#### CHARACTERISTICS OF MAJOR CARBON ACCUMULATIONS

With the uraninite fraction mostly concentrated at the reef footwalls as part of the heavy suite, the bulk of the organic materials were necessarily trapped at the very base of the conglomerate horizons. When the uraninite grains happened to lie close to each other, there would result chainlike aggregates of carbon, as depicted in figure 13B and C. In this way, continuous bands of carbonaceous matter may appear at the footwalls of reefs (fig. 15). As already noted by previous students, these carbon bands may vary in thickness from fractions of a millimeter to some centimeters, giving rise to a special type of "carbon reef," the prototype of which is represented by the economically important "Carbon Leader" of the West Rand. The carbonaceous material of such bands is sometimes massive, but often displays a characteristic fibrous texture perpendicular to the bedding, as noted by Liebenberg (1955, pl. 28). Figure 16 gives a schematic picture of a typical carbon reef with a seam of fibrous (or columnar) thucholite at its base.

Microscopic inspection of such aggregates (Schidlowski, 1966b) reveals that the fibrous carbon in many places is heavily mineralized, as might be expected inasmuch as it occupies those sites of the conglomerates where the bulk of the economic minerals have been accumulated. The bands are particularly rich in redeposited gold (fig. 15); locally it preserves as casts the orthogonal pattern formed by the intracolumnar interstices, and elsewhere it fills wedgelike cracks transecting the individual fibres perpendicular to their long axes (fig. 17A-C). With the exception of fragments of the primary  $\text{UO}_2$  particles, which are spread throughout the carbon matrix, all allogenetic components are practically confined to the interstices between the fibres.

Rounded grains of pyrite, quartz, iridosmine, and other minerals are engulfed by strands of carbon, and this feature accounts for a typical augen texture, with gold and reconstituted sulfides accumulated in the pressure shadows on either side of detrital minerals (fig. 17D, E). Excellent flow lamination in carbonaceous material has formed around inclusions of authigenic sphalerite particles, with the latter often dragged apart ("boudinaged") and elongated fragments tending to arrange themselves parallel to the laminae (fig. 17F). The marked plasticity of the carbonaceous material is also demonstrated in figure 18C.

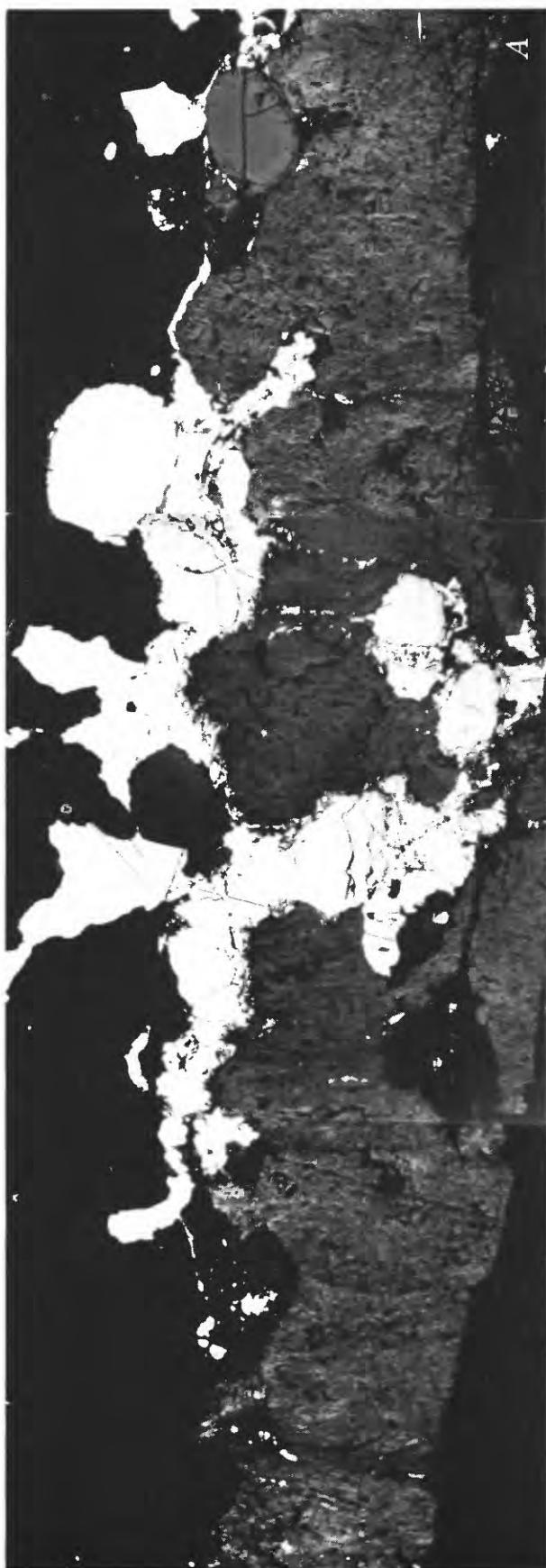
A section of fibrous carbon cut in a skew angle to the fibre direction is presented in figure 18A, the ends of the individual fibres showing up as lenses often centered by small uraninite inclusions. These inclusions have given rise to radioactive haloes within the surrounding carbonaceous matter in which the reflectivity is markedly increased. With crossed nicols (fig. 18B), a certain pregraphitization of the material is obvious, although it has been demonstrated by various investigators that the carbonaceous material is basically amorphous (Davidson and Bowie, 1951; Liebenberg, 1955). Actually, graphite is an extremely rare constituent of the blanket (fig. 18D).

From the fabric of these fibrous aggregates it is evident that the carbonaceous matter in its present form is clearly younger than most minerals of the detrital suite. On the other hand, there can be no doubt that the bulk of the gold that also formed part of the original heavy fraction of the conglomerates has undergone large-scale reconstitution at a relatively late stage, being texturally fully integrated in the kinematic pattern displayed by the carbon aggregates.

#### ORIGIN OF COLUMNAR THUCHOLITE

In an attempt to account for columnar thucholite, the writer (Schidlowski, 1966b), has drawn analogies to the mode of origin of fibrous mineral aggregates grown at right angles to the bedding planes within intrastratal partings. Such phenomena are well known from minerals like gypsum, rock salt, calcite and many others. According to Mügge (1928), these aggregates may be visualized as having resulted from the mutual lateral impediment of minerals growing in the limited space of a rock fissure, with crystal growth ultimately possible only in the direction of the parting walls of the crevice. Although applicable only with certain modifications to the Witwatersrand carbon, this model is likely to give a satisfactory explanation for the occurrence of the columnar thucholite bands, which preferentially came to occupy so-called "footwall partings" at the base of individual conglomerate horizons.

Recently, however, Hallbauer and van Warmelo (1974) have offered an alternative explanation, relating



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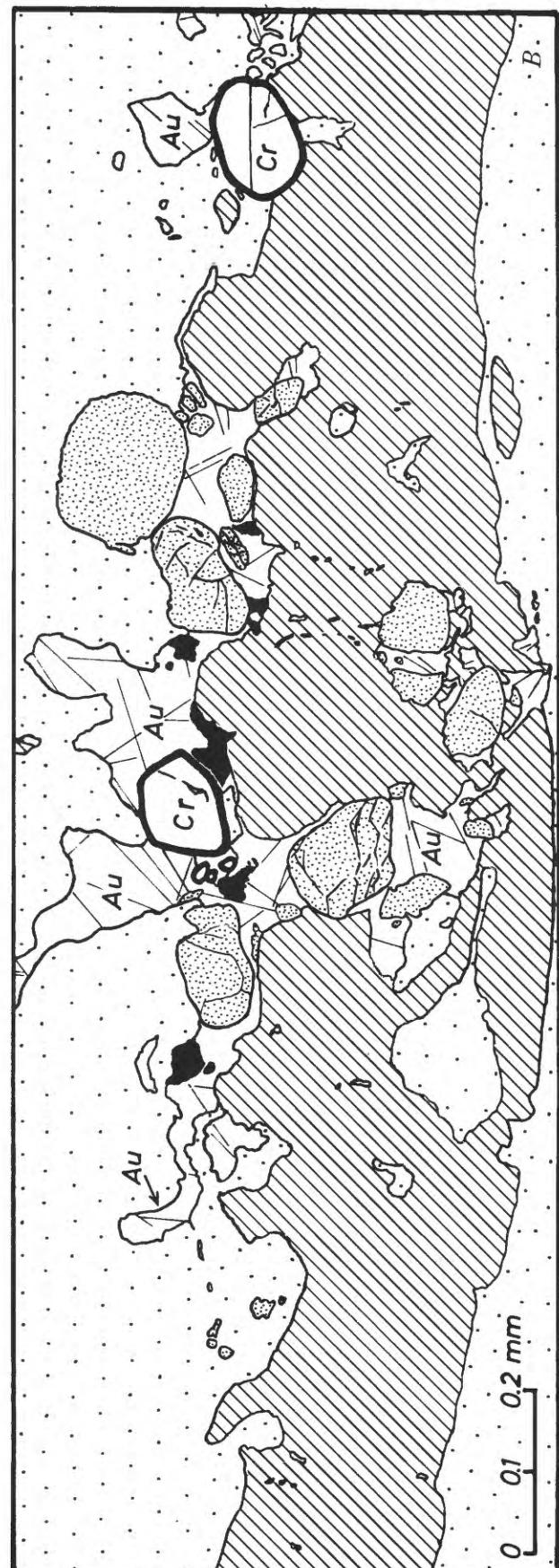


FIGURE 15.—Typical "carbon" footwall of a gold and uranium-bearing conglomerate horizon, showing distribution of minerals (Basal Reef, from Lorraine Gold Mines, Orange Free State). A, The carbonaceous material, gray displaying faint indications of a fibrous texture perpendicular to the bedding; at this low magnification ( $\times 120$ ), the angular bodies of uraninite sprinkled through the carbon matrix cannot be recognized. Oil immersion. B, Explanatory sketch shows distribution of principal minerals: heavy lined, carbonaceous matter; black, limnaeite; dense stippling, quartz; wide with scratches, gold (Au); strongly outlined, chromite (Cr).

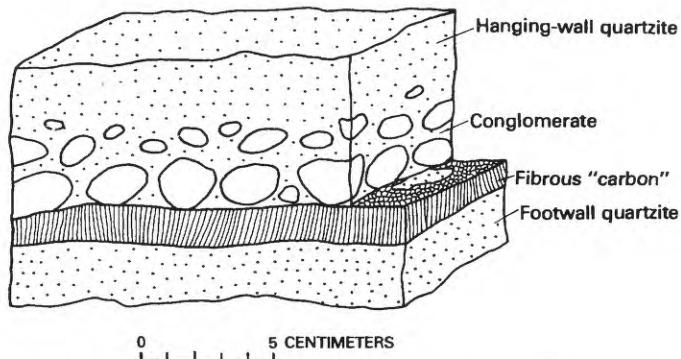


FIGURE 16.—Schematic block diagram of a "carbon reef" characterized by a seam of fibrous thucholite at the base of a conglomerate layer. The carbon fibres are approximately perpendicular to the bedding planes. Note that the pebbles are relatively loosely packed within the quartzite matrix, as is often typical of this particular reef type.

these aggregates to colonies of a putative symbiotic community assumed to consist of an algal partner and a fungus. A reconstruction of Hallbauer (1975) of this plant (fig. 19) does indeed suggest affinities to modern lichens. According to these authors, the carbon fibres represent the fossil remains of the columnar isidia of a lichenlike symbiont believed to have carpeted the floor of the primary depositional basin as extended colonies and to have trapped gold and other detritals by a sort of corduroy-table action.

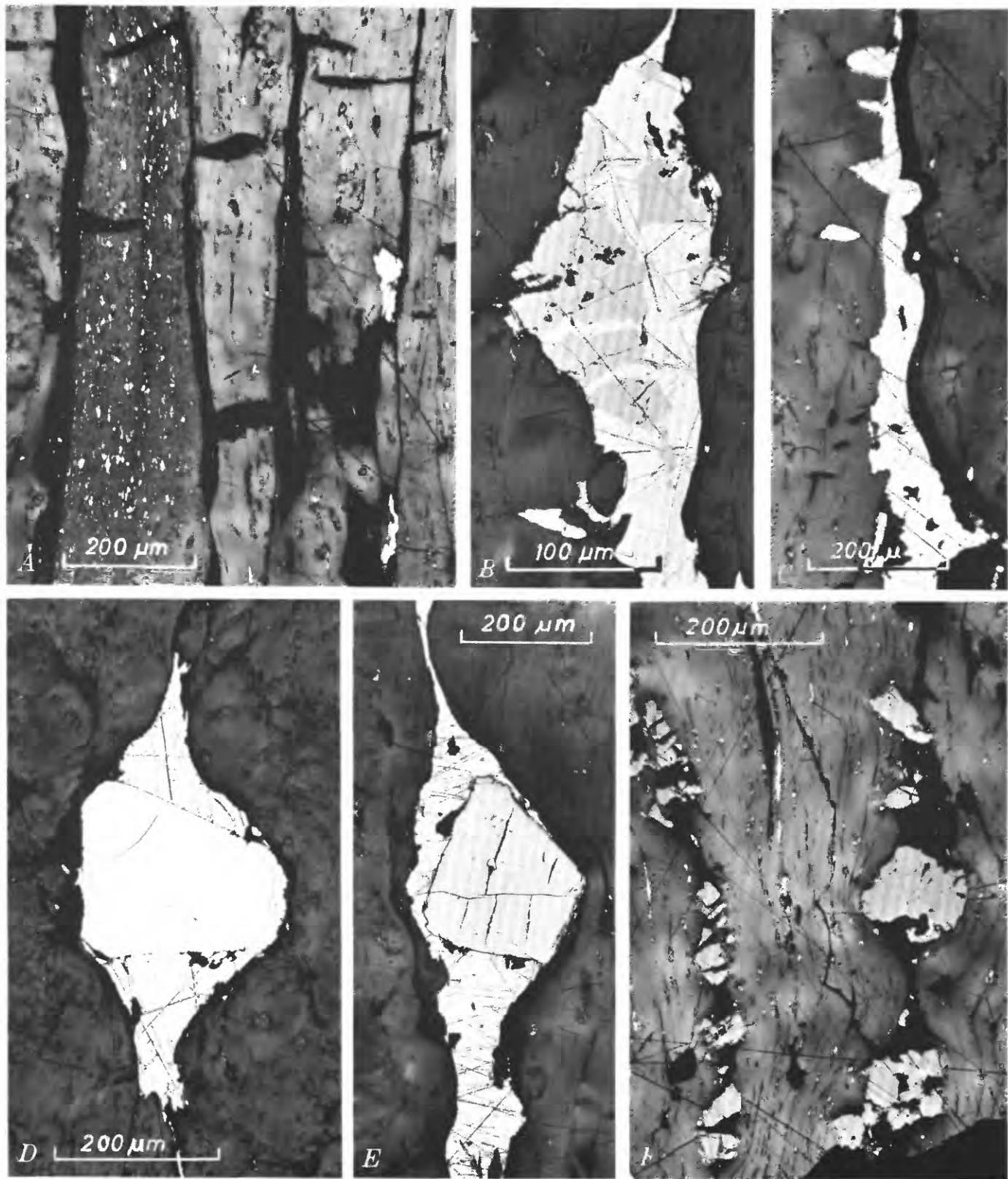
Contrary to the strong points raised by Hallbauer (1975) in support of this hypothesis, the student of the Precambrian familiar with both the topic and the evolutionary history of ancient life will have certain reservations as to the concept as a whole. These reservations may be summarized as follows:

1. The fact that this putative symbiont appears just once in the Earth's history must necessarily raise suspicion, since lichens could be expected to stand for a rather "conservative" type of organism, which should have appeared repeatedly in a similar morphological configuration in the paleontological record. This contrasts markedly with some organo-sedimentary structures attributable to algae, which have been found from almost the start of the sedimentary record until recent time.
2. Both the individual carbon fibres and their aggregates do not display a morphological differentiation suggestive of biological affinities when sub-

jected to reflected-light microscopy (figs. 17, 18). In particular, the rheologic fabric of the fibrous seams (fig. 17D-F) almost excludes the possibility that the fibres are structural remains of ancient plants. Ultramicroscopic evidence claimed to favor a biologic nature of the columnar carbon appears to be either inconclusive or largely overinterpreted. There is, furthermore, no compelling evidence as yet that the organic materials were primarily responsible for the economic mineralization of the bank. Notably, the textural relation between gold and carbon as presently observed (fig. 17) is indicative of a diagenetic and (or) metamorphic redistribution of the gold fraction within the carbon bands but definitely not of a primary precipitation of the metal by biologic agents. In general, both gold and uraninite are always found at the reef footwalls, irrespective of the presence or absence of carbon within a particular conglomerate horizon.

3. The putatively biogenic structures are preserved as massive coaly matter. If these fibres really represent structurally preserved remains of a lichenlike symbiont, the carbon content of the primary plant tissue was certainly smaller by at least one or two orders of magnitude than the carbon content of the present-day coaly layer. An original lichen carpet a few centimeters thick undergoing coalification would have been reduced to a carbon seam of millimeter scale, with any fibrous texture largely obliterated during the process of compaction. The strongly negative  $\delta^{13}\text{C}$  values of the carbonaceous material,  $-30$  permil and less relative to the National Bureau of Standards (NBS) isotope reference Sample No. 20 (Schidlowski, 1968), would furthermore suggest that the carbon of these layers derived, at least in part, from mobilized fractions of sedimentary organic material in which the light isotope,  $^{12}\text{C}$ , had been increased as a result of a "chromatographic" effect. Sedimentary organic materials normally display  $\delta^{13}\text{C}$  values close to  $-25$  permil (Degens, 1969; Eichmann and Schidlowski, 1975). Hence the carbon compounds accumulated in these bands are unlikely to represent isotopically undifferentiated, primary organic matter.
4. The occurrence of relatively advanced organisms such as lichens in the Witwatersrand System,

FIGURE 17.—Photomicrographs showing fibrous thucholite with characteristic mineral inclusions. A, Aggregate of columnar carbon; the individual columns are clearly separated by intracolumnar interstices and crosscut by wedgelike drag fissures. With the exception of numerous specks of galena within the second-from-left fibre, the aggregate is almost devoid of economic materials. B, C, Intracolumnar interstices filled with gold (white, scratched) and chalcopyrite (light gray); the gold also occupies wedgelike drag fissures perpendicular to the fibrous texture. D, E, Detrital pyrite grains in intracolumnar fissures; strands of carbon have flowed around the pyrite grains; gold (white, scratched) redeposited in the pressure shadows at each side of the pyrite inclusions. F, Sphalerite dragged apart ("boudinaged") in intracolumnar fissure. Note flow lamination within thucholite, with an elongated sphalerite fragment in the upper right corner tending to arrange itself parallel to the laminae. All examples from B Reef, Lorraine Gold Mines, Orange Free State. Oil immersion.



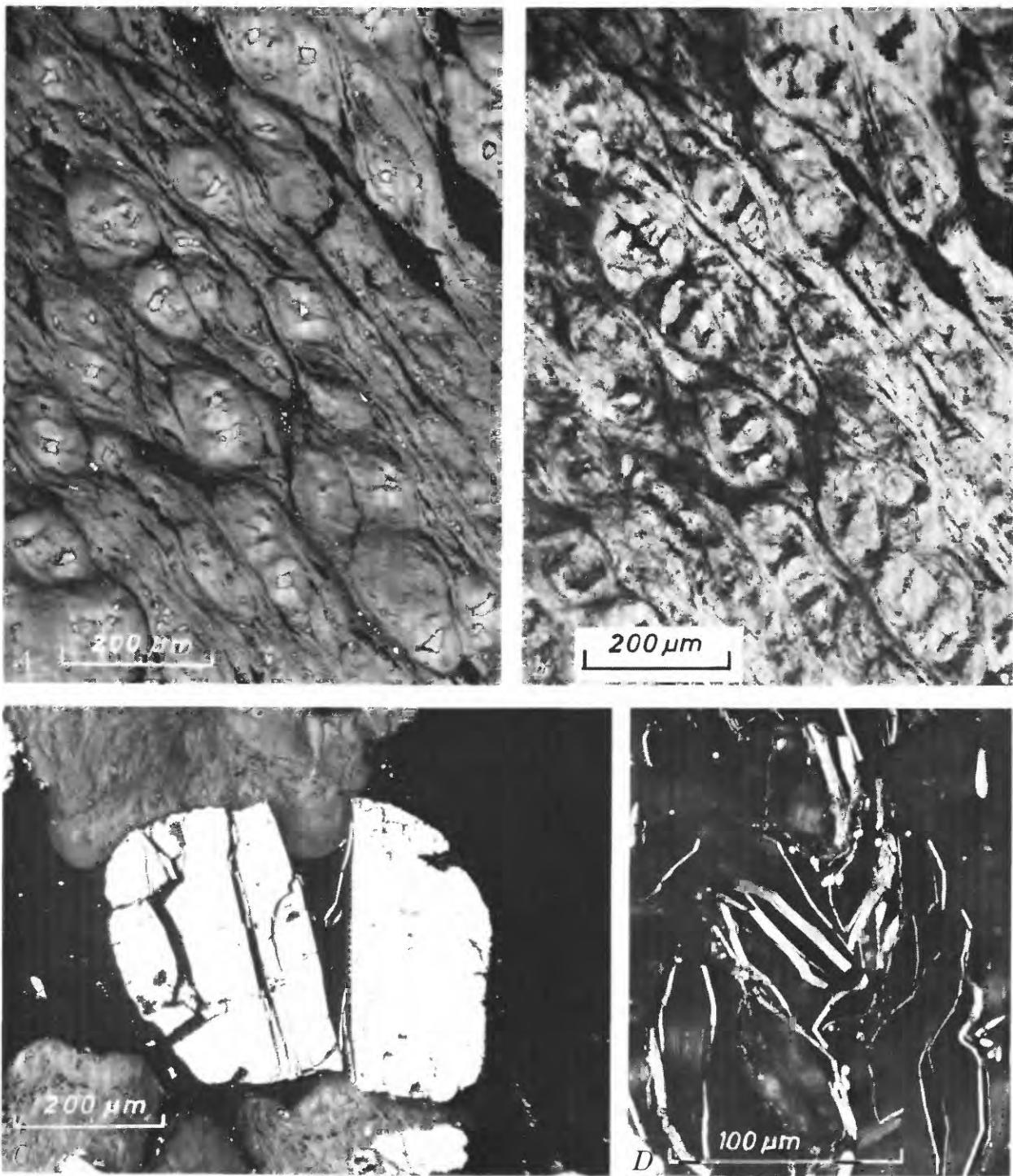


FIGURE 18.—Photomicrographs showing examples from various Orange Free State conglomerates. *A*, Fibrous carbon aggregate cut at an angle to main fibre direction; the outlines of the fibres show as lenticles on the surface of the polished section. Numerous relics of uraninite give rise to radioactive haloes within which the reflectivity of the carbonaceous material is markedly increased. *B*, With crossed nicols, a certain "pregraphitization" of the material is optically evident. *C*, Demonstration of extreme mobility of the carbonaceous matter; taking advantage of a pre-existing cleavage fissure, the carbon has wedged apart a detrital pyrite grain. *D*, Assemblage of scaly graphite in a quartz matrix. Oil immersion.

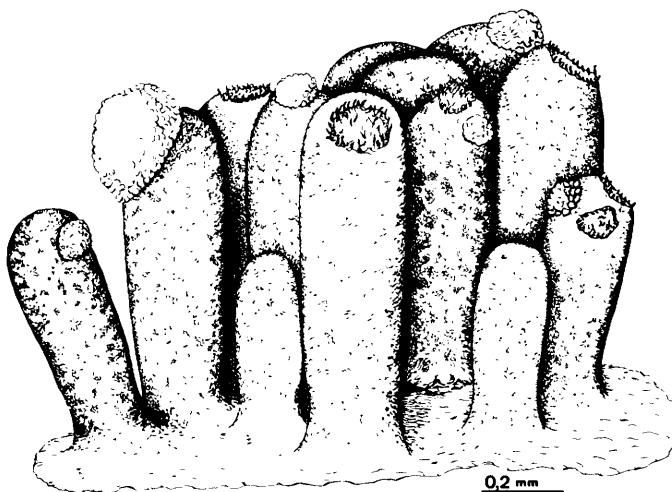


FIGURE 19.—Reconstruction of the lichenlike symbiont putatively responsible for the fibrous carbon aggregates at the base of reefs (Hallbauer, 1975, fig. 35; published with permission of the National Institute for Metallurgy, Minerals Science and Engineering). The present carbon fibres are believed to represent the fossilized columnar isidia of the original plant.

about  $2.5 \times 10^9$  years B.P., is incompatible with the currently established timetable of evolutionary diversification of ancient life. Although the timing of the advent of eucaryotic organization is presently under dispute (Knoll and Barghoorn, 1975), there is general agreement that this quantum step in evolutionary history cannot have substantially antedated the benchmark of some  $1.5 \times 10^9$  years B.P. (Schopf, 1974). It might, of course, be argued that this benchmark is not yet the definite one, but, for a variety of reasons, a major shift towards an earlier age is unlikely to be expected. It seems justifiable, therefore, to state at this stage that the appearance some  $2.5 \times 10^9$  years ago of a symbiotic association involving a fungal (that is, eucaryotic) partner is as improbable as the discovery of a dinosaur in the early Paleozoic.

In conclusion, the interpretation of the fibrous thucholite aggregates as fossilized plants must be rated as almost certainly incorrect and, in the opinion of this writer, may safely be discarded. The same applies to metallogenic concepts based on a large-scale participation of living organisms in the process of concentration of the principal pay metals, notably gold and uranium, in the blanket. Unless they are substantiated by clear-cut paleontological or biogeochemical evidence, restraint should be practiced in advocating such hypotheses, as the economic mineralization of the Witwatersrand may be reasonably explained in terms of "conventional" processes operative in the formation of placer deposits.

#### SOURCE OF THE CARBON CONSTITUENTS

It is generally agreed by most students of the Rand that the mobile precursors of the present hydrocarbons were, in all likelihood, indigenous to the sequence, stemming from the decomposition of contemporaneous organic material buried in the sediments. A derivation from locally overlying coal measures of the Karroo System, as envisaged by Davidson and Bowie (1951), may be ruled out primarily because of the buoyancy of migrating hydrocarbons. With the underlying basement rocks constituting a very unlikely source, there is no reason to seriously dispute the indigenous character of the carbonaceous substances.

Consistent with the assumed biogenicity of the material is its isotopic composition (Hoefs and Schidlowski, 1967; Hoering, 1967), the strongly negative  $\delta^{13}\text{C}$  values with a minimum at  $-37.1$  permil relative to the NBS standard suggesting genetic affinities to oil and asphalt rather than to "normal" kerogens. The  $\delta$ -values of carbon from Blind River fall within the same range (Eichmann and Schidlowski, 1974). The reported presence of a suite of amino acids (Prashnowsky and Schidlowski, 1967) requires confirmation by more sophisticated techniques allowing a differentiation between indigenous substances and possible contaminants. A biogenic derivation of the carbon is also supported by the occurrence within Witwatersrand rocks of microstructures suggestive of biologic affinities, as previously noted (Schidlowski, 1965, 1970b; Oberlies and Prashnowsky, 1968; Pflug and others, 1969; Hallbauer and van Warmelo, 1974). The most convincing evidence for biologic structures are myceliumlike filamentous aggregates described by Hallbauer and van Warmelo from ashed residues of carbonaceous matter. With such aggregates explained by their discoverers as fossil fungi (that is, eucaryotes) and their occurrence within the rock not yet confirmed by petrographic slides, there were necessarily strong doubts as to the indigenous character of these structures. However, according to E. S. Barghoorn (personal commun., 1975), the indigenous character of these aggregates cannot be doubted, but their interpretation as fungi is incorrect. If a tentative interpretation of these remains as fossil bacteria (Actinomycetes) turns out to be correct, these forms should contribute substantially to the stock of excellently preserved microfossils hitherto described from Precambrian rocks.

#### PROBLEMS OF URANIUM MINERALIZATION IN PRECAMBRIAN CONGLOMERATES

With our present knowledge of the regional geology of the Witwatersrand ore province (Brock and others, 1957; Brock and Pretorius, 1964) and with crucial

sedimentologic and geochemical evidence having piled up in support of a detrital origin of its economic conglomerates, the hydrothermal genetic concept (Davidson, 1957, 1965) for uraninite in the blanket may now be safely dismissed. Nevertheless, the occurrence of uraninite constituents in a detrital assemblage raises some questions, which are briefly addressed in the following discussion.

It is well known that uraninite is thermodynamically unstable in the present weathering cycle (Davidson and Cosgrove, 1955; Zeschke, 1959a; Dybek, 1962). Tetravalent uranium in uraninite is readily oxidized to the hexavalent state in oxygenic environments, thereby giving rise to the soluble uranyl complex  $[U^{6+}O_2]^{2+}$ , which is subsequently carried away by weathering solutions. Accordingly, uraninite is practically nonexistent in modern placers, the few noteworthy exceptions described (Steacy, 1953; Zeschke, 1959b; Trushkova and Kukharenko, 1961; Lang and others, 1962; Darnley, 1962) being quantitatively unimportant and mostly restricted to cold climates, where chemical weathering is retarded as a result of low mean temperatures. Moreover, with the exception of extremely old formations such as the Dominion Reef, the Witwatersrand reefs, and the conglomerates at Blind River and Serra de Jacobina, fossil uraninite placers are practically unknown throughout the sedimentary record. The occurrence, therefore, of uraninite in economic grades in lower and middle Precambrian detrital rocks must necessarily appear enigmatic.

Various efforts have been made to reconcile the presence of detrital uraninite in these ancient formations with the chemical instability of the mineral in the exogenous cycle. Ramdohr (1958a, b) conjectured that the  $UO_2$  constituents of the reef were still geologically young and relatively refractory when subjected to the stress of weathering and transport, because their lattices had not yet been weakened by endogenic  $\alpha$ -bombardment. Wiebols (1955), remarking on the local occurrence of tillitelike horizons within the Witwatersrand System, invoked the effects of a cold climate for the survival of the uraninite fraction. Several investigators have, on the other hand, entertained the view that the persistence of uraninite as a detrital mineral might indicate an extremely low redox potential of the ancient environment—that is, an anoxic continental weathering cycle (Louw, 1954; Liebenberg, 1955; Ramdohr, 1958b; Schidlowski, 1966a).

The existence of a basically anoxic atmosphere during the time of deposition of Precambrian uranium conglomerates,  $T \geq 2.0 \times 10^9$  years B.P. (fig. 20), is largely consistent with current concepts of atmospheric evolution (Berkner and Marshall, 1967; Cloud, 1968; Schidlowski and others, 1975; Junge and others, 1975;

Schopf, this volume, chap. B). Although uraninite is thermodynamically stable only at a  $P_{O_2} \leq 10^{-21}$  atm, Grandstaff (this volume, chap. C) has concluded that, at partial pressures of oxygen between  $10^{-2}$  and  $10^{-6}$  of the present level, the oxidation rate may be sufficiently slow for uraninite to survive under the set of environmental and physiographic conditions that most probably governed the deposition of the Witwatersrand rocks and related uranium-bearing conglomerates. The ubiquitous presence of detrital pyrite in the reef could be likewise ascribed to low environmental oxygen pressures, since sulfides are also rapidly destroyed by oxidation.

Oxygen concentrations of the above range, particularly those close to the so-called "Urey level" of  $10^{-3}$  of the present partial pressure of oxygen, have long been regarded as typical for the geological time span prior to  $2 \times 10^9$  years ago. The concept of an ancient anoxic weathering cycle would therefore provide a general explanation for the worldwide occurrence of pyrite-uraninite placers during the early history of the Earth and is also consistent with an impressive array of other relevant evidence (fig. 20, lower part). At this stage, the postulate of a substantially lower redox potential in the ancient environment is probably the most reasonable way to account for the origin of these deposits.

It might be argued, of course, that during processes of rapid sedimentation any debris will stand a fair chance of survival, irrespective of its thermodynamic properties. Hence, an ultimate acceptance of the "placerist" concept for the uranium mineralization of Precambrian conglomerates would not necessarily imply that the occurrence of uraninite among the detrital minerals is accepted as conclusive evidence of an anoxic continental weathering cycle. Such reservations apparently would be supported by the fact that the Witwatersrand System exhibits, at least in part, obvious molasse features, intercalated graywackes, unsorted boulder beds, and the like, indicative of rapid burial of weathered debris from the surrounding borderlands.

However, in the opinion of this writer, these arguments may be severely weakened by the following considerations: With the age of all known uranium-bearing conglomerates more than  $2 \times 10^9$  years and the mass half-age (life) for sedimentary rocks lying in the range of 500 to  $600 \times 10^6$  years (Garrels and MacKenzie, 1971, p. 270), only between 6 and 12 percent of the total sedimentary column deposited  $2 \times 10^9$  years ago can be expected to have escaped subsequent destruction. For sediments deposited during Witwatersrand time,  $2.5 \times 10^9$  years ago, the remaining proportion would approach 3 percent as a minimum. Within these scanty relics of an originally huge sedimentary column, detrital uraninite is apparently well represented on a worldwide scale, whereas all younger sediments, whose chances of

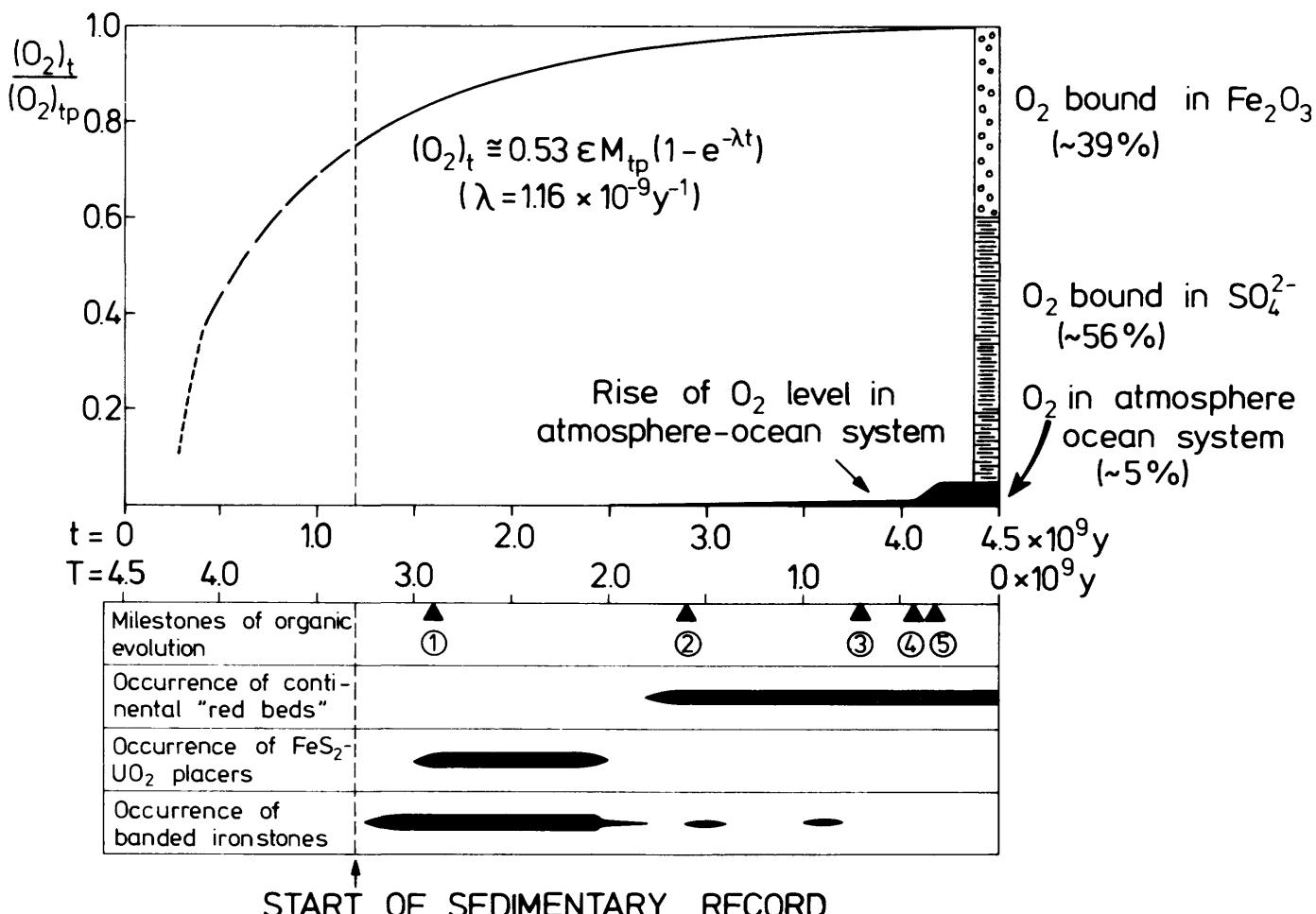


FIGURE 20.—Evolution of the Earth's total budget of photosynthetic oxygen, based on the terrestrial  $^{13}C$  balance and current concepts for the accumulation of the stationary sedimentary mass as a function of time (Schidlowski and others, 1975, fig. 14; published with permission of Elsevier Scientific Publishing Co.). Stationary oxygen reservoirs existing at times  $t$  are expressed as fractions of the present reservoir  $(O_2)_{tp}$ .  $M_{tp}$  is the present sedimentary mass with  $tp = 4.5 \times 10^9$  years.  $T$  is time before present in years. Column on right side shows partitioning of oxygen between the "bound" and "free" reservoirs in the present budget (Li, 1972), with molecular oxygen accounting for about 5 percent only. Note disproportionation between bound and free reservoirs during the Precambrian and early Paleozoic, most probably due to a short circuit of the ancient oxygen cycle in the Precambrian seas. Tentative rise of free oxygen level in atmosphere-ocean system (black field) is inferred from paleontological and geological evidence shown. Milestones of organic evolution indicated are (1) appearance of oldest algal bioherms (photoautotrophic blue-green algae), (2) appearance of eucaryotic biota, (3) appearance of oldest eumetazoan faunas, (4) life conquering continents (Upper Silurian); (5) appearance of exuberant continental floras (Upper Carboniferous).

preservation grow exponentially with decreasing geological age, definitely lack these deposits. Because the Phanerozoic record, which accounts for roughly 50 percent of the total sedimentary mass, has been extensively examined by generations of field geologists and because prospecting for uranium has been carried on at a large scale for decades, it seems justified to conclude that uraninite placers did not form during the later history of the Earth. Likewise, pyrite conglomerates have not been reported from younger geological formations, whereas the association sulfide-uraninite is characteristic of Witwatersrand-type detrital gold-uranium deposits.

It is hard to conceive that geological environments favorable for the formation of pyrite-uraninite placers should not have existed repeatedly in the geological past; notably, rapid burial of weathered debris has occurred during numerous sedimentary cycles, as illustrated by the presence of numerous graywackes and other poorly sorted sediments in younger geological formations. Accordingly, it is very difficult to account for the complete absence of pyrite-uraninite deposits from younger sediments without invoking changes in the redox potential of the environment. Confinement of pyrite-uraninite placers to the early and middle Precambrian would certainly be too much of a coincidence if the

formation of such deposits were governed exclusively by geological factors.

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**DISCUSSION FOLLOWING PAPERS BY DRs.  
SCHIDLOWSKI, HALLBAUER (PAPER 3<sup>4</sup>)  
AND LOIS NAGY**

[At the Workshop the paper by Zumberge, B. Nagy, and L. Nagy was presented in two parts. Lois Nagy spoke first and confined her remarks to the Precambrian micropaleontology of the Vaal Reef. Bartholomew Nagy followed and covered the rest of the subject matter.]

*Schopf:* I would like to briefly address myself to the last two papers in particular. It seems to me that those of us who are concerned with the early history of life have two primary tasks incumbent upon us if we are reporting objects that we interpret as Precambrian fossils. First, it must be established that these objects are indigenous to the rock, that they are syngenetic with deposition, that they are part of the rock and date from the time of original sedimentation. And secondly, we must establish that these objects are of biological origin; regardless of whether we interpret them as fungi, algae, or what have you, there must be a set of criteria that allows us to say without doubt that these are biologic objects, rather than aggregations of organic matter of some other origin. Finally, I would point out that, as I think is probably obvious to most of us, this is a very difficult sort of game to play. It is often, or at least in some cases, not easy to know whether the structure you are looking at is biological in origin or not: it is usually somewhat easier to establish that the object is indigenous to the sediment. It should be said that this field is only about ten years old, that there is very much to be learned, and the generalizations are not particularly well established. So it is possible to have strange things turning up.

With regard to Dr. Hallbauer's paper, it seems to me that he has established that there are fibrous aggregates of organic matter within the Witwatersrand rocks. I think that is well established indeed. Personally, I do not believe that he has established at all that these structures are biological. Without getting into the great details involved, it is my opinion that the branching structure that he has shown differs from all biological branching structures of which I am aware. It is not a fungal type of branching structure, at least those of which I am aware, and I have looked at quite a number of modern fungi. These tubes that he has shown are anastomosing, they are of variable diameter, sometimes you have large ones, sometimes you have small ones, sometimes you have multiple branches, but, as many of you know, in general in biological structures you get sort of a V-shaped branch, and these tubes do not generally anastomose back together again. On the slides that I saw, it seem-

ed to me that I could see several examples in which there were multiple tubes coming off and then merging back together. Well, maybe that's the way primitive organisms were, but there is nothing like that known today as far as I am aware. Secondly, I do not understand at all, as Dr. Schidłowski pointed out, how such a presumably delicate structure can be preserved. These are not permineralized organisms, if they are organisms. They are preserved in a sedimentary environment, I guess mostly analogous to a coal, and in coals one does not see structures of this type either. Even in coals that you know are of plant origin, one doesn't get such delicate things as presumably these organisms were. So it is my guess at this time, that although these are assuredly indigenous to a Precambrian sediment, I believe that the case is very far from established that they are biological.

With regard to the interesting things that Dr. [Lois] Nagy has presented, I believe that neither the biogenicity nor the Precambrian age of these structures has been established. We see interesting particles in scanning electron micrographs. The carbonaceous nature of these structures is very well established, but how do we know that they are of Precambrian age? This is a problem with looking at microscopic objects with a scanning electron microscope: it is a terrific tool for certain sorts of studies. But if you want to prove those objects are of Precambrian age, it seems to me that the best way to study them is in thin sections. Furthermore, it seems to me that if they are biologic in origin they ought to have the sort of regularity that one sees in modern biologic systems. I would tend to have interpreted the objects that Dr. Nagy showed as probably bits and pieces of organic material from the Witwatersrand rocks rather than being demonstrably remains of micro-organisms. And I think this is also true about Dr. Hallbauer's. In my opinion they seem to me to lack the type of biologic regularity that one sees in biologic systems. I am sorry to say all these things because I do not appreciate standing up and pretending that I am the great expert on all these types of things, and I don't want to be impolite to either my colleagues or anybody else. It's darned hard to be sure about these sorts of things, and I think it behooves those of us who deal with Precambrian fossils to nail the case down just as firmly as we possibly can, to establish that these things are both Precambrian in age and of biologic organization.

*Hallbauer:* I just want to point out that in scanning electron photomicroscopy if objects overlie each other, especially filaments, in the photograph it looks as if they come together again. This I know is very difficult to show in a set of photographs. In addition, I want to

<sup>4</sup> See editors statement at start of Discussion, Chapter M.

say that to me there is no doubt that these things are of Precambrian origin. They are in the rocks, and there is no reason to believe that they were introduced at a later stage. With regard to the other structures, I am not a biologist, I can only say what other biologists have told me with regard to the filamentous structures. They have been identified as organisms by Preston Cloud, although he had reservations and said, "How did they get into the rock?" Some biologists say they are biological structures, some others have reservations, so it is very difficult to decide from my point of view as a mineralogist.

*L. Nagy:* I must say that this is one of the nice things about small scientific meetings, when you can get together with your colleagues and they sort of disagree with you. This helps keep you on your toes, and I think that what Bill [Schopf] said about my work is indeed quite true. One of the things that I should have pointed out is that I have looked at thin sections of the carbonaceous matter, but unfortunately it is so very opaque you really can't see too much in it. So I've got to scratch my head and go back to the drawing board to see how we can get petrographic thin sections that are made in such a way that we can see structures in them. He had a very valid point. I have never published a paper of any structures that have not also been found in thin sections. And this work will not be published until structures are indeed found in petrographic thin sections.

*Robertson:* I have a comment for Dr. Schidlowski with reference to the brannerite reaction. Ramdohr called that the "Pronto reaction" from the Pronto mine in Canada. That's the farthest south of the Elliot Lake deposits and is the one which would be more subject to later metamorphic events. In the deposits farther north there are a number of uranium-bearing beds. Information is really rather scanty, but I think the indication that we have is that the brannerite content varies within the bed relative to uraninite and it varies from bed to bed, so that where beds are stacked upon one another, we do not in fact find the same brannerite-uraninite ratio. This might suggest that some mechanism other than metamorphism was responsible.

*Schidlowski:* I would suggest that the amount of  $TiO_2$  present is also one of the prerequisites. So metamorphism is probably only one factor. That would be my answer to this.

*Skinner:* I am not a biologist either, but I think that I have seen Dr. Hallbauer's structures before. I have seen them in coke. I would submit that perhaps much of what he is seeing is an artifact of vesiculation. As for the filamentous structures, I wonder if one should not look for an origin for them in fibrous growths of

the kind that one sees in asbestos. And I would suggest that perhaps a line of research to follow on that would be single crystal X-ray crystallography and see if some of your filamentous structures are not in fact single crystals.

*Hallbauer:* Well, there again I can only repeat what I was told by biologists. If you can show microscopic preparations of these filaments to a mycologist and he recognized them immediately as fungal structures, then there is nothing for the geologist to do.

*Skinner:* Would you permit me to tell a short story? I was working one time in Australia in some shallow lakes, and it was in a region where a number of algal colonies form little biscuit-shaped calcium carbonate growths. In the lake I was working in I saw something that looked a little bit familiar. It was about the size of a thumbnail and had a rounded shape and I thought, "Hello, here's another one of these things, but a different shape." So I gathered a bunch of them, took them back to the university, and gave them to the biologist who looked at them and said, "Yes that's true. That's just what they are. They are some kind of algal growth. They are new, and we'd like to describe them." They took about about a dozen of them and dissolved the calcium carbonate away, and then described the gelatinous cellular structures that were left. They had even gone to the point of writing a paper and submitting it to the Royal Society of South Australia, at which it had been accepted. I mentioned this one day to Martin Glassner, who many of you know is a paleontologist, and he said, "What did that thing look like?" So I described it. He said, "Have you got any more?" And I pulled one out of the drawer, and he said, "Well, I know what that is, that's a gastrolith from a crawfish, and the cormorants have been eating those things." And true enough, I'd picked these up underneath a log where the cormorants had been sitting and passing them through their bodies and dropping them into the lake. So I am not sure that showing it to a biologist always gives you a definitive answer.

*Hallbauer:* In that case, to what authority do you go?

*Schidlowski:* Originally I wanted to briefly comment on Dieter's [Hallbauer] talk, but when Bill [Schopf] stood up I got a little bit mixed up, so I propose to give my contribution as if I hadn't heard Bill's comment. I would have said, . . . I would personally differentiate clearly, and I am still convinced that the fibrous stuff is not biogenic, at least the pictures which I have seen have not yet convinced me. Whereas, in contrast to Bill, I would have been inclined to acknowledge the other stuff. I also remember that Pres Cloud when we met him—Bill, you were present in Leicester—that Pres said this is certainly biogenic stuff. And maybe

you have not seen Dieter's last published pictures in that South African journal. Perhaps you should give it a look and think about it. But, anyhow, you understand more of that than I do, so it would be just a row between Pres Cloud and you.

*Schopf*: You know, this is getting to be a little bit like what I refer to as anecdotal science. If I recall the conversation of which I believe you are speaking, Pres Cloud's point was that in Dr. Hallbauer's paper in Precambrian Research, Cloud interpreted the filaments shown in optical photomicrographs purporting to show cells to be certainly biological and certainly contamination.

*Schidlowksi*: Yes. He said they looked too modern, and he suspected they were contaminations, but anyhow they were biologic. We should perhaps split the problem in two portions. First, is that stuff biogenic, and second, is it indigenous? I would yield to the first one, and I will concur with you that perhaps the second point needs to be proved.

*Myers*: I would simply like to ask how the temperatures of oxidation were measured in Dr. Hallbauer's oxidation procedure.

*Hallbauer*: There was no accurate measurement of temperature. It was measured on [some?] element nearby. So it was plus or minus 500°C, say plus or minus 10 to 20 degrees.

*Ruzicka*: Dr. Schidlowksi mentioned extinction of hydrocarbon within one or around one grain of uraninite.

*Schidlowksi*: What do you mean by "extinction"?

*Ruzicka*: This is what I am going to ask you, because we observed that if you turn the table under crossed nicols . . .

*Schidlowksi*: Oh, you refer to the reflection pleochroism.

*Ruzicka*: Yes. You get simultaneous extinction in large areas. How do you explain it?

*Schidlowksi*: I would say there must be a certain pre-graphitization stage in which the carbonaceous material tries to approach a certain order, finally, of course, aiming at graphite but not reaching it at the moment.

*Ruzicka*: This is your explanation?

*Schidlowksi*: Yes. But I think there are other experts who have better explanations to offer, but I think as a first approximation I should be right. Actually, the stuff I showed you is not graphite at all. Many people have found traces of graphite in the reefs and usually associate it with that carbonaceous stuff, but if you would do some X-ray work you would not get graphite patterns. By the way, in reflected light microscopy it is very easy to distinguish between carbon in the widest sense and graphite in the proper sense, as anybody familiar with this type of work will admit.

*Hallbauer*: In some of Dr. Schidlowksi's recent papers he describes the occurrence of pristane and phytane as chlorophyll derivatives in the carbonaceous material. Now, how does he explain the presence of these things in hydrocarbons which can be floated through the rock and were polymerized by uranium?

*Schidlowksi*: Pyrolysis gas chromatography was done on the carbonaceous matter, and we got a whole pattern of hydrocarbons. We used mild pyrolysis, so we got all this stuff which is volatile up to let us say 250°C. Among these graphs we got small peaks in the place where one could expect phytane and pristane. We took these peaks as real, but we did not confirm them with mass spectrometry, so I wouldn't put my head on the block for this. When we get a chance to go deeper into the matter, we will certainly come nearer to the truth. I think it is justified to record that at least traces of these degradation products are actually present. If that stuff is mobilized, it must have been taken along with the other hydrocarbons when they migrated through the sequence.

*Paul L. Weis, U.S.G.S., Reston, Va.*: Dr. Schidlowksi, one gets the impression in looking at your photomicrographs of the progressive decomposition, disintegration, or disaggregation of uraninite grains that this is a process that takes place over a considerable period of time. The implication may be that this is not a syn-genetic effect, but something that takes place afterwards, in which case it answers at least part of your comment that, whether it is biogenic or not, it can't be an original constituent of the rock. The other aspect of this question is, as the uraninite is attacked by the hydrocarbons, where does the excess uranium go?

*Schidlowksi*: Into the brannerite, for instance, but here reservation is justified, of course. If we admit that the [carbonaceous] stuff has migrated, any one is justified in asking, "Where does it come from?" And those familiar with the whole story know that Davidson originally contended that the stuff came from overlying Karroo coal measures. But hydrocarbons are light, and because of that they do not sink into basins, and there are some other points against a source in the coal measures. If you rule out the Karroo measures, we must look for sources of the carbon. We know what is below and we also know what is above in the sequence, and there are actually no strata which contain massive concentrations of reduced carbon in any modification. So I think, although we could not settle the topic definitely, we can narrow it down and say that, with 90 percent probability, the stuff is indigenous in the widest sense, although I must admit there is a slight possibility still open for extraneous introduction.

*Irving Breger, U.S.G.S., Reston, Va.:* I'd like to make a few comments for all that they are worth. The organic matter associated with the actual uraninite grains and the nature of that type of material is in question on the basis of what was said here. First of all, what we have found is that organic matter that has been subjected to lengthly irradiation by uranium and its daughter products tends not to go to graphite, but instead takes on a diamond-type structure, for the simple reason that the graphite type lattice tends to buckle and you get a more diamond-like structure, which is harder. I suggest also that when we talk about organic matter and this type of structure in the future, it would be very nice to know that this organic matter contains more than just carbon. Is there oxygen in it, too? How much hydrogen? Just knowing that it contains carbon really doesn't give us much to work on.

The second thing I would like to say is really a suggestion for your consideration. Let's assume that we have a sedimentary situation in which we have been washing down, gold, uraninite, and other heavy minerals, and we have a rather quiet situation in which we can also get perhaps some life forms developing. Let's assume then that these life forms were decomposing, because obviously they decomposed, too. If you are looking for a source of methane, it must be there because we had reducing conditions. I think we are all agreed that there was no oxygen about at the time, so that if you are looking for methane to polymerize you don't have to go to the Karroo Formation or anywhere else; it was indigenous. Although I don't think that's the answer, either. But let's assume that the uraninite and other mineral grains have now come down, they are buried with this organic matter at the bottom of a shallow basin, and the organic matter, being what it is, probably acidic in nature, tends to attack the mineral grains, perhaps through crevices, and it seeps up into them in such a way that it makes up those channels that are backfilled that you have shown us in your excellent pictures. What more does one need? Do you have to go to very sophisticated explanations? It's the same material, as far as I can tell.

There is another question I'd like to raise here with you and that is: In some of the pictures you have shown, you very clearly indicated that the organic matter was highly metamorphosed in the sense it was much more highly reflective than you would have expected. Why wasn't the same organic matter in the crevices of the actual uraninite grains just as equally reflective as that that was not associated with uraninite grains?

*Schidlowski:* I think they are photographed in a position where the reflection pleochroism was responsible

for a dark strip, so if you turned your microscope stage it would have cleared up.

*Breger:* I don't think so. I think you have picked an easy answer out of it.

Another thing I would like to bring up is the time sequence. What is the paragenesis of all this? Everybody assumes that it's happening today, just this minute. What is the time sequence in terms of the introduction of the uraninite grains into the sedimentary basin, the time that it has taken for degradation of the uraninite grains by the decomposing acidic organic matter so that perhaps the grains are cracked and carbonaceous material has moved up into and backfilled it? I'm not sure that this is a replacement in the truest sense of the word.

*Schidlowski:* How would you account for that? What alternative would you offer?

*Breger:* Let's take a uraninite grain that is immersed in a colloidal type organic goo, sitting at the bottom of a basin, and this uraninite grain has a few little microcracks in it and attack occurs in this goo at the cracks and the organic matter seeps into the cracks and breaks up the uraninite and perhaps dissolves it.

*Schidlowski:* Just in a mechanical way?

*Breger:* Mechanical and partially chemical, too.

*Schidlowski:* Why does the organic matter not penetrate into the cracks of other minerals, for instance, the ubiquitous pyrite, the chromite, the zircon, and so on? I think this imposes certain limitations.

*Breger:* I think we are missing the chemistry. First of all, I don't think organic matter attacks zircon. Secondly, I don't think it attacks pyrite, but it does attack uraninite.

*Schidlowski:* It may invade in cracks in any other material. So if it invades in cracks, if it were a purely mechanical process, I would suggest that all minerals were in equal position to get a little bit of that stuff.

*Breger:* But I didn't say purely mechanical; I said mechanical and chemical. I said that the acidic environment will lead to an attack on the uraninite grains.

*Schidlowski:* Actually, the point is that only in the uraninite particles do we have that stuff and no other cracks. So, okay, any other questions?

*Breger:* I don't know if there were so many questions as statements, maybe not even of fact. I would also like to suggest that if we had uranium in some of these plants—we'll assume that they are plants for the moment—it sure would mess up their life process, so that all I can suggest here is that if the uranium did get involved with these plants that it had to come after they were dead. They served as a hollow zone in which precipitation, either chemical or mechanical, could

take place. I do want to say, though, that I thought these two papers were just excellent.

*Schidlowksi:* The mechanism which you alluded to now is, I think, by far the most probably reason for bringing about a uranium-carbon association. It worked, for instance, in the Colorado-type deposits, but my personal opinion is that in the Wits deposits things were a little bit different. So I would differentiate between Colorado-type uranium-carbon associations and Witwatersrand type.

*Breger:* I wish you wouldn't even mention them in the same breath.

*Hallbauer:* There is one remark that was made about these structures being similar to coke. We have thought about this possibility, and we've done some work and we are convinced that coke looks totally different. The pores in coke have a totally different structure from the ones shown here. They are irregular and you never have an isotropic structure as the one shown. Then about the preservation of these, let's call them organisms, there is no reason why they shouldn't be preserved in such an excellent state because the Witwatersrand rocks have a history of very minor tectonic movements and most of the rocks have not been folded. There is only very slight metamorphism, and most of the organisms have been preserved in the matrix of the quartz-pebble conglomerate, which acted as a shield, so there is no reason why they should be squashed.

*Whiteside:* I would just like to make one comment, and that is that there has been quite a lot of tectonism in South Africa. In the Witwatersrand one gets faults of displacements of two miles; around the Free State Goldfield there are others probably with even more displacement. There is a horizontal displacement I know of of 3,000 feet in the East Rand, and so on. I think the rocks have been subjected to quite a lot of tectonism. And along the edge of the Free State basin the lower portion of the upper Witwatersrand is overturned.

*Hallbauer:* What I wanted to say was that there was very little folding of the Witwatersrand strata, and Dr. Whiteside is quite right in pointing out these various structures. Indeed, in the vicinity of faults, the carbonaceous material is completely powdered and does not show these structures. I have just selected parts which have been preserved better than the other ones.

*Graham Curtis, Gulf Mineral Resources:* Dr. Hallbauer, I understand when you processed your carbon you heated it up to 500°C?

*Hallbauer:* Yes.

*Curtis:* Well, did you gather any of the gases or any of

the materials that might have been driven off of the carbon at that time?

*Hallbauer:* No.

*Curtis:* In coalification, either natural or in making coke, there is always something left over to gather. It would seem that collecting these residues would be important and might be a technique for determining whether or not the carbonaceous material is biological.

*Hallbauer:* That is a technique that is to some extent used by Dr. Schidlowksi's team; they heat the carbonaceous material and recover the volatiles.

*B. Nagy:* The first paper after lunch, I presume, is going to be given by me, which will be a continuation of the last paper. I am going to report on the combined pyrolysis, gas chromatography, and mass spectrometry of Vaal Reef samples heated to 450°C in  $10^{-6}$  torr vacuum.

*Armstrong:* I would like to ask about the radiation damage to the carbon. Is there any technique to measure the amount of radiation to be expected over a certain period of time or a method for measuring the amount of radiation damage to carbon?

*Schidlowksi:* To whom was the question addressed? The only radiation damage I could think of is just the increase in reflectivity. At the moment I can't recall any other. Can you?

*Armstrong:* There was a slide I thought which showed some radiation damage.

*Schidlowksi:* Yes, you get cracking halos, but they are a little bit different. Mostly they must be ascribed to zircons, because if zircons undergo metamictization that results in a volume increase, then all the neighboring stuff is blasted. As for radiation alone, some little blasting halos have been observed around uraninite grains, but I think these phenomena are not so distinct.

*Charles Miles, Protocol Corp., Denver, Colo.:* For Dr. Schidlowksi. You studied uraninite with carbon. Was there uraninite elsewhere, but nearby, in the samples you studied that was not associated with carbon?

*Schidlowksi:* Yes.

*Miles:* Is it the same size material?

*Schidlowksi:* They are all about the same. I would say between 80 microns and 160 with the average about 120. Fractured grains are, of course, smaller, and the grains which I referred to as relict bodies within the carbonaceous matter, they are between 40 and 50.

*Sims:* The aspect of preservation potential of these carbon seams seems to be bugging a lot of people. I'd like to suggest that the process of sand-wave migration, accompanied perhaps by rapid silicification of the matrix of the sand, could in actual fact contribute to

the preservation and prevent subsequent destruction of fine filamentous, perhaps organic, products preserved in these carbon seams. Perhaps this thought is way out.

*Joel Leventhal, U.S.G.S., Denver, Colo.:* Since there seems to be a lot of doubt about the biological nature of these things, I would like to throw a little bit of doubt into the hydrocarbon situation. Living organisms in general don't show large amounts of hydrocarbons, so I would say that's not a very good source for hydrocarbons in Dr. Schidlowksi's presentation. Methane is the abundant hydrocarbon that micro-organisms produce. The problem is how do you get enough of this gaseous methane to stick around long enough to be polymerized? It's going to be in aqueous solution, and it's probably going to float out again. So I think we may be back to the kind of thing that Irving Brager is suggesting, that the stuff is just setting there in a soup of carbon. The -30 permil carbon values don't sound particularly unusual; they are not like the -80 that's often quoted for natural gases.

Let me then ask a question: Is there carbonaceous material without uranium? If so, perhaps the comparison of the carbonaceous material without the uranium compared to the carbonaceous material with the uranium can be used as an index of the origin, or the radiation damage, or the diagenesis, or any of these things.

*Schidlowksi:* I have never seen carbonaceous matter without even at least faint relicts of uraninite, but I recall that Ramdohr once mentioned that he had seen such stuff but not in a proper carbon footwall of the reef but from somewhere higher in the reef. I am not sufficiently familiar with the occurrence to comment on it. At least we cannot rule out that some carbonaceous material occurs without uranium, so there must have been other fixation processes.

You also mentioned the isotopic composition of the carbon. I think there is ample evidence by now that undifferentiated organic matter on average yields around -25 permil. If we get values less than -30, if I remember correctly the Witwatersrand goes to -37,

then I think the likelihood is rather large that the carbon is no longer undifferentiated and that the light-carbon isotope has been increased by what I called a "chromatographic effect."

*Leventhal:* I would say -37 is definitely not light enough to prove that it hasn't gone through some kind of cracking rather than a biological process. I think most petroleum people would say -37 just looks like metamorphosed organic matter, not freshly deposited material.

*Schidlowksi:* Just to illustrate this point, we had a core from the Transvaal Dolomite, and there were carbon partings in it. We sampled the carbon partings and the rock between the partings, which contained about 0.5 percent organic matter. The finely dispersed carbonaceous material within the rock yielded about -26 permil,  $\pm 4$ , whereas from the carbon partings we got values as low as -40 permil. We concluded that a secondary effect must have been responsible for this accumulation of carbon twelve. I think our conclusion is fairly reasonable, at least in this case.

*Hallbauer:* I just want to mention that there are carbonaceous seams without gold and with very low uranium content. Normally they are of no interest to the mining industry, and therefore they are quite easily overlooked.

*Schidlowksi:* The example I was talking about was in the Transvaal Dolomite, not in the Witwatersrand.

*Ruzicka:* We analyzed hydrocarbon in which were embedded idiomorphic grains of uraninite, and the only component we found in this hydrocarbon was sulfur, about 24 percent sulfur but nothing else, not even small amounts of uranium or thorium.

*Schidlowksi:* Elemental sulfur?

*Ruzicka:* Yes.

*Schidlowksi:* At 25 percent it must have looked yellow all over. That is in the Blind River carbon?

*Ruzicka:* Yes.

*Schidlowksi:* Is somebody working on that at the moment?

*Ruzicka:* Yes.



# Some Aspects of the Development of the Vaal Reef Uranium-Gold Carbon Seams, Witwatersrand Sequence: Organic Geochemical and Microbiological Considerations

By JOHN E. ZUMBERGE, BARTHOLOMEW NAGY, and LOIS ANNE NAGY

GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN  
QUARTZ-PEBBLE CONGLOMERATES

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## GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN QUARTZ-PEBBLE CONGLOMERATES

### SOME ASPECTS OF THE DEVELOPMENT OF THE VAAL REEF URANIUM-GOLD CARBON SEAMS, WITWATERSRAND SEQUENCE: ORGANIC GEOCHEMICAL AND MICROBIOLOGICAL CONSIDERATIONS

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#### ABSTRACT

The solvent-insoluble organic matter (kerogen) that is the predominant syngenetic component of the Vaal Reef carbon seams was analyzed by combined high-vacuum pyrolysis-gas-chromatography mass spectrometry, combined electron microprobe-scanning electron microscopy and electron paramagnetic resonance spectroscopy. The pyrolysis results indicate that the Vaal Reef "carbon" is a random polymer-type matter consisting mainly of aromatic and short-chain aliphatic hydrocarbons, as well as aromatic and other organic-sulfur constituents. Scanning electron microscopy revealed coccoid and filamentous microstructures in the carbonaceous seams. These structures are conceivably of biological affinity and may be fossil blue-green algae and (or) bacteria. Gold is often concentrated at the interfaces of admixed clay-carbon microenvironments. This implies physical and chemical interactions between clays and organic matter conducive to the entrapment of detrital gold during deposition and the crystallization of colloidal/dissolved gold in the carbon seams. High concentrations of organic free radicals (molecules with unpaired electrons) in the carbonaceous matter, deduced from electron paramagnetic resonance spectroscopy, suggest that radiation emitted from uranium played a unique role in the reconstitution and polymerization of the progenitor biochemicals.

#### INTRODUCTION

The Vaal Reef carbon seams are part of the extensive gold-uranium deposits of the Witwatersrand Sequence, which is between 2,500 and 2,750 m.y. (million years) old (Pretorius, 1974). These thin carbon seams, rich in uranium and gold, are commonly found along the basal contact of the corresponding pebbly quartz-arenite bed. The Vaal Reef stratiform deposit is as much as ~1.7 m thick and covers an area of approximately 260 km<sup>2</sup>. It is part of the Main-Bird Series of the Upper Witwatersrand Sequence and is approximately 2,500 m.y. old. The petrology of the Vaal Reef deposit, inclusive of stratigraphical, structural, and mineralogical data, has been studied in detail by Minter (1976). The samples studied were obtained from the Klerksdorp area, which

is approximately 160 km southwest of Johannesburg, South Africa.

Solvent-insoluble organic matter, which is called kerogen, commonly occurs finely disseminated in Precambrian sedimentary rocks such as the Swaziland Sequence (Nagy and Nagy, 1969; Dungworth and Schwartz, 1974), the Transvaal and Bulawayan stromatolites in southern Africa (Zumberge and Nagy, 1975; Nagy and others, 1977), and the McMinn shale, Australia (Hoering and Abelson, 1965). It is generally accepted that kerogen in Precambrian sedimentary rocks evolved from the biochemical constituents of bacteria and simple algae.

The origin of the Witwatersrand carbonaceous matter has long been the subject of discussion. Yet, Garnier (1896) and Spilsbury (1908) had already considered that the "carbon" was derived from organisms (algae). Sharpe (1949) concluded that the progenitors were most likely bacteria and primitive algae along ancient shorelines, intertidal zones, stream channels, and lagoonal deposits. This conclusion was based on studies of the Main Reef, Main Reef Leader, Black Reef, and other reefs of the East Rand Goldfield. MacGregor (1953) considered that the Witwatersrand basin was surrounded by algal "peat bogs" and that reducing conditions induced by decaying organic matter helped to precipitate gold and uranium. Miholic (1954), Brock and Pretorius (1964) and Snyman (1965) also proposed that primitive organisms, such as algae, gave rise to the carbonaceous matter. Studies have been made of the microbiology of modern algal mats (for example, Golubic, 1976) and their organic chemistry, including the newly formed kerogen, or protokerogen (for example, Philp and Calvin, 1976). These modern analogs support the contention that the carbon seams are ancient algal and (or) bacterial mats and their rapidly evolved protokerogen could well have played a significant role in the accumulation of gold and uranium. On the basis of these considerations, it appears that the nature and history of the auriferous-uraniferous Witwatersrand

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carbon seams cannot be fully understood without studying the molecular composition and paleobiology of the "carbon."

#### MATERIALS AND METHODS

The molecular composition of seventeen Vaal Reef "carbon" samples from the Klerksdorp area has been determined. Before molecular analysis, the carbonaceous matter was separated from the host rocks with a solvent-cleaned scalpel. The carbonaceous separates were powdered in acid-cleaned ball mills. To exclude or minimize laboratory contamination, all glassware used during analysis was first cleaned in a mixture of hot, concentrated 85-percent  $H_2SO_4$ -15-percent  $HNO_3$ ; all solvents were freshly distilled. Procedure blanks (the entire analytical procedure without samples) were repeatedly performed to search for laboratory contaminations, but none were detected.

Next, the powdered Vaal Reef kerogen was consecutively extracted by ultrasonication with benzene/methanol (3:2 v/v), methanol, and water prior to analysis by combined high-vacuum pyrolysis gas-chromatography mass spectrometry. This solvent extraction process removes soluble organic compounds, which can be postdepositional contaminations introduced into sedimentary rocks and the insoluble polymeric kerogen (Abelson and Hare, 1968; Nagy, 1970; Smith and others, 1970; Sanyal and others, 1971). After drying, the decontaminated samples were first degassed for 1/2 hour at 100°C at  $\sim 10^{-6}$  torr vacuum in the pyrolysis apparatus and subsequently were pyrolyzed at 450°C for 1/2 hour again under  $10^{-6}$  torr vacuum (Bandurski and Nagy, 1976). Pyrolysis was selected to degrade the solid kerogen to low-molecular-weight fragments because it is a relatively simple and effective technique. Under optimum conditions pyrolysis liberates the components of polymers unaltered or only minimally altered by inter- and intramolecular rearrangements. Thermal energy is used to fragment the polymer, and high vacuum minimizes the recombination and alteration of the breakdown products. Pyrolysis under  $N_2$  or He at atmospheric pressures may yield a variety of products that are absent, as such, in the polymer. Degassing prior to pyrolysis was necessary to remove traces of solvents and (or) contaminations that may have remained in the samples after solvent extraction and drying. The pyrolyzates (breakdown products) were directly introduced and separated on a polyphenyl ether (OS-138) capillary gas-chromatographic column (46 m  $\times$  0.5 mm int diam) in a Perkin-Elmer Model 226 gas chromatograph, which was, in turn, connected to a Hitachi RMU-6E mass spectrometer. Mass spectrometry was used for the identification of the pyrolysis products. It needs to be stressed that although pyrolytic

conditions have been optimized ( $\sim 10^{-6}$  vacuum, small sample size), some of the pyrolyzates might have been somewhat altered from the original kerogen components by thermal energy.

The electron paramagnetic resonance spectra of powdered Vaal Reef samples were obtained with a Varian E-3 spectrometer at room temperature. The microwave and modulation frequencies were set at 9.493 GHz and 100 Hz, respectively; the magnetic field was 3,380 Gs, and the scan range was  $\pm 10$  Gs. The scan range was expanded to observe the presence of any inorganic free radicals. The organic free-radical abundances were determined by comparison with a standard (diphenylpicrylhydrazyl) of known free-radical content.

The scanning electron micrographs and electron-microprobe analyses were obtained using an Applied Research Laboratory Scanning Electron Microprobe Quantometer. This is a single instrument designed to perform both as a scanning electron microscope and an electron microprobe. By increasing the beam current and changing other instrumental parameters, electron-microprobe analyses can be performed on the very same particles detected by scanning electron microscopy. This instrument can detect light elements such as carbon, nitrogen, and oxygen.

#### RESULTS

The pyrolysis results show that the Vaal Reef "carbon," or, according to a more appropriate terminology, kerogen, is a random polymeric solid. It consists of clusters of condensed aromatic hydrocarbons, as well as aromatic sulfur components connected by short-chain aliphatic hydrocarbon and alkyl sulfur bridges. The polymeric structure is relatively tight and stable since it is not broken up in vacuum below  $\sim 450^\circ C$ . Note in this connection that pyrolysis at 300°C yielded predominantly low-molecular-weight alkyl compounds. A typical gas chromatogram showing the separated pyrolysis breakdown products of one of the Vaal Reef kerogen samples is shown in figure 1. The mass spectrometric identifications of peaks with numbers assigned on the gas chromatograph are listed in table 1. The pyrolysis products of the other Vaal Reef kerogen samples were closely similar to the products listed in table 1. Alkyl substituted aromatic components and some aromatic sulfur compounds constitute the majority of the pyrolyzates listed in the table. There is no evidence that the sulfur compounds are pyrolytic artifacts. Pyrolysis of finely powdered pyrite with an aromatic hydrocarbon yielded no organic-sulfur compounds under experimental conditions identical with those used for the kerogen pyrolysis. The few pyrolyzates that emerged from the gas-chromatographic column below 60°C

are not shown in figure 1 and table 1. These compounds were mainly C<sub>2</sub>-C<sub>4</sub> alkanes, alkenes and benzene, and minor quantities of low-molecular-weight alkyl sulfides, acetaldehyde, acetone, and methanol. No nitrogen-containing organic compounds have been detected. It is noteworthy that X-ray diffraction analysis revealed no evidence that these Vaal Reef "carbon" samples contain crystalline carbon—for example, graphite.

TABLE 1. *Mass-spectral identifications of the major gas-chromatographic peaks*

[Peak numbers correspond to those in fig. 1]

Peak	Compound	Peak	Compound
1	toluene.	18	methylidene.
2	2-methylthiophene.	19	Do.
3	3-methylthiophene.	20	naphthalene.
4	ethylbenzene.	21	benzothiophene.
5	p-xylene.	22	methyl-1, 2-dihydronaphthalene.
6	m-xylene.	23	2-methylnaphthalene.
7	o-xylene.	24	1-methylnaphthalene.
8	n-propylbenzene.	25	ethylnaphthalene.
9	methylthylbenzene.	26	Do.
10	Do.	27	dimethylnaphthalene.
11	trimethylbenzene.	28	Do.
12	methylstyrene.	29	Do.
13	methylpropylbenzene.	30	Do.
14	C <sub>1</sub> -benzene.	31	Do.
15	indene.	32	propynaphthalene.
16	methylindane.	33	trimethylnaphthalene.
17	Do.	34	Do.

The results of the electron paramagnetic resonance-spectroscopy studies on the Vaal Reef kerogen show an unusually high concentration of organic free radicals

(3.4–5.5 × 10<sup>20</sup> spins/gram carbon in the kerogen components; the *g* value was 2.0067). This free radical concentration is greater by 1 to 2 orders of magnitude than the usual abundance of organic free radicals in coals (for example, Austen and others, 1966). This excess of free radicals may be attributed to the high abundance of uranium in the Vaal Reef kerogen (as much as 10 percent by weight uranium). Radiation emitted by uranium isotopes is known to affect organic matter, resulting, in part, in free-radical production.

The uranium contents of ten separate Vaal Reef kerogens show an inverse linear correlation with the quantities of dimethylnaphthalenes identified during pyrolysis. With increasing uranium content, the dimethylnaphthalenes decreased in abundance relative to the other naphthalenes. This correlation is shown in figure 2. Since it is more difficult to thermally cleave the aromatic carbon-carbon double bonds than the alkyl bridges during pyrolysis, increasing abundances of dimethylnaphthalenes may indicate somewhat less of an aromatic polymer matrix. Thus, the kerogen matrix is less aromatic at relatively low uranium abundances than at higher concentrations of this element. This is consistent with the effect of prolonged radiation from uranium on organic matter, first producing free radicals and subsequently, aromatization (Huang and others, 1974).

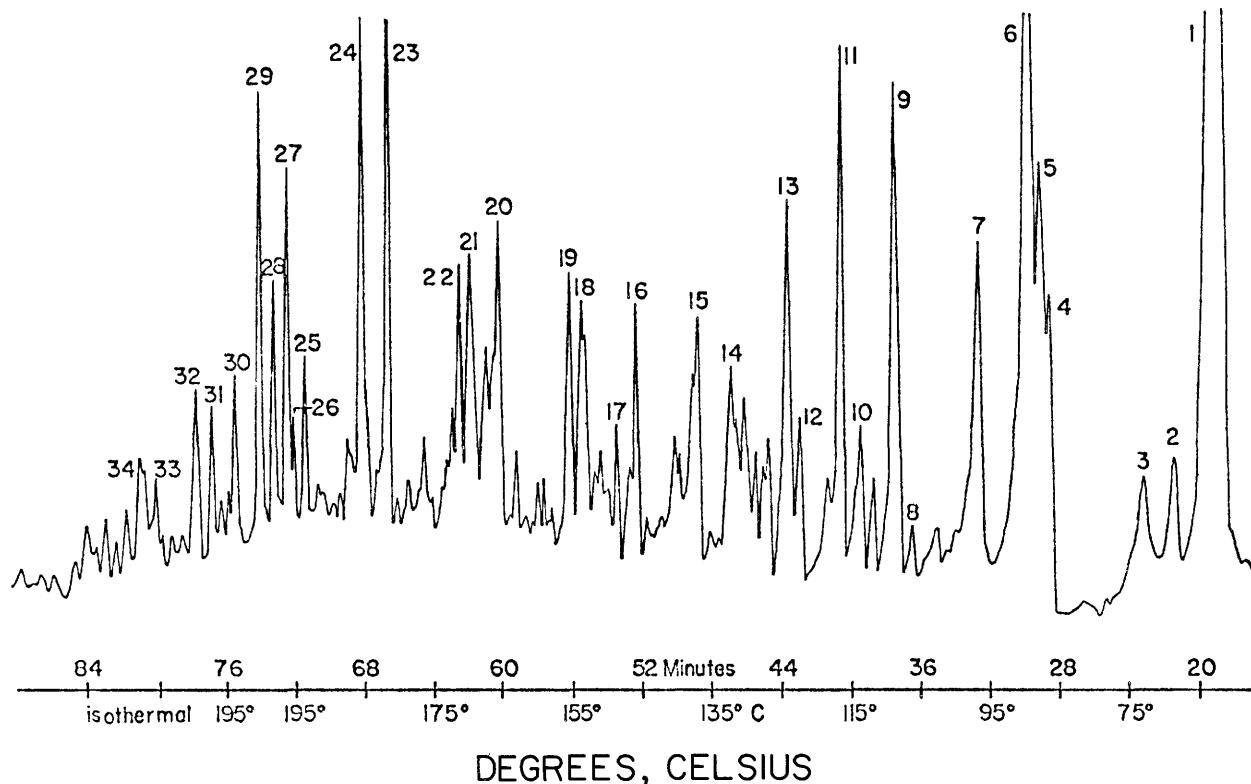


FIGURE 1.—Gas chromatogram of a Vaal Reef "carbon" sample showing separated breakdown products obtained by vacuum pyrolysis at 450°C.

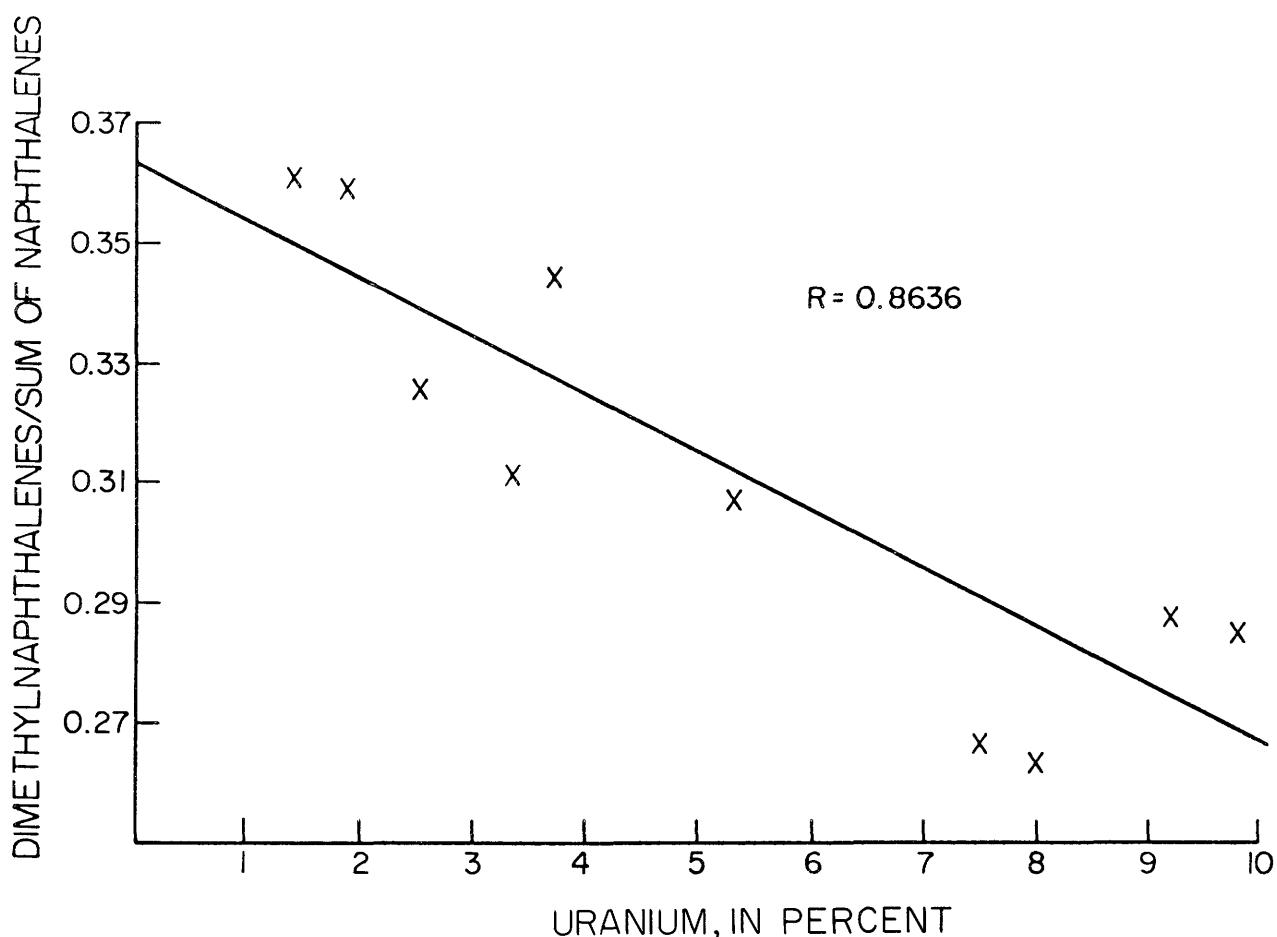


FIGURE 2.—Correlation between the ratio of dimethylnaphthalenes to other naphthalenes and uranium content.  $R$  is the coefficient of correlation.

The results of the electron microprobe analysis show that the Vaal Reef carbon seams contain two finely admixed substances with different compositions. The first component contains mainly carbon and sulfur from the kerogen; hydrogen cannot be detected by the electron microprobe. The second substance consists mainly of silicon, aluminum, iron, and sulfur; the sulfur was associated with iron. This second component is apparently an iron-rich clay together with some pyrite. The presence of clays was confirmed by the flaky mineral morphology observed by the scanning electron microscope and by X-ray diffraction. Gold was commonly found between and near the interfaces of kerogen and clay clusters. It is interesting that one gold particle within a clay cluster exhibited well-developed crystalline morphology. The majority of the gold particles in both kerogen and clay shows detrital morphologies. Well-preserved microstructures of probable biological affinity were also observed under the scanning electron

microscope (Nagy, 1975). These filamentous and coccoid forms were found in powdered preparations and contained carbon.

## DISCUSSION

The most likely source for the extensive Vaal Reef kerogen deposits was ancient, primitive mat-building prokaryotes. It is not known if the Vaal Reef mats were built by filamentous bacteria or blue-green algae, and it is possible that it was a combination of both. The Vaal Reef fossil-like microstructures have so far been found only in the carbonaceous seams and not in the adjacent quartzite host rock. These microstructures are unornamented coccoid and tapered filamentous forms. Modern algal mats consist of a tight, often weblike matrix of filamentous microorganisms (together with coccoid forms). Because there is no compelling evidence that Precambrian mats had a different texture, it is

reasonable to assume that uranium, gold, and clays became concentrated in the original Vaal Reef mats through sedimentary processes, as was suggested by, for example, Minter (1976).

It has been established that many species of mat-building modern (extant) blue-green algae and bacteria can live under anaerobic conditions. However, since blue-green algae give off  $O_2$  as a result of photosynthesis, it is not clear whether blue-green algal cells are ever completely anaerobic (Brock, 1976). Photosynthetic bacteria do not release  $O_2$ . A cross section of a mat from the Persian Gulf showed several distinctive layers 1 mm thick caused by specialized microbial populations. The brown surface layer consisted of organisms with a brown extracellular sheath pigment, which protected the underlying layers from extensive solar radiation. The transitional zone, where both aerobic and anaerobic bacteria have been found, was pink. The bottom area contained black iron-stained layers of mainly non-photosynthetic anaerobic bacteria (Golubic, 1976). This illustrates the complexity of mat matrices.

Stromatolites are formed by mat-building blue-green algae and bacteria, which can trap and bind sedimentary particles. These sedimentary structures can form dome or pancakelike structures from the blue-green algal mats. However, in coastal marshes algal domes may become partially buried, and thus their textures and fabric will be different from stromatolites formed in other environments (Monty and Hardie, 1976). The well-developed Belingwe (Bickle and others, 1975) and Bulawayan (MacGregor, 1941) stromatolites (2.7 billion years old; Wilson and others, 1978) are known from Zimbabwe (Rhodesia). The Transvaal Sequence stromatolites in South Africa (for example, Young, 1933) occur extensively and are also well developed. Their age is ~2.3 billion years. These latter stromatolites have yielded the first known evidence for algal cell diversification (Nagy, 1974, 1978). Therefore, it is somewhat surprising that there are no known stromatolites associated with the extensive Vaal Reef sediments and carbon seams. However, one of us (B. Nagy) has seen an in situ, slightly domelike, semiconvex structure in a carbonaceous seam on the wall of a Vaal Reef mine tunnel at 2,600 m depth. It is interesting to speculate whether this kerogen structure could have become a stromatolite by trapping more mineral particles had the sedimentary and environmental conditions been different before burial.

Current studies of algal mats from Baja California provide some clues to the possible precursors of kerogen. Philp and Calvin (1976) found that a kerogenlike substance forms in algal-bacterial mats relatively soon after burial. The chemical composition of this protokerogen is only partly known; it contains

aliphatic compounds, including alkanes. The major cellular biochemical components of blue-green algae and bacteria are proteins, carbohydrates, and lipids. The degradation products of these biochemicals can give rise to a large number of organic compounds under anaerobic conditions. Some of these organic substances could have played a role in the crystallization and accumulation of gold and (or) uraninite. The initial degradation products of the algal and bacterial biochemicals form a humic-type substance. This matter can stabilize gold colloids in mats and thus preserve them for crystallization during diagenesis. A similar process was illustrated by Ling Ong and Swanson (1969).

These essentially nonaromatic biochemicals must have been also converted to the random aromatic polymer of the Vaal Reef kerogen, and, in the process, the proteins lost their nitrogen content (Zumberge, 1976). One should note that most common coals are also random aromatic polymers; however, their origin can be traced to higher plants, which contain an abundance of lignin (a highly aromatic biopolymer), which did not evolve until the Paleozoic.

Generally, exposure of organic chemicals to  $\alpha$ ,  $\beta$ , and  $\gamma$  irradiation tends to produce unsaturated molecules and subsequent polymerization with the release of volatiles such as  $H_2$ ,  $CH_4$ ,  $CO_2$ , and  $NH_3$  (see Bovey, 1958.) For example, when amino acids and proteins are irradiated in the laboratory, they undergo deamination - that is, loss of  $NH_3$  (see Arena, 1971). Similarly, fatty acids often decarboxylate - that is, lose  $CO_2$  - when irradiated (Desrosier and Rosenstock, 1960). Thus, the reason why nitrogen compounds were not detected among the pyrolysis products of the Vaal Reef kerogen may be because the major nitrogen source, the algal and bacterial proteins, were deaminated by radiation from uranium in the original mats and (or) subsequently in the (proto)kerogen. Older and younger Precambrian kerogens of comparable biological origin released organic nitrogen compounds upon pyrolysis (Nagy, 1976). These rocks, and their kerogen, however, lack the uranium found in the Vaal Reef kerogen. Various organic free-radical reactions were probably responsible for the increasing aromaticity during the development of the Vaal Reef kerogen. The aromatic hydrocarbons are more stable to irradiation than are saturated compounds - for example, alkanes - and this increase in stability was a likely driving force to the final aromatization of the Vaal Reef kerogen. It should be noted, however, that irradiation most likely was not the only cause for the aromatization of the kerogen. Precambrian kerogens, particularly those in southern Africa, are basically aromatic in composition, although to varying extents. Temperature and time may also be major factors causing aromatization.

It appears that uranium has influenced to a considerable extent the diagenetic evolution of the Vaal Reef carbonaceous matter by first aiding in the alteration of microbial biochemicals and then, through free-radical reactions, facilitating the evolution of the random aromatic polymer. The Vaal Reef kerogen is thus both diagenetically and chemically unique, first, because it has been substantially affected by radiation during its history and, second, because it lacks the common oxygen- and nitrogen-containing kerogen components.

This study shows that, for the Witwatersrand carbon seams and other ore deposits associated with carbonaceous matter, organic geochemistry together with micropaleontology may help to elucidate certain problems in economic geology.

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# The Distribution and Sedimentary Arrangement of Carbon in South African Proterozoic Placer Deposits

*By* W. E. L. MINTER

GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN  
QUARTZ-PEBBLE CONGLOMERATES

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## GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN QUARTZ-PEBBLE CONGLOMERATES

# THE DISTRIBUTION AND SEDIMENTARY ARRANGEMENT OF CARBON IN SOUTH AFRICAN PROTEROZOIC PLACER DEPOSITS

By W. E. L. MINTER<sup>1</sup>

### ABSTRACT

Carbon, which occurs as grains, films, and thin seams in Witwatersrand Proterozoic placer deposits, is generally confined to carbon-seam reefs that were deposited in distal environments. The distribution of carbon on paleosurfaces, on sedimentary accumulation surfaces like pebble layers, on trough-shaped bedforms of pi-crossbedded units and foresets, and on the winnowed top of placer sediments implies that its growth took place contemporaneously with placer deposition in an aquatic fluvial environment. The areal distribution of carbon seams in distal environments is patchy, and its sparsity or total absence in some areas does not affect either the gold or the uranium content of the placer. High gold and uranium contents that appear to be associated with carbon seams are at the base of the reef because that position represents both the stable consolidated paleosurface upon which the plant material anchored itself and also the surface of bedload concentration.

### INTRODUCTION

The presence and nature of carbon that occurs in some Witwatersrand placer deposits have caused much speculation and encouraged investigations into its possible origin. It occurs in the form of fine grains, films, and seams, as much as 5 cm thick. Before the results of microscopic and chemical investigations, based on relatively few samples, are examined, it is important to consider the distribution and arrangement of the carbon relative to the sedimentary nature of the placer deposits.

### THE DISTRIBUTION OF CARBON

In basket reefs, where the placer sediment is composed of coarse conglomerate, carbon is rare and generally occurs as fine, sparsely scattered grains. In shallow but proximal braided-channel deposits, like the B Reef in the Orange Free State, fine grains within large pebble lags are more abundant and thin patchy seams occur (fig. 1). Carbon is very much more common in distal sedimentary environments, where the placer deposits were composed of small-pebble pebbly sand-

bodies and have consequently become known as carbon-seam reefs. However, in the distal environments, the carbon seams are neither uniformly distributed nor always present. For instance, an isopach map of the carbon seam in the Vaal Reef, which was constructed from approximately 500 stations in the Klerksdorp Goldfield, indicated that thicker carbon (fig. 2) was more widespread along the western flank of the deposit (Minter, 1972).

### THE ARRANGEMENT OF CARBON

In distal deposits the most common position occupied by carbon is directly upon the paleosurface represented by the surface contact of the placer footwall (fig. 3A). The carbon is generally between 1 and 5 mm thick and exhibits a bladed or columnar structure at right angles

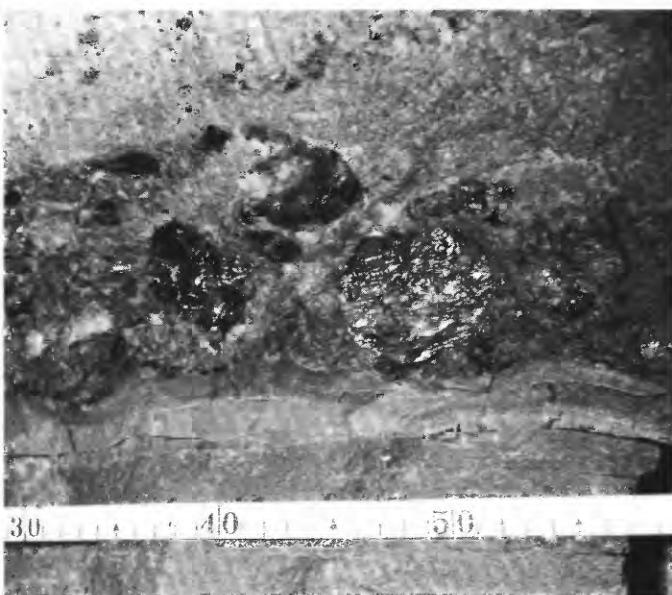


FIGURE 1.—A thin discontinuous carbon seam on rippled paleosurface sediment beneath a thin gravel lag of B Reef in proximal environment. Freddie's Mine in the Orange Free State Goldfield. (Scale is in centimeters.)

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FIGURE 2.—A thick carbon seam with columnar structures that enclose sand grains, pebbles, and detrital minerals in distal environment. Western flank of Vaal Reef, Vaal Reefs Mine, Klerksdorp Goldfield.

to bedding. Basal lag concentrates of small pebbles and detrital heavy minerals occur on top of, and in some instances are depressed or embedded in, the seam. When these basal lags represent the pebbly base of a thicker quartzite placer unit, an additional carbon seam has been seen to occur on top of the pebble layer (fig. 3B). Individual pebbles coated with carbon have also been observed in the quartz-arenite matrix of the Basal Reef (fig. 3C), without an associated carbon seam.

Carbon seams also occur on the top surface of placer units, as in distal parts of the Steyn Reef in the Orange Free State, and on primary structures within the reef unit. These may be bedding planes of laminar layers, channel surfaces, and trough-crossbedded foresets. In figure 3D, a trough-crossbedded unit in the Steyn Reef contains carbon seams 5 mm thick on the bedform and on foresets (fig. 3E) of a trough-crossbedded unit that is 40 cm thick.

## CONCLUSIONS

The distribution and arrangement of carbon in Witwatersrand placer deposits seems to be consistent with that of an aquatic algal-like plant. The sparsity of carbon in proximal environments could have been a consequence of the low preservation potential expected of algal mats under high velocity conditions. Thicker carbon in the Vaal Reef along the western flank of the deposit seems to have been similarly related to relative hydraulic-energy levels.

During the deposition of placer sediment in distal environments the dominance of sediment migration fluctuated with sediment accumulation. During dominant sediment migration, in the form of trains of small-scale lunate sand dunes, small pebbles and detrital minerals accumulated on a practically stable sediment surface upon which an algal plant could also have thrived. These conditions could account for the carbon on the paleosurface and overlying planes (figs. 3A and 3B). During interruptions in flow, sand dunes may become stationary and subaerial, as illustrated in figure 4. In this way, algal material may grow on foresets and bottomsets and be preserved in situ within the internal geometry of the placer unit, as in figures 3D and 3E. Pebbles submerged in these environments would acquire an algal coating, which could be retained during subsequent transport and preserved, as shown in figure 3C. Carbon preserved on layers other than the paleosurface is seldom associated with gold contents as high as found on the paleosurface. This seems to be a factor of the lowest and optimum bedload position for gold.

## REFERENCE

Minter, W. E. L., 1972, The sedimentology of the Vaal Reef in the Klerksdorp area: University of the Witwatersrand, Johannesburg, unpublished Ph.D. dissertation, 170 p.

## DISCUSSION FOLLOWING PAPERS BY DRS. B. NAGY AND MINTER

*Hallbauer:* I have a question for Nagy. How did you determine the clay content in the carbonaceous material?

*Nagy:* You take a fraction from the total sample and go for the major X-ray peaks to get the area under the peaks and then correct for quartz and pyrite and other minerals which are present in large quantities. In other words, the clay content which I presented was a clay content corrected for the carbon, quartz, and pyrite.

*Simpson:* I would like to ask Dr. Minter whether he thinks the outer envelopes that he sees around pebbles

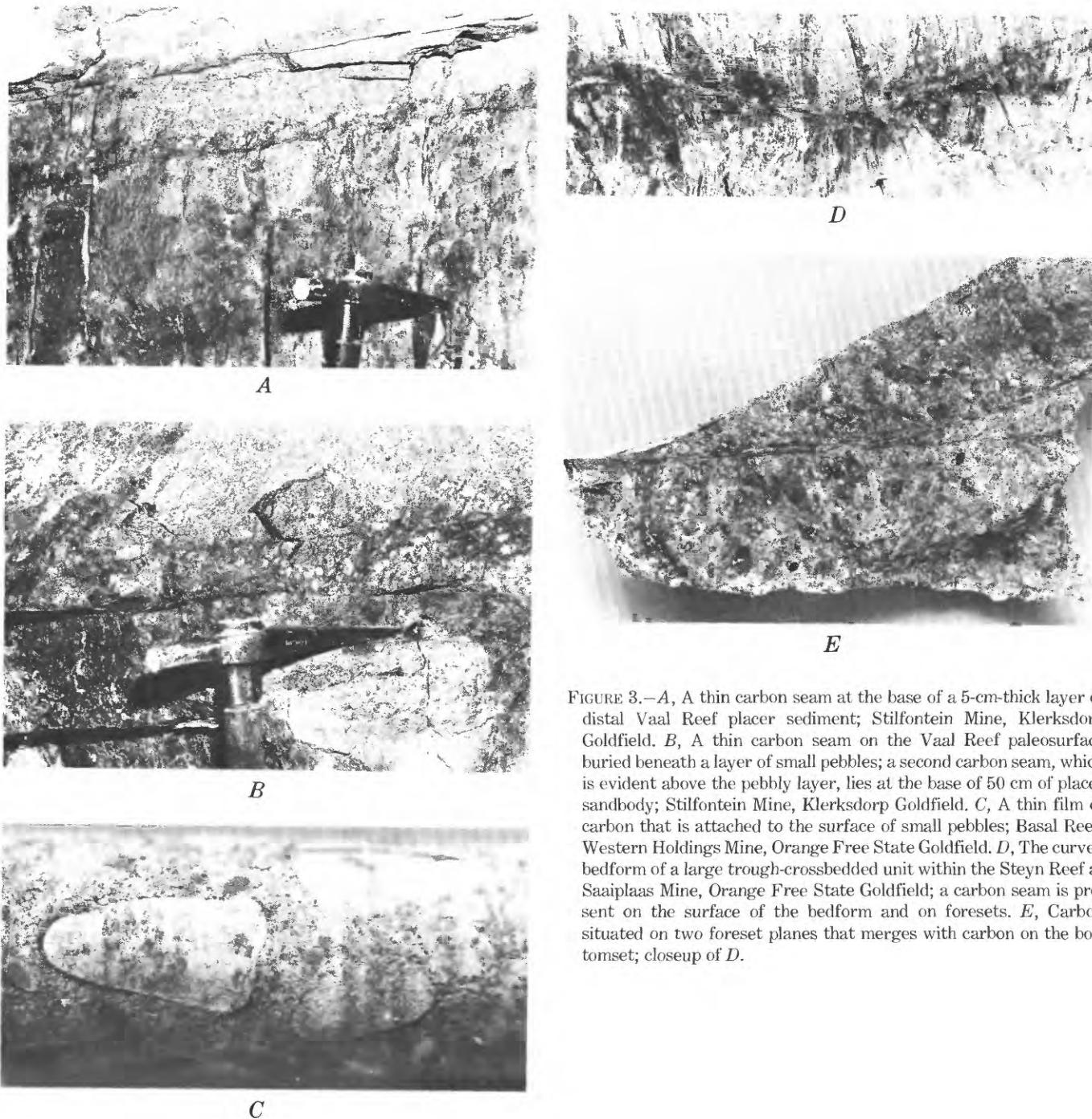


FIGURE 3.—*A*, A thin carbon seam at the base of a 5-cm-thick layer of distal Vaal Reef placer sediment; Stilfontein Mine, Klerksdorp Goldfield. *B*, A thin carbon seam on the Vaal Reef paleosurface buried beneath a layer of small pebbles; a second carbon seam, which is evident above the pebbly layer, lies at the base of 50 cm of placer sandbody; Stilfontein Mine, Klerksdorp Goldfield. *C*, A thin film of carbon that is attached to the surface of small pebbles; Basal Reef, Western Holdings Mine, Orange Free State Goldfield. *D*, The curved bedform of a large trough-crossbedded unit within the Steyn Reef at Saaiplaas Mine, Orange Free State Goldfield; a carbon seam is present on the surface of the bedform and on foresets. *E*, Carbon situated on two foreset planes that merges with carbon on the bottomset; closeup of *D*.

could account for the carbonaceous material that Dr. Schidlowski has described around uraninite?

*Minter:* I certainly think that it could account for that, because the uraninite is also concentrated as a heavy mineral on toe sets and would be incorporated and could be wrapped up in the algal mat. I would also like to draw your attention to a previous hypothesis, I think suggested by Snyman or Liebenberg, that some of these uraninite grains might have actually decomposed to produce the carbon rather than carbon filtering into the cracks; that the carbon represents decomposition products. We hadn't mentioned that before and that has been published.

*Nagy:* Now that the word algae has become respectable again, I would like to make a comment. Algae that we know of from modern blue-green algae, particularly those which form stromatolites, secrete a large amount of polysaccharides. They also decompose. So wherever you have algae you won't have a lot of decaying organic material which is going to be a green mass. I think this is a very important consideration to take into account in evaluating the origin of the Vaal Reef carbon.



FIGURE 4. –Halted sand dunes, which produce trough-crossbedded units. Dark algal growth that is evident in the pool that fills one of the troughs is growing on the base and on the leeside foreset plane; current is from right to left. White Umfolozi River, Swaziland.

# Some Aspects of Witwatersrand Mineralization, With Special Reference to Uranium Minerals

*By* CLIVE E. FEATHER

GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN  
QUARTZ-PEBBLE CONGLOMERATES

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GEOLOGICAL SURVEY PROFESSIONAL PAPER 1161-Q





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## GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN QUARTZ-PEBBLE CONGLOMERATES

### SOME ASPECTS OF WITWATERSRAND MINERALIZATION, WITH SPECIAL REFERENCE TO URANIUM MINERALS

By CLIVE E. FEATHER<sup>1</sup>

#### ABSTRACT

Evidence is presented that suggests that temperatures of metamorphic activity in the vicinity of 400-600°C have been achieved in the Witwatersrand gold-bearing reefs, based on the chemical composition of mixed crystals of gersdorffite and cobaltite that are undoubtedly of authigenic origin, being deposited at the same time as the gold was redistributed and uraninite was probably recrystallized.

Sedimentological studies support the view that uraninite is of detrital origin, but it has been suggested that the mineral resulted also from precipitation from solution, since the "lichenlike plant," which is seen today as carbon ("thucholite") possibly also absorbed uranium from solution during its growth. However, the  $UO_2/ThO_2$  ratio varies over a wide range in the grains from single reef samples, suggesting a multiple granitic or pegmatitic detrital source.

All original features of the uraninite grains have been destroyed by subsequent partial chemical dissolution and alteration to the uranium titanate mineral brannerite (confirmed by electron microprobe and X-ray diffraction studies).

Age determination by ion microprobe on minute volumes within the metamicr crystals gives the same age (about 1,800 million years) for both uraninite and brannerite, suggesting that recrystallization and alteration took place at the same time. This is younger than results obtained on whole grains as determined by other investigators (about 2,000 million years), possibly because whole grains may be contaminated with some radiogenic lead of a greater age, which occurs in pits and cracks in the grains and thus was not completely purged from the grains at the time of recrystallization.

Electron microprobe analyses of a large number of gersdorffite, cobaltite, uraninite, and brannerite grains are presented.

Losses to uranium-extraction-plant residues are essentially due to enclosure of minute uraninite crystals in carbon and, especially in the west of the Witwatersrand Basin, to enclosure of minute brannerite crystals in acid-insoluble hosts, mainly phyllosilicates and leucoxene.

#### INTRODUCTION

In recent years, mainly as a result of the development of sophisticated instrumentation such as the scanning electron microscope and electron microprobe, considerable advances have been made in our understanding of the processes responsible for the mineralization of the conglomerate reefs of the Precambrian Witwatersrand

deposit. Our present knowledge of the ore mineralogy of the reefs has been collated and reviewed by Feather and Koen (1975), who indicated certain areas where further research is necessary.

With this in mind, in the present paper certain aspects of mineralization are considered in detail; *inter alia*, uranium mineralization and the temperature to which the reefs were subjected during the major period of metamorphic activity that they underwent are discussed.

Chemical compositions were determined with a JEOL JXA-5A Electron Probe X-ray microanalyser using pure metals, synthetic compounds, and analysed minerals as standards. Data refinement was achieved by the computer program of Duncumb and Jones (1969). Iron-filtered  $CoK\alpha$  radiation was used for all 57.3 mm Debye-Scherrer camera X-ray diffraction studies.

Samples of all the economically important reefs of the Witwatersrand stratigraphic sequence were examined. Polished sections of reef and heavy concentrates were prepared. In addition, a large number of mine recovery-plant products were examined.

#### TEMPERATURES OF METAMORPHIC ACTIVITY

##### INTRODUCTION—THE MODIFIED PLACER THEORY

It is not intended that the arguments that were put forward for a hydrothermal (for example, Davidson, 1960) or a placer origin for the mineralization of the conglomerates be discussed here. Recent evaluations by Pretorius (1974a, b, 1975), among others, especially with regard to the sedimentology of the Witwatersrand Basin, clearly support a placer origin for the mineralization of the reefs. In brief, it is believed that deposition took place along the interface between a fluvial system that brought sediments and heavy minerals from an elevated source area to the northwest of the basin and a lacustrine littoral system that reworked the material and redistributed the finer sediments along the shore-

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line of an ancient inland sea. The heavy minerals were concentrated by gravity settling and subsequent winnowing by wave and current action. Some investigators, such as Hallbauer (1975) and Riemer (1975), support the view that some of the gold and uranium was carried into the basin in solution and deposited biochemically by an ancient lichenlike life form that preferentially developed in quieter water conditions at the mouth of major rivers at the end of cycles of sedimentation.

There is little doubt that some of these ore minerals, notably the gold, base-metal sulphides and arsenides, and uraninite, underwent subsequent alteration and redistribution in situ in the reefs after burial during at least one major epoch of metamorphic activity. The date of this epoch is set by the age of the uranium-bearing minerals, which would have been purged of their radiogenic lead at this time.

This is the modified placer theory, which enjoys a fair degree of consensus, especially among those who are familiar with the geology of the basin through mining activities.

#### PREVIOUS WORK

Fuller (1958), using the method of Kullerud (1953), in which the iron content of sphalerite is used as a geothermometer to determine the temperature at the time of deposition, obtained an average temperature of 450°C for undoubtably authigenic sphalerite in samples from the Bird Reefs Group taken just south of Krugersdorp on the Witpoortjie fault. Similarly, von Rahden (1964) obtained a temperature of 400°C on a grain of authigenic sphalerite from the Bird Reefs Group at Luipaardsvlei gold mine.

Barton and Kullerud (1958), however, point out that this method gives only a minimum temperature of formation, and therefore the temperatures in the reefs may have been still greater than those indicated. However, the iron content has been shown to be not only dependent upon temperature. This is borne out by the fact that the iron content of secondary sphalerite in the reefs varies from 1 to 7 percent (table 1), even when secondary pyrite and pyrrhotite are abundantly present.

The presence of various other secondary minerals in the reefs has been cited as giving information as regards temperature of formation. Pyrrhotite, which occurs sporadically throughout the reefs, occurs in occasional abundance in the vicinity of major dykes. Most investigators believe it to have been derived from pyrrhotitization of primary pyrite. Exsolved pentlandite lamellae are common, and both monoclinic and hexagonal pyrrhotite occur in the reefs. Both these facts are indicative of fairly high temperatures of pyrrhotitization (Ramdohr, 1955; von Rahden, 1964). Similarly, the

TABLE 1.—Composition, in percent, of sphalerite in the Witwatersrand reefs

[Names in parentheses are the abbreviated names of gold mines, as used in all the tables. The numbers in parentheses refer to sample numbers of the Witwatersrand collection of the X-ray Fluorescence Section, Anglo American Research Laboratories]

Sample	Grain	Zn	Fe	S	Total
Vaal Reef (Vaal Reefs) (25) —	S1	64.3	1.2	33.0	98.5
	S2	64.1	1.3	33.3	98.7
	S3	65.4	1.3	33.1	99.8
	S4	64.8	1.1	32.6	98.5
	S5	65.4	1.2	33.0	99.6
	S6	64.8	1.1	32.9	98.8
	S7	65.0	1.2	33.8	100.0
	S8	66.0	.9	33.0	99.9
	S9	64.6	1.1	32.8	98.5
Average —————	---	64.9	1.2	33.1	99.2
Ventersdorp Contact Reef (Western Deep Levels) (32)	S10	60.7	7.3	33.0	101.0
Basal Reef (Welkom) (184) —	S11	62.3	3.7	34.0	100.0
	S12	62.6	3.4	34.0	100.0
	S13	63.6	3.8	32.6	100.0
	S14	62.9	3.5	33.5	99.9
	S15	62.2	3.9	33.7	99.8
Average —————	---	62.7	3.7	33.6	100.0

presence of chloritoid replacing sericite in the matrix of the reefs has been considered by many (for example, Ramdohr, 1955) to be an indicator of high temperature and pressure conditions. Mackinawite, although very rare, must have been crystallized at a minimum temperature of 210–250°C, as cited by Schidłowski (1968). Graphite, occurring as bent and broken flakes, apparently formed in situ as a result of graphitization of carbon (Saager, 1969; Schidłowski, 1967). X-ray diffractometric analyses of columnar and massive carbon, believed to be the fossil remains of an ancient lichenlike plant, frequently shows signs of incipient graphitization (Feather and Koen, 1975), indicating that fairly widespread elevated temperatures were achieved during metamorphism.

These observations are, however, of a qualitative nature. Consequently, in the present investigation, quantitative information was sought of a more suitable mineral assemblage, notably of the cobaltite and gersdorffite, which are widespread in the reefs.

#### THE EVIDENCE OF COBALTITE AND GERSDORFFITE

The cobaltite-gersdorffite grains are of two distinct generations, allogenic and authigenic. The allogenic grains, all with cobalt>nickel>iron—that is, cobaltite—are well rounded (fig. 1), may contain pyrrhotite inclusions, and have undergone little or no alteration since burial in the reefs.

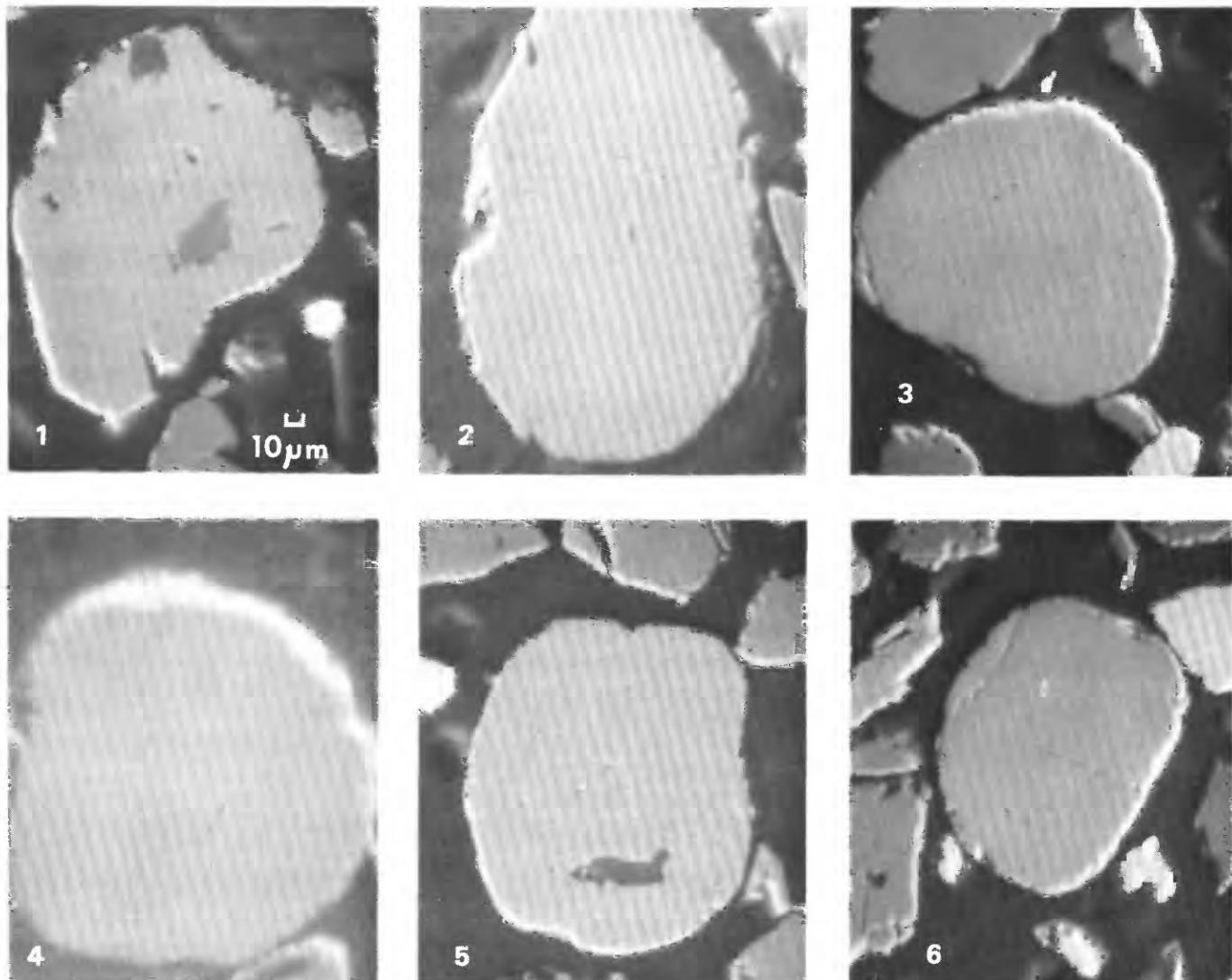


FIGURE 1.—Typically rounded detrital cobaltite grains, two with somewhat rare pyrrhotite inclusions. This type of cobaltite is associated very rarely with gold. Scanning secondary electron photomicrographs,  $\times 250$ . Cristaalkop Reef, Vaal Reefs Mining and Exploration Co., Ltd.

Electron-microprobe analyses of a large number of grains are plotted in the ternary diagram FeAsS-CoAsS-NiAsS (table 2; fig. 2). Superimposed on this diagram are the chemical isotherms of mixed crystal formation of Klemm (1965). Different symbols are used to identify each reef. The temperatures at which the grains formed vary widely over the range of  $<300^{\circ}\text{C}$  to about  $600^{\circ}\text{C}$ , and the grains may have been derived from one or more possible hydrothermal deposits situated in the primary source rocks to the northwest.

Further evidence may be derived from the crystal structure of the cobaltite. X-ray diffraction analyses of a large number of grains of varying composition consistently gave the  $\text{Pca}_2_1$  space group of the orthorhom-

bic system. According to Giese and Kerr (1965) and Gammon (1966), this is the low temperature ordered crystal form of cobaltite, having crystallized at temperatures below about  $800^{\circ}\text{C}$ .

The second (authigenic) (Ni, Co) sulpharsenides (fig. 3) are more variable in chemical composition (table 3; fig. 4). The majority have  $\text{Ni} > \text{Co} > \text{Fe}$  and thus may be classified as gersdorffite. Indicated temperatures of formation of individual grains (table 3; fig. 4) vary within the individual reef samples, and in the Witwatersrand System as a whole, over the range of about  $400$ – $600^{\circ}\text{C}$ . Variation within reefs is explained by slow and zoned crystal growth. Although not always present, the compositions for a particular reef sample may be plotted

TABLE 2.—*Electron-microprobe analyses, in percent, of detrital cobaltite grains in some Witwatersrand reefs*

[The numbers in parentheses refer to sample numbers of the Witwatersrand collection of the X-ray Fluorescence Section, Anglo-American Research Laboratories]

Grain	Ni	Co	Fe	As	S	Total	Atomic proportions (based on cations)		
							NiAsS	CoAsS	FeAsS
<b>Main Reef, South African Lands (150)</b>									
A63	8.2	19.3	3.0	48.3	19.6	98.4	0.268	0.630	0.101
A70	2.8	24.9	2.9	47.5	20.0	98.1	.091	.811	.098
A75	3.4	25.0	3.8	40.3	24.4	96.9	.105	.773	.122
<b>Vaal Reef, Vaal Reefs (North Section) (5)</b>									
A103	5.1	27.2	4.6	44.5	20.3	101.7	0.138	0.734	0.129
A104	3.4	30.7	3.3	44.2	20.1	101.7	.091	.818	.091
A105	8.3	25.4	3.5	43.1	19.8	100.1	.223	.680	.097
<b>Vaal Reef, Vaal Reefs (186)</b>									
C1	2.0	31.9	2.0	45.5	18.9	100.3	0.056	0.887	0.058
C2	9.5	19.8	4.5	45.1	19.2	98.1	.280	.583	.137
C3	12.1	17.0	6.9	44.2	19.8	100.0	.334	.469	.197
C4	8.9	21.3	6.1	44.3	19.3	101.2	.244	.583	.173
C5	8.3	19.2	6.1	45.2	19.7	98.5	.246	.567	.187
C6	15.0	14.6	6.6	44.1	18.9	99.2	.412	.400	.187
C7	10.5	21.5	3.9	44.8	18.6	99.3	.292	.596	.112
C8	7.2	25.9	2.9	45.0	18.8	99.8	.200	.717	.083
C9	7.2	25.3	3.7	44.6	19.1	99.9	.199	.696	.105
C10	9.4	22.7	3.5	44.2	19.3	99.1	.264	.635	.101
<b>Leader Reef, Free State Geduld (200)</b>									
A111	3.8	30.2	2.1	44.0	19.5	99.6	0.105	0.835	0.060
A112	3.5	28.5	3.4	44.5	19.2	99.1	.099	.802	.099
A114	3.5	35.4	3.5	41.6	19.7	103.7	.083	.832	.085
<b>Cristaalkop Reef, Vaal Reefs (171)</b>									
A33	7.2	24.1	6.4	47.0	18.6	103.3	0.190	0.635	0.175
A37	11.5	17.9	4.5	49.3	19.3	102.5	.338	.525	.137
A38	5.2	26.5	2.7	48.1	19.8	102.3	.151	.768	.081
A41	7.6	21.4	5.1	50.2	18.8	103.1	.222	.624	.154
A43	5.5	24.4	5.0	45.2	20.6	100.7	.157	.695	.148
A46	5.7	27.0	2.7	48.0	19.1	102.5	.161	.760	.079
A48	8.1	24.0	2.0	48.9	18.5	101.5	.238	.702	.061
<b>Carbon Leader, West Driefontein (167)</b>									
A12	4.0	23.5	4.0	41.2	21.3	94.0	0.127	0.742	0.131
A13	8.7	21.9	2.4	44.7	20.0	97.7	.263	.662	.075
A14	4.3	26.2	3.3	44.1	20.7	98.6	.127	.772	.101
A16	8.9	20.5	3.6	44.5	20.0	97.5	.269	.618	.113
A17	4.4	28.3	1.2	45.1	20.0	99.0	.129	.826	.045
A18	2.9	28.9	2.4	44.5	20.3	99.0	.085	.843	.073
A20	5.0	30.2	1.9	44.8	19.7	101.6	.135	.812	.053
A21	8.1	26.8	2.8	46.0	19.7	103.4	.215	.709	.077
A22	10.3	23.0	4.5	46.3	19.5	103.6	.272	.606	.123
A23	9.7	17.7	7.0	43.0	20.6	98.0	.281	.511	.209
A24	1.6	32.8	2.6	43.2	20.3	100.5	.043	.884	.073
A28	8.5	19.2	6.7	44.0	18.7	97.1	.246	.554	.200

TABLE 2.—*Electron-microprobe analyses, in percent, of detrital cobaltite grains in some Witwatersrand reefs—Continued*

Grain	Ni	Co	Fe	As	S	Total	Atomic proportions (based on cations)		
							NiAsS	CoAsS	FeAsS
<b>Carbon Leader, Doornfontein (169)</b>									
A82	8.8	17.5	6.9	43.8	21.7	98.7	0.264	0.523	0.214
A85	5.8	26.7	1.6	45.7	19.7	99.5	.170	.781	.049
A86	6.6	25.3	2.7	44.9	19.5	99.0	.191	.729	.081
A87	3.5	27.2	5.4	42.3	21.7	100.1	.097	.749	.154
A90	7.3	24.7	3.7	47.0	19.4	102.0	.204	.689	.107
A93	4.6	29.4	2.3	47.8	19.3	103.4	.167	.808	.065
A94	8.0	19.8	7.1	46.0	18.7	99.6	.228	.563	.209
A95	6.0	27.2	3.1	46.7	18.8	101.8	.165	.747	.088
A96	6.5	22.8	5.4	45.3	19.5	99.5	.187	.653	.160
A97	2.5	30.8	3.1	47.1	19.1	102.6	.069	.844	.088
A99	7.0	22.8	5.7	47.1	18.3	100.9	.154	.717	.129
<b>Ventersdorp Contact Reef, West Driefontein (168)</b>									
A53	2.2	30.4	1.5	45.4	20.0	99.5	0.065	0.890	0.046
A55	.6	33.1	1.9	44.5	21.4	101.5	.017	.928	.055
A56	.8	32.1	1.4	47.2	19.7	101.2	.023	.934	.042
A57	9.8	20.1	4.5	46.4	19.8	100.6	.284	.581	.135
A58	7.1	25.9	1.7	46.8	19.4	100.9	.205	.745	.051
A59	2.2	31.0	2.1	47.2	19.6	102.1	.062	.876	.062
A51	4.3	28.1	1.2	45.8	19.8	99.2	.128	.835	.037

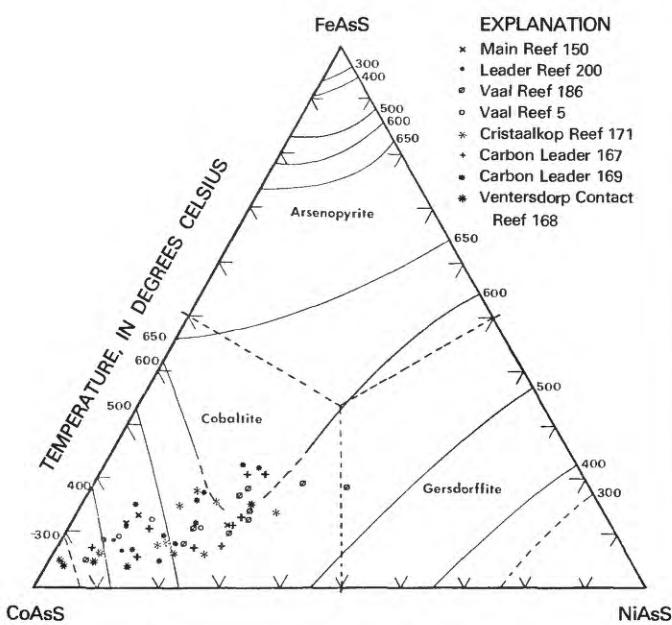


FIGURE 2.—Electron microprobe analyses of Witwatersrand cobaltite grains of detrital origin plotted in the diagram FeAsS-CoAsS-NiAsS. Superimposed on the diagram are the experimentally determined chemical isotherms of mixed crystal formation of Klemm (1965), indicating that temperatures of crystallization in the source rocks ranged from < 300°C to about 600°C. This wide range is indicated by the reefs as a whole and even by individual reefs suggesting multiple sources for detrital grains. Sample numbers in explanation from the Witwatersrand Collection of the X-ray Fluorescence Section, Anglo American Research Laboratories, South Africa.

along a line that extends toward pure NiAsS, shown by dotted lines in figure 4. Zoned crystal growth is commonly visible. For example, grain A 201 from the Basal Reef has a core that is optically silver-white with a greyish tinge and a composition indicating a temperature of crystallization of 450°C. It is immediately enclosed by a zone of higher nickel content, blueish in colour, which indicates a temperature of < 300°C. The outermost zone is even more blueish, has a higher nickel content, and thus was formed at an even lower temperature.

The foregoing deductions are based on the premise that cobalt, nickel, and iron were freely available in the reefs at the time of crystallization. The zoned structure and crystallization trends (dotted lines in fig. 4) may also be explained as due to depletion of cobalt and iron during crystal growth. For this reason, in all cases, the temperatures indicated must be regarded as minimum temperatures of formation.

Additional information was sought in the crystal structure of the authigenic gersdorffite. Both  $P_{a3}$  (pyrite) and  $P_{213}$  (ullmannite) space groups of the cubic system are present. According to Bayliss and Stephenson (1967) and Bayliss (1969), these are the high and intermediate temperature forms of gersdorffite and, as may be deduced from figure 5, resulted from crystallization at temperatures in the range of 450–600°C. This is further confirmed by the fact that mixed crystals of the authigenic sulpharsenide, with  $Co \approx Ni$ , gave the  $P_{ca21}$  low temperature form of cobaltite, indicating that this

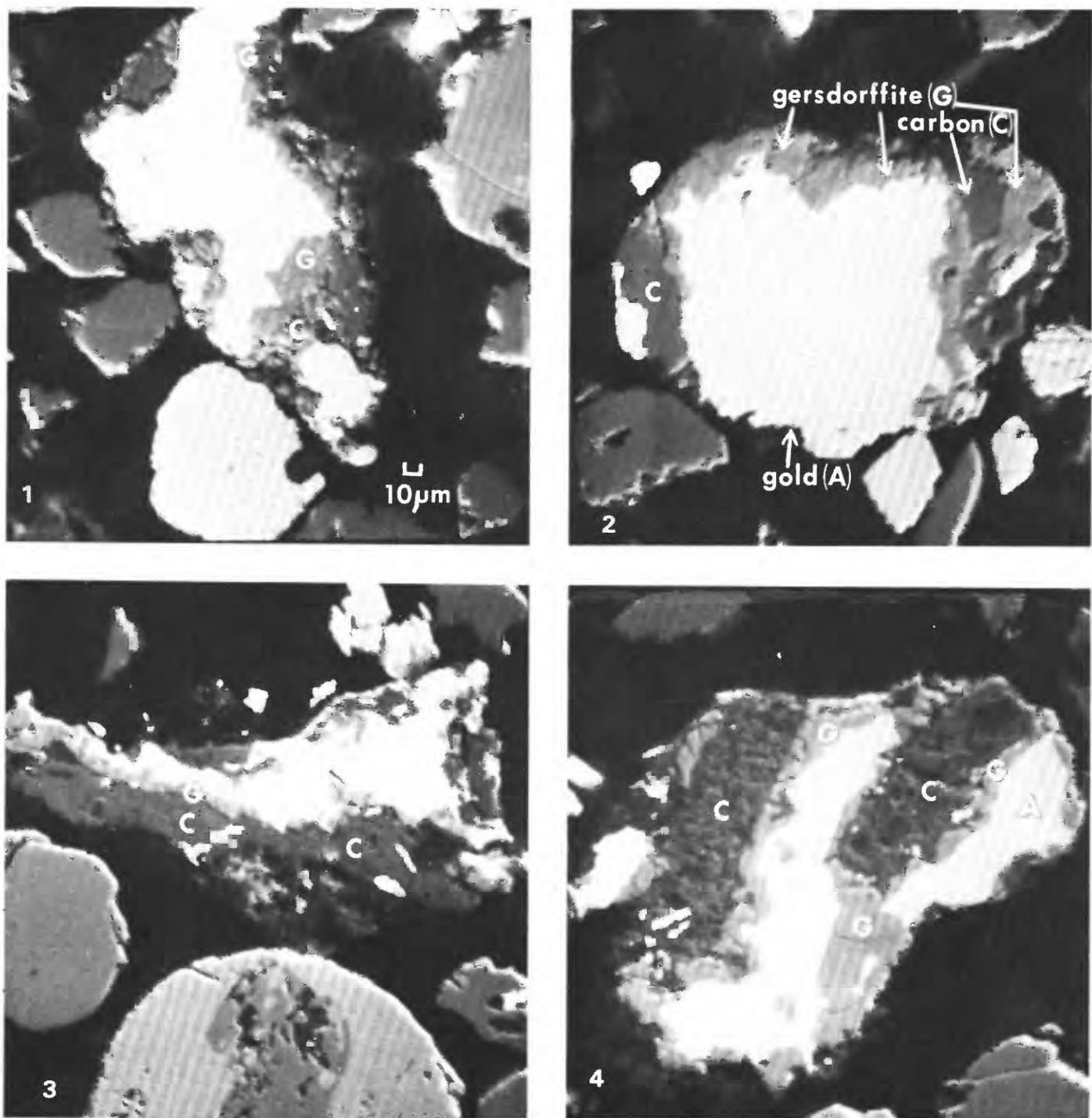


FIGURE 3.—In contrast to detrital cobaltite, gersdorffite (G) occurs as an authigenic mineral, as irregular masses commonly intergrown with gold (A) and frequently associated with carbon (C). It may contain galena, chalcopyrite, and pyrrhotite, all these minerals being of common paragenesis. These grains occur in the same reef sample as the cobaltite grains illustrated in figure 1. Scanning back-scattered electron photomicrographs,  $\times 300$ . Cristaalkop Reef, Vaal Reefs Mining and Exploration Co., Ltd.

TABLE 3.—*Electron microprobe analyses, in percent, of authigenic arsenosulphides of nickel and cobalt, mainly gersdorffite, in some Witwatersrand reefs*

[The numbers in parentheses refer to sample numbers of the Witwatersrand collection of the X-ray Fluorescence Section, Anglo-American Research Laboratories]

Grain	Ni	Co	Fe	As	S	Total	Atomic proportions (based on cations)			Approximate temperature (°C)	
							NiAsS	CoAsS	FeAsS	Grain	Average for sample
<b>Vaal Reef, Vaal Reefs (North Section) (5)</b>											
A101	21.4	9.2	5.4	45.1	19.0	100.1	0.592	0.254	0.154	460	---
A102	25.1	8.4	2.4	43.5	19.2	98.6	.698	.233	.069	380	---
A107	15.5	12.2	7.3	46.8	16.8	98.6	.440	.356	.214	550	460
<b>Vaal Reef, Vaal Reefs (186)</b>											
N2	23.0	3.8	7.9	45.0	20.8	100.0	0.658	0.108	0.234	460	---
N3	25.6	4.7	5.2	44.7	19.4	99.6	.718	.131	.151	400	---
N4	23.0	9.2	4.8	44.4	19.2	100.6	.619	.247	.134	450	---
N5	22.3	7.9	6.4	44.6	19.7	100.9	.606	.214	.180	460	---
N6	19.6	4.3	10.1	45.5	19.8	99.3	.571	.125	.304	510	---
N8	23.7	7.5	3.9	45.5	18.5	99.1	.673	.212	.114	410	---
N10	25.0	6.8	4.6	45.0	19.0	100.4	.684	.186	.130	410	---
N11	20.5	8.2	6.3	45.6	18.8	99.4	.583	.232	.185	470	---
N12	21.7	4.2	8.5	45.8	19.2	99.4	.626	.121	.253	470	450
<b>Basal Reef, near dyke contact, Welkom (192)</b>											
G1	14.9	11.0	7.4	45.9	19.8	99.0	0.445	0.327	0.228	540	---
G3	17.3	10.2	7.2	45.4	19.1	99.2	.495	.291	.213	520	---
G4	12.9	15.8	7.2	44.8	19.3	100.0	.357	.437	.206	580	---
G5	16.8	10.9	7.3	45.6	19.1	99.7	.477	.309	.214	530	---
G6	17.5	9.2	6.9	45.0	19.7	98.3	.518	.271	.211	510	---
G7	14.3	11.3	6.6	44.0	19.9	96.1	.441	.348	.211	540	---
G8	13.4	13.9	6.9	45.7	18.2	98.1	.390	.403	.207	570	---
G9	10.2	13.1	6.7	45.5	20.0	95.5	.338	.433	.229	590	---
G10	10.7	18.3	6.2	45.6	19.3	100.1	.303	.516	.181	600	---
G11	17.3	10.2	7.9	44.9	19.9	100.2	.485	.286	.229	530	---
G12	17.8	9.6	7.8	45.9	19.7	100.8	.502	.270	.227	520	---
G13	17.4	10.3	7.7	46.0	19.5	100.9	.488	.288	.223	530	---
G14	15.0	10.6	6.8	45.8	19.0	97.2	.460	.324	.216	540	---
G16	17.8	10.3	7.8	44.6	20.0	100.5	.493	.284	.223	520	---
G17	18.1	9.6	7.2	44.7	19.7	99.3	.515	.272	.212	510	---
G18	15.9	11.1	7.5	46.0	19.3	99.8	.458	.319	.223	540	---
G19	18.6	9.4	7.3	44.0	19.4	98.7	.524	.264	.212	505	---
G20	18.5	10.1	8.5	42.8	19.8	99.7	.495	.270	.235	520	540
<b>Leader Reef, Free State Geduld (200)</b>											
A113	20.1	6.1	7.1	48.2	17.9	99.4	0.600	0.182	0.219	470	---
A116	22.5	4.2	6.4	47.9	18.3	99.3	.676	.126	.199	425	---
A117	15.4	15.7	5.0	45.3	19.1	100.5	.425	.432	.143	540	---
A118	15.6	13.5	6.0	46.2	19.4	100.7	.442	.382	.176	540	---
A119	20.1	9.0	7.6	49.0	17.4	103.1	.544	.243	.213	500	---
A120	6.2	25.8	5.4	44.5	20.7	102.6	.165	.686	.149	610	510
<b>Zoned grain (A 201)</b>											
Center	23.5	5.4	7.8	42.9	20.3	99.9	0.636	0.146	0.218	450	---
Intermediate	30.1	2.4	2.6	46.2	18.7	100.0	.856	.068	.076	<300	---
Outside	34.6	<.1	1.7	43.7	20.0	100.0	.952	.000	.048	<300	---

TABLE 3. - *Electron microprobe analyses, in percent, of authigenic arsenosulphides of nickel and cobalt, mainly gersdorffite, in some Witwatersrand reefs - Continued*

Grain	Ni	Co	Fe	As	S	Total	Atomic proportions (based on cations)			Approximate temperature (°C)	
							NiAsS	CoAsS	FeAsS	Grain	Average for sample
<b>Cristaalkop Reef, Vaal Reefs (171)</b>											
A31	15.7	8.9	7.7	45.4	19.9	97.6	0.483	0.272	0.245	530	---
A36	13.7	14.9	5.2	51.5	18.3	103.6	.404	.438	.158	550	---
A39	15.3	11.1	5.5	51.1	15.7	98.7	.477	.345	.177	520	---
A40	19.5	4.6	7.9	45.5	19.6	97.1	.605	.142	.253	485	---
A42	18.7	6.9	6.1	53.1	15.5	100.3	.587	.216	.198	480	---
A44	14.0	13.6	4.1	46.1	19.2	97.0	.440	.427	.133	520	---
A45	18.5	6.4	7.4	49.6	17.8	99.7	.569	.196	.235	485	---
A47	21.1	3.9	6.6	50.6	17.2	99.4	.663	.122	.214	440	---
A50	14.8	8.9	8.6	47.6	18.3	98.2	.454	.273	.273	540	510
<b>Carbon Leader, West Driefontein (167)</b>											
A11	16.7	10.8	5.2	45.8	19.0	97.5	0.509	0.328	0.164	505	---
A15	16.7	12.6	3.9	44.9	19.7	97.8	.502	.378	.121	500	---
A19	20.1	5.4	6.6	47.5	17.6	97.2	.622	.167	.211	460	490
<b>Carbon Leader, Doornfontein (169)</b>											
A83	14.1	9.1	8.9	45.9	20.0	98.0	0.436	0.280	0.284	550	---
A84	17.2	14.5	1.0	45.0	20.1	97.8	.526	.442	.032	480	---
A88	15.3	9.6	6.5	47.8	17.5	96.7	.484	.303	.213	520	---
A89	17.8	9.8	5.0	47.2	19.0	98.8	.544	.299	.158	485	---
A91	18.7	4.5	9.3	46.7	19.4	98.6	.570	.137	.293	500	---
A98	18.3	11.5	8.0	48.1	18.8	104.7	.481	.302	.217	520	---
A81	15.5	12.8	4.4	45.1	20.7	98.5	.472	.389	.139	510	510
<b>Ventersdorp Contact Reef, West Driefontein (168)</b>											
A54	20.4	8.2	5.1	46.0	20.0	99.7	0.603	0.242	0.156	460	---
A50	25.5	3.1	5.2	46.8	19.4	100.0	.751	.091	.158	360	410

cobaltite crystallized at temperatures of about 600°C (see fig. 5).

To summarize, this study of the chemistry and crystal structure of secondary gersdorffite-cobaltite has indicated that minimum temperatures in the range of 400–600°C, and probably closer to 600°C, have been achieved in the Witwatersrand reefs during metamorphism of the reefs. Samples that were taken close to dykes—for example, 192-Basal Reef (fig. 4)—indicated only slightly higher temperatures, about 500–600°C, than did reef samples not associated with dykes—the rest in figure 4, about 400–550°C. It appears, therefore, that these temperatures were widespread and not localized to the vicinities of crosscutting dykes and related hydrothermal veins. However, a word of caution should be noted here. The work of Klemm (1965) has not been independently and critically examined. In addition to temperature, a variety of other factors might influence the mixed crystal formation. However, the excellent correlation with the temperatures indicated by

the crystal structures does suggest that Klemm's isotherms are applicable to the metamorphism of the Witwatersrand System.

#### OPTICAL CHARACTERISTICS OF GERSDORFFITE

Witwatersrand gersdorffite is cubic, thus shows no anisotropism or birefringence, and varies in colour from silver-white to white with a blueish tinge as nickel content increases. The NiAsS end member is strongly blueish and readily confused with skutterudite, which has almost identical optical properties. There is additional chance of misidentification resulting from descriptions of gersdorffite in the literature. For example, according to Uytendbogaardt and Burke (1971), gersdorffite is white with a yellowish or pinkish tint.

In the past, a number of references to the presence of skutterudite in the reefs have been made. Among others, Ramdohr (1955) found that the redistributed gold was often associated with skutterudite, which is of

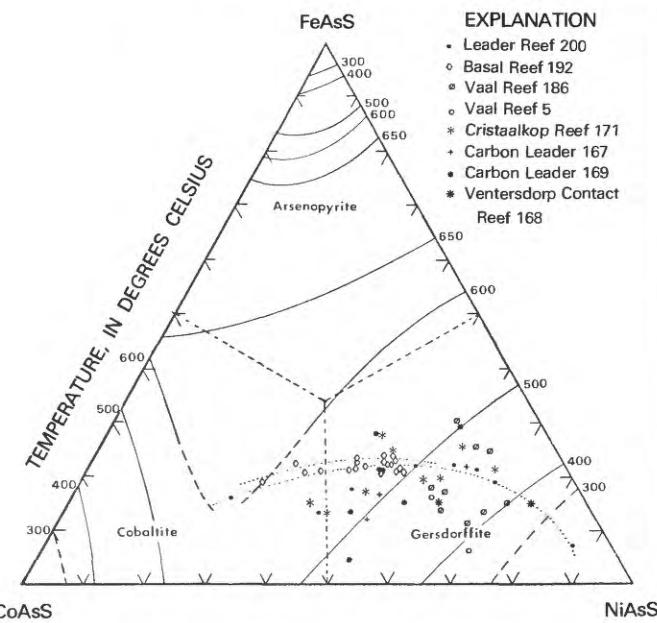


FIGURE 4.—Electron microprobe analyses of authigenic sulpharsenide grains (mainly gersdorffite) plotted in the diagram FeAsS-CoAsS-NiAsS. Superimposed on the diagram are the experimentally determined chemical isotherms ( $^{\circ}\text{C}$ ) of Klemm (1965), which indicate the limits of mixed crystal growth. The analyses indicate that minimum temperatures in the range of 400–600 $^{\circ}\text{C}$  were present in the reefs during the period of metamorphism that remobilised the gold and crystallized the secondary sulphides, sulpharsenides, and arsenides. Sample numbers in the explanation from the Witwatersrand Collection of the X-ray Fluorescence Section, Anglo American Research Laboratories, South Africa.

about the same age, though sometimes distinctly younger. Viljoen (1963) also noted the close association of gold and skutterudite.

In the present investigation, skutterudite could not be found, despite an extensive search by electron microprobe, and it is concluded that previous investigators have confused gersdorffite with skutterudite. The descriptions that both these authors give for skutterudite could apply equally to gersdorffite.

### URANIUM-BEARING MINERALS

The important uranium-bearing minerals in the Witwatersrand and related reefs are uraninite and the alteration product of uraninite. Lesser uranium-bearing minerals include zircon, monazite, columbite, betafite, uranophorite, euxenite, and xenotime.

In this section, the result of an extensive electron-microprobe examination of these minerals is presented, intending to complement the findings of previous investigators, to present new information, and notably to present quantitative microprobe analyses of the important uranium-bearing minerals.

Evidence is presented to confirm that the alteration product of uraninite is brannerite. Particular attention is drawn to this mineral, as it may be the main uranium-bearing mineral in the reefs, especially in the Vaal and Basal Reefs.

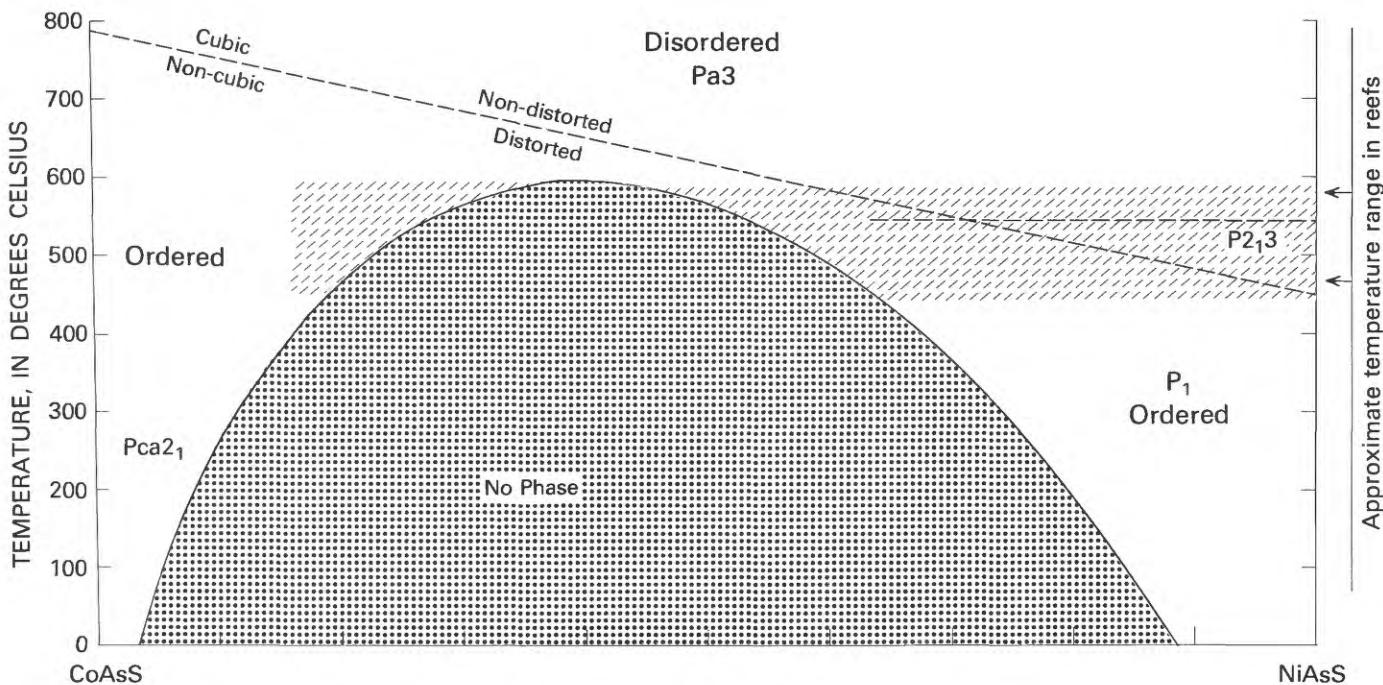


FIGURE 5.—Sketch relating crystal structure ordering with temperature for the system CoAsS-NiAsS (atomic proportions) (after Bayliss, 1969). The structures of authigenic sulpharsenides in the Witwatersrand indicate that temperatures in the range of 450–600 $^{\circ}\text{C}$  were attained during metamorphism of the reefs, in agreement with the temperatures of mixed crystal formation given in figure 4.

## URANINITE

## INTRODUCTION

Cooper (1923) was the first to record the presence of uraninite in the reefs. Ramdohr (1955) and Liebenberg (1955) published the first detailed studies of the mineral, which, in general, investigators have regarded as being essentially of detrital origin. It invariably occurs in greatest concentration close to the footwalls of the reefs, or on the false footwalls (Ramdohr, 1955) within them, in apparent hydraulic equilibrium with other heavy minerals that are definitely of detrital origin.

The grains have a limited size range. Liebenberg (1955) records that the average diameters range from 75–100  $\mu\text{m}$ . They contain abundant galena inclusions and may enclose gold, gersdorffite, pyrrhotite, and chalcopyrite.

The grains are commonly rounded, tending to be oval in shape (Liebenberg, 1955), and are surrounded by phyllosilicates, mainly sericite. Euhedral and subhedral grains, however, are not uncommon. During the present investigation, fractured sections of the reef (simply reef samples that were broken open and the freshly exposed surfaces studied) revealed the presence of uraninite grains that were often bounded by some or all crystal faces, as in figure 6. This might be expected from the fact that the grains were recrystallized and were purged of radiogenic lead during metamorphism. Rounded grains were invariably found to be accompanied by traces of minute crystals of an alteration product.

It is suggested, therefore, that the rounding is due to partial chemical dissolution rather than detrital rounding.

Uraninite also occurs as minute anhedral and subhedral inclusions in carbon. The carbon is believed to be the fossil remains of an ancient lichenlike plant (Hallbauer, 1975). Thus two distinctly different generations of uraninite, which will be referred to as free and included, respectively, occur in the reefs, but each generation is remarkably uniform in texture throughout the reefs.

THE  $\text{UO}_2/\text{ThO}_2$  RATIO

Whereas most investigators of the reefs support a detrital origin for the uraninite not associated with carbon, some, such as Koen (1961) and Riemer (1975), have suggested that the uraninite may have precipitated from solution, aided by biochemical activity of the "lichen" or similar organisms.

Hallbauer and van Warmelo (1974) have pointed out that the "lichen" has certainly absorbed both uranium and gold, and other inorganic materials, during its

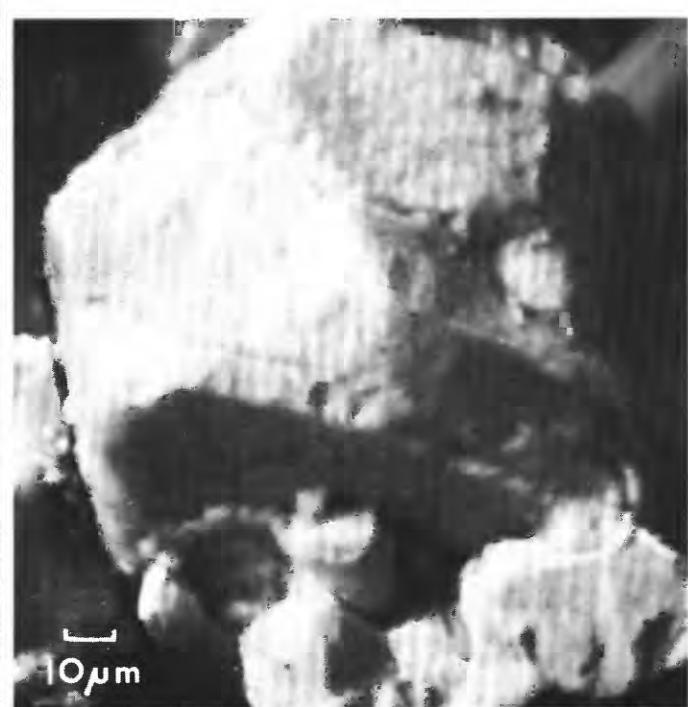


FIGURE 6.—Uraninite grains frequently display one or more crystal faces, and euhedral grains like the one illustrated here, exposed on the face of a fractured section of reef, are not as rare as believed by previous investigators. Scanning secondary electron photomicrograph,  $\times 600$ . Basal Reef, President Brand Gold Mining Co., Ltd.

growth. The question arises, however, as to whether the "lichen" "digested" detrital matter or whether it actually absorbed these elements from solution. It is a common observation that the carbon, the fossil form of the "lichen," replaces the uraninite grains partially or almost wholly.

Recrystallization during metamorphic activity has destroyed all the primary features of the uraninite. As noted above, the present state of rounding of the grains is probably due to partial dissolution rather than attrition, and evidence of crystallization as colloidal pitch-blende—for example, radial textures and cracks—is also absent.

During the progress of this investigation, Grandstaff (1974) published electron-microprobe analyses of free uraninite grains in a sample of reef from Vaal Reefs West mine. In common with the present author's findings at the time, Grandstaff found that although the individual grains were fairly homogeneous, the overall  $\text{UO}_2/\text{ThO}_2$  ratio of grains varied vastly in the assemblage of uraninite grains in the reef sample. He concluded that such heterogeneity favours a detrital, placer origin for the uraninite, which has been derived from a number of

primary sources. He points out that the Witwatersrand grains contain appreciable thorium (up to 10 percent  $\text{ThO}_2$ ), in common with a pyrogenetic (granitic or pegmatitic) source, rather than low-temperature, hydrothermally derived uraninite, which is normally very low in thorium content.

In the present investigation, uraninite from a number of reefs was analysed for uranium, thorium, lead, iron, calcium, and titanium by electron microprobe, using, as standards, sintered  $\text{UO}_2$  and thorium (prepared by the Atomic Energy Board, Pelindaba), lead-metal, and analysed hematite, wollastonite, and rutile. The results are summarised in tables 4 and 5, and in figure 7 the  $\text{UO}_2/\text{ThO}_2$  ratios of the grains are plotted against  $\text{ThO}_2$  content. The data for free uraninite and uraninite included in carbon are plotted separately.

Within all the reefs examined great variation in the  $\text{UO}_2/\text{ThO}_2$  ratio occurs in the free uraninite, favouring, in each reef, a multiple primary source for the uraninite, in keeping with Grandstaff's (1974) findings. In addition, a similar variation occurs in the uraninite enclosed within the carbon. Although it may be possible that the  $\text{UO}_2/\text{ThO}_2$  contents of the water that came into contact with the "lichen" varied so vastly, it seems more likely that the "lichen" "digested" detrital grains, hence the large and similar variation in  $\text{UO}_2/\text{ThO}_2$  ratio.

The overall compositions of the free and included uraninites are very similar—compare tables 4 and 5—but before it could be proved that they are definitely from a common source, variations in all elements within each reef would have to be determined and the data would have to be treated statistically.

TABLE 4.—Results, in percent, of the electron-microprobe analysis of uraninite not associated with carbon in some Witwatersrand ores

[The numbers in parentheses refer to sample numbers of the Witwatersrand collection of the X-ray Fluorescence Section, Anglo-American Research Laboratories]

Source	Grain	$\text{UO}_2$	$\text{ThO}_2$	$\text{PbO}_2$	$\text{FeO}$	$\text{TiO}_2$	$\text{CaO}$	Total	$\text{UO}_2/\text{ThO}_2$ ratio
Cristaalkop Reef (Vaal Reefs South (171).	T21 -----	65.8	5.3	23.3	0.8	<0.01	0.6	95.8	12.4
	T22 -----	66.5	6.1	21.9	.5	.02	.7	95.7	10.9
	T23 -----	70.5	1.7	23.8	.5	.04	.8	97.3	41.5
	T24 -----	62.4	6.1	24.7	.4	<.01	1.1	94.7	10.2
	T25 -----	63.0	8.0	23.0	.5	<.01	1.0	95.5	7.9
	T27 -----	66.6	3.2	23.3	1.0	.04	.9	95.0	20.8
	T28 -----	66.3	1.4	28.0	.6	.02	.9	97.2	47.4
	T29 -----	63.5	3.9	28.5	.6	<.01	.8	97.3	16.3
	T30 -----	65.7	10.2	18.9	.7	.08	1.1	96.7	6.4
Average -----		65.6	5.1	23.9	.6	.02	.9	96.1	12.9
Carbon Leader (Western Deep Levels) (135).	T13 -----	69.6	2.7	26.1	0.2	0.08	1.0	99.7	25.8
	T14 -----	66.4	2.5	27.8	.2	.12	.7	97.7	26.6
	T15 -----	63.8	2.0	30.3	.2	.06	.7	97.1	31.9
	T16 -----	62.6	7.0	24.5	.2	.04	.8	95.1	8.9
	T17 -----	67.1	5.2	21.1	.2	.06	.7	94.4	12.9
	T18 -----	69.2	2.1	28.0	.2	.10	.7	100.3	33.0
	T19 -----	67.9	7.2	18.3	.2	.06	.6	94.3	9.4
Average -----		66.7	4.1	25.2	.2	.07	.7	97.0	16.3
Carbon Leader (West Driefontein) (167).	B43 -----	61.1	5.4	27.9	1.1	0.25	0.4	96.2	11.3
	B44 -----	71.3	2.6	24.2	.6	.25	.6	99.6	27.4
	B46 -----	68.2	4.3	28.2	.4	.10	.5	101.7	15.9
	B47 -----	70.0	2.1	23.6	.4	.10	.6	96.8	33.3
	B48 -----	68.2	3.5	24.7	.5	.12	.3	97.3	19.5
	B49 -----	67.3	6.4	21.6	.4	.16	.5	96.4	10.5
	B52 -----	67.6	9.2	22.8	.9	.14	.3	100.9	7.3
Average -----		68.1	4.2	24.7	.6	.16	.5	98.3	16.2
Main Reef (South Africa Lands (151).	B31 -----	68.7	3.3	14.7	2.3	1.20	0.4	90.6	20.8
	B32 -----	69.1	2.1	21.0	1.1	.50	.4	94.2	32.9
	B36 -----	67.3	1.5	24.4	.9	.19	.3	94.6	44.9
	B37 -----	63.5	2.9	17.3	5.0	2.03	.5	91.2	21.9
Average -----		67.2	2.5	19.4	2.3	.98	.4	92.8	26.9
Basal Reef (Welkom) (184)	U4 -----	68.2	6.3	19.6	2.4	<0.1	0.4	96.9	10.8
	U5 -----	70.5	3.3	16.8	4.1	.2	.4	95.3	21.4
	U7 -----	70.1	5.2	24.8	1.1	.1	.5	101.8	13.5
	U8 -----	66.6	4.5	25.2	.9	<.1	.3	97.5	14.8
	U9 -----	72.5	2.6	26.1	1.6	<.1	.5	103.3	27.9
	U10 -----	64.8	3.3	23.7	5.2	.2	.5	97.7	19.6
Average -----		68.8	4.2	22.7	2.6	.1	.4	98.7	16.4
Overall Average -----		67.2	3.9	23.6	1.0	.16	.6	96.8	17.2

TABLE 5.—*Electron-microprobe analysis, in percent, of uraninite inclusions in selected samples of uraniferous carbon*

[The numbers in parentheses refer to sample numbers of the Witwatersrand collection of the X-ray Fluorescence Section, Anglo-American Research Laboratories]

Source	Grain	UO <sub>2</sub>	ThO <sub>2</sub>	PbO <sub>2</sub>	FeO	TiO <sub>2</sub>	CaO	Total	UO <sub>2</sub> /ThO <sub>2</sub> ratio
Vaal Reef (Vaal Reefs North) (25)	L1	66.0	2.4	28.0	0.1	0.1	0.2	96.8	27.5
	L2	74.0	3.2	18.0	.2	.2	.7	96.3	23.2
	L6	71.5	1.0	20.1	.2	.1	.6	93.5	71.5
	L7	76.6	2.5	13.6	.3	.1	.6	93.7	30.6
	L8	70.0	6.2	18.7	.3	.3	.5	96.0	11.3
	L9	68.1	2.7	24.2	.3	.1	.6	96.0	25.2
	L10	67.0	4.4	22.2	.2	.3	.7	94.8	15.2
	Average	70.5	3.2	20.7	.2	.2	.6	95.4	22.0
Carbon Leader (Western Deep Levels) (34).	T1	69.7	2.6	19.7	0.4	0.3	1.0	93.7	26.8
	T3	66.4	.9	26.8	.3	.1	.5	95.0	73.8
	T4	65.2	7.8	18.8	.2	.4	.8	93.2	8.4
	T5	68.1	1.8	24.6	.3	.2	.7	95.7	37.8
	T6	70.0	1.5	23.4	.3	.3	1.0	96.5	46.7
	T7	65.8	4.4	22.9	.2	.2	.5	94.0	15.0
	T8	71.4	2.4	22.9	.2	.4	.7	98.0	29.8
	T9	69.1	3.7	18.7	.4	.2	.4	92.5	18.7
	T10	69.4	4.7	17.7	.1	.4	.4	92.7	14.8
	Average	68.3	3.3	21.7	.3	.3	.7	94.6	20.7
Carbon Leader (Doornfontein) (169).	B53	71.2	1.3	24.7	0.6	0.2	0.5	98.5	54.8
	B55	71.2	1.3	24.6	.4	.3	.7	98.5	54.8
	B56	70.8	1.5	24.8	.6	.1	.6	98.4	47.2
	B57	73.0	3.2	19.1	.4	.6	.7	97.0	22.8
	B58	75.8	8.6	9.8	.6	.7	.8	96.3	8.8
	B59	69.0	8.3	19.5	.5	.3	.7	98.3	8.0
	B60	71.4	2.0	23.6	.3	.2	.6	98.1	35.7
	B65	69.7	.4	26.1	1.1	.1	.6	98.0	174.3
	Average	71.5	3.3	21.5	.6	.3	.7	97.9	21.7
	Basal Reef (Welkom) (184)	U17	63.8	1.7	23.9	0.4	<0.1	0.3	90.1
	U20	67.0	6.7	23.6	.3	<.1	.3	97.9	10.0
	U23	70.2	2.0	28.1	1.2	<.1	.4	101.9	35.1
	U24	68.6	2.2	29.8	.8	<.1	.7	102.1	31.2
	U25	68.6	2.5	30.4	1.3	<.1	.5	103.3	27.4
	U26	71.0	2.5	27.5	.9	<.1	.6	102.5	28.4
	U30	63.8	2.3	27.2	1.1	<.1	.5	94.9	27.7
	U31	71.6	2.4	27.9	.9	<.1	.3	103.1	29.8
	Average	68.1	2.8	27.3	.9	<.1	.5	99.6	24.3
	Overall Average	69.5	3.2	22.8	.5	.2	.7	96.9	21.7

Although the evidence in favour of a detrital source for the uraninite appears to be most convincing, it is difficult to explain why only the well-mineralized reefs contain the bulk of the uraninite. There are numerous conglomerate and sandstone layers in the succession that may contain other typical detrital grains, for example, chromite and zircon, but that contain neither uraninite nor gold. In addition, in the reefs, uraninite is much more abundant than zircon by several tens of times. This is contrary to what might be expected from granitic or pegmatitic source rocks, which are expected to have contained more zircon than uraninite.

The mineralized reefs do contain one common ingredient—that is, carbon—that is virtually absent in the barren sediments. Despite the foregoing evidence it does seem possible that the ancient “lichen” played an obscure role in aiding the concentration of both gold and

uranium. However, it may be argued that the presence of detrital uraninite was essential to the “lichen’s” origin and development.

#### BRANNERITE

#### INTRODUCTION

As previously noted, the free uraninite grains are invariably partially dissolved and altered. Both Ramdohr (1955) and Liebenberg (1955) noted the presence of an alteration product. Liebenberg distinguished between two products: (1) a common microcrystalline, relatively homogeneous phase somewhat similar in appearance to parent uraninite, which he called “secondary uraninite,” and (2) uraniferous leucoxene. The “secondary uraninite” forms encrustations (fig. 8) on or may partial-

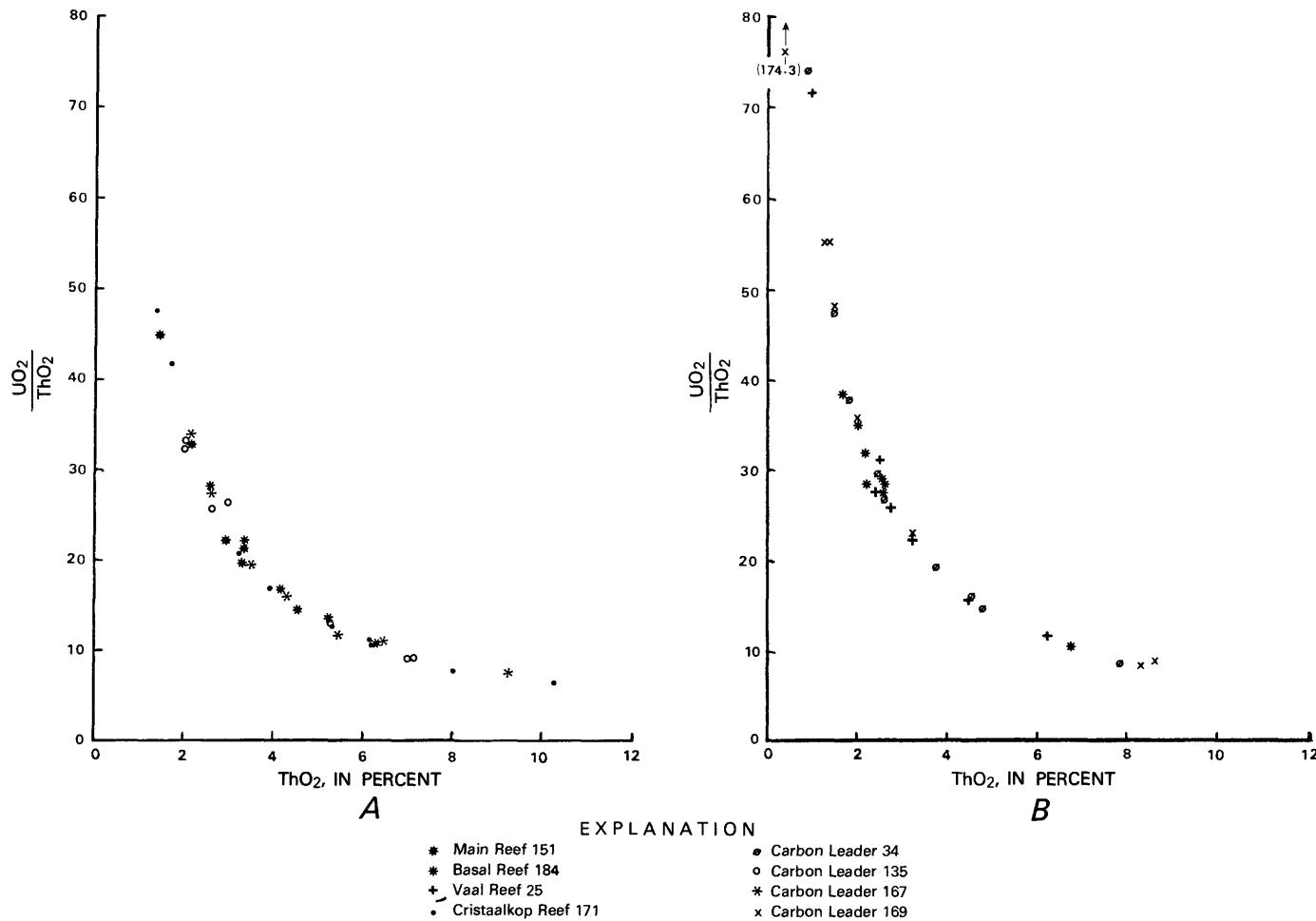


FIGURE 7.—Variation of  $\text{UO}_2/\text{ThO}_2$  ratios. *A*, In free grains of uraninite. *B*, In uraninite associated with carbon. Grandstaff (1974) believed that such variation could result only if the uraninite were from multiple sources and suggested that it is of detrital origin, having been derived from a number of granitic or pegmatitic bodies. The variation in carbon-included uraninite supports the theory that the “lichen,” fossilised as carbon, “digested” detrital uraninite grains rather than absorbed these elements from solution. Sample numbers in the explanation, from the Witwatersrand collection of the X-ray Fluorescence Section, Anglo American Research Laboratories, South Africa.

ly or wholly replace grains. Ramdohr called this material “ghosts,” since the outline of the parent grain is frequently discernible in the crystal orientation of the altered material. Both “secondary uraninite” and uraniferous leucoxene may also be seen infiltrating neighbouring quartz, sericite, clay, and sulphide grains (fig. 9).

When it was established that the “secondary uraninite” contained abundant titanium, it was speculated (for example, Davidson, 1957; Davidson, 1960; Jacob, 1966; Schidlowski, 1966) that it might be the uranium titanate mineral called brannerite, derived from the combined reaction of uraninite and ilmenite or titaniferous magnetite. Pyritization of ilmenites is abundantly evident in the reefs, and titanium would have been freely available for reaction. Although Jacob (1966)

obtained a refractive-index value that was similar to that of brannerite, attempts to identify positively the mineral by X-ray diffraction failed, even when heating experiments were carried out (von Rahden and Hiemstra, 1967; Mihálik, 1968; von Rahden, 1970).

#### ELECTRON MICROPROBE AND X-RAY DIFFRACTION INVESTIGATION

Using back-scattered electron images, the mineral appears to be homogeneous, despite abundant galena inclusions. A number of grains were analysed for uranium, thorium, lead, iron, titanium, calcium, and silicon, and the results are cited in table 6. As in the uraninite analyses (tables 4, 5), the elements are expressed as oxides. The low totals probably result from metamictiza-

TABLE 6.—*Results of electron-microprobe analysis, in percent, of brannerite in some Witwatersrand reefs and recovery-plant samples*

[Ideal  $\text{UO}_2 \cdot 2\text{TiO}_2$  gives  $\text{UO}_2/\text{TiO}_2 = 1.69$  and  $\text{U/Ti} = 2.48$ . X-ray diffraction analysis of grain B6 is given in table 7. n.d., not determined. The numbers in parentheses refer to sample numbers of the Witwatersrand collection of the X-ray Fluorescence Section, Anglo-American Research Laboratories]

Source	Grain	$\text{UO}_2$	$\text{ThO}_2$	$\text{PbO}_2$	$\text{FeO}$	$\text{TiO}_2$	$\text{CaO}$	$\text{SiO}_2$	Total	$\text{UO}_2/\text{TiO}_2$ ratio	$\text{U/Ti}$ ratio	$\text{UO}_2/\text{ThO}_2$ ratio
<b>A. Recovery-plant samples</b>												
Head Sample 1 (Vaal Reefs).	B1	40.1	3.1	0.3	0.9	36.5	0.6	8.0	89.5	1.10	1.6	12.9
	B3	37.0	4.5	1.0	1.3	42.6	.5	12.1	99.0	.87	1.3	8.2
	B4	42.2	4.6	8.1	.8	33.9	.6	8.0	98.2	1.24	1.8	9.2
	B5	26.6	2.3	.3	1.6	45.1	.3	13.7	89.9	.59	.9	11.6
Average		36.5	3.6	2.4	1.2	39.5	0.5	10.5	94.2	0.92	1.4	10.1
Head Sample 2 (Vaal Reefs).	B6	33.6	5.1	14.1	1.0	34.8	0.4	6.4	95.4	0.97	1.5	6.6
<b>B. Rock samples</b>												
Leader Reef (Free State Geduld) (134).	B21	33.6	1.3	9.0	1.1	45.2	0.2	4.7	95.1	0.74	1.1	25.8
	B22	33.1	2.5	4.7	1.6	46.2	.4	6.9	95.4	.73	1.1	13.2
	B23	38.4	2.5	22.1	<.1	32.6	<.1	1.0	96.6	1.18	1.8	15.4
	B24	35.0	3.1	4.8	.7	25.0	.6	9.6	78.8	1.40	2.1	11.3
	B25	31.3	4.6	2.3	1.0	31.7	.2	17.3	88.4	.99	1.5	6.8
	B26	43.6	3.7	6.5	3.5	11.5	.6	11.3	80.7	3.79	5.8	11.8
Average		35.8	3.0	8.2	1.3	32.0	0.3	8.5	89.1	1.12	1.7	11.9
Vaal Reef 1 (Vaal Reefs) (72).	B7	32.5	3.4	10.7	1.1	37.3	1.0	5.7	91.7	0.87	1.3	9.6
	B8	27.2	2.0	11.4	7.1	24.8	.5	11.7	84.7	1.10	1.7	13.6
	B10	31.9	.9	29.2	2.3	18.7	1.1	7.8	91.9	1.71	2.6	35.4
	B14	26.4	2.8	11.8	2.2	45.5	.2	8.7	97.6	.58	.9	9.4
	B16	25.2	3.1	50.4	6.2	15.4	.3	4.7	102.3	1.64	2.5	8.1
Average		28.6	2.4	22.7	3.8	28.3	0.6	7.7	94.1	1.01	1.5	11.9
Vaal Reef 2 (Vaal Reefs) (25).	U1	26.2	0.9	17.0	14.1	30.5	0.2	n.d.	88.9	0.86	1.3	29.1
	U3	39.9	3.2	37.8	11.3	21.7	.3	n.d.	114.2	1.84	2.7	12.5
	U5	38.3	1.8	32.9	14.3	27.0	.5	n.d.	114.8	1.42	2.1	21.3
	U9	34.4	5.4	10.9	9.1	24.7	.2	n.d.	84.7	1.39	2.0	6.4
	U10	31.2	3.0	8.8	14.8	25.7	.2	n.d.	83.7	1.21	1.8	10.4
Average		34.0	2.9	21.5	12.7	25.9	0.3	n.d.	97.3	1.31	2.0	11.7
Carbon Leader (Western Deep Levels) (74).	U15	43.0	3.8	9.2	6.0	33.3	0.7	n.d.	96.0	1.29	1.9	11.3
	U18	45.9	1.7	7.7	5.7	34.2	1.0	n.d.	96.2	1.34	2.0	27.0
	U19	47.1	2.8	12.0	4.7	29.5	.4	n.d.	96.5	1.60	2.4	16.8
	U20	43.9	.9	6.7	5.1	30.6	.5	n.d.	87.7	1.43	2.1	48.8
Average		45.0	2.3	8.9	5.4	31.9	.7	n.d.	94.2	1.41	2.2	19.6
Main Reef (South Africa Lands) (151).	B27	33.0	0.2	2.5	0.6	52.5	0.8	3.7	93.3	0.63	1.0	165.0
	B28	38.1	.9	.9	.7	37.1	.9	5.6	84.2	1.03	1.6	42.3
	B29	49.5	2.8	17.6	1.2	15.7	.3	5.4	92.5	1.33	1.8	17.7
	B33	40.1	.3	<.1	1.0	34.6	1.1	3.5	80.6	1.16	1.8	133.7
	B35	31.3	3.1	<.1	1.0	34.0	.4	12.3	82.1	.92	1.4	10.1
Average		38.4	1.5	4.2	0.9	34.8	0.7	6.1	86.6	1.10	1.7	25.6
Overall		36.0	2.7	11.7	4.1	31.9	0.5	8.0	94.9	1.13	1.7	13.3

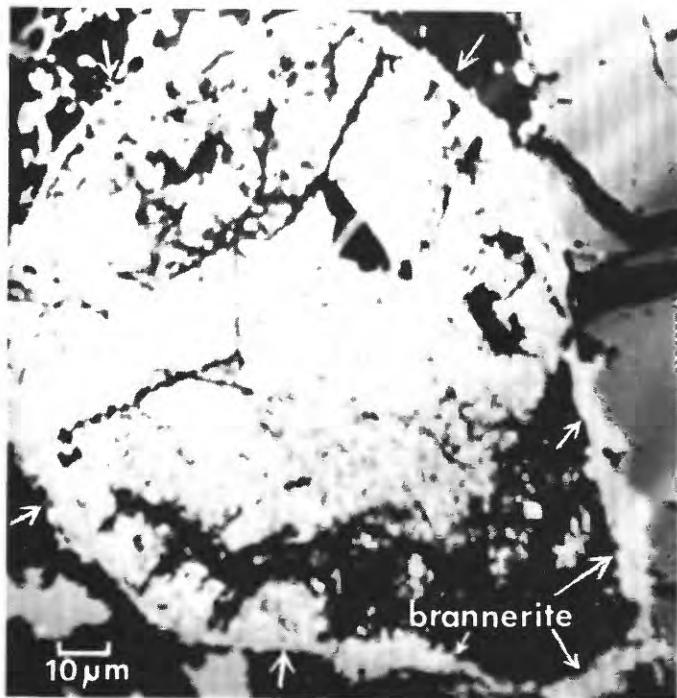


FIGURE 8.—Encrustation of brannerite on a partially altered uraninite grain being replaced by quartz (black). Scanning back-scattered electron photomicrograph,  $\times 600$ . Vaal Reef, Vaal Reefs Mining and Exploration Co., Ltd.

tion and voids in the crystal lattice structure and possibly also from the fact that iron may be present as  $\text{Fe}_2\text{O}_3$  instead of  $\text{FeO}$  and lead may be present as a sulphide rather than an oxide, because galena inclusions are abundant, although visible inclusions were avoided during analyses.

In table 6, the  $\text{SiO}_2$  content of the grains varies from 1 to as much as 18 percent. It is probable that it is derived from the abundant authigenic silica, which has invaded all fractures and pores in the reefs, having entered the structure of the uranium titanate either at the time of formation or later when the grains became metamict and somewhat porous (in the same way, for example, as sulphur entered the grains to form galena of radiogenic lead).

As in uraninite, the  $\text{UO}_2/\text{ThO}_2$  ratio varies extensively in the alteration product (fig. 10), helping to confirm its derivation from the uraninite.

Selected grains were heated to  $1,000^\circ\text{C}$  for one hour and examined by X-ray diffraction analysis. The results are given in table 7. Although the diffraction data may partially fit a mixture of  $\text{Pb}_3\text{O}_4$ , quartz, and rutile or the mineral may be contaminated with these three, there are a number of important lines of these proposed impurities that are absent (italic in table 7). Ignoring the lines of an unknown impurity, the best fit is with the data for brannerite given by Patchett and Nuffield (1960).<sup>2</sup>

It may be argued that a metamict microaggregate of  $\text{TiO}_2$  and  $\text{UO}_2$  would, on heating, synthesize brannerite and that the original mineral may not have been brannerite at all. However, this is a problem inherent in all studies of metamict minerals.

As Ferris and Ruud (1971) have pointed out, because of the uncertainty of heating experiments and X-ray diffraction analyses, structure analyses cannot be considered essential for positive identification of metamict minerals such as brannerite.

What we do know is that we have in the Witwatersrand a homogeneous mineral similar in appearance—medium grey, medium reflectance—to descriptions of brannerite given in the literature (Pabst, 1954; Patchett and Nuffield, 1960; Ferris and Ruud, 1971). It is similar in composition to brannerite from other sources (table 8; Ferris and Ruud, 1971). For the overall average in table 6, if the uranium content is increased to what it would have been before radioactive decay, if the reasonable assumption is made that thorium, calcium, and iron substitute for uranium in the structure, and if  $\text{SiO}_2$  is ignored, the average normalised composition would then be approximately 65 percent  $\text{UO}_2$  and 35 percent  $\text{TiO}_2$ , close to the ideal formula  $\text{UO}_2 \cdot 2\text{TiO}_2$  (62.8 percent  $\text{UO}_2$  and 37.2 percent  $\text{TiO}_2$ ).

It is the opinion of the present author that the uranium titanate mineral in the Witwatersrand is indeed brannerite.

The uraniferous leucoxene described first by Liebenberg (1955) is found to be an admixture of leucoxene and varying amounts of tiny needles of brannerite (fig. 9, grain 5).

#### ECONOMIC IMPORTANCE OF BRANNERITE

Brannerite is most abundant in the reefs mined in the West and Far West Witwatersrand, where it may be the most important uranium-bearing mineral, and is of lesser abundance in the Orange Free State. In the Middle, East, and Far East Rand it is relatively rare.

According to Liebenberg (1955), referring to brannerite as "secondary uraninite," its rate of dissolution in the oxidising dilute sulphuric-acid leach solutions of the uranium-extraction plants is slightly greater than that of uraninite. However, on examination of many leach residues during the present investigation, in addition to uraninite enclosed in carbon, brannerite was frequently the most abundant uranium-bearing mineral in the plant tailings. This is due to enclosure of the brannerite by minerals that are themselves insoluble. Phyllosilicates, pyrite, quartz, and leucoxene are common refractory

<sup>2</sup> Since this paper was submitted for publication, numerous Debye-Sherrer diffraction patterns confirming brannerite have been obtained from samples collected in several parts of the Witwatersrand.

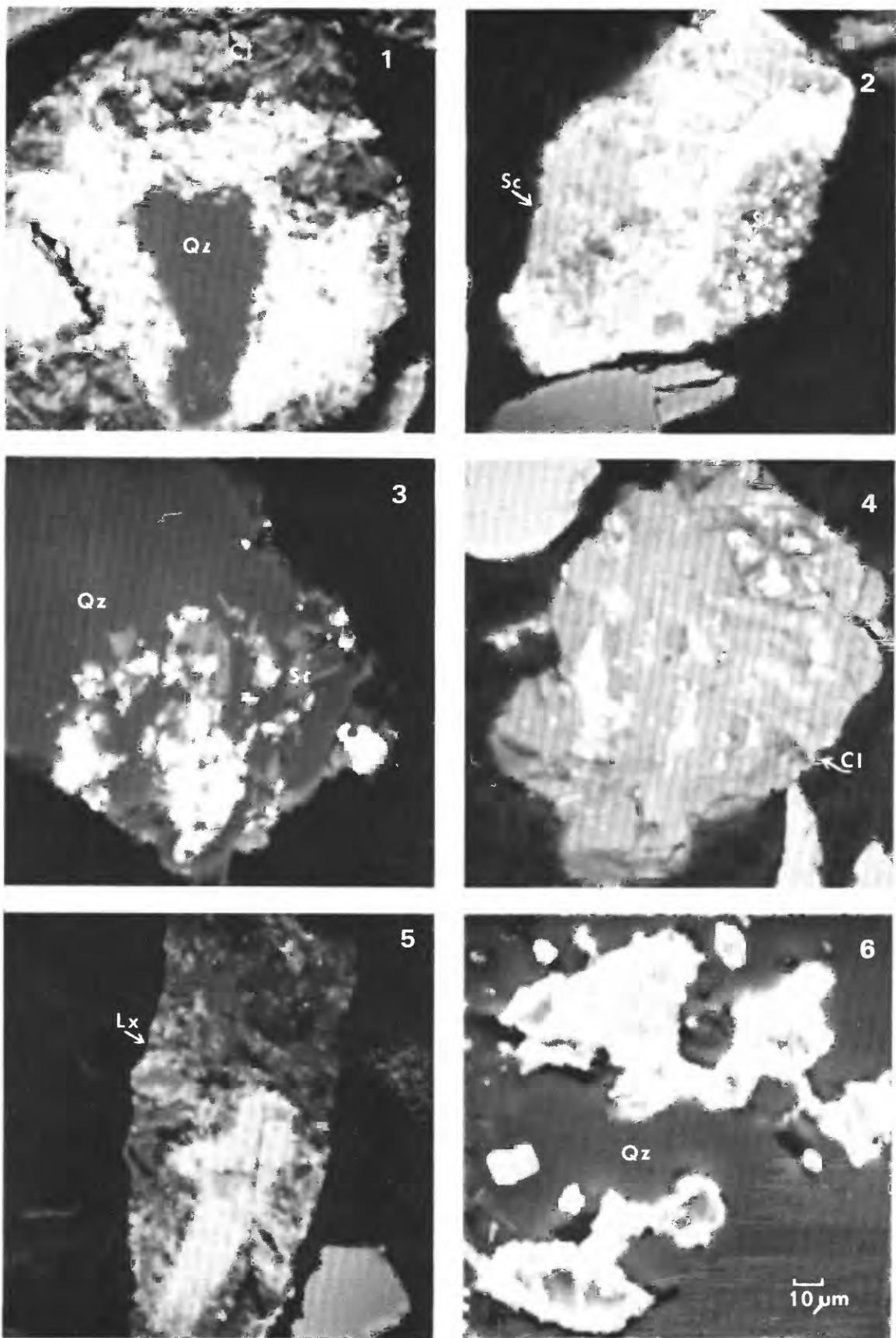


TABLE 7.—*Best obtained X-ray powder-diffraction data for Witwatersrand brannerite, compared with previously published information on brannerite from other sources*

[ASTM diffraction data for  $\text{Pb}_3\text{O}_4$ , quartz, and rutile are listed for comparison, since the Witwatersrand data may be interpreted as a mixture of these three, although some important lines are absent (italics) or may be contaminated with one or more of these materials (there is no fit with the powder diffraction data for  $\text{PbO}_2$  (ASTM 11-549), high temperature  $\text{PbO}_2$  (ASTM 22-389), tridymite (ASTM 18-1170), and cristobalite ( $\alpha$ - or  $\beta$ -; ASTM 11-695 and 4-359)). In addition, the grain contains an unidentified substance, diffraction lines of which are given in parentheses. d, lattice spacing, in angstroms; I, relative intensity; b, broad; f, faint; vf, very faint]

<i>hkl</i>	Witwatersrand		Brannerite Blind River <sup>1</sup>		Synthetic <sup>1</sup>		$\text{Pb}_3\text{O}_4$ ASTM 8-19		Quartz ASTM 5-490		Rutile ASTM 21-1276	
	d	I	d	I	d	I	d	I	d	I	d	I
---	(8.78)	6	---	---	---	---	---	---	---	---	---	---
---	(6.45)	1	---	---	---	---	---	---	---	---	---	---
001	Black	---	6.03	3	6.04	36	6.23	11	---	---	---	---
201	4.84	10	4.77	8	4.74	96	---	---	---	---	---	---
200	4.36	1	4.31	2	4.29	19	---	---	4.26	35	---	---
---	(3.78)	---	---	---	---	---	---	---	---	---	---	---
---	---	---	---	---	---	---	3.66	3	---	---	---	---
110	3.47	4	3.43	10	3.44	>100	---	---	---	---	---	---
202	3.35	5	3.33	4	3.35	>100	3.38	100	3.343	100	---	---
111	3.26	3	3.27	4	3.28	13	3.28	7	---	---	3.25	100
---	3.16	1	---	---	---	---	3.11	19	---	---	---	---
002	3.02	3	3.02	3	3.02	36	---	---	---	---	---	---
201	2.87	4	2.92	5	2.90	36	2.903	48	---	---	---	---
111	2.78	2	2.76	5	2.77	33	2.787	45	---	---	---	---
---	(2.69)	2	---	---	---	---	---	---	---	---	---	---
---	2.60	.5	---	---	---	---	2.632	30	---	---	---	---
112	2.51	4	2.51	4	2.53	31	---	---	---	---	2.487	50
311	2.45	4	2.46	4	2.47	33	2.444	2	2.458	12	---	---
401(?)	---	---	---	---	2.41	17	---	---	---	---	---	---
203	2.33	1	---	---	2.303	8	---	---	---	---	---	---
312	2.24	2(b)	---	---	2.292	27	2.289	4	2.282	12	2.297	8
310	---	---	2.281	6	2.276	12	2.205	1	2.237	6	---	---
400	2.12	1	2.156	vf	2.144	9	---	---	2.128	9	---	---
---	2.05	3	2.078	vf	2.080	7	2.076	1	2.054	10	---	---
---	2.02	4	2.023	3	2.043	36	2.032	12	---	---	---	---
---	---	---	---	---	2.015	23	1.970	12	1.980	6	---	---
---	1.916	2	---	---	1.911	5	---	---	---	---	---	---
---	1.890	2	1.903	4	1.903	19	1.903	22	---	---	---	---
---	1.861	2	1.866	2	1.881	24	1.887	<1	---	---	---	---
---	---	---	---	---	---	---	1.829	<1	---	---	---	---
---	---	---	---	---	---	---	---	---	1.817	17	---	---
---	---	---	---	---	---	---	---	---	1.801	<1	---	---
---	1.775	1(b)	1.778	vf	1.798	4	---	---	---	---	---	---
---	---	---	---	---	1.749	8	1.755	30	---	---	---	---
---	---	---	1.737	f	1.732	5	---	---	---	---	---	---
---	---	---	---	---	1.723	3	---	---	---	---	---	---
---	1.699	8	1.695	3	1.709	20	1.7025	2	---	---	---	---
---	1.678	1	1.667	vf	1.675	5	1.6897	2	1.672	7	---	---
---	---	---	---	---	1.642	7	1.6417	8	1.659	3	---	---
---	1.633	1	1.621	4	1.630	19	1.6302	<1	---	---	1.6237	20
---	---	---	---	---	1.609	18	---	---	1.608	<1	---	---
---	---	---	---	---	1.597	6	1.5876	12	---	---	---	---
---	(1.582)	1	---	---	---	---	---	---	---	---	---	---
---	---	---	1.568	4	1.569	7	---	---	---	---	---	---

FIGURE 9.—Especially in the Vaal Reef in the Far West Witwatersrand, brannerite is an important, often the most important, uranium-bearing mineral present in uranium-extraction-plant residues. Its presence is by virtue of its common enclosure, as minute crystals, in quartz (Qz), sericite (Sc), chlorite (Cl), and leucoxene (Lx), all of which are insoluble in oxidizing dilute sulphuric acid leach solution, thereby indirectly rendering refractory a substantial proportion of the normally soluble brannerite present. Scanning back-scattered electron photomicrographs,  $\times 480$ . Vaal Reef, Vaal Reefs Mining and Exploration Co., Ltd.

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TABLE 7. - *Best obtained X-ray powder-diffraction data for Witwatersrand brannerite, compared with previously published information on brannerite from other sources -Continued*

[ASTM diffraction data for  $\text{Pb}_3\text{O}_4$ , quartz, and rutile are listed for comparison, since the Witwatersrand data may be interpreted as a mixture of these three, although some important lines are absent (italics) or may be contaminated with one or more of these materials (there is no fit with the powder diffraction data for  $\text{PbO}_2$  (ASTM 11-549), high temperature  $\text{PbO}_2$  (ASTM 22-389), tridymite (ATSM 18-1170), and cristobalite ( $\alpha$ - or  $\beta$ -: ASTM 11-695 and 4-359)). In addition, the grain contains an unidentified substance, diffraction lines of which are given in parentheses. d, lattice spacing, in angstroms; I, relative intensity; b, broad; f, faint; vf, very faint]

hkl	Witwatersrand		Brannerite		Blind River <sup>1</sup>		Synthetic <sup>1</sup>		$\text{Pb}_3\text{O}_4$ ASTM 8-19		Quartz ASTM 5-490		Rutile ASTM 21-1276	
	d	I	d	I	d	I	d	I	d	I	d	I	d	I
---	---	---	---	---	1.561	6	---	---	---	---	---	---	---	---
---	1.554	1	---	---	1.558	6	1.5580	7	---	---	---	---	---	---
---	---	---	---	---	---	---	1.5292	8	---	---	---	---	---	---
---	---	---	---	---	---	---	1.5116	2	---	---	---	---	---	---
---	1.494	2	---	---	1.493	1(b)	---	---	---	---	---	---	---	---
---	---	1.482	vf	1.486	3	---	---	---	---	---	---	1.4797	10	---
---	---	---	---	---	---	---	1.4687	4	---	---	---	---	---	---
---	---	1.458	vf	1.450	4	1.4521	2	---	---	---	---	1.4528	10	---
---	---	1.441	vf	1.431	2	---	---	---	---	---	---	---	---	---
---	---	---	---	---	---	---	---	---	---	---	---	1.4243	2	---
---	---	1.410	vf	1.415	2	---	---	---	1.418	<1	---	---	---	---
---	---	---	---	1.384	3	1.3944	<1	1.382	7	---	---	---	---	---
---	1.374	1	1.367	2	1.376	4	1.3728	1	1.375	11	---	---	---	---
---	---	---	---	---	---	---	---	1.372	9	---	---	---	---	---
---	---	---	---	---	---	---	1.3471	4	---	---	---	1.3598	20	---
---	---	---	---	1.308	3	1.3109	4	---	---	---	1.3041	2	---	---
---	---	---	---	---	---	---	---	1.288	3	---	---	---	---	---
---	---	---	---	1.266	2	---	---	---	---	---	---	---	---	---
---	---	---	---	1.263	5	---	---	---	---	---	---	---	---	---
---	---	---	---	1.242	2	---	---	---	---	---	1.2441	4	---	---
---	---	---	---	1.239	2	---	---	---	---	---	---	---	---	---
---	---	---	---	1.234	5	---	---	1.228	2	---	---	---	---	---
---	---	---	---	1.218	3	---	---	---	---	---	---	---	---	---
---	---	---	---	1.200	3	---	---	1.1997	5	1.2006	2	---	---	---

<sup>1</sup> Patchett and Nuffield (1960).

TABLE 8. - *Some analyses of brannerite, in percent, reported in the literature*

[See Pabst (1954) and Frondel (1958)]

Origin	$\text{UO}_2$	$\text{ThO}_2$	$\text{PbO}_2$	$\text{FeO}$	$\text{TiO}_2$	$\text{CaO}$	$\text{SiO}_2$	Total <sup>1</sup>	$\text{UO}_2/\text{TiO}_2$ ratio	$\text{U/Ti}$ ratio
Kelly Gulch, Idaho	41.93	4.10	0.2	2.9	39.0	2.9	0.6	91.63	1.08	1.59
Cordoba, Spain	49.79	1.20	2.51	3.11	32.45	2.74	.16	91.96	1.53	2.25
Crocker's Well, South Australia	30.05	12.81	2.99	1.23	35.13	---	---	82.21	.86	1.26
Vosges, France	41.65	.30	3.43	5.4	35.2	2.7	3.3	91.98	1.18	1.74
Mono County, Calif.	38.41	5.00	---	2.4	32.9	2.8	.5	82.01	1.17	1.72

<sup>1</sup> Totals are incomplete; refer to Frondel (1958) for complete analyses.

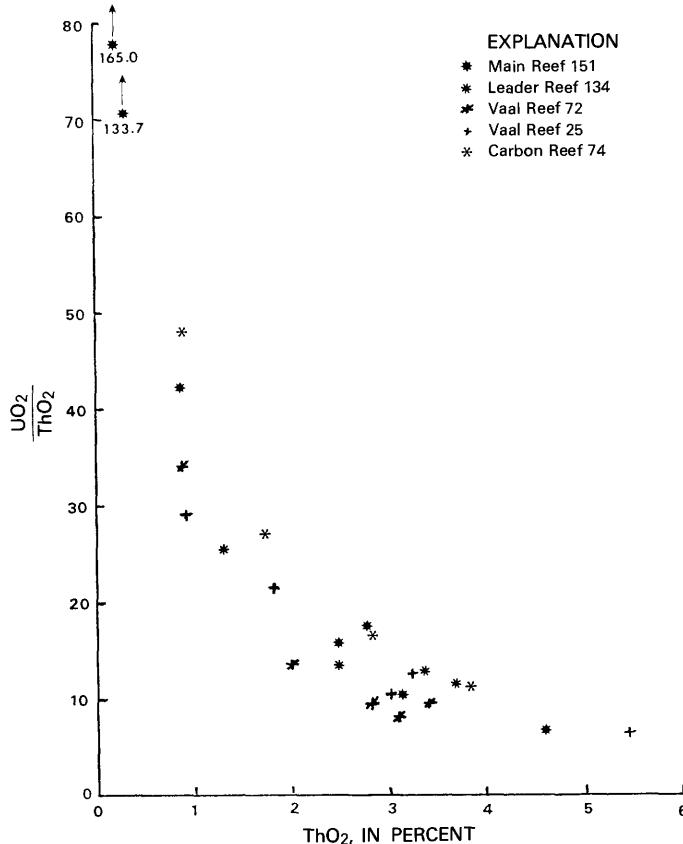


FIGURE 10.—Variations in thorium content of brannerite. The  $\text{UO}_2/\text{ThO}_2$  ratio varies as widely in brannerite as it does in uraninite, which is to be expected, as it is the alteration product of uraninite. Sample numbers in the explanation are from the Witwatersrand Collection of the X-Ray Fluorescence Section, Anglo American Research Laboratories, South Africa.

hosts (fig. 9). Finer milling of the ore to release the tiny crystals of brannerite (generally  $<5\mu\text{m}$ ) would be impractical. Only by dissolution of these refractory host minerals will recovery efficiencies significantly improve.

It has been suggested by Laxen (1973) that (U, P) compounds, if present, may be relatively refractory and slow to react with the leach solutions. Although Mihálik was able to identify a (U, P) compound in the Dominion Reef, no similar compound could be found during the present study of the reefs of the Witwatersrand and Ventersdorp Systems.

In an intensive search for phosphorus compounds associated with uranium, only monazite and a calcium phosphate mineral, presumably apatite, could be found, occurring as minute crystals in the matrix of the conglomerates and sometimes associated as discrete grains, with brannerite, as illustrated in figure 11. These discrete phosphate grains are not expected to interfere with the dissolution of the brannerite. Also, there is no evidence that the brannerite in the residues, compared

with that in the plant feed, is in any way more closely associated with these phosphate minerals.

#### UO<sub>2</sub>/ThO<sub>2</sub> RATIO IN THE REEFS AS A WHOLE

In table 9 a comparison is made of the overall  $\text{UO}_2/\text{ThO}_2$  contents of rock samples of the reefs, determined by X-ray fluorescence spectrometry, with the average obtained on uraninite and (or) brannerite grains in the same reef samples by electron microprobe. The ratio is always lower in the case of the overall rock samples, which suggests that there are other thorium-bearing minerals in the reefs.

TABLE 9.—Comparison of uranium/thorium ratios of reef rock samples as a whole, analysed by X-ray fluorescence spectrometry, and the average ratios of uraninite and (or) brannerite grains, analysed by electron microprobe

Sample	Reef	Whole rock		
		$\text{UO}_2$ (ppm)	$\text{ThO}_2$ (ppm)	$\text{UO}_2/\text{ThO}_2$ (ratio)
25	Vaal Reef	1,125	199	5.7
27(72)	Vaal Reef	442	166	2.7
34	Carbon Leader	914	61	15.0
151	Main Reef	19	5	3.8
134	Leader Reef	4,892	570	8.6
171	Cristaalkop Reef	5,243	468	11.2
184	Basal Reef	4,040	1,530	2.6

<sup>1</sup> Ratio of averages.

Investigation shows that zircon, monazite, and uranothorite are present, although relatively rare, and all three contain varying and sometimes appreciable amounts of thorium and relatively lesser uranium.

#### AGE OF WITWATERSRAND URANIUM-BEARING MINERALS

Polished sections containing Witwatersrand uraninite, brannerite, and zircon were sent to Drs. Hinckley and Anderson at the ARL Hasler Research Center, Goleta, Calif., for age determination by ARL IMMA Ion Microprobe. (For a description of the instrument and technique see, for example, Andersen and Hinckley, 1972.) The results that they obtained are as follows:

Mineral	$\text{Pb}^{207}/\text{Pb}^{206}$ ratio	Age (million years)
Uraninite (average)	$0.1078 \pm 0.0002$	$1,770 \pm 60$
Brannerite	$0.1103 \pm 0.007$	$1,810$
Zircon	$0.2017$	$2,845$

The age values were calculated using the newly recommended decay constants of Tatsumoto and others (1973). The average value for uraninite ( $1,770 \pm 60$  m.y. (million years)) is somewhat lower than previously determined values of approximately 2,000 m.y. (Davidson,

1960). The difference might lie in the fact that the ion beam sampled the lead within the crystal structure of the uraninite, eliminating any contamination from the environment of the grains.

The fact that the brannerite and uraninite are practically of the same age (limits of accuracy overlap) may indicate that the formation of brannerite by alteration of uraninite coincided with the period when the remaining uraninite recrystallized. This age, 1,800 m.y., agrees with the last stages of emplacement of the Bushveld Igneous Complex (Davidson, 1960), which thus may have been the source of the metamorphic activity.

The zircon age, 2,845 m.y., is somewhat older than that of the Witwatersrand System itself, approximately 2,500 m.y., and may be indicative of the age of crystallization or recrystallization of the source rocks.

#### PARAGENESIS OF GERSDORFFITE AND OTHER SECONDARY MINERALS

In geological terms, temperatures in the vicinity of 600°C are not very high, and relatively low grade metamorphism is expected to have occurred. Apart from very thorough cementing of the pebbles and matrix, mainly through the redistribution of quartz, a proportion of, if not all of, the gold was redistributed, virtually *in situ*, and gersdorffite and other secondary minerals were formed from the alteration of pyrite and less stable detrital minerals that were present.

Gersdorffite is invariably associated with gold, as noted by Saager (1969), with which it is commonly intergrown. So regular is this association that gersdorffite may be used as an indicator of gold content. The association is even closer than that of carbon and gold, where carbon may be present in abundance without the gold content being sympathetically high. (In mining practice, carbon content is commonly used as an indicator of gold content.)

Intergrowths of gersdorffite and gold were seen often to contain chalcopyrite and, more rarely, pyrrhotite, sphalerite, tučekite ( $Ni_9Sb_2S_8$ ), pentlandite, stibiopalladinite, and sudburyite, all clearly secondary minerals of the same generation. These intergrowths are notably xenomorphic, unlike secondary pyrite and galena of the same age, which invariably show distinct cubic crystal outlines.

At the same time, much rarer sperrylite, hollingworthite, ruthenarsenite, and iridarsenite are believed to have formed from reconstitution of the remains of

highly altered primary platinum group mineral grains. Alteration of phyllosilicates in the matrix, pyritization of iron oxides and ilmenite leading also to abundant needles of rutile, recrystallization of uraninite and crystallization of brannerite, and the growth of tourmaline, bravoite, cubanite, proustite, stromeyerite, tennantite, and mackinawite, all very rare, are expected to have taken place at this time.

#### GENERAL CONCLUSIONS

Investigation of the Witwatersrand cobaltite-gersdorffite assemblage has revealed, on the basis of crystal structure and chemistry, that temperatures in the vicinity of 400–600°C were achieved during metamorphism of the reefs.

It is apparent that the gold was remobilised and secondary base metal sulphides, sulpharsenides, and arsenides were formed during metamorphism. It appears that at the same time the uraninite was recrystallized and was partially altered to a uranium titanate mineral, which is confirmed to be brannerite.

Age determinations by ion microprobe show that the last period of recrystallization of uraninite, presumed to be the same period as gave rise to the metamorphic activity during which gersdorffite and related minerals were formed, took place about 1,800 m.y. ago. This coincides with the last stages of emplacement of the Bushveld Igneous Complex.

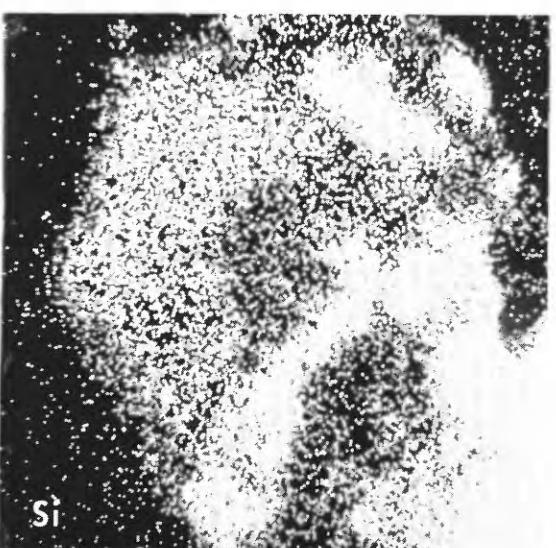
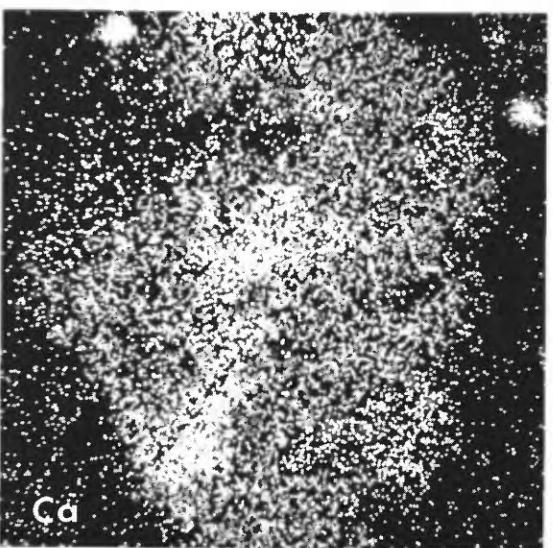
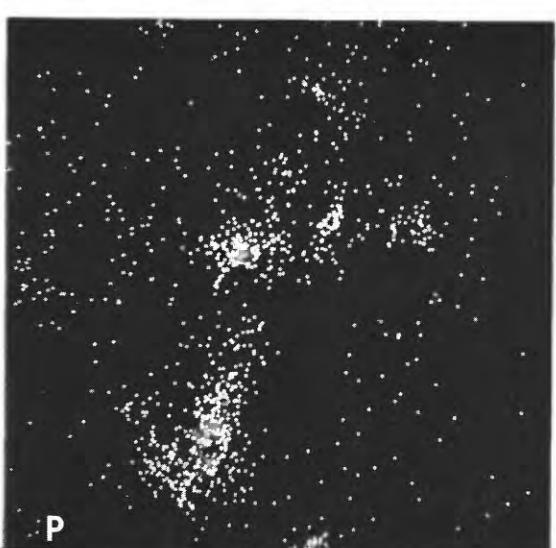
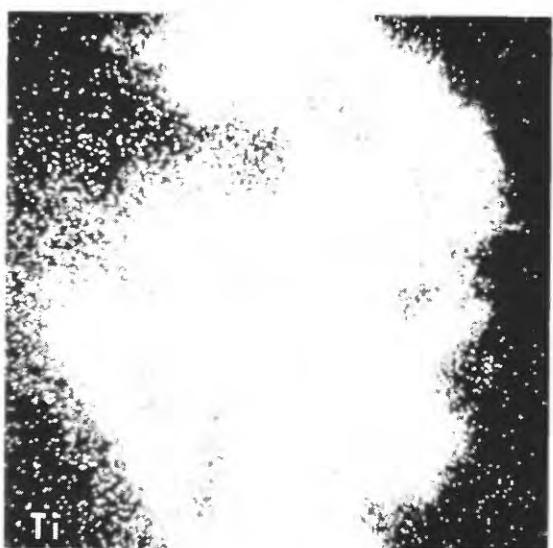
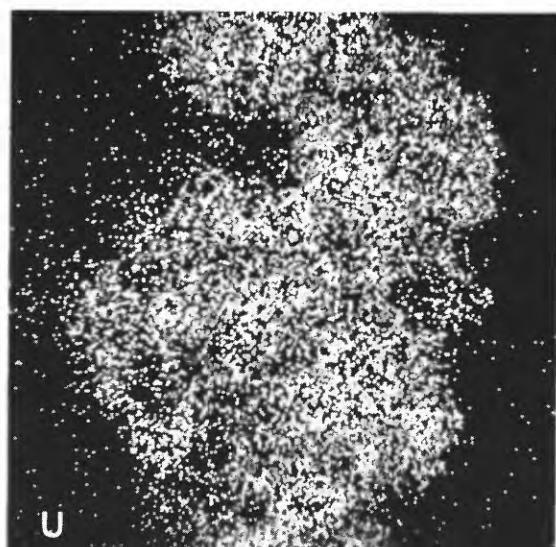
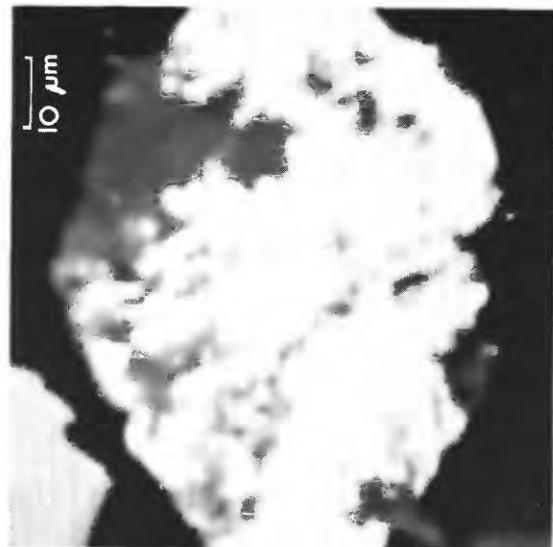
The variable  $UO_2/ThO_2$  ratios in uraninite grains not associated with carbon suggest that the grains are of detrital origin, having been derived from a number of granitic or pegmatitic sources. A similar variation in uraninite included in carbon suggests that the ancient "lichen," which is fossilised as the carbon, "digested" the detrital uraninite, rather than absorbed the  $UO_2$  and  $ThO_2$  from solution.

In addition to uraninite included in carbon, brannerite enclosed by refractory silicate minerals is an important source of uranium losses to uranium extraction plant residues.

#### ACKNOWLEDGEMENTS

The author is grateful to Anglo American Research Laboratories for having made this research project possible, to all his colleagues in the various sections of the Laboratories for their guidance and assistance, to gold mines of Anglo American Corporation of South

FIGURE 11. - Minute crystals of a calcium phosphate occurring as a discrete phase in an aggregate particle of brannerite, quartz, and chloritoid. No uranium-phosphorus compound was detected in the reefs examined. Scanning back-scattered electron and X-ray photomicrographs,  $\times 1,000$ . Vaal Reef, Vaal Reefs Mining and Exploration Co., Ltd.



Africa, Ltd., and Gold Fields of South Africa, Ltd., for samples, and to Applied Research Laboratories for the ion-microprobe determination of the age of Witwatersrand uranium-bearing minerals.

Finally, the author wishes to pay tribute to Mr. E. J. J. van Vuuren who, as the enthusiastic and inspiring coordinator of this project during his term of office at the Laboratories, was a fount of information by virtue of his long experience on gold and uranium extraction plants.

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## DISCUSSION

*Cannon*: I would like to ask you about the metamorphic temperatures that you quoted. They strike me as being quite high, and I wonder if you see this metamorphism in some of the more pelitic beds. You might have garnet or even staurolite porphyroblasts in pelitic material at those temperatures. Do you find those in the area?

*Feather*: No, they are extremely rare. These temperatures do seem to be higher than I think most people would like to see.

*Skinner*: I think the temperatures are too high because the ways in which you estimated them are not really valid ways to do it. The sphalerite temperature has for a long time now been shown to be invalid as a temperature measure. It is a function both of sulfur pressure and of temperature. The old curve, that presumably was used to get a minimum of 400 degrees, is itself wrong and has been replaced for perhaps eight or nine years now by much more accurate curves. The temperature that would be indicated if the sulfur pressure were controlled by pyrite, which one must assume it is, probably validly in the Witwatersrand, is going to be somewhere between zero and about 300 to 350 degrees, in that range. It's not going to be up to 400 or 500 degrees. The second way that you estimated temperature was with gersdorffite, cobaltite, and arsenopyrite. That work is

very suspect. The solvus determinations that you used to base the estimates on are very suspect because of extreme problems in kinetics in trying to get homogenization in those materials. Using aqueous synthesis, you can in fact make complete solid solution all the way across from gersdorffite to cobaltite with up to about 15 percent iron. You can do that as low as 150°C, which suggests that you could form all of those at 150 degrees and perhaps lower. I think the caution you threw out that you should await final decision until somebody else has looked at the diagram is probably a very wise caution. I personally think that that work is of little or no value at all in estimating temperature.

*Feather*: Thank you. I am very pleased for your comments. They certainly do throw out a word of very strong caution.

*Schidlowski*: I address a question to the expert. Does the occurrence of mackinawite narrow down things a little bit?

*Skinner*: It depends in part on the composition of mackinawite. If it were a very nickel-rich mackinawite, you might be able to push the temperature up to around 250 degrees. If it's just a straight iron mackinawite, it probably is below 200 degrees for it to remain stable.

*Schidlowski*: Just another comment. I was particularly happy to hear that the thorium tenor of the uraninite fragments within the carbon have the same values as the big grains. So I think that should dispose more or less definitely of all ideas that this uraninite has been precipitated the cold way.

*Simpson*: Just want to comment on those data. The ranges are the same but the distribution of the data within those ranges is different for the two environments.

*Feather*: I don't feel that we've analyzed sufficient grains to statistically treat the data and to draw any conclusions of that type.



# Uranium Mineralization of the Witwatersrand and Dominion Reef Systems

*By* P. R. SIMPSON *and* J. F. W. BOWLES

GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN  
QUARTZ-PEBBLE CONGLOMERATES

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## GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN QUARTZ-PEBBLE CONGLOMERATES

# URANIUM MINERALIZATION OF THE WITWATERSRAND AND DOMINION REEF SYSTEMS

By P. R. SIMPSON and J. F. W. BOWLES<sup>1</sup>

### ABSTRACT

Uranium-bearing minerals in the Witwatersrand and Dominion Reef sediments have been studied by ore microscopic, electron microprobe, fission track, and neutron activation analytical methods to determine the controls of uranium mineralization. In the Dominion Reef, which represents a high-energy blanket type of depositional environment, allogenic thorian uraninite occurs in hydraulic equivalence with allogenic pyrite, quartz, and possibly also gold in the sediments that have uranium-thorium ratios between 3.1 and 5.6, indicating substantial amounts of thorium-rich resistate minerals.

The Witwatersrand sediments have uranium-thorium ratios ranging between 7.1 and 19.6, indicating lesser amounts of resistates, which is consistent with the lower-energy depositional environment. The proximal or nearshore deposits are of blanket type but are distinguished from the Dominion Reef by the abundance of concretionary pyrite formed within the basin and the presence of carbonaceous matter. The distal deposits formed at greater distance from the shoreline contain decaying organic material, which has precipitated both uranium and gold from solution. Subsequent metamorphism has resulted in the formation of carbonaceous material bearing a finely disseminated low-thorium pitchblende and a fine dissemination of gold associated with sulphides and arsenides.

Further evidence of the existence of uranium in solution is to be found in the blanket deposits. In this case fine disseminations of uranium (>500 parts per million) occur in clay minerals within concretionary pyrite nodules and in lenses formed of clay minerals in Witwatersrand blanket deposits. They represent reduction-deposition of the soluble uranyl ion below the sediment-water interface where conditions are reducing.

Allogenic thorian uraninite from the present-day Indus River has a texture, composition, and association with gold and pyrite similar to allogenic uraninite in the Witwatersrand and Dominion Reef Systems. Thorian uraninite is a stable phase over large distances in this river. Hence it would appear quite unnecessary to postulate a reducing atmosphere for the transportation of detrital uraninite. Moreover the retention of sulphate and uranyl ions in solution in the model proposed here suggests that the atmosphere was oxidizing at the time of deposition. This conclusion indicates the likely occurrence in younger sediments of mineralization of this type provided the necessary geological criteria are met.

### PREFACE

By S. H. U. BOWIE, F.R.S.

The problem of the origin of the mineralization of the Witwatersrand and Dominion Reef has been one of the great controversies of the past 50 years. Any new attempt to solve it requires the application of the most up-to-date mineralogical techniques. Although unequivocal results have not yet been obtained, important new information on the bimodal nature of the uranium mineralization has been established.

The significance of this study, which illustrates the application of techniques that will become more commonplace as the 21st century approaches, lies in providing essential information on the origin of uranium in quartz-pebble conglomerates. More than 35 percent of known uranium reserves occur in such rocks, particularly in Canada and South Africa but also, as recently reported, in Algeria. Previously many geologists have held that uranium can only be enriched to economic grades in Precambrian rocks if anoxic atmospheric conditions prevailed at the time of sedimentation. However, the remarkable similarity between the subeconomic concentrations of thorian uraninite in the present-day Indus Valley and that of the Dominion Reef and Witwatersrand Systems, as well as other evidence, invalidates any such concept.

Further work is required to complete the story of the Witwatersrand mineralization, and it is hoped that multidisciplinary investigations will continue. The full resolution of the genesis of the mineralization could result in new discoveries that are essential to the availability of uranium for future energy needs.

### INTRODUCTION

Uranium mineralization in the Witwatersrand and Dominion Reef sediments is presently and has been,

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since its discovery by Cooper in 1923, the subject of debate and speculation as to its origin.

Mineralization has been described variously as the product of hydrothermal processes by Graton (1930) and sedimentary processes by Ramdohr (1958) and others. The detrital hypothesis for the origin of uraninite has been taken to account for all primary mineralization in the basin by some workers. This idea has been further developed to the point where a reducing atmosphere was invoked as an essential prerequisite to the transportation of detrital uraninite grains (Liebenberg, 1955; Grandstaff, 1975). This has led to the notion that uraniferous conglomerates are thereby restricted by these atmospheric conditions to the Archaean or early Proterozoic (Robertson, 1974).

The purpose of this study is to re-evaluate the uranium mineralization using mineralogical methods since the implications of the earlier work for the further discovery of uraniferous conglomerates would appear to be unduly restrictive.

All specimens available to the authors were sectioned for study to obtain as broad a view of the principal types of mineralization as possible, though it is appreciated that the coverage of this basin is by no means as complete as could be wished. The outline geology, principal mining areas, and section numbers of specimens studied are shown in figures 1 and 2.

In pursuing this investigation, two questions have been uppermost in our minds: firstly, does the formation of these and similar deposits elsewhere require a reducing atmosphere or merely reducing conditions of deposition beneath the sediment/water interface; and secondly, what are the respective roles of uranium as detrital grains of uraninite and as uranyl ions in solution. Answers to these two questions will have considerable relevance to the understanding of the mode of formation and estimating the potential for new discoveries of this type of mineralization.

## EXPERIMENTAL METHODS

The details of the complex mineralization and the distribution of uranium were elucidated by ore microscopy, electron probe microanalysis, and fission track registration. The techniques of the first have been recently described by Bowie and others (1975), and the last is discussed in the section on uranium micromapping.

The electron-probe analyses were undertaken with a Cambridge Microscan-5 instrument fitted with a Canberra energy dispersive analytical system. The latter was used for the analysis of titanium, manganese, iron, lead, thorium, uranium, and yttrium, whilst calcium and cerium were analysed simultaneously by the Bragg spec-

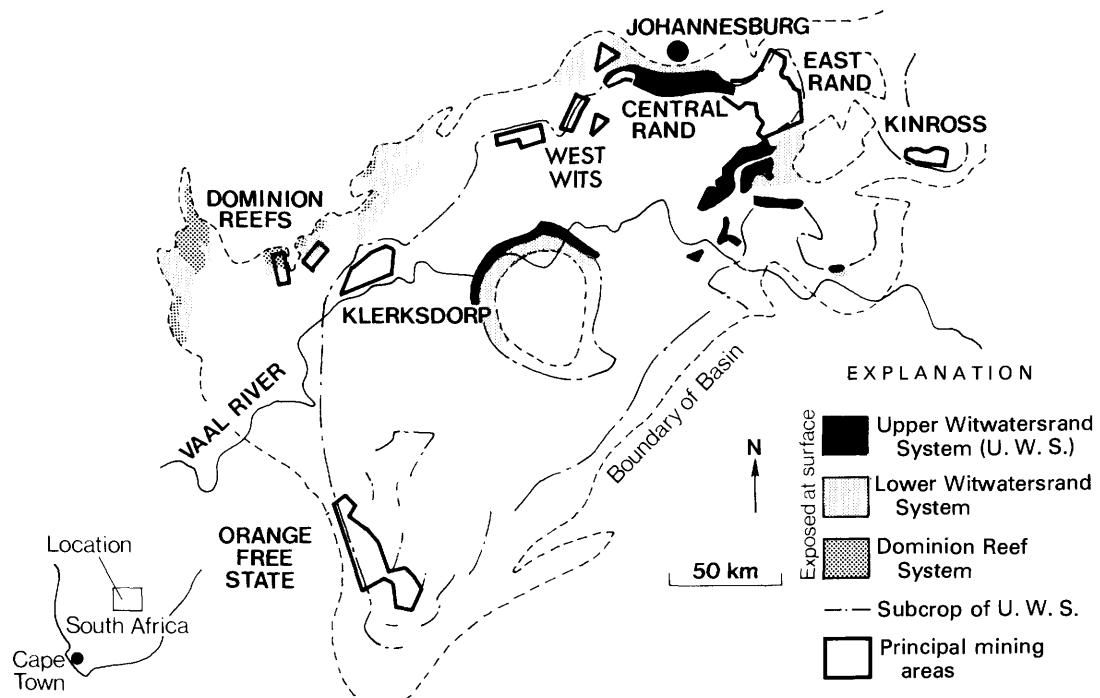


FIGURE 1.—Map of the Witwatersrand Basin illustrating sub-Ventersdorp geology and showing principal mining areas.

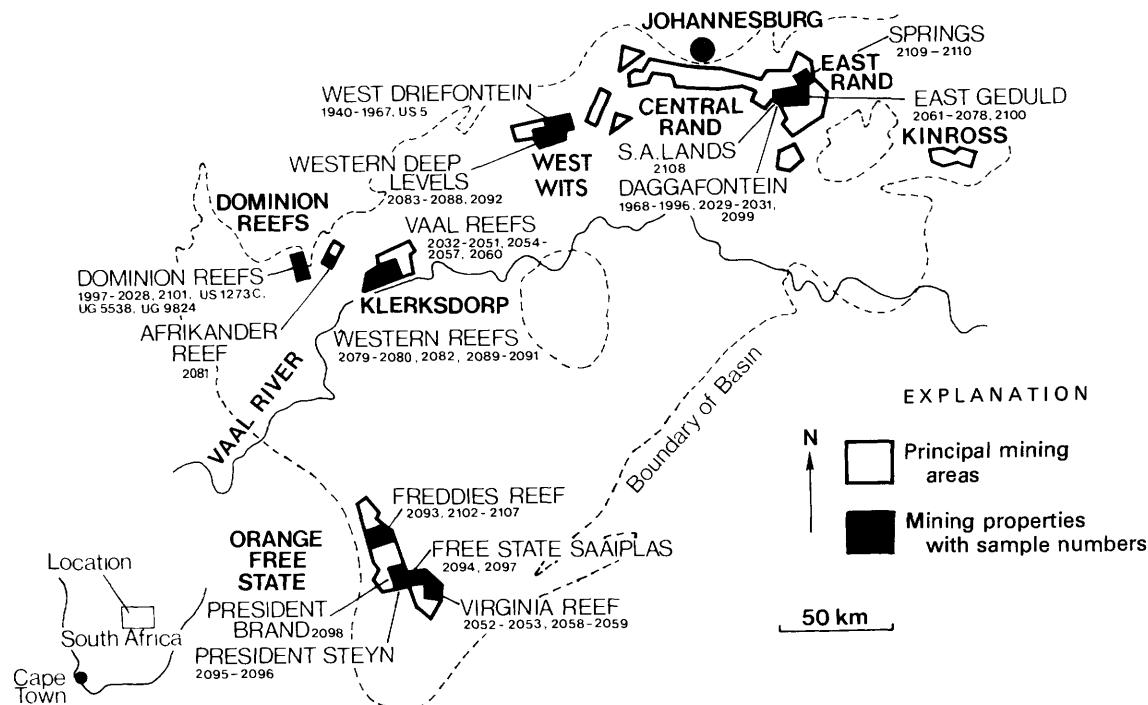


FIGURE 2.—Map of the Witwatersrand Basin illustrating principal mining areas and sample localities. The numbers in small print refer to polished thin sections. Polished sections from the reference collection are prefixed **UG** or **US**.

trometers of the microprobe. X-rays from each specimen were counted for 250 s at a low beam current of  $1 \times 10^{-9}$  A, which conditions were suitable for both analytical systems. Data were reduced by a PDP11 computer, which calculates concentrations of the elements present at one analytical point while the next is being analysed. This technique enables an analysis for nine elements to be produced every 4–5 min. Pure metals were used as standards for manganese, iron, thorium, and uranium; a rare-earth glass REE-3, kindly provided by the University of Oregon (Drake and Weill, 1972), for calcium, yttrium, and cerium; and chemically analysed rutile and galena, for titanium and lead, respectively. The analytical methods for wavelength-dispersive analysis described by Bowles (1975) were used for calcium and cerium. In another paper, Bowles (in preparation) describes the quantitative analytical programme for the energy-dispersive equipment and compares selected results with those obtained with the Bragg spectrometers. The results are closely comparable for all elements with the exception of uranium, for which the energy-dispersive results are consistently 3 percent lower than the Bragg spectrometer results. Investigation has shown that as between the pure metal and the oxide there is a change in the ratio of the  $L\alpha_1$  to  $L\alpha_2$  intensities. The lower resolution of the energy-dispersive equipment records both X-ray emissions as a single

peak, so that a variation in the ratio does not affect the results. The analyses presented here for uranium and thorium are those obtained using the energy-dispersive equipment. The measured concentrations were corrected using the quantitative correction programme devised by Mason and others (1969).

## MINERALOGICAL EVIDENCE

### PYRITE

Pyrite is the principal opaque mineral of the Witwatersrand and Dominion Reef Systems. In hand specimen rounded grains of pyrite, commonly known as "buckshot" (Feather and Koen, 1975), display clearly defined sedimentary features with well-developed bedding and crossbedding in layers also containing sand-sized grains of quartz. In the pebble-size conglomerate, pyrite occurs as a discrete disseminated phase together with other sand-size constituents, principally quartz, in the matrix. Radioactivity is closely associated with pyritic zones in the hand specimen, and those were therefore selected for more detailed investigation. Indeed, the presence of pyrite in the hand specimen is one of the clearest indications of the likely presence of anomalous radioactivity in the suite of specimens studied.

Ramdohr (1958), Saager (1970), and Köppel and Saager (1974) distinguished three types of pyrite in the Witwatersrand ore: (I) allogenetic, (II) concretionary authigenic, and (III) reconstituted authigenic. Type I pyrite has rounded and abraded outlines; it is completely fresh, is uniformly compact and homogeneous, and has a distinctive range of inclusions (fig. 3A). There is no doubt it is correctly described as allogenetic. It is recognized in this study as the dominant pyrite type of the Dominion Reef System, whereas in the Witwatersrand any one of the three types may be dominant. Pyrite of this type also occurs partially or completely enclosed within very coarse quartz grains; it displays euhedral grain boundaries towards the host quartz but has a subrounded and abraded margin where it forms the exterior of the composite grain (fig. 3A). Optical study and electron-probe microanalysis indicate that galena is the commonest phase included in allogenetic pyrite. The exsolution blebs of galena are distinct from marginal intergrowths of pyrite and galena resulting from the migration of radiogenic lead from coexisting uraninite grains (fig. 4 A, B).

Pyrite of type II is present in variable amounts in all samples studied from the Witwatersrand System. It is readily distinguished from type I by its porous nature and from type III by its rounded outline. The concretions are generally coarser than coexisting allogenetic pyrite, ranging from granular to coarse sand size, although the individual pyrite crystallites that form the concretions are often 10  $\mu\text{m}$  in diameter or less. However, in any one specimen the concretions are heterogeneous in nature with regard to shape, size, and crystallite composition. Some concretions have spherically zoned alternating sulphide and silicate shells; others exhibit a layered structure. Some are so dark and fine grained as to be indistinguishable from melnikovite or colloform pyrite, which is thought to represent a crystallized  $\text{FeS}_2$  gel.

In the concretionary pyrite, the coarser crystallites, which are better crystallized and brighter, are texturally very similar to framboidal pyrite. The pore spaces between crystallites represent about 10 percent by volume of the concretions, although zones occur where the pyrite content may be as low as 10 percent. Even within one concretion the texture may be very variable, ranging from fine-grained disseminated pyrite to a more compact equigranular variety without pore spaces between crystallites. Saager and Mihálik (1967) proposed that these concretions formed *in situ*, since they claimed the "extremely delicate nature of the concretions exclude any long transport distance." However, it should be noted that the crystallites forming the concretions are in most cases interconnecting, thereby providing a rigid skeleton, and the pore spaces between crystallites

are filled by a matrix comprising quartz and a 7 Å clay mineral (unidentified), which provides additional strength to the concretions. Most of these composite structures have survived later metamorphism and deformation relatively unaltered. Moreover, coexisting detrital quartz granules show appreciable pressure solution effects along their margins, though both allogenetic and concretionary grains occurring in close proximity are underformed, testifying to their relative rigidity and strength.

In our view the heterogeneity of the concretionary pyrite in any hand specimen and the evidence of some rounded granules that have apparently been fractured and abraded before deposition indicate that most of the concretionary pyrite has been transported to its present location and did not form *in situ* (fig. 5). The variability of this pyrite in any particular sample is considered to reflect a source area and selective effects of transportation processes. The intimately intergrown nature of the sulphide and silicate phases in the concretions is most probably a primary feature inherited from the source area and is not due to subsequent introduction of silica, since the regular fine-scale laminations observed in some cases preclude the possibility of later introduction (fig. 6). Hallbauer (1975) has described subspherical concretions of this type extracted for study by scanning electron microscopy. However, there are rare occurrences of zoned spherical concretions. The example shown (fig. 3B) has a thin inner zone of chalcopyrite with rare grains of gold and a siliceous rim partially overgrown by pyrite. The spherical outline is clearly a primary feature, and deductions about the extent to which transport may or may not have occurred cannot in general be made from a study of external form alone.

Electron probe scans of the concretionary pyrite in one specimen (2463A) indicate that the lead content, represented by galena, varies widely in amount and distribution from concretion to concretion. In some (fig. 7A) galena is rare. Others have a high content of disseminated galena in 10  $\mu\text{m}$  grains uniformly distributed throughout the concretion (fig. 7B), and some have fine veinlets of galena probably representing later introduction of lead.

Pyrite of type III occurs as euhedral grains in the conglomerate matrix or forms overgrowths on detrital components. It is clearly authigenic in origin. Köppel and Saager (1974) report enrichments of chalcopyrite, sphalerite, and galena associated with the development of this pyrite type and also remark that the presence of dykes intrusive into the Witwatersrand rocks has influenced its development. However, these authors attribute the bulk of this reconstituted pyrite to regional metamorphism. The presence of grains of gold, in some

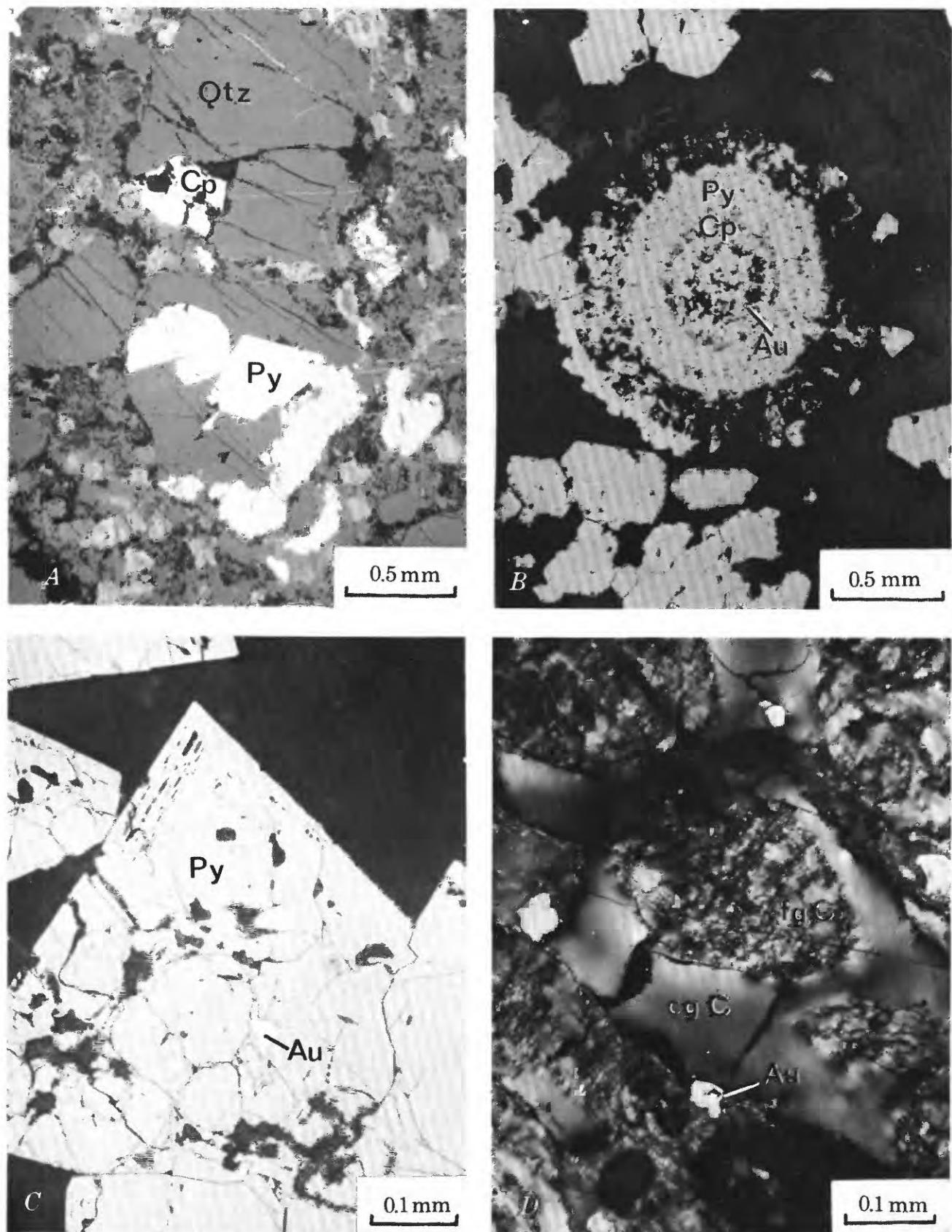


FIGURE 3.—A, Banket ore sample showing allogenic pyrite (Py), chalcopyrite (Cp), and quartz (Qtz); Dominion Reef, (US 1273C), reflected light, air. B, Concretionary pyrite of spherical type composed of alternate layers of pyrite (Py) and chalcopyrite (Cp), with a grain of gold (Au). Silicates occur in the body of the concretion and particularly in the rim, as well as in the porous matrix (black). West Driefontein, West Wits (PTS 1966); reflected light, oil. C, Authigenic reconstituted pyrite (Py) containing gold (Au). West Driefontein, West Wits (PTS 1958); reflected light, oil. D, Carbonaceous matter showing coarse (cgC) and fine-grained (fgC) carbon, with gold (Au). Carbon Leader, West Wits (US 5); reflected light, oil.

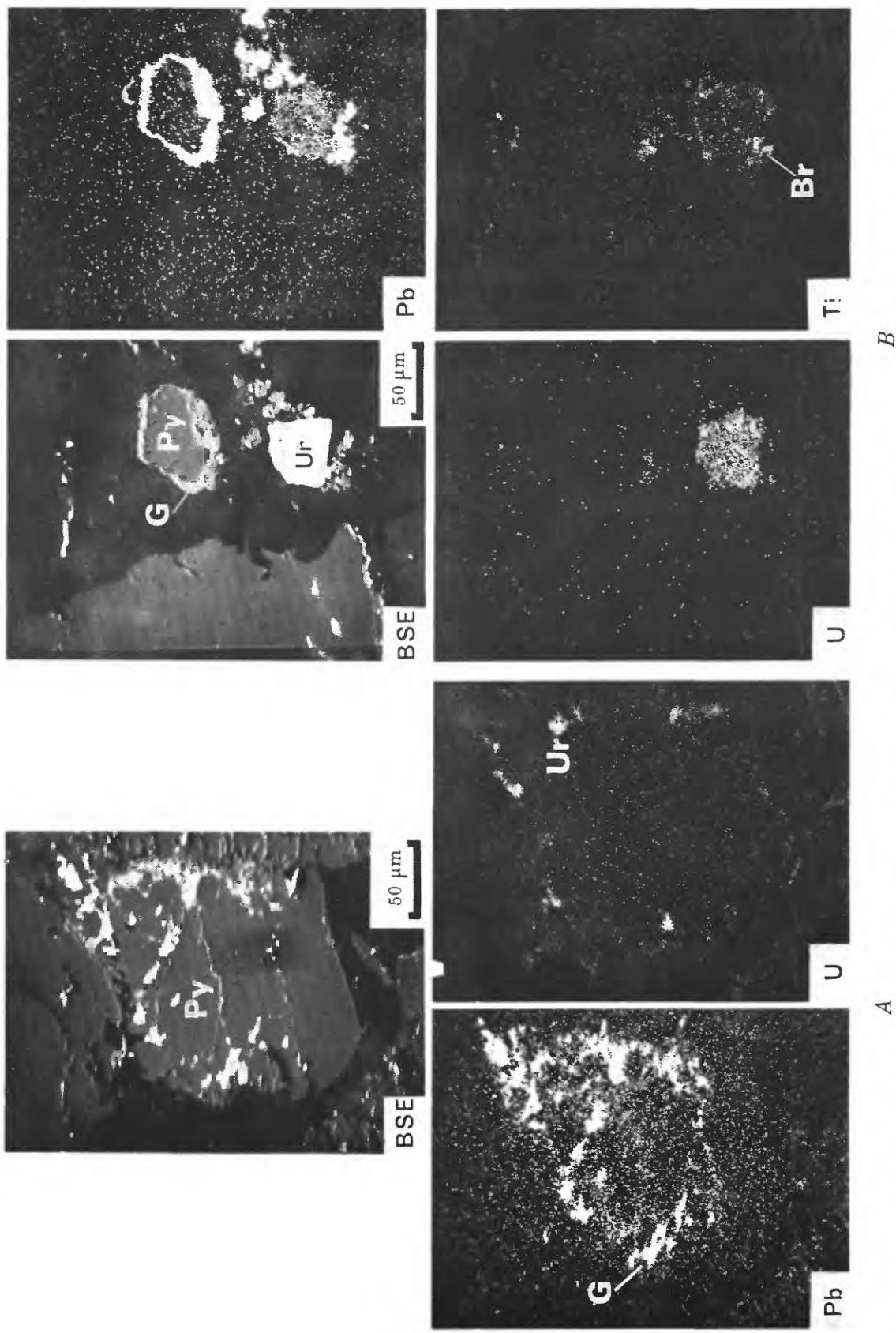


FIGURE 4.—Electron-probe scanning images showing (A) allogenic pyrite (Py) containing galena (G) and rimmed by uraninite (Ur) and (B) localized migration of radiogenic lead from uraninite (Ur) to the rim of an adjacent allogenic pyrite (Py) grain, with formations of minor amounts of brannerite (Br); both from Basal Reef, President Brand, Orange Free State (PTS 2463A).

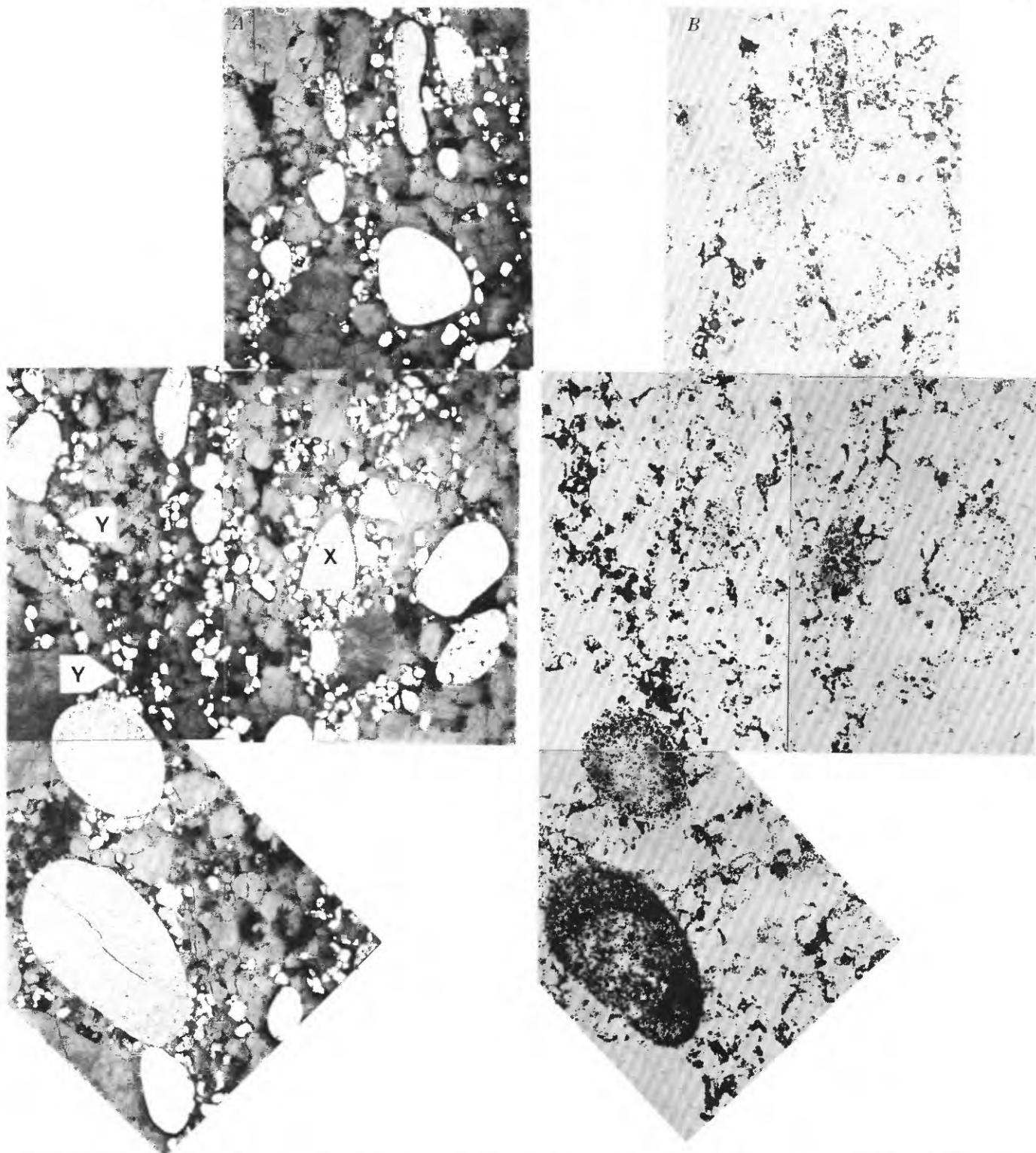


FIGURE 5.—A uraniferous pyritic band consisting of larger concretionary and smaller allogenic pyrite (white). *A*, Reflected light. *B*, Corresponding Lexan print. Uranium is enriched in some concretionary pyrite grains but not in others (see Lexan print). One concretionary pyrite grain (*X*) has been fractured and further worn before deposition. Uranium present in the matrix is accounted for by allogenic grains of uraninite (*Y*) and interstitial clay minerals. Basal Reef, President Brand, Orange Free State (PTS 2463B).

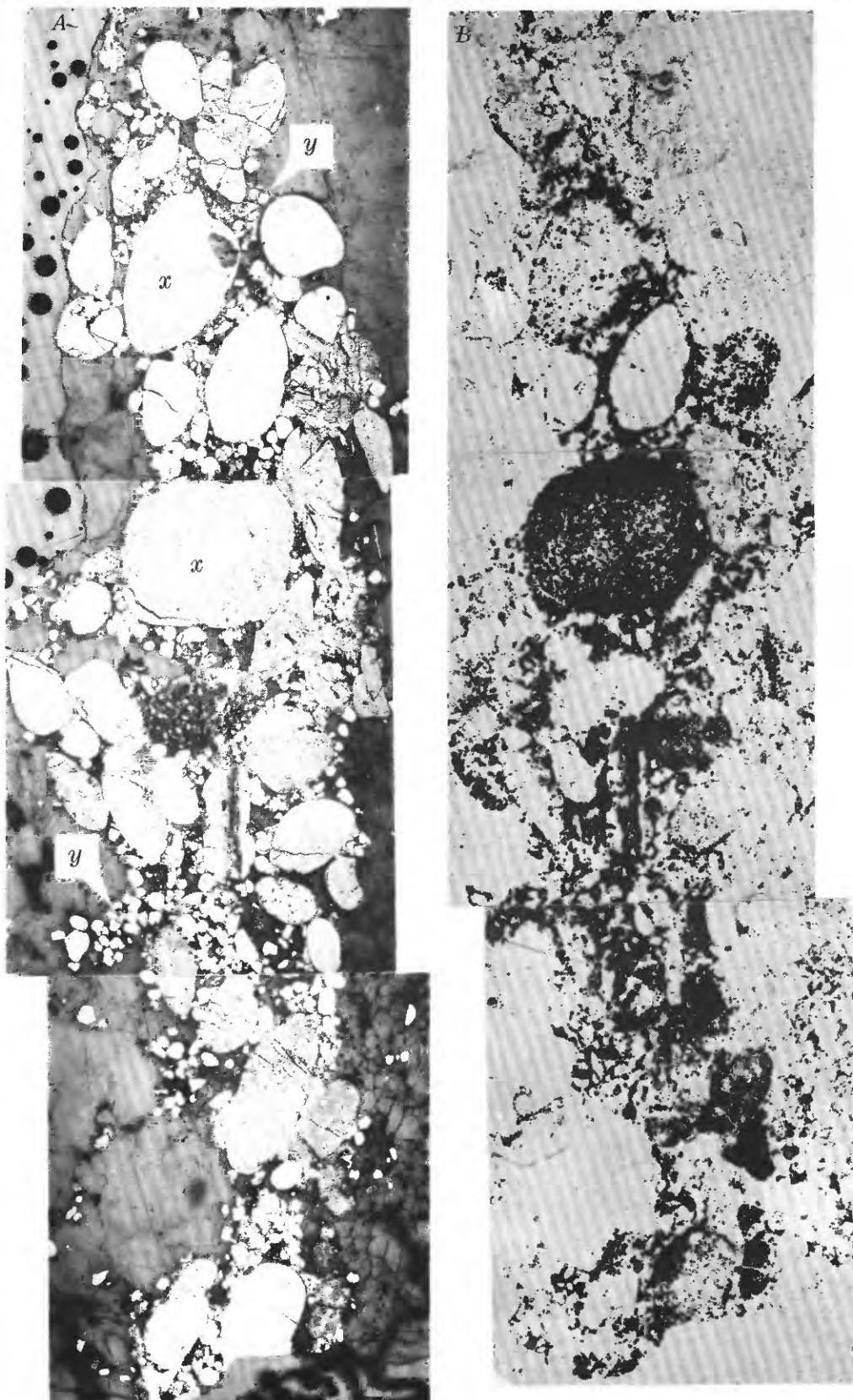


FIGURE 6.—Uraniferous pyrite band consisting of mainly concretionary pyrite grains (white), which are a heterogeneous assemblage with regard to uranium enrichment (see Lexan print and compare grains marked *x*). *A*, Reflected light. *B*, Corresponding Lexan print. Uranium present in the matrix is accounted for by the presence of allogenetic grains of uraninite (*y*) and interstitial clay minerals. Basal Reef, President Brand, Orange Free State (PTS 2463A).

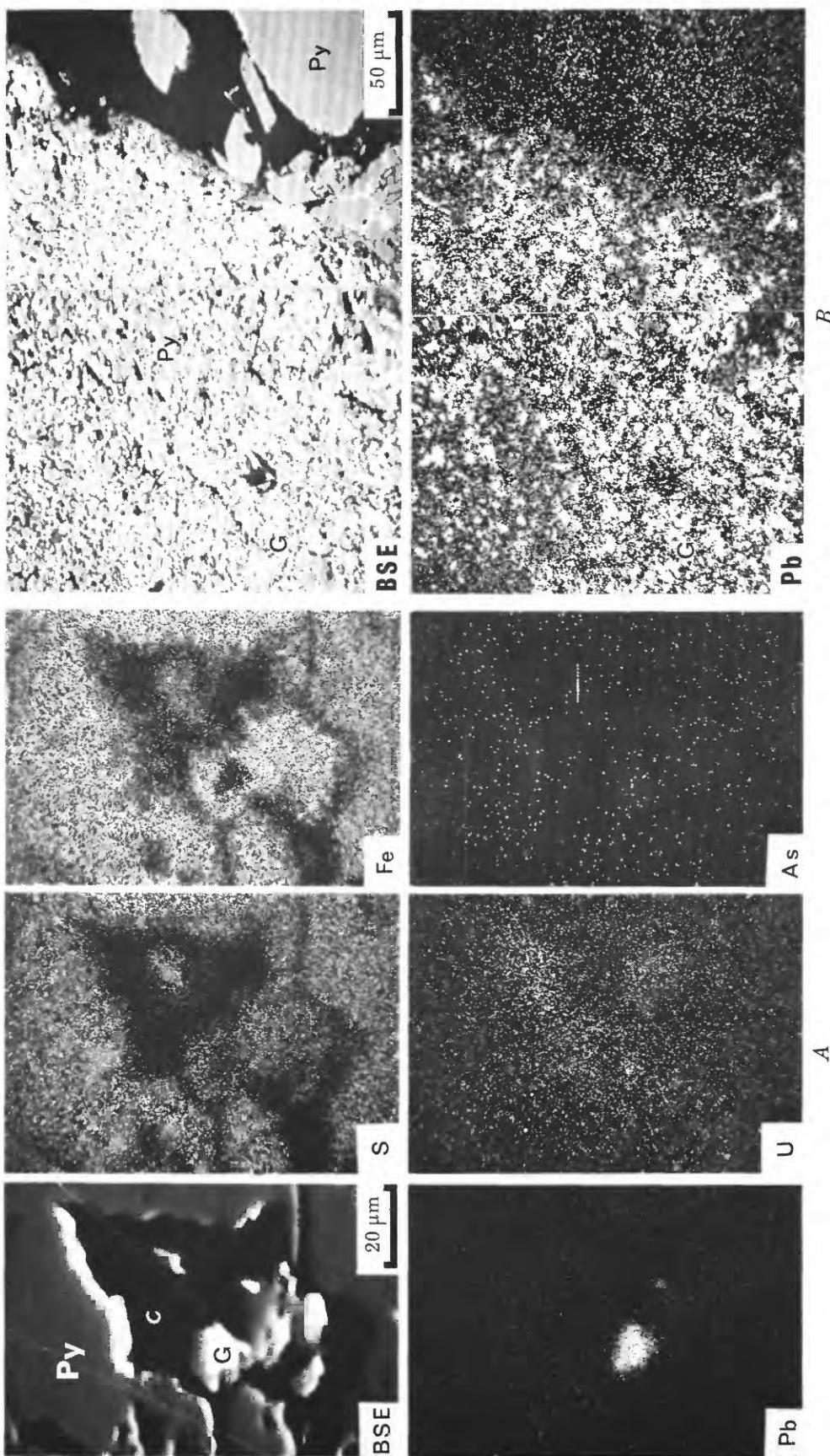


FIGURE 7.—Electron-probe scanning images showing (A) a discrete galena (G) grain occurring in a low-lead type pyrite concretion (Py). Uranium is present in the interstitial clay phase (C). (B) galena (G) distribution in concretionary pyrite of high-lead type is shown for comparison with low-lead type; both from Basal Reef, President Brand, Orange Free State (PTS) 2463 A.

cases abundant, noted in pyrite of this type during the present study (fig. 3C), indicates that gold and pyrite were locally remobilized and redeposited together.

#### URANINITE

Before describing the details concerning the analysed material, it is necessary to comment on the use of the terms uraninite and pitchblende. Uraninite is the name for the species as a whole, including all of its varieties (Frondel, 1958). Pitchblende is a variety of uraninite distinguished primarily by its appearance. In the Witwatersrand all detailed work to date shows that uraninite found there can be divided into two groups: (1) a well-crystallized variety with significant thorium content and (2) a poorly crystallized variety, which occurs only in association with organic matter and has a low thorium content. As these tend to be found in different environments it is important to distinguish between them, so in this paper "uraninite" is restricted to the well-crystallized variety and "pitchblende" to the other.

Uraninite occurs as discrete allogenic grains varying in shape from euhedral to subrounded and rounded grains, usually within a limited size range in the fine to very fine sand sizes (0.06–0.25 mm), and is often fractured. It occurs in this form both in the Dominion Reef and Witwatersrand Systems. Thorium content is the best indicator of primary differences in populations of uraninite grains since lead content within and between grains may be quite variable owing to exsolution and migration of radiogenic lead. This lead mobility has a greater effect on the uranium content than on thorium, which has a more homogeneous distribution and is less readily displaced. It is possible to observe in some cases the migrations and reaction of radiogenic lead with nearby allogenic sulphide to form a galena rim (fig. 4B). The same section the close proximity of brannerite to decaying uraninite, with nonuraniferous titanium minerals at a greater distance, is indicative of local migration and reaction of uranium with titanium minerals in the sediments.

Electron microprobe analyses show in this study that the Dominion Reef uraninite has a median  $\text{ThO}_2$  content of 6.6 percent with a range from 5.4 to 8.4 percent (fig. 8C) and a median  $\text{UO}_2/\text{ThO}_2$  ratio of 8.5 with a range from 6.4 to 10.9 (fig. 9).

In the Vaal Reef of the Witwatersrand System, allogenic uraninite analysed in a columnar carbonaceous band (PTS 2034) has a median  $\text{ThO}_2$  content of 3.5 percent with a range from 0.9 to 9.1 (fig. 8B) and a median  $\text{UO}_2/\text{ThO}_2$  ratio of 19 with a range of 8.3 to 48 (fig. 9). The separate populations of  $\text{ThO}_2$  in uraninite grains from the Dominion and Vaal Reefs were tested by analysis of variance methods and found to be significantly dif-

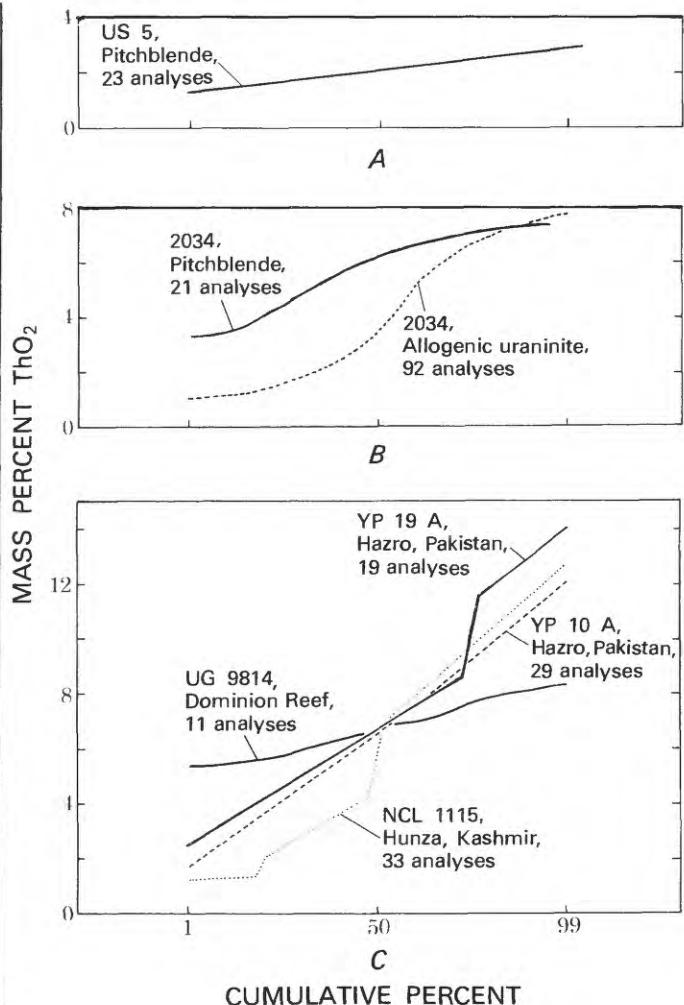


FIGURE 8.—Cumulative-frequency diagrams for  $\text{ThO}_2$  obtained by electron probe microanalysis. A, Mass percent  $\text{ThO}_2$  of fine pitchblende grains in granular 'flyspeck' carbon, Carbon Leader, West Wits (US 5). B, Mass percent  $\text{ThO}_2$  of coexisting granular allogenic uraninite and fine-grained pitchblende in columnar carbon, Vaal Reefs, Klerksdorp (PTS 2034). C, Mass percent  $\text{ThO}_2$  of allogenic uraninite for one sample from the Hunza River, Kashmir, a tributary of the Indus River, two samples from Hazro, Pakistan, on the Indus, and one sample from the Dominion Reef.

ferent at the 0.001 level, but variations within these populations are not significant. Significant positive correlation between calcium and manganese and between iron and manganese is common to both uraninite populations.

The  $\text{ThO}_2$  content,  $\text{UO}_2/\text{ThO}_2$  ratio, and correlation between calcium and manganese and iron and manganese probably reflect conditions of crystallization in the source area. The higher median  $\text{UO}_2/\text{ThO}_2$  ratios for the Vaal Reef uraninite and the lower thorium content compared with uraninite from the Dominion Reef indicate some variation in the source area for the two horizons.

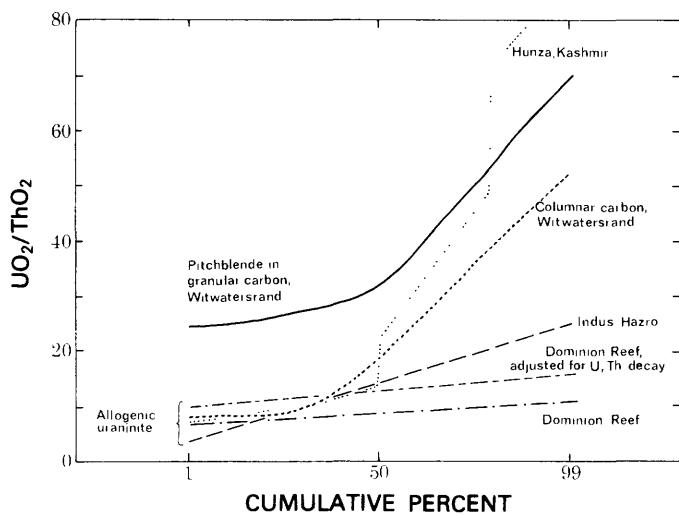


FIGURE 9.— $\text{UO}_2/\text{ThO}_2$  ratios for separate populations of uraninite and pitchblende plotted on a cumulative frequency diagram. The highest median values are given by fine-grained pitchblende in granular carbon from the Carbon Leader (US 5). Allogenic uraninite from the Vaal Reefs in the Witwatersrand (PTS 2034) has a lower ratio, which is still higher than that of uraninite from the Dominion Reef (UG 9824). Detrital uraninite from the Indus River at Hazro (YP 10A and YP 19A) and the Hunza River, Kashmir, a tributary of the Indus, is plotted here for comparison with the Dominion Reef and Witwatersrand samples as they occur at the present time. The Dominion Reef samples have also been recalculated to take account of decay of uranium and thorium since crystallization at  $\sim 3,000$  million years ago, and this correction tends to reduce the small difference between populations.

However, fluid-inclusion studies do not reveal any differences in the quartz pebbles (Shepherd, 1977).

#### GRAIN SIZE OF URANINITE AND ASSOCIATED PHASES

A typical sample (UG 5538) from the Bramley section of the Dominion Reef Mine, Klerksdorp, was examined in detail by grain-size analysis. Coexisting allogenic grains of quartz, pyrite, uraninite, and gold were measured by means of the Humphries eyepiece micrometer, which can be used to classify grains directly according to the phi scale. Each population, except for gold, consisted of a hundred grains measured along the Feret diameter, which is the maximum grain length projected onto a fixed line. The populations appear to be single populations in each case resulting in linear cumulative-frequency curves. Sample means and standard deviations were calculated in terms of phi units, from which the standard error of the mean was obtained. This was transferred to a chart containing graphical results calculated for Stokes law and the square-root law combined (Tourtelot, 1968), which indicates that quartz, pyrite, and uraninite closely approximate to a hydraulically equivalent assemblage (fig. 10). The gold grains counted were too few to be statistically significant but

nevertheless appear too small for exact hydraulic equivalence. This point was noted by Coetzee (1965), but no satisfactory explanation has yet been proposed.

#### URANIFEROUS AND AURIFEROUS CARBONACEOUS MATERIAL

Uranium and gold in association with carbonaceous material is one of the most important modes of occurrence of these metals in the Witwatersrand Basin, particularly in view of the uniformly high grades of mineralization encountered and the wide lateral extent of such horizons as the Carbon Leader in the Far West Rand (Davidson and Bowie, 1951; Bourret, 1973). Any satisfactory explanation of the nature of this metal-rich carbonaceous matter should take account of the following observations:

1. The Carbon Leader, which represents a most striking enrichment of uranium and gold, forms a very restricted horizon in the reef never greater than 13 mm thick (Davidson and Bowie, 1951).
2. The levels of both uranium and gold over a wide lateral extent are consistently high.
3. The  $\text{UO}_2/\text{ThO}_2$  ratios in the carbonaceous type of mineralization are higher compared with the blanket type of both the Witwatersrand and Dominion Reef Systems, and the absence of the carbonaceous type of mineralization in the latter.
4. Mineralization is absent in certain occurrences of carbonaceous material.

Some authorities consider the carbonaceous material represents fossilized remains of Precambrian plants (Hallbauer, 1975). Whether such is its origin or not, there is little doubt that it has exercised a key role in the formation of these deposits.

The carbonaceous material comprises granular and columnar varieties, which have many features in common whilst differing in certain important aspects (Davidson and Bowie, 1951). According to the analyses of Davidson and Bowie this material contains 43 percent carbon and 2 percent hydrogen, and they called this phase hydrocarbon. However, we have not attempted to define the organic constituents of all the carbonaceous materials studied here and have therefore retained a more general nomenclature used by Vine and others (1958).

The granular or "fly speck" material (fig. 11) consists of ovoid to rounded grains (100–400  $\mu\text{m}$ ) with an interstitial matrix of subrounded allogenic pyrite, chromite, and concretionary pyrite ( $\sim 50 \mu\text{m}$ ). The granules are partially moulded around interstitial grains, particularly where they lie along grain boundaries between granules (fig. 11), but the carbonaceous matter at no point either replaces or invades the opaque

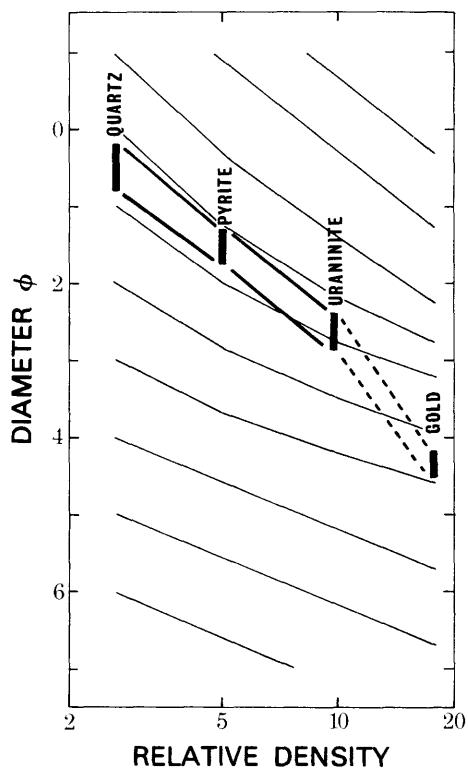


FIGURE 10.—Standard error of the mean size for coexisting quartz, pyrite, and uraninite plotted on the phi scale against specific gravity. The light curves are solutions for Stokes' law and the square-root law combined where they overlap. The relative grain sizes of the suite of minerals plotted in heavy type indicate that quartz, pyrite, and uraninite are hydraulically equivalent. The gold is smaller than would be expected for hydraulic equivalence, but the number of grains measured is inadequate for full statistical treatment. Bramley high grade, Dominion Reef (UG 5538). (Diagram after Tourtelot, 1968.)

minerals. The granules have mottled extinction under crossed polars in reflected light and contain two main phases of carbonaceous matter (fig. 3D), consisting of relatively coarse-grained smooth areas with undulose extinction interspersed with fine-grained material having a similar reflectance of 14.3 percent (Type I of Davidson and Bowie, 1951, p. 6). The fine-grained material is veined along grain boundaries by a dendritic system containing pitchblende, gold, pyrite, galena, and gersdorffite. The gold and gersdorffite are commonly intergrown, as are the gold and pitchblende. Galena tends to form separate grains but is also intergrown with pyrite. These textural and mineralogical relationships have been confirmed by both scanning and quantitative electron-probe analysis. The pitchblende has a median

$\text{ThO}_2$  content of 2.0 percent with a range from 1.1 to 2.9 percent (fig. 8A) and a median  $\text{UO}_2/\text{ThO}_2$  ratio of 33 with a range from 25 to 69 (fig. 9). Analysis of variance shows that this population is significantly lower in its  $\text{ThO}_2$  content at the 0.001 level than are all the other uraninite and pitchblende phases in the Witwatersrand and Dominion Reef. There is significant correlation between uranium and calcium (0.01 level), uranium and cerium (0.01 level), and  $\text{PbO}$  and  $\text{ThO}_2$  (0.001 level). However, there is no correlation between calcium and manganese or between iron and manganese as for the allogeic uraninite, pointing to a distinct origin for the pitchblende in the granular carbon, which probably developed *in situ* during diagenesis and metamorphism.

The carbonaceous seam of the Carbon Leader usually occurs along the basal parting in quartz-pebble conglomerate, and has a well-developed columnar structure normal to the bedding. It is linked with, but different from, the granular form described. It consists of the same two carbon phases, occurring as lenticular to ovoid bodies with their long axes normal to the bedding. Individual bodies are cut by discontinuous vertical sutures, which emphasize the overall columnar effect (fig. 12). Scanning electron-probe studies demonstrate that these sutures carry fine-grained pitchblende, brannerite, gold, and other phases (fig. 13).

The inclusions within the columnar carbonaceous material are similar mineralogically to those in the granular material, with one notable exception, namely the occurrence of coarse-grained uraninite, often surrounded and partly replaced by coarse-grained carbonaceous matter (fig. 14). This uraninite is concentrated along one margin of the carbon seam though not restricted to that margin. Analyses of neighbouring fragments show that they belong to groups within which the  $\text{ThO}_2$  content is similar but may differ very considerably from that in adjacent groups, indicating a chemically heterogeneous assemblage of uraninite crystals subsequently fragmented and replaced by carbonaceous matter (fig. 15). The chemistry of the allogeic uraninite grains has been previously described and is shown in figures 8 and 9. Pitchblende, associated with partially replaced allogeic uraninite has a much higher thorium content than the pitchblende in the granular carbonaceous matter; it has a median  $\text{ThO}_2$  content of 6.2 percent with a range from 3.17 to 6.9 percent (fig. 8B) and a median  $\text{UO}_2/\text{ThO}_2$  ratio of 9.0 with a range from 6.2 to 12.2. This population is significantly different from the coexisting coarse-grained allogeic uraninite (0.001 level) but not from the uraninite in the Dominion Reef. It is possible that the high thorium content is attributable to solution and redeposition of both uranium and thorium from the allogeic uraninite undergoing attack by carbonaceous matter.

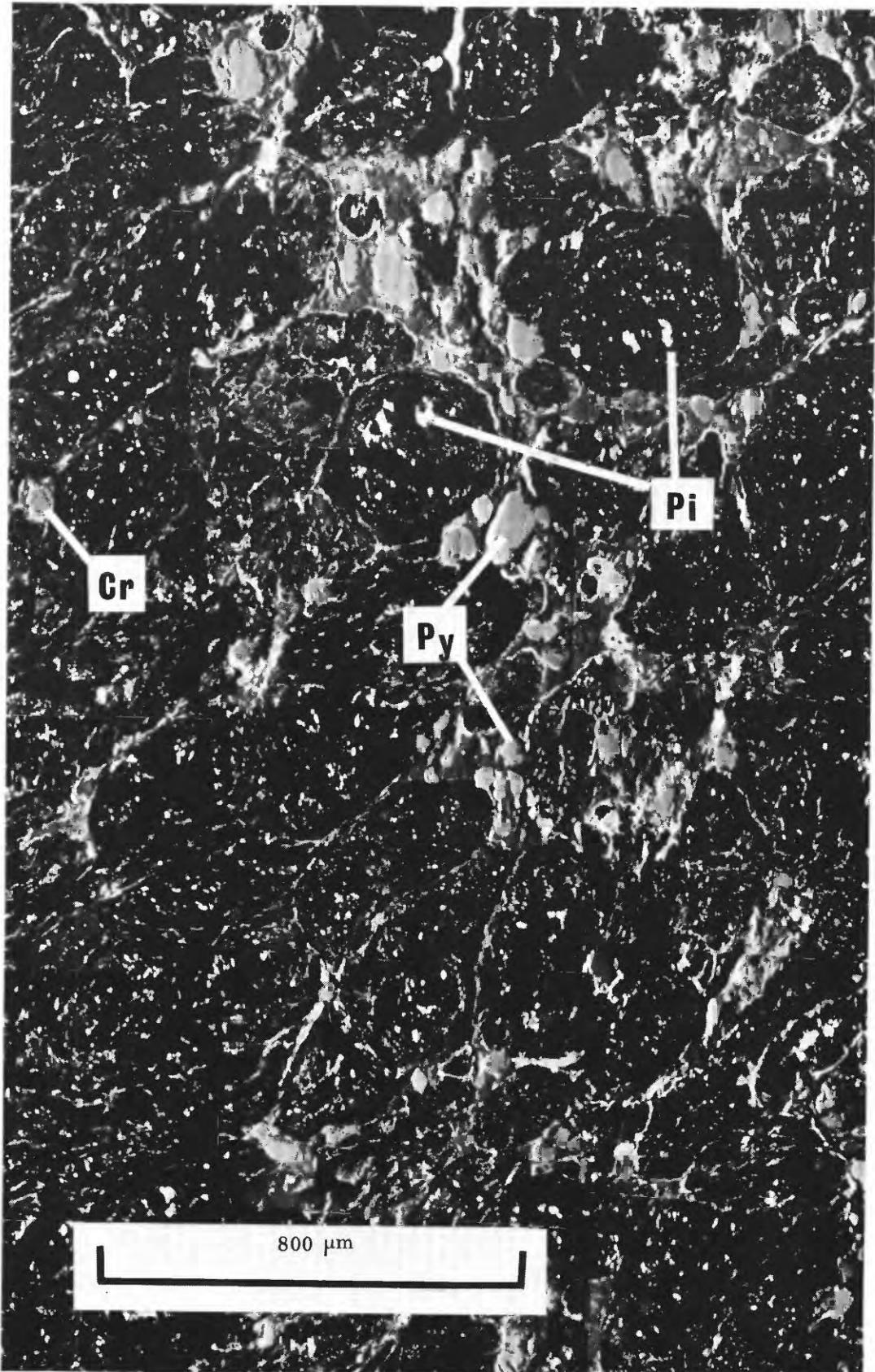


FIGURE 11.—Electron-probe backscattered electron image of granular carbonaceous material containing fine-grained particles of pitchblende (Pi) and gold in a carbonaceous matrix (black). Allogenic grains of pyrite (Py) and chromite (Cr) occur interstitially to the granular carbon, but allogenic uraninite was not detected. Carbon Leader, West Wits (US 5)

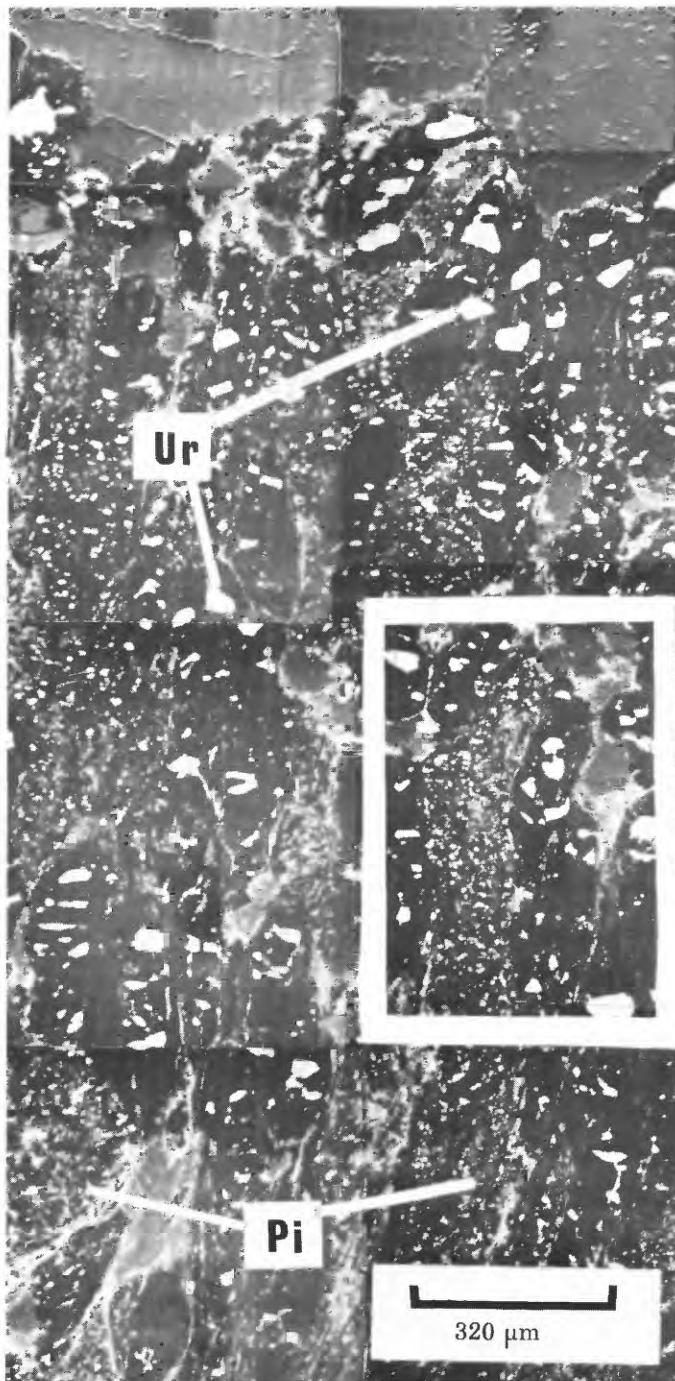


FIGURE 12.—Electron-probe backscattered electron image of columnar carbon showing zones of coarse fragmented allogenic uraninite (Ur) and fine-grained disseminated pitchblende (Pi) in a carbonaceous matrix (black). Vertical lineations of a strongly reflecting phase between these zones consist of fine disseminations of gold associated with brannerite, pitchblende, and churchite. Vaal Reefs, Klerksdorp (PTS 2034).

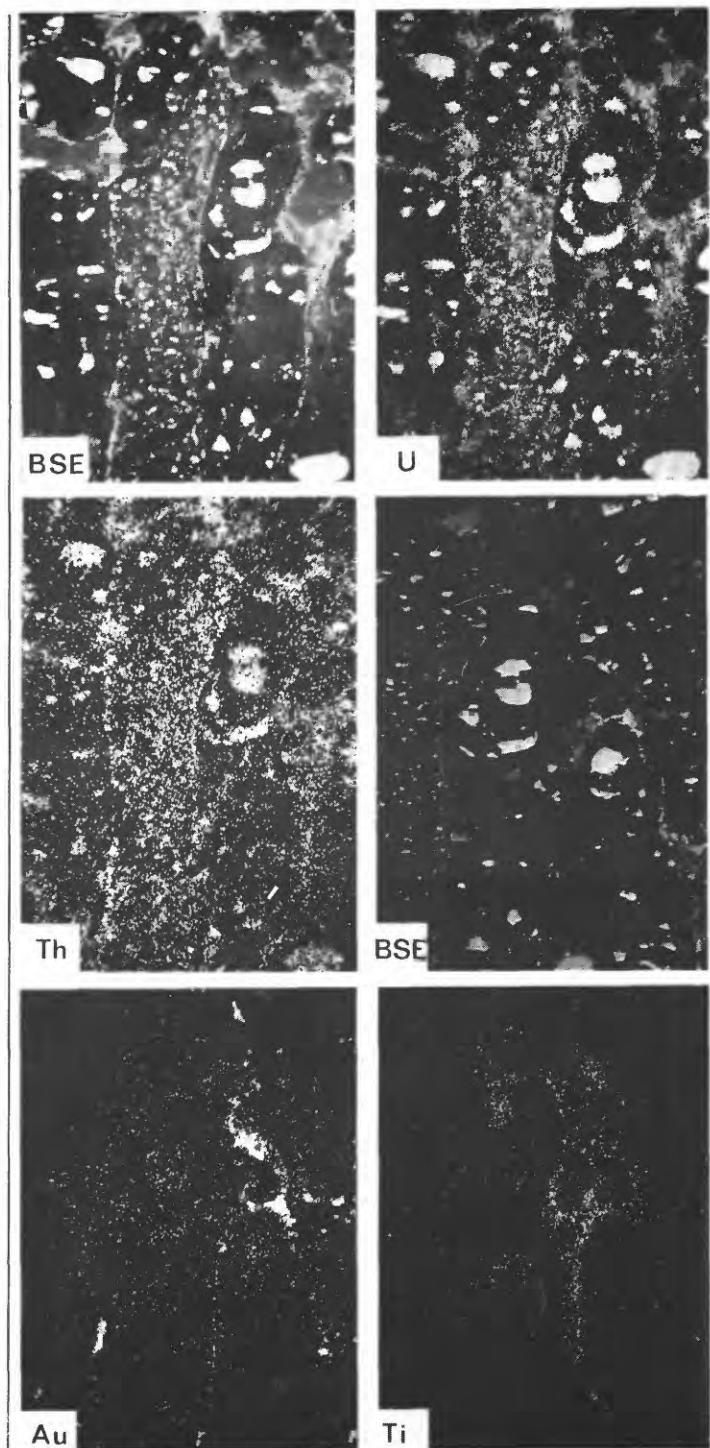


FIGURE 13.—Electron-probe scanning images from two overlapping areas of columnar carbon within inset on figure 12 showing backscattered electron images and distribution of uranium and thorium corresponding to coarse-grained uraninite and fine-grained pitchblende. The latter is intimately associated with gold and a titaniferous phase, probably brannerite, in a carbonaceous matrix (black). Vaal Reefs, Klerksdorp (PTS 2034).

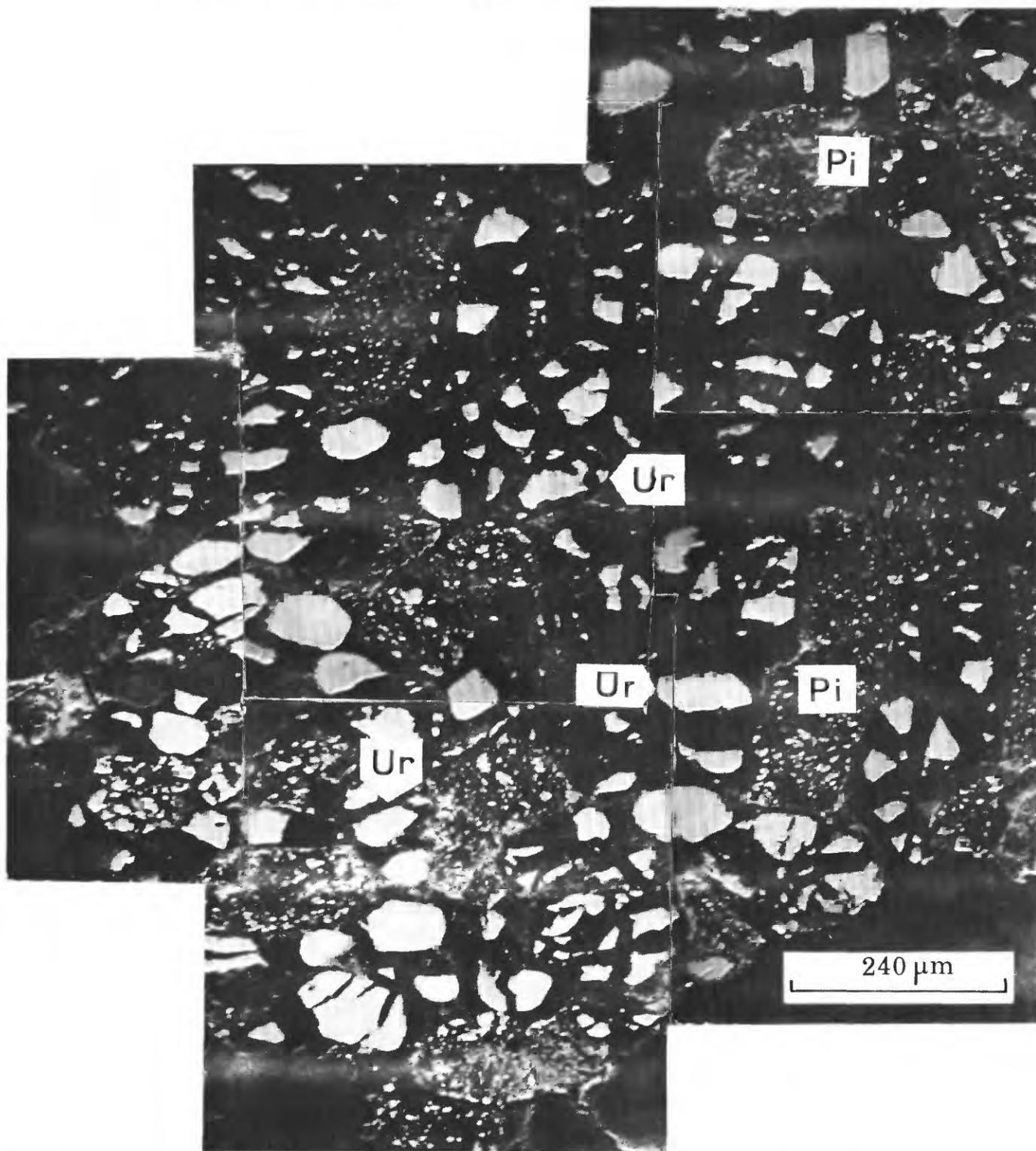


FIGURE 14.—Electron-probe backscattered electron image of columnar carbonaceous material showing clusters of coarse-grained allogenetic uraninite crystals (Ur) partly digested by carbonaceous matter (black) with lobate zones of fine-grained pitchblende (Pi) in a carbonaceous matrix. Vaal Reefs, Klerksdorp (PTS 2031).

The disseminated pitchblende in fine-grained columnar carbonaceous matter is optically and texturally very similar to that in the granular variety and is also intimately associated with galena, gersdorffite, gold, and pyrite, forming an interconnecting dendritic texture of microcrystals in the manner previously described.

The outer margin of the carbonaceous matter, where it is in contact with allogenetic pyrite, is a favoured site for gold concentration in the form of fine platelets. Internal pore spaces within carbonaceous matter are also an important site for the formation of complex intergrowths of uraniferous silicates, gold, and gersdorffite. Partially replaced uraninite within carbonaceous mate-

rial is also a common site for gold overgrowths. All these textures suggest that much of the gold, pitchblende, churchite (see next section) and associated sulphides crystallized in their present position as a result of diagenetic or later metamorphic processes.

#### CHURCHITE

A tenuous phase surrounds the fine-grained pitchblende of the columnar carbon (fig. 16). It contains major yttrium and phosphorous with the heavy rare-earth elements gadolinium, terbium, dysprosium, erbium, and ytterbium, corresponding to either xenotime ( $YPO_4$  or

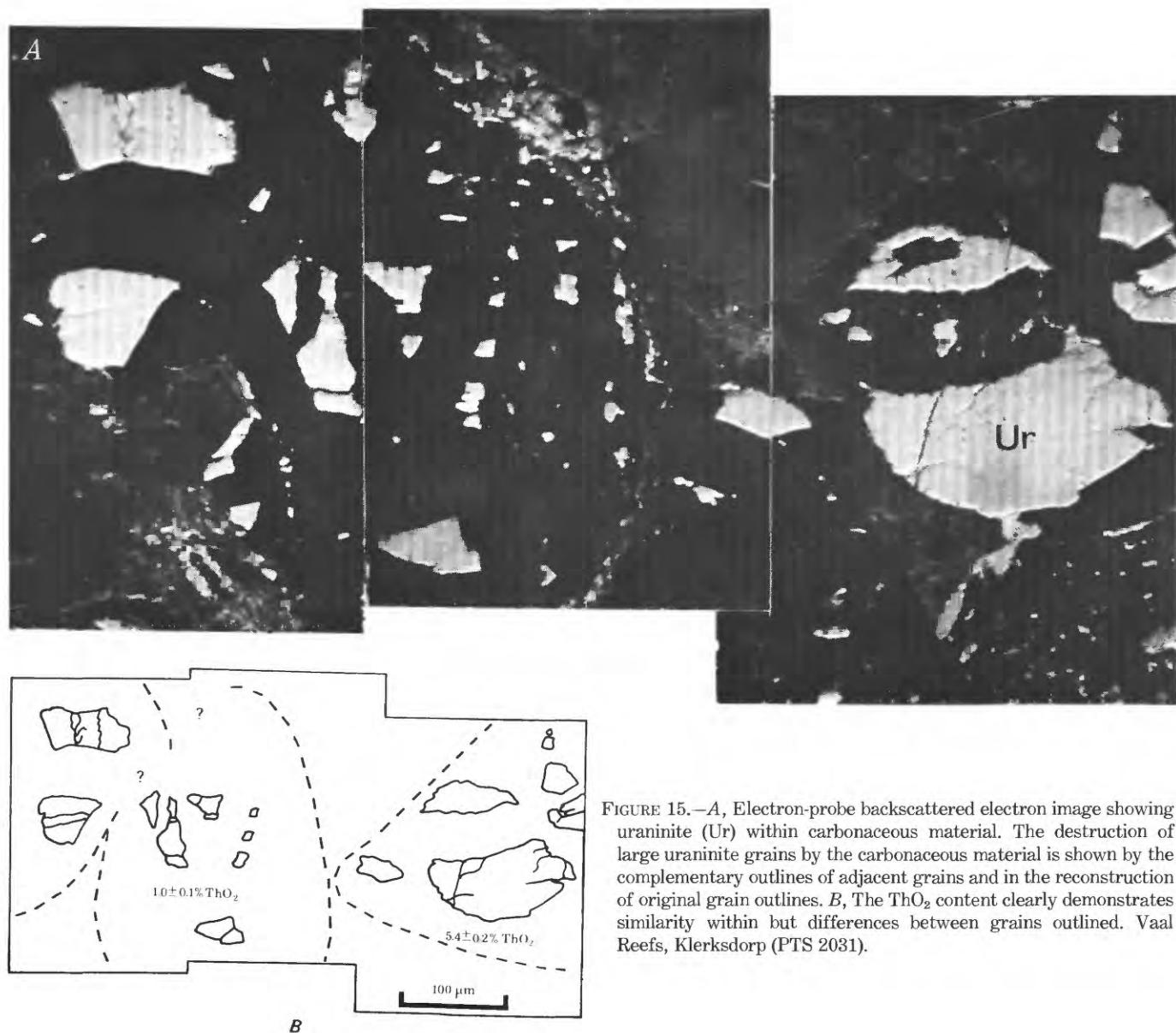


FIGURE 15.—A, Electron-probe backscattered electron image showing uraninite (Ur) within carbonaceous material. The destruction of large uraninite grains by the carbonaceous material is shown by the complementary outlines of adjacent grains and in the reconstruction of original grain outlines. B, The ThO<sub>2</sub> content clearly demonstrates similarity within but differences between grains outlined. Vaal Reefs, Klerksdorp (PTS 2031).

churchite ( $\text{YPO}_4 \cdot 2\text{H}_2\text{O}$ ). Comparison of the rare-earth distribution (table 1) with that reported in the literature for xenotime and churchite points to an identity with the latter because of the distinct enrichment of the former in dysprosium, erbium, and, particularly, ytterbium relative to gadolinium.

This is particularly interesting in the present context, since xenotime is found principally in granitic pegmatites whereas churchite<sup>2</sup> can be formed by a combination of biochemical processes and diagenetic reactions close to the surface (Milton and others, 1944). In the occurrence described by these authors, spherulites of churchite are formed as a coating on siliceous limonite or black manganese oxide. They suggest that the heavy rare earths were concentrated biochemically in the leaves of certain deciduous trees. Stability of the forest floor permitted concentration in the humus layers, from which the churchite coating on existing iron and manganese oxides was derived diagenetically. It seems possible, therefore, that in the Witwatersrand some combination of biochemical and diagenetic processes may have led to the formation of churchite around the fine pitchblende grains in the columnar carbon.

#### ORIGIN OF THE CARBONACEOUS MATERIAL

Several authors have reported carbonaceous material devoid of mineralization, so neither uranium nor gold performs an essential role in its formation. The columnar structures are developed normal to the bedding, as in stromatolites, and it is unlikely therefore that they are metamorphic structures as suggested by Liebenberg (1955). Hallbauer and van Warmelo (1974) later proposed that the structures are a primary feature of the carbonaceous matter. They advanced a theory, further elaborated in Hallbauer and others (1975) and Hallbauer (1975), that the columnar carbonaceous material is the fossilized residue of a Precambrian symbiotic association (designated *Thycomyces lichenoids*) consisting of an algal partner and a fungal organism and occupying areas between a few square centimetres and a few

square metres. Treatment of the carbon revealed a skeletal framework of uranium and thorium oxides and of gold, and slow oxidation of the carbon at 500°C left a heavy-metal fabric, which appears to correspond to the structures described in the present study. Since these metals are most probably deposited in pore spaces in the carbonaceous matter, it is unlikely that they represent the actual skeleton of the "plant" or are a replacement of any fibrous structure as Hallbauer (1975) suggests. However, the observation that several fine-grained phases containing heavy metals, notably lead, uranium, thorium, gold and r.e.e., combined with arsenic or sulphur, are closely associated with possible stromatolitic algal mats or related structures may have genetic significance for the origin of mineralization in the Witwatersrand, which is discussed later.

#### INDUS RIVER URANINITE

Samples of uraninite from the Indus River were analysed by electron probe for comparison with the allo-genic uraninite of the Dominion Reef and Witwatersrand Systems and with a view to assessing the stability of uraninite as a detrital mineral in recent alluvial deposits. The samples had been collected from the Hunza River, a tributary of the Indus River in Kashmir (Darnley, 1962), and from the Indus alluvium near Hazro, Pakistan (Miller, 1963).

The Hunza sample consists of a 99-percent pure concentrate in which nearly all the uraninite grains have at least one well-defined crystal face and some show cubic form with little evidence of corrosion. They are in the very fine to fine range of sand size (0.05–0.20 mm) (fig. 17). Pyrite is present in the concentrate as euhedral cubes, fresh and untarnished, and as concretions. An extensive assemblage of radioactive resistate minerals and sulphides has also been reported (Darnley, 1962).

The Hazro samples, which Miller (1963) had reported to contain both uraninite and gold, were also re-examined, involving the preparation of concentrates by vanner, superpanner, and heavy liquids. The final concentrate contained—in addition to uraninite—pyrite, arsenopyrite, uranothorite, and a grain of gold. The

<sup>2</sup> Described as weinschenkite by Henrich (1935), Milton and others, (1944), and Pokrovskiy and others, (1965) but shown to be churchite by Claringbull and Hey (1953).

TABLE 1.—Comparison of heavy rare-earth distribution in xenotime, churchite, and yttrium phosphate phase in Witwatersrand columnar carbon relative to gadolinium

	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu
Yttrium phosphate phase in carbon <sup>1</sup>	1	0.49	1.42	—	1.19	—	0.88	—
Xenotime <sup>2</sup>	1	.53	5.57	0.72	5.41	1.26	13.03	1.07
Churchite <sup>3</sup>	1	—	1	.72	1	trace	trace	—
Churchite <sup>4</sup>	1	—	.93	.16	.53	—	.60	.10

<sup>1</sup> Ratio of  $L\alpha$  X-ray peaks from present study.

<sup>2</sup> From an analysis by Jefford (1962, table 2, no. 1), who quotes similar analyses for other specimens.

<sup>3</sup> Ratio of  $L\alpha$  X-ray peaks on Bavarian churchite by Henrich (1935).

<sup>4</sup> From the average of two analyses by Pokrovskiy and others (1965, table 2).

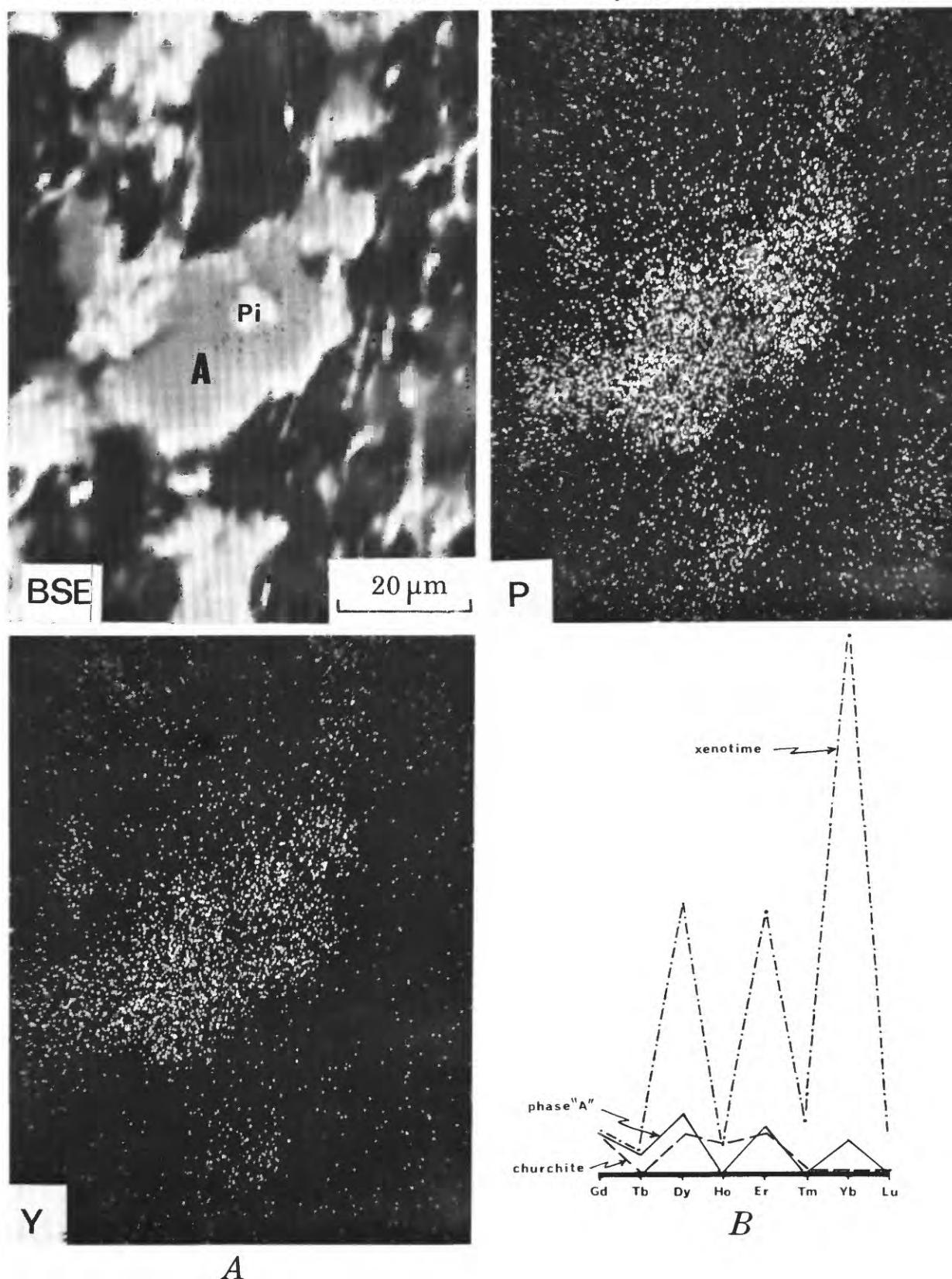


FIGURE 16.—A, Electron probe scanning images of phase "A", probably churchite, containing major yttrium and phosphorus, which overgrows pitchblende (Pi); the example shown occurs in fibrous material within columnar carbon shown in figure 12. B, The distribution of heavy rare-earth elements in phase "A" relative to gadolinium is compared here with examples of churchite and xenotime from the literature. Vaal Reefs, Klerksdorp (PTS 2023).

sulphides were fresh, but many of the uraninite crystals, with at least one crystal face, had a distinct outer weathered zone, though beneath this they were fresh and homogenous.

The  $\text{ThO}_2$  content of one sample from the Hunza and two from Hazro are shown in figure 8C. The three populations exhibit no significant difference, indicating constancy in uraninite composition between sites 300 km apart. The mean  $\text{UO}_2/\text{ThO}_2$  ratio for the Indus samples is shown in figure 9, to which has been added the ratio for the Dominion Reef uraninite adjusted for uranium and thorium decay to eliminate the effect of the greater age. There is no significant difference between the two uraninite populations after adjustment.

Viljoen (1963) regarded the lack of oxidation of detrital pyrite in the Witwatersrand System as evidence of an atmosphere free of oxygen, and Liebenberg (1955) and Grandstaff (1975) considered a nonoxidizing atmosphere was necessary for uraninite transportation. Neither of these conclusions is supported by the evidence from the present-day Indus River, in which, as

we have seen, uraninite, gold, pyrite, and other sulphides form a stable assemblage over several hundred kilometres.

## GEOCHEMICAL EVIDENCE

### LEAD CONTENT OF URANINITE AND PITCHBLEND

Cumulative frequency curves for the lead content of selected uraninite and pitchblende phases are shown in figure 18. The data fall on four main distributions as follows: (1) relatively young (approximately 90 million years) low-lead allogenetic uraninite from the Indus, (2) pitchblende in granular carbon from the Carbon Leader in the Witwatersrand with intermediate lead content, (3) allogenetic uraninite in the Witwatersrand and Dominion Reef with high lead content, and (4) pitchblende with high lead content intimately associated with allogenetic uraninite in the Witwatersrand.

The intermediate lead content of the pitchblende sample US 5 is consistent with the textural observation that this phase is younger than the allogenetic uraninite and is

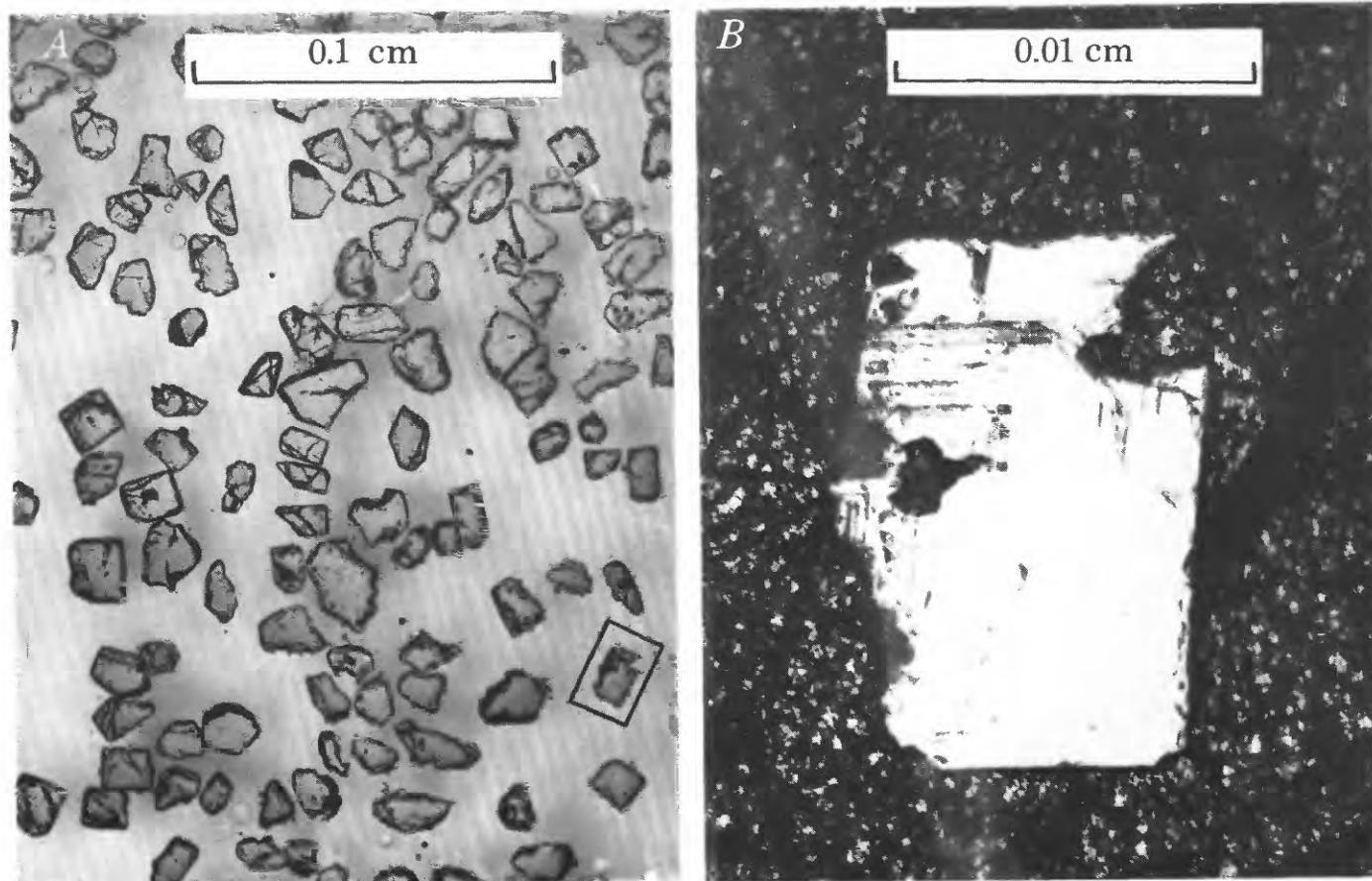


FIGURE 17.—Indus River uraninite. *A*, Uraninite grains from the Hunza River, Kashmir, a tributary of the Indus River; the uraninite grains usually show at least one crystalline face and several cubes are present. *B*, The grain shown in detail exhibits alteration along discrete zones parallel to well-developed crystal faces. Most of the grains in the sample, however, are fresh and unaltered. No correlation between degree of alteration and  $\text{ThO}_2$  content has been detected.

## URANIUM MICROMAPPING WITH THERMAL NEUTRONS

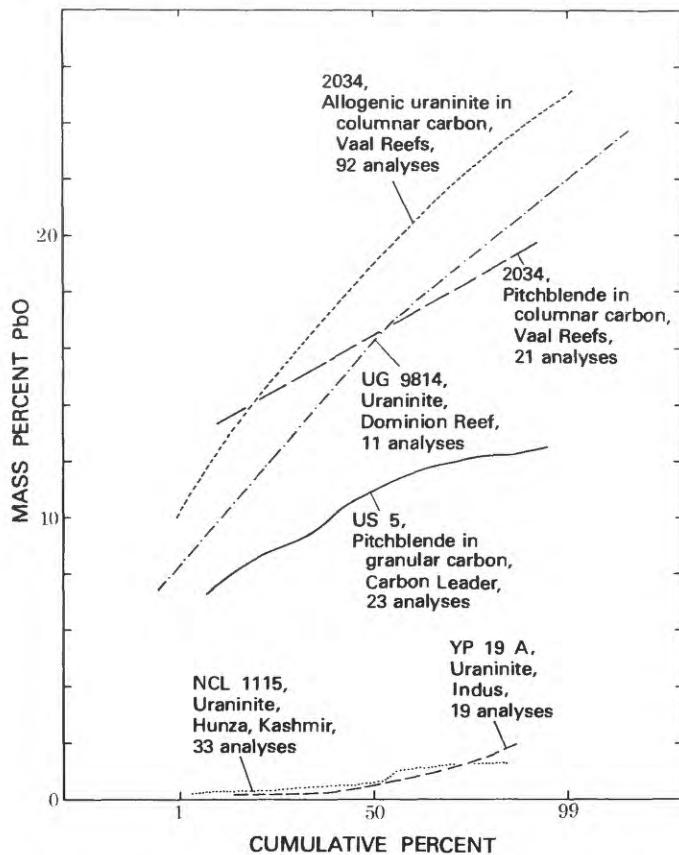


FIGURE 18.—Cumulative frequency diagram of the PbO content of uraninite and pitchblende from Dominion Reef, Witwatersrand, and Indus River sediments.

formed in situ from the diagenesis and metamorphism of the carbonaceous host. The low thorium content of this phase is indicative of a low-temperature solution (thorium being relatively insoluble), from which uranium was precipitated by absorption on decaying organic matter represented by the carbonaceous material in the section. The highest lead contents are given by the allogenic thorian uraninite phases, which are probably a reflection of their greater age. Partial rejuvenation of the allogenic uraninite from the Witwatersrand resulted in the formation of a lead-rich pitchblende in sample 2034. However, the evidence from sample US 5 indicates that the presence of allogenic uraninite is not essential for the formation of younger pitchblende, since it may also form directly from the metamorphism of uranium-enriched carbonaceous material. Younger ages exhibited by pitchblende in the absence of allogenic uraninite should not be generally interpreted therefore as rejuvenation of an older allogenic uraninite, since in the example studied here, pitchblende represents direct formation of this phase from a metal-enriched organic substrate; allogenic uraninite was not detected.

The distribution of uranium in Witwatersrand and Dominion Reef samples was studied by the fission-track method using Lexan polycarbonate for the registration of fission fragments from the induced fission of  $^{235}\text{U}$  by thermal neutrons, as described by Price and Walker (1963), Kleeman and Lovering (1967), and Bowie and others (1973). The method is ideal for providing information on the location of uranium in basket-type occurrences. The radioactive bands of interest can usually be included within the breadth of a single polished thin section and radiation doses can be adjusted so as to span concentration ranges. The samples studied here have been subjected to a dose of approximately  $5 \times 10^{16}\text{n cm}^{-2}$  in order to obtain a lower detection limit of 0.01 ppm (parts per million) and track saturation at about 40 ppm. This enables associations of uranium with mineralogical phases to be readily determined when the uranium is below the limits of detection by microprobe.

The results show that uranium is closely confined to discrete horizons both in the basket-type and carbonaceous reefs and that it not only occurs as grains of uraninite but also in a finely disseminated form. Thus uranium is present in pyrite concretions; within lenticular fragments comprising clay-size quartz, kaolinite, and pyrophyllite; as a fine disseminated coating on allogenic pyrite grains otherwise uranium free; and, rarely, within dislocations and shears (figs. 5, 6, 19).

Both microprobe and fission-track studies show the presence of uranium in the interstitial 7-Å clay phase in the pyrite concretions (fig. 7A) but fail to detect it in the pyrite itself. A combination of these techniques shows that the uranium content of individual concretions is variable within a single reef over a distance of a few millimetres. Whereas some are virtually uranium free, others are enriched in amounts readily detected by electron microprobe ( $>500$  ppm). Uranium enrichment occurs only in the rims of some concretions, whereas in others, particularly the finer grained melnikovitic types, the enrichment usually occurs throughout the concretion. This association of uranium with pyrite is particularly striking where it occurs surrounded by an essentially barren quartzitic groundmass, and in cases such as this the evidence for a primary association of uranium with the concretion is strong and probably related to deposition of uranium during the formation of the concretion. The uranium in the rims of concretions was probably absorbed during transport or diagenetically prior to lithification.

Allogenic pyrite is also closely associated with uranium, but the uranium in the pyrite is below detection limit, and in this case the uranium occurs in a secondary clay mineral forming a marginal overgrowth to the pyrite. This association is particularly well developed in

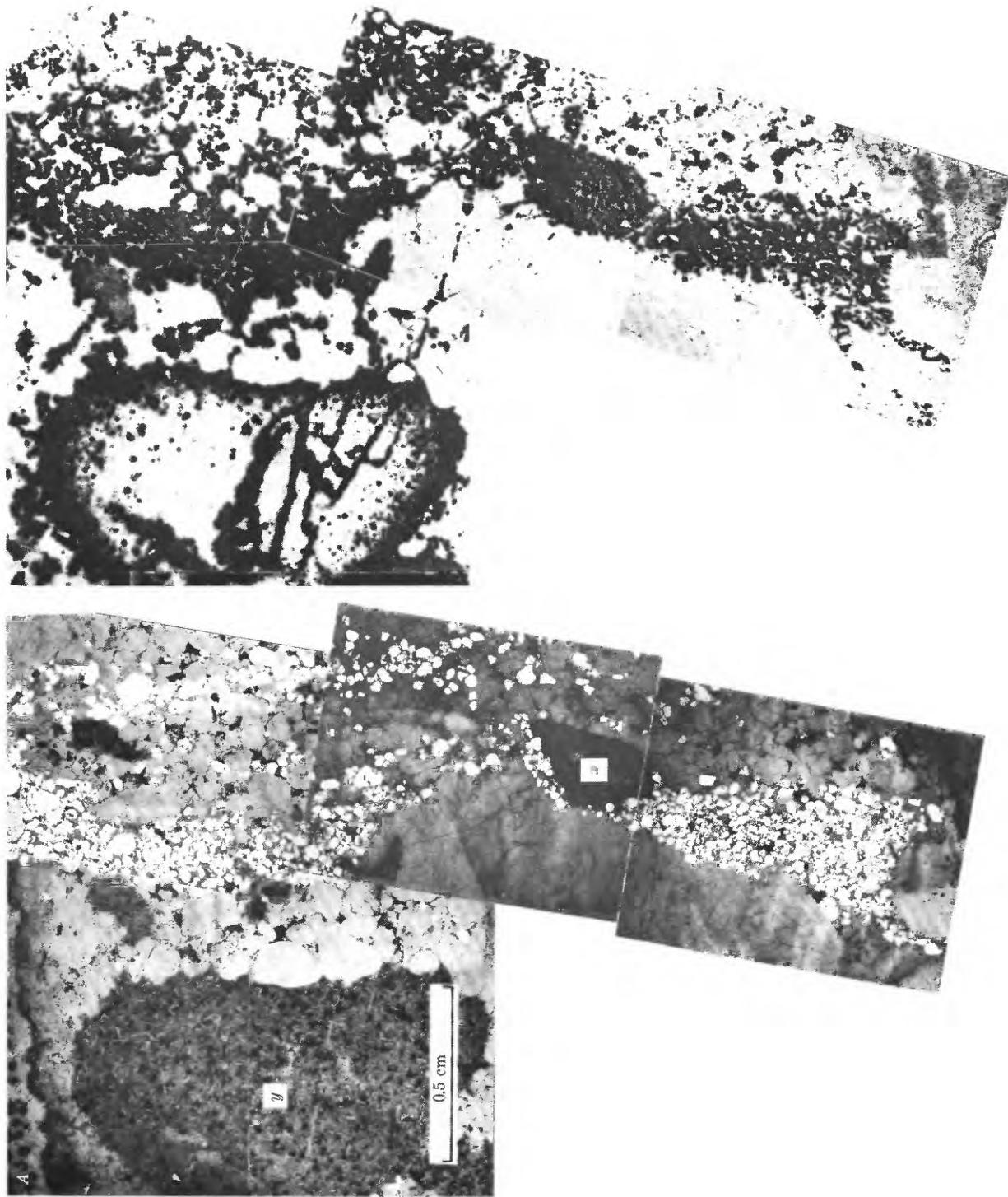


FIGURE 19.—A uraniferous pyrite band consisting principally of allogenetic pyrite with composite lenticular grains of quartz, kaolinites and pyrophyllite ( $x$ ), and quartz, pyrophyllite, and kyanite ( $y$ ). Uranium enrichment is recorded in grain  $x$ , in the rim and vermicular veinlets in grain  $y$ , and as a fine dispersion associated with chlorite interstitially to allogenetic pyrite grains (see Lexan print). Rare grains of allogenetic uraninite are also present. A, Reflected light. B, Corresponding Lexan print Basal Reef, President Brand, Orange Free State (PTS 842).

allogenic pyrite occurring in close proximity to a carbonaceous band rich in uranium and gold (PTS 1944, West Driefontein). In this case the uranium was probably deposited *in situ* and subsequently incorporated into the clay mineral overgrowth, which formed during diagenesis and metamorphism. This relationship between allogenic pyrite and overgrowths of fine-grained clay minerals and uranium has been recorded widely and is an important control in the deposition of uranium in the Witwatersrand System. The control is specific since uraniferous silicates overgrowing allogenic pyrite occur dispersed through a barren quartzite matrix and are readily identifiable and distinguishable from the textures produced by shearing and later introduction of uranium.

The association of uranium with lenticular to rounded granules consisting of quartz, kaolinite, and pyrophyllite has been recognized in many of the Witwatersrand banket-type ores studied here. The uranium occurs disseminated throughout the granules and produces point sources of fission tracks, which have not yet been correlated with any specific uranium mineral (fig. 19). In some specimens this is the only uranium-bearing mode. The occurrence of these enriched granules in a barren groundmass indicates that uranium enrichment most probably occurred either before deposition or at the latest during diagenesis. Larger rounded pebbles also occur, some consisting of quartz, pyrophyllite, kyanite, and possible illite with secondary spherulitic structures.

In this case uranium occurs on what appears to be a leached and weathered margin and along a vermicular veinlet structure within the pebble. Enrichment in uranium during transport and diagenesis related to weathering of the pebble rim (fig. 19) seems to be the most likely explanation of this occurrence. Regional metamorphism has tended to reduce the thickness of the uraniferous rim, which is best developed where weathered pebbles are relatively unaltered.

#### NEUTRON-ACTIVATION ANALYSIS OF WHOLE-ROCK SAMPLES

Instrumental neutron-activation analysis of whole-rock samples was undertaken as an adjunct to the mineralogical studies utilizing a method developed by Plant and others (1976).

The results are given in table 2 and demonstrate the increase in U/Th ratios from the Dominion to the Vaal Reefs in the Witwatersrand Basin. Freddies Reef, East Geduld, and Daggafontein are somewhat higher and similar to one another, whereas samples from West Driefontein, dominated by carbonaceous matter, have the highest ratios. Twenty-four additional elements obtained by this method on the same sample are presented

for comparison with uranium and thorium. This gradational variation in the U/Th ratio is an approximate measure of the depositional environment. The high-energy environment has a low ratio, and the low-energy environment a high ratio, with continuous variation between the two.

#### BIMODAL NATURE OF THE URANIUM MINERALIZATION

The occurrence of allogenic grains of uraninite in the Dominion Reef (Taylor and others, 1962) and the Witwatersrand (Liebenberg, 1955; Grandstaff, 1975) is well established. This phase occurs in the Dominion Reef in association with resistate phases such as garnet, monazite, cassiterite, zircon, allogenic sulphides, rare authigenic sulphides, and gold. The similarity of this assemblage with modern gravels on banks in the Indus is notable, although uraninite in the Indus is at present subeconomic (Miller, 1963). In the Witwatersrand System the characteristically lower content of allogenic uraninite and rounded resistate phase, the relatively greater importance of concretionary pyrite, and the appearance of carbonaceous matter as an important accessory or even dominant phase represent a markedly different style of mineralization.

The general similarity of the median values of the  $\text{UO}_2/\text{ThO}_2$  ratios between 14 and 20 for allogenic uraninite from the Indus Valley, Dominion Reef, and Witwatersrand is most significant. However, the pitch-blende associated with granular carbonaceous material in the Witwatersrand has a much higher median ratio of about 32, and this is a distinct and separate phase of mineralization differing from the allogenic uraninite texturally, compositionally, and in its close association with carbonaceous matter and gold.

The fission track studies have clearly indicated an association of uranium in Witwatersrand sediment with clay minerals occurring individually or intergrown with, or overgrowing, pyrite, and in many cases this association has the appearance of a primary, essentially syngenetic, relationship unaffected by appreciable uranium remobilization.

Evidence of this type from Witwatersrand sediments provides support for the existence of some uranium in solution in the basin during sedimentation and diagenesis, as discussed by Pretorius (1975). The uranium would be highly mobile as the soluble uranyl ion,  $\text{UO}_2^{2+}$ , which would subsequently be absorbed, reduced, and precipitated by organic-rich muds probably aided by and associated with the simultaneous reduction of sulphate by bacteria, as proposed by Trudinger (1971):



The hydrogen sulphide formed would subsequently react with other metals in solution such as iron and lead to form the authigenic intergrowths of pyrite and galena with clay minerals, gold, and uranium observed in the sediments.

The most likely modes of introduction of uranium, gold, and sulphate in solution are the complex fault system of the Witwatersrand Basin and dissolution of coexisting detrital minerals. Some of the sulphate would have been precipitated in close proximity to hot springs at the margin of the basin, but some gold and uranium would have remained in solution, perhaps with the aid of metal-organic complexes (Vine and others, 1958; Ling Ong and Swanson, 1974; Boyle and others, 1975). The gold and uranium in solution would have been carried out into the basin to be precipitated together on the distal portions of the alluvial fans by the action of decaying organic matter.

This process accounts for the observed primary distribution of uranium and gold, other than the purely detrital material, and explains the separation of uranium and thorium in the progression from the higher energy Dominion Reef to the lower energy Witwatersrand environment. Though relatively unimportant in the higher energy blanket type of mineralization, deposition from solution through the agency of organic matter would seem to be responsible for major economic concentrations of both uranium and gold over large areas in the Witwatersrand Basin in the lower energy Carbon Leader type of mineralization.

### CONCLUSIONS

The overall control of the uranium mineralization in the Witwatersrand and Dominion Reef Systems is the presence of a depositional basin overlying an Archaean craton in which very early differentiation on the model proposed by Fyfe (1970, 1973) and amplified by Moorbat (1975) enriched the upper crust in oxyphile metals including uranium. This process has been considered by Bowie (1970) as a fundamental control in the delineation of metallogenic provinces and hence in the distribution on a global scale of economic concentrations of uranium.

In this basin, uranium was deposited in two ways, as detrital alloigenic uraninite and by precipitation from solution. The detrital components were concentrated in the higher energy conglomerate environment of the Dominion Reef and proximal parts of the Witwatersrand Basin. The solution components including uranium and gold were precipitated under reducing conditions through the agency of decaying organic matter in the lower energy or distal regions of the basin, giving rise through diagenesis and metamorphism to such uranif-

erous horizons as the Carbon Leader of the West Wits, which are not directly related to known palaeogeographical channels. It can now be clearly demonstrated that relatively small amounts of thorium increase the resistance to attrition and oxidation to the extent that a thorian uraninite with 1 percent or more thorium oxide can survive as river detritus under present-day atmospheric conditions. All of the uraninite grains from the Witwatersrand Basin considered to be of detrital origin contain 1 percent thorium oxide or more, hence it is unnecessary to postulate a reducing atmosphere; indeed, the retention of sulphate and uranyl ions in solution would seem to require the existence of an oxidizing atmosphere in the model proposed here for the deposition of uranium from solution.

This conclusion therefore suggests that the occurrence of pebble conglomerates with detrital uraninite and pitchblende deposited from solution may have a more widespread occurrence throughout geological time than hitherto suspected, provided the necessary geological criteria for their formation are met.

### ACKNOWLEDGMENTS

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TABLE 2.—*Neutron-activation analysis, in parts per million, except where stated otherwise*

Sample location	Rock sample	Na	K	Sc	Cr	Fe <sup>1</sup>	Co	Zn	As	Rb	Ag	Cd	Sb	Cs
Dominion Reefs	5A: 2006/2133	1,219	<2,000	38	<400	10.2	570	<300	3,800	<100	<40	<100	<28	<5
	BR8N2(B): 2004/2134	<200	6,600	24	249	7.31	260	<300	340	<200	<35	<100	<6	<5
	BR6N4(A): 2018/2135	<200	<2,000	30	374	4.96	425	<300	3,100	<200	<40	<100	<10	<6
	BR7N1(B): 1998/2136	<1,000	<2,000	32	297	7.63	420	<300	3,600	<200	<40	<100	19	<10
Vaal Reefs	UR21A: 2055/2121	<800	<2,000	10	1,400	6.56	260	344	605	<100	<20	<100	25	<5
	UR3: 2034/2122	<1,000	<2,000	13	450	2.80	216	<350	1,200	<200	<40	<100	53	<6
	UR5: 2039/2123	980	<2,000	7	890	4.89	229	<160	670	<200	<40	<100	27	7
	UR16: 2049/2124	990	<2,000	8	315	1.65	120	<160	370	<100	<30	<100	21	5
Freddies Reef	FW4: 2105/2137	52	<2,000	4.2	850	1.77	108	440	430	<100	<15	<100	6	<5
	FW2: 2103/2138	66	<2,000	4.6	760	3.17	123	354	620	<80	22	<100	10	<5
East Geduld	EG4: 2064/2139	330	5,900	6.7	2,300	5.09	200	366	640	<100	<15	<100	15	<5
Daggafontein	EG18: 2078/2140	190	930	4.6	560	3.63	117	295	140	<100	<40	<100	<5	<5
	26: 1995/2128	94	<2,000	6	152	6.76	312	239	336	<100	24	<100	<5	<3
	27: 1996/2129	256	<2,000	18	1,200	7.67	219	<800	296	<100	<40	<100	<5	<3
	KE4/7: 1975/2130	220	<2,000	3	115	.21	21	220	29	<50	<15	<100	<5	<3
	K6F12/11: 1979/2131	86	<2,000	8	244	1.19	43	818	134	<100	<15	<100	<5	<3
	KG6/10: 1978/2132	520	2,200	11	650	2.79	55	<100	120	<70	<15	<100	6	<3
West Driefontein	8A: 1959/2125	387	<2,000	7.5	1,040	4.98	140	360	450	<100	46	<100	9	<3
	4B: 1950/2126	204	<2,000	4	235	4.34	59	264	46	<50	39	<100	<5	<3
	5: 1951/2127	<400	<2,000	4	84	2.38	14	463	60	<150	130	<100	7	<3

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TABLE 2.—*Neutron-activation analysis—Continued*

Sample location	Rock sample	Ba	La	Ce	Nd	Sm	Eu	Tb	Lu	Hf	Ta	Au	Th	U	U/Th ratio
Dominion Reefs -----	5A:														
	2006/2133	< 500	1.3	1,200	1,000	445	< 2	20	14	< 10	410	3	809	3,900	4.8
	BR8N2(B):														
	2004/2134	< 1,000	906	1,200	850	285	12	16	10	15	240	9	620	2,300	3.7
	BR6N4(A):														
	2018/2135	< 1,000	1,400	2,200	910	335	12	17	11	19	450	2	753	2,300	3.1
	BR7N1(B):														
	1998/2136	< 1,000	1,100	1,800	1,270	600	20	15	18	18	450	5	970	5,400	5.6
	UR21A:														
	2055/2121	< 1,000	260	700	1,500	450	20	8	13	22	8	154	477	4,500	9.4
Vaal Reefs -----	UR3:														
	2034/2122	< 1,000	475	900	2,800	850	37	24	22	30	< 100	218	821	8,400	10.2
	UR5:														
	2039/2123	< 1,000	248	350	860	200	17	6	6	36	7	69	298	2,100	7.1
	UR16:														
Freddies Reef -----	2049/2124	< 1,000	172	500	940	250	15	6	8	11	< 5	214	273	2,700	9.9
	FW4:														
	2105/2137	< 500	59	260	170	60	4.7	< 5	< 3	18	30	90	52	685	13.2
	FW2:														
East Geduld -----	2103/2138	< 500	68	135	195	65	5.3	< 5	< 3	< 20	30	115	80	765	9.6
	EG4:														
	2064/2139	< 500	41	< 200	280	< 10	3.6	5	< 3	17	6	118	40	510	12.8
Daggafontein -----	EG18:														
	2078/2140	< 500	57	80	73	12	1.2	2	< 3	11	4	1.5	20	75	3.8
	26:														
	1995/2128	< 1,000	44	< 100	227	75	1.5	< 2	< 1	4	15	173	71	895	12.6
	27:														
West Driefontein -----	1996/2129	< 1,000	42	< 10	400	160	8	3	< 3	8	20	132	102	2,000	19.6
	KE4/7:														
	1975/2130	< 1,000	20	22	9	6	1	< 2	< 1	< 2	< 5	.4	< 5	43	---
	K6F12/11:														
	1979/2131	< 1,000	16	45	112	30	1.5	< 2	< 1	2	2	28	10	337	---
	KG6/10:														
	1978/2132	< 300	22	55	43	20	2	< 2	< 1	2	30	11	15	180	12.0
	8A:														
	1959/2125	< 1,000	47	< 300	88	40	3	< 2	< 5	19	50	428	40	460	11.5
	4B:														
5:	1950/2126	< 1,000	25	< 10	340	100	6	3	< 3	7	< 5	221	75	1,400	18.7
	1951/2127	< 1,000	69	< 30	454	140	7	16	4	< 5	< 5	444	99	1,500	15.2

<sup>1</sup> Percent.

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# Detrital Uraninite and Pyrite: Are They Evidence for a Reducing Atmosphere?

By P. R. SIMPSON and J. F. W. BOWLES

GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN  
QUARTZ-PEBBLE CONGLOMERATES

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## GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN QUARTZ-PEBBLE CONGLOMERATES

### DETritAL URANINITE AND PYRITE: ARE THEY EVIDENCE FOR A REDUCING ATMOSPHERE?

By P. R. SIMPSON and J. F. W. BOWLES<sup>1</sup>

#### ABSTRACT

Many occurrences of uraniferous quartz-pebble conglomerates from widely separated regions are known to date from the period 2.8–2.0 billion years ago. The deposits of the Witwatersrand System are among the most important of this type, and the occurrence in them of allogenetic uraninite and pyrite has been widely used as evidence that the atmosphere during that period of the Earth's history was reducing or anoxicogenic.

Comparison of modern alluvial sediments of the Indus River with those of the Dominion Reef and Witwatersrand Systems indicates stable assemblages of detrital thorian uraninite and pyrite of similar grain size and composition. Authigenic pitchblende and pyrite provide indirect evidence of uranyl and sulphate ions coexisting with detrital thorian uraninite and pyrite during the formation of Witwatersrand sediments. It is suggested that this bimodal form of transport and deposition can be most readily accounted for by a model in which the sulphate and uranyl ions exist in superficial, oxidised, active sediments and river waters draining into the Witwatersrand Basin while detrital grains of uraninite were contained within the bedload of streams in reducing conditions below the sediment/water interface. The persistence of uraninite during burial and diagenesis would be favored by catastrophic burial of the sediments in a subsiding basin. This would prevent oxidation of the abundant organic matter, now represented by carbonaceous material, to give carbonate and bicarbonate ions, which form uranyl carbonate complexes that are highly soluble even in reducing conditions.

The occurrence of Precambrian uraninite-bearing conglomerates is therefore controlled by geological and not atmospheric conditions. It is suggested that the deposits are the erosion products of the upper layers of a primitive, layered Archaean crust containing granitic rocks enriched in thorian uraninite. The period 2.8–2.0 billion years ago is thought to represent a unique phase of crustal evolution, when extensive erosion of uranium-rich upper crust resulted in the deposition of uraniferous sediments in enclosed basins.

#### INTRODUCTION

Many occurrences of uraniferous quartz-pebble conglomerates from widely separated regions of the Earth such as Witwatersrand in Southern Africa and Elliot Lake in North America are known to date from the period 2.8–2.0 b.y. (billion years) ago. The deposits of the Witwatersrand are economically among the most important of this type. Gold and uranium are contained

in conglomerate horizons in the lower portion of the Dominion Reef system and in conglomerate and carbonaceous reefs within the upper portions of the Witwatersrand System. The uranium has two principal modes of occurrence, consisting principally of uraninite grains in conglomerate and uraninite and pitchblende in carbonaceous horizons. Fresh unoxidised pyrite is present as coarsely crystalline grains and as fine-grained concretions in conglomerate. Gold is present as grains in conglomerate and intergrown with pitchblende in carbonaceous horizons (Simpson and Bowles, 1977). Uraninite, pyrite, and gold thus occur as both allogenetic and authigenic minerals in this deposit.

According to Köppel and Saager (1974), the Witwatersrand rocks are flanked by a number of domelike small inliers of granite-greenstone basement, which form the elliptical frame defining the depositional basin of the Dominion Reef and Witwatersrand sediments. These inliers are considered to be the provenance area for allogenetic uraninite, gold, pyrite, and associated heavy minerals found in the basin, and direct connections have been established for gold and pyrite. The allogenetic uraninite is similar in age to the granites (Rundle and Snelling, 1977).

The presence of allogenetic uraninite and pyrite in the Witwatersrand sediments has been widely used as one of the principal items of geological evidence that the atmosphere during the period 2.8–2.0 b.y. ago was reducing or nonoxidising (Liebenberg, 1955; Ramdohr, 1955, 1958; Holland, 1962; Schidlowski, 1966, 1970; Schidlowski and others, 1975). Robertson (1974) considers that the occurrence of uraninite-bearing quartz-pebble conglomerates is restricted to a period in time following the development of extensive acid crust but prior to the evolution of an oxidising atmosphere. This suggestion depends heavily, however, on the belief that uraniferous conglomerates provide evidence for a reducing atmosphere in the first instance. There is no dispute as to the relative abundance of these deposits in the early Precambrian, which contains all the presently known occurrences of uraniferous conglomerate of

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economic grade. However, the new mineralogical evidence for the origins of uraninite and pyrite is not in agreement with anoxogenic or reducing atmospheric conditions 2.8–2.0 b.y. ago. Thus an alternative reason is necessary to explain the concentration of uraniferous conglomerates in this period of the Earth's history.

Uraninite-, pyrite- and gold-bearing allogenic ore-mineral assemblages are known to occur in modern sediments at several localities. For example, in the Indus River, where the concentrations are presently regarded as subeconomic (Miller, 1963; Darnley, 1962), uraninite is present in adequate quantity for study; and it can be demonstrated that it is sufficiently stable under present-day atmospheric conditions to survive exhumation, surface weathering, and transport in the river system over considerable distances (Simpson and Bowles, 1977). Allogenic uraninite is also apparently stable in other recent sediments. For example, Steacy (1953) has reported uraninite of unknown thorium content in a black sand with notable development of cube faces and brilliant lustre, and Coppens and Mayanda (1969) have also reported uraninite in beach sand derived from the erosion products of the Quiberon granite nearby. The purpose of this contribution therefore is to re-evaluate selected aspects of uraninite and pyrite distribution in Precambrian and comparable modern sediments in relation to atmospheric and hydropheric evolution.

#### EVIDENCE FROM ALLOGENIC URANINITE IN THE WITWATERSRAND AND INDUS RIVER

The close similarity in size (0.05–0.25 mm) and compositional range of allogeic uraninite from the Dominion Reef, Witwatersrand, Indus River alluvium near Hazro, Pakistan, and alluvium in the Hunza River, Kashmir, which is a tributary of the Indus, has been reported previously (Simpson and Bowles, 1977). The  $\text{ThO}_2$  content and  $\text{UO}_2/\text{ThO}_2$  ratio of uraninite from the Dominion Reef correspond closely to those for uraninite from Hazro, and the  $\text{ThO}_2$  content and  $\text{UO}_2/\text{ThO}_2$  ratio of uraninite from the Vaal Reef correspond closely to those for uraninite from the Hunza River. Calculation of the uranium and thorium content of the Precambrian uraninite to account for radiogenic decay of uranium and thorium since crystallization about 3.0 b.y. ago improves this correlation. The uranium and thorium content of the uraninite populations from the Indus and its tributaries span the full range of uraninite compositions of the Dominion Reef and Witwatersrand. It has previously been suggested that since thorianite is resistant to oxidation and exists as a detrital mineral (Frondel, 1958), the thorium content of uraninite may increase its stability as a detrital mineral (Davidson, 1960).

In order to test this suggestion and to investigate the possible role of other cations in stabilizing allogeic uraninite, 33 grains of uraninite from the Indus at Hazro were analysed on an electron microprobe using both energy-dispersive and wavelength-dispersive techniques. The microprobe analyses are presented in table 1. A correlation matrix for the electron-microprobe data is given in table 2, which indicates very significant negative correlation between  $\text{UO}_2$  and  $\text{Y}_2\text{O}_3$ ,  $\text{UO}_2$  and  $\text{Ce}_2\text{O}_3$ ,  $\text{TiO}_2$  and  $\text{ThO}_2$ ,  $\text{TiO}_2$  and  $\text{Ce}_2\text{O}_3$ . Significant positive correlations exist between  $\text{UO}_2$  and  $\text{TiO}_2$ ,  $\text{ThO}_2$  and  $\text{Ce}_2\text{O}_3$ ,  $\text{CaO}$  and  $\text{Ce}_2\text{O}_3$ ,  $\text{Y}_2\text{O}_3$  and  $\text{Fe}_2\text{O}_3$ .  $\text{PbO}$  and  $\text{MnO}$  do not correlate with any other oxides. The minor element correlations suggest that the grains represent a single population in the bedrock and are probably not derived from multiple unrelated phases of mineralisation in which correlation would be less likely. The lack of correlation of  $\text{PbO}$  is accounted for by radiogenic origin and consequent uneven distribution within uraninite grains.

Comparison of  $\text{ThO}_2$  content with size, shape, roundness, degree of alteration, and sphericity of the uraninite grains suggests that there is no correlation between the principal compositional variable,  $\text{ThO}_2$  (1.20–12.08 percent), and the sedimentary features of uraninite grains that are likely to have resulted from weathering, transportation, and diagenesis. It is not known, however, whether uraninite containing less than 1.2 percent  $\text{ThO}_2$  is either relatively soluble or is unavailable in the source regions. From the textural information, uraninite containing as little as 1.2 percent  $\text{ThO}_2$  seems as likely to survive in the Indus as uraninite containing ten times as much. It is therefore suggested that in this population the compositional variation within the range studied has no measurable effect on survival. On the other hand, uraninite grains collected as a concentrate far from their primary sources will have encountered variable degrees of weathering, which differ for each grain during several cycles of erosion and deposition during downstream progression. The variation in attrition suffered by grains during transport therefore is the main factor controlling variation in grain characteristics and is unrelated to grain composition within the compositional range studied.

#### EVIDENCE FROM THEORETICAL MODELS OF URANINITE STABILITY

Errors in the assumptions used in designing models of the natural environment are likely to lead to false estimates of uraninite stability. For example, Grandstaff (1973) has calculated that for uraninite from Blyvooruitzicht in the Witwatersrand to survive erosion, transportation, deposition, and burial, "oxygen levels must have been less than approximately  $10^{-2}$  to  $10^{-6}$

TABLE 1.—*Electron microprobe analyses, in percent, of 33 detrital uraninite grains from a sample of Indus alluvium, near Hazro, Pakistan*

[These grains were analysed by a combination of wavelength- and energy-dispersive techniques utilising correction procedures described by Mason, Frost, and Reed (1969) and to be described by Bowles in a report in preparation. The samples were collected by Miller (1963) and further described by Simpson and Bowles (1977)]

Analysis No.	TiO <sub>2</sub>	MnO	Fe <sub>2</sub> O <sub>3</sub>	PbO	ThO <sub>2</sub>	UO <sub>2</sub>	Y <sub>2</sub> O <sub>3</sub>	CaO	Ce <sub>2</sub> O <sub>3</sub>	Total
1	.28	.08	.17	.66	1.20	99.89	.10	.02	---	102.40
2	.24	.10	.12	1.25	1.27	98.40	.14	.02	---	101.54
3	.30	.04	.14	1.32	1.38	98.54	.05	.02	---	101.79
4	.26	.09	.18	.50	2.19	97.95	.13	.02	---	101.27
5	.24	.09	.18	.75	2.27	97.71	.05	.02	---	101.31
6	.25	.07	.12	1.31	2.35	96.63	.15	.02	---	100.90
7	.29	.07	.14	.29	2.78	98.44	.14	.02	.03	102.20
8	.28	.08	.09	.47	2.79	97.13	.10	.02	---	100.96
9	.24	.04	.18	1.31	2.89	96.57	.20	.02	---	101.45
10	.26	.05	.21	.35	3.16	95.35	.72	.08	.03	100.21
11	.23	.08	.21	.30	3.19	98.31	.05	.04	.03	102.44
12	.24	.04	.10	.43	3.69	96.57	.19	.24	---	101.50
13	.21	.07	.17	.60	3.82	96.89	.11	.03	---	101.90
14	.27	.06	.16	.39	3.92	96.01	.46	.05	.03	101.35
15	.21	.07	.20	1.43	3.93	93.44	.59	.04	.10	100.01
16	.22	.09	.19	1.34	4.09	91.91	1.16	.03	.04	99.07
17	.28	.08	.24	1.15	6.32	87.87	1.65	.04	.07	97.70
18	.24	.06	.17	1.19	6.43	89.80	.88	.02	.05	98.84
19	.16	.06	.12	.59	6.68	88.91	1.32	.11	.10	99.05
20	.20	.07	.28	.85	7.06	82.53	2.52	.08	.07	93.66
21	.19	.04	.17	1.20	7.11	88.78	1.39	.02	.09	98.99
22	.22	.04	.17	.24	7.24	89.14	1.65	.04	.01	98.75
23	.17	.07	.19	.52	8.24	89.97	.99	.02	.02	100.19
24	.16	.09	.18	.52	8.40	90.67	---	.40	.23	100.60
25	.12	.10	.26	1.06	8.65	86.84	1.40	.02	.08	98.53
26	.20	.07	.16	1.10	8.69	88.42	.62	.09	.05	99.40
27	.23	.07	.26	.57	8.70	88.53	.59	.03	.10	99.08
28	.20	.05	.14	1.06	9.00	84.68	1.61	.11	.14	96.99
29	.13	.10	.27	.59	9.05	85.58	1.58	.06	.13	97.49
30	.14	.10	.16	.41	9.08	88.81	.14	.31	.32	99.47
31	.19	.04	.14	.37	9.20	90.36	.05	.02	.04	100.41
32	.08	.10	.18	1.17	10.69	86.77	.05	.34	.42	99.75
33	.09	.07	.15	.59	12.08	83.75	.53	.06	.24	97.56

that of the present level in the mid-Precambrian." This conclusion is evidently incorrect since the presence of detrital uraninite of similar composition in the Indus River alluvium at the present time requires no special atmospheric conditions.

When detrital uraninite grains enter a river system they are unlikely to suffer either oxidation below the sediment/water interface, where conditions are generally reducing, or chemical attack and dissolution, provided carbonate and bicarbonate ions are lacking (Garrels and Christ, 1965). In these conditions the grain may survive for long periods and be transported over long distances if periods of transport are short relative to total residence time, thus reducing direct contact with oxidised river water.

The greatest problem in any model of detrital uraninite stability arises with the need to deposit relatively fresh and untarnished euhedral grains into a river system since the greatest exposure to oxidation is likely to occur at this point as the grains come in direct contact with the atmosphere. In rapidly exhumed, eroded, and glaciated mountainous terrain this is probably easier to achieve than in a more mature drainage system, and these conditions help to account for the presence of detrital uraninite in the Indus alluvium and probably also by analogy in the Witwatersrand. In Grandstaff's (1973) model the assumptions used to estimate the degree of exposure to oxidation at different stages in the erosion cycle probably bear little relation to the conditions in the present day Indus River basin or the Witwatersrand.

TABLE 2.—*Correlation matrix for the chemical composition of 33 detrital uraninite grains determined by electron-probe microanalysis and shown in table 1*

[The uraninite grains were obtained from a sample of Indus alluvium near Hazro, Pakistan, collected by Miller (1963) and further described by Simpson and Bowles (1977)]

	TiO <sub>2</sub>	MnO	Fe <sub>2</sub> O <sub>3</sub>	PbO	ThO <sub>2</sub>	UO <sub>2</sub>	Y <sub>2</sub> O <sub>3</sub>	CaO	Ce <sub>2</sub> O <sub>3</sub>
TiO <sub>2</sub> -----	1.000	---	---	---	---	---	---	---	---
MnO -----	-.341	1.000	---	---	---	---	---	---	---
Fe <sub>2</sub> O <sub>3</sub> -----	-.124	.190	1.000	---	---	---	---	---	---
PbO -----	.015	.043	.067	1.000	---	---	---	---	---
ThO <sub>2</sub> -----	-.805	.082	.238	-.090	1.000	---	---	---	---
UO <sub>2</sub> -----	.697	-.061	-.413	-.103	-.910	1.000	---	---	---
Y <sub>2</sub> O <sub>3</sub> -----	-.205	-.111	.586	.156	.406	-.711	1.000	---	---
CaO -----	-.439	.149	-.247	-.070	.349	-.216	-.204	1.000	---
Ce <sub>2</sub> O <sub>3</sub> -----	-.752	.374	-.007	.036	.715	-.602	.025	.691	1.000

### EVIDENCE FROM CARBONACEOUS MATTER IN THE WITWATERSRAND

Evidence has previously been given (Pretorius, 1975; Hallbauer, 1975; Simpson and Bowles, 1977) that the carbonaceous matter in the Witwatersrand represents organic plant remains (fig. 1A, B). These remains most likely represent the metamorphosed and fossilised remnants of stromatolitic algal mats (Mendelsohn, 1976).

The Carbon Leader in the Upper Witwatersrand System is an example of this type of formation covering several hundred square miles of the West Witwatersrand (Bourret, 1973). It therefore provides good evidence for the establishment of photosynthesis and generation of atmospheric oxygen in sufficient quantity to provide a shield from ultraviolet radiation, which would otherwise destroy plant life (Fyfe, 1974).

The presence of atmospheric oxygen is also supported by carbon-isotope evidence from Precambrian limestones and dolomites, from which it has been concluded that close to 80 percent of the amount of oxygen contained in the present oxygen budget should have been released prior to 3.0 b.y. ago (Schidlowski and others, 1975).

### EVIDENCE FROM AUTHIGENIC PITCHBLENDÉ AND DISSEMINATED URANIUM IN THE WITWATERSRAND SYSTEM

The existence of fine-grained low-thorium pitchblende in carbonaceous material in the Witwatersrand probably results in part from deposition of uranium in solution onto organic matter. Lower grade concentrations of uranium have also been identified, by fission track methods, in association with phyllosilicates, which are finely intergrown with authigenic pyrite and also form lenticular mudstone pellets. These occurrences are also

considered to represent deposition of uranium from solution (Simpson and Bowles, 1977). This evidence indicates that detrital uraninite in sediments upstream underwent partial oxidation to form the soluble uranyl ion, which was then reprecipitated downstream below the sediment/water interface in association with organic-rich mud, authigenic pyrite, and decaying organic remains of stromatolitic algal mats (fig. 1B). These reactions require oxidising atmospheric conditions in order to function in the manner suggested, except where the concentration of bicarbonate is high, since the uranium-carbonate complex ions can exist in aqueous solution under reducing alkaline conditions (Garrels and Christ, 1965). However, no carbonate has been described from the sediments of the Witwatersrand succession.

The evidence from the stromatolitic algal mats is indicative of a sufficient supply of atmospheric CO<sub>2</sub> to promote supratidal growth, and the reactions described in this paper assume that the atmosphere was well mixed and that water washed detrital grains into the site of deposition. The discussion on the sulphur cycle, in the next section, indicates that pH conditions in the mineralising surface water solutions were probably neutral to acidic rather than alkaline as a result of the activity of bacteria catalysing the production of relatively large amounts of sulphate from allogenic pyrite.

Under these conditions the uranyl ion and organometallic complexes were probably the principal uranium species in solution during deposition of the sediments. The lack of carbonate during diagenesis is probably attributable to rapid burial, which prevented reaction between organic-rich sediments and the coexisting oxidising atmosphere. The nature and thickness (8 km) of the Witwatersrand sediments and the preservation of large amounts of carbonaceous matter are good evidence for rapid burial.

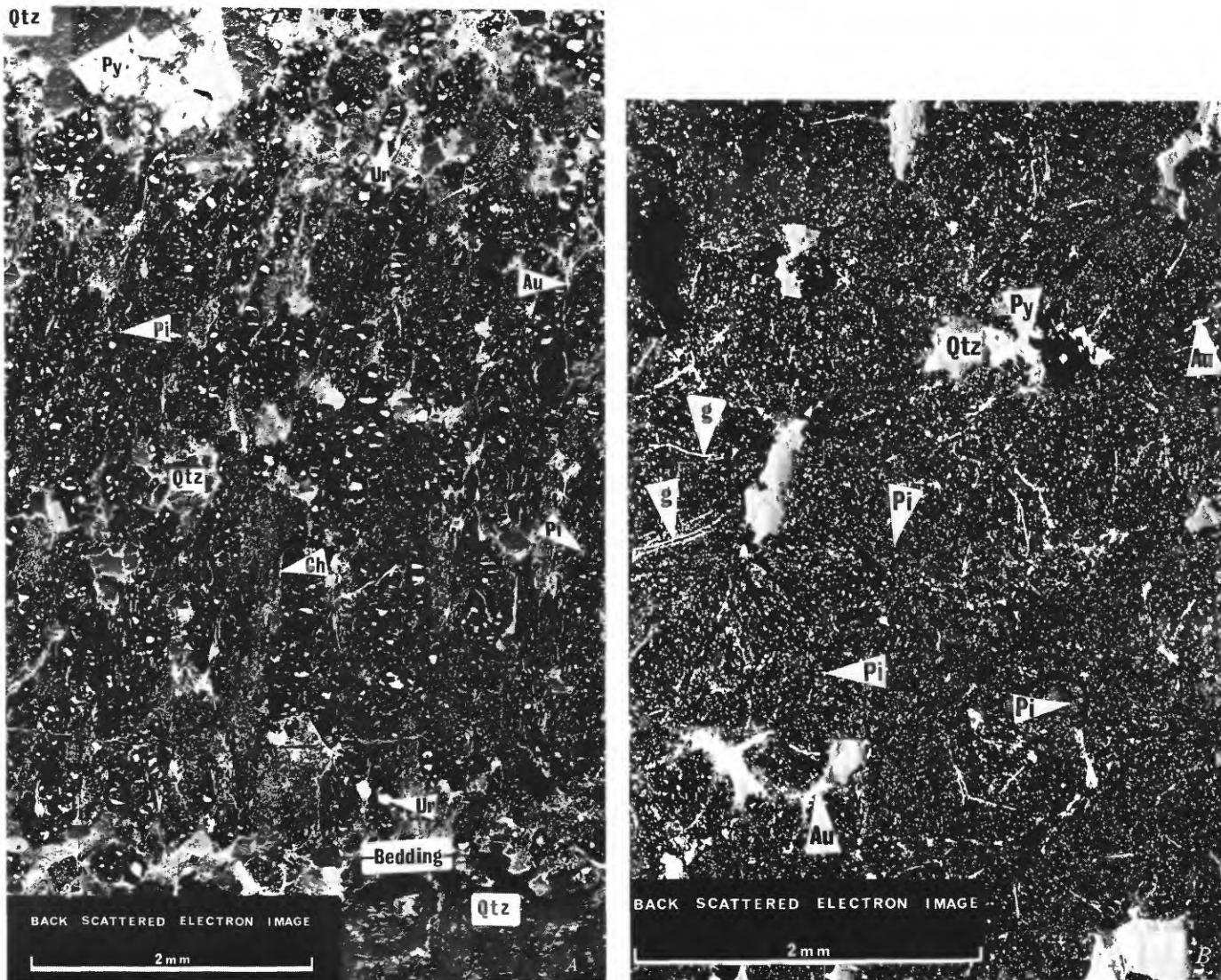


FIGURE 1. - Photographic mosaics obtained by use of computer-controlled stepping motors, showing macrostructure of specimens. Carbonaceous matter representing fossilized stromatolitic algal mats contains allogenetic uraninite associated with authigenic pitchblende, intergrown with finely divided authigenic gold. These relations are most clearly observed in the back-scattered electron image of the electron microprobe, which exploits the difference in mean atomic weight between carbonaceous matter on the one hand and gold, uraninite, and pitchblende on the other.

A, Polished thin section no. 2084, Vaal Reef. This is a section of an 8-mm thick horizon of columnar carbonaceous material (black) from a quartz-pebble conglomerate. Some quartz pebbles (Qtz) in the conglomerate layers at the upper and lower margins of the figure contain euhedral pyrite (Py). Detrital grains of uraninite (Ur) and quartz (Qtz) are overgrown or entrapped by the mat (black). Uranium and gold are also deposited diagenetically as pitchblende (Pi) and authigenic gold (Au) and together with churchite (Ch) these authigenic phases outline the vertical columnar structure of the carbonaceous matter (black) (Simpson and Bowles, 1977).

B, Polished thin section no. 1952, West Driefontein. Massive carbonaceous matter in thin section (black) contains uniformly dispersed fine-grained authigenic pitchblende (Pi) and coarser grained authigenic gold (Au), which are often intergrown, with minor amounts of much coarser allogenetic quartz (Qtz) and pyrite (Py). No heavy-metal detrital phases are present; hence the low-thorium pitchblende and gold represent deposition of uranium and gold from solution onto stromatolitic algal mats (black), followed by recrystallisation during diagenesis and metamorphism. These processes have also remobilised lead of probably radiogenic origin into cracks where it is present as galena (g).

## EVIDENCE FROM THE SULPHUR CYCLE IN THE WITWATERSRAND

The existence of abundant authigenic pyritic concretions coexisting with allogenic pyrite grains (Simpson and Bowles, 1977, figs. 6, 7) indicates that reactions of the following type have probably occurred during formation of the sediments:

1. Partial oxidation of detrital allogenic pyrite to generate sulphate in solution
2. Deposition of authigenic pyrite concretions and overgrowths in reducing environments below the sediment/water interface, a reaction that takes place in unconsolidated silts and muds now represented by interstitial silicate phases in the concretions
3. Reworking of these sediments and downstream transport of secondary authigenic pyrite washed out of the unconsolidated muddy sediment and unoxidised allogenic pyrite, which were then redeposited together.

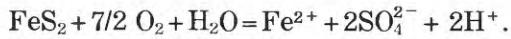
All three stages were repeated in whole or in part until final burial and diagenesis in subsiding portions of the basin, where pyrite and uraninite were maintained in a reducing environment below the sediment/water interface and out of contact with the oxidising atmosphere. It is important to note here that fresh authigenic and allogenic pyrite are also identified in samples from the Indus River by Simpson and Bowles (1977).

The reactions described are probably due to combined inorganic and bacterial surface oxidation of pyrite to form sulphate, associated with bacterial reduction of sulphate below the sediment/water interface. These processes are summarised in more detail in the following sections.

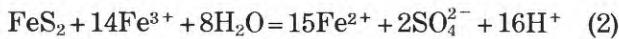
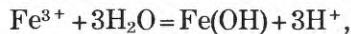
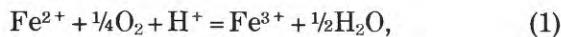
### INORGANIC SURFACE OXIDATION OF PYRITE

Inorganic oxidation of pyrite under surface conditions in equilibrium with atmospheric oxygen has been described by Stumm and Morgan (1970) and can be summarised as follows:

Initial step:



Rate controlling step:



The slowest reaction or rate controlling step is the oxidation of  $\text{Fe}^{2+}$ ; the fastest reaction is the oxidation of pyrite by  $\text{Fe}^{3+}$  ion. Therefore, once the reaction is

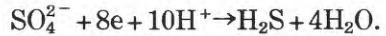
started the oxidation of pyrite cycles through reactions (1) and (2).

### BACTERIAL SURFACE OXIDATION OF PYRITE

At the present time pyrite oxidation rates are greatly accelerated by the activities of acidophilic iron- and sulphur-oxidising bacteria (Silverman, 1972). Several bacteria may be involved, but the most common and most effective are *Ferrobacillus ferrooxidans*, which oxidises pyrite by direct attack, and *Thiobacillus ferrooxidans*, which are thought to catalyse the oxidation of  $\text{Fe}^{2+}$  to  $\text{Fe}^{3+}$  in the rate controlling reaction (1). Microbial activity by these and other bacteria is predominant only under ideal conditions of low pH, a temperature of about  $30^\circ\text{C}$ , and the presence of oxygen, iron, sulphur, and nutrients ( $\text{N}, \text{CO}_2$ ), according to Hoag and Webber (1976).

### BACTERIAL REDUCTION OF SULPHATE BELOW THE SEDIMENT/WATER INTERFACE

Bacteria also take part in reducing reactions, especially those belonging to the genera *Desulfovibrio* and *Desulfotomaculum*; they are anaerobic and reduce sulphate to  $\text{H}_2\text{S}$  as follows:



Iron in solution would form pyrite by reaction with  $\text{H}_2\text{S}$  (Trudinger, 1971). The abundant authigenic pyrite concretions from the Witwatersrand were presumably formed as the result of bacterial activity of this type in organic-rich muddy sediments (Saager, 1970).

In summary the evidence from the sulphur cycle shows that the occurrence of pyrite in the Witwatersrand is more readily explained by assuming oxidising rather than reducing atmospheric conditions during deposition, probably in association with active bacterial processes aiding dissolution and reprecipitation of pyrite.

### SUMMARY

The evidence considered in this study indicates that the terrestrial atmosphere at the time of formation of Witwatersrand sediments was not detectably different from that of the present day. The reasons for the earlier suggestion that the atmosphere must be reducing or nonoxidising to account for the presence of uraninite and pyrite in the Witwatersrand are probably due to overestimation of the extent of atmospheric exposure of these phases. Uraninite and pyrite stabilities and their environmental significance must be considered in relation to their geological context if the deductions made

are to be realistic. The coexisting phases and style of mineralisation in both ancient and modern river systems are best explained by oxidising atmospheric conditions and reducing conditions below the sediment/water interface.

Comparison of modern alluvial sediments of the Indus River with those of the Dominion Reef and Witwatersrand Systems has shown that stable assemblages of detrital thorian uraninite and pyrite of similar grain size and composition can be found in both ancient and modern sediments. Authigenic minerals in the Witwatersrand provide indirect evidence of uranyl and sulphate ions coexisting with detrital thorian uraninite and pyrite during formation of the Witwatersrand sediments. It is suggested that this bimodal form of transport and desposition can be most readily accounted for by a model in which sulphate and uranyl ions exist in superficial, oxidised, active sediments and river waters draining into the Witwatersrand Basin while detrital grains of uraninite were contained within the bedload of streams in reducing conditions below the sediment/water interface. The persistence of uraninite during burial and diagenesis would be favoured by catastrophic burial of the sediments in a subsiding basin. This would prevent oxidation of the abundant organic matter, now represented by the carbonaceous material, to give carbonate and bicarbonate ions, which form uranyl carbonate complexes and which are highly soluble even in reducing conditions.

## CONCLUSIONS

The great preponderance of economic uraniferous conglomerates in the time span 2.8–2.0 b.y. ago is controlled by geological conditions and not by atmospheric factors. This close association between uraniferous conglomerate and Precambrian crust has previously been pointed out by Bowie (1970), who considers this to be a significant factor and guide for prospecting.

According to Moorbath (1975, 1976), continental growth occurred on a major scale during the early Precambrian. Juvenile plutonic material crystallised slowly at depth and differentiated to form a compositionally layered lower crust of the type described by Fyfe (1970, 1973), Brown and Fyfe (1970), Heier (1973a, b), and Smith (1976), in which granitic components and incompatible elements migrate upwards to form calc-alkaline plutonic rocks, leaving behind a thick residue of depleted granulite facies rocks at greater depth. Highly complex gradation crustal layering results. The period 2.8–2.0 b.y. ago therefore represents a unique phase of crustal evolution, when extensive erosion of uranium-rich upper crust resulted in the deposition of uraniferous sediments in enclosed basins, under atmospheric conditions similar to those of the present day.

This conclusion is compatible with the findings of other studies, notably those of Dimroth and Kimberley (1976). From physical considerations, Shimizu (1976) concluded that an initial highly reducing atmosphere on the Earth's surface would be highly unstable. Hence from physical and geological considerations the evidence indicates that the atmosphere in the period 2.8–2.0 b.y. ago was oxidising.

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#### DISCUSSION FOLLOWING PAPERS BY DRS. FEATHER, SIMS, AND SIMPSON AND BOWLES

*Whiteside*: May I just make a comment on not these two last papers but the one previous to that by Mr. Feather. He mentioned that the uraninite and brannerite were probably about 1,800 million years old. Now, the Witwatersrand itself has been dated between 2,300 and 2,800, around about 2,500, which means that there has been presumably some metamorphic event which has been stamped on these uraninites. I believe that 1,800 million years is the best age for the Bushveld Complex. Could there be any connection, as a heat source especially?

*Feather*: I wanted actually to point that out in the course of my talk. That I had in fact noted that the Bushveld Igneous Complex was of the same age.

There is just something that I would like to comment on from Peter Simpson's paper. I am very interested in the uranium which is distributed in minerals other than the uraninite and brannerite and zircon and so on. We ran into a strange situation in analysis for uranium before X-ray fluorescence was used and that was the chemical dissolution of uranium in the samples using nitric acid. We found that using a sample containing about 400 parts per million we were leaving behind about 10 parts per million after leaching with hot nitric acid for some time. In a search on concentrates which I derived from the residue material I had great difficulty finding the uranium. Now you have possibly pointed to the areas where the uranium actually did exist, but of course the nitric acid would have dissolved certain things such as the pyrite. Are you suggesting that the uranium occurs actually in the pyrite in those concretionary pyrite nodules, or are you stating that the uraninite occurs in some interstitial phase or in the holes in those concretionary bodies?

*Simpson*: I think you've put your finger right on it there. It's very difficult to demonstrate this on a slide because the resolution isn't good enough, but if you

would like to have a look at the sections under the microscope and compare the Lexans directly with the sections, I think you will see for yourself that the uranium is not in the pyrite. It is related to the interstitial matrix, which I'll have to confess I haven't put on the probe yet and haven't yet identified, but it looks similar to the phyllosilicates in that lenticular body which I described and out of which I got pyrophyllite, kaolinite, and quartz. They are very small, and there is an exact correlation of the fission tracks in the Lexan with the interstitial phase and not with the pyrite. It isn't in the pyrite phase, although the spherical grains are in the pyrite.

*Saager:* I think I would agree that the uranium is in that interstitial phase. During our lead-isotopic measurements we did some uranium determinations. I do not agree with your idea that there is a lot of uranium around; the amounts we did find were between 15 and 70 ppm uranium in the buckshot pyrite. I think this is a very small amount of uranium.

*Simpson:* The sections shown there have very few detrital grains in them. They were picked out to work on because they were radioactive; we couldn't understand why they were radioactive because we couldn't see any detrital grains. I think you'll find that in toto there is a fair amount of uranium. There is a lot more uranium in the phyllosilicates than in the buckshot pyrite, actually, and that phyllosilicate phase seems to be a great deal richer. You must remember that the uranium in the pyrite which is porous is diluted by nonuraniferous pyrite, and the actual point where the uranium occurs is very much richer. If you've got an overall value of 70 ppm, that's for the pyrites and phyllosilicate combined, and if you recognize that the phyllosilicates may be only 20 percent of the total buckshot pyrite, you have got to multiply by a factor of five to get the actual concentration at that point, haven't you? Because the pyrite itself is barren. So it's going to be 350 ppm for the phyllosilicate.

*Saager:* That's, of course, taking the higher value. I think more of our values were closer to 15 or even 20.

*Simpson:* The Lexan track registration is saturated at 40 ppm, and all the tracks I showed you, where uranium does occur, are well saturated. So the uranium content would be 40 ppm or more for the conditions of radiation employed for those particular sections. I have to admit, however, that we haven't done this quantitatively. We've only done it qualitatively up to this stage, but we could make much more detailed quantitative assessments if this was worthwhile doing. Or you could do it if you wanted to.

*Skinner:* I have two questions, Peter [Simpson]. First of all, would you hazard a guess, or if you have a measurement would you give us your estimate of how

much of the uranium in the Witwatersrand is in these phyllosilicates?

*Simpson:* I wouldn't like to be quantitative about this, but I would say that a great deal more of it is there than anybody has yet ever mentioned or possibly even appreciated.

*Skinner:* Are you talking about 90 percent of it?

*Simpson:* No. I think a great deal of it is in the carbonaceous material. There is a great deal of it in discrete detrital phases. Just hazarding a guess, it might be as high as 30 percent, but I haven't done it quantitatively yet.

*Skinner:* Okay. My second question arises from an observation I made from the slides that you showed us. All of your pyrites are shattered, broken by brittle fracture. I noticed the same things in Dr. Schidlowski's excellent slides. I wonder if this is a uniform feature of the Witwatersrand. Pyrite is a strong mineral, as you know, and requires quite a stress before it's going to break. If the pyrite is all shattered throughout the Witwatersrand, then it has undergone considerably more deformation than I would have thought from Dr. Hallbauer's discussion of the carbon this morning.

*Simpson:* I would agree with that. In general, my experience is that pyrite is shattered in many, many cases. I don't know the exact structural locations of the samples I am working on, and it's very difficult to interpret that type of information other than to make the observation that, yes, in many cases the pyrite is shattered.

*Skinner:* Would one of the others know?

*Saager:* I think that the shattering is actually a factor of the size of the pyrite; very small, compact detrital pyrites are definitely not shattered. The stuff we have been working on for the lead-isotope study we specifically collected only pyrite that was not shattered at all. These were very small detrital compact pyrites, so I think it actually depends upon the size of the pyrite.

*Feather:* Peter, two questions: What is the possibility that the uranium in the phyllosilicates may actually be present as very tiny needles of brannerite? Have you been able to examine them in the scanning electron-microscope?

*Simpson:* No, we haven't examined them by any instrumental method other than by this method. The trouble arises that when you put a thing in a reactor, you are then very reluctant to put it in your probe for danger of contamination of the probe, so you have to make up one section to go in the probe, one section to go in the reactor. We still are at an early stage in our work, and we haven't yet probed the sections corresponding to those that have been in the reactor. So I

don't have that information. Optically, I cannot see any opaque phases in the phyllosilicate-uranium association. There is nothing in the rock which is opaque. There is nothing there to lead one to suspect, optically or mineralogically, that there is any reason for uranium to be there. But this is something that we have come across over and over again in our uranium-prospecting activities in Britain, where time and time again geologists have sent in samples which they claim to be radioactive and there are no visible uranium minerals to be seen, no secondaries even. That is why we adopted this technique as a routine procedure because it does at least show you where the uranium is at low levels when it is associated with phases you don't recognize as uranium minerals. For example, hematite or fine-grained limonite can contain up to a thousand ppm uranium without showing any visible signs and that that is the case. Having learned that, we now use this technique routinely. So I don't see any brannerite, but you are welcome to look at the slides and examine them yourself if you wish.

*Feather:* I do find your observations very interesting. If there was uranium in solution and if you assume that it was in solution at the time that the reefs were laid down, it doesn't seem unlikely that by an ion-exchange reaction clay minerals could have absorbed uranium. Another question that I would like to ask you is whether it is not possible that uranium was absorbed by these minerals, not at the time of deposition, but during some subsequent metamorphic epoch when there was a lot of uranium redistributed in the reefs in the form of brannerite, because brannerite occurs not only in the way that we have seen as ghosts in the original site of the uraninite grain, but it often migrates considerable distances away from where the parent obviously was.

*Simpson:* Well, this is a very good question, and it's a point that obviously one bore in mind in studying these rocks. The impression you get is that the uranium, in this loosely bound form, is very closely confined to the site of original deposition of phyllosilicates and pyrite. It doesn't deviate even 1 or 2 mm away from these very fine layers of pyrite, and one would suspect that if it had been introduced later at least some of the uranium would have found its way out of these layers. The fact that it hasn't done so, of course, isn't absolutely conclusive. I mean, it is possible, I suppose, that these, for some reason or another we don't understand, could have just been the pathways along which uranium came. What I would encourage you to do is to look at these things for yourself and find out to what extent you can find evidence either way. The time of introduction of the uranium is a bit of an open question at the present, but because it's confined in these very narrow layers I am inclined to suspect it's a

primary feature. And it's not surprising, after all, because this is exactly what happens in stream sediments today.

There is one other point I should mention. If this uranium was introduced later, this would require oxidizing solutions, no question about that. If the solutions were oxidizing then you would expect to see some effect on the pyrite. You would expect the pyrite to be oxidized. The pyrite is not oxidized; it is not affected in any way at all by the presence of this uranium. It seems to be quite happy there, and therefore I have concluded on balance that this uranium was fixed at or around the time of deposition of the phases with which it is associated.

*Feather:* Well, I might differ with you there, but I don't think we should go into it now.

*Roscoe:* Have you tried these same techniques with Dominion Reef material?

*Simpson:* Unfortunately, we haven't got around to that yet, and we don't have any appreciable quantity of samples. If you would like to send us a few samples to try it on \* \* \*.

*Roscoe:* Well, I have no Dominion Reef samples.

*Simpson:* Sorry, did you say Dominion? Oh, I beg your pardon. I mistook the question. I immediately assumed you were talking about Canada. Dominion Reef samples. Yes, we have tried this with Dominion Reef samples, and we have found that the track distribution corresponds to the sites of detrital minerals.

*Roscoe:* Well, of course the Elliot Lake ores are more like the Dominion Reef type and the bulk of the uranium is in discrete heavy grains. I have never seen an alpha track plate of any Elliot Lake ore that didn't have a great deal of uranium in matrix material in the fine-grain muscovitic matrix, which usually has smeared out leucoxene in it, too. There always are tracks in there. Your procedure, of course, is so sensitive that you do swamp any uranium-bearing materials so you are exaggerating the amount in the matrix in this case. I think you may see the same type of thing in the Dominion Reef and Elliot Lake ores, maybe not to the same extent.

*Schidlowksi:* I principally agree with most of your points. I am just a little bit suspicious about your secondary uraninite, those tiny specks associated with gold. I am thinking of possible criteria to narrow these down. My question goes rather to Dave [Grandstaff] than to you. Dave, if this was secondary uraninite, shouldn't the thorium tenor be considerably lower?

*Grandstaff:* Yes, I think that this low-temperature aqueous uraninite should have low thorium values. I think this would be a good distinguishing criterion to distinguish the detrital from the replacement secondary uraninite. The uraninite should be probed before it can be stated conclusively which it is.

*Simpson:* I agree.

*Grandstaff:* The second point I'd like to make is on reaction kinetics of pyrite. The reaction kinetics of pyrite are not known at this point, but you can't necessarily assume that conditions under which uraninite would break down, would be oxidized and dissolved, would necessarily cause appreciable oxidation and dissolution of pyrite.

*Simpson:* There has been very little said about oxidizing solutions in these rocks at all up to now, but there does seem to be some evidence for them. The point is, if there was going to be something to be seen in the rock, I would expect to see some effect on the pyrite. That's all.

*von Backström:* I'd like to make a comment to keep the record straight. I think most South African geologists would bear me out that the Witwatersrand Supergroup has been subjected to a considerable amount of tectonic disturbance, not only faulting but also very pronounced angular unconformities. There is also a certain amount of folding. I think it stands to reason with a very old formation like that, followed by the Ventersdorp volcanicity (outside Klerksdorp, boreholes of over 10,000 feet failed to drill through the Ventersdorp into the Witwatersrand below), there must have been an extreme disturbance in the floor of the basin and very deep valleys developed. Then there was the Bushveld Complex episode, which as you know is an enormous complex, that was emplaced in the immediate vicinity of the Witwatersrand and created a space problem. It certainly had severe effects on the Transvaal System, which covers the Ventersdorp. So my proposition is that I think that we must get away from this idea that the Witwatersrand has not been severely disturbed tectonically. I think that the fact that most of the mines have one or two geologists stationed at each mine and a lot of their work concerns displacement of the reef bears this out. The other point that I'd like to make is that in the Western Transvaal, certainly in the Ottosdal area where I worked and mapped, the pebbles in the lower part of the Dominion Reef were badly disturbed. They were shattered, and there is a terrific amount of shearing and displacement within the conglomerates within the reef, even a certain amount of streaking out. This is further evidence of tectonic disturbance, and I think that this would account for the fact that many of the pyrite pebbles have also been shattered.

*Bourret:* I would like to add one further comment to what Drs. Whiteside and von Backström have said in that we are carried away a little bit with an oversimplification of the tectonic picture. Dennis Pretorius continually brings out the fact that there are numerous sills and dikes and that 10 to 15 percent of

the basin actually is occupied by sills and dikes which have followed the fault patterns. There are three periods of igneous activity which are accounted for by the sills and dikes. The first is the post-Ventersdorp, then the post-Transvaal, and finally the post-Karoo periods of intrusive activity, and of course these tie in with the Bushveld Complex period of about 1,900 million years ago. I submit that not enough attention has been given to the tectonic disturbances that have taken place and are stamped in the record by these sills and dikes.

*Armstrong:* I have a question that I'd like to ask the South African contingent. If I understand the model in its broader aspects, the present thinking is that the basin was filled by coalescing fans that came from the north, the northwest, and the southwest, and that there were five or six separate source areas. No one has touched on the fineness of the gold. For you non-gold-miners, I am not talking about size, I'm talking about the purity of the gold. I would think, to support a placer genesis for the gold, that it would be highly unlikely that there would be a uniform fineness of the gold if it's derived from five separate source areas. Are there any maps showing the fineness distribution of the gold of the entire Rand, the distribution of the trace elements, the copper content, the silver content, or the ratios of different elements in the gold? Can any of you help me on this?

*Whiteside:* As far as I remember, Hargraves did some work some time ago on the silver content. He tried to plot the silver content of various parts of the Witwatersrand and I don't think the variation was very great. He tried to equate the silver content of the gold with depth, and I am not sure that he proved or disproved his point. I think you will find that report in one of the publications of the Economic Geology Research Unit of Wits University, one of the very early ones.

*Schidlowski:* As far as I remember, Hargraves worked on bullion, whereas Rudolf Saager and I, more or less simultaneously, applied microprobe techniques. Speaking for myself, I think I found on the average a silver content of roughly 10 percent. I think it was between 11 and 8. And I think Rudolf Saager found something similar. Is that correct?

*Saager:* Yes.

*Schidlowski:* I worked only on Orange Free State reefs.

*Saager:* Unfortunately, I also worked only on Orange Free State reefs, but I think there is a recent paper by Liebenberg, and there's an internal report of the National Institute of Metallurgy by [Viljoen?]. He found a greater variation than we did in the Basal Reef, and he had samples, I think, from about three or four of the goldfields.

*Armstrong:* I would think that you could sample the East Rand and the Central Rand and the far west and the Orange Free State and that, if indeed you do have five separate sources for the sediments and the gold, I would think that the character of the gold would be finger-printed by its trace-element content. A good many years ago I worked in the Porcupine district, Ontario, Canada, and there was a theft from the Dome Mine. Half the brick that was stolen was recovered by the gold squad, and they were able to identify it by the trace elements in it, and there was no doubt about it. The gold from each mine in the district had distinctive suites or amounts of trace elements in it. I would think that this type of study, if it hasn't been made, could lend great support to the detrital or nondetrital origin of Rand gold and might be an important feature to be studied. Is it that difficult to study?

*Myers:* Zemansky and Desboro wrote a paper a couple of years ago in which they show that gold subjected to the temperatures that the Rand simply had to be subjected to will homogenize in a relatively short time geologically. This complicates the problem. It is further complicated by the evident fact that there is silver somewhere other than in the gold. It just makes it a can of worms.

*Saager:* We analyzed gold on the microprobe, and we also had assays run on the same ore samples. We found almost no variation of fineness of the actual gold samples, but we found an increase of the silver content of the total ore sample. Also, you find a definite increase of silver in the ore samples down slope, whereas the fineness of the actual gold remains more or less constant. So silver must be floating around somewhere else.

*Grandstaff:* Dimock and Hargraves did a little bit of microprobe work for silver on the uraninites and found very low levels of silver in the uraninite. This could be an additional source of silver.

*Bourret:* Going back to this very famous volume by Pretorius, I'd like to read the comment he makes on page 42 with respect to silver where he states, "The percentage [of] silver in the [gold] bullion is lowest in the center of the lobe [of the conglomerate distribution,] as would be expected in a typical alluvial deposit where more pronounced movement of water down the main channel would have tended to leach out greater amounts of silver." I'm sorry he doesn't go on and explain that further.

*Skinner:* It's a well-known fact that's been known for a long, long time that detrital gold grains have gold-rich rims and silver-rich cores. So there is nothing mysterious about this.

*Hallbauer:* We have just started with analysis of

selected gold particles, freed from the rock by hydrofluoric acid treatment, for silver and trace elements by neutron activation analysis, but so far we haven't anything to report.

*Feather:* Just one final comment about the detrital gold grains being depleted in silver around the rims. This is absent in the Witwatersrand; the grains are remarkably homogenous. Also we've analyzed a large number of gold grains, and just to follow on what Rudolf [Saager] and Manfred [Schidlowski] have said, we do find that in the Vaal and Basal Reef samples there is very little variation in the silver content, but when we come to the Central Rand and the East Rand, the overall silver contents of the samples from different reefs vary vastly, suggesting multiple source for the gold in that northern area as opposed to the west and south area, but we haven't gone into this in any detail yet.

*Armstrong:* Did you analyze the gold for copper?

*Feather:* No, nickel and iron, and I haven't drawn any conclusions from those two elements.

*Skinner:* Brad Myers, I think, already gave the answer to the reason why you wouldn't expect to see the rims in the Witwatersrand. The Zamansky and Desboro paper he referred to is a study on the diffusion rate of silver in gold and gold in silver, and at a temperature like 150 or 200, you simply couldn't preserve a rim, you would just homogenize the whole grain.

*Whiteside:* May I just say one last word. In the assay offices on the East Rand, they used to deduct 10 percent from gold assays to allow for the silver content of samples from the Main Reef Leader, and 15 percent for samples from the Kimberley Reef, which is quite an appreciable difference. I have had gold assays made of some of the very small and narrow reefs high up in the sequence, right at the top of the Witwatersrand in the West Rand, that showed a silver content of almost 50 percent. So there is quite a considerable variation in the silver content of Rand gold, and the silver content appears to increase as you go up the geological column.

*von Backström:* Ladies and Gentlemen, it only remains for me as convener of the South African contribution to thank each and every one of you for your cooperation and the papers you have presented, and the way that you have presented them, and also, the participants in the discussions. And on behalf of all of us, a special word of thanks to the United States Geological Survey who made this wonderful trip possible. And then also, of course, the organizing committee that did such a vast amount of work and those behind the scenes, thanks ever so much. Also, all of our American friends for your hospitality. Thank you.

**BRAZIL**



## OPENING REMARKS

*Moura:* Ladies and gentlemen:

I'm glad to be again in this nice country of yours attending this meeting. Unfortunately, Dr. Villaça could not come to talk about uranium in conglomerates of the Precambrian Moeda Formation in Minas Gerais State. I'm going to try to do my best in spite of my problems in understanding and talking your language. First of all I'd like to thank you all on my company's behalf and on my own for the invitation to participate in this Workshop.

You are aware of the problems the world is facing now due to the shortage of energy and the increasing price of fuels. Of course, we realize that something has to be done about it.

I had originally intended only to present some information on work that is being done in Brazil in prospecting for uranium-bearing conglomerates. After hearing the excellent papers that have been presented during the last two days, however, I thought it would be of interest to make a few brief introductory remarks about the problems we are facing in Brazil, where we are in the earliest stages of prospecting.

It seems to me that, because of the world energy situation, new districts of uranium- and gold-bearing conglomerates should be found as quickly as possible. As a field geologist I have been wondering how could the detailed studies that we have heard these last days be used in finding new areas of interest. I have to confess that I did not succeed in getting any answer. Maybe I didn't understand the papers; I don't know.

I did get some scattered hints, however, that may make my work in Brazil become even harder. Dr. Minter, for instance, pointed out that the mean gold

content in conglomerate from several cores of the same reef could be something like 200 to 300 percent different from the true mean. If our mean grade is almost at the cutoff grade, how can we decide to put down a shaft where the decision is based on drill hole results in a completely new area? Of course, I know that in Canada and in South Africa the deposits are well known and the models are already developed.

How can we recover thin layers of carbonaceous material in quartzite without enough stratigraphic control? How can we use mineralogical associations to help in prospecting new areas? I understand that those data are usable in Canada where the stratigraphy is well established and one can know when he reaches the footwall. One need only back up and wedge the hole and drill more carefully. The potential of those areas is already known. But how does one do the same thing in new areas? I understand also that the mineralogical associations are different in Canada from those of the Witwatersrand and that they depend principally upon the rocks in the source areas. In Brazil the Jacobina as well as the Quadrilátero Ferrífero conglomerates are different from those of Canada and South Africa.

In the prospecting stage one hardly can afford detailed research. I think it is more useful to take models that have already been developed and to select those features that better fit our own problems, trying to use them as a first approach. Later on, when mines are in operation, then detailed research can be carried out.

As we are short of time, I think I'd better present the paper.



# Uranium in the Precambrian Moeda Formation, Minas Gerais, Brazil

*By* JOSÉ NILSON VILLAÇA *and* LUCIANO A. M. MOURA

GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN  
QUARTZ-PEBBLE CONGLOMERATES

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GEOLOGICAL SURVEY PROFESSIONAL PAPER 1161-T





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## GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN QUARTZ-PEBBLE CONGLOMERATES

### URANIUM IN PRECAMBRIAN MOEDA FORMATION, MINAS GERAIS, BRAZIL

By JOSÉ NILSON VILLAÇA and LUCIANO A. M. MOURA<sup>1</sup>

#### ABSTRACT

The Quadrilátero Ferrífero with an area of about 7,000 km<sup>2</sup> is located south of Belo Horizonte in Minas Gerais, Brazil. Precambrian metaconglomerates (Proterozoic) of the Moeda Formation and Maquiné Group are believed to be favorable host rocks for uranium deposits. Some areas are now being studied or have work planned for next year.

Drilling succeeded in detecting at least three channels in different areas with ore-grade uranium-bearing oligomictic conglomerates. Reserve calculations require additional detailed work in those areas.

Some models indicate that the sediments came from the São Francisco Craton, but paleocurrent directions in the Gandarela Syncline, as well as at Jacobina, indicate that the detritus came from the east, at least at these sites. This means that other cratonic areas must exist to the east of these outcrops.

The Quadrilátero Ferrífero (about 7,000 km<sup>2</sup>) is mostly included between lat 19°45' and 20°30' S. and long 43°22'30" and 44°7'30" W. and lies near Belo Horizonte, Minas Gerais State, Brazil.

#### INTRODUCTION

The Quadrilátero Ferrífero (about 7,000 km<sup>2</sup>) is mostly included between lat 19°45' and 20°30' S. and long 43°22'30" and 44°7'30" W. and lies near Belo Horizonte, Minas Gerais State, Brazil (fig. 1).

Uranium-bearing conglomerates in the Moeda Formation are known to crop out far beyond the limits of the Quadrilátero.

#### TRANSPORTATION AND COMMUNICATION

Some paved highways give access to the neighborhood of prospecting sites. Usually it is not necessary to travel more than 20 km from paved roads to reach the prospects.

A railroad network built for iron-ore transportation is in operation. Passenger transport within the region is by bus or car.

Frequent airplane service from Belo Horizonte is available to the main cities of Brazil.

Long-distance dialing telephone systems connects Belo Horizonte with other places of the country and abroad.

Electricity is available for mining operation, with powerlines near most of the sites of Quadrilátero Ferrífero.

#### CLIMATE

The climate in Quadrilátero Ferrífero, as well as in all central Brazil, is semitropical. Extreme temperatures are not known. The maximum is rarely over 30°C, and the minimum is usually above 0°C. The mean annual temperature is slightly below 20°C. Mean annual rainfall varies from place to place, from somewhat less than 1,500 mm to slightly over 2,000 mm.

#### TOPOGRAPHY

The topography is marked by high and rugged peaks, long chains of mountains, with some deep canyons and valleys. The maximum elevation in Quadrilátero Ferrífero is 2,100 m at the top of Serra do Caraça, and the minimum, about 600 m, where the Paraopeba River leaves the region.

#### HISTORY

The first prospecting work for uranium in Quadrilátero Ferrífero was done in 1971 by carborne survey, when more than 100 radioactivity anomalies were found. Of those anomalies, 27 percent were in granitic and gneissic rocks, 37 percent in schists of the Rio das Velhas Series, 30 percent in the Minas Series, mostly in conglomerates, and 6 percent in other rocks (fig. 2).

Due to the deep weathering in the area, chemical assays usually showed low uranium values in surface samples of conglomerates, 20 to 30 ppm for anomalies that give  $2 \times 10^3$  to  $5 \times 10^3$  cps (SPP-2). The chemical values increase with depth.

<sup>1</sup> NUCLEBRÁS, Distrito Belo Horizonte, Rua Washington, 801-S10N, Belo Horizonte, M. G., Brazil.

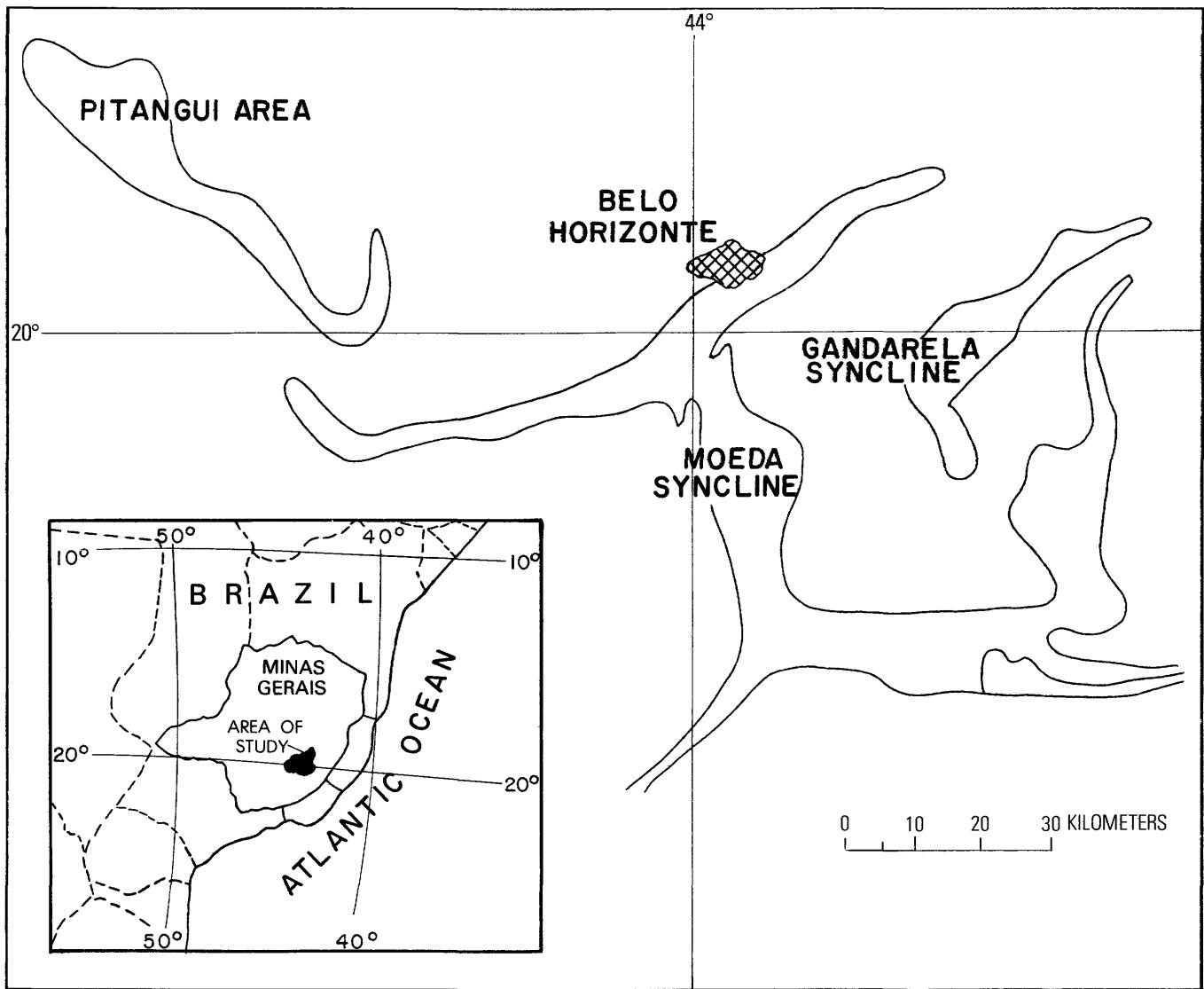


FIGURE 1.—Location and study area.

After preliminary mapping and radiometric studies, areas were selected for drilling. Encouraging results, with some ore-grade intersections in drill holes, indicated the need of more detailed mapping for better evaluation of the areas.

#### STRATIGRAPHIC SECTION

The stratigraphy used in the Quadrilátero Ferrífero is shown in table 1, which is taken from Dorr (1969, pl. 12).

Robertson (1974) correlated the Moeda Formation of the Caraça Group with other uranium-bearing Precambrian conglomerates of the world as shown in table 2. The Maquiné Group was added to the correlation by Villaça (1974) owing to its radioactive conglomerates.

The correlation (table 2) gives 2,200 million years as the age of the Moeda Formation, but rubidium-strontium determinations indicate an age of about 1,350 million years. It is thought that the radiometric age reflects later events rather than the age of deposition.

Around the São Francisco Craton (fig. 3) are Precambrian rocks that are potential hosts for uranium deposits.

The vectors on figure 3 show known paleocurrent directions in the area resulting from crossbedding measurements plotted on polar diagrams. In the western Quadrilátero Ferrífero as well as in the Pitangui area the sediments came from the São Francisco Craton. In the Gandarela Syncline, however, the source is located to the east (Lindsey, 1974).

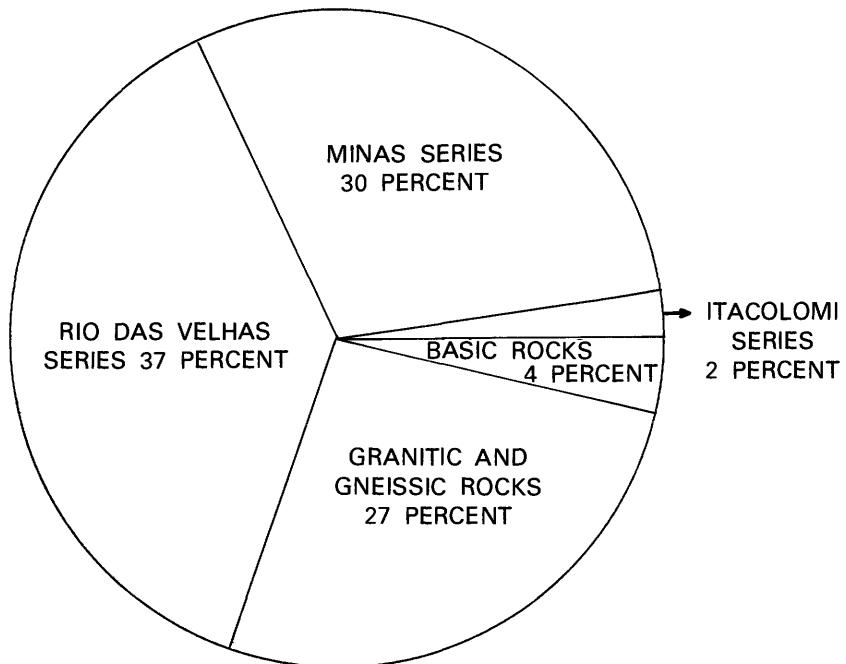


FIGURE 2.—Stratigraphic distribution of radioactive anomalies.

The paleocurrent directions in the Jacobina area (fig. 4) also indicate that the source lies to the east (Bastos and Moura, 1974).

The conglomerate pebble sizes both in the Jacobina area and in the eastern part of the Gandarela Syncline indicate a short distance of transport.

Jacobina, in the northeast corner of figure 3, is being studied by Unigeo, whereas Quadrilátero Ferrifero and Pitangui are almost unknown geologically. Reconnaissance work is necessary in order to locate where conglomerates occur.

#### AREAS BEING PROSPECTED

NUCLEBRÁS intends to continue work in the Moeda Formation at Pitangui and in the Ouro Fino Syncline during 1976.

Now the Company is studying the Gandarela structure in sites already drilled. A mapping program is underway in the Maquiné Group, where some radioactive conglomerates crop out. Included in the program are detailed geological mapping of some selected areas, surface sampling, and re-evaluation of the previous results of drilling programs. At least two years will be necessary to finish the work presently scheduled.

#### DISTRIBUTION OF CONGLOMERATES IN GANDARELA SYNCLINE

The maximum known thickness of the Moeda Formation is about 350 m in the southern part of the Gandarela

Syncline. Its thickness decreases northward and northwestward (fig. 5), and the total thickness of conglomerate in the Moeda also decreases northward and northwestward (fig. 6). Both maps are based on reconnaissance traverses made in 1974 along the entire length of the outcrop of Moeda Formation around the syncline and on data from drill holes.

The thick conglomerates in the southeast area are coarse conglomerates; some reefs contain boulders and cobbles (figs. 7, 8). This type of conglomerate is usually polymictic and immature, with cobbles of schist, indicating a short distance of transport.

Along the western side of the basin the conglomerates are thin, pebbles are rarely bigger than 3 cm in diameter (figs. 9, 10), and the channels pinch out quickly. The mean paleocurrent direction indicates a source area to the east. These conglomerates are a distal facies of the conglomerates that crop out to the east.

These data suggest that the conglomerates are present in subsurface over a large area of the syncline. If conglomerates are present, this is a favorable area in which to explore for uranium deposits.

#### MINERALIZATION

Both polymictic conglomerates and oligomicic quartz-pebble conglomerates occur in the western part of the Gandarela Syncline.

Southeast of this syncline at least two channels have been found, one of which is in the basal Moeda Forma-

TABLE 1.—Stratigraphic section in the Quadrilátero Ferrífero, Minas Gerais, Brazil

[Only Minas and Rio das Valhas Series]

AGE	SERIES	GROUP	FORMATION	SEDIMENTARY ENVIRONMENT
PRECAMBRIAN	MINAS	PIRACICABA	SABARA	FLYSCH
			BARREIRO	
			TABÓES	
			FECHO DO FUNIL	
			CERCADINHO	
		ITABIRA	GANDARELA	STABLE SHELF
			CAUÊ	
		CARACA	BATATAL	
			MOEDA	
	RIO DAS VELHAS	TAMANDUA	(UNNAMED)	STABLE SHELF
			CAMBOTAS	PARALIC
		MAQUINÉ	CASA FORTE	
			PALMITAL	MOLASSE
		NOVA LIMA	(UNDIVIDED)	FLYSCH
			BASEMENT NOT EXPOSED	

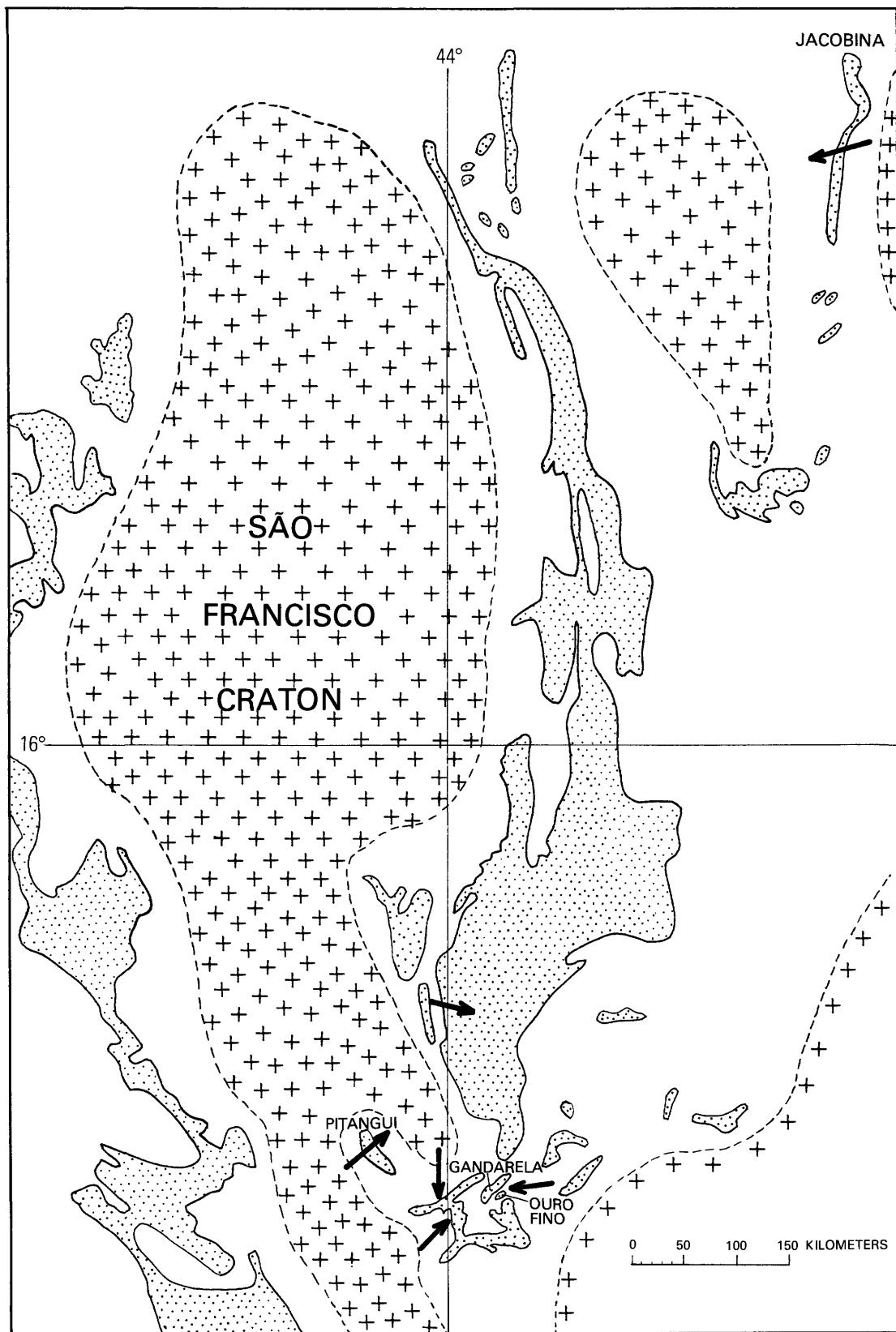


FIGURE 3.—Precambrian rocks (dots) around São Francisco Craton (crosses). Vectors show paleocurrent directions.

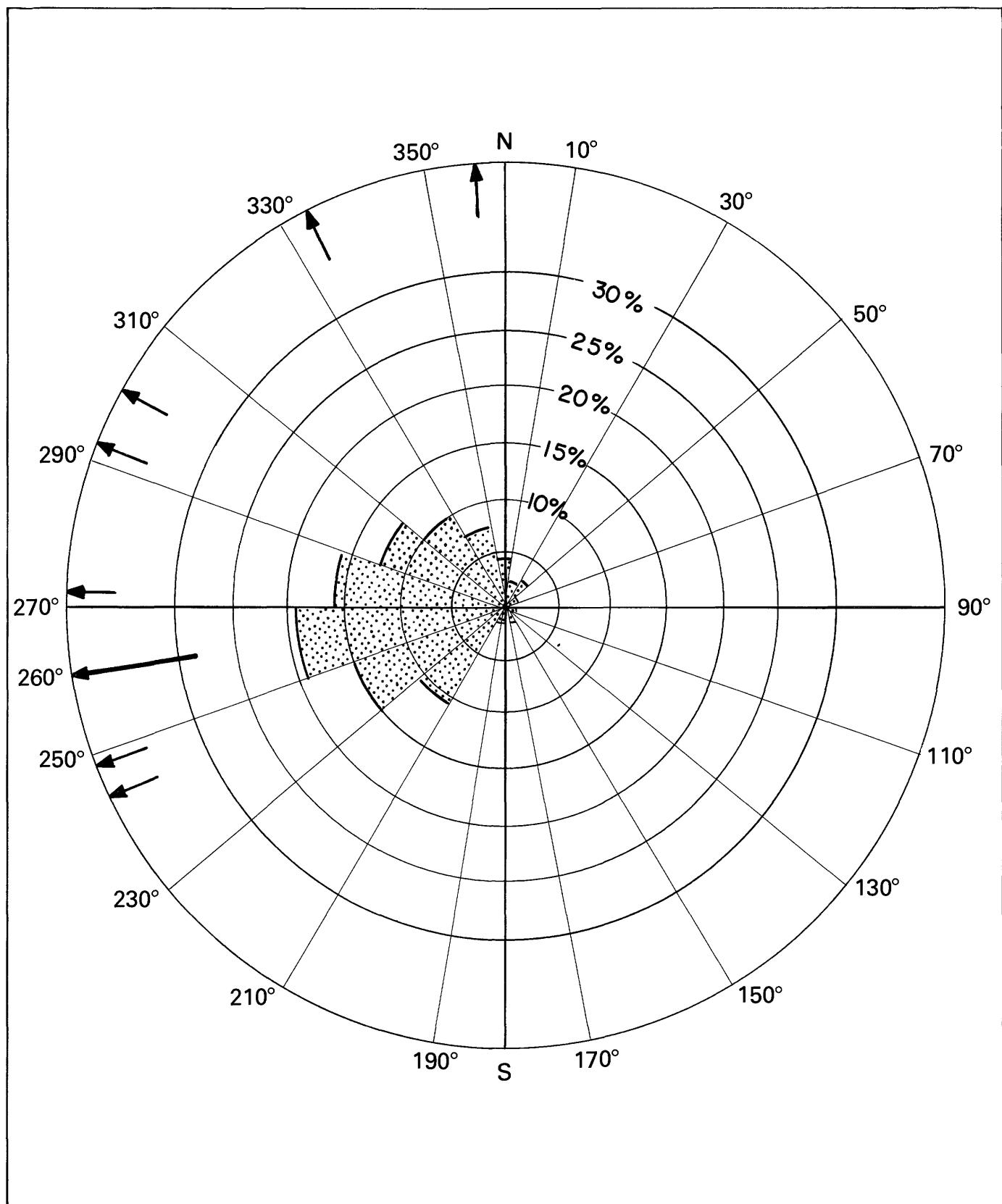


FIGURE 4.—Paleocurrent directions in Jacobina area. Long vector indicates paleocurrent direction for the area; short vectors indicate mean paleocurrent direction for each block.

TABLE 2.—*Stratigraphic correlations of Robertson (1974) with Maquiné Group added by Villaça (1974)*

ENVIRONMENTS	CANADA		SOUTH AFRICA	BRAZIL	AUSTRALIA
	ELLIOT LAKE	MONTGOMERY		QUADRILÁTERO FERRÍFERO	NULLAGINE
OXIDIZING (Red rocks)	LORRAINE FORMATION	HURWITZ GROUP	TRANSVAAL SUPERGROUP	ITABIRA GROUP	HAMERSLEY GROUP
			2200 M.Y.		
REDUCING (Sediments with pyrite and uranium)	ELLIOT GROUP	MONTGOMERY LAKE SEQUENCE	WITWATERSRAND SYSTEM DOMINION REEF SYSTEM	MOEDA FORMATION MAQUINÉ GROUP	GREEN HOLE FORMATION (U)
	(U)	(U+Au)	(Au+U)	(U+Au)	(U)

tion. Uranium may be present with some gold, and some drill holes cut ore-grade material (at the present price of  $U_3O_8$ ).

Thinning of conglomerates, imbricate thrust faults, and an overturned fold are shown in the cross section (fig. 11).

The structural complexity of the area and the great variation in grade require more detailed work to be done in order to delimit conglomerates and to evaluate reserves.

#### MINERALOGICAL ASSOCIATION

The matrix of the conglomerate is composed dominantly of fine quartz, pyrite, and rutile, with small amounts of zircon, kyanite, muscovite, molybdenite, and radioactive minerals. Amorphous carbonaceous material similar to thucholite has been found.

The thorium content of the conglomerates is low, and the  $ThO_2/U_3O_8$  ratio is variable. Great lateral variations of the ratio occur within the same conglomerate, and the ratio varies from one conglomerate to another. Figure 12 illustrates this variation in the basal conglomerate of the Moeda Formation in the southwest side of the Gondarela Syncline.

The relation between the  $U_3O_8/TiO_2$  and the  $ThO_2/TiO_2$  ratios in the basal conglomerate, based on drill core assays, is shown in figure 13. The uranium content increases where  $TiO_2$  exceeds 150 ppm, and it decreases where  $TiO_2$  exceeds 600 ppm. The pronounced decrease in the uranium content where the  $TiO_2$  reaches 350 ppm is related to insufficient data.

Insufficient gold analyses were available for valid conclusions to be drawn about its relation to uranium, thorium, and other elements present.

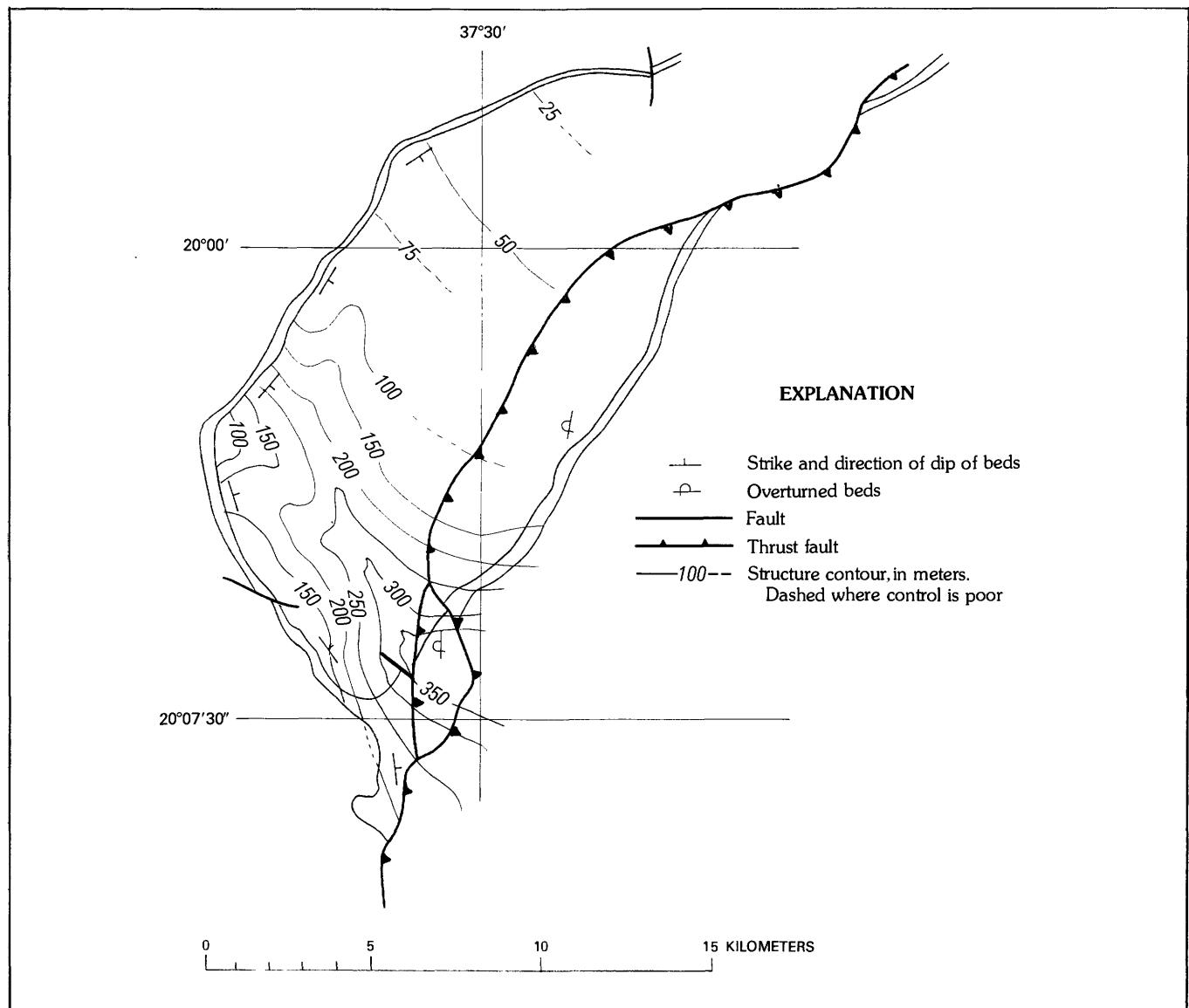


FIGURE 5. Isopach map of Moeda Formation in the Gandarela Syncline.

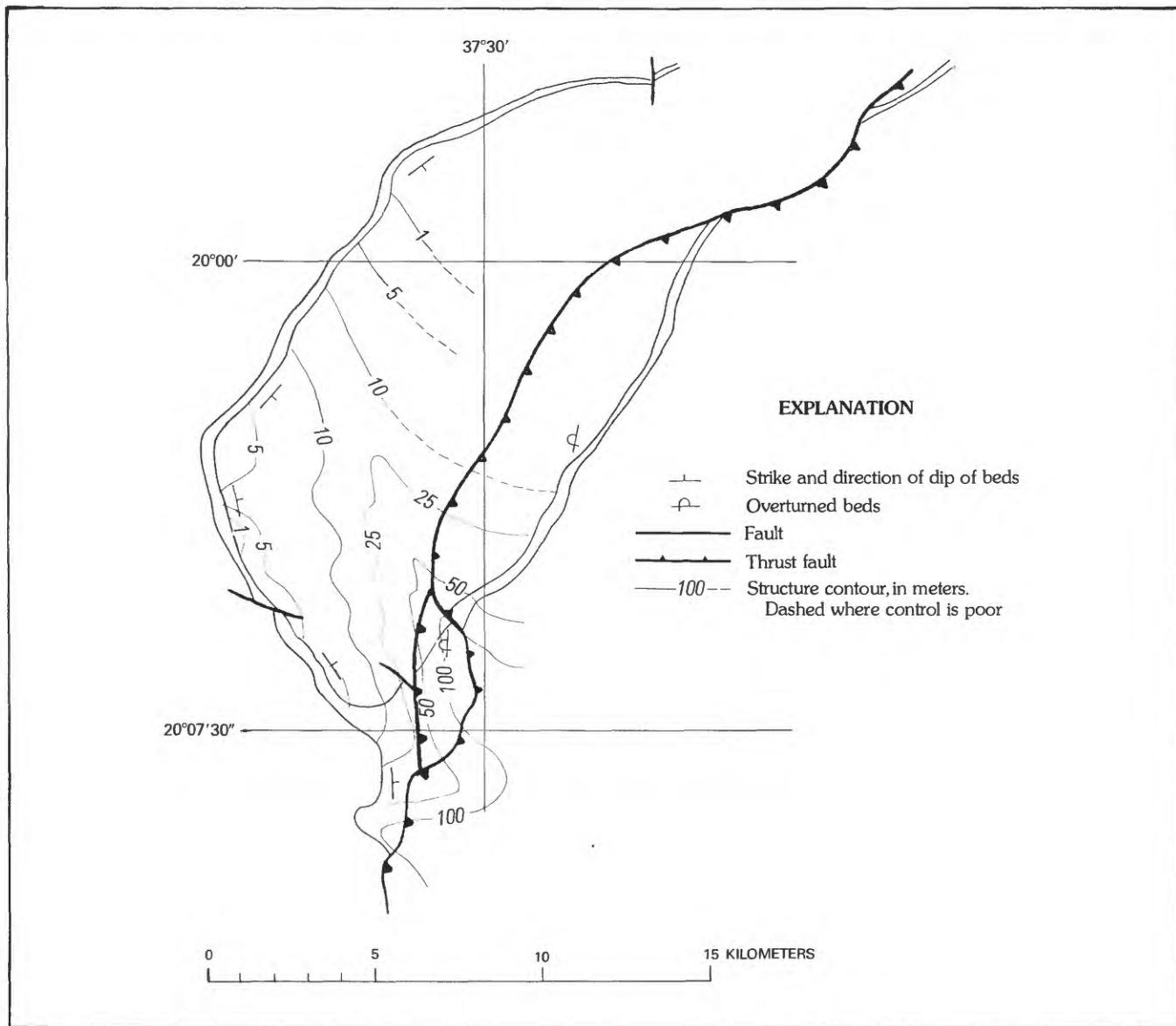


FIGURE 6.—Isopach map of total conglomerate in the Gandarela Syncline.



FIGURE 7.—Quartzite boulder, east of the Gandarela Syncline.



FIGURE 9.—Conglomerate with black quartz pebble, in the west of the Gandarela Syncline.



FIGURE 8. Polymictic conglomerate, cobble size, east of the Gandarela Syncline.



FIGURE 10.—Conglomerate with iron staining due to pyrite weathering.

### WORKING METHODS

Due to the structural and stratigraphic complexity of the area, a geologic mapping program covering 52 km<sup>2</sup> of the Gandarela Syncline was approved and is being carried out at the moment. East-west geologic traverses 200 m apart are being run from north-south base lines; about 260 km of geological traverses are planned for the Gandarela Syncline. Geologic mapping is being done at a scale of 1:2,500.

Trenches will be opened on the conglomerate zones for channel sampling. Mineralized conglomerates will be mapped at a scale of 1:500 and more detailed sampling done.

Diamond-drill cores, after being logged and systematically sampled, will be correlated with the mapping data in order to evaluate areas and to guide additional exploration.

### CONCLUSIONS

Many data are needed to estimate the potential of conglomerate zones in areas of strong tectonism.

Although high uranium grades have been found in some holes, following favorable host rocks requires a good knowledge of the tectonics and facies variations

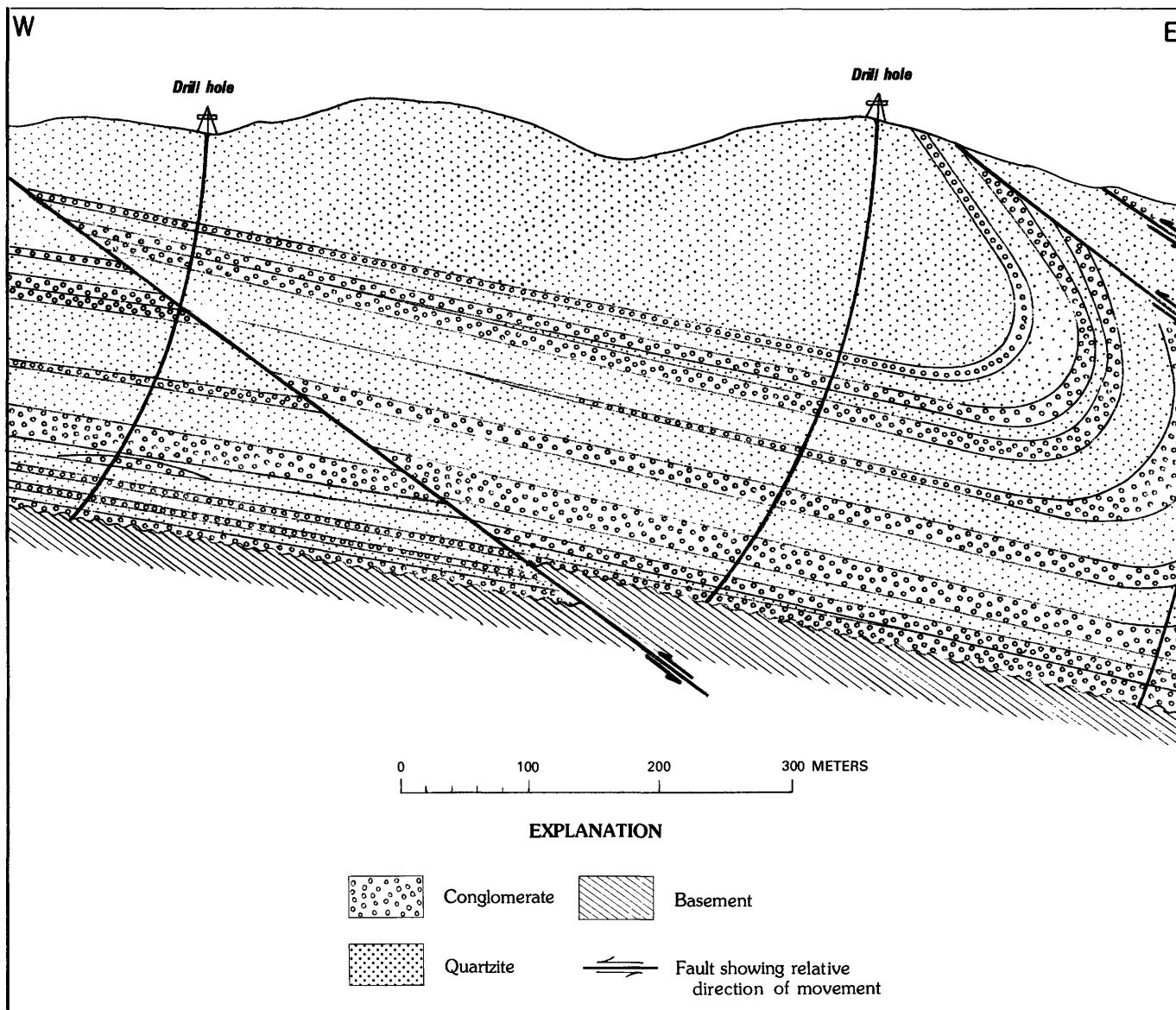


FIGURE 11.—Cross section of southeast Gandarela Syncline.

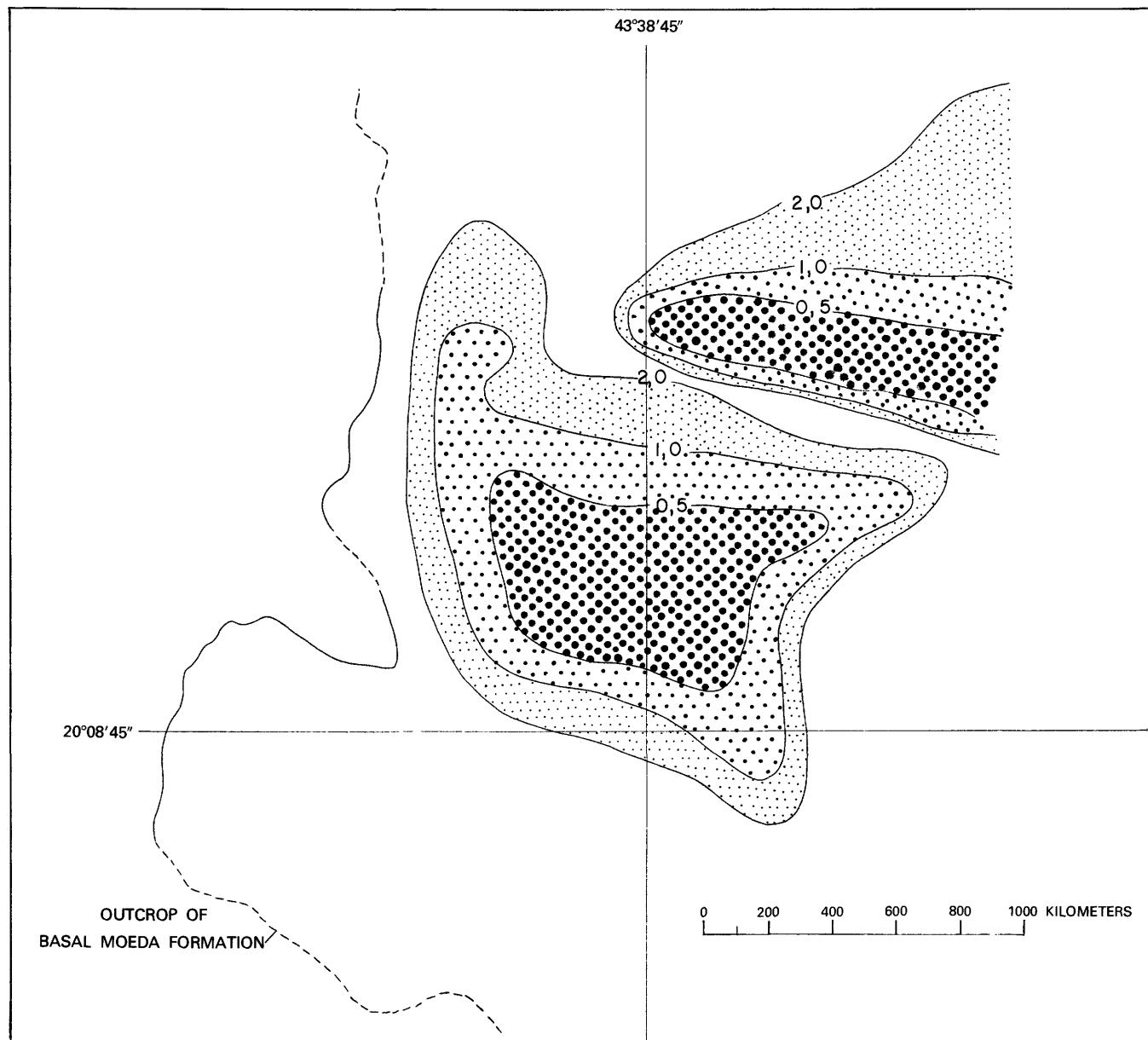


FIGURE 12.— $\text{ThO}_2/\text{U}_3\text{O}_8$  ratio distribution in the basal conglomerate in the southeast Gandarela Syncline.

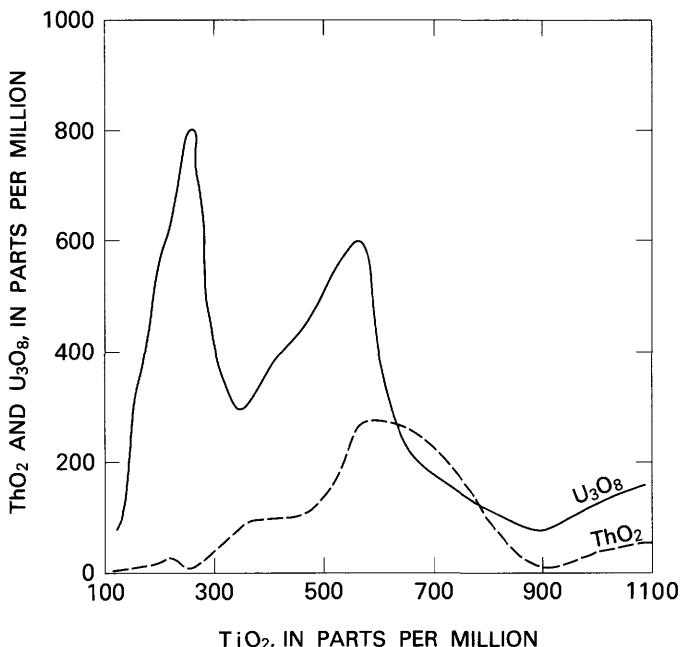


FIGURE 13.—Relationship between titanium oxide ( $TiO_2$ ) and uranium ( $U_3O_8$ ) and thorium ( $ThO_2$ ) oxides.

within the area so that the holes can be located in the most favourable sites.

Grade variations within ore horizons demand wedging of the drill holes to provide many intersection of the reefs, since a simple intersection is not able to collect a representative sample of large areas.

The extensive Proterozoic rocks with lithology similar to that of the Moeda Formation that crop out around São Francisco craton are hosts suitable for the occurrence of uranium deposits and as the result of more work may become favorable exploration targets.

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#### DISCUSSION

Cannon: Do you have any radiometric data on what the age of the Moeda Formation is?

Moura: There are some strontium-rubidium measurements that give less than 1,800 million years, but we usually consider that these ages might reflect later events and be due to metamorphism. We don't know yet. We have red-bed formations right above the reducing environment of the Moeda Formation; the Caué Formation is itabirite iron ore. In other areas of the world this change from reducing to oxidizing conditions is thought to have occurred about 2,200 million years ago.

Robertson: Along the same lines, do you have an age on the São Francisco Craton?

Moura: No. We are just in the early stages of prospection.

Whiteside: If I may just comment a little bit. I was very lucky. I visited Belo Horizonte about 1971 when they first found uranium and I saw some of the outcrops. They have great difficulties there because weathering is extremely deep and it's only a few of the more resistant beds that crop out, mostly quartzites. They stick up as hogback or sawtooth ridges. The few outcrops I did see of conglomerate were small pebble, I would say about a half an inch, pyritic, and looked very like some of the Witwatersrand conglomerates, but those did not carry either gold or uranium. I think it's mostly towards the base of the Moeda that mineralization occurs, and I think the ones I saw must be somewhere near the top. The only piece of real mineralization, if I may say so, that I saw was a black mineral in a little bit of core. They asked me what it was, and I said, "I don't know." Actually, it was a piece of uraninite; it was about 2 mm in length and 1 mm broad, and it looked as though it was rounded. There is not very much more I can say, but on the whole, I don't think I would have correlated what I saw in the Moeda with what I know of the Witwatersrand. (This is a very long-range correlation, of course, in spite of continental drift.) I don't think there would be anything in Brazil equivalent to the division of argillaceous-arenaceous in the lower Witwatersrand and arenaceous-rudaceous in the upper Witwatersrand. I never saw anything like that in Brazil.

Ruzika: Can you tell us something about the carbon content in the Moeda?

Moura: Some analyses were made by Robertson and Associates of carbon seams and things like that, that Robertson suspected could be thucholite. In his report he reported two or three samples as thucholite, but I don't think that it has been established yet.

Skinner: With so much titanium around, did you see any brannerite? Is there much brannerite in the deposit

*Moura:* I haven't seen any, but mineralogical studies have found uraninite and brannerite, sometimes associated with pyrite and sometimes with what was called thucholite, but we are not sure of the last one. In the matrix is fine quartz, pyrite, and rutile. Fine quartz grains and pyrite are the most abundant minerals, and small amounts of zircon, kyanite, muscovite, and molybdenite are also present in the area.

*Armstrong:* On your map you showed the area south of Belo Horizonte and Jacobina, and you said that in the intervening area there are rocks of the same age and lithology and that this intervening area would be a good one to prospect. If I recall correctly, rocks of that same age also extend an equal distance to the northwest along the western flank of the São Francisco Craton. Is that an equally good prospecting area?

*Moura:* Yes, we think so. You saw the São Francisco Craton and on both sides of it there were some data points. We think that both areas are favorable for prospecting, but we have only scattered data on the western area. On the southwest side of the craton in the Pitangui area we have found uranium- and gold-bearing conglomerate, but we don't know if it will make a mine.

*Armstrong:* Yes, but I really meant a long way northwest of the Pitangui area.

*Moura:* Yes, it continues over there. Sometimes it's buried by younger rocks but it appears some places.

*Meyer, Falconbridge Nickel Mines, Ltd., Canada:* May I just echo some of Mr. Moura's words earlier in his talk? The excellent presentations of the last two days will give valuable direction to any geologist who has an established ore body in the ground, but the basic

question in the minds of many of us here "How do we find another Witwatersrand or another Blind River?" - remains unanswered. We are two-thirds of the way through our time here and may I ask the remaining speakers to give some thought to this most important question.

*Moura:* Excuse me, I didn't get the question he made. Could you translate?

*Armstrong:* It doesn't require an answer. All he said was that he agrees with your opening remarks. How do we find other deposits?

*Thorman, U.S.G.S.:* I would like to echo what Dr. Whiteside said, probably a little louder, in that if you haven't been there you can't appreciate the exploration problems. Weathering is intense, the area is enormous, in fact it's huge, and the work they are doing now is quite good in view of the manpower that's available and the area that you have to cover. I spent almost two years down there mapping the Quadrilátero Ferrífero, and I can confirm that the stratigraphic and structural problems are complex. We'd really like to start mapping all over again because now we know where the outcrops are, and with a better knowledge of the stratigraphy we could do a better job. So I think I'll echo Luciano's [Moura] statement that the microprobe and other things are great tools once you know where you are geologically, but it takes a tremendous amount of basic work before you can start using microprobes and that sort of thing beneficially.

*Armstrong:* From a few days inspection I can state that the structure is more complicated than was indicated on the cross sections. The thrust faults have been cut by later high-angle faults so that the rocks are cut up into fault-bounded blocks.

**CANADA**



# The Blind River Uranium Deposits: The Ores and Their Setting

*By* JAMES A. ROBERTSON

GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN  
QUARTZ-PEBBLE CONGLOMERATES

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GEOLOGICAL SURVEY PROFESSIONAL PAPER 1161 - U





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## GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN QUARTZ-PEBBLE CONGLOMERATES

### THE BLIND RIVER URANIUM DEPOSITS: THE ORES AND THEIR SETTING<sup>1</sup>

By JAMES A. ROBERTSON<sup>2</sup>

#### ABSTRACT

In the Blind River area, Proterozoic clastic sedimentary and minor volcanic rocks (Huronian Supergroup) unconformably overlie and transgress northward over dominantly granitic Archean terrane (2,500 million years) and are intruded by Nipissing Diabase (2,150 million years). Later deformations and metamorphic events are recognized.

The Matinenda Formation (basal Huronian) comprises northward-derived arkose, quartzite, and pyritic, uraniferous oligomictic conglomerates, which contain 75 percent of Canada's uranium reserves. Historic grades approximate 2 pounds  $U_3O_8$ /ton (1 kilogram/metric ton), but lower grade material can be mined with increasing price. Some thorium and rare earths have been marketed. The general absence of gold reflects lack in the source area.

The conglomerate beds lie in southeasterly striking zones controlled by basement topography down-sedimentation from radioactive Archean granite. Distribution of monazite relative to uraninite and "brannerite" and the presence of uranium values in overlying polymictic conglomerates, which truncate the ore beds, indicate that the mineralization is syngenetic, probably placer. The role of penecontemporaneous mafic volcanics is problematical, but these could have been a source for sulphur in the pyrite.

Drab-coloured rocks, uranium and sulphide mineralization, and a post-Archean regolith formed under reducing conditions all suggest a reducing environment. Sedimentary features indicate deposition in fast-flowing shallow water and possibly a cold climate. In the upper Huronian (Lorrain Formation), a monazite and iron oxide assemblage associated with red beds suggests a change to oxidizing conditions.

Similar deposits occur at Agnew Lake, and polymictic conglomerate and siltstone found north of Sudbury carry minor uranium and locally gold values. Provenance, lack of oxygen, transportation and depositional processes, and no major modification by later events are the dominant factors in the formation and preservation of the ore bodies.

Research is needed on mineralogical relations, detailed sedimentology of the conglomerates, and the volcanic rocks, their nature and role.

#### INTRODUCTION

Economic Blind River uraniferous pyritic oligomictic conglomerates were found in 1953; the presence of uraniferous conglomerate had been known since 1948. Since 1953, the activities of mining companies, of uni-

panies, of universities, and of both Canadian and Ontario government geologists have led to a large body of literature and other data. This paper is an attempt to summarize the present state of knowledge and to provide guidelines for the exploration geologist or the geologist in the field who may find himself in favorable terrane; it also serves as background for the more detailed papers that follow. The paper is in three sections: first a description of the regional geology, second a description of the ore-bearing conglomerates, and third a discussion of those factors of genetic significance.

#### REGIONAL GEOLOGY

Blind River (fig. 1) lies on the north shore of Lake Huron, halfway between Sudbury and Sault Sainte Marie. The town of Elliot Lake, built to service the uranium mines, lies 20 miles northeast of Blind River.

The region lies on the boundary between the Southern and Superior Province of the Canadian Shield. The Superior Province (Goodwin and others, 1972) comprises Archean rocks that were affected by the Kenoran Orogeny (2,500 m.y. (million years)), and the Southern Province (Card and others, 1972) includes Proterozoic rocks affected by the Penokean Orogeny (1,750 m.y.). The Blind River-Elliot Lake area lies at the margin both of early Proterozoic sedimentation and of the mid-Proterozoic metamorphism.

Table 1 is a table of formations using the nomenclature recommended by the Federal-Provincial Committee on Huronian Stratigraphy (Robertson and others, 1969), and table 2 permits comparison with schemes used in publications prior to 1969. Table 3 provides a synopsis of stratigraphy and its relation to mineralization.

The bedrock of the area falls into three broad units, the distribution of which is shown on figure 2. These are (1) the Archean basement, consisting of Keewatin-type greenstone, Algoman granite, and minor mafic intrusives, (2) the Huronian sedimentary and metasedimentary rocks, with local minor mafic volcanics, and (3)

<sup>1</sup> This paper was presented at the workshop by James A. Robertson. In 1976 it was published under the same title by the Ontario Division of Mines as Miscellaneous Paper 65, 45 pages. It is reprinted here, with some minor changes, with the permission of the Director, Ontario Geological Survey, Ministry of Natural Resources.

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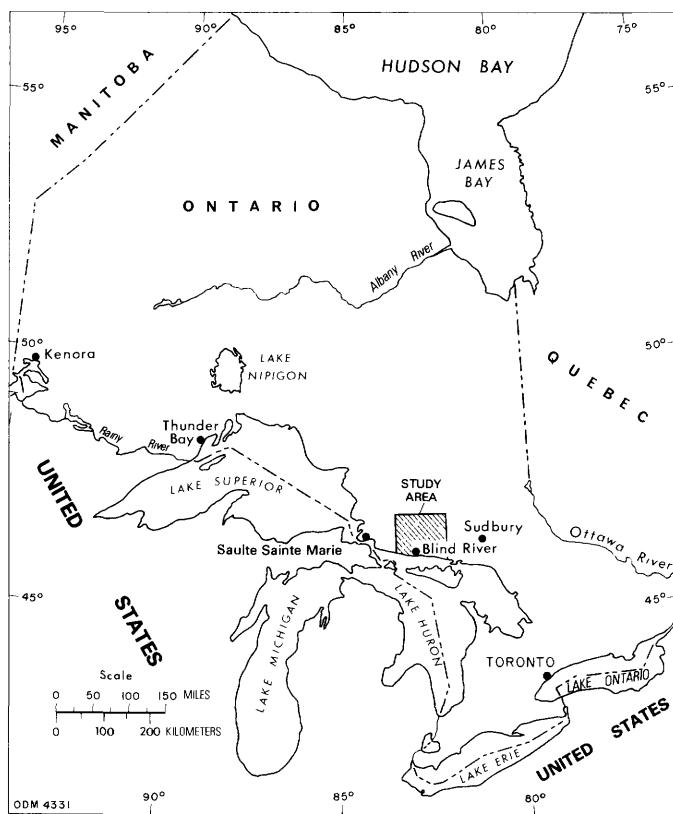


FIGURE 1.- Location of Blind River-Elliot Lake area.

the post-Huronian intrusive rocks, comprising the Nipissing diabase sills, diabase dikes, the Cutler Granite, the Croker Island Complex, and olivine diabase believed to be Keweenawan in age (only the Cutler Batholith and the Croker Island Complex are shown in figure 2). The structure is also illustrated in figure 2. In the north-central zone lies the Quirke Syncline; farther north is the Flack Lake Fault, with upper Huronian rocks exposed on the downthrown (north) side; and in the center is the Chiblow Anticline, the south limb of which is repeated by a major east-striking fault—the Murray Fault. The axes of these folds strike slightly north of west and plunge gently west, giving the sedimentary units a reverse S-shaped outcrop. To the south of the Murray Fault, east of Algoma, the folding and metamorphism are more intense and complex. To the east of the Cutler Batholith, fold axes and other linear features generally plunge eastward, but to the west of the batholith the linear structures plunge westward. Only the most clearly defined folds are shown in figure 2. The east end of the Cutler Batholith is controlled by the Spanish Anticline (Robertson, 1966a, 1975a; Cannon, 1967). The La Cloche Hills are defined by the Lorrain quartzite, which forms the limbs of the La Cloche Syncline. The Croker Island Complex lies near the axis of the McGregor Bay Anticline. When traced eastward,

TABLE 1. *Formations for the Blind River-Elliot Lake area*

[Codes keyed to fig. 2 are in parentheses. Geologic ages given are from a variety of sources]

Unit	Dominant lithology	Age (million years)
<b>Phanerozoic</b>		
Cenozoic		
Pleistocene and Recent	Sand, gravel, till	
<i>Unconformity</i>		
Paleozoic		
Ordovician (20)	Limestone	
Precambrian		
Proterozoic		
Keweenawan Supergroup		
Sudbury dikes (19)	Olivine diabase	1,225
<i>Intrusive contact</i>		
Mount Lake Dike	Quartz diabase	
<i>Intrusive contact with Nipissing Diabase</i> <sup>(1)</sup>		
Hudsonian		
Croker Island Complex (18)	Gabbro, granite	1,445
Cutler Batholith (18)	Granite	1,750
<i>Intrusive contact</i>		
Penokean		
Nipissing (17)	Quartz diabase, diorite	2,155
<i>Intrusive contact</i>		
Huronian Supergroup		
Cobalt Group		
Bar River (16)	Quartzite	
Gordon Lake (15)	Siltstone, sandstone	
Lorrain (14)	Quartzite, conglomerate arkose	
Gowganda (13)	Conglomerate, greywacke, quartzite	
<i>Unconformity-disconformity</i>		
Quirke Lake Group		
Serpent (12)	Quartzite	
Espanola (11)	Limestone, siltstone	
Bruce (11)	Conglomerate	
<i>Local disconformity</i>		
Hough Lake Group		
Mississagi (10)	Quartzite	
Pecors (9)	Argillite	
Ramsay Lake (8)	Conglomerate	
<i>Local disconformity</i>		
Elliot Lake Group <sup>2</sup>		
McKim (7)	Argillite	
Matinenda (5)	Quartzite, with or without uranium conglomerate	
	Conglomerate	
	Arkose, with or without uranium-conglomerate regolith	
<i>Unconformity</i>		
Archean		
Late Archean intrusives	Diabase	2,500
<i>Intrusive contact</i>		
Kenoran (Algoman) (3)	Granite	2,500+
<i>Intrusive contact</i>		
Early Archean intrusives	Gabbro	
<i>Intrusive contact</i>		
Keewatin (1)	Volcanic and sedimentary rocks	

<sup>1</sup> Mount Lake Dike may be 1,795 m.y. old.

<sup>2</sup> Volcanic rocks are found locally in the Elliot Lake Group (6); each occurrence has been given its own name; age about 2,400 m.y.

TABLE 2. -*Systems of Huronian stratigraphic nomenclature*

COLLINS (1925)		ROBERTSON (1967a)		ROSCOE (1960)		ROBERTSON AND OTHERS (1969)		
Cobalt Series	Upper White Q <sup>1</sup>	Cobalt Group	Bar River		Cobalt Group	Upper White Q <sup>1</sup>	Cobalt Group	
	Banded Cherty		Gordon Lake			Banded Cherty		
	Lorrain		Lorrain			Lorrain		
	Gowganda		Gowganda			Gowganda		
Bruce Series	Serpent	Bruce Group	Serpent		Quirke Lake Group	Serpent	Quirke Lake Group	
	Espanola		Espanola			Espanola		
	Bruce		Bruce			Bruce		
	Mississagi (Ramsay Lake)		Upper Mississagi		Hough Group	Mississagi	Hough Lake Group	
			Middle Mississagi argillite			Pecors		
	Granites		Conglomerate			Whiskey		
			Lower Mississagi argillite		Elliot Group	Nordic	Elliot Lake Group <sup>2</sup>	
			Quartzite			Matinenda		
Archean	Sudbury Series	Archean	Algoman		Archean	Archean		
	McKim		Keewatin			Archean		
	Schist Complex							

<sup>1</sup>Q. equals Quartzite<sup>2</sup>Volcanic rocks are found locally in the Elliot Lake group.

the La Cloche Syncline and the McGregor Bay Anticline are part of a series of folds trending easterly between Espanola and the Grenville Front (Card, 1967a-d; Young, 1966; Young and Church, 1966).

Bedding-plane slips, thrust faults, and near-vertical faults that strike either northwest or parallel to the axial planes of the folds are common. The fault pattern, jointing, slickensides on bedding planes, and drag folds suggest that north-south compression formed the folds. Several periods of intensity of deformation and metamorphism can be demonstrated.

#### ARCHEAN

##### KEEWATIN-TYPE ROCKS

Keewatin-type rocks are found locally in the Archean of the North Shore of Lake Huron and particularly underlie the Huronian rocks near Elliot Lake. The rock-types found include massive and pillow lavas, pyroclastic rocks, and sedimentary rocks including lean iron-formation. The strike is northwest, and dips are generally steep in the northeast. Metamorphism is of the chlorite grade, rising to the amphibolite grade in hybrid zones close to contacts with the Algoman granite. The general lack of greenstone in the cratonic block north of the Huronian basin and a low frequency of known gold

occurrences in the area believed to be the source area are mirrored in the lack of gold in the uraniferous conglomerates.

#### ALGOMAN GRANITIC ROCKS

Granitic rocks of Algoman age (2,500 m.y. (Lowdon, 1960, 1961; Lowdon and others, 1962, 1963; Van Schmus, 1965; Wanless and others, 1965)) underlie approximately half the area shown on figure 2. These granitic rocks may be divided into two broad groups: (1) medium- to coarse-grained, gneissic to massive granodiorite, generally grey to pink in colour with abundant inclusions of Keewatin-type rocks; and (2) massive red quartz monzonite, generally without inclusions and slightly radioactive. Bodies of the second type are found in the Quirke Lake, Aubrey Falls-Ranger Lake, and Elliot Lake areas (fig. 2; Robertson, 1960, 1961, 1967, 1968a).

It is of interest that in the Montreal River area north of Sault Ste. Marie and eastwards as far as Aubrey Falls there are occurrences of pitchblende coating joints in the granitic rock or associated with the contact of late Precambrian diabase and early Precambrian granite. In the same area uraninite occurs in Archean pegmatite (Nuffield, 1955; Robertson, 1968b, 1975a). A statistical analysis of crossbedding and size distribution of chert

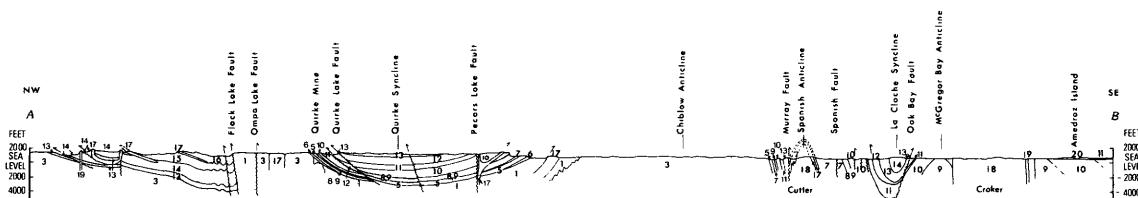
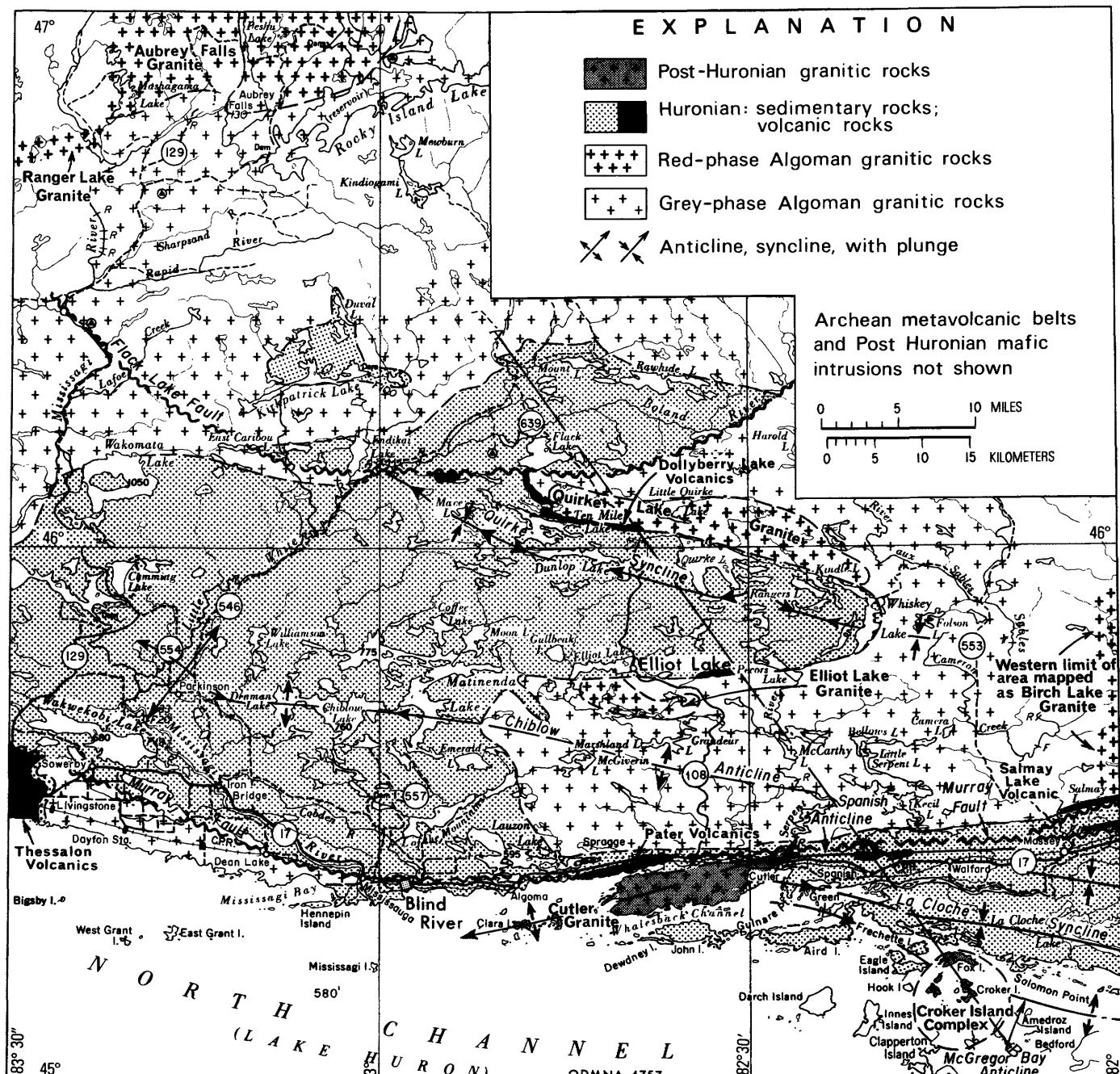


FIGURE 2.—Generalized geologic map of the Blind River-Elliot Lake area, with schematic cross section of the Elliot Lake area (line A-B). Units on the cross section are keyed to table 1.

TABLE 3.—*Summary of Huronian stratigraphy in the Blind River-Elliot Lake area*

Group	Formation	Lithology	Thickness (in feet)	Depositional environment	Source	Mineralization
Cobalt	Bar River	Quartzite	At least 1,000 at Flack Lake; at least 4,000 at Willisville.	Shallow water	Source north, but currents variable.	
	Gordon Lake	Siltstone, sandstone.	1,000 at Flack Lake; 3,000 at Willisville.	Shallow water		
	Lorrain	Quartzite, congo- merate, arkose.	2,000-6,000	Shallow water	North-northwest	Th-U in north; Cu?
	Gowganda	Conglomerate, grey- wacke, quartzite, siltstone.	500-4,200	Glacial in north; glacial-marine in south.	North-northwest	
Quirke Lake	Serpent	Quartzite	0-1,100	Shallow water	Northwest	
	Espanola	Limestone, dolo- stone, siltstone.	0-1,500	Shallow water	Northwest	
	Bruce	Conglomerate	0-200	Glacial shallow water.	North?	
Hough Lake	Mississagi	Quartzite	0-3,000+	Shallow water	West-northwest in west; north in southeast.	U near base- ment highs.
	Pecors	Argillite	40-1,000+	Shallow water	North-northwest	Traces U near basement high.
	Ramsay Lake	Conglomerate	5-200	Glacial shallow water.	Northwest?	Traces U where un- conformable on Matin- enda Forma- tion.
Elliot Lake	McKim	Argillite-grey- wacke.	0-2,500+	Shallow water (turbidite).	Northwest	Traces U near basement highs.
	Matinenda	Quartzite, arkose, conglomerate.	0-700+	Shallow water	Northwest	U-Th-rare earths in con- glomerate in basement lows. <sup>1</sup>
Volcanic rocks	Andesite-bassalt (fel- sic volcanics).	Local piles	Subaerial	Flack Lake Fault; Murray Fault.		U-Th in con- glomerate interbeds.

<sup>1</sup> All uranium deposits of commercial importance in the Blind River-Elliot Lake area are in the Matinenda Formation.

pebbles in the Mississagi Formation (McDowell, 1957) indicated that the source area for the lower Huronian lay 130-250 miles (210-400 km) west-northwest of Thessalon.

More recently airborne  $\gamma$ -ray-spectrometer surveys have confirmed that much of the granitic terrane north and northwest of the Elliot Lake area contains anomalous amounts of uranium (Darnley and Grasty, 1971; Richardson and others, 1975). In general, those areas containing the massive red quartz monzonite exhibit anomalous uranium content.

The massive red granitic rocks of the Archean basement are held to be the source of the uranium found in the Huronian conglomerate.

After the peak of the Kenoran Orogeny the area was fractured and numerous diabase dikes were intruded.

The region was then subjected to a prolonged period of weathering and peneplanation. During this period hollows or valleys developed over the more susceptible rocks, such as those composing the greenstone belts. Throughout the Blind River area partially weathered zones have been preserved under the Huronian-Archean unconformity and, particularly, over the granitic rocks.

At distances of greater than 100 yards (90 m) from the contact with the overlying Huronian, the granitic rocks exposed at the surface are of the normal type. However, as the actual contact is approached, the plagioclase feldspar grains become yellow, owing to the development of sericite, and the ferromagnesian minerals are no longer present; the granitic texture is preserved by the quartz and microcline crystals. This material passes into an unsorted aggregate of partly corroded quartz

and microcline grains in a yellow-green matrix of sericite. Rarely, angular fragments of vein quartz and patches of less-altered granite may be present. This material is overlain by sorted sericitic arkose, in which bedding and crossbedding are generally visible. In some outcrops the actual contact may be marked by a band of angular to subangular quartz fragments up to an inch across.

This transition zone is generally interpreted as a regolith or fossil weathered zone developed during the Archean-Proterozoic interval. Such weathered zones have been observed over both red and grey phase Algoman granites. Core sections also show the development of crumbly chloritic material between the greenstone and the overlying sedimentary rocks (Rice, 1958). This has also been regarded as regolith. Both Pienaar (1963) and J. A. Robertson (1960 and later) have analysed samples of regolith. Figure 3, derived from their analyses, is a straight-line diagram illustrating development of regolith. The following features are apparent:

1. Silica is a constant, and there is a slight gain in alumina.
2. Zirconia is constant, reflecting stability of zircon.
3. Total iron and both ferrous and ferric iron show marked loss; ferric iron is lost more completely than ferrous.
4. Magnesia and manganese have been partially lost.
5. Lime and strontium and soda have been markedly reduced.
6. Potash, rubidium, and water (+) are strongly increased.

These observations reflect (1) the destruction of the plagioclases and the removal of the soluble constituents, (2) the stability of the potassic feldspars (microcline), and (3) the formation of hydrated clay minerals represented by sericite. The trace elements follow the major elements in pairs: Mn-Mg, Rb-K, and Sr-Ca.

Total iron has been lost, suggesting that  $Fe_2O_3$  has been converted to FeO and removed by leaching. Such reduction may have been owing to the exclusion of the iron from the atmosphere by overlying material or to an atmosphere deficient in oxygen.

Roscoe and Steacy (1958, p. 4-5) studied the distribution of uranium and thorium in two series of saprolite (regolith) samples from the Quirke Lake area of Township 144. They stated, "Both show uranium to be about one-third less in the most altered samples than in the freshest granite. One shows a proportional loss of thorium which is only slightly less than the loss of uranium. The other shows a net gain of thorium."

As regolithic material contributed to the formation of the uraniferous, pyritiferous, oligomictic conglomerates of the basal Lower Huronian, the presence in the

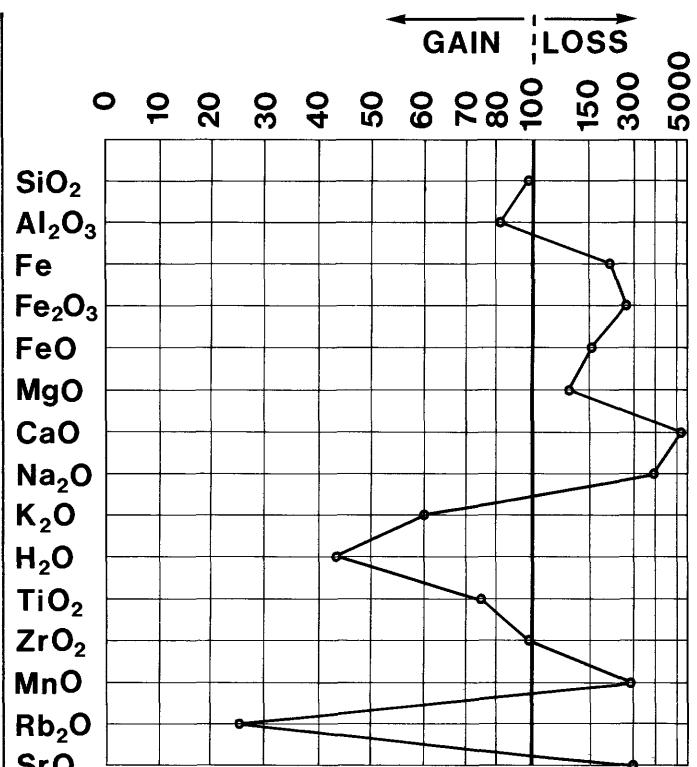


FIGURE 3.- Formation of regolith.

regolith of FeO (ferrous iron) and the persistence of uranium normally soluble in an oxidizing environment are of interest, whatever the cause of reduction.

The source area for the uranium-bearing rocks was subjected to prolonged weathering with development of residual regolith under reducing conditions. Slight topographic variation was developed.

#### HURONIAN SUPERGROUP

The Huronian Supergroup unconformably overlies the Archean rocks. The classical descriptions were given by Logan (1863) and Collins (1925). Recent work has resulted in several revisions of Huronian stratigraphy (Robertson and others, 1969), including the recognition of the cyclical nature of much of the sequence and the presence of mafic volcanic rocks near the base of the sequence at many localities (Roscoe, 1969; Robertson, 1971b).

#### ELLIOT LAKE GROUP

The lowermost group of the Huronian is the Elliot Lake Group, comprising the Matinenda and McKim Formations and local mafic volcanic assemblages.

#### MATINENDA FORMATION

The Matinenda Formation consists of greenish arkose, with or without uraniferous quartz-pebble conglomerate

beds, followed by grey quartzite. Crossbedding and pebble-orientation studies by McDowell (1957, 1963) and Pienaar (1963) indicate that the currents flowed from the northwest but were markedly influenced by basement topography, which in turn reflects the Archean structure. The ore conglomerates occur mainly in valleys in the basement surface (see fig. 7). The thickness of the Matinenda Formation increases from 0 at the north shore of Quirke Lake through 600 ft (180 m)

near Elliot Lake to 700 ft (210 m) on the north side of the Murray Fault at the Pronto Mine. Superimposed on the regional pattern are the effects of basement highs and lows. The formation has not been recognized south of the Murray Fault east of Algoma, but it does lie under Lake Huron west of Blind River. As northward overlap is pronounced (fig. 4) the ore-beds at Nordic are older than those at Quirke. If the northward overlap were not disrupted by basement highs, the ore beds at Pronto, the

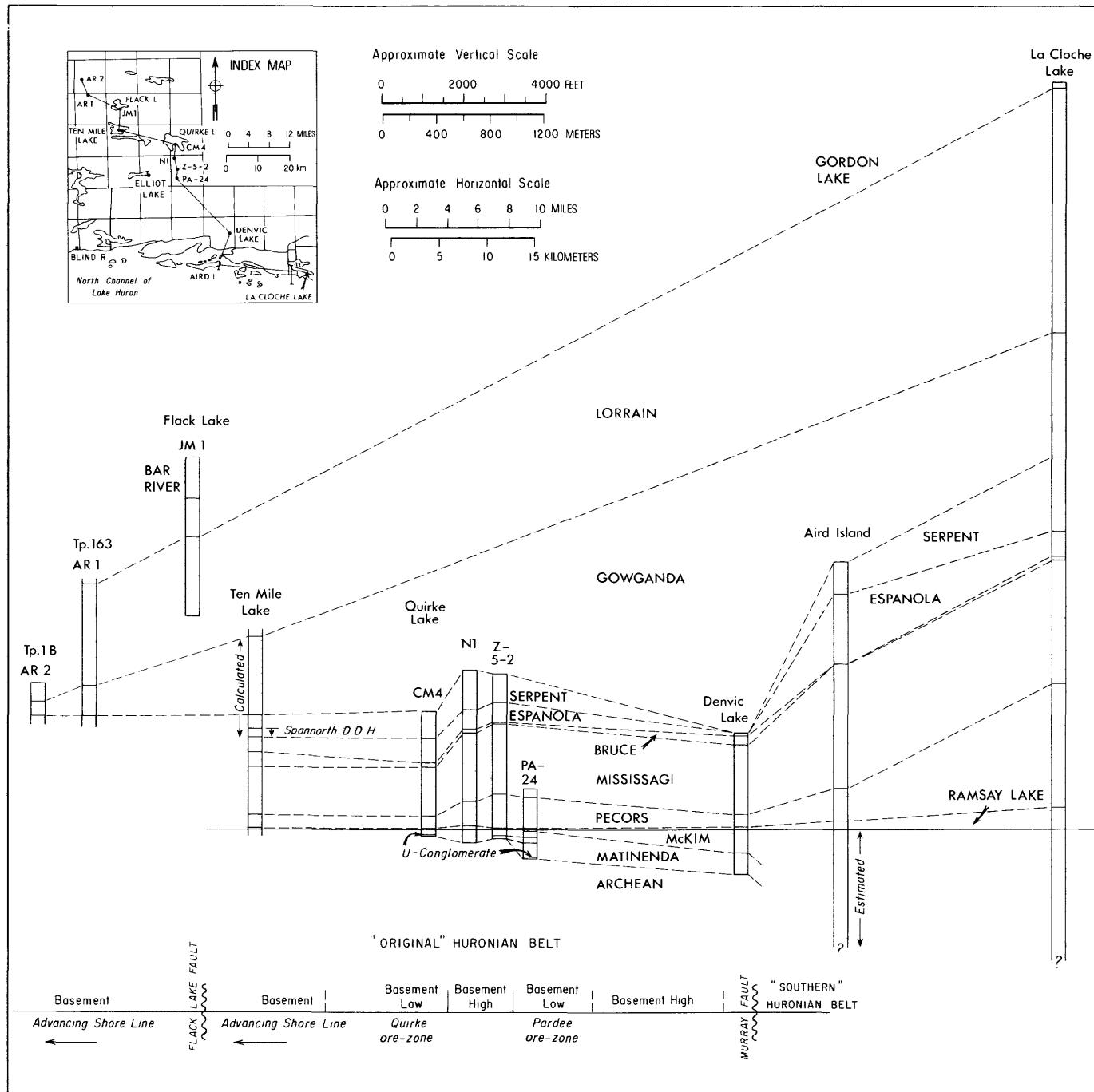


FIGURE 4.--Lateral variation of the Huronian Supergroup, Elliot Lake area.

most southerly known occurrence, would be the oldest, but in the absence of a regional marker horizon this cannot be established. The quartz clasts at Pronto are cobbles, partly sorted and moderately rounded; those at Nordic are large pebbles, well sorted and rounded; those at Quirke are small- to medium-sized pebbles, well sorted and rounded. Similar relations are seen going up sequence at both Nordic and Quirke, and packing becomes less pronounced.

#### MCKIM FORMATION

Argillaceous strata constituting the McKim Formation thicken southeasterly and form the most important formation of the Elliot Lake Group on the south limb of the Chiblow Anticline north of the Murray Fault between Sheddron Township and the east limit of the area shown on figure 2 (Robertson, 1965a, 1966a, b, 1975a). To the east of Algoma, this unit, but without any underlying arkose or Archean basement rocks, is repeated south of the Murray Fault. Near the fault, it is represented by schists of the almandine-staurolite grade, but farther south it is represented only by poorly exposed less-metamorphosed rocks. The unit south of the fault was formerly believed to be Archean in age (Collins, 1925, 1936).

#### VOLCANIC ROCKS

In the Blind River area (fig. 2) mafic volcanic rocks are found at Thessalon, Dollyberry Lake, near Nordic Mine, south of Pronto Mine, and in the vicinity of Massey. Thick volcanic assemblages have also been recognized northeast of Sault Ste. Marie (Duncan and Aberdeen Townships) and between Agnew Lake and Sudbury. Characteristically the rocks are massive and dark green to black and only locally show feldspar phenocrysts and amygdules. Ropy texture and flow-top breccias are recognised, and pillows are very rare or absent, suggesting a subaerial environment. Pillows have been observed in the upper part of the Duncan sequence (Bennett, 1975) and in the Sudbury area (Innes, 1975), suggesting transition to submarine deposition. Locally, arkose and uranium-bearing pyritic quartz-pebble conglomerate are found either interbedded in the fringe areas of the piles or between the volcanic rocks and the Archean rocks (Robertson, 1968b; Innes, 1975; Bennett, 1975). Locally at Thessalon and Massey, but better developed at Sudbury, intermediate or felsic flows are found. So far, attempts to date these suggest a date of about 2,400 m.y., but a reliable isochron needs to be established (Fairbairn and others, 1969). J. A. Robertson (1969a) has pointed out the close spatial relations to two major fault zones, the Murray and the Flack, which are also zones of marked change in sedimentation.

Innes (1974, 1975) has studied the volcanic rocks at Sudbury. These data and limited work on the rocks at Massey (Robertson, 1976) and Dollyberry Lake (J. A. Robertson, unpublished) show the rocks are subalkaline tholeiites and host iron and copper sulphide. Between Massey and Sudbury, layered gabbro-anorthosite intrusive bodies are found cutting Archean rocks; these are probably part of the same igneous cycle as the volcanic rocks and have similar chemical affinities. No age dates have been attempted.

The known uranium ore bodies are marginal to the volcanic piles, which may have physically controlled the current depositing the uraniferous conglomerates in the same way as the topographic highs reflecting basement geology.

Emanations from the volcanism may also have contributed sulphur to convert detrital magnetite to pyrite in the sediments.

#### HOUGH LAKE GROUP

The Hough Lake Group comprises the Ramsay Lake Formation, polymictic paraconglomerate; the Pecors Formation, argillite; and the Mississagi Formation, quartzite.

#### RAMSEY LAKE FORMATION

The Ramsay Lake conglomerate consists of pebbles and cobbles of grey granite, quartz, and mafic igneous rocks scattered in a dark grey greywacke-to-quartzite matrix characterised by large grains of smoky quartz and pyrite. The lithology indicates mass transportation, and an association with graded greywackes with dropped clasts suggests ice rather than mudflow as the transportation medium. A crude bedding, particularly in the south, the presence of mud cracks and ripple marks on the upper surface, and crossbedding in quartzite or greywacke lenses indicate deposition in shallow water.

On the north shore of Quirke Lake the Ramsay Lake Formation rests directly on basement (Robertson, 1961, 1968a), but in the Quirke, Denison, and Panel Mines it truncates the uraniferous horizons in the Matinenda Formation and is itself slightly radioactive. Elsewhere, the Ramsey Lake Formation truncates arkose; the basal part of the formation is both lighter in colour and more potassic than the upper part, reflecting incorporation of material from the Matinenda Formation.

The formation thickens from 0-15 ft (0-4.5 m) at northernmost outcrops to more than a thousand feet (300 meters) in the Espanola-Sudbury area (Card and others, 1972). The Ramsay Lake Formation indicates that the general climate was cold and that the uranium mineralization was syngenetic.

## PECORS FORMATION

The Ramsay Lake Formation is followed conformably by the Pecors Formation, which comprises a sequence of argillites in the Quirke Lake-Elliot Lake area, of argillites, siltstones, and quartzites on the south limb of the Chiblow Anticline, and of siltstones and argillaceous quartzite where found to the south of the Murray Fault. The transition zone from the Ramsay Lake conglomerate comprises argillite, which may include varvite with dropped clasts (Robertson, 1968a), but locally a quartzite bed may be present. The argillites are frequently ripple marked. On the south limb of the Chiblow Anticline and south of the Murray Fault, ripple marks, microcrossbedding, and slumping structures are characteristic.

The thickness ranges from 100 ft (30 m) at Quirke Lake (Robertson, 1961, 1968a) to about 1,000 ft (300 m) south of the Murray Fault (Robertson, 1976), but the formation may be missing over well-developed basement highs, and its thickness is markedly reduced over the crest of the Chiblow Anticline. This reduction is in sedimentation thickness and not due to folding. On the flank of a basement high running from Chiblow Lake to the Cutler-Massey area some of the argillaceous beds of the Pecors Formation are moderately radioactive (Robertson, 1975a, 1976, p. 46).

## MISSISSAGI FORMATION

The Mississagi Formation consists of greenish arkose on the north limb of the Quirke Syncline. Elsewhere it is normally made up of well-bedded grey quartzite, but adjacent to basement highs the yellow-green colour and increased feldspar content are again characteristic. Thicknesses range from 600 ft (180 m) at Quirke Lake to 1,500 ft (460 m) near Elliot Lake (Robertson, 1961, 1968a) and to 2,700 ft (820 m) on the south limb of the Chiblow Anticline, as exposed both north and south of the Murray Fault at Blind River (Robertson, 1964). Current direction was from the northwest, and the influence of basement topography was much diminished.

Along the north limit of the Quirke Syncline and again adjacent to the Chiblow-Cutler basement high the yellow-green arkosic phase is characterised by radioactivity. Uranium and thorium, as well as potassium, is revealed by  $\gamma$ -ray spectrometry (Darnley and Grasty, 1971). In the northern section thin pyritic quartz-pebble bands, carrying sulphide and trace uranium and thorium mineralization, are found (Robertson, 1963).

## QUIRKE LAKE GROUP

The Quirke Lake Group comprises polymictic paraconglomerate, Bruce Formation; carbonate and

siltstone, the Espanola Formation; and quartzite, the Serpent Formation.

## BRUCE FORMATION

The Mississagi Formation is overlain disconformably by the Bruce Formation - a conglomerate that consists of boulders of white granite and greenstone in a partly sorted, slightly pyritic, siliceous greywacke matrix. The Bruce Formation is probably a tillite that has been subjected to some sorting. Locally, dropped clasts have been seen in varvite lenses. The conglomerate can be traced throughout the entire district. There are marked local variations in thickness, but to the north of the Murray Fault the unit is generally less than 200 ft (60 m) thick (fig. 4). South of the Murray Fault, the conglomerate is represented by a few feet of conglomeratic grit. It is missing in places, and elsewhere there are channels of boulder conglomerate up to several hundred feet in thickness.

The close similarity to the Ramsay Lake Formation should be noted (table 1). The matrix is more siliceous and is characterised by more pyrite and the lime and soda contents are greater than in the Ramsay Lake Formation. Where metamorphosed, the pyrite is replaced by pyrrhotite, and the formation has a magnetic expression. The basal part of the formation contains reworked material from the underlying Mississagi Formation.

## ESPAOLA FORMATION

In the Blind River area the Espanola Formation consists of three units: a lower unit, characterized by limestone, the Bruce limestone; a middle unit, characterized by mudstone and greywacke, the Espanola greywacke; and an upper unit having a marked development of ferruginous dolomite, the Espanola limestone. This three-fold division is not possible in the southeastern part of the area mapped, and the formation was not subdivided either north or south of the Murray Fault (fig. 4; Robertson, 1965a, b, c., 1966a; Robertson and McCrindle, 1967). Near Espanola an upper crossbedded quartzite member is present (Card, 1976).

The limestone member consists of thinly interbedded cream-coloured limestone and siltstone. Differential weathering and drag folding give the rock a spectacular appearance. Where the unit is complete, the thickness is generally 100 ft (30 m).

The siltstone and dolomite members can be distinguished from each other only by the brown-weathering ferruginous dolomite bands in the latter. Both members are characterized by intraformational breccias, siltstone and conglomerate dikes, mudcracks, and ripple marks. These features indicate shallow-water

deposition and tectonic disturbance. Occasional quartzite beds, which are more common to the northwest, show crossbedding that is indicative of a northwesterly derivation. Where complete, in the Quirke Syncline, the thickness of the siltstone member is 300–400 ft (90–120 m) and that of the dolomite member is 150 ft (45 m).

This unit is of considerable interest, as it represents an early carbonate. The presence of carbonate and of iron suggests that the environment contained some free oxygen.

#### SERPENT FORMATION

The Espanola Formation is overlain by the Serpent Formation, a white feldspathic quartzite, which is best exposed in the northern and eastern sections of the Quirke Syncline. The maximum known thickness of the Serpent Formation is 1,100 ft (330 m). Crossbedding, and lithology and thickness changes in individual members, again indicate a derivation from the northwest. Ripple marks and mudcracks indicate shallow-water conditions. The unit is also exposed in Shedden Township north of Walford Lake and on islands in the north channel of Lake Huron at the south limit of the area mapped (fig. 2; Robertson, 1965a, b, c, 1966a, 1975b, d; Robertson and McCrindle, 1967).

The lateral variation in thickness of the lowermost formations of the Huronian is illustrated in figure 4.

#### COBALT GROUP

The Quirke Lake Group is overlain unconformably by the Cobalt Group, which, in the Blind River-Elliot Lake area, consists of the Gowganda Formation and the Lorrain Formation, the Gordon Lake Formation, and the Bar River Formation.

#### GOWGANDA FORMATION

Within the map area (fig. 2), the Gowganda Formation rests on all formations between the upper Mississagi and the Serpent; in the Mount Lake and Kirkpatrick Lake area, north of the Flack Lake Faults (Roberston, 1971b; Siemiatkowska and Guthrie, 1975), it rests on the Archean basement. Locally, the contact can be seen truncating the bedding of the underlying formations, and consolidated or partly consolidated fragments of the underlying rocks are found in the lowermost beds of the Gowganda Formation; elsewhere, the contact is a "soft-sediment" contact, and in yet other localities it is a knife-sharp contact.

The Gowganda Formation is a heterogeneous assemblage of conglomerate, greywacke, quartzite, and argillite. These rock types are found throughout the sequence, although the lower part is characterized by

boulder conglomerate in which red granitic clasts predominate, and the upper is characterized by quartzite and argillite. Within the Quirke Syncline, as mapped by the writer, the Gowganda Formation is about 1,700 ft (510 m) thick (Robertson, 1963). To the west, in areas mapped by Frarey (1959, 1962), it is at least 3,000 ft (900 m) thick. To the north near Mount Lake, the formation lies on basement rocks and is less than 500 ft (150 m) thick (Wood, 1968a, b, 1975). To the south in the La Cloche Syncline, the Gowganda Formation is some 4,200 ft (1,260 m) thick (Robertson and McCrindle, 1967) at La Cloche Lake and in the Willisville-Espanola area (Casshyap, 1966, 1968). The conglomerate matrix is fine grained, chloritic, and characterized by high soda content relative to the lower Huronian polymictic conglomerate (fig. 5).

The origin of the Gowganda Formation has been much discussed. Dense boulder conglomerates, quartzites, and argillites were definitely water laid; varved conglomerates and greywackes probably formed under conditions characterized by alternate freezing and thawing, although some authorities would ascribe these rocks to turbidity currents; and sparse boulder conglomerates with a disrupted greywacke matrix may be either tillites or mudflow deposits. Ovenshine (1965a, b) has described the structures of the Gowganda Formation of the Elliot Lake-Blind River area and has endorsed a glacial environment, particularly on the basis of numerous dropped clasts in varved greywackes. In the southeast part of the area shown in figure 2 and in the areas to the east mapped by Card (1967a-d) and Casshyap (1966, 1968), a marine rather than a continental glacial environment is indicated (Lindsey, 1966). Some clasts are

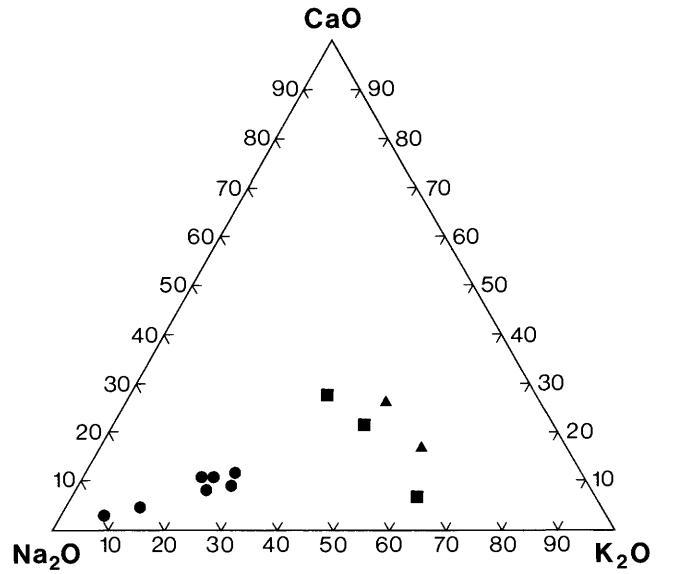


FIGURE 5. - Alkali content, Huronian polymictic conglomerates.

striated, but J. A. Robertson (1971a) has questioned the use of this feature as a criterion for glacial action (Lindsay and others, 1970) and suggests that the striae are due to postdepositional tectonism, as described by Bielenstein and Eibacher (1969). Of particular interest is the presence of iron oxide in the Gowganda Formation. The arkosic quartzites are red to pink in colour, reflecting the preservation of hematitic dust in the feldspar and interstitial material. Some thicker beds of argillite or siltstone contain magnetite and zones of such material can be traced on regional magnetic maps. No significant uranium anomalies have been identified, but scattered thorium and potassium values (Richardson and others, 1975) reflect the granitic content of the conglomeratic lower portion of the formation. The lack of a preserved regolith at the Archean surface (Robertson 1970c; Wood, 1970a) may indicate oxidizing rather than the reducing conditions that prevailed during the deposition of older lower Huronian sediments elsewhere in the district (Robertson, 1969a; Roscoe, 1969; Wood, 1970a).

#### LORRAIN FORMATION

The Lorrain Formation conformably overlies the Gowganda Formation. In the Flack Lake area several units have been mapped within the Lorrain Formation in the following ascending stratigraphic order: (1) pink ferruginous quartzite and minor siltstone, (2) coarse-grained green arkose, (3) pink coarse hematitic arkose with radioactive (thorium:uranium  $>10:1$ ) quartz-pebble conglomerates (Robertson 1970c; Robertson and Johnson, 1970); (4) interbedded pink and buff quartzite and, in a few places, greenish quartzite (quartz-chert-jasper pebble bands are characteristic of the upper part of this member), (5) massive white quartzite, with quartz-chert-jasper pebble bands in the lower part. Sericite, kaolinite, pyrophyllite, and diasporite are found in the nonfeldspathic beds of members (4) and (5) (Chandler and others, 1969) and indicate the onset of warm rather than frigid conditions (Wood, 1970a; Young, 1970). Crossbedding is common in the Lorrain Formation in the Flack Lake-Mount Lake-Rawhide Lake area but is variable; southwesterly current directions have been interpreted by Hadley (1969) and by the writer, although the source area was probably to the north. North of Bruce Mines, Hadley (1969) deduced southeasterly current directions. The five members can be traced throughout the Flack Lake-Mount Lake-Rawhide Lake area (Robertson, 1971b; Wood, 1975; Siemiatkowska and Guthrie, 1975). Their equivalents can also be recognized and traced in the La Cloche Lake-Whitefish Falls area (Card, 1971a; Chandler, 1969), but the individual units are both finer grained and thicker, indicating deposition farther from source. They have

been strongly folded, foliated, and somewhat metamorphosed; the clay minerals are represented by kyanite and andalusite (Church, 1967; Chandler, 1969; Robertson, 1970b, 1976).

#### GORDON LAKE FORMATION

The Lorrain Formation is overlain, apparently conformably, by the Gordon Lake Formation, a 1,000-ft (300-m) sequence of well-bedded siltstone, argillite, chertstone, and fine- to medium-grained sandstone. There are three members: (1) a lower member comprising reddish sandstone and siltstone with anhydrite and gypsum nodules (and salt casts?), (2) a middle member of dark green siltstone, argillite, and minor sandstone, and (3) an upper member of reddish siltstone, argillite, and chert (Eibacher and Bielenstein, 1969; Bottrill, 1970; Robertson, 1969b, 1970c). The middle member tends to form a scarp in the Flack Lake area that corresponds to a moderate magnetic anomaly (Robertson, 1971b). Current ripples, microcrossbedding, slumping structures, and desiccation cracks indicate deposition in very shallow water (Young, 1969; Robertson, 1969b, 1970c; Robertson and Johnson, 1969, 1970; Wood, 1970a, b). In the core of the La Cloche Syncline a few hundred feet of siltstone and quartzite that have been correlated with the Gordon Lake Formation also coincide with a magnetic anomaly (Robertson, 1976).

#### BAR RIVER FORMATION

The Gordon Lake Formation is overlain by the Bar River Formation, which comprises at least 1,200–1,500 ft (360–450 m) of massive to well-bedded orthoquartzite, generally crossbedded and ripple marked (Robertson, 1969b; Robertson and Johnson, 1969; Woodward, 1970; Eibacher and Bielenstein, 1969). Muderacks and desiccation features are also found, particularly in finer grained sandstones and intercalated siltstone bands. One unit is characterized by thin bedding and ferruginous siltstone and quartzite. The iron has been partly redistributed, and small-scale solution depositional fronts adjacent to joints and fractures are common. Some ripple-marked surfaces at this horizon are covered with segmented, tapering, sinuous structures of possible organic origin (Hoffman, 1967; Young, 1967; Donaldson, 1967). Young (1967) and Hoffman (1967) favoured metazoans (worms) as the organism involved, but Donaldson (1967) suggested that the structure represents the infilling of desiccation cracks in algal mats. In the same area there are many rather similar structures that are undoubtedly desiccation features. The desiccation features of the Gordon Lake Formation (Young, 1969; Robertson, 1969b) are so similar to the most organic-looking structures of the Bar River For-

mation that the organic nature of the latter remains in doubt. Wood (1970a, b) has recorded oolites in the ferruginous rocks, and their presence along with the many primary structures indicated deposition in very shallow water. In the ferruginous rocks Bottrill (1970, personal commun.) has observed pyrite and in polished section has identified the major iron mineral as maghemite rather than hematite. The conditions of deposition, sedimentation, climate, and state of atmospheric oxidation represented by the upper Huronian rocks of the Flack Lake are discussed in more detail by Wood (1971).

#### SUMMARY OF HURONIAN

The Huronian rocks of the area thus comprise thick sequences of clastic sediments and minor tholeiitic basalts. The sediments were derived from the adjacent Archean Craton and were deposited as migrating diachronous facies with cyclical rejuvenation. The depositional basin extended east-west and deepened towards the southeast. The distribution of volcanic rocks and marked changes in thickness and facies of sedimentary units were controlled by hinge zones, which later became regional fault zones. Minor changes in composition of similar rock types are due to shifts in provenance and to the efficacy of weathering and winnowing processes. Radioactivity and heavy minerals are characteristic of quartz-pebble conglomerates in the fluvial near-shore facies. In the lower Huronian the association is of uranium and sulphides, predominantly pyrite, and in the upper Huronian it is of thorium and iron oxides. In the middle Huronian conditions became favourable for preservation of iron oxides. Glacial deposits are characteristic of the lower and middle Huronian, whilst the uppermost Huronian was apparently deposited in a dry and hot environment. Figure 5 illustrates the alkali content of the matrices of the Huronian polymictic paraconglomerates; in figure 6 three compositional ratios for Huronian argillaceous rocks are shown. Of particular interest in figure 5 is the variation in maturity of the paraconglomerate matrices, which decreases towards the Gowganda Formation, the marked increase in soda, and, in the lower part of the paraconglomerate sequences, the incorporation of potassic material from the underlying arenite. Figure 6 shows an increase in ferric/ferrous iron ratio in the argillaceous rocks, reflecting change in general oxidation conditions; an increase in the  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio, reflecting increased efficacy of chemical weathering in the upper Huronian; and variations in the  $\text{Na}_2\text{O}/\text{K}_2\text{O}$  ratio, reflecting changes in provenance from a granitic to a granodioritic-greenstone terrane. The arenaceous rocks show similar changes in provenance and efficacy of chemical weathering. The role of oxidation is apparent in the frequency of red beds from the Gowganda Formation up-

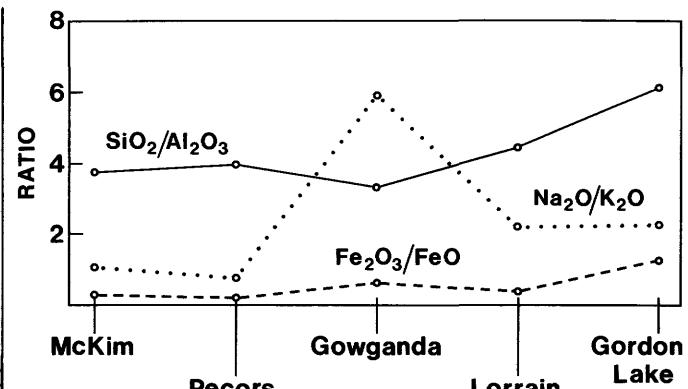


FIGURE 6.—Silica/alumina, ferric iron/ferrous iron, and soda/potash ratios for Huronian argillaceous rocks.

wards contrasted to the total lack of red beds in the lower formations.

#### POST-HURONIAN EVENTS

Following the close of sedimentation, the area was subjected to a long history of structural deformation, igneous intrusion, and, in the Penokean fold belt, regional metamorphism, which locally attained amphibolite facies (Card, 1964; Card and others, 1972; Robertson, 1972). These events do not concern the formation of the uranium deposits. The oldest intrusion, the Nipissing diabase, took place at  $2,155 \pm$  m.y. ago (Van Schmus, 1965), and this serves to place a minimum age on the Huronian sedimentary rocks. Alteration (albitization, chloritization, and carbonatization) associated with dikes or sills has locally affected the uranium deposits, which were clearly in place prior to the intrusion (Robertson, 1968a, 1970a).

In general, deformation and metamorphism of that part of the Blind River area in which uranium ores are found is minimal. The lack of intense metamorphism may be regarded as a factor in the preservation of the ore bodies. Age data on minerals, however, reflect some resetting of the decay clocks at times corresponding to known peaks of regional metamorphism or thermal events rather than episodic introduction of more uranium (Roscoe, 1969).

#### THE URANIUM DEPOSITS

Uranium and thorium-uranium deposits are found in conglomerates at a number of localities in the Blind River-Elliot Lake and Agnew Lake areas, as well as throughout the Huronian belt (Robertson, 1968a, 1975a; Thomson, 1960).

### DISTRIBUTION OF URANIUM ORE DEPOSITS—BLIND RIVER-ELLIOT LAKE

The workable uranium deposits of the Blind River camp are found as quartz-pebble pyritic conglomerate beds in zones controlled by basement topography (fig. 7). In the Quirke Syncline the relation of the uraniferous conglomerates to granite-greenstone contact areas and valleys over softer zones in the greenstone belt is clearly demonstrated (fig. 7). At Pronto, however, there is no clear relation to basement geology, which raises the possibility that there may be other economic uranium deposits underlain by granite. It is clear, however, that Pronto is located on the flank of a regional basement high.

The ore zones strike northwest-southeast and are controlled by basement structures. The Quirke zone (the largest in the area) is 32,000 ft (9,600 m) long and from 6,000 ft (1,800 m) to 9,000 ft (2,700 m) wide, and the Nordic zone is 19,000 ft (5,700 m) long and from 4,400 ft (1,320 m) to 6,000 ft (1,800 m) wide (Robertson, 1967, 1968b). The Pronto deposit (Robertson, 1970a) and the unworked zones are of smaller dimensions.

At Quirke No. 1 the main ore zone is approximately 30 m (100 ft) above basement and is approximately 3.5 m (12 ft) thick. Toward the east the ore zone is truncated by an unconformity at the base of the Ramsay Lake conglomerate.

At Quirke No. 2 and Denison, the best ore development is in the Denison Reef some 30 m (100 ft) below the Quirke Reef. The Denison Reef normally comprises two conglomerate zones each 1.8–3.6 m (6–12 ft) thick separated by barren arkose 0.6–2.4 m (2–8 ft) thick. Throughout much of the Denison Mine, ore grade was sufficient to permit mining of both conglomerate beds and the intervening quartzite as one unit using large-scale equipment and trackless haulage. Quirke No. 2 has been developed using conventional haulage. Other conglomerate beds 0.6–3 m (2–10 ft) thick separated by quartzite beds 3.6–6 m (12–20 ft) thick are known on both the Quirke and Denison properties, but these have not been mined. At Stanrock Mine another reef was found under the Denison Reef in the southeastern part of the mine. The en echelon pattern with the oldest reef to the southeast conforms to the regional overlap pattern. The nomenclatures used in the various mines of the Quirke zone and their stratigraphic relations are illustrated schematically in figure 8.

At the Nordic Mine the main ore bed comprised conglomerate or conglomerate with quartzite over a width of 3 m (10 ft) with a grade of 2.5 lbs  $U_3O_8$  per short ton (1.25 kg per metric ton). Locally another reef lower in the sequence, the Lacnor Reef, was mined where grade attained 2.0 lbs  $U_3O_8$  per short ton (1 kg per metric ton). In the eastern part of the mine a third reef, the Pardee

Reef, attains ore grade, 2.3 lbs  $U_3O_8$  per short ton (1.15 kg per metric ton) over 5 ft (1.5 m). This reef extends eastward over a basement ridge into the Pardee and Pecors mineralized zones (fig. 7). These reefs extend down rake to the Stanleigh Mine. Operations were largely carried out over the Lacnor and Nordic Reefs. As with the Quirke zone, other reefs of conglomerate of submarginal grade or minor extent are known.

Five types of boundary to the ore beds are known:

1. Outcrop of the conglomerate bed.
2. Wedging due to contact with either regional or local basement "highs". To the west of Quirke No. 1 the "basement" may be the edge of a pile of Huronian volcanics rather than Archean.
3. Lateral thinning of conglomerate and thickening of the intervening quartzite beds accompanied by drop in grade of the radioactive units. This is probably the typical boundary and the definition is arbitrary.
4. Removal by erosion subsequent to deposition, as for example in Quirke, Denison, and Panel, where there is an unconformity at the base of the Ramsay Lake conglomerate. Where material from conglomerates of the Matinenda Formation has been incorporated in the Ramsay Lake conglomerate, the latter contains anomalous but minor amounts of uranium radioactivity.
5. Fault contact. Locally, areas were not mined either because of unfavourable mining conditions caused by faults, extensive fracturing, or diabase dikes or because of unfavourable milling conditions caused by albite, chlorite, or carbonate alteration.

Some of these relationships are illustrated in figure 9, a cross section through the New Quirke (Quirke No. 2) Mine of Rio Algoma Mines, Ltd.

### LITHOLOGY AND MINERALOGY OF URANIUM ORE DEPOSITS

The typical ore-bearing conglomerates of the Matinenda Formation consist of well-rounded, well-sorted quartz pebbles in a matrix of quartz, feldspar, and sericite and have an average pyrite content of 15 percent. Monazite and zircon are characteristic heavy minerals. Brannerite and uraninite are found in the matrix. Thucholite is found locally and may line fissures in the ore beds. The ore minerals are brannerite, uraninite, and monazite (Robertson, 1960 and later; Roscoe, 1957 and later; Pienaar, 1963; Roscoe and Steacy, 1958).

Thucholite is present in the ore but is also in fractures as a postore secondary mineral. Almost no work has been done on thucholite at Blind River. The recent interest in hydrocarbon at the Rand has prompted Ruzicka

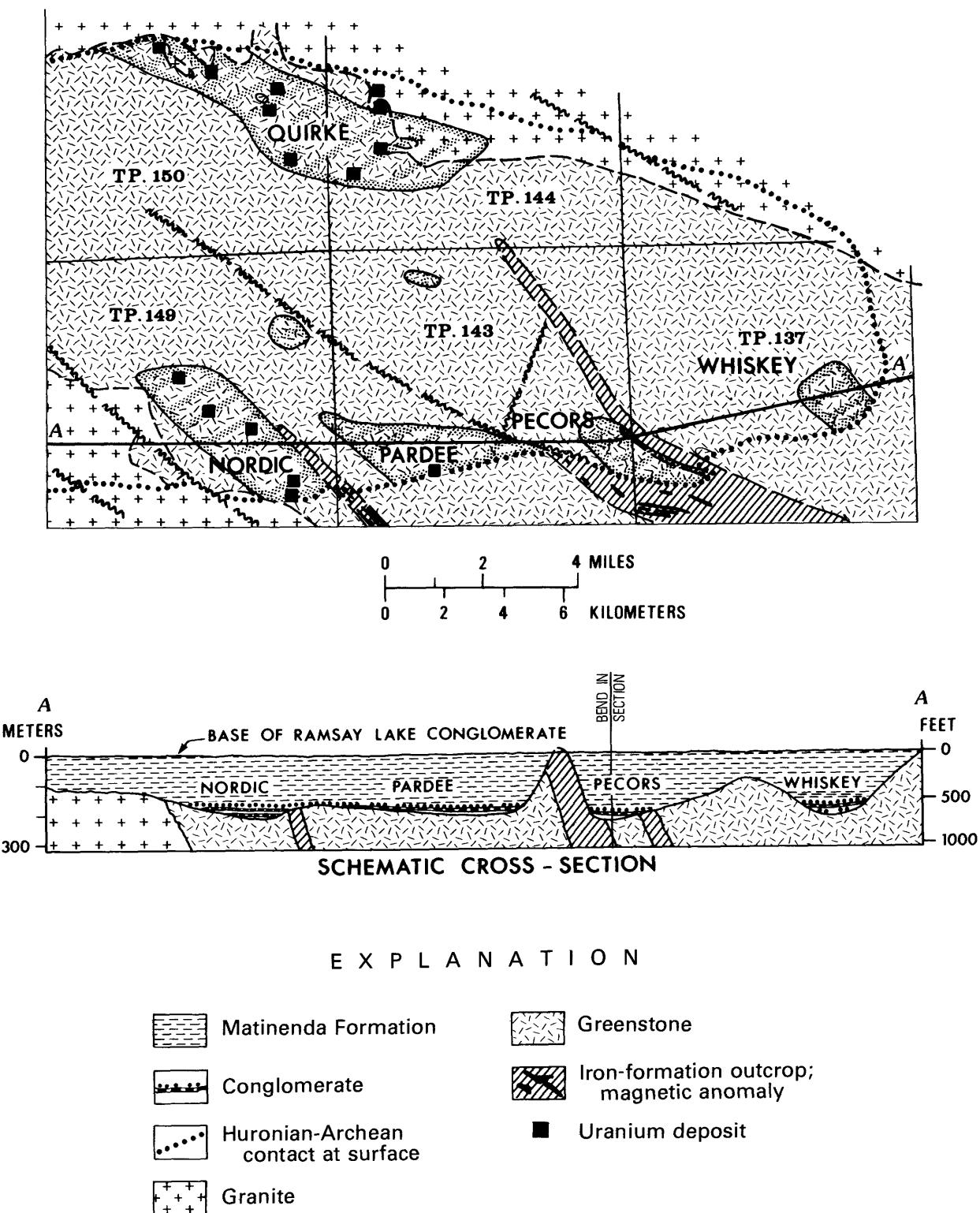


FIGURE 7. Uranium deposits in the Quirke Syncline.

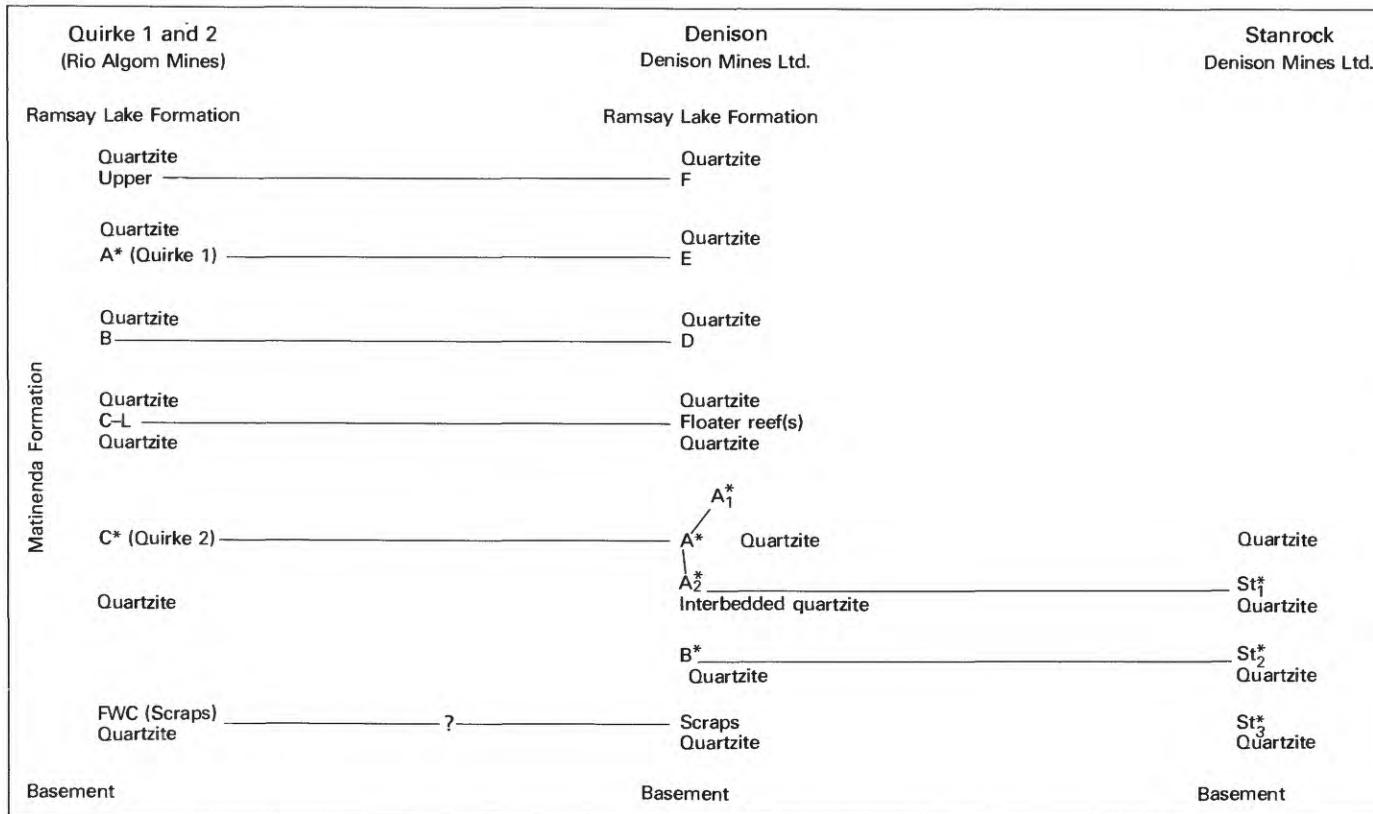


FIGURE 8.—Correlation and nomenclature of quartz-pebble conglomerate reefs, as used in the mines of the Quirke zone. Asterisks indicate beds having supported production.

to initiate comparative studies on Blind River material; an initial progress report is included in Ruzicka's paper in this volume (chap. V).

Gummites (soddyite and uranophane) are rare, but Rice (1958) stated that they form a significant proportion of the ore mineralization at the Spanish American Mine. Uranothorite has also been identified (Roscoe and Steacy, 1958, p. 14) and Patchett (1959, 1960) identified coffinite in altered material from the Nordic Mine.

Pyrite is the commonest sulphide mineral; it usually constitutes 10–15 percent of the matrix, but it may rarely be as much as 30 percent. Pyrite is concentrated in the matrix, and only rarely is there indication of replacement or fracture filling in the quartz pebbles. Individual grains may be rounded ("buckshot" pyrite) or subhedral to massive. R. G. Arnold (1954) suggested that the pyrite formed by sulphidization of detrital magnetite, and he described grains showing cores rich in leucoxene that he considered to have developed from ilmenite exsolved from the original magnetite. More recently, Bottrell (1971) has suggested that the sulphur required was derived from the lower Huronian volcanic rocks. However, no laboratory studies have been undertaken

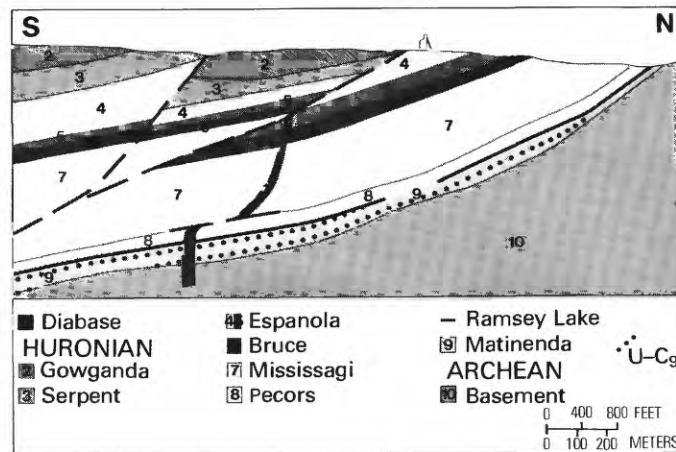


FIGURE 9.—Cross section, New Quirke mine.

to test this hypothesis. Pienaar (1963), in a trace-element study, could not distinguish between the pyrite of the ore beds and the pyrite found in other rocks of the district.

Other sulphide minerals found are pyrrhotite (occasional scalenohedral pyrrhotite crystals have been found

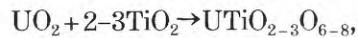
in vuggy cavities), chalcopyrite and minor arsenopyrite, galena (probably radiogenic), cobaltite, and marcasite, molybdenite, and sphalerite as coatings in late fractures (often with thucholite). At Denison Mine, tourmaline crystals were observed in a late quartz veinlet. The following detrital heavy minerals have also been identified: Anatase, amphibole, barite, apatite, cassiterite, chromite, diopside, garnet, epidote (allanite?), gold, fluorite, hematite, ilmenite, magnetite, monazite, rutile, scheelite, sphene, spinel, tourmaline, xenotime, yttriofluorite, and zircon, partly as malacon and more rarely as cyrtolite (Abraham, 1953; Arnold, 1954; Holmes, 1956; Milne, 1959; Patchett, 1960; Pienaar, 1958, 1963; Ramdohr, 1957, 1958a, b; Rice, 1959; Roscoe, 1957; Roscoe and Steacy, 1958, p. 13-15; Thorpe, 1963; Canadian Institute of Mining and Metallurgy, 1957).

In addition, Milne (1959), in samples collected for Patchett (1960) at the Nordic Mine, identified chamosite, greenalite, and grunerite. These iron silicates were probably derived from the ridge of iron-formation forming the eastern boundary of the Nordic channel (fig. 7).

Although a wide variety of detrital minerals have been identified and described by the authors listed, it should be stressed that these occur in very small quantities and are not necessarily present in all samples that have been studied.

There has been much discussion and description of the main minerals in the ore, mainly from polished-section work. The similarity to the gold- and uranium-bearing bankets of the Witwatersrand (Ramdohr, 1958a, b), radioactive conglomerate at Jacobina in Brazil (Bateman, 1958; Gross, 1968), and some deposits in Australia and Russia has been stressed in the literature (Davidson, 1957; Derry, 1960).

The major ore mineral at the Pronto Mine and in the Quirke Mine A Reef (Theis, 1973) is brannerite, first described from the area by Nuffield (1954). Although some relatively fresh material has been observed (Rice, 1958), brannerite is typically found as ovoid red-brown to black grains in the metamict state showing bladed rutile surrounded by a uranium oxide and rare-earth oxides, the two-phase uranium-titanium compound of Pienaar (1958, 1963). Pienaar (1958, 1963), Roscoe (1969), and Robertson and Steenland (1960) suggested that the rounding is due to transportation and that the material is detrital. Ramdohr (1957) has suggested that the brannerite rather than decomposing is authigenic and has proposed the "Pronto Reaction,"



which he held took place during metamorphism. Experimental efforts to reproduce this reaction indicate that it takes place at temperatures in excess of those to which the rocks have been subjected (Gruner, 1959, p.

1315; Patchett, 1959). The brannerite generally contains small inclusions of pyrrhotite and of radiogenic galena (Thorpe, 1963, p. 39). Table 4 (from Robertson, 1968b) lists the published partial analyses of brannerite from the Blind River area along with the regional value selected by Thorpe.

Blind River brannerite has been recently studied by Ferris and Ruud (1971) and by Theis (1973). These authors concluded that Blind River brannerite, and the brannerite or uraniferous leucoxene of the Rand, formed at low temperatures during diagenesis as a result of uranium migrating to decomposing ilmenite.

Uraninite is the second most important mineral at Pronto and apparently the most important in the Nordic zone and in the C Reef at Quirke Mine (Theis, 1973). Generally it occurs as black subhedral grains approximately 1 mm across. Ramdohr (1958a, b) and D. S. Robertson (1962a, b, 1974) have described rounded grains. Derry (1960) and Thorpe (1963) have both selected 6 percent as the regional value of  $\text{ThO}_2$  content. Table 5 (from Robertson, 1968b) summarizes the early published data on Blind River uraninite, since confirmed by Grandstaff (1974) and reported in this volume (chap. J). This corresponds to the composition of pegmatic rather than hydrothermal uraninite. The rounding and thorium content may indicate a detrital origin. Rice (1958), however, believed that uraninite formed as a result of leaching from brannerite by hydrothermal solutions emanating from diabase; diabase is more common in the Nordic Mine than elsewhere. Patchett (1960) also supports a secondary origin for uraninite and stated that the cores are carbon-rich. Davidson (1957, 1965) has used the general lack of uraninite in modern placers and geochemical principles along with the Lyellian concept of uniformitarianism (actualism) as argument against the detrital origin of uraninite.

Roscoe (1959b) has described the monazites from the Blind River ores and has pointed out that monazite can contain considerable uranium and is, therefore, one of the ore minerals. Grains are normally rounded to subangular and less than 0.3 mm in diameter. Grey varieties of monazite are strongly radioactive, may contain uranothorite or thorite (Roscoe, 1959b; Patchett, 1959), and have pyrite inclusions. Table 6 (from Robertson, 1968b) gives Roscoe's (1959b) uranium-thorium analyses of monazites and the values selected by Thorpe (1963). Roscoe has shown (1959a, b) that the uranium-thorium ratios in monazite are comparable to that of the basement. The lateral variation in the ore-mineral and uranium-thorium ratios, as studied by Roscoe (1959a, b), D. S. Robertson (1962, 1974), J. A. Robertson (1968a), and Thorpe (1963), are best explained by the relative stability of monazite during transportation; monazite is the chief radioactive mineral in the occurrences in the Lorrain Formation.

TABLE 4.—Uranium, thorium, and titanium contents, in percent, of *Blind River brannerite*

[From Robertson (1968a, p. 92)]

Mine	Author	Colour	Comments	$U_3O_8$	$ThO_2$	$TiO_2$	$U_3O_8/ThO_2$ ratio
Others than Blind River.	Thorp (1963)			40-52	0.3-15	32-40	40-52
Nordic	Patchett (1960)	Reddish brown	Altered	32.6	2.6	30.3	12.5
Do	do	Darker	More glassy, less altered.	32.2	1.8	30.5	17.9
Denison	Roscoe (1959a)	Black		41	6.1	20	6.7
Do	do	Dark brown		36	1.8	30	20.0
Quirke	do	do		24	1.7	37	14
Do	do	Brown		20	2.2	30	9.1
Do	do	Cream		6	1.2	27	5.0
Can-Met	Pienaar (1963)	Veinlet material		8.70	1.14	n.d.	7.6
Regional	Thorpe (1963)			30	2.3	30	13

TABLE 5.—Uranium and thorium contents in *Blind River uraninite*

[From Robertson (1968a, p. 93)]

Mine	Author	$U_3O_8$	$ThO_2$	$U_3O_8/ThO_2$
Denison	Pienaar (1963)	48.4	5.6	8.65
Do	do	39.0	7.2	5.4
Do	do	55.5	5.4	10.3
Nordic	Patchett (1960)	55.25	6.25	8.85
Quirke	Wanless and Traill (1956)	51.7	4.1	12.6
Pronto	do	57.0	5.12	11.1
Nordic	Roscoe (1959a)	64	6.8	10.15
Denison	do	60	5.9	10.2
Panel	Thorpe (1963)	<sup>1</sup> 29.6	<sup>1</sup> 3.84	<sup>1</sup> 7.7
Regional	do	60	6	10

<sup>1</sup> Contaminated?

Locally, individual conglomerate bands may assay as high as 20 lbs or more  $U_3O_8$  per short ton (10 kg per metric ton), but over mining widths of the order of 9-30 ft (3-9 m) the average grade of those beds that are, or have been, mined is 2-3 lbs  $U_3O_8$  per short ton (1-1.5 kg per metric ton). The fringe areas to mined beds, the lateral extension to the Quirke zone, and the Pardee zone, however, contain considerable amounts of material with average grades of 0.5-1.5 lbs (0.25-0.75 kg).

The arkose interbedded with the ore conglomerate is generally greenish in colour and is crossbedded (Roscoe 1966; Robertson, 1968a, b, 1975a). Normally this crossbedding is of the festoon type, indicative of a fluvial environment.

#### AGNEW LAKE AREA

The deposits at Agnew Lake (Little and others, 1972), currently under development, comprise gritose oligomictic conglomerate interbedded with arkose in a quartzite sequence probably equivalent to the Matinenda Formation. The pebbles are smaller and more sparse than in the Blind River ore, and pyrite is less conspicuous than at Blind River-Elliot Lake. Thorium-to-

TABLE 6.—Analyses of *Blind River monazites*

[From Robertson (1968a, p. 93)]

Colour	Roscoe (1969b)				Thorpe (1963)	
	1	2	3	4	Range	Average
Orange						
$ThO_2$	5.9	3.9	6.7	5.9	5.6-6.2	5.6
$U_3O_8$	.31	.95	1.65	2.9	1-2	1.4
$U_3O_8/ThO_2$	.0525	.244	.246	.492	.18-.32	.25

uranium ratios are higher than at Blind River-Elliot Lake, reflecting the presence of uranothorite and monazite as the dominant radioactive minerals. Post-Huronian deformation is considerably more intense, resulting in steep dips and in development of cleavage and stretched pebbles. Little and others (1972) estimated the reserves at 8,000 short tons (7,250 metric tons)  $U_3O_8$  in material grading 1.88 lbs  $U_3O_8$  per short ton (0.940 kg per metric ton).

#### COBALT EMBAYMENT

That part of southern Ontario lying north of Sudbury is known as the Cobalt Embayment, it should be noted, however, that neither thrust faulting nor plate tectonics have been recognised as significant factors in the regional geology. Lower Huronian rocks are intermittently exposed in the southern portion of the Cobalt Plate but are largely concealed by Nipissing diabase and upper Huronian rocks. The regional geology has been discussed by Thomson (1960) and by Meyn (1972). Precise stratigraphic correlation with Blind River is unclear, but rocks equivalent to the Mississagi Formation are present. At the base of these, wherever exposed, there are anomalous concentrations of uranium and, at some localities, gold in conglomerates and uranium in argillaceous quartzites.

Early descriptions of the conglomerates (Thomson, 1960; Meyn, 1972; Robertson, 1968a) compared these rocks with the Blind River oligomictic conglomerates. On closer examination, however, this comparison is invalid. Many of the conglomerates are polymictic, con-

taining fragments of granite, greenstone, and iron-formation sparsely distributed in an argillaceous quartzite matrix. Many of the so-called quartz pebbles are actually quartzite of unknown provenance or iron-formation similar to that of nearby Archean greenstone belts. Outcrops of these rocks in Vogt Township and Turner Township reportedly contain trace amounts of gold (Robertson, 1968b).

These deposits are clearly derived from Archean terrane but are of a provenance different from those at Blind River and Elliot Lake. Although these rocks are perhaps slightly younger, they must be considered part of the same metallogenic province. Additional prospecting and exploration in the southern part of the Cobalt Embayment should yield more data of stratigraphic and, hopefully, of economic value.

#### PRODUCTION

Between 1955 and 1973 the Blind River-Elliot Lake camp has produced, from 12 mines, approximately 1.5 billion dollars worth of uranium, and minor amounts of thorium and yttrium, from material grading 2 lbs  $U_3O_8$  per short ton (1 kg per metric ton). The maximum production was in 1959, when 12,150 tons (11,020 metric tons)  $U_3O_8$  were produced. In 1973, two mines were operating: the Denison Mine produced 1,712 short tons (1,552 metric tons)  $U_3O_8$  from ore grading 2.57 lbs  $U_3O_8$  per short ton (1.28 kg per metric ton), and the New Quirke Mine produced 2,409 short tons (2,185 metric tons)  $U_3O_8$  from ore grading 3.4 lbs  $U_3O_8$  per short ton (1.7 kg per metric ton) (1973 Annual Reports for Denison Mines Limited and Rio Algom Mines Limited). The Agnew Lake Mine north of Espanola has a developed ore body, but the decision to start production would require a suitable sales contract.

Ontario's current production is more than 4,000 short tons (3,600 metric tons)  $U_3O_8$  per year, but the known deposits can support a production of between 11,000 and 14,000 short tons (10,000 and 12,700 metric tons)  $U_3O_8$  per year, which can be obtained by expanding operating plants, reopening closed plants, and some construction of new plants.

#### RESERVES

It was estimated (Robertson, 1975a) from data available to the public in January 1973, that Ontario deposits, predominantly the sedimentary deposits of Blind River-Elliot Lake-Agnew Lake areas, contain approximately 200,000 short tons (180,000 metric tons) of recoverable  $U_3O_8$  from material with a millhead grade of 1.8 lbs  $U_3O_8$  per short ton (0.90 kg per metric ton) and an additional 150,000 short tons (136,000 metric tons) from material with a millhead grade of 1.4 lbs  $U_3O_8$  per short

ton (0.70 kg per metric ton) and a cutoff grade of 1 lb  $U_3O_8$  per short ton (0.5 kg per metric ton) over mining widths. The higher grade uranium reserves also contain at least 100,000 short tons (90,000 metric tons) of recoverable  $ThO_2$ . The higher grade material constitutes some 75 percent of the free world's reserves recoverable at less than \$10.00 U.S. (1973) per lb  $U_3O_8$  (Little, 1974; Williams and Little, 1973; Organization for Economic Co-operation and Development, 1973). Estimated additional ore (possible ore and prognosticated ore) in the localities of the Blind River-Elliot Lake-Agnew Lake areas may be as much again, but extensive and costly exploration must be undertaken before existence of this material can be confirmed and it can be considered part of the reserves.

#### ORIGIN OF THE URANIUM DEPOSITS OF BLIND RIVER TYPE

The following discussion is largely taken from earlier summaries by the author (Robertson, 1968a, 1969a).

Uranium and thorium mineralization occurs at a number of localities throughout the world in quartz-pebble conglomerates bearing appreciable pyrite and, especially at the Witwatersrand in South Africa, gold in the matrix. The origin of these conglomerates has been much debated. The similarity of the deposits at Blind River to one or another of several of the well-known deposits at Witwatersrand, South Africa, and Jacobina, Brazil, have been pointed out by Bateman (1958), Davidson (1957, 1965), Derry (1960, 1961), Joubin (1960), and Gross (1968). Davidson also mentions similar deposits in Australia and Russia. This characteristic assemblage and its distribution is a major theme of this volume. The relation to major unconformities, particularly those marking the Proterozoic-Archean boundary, has been emphasized (Derry, 1961; Davidson, 1965; Robertson, 1960 and later).

In the Blind River camp this unconformity can be placed at 2,500 m.y. ago; it is clear that the conditions did not persist beyond a minimum of 2,155 m.y. ago and that the period 2,500–2,000 m.y. ago seems the most favourable.

Bateman (1955, p. 371), Joubin and James (1957), Davidson (1957, p. 668), and Heinrich (1958) have cited as evidence of a hydrothermal origin the supposedly high uranium-to-thorium ratios, the high titanium-to-iron ratio, and the association of titanium, cobalt, nickel, thorium, and uranium in a deposit carrying the characteristic minerals gold, brannerite, uraninite, and pyrite. Patchett (1960), after a detailed study of only three samples from the Nordic Mine, regarded the ores as epigenetic. Joubin (1954) suggested the "Keweenawan" diabase as a source but in a later paper (1960) admitted that mining evidence clearly indicated

that the diabase postdated the uranium mineralization. Davidson (1957) suggested that the (supposedly) post-Huronian granite lying to the southeast was the probable source; however, much of the granite formerly considered to be of possible post-Huronian age and shown as such on the Lake Huron Sheet (Canada Geological Survey, 1933) has since been proved to be older than the Huronian (Robertson, 1972; Card and others, 1972). Only the Cutler granite, exposed south of the Murray fault 20 miles (32 km) south of Elliot Lake, is now considered to be of post-Huronian age (Robertson, 1964; Van Schmus, 1964b, 1965).

In 1965, Davidson (1965) returned to the question of the origin of blanket ore bodies and considerably modified his earlier views. The revised hypothesis may be summarized as follows:

1. Deposition of molasse-type sediments in deep basins with conglomerate near the basal unconformity
2. Leaching of the metal content (for Blind River uranium and thorium) by groundwater
3. Prolonged series of intrastratal migration, allowing mineralized groundwater to sink to the lowest permeable horizons, the oligomicitic conglomerate beds, where the metals would be reprecipitated
4. The thermal energy for cycling groundwater would be derived from postsedimentation intrusions. For the Blind River area, the final cycle of groundwater took place during the Hudsonian (Penokean) orogeny, as evidenced by the 1,700-m.y. age for uraninite and brannerite obtained by Mair and others (1960). However, Roscoe (1969) showed that the isotopic compositions are best explained by resetting rather than introduction of new material.

Abraham (1953) and McDowell (1957, 1963) regarded the ores as fossil placer deposits. Pienaar (1958, 1963) and Roscoe (1957 and later) have also indicated a preference for a placer origin. D. S. Robertson (1962a, b) has also assembled many data, particularly on uranium/thorium ratios, that is suggestive of a placer origin. Holmes (1957) suggested that the ores were of syngenetic (placer) origin but were modified by later events.

Derry (1960) has suggested a syngenetic origin for the uranium mineralization but has raised the possibility that the uranium was largely carried in solution and reprecipitated in gravel banks by bacterial agencies. Joubin (1960) has published similar ideas but has not included bacterial precipitation.

It may be noted that other conglomerates in the district, for example, the Matinenda polymictic conglomerate and the Ramsay Lake, Bruce, and Cobalt conglomerates, do not carry markedly high uranium values, although all, particularly the Bruce, carry pyrite and

pyrrhotite. An exception to this occurs when such a conglomerate unconformably overlies the uranium-bearing sequence or infringes on the basement. It should be noted that the red granitic bodies in the basement are radioactive (Robertson, 1960, 1967).

The sericitic matrix of the ore-bearing conglomerates is similar to the sericitic paleosol developed on the Archean-Huronian unconformity and was probably derived from that regolith; there is no reason to suppose that it was produced by the passage of hydrothermal solutions. *Uraniferous oligomicitic pebble conglomerates and a green arkose sequence* are characteristic of whichever part of the Huronian overlies the basement in the area of the north shore of Lake Huron, and these rocks show a progressive northward overlap. Significant thicknesses and grades have so far been found only in the Matinenda Formation.

Quartz veins and other evidence of intense hydrothermal activity are not conspicuous in the rocks of the area and bear no sympathetic relation to the uranium deposits. Where found, the associated mineralization is of copper and other sulphide minerals. Within the Blind River-Elliot Lake camp, there is no indication that either the Nipissing (formerly included with the Keweenawan) or the olivine-diabase intrusions are a possible source of major radioactive mineralization. There is, however, evidence that ores were subjected to intense local alteration and that all rocks suffered sulphide mineralization at the time of intrusion of the Nipissing diabase and the time of regional folding. Age-determination data are consistent with this concept.

The overall distribution of beds and the behaviour of thickness and grades are more consistently explained by the modified placer hypothesis, which the author has supported (Robertson, 1960 and later). The recent mineralogic studies of Ferris and Ruud (1971) and Theis (1973) have added cogent additional support to the modified placer hypothesis.

The deposits were derived from Archean terrane to the north and northwest, transported by rapidly moving water, deposited in a near-shore fluviatile environment under reducing conditions about 2,500 m.y. ago, and, later, subjected to diagenesis and to minor alteration during subsequent intrusive metamorphic and tectonic events.

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the uranium deposits. R. Balgalvis and J. Michalik of the Geological Branch of the Ontario Division of Mines, Ministry of Natural Resources, provided assistance in the preparation of the illustrative material, and Miss T. Abolins and Miss C. Elliott helped in the production of the manuscript. Permission to participate in the workshop and to publish this paper was given by the Director of the Geological Branch, Ontario Division of Mines.

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# Some Metallogenic Features of the Huronian and Post-Huronian Uraniferous Conglomerates

By VLADIMIR RUZICKA

GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN  
QUARTZ-PEBBLE CONGLOMERATES

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## GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN QUARTZ-PEBBLE CONGLOMERATES

# SOME METALLOGENIC FEATURES OF THE HURONIAN AND POST-HURONIAN URANIFEROUS CONGLOMERATES

By VLADIMIR RUZICKA<sup>1</sup>

### ABSTRACT

Two genetic types of uraniferous conglomerates can be distinguished: (1) those with syngenetic mineralization, and (2) those with epigenetic mineralization.

Uraniferous conglomerates with syngenetic mineralization are restricted to lower Proterozoic sediments, such as those in the Huronian Supergroup in Canada. It is postulated that deposits with this type of mineralization can occur only in regions where above-normal concentrations of radioelements and uranium minerals were present in the source rocks.

Liberation of uranium minerals, their transportation to the site of deposition, and their deposition along with other heavy minerals took place under oxygen-deficient conditions but at a time when primitive organisms existed.

The depositional environment was near volcanic centres; excessive sulphur, a product of volcanic and postvolcanic activity, apparently caused sulphurization of some minerals and of hydrocarbon, the latter derived from organic and possibly inorganic material.

Concentration of uranium minerals during transportation and deposition was governed by hydraulic and hydrodynamic conditions.

The sedimentary syngenetic origin of uranium mineralization may be illustrated by a portion of an ore deposit that corresponds to a complete cycle of sedimentation. The cycle starts with deposition of coarse grains of quartz, continues with the deposition of a mixture of finer grains of quartz and pyrite with distinct graded bedding, is followed by deposition of monazite and brannerite, and ends with deposition of fragmented fine grains of uraninite embedded in sulphur-rich hydrocarbon. The bed containing hydrocarbon and uraninite is locally depressed by impact of an exotic pebble of quartz. This situation apparently testifies to the fact that the carbonaceous material is a product of algal mats that existed in quiet times between cycles of sedimentation. The succeeding cycle again starts with the deposition of coarse grains of quartz.

Preservation of uranium mineralization during and after diagenesis was possible owing to reducing conditions of the depositional environment.

Uraniferous conglomerates with epigenetic mineralization are localized in regions containing rocks with higher contents of uranium. Primary concentration of uranium minerals in these source rocks can also be syngenetic.

Transportation of uranium from the site of original liberation to the site of deposition was apparently mainly in solutions and in refractory minerals by hydraulic action. Mobilization of uranium from the source rocks and its redistribution were governed by hydrodynamic gradients related to erosional, tectonic, magmatic, or metamorphic processes in the Earth's crust. The conglomerate beds apparently served as channels for penetration of uranium-bearing fluids.

Deposition of uranium took place by one or more epigenetic processes: adsorption, complexing, precipitation, or redox changes according to the degrees of solubility and stability of uranium compounds.

The mineralization in this type of deposit has been preserved beneath the zone of oxidation.

Evidence of these mentioned features is based upon studies on the Huronian uraniferous conglomerate from Elliot Lake, Canada, and on the late Precambrian and Paleozoic radioactive conglomerates from the eastern Ural Mountains and Enisey Crest region, U.S.S.R.

### INTRODUCTION

At the symposium of the International Atomic Energy Agency (1974) dealing with the formation of uranium deposits, fundamental criteria on the metallogeny of uranium were formulated, among which special attention was paid to those related to the genesis of uranium mineralization in quartz-pebble Huronian conglomerates.

It was suggested that reports of younger conglomerates of this type such as Cambrian conglomerate, described in the Soviet literature, are in error.

Criteria related to some metallogenic features of the quartz-pebble conglomerates were derived from studies of the stratigraphic setting, sedimentary nature, mineral composition, and geochemical features of the Rand-Elliot Lake type conglomerate. No further description of this type of mineralization was given in the International Atomic Energy Agency report (1974).

The genesis of uranium mineralization in post-Huronian conglomerates was discussed only briefly in the subsequent discussion, which showed that this topic needs more clarification and the deposit types require more exact classification.

Therefore this study concentrates on metallogenic features of the Huronian and post-Huronian conglomerates in order to contribute to the solution to the mentioned problems. The author's views on the long-standing question of a syngenetic versus epigenetic origin of various kinds of conglomerate uranium deposits are also presented.

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The conclusions are based upon studies of the Huronian uraniferous conglomerates, Elliot Lake, Canada, and of the late Precambrian and Paleozoic radioactive conglomerates of the eastern Ural Mountains and the Enisey Crest region, U.S.S.R. In all instances the host for the uranium mineralization is quartz-pebble conglomerate; in addition, the Russian deposits are also in uraniferous polymictic conglomerates.

### SOME FEATURES OF THE URANIUM MINERALIZATION AT ELLIOT LAKE, CANADA

The Geological Survey of Canada airborne gamma-ray spectrometric map of the Elliot Lake-Blind River region (Richardson and others, 1975) shows that the Elliot Lake and Blind River uranium deposits are localized within a terrane exhibiting abnormal concentrations of radioelements. The deposits are confined to lower Proterozoic sediments of the Huronian Supergroup whose provenance was the pre-Huronian basement rocks: granites, granite gneisses, and other felsic rocks containing enclaves of metavolcanics and metasediments with banded iron-formations. The granitic rocks that occur in areas from which clastic material was supplied to the Huronian sediments locally contain concentrations of uranium exceeding several times normal clarke for such rocks (Richardson and others, 1975; Roscoe and Steacy, 1958). Samples of these rocks contain between 5 and 10 ppm (parts per million) eU (Darnley and others, 1977). This is considered a fundamental metallogenic feature and is a necessary condition for the formation of an economic uranium deposit of this type.

The source rocks underwent intensive weathering during pre-Huronian time. It is postulated that weathering and disintegration of the source rocks took place under oxygen-deficient conditions (Roscoe, 1969, 1973). It is also postulated that the transportation of the detritus from the site of origin to the site of deposition occurred under conditions similar to those for the disintegration of the source rocks (Roscoe, 1969). Evidence for this postulate, among other features, is the presence of subrounded grains of pyrite and other ore-forming minerals, which are apparently of detrital origin (fig. 1). Deficiency of free oxygen might also have been partially related to volcanic activity. It is the author's opinion that both an oxygen-deficient atmosphere and volcanic activity were necessary factors in formation of the Elliot Lake deposits.

Concentration of uranium minerals was governed by hydraulic action of the streams.

Elliot Lake ore was deposited in sedimentation cycles. One such cycle is illustrated in figure 2. It starts with deposition of a coarse fraction of light minerals (quartz) and continues with deposition of a gradationally finer and finer mixture of quartz and pyrite grains. Farther

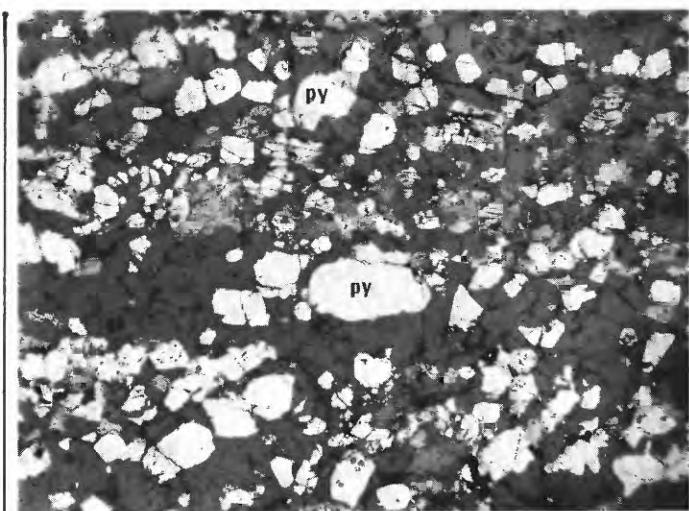


FIGURE 1.—Photomicrograph of a uranium ore sample collected in Denison Mine, Elliot Lake, containing rounded grains of pyrite (py) ( $\times 3$ ). Reflected light.

upward in the section the graded bedding encompasses concentrations of finer grains of pyrite, monazite, and brannerite. The cycle ends with deposition of microscopic fragmented grains of uraninite embedded in hydrocarbon (fig. 3) (Ruzicka and Steacy, 1976).

The upward fining of the mineral grains in these cycles is proportional to their absolute weight and inversely proportional to their density. Figure 4 shows a quartz pebble of the subsequent cycle depressing the uraninite-hydrocarbon-rich band of the previous cycle.

The deposition of uranium was governed by sedimentary processes. There is no doubt about the detrital nature of the quartz grains; the pyrite occurs as angular to subrounded grains that show a decrease in size toward the top of the bed; the brannerite is present mainly as ragged grains, and the monazite, as rounded grains. The uraninite contains several percent of thorium (Ruzicka and Steacy, 1976). This testifies to the origin of the uraninite in a high-temperature environment.

Roscoe (1969) and Theis (1976) observed composite grain aggregates containing titania and uranium that, according to Theis, are enriched by uranium only in the vicinity of uraninite grains. Since this enrichment is a local process, involving distances of only a few millimetres, it can be postulated that the formation of these aggregates occurred *in situ*—that is, during diagenesis. Thus, this phenomenon can be called “uranization.” It apparently corresponds to Ramdor’s “Pronto-reaction” (Ramdohr, 1958). Because no additional uranium has been introduced into the sediment, such a process may be regarded as redistribution of syngenetic uranium.

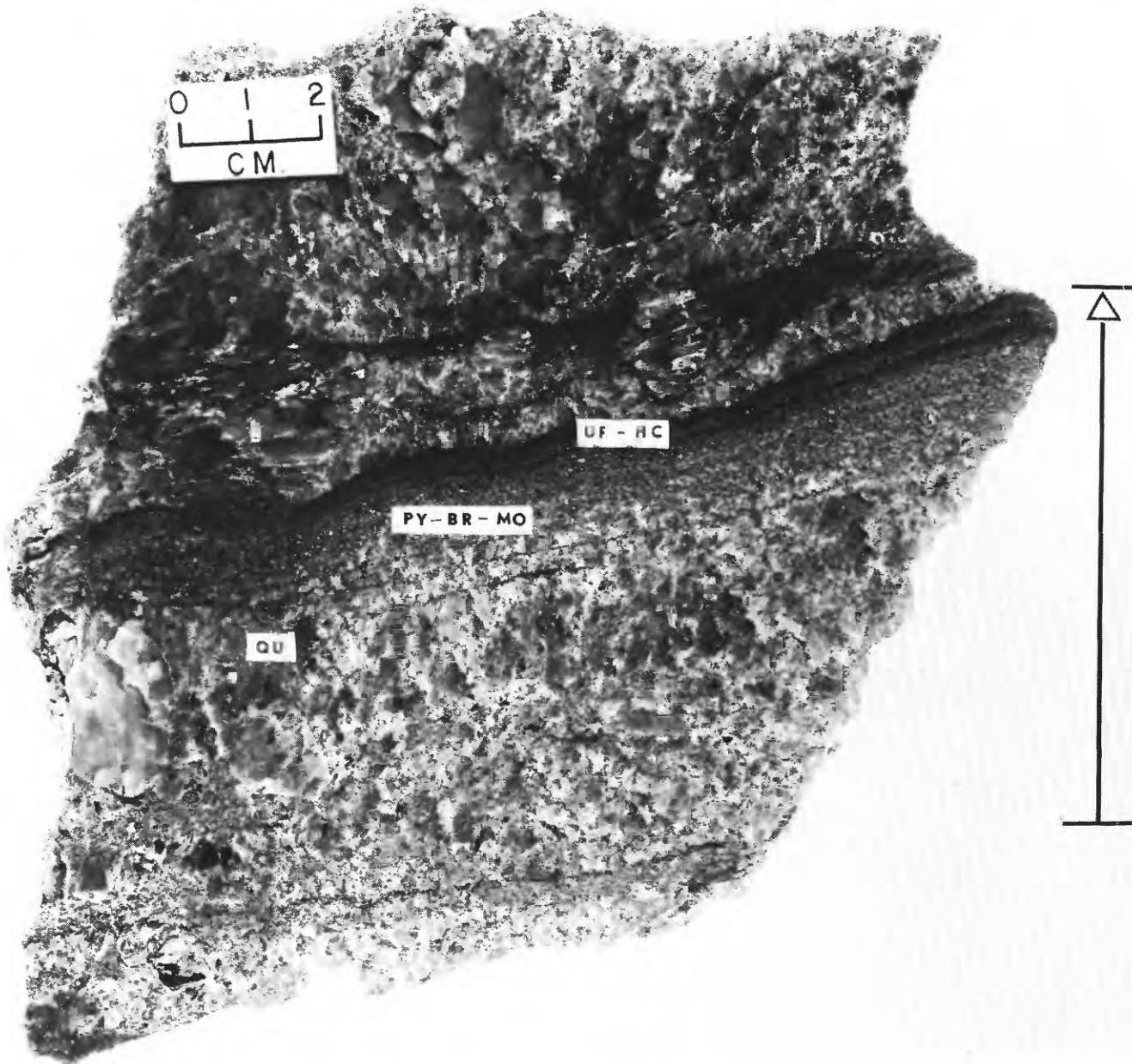


FIGURE 2.—A sample of uranium ore from Denison Mine, Elliot Lake, Ontario; the portion within the arrow area contains bedded grains of quartz, pyrite, brannerite, monazite, uraninite, and hydrocarbon and represents one sedimentation cycle. The arrow points towards the top of the bed. UR-HC=uraninite-hydrocarbon-rich section; PY-BR-MO=pyrite-brannerite-monazite-rich section; QU=quartz-pebble-rich section.

The hydrocarbon in the Elliot Lake ores, variously described as "radioactive hydrocarbon," "uranium hydrocarbon," or more generally as "thucholite" might be of biogenic origin (Ruzicka and Steacy, 1976). A similar phenomenon has been studied by Hallbauer (1975) in the Witwatersrand uranium ores and interpreted as evidence for plant origin of the Witwatersrand

"carbon." Pretorius (1975) interpreted the carbonaceous material arranged in narrow layers in the gold-uranium conglomeratic ores as a product of "algal or lichen colonies preferentially developed about the mouths of the major rivers and at the end of certain cycles of sedimentation."

The hydrocarbon usually exhibits uniform extinction

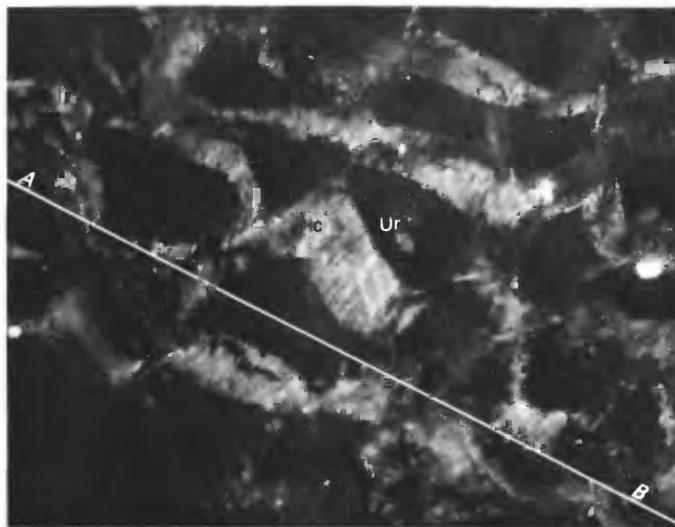


FIGURE 3.—Photomicrograph showing arrangement of detrital grains of uraninite (Ur) embedded in hydrocarbon (Hc). Line A-B denotes the plane of bedding ( $\times 163$ ). Reflected light.

and a columnar structure under crossed nicols (figs. 5, 6). The fibres (columns) are oriented normal to the bedding plane. This corresponds with the orientation of the columnar carbon in ores from the Orange Free State Goldfield (Basal Reef), as reported by McKinney (1964). Hallbauer (1975) postulated an organic origin of the columnar carbon. Some of the hydrocarbon heals fractures in uraninite (fig. 7). Uraninite embedded in the hydrocarbon can be interpreted as evidence of transportation of uraninite in organic-supported colloids. Furthermore, if one accepts that the organisms were also active in the provenance area of the detrital material, then the possibility that the biological agents participated in liberation of uranium minerals from source rocks cannot be excluded. Therefore, accepting Silverman and Minoz' (1970) conclusions on similar situations, one can postulate that the liberation of uraninite from the source rocks might also be caused by fungal attacks.

The abundant pyrite is apparently partly of detrital and partly of epigenetic origin (Roscoe, 1969). In addition to pyrite derived from Archean rocks, the sulphur-saturated fluids from contemporaneous volcanic activity apparently enabled crystallization of pyrite in or immediately adjacent to the volcanic centres. Such fluids may also have introduced pyrite into the transported detritus, allowed crystallization of pyrite in situ within the deposit, and affected the detrital minerals by sulphurization during transportation and deposition. Some sulphur could also have been derived from organic matter or from meteoric sulphate solutions (Ferris and Ruud, 1971). It is the author's opinion that some of the pyrite exhibiting abnormal magnetic susceptibility in the

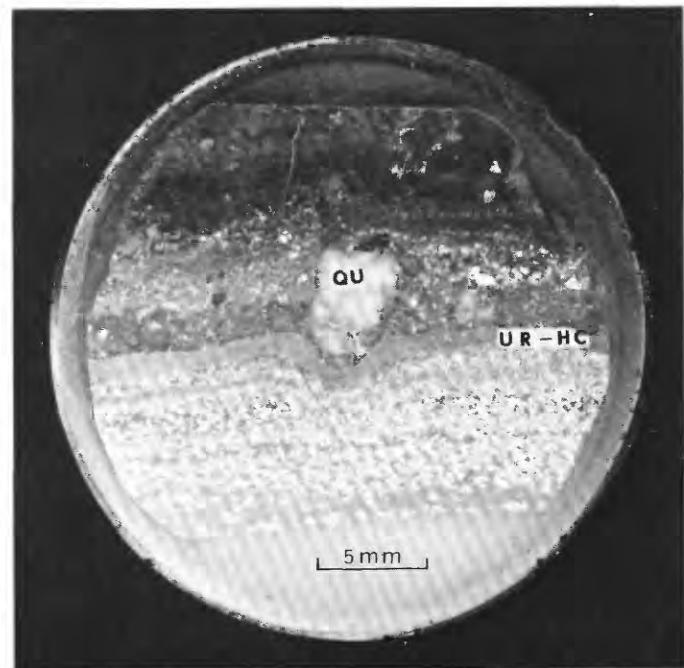


FIGURE 4.—A quartz pebble depressing a uraninite-hydrocarbon-rich band. A polished section of the Elliot Lake ore, Denison Mine (Geological Survey of Canada Photograph 203006M).

Elliot Lake ore (Hartman and Wyman, 1970) originated by sulphurization of magnetite. Sulphurization is apparently the cause of abnormally high content of sulphur in the hydrocarbon. H.R. Steacy of the Geological Survey of Canada investigated (personal commun.) "thucholites" from several types of environments and found the highest sulphur concentrations (more than 20 percent sulphur) only in Elliot Lake hydrocarbons. The sulphides acted also as agents for preserving detrital uranium minerals during transportation, deposition, and diagenesis.

These characteristics support the idea of a sedimentary-syngenetic origin for the Elliot Lake ores. However, more detailed studies are necessary to obtain a complete model of the physical, chemical, and organic mechanisms involved in formation of this type of deposit.

The author proposes a term for uranium deposits of this type as "lower Proterozoic oligomictic pyritic conglomerates with syngenetic mineralization."

#### SOME FEATURES OF THE POST-HURONIAN CONGLOMERATES IN THE URAL MOUNTAINS AND ENISEY CREST, U.S.S.R.

Auriferous and uraniferous conglomerates in the U.S.S.R. have been recently most comprehensively described by Krendelev (1974), Khvostova (1969), Oslopovskikh (1968) and Krendelev and Zlobin (1972).

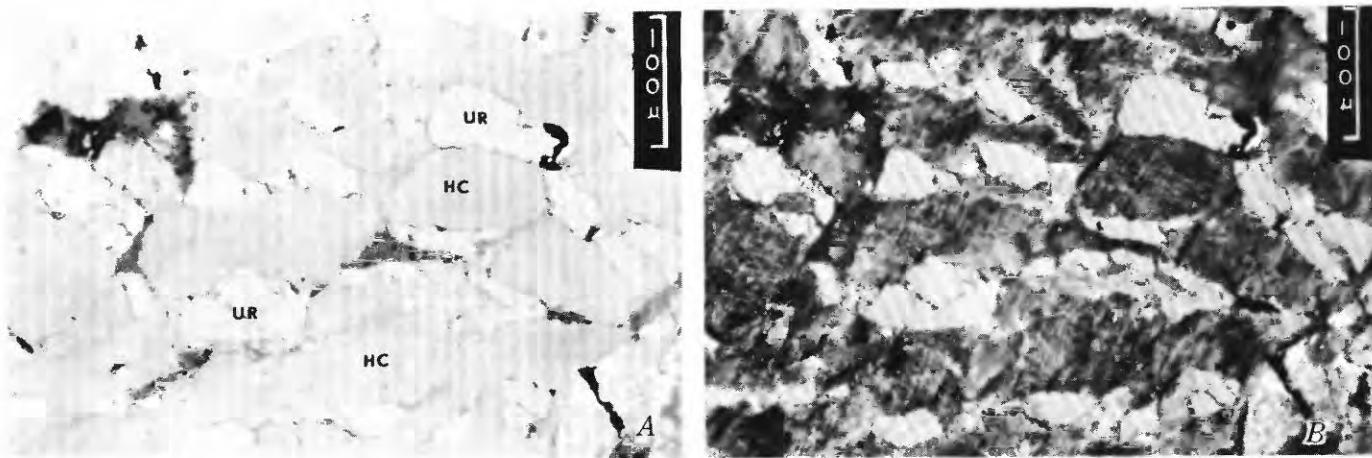


FIGURE 5.—Photomicrographs of a portion of the uraninite-hydrocarbon band labeled UR-HC on figure 4 (UR=uraninite, HC=hydrocarbon). A, under parallel nicols. B, under crossed nicols. Reflected light. (Identification and photos by S. Kaiman, Canada Centre for Mineral and Energy Technology.)

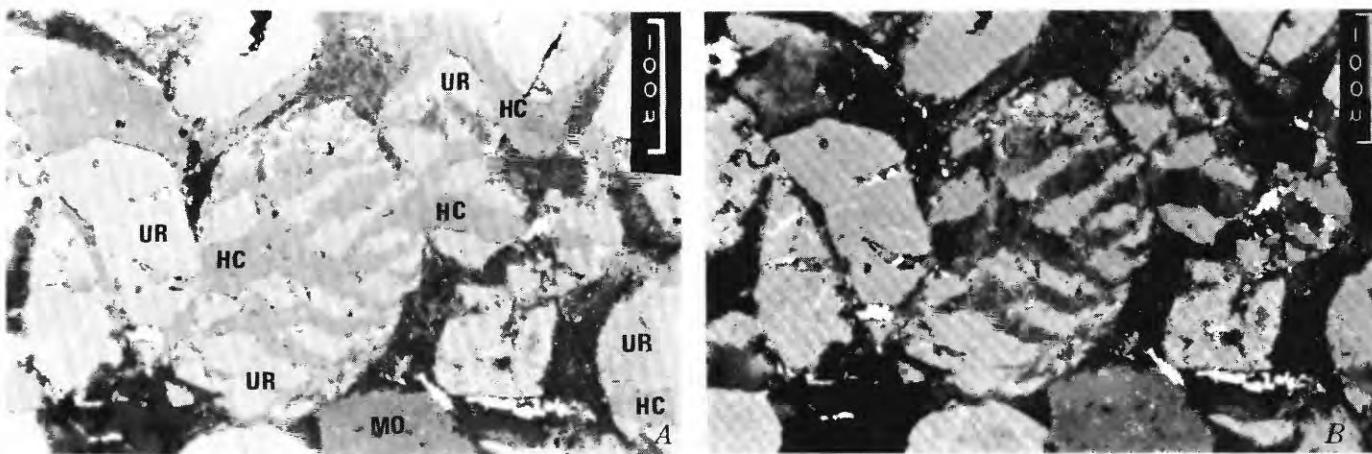


FIGURE 6.—Photomicrographs of a portion of the uraninite-hydrocarbon band labeled UR-HC on figure 4 (UR=uraninite, HC=hydrocarbon, MO=monazite). A, under parallel nicols. B, under crossed nicols. Reflected light. (Identification and photo by S. Kaiman, Canada Centre for Mineral and Energy Technology.)

Several deposits containing gold or uranium, such as Laba, Baley, Kara-Su, Khuzhur, Timan, Panimba, and Eastern Ural Mountain, have been described in the Russian literature. Among these deposits, the Eastern Ural Mountain deposit and the Panimba occurrences contain some uranium.

The Eastern Ural Mountain deposit occurs in the Lower Cambrian. The ore-bearing horizon unconformably overlies Precambrian gneisses, which have been metamorphosed to the garnet facies. The basement rocks also comprise effusives of the rhyolite-dacite group and intrusives of the biotite-microcline granite group. A regolith as much as 10 m thick is developed on the basement rocks (Krendelev, 1974).

The Cambrian sediments that host the uranium mineralization are usually classified into two groups: the lower, approximately 1,000 m thick, consisting of quartzites with intercalations of argillites and conglomerates, and the upper, 500–1,000 m thick, consisting of argillites with intercalations of quartzites and tuffs.

Uranium mineralization is confined mainly to basal and intraformational conglomerates. The basal conglomerates (0.3–1.0 m thick) are polymictic, carrying dominant quartz pebbles and minor granite and quartzite pebbles. The intraformational conglomerates are mainly oligomictic and monomictic with local thick masses ranging from 3.0 to 9.0 m. The monomictic portions of the conglomerates are, as a rule, more

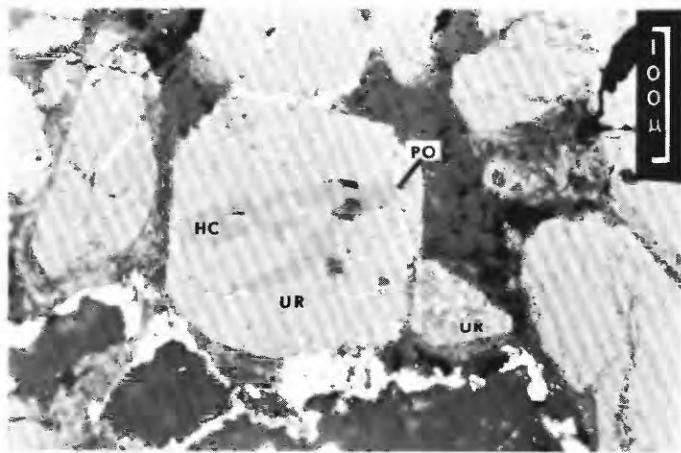


FIGURE 7.—Photomicrograph of a portion of the uraninite-hydrocarbon band labeled UR-HC on figure 4 (UR = uraninite, HC = hydrocarbon, PO = pyrrhotite). Reflected light. (Identification and photo by S. Kaiman, Canada Centre for Mineral and Energy Technology.)

mineralized than are the oligomicitic parts. In both cases uranium, thorium, rare-earth elements, niobium, titanium, and zirconium mineralization is exclusively confined to the matrix.

The heavy-mineral assemblage of the conglomerates consists of zircon, malacon, thorite, uranotiorite, uranotorianite, allanite, ilmenite, rutile, brannerite(?), hematite, magnetite, pyrite, and other minerals, pyrite being only a minor constituent. Pitchblende and gold have not been reported. The uranium/thorium ratios range generally from 1:1 to 1:18, and in those parts directly overlying the basement granites, from 3:1 to 1:10. Concentrations of the ore minerals also occur in veinlets and in metasomatic disseminations. Tonnages and grades of the deposit have not been disclosed.

The genesis of the Eastern Ural Mountain deposit has been a subject of discussion. Shcherbin (1968a, b) correlated genesis of the deposit with the genesis of the "ancient" (that is, lower Proterozoic) uraniferous pyritic quartz-pebble conglomerates and classified the deposit as an "ancient placer" type.

On the other hand, Krendelev (1974) opposed Shcherbin and summarized his own opinion as follows:

1. The mineralization is confined to lower Paleozoic sediments (and in this respect differs from the Elliot Lake-Rand type).
2. The area with the mineralization is spatially related to a deep-seated fault.
3. Volcanic activity affected the ore-bearing beds as well as the older (foot-wall) and younger (hanging-wall) rocks.
4. The host rocks and the underlying complexes, which contain intrusive and extrusive rocks, have been regionally metamorphosed. Metamorphic events

caused mobilization and redistribution of all rock-forming components.

5. Locally, contact metamorphism took place.
6. Ore mineralization in the conglomerates, quartzites, and acidic extrusives is apparently related to the epidote-amphibolite state of metamorphism. The metamorphic events took place  $239 \pm 9$  million years ago (and are therefore younger than the host rocks).
7. Distribution of secondary rock-forming minerals, such as twinned microcline, albite, muscovite, and blue quartz, coincides apparently with the distribution of the ore mineralization.
8. Because of these facts and certain typomorphic features of the ore-forming minerals, the ore-forming processes can be interpreted as epigenetic. Thus the syngenetic theory, as formulated by Shcherbin (1968b), should be considered as not proven.

The second deposit, Panimba, occurs within the central auriferous belt of the Enisey Crest in the Central Asian part of the U.S.S.R., where other uranium occurrences are also known (Ruzicka, 1971).

The uraniferous and rare-earth-element-bearing conglomerate occurs in a limb of an anticline, the core of which consists of a sedimentary sequence of limy shales, quartzites, and breccias. The limb contains beds of conglomerates intercalated in quartzites and black shales. The rocks are of upper Proterozoic age and are highly metamorphosed.

The mineralized conglomerate is between 5 and 10 m thick and extends along strike for several kilometres. The most abundant minerals present are pyrite, pyrrhotite, marcasite, chalcopyrite, and sphalerite. Gold in individual samples grades 1.55 gram per ton; nickel, 0.045 percent; and cobalt, 0.059 percent. Uranium and thorium are present, but their amounts have not been disclosed.

The genesis of the Panimba deposit has been interpreted by Krendelev (1974) and can be summarized as follows:

1. The origin of the mineralization has been apparently related to the magmatic processes of a granite intrusion.
2. During the period of magmatic and postmagmatic activity the rocks have been decarbonatized and altered by magnesian metamorphism, during which sulphur was mobilized from organic matter and redistributed.
3. Permeable rocks, such as conglomerates, served as channels for mineralizing fluids.
4. Mineralization is therefore apparently of epigenetic origin and not necessarily of syngenetic-sedimentary (that is, placer) type.

The author of this paper proposed the definition of the Panimba and similar uranium conglomeratic deposits as "post-lower Proterozoic conglomerates with epigenetic mineralization."

## CONCLUSIONS

Comparisons between the two genetic types of uraniferous conglomerates allows generalization of their basic features as follows:

A. Lower Proterozoic pyritic oligomicitic conglomerates with syngenetic mineralization:

1. The source area contains rocks with abnormal clarkes of radioelements and with uranium-bearing minerals.
2. Liberation of the uranium minerals took place by physical, chemical, and possibly also biological mechanisms under oxygen-deficient conditions.
3. Transportation of the uranium minerals in the detritus occurred under oxygen-deficient conditions, namely in an oxygen-deficient atmosphere combined with volcanic and postvolcanic exhalation. Some uranium minerals were probably transported as suspensions in colloidal organic matter.
4. Concentration and deposition of uranium and accompanying minerals were governed by hydraulic action of streams. Sedimentation took place at a time when primitive organisms already existed.
5. Excessive sulphur, a product of the associated volcanic activity, caused sulphurization of some minerals and hydrocarbon, the latter being derived from organic and possibly also inorganic material.
6. Preservation of concentrations of uranium minerals was possible, with local redistribution of uranium due to the presence of reducing agents in the deposit and by burial of the mineralized beds.

B. Post-lower Proterozoic conglomerates with epigenetic mineralization:

1. Deposits occur in areas containing rocks with higher uranium clarkes. Deposition of uranium in some of the source rock units could have been originally syngenetic.
2. The first stage in redistribution of uranium from the source area apparently involved transport of uranium from source to the site of deposition in solutions or in refractory minerals. The process presumably resulted in a further local concentration of uranium in the environment.

3. The critical stage in the epigenetic process involved mobilization of stage 2 uranium and its redeposition, governed by hydrodynamic gradients, related to erosional, tectonic, magmatic, or metamorphic processes active in the Earth's crust. During this process the conglomerates served as channels for penetration of the uranium-bearing fluids.
4. Deposition of uranium took place by one or more of the epigenetic processes: adsorption, complexing, precipitation, or redox changes according to solubility and stability degrees of uranium compounds.
5. Preservation of primary mineralization was possible beneath the zone of oxidation.

These conclusions fully support criteria on metallogeny of uranium as formulated by the symposium of the International Atomic Energy Agency (1974) on the formation of uranium deposits. Furthermore, they allow formulation of definitions distinguishing syngenetic and epigenetic mineralization in uraniferous conglomerates.

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# Temporal and Other Factors Affecting Deposition of Uraniferous Conglomerates

*By* STUART M. ROSCOE

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QUARTZ-PEBBLE CONGLOMERATES

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GEOLOGICAL SURVEY PROFESSIONAL PAPER 1161-W





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## GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN QUARTZ-PEBBLE CONGLOMERATES

### TEMPORAL AND OTHER FACTORS AFFECTING DEPOSITION OF URANIFEROUS CONGLOMERATES

By STUART M. ROSCOE <sup>1</sup>

#### ABSTRACT

Uraniferous, auriferous, pyritic quartz-pebble conglomerates, unknown in strata less than 2.3 billion years old, are considered preox-yatmoversian analogues of black-sand placers common in younger terrestrial sediments. Radioactive conglomerates in Huronian rocks near Elliot Lake, and others, are within the most proximal facies of once-extensive sheets of terrigenous clastics derived from soil mantles and chemically mature colluvium that covered Archean protocontinents weathered under an atmosphere that lacked free oxygen.

The initial entrapment of quartz-gravel-bearing grits in Elliot Lake Group, as well as subsequent deposition, deformation, and preservation of Huronian rocks, was governed by the development of tectonically unstable conditions in an epicontinental sector of the Superior Craton. Warping of the southerly inclined paleoslope, adjustments along old fractures, and eruptions of basalt were accompanied by deposition of coarse clastic wedges or alluvial fans atop soil-mantled Archean basement rocks, weathered Huronian basalt flows, and finer grained sands that flanked or overlay other gritty conglomerate-bearing lenses.

Successions resembling the Huronian Supergroup are found west of Hudson Bay, in Wyoming, and in central Quebec, suggesting that tectonic traps for coarse clastics with pyritic conglomerates developed extensively around a more stable core of the Archean protocontinent about 2.5 billion years ago. Comparable tectonic conditions developed earlier in South Africa, where bankets occur in strata 2.8 to 2.3 billion years old. Possibilities of discovering other occurrences of this type in North American rocks older than 2.6 billion years are remote, but one occurrence in Quebec may be in this category. Favourable areas are the Huronian Supergroup, other remnants of Paleoaphebian strata (2.6 to 2.3 billion years), and highly metamorphosed equivalents of these. Many of these remnants are buried under more extensively preserved younger Proterozoic strata, but the Paleoaphebian rocks should not be regarded as basal phases of the Proterozoic tectonostratigraphic units.

#### INTRODUCTION

The pyritic quartz-pebble conglomerates, or bankets, considered in this publication have been reported in fewer than a dozen separate districts in the world, yet they include the most extensive known deposits of

uranium and gold, aggregating very large proportions of the world's resources of these metals. This is remarkable, as other types of uranium and gold deposits are vastly more numerous and occur in geological situations that are widespread. Moreover, all known bankets, invariably containing a distinctive suite of enriched constituents including uranium and gold, are in successions that are either reasonably reliably dated or interpretable as more than about 2.3 aeons (10<sup>9</sup> years) old. They contain the coarsest fractions of detritus derived from weathering of extensive granitoid terrains. Any such sediments this old contain disseminated pyrite, and pebbly layers within them contain coarser grains of pyrite along with rutile, zircon, monazite, other undoubtedly detrital heavy minerals, and gold and uranium minerals. Pyrite has not been found to occur in this manner in otherwise-similar younger sediments although the younger sediments are collectively many orders of magnitude more abundant than the scattered remnants of pre-2.3 aeon quartzose sandstones.

Quartzose clastics deposited less than 2.3 aeons ago contain fine-grained interstitial ferric oxide and heavy detrital iron oxides in lieu of pyrite. Clearly, these younger deposits have had different histories of weathering, sedimentation, diagenesis, metamorphism, or epigenetic modifications than have the older ones. The change in sediment type is coincident with a change in character of paleosols from pyritic ones depleted in iron and particularly ferric oxide to ones showing an upward enrichment in ferric oxide. This change preceded the first widespread depositions of dolomite and the most voluminous world-wide depositions of ferric iron in banded iron-formations. Many authorities interested in genesis of iron-formations (Bayley and James, 1973; Beukes, 1973), in atmospheric and biological evolution (Oparin, 1938; Rubey, 1955; Cloud, 1973; Holland, 1973; and others), and in uranium deposits (Roscoe, 1969)

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have considered that these phenomena reflect a change in the Earth's atmosphere from one lacking free oxygen to one containing free oxygen. I have suggested the term "oxyatmovation" for this change (Roscoe, 1973).

Skeptics hinge their doubts about this hypothesis on several incorrect ideas. Foremost amongst these is the belief that an atmosphere containing free oxygen is as essential a condition for the chemical precipitation of ferric iron in Archean iron-formations as it is for the survival and deposition of detrital and authigenic hematite in continental red beds (see Davidson, 1965). This belief requires a further unwarranted assumption that the presence of pyrite in lieu of hematite is in itself sufficient reason to suppose that basket-bearing successions were deposited not subaerially under high-energy conditions but in euxinic marine environments (see Pettijohn, 1970). Alternatively, it has been supposed that the pyritic rather than hematitic character of the strata must be due to metamorphism or regionally pervasive metasomatic processes. Other critics purport that the hypothesized change in the atmosphere is based only on inconclusive evidence that uraninite and pyrite occur in forms resembling detrital grains. This attack can be discounted, as it ignores other evidence such as hydraulic equivalent sizes of minerals in bankets, variations in compositions amongst associated uraninite and pyrite grains, penecontemporaneous displacements of some of these grains during compaction, the iron-depleted character of paleosols, and the pyritic and nonhematitic character of host strata evidently deposited subaerially.

Knowledge of environments of deposition of various pyritic conglomerates is thus important not only as an exploration guide but also as a critical factor in deciphering atmospheric evolution. New data presented herein show that major conglomeratic zones near Elliot Lake were deposited subaerially in headward, largely distributary, sections of swift, necessarily aerated streams—a most inhospitable environment for accumulation of organic material. Pyritic conglomerates occur in a range of geological environments, as do black-sand placers. Differences in chemical and textural maturity of sediments, associations with shale, greywacke, or volcanics, or differences in metamorphism have nowhere been found to determine that pyrite rather than iron oxides dominate the heavy-mineral or diagenetic-mineral assemblages.

#### BANKET-BEARING SUCCESSIONS

##### DISTRIBUTION AND LITHOLOGIES

Uraniferous and auriferous pyritic conglomerates are known in South Africa, in the Fortesque Group in Western Australia, at Serra de Jacobina in Brazil, in the Huronian Supergroup north of Lake Huron, at Sakami Lake 90 miles (145 km) east of James Bay, and at Padlei

(Montgomery Lake) 100 miles (160 km) west of Hudson Bay. In addition, the Deep Lake Formation in the Medicine Bow Mountains in southeast Wyoming contains limonitic, slightly radioactive, slightly auriferous quartz-pebble layers that represent bankets in which pyrite has been almost entirely destroyed by weathering and uranium minerals leached in surface exposures. The North American localities are shown in figure 1.

Stratigraphic data for a number of these areas have been summarized by Roscoe (1973, fig. 3), with available isotopic dates of basements, basket host strata, and of red beds, black sands, iron-formations, and unconformities. Some of this information showing common mities. Some of this information showing common features amongst basket-bearing successions is outlined more specifically below.

The bankets occur in slightly feldspathic to highly feldspathic argillaceous quartzites, which dominate the host successions. Volcanics form important parts of most successions and are present in all, with the possible exception of Jacobina, which contains serpentinite and amphibolite bodies. Serpentinite is also present at Sakami Lake. All successions contain argillaceous units. Tillites are present in the Huronian, Medicine Bow, Padlei, and possibly the Witwatersrand sequences. The Witwatersrand is particularly thick and varied. It is underlain by the dominantly volcanic Dominion Reef and overlain by Ventersdorp lavas; both contain bankets.

#### CORRELATIONS IN NORTH AMERICA

The Huronian Supergroup (A), the Deep Lake Formation and an overlying section of the Libby Group in Wyoming (B), and the Montgomery Lake Group together with the lower part of the Hurwitz Group in District of Keewatin (C) are compared in table 1. Blackwelder (1926) noted similarities between Medicine Bow and Huronian rocks and, in particular, considered the Headquarters Schist to be a tillite likely correlative with the Gowganda Formation. Huston (1968), on the basis of such comparisons, suggested that economically significant paleoplacers might be found in the Deep Lake Formation. Young (1973) concluded that the upper groups in the three areas showed evidence of climatic changes so similar and uncommon that he believed they must be coeval. In 1973, I found that limonitic quartz-pebble layers at several places and at different stratigraphic positions within the Deep Lake Formation contained uranium, thorium, gold, titanium, and other minor constituents in amounts much like those in weakly radioactive pyritic small pebble beds of the Mississagi Formation but in amounts different from those in black-sand-bearing pebble beds. Thus the Deep Lake Formation contains bankets, although pyrite and no doubt

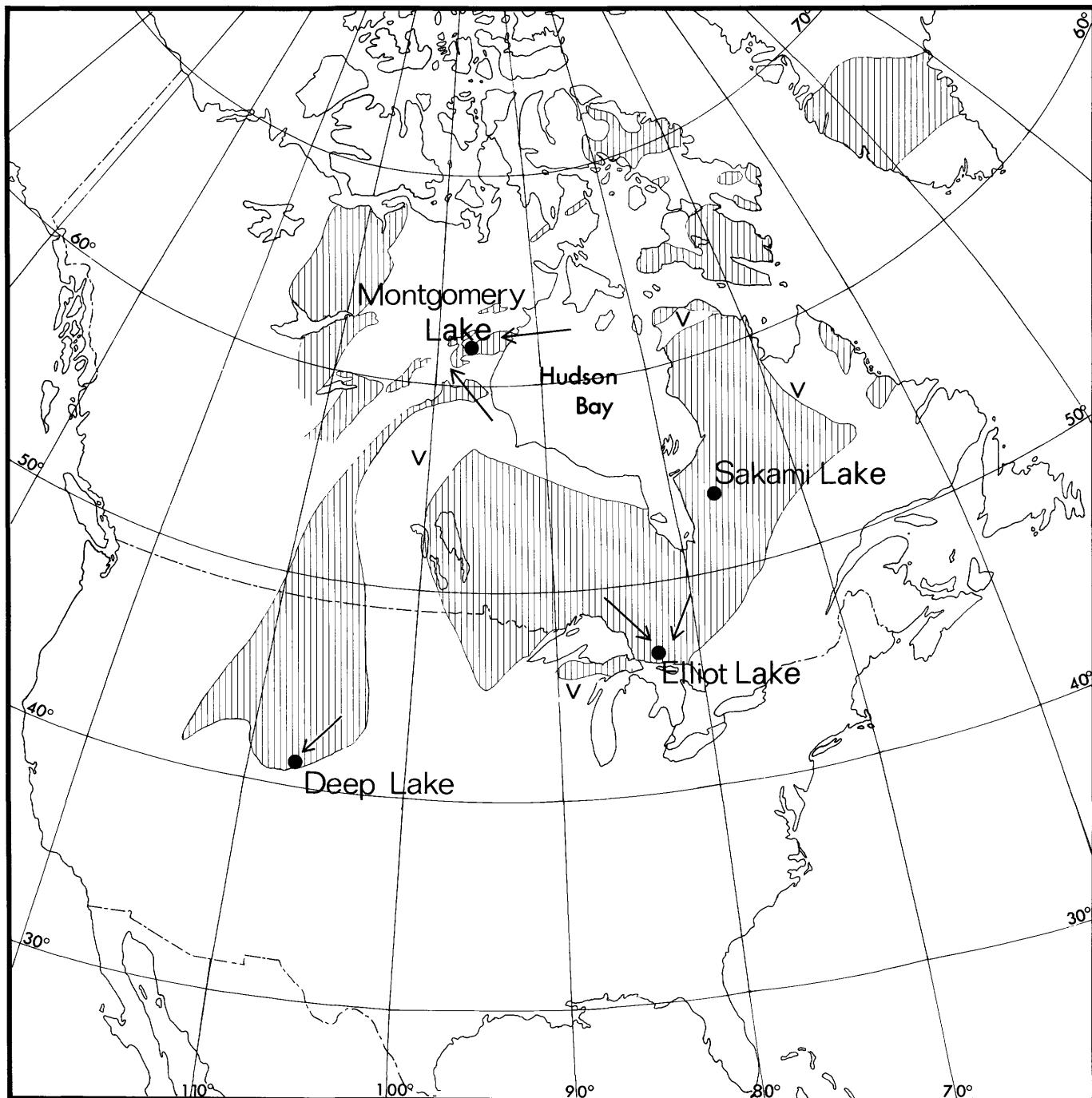


FIGURE 1.—Occurrences of pyritic uraniferous quartz-pebble conglomerates in North America. The distribution of Archean rocks at time of deposition of earliest Proterozoic rocks is indicated by the vertical pattern on the Superior, Slave, and Wyoming provinces and on other areas where rocks metamorphosed in Proterozoic time evidently included Archean basement. Most intervening blank areas were likely also part of the Archean craton, with possible exceptions of areas containing Aphebian volcanic rocks (V). Arrows indicate directions of sediment transport.

TABLE 1.—*North American successions containing bankets*

Unit	Thickness (kilometers)	Lithology	Unit	Thickness (kilometers)	Lithology	Unit	Thickness (kilometers)	Lithology
Huronian Supergroup, Ontario			Medicine Bow Mountains, Wyoming			Padlei area, Keewatin District		
Cobalt Group	4 -9		Libby Group (Lower part)	3-7		Hurwitz Group (lower part)	1.0 -3.5	
Bar River Formation	1	Orthoquartzite.	Surgarloaf Formation	~1.5	Orthoquartzite.	Kinga Formation:		
Gordon Lake Formation	.3 - .9	Varicolored, siliceous siltstone.	Lookout Formation	~.5	Varicolored, siltstone.	Whiterock Member	.2 - .3	Fine-grained orthoquartzite, white, pink.
Lorrain Formation	.9 -2.0	Orthoquartzite. Subarkose. <sup>1</sup>	Medicine Peak Formation.	~2.5	Quartzite <sup>1</sup> .	Maguse Member	.3 -1.0	Orthoquartzite, subarkose. <sup>1</sup>
Gowganda Formation	.9 -1.5	Arkose, siltstone <sup>1</sup> . Orthoconglomerate. Paraconglomerate.	Heart Formation	~1.0	Arkose. <sup>1</sup>	Padlei Formation	.3 - .5	Paraconglomerate. Siltstone.
			Headquarters Formation	~1.0	Siltstone. Paraconglomerate.			
Quirke Lake Group	.5 -1.5		Deep Lake Formation	>1.0		Montgomery Lake Group.	0 -1.6	
Serpent Formation	.25- .9	Arkose. Subarkose.	(upper part)	---	Quartzite. Pebby quartzite <sup>1</sup> .			Quartzite. Subarkose. <sup>1</sup>
Espanola Formation	.2 - .5	Siltstone. Limestone.			Limestone and. Paraconglomerate.			Siltstone. Subarkose. <sup>1</sup>
Bruce Formation	.05- .2	Paraconglomerate.						Para- and orthoconglomerate.
Hough Group	.3 -2.0		(central part)	---	Quartzite. Subarkose. <sup>2</sup>			
Mississagi Formation	.2 -1.2	Subarkose.			Siltstone.			
Pecors Formation	.03- .9	Siltstone.			Paraconglomerate.			Quartzite. Subarkose. <sup>1</sup>
Ramsey Lake Formation	.01- .2	Paraconglomerate.						
Elliot Lake Group	0 -2.5		(lower part)	---	Basalt.			
McKim Formation	0 -1.0	Siltstone.			Basalt conglomerate.			
Metunenda Formation	0 - .3	Subarkose. <sup>1</sup>			Paraconglomerate.			Acid volcanics.
Various volcanic units	0 -1.2	Basalt <sup>1</sup>			Siltstone.			
Various arenite units	0 - .5	Subarkose. <sup>1</sup>			Subarkose. <sup>1</sup>			

<sup>1</sup> Red beds or black sands (generally both) present.<sup>2</sup> Bankets (radioactive pyrite quartz-pebble beds) present.

much uranium have been weathered and leached from outcrops. Furthermore, Deep Lake lithologies are indistinguishable from those in the Huronian sequence beneath the Gowganda Formation, and they are also stratigraphically ordered in the same sequence. The two successions, dominated by various feldspathic quartzites, are both interrupted sequentially first by basalt, then by tillitelike paraconglomerate with argillite, and then by another paraconglomerate unit with limestone and siltstone, and finally both are capped by a thick tillite-bearing unit. No limestone unit has been found in Montgomery Lake rocks (fig. 2), but otherwise these are not much different.

Insofar as these very distinctive successions were produced by effects of sequential changes in climate in combination with epeirogenic pulses, it seems most unlikely that they could have been formed at different times.

#### AGE

The basket-bearing lower parts of the Huronian succession (Elliot Lake and Hough Groups) were deposited during an interval that began more than 2.35 aeons ago (Fairbairn and others, 1965) and ended no less than

$2.29 \pm 0.09$  aeons ago (Fairbairn and others, 1969) according to Rb:Sr isochron dates (1.39 decay constant) on volcanics in the Elliot Lake Group and argillaceous sediments in the stratigraphically higher Cobalt Group. The 2.35-aeon date is on Coppercliff rhyolite and it is considered a minimum date, as several Rb:Sr isochrons on basalts in the Elliot Lake Group yield poorly defined dates ranging from 2.2 to 1.9 aeons (Knight, 1966)—doubtlessly variably reduced due to metamorphism that must have affected the more rubidium-rich rhyolite as well, but to a lesser extent. The Huronian rocks are intruded by Nipissing diabase dated as 2.16 aeons old (Van Schmus, 1965; Fairbairn and others, 1969) and by the Murray granite, 2.23 aeons old (Gibbins and others, 1972). The 2.29-aeon date on Gowganda argillite is especially important as the Gowganda Formation contains the oldest reported red beds—hematitic shales and arkoses—and it is overlain by Lorrain quartzite, which is also hematitic, not pyritic, and which contains the oldest reported black sands.

Banket-bearing strata unconformably overlie Archean rocks 2.5-3.0 aeons old, excepting the vertically dipping metastrata at Sakami Lake whose relation to adjacent

rocks, which are generally believed to be 2.45–2.75 aeons old, has not been described. The basement in South Africa is at least 0.1 aeon older than basements beneath the Huronian, Deep Lake, and Padlei successions. Bankets in the African successions were deposited through an interval extending from 2.8 to 2.3 aeons (Niekerk and Burger, 1966, 1969). Volcanics overlying bankets in the Fortesque Group of Australia have been dated isotopically at approximately 2.2 aeons (Trendall, 1969).

Evidence that the Deep Lake Formation and the Montgomery Lake Group are correlative with the lower groups of the Huronian Supergroup has been presented above. If this is accepted, it can be concluded that all North American as well as South African and Australian bankets are in strata more than 2.2–2.3 aeons old. There is no reason to suppose that the remaining known bankets—those in Brazil—are any younger. They are certainly more than 1.8 aeons old (Ferreira, 1972) and are most likely older than nearby sequences containing extensive banded iron-formations like those in Australia, North America, and South Africa, which are about 2.0 aeons old.

#### EARLY PROTEROZOIC SUCCESSIONS LACKING BANKETS

The Cobalt, lower Libby, and lower Hurwitz Groups contain minor black sands and red beds but lack bankets and pyritic arenites. Otherwise, they are similar to lower Huronian Groups and Deep Lake cycles, which consist of basal paraconglomerate units overlain by fine-grained sediments and then by pyritic quartzites with bankets. The difference cannot be attributed to metamorphism as both upper and lower sequences have been metamorphosed together to greatly different extents in different areas. Thus we have evidence in three widely separated areas that coarse arenites deposited more than about 2.3 aeons ago contain interstitial pyrite rather than hematite and contain pyritic—or “yellow sand”—heavy-mineral accumulations (bankets) rather than black sands, whereas the reverse is true of younger arenites. The only cause that has been suggested for this is a change from an atmosphere lacking stable free oxygen to one containing significant amounts of oxygen (Roscoe, 1969, p. 83). Such a change should have been worldwide and probably instantaneous in terms of isotopically resolvable geological time. To test this we must examine the important early Proterozoic successions that were deposited after the basket-bearing successions and that are preserved through much more extensive areas.

The oldest post-Archean strata, apart from the few known small remnants of 2.8–2.2-aeon basket-bearing strata discussed above, are dominantly marine successions including dolomite, shale, banded iron-formation,

quartzite, and volcanics deposited and deformed more than 1.8 aeons ago. Clastics with red beds and black sands are present in many of these successions, generally in upper parts but in some cases in middle or basal sections. These rocks form some of the most extensive belts of folded Precambrian rocks. They include the Circum-Ungava (Kaniapiskau, Mistassini, Belcher), Marquette Range, Hamersley, Minas Gerais, Transvaal, and other successions with iron-formations, which collectively contain the bulk of the world's iron resources. Some extensive Aphebian successions 2.3–1.8 aeons old, however, lack significant iron-formations. Among these are volcanic-sedimentary strata in Manitoba believed to be about 1.85 aeons old, strata in the Coronation Geosyncline in the northwesternmost part of the Canadian Shield, and other sequences in western Canada. Lack of banded iron-formation is no doubt due to unfavourable environmental factors in many cases, but it is difficult to identify these. It is possible that some “barren” successions were deposited before or after an optimum time period for deposition of banded iron-formation, perhaps 2.1–2.0 aeons ago, as suggested by isotopic dates on volcanics associated with major iron-formations (for example, Hamersley). Wherever adequate geological or isotopic evidence is available, the relatively common Aphebian rocks are found to be significantly younger than the basket-bearing successions. Important examples are given in succeeding paragraphs.

Huronian strata at Sudbury were folded, metamorphosed, intruded by Murray-Creighton granite 2.2 aeons ago (1.39 decay constant, Gibbons and others, 1972) and deeply eroded prior to deposition of the Whitewater Group, which was intruded by the Sudbury Irruptive at 1.87 aeons (Krogh and Davis, 1974). In Michigan, 300 miles (480 km) west of Sudbury, the Marquette Range Supergroup was deposited locally atop metamorphic and granitic rocks about 2.1 aeons old, as well as on older Archean rocks (Banks and Van Schmus, 1972). In central Quebec, 400 miles (640 km) northeast of Sudbury, the Chibougamau Group, which is almost certainly correlative with the Gowganda Formation, was faulted, highly deformed, and eroded prior to deposition of the Mistassini Group (Roscoe, 1969, p. 116, 117, fig. 9). The upper part of the Libby Group in Wyoming contains dolomite, black shale, shaly magnetite iron-formation, and volcanics, all similar to rocks in the Marquette Range Supergroup. The metamorphosed basket-bearing rocks at Sakami Lake are older than arkosic red beds of the nearby, little-disturbed Sakami Formation, which is probably correlative with red-bed- and black-sand-bearing clastics composing the lowest unit of the Kaniapiskan Supergroup. The Transvaal System in South Africa unconformably overlies the basket-bearing

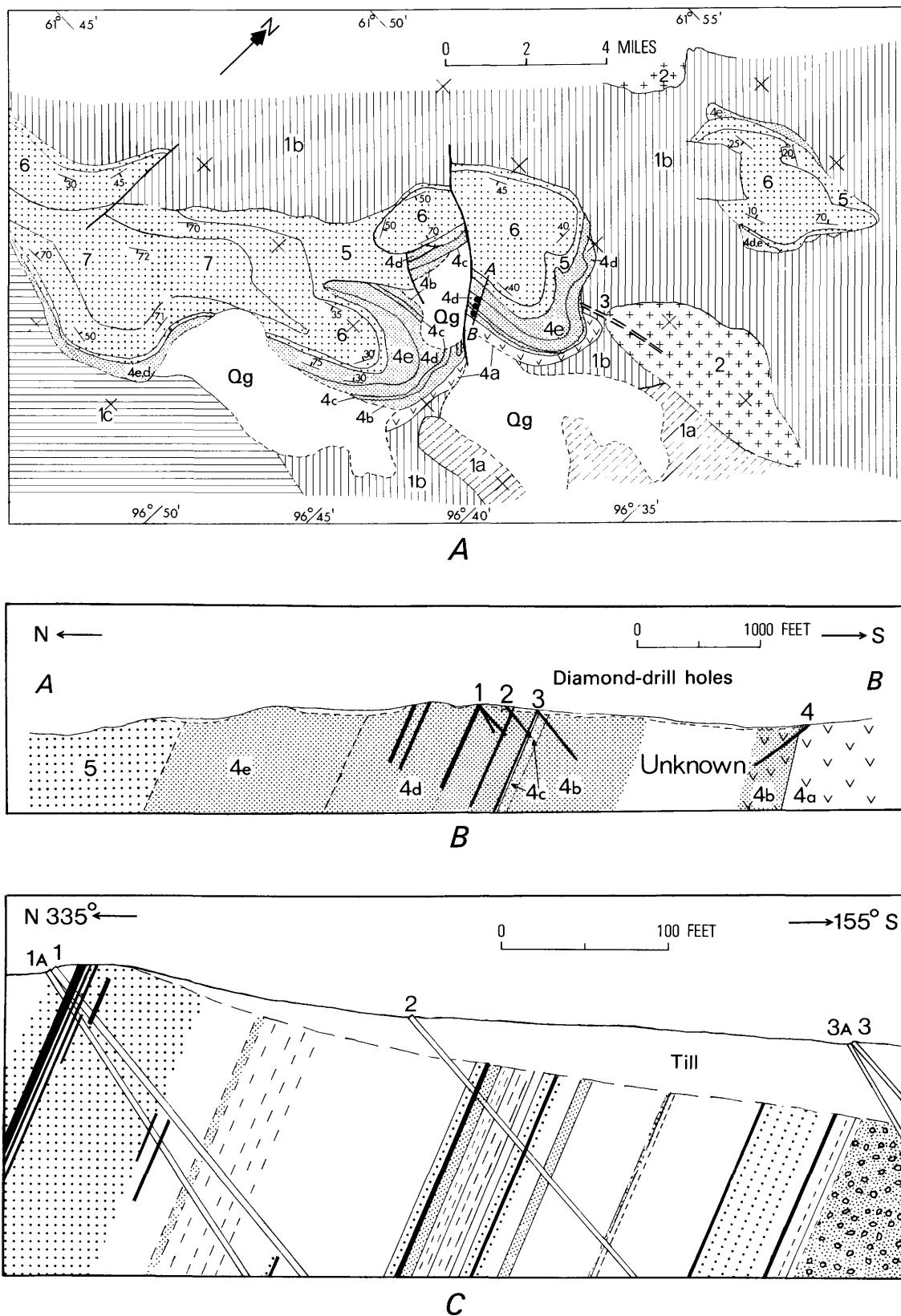


FIGURE 2.—Pyritic conglomerate beds in the Montgomery Lake Group, Keewatin District, N.W.T., Canada. A, Geologic map. B, Cross section A-B. C, Cross section showing detail for diamond-drill holes 1, 2, and 3.

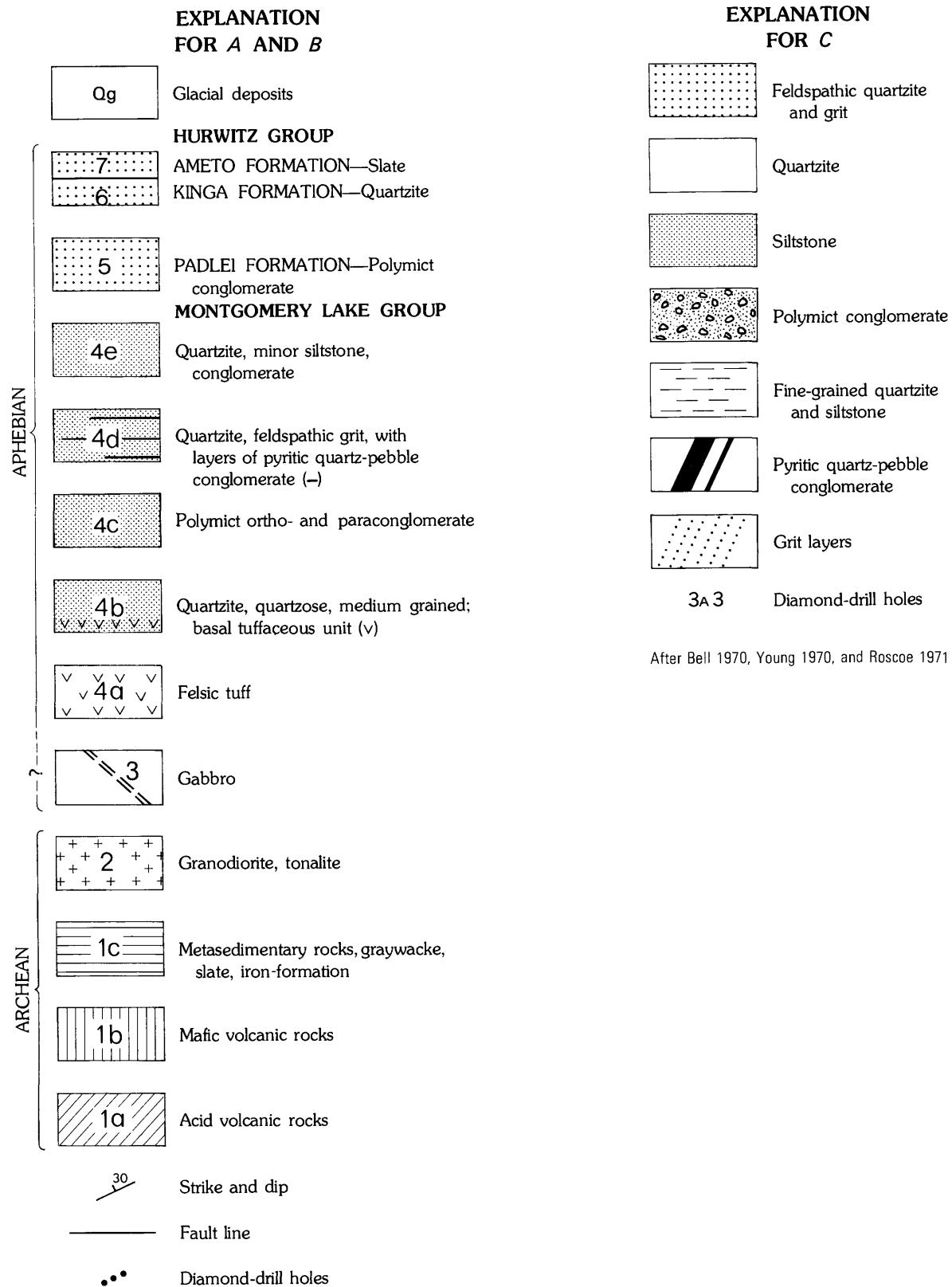


FIGURE 2.—Continued.

groups. In Australia the Hamersley Group overlies the Fortesque Group and contains volcanics dated at 2.0 aeons (Trendall, 1969).

## DEPOSITIONAL ENVIRONMENTS OF BANKETS NEAR ELLIOT LAKE

### LITHOSTRATIGRAPHY OF THE ELLIOT LAKE GROUP

This volume provides an opportunity to outline some previously unpublished concepts about the depositional environments of uranium-ore-bearing units near Elliot Lake. Data from deep exploratory drill holes west of the mines, in particular, have shed new light on the history of deposition of the Elliot Lake Group, strengthening the conclusion that the rocks were deposited subaerially. The data cannot be outlined in detail here, nor is it possible to repeat stratigraphic and petrographic descriptions that may be found elsewhere. A brief outline of stratigraphic relations is presented, however, to set descriptions of relations in the Elliot Lake area in perspective.

The uranium ores and principal prospects in the Huronian belt are in the Matinenda Formation of the Elliot Lake Group (table 1); pyritic uraniferous conglomerate layers are also present, sparingly, in the Livingstone Creek Formation, which is in the lower part of the Elliot Lake Group west of Elliot Lake, and within sequences of basalt flows in the Elliot Lake Group. The Mississagi Formation of the overlying Hough Group also contains pyritic uraniferous pebbly layers. The two basket-bearing groups extend through a distance of some 200 miles (320 km) in the Sault Ste. Marie-Sudbury region, and their combined thickness is as great as 20,000 ft (6,100 m) in the southeastern part of the belt.

The Elliot Lake Group includes all strata that nonconformably overlie Archean rocks and underlie the regionally extant Ramsay Lake Formation, a paraconglomerate that forms the basal unit of the Hough Lake Group. Major units of the Elliot Lake Group—volcanic rocks, coarse-grained subarkose and argillaceous rocks—are lenticular. Many of the outcrop areas of the group are separated, one from the other, so that it is not possible to establish the sequence of deposition of all volcanics and arenites throughout the belt.

The McKim Formation, comprising fine-grained argillaceous quartzite, siltstone, and argillite, forms the uppermost unit of the Elliot Lake Group in the eastern part of the belt near Sudbury. Here it is several thousand feet thick, and it overlies several thousand feet of volcanic rocks, including the Stobie Formation—basalt with some intercalated sediments—and the less extensive Coppercliff Rhyolite Formation. Subarkose called the Matinenda Formation (Roscoe, 1957a, b) forms most or all of the group in some central parts of the belt near

Blind River, Elliot Lake, and Quirke Lake, but sequences in flanking areas include basalt flows. A tongue of McKim Formation extends westward into this area over some parts of the Matinenda Formation, and a wedge of Matinenda Formation extends easterly towards Sudbury between the volcanics and the McKim Formation. In the western part of the belt, basalt flows of the Thessalon Formation overlie subarkose of the Livingstone Creek Formation. The Elliot Lake rocks are bevelled by an erosion surface and are overlain by the Ramsay Lake Formation, which is considered to be a tillite.

### MATINENDA FORMATION

In the Elliot Lake area the Matinenda Formation has been subdivided into three coarse-grained subarkose units, successively the Ryan, Stinson, and Manfred Members (Roscoe, 1969). A fourth lenticular subarkose unit, the Keelor Member, has been identified 15–20 miles (23–32 km) west of Quirke Lake (figs. 3, 4). The Ryan, Manfred, and Keelor Members contain abundant pyritic, radioactive quartz-pebble beds and are gritty, variegated, greenish coloured, and pebbly throughout. The Stinson Member is pale grey, relatively well sorted, and contains conglomerate beds only at its base. These beds include polymict, as well as oligomict, orthoconglomerates and they contain little pyrite or radioactivity. Some of the conglomerates, evidently deposited as open-framework cobble gravels, contain authigenic quartz, carbonate, pyrrhotite, and pyrite. The subarkose units are built of crossbeds that filled troughs, generally 1–4 ft (0.3–1.2 m) wide. Trends of troughs and dips of foresets are unimodally southeast in the eastern part of the area but are more southerly in outcrops 15 miles (24 km) west of Elliot Lake.

Basalt flows in the Elliot Lake Group west of Quirke Lake have been called Dollyberry Lake volcanics (Robertson, 1969). They are believed to have been erupted after deposition of the Ryan Member but prior to deposition of other Matinenda members. If the general practice of assigning formation rank to volcanic units in the Elliot Lake Group, and of excluding nonarenaceous rocks from the Matinenda Formation, were to be followed, it would create some confusion in discussions. For this reason, the Dollyberry Lake volcanic unit is referred to herein as "Dollyberry basalt" and treated, informally, as a member of the Matinenda Formation. Two other significant units are present in the area, but neither is along the line of section shown in figure 3. One is a wedge of volcanic rocks overlain by the Ryan Member east of the Nordic ore zone. Robertson (1961) mapped this wedge as questionable Archean but later (Robertson, 1973) he suggested that it is probably part of the Elliot Lake Group. The other additional unit

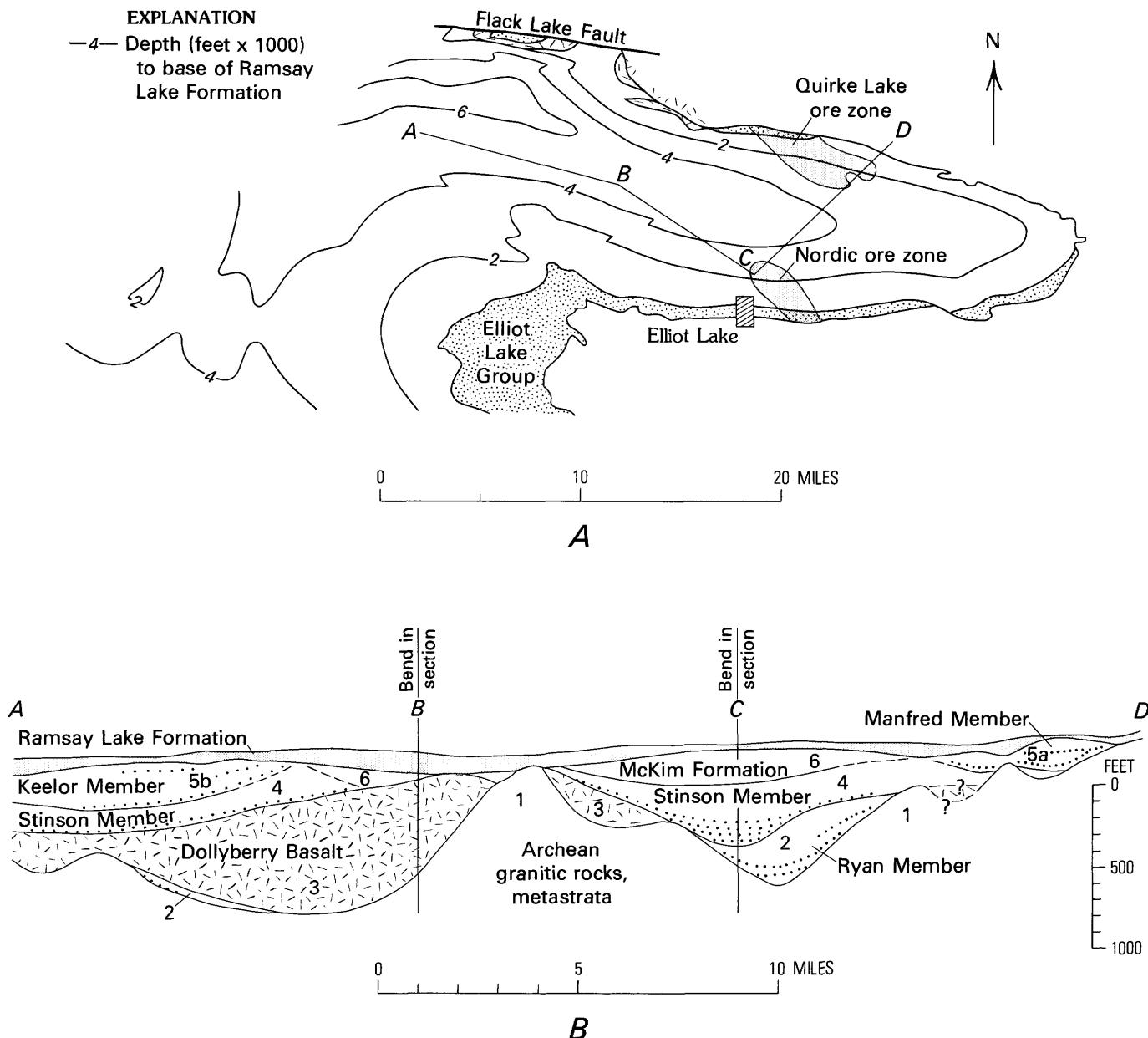


FIGURE 3.—Stratigraphic relations in Elliot Lake Group near Elliot Lake. A, Map showing outcrops of Elliot Lake Group and depth to the base of the Ramsay Lake Formation. B, Schematic cross section near Elliot Lake showing relations among arenaceous members of the Matinenda Formation, Dollyberry basalt, and McKim Formation in the Elliot Lake Group.

is a lens of poorly sorted arkosic rocks that outcrop through a length of 5,000 ft (1,500 m) at Crazy Lake, 17 miles (27 km) west of Quirke Lake, along the north limb of the syncline. The Crazy Lake unit is truncated by the Flack Lake Fault so its base is unknown, but it is as much as 500 ft (150 m) thick. It is capped by erratically uraniferous, poorly sorted, pyritic oligomictic conglomerate and overlain by Dollyberry basalt.

#### CONGLOMERATIC ZONES

The uranium-bearing conglomeratic zones in the Ryan, Manfred, and Keelor Members consist of quartz-pebble conglomerate, pebbly grit, and coarse subarkosic interlensed through as much as 100 ft (30 m) of strata. Pebble-rich sections, with lenses containing well-packed, well-sorted pebbles, alternate with sections con-

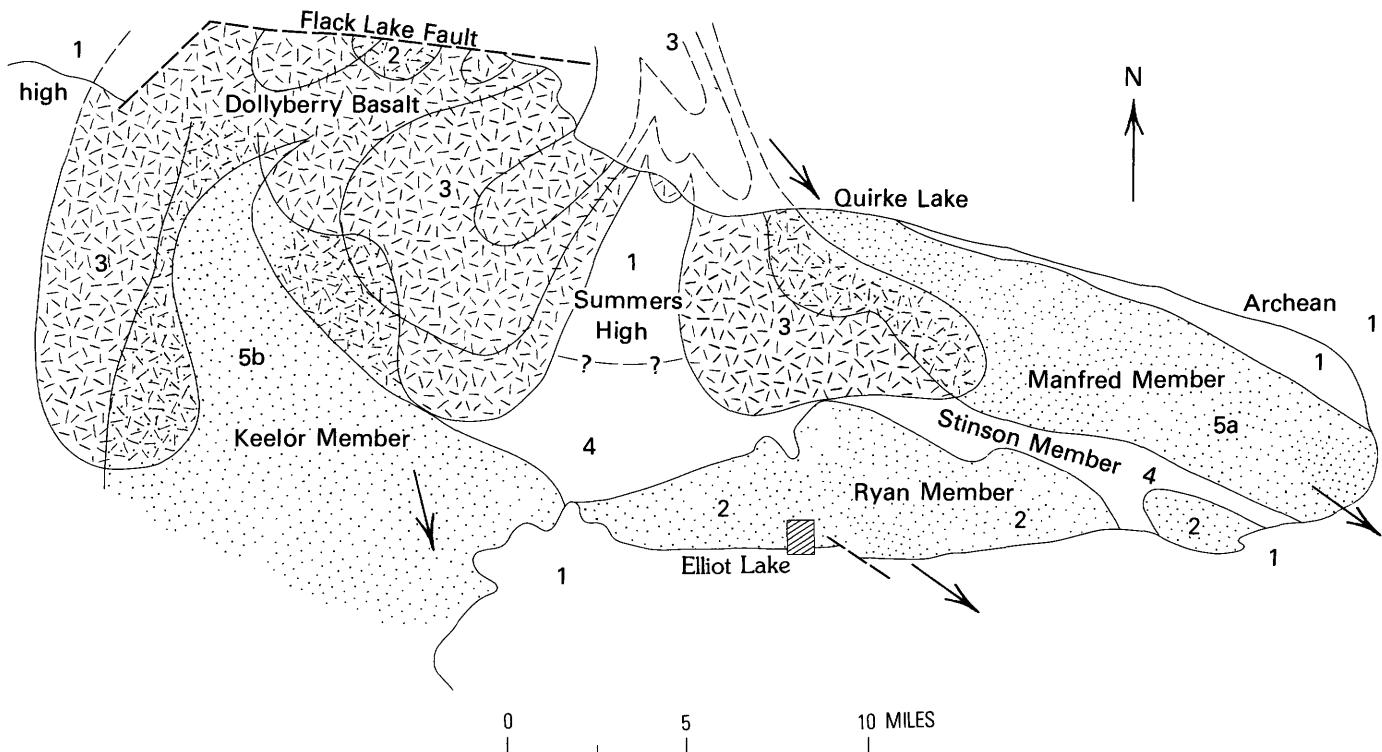


FIGURE 4.—Subsurface distribution of conglomeratic and volcanic strata in the Elliot Lake Group near Elliot Lake. Designations of units by number, their stratigraphic positions, and depths to the base of the Ramsay Lake Formation are as shown in figure 3. Unit 4 is shown only where it overlies Archean basement (1); elsewhere, it overlies unit 2 and southerly areas of unit 3, and it underlies southerly areas of units 5a and 5b. The latter units overlap unit 3. The most closely spaced dots indicate areas with the most abundant conglomerate beds. Arrows indicate paleocurrent directions. Contour-like lines of unit 3 approximate 500-ft (152-m) thickness intervals.

taining few pebbles. Some of the latter sections can be traced great distances and define stratigraphic levels within the zones, perhaps better than the pebble-rich sections, which locally are called "reefs." The reefs are not layers but, rather, are concentrations of lenses containing well-packed pebbles. The conglomerate lenses are crossbedded and are similar to troughs in pebble-free subarkose. They also have similar trends and foreset dips—southeasterly in the Ryan and Manfred Members. Cross laminae are commonly graded, with pebbles at their bases. In some cases, pebbles evidently rolled down successive foresets to form a basal zone of pebbles imbricated downstream. Most of the longest axes of pebbles are either normal or parallel to foreset dips, indicating that many were "weathercocked," rotated parallel to current flow, after they ceased rolling. Pebbles are similarly oriented where they are tightly packed in lenses that lack arenaceous or pyritic cross laminae (Pienaar, 1963). It seems likely that these represent the lower parts of troughs whose upper, more arenaceous, parts were planed off. The richest reef sections, containing the greatest concentrations of pyrite and other heavy-mineral grains, probably represent suc-

cessions of such remnants of basal pebble-rich parts of troughs.

Pebbles are generally smaller in successively higher reefs and in each reef become progressively smaller in the downstream direction. In reefs mined at Quirke Lake they decrease from 5 cm or more in the most proximal areas to 2 cm in distal areas less than 3 miles (5 km) to the southeast. Pyrite grains show corresponding decreases from about 1 mm or more to less than 0.3 mm. Uraninite grains 0.05-0.20 mm (mostly 0.1 mm) in diameter are restricted to proximal conglomerates with pebbles larger than 3 cm. Other heavy minerals, mainly titaniferous grains, monazite, and zircon, constitute less than 5 percent of total heavy minerals in the most proximal, largest pebble conglomerates but increase in progressively more distal conglomerates to more than 15 percent. Titaniferous grains are as much as 1.5 mm in diameter in proximal conglomerates, less than 0.3 mm in the most distal ones. Monazite grains, about 0.3 mm in diameter, are most abundant, as abundant as titania grains, in upper reefs and near the distal margin of the Quirke Lake ore zone. Zircon grains, less than 0.3 mm in diameter, are most abundant in yet more distal, sub-

marginal parts of the zone and in uppermost small-pebble conglomerate beds. Granules and smaller particles of quartz, potash feldspar, and mica in matrices of large-pebble, as well as small-pebble, conglomerates are notably poorly sorted in contrast to pebbles and heavy minerals.

The conglomerate zones represent sites of remarkably efficient hydrodynamic sorting, wherein the largest and heaviest mineral grains, including pyrite and uraninite, were concentrated in the most energetic environments along with the largest quartz pebbles (Roscoe, 1969; N. J. Theis, personal commun.). Individual rich conglomerate lenses were likely formed in part from reworked detritus initially deposited upstream as pebbly grit, and much fine detritus was undoubtedly winnowed out of the conglomeratic zone as it was built up. Nevertheless, the conglomerates do not represent lag gravels in any strict sense. All size ranges of detritus were transported and deposited together. The tremendous overall abundance of pebbles and heavy minerals in the zones must have been produced by concentration of these materials throughout prolonged transport.

Proportions of gravel to sand carried into the depositional area fluctuated and diminished as the conglomeratic zones were built up. Initially, floods carrying abundant coarse gravel alternated with floods carrying only small pebbles in addition to abundant sand and finer detritus. Stream competency waned as the zones built up and finally only infrequent, exceptional floods carried gravel onto upper parts of deposits as much as 300 ft (90 m) thick.

Two different types of conglomeratic zones can be distinguished in the area, despite the fact that erosion in Matinenda time, as well as later, has removed important parts of all zones explored by drill holes. The Nordic (Ryan) zone rests directly on basement rocks and appears to have been deposited along a valley controlled by basement topography and perhaps also by syndepositional faulting. The small Pronto deposit 15 miles (24 km) south of Elliot Lake is probably a remnant of a similar "valley" deposit. The much larger Quirke Lake (Manfred) zone heads atop Archean rocks and the eastern margin of Dollyberry lava fields, extends southward overstepping the northern limit of the Stinson Member, and spreads out and thins in a fanlike manner over the relatively smooth top surface of the Stinson. The conglomerate-rich Keelor Member has relations similar to the Manfred Member at Quirke Lake. It was evidently deposited as a series of coalescent fans where streams debouched from lava fields and Archean uplands onto a more gently sloping surface of a Stinson sand "plain," which was probably an older bajada deposited by streams that did not carry gravel.

The Nordic zone, as outlined by the area mined within

two closely spaced reefs near its base, is about 4.5 miles (7.2 km) long and about 1 mile (1.6 km) wide, trending southeast parallel to the paleocurrent direction indicated by foreset dips. It is bounded to the east by a paleoridge formed by a band of Archean cherty iron-formation. The main ore reefs abut against this paleorise. A stratigraphically higher reef with economic potential, however, extends easterly over the higher paleosurface and is overlain by subarkose containing many thin subeconomic reefs. All of the conglomerate beds feather out rapidly towards the west in the downstream shadow of the "Summers High," a considerable hill of Archean granite that was not covered by an appreciable thickness of Elliot Lake strata.

Sizes and proportions of heavy minerals vary with sizes of pebbles in the Nordic zone as they do in the Quirke Lake zone, but the patterns of pebble-size changes within individual reefs in the two zones are significantly different. There is little decrease in pebble size from the most proximal to the most distal part of the Nordic ore zone. Instead, pebble sizes and uraninite abundances decrease laterally both east and west from the central axis of the zone. Ore throughout the length of the chutelike zone is like that found through a much broader, entirely proximal, part of the Quirke zone. The eroded southerly extension of the zone, like its flanks, must have included abundant smaller pebble conglomerate with heavy-mineral suites relatively rich in titania, monazite, and zircon, similar to conglomerates in the medial and distal parts of the Quirke Lake zone. This suggests that a zone like the Nordic zone could have changed downstream into one like the Quirke Lake zone.

#### DOLLYBERRY BASALTS

The Dollyberry basalts require special consideration because of their close spatial and temporal relation to the uranium ore deposits near Elliot Lake. Rifting was likely the main link between basalt eruptions and the beginning of deposition of Huronian clastics. Tectonic movements accompanying eruptions, including development of fault scarps, as well as the lava fields or volcanoes themselves, likely had important influences on drainage systems and on locations of sites of gravel deposition. The distribution of the lavas provides clues concerning topography both prior and subsequent to their eruption and thus contributes to interpretations concerning locations and histories of gravel-bearing paleostream systems. Some workers have suggested that the uranium ores, or pyrite within them, might be genetically and chemically related to contemporaneous volcanism (Robertson, 1973; Ruzicka, this volume, chap. V). This idea can be discounted for many reasons,

among them the evidence presented in following paragraphs, that the Dollyberry eruptions were not contemporaneous with deposition of any of the important bankets at Elliot Lake. Moreover, bankets were deposited in Hough Group rocks long after the last eruptions occurred in Huronian rocks.

The lavas are amygdaloidal, and closely spaced scoriaceous and brecciated zones suggest that many flows are thin. No pillows have been noted. The rocks are dark grey to dark green for the most part, but bleached and argillized zones are found in many scoriaceous and brecciated sections, presumably flow tops. This alteration is similar to that found in greenstone at the Archean-Huronian nonconformity and to that found rimming greenstone and basalt clasts within Huronian strata. It is considered to represent interflow weathering of lava surfaces.

Dollyberry basalts outcrop along the north limb of the Quirke Lake Syncline 4–15 miles (6.5–24 km) northwest of Quirke Lake. They overlie paleosol-mantled basement rocks in this outcrop belt, except in its northwesternmost part, where they overlie more than 500 ft (150 m) of Crazy Lake arkose and are truncated by the Flack Lake Fault. The basalt pile is thicker, locally more than 1,000 ft (300 m) thick, along the outcrop belt than in subsurface drill intersections to the south. A main lobe extends 8 miles (13 km) south along the west flank of the Summers High, and a thinner lobe of flows extends south along the east flank of this paleohigh (fig. 4). The eastern lobe, as presently outlined, ends very near the northernmost known limit of the Ryan Member and the Nordic ore zone. The lavas must have flowed southward around the Summers High from eruptive centres near or north of the Flack Lake Fault.

West of the Summers High, Dollyberry basalts thin southward at an average rate of more than 200 ft (60 m) per mile. The surface of the lava fields, barring northerly tilting, must have had a correspondingly high slope gradient toward the south when Stinson sands and Keelor gravels were deposited on their lower reaches. It also had considerable local relief, as revealed by intersections in deep drill holes. In some of these, Dollyberry lavas are overlain by Stinson subarkose containing few pebbles. In others, as much as 70 ft (21 m) of basalt breccia or sharp-stone conglomerate grade up into basalt- and quartz-cobble conglomerate. The conglomerate matrix contains quartz and carbonate, as well as granules and argillitic detritus. It also contains concentrations of pyrite or pyrrhotite with traces of chalcopyrite not accompanied by radioactive minerals. The sulphides are clearly authigenic and are most commonly found at the top of the conglomerate or in separate beds a few feet above it within Stinson subarkose. Many basalt clasts are variably argillized. In

places, conglomerate with intensely altered clasts was covered by a thin layer of green argillite, representing redistributed residual clay, prior to deposition of Stinson sand. It is noteworthy that iron sulphide, not ferric oxide, is concentrated at these obviously weathered surfaces and that uranium is not concentrated along with sulphides, as it is in bankets.

The basal conglomeratic section of the Stinson Member contains abundant basalt clasts where it overlies and channels the Ryan Member, as well as where it overlies Dollyberry basalts. The abundant conglomerates in the Ryan Member do not contain basalt clasts, so the Ryan is considered older than the Dollyberry basalts. Only a few holes have been drilled through the basalts. Most of these passed directly into paleosol-mantled granite, but several holes intersected a few feet of sorted arkosic sediments with quartz clasts, pyrite, and radioactive minerals between the basalt and the paleosol. These sediments, and the thick Crazy Lake arkose, are likely remnants of alluvial deposits correlative with the Ryan Member.

The eastern lava lobe is believed to have filled a broad valley previously occupied by the stream system that deposited the Ryan Member. The northern limit of this member is not well defined; it is largely an erosional rather than a depositional limit. Evident post-Dollyberry erosion may have obliterated evidence of more extensive pre-Dollyberry erosion of the Ryan Member.

#### DEPOSITIONAL HISTORY: A CONCEPTUAL MODEL

Interlenticulated units forming the Elliot Lake Group were deposited about 2.4 aeons ago on successive eroded paleosurfaces that were shifted and warped in an easterly-trending fissured zone within the Superior Craton. The clastic sediments, largely clay and quartz clasts, were shed southward from weathered crystalline rocks in a more central part of the rising craton. The coarsest detritus was entrapped in the rifted zone, and volcanic rocks, principally basalts, were extruded along fissures.

The Ryan Member, the oldest clastic unit in the Elliot Lake mining area, was deposited by braided streams in a valley that trended and broadened southeastward. Early Ryan deposition drew on a plentiful nearby quartz-gravel supply that was then exhausted and not replenished from distant sources for a very long time. The coarsest gravels, containing abundant detrital pyrite and uraninite, were deposited early along a chutelike central section of the valley. Subsequently, coarse sand and fine gravel were deposited over a broader area.

Uplift to the north cut off the supply of sand and resulted in scouring of Ryan sand and gravel from the

northern part of the valley. This was followed by eruptions of Dollyberry basalt 7-15 miles (11-24 km) northwest of the present position of Quirke Lake, producing a volcanic edifice built up as much as 1,500 ft (450 m) above its Archean floor. Lavas flowed southward onto lower ground, filling the northern part of the Nordic valley. The rough, locally steeply-sloping, lava surfaces were weathered and eroded under anoxic conditions during a period when little or no detritus was carried into the area.

Resumption of southerly detrital transport was accompanied by washing, or continued washing, of petromict gravels from nearby Dollyberry and Archean uplands onto the scoured and locally channelled surface of the Ryan Member, but little gravel was carried from the main provenance. Deposits of coarse, sorted Stinson sands were built up, culminating in development of a smooth surface sloping gently southward from bases at steeper slopes and spurs of weathered Archean rocks and Dollyberry lavas.

Deposition of the Manfred and Keelor Members began abruptly when streams suddenly began to carry abundant quartz gravel in addition to finer detritus southward over the Archean surface and the topographically lower parts of Dollyberry lava fields. Gravels were deposited where streams debouched from confined watercourses at the piedmont angle and spread out over the Stinson bajada. Distributary fans built up, out onto the sands and back onto the igneous rocks. The finest gravel and lightest grains of heavy minerals were carried to the most distal parts of fans. Slopes decreased as the deposits aggraded. Stream competency waned, and the proportion of sand to gravel in flood deposits increased.

Medium- to fine-grained argillaceous clastic sediments of the McKim Formation were deposited atop the Stinson Member where it overlies the Ryan Member. The lack of recognized Manfred sands in this area is noteworthy (fig. 3). Manfred outwash may have been diverted entirely to the southeast, so that no fine gravels were deposited near the Nordic zone, or Manfred sand and fine gravel may have been eroded off the Stinson surface here, or—least likely—Manfred subarkose with beds of small-pebble conglomerate may grade abruptly into much finer grained argillaceous quartzite of the McKim Formation. The McKim Formation was likely deposited partly in a flooded eastern section of the rifted zone near Sudbury, but a fluvial depositional environment seems more likely for the tongue at Elliot Lake.

The Elliot Lake strata, particularly those in proximal localities, as at Quirke Lake, were tilted, warped, and bevelled prior to glaciation and deposition of Ramsay Lake tillite, which forms the basal unit of the Hough Group. It is noteworthy that the Ramsay Lake Formation is thinner where it overlies the richest parts of the

Quirke Lake and Nordic ore zones than it is in areas flanking the conglomeratic zones. This is consistent with the idea that the ore zones were parts of fans with positive relief.

This conceptual model will no doubt require important modifications. Nevertheless, it serves to dramatize some critical limitations that can be imposed on choices of possible ways that the nonoxidized uranium deposits, sediments, and paleosols could have been deposited and modified. The depositional environment could not have been more different from environments of deposition of younger euxinic deposits. It was not at all different physically from environments of deposition of some of the most oxidized younger sediments. The sedimentary particles were exposed to chemical attack by surface and ground water saturated with atmospheric gases throughout a history beginning with their births in well-drained soils lacking vegetative cover and continuing even after their final burial beneath channels of swiftly flowing streams. Any organisms that might have been present would have been subject to similar attack. The deposits were not laid down and buried near areas where biotic or volcanic activity might have produced vast quantities of reductants capable of scavenging oxygen from all aerated surface and ground waters. Neither were they formed in an extremely unusual tectonic environment that has not recurred on a significant scale since early Proterozoic time. Clearly, the widespread paleosols were formed and the sediments were deposited subaerially under an atmosphere that contained no significant amount of free oxygen.

#### COMPARISON OF OLDER AND YOUNGER TERRIGENOUS SEDIMENTS

Extreme proximal depositional environments like those of Elliot Lake ores were not an essential condition for the formation of bankets. Witwatersrand ores, stratigraphically far above basement, may have been formed in more distal parts of fluvial systems. Subeconomic bankets were evidently deposited in a wider range of environments similar to various environments of deposition of black sands in hematitic clastics less than 2.3 aeons old. Some examples of varied pyritic suites are outlined in following paragraphs. Obviously, it is not possible to prove that the pyritic suites and their closest nonpyritic counterparts are mutually exclusive and time bound, but extensive exploration of terrigenous sediments throughout the world has not provided evidence to the contrary.

Weakly radioactive layers with small pebbles are found through several thousands of feet of Mississagi, Deep Lake, and Montgomery Lake strata, so depositional sites must have extended great distances downstream from fall lines. Beds with quartz pebbles as

much as an inch or more in diameter in the Montgomery Lake Group (fig. 2) are interlayered with coarse- to fine-grained quartzite and with siltstone. Some beds overlie siltstone and form bases of upward-fining cycles likely deposited in a relatively distal fluvial environment. Pebble beds atop the upward-coarsening transition zone between Pecors argillite and Mississagi subarkose may have been deposited in topset beds of a prograding delta. Conglomerate beds interlayered with Huronian basalt flows represent yet another depositional environment.

Other variations amongst basket associations include differences in composition of detritus other than pebbles. Sand in conglomerate matrices and in associated beds range from protoquartzite to arkose (for example, locally in Ryan Member) and to subgreywacke (for example, at Manfred-Ramsay Lake contacts and perhaps in some Montgomery Lake beds). Although volcanics are found in most host successions, their associations with baskets range from intimate to remote. The Manfred and Keelor Lake conglomeratic zones were deposited considerably later than were lavas in the Elliot Lake district. Pyritic beds in the Mississagi Formation, and through the greater parts of other host formations, were not coeval with any known volcanism. Volcanic processes or rocks thus cannot be imagined possible sources of sulphur, other basket constituents, or pervasive reducing agents.

Younger quartzose arenites deposited in various terrestrial environments are characterized by red beds, and they contain detrital heavy-mineral suites dominated by iron oxides. Concentrations of heavy minerals are black sands rather than baskets. The difference cannot be attributed to differences in depositional environments or to differences in postdepositional histories. Black-sand-bearing successions, like older pyritic successions, were deposited in a range of environments—alluvial fans, braided streams, flood plains, deltas, beaches, volcanically active areas, and tectonically stable and tectonically unstable areas. They are found in rocks that have been subjected to all sorts and degrees of alterations and metamorphic processes. No explanation more plausible than oxyatmovation has been offered for the time-bounded character of the two suites of terrestrial sedimentary rocks.

The concept that baskets are heavy-mineral concentrations deposited under an anoxic atmosphere provides a rationale for the occurrence of abundant detrital pyrite and locally abundant detrital uraninite within them, but it does not automatically dispel all mysteries concerning the origin of these and other minerals in the baskets. These problems do not fall within the scope of this paper, but one important phenomenon must be mentioned briefly.

A considerable proportion of the pyrite must have originated as iron oxides and iron-titanium oxides in Archean rocks in addition to larger amounts that originated as pyrite, and some was formed authigenically within the baskets (Roscoe, 1969). The scarcity of iron oxides compared to pyrite in paleosols and the lack of coarsely bladed integrowths of pyrite and anatase in the baskets suggest that iron oxides were sulphidized in the soils and that ilmenite-bearing grains were altered before rather than after deposition of the baskets. The excess sulphur required for the pervasive sulphidization must have been of atmospheric origin and, unless unreasonably high concentrations of hydrogen sulphide are postulated in the paleoatmosphere, anaerobic bacteria must have played an important role in the transformations. Some of the uranium minerals also might have been formed originally in the soils, but this seems unlikely in the case of uraninite that contains thorium and rare earths in amounts similar to those in uraninites in igneous rocks.

## REGIONAL TECTONIC SETTINGS OF BANKETS

The depositional and erosional features described above reflect tectonic movements in combination with climatic changes and volcanic eruptions. Matinenda alluvium was likely collected and transported southerly over an extensive land surface following, perhaps, a slight increase in regional slope. We see in the formation only the coarsest fraction of this weathered detritus—mainly the largest quartz grains that were present in eroded Archean granitoid rocks. These, along with heavy minerals and pebbles of vein quartz, were deposited in some sort of tectonic trap likely formed along ruptures. The bulk of material eroded from the uplifted continental core, consisting of fine-grained siliceous, feldspathic and argillaceous detritus, and dissolved salts, was carried farther south down the continental paleoslope.

Cyclic similarities between Huronian Groups above the Elliot Lake Group have been emphasized by Roscoe (1957a, b, 1969, 1973) and by Frarey and Roscoe (1970) and have been attributed to epeirogenic movements attendant on glacial loadings and unloadings superimposed on more general uplift of a central part of the Archean craton. The uplifts may have initiated the glacierizations marked by tillites and by some polymict orthoconglomerate and arkose, but the preponderance of submature and even supermature clastics—mainly coarse quartzites—through most of the Huronian succession militates against any possibility that mountainous terrains were carved in Archean rocks near the Huronian belt. The tillites are overlain by fine-grained sediments and, in the Quirke Lake Group, limestone.

These were likely deposited in large lakes and short-lived seas flooding the depressed craton. Subsequent rebound resulted in resumption of deposition of coarse clastics in the Huronian belt, with the bulk of detritus and dissolved cations (of sodium, calcium, magnesium, iron) and silica carried farther down the continental paleoslope. The arenites that dominate the Huronian succession thicken southward through the Huronian belt, so the Archean platform must have extended well to the south into what is now the tectonically younger Grenville Province (Frarey and Roscoe, 1970).

The distribution of Archean rocks in North America, as well as basket localities and general sediment transport directions therein, is shown in figure 1. The Superior, Slave, and Wyoming provinces, where rocks were not significantly metamorphosed more recently than 2.5 aeons ago, are not distinguished from remaining areas where Archean rocks were metamorphosed or remetamorphosed about 1.8 aeons ago (Hudsonian and Penokean orogenies). Intervening areas were likely also cratonized in Archean time. Moreover, Archean basements to Huronian and Medicine Bow strata must have once extended well south of Superior and Wyoming provinces. A few areas containing volcanic rocks about 1.9 aeons old, designated V in figure 1, may represent rifts wherein some Aphebian rocks were deposited on oceanic rather than continental crust.

Crossbed dips, southeasterly in Huronian quartzites, southwesterly in Deep Lake quartzites and westerly in Montgomery Lake quartzites, suggest a drainage divide or uplift axis extending north- to northeastward through western Ontario. The North American basket-bearing belts trend northeast, excepting the western part of the arcuate Huronian belt. They are at an angle to the hypothesized uplift axis, not peripherally disposed around the uplift or around any likely cratonic block. It is suggested, therefore, that they mark intracratonic ruptures that were sites of vulcanism and of crustal flexuring that entrapped and eventually folded continental sediments.

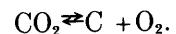
The known Paleoaphebian belts were no doubt very much more extensive originally, and there were probably others. Great volumes of Archean granitic rocks must have been eroded to provide coarse quartz grains in Huronian and other thick quartzite wedges (Frarey and Roscoe, 1970, p. 154). If provenance areas were limited and submountainous as suggested here, differential uplift and erosion must have been very considerable and occurred through a prolonged period.

## DISCUSSION

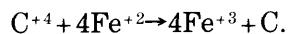
The proposition that banks were deposited subaerially under an anoxic atmosphere is susceptible to the criticism that some of the evidence for this is

negative. For instance, the observation that basket-bearing pyritic quartzites and black-sand-bearing red beds are mutually exclusive and separated in the geological column by a time barrier at 2.3 aeons carries risks that exceptions may be found. Some have in fact been suggested (for example, Ruzicka, 1971, p. 148-158; Bottrill, 1971, p. 82). However, none of these has been shown to have diagnostic features shared by all the ancient banks but not found in indisputably epigenetic deposits. Most allegedly basketlike occurrences of pyrite and uranium in younger rocks are irrelevant by the advocate's own account. So that I may be absolved from suspicion of attempting "proof by definition," let us consider a comparison that C. F. Davidson (1965, p. 1198) made between locally "somewhat metamorphosed" Permian conglomerates "locally mineralized with uraninite, pyrite, and other sulphides" and Witwatersrand ores. In this, he ignored the facts that ancient banks are all found through great successions of strata, not as local impregnations of pyrite in normally nonpyritiferous beds, and that alleged younger analogs are never extensive, never geochemically or mineralogically similar to banks, and never difficult to recognize as epigenetic.

Doubts about an early Proterozoic oxyatmosphere stem most commonly from similarities between Archean and younger chemical and biogenic sediments. Carbonaceous material in older rocks is considered evidence that oxygen was produced photosynthetically from carbon dioxide. Sedimentary iron oxides indicate that ferrous iron was oxidized to ferric iron in ancient seas, and rare local occurrences of sulphates apparently required oxidizing conditions. The existence of these phenomena, however, do not refute evidence that atmospheric gases were incapable of oxidizing ancient surficial materials. Anaerobes utilize oxidation-reduction reactions. As outlined by Cloud (1973), the accumulation of free oxygen in the atmosphere must have been dependent on evolution of photosynthetic organisms, initially in an anoxic environment. Photosynthesis and oxidation of organic material can be represented by the reactions



If ferrous iron is present, and if carbonaceous material or iron oxide is buried or otherwise removed as produced, reactions can be summarized as



There is no requirement that the atmosphere contain oxygen, although if oxygen-tolerant organisms are involved, this would enhance the process.

Red beds and black sands in the Cobalt Group and other Paleoaphebian strata indicate that free oxygen began to accumulate in the atmosphere not synchronously with but well in advance of deposition of the

great circum-Ungava and Lake Superior iron-formations. This is contrary to beliefs held by Cloud (1973), Garrels and others (1973), and others who have considered the probable course of atmospheric evolution. It does not damage their hypotheses in any important way, but it may place constraints on models for deposition of the early Proterozoic iron-formations. Another constraint, dealt with most effectively by Holland (1973), is a requirement for an enormous supply of iron to some sites such as the Hamersley Basin. Taking some liberties, perhaps, in applying and extending the model he has suggested, we can postulate the sequence of postoxyatmovation events outlined in the following paragraphs.

Prolonged deep preoxyatmovation erosion of continents supplied abundant ferrous iron, along with other cations and silica, to the ocean. Despite continual deposition of much iron as sulphide, magnetite, carbonate, silicates, and some hematite, sea water below the photic zone and an intermediate mixing zone remained a reservoir of dissolved ferrous iron even after oxygen began to accumulate in the atmosphere. Deposition of thick, extensive iron-formations in special environments and more widespread deposition of dispersed iron minerals depleted the reservoir significantly during a comparatively short time. The most favourable environment for deposition of banded iron-formation would be developed where a strong current of "reservoir water" agitated by sea-floor volcanic eruptions flowed past shallows with extremely high levels of photobiotic activity. A strait or seaway formed by rifting and marginal flooding would be ideal. The Labrador Trough could represent such a situation. None of the North American belts with major iron-formations are definite pericratonic orogens; all might be intracratonic.

The considerable interest shown in hydrocarbon occurrences in Elliot Lake ores perhaps reflects some degree of suspicion that these represent a significant part of the story, as they may in the Witwatersrand. We have no data on abundance of thucholite in North American bankets. Possibly it is not quite as rare in Elliot Lake ores as I have imagined. Thin seams of black thucholite in dark uraninite-rich layers are difficult to recognize underground. Globules and sooty incrustations of very weakly radioactive hydrocarbon are more conspicuous but are far from common. These coat fractures and minerals in vuggy veinlets that are not restricted to bankets; in fact, they have been intersected in drill cores away from ore zones and hundreds of feet above target strata. It seems most reasonable to suppose that scarce hydrocarbon found in ore is also epigenetic. Certainly there would have been more hospitable places for organic material to grow than in coarse gravels in rapids.

Mineralographic evidence and other specific evidence for detrital origin of uraninite and pyrite grains in bankets have not been reviewed in this paper because these have been well documented elsewhere (Liebenberg, 1955, 1960; Ramdohr, 1958; Hiemstra, 1968; Roscoe, 1969). These observations seem to be generally regarded as the only direct evidence that the atmosphere was anoxic until the beginning of Proterozoic time, although other evidence has been presented (Roscoe, 1969). The timing of the oxyatmovation is too important a matter to leave standing on one foot (see Davidson, 1965, p. 1195). Perhaps evidence given in this paper for a transition in weathering and subaerial sedimentation processes from anoxic to oxidizing 2.3-2.2 aeons ago will encourage research leading to a better understanding and more precise dating of this transition.

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# Genesis of Canadian Uraniferous Quartz-Pebble Conglomerate Ores

By H. W. LITTLE

GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN  
QUARTZ-PEBBLE CONGLOMERATES

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## GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN QUARTZ-PEBBLE CONGLOMERATES

### GENESIS OF CANADIAN URANIFEROUS QUARTZ-PEBBLE CONGLOMERATE ORES

By H. W. LITTLE<sup>1</sup>

#### ORIGIN OF THE ORE

As in the Rand district of South Africa where controversy over the origin of the deposits raged violently for some years, at Blind River controversy began almost at the outset of production from the Early Aphebian conglomerates (fig. 1). The principal controversy was between the syngeneticists and the hydrothermalists, the latter being greatly in the minority. Their main arguments centered on permeability of the conglomerate, concentration of heavy minerals at the base of the conglomerate, rounding of uranium minerals, whether due to erosion or colloidal precipitation, and thorium:uranium ratios.

The primary dispute of syngenetic versus hydrothermal origin for the ore has continued for many years. J. A. Robertson (1968) has pointed out that metasomatic effects are greatest in the Pronto and Can-Met Mines in the Elliot Lake area (fig. 1), where the hydrothermalists have marshalled most of their supporting data. Derry (1960) believed that the albitic metasomatism at Pronto is later than both radioactive and sulphide mineralization. Heinrich (1975) further emphasized the effects of metasomatic alteration there by classifying the types of alteration and their effects upon the ore and associated rocks.

At the present time, those geologists who have worked most closely with the deposits seem to be unanimous in the opinion that the ores are syngenetic. Some geologists believe that the ores may be entirely or partly formed by chemical or biochemical precipitation, Derry (1960) being the initial proponent of the latter, which is supported in part by Ruzicka (this volume, chap. W). The relatively high thorium content of the uraninite, however, weighs against this possibility. Grandstaff (this volume, chap. J) offered further evidence in this regard. Moreover, at Agnew Lake (fig. 1) uranothorite

is the predominant ore mineral, and it cannot have been formed in this way; it is formed only in a high-temperature environment such as pegmatites and granitic or metamorphic terranes. It is therefore difficult to arrive at a conclusion other than that the Agnew Lake quartz-pebble conglomerate uranium ores are of placer origin, for the metamorphism there is dynamic and the granites are older than the conglomerate. Indeed, the unconformity is folded by plastic deformation of the granite, and the pebbles are greatly elongated.

Roscoe (1969) has demonstrated that some uraninite grains are detrital, that they are commonly concentrated at the base of conglomerate layers, where one appeared to be pressed down by a quartz pebble into the argillite below, and that in places they show graded bedding, and Ruzicka has given strong supporting evidence (see chap. W). Roscoe's belief that at least some brannerite as well is detrital has been disputed by Ferris and Ruud (1971), who supported Derry (1960). Ferris and Ruud did not believe that leached brannerite derived from crystalline brannerite would form an internal network of needles and blades of anatase and rutile such as is found in the Blind River and Elliot Lake ores. Furthermore, brannerite is relatively rare in hydrothermal and pegmatitic occurrences that would be expected to be the source of detrital brannerite.

It was noted long ago that ore is not concentrated near faults or dykes. One of the most important observations was reported by J. A. Robertson to Roscoe (1969) and later published by himself (Robertson 1970), that in places where the polymictic Ramsay Lake Conglomerate truncates the ore-bearing conglomerate it is uraniferous. D. S. Robertson and Douglas (1970) commented at greater length upon this feature, implying that the uranium content is found only down the paleoslope and comprises radioactive debris from the erosion of the quartz-pebble conglomerate. This not only proves a pre-Ramsay Lake age for the ore but is strong evidence in favour of a syngenetic origin.

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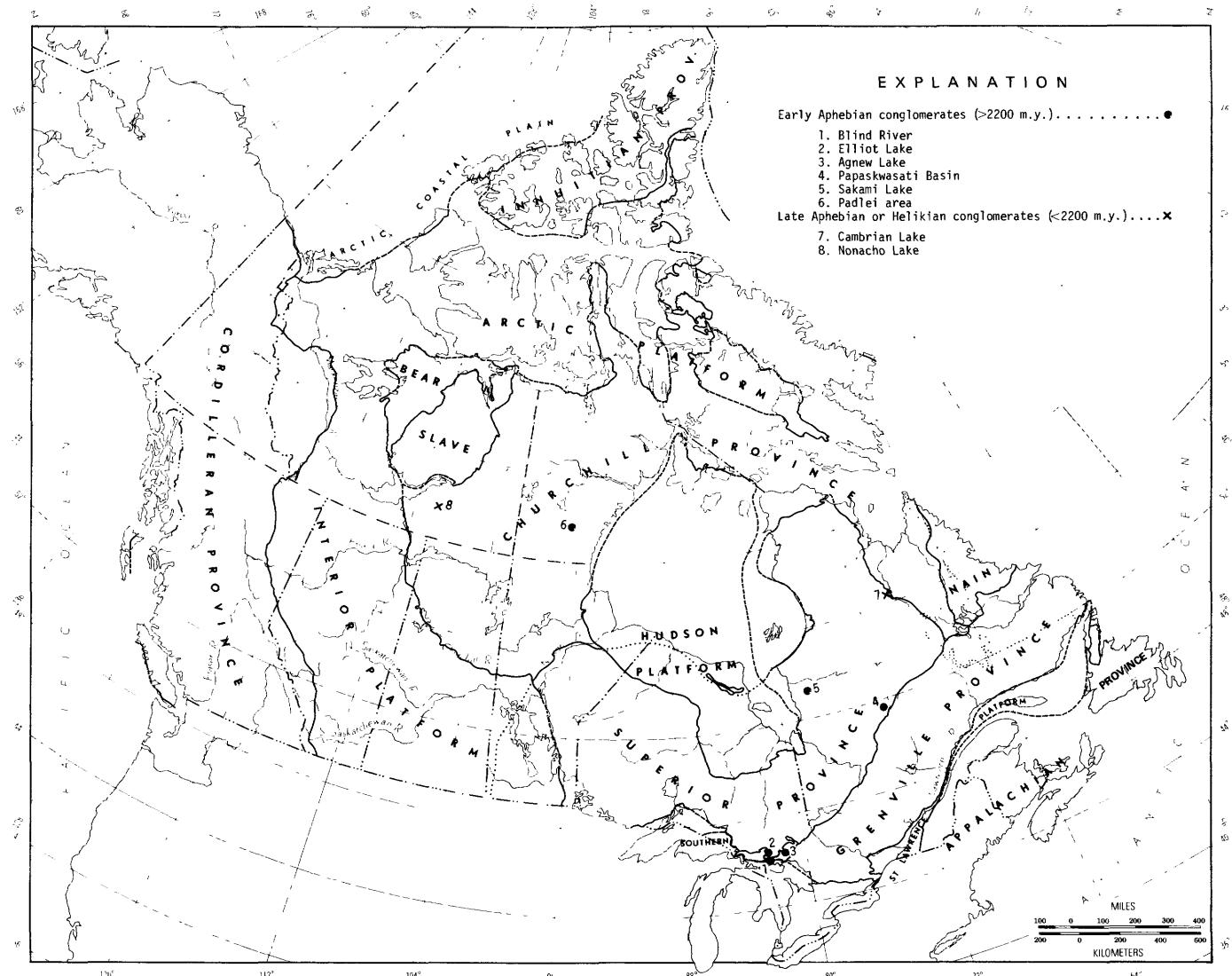


FIGURE 1.- Location of uraniferous quartz-pebble conglomerates in Canada.

#### PROVENANCE

The question of provenance is to me a taxing one in that, although the subarkoses that are intercalated with the quartz-pebble conglomerate reefs are evidence for a granitic provenance, granitic pebbles in the reefs are extremely rare. Furthermore, quartz veins are not abundant in the granites northwest of the Elliot Lake area, which paleocurrent data show to be the source of the sediments. Roscoe (1969) has suggested that erosion of a greenstone belt might have provided a prolific local source for pebbles both of chert and of vein quartz.

Roscoe and Steacy (1958), from a sampling of the porphyritic, red Archean quartz monzonite northwest of the ore-bearing sediments, concluded that the uranium content of the quartz monzonite was above normal and that

the thorium:uranium ratios were similar to those in the ores. Subsequent testing of these granitic rocks by Bottrill (1970) with a portable gamma-ray spectrometer, which enabled wider sampling, showed, as a result of 32 readings, that the uranium and thorium values are close to the clarke values. The thorium:uranium ratio was 7:1, dissimilar to that of 2:1 obtained by Roscoe and Steacy. More recently, airborne gamma-ray spectrometry by the Geological Survey of Canada northwest of the Quirke ore zone has shown a relatively high uranium content in the supposed source rocks. It is, however, unlikely that the area sampled is the same area of provenance that existed 2,500 m.y. (million years) ago; in the writer's opinion, the provenance area must have been farther northwest and somewhat higher than the present erosion surface.

### SEDIMENTATION

Originally it was believed that the Matinenda Formation was deposited in a deltaic environment, and later, that the strandline retreated northward during sedimentation, so that the more northerly reefs are stratigraphically higher. Beginning with Pienaar (1963), recent authors have emphasized a fluvial origin, particularly for the coarser clastics, but at the same time have retained the concept of a marine origin for parts of the succession. At Agnew Lake a northerly retreating shoreline has not been demonstrated. During the early stages of exploration it was thought to be the case (Robertson and Douglas, 1970) but subsequently, after the structural picture had been largely resolved, it became questionable whether more than one conglomerate reef is there (Barnes, 1972).

There was some speculation whether the sedimentation took place in a marine or nonmarine environment. In a recent symposium on basins and geosynclines in the Canadian Shield, Card, discussing a paper by Frarey and Roscoe (1970), offered evidence that in the northern part of the Huronian geosyncline, deposition was non-marine or in shallow epicontinental seas, whereas to the south it was in a deep-water marine environment. Frarey and Roscoe disputed this contention and maintained that deposition of coarse clastics was entirely fluvial.

In recent years there has been a suggestion that there is an east-west orientation within the ore zones of the Quirke, Nordic, and Pronto Mines. This idea has been expressed recently by Rupert and others (1972) in the following terms: "It is suggested that (paleocurrent) data are best interpreted as evidence for eastbound longshore currents in this basin of deposition rather than as the product of river channels running into a basin. In this environment, lag deposits of scoured pebble and conglomerate beds would form over or on the southwestern and western flanks of stratigraphically higher structures." They go on to suggest that in prospecting for deeply buried uranium deposits the possibility of finding point-bar or depositional types of placers at ancient channel mouths should be considered.

With a view to establishing more coordinated data, the Uranium Resource Evaluation Section of the Geological Survey of Canada, in cooperation with the Ontario Division of Mines, has set up a computerization system whereby paleogeographic and isopach maps can be produced to aid in the prediction of the location of undiscovered orebodies.

### EFFECT OF LAVAS

Roscoe (1969), and subsequently others, correlated his Elliot Lake Group with mafic lava-bearing formations to the west in the Thessalon area, to the south in the

Spragge area, and to the east in the Sudbury area. Roscoe suggested that the volcanics would have a marked physical effect on Matinenda sedimentation. Subsequent to his report, mafic volcanic rocks interbedded with Matinenda sediments, as Roscoe had predicted, were reported by J. A. Robertson (1969) at several places along the Quirke Syncline, particularly at Dollyberry Lake, west of the Quirke ore zone.

The problem of lack of magnetite in the Aphebian paleoplacers has been largely resolved. Garrels and Christ (1965) from experimental data showed that magnetite derived from erosion of the Archean basement would have been converted to pyrite in the low Eh environment of an anoxic atmosphere and in a hydrogen-sulphide-rich environment. The discovery of the Dollyberry volcanics by J. A. Robertson (1969) in Township 157, west of the Quirke ore zone, interbedded with Matinenda strata, indicated a rich source of hydrogen sulphide. In a written communication, T. J. Bottrell suggested to the writer in 1970 that this could be a source of the abundant pyrite in the uraniferous conglomerate ores. Ruzicka (chap. W, this volume) has also shown this as a likely source of pyrite and added further evidence by showing, with H. R. Steacy, that thucholite present in the ores is abnormally high in sulphur. In the Sakami Lake deposits (fig. 1), where lavas were probably present, sulphur is fairly abundant; in Papaswasati Basin, where no lavas are known, it is not.

The discovery of Huronian lavas in the Quirke Syncline points up the need for careful reappraisal of drill holes that were stopped in greenstone in the belief that the Archean basement had been penetrated. In some cases the greenstone may have been Huronian lava or even sheared diabase; in either case, deepening of the drill hole would be advisable, because quartz-pebble conglomerate may be intersected at a lower horizon.

### ANOXYGENIC ATMOSPHERE

The concept of an anoxic atmosphere early in Precambrian time was proposed at least as early as 1938, and in 1955 it was applied specifically to explain the origin of uraniferous placers by Liebenberg (1955). A number of papers supporting this theory have appeared in print in recent years and many are reviewed by Roscoe (1969) and Myers (1970). Frarey and Roscoe (1970) summarized the geological data that support the argument for an anoxic atmosphere during early Huronian time. Briefly, they state that it is generally believed that an anoxic atmosphere existed and that, according to Preston Cloud, there is reason to believe that life initially developed at this time. From a study of the paleosol existing on the Archean basement it was shown that iron was lost rather than accumulated

and the ferric:ferrous ratio decreased rather than increased. Frarey and Roscoe believed that the drab colour of most Huronian rocks, the lack of hematite below the Cobalt Group, in which hematite is present, and the presence of abundant pyrite in the conglomerate of the Matinenda Formation all indicate that nonoxidizing conditions existed. The presence of detrital uraninite and pyrite in the quartz-pebble conglomerates was cited as further evidence of the lack of oxygen in the atmosphere. Pettijohn (1970) objected to the concept but solely on the grounds that absence of red colour and presence of detrital pyrite do not necessarily indicate an oxygen-free atmosphere, for in the Appalachians all pre-Silurian Paleozoic sediments are drab and detrital pyrite of mine tailings is transported many miles downstream.

Further support for an anoxygenic atmosphere prior to 2,200 m.y. ago has been offered by Bain (1972), who pointed out that triploblasts and diploblasts, which required oxygen for their metabolism, first appeared 2,200 m.y. ago. He also contended that the pH of runoff from the land was lowered about 2,050 m.y. ago, for the first time below the uraninite solution value. Tugarinov (1972) agreed that such an atmosphere change took place but that it occurred 3,000 m.y. ago. However, his time scale does not agree with that of other writers, for he maintains that sedimentary iron ores were deposited 2,500 to 2,300 m.y. ago, followed immediately by precipitation of carbonates.

More recent papers, based upon more isotopic age determinations, have centered in upon an age of 2,200 m.y. ago as the time of the critical point of oxygen enrichment of the atmosphere.

From India we have had recent apparent confirmation of this hypothesis in a recent announcement that quartz-pebble conglomerate containing pitchblende, monazite, and zircon and carrying interesting values in uranium has been discovered in the Dharwar metamorphic complex, which was affected by metamorphism dated at 2,600 m.y. (Rao, 1975). Pyrite is reported to be present, but the quantity is not given; other minerals present are chalcopyrite, covellite, ilmenite, rutile, and magnetite.

On the other hand, many pyritiferous quartz-pebble conglomerates of early Aphebian age are low or lacking in uranium values, whereas one that is Helikian contains interesting values. It will be described later.

W. B. Myers' paper (1970) dealt largely with the Witwatersrand but has application to the Blind River-Elliot Lake deposits. He pointed out that in the Rand both detrital and oolitic pyrite occur in shallow-water sediments, together with the general absence of pyrite from graded-bedded siltstones and shaly sediments, and suggested that shallow-water sedimentation was influenced by an anoxygenic atmosphere, whereas in

deeper waters biologic photosynthesis by anaerobic organisms may have been taking place. Myers contend that the preservation of gold in a fossil placer deposit, like uraninite, would require an atmosphere of low oxygen concentration, particularly where the particle size is less than 40  $\mu\text{m}$ , which size particles are subject to rapid solution.

#### MINERALOGY AND ZONING

In many papers the mineralogy of the quartz-pebble conglomerate ores has been described. One of the best and most detailed is that by Roscoe (1969), who includes all previous work in his discussion. Pyrite is the most abundant sulphide, ranging from 5 to 10 percent of the ore. In the mines the abundance of pyrite is generally thought to be in direct proportion to the grade of the ore, but Roscoe stated that although pyrite is generally more abundant in highly radioactive conglomerates, there is no positive correlation between pyrite and uranium content.

Sulphur isotope compositions have been determined for a number of pyrite crystals from the ore conglomerates. These show a  $\delta$ -range (Roscoe, 1969) from 0.0 to +2.7 per mil, which is comparable with the range reported in Witwatersrand ores (Hoefs and others, 1968), and strongly indicates a magmatic or hydrothermal origin rather than an origin by biochemical precipitation. It does not, of course, preclude a detrital origin.

In Blind River-Elliot Lake ores brannerite, uraninite, and uranoan monazite, in order of decreasing importance, are the main ore minerals. Many silicates, oxides, and other minerals have been reported in minor quantity at Blind River and Elliot Lake; in all, some 44 such minerals have been identified. On the other hand, at Agnew Lake, in addition to pyrite, only uranothorite and monazite have been reported.

Studies of variations in the  $\text{ThO}_2:\text{U}_3\text{O}_8$  ratios have been of interest with respect to zoning and its bearing on origin of the ores. Data have been obtained from mill-heads, which represent very large samples, and drill-hole information, representing very small samples. In one mine, records of  $\text{ThO}_2:\text{U}_3\text{O}_8$  ratios of carload lots from all parts of the mine were plotted. In one reef the ratios varied from 1:1 to 1:30; in another reef they ranged from 1:1 to 1:53. Furthermore, in the Quirke property the  $\text{ThO}_2:\text{U}_3\text{O}_8$  ratio increases from northeast to southwest (D. Sprague, oral communication, 1970) instead of from northwest to southeast, as has been claimed for the district in earlier papers. A re-examination of old data and the inclusion of new data is recommended.

**CLIMATE**

Roscoe (1969) suggested that another environmental factor in the deposition and preservation of uraniferous placers in the lower Huronian succession was that of climate. He gave evidence that glacial conditions prevailed not only during Gowganda time but also in earlier times, particularly during deposition of the Ramsay Lake and Bruce Formations. Other writers have supported a tilloid and glaciolacustrine origin for these formations. This thesis was further elaborated by Frarey and Roscoe (1970), who indicate that evidence for a glacial origin for Ramsay Lake and Bruce paraconglomerates is more compelling than for the well-publicized Gowganda. The effect of such temperature conditions on the uraniferous conglomerate may be a factor in the concentration of uranium, as the paper by Grandstaff in this volume (chap. C) has shown.

**PAPASKWASATI BASIN**

In the Papaskwasati Basin, Quebec (fig. 1), sedimentation similar to that in the Lower Huronian seems to have gone on contemporaneously. Sediments, including a basal quartz-pebble conglomerate, were deposited in a downfaulted basin that has been little deformed since the time of deposition (fig. 2). There were, however, no lavas interbedded with the basal part of the succession, and as an apparent consequence, pyrite is far from abundant in the quartz-pebble conglomerate, which ranges in thickness from a few to more than 200 ft (60 m) (fig. 3). Flavioglacial processes have not been identified here, and perhaps this, too, prevented concentration, but source rocks, which are to the east, are not abnormally rich in uranium. Some 69 diamond-drill holes were drilled to intersect the conglomerate, and much surface trenching was done. However, uranium values ranged from a trace to about 0.03 percent  $U_3O_8$ , with a few local assays somewhat higher. Results of exploration were therefore not encouraging and no further work has been done.

**SAKAMI LAKE**

D. S. Robertson (1974; Robertson and Lattanzi, 1974) described an interesting occurrence of uraniferous quartz-pebble conglomerate on the west side of Sakami Lake (fig. 1), which is in Quebec, east of James Bay. A thin succession of feldspathic quartzite, which has been traced intermittently for about 5 miles (8 km), contains at least three thin interbeds of quartz-pebble conglomerate and some of amphibolite and chloritic schist that probably are metamorphosed lava. The pebbles are all of quartz, and most are less than 1 in. (2.5 cm) in diameter but are much stretched along the direction of the strike of the beds. Pyrite is quite abundant in the

matrix, and is commonly in cubic form. In general, radioactivity seems roughly proportional to the pyrite content. Once again, the lavas seem a likely source for the pyrite in the sediments.

D. S. Robertson has speculated that the age of those deposits is Archean because they have been involved in the Kenoran orogeny. On the other hand, he has remarked that the succession is remarkably similar to that of the Lower Huronian and has suggested that sedimentological conditions rather than time limits set by geochronology should govern the age classification. The sedimentation at Sakami Lake seems to have occurred at about the same time as the Lower Witwatersrand—that is, prior to 2,500 m.y. The thorium:uranium ratio seems to be higher than in Elliot Lake ores but lower than in those of Agnew Lake Mine.

**MONTGOMERY LAKE**

Uraniferous quartz-pebble conglomerates in the Hurwitz Group at Montgomery Lake, Padlei area (figs. 1, 4), have been covered in chapter W, this volume, by Roscoe and need not be further discussed here. Metamorphic equivalents of the Hurwitz Group extend for a great distance to the south, and similar ones may be found there, perhaps reconstituted.

**DEPOSITS IN POST-EARLY APHEBIAN QUARTZ-PEBBLE CONGLOMERATES**

The uraniferous deposits in the Cambrian Lake area (fig. 1), northern Quebec, have been known for several years. Quartz-pebble conglomerate intercalated with arkosic rocks has been traced along the strike for several miles. This formation lies at the base of the Kaniapiskau Supergroup on the west side of the Labrador Trough and is of Aphebian age. It is regarded as late Aphebian because the basal formation contains hematitic beds. The heavy minerals identified in the conglomerate are pyrite, monazite, and minor uraninite. In this respect they resemble conglomerates of the Lorrain Formation, which are relatively rich in thorium and low in uranium.

The Nonacho Group (fig. 1) in the District of Mackenzie has been described by McGlynn (1966, p. 32) as follows:

The Nonacho Group comprises a conformable sequence of polymictic conglomerates, conglomeratic arkoses, arkoses, and shales. Along the south part of the east contact the basal units consist of a variable thickness of sedimentary breccia that comprises angular boulders of the underlying granitic rock in a matrix of arkose. This rock grades upward and possibly outward from the contact to arkoses containing the odd angular boulder of granite gneiss and beds and lenses of disintegrated granite. Between the breccia and basement is a zone consisting of granitic gneiss broken into large blocks, measuring as much as 10 feet across, that have been only slightly moved from their

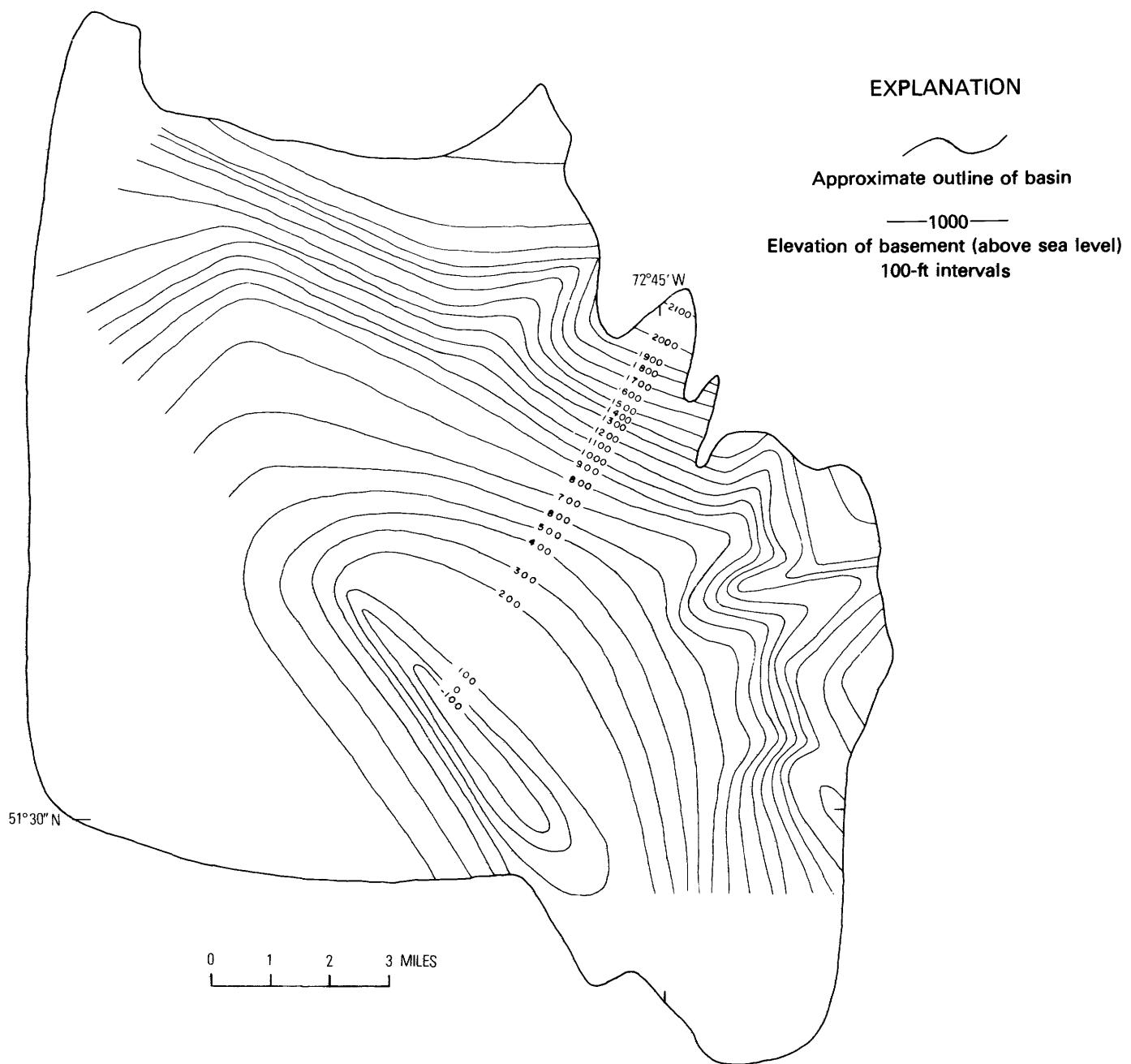


FIGURE 2.—Basement topography beneath the Papaskwasati Formation (compiled by R. A. Boulay).

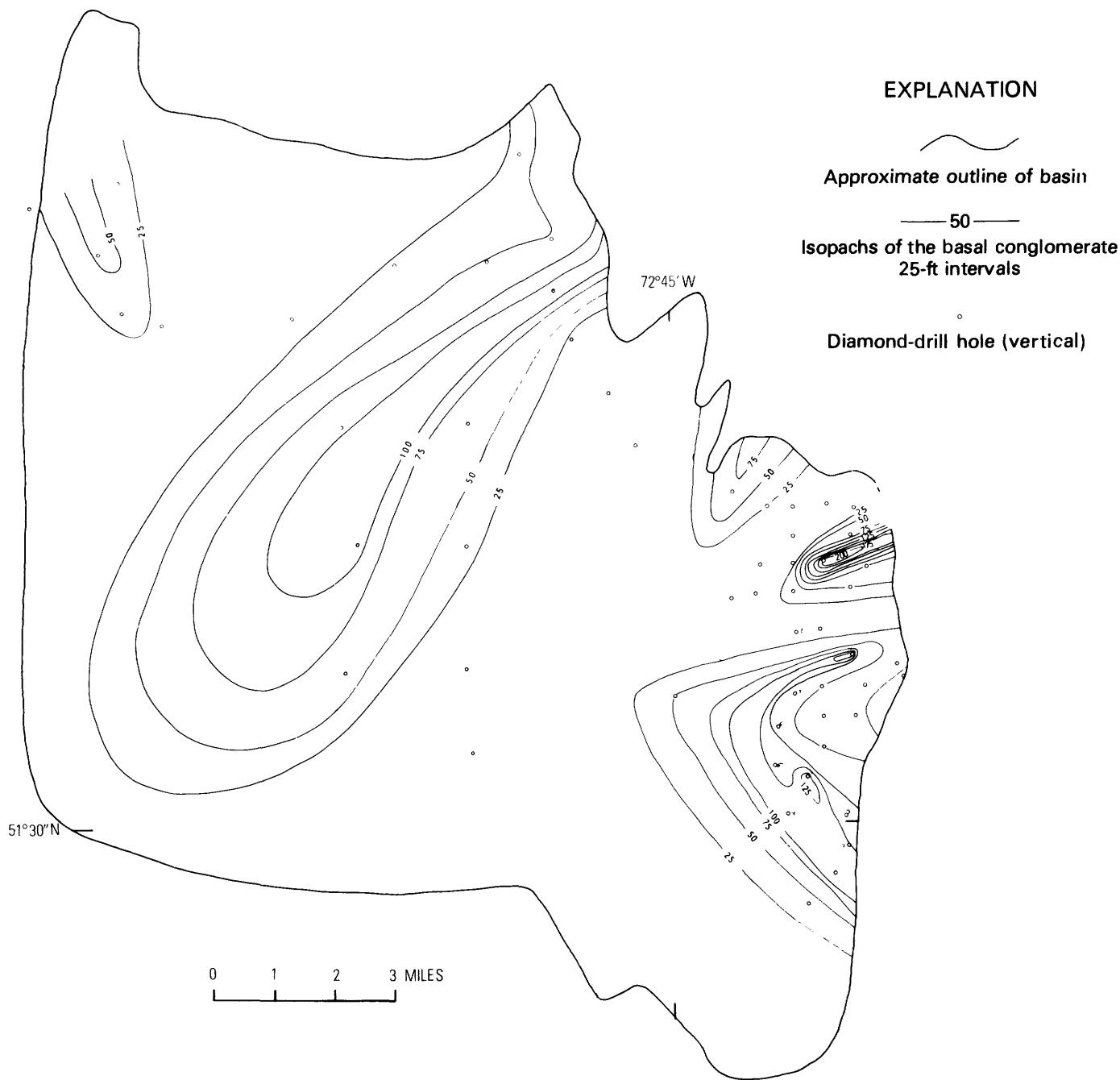


FIGURE 3.—Isopach map showing the basal quartz-pebble conglomerate of the Papaskwasati Formation (compiled by R. A. Boulay).

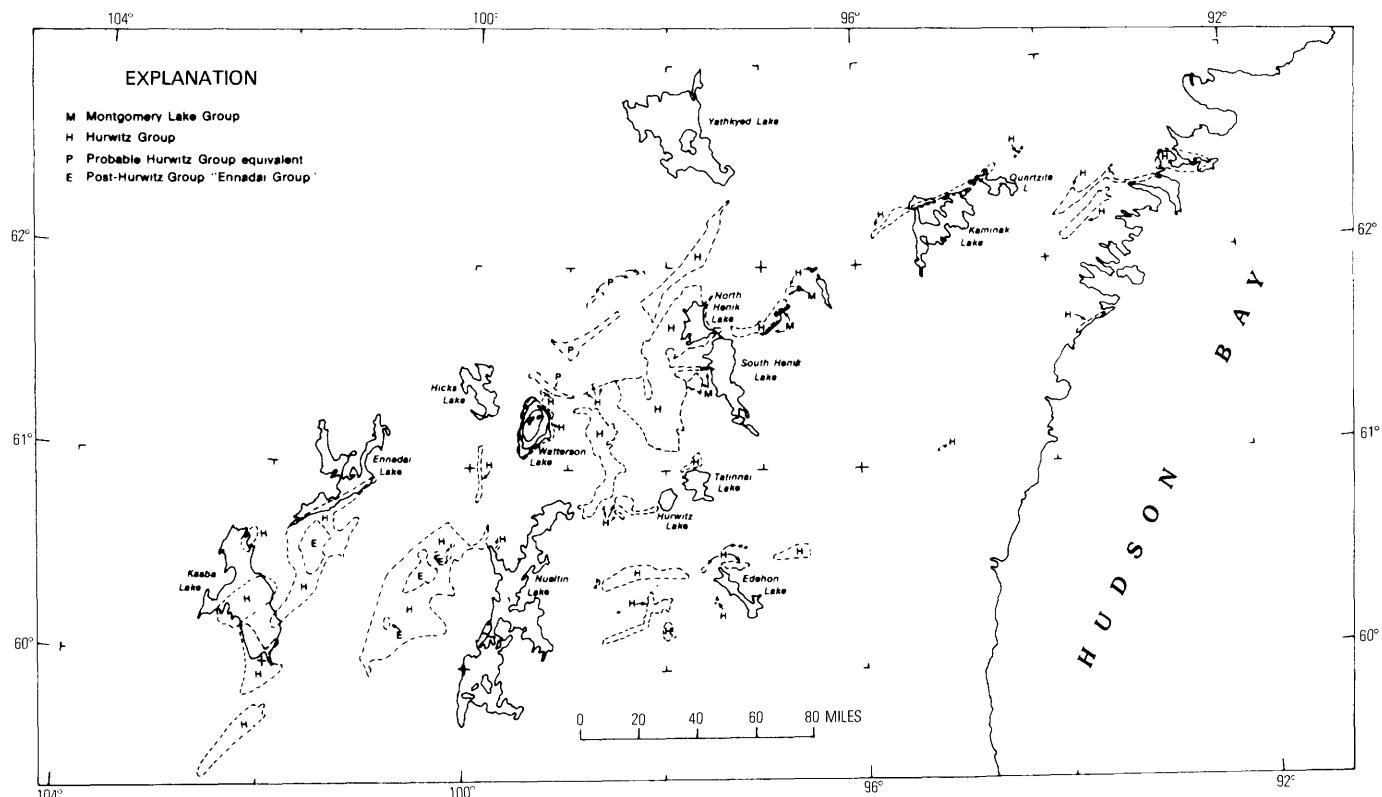


FIGURE 4. - Distribution of Hurwitz Group and associated rocks (from Wanless and Eade, 1975). Reproduced with permission of the National Research Council of Canada (from the Canadian Journal of Earth Sciences, v. 12, p. 96).

original positions, separated by seams of muddy arkosic sand. This rock grades to fractured granite with the odd sand seam that in turn grades to unfractured gneisses, migmatites, etc. Locally these basal units are overlain by polymictic conglomerates rather than arkose. In several places the contact zone is intensely sheared and the above relationships are thereby obscured. A similar unconformity is observed in several places along the west contact of the sediments, but in large part the relationships on the west contact so far mapped are obscured by intense shearing, crushing and brecciation related to faulting in the contact area. Observations made to date indicated that the sediments were deposited on a basement of considerable relief.

In the southeast part of the area the basal arkoses are overlain conformably by widely bedded polymictic conglomerate. This rock contains closely packed boulders ranging in size from fractions of inches to 2 feet, of granitic rocks, vein quartz, quartzite, basic and acidic volcanic rocks, and fine-grained porphyritic rocks with a matrix of arkosic sand. Lenses and beds of arkose are common, and the transition to the overlying arkoses is marked simply by increasing amounts of arkose. The arkoses are buff, pink, or light grey rocks, which in places are conglomeratic in that they contain lenses and beds of conglomerate, beds of conglomerate that are the thickness of one pebble, and single pebbles or clusters of pebbles. Beds of red or green shale up to 10 inches thick and beds of ill-defined zones of shale chip conglomerate also occur. Crossbedding, parallel laminations, and ripple laminations are found in some horizons in the arkoses. These arkoses grade into a second band of polymictic conglomerate in which very large boulders of white quartzite are common. This unit is overlain by arkose.

The Nonacho Group is regarded as Helikian in age and so is younger than other successions that contain

uraniferous quartz-pebble conglomerates. The basal unit is pink feldspathic quartzite, succeeded by a polymictic, nonradioactive conglomerate. Above this is 200-300 ft (60-90 m) of white to greenish feldspathic quartzite containing thin beds that are rich in specular hematite and are faintly radioactive. This is succeeded by a nonradioactive bed of conglomerate and some 200 ft (60 m) of feldspathic quartzite. This, in turn, is overlain by paraconglomerate 40-50 ft (12-15 m) thick and a zone of interbedded quartz-pebble conglomerate and feldspathic quartzite some 30 ft (9 m) thick.

The feldspathic conglomerates are as much as 4 ft (1.2 m) in thickness and contain magnetite and minor pyrite. This zone is overlain by greenish brown argillite that in places contains malachite. Uranium is most abundant in the zone of quartz-pebble conglomerates, and, from a number of samples, it has been said to average close to 1 lb per short ton (0.5 kg per metric ton) (0.05 percent  $U_3O_8$ ) across 6 ft (1.9 m) and to extend several thousand feet along the strike. The horizon has been traced intermittently for miles. No gold or tin is reported from the conglomerates, although both have been found in the radioactive hematitic beds.

Unfortunately, no samples of this material have as yet been obtained by the Geological Survey of Canada nor

has any mineralogical work been done on the uraniferous zone. But this deposit is of more than casual interest to this group because it is of much higher grade than quartz-pebble conglomerates that are younger than early Aphebian age elsewhere.

In summary, I would like to propose that in searching for quartz-pebble conglomerate deposits, the best place to look is in the lower part of early Proterozoic basins, but it is unwise to ignore the younger basins, for viable ores may be found there, and certainly vein and replacement-type deposits do occur in such an environment.

From a consideration of the criteria outlined in this discussion a map of Canada has been drawn that indicates areas in which additional quartz-pebble uranium deposits may be discovered (fig. 5).

## EFFECTS OF METAMORPHISM ON HURONIAN ROCKS IN THE GRENVILLE PROVINCE

Finally, we might consider the effects of high-grade metamorphism and ultrametamorphism. Within the assemblage of paragneisses, amphibolites, schists, migmatites, and granitic rocks that compose the Grenville Province and other metamorphic terranes, beds of quartzite and quartz-pebble conglomerate, some of which are uraniferous, may be found from place to place. In some of these metamorphic terranes pods of white pegmatites containing biotite, magnetite, and uraninite are abundant. These pegmatites may represent remobilization and limited migration of uraniferous quartz-pebble conglomerate deposits, thus completing the full cycle by forming material that is generally regarded as typical of the area of provenance of the con-

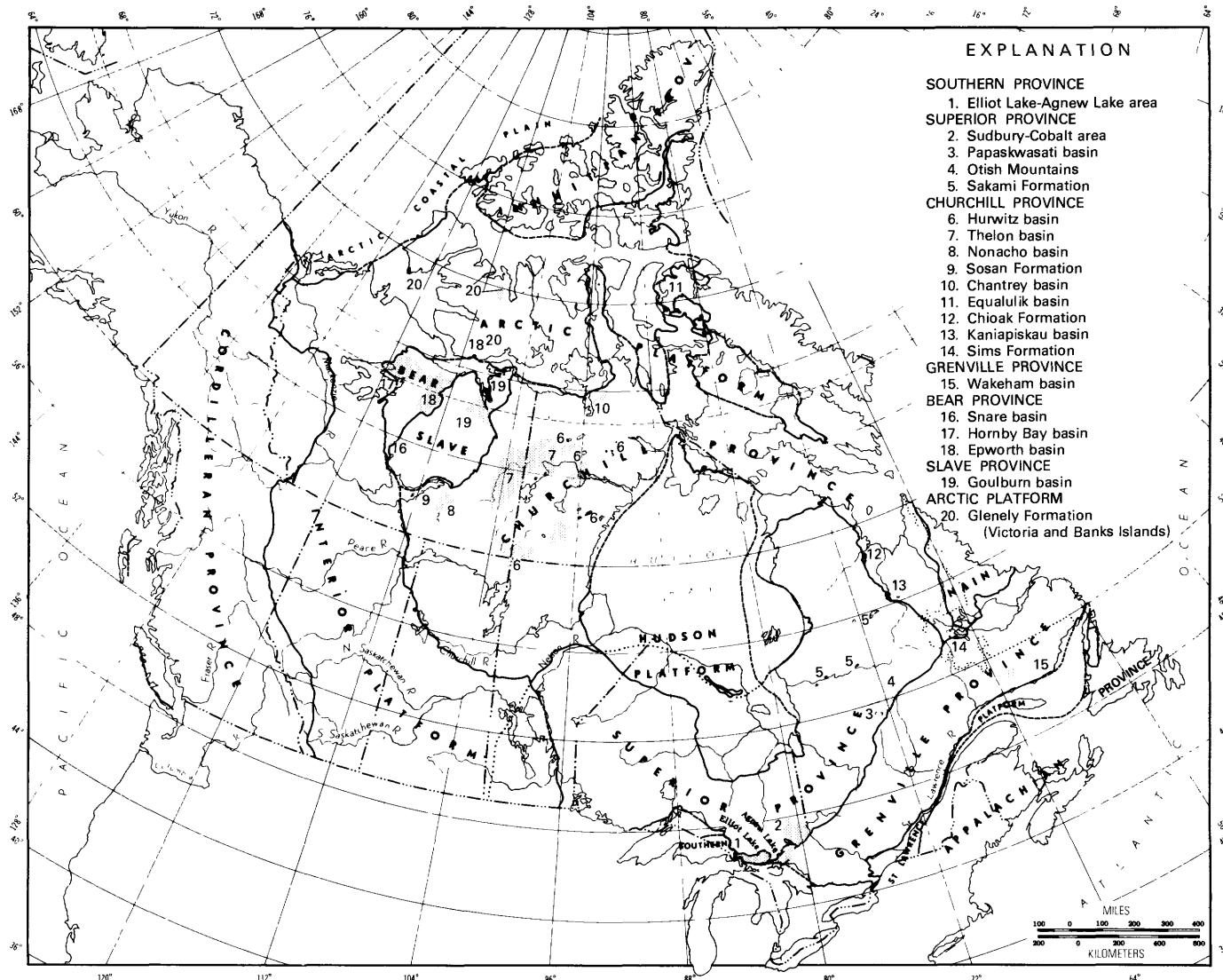


FIGURE 5.—Potential areas for uraniferous quartz-pebble conglomerate deposits (from Little, 1974, fig. 3). Reproduced with permission of the Canadian Institute of Mining and Metallurgy.

conglomerate deposits. The red intrusive pegmatites of the Bancroft area may therefore be the product of ultrametamorphism of uraniferous quartz-pebble conglomerate deposits, accompanied by soda and potash metasomatism. Either process might produce the so-called porphyry uranium deposits of Frank Armstrong (1974). Careful working out of the stratigraphy in metamorphic terranes, therefore, could be a useful guide in tracking down these possible byproducts of uraniferous quartz-pebble conglomerates.

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### DISCUSSION FOLLOWING PAPERS BY DRs. ROBERTSON, RUZICKA, ROSCOE, AND LITTLE

**Schidlowski:** Just a quick comment on the ore microscopic slides shown during the last presentation. I was really puzzled to see that, for instance, the pyrite is in a virtually unabraded state. If I didn't know the whole geological background story I would not be prepared to admit that these are detrital pyrites. What are the exact reasons for the extreme immaturity of these detritals in the Blind River area?

**Armstrong:** To whom is this addressed?

**Schidlowski:** To all Canadians.

**Roscoe:** Well, I don't know which pyrite grains you're speaking about there.

**Schidlowski:** Some of yours. I think you showed us one slide, and they looked reasonably good. I even spotted one single muffin.

**Roscoe:** Yes, there are some of them all right.

**Schidlowski:** But particularly the grains shown by Dr. Ruzicka. I think they were in an unrounded state.

*Roscoe:* They may have been recrystallized. They may have been overgrown.

*Schidlowksi:* So they should have been buried very, very quickly. Are these bands associated with lots of gray-wackes or subgraywackes? What is the exact nature of the arenitic component of the sequence?

*Ruzicka:* It's a little bit arkosic.

*Schidlowksi:* Okay, that would fit in with it. The next part of my question goes to you specifically. Did I understand you correctly that you would prefer to explain the uraninite particles within the carbon as having precipitated from soluble uranium compounds carried into the basin?

*Ruzicka:* No.

*Schidlowksi:* No?

*Ruzicka:* Because of the very high thorium content in these uraninites.

*Schidlowksi:* Okay, then I did not understand that too well. Okay, that was all.

*Nagy:* I think that by now it seems to be rather clear that the association of carbonaceous matter with uranium minerals and gold is a very important association. I'd like to make a comment and ask a question of Drs. Ruzicka and Schidlowksi, and perhaps even Brian Skinner would like to comment on this. Micropaleontologists generally believe that by two and a half billion years ago life was present on Earth only as very simple, nonnucleated cells, bacteria, and blue-green algae. Blue-green algae and bacteria, like all living things, die. Now, if they die under anaerobic conditions, then the decomposition decay product which is going to be formed is going to be methane. Now I wonder, do we have to introduce any kind of fancy theories about polymerization of abiological methane or of very complex organisms recorded by the columnar structure which surrounds the uranium grains? Could these not be simply polymerization products by radiation from the uranium minerals of methane of biological origin?

*Ruzicka:* I think this is quite possible. The hydrocarbon occurs in several forms. The one I showed you reminds me of algal matter or stromatolites, but quite often you can find hydrocarbon in very young fractures and evidently it has moved.

*Schidlowksi:* I would principally concur to this statement.

*Nagy:* In other words, you pretty much agree with what I am saying that the hydrocarbon could have come from biological material.

*Schidlowksi:* Yes, the decomposition of organic material in the widest sense. So since we had only bacteria and blue-greens, these should have constituted the probable source.

*Nagy:* I'm glad you used that word "organic."

*Grandstaff:* Dr. Ruzicka, I wonder if you could clarify a point on the extinction of the organic matter. I was wondering if the extinction bends around the pebble, where the pebble passes down into the underlying layer? Bending or disruption of the extinction might suggest whether the organic matter or the organization was present before or after compaction.

*Ruzicka:* It is a very good idea to observe this, but it hasn't been done, unfortunately.

*Myers:* Dr. Ruzicka, I didn't quite get it clear, but did that thucholite that you show come from the specific locality beside the pebble?

*Ruzicka:* Yes. I mentioned that this sample was taken from the lower reef of the Denison Mine.

*Roscoe:* Dr. Ruzicka, I wonder if you would elaborate on your proposal that eruption of volcanic rocks was a necessary part of the concentration of the pyritic conglomerates. I think you probably said Elliot Lake ores, but what is the distinction between ores and pyritic conglomerates which occur stratigraphically far above ores in the Mississagi Formation? I might add, too, that I have looked at Aphebian conglomerates near volcanic rocks, like at Flin Flon, Wilson Island [Nairn?], and there again they are hematitic and high thorium.

*Ruzicka:* I think you must have uraninite present and afterwards the conclusion is drawn that it is related to the volcanic activity. It is practically a deduction of the observation. It's a conclusion, it's a fact, it is there.

*Hallbauer:* I want to make a remark about this carbonaceous material. If it is assumed that it is a polymerization product, then it should be plain hydrocarbon, without any inorganic inclusions, but all the carbonaceous material from Blind River, as well as from South Africa, includes filamentous inorganic material which cannot be explained by just polymerization. I have shown a few examples from Blind River where the filaments have all the same diameter, all the same structure, so polymerization only is not the answer. I think that it's possibly too easy to say that paleobiologists have agreed that there were only simple organisms in the Precambrian rocks; therefore, nothing else may exist. I think this is a bit too easy.

*Schidlowksi:* May I just comment on that briefly. I would say, Dieter [Hallbauer], that the reconstituted minerals within the interfibral spaces just constitute redeposited material in the widest sense, so they just occupied those places which were allowed by the carbonaceous structures and they just occupied the little spaces. That would be my explanation for that.

*Hallbauer:* What do you understand on the carbonaceous structure, first of all?

*Schidlowski:* I just mean that the little space between the individual carbon seams.

*Hallbauer:* No. The mineral matter is inside the carbonaceous matter.

*Schidlowski:* Yes, of course, the uraninite relics.

*Hallbauer:* No, it's even finer. The filaments have a diameter of about 1 or 2 microns, so how do they fit into this? I've got examples of real polymerized carbonaceous matter from the Witwatersrand which occurs in voids near the Carbon Leader Reef where it was heated and was redeposited. That is real polymerized carbonaceous matter which does not contain any inorganic material whatsoever. You can heat it, and it just is vaporized. This carbonaceous material occurs as crusts on quartz crystals and others.

*Armstrong:* We've had a lot of discussion about this carbonaceous material. I don't think we're going to be able to decide it. And time is running short. If there's some new aspect to it that can be brought up for discussion, fine, but maybe there are some other questions that people would like to ask.

*Myers:* I would like to make just one comment. If you go and look in a stope of the Carbon Leader, at least the stopes that I have seen, the texture of the carbonaceous material is variable inch by inch. You can see repeated gradations between fly-speck carbon and individual granules which presumably contain remnants of detrital uraninite. They get more and more concentrated, and finally the point is reached where you get a silicate-rich thucholite, mechanically incorporating a bunch of the matrix; then this goes in another half inch to beautifully columnar stuff, and then in a few more inches you may have pebbles. All you've got to do is go look in the stopes.

*Schidlowski:* I would completely subscribe to that.

*Myers:* I would like to ask Stu Roscoe, has he seen the contact between the top of the Dollyberry basalt and the overlying gravels? Does it have a regolith on it?

*Roscoe:* Yes, there are regoliths on top of the Dollyberry; that's common.

*Bourret:* My question is directed principally to Dr. Ruzicka. Yesterday, in discussing the Rand, we learned and were told repeatedly about the tectonic activity, rejuvenation, and the raising of the source areas, and the repeated energy events which caused recycling of the material flowing into the Rand basin. My question is: Is there any evidence in the Elliot Lake deposits that there have been repeated tectonic events to supply energy for reconstitution and reworking?

*Ruzicka:* I would say there are two such effects. One is the alteration and introduction of uranium remobilized from other portions of the conglomerate around certain diabase dikes. For example, in the vicinity of these dikes you find uranium enriched as much as ten

times, and uranium content decreases with the distance from the dikes. But not all of these dikes have such an effect. Second, I would say that the influence of faults, for example, is not much in the Elliot Lake deposit.

*Robertson:* I would like to comment on the last two questions, first of all, on the Dollyberry: The structures of the volcanics suggests that they were in fact probably subaerial rather than subaqueous. As far as breaks in the conglomeratic sequence are concerned, we don't have quartz-pebble conglomerate reworked into an overlying quartz-pebble conglomerate, the situation corresponding to, say, the Witwatersrand. We don't seem to have that. We have small-scale crossbedding and so on, and obviously coalescence of beds, but we don't have the major reworking that you seem to have on the Rand. The other comment I would like to make is something that I was supposed to say and didn't. There is in fact very little gold in the Blind River deposits. I would suggest that this is because in the hinterland of the area I was describing and the area shown in the gamma-spectrometry map, there is very little greenstone. When you go east into the bottom part of the Cobalt Plate, where the beds carrying mineralization are probably somewhat younger than those at Blind River (we don't have a good correlation on them), there is a certain amount of gold along with uraninite in some beds. We don't really know whether it is free gold or whether it's gold in sulfides, and in that area the basement rocks contain much more in the way of greenstone. I would relate the gold content in conglomeratic ores in the Huronian to the provenance.

*Roscoe:* I'd like to add another postscript on what Jim [Robertson] said. I have seen a composite pebble such as described in the Rand, a pebble of conglomerate within conglomerate, in the Montgomery Lake area but never in the Elliot Lake area.

*Bourret:* If the Blind River-Elliot Lake conglomerates have not been reworked, very many times or perhaps only slightly, then how do you account for the very high uranium content compared to Witwatersrand reefs, which have been reworked a great many times?

*Ruzicka:* I would say this is a concentration in the source area and the concentration effected during the transportation and deposition.

*Saager:* Dr. Ruzicka, do you find any cobalt and nickel sulfides in the Blind River conglomerates, and do you find any chromite in the Blind River?

*Roscoe:* Yes you do. You find millerite and arsenopyrite.

*Ruzicka:* But very little.

*Roscoe:* Chromite, cassiterite. There's a very rich suite of heavy minerals, but quantitatively they are very small.

*Whiteside:* I'm sorry to jump up and down like this but

just for the record, there are quartz-pebble conglomerates in Ghana which more or less fit in with this suite of conglomerates that we've been discussing. The interstitial mineralization, as far as I know, is hematite and there is no radioactivity, there's just gold.

*Staatz:* Both in the South African talks and in the Canadian talks we have discussed a lot about having brannerite. Brannerite is an exceedingly rare mineral except in this type of deposit. Now my question, I think I'll give it to Vlad [Ruzicka] there, is whether he believes that brannerite is derived like most of the other minerals, that it is brought in like the pyrite and the uraninite, are brought in, or whether brannerite is indeed formed within the formation?

*Ruzicka:* In my talk I mentioned Theis, who studied this problem, and I also mentioned the "Pronto reaction" of Ramdohr. I would say the "brannerites" are uranium-rich titania aggregates, which are relatively common, but according to Stacey there are also a few grains of true brannerite.

*Staatz:* Yes, but are these grains of "true brannerite" or "true aggregates," are they brought in from the outside or are they derived later in the rock?

*Ruzicka:* In this case I would say the true brannerites are of detrital origin.

*Nick Ferris, Urania Exploration, Golden, Colo.:* I will attempt to elaborate on brannerite a little bit. At first at Blind River it was thought to be an entirely detrital grain; however, Paul Ramdohr in the mid-fifties noted the delicate complex texture inside the small, rounded grains and concluded that brannerite might be authigenic, having been formed from a pre-existing mineral. Because of his observations at the Pronto Mine at the southern part of the district, he suggested the so-called "Pronto reaction," in which titania migrates to uraninite and combines with it to form brannerite, a uranium titanate. Ramdohr also felt the same reaction was responsible for brannerite in the South African ores, although he referred to the mineral as "uraninite ghosts." This has also been mentioned by Schidlowski, in 1966 I believe, with excellent photographs and scan photos from the microprobe. Schidlowski also subscribed, at that time at least, to the "Pronto reaction" as the explanation. The texture of brannerite from several Blind River localities has convinced me that brannerite from quartz-pebble conglomerates is an authigenic mineral originating as a replacement of ilmenite when ilmenite altered to rutile in the presence of uranium in solution. There is evidence for this in the texture if you compare the textures of the Blind River brannerites, the needle network and blades (Nick Theis has some excellent photos of this and has done a lot of work on it) with the textures you get when you do indeed alter beach sands or

rocks containing ilmenite to rutile. This texture is what's suggestive of this process, as well as the fact that you should have ilmenite at Blind River and you don't have any. So presumably it was there and something happened to it. What I am suggesting here is that the "Pronto reaction" really is not valid; in it titania from an undisclosed source goes to and in part replaces uraninite. I believe instead that uranium in solution from any of several possible sources, some of which were mentioned yesterday, goes to a titania mineral and alters it to this delicate needle network and mixture of various things in the small, rounded grains which we now call brannerite and is found at Blind River and perhaps also at the Rand. I conducted as many tests as I could on some other brannerites from around the world using the microprobe and observing them with the microscope as well. The types of brannerites that we've been talking about these past two and a half days are decidedly different texturally; they also are different compositionally because they show a large variation in the amount of uranium which occurs in them, not only in the entire grain but within those parts of the grain which can be analyzed by microprobe. I guess my main point here is that uranium migrated to a titanium mineral, I believe ilmenite, and formed uraniferous rutile, or uraniferous leucoxene as Von Rahden and Hiemstra have said, which can in fact become rich enough to grade compositionally to true brannerite, if there is such a thing, when the ilmenite has decomposed. This is the twist, then, on the "Pronto reaction." Now, this is also not the question that I raised my hand for. My other question I'll direct to any of the Canadian contingent. In the Blind River ores there is a greenish-yellow sericitic matrix. I'd like to know the origin of this. I've had various responses to this question before: that it is metamorphosed in-filling clay, a volcanic material, or decomposed, redistributed and altered feldspar, but I'm still curious.

*Roscoe:* I am, too. I assume it was a clay, it was detrital, and that there was potash restoration in it. It does incorporate the finest detrital fraction within the conglomerate, and it must have been in-filling. I assume that there was alteration subsequently.

*Meyer, Falconbridge Nickel Mines, Toronto, Canada:* I'd like to make a comment first on Brad Myers' question, which has already been answered by Jim Robertson and Stu Roscoe. The volcanics at the base of the Huronian do seem to be subaerial, and in fact there seems to be a considerable unconformity, or certainly a period of erosion, between the volcanics and the basal Matinenda. Our drilling has shown this quite conclusively. When I say ours I mean Falconbridge Nickel Mines, back in the midsixties. So much for a

comment. Second, the question, addressed to Dr. Ruzicka. Would it be possible that the uranium in the upper Proterozoic and Cambrian conglomerates of Russia is genetically more akin to the Colorado Plateau type rather than the Lower Proterozoic conglomerates in South Africa and Blind River? If your answer is "yes" on that, then what bearing does that have on exploration in sedimentary rocks in this vast time span between the two?

*Ruzicka:* I would say, and as I mentioned in my talk, the mineralization is more epigenetic than syngenetic, so it is more similar to the Colorado Plateau type than Elliot Lake type. You have many occurrences of uranium mineralization in young conglomerates. For

example, you have North Carolina, the Ningyo-toge deposit in Japan; you also have mineralization in the Permo-Triassic conglomerates of the west Carpathians, and so on.

*Meyer:* My question was, what bearing on sedimentary rocks between the lower Proterozoic and, say, the pre-Devonian? In other words, before you got any plant cover, which seems to have been important to deposition in post-Devonian rocks. I want to know specifically pre-Devonian but post-lower Proterozoic.

*Ruzicka:* You can have uranium mineralization in sandstone in that time span, but this type of mineralization is different from the Elliot Lake type.

**UNITED STATES**



# Thorianite from the Hogatza Placer, Alaska

*By* MORTIMER H. STAATZ

GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN  
QUARTZ-PEBBLE CONGLOMERATES

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GEOLOGICAL SURVEY PROFESSIONAL PAPER 1161-Y





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## GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN QUARTZ-PEBBLE CONGLOMERATES

### THORIANITE<sup>1</sup> FROM THE HOGATZA PLACER, ALASKA

By MORTIMER H. STAATZ<sup>2</sup>

The Hogatza placer lies along Bear Creek, a tributary of the Hogatza River, where it emerges from the Zane Hills. This locality is about 240 km west of Fairbanks and just east of the Seward Peninsula. A dredge has mined gold from this placer since at least the early 1950's. Thorianite was found in a dredge sample collected by Thomas Miller during the mid-1960's, when he was studying the precious and nonferrous metals of this area for the U.S. Geological Survey. In 1974 Miller and the writer spent several days in the area collecting samples of both placer material and the source rocks. Concentrate from the sluice boxes on the dredge are abnormally radioactive. Two-thirds of the heavies in this concentrate are magnetite. The next most abundant mineral is thorianite, which commonly occurs in small black sand-size cubes (fig. 1). Other minerals identified, in approximate order of abundance, are zircon, sphene, amphibole, spinel, garnet, ilmenite, hematite, gold, epidote, and goethite. Thorianite is both isostructural and isomorphous with uraninite, and a complete solid solution exists between  $\text{ThO}_2$  and  $\text{UO}_2$  (Frondel, 1958, p. 47-49). The names thorianite and uraninite are restricted to the halves of the series with  $\text{Th} \square \text{U}$  and  $\text{U} \square \text{Th}$ , respectively, in atomic percent. A sample of the thorianite from the Hogatza placer was analyzed by J. N. Rosholt, Jr., of the U.S. Geological Survey in Denver, by the isotope-dilution method. It was found to contain 47.3 percent  $\text{ThO}_2$  and 36.8 percent  $\text{UO}_2$ .

The thorianite in the Hogatza placer was derived from a gneissic monzonite (fig. 2) that forms a border facies of the otherwise rather massive granodiorite of the Zane Hills pluton. The border facies is 5 to 10 times as radioactive as the main igneous mass. The uranium content of 11 samples of the gneissic monzonite ranged from 11 to 129 ppm (parts per million) and averaged 47



FIGURE 1.—Cubes of thorianite from the Hogatza placer.

ppm. The thorium content of these samples ranged from 46 to 268 ppm and averaged 128 ppm. Concentrates of the heavy minerals from these rocks vary somewhat, but amphibole, pyroxene, magnetite, zircon, and sphene are abundant in most. Allanite and biotite occur in many samples, and thorianite, garnet, pyrite, molybdenite, muscovite, goethite, betafite, and thorite occur sparsely in some samples. The thorianite occurs as small cubes similar to those found in the Hogatza placer.

Much of the material in the Hogatza placer is glacial till. During the Pleistocene a glacier came down Clear Creek (fig. 2). Part of this glacier came over a saddle separating this creek from the headwaters of Bear Creek, scouring the dividing ridge of gneissic monzonite, and pushed down Bear Creek, where the material was deposited. The glacial till deposited along Bear Creek has been reworked by the stream, but beyond the eastern front of the Zane Hills where the glacier spread out, much of the glacial material does not appear to have been reworked.

<sup>1</sup>[This mineral prior to analysis was thought to be uraninite and was so reported at the workshop.]

<sup>2</sup>U.S. Geological Survey, Denver, Colo.

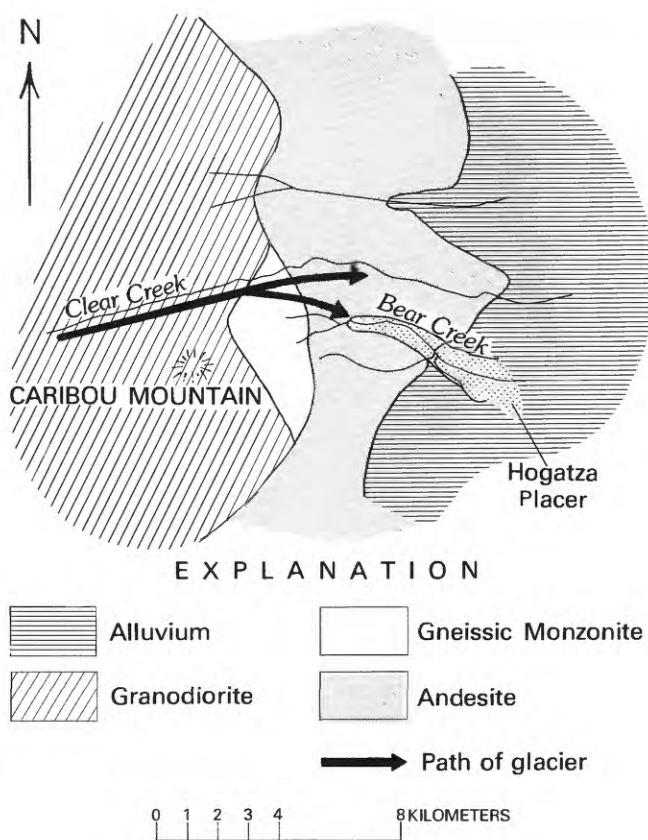


FIGURE 2.—Generalized geology in the vicinity of the Hogatza placer. Derived from figures 3 and 4 of Miller and Ferrians (1968, p. 7, 9).

Uraninite and thorianite are rare in modern placers. The Hogatza placer is not unique, however, as uraninite was reported in a black sand in British Columbia by Hal Steacy (Steacy, 1953, p. 549). Whether conditions have to be extremely favorable or not for uraninite or thorianite to occur in modern placers is not known. The rarity of uraninite and thorianite in post-Precambrian placers may not be a function of its preservation but rather of its rarity in source rocks, such as granitic rocks and veins.

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Basal Conglomerates and Weathered Zones in the  
Marquette Range Supergroup, Northern Peninsula of Michigan—  
Age, Indications of Atmospheric Oxygen, and Uranium Potential

*By* WILLIAM F. CANNON

GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN  
QUARTZ-PEBBLE CONGLOMERATES

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## GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN QUARTZ-PEBBLE CONGLOMERATES

### BASAL CONGLOMERATES AND WEATHERED ZONES IN THE MARQUETTE RANGE SUPERGROUP, NORTHERN PENINSULA OF MICHIGAN— AGE, INDICATIONS OF ATMOSPHERIC OXYGEN, AND URANIUM POTENTIAL

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#### ABSTRACT

The Marquette Range Supergroup is a thick accumulation of sedimentary and volcanic rocks deposited between 2.6 and 1.9 billion years ago. Conglomerates and quartzites that are basal in the sense that they lie directly on Archean rocks, are of three ages, and crop out along a strike length of about 600 kilometers. The conglomerates have been considered possible hosts for uranium ore because of close geographic proximity, approximate age equivalence, and certain lithologic similarities to uraniferous quartz-pebble conglomerates in the Huronian of Canada. To date, no uranium deposits are known in conglomerates of the Marquette Range Supergroup, but extensive cover by glacial deposits is a severe hindrance to exploration. There is a substantial possibility that even large deposits would not be found in outcrop.

Radiometric dating suggests that the Marquette Range Supergroup is at least in part younger than the Huronian, but results are not conclusive. The best guess as to correlation is that the lowest units of the Marquette Range Supergroup are equivalent to the uppermost units of the Huronian.

Iron-formations and weathered zones near the middle of the Marquette Range Supergroup contain indications of an oxygen-rich atmosphere. Because uraninite is not stable in the presence of more than traces of free oxygen, it may have broken down too rapidly to be concentrated in placers during deposition of the Marquette Range Supergroup, but the possibility of its metastable survival under an atmosphere that was almost certainly less oxygen-rich than at present seems quite possible.

A favorable uranium potential for conglomerates of the Marquette Range Supergroup is indicated by the widespread presence of anomalously uraniferous Archean basement rocks, abundant quartz-pebble conglomerate in basal units, uranium concentrations in black slates and iron-formations in slightly younger parts of the supergroup, and monazite-rich placer deposits in some of the quartz-pebble conglomerates.

Exploration of the area is hindered by very extensive drift cover and very few outcrops. Exploration programs should not be designed so as to be dependent on outcrop data but rather should employ techniques capable of detecting ore beneath several tens of feet of drift. Direct sampling of bedrock by diamond drilling may, in the long run, be the most efficient exploration tool.

#### INTRODUCTION

Vast deposits of uranium occur in fossil placer deposits in conglomerates older than 2 b.y. (billion years) in many parts of the world, the Witwatersrand of South Africa and Huronian of Canada being the best known and economically most important. The Marquette Range Supergroup in northern Michigan and northern Wisconsin is similar to the Huronian in age and geographic location, as well as in certain lithologic and stratigraphic features. This paper considers the possibility that deposits of uraniferous quartz-pebble conglomerate like those in the Huronian might be discovered in the Marquette Range Supergroup. This possibility is assessed in terms of the possible correlation of the two sequences, the genesis of ores in the Huronian, and the possibility that similar genetic conditions prevailed during deposition of the Marquette Range Supergroup and in terms of local geologic features of the Marquette Range Supergroup.

#### GEOLOGY OF THE MARQUETTE RANGE SUPERGROUP

##### GENERAL

The Marquette Range Supergroup is a thick sequence of metasedimentary and metavolcanic rocks in the central and western parts of the upper peninsula of Michigan and northern Wisconsin (fig. 1). The age of the rocks is firmly bracketed between 2.6 b.y., the age of the basement on which they unconformably lie, and 1.9 b.y., the age of the Penokean orogeny during which they were deformed and metamorphosed.

The supergroup was deposited unconformably on an Archean granite-greenstone terrane. Little direct evidence of the nature of the land surface is known because of later deformation, but because a relatively long time span separates the late Archean orogenic ac-

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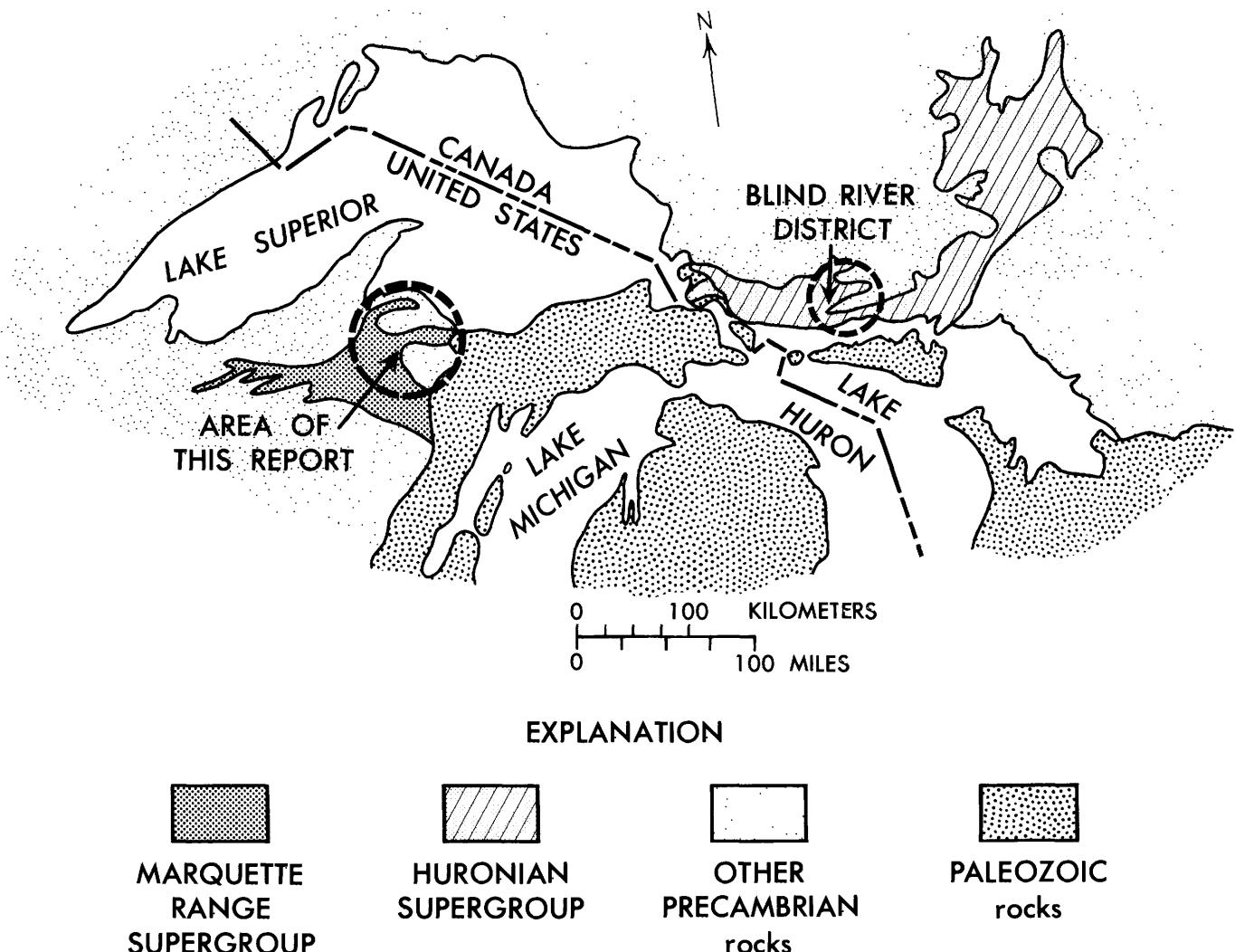


FIGURE 1.—Index map showing generalized distribution of rocks of the Marquette Range Supergroup and Huronian Supergroup.

tivity from the later sedimentation and because the basal Marquette Range sediments are similar to blanket-type clastic-carbonate sequences, the surface is presumed to have been of low relief.

In addition to the unconformity at the base of the section, two major unconformities are within the section as shown in figure 2 and are used to separate groups. Because of erosion of older sediments along these unconformities, and perhaps because of onlap relations, the basal clastic units of three different ages lie on Archean basement at various places. Because uraniferous conglomerates in the Huronian are confined to conglomerates that lie on or near the Archean rather than those within the sedimentary sequence, the discussion of conglomerates in the Marquette Range Supergroup emphasizes those that lie on or near Archean basement rather than those within the section. The discussion is

also confined to the central part of the upper peninsula of Michigan and does not include correlative rocks in the Gogebic district of the western upper peninsula and possibly correlative rocks in northern Wisconsin.

#### CHOCOLAY GROUP

The Chocolay Group, the oldest unit of the Marquette Range Supergroup, is an orthoquartzite-carbonate sequence deposited in a stable tectonic environment. In general, the basal unit is marine quartzite with local conglomerate beds, but tillites and waterlaid conglomerate, arkose, and argillite of glacial affinity are known in the Menominee district (Pettijohn, 1943), the Calumet trough (James and others, 1961), the eastern Marquette district (Gair and Thaden, 1968; Gair, 1975), and Dead River basin (Puffett, 1969; Clark and others, 1975) (fig.

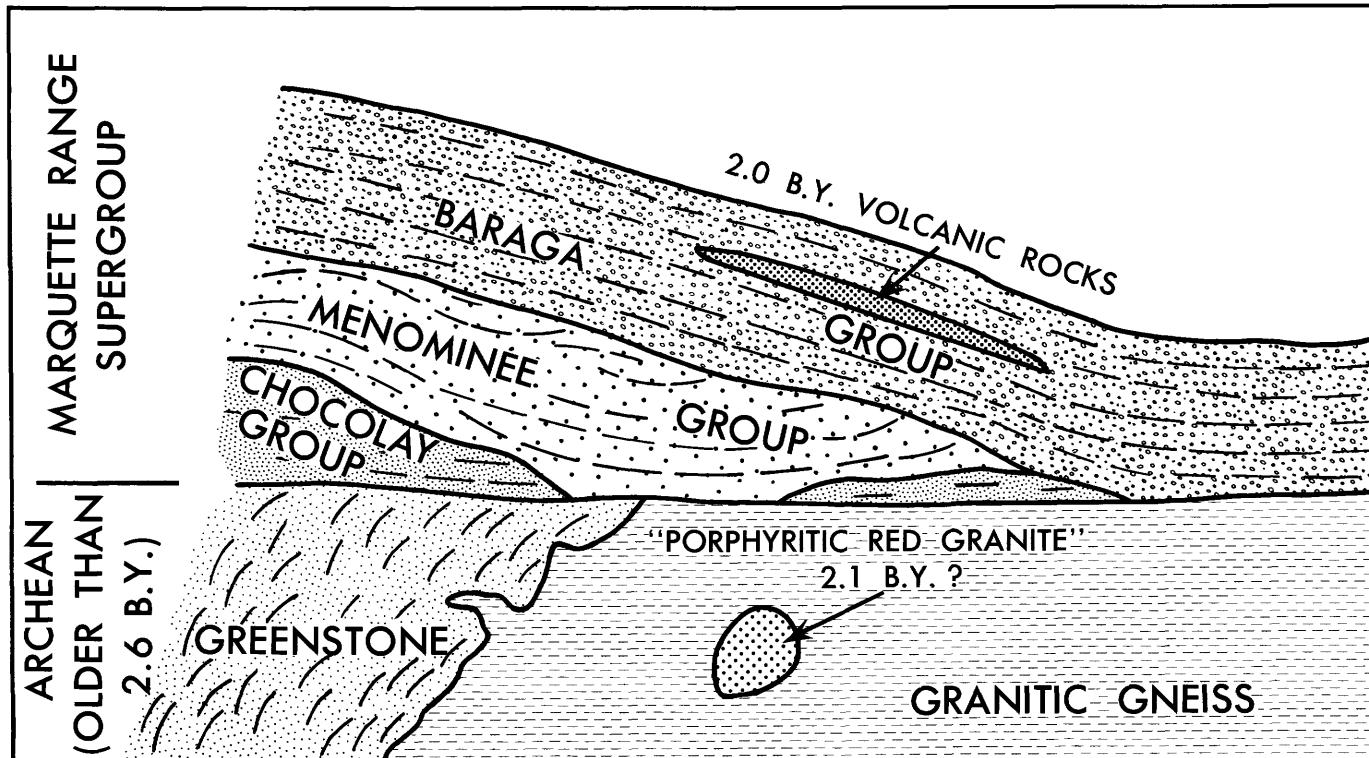


FIGURE 2.—Schematic stratigraphic section showing unconformable relations between the Chocolay, Menominee, and Baraga Groups. Radiometric age determinations are discussed in the text.

3). The rocks of glacial affinity all lie directly on Archean rocks and are believed to be of equivalent ages. Clasts are mostly granitic rocks, with other Archean rocks also present. The matrix generally is chloritic, and the rocks typically are poorly sorted (fig. 4). Their wide distribution and the lack of high relief on the Archean surface suggest that these rocks are a product of continental glaciation and originally were probably much more widespread than indicated by their present degree of preservation. The glacial deposits grade upward into marine deposits of sericitic schist and orthoquartzite; where glacial deposits are absent the marine sequence lies directly on the Archean. In the Menominee district (Bayley and others, 1966) the lowest 30–60 m of the marine sequence are pale-green sericitic quartzite and schist that is overlain by white, gray, and pink vitreous quartzite. Some basal beds contain abundant magnetite, tourmaline, and some pyrite. In central Dickinson County (James and others, 1961) as much as 100 m of gray and green sericitic quartzite and quartzose schist (25 percent sericite, 75 percent quartz) are at the base of the section and are overlain by vitreous quartzite containing zones of quartz-pebble conglomerate as much as 1 m thick. In the eastern Marquette Range (Gair and Thaden, 1968; Puffett, 1974) sericitic and feldspathic schist and quartzite underlie vitreous white, gray, and

pink quartzite containing minor pebbly zones bearing pebbles of vein quartz, chert, and quartzite. Paleocurrent measurements (Pettijohn, 1957) indicate a source to the northwest for the marine clastic rocks of the group.

#### MENOMINEE GROUP

The Menominee Group is a marine sequence generally with basal quartzite that grades upward into finer grained clastic rocks that in turn grade upward into iron-formation. The group contains the major economic iron-formations of the area. Commonly the Menominee Group lies unconformably on the Chocolay Group, but in much of the western Marquette district it lies directly on Archean rocks (Puffett, 1974; Cannon and Klasner, 1974, 1975a, b; Klasner and Cannon, 1975a, b; Clark and others, 1975). There, the group contains a basal orthoquartzite as much as 150 m thick with conglomerate zones near the base. Most conglomerates bear pebbles of vein quartz and chert, but other rock types are also present, including carbonate rocks, older quartz-pebble conglomerate, and granitic rocks. Clasts vary from rounded to subrounded and range in diameter up to about 30 cm (fig. 5). Locally the conglomerate matrix contains small amounts of pyrite, some of which is in rounded, possibly detrital grains.

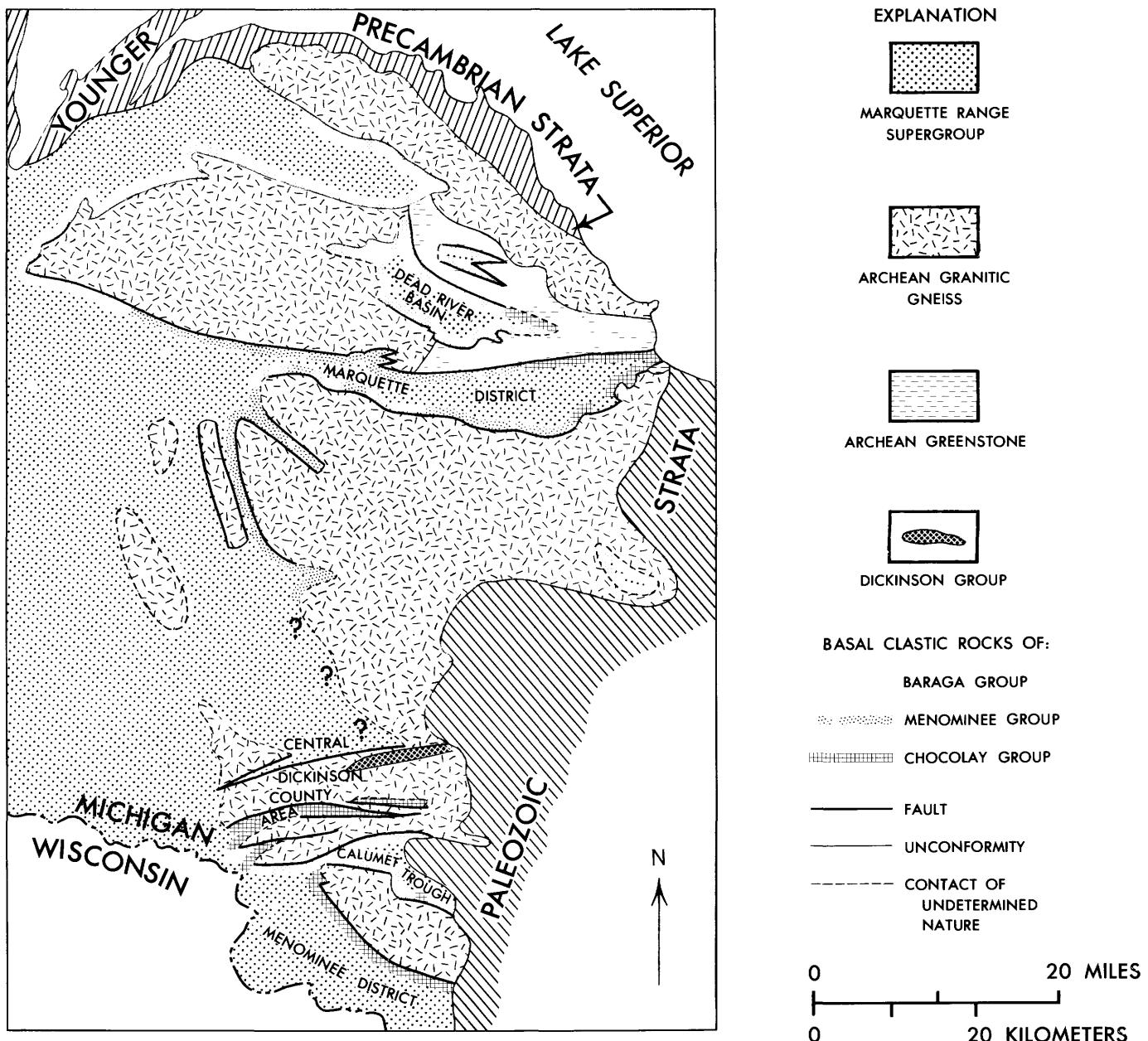


FIGURE 3.—Geologic map of the central part of the northern peninsula of Michigan showing the distribution of basal clastic rocks of the Chocolay, Menominee, and Baraga Groups. Where no pattern is shown, basal rocks are volcanic, iron-formation, or dolomite.

#### BARAGA GROUP

The Baraga Group is a very thick accumulation of eugeosynclinal graywacke, shale, iron-formation, and volcanic rocks, but the basal part of the section is generally a shallow-water transgressive sequence that contains the most extensive basal conglomerates in the Marquette Range Supergroup. Typically, coarse clastic rocks at the base grade upward into slates and intercalated iron-formation. Less commonly, slate and iron-

formation lie directly on Archean rocks with only local lenses of basal conglomerate.

Three types of conglomerate are distinguished:

1. Thin (3 m or less) oligomictic ferruginous chert-pebble conglomerate derived mostly by reworking of iron-formation of the Menominee Group. Such rocks are most common where the Baraga Group lies directly on the Menominee Group but are also found locally lying on Archean rocks.

2. Thin, locally derived lenses of conglomerate containing mostly fragments of immediately underlying Archean rocks in a quartzose or chloritic matrix (fig. 6A). Some lenses are demonstrably related to the flanks of local topographic highs on the Archean surface and are most likely wave-washed bedrock rubble.

3. Wedges as much as 100 m thick of polymictic conglomerate with clasts generally 1-10 cm in diameter but locally as much as about 1 m in diameter (fig. 6B). Clasts are mostly iron-formation, vein quartz, and older sedimentary rocks. The matrix is quartzose and typically very hematitic. Four such wedges are known, all in the

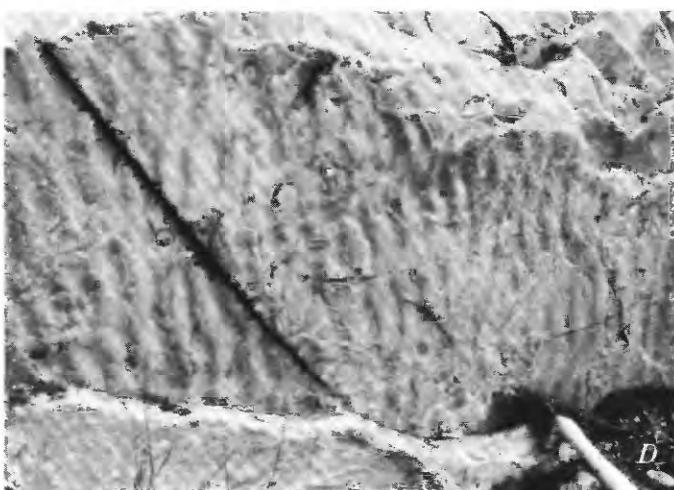


FIGURE 4.—Basal clastic rocks of the Chocolay Group. A, Tillite composed of large clasts of granitic rock in chloritic matrix. B, Fluviaatile deposits of granitic pebble conglomerates interbedded with finer grained arkose and argillite. C, Dropstone clast of granite in chloritic argillite. D, Ripple-marked marine orthoquartzite.



FIGURE 5.—Quartz-pebble conglomerate at base of Menominee Group. Subangular to rounded pebbles of vein quartz are in arkosic matrix. Magnet is about 4 cm in diameter.

Marquette district. Two are demonstrably positive depositional forms, deltas or alluvial fans, rather than channel deposits, and their spatial distribution suggests that they were derived from an early uplift of the Archean rocks that now bound the Marquette trough on the south. One wedge, in the eastern Marquette district, contains monazite-rich placers within quartz-pebble conglomerate; grades are locally as high as 110 lb of monazite per short ton (55 kg per metric ton) (Vickers, 1956a). All four wedges lie unconformably on the Menominee Group, but the abundance of vein-quartz pebbles in the conglomerates indicates that Archean basement rocks were being eroded and transported to the depositional basin.

#### RELATION TO THE HURONIAN SUPERGROUP

The relation of the Marquette Range Supergroup to the Huronian Supergroup has been in dispute for nearly a century and is still not known with certainty in spite of

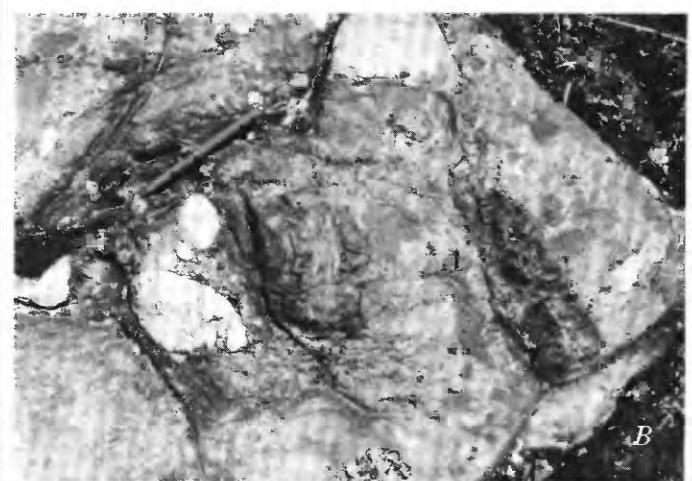


FIGURE 6.—Basal conglomerate of Baraga Group. A, Conglomerate composed of angular fragments derived from underlying granitic gneiss and rounded fragments of vein quartz (lighter colored clasts). Contact of conglomerate with gneiss (gr) shown by dotted line. B, Conglomerate with fragments of iron-formation from underlying Menominee Group and well-rounded vein quartz fragments derived from a more distant Archean source area.

numerous radiometric age determinations. This problem is especially important in assessing the uranium potential of conglomerates in the Marquette Range Supergroup because of the large uranium deposits of the Blind River district in basal rocks of the Huronian.

For many years the rocks of the Marquette Range Supergroup were called Huronian. Early workers suggested the correlation based on similarities in rock types, approximately equivalent age, and close geographic proximity. In 1958, James (1958) suggested that the rocks in Michigan be called Animikie Series because correlation with the Animikie Group in Minnesota seemed more certain than with the Huronian. In 1970 (Cannon and Gair, 1970) the term "Animikie" was abandoned and the rocks were given the local name of Marquette Range Supergroup, primarily to avoid conflict with the stratigraphic code of the U.S. Geological Survey.

Over the past ten years radiometric age determinations have helped to define the absolute ages of the Huronian Supergroup and the Marquette Range Supergroup. Both sequences lie unconformably on Archean rocks at least 2.6 b.y. old. In Ontario, dikes and sills of Nipissing diabase were intruded after Huronian sedimentation and probably after early phases of regional deformation of the Huronian. An age of about 2.15 b.y. is well established for the Nipissing (Van Schmus, 1965; Fairbairn and others, 1969) and seems a reliable minimum age for the Huronian. Fairbairn and others (1969) determined an age of 2.29 b.y. for argillite in the Gowganda Formation, which may be the time of deposition or diagenesis. Thus deposition of the Huronian Supergroup is rather reliably bracketed between 2.6 and 2.15 b.y. ago.

In Michigan, the Marquette Range Supergroup was deformed and metamorphosed about 1.9 b.y. ago (Banks and Van Schmus, 1971; Goldich and others, 1961; Aldrich and others, 1965). Banks and Van Schmus (1971) report a zircon concordia age of 2.1 b.y. for "porphyritic red granite" that forms two small plutons surrounded by Archean rocks in central Dickinson County. These plutons were interpreted to have been emplaced prior to deposition of the Marquette Range Supergroup (James and others, 1961) on the basis of internal structures that were not believed consistent with post-Marquette Range structures. Banks and Van Schmus therefore considered that 2.1 b.y. was a maximum age for the Marquette Range Supergroup, an age which, if correct, invalidates any correlation with the Huronian.

It is my opinion that enough uncertainties exist in the data that the radiometric age should not be considered a maximum age for the Marquette Range Supergroup for several reasons. First, the porphyritic red granite has had a complex postemplacement history, including granulation and recrystallization of minerals (James and

others, 1961), and the radiometric age may not be the time of emplacement. No intrusive rocks or thermal events of this age are known elsewhere in the area. Second, radiometric determinations on other rocks only a short distance from the porphyritic red granite yield spurious results and in places do not define an Rb-Sr isochron (Banks and Van Schmus, 1971), suggesting that isotopic systems have been disturbed, perhaps over a large area. Finally, the porphyritic red granite is not proved to be older than the Marquette Range Supergroup and is nowhere in contact with it; the proposed age relations are based solely on structural arguments. Thus, even if 2.1 b.y. is the age of emplacement of the porphyritic red granite, it cannot, with complete confidence, be considered to define the age of the Marquette Range Supergroup.

A single age on volcanic rocks in the Baraga Group is about 2 b.y. (Banks and Van Schmus, 1971). If this age is correct, the Baraga Group is younger than the Huronian, but correlation of the Menominee and Chocolay Groups with the Huronian is still possible. A correlation of the Gowganda Formation of the Huronian with basal Chocolay Group tillites of the Marquette Range Supergroup and of the Lorrain Quartzite with quartzites of the Chocolay Group has been suggested by Young (1973). This correlation is attractive in light of striking lithologic similarities of these parts of the section and of the relative "uniqueness" of continental glacial deposits in time, but it is weakened by the presence of other conglomerates in the Huronian, also believed to be of glacial origin (Young and Chandler, 1968; Roscoe, 1969; Frarey and Roscoe, 1970), which could also be correlated with the Chocolay Group.

In my opinion, the question of correlation cannot be answered adequately with existing data, and the possibility should be held open that part or all of the Marquette Range Supergroup is equivalent in age to the Huronian Supergroup. Surely, in evaluating the possibility of the occurrence of uraniferous quartz-pebble conglomerates in Michigan, the Marquette Range Supergroup should not be excluded solely on the basis of the scant radiometric data suggesting that it might be younger than rocks in the Huronian to which known occurrences of such deposits seem to be restricted.

In the following sections are listed some major similarities and differences between the Marquette Range Supergroup and Huronian Supergroup that illustrate the geologic grounds on which the question of correlation can be argued.

#### SIMILARITIES

1. Both supergroups are thick sedimentary accumulations deposited sometime during the interval 2.6 to 1.9 b.y. ago.

2. Both contain relatively uncommon Precambrian rock types, such as glacial deposits, thick orthoquartzites, and carbonate rocks.
3. Both were intruded by major systems of mafic dikes and sills. The Huronian was intruded after sedimentation about 2.15 b.y. ago. The Marquette Range Supergroup was probably intruded several times, the major event being during volcanism during Baraga Group deposition (Cannon, 1973), for which one age of 2 b.y. has been obtained (Banks and Van Schmus, 1971).
4. The two supergroups are in close geographic proximity and approximately on strike with each other (fig. 1). In outcrop they can be traced to within about 250 km of each other, the intervening area being covered by Paleozoic rocks or the waters of Lake Superior. On aeromagnetic maps, trends of the Marquette Range Supergroup can be traced eastward beneath Paleozoic cover for several tens of kilometers before being truncated by trends of younger Keweenawan rocks. The Keweenawan rocks are now believed by many to occupy an arcuate rift with roughly the shape of a southward opening U, the northern apex being approximately in the center of Lake Superior. The interior portion, including the Marquette Range Supergroup, is believed to have moved southward as the rift opened about 1.1 b.y. ago. If this is correct, the Marquette Range Supergroup was deposited somewhat north of its present location with respect to the Huronian and was, prior to Keweenawan rifting, even closer to the Huronian than at present. Taking these factors into consideration, it is likely that no more than 150 km separated the two supergroups prior to 1.1 b.y. ago. Considering the likelihood that the sedimentary rocks originally extended well beyond their present outcrop areas, it is not unlikely that both could have formed in the same sedimentary basin.

#### DIFFERENCES

1. The Marquette Range Supergroup contains thick deposits of iron-formation at several stratigraphic horizons, whereas iron-formation is virtually unknown in the Huronian.
2. Deposition of the Marquette Range Supergroup culminated in accumulation of a very thick sequence of eugeosynclinal deposits. A similar tectonic environment of sedimentation is not recognized in the Huronian.

#### ORIGIN OF URANIFEROUS QUARTZ-PEBBLE CONGLOMERATES AND THE POTENTIAL FOR THEIR OCCURRENCE IN MICHIGAN

##### ORIGIN OF BLIND RIVER DISTRICT URANIUM ORE

Because of the many similarities between the Huronian rocks that contain the Blind River uranium deposits and the Marquette Range Supergroup, considering the origin of Blind River ore is one means of assessing the possibility that similar ore might be found in Michigan.

The mode of origin of Blind River uranium ore has been controversial, virtually since the deposits were discovered more than 20 years ago. It is not my purpose to discuss these arguments in detail. Many of my co-authors in this volume are much more qualified than I, and the subject is thoroughly covered. In general, opinion has been divided between two points of view, one that favors an epigenetic (hydrothermal or groundwater) origin for uranium mineralization and the invariably associated pyrite and a second that favors a syngenetic (clastic) origin for uranium minerals and pyrite. Among supporters of the latter view are included J. A. Robertson (1968), D. S. Robertson (1974), and Roscoe (1969, 1973), all of whom are intimately acquainted with Blind River deposits through many years of research. Judging from the most recent literature, opinion is approaching a consensus in favor of a clastic origin.

Recently, Roscoe (1969, 1973) and D. S. Robertson (1974) have expanded the theory of placer origin to include implications on the evolution of the atmosphere. In their view, the atmosphere contained no free oxygen prior to about 2.2 b.y. ago, thus allowing minerals such as uraninite and pyrite, unstable in an oxygen-rich atmosphere, to be eroded, transported, and accumulated into placers. They believe the atmosphere began to accumulate free oxygen shortly after deposition of the basal Huronian units, thus inhibiting further development of such placers. They based their arguments on the drab color and generally unoxidized nature of lower Huronian rocks, including the uraniferous conglomerates, and of underlying fossil soils, in contrast to more oxidized units in the upper parts of the Huronian and in other sequences believed to be slightly younger than the Huronian. D. S. Robertson described such sequences on a worldwide scale and suggested that the change in oxidation state of the sediments reflects a unique point in atmospheric evolution that can be used as a world-wide time marker. In the view of Roscoe and D. S. Robertson, uraniferous conglomerates must be restricted to rocks older than the oldest rocks in the Marquette Range Supergroup.

#### THE PROBLEM OF ATMOSPHERIC OXYGEN

The question of when free oxygen first appeared in the atmosphere has now become a principal issue in considering the origin and stratigraphic distribution of uraniferous quartz-pebble conglomerates, and I would like to discuss briefly some general points before discussing evidence from the Marquette Range Supergroup in more detail. Many researchers have concluded that the presence of vast iron-formations about 2 b.y. old indicates that the atmosphere was anoxic at that time so that iron could be transported in the relatively soluble ferrous form. An oxygen partial pressure of about  $10^{-70}$  atm is required to permit significant concentrations of dissolved ferrous iron in water. The presence of oxide-rich iron-formation throughout much of recorded geologic time prior to 2 b.y. ago requires a source to supply oxygen during deposition if ferric iron minerals were precipitated from dissolved ferrous iron. If the atmosphere were anoxic, some alternate source is needed, and Cloud (1965, 1973) has suggested that primitive photosynthetic microorganisms produced oxygen that was immediately consumed by ferrous iron in sea water. Thus, oxygen partial pressure was buffered at about  $10^{-70}$  atm as long as sufficient ferrous iron was in the oceans and iron-formation was precipitated in the process. In Cloud's view, this process culminated about 2 b.y. ago in a great episode of iron-formation deposition, consuming the supply of ferrous iron and allowing oxygen to begin to accumulate in the atmosphere.

Recently, Holland (1973) and Drever (1974) have proposed models that can account for iron-formations with an atmosphere containing substantial free oxygen. Drever suggested that with an atmosphere containing about one-fifth or less of present oxygen levels and with sufficient biologic productivity to supply organic carbon, ocean water below the thermocline would be anoxic because of uptake of oxygen by free carbon. Iron in detrital sediments deposited below the thermocline would be partially dissolved to ferrous iron and accumulate to a concentration of about 10 ppm (parts per million). As this water welled up into shallower levels, it became oxidized by contact with the atmosphere and precipitated iron minerals as the solubility of iron was diminished.

Although this model is not demonstrably the means by which iron-formation was deposited, it does, nevertheless, provide a plausible means of forming iron-formation under an oxygen-rich atmosphere, and the presence of iron-formation cannot be taken as evidence of an anoxic atmosphere with as complete confidence as has been done by many workers.

These arguments concerning iron-formation have important bearing on the possible occurrence of uraninite and pyrite as placer minerals and should be considered

along with more direct evidence from the uraniferous conglomerates themselves. If one accepts models for the origin of iron-formation requiring an anoxic atmosphere, with oxygen partial pressures about  $10^{-70}$  atm, then one must also accept that uraninite and pyrite are stable so long as iron-formations are being deposited, because both minerals remain stable at oxygen pressures much greater than  $10^{-70}$ . For instance, Holland (1962) has calculated that uraninite remains stable until oxygen partial pressure reaches about  $10^{-21}$  atm. Thus, the presence of iron-formation could be taken to indicate that both pyrite and uraninite were stable under the atmosphere existing at the time when iron-formations were deposited.

If, on the other hand, one accepts a model such as Drever's, in which iron-formations are formed under a relatively oxygen-rich atmosphere, then the presence of iron-formation cannot be used to conclude that pyrite-uraninite placers could be formed stably. Rather, it would be likely that both minerals would be unstable, at least in a strict thermodynamic sense, under such an atmosphere.

#### EVIDENCE FOR AN OXIDIZING ATMOSPHERE DURING DEPOSITION OF THE MARQUETTE RANGE SUPERGROUP

Pyrite and uraninite have never been found in large quantities in conglomerates in the Marquette Range Supergroup, and there have been no studies bearing directly on the stability of uraninite and pyrite during deposition. Some features in both iron-formation and weathered zones along unconformities, however, do contain evidence of an oxygen-rich atmosphere during at least part of the time of deposition of the supergroup and indirectly bear on the potential for pyrite-uraninite placers in Michigan.

#### EVIDENCE FROM IRON-FORMATION

The Negaunee Iron-formation, the principal iron-formation of the Menominee Group in the Marquette district, contains textural and mineralogical features that suggest an oxygen-rich atmosphere during deposition. The Negaunee, which contains many lithologic types of iron-formation, includes persistent stratigraphic units of wavy-bedded jaspilite containing irregular, lenticular, commonly oolitic beds of jasper that are interbedded with specular hematite (fig. 7A). The wavy bedding and oolitic character indicate a high-energy and presumably shallow-water environment of deposition. Other units contain more reduced assemblages that, where not strongly altered by metamorphism, include siderite, minnesotaite, and magnetite. They typically are rather even bedded, suggesting quiet water during deposition (fig. 7B).

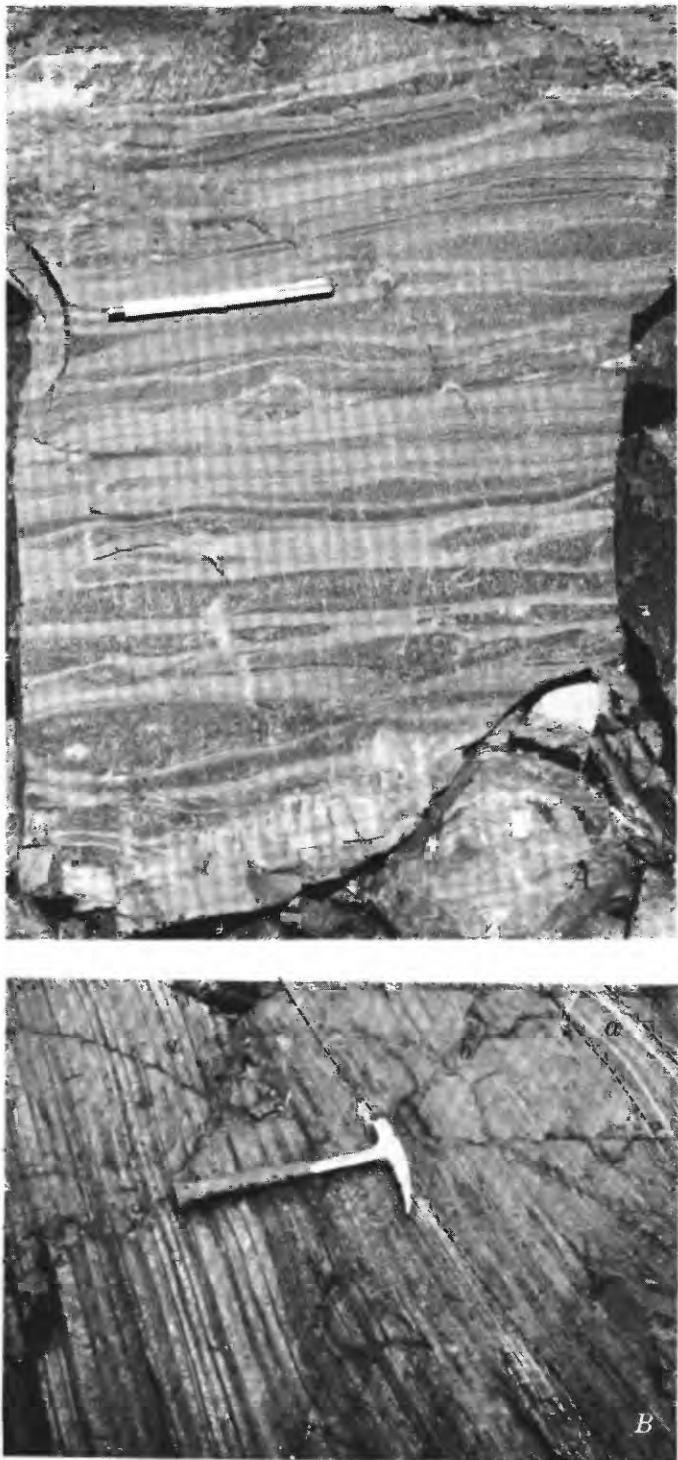


FIGURE 7.—Iron-formation of the Menominee Group. *A*, Wavy-bedded jaspilite with irregular beds of jasper (darker layers) and interlayers of specular hematite (lighter layers). *B*, Even bedded chert-magnetite-iron silicate iron-formation (units *a*) in contact with wavy bedded jaspilite (units *b*).

In much of the district the correlation of bedding characteristics with oxidation state of iron is strikingly consistent. Even-bedded iron-formation, presumably of deep-water deposition, contains reduced mineral assemblages, whereas wavy-bedded oolitic iron-formation, of shallow-water deposition, generally contains thoroughly oxidized minerals. One outcrop shows an interlayering of these lithologic types in beds a meter or less thick (fig. 7*B*), and the abrupt change in both bedding characteristics and oxidation state is repeated many times across the outcrop. These lithologic changes must reflect slight variations in water depth or depth of wave action, so that wavy-bedded hematitic iron-formation was deposited above wave base, whereas even-bedded, more reduced iron-formation was deposited below wave base. It follows that the source of oxygen to form the thoroughly oxidized wavy-bedded jaspilite was the atmosphere. The agitated water in which wavy-bedded jaspilite was deposited brought oxygen-rich water to the sediment-water interface, so that all iron was oxidized to the ferric state. The quieter water in which even-bedded iron-formation was deposited did not maintain sufficient chemical communication with the atmosphere to provide enough oxygen to oxidize all available iron, and magnetite, iron carbonate, and iron silicates were therefore stable.

#### EVIDENCE FROM WEATHERED ZONES

Northern Michigan is an area of very poor bedrock exposure. Consequently, exposures of unconformities are rare. Rarer still are exposures showing effects of weathering in rocks beneath unconformities. Nevertheless, enough data have been gathered over many years of study to substantiate some rather firm conclusions.

The most extensive weathered zone known is developed beneath the unconformity separating the Menominee and Baraga Groups and is best developed in the Negaunee Iron-formation in the Marquette district (fig. 8). Van Hise and Leith (1911) proposed that jaspilite near the top of the Negaunee Iron-formation formed by weathering of originally sideritic iron-formation. The presence of fragments of jaspilite in basal conglomerates of the overlying Baraga Group was taken as evidence that weathering had taken place before Baraga Group deposition. James (1954), who argued that jaspilite was a primary facies of iron-formation and had not formed by oxidation of sideritic iron-formation, cast doubt on Van Hise and Leith's weathering hypothesis. Certainly the features discussed in the previous section

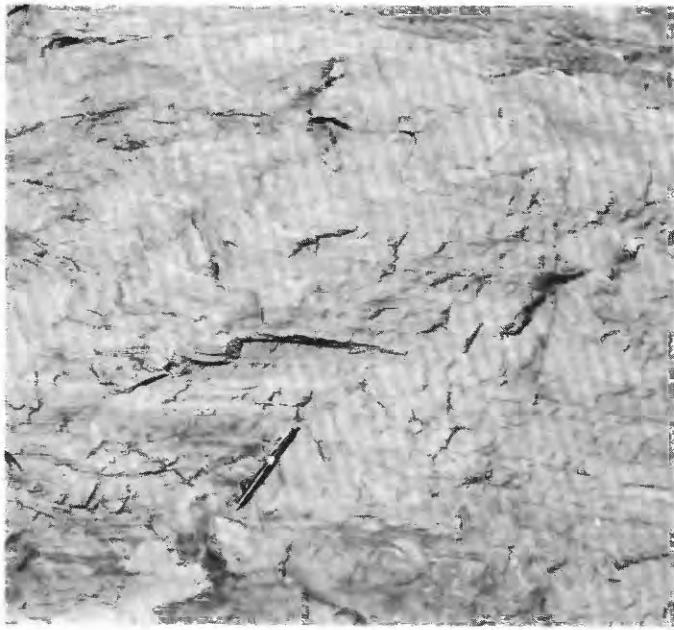


FIGURE 8.—Even-bedded jaspilite with uniformly layered jasper (darker layers) and interbeds of specular hematite (lighter layers).

indicate that some jaspilite is a primary sediment, yet other evidence strongly suggests that some jaspilite has formed by weathering and oxidation of sideritic iron-formation. Gair (1975) describes exposures in the Cliff Shaft Mine in the eastern part of the Marquette district, in which a thick jaspilite unit contains "islands" of sideritic iron-formation and jaspilite grades on strike into sideritic iron-formation. These changes undoubtedly occurred before regional deformation and metamorphism about 1.9 b.y. ago, and because such rocks are restricted to a stratigraphic interval immediately below the pre-Baraga Group unconformity, they seem likely to have formed by oxidative weathering as originally proposed by Van Hise and Leith.

In many places the Baraga Group lies directly on Archean granitic rocks or greenstone (fig. 3), and during the past year I have searched for evidence of weathering in these rocks beneath the unconformity. At only two localities, both in the Dead River basin, have I found features that might be caused by oxidative weathering. At both localities, gray granitic gneiss beneath the unconformity has a thin pink zone developed on it immediately beneath the unconformity (fig. 9A). The zone, which varies from about 1 cm to several centimeters in thickness, may be caused by oxidation of the gray gneiss. The change in color is caused primarily by a color change in plagioclase. White to gray plagioclase, typical of most of the gray gneiss, is altered to pink and orange near the unconformity. In addition, mafic components, mostly biotite, apparently were leached from the pink zone. The basal sediments of the Baraga Group are black

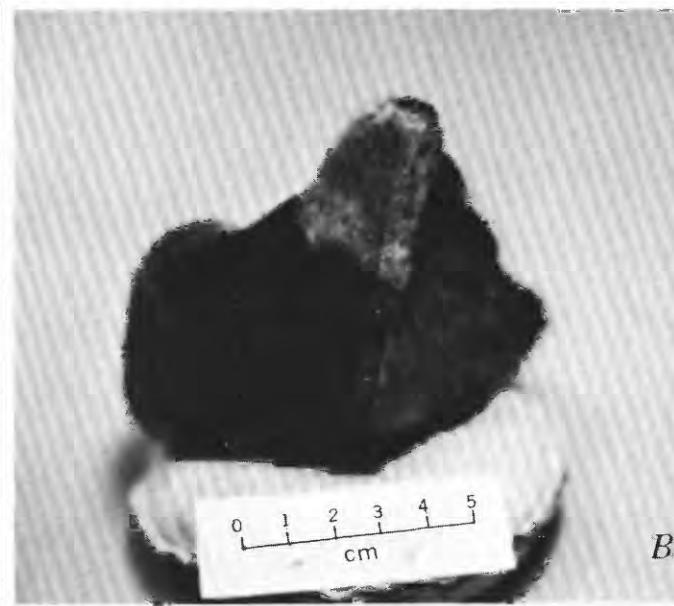


FIGURE 9.—Oxidized granitic rocks at base of Baraga Group. A, Gray granitic gneiss (a) converted to pink granitic gneiss (b) beneath unconformable contact with black pyritic shale (c) at base of Baraga Group. B, Pebble of gray granitic gneiss (upper part of specimen) with rind of pink oxidized granite in black pyritic shale of basal Baraga Group.

pyritic shale that shows little or no oxidation, so the pink zone beneath the unconformity must have formed before sedimentation. A few pebbles of the gray gneiss in local conglomerate lenses have similar pink rinds (fig. 9B) that also were probably formed by oxidative weathering prior to sedimentation.

At the same localities, pockets of deeply weathered limonitic material are along the unconformity. The

nature of this material prior to recent oxidation to limonite is not known but it may be a remnant of iron-rich residual soil that would be expected to form under an oxidizing atmosphere.

Elsewhere, Archean rocks beneath the unconformity show no effects of weathering or oxidation. If weathered zones or residual soils had formed, they were apparently totally removed and reworked during Baraga Group transgression. Thus weathering and oxidation appear to have been more extensive in the iron-formation, which is similar to the case prevalent today. Weathering during late Precambrian or Phanerozoic time has, in places, oxidized iron-formation for hundreds of meters beneath the surface, whereas weathering of surrounding rocks was not deep enough for weathered zones to have survived Pleistocene glaciation.

In summary, two independent lines of evidence lead to the conclusion that the atmosphere contained abundant free oxygen at least as early as the close of Menominee Group sedimentation, at least 2 b.y. ago. Iron-formation was apparently readily formed under such an atmosphere. Extensive iron-formations were deposited in at least four younger stratigraphic intervals of the Marquette Range Supergroup. From this I conclude that the presence of iron-formation cannot be used to infer an anoxic atmosphere and is therefore not a reliable guide in assessing the potential for placer accumulation of uraninite.

Data on weathering along unconformities at the base of the Chocolay and Menominee Groups are very scant, but no evidence of oxidation along these unconformities has been found. The only occurrence of a regolith, to my knowledge, is beneath the Menominee Group in the western Marquette district. At two localities the basal conglomerate of the Menominee Group is separated from underlying Archean granitic gneiss by a zone of gray-green chloritic and sericitic material believed to be a regolith. The material forms a unit from 1 to about 3 m thick that grades downward into granitic gneiss and lies beneath conglomerate composed of pebbles of vein quartz, chert, and older sedimentary-rock fragments. The base of the conglomerate and upper parts of the regolith contain minor pyrite and magnetite. The rocks were metamorphosed to staurolite grade, and pyrite is generally recrystallized to well-formed cubes, but, rarely, rounded, possibly detrital grains are preserved. Although these rocks might be interpreted to have formed in an anoxic atmosphere, data are too sparse to justify any firm conclusions.

#### SIGNIFICANCE OF OXIDIZED ROCKS

The two independent lines of evidence just discussed indicate that by the close of deposition of the Menominee

Group the atmosphere contained sufficient free oxygen to oxidize a substantial amount of iron. A problem arises of how to interpret this in terms of oxygen partial pressure in the atmosphere and its bearing on the ability of uraninite to persist in a clastic sedimentary environment. The stability relations of the minerals involved are rather well known and provide a first step in solving this problem. Hematite becomes the stable iron oxide at vanishingly small oxygen partial pressures ( $\sim 10^{-65}$  atm) and has a very broad stability field. Uraninite remains stable at oxygen partial pressures up to about  $10^{-21}$  atm (Holland, 1962) and has a large overlap with the stability field of hematite. Thus, uraninite and hematite can be in equilibrium over a wide range of partial pressures, and the presence of hematite in a rock does not automatically preclude the presence of uraninite under equilibrium conditions.

The problem is complicated, however, by two considerations: (1) It is possible that the atmosphere contained free oxygen far in excess of minimum amounts required to stabilize hematite; (2) concentration of a mineral in placers may reflect its ability to persist metastably rather than its true thermodynamic stability.

Considering first the amount of free oxygen required to form the oxidized rocks in the Menominee Group, it is necessary to consider some quantitative and kinetic aspects of oxidation as well as intensive variables that govern mineral stabilities. The formation of oxide facies iron-formation and of extensive oxidized zones along the post-Menominee Group unconformity requires that a large amount of oxygen be delivered to the site of oxidation in a geologically short period of time. The erosional break between the Menominee and Baraga Groups cannot be a long time span, because Gair (1975) has found that the two groups are locally conformable and interbedded. The rate of sedimentation of oxide facies iron-formation cannot be accurately estimated, but assuming that it is not greatly dissimilar from rates determined from varved layers in iron-formation in the Hammersley Range, Australia, by Trendall and Blockley (1970), it, too, represents a short time period. An atmosphere containing only the minimum oxygen level to form hematite ( $\sim 10^{-70}$  atm) is for practical purposes anoxic and seems very unlikely to be capable of supplying sufficient oxygen in the required time span to form the hematitic rocks. Even at the upper stability limit of uraninite ( $\sim 10^{-21}$  atm), the entire atmosphere probably contained no more than a few tens of grams of oxygen. The entire atmospheric oxygen supply could oxidize literally no more than a handful of uraninite during oxidative weathering.

In my opinion, rocks such as those in the Menominee Group that appear to have been oxidized as a result of contact with the atmosphere must have formed under ar-

atmosphere with oxygen levels well in excess of minimum amounts required to stabilize hematite and oxidize uraninite. If this were not the case and the atmosphere contained only a few tens of grams of oxygen, that gas would be so widely dispersed that even in a dynamic system in which  $O_2$  is continually being supplied by photosynthesis or photodissociation of water and consumed by oxidation of reduced compounds, it seems unlikely that the large quantities of  $O_2$  that were evidently supplied to weathered zones in the Marquette Range Supergroup could be delivered to the site of oxidation in a short time span. The preserved portions of the oxidized rocks contain many thousands of tons of  $O_2$  presumably derived from the atmosphere, and the original zone of oxidized rocks may have contained literally billions of tons of  $O_2$ . The principal question is, could an atmosphere that at any one moment contained only a fraction of a ton of  $O_2$  produce so much oxidation? I believe the answer is no; the atmosphere probably contained a great deal more  $O_2$ .

This, then, brings us to the second consideration of whether uraninite could persist to form placer deposits under such an atmosphere. Although uraninite may be well removed from its stability field, it might persist metastably. Magnetite, for instance, has an even lower oxygen tolerance than uraninite yet is a common detrital mineral even today as a result of its relatively slow reaction rate. The preservation of uraninite, like any other metastable detrital mineral, depends on rates of weathering, erosion, and burial relative to the rate of breakdown. The rate of oxidative breakdown of uraninite should be controlled by the partial pressure of oxygen and should therefore increase with time as the atmosphere evolved from oxygen-free toward the present oxygen levels. I would suggest, therefore, that the restriction in time of uraniferous and pyritic conglomerates cannot be defined by an absolute time marker about 2.2 b.y. ago, as has been suggested, particularly if the first appearance of hematite is used to define the onset of oxidizing conditions. Rather, it seems more likely that such deposits would gradually become scarcer in younger rocks as oxygen levels increased, requiring progressively more favorable conditions to preserve uraninite or requiring the occurrence of uranium in more thorium-rich and hence more oxygen-tolerant minerals. The presence of modern-day uraninite-bearing placers in the Indus River (Miller, 1962; Zeshke, 1961) supports this view.

In light of these considerations, uranium-bearing conglomerates might be less likely to have formed during deposition of the Marquette Range Supergroup than during the Huronian if the Marquette Range Supergroup is partly younger than the Huronian, but their presence is certainly not precluded.

#### POTENTIAL FOR URANIUM-BEARING QUARTZ-PEBBLE CONGLOMERATE IN THE MARQUETTE RANGE SUPERGROUP

The foregoing arguments in favor of a strongly oxidizing atmosphere during deposition of at least part of the Marquette Range Supergroup suggests that the conditions may have been less favorable for formation of quartz-pebble uranium ore than they were during deposition of the Huronian. Nevertheless, our understanding of the requisite conditions for their formation and of the age relations between the Marquette Range Supergroup and Huronian are still uncertain enough so that the uranium potential of quartz-pebble conglomerates in the Marquette Range Supergroup is best judged directly by considering features of the rocks themselves.

A few positive and negative factors based on local geologic features are discussed in the following paragraphs.

Favorable factors include:

1. Lenses of quartz-pebble conglomerate are relatively common near the base of the sedimentary section and provide a large volume of potentially mineralized rock.
2. Archean granitic rocks in many areas of northern Michigan are anomalously rich in uranium (fig. 10) and could have provided a source for radioactive minerals for overlying sediments. One bulk sample of Archean granitic gneiss contains 15 ppm  $U_3O_8$  and 102 ppm  $ThO_2$  (Malan and Sterling, 1969), and 27 samples collected throughout northern Michigan average 16 ppm  $U_3O_8$  and range from 3-40 ppm (Kinnaman and Illsley, 1962).
3. Uranium concentrations in slate and in iron-formation in several areas within higher stratigraphic units of the Marquette Range Supergroup (fig. 10) indicate that uranium was being supplied to the sedimentary basin (Vickers, 1956b).
4. The presence of monazite-rich placers in parts of the basal Baraga Group (Vickers, 1956a) indicates that physical conditions of sedimentation were suitable to form significant placer accumulations of minerals that were originally disseminated in older rocks.
5. Fragmentary evidence suggests that basal sediments of the Chocolay and Menominee Groups are not strongly oxidized. The only known regolith is drab-colored magnetite and pyrite-bearing rock. Pyrite, although not necessarily detrital, is a minor but widespread mineral in basal quartzites of both the Chocolay and Menominee Groups (Tyler and others, 1940).

6. Glaciation at the beginning of Chocolay sedimentation should have provided a large volume of freshly ground and, at least locally, uranium-rich Archean rocks that may have been reworked and incorporated into overlying rocks, possibly with resultant concentration in placers. Where glacial deposits lie on anomalously radioactive rocks, they, too, are anomalously radioactive, locally somewhat more radioactive than are the underlying gneisses.
7. Scant data from the Chocolay and Menominee Groups indicate the Th/U ratios of the conglomerates are low; most are in the range 2:1 to 4:1. These are comparable to or somewhat lower

than ratios from Archean rocks and indicate that thorium was not being preferentially retained over uranium in the clastic sedimentary environment. Ratios from the Baraga Group are much higher but are strongly influenced by a relatively large number of samples from monazite-rich placers.

Unfavorable factors include:

1. The contact between the Marquette Range Supergroup and the Archean basement is a fault in many areas. The tectonic style of the area, which I have discussed in more detail elsewhere (Cannon, 1973), consists basically of upfaulted blocks of Archean basement rocks with the sedimentary rocks

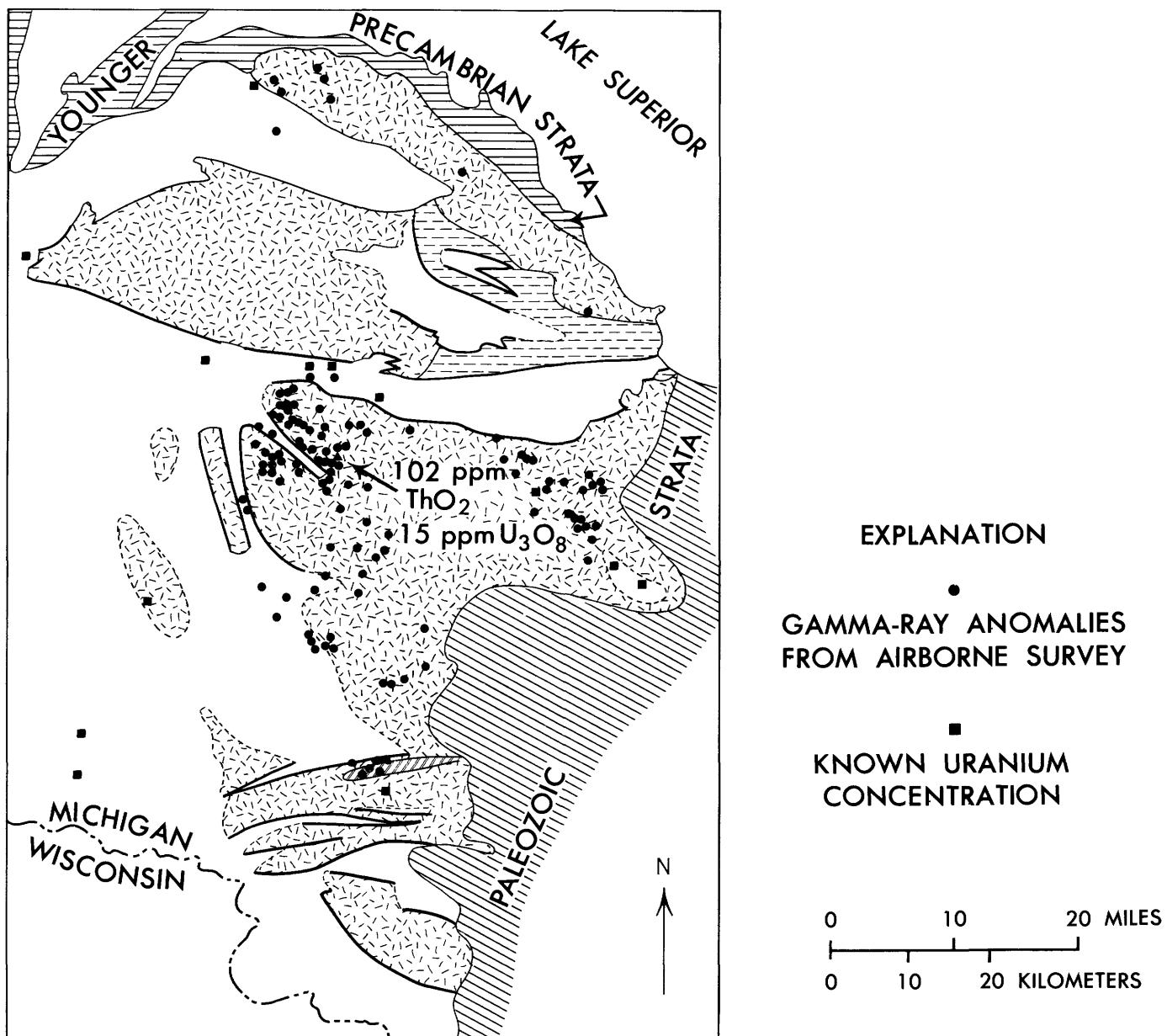


FIGURE 10.—Map showing distribution of gamma-ray anomalies from airborne survey (Balsley and others, 1950) and known uranium concentrations (Vickers, 1956b).

of the Marquette Range Supergroup draped over the uplifts. The contact is intensely sheared in many places, and the lowest part of the section is eliminated by faulting. Thus, the basal units of principal interest for uranium exploration are not present in some areas. Because of poor exposure and the general conformity of bedding with the basal contact, even where the contact is a fault, it is commonly difficult to judge the extent of fault truncation of the base of the section. Figure 3 shows my best estimate of the nature of the Marquette Range Supergroup-Archean contact and indicates which segments of the contact are relatively undisturbed unconformity and which are faults.

2. In spite of a great deal of geologic mapping and exploration in the area, albeit most was not concerned with uranium exploration, no significant deposits have been found. Radioactivity of most conglomerates is only slightly higher than that of adjacent Archean rocks. Rarely, radioactivity reaches three times that of the adjacent Archean rocks. As discussed in more detail in the following section, this may not be an overriding negative factor. Because of the very limited exposures of bedrock there is a substantial probability that even a major district would not be discovered in outcrop.

#### **EXPLORATION FOR URANIFEROUS QUARTZ-PEBBLE CONGLOMERATE IN MICHIGAN—PROBLEMS AND DISCOVERY PROBABILITIES**

In the previous discussions many similarities and differences between the Marquette Range Supergroup and the Huronian have been considered. One additional important difference is the amount of Pleistocene drift cover on bedrock. The Huronian rocks near Blind River are very well exposed because of thin drift cover; indications of several of the known ore deposits were seen initially in outcrop. In Michigan, on the other hand, glacial deposits are thick and bedrock exposures are rare. Much less than one percent of the outcrop belt of the basal clastic units is exposed. Exploration of these units therefore requires a technique capable of detecting mineralized rock through drift cover generally several meters thick and commonly several tens of meters thick. A number of standard geochemical and geophysical techniques might detect buried ore bodies, but I would like to illustrate here that blind diamond drilling might also be a valuable and relatively inexpensive technique. By blind drilling I mean that drill holes would not be located to explore known anomalies, but rather would simply be located so as to intersect known basal clastic zones. There are about 650 km of strike length of basal clastic units lying on Archean basement. The location of

the contact is rather accurately known from detailed mapping for most of that length. By analogy with Blind River, uranium ore is most likely to be in the lowest 50 m of the sedimentary section. Thus the total exploration target is relatively small and well defined. Maps of the Blind River district (J. A. Robertson, 1968) indicate that ore bodies or mineralized rock typically have a strike length of about 1.5 km. If deposits of that size were in Michigan, a single deposit would occupy about one four-hundredths of the strike length of the outcrop belt to be explored, and the probability of hitting it with a randomly placed drill hole would be one in 400. On the other hand, to be certain of hitting it would require 400 drill holes spaced about 1.5 km apart along the outcrop belt. The discovery probability for any other small number of drill holes is a straight-line function between these two extremes. If more than one deposit exists in the area the probability of making a discovery increases exponentially with the number of deposits. Figure 11 shows curves relating number of drill holes to probability of making a single discovery for cases in which 1, 2, 5, and 10 deposits exist. It shows, for instance, that if a major district (10 deposits) comparable to Blind River exists in Michigan, the probability of discovering one deposit would be 0.5 with only 25 drill holes and 0.75 with 50 holes. If a much smaller district (2 deposits) were in Michigan, 200 drill holes would have a 0.75 probability of finding one deposit.

I would suggest, therefore, that blind diamond drilling is a realistic means of conducting a major exploration program in the area. Two hundred drill holes, each about 50 m long, would have a probability of about 0.5 of finding a single existing deposit, about 0.75 of finding one of two existing deposits, and a much higher probability of finding one of a larger number of deposits. Such drilling is modest in comparison to petroleum wildcat exploration, where expected success is commonly no better than 1 in 10.

Figure 11 can also be used to estimate the probability of discovery of a deposit in outcrop. Most of the strike length of basal clastic units has been mapped in detail, and maps are available showing outcrop distribution. Using these data, I have estimated that the total amount of outcrop of the lower 50 m of the section provides information equivalent to about 25 diamond-drilling holes. If a major district comparable in size to Blind River were in Michigan, chances of discovery of a single deposit in outcrop are only about 0.5 after all outcrops have been thoroughly explored. Because all outcrops have not been thoroughly explored to date, the probability that a deposit would be discovered with current knowledge is considerably less than 0.5. If only one major deposit is in Michigan, the probability of finding it in outcrop is very low, only about 0.05.

These relations illustrate the necessity to explore the area in a manner that is not greatly dependent on outcrop information, and they suggest that the failure to find deposits to date should not be viewed as a greatly discouraging factor in considering future exploration.

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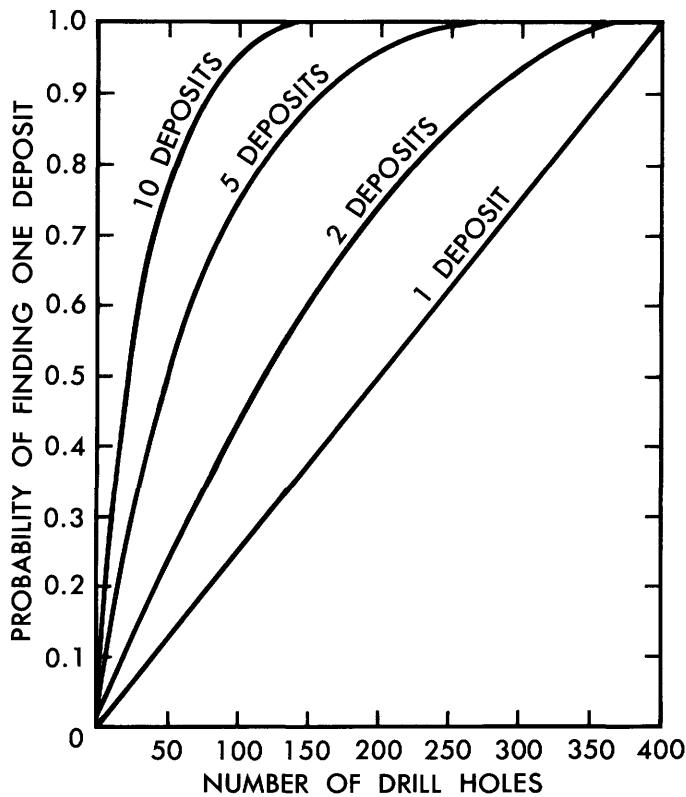


FIGURE 11.—Curves relating probability of discovering one deposit to number of holes drilled for cases in which 1, 2, 5, and 10 undiscovered deposits exist in the central northern peninsula of Michigan. Curves are drawn on the assumption that 650 km of basal clastic rocks are in the unexplored (drift-covered) outcrop belt and a single deposit would extend for 1.5 km along the belt.

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# Genesis of Uranium-Gold Pyritic Conglomerates

*By* W. BRADLEY MYERS

GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN  
QUARTZ-PEBBLE CONGLOMERATES

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GEOLOGICAL SURVEY PROFESSIONAL PAPER 1161-AA





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## GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN QUARTZ-PEBBLE CONGLOMERATES

### GENESIS OF URANIUM-GOLD PYRITIC CONGLOMERATES

By W. BRADLEY MYERS<sup>1</sup>

#### ABSTRACT

The ancient pyritic ore conglomerates have a common origin best exemplified by the Witwatersrand deposits. All contain detrital pyrite and uraninite, which are unstable in modern oxygenated environments and were deposited in a reducing atmosphere. The Rand reefs are not similar to modern gold placers. Placers result from the near incapacity of streams and currents to transport coarse gold. Placers as rich as Rand reef occur only in narrow paystreaks within 15 kilometers of a coarse-gold source.

The broad dispersion of gold in the reefs is due to solution transport of metal complexed as aurous sulfide, leached anoxigenically from crustal rocks, probably from sea-floor basalt, and precipitated by a slow reaction driven by the radioactive decay of detrital uraninite. Radiolysis of water on shallow marine unconformities resulted in diffusion of hydrogen to the atmosphere and a slight excess of hydroxyl free radical in the reef environment. The mild oxidizing tendency slowly dissolved uraninite, precipitated gold, and oxygenated thucholite. These actions define a maturing process. A uraninite placer accumulating on an unconformity becomes progressively converted to a gold reef with little residual uraninite. The most mature reefs tend to grade toward the thucholite-seam type, very thin but exceedingly rich in gold. A combination of chemical attack and physical reworking accounts for the general thinness of mature reefs.

Pyrite, like uraninite, decreases in abundance with increasing maturity; "buffering" by pyrite moderated the oxidative depletion of uranium. Where pyrite was scanty or absent, uraninite was completely dissolved by the effects of radiolysis and no ore formed.

#### INTRODUCTION

Probably 75 percent of the world's gold resources and at least 50 percent of its uranium resources are contained in Precambrian pyritic conglomerates  $2.0-3.0 \times 10^9$  yr old (table 1). These conglomerates contain detrital uraninite and pyrite, which are readily oxidized and are unstable in modern oxygenated environments and were deposited in a reducing atmosphere. Gold in the ore conglomerates is crystalline, and in the conventional view it is assumed to have been initially detrital. It is deduced here that, on the contrary, the gold was transported as complexed ions in anoxic solution and was precipitated on long-lasting unconformities on the floors of shallow seas by a slow reaction driven by the radioactive decay of uranium.

TABLE 1. -Ages, in billions ( $10^9$ ) years, of pyritic uranium-gold conglomerates

Locality	Age	Reference
Serra de Jacobina, Brazil	>2	Cordani and others (1973).
Moeda conglomerate, Brazil	>2	Do.
Blind River-Elliot Lake, Canada	2.3-2.5	Roscoe (1969, 1973).
Kriivoi Rog Series, Ukraine	~2.5	Chukhrov and others (1970).
Kursk Magnetic Anomaly Series, Ukraine	~2.5	Do.
Witwatersrand, South Africa	<2.3-2.8	Nierkerk and Burger (1964, 1969).
Pongola Basin, South Africa	<2.85-3.35	Beukes (1973).

I wish to thank the South African mining houses for their many courtesies and their unfailing hospitality during my stay there. Especial thanks are due to Dr. E. S. A. Antrobus of Anglo American Corp. and Dr. D. A. Pretorius, Witwatersrand University, who made many arrangements for me. Dr. W. E. L. Minter kindly guided me on numerous mine visits. The staffs of the Denison and New Quirke Mines were very helpful during my visit to the Elliot Lake district, Ontario. I am indebted to D. A. Singer and J. DeYoung of the U.S. Geological Survey for their help in statistical problems. This paper has benefited greatly from reviews by colleagues Warren Hamilton and Paul B. Barton, Jr.

#### GEOLOGIC SETTING OF THE WITWATERSRAND REEFS

The Rand pyritic conglomerates (Witwatersrand, South Africa) support the world's largest gold production and have been extensively studied. Six or so major productive conglomerates and about twice as many minor ones are scattered through a cratonic section 9 km thick (Dominion Reef, Witwatersrand, and Ventersdorp Lava Systems; Haughton, 1969; Pretorius, 1964).

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The sedimentary rocks are mostly feldspathic quartzite (Fuller, 1958), typically cross bedded; they also include much iron-formation (Beukes, 1973) and barren conglomerate but virtually no carbonate rocks; and they lie on a dominantly granitic Archaean basement. The section is overlain unconformably by the stromatolitic Dolomite Series of the Transvaal System, whose basal carbonaceous beds contain the youngest well-documented pyritic reef (Haughton, 1969). The surmounting shaly Pretoria Series of the Transvaal System ( $2.0 \times 10^9$  yr) contains hematite-chamosite oolites (James, 1966), in contrast to the more reduced iron-formation of the underlying sequences. The massive release of oxygen to the previously anoxygenic atmosphere by algal photosynthesis, such as that which here deposited 1,200 m of Dolomite Series carbonate, may have raised the ambient oxygen pressure to levels generally beyond that permitting development of reduced iron-formation (French, 1973; Cloud, 1973; Garrels and others, 1973) or preservation of detrital pyrite.

Rand and other pyritic conglomerates consist of well-rounded resistate clasts, mostly quartz and chert (Roscoe, 1969; Liebenberg, 1955). In the Rand the productive conglomerates, referred to by the mining term "reefs," lie on unconformites (Haughton, 1969) within the sequences and, unlike the barren conglomerates that lie conformably with the sections, are characteristically thinner than 1 m and may be only single layers of pebbles (Pretorius, 1974). The clasts are typically small. The average pebble diameter of the reefs that have yielded the greater part of the Rand gold is between 1 and 5 cm (Pretorius, 1964; Antrobus and Whiteside, 1964; de Kock, 1964; Winter, 1964a, b; McKinney and others, 1964).

Sericitic alteration characterizes weathering zones on the eroded Archaean floors beneath the pyritic conglomerate sequences of the Rand, the Pongola Basin (Mathews and Scharrer, 1968), Blind River-Elliot Lake (Roscoe, 1969), the Krivoi Rog (Gershoyg and Kaplum, 1970), and probably the Serra de Jacobina (Leo and others, 1964; Cox, 1967). At both Blind River-Elliot Lake and the Krivoi Rog localities the sericitization of greenstone indicates the marked addition of potassium. A similar addition during modern seafloor alteration of spreading-ridge basalts to potassic mixed-layer clay (Hart, 1973) implies that the ancient pyritic conglomerate sequences also were marine. A marine source is also implied for the authigenic gibbsite, kaolinite, potassic mixed-layer clay and (or) chlorite (Helgeson and Mackenzie, 1970) needed as the substrates of recrystallized phyllosilicates in the reef conglomerates. The cleanly washed, resistate, heavy-mineral-rich matrix could not have contained detrital clay; feldspar or mica are scarcely more likely. Thus reef phyllosilicates were

likely authigenic, growing in voids between siliceous clasts.

The Witwatersrand System underwent low-grade burial metamorphism. Diagenetic reconstitution of the feldspar in the productive upper section is much more complete than that in the nearly barren lower section (Fuller, 1958; de Jager, 1964), and therefore some investigators have interpreted it as the result of magmatism-related hydrothermal alteration. Detrital feldspar in the lower Witwatersrand has been only partly sericitized, whereas that in the upper part has been virtually completely changed to muscovite, pyrophyllite, and chlorite (Fuller, 1958). The same phyllosilicates have also recrystallized in the reefs from the supposed authigenic minerals described. Chloritoid has developed in the upper rocks of appropriate composition, and, exceptionally, kyanite (Liebenberg, 1955) has recrystallized from pyrophyllite and more aluminous minerals.

#### DETTRITAL MINERALS UNSTABLE IN AN OXYGEN ATMOSPHERE

Pyrite is overwhelmingly the most abundant opaque mineral in most of the Rand reefs (Saager, 1970) and, though widely variable in amount, might average 3 wt percent (weight percent) of the mined gold ore (Pretorius, 1964). Part of the pyrite is clearly detrital from the very old crustal rocks and locally has a  $^{207}\text{Pb}/^{206}\text{Pb}$  age of  $3.36 \times 10^9$  yr (Koppel and Saager, 1974). Pyrite is present as single crystals and granular aggregates, replacements of various iron oxides, and porous concretionary nodules. All of these characteristically occur as water-worn remnants (Ramdohr, 1958; Saager, 1970; Liebenberg, 1960) (fig. 1). The rounded clasts of pyrite are in general hydraulic equivalence with undisputed clasts of other minerals of various densities (Pretorius, 1974; Coetzee, 1965).

Crystalline uraninite, in closely sized and generally well-rounded clastic grains (median grain size: avg, 0.103 mm; range, 0.083–0.136 mm) hydraulically equivalent with detrital grains of chromite, zircon, and quartz, occurs sparingly in the matrix of all Witwatersrand reefs (Liebenberg, 1955, 1960; Ramdohr, 1958; Coetzee, 1965). The lead-isotope age of the uraninite is about  $3.0 \times 10^9$  yr (Burger and others, 1962), older than the lead-isotope age of  $2.8 \times 10^9$  yr (Nierkerk and Burger, 1969) of the oldest reef series, further indicating a detrital origin. Electron microscope studies by Grandstaff (this volume, chap. J) and by Feather (this volume, chap. Q) show widely varying thorium contents for different grains in single samples, conclusively proving detrital derivation from multiple sources.

Primary inclusions of zircon in uraninite (Hiemstra, 1968a) and the relatively high thorium contents of the

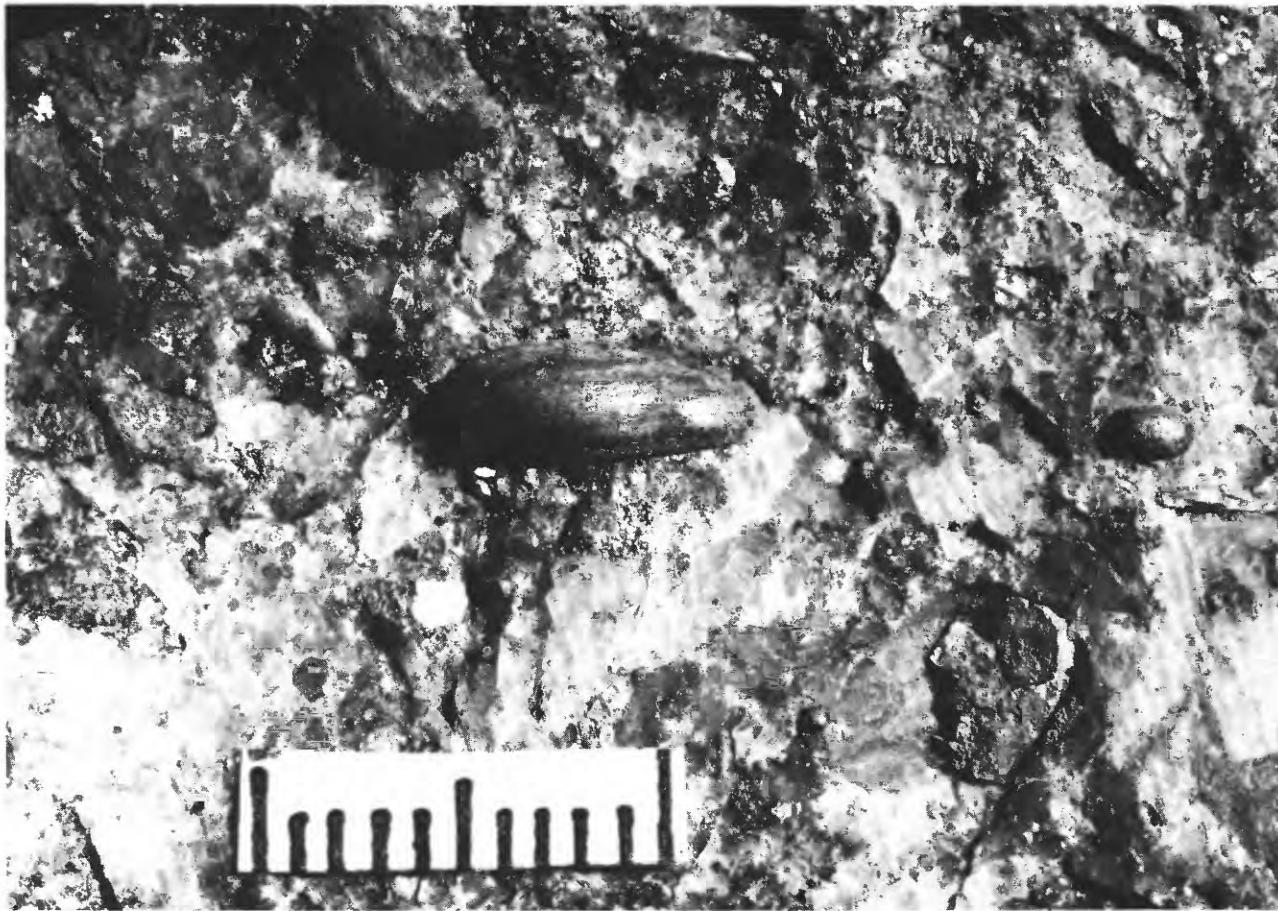


FIGURE 1. - Pyrite pebbles from the Basal Reef, Western Holdings Mine, Orange Free State. 1-cm scale ( $\times 5.4$ ). The shape of the large pebble is controlled by layering in the pyrite; freshly mined pebbles are bright, untarnished. Pyrite occurs as rounded sand grains or pebbles in all reefs.

uraninite show derivation from a high-temperature granitic-pegmatitic source. A modern equivalent of the source rock may be certain Phanerozoic French granites where uraninite occurs as tiny accessory crystals within rock mineral grains (Barbier, 1972).

The lack of Rand uraninite clasts larger than 0.4 mm (Liebenberg, 1955) suggests such a source for the reef  $UO_2$ .

The reefs of the Blind River-Elliot Lake deposits (Roscoe, 1969) and the Rand (Liebenberg, 1955) contain impoverished suites of the detrital minerals expectable from the underlying basements. The surviving minerals are those chemically and physically resistant; notable is the ultrastable mineral zircon. Waterworn minerals of the Rand reefs have high Vickers microhardness (Young and Millman, 1964; McLeod and Chamberlain, 1969); except for a few diamonds and rare corundum (Liebenberg, 1960; Raal, 1969), the hardest is pyrite.

Eh-pH diagrams of the stability fields of pyrite and uraninite in an aqueous system (fig. 2) show that neither

of these minerals is stable in the present oxygenated atmosphere. This conclusion is upheld by the rarity of pyrite and the virtual absence of uraninite as detrital minerals in modern sediments. The diagrams indicate that in an anoxic environment abundant pyrite would tend to protect uraninite in the presence of weak oxidizing tendencies by "buffering" Eh.

Bezgubov and others (1970) found that there are two types of Precambrian uraniferous quartz-pebble conglomerates: (1) pyritic conglomerates containing uraninite and (2) magnetic or martitic conglomerates containing thorite, monazite, and zircon. The conglomerates of the first type contain Eh-sensitive minerals, in contrast to those of the second type, and are confined to the older Precambrian. The lack of Eh-sensitive uraninite from the second type, pyrite-free conglomerate, also found in older Precambrian deposits, suggests that the pyrite-uraninite association is not simply that of two Eh-sensitive minerals unstable in an oxygenated atmosphere.

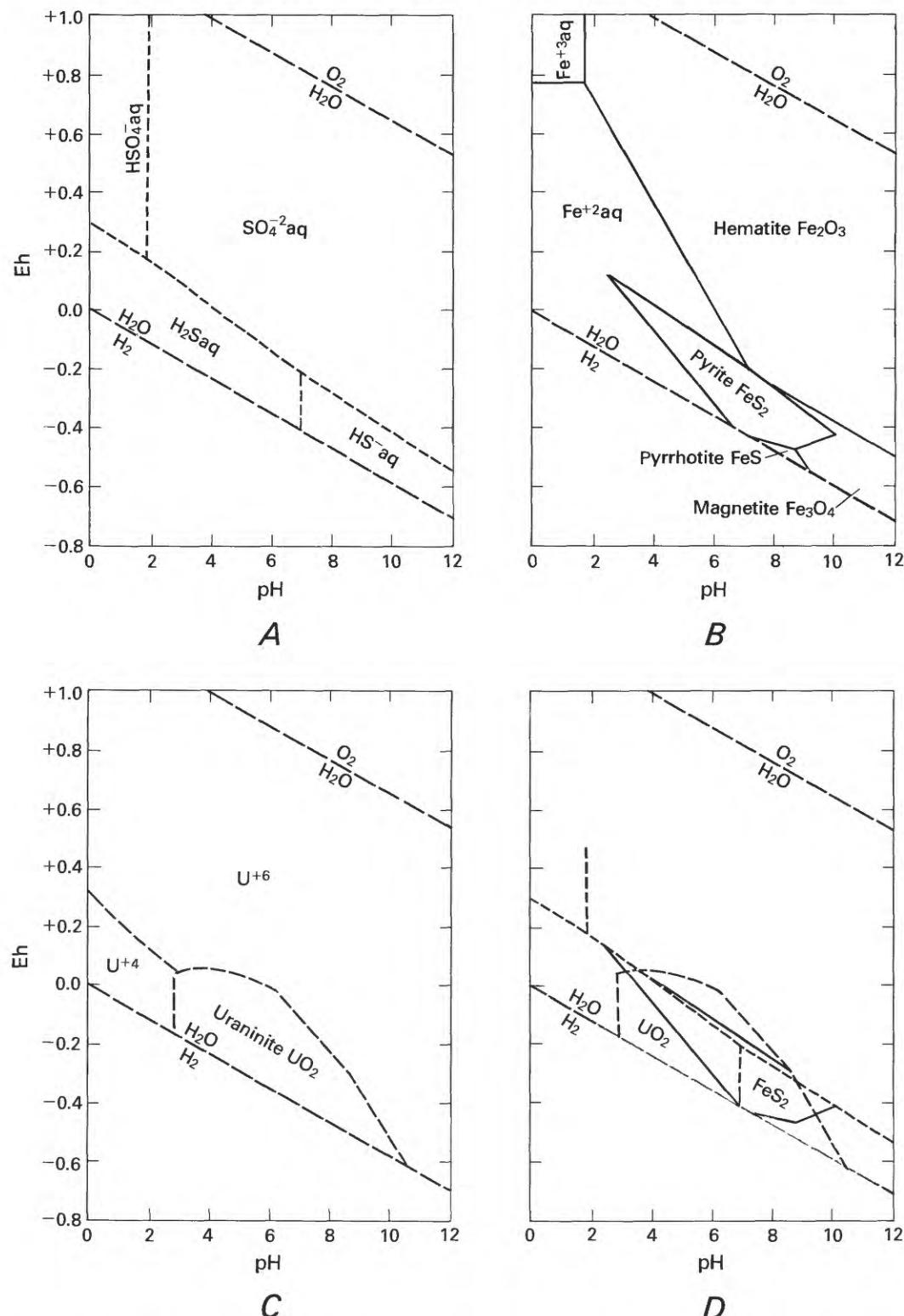


FIGURE 2. - Eh-pH diagrams of the stability fields of pyrite and uraninite and the equilibrium distribution of soluble sulfur species in water at 25°C and one atmosphere total pressure. *A*, Equilibrium distribution of soluble sulfur species; the ratios of the sulfur species are unity at the field boundaries. *B*, Stability relations of iron oxides and sulfides at an activity of dissolved sulfur of  $10^{-5}$ . *C*, Stability relations of uraninite and uranium ions with  $PCO_2 = 10^{-3.4}$  atm, the partial pressure in the modern atmosphere. *D*, Combined diagram showing stability fields of pyrite and uraninite in relation to the triple point,  $(SO_4^{-2} \text{ aq}) / (H_2S \text{ aq} | HS^{-} \text{ aq})$ . Boundaries of solids, at activities of  $10^{-6}$  of dissolved iron and uranium species. Adapted from Garrels and Christ (1965) and Hostetler and Garrels (1962).

## GOLD

The Witwatersrand gold is crystallized in irregular particles normally from 0.002 to 0.5 mm, exceptionally as much as 1-2 mm, in length (Liebenberg, 1955; Hallbauer and Joughin, 1973). Nuggets are lacking, and, save for rare late veinlets, the quartz and chert pebbles are barren (Pretorius, 1974; Liebenberg, 1955). Though the vast majority of particle shapes are not recognizably detrital, a placer origin has been widely accepted because the gold is syngenetic (Antrobus and Whiteside, 1964) and because the bulk of the reef components are generally accepted as waterworn.

### PLACER ORIGIN UNLIKELY

But the ancient conglomerates simply are not comparable to known gold placers (Joralemon, 1952). Summarizing the Rand reefs, Pretorius (1974, p. 40) stated:

The average size of the gold grains is 0.035 mm; of the uraninite grains, 0.065 mm; and of the platinoid grains, 0.055 mm. All these figures illustrate the extremely small size of the minerals of economic importance in the reefs. No medium- to large-sized nuggets have been found during mining operations, and it can be concluded that, in regard to gold-particle size, the detrital deposits of the Witwatersrand are not comparable with modern-day placers.

The economy with which gold is won from gravels, the unfamiliar units in which it is reported, and the romance surrounding rich placer pockets combine to obscure the evident fact that the Witwatersrand reefs are richer<sup>2</sup> and far more extensive than any modern gold placers. Even the notoriously gold-poor reefs of the Blind River-Elliot Lake district in Canada contain twice the gold content yielded by average gold-dredge gravels (see following paragraphs). This paradox underscores the lack of quantitative analysis in the easy acceptance of a placer origin for the gold of pyritic conglomerates.

As part of the Heavy Metals program of the U.S. Geological Survey, modern studies have been conducted on the Tertiary gravels of the Sierra Nevada (Yeend, 1974), stream placers in Oregon (Boggs and Baldwin, 1970), and the beach placers at Nome, Alaska (Nelson and Hopkins, 1972). This work has reestablished the validity of the lessons of prior workers (Lindgren, 1911; Janin, 1918). Placers do not depend fundamentally on the functioning of some special winnowing process but on a nearby source of coarse gold. The United Nations Economic Commission for Asia and the Far East has counselled that marine gold placers are inherently unlikely because of the proximity of gold placers to source (Emery and Noakes, 1968).

Economic gold placers result from the near-incapacity of streams and currents to transport coarse gold (Tuck, 1968; Shilo and Shumilov, 1970). Such placers as

remotely approach the richness of representative Rand reef (7-21 ppm (Pretorius, 1974)) occur only in narrow paystreaks (Lindgren, 1911), which are generally within a few kilometers, at most, say, 15 km, of a coarse-gold source (Nelson and Hopkins, 1972; Emery and Noakes, 1968). With few exceptions, rich gravel is bouldery, poorly sorted, and compositionally immature (Yeend, 1974) and contains sporadic nuggets. Grain size and grade rapidly decrease downstream (Boggs and Baldwin, 1970), as area of the deposits increases, just as the yield of gold decreases exponentially away from the upper end of a dredge table (Janin, 1918, p. 182-184). But even the most extensive of modern gold placers, the dredge fields, are small (5-50 km (Lindgren, 1911)) compared with the reefs (as much as 400 km<sup>2</sup> (Antrobus and Whiteside, 1964)), whereas dredge-field average grade (0.10-0.13 ppm (Lindgren, 1911; Janin, 1918)), though not directly comparable, is but one percent of that of the reefs! In contrast to modern placers, the Main Reef Leader of the East Rand has been mined throughout a downcurrent distance of 30 km (Pretorius, 1974), with the gold content and the pebble diameter of the mature resistate assemblage paralleling a slow decrease in size of detrital zircons (1.4 percent/km (Viljoen, 1968)).

According to Pretorius (1974), 55 percent of the world's total gold won throughout recorded history has been mined from the Witwatersrand. If the Rand gold were detrital, an unequaled concentration of giant parent deposits, now vanished, must have closely rimmed this local basin.

But what if such unequaled concentration of deposits had rimmed the basin? Judging from modern gold placers, it would not have been possible to transport gold clasts in running water, in whatever amount imaginable, so as to conform to the mined distribution of the Main Reef Leader. For, regardless of the amount of gold available, the evidence points to a steeper grade and grain-size gradient in the down-current direction than is developed in the East Rand Basin.

Figure 3 shows that the grain size of the predicted gold particles of the average Rand reef lies well below the lower limit of those in Siberian placers. In the Siberian placers the minimum gold particles have a constant weight (~1.5 mg), regardless of the degree of flattening. So do those of the U.S., dominantly Alaskan, placers (~1 mg) described by Tuck (1968). This indicates that resistance to transport, rather than some winnowing effect, is the prime factor in the formation of gold placers.

If the gold was carried in complexed solution, its broad dispersion in the Rand reefs becomes understandable. The equally broad dispersion of detrital uraninite ( $d \sim 9$ ) is, to begin with, much less a problem than for gold ( $d \sim 17.5$ ). Beyond that, the uniformly small size (average

<sup>2</sup> [In 1976 the average gold recovery from Rand ores was 0.2686 troy ounces per short ton (9.21 grams per metric ton) milled (Mining Survey, 1977).]

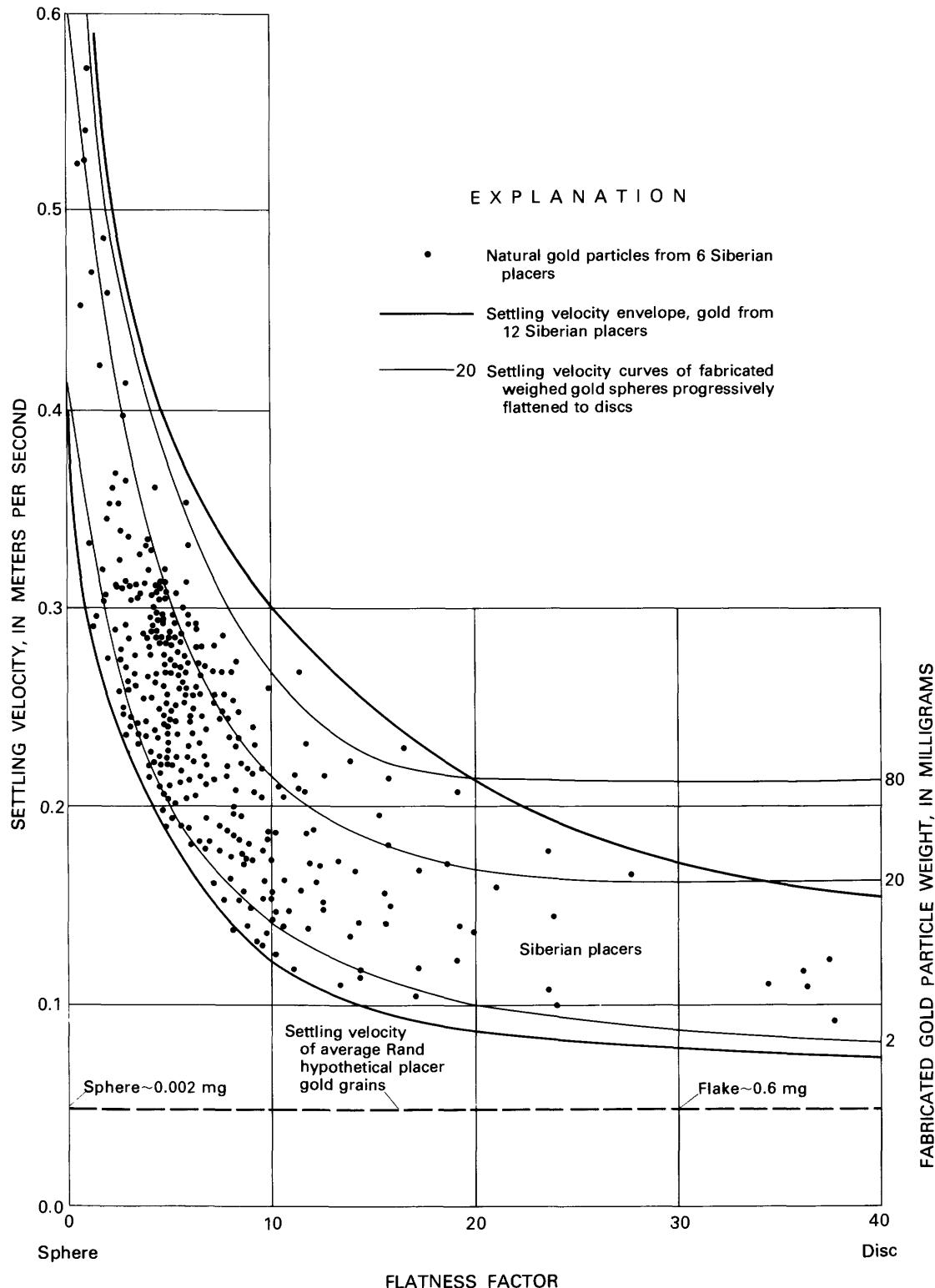
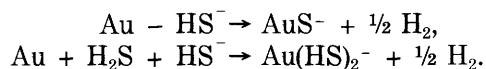


FIGURE 3.—Schematic comparison of settling velocities of natural gold particles from Siberian placers with similar fabricated gold particles and average Rand hypothetical-placer gold grains. The settling velocity of the Rand hypothetical-placer gold was determined from the average size of undoubtedly clastic mineral grains in the reefs by experimental settling velocities from Gibbs and others (1971). The Rand clast sizes were averaged from data in Liebenberg (1955) and Coetze (1965). Graph modified from Shilo and Shumilov (1970, figs. 1, 2, table 1). Flatness factor determined from equation  $((\text{length} + \text{width})/(2 \text{ thicknesses})) - 1$ .

median grain size, 0.103 mm) of the uraninite from granitic sources suggest that it may have been largely transported to the reef site in sand-sized clasts of granitic rock minerals. Hydrogen-ion metasomatism (discussed in the section, Bacterial Sulfur Cycle) of the feldspathic debris would have freed the tiny uraninite crystals at the reef site and simultaneously obliterated any evidence of this means of transport. In a similar fashion the rare grains of detrital platinoids ( $d \sim 17.5$ ) found in the reefs (see section, Orange Free State Goldfield) were probably transported in sand-sized fragments of serpentinized ultramafic rock.

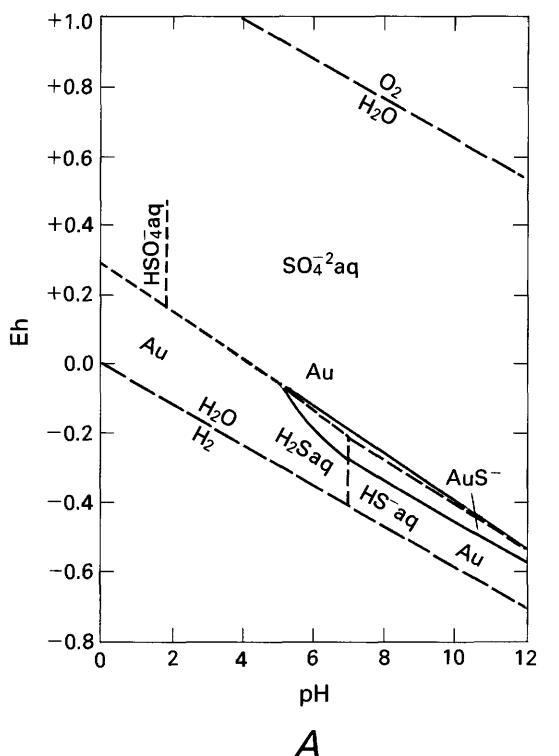
#### TRANSPORT IN COMPLEXED SOLUTION

The Rand gold is believed to have been transported in complexed solution (Myers, 1971). It is thought to have been leached anoxygenically from ordinary crustal rocks, most likely from sea-floor basalt. The aurous thiocomplexes (Krauskopf, 1951; Seward, 1973) are obvious gold solvents in an aqueous pyritic environment at room temperature. The reactions for two of the more likely complexes are:

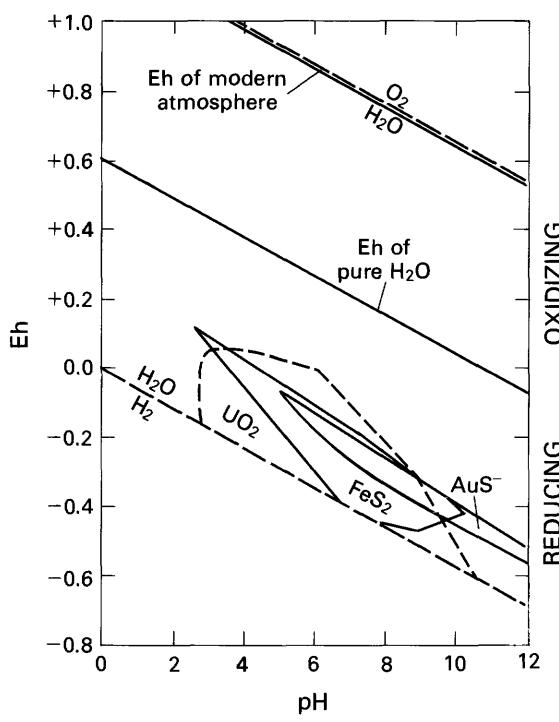


In figure 4, Eh-pH diagrams of the stability field of the  $\text{AuS}^-$  complex are compared with others of interest. Other thiocomplexes have similar stability fields. At near-neutral pH the narrow stability fields of the complexes lie at the top of the pyrite field, straddling the dissolved sulfide-dissolved sulfate join. Figures 2 and 4 show that the association of pyrite, detrital uraninite, and aurous thiocomplex constitutes a system that may explain the Rand reefs. Pyrite and uraninite were transported as clasts and gold moved in complexed solution. The gold complexes would have been stable either in a system held generally at the upper limit of the pyrite field by oxidizing tendencies or in a system dominated by the bacterial sulfur cycle. Gold would have been deposited with Eh shifts in either direction; it is likely that an oxidizing reaction originally precipitated the gold of the reefs.

FIGURE 4.—Eh-pH diagrams showing the  $\text{AuS}^-$  solubility window in the aqueous Au-S system and its relation to the stability fields of pyrite and uraninite. A, Stability relations of gold and aurous sulfide complex in water at 25°C and one atmosphere total pressure at an activity of dissolved sulfur of  $10^{-5}$  and the controlling equilibrium distribution of soluble sulfur species (see fig. 2A). Boundary of field of  $\text{AuS}^-$  complex at an activity of  $10^{-8}$  dissolved gold. B, Stability fields of aurous sulfide complex, pyrite (see fig. 2B), and uraninite (see fig. 2C). Adapted from Garrels and Christ (1965) and Hostetler and Garrels (1962).



A



B

### BACTERIAL SULFUR CYCLE

The writer has proposed (Myers, 1970, 1971) that an anaerobic bacterial sulfur cycle existed in the Rand reef environment. Colored sulfur bacteria, primitive (Margulis, 1969) photosynthetic sulfide-oxidizing prokaryotes now symbiotic with sulfate-reducing bacteria in restricted niches (Kondrat'eva, 1965; Takahasi and Ichimura, 1970; Trüper and Genovese, 1968), would have flourished widely in the photic zone in an oxygen-free system (fig. 5). As in figure 5, the postulated ancient bacterial cycle balanced organic matter and  $\text{CO}_2$ , as well as dissolved sulfide and sulfate. At a near-neutral pH, the cycle would have regenerated pyrite (fig. 2A, B, D), generally keeping the concentration of soluble sulfur species low ( $10^{-5}$  m) where isotopic fractionation is slight (Harrison and Thode, 1958; Perry and others, 1971; Hoefs and others, 1968). The formation of new biogenically mediated pyrite was dependent on the relatively high activity of dissolved  $\text{Fe}^{+2}$  adjacent to original detrital pyrite and other iron minerals concentrated on the reef surface in the infralittoral zone.

The bacterial sulfur cycle must have produced significant amounts of  $\text{H}_2\text{SO}_4$ , which would have reacted immediately with the feldspathic debris in the sedimentary environment to produce authigenic clays and (or) chlorite (Helgeson and Mackenzie, 1970). Buffering by the original silicates would have kept the pH near neutral. The complete conversion of this debris in the upper productive part of the Witwatersrand system (see the section, Geologic Setting of the Witwatersrand

### SUNLIGHT

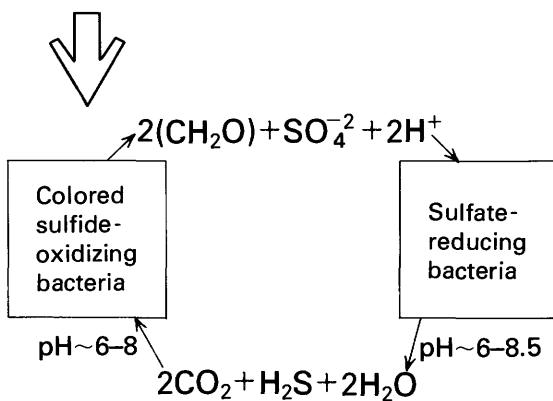


FIGURE 5.—Modern anaerobic bacterial sulfur cycle. The cycle occurs in stagnant ponds on the underside of algal mats and at shallow depth in certain lakes under anaerobic conditions. Bacteriochlorophyll of the colored sulfur bacteria, with an absorption spectrum utilizing light energy not absorbed by algae, photosynthesizes organic matter from  $\text{CO}_2$ . Sulfate-reducing bacteria consume organic matter with oxygen extracted from sulfate.

Reefs) is believed dependent on bacterially mediated hydrogen-ion metasomatism during deposition.

Oolitic textures in Rand concretionary pyrite have been interpreted as biogenic (Schweigart and Rahden, 1965); indeed, much smaller microstructures in porous and concretionary reef pyrite have been considered to represent mineralized bacteria (Ramdohr, 1958; Saager, 1970; Schidlowski, 1966). Concretionary pyrite also contains significant nickel and cobalt (Saager and Esselaar, 1969). Pyrite concentrates from the Vaal Reef locally contain 1-3 percent nickel, the bulk of it in the crystal lattice (F. H. S. Vermaas, unpublished data). Authigenetic cobalt, copper, and nickel ore minerals are minor reef accompaniments. The assemblage suggests that the ancient sulfide nodules were analogues of the porous, bacterially mediated (Ehrlich, 1971) manganese oxide nodules of the present seafloor, which are potential nickel-copper-cobalt ores (Cronan and Tooms, 1969; Tinsley, 1975).

Like the modern oxide nodules, the ancient pyrite concretions may also have contained significant amounts of silver. Though available data are not conclusive, the weight of the evidence suggests that much of the silver of the reefs was once contained in the virtually ubiquitous pyrite.

### THUCHOLITE

Uraniferous thucholite, a jet-black, brittle asphaltite, is closely associated with detrital uraninite in the ore conglomerates and must be related genetically to it. Perhaps one-fourth of the total uranium in the Rand reefs is disseminated in this oxygenated bitumen, which has variably replaced uraninite. Thucholite envelops detrital uraninite grains and wedges them apart along fractures. The asphaltite progressively separates uraninite shards and digests them so that little remains of the original grains (Liebenberg, 1955; Ramdohr, 1958; Schidlowski, 1969). Much of the Rand thucholite occurs as tiny granules, "fly-speck carbon"; at many localities granules grade by coalescence into bedding-plane sheets of thucholite that vary from a fraction of a millimeter to several centimeters in thickness (de Kock, 1964).

Thucholite also replaces uraninite in Canadian Precambrian pegmatites (Ellsworth, 1928) and in the Precambrian high-temperature complex-metal ore deposit of Boliden, Sweden (Grip and Ödman, 1944). In these settings and in the Rand, the mineraloid is thought to have been formed by polymerization of fluid hydrocarbons by uraninite radiation (Liebenberg, 1955; Schidlowski, 1969). Experimental radiation of any type of *n*-paraffins of intermediate chain length has three products (Charlsby, 1954): (1) long-chain infusible molecules with lowered H/C ratios (graphite is formed

with very high radiation dosages), (2) gaseous short-chain paraffins with increased H/C ratios, particularly methane, (3) free hydrogen. All three products are present in the Rand goldfields. Methane is present in many of the mines, and gases containing as much as 77.5 percent H<sub>2</sub> have been reported from several mines (Liebenberg, 1955). Thucholite locally contains graphite (Schidlowski, 1967).

Hydrocarbons could be formed by alpha-particle bombardment of the fatty acids present in organic sediments (Liebenberg, 1955; Whitehead, 1954). Yield estimates by Whitehead (1954) suggest the uranium here believed originally present in the Rand reefs was adequate to produce the thucholite substrate-hydrocarbons from bacterial organic matter.

The hypothesis that sheets of cross-fibered thucholite represent Precambrian algal coal has been disproved by Schidlowski (1969). The lenticles, which some investigators thought to be compaction-flattened algal bodies, are in fact oblique sections of cross-fibers in thucholite sheets. The <sup>13</sup>C values in thucholite are typical of petroleum, not of coal (Schidlowski, 1969; Eichmann and Schidlowski, 1974). A petroleum origin is further indicated by the systematic relation between H/C ratios and total radiation dosage in a series of uranium-bearing petroleum asphaltites and thucholites, two of the latter from the Witwatersrand (Pierce and others, 1958).

Hallbauer (1975; this volume, chap. M) has proposed that the thucholite represents the remains of primitive lichen. The characteristic cross-fibered habit of the asphaltite is taken by him to represent fungal hyphae; he interprets the "fly-speck carbon" granules as fruiting bodies. After ashing the thucholite at 500°C, he finds microscopic filamentous structures, which he believes are of plant origin. It is hard to see why the fragile fungal structures he proposes would not have been flattened to carbon films by rock compression. It is even more difficult to imagine why the delicate microscopic filamentous structures would be preserved after undergoing not only strong compression but also very high radiation dosages from the ubiquitous detrital uraninite and why these structures are seen only in the severely oxidized material. Hallbauer has acknowledged (oral commun., Oct. 14, 1975) the possibility that the thucholite might have been converted to coke in the early stages of the ashing treatment at 500°C. It seems likely that the filaments are coke structures whose orientation was controlled by the original fibrous texture of the thucholite and which were preserved during the subsequent oxidation. According to this interpretation, the gold filaments figured by Hallbauer (1975) were drawn out by deformation of heat-softened gold at 500°C.

A modified paragenesis of thucholite is sketched here. It is based on the thesis that a soluble organic substrate

of thucholite was everywhere present in the waters of the Rand reefs. It is believed to have been converted by stepwise reactions directly to protothucholite on the site by a combination of ionizing radiation and the presence of dissolved uranium adjacent to detrital uraninite. (The oilfield uraniferous asphaltites of the Texas Panhandle formed from the abstraction of dissolved uranium by petroleum, according to Pierce and others (1964), and contain no particulate uraninite larger than 2  $\mu\text{m}$ .) It is maintained here that the apparent "digestion" of detrital uraninite by thucholite is illusory. The detrital uraninite was instead dissolved along minute flaws and fractures by an aqueous oxidation (see the section, Genesis of Gold in Reefs, Oxidizing Reactions) – the thucholite resulted from solution of the uraninite, but did not cause it.

It seems likely that, for the most part, uraninite grains were thucholitized while immobilized in the matrix of the reef. Coatings of protothucholite were probably rubbed off most of the active grains of uraninite in intermittent motion on the reef surface. Those uraninite grains that escaped thucholitization after being finally immobilized in the reef matrix are thought to have been sealed off from effective contact with water and dissolved organic matter by the development of authigenic clays and (or) chlorite (see the section, Bacterial Sulfur Cycle) and by the deposition of silica within the reef (Liebeberg, 1955, p. 191). The abundant uraninite of the Dominion Reef has not been thucholitized because its high thorium content (Grandstaff, this volume, chap. J; Feather, this volume, chap. Q) renders it resistant to oxidation and solution. Elsewhere in the Rand, high-grade concentrations of uraninite are invariably thucholitized (Liebenberg, 1955). That such areas were nowhere sealed off by clay minerals or silica is probably owing to a side effect of the strong radioactivity (see the section, Genesis of Gold in Reefs, Oxidizing Reactions).

## INVERSE RELATION OF URANIUM AND GOLD

This investigation has recognized that gold and uranium vary inversely in the pyritic conglomerates. The relation is held to be due to the progressive solution of uraninite and precipitation of gold in the reef environment. The uranium-gold ratio is considered to be an index of maturity of individual conglomerates (for another explanation of the varying uranium-gold ratios, see Minter, this volume, chap. K).

Earlier, Winter had hinted broadly at the inverse relation and its meaning in his descriptions of the Orange Free State Goldfield (1964a, p. 436, 442; 1964b, p. 532, 545–546). Very recently, an instance of the inverse relation was described without comment. Of the new Cooke Section of the Randfontein Mine, Mining Magazine wrote (1976, p. 263), "From north to south the gold value generally decreases while the uranium value increases."

Most comprehensively, Cousins, who had access to a considerable body of assay results not available to me, stated (1956, p. 106-108):

Uranium/gold ratios show only minor variations within a block of ore and show regular variations from locality to locality \* \* \* Very wide variations in the ratio have been observed; the highest ratio being 3,000 times as great as that of the lowest. It appears rare, however, for reefs very rich in gold to have very high uranium/gold ratios. Such high ratios are found usually in reefs with low gold values and indicate a more widespread distribution of uranium than gold.

The South African points shown on figure 6 and in table 2 represent averages made up of very large quantities of mined ore or developed reserves. All mines are listed for which production figures for both uranium and gold are available.

TABLE 2.—Uranium and gold contents, in parts per million, and the  $U_3O_8/Au$  ratios of pyritic conglomerates in South Africa, Canada, and Brazil

District, mine, or single reef	$U_3O_8$	Au	$U_3O_8/Au$
1 Blind River-Elliott Lake district, Canada, 34 samples <sup>1</sup> -----	1,777	0.262	6,780
2 Upper Monarch Reef, Zone 2, West Rand Consolidated Mine, <sup>2</sup>	1,331	.700	1,900
3 Blind River-Elliott Lake district, Canada <sup>3</sup> -----	1,400	.857	1,750
4 Monarch Reef, West Rand Consolidated Mine <sup>2</sup> -----	1,250	1.720	727
5 Upper Monarch Reef, Zone 4, West Rand Consolidated Mine, <sup>2</sup>	954	1.320	723
6 Luijpaardsvlei Mine, uranium section, 1956-1964 <sup>4</sup> -----	791	1.330	595
7 West Rand Consolidated Mine, uranium section, 1959-1972, <sup>5</sup> -----	712	1.280	556
8 East Champ d'Or Mine, uranium section, 1959-1964 <sup>4</sup> -----	402.3	1.000	402
9 Randfontein Mine, uranium section, 1959-1964 <sup>4</sup> -----	578.2	2.040	283
10 Randfontein Mine, SD 32 Shaft area <sup>6</sup> -----	900	5.00	180
11 White Reef, West Rand Consolidated Mine <sup>2</sup> -----	627	5.010	125
12 Randfontein Mine, Cooke Section, No. 3 Shaft area <sup>5</sup> -----	450	6.00	75.0
13 Randfontein Mine, Cooke Section, No. 2 Shaft area <sup>5</sup> -----	400	7.00	57.1
14 Vogelstruisbult Mine <sup>4,6</sup> -----	243	8.280	29.3
15 Virginia Mine <sup>4,6</sup> -----	243.5	8.420	28.9
16 Zandpan Mine <sup>4,6</sup> -----	343	13.050	26.3
17 Western Reef Mine <sup>4,6</sup> -----	239	9.210	26.0
18 Daggafontein Mine <sup>4,6</sup> -----	193	8.350	23.1
19 Vaal Reefs Mine <sup>4,6</sup> -----	321	16.100	19.9
20 Hartebeestfontein Mine <sup>4,6</sup> -----	296.5	15.450	19.2
21 Randfontein Mine, Cooke Section, No. 1 Shaft area, <sup>5</sup> -----	200	11.00	18.2
22 Buffelsfontein Mine <sup>4,6</sup> -----	262	14.580	18.0
23 Ellaton Mine <sup>4,6</sup> -----	153	8.600	17.8
24 Harmony Mine <sup>4,6</sup> -----	217.5	12.960	16.8
25 Welkom Mine <sup>4,6</sup> -----	142.5	10.610	13.4
26 Freddie's Consolidated Mine <sup>4,6</sup> -----	132	11.330	11.7
27 President Steyn Mine <sup>4,6</sup> -----	138	12.000	11.5
28 Lorraine Mine <sup>4,6</sup> -----	117	10.720	10.9
29 Canavieras Mine, Serra de Jacobina, Brazil <sup>7</sup> . -----	150	15.430	9.7
30 Doornfontein Mine <sup>4,6</sup> -----	145	15.170	9.6
31 Stilfontein Mine <sup>4,6</sup> -----	135	14.940	9.0
32 Eastern section, Free State Geduld Mine <sup>8</sup> -----	109.5	12.620	8.7
33 Blyvoortuizicht Mine <sup>4,6</sup> -----	168	21.370	7.9
34 West Driefontein Mine <sup>4,6</sup> -----	164.5	28.660	5.7
35 President Brand Mine <sup>4,6</sup> -----	131	23.950	5.5
36 Central section, Free State Geduld Mine <sup>8</sup> -----	67	23.920	2.8
37 Western section, Free State Geduld Mine <sup>8</sup> -----	115	46.870	2.5

<sup>1</sup> Roscoe (1969, Appendix, table C).

<sup>2</sup> London Mining Journal (1972).

<sup>3</sup> Finch and others (1973).

<sup>4</sup> Chamber of Mines of South Africa, 1967-1972, Annual Reports 78-83; Transvaal and Orange Free State Chamber of Mines, 1955-1966, Annual Reports 67-77.

<sup>5</sup> Mining Magazine (1976).

<sup>6</sup> Through 1969.

<sup>7</sup> Bateman (1958).

<sup>8</sup> K. J. Bell (Anglo American Corp. of S. Africa, Ltd, written commun., October 20, 1971); 1971 ore reserves.

The mine-wide averages (open circles on fig. 6) represent all uranium ore milled and all gold ore milled from a single mine. In all such cases the uranium is a coproduct, and the amount of uranium ore milled is less than that of gold ore, ranging from 10 percent to 98 percent of the

tonnage of the primary product. The amount of uranium ore represented by individual points ranges from 1.8 million metric tons to 32.2 million metric tons. Evidently, if the uranium ore makes up but a small percentage of the gold ore, the uranium grade of the plotted point on figure 6 will be higher than the true average would have been for all gold ore. Yet the distribution of such points shows that this factor is not a dominant one in determining whether the mine in question lies above or below the calculated curve.

Most of the mine-wide averages include ore from several reefs. Though the grade and  $U_3O_8/Au$  ratio in such cases will differ from those of the component beds, the composite average is as valid in determining the inverse relation between uranium and gold as an average made up of ore from a single reef.

The points for single reefs and single-reef zones (solid circles) or annual plots of single mine sections (line plots) represent gold and uranium values for a given tonnage of ore. The amounts of ore represented are comparable to those of the mine-wide averages, commonly involving many millions of metric tons. An inspection of figure 6 will show that although the mine-wide averages must be classed as inferior to those from single reefs, reef zones, or mine sections, the distribution of points around the calculated curve does not appear to support this conclusion.

The three points plotted for Canada and Brazil (solid triangles) do not represent formal averages of specific amounts of ore. One of the Blind River-Elliott Lake averages represents the arithmetic mean of all samples (34) for which combined uranium-gold assays were available, regardless of reef or location. The other point is a published estimate of district-wide average grade. The Brazilian point is a published estimate of average grade based on the milling of a few thousand metric tons of ore from a single stope.

## GENESIS OF GOLD IN REEFS

The present thesis holds that the gold content of a reef increases with the duration of the unconformity on which the conglomerate was reworked. When figure 6 is read as a diagram of reef maturity, with the abscissa as a crude scale of age, both absolute and relative amounts of uranium are seen to decrease with age. This accords with descriptions of thucholitization of Rand detrital uraninite and of large uraninite crystals in a Canadian pegmatite (Liebenberg, 1955; Ramdohr, 1958; Schidlowski, 1969)—thucholitization apparently both digested uranium and expelled it from the system.

All gold-bearing Rand reefs contain thucholite, with which a widely variable proportion of the gold is directly associated (fig. 7) (Horwood, 1911; Liebenberg, 1955; Hallbauer and Joughlin, 1973). Abundances of gold and

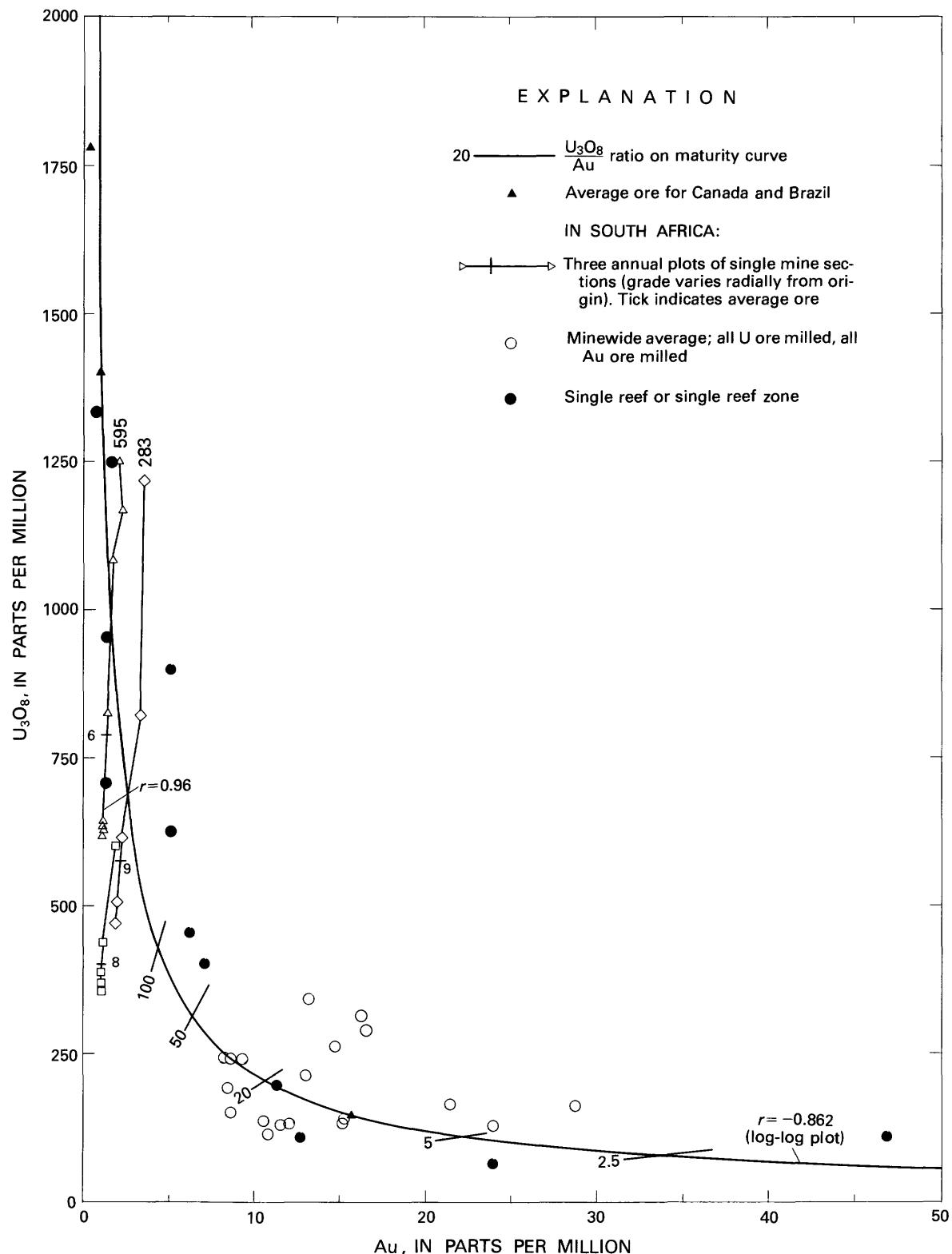


FIGURE 6. – Uranium and gold content of reefs in South Africa, Canada, and Brazil. Data points are listed in decreasing order of  $\text{U}_3\text{O}_8/\text{Au}$  ratio (table 2). Within a single mine, ore grade fluctuations in a reef of nearly constant maturity give rise to the widely recognized sympathetic variations in gold and uranium content (data points 6, 8, and 9). Among different mines on the same reef, or among different reefs an inverse relationship generally exists between uranium and gold. The symbol  $r$  is the linear coefficient of correlation, explained more fully in the section, Genesis of Gold in Reefs, Degrees of Association of Uranium and Gold. The curve is from the best-fit linear regression calculated by the method of least squares on the logarithms of the data points.

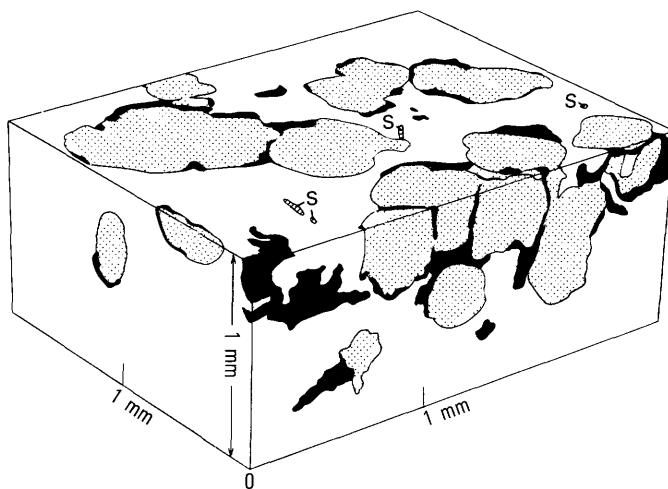
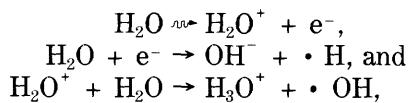


FIGURE 7.—Gold (black) with thucholite (stippled) and very minor sulfide (S), probably pyrite, in quartzite gangue (blank). Gold encases but does not crosscut thucholite granules, indicating that the gold was introduced early, not after the formation of fractures. Horizontal face of the block is parallel to reef, a single lamina on a minor unconformity; South Reef, West Rand. Bar scales 1 mm long.

thucholite tend to vary together (de Kock, 1964), and by far the richest gold ores are thucholite laminae such as the Carbon Leader, only a few millimeters or centimeters thick but containing up to several weight-percent gold (Liebenberg, 1955; de Kock, 1964). The notable association of gold with thucholite is not due to a property of the bituminous material itself; instead, it occurs because thucholite surrounds highly radioactive uraninite whose nuclear decay resulted in the precipitation of gold.

#### RADIOLYSIS OF WATER

The radioactive decay of detrital uraninite accumulating on the Rand reefs caused the radiolysis of water and the formation of free radicals according to the following reactions:



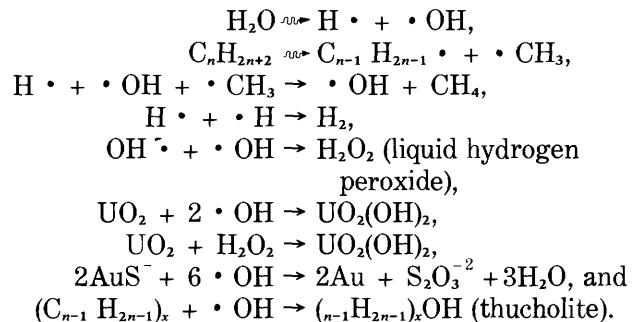
where  $\rightsquigarrow$  indicates ionizing radiation and  $\cdot \text{H}$  and  $\cdot \text{OH}$  denote charge-free free radicals (Dragonić and Dragojić, 1971, p. 10-11).

When water is irradiated with X-rays or  $\gamma$ -rays in a completely water-filled vessel and the ionizing radiation is then removed, it is found that the free radicals recombine and only water remains. Contrastingly, if the same radiolysis takes place in the presence of a large volume held at a vacuum, there is a continuous evolution of free hydrogen (Allen, 1961, p. 24, 75). From this it is clear that under low-pressure conditions where hydrogen can diffuse from the system, a fractionation of the reducing

and oxidizing free radicals will occur. Such a fractionation must have occurred on the shallow unconformities on which the reefs certainly accumulated, whether they were marine, as favored here, lacustrine, or fluvial. A calculation was carried out to determine whether the fractionation would have been effective. A hydrogen pressure in the ancient atmosphere of  $10^{-4}$  atm and a reef 0.9 m thick with a uraninite content of 2,000 ppm (parts per million)  $\text{U}_3\text{O}_8$  were assumed. The diffusion system would have kept the hydrogen pressure of the reef at  $10^{-3}$  atm with a reef depth of 20 m. As the reefs probably accumulated at depths considerably less than 20 m, the effective reef pressure would have been even less. Thus, the calculation shows that diffusion of hydrogen would have been an effective fractionating mechanism.

#### OXIDIZING REACTIONS

Radiolysis fractionation would have resulted in an excess of hydroxyl free radicals in the reef environment. In the presence of detrital uraninite, pyrite, aurous sulfide complex, and hydrocarbons, hydroxyl free radicals would have given rise to the following oxidizing reactions:



These oxidizing reactions would have dissolved uraninite, precipitated gold, and polymerized oxygenated thucholite from hydrocarbons.

Although hydrocarbons polymerized anhydrously by nuclear reactors (Charlsby, 1954) grossly simulate thucholite, the natural mineraloid is oxygenated (Ellsworth, 1928; Liebenberg, 1955; Pierce and others, 1958). Increases in the oxygenated asphaltite fractions of crude oils have been recorded during experimental aqueous irradiations that freed hydrogen and methane (Colombo and others, 1964). Part of the gas was generated by anhydrous degradation reactions (Charlsby, 1954) within oil globules in an oil-water emulsion, but part was produced within the same test vial by reactions involving water (Colombo and others, 1964). Water served as hydrocarbon oxidant because the bubble of reducing gases in the test vial did not equilibrate with oxidizing hydroxyl radicals in the subjacent hydrous mixture.

Similar oxidizing reactions must have taken place during the formation of natural uraniferous petroleum asphaltites investigated by Pierce and others (1958, table 1). Plotting of their data on 23 asphaltites and one Rand thucholite shows a direct relation between the uranium content and oxygen content. This correlation is best displayed as an inverse relation between uranium and the C/O ratio (fig. 8.).

Such reactions would have been very slow because the radioactive decay of natural uranium is itself a very slow process. But the productive Rand reefs range in age from  $2.8$  to  $2.3 \times 10^9$  yr, an interval nearly as long as the Phanerozoic eon. In the Witwatersrand Basin this interval of 500 million years was one of uncommon crustal stability. The unconformities on which the reefs occur seem likely to have endured for several million years. Thus secular reactions that would have been grossly incompetent in many environments may plausibly be evoked to explain the relation shown by figure 6.

Locally, these slow reactions appear to have been more brisk. High-grade concentrations of uraninite in the Rand are almost invariably thucholitized (see the section, Thucholite). Lower-grade accumulations commonly contain unthucholitized grains, implying that these areas were sealed off by authigenic silica or clay minerals from chemical interaction. It seems likely that the high-grade concentrations gave off enough hydrogen that they were kept permeable to aqueous solutions.

#### MATURATION OF REEFS

Several processes working concurrently during the formation of the reefs control the inverse relation between uranium and gold:

1. Chemical reactions driven by the nuclear decay of uranium:

a. A rate of uranium solution that depended, via the uraninite-driven radiolysis of water, on the amount of uranium present, and hence decreased exponentially with time from the onset of thucholitization to the time of effective burial. (Radioactive decay is a first-order reaction—that is, the number of atoms that decompose in unit time will be proportional to the number present.)

b. A linear precipitation of gold, the rate determined at first by the concentration of the scarce aurous complex, later by the diminished availability of the precipitant, which in turn was controlled by the concentration of uraninite.

2. Concentration due to reworking:

a. The gold content increased exponentially with time because of concentration into an ever-thinning reef.

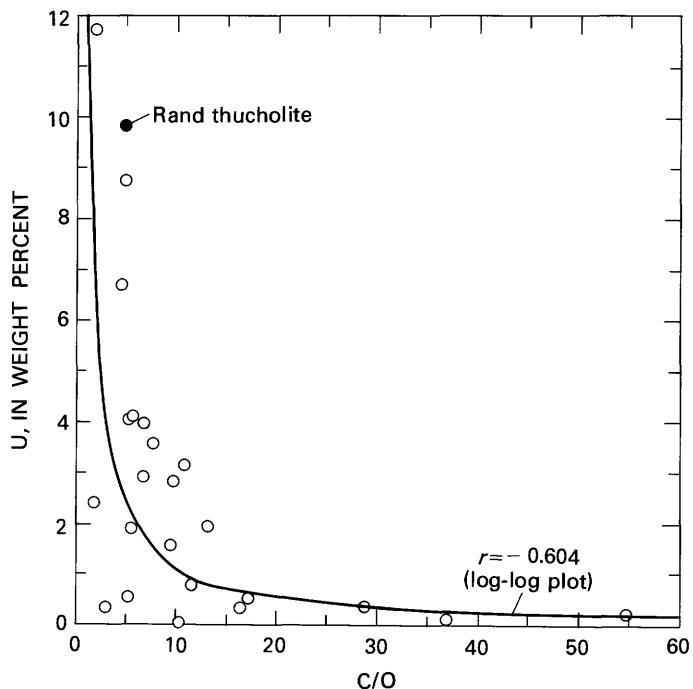


FIGURE 8.—Uranium content and C/O ratio for 23 uraniferous petroleum asphaltites and a Rand thucholite. Data from Pierce and others (1958, table 1) recalculated to an ash and sulfur-free basis. The curve is from the best-fit linear regression calculated by the method of least squares on the logarithms of the data points.

b. The rate of the chemically controlled exponential decrease in uranium was slowed by the opposed process of concentration.

Together, these actions define a maturing process. A uraninite placer, accumulating on an unconformity, became progressively converted to a gold reef with little residual uraninite. Immature uraninite-rich reefs tend to be relatively thick conglomerates with dispersed metal content (Roscoe, 1969; Pretorius, 1964). The most mature reefs tend to grade toward the thucholite-seam type, very thin but exceedingly rich in gold (Liebenberg, 1955; de Kock, 1964). A combination of chemical attack and physical reworking accounts for the general thinness of mature reefs and for the detrital implication of such features as cylindrically rolled gold particles encased in pyrite (de Kock, 1964, p. 368). A maturing reef at any given moment is the sum of the chemical processes that make it progressively more uranium-poor and gold-rich and the reworking that redistributes the chemical products as it concentrates them by thinning the assemblage.

As is more fully explained in the section on Degrees of Association of Uranium and Gold, mature reefs (table 2) are those whose rates of gold precipitation are controlled by the amount of uraninite present—that is, those reefs in which the amount of uraninite is so

diminished that the supply of uranium, rather than the supply of gold, controls the rate of precipitation of gold. Immature reefs, by contrast, are those in which uranium is in excess, and the supply of gold controls the rate of gold deposition.

Hydrogen-ion metasomatism of feldspathic debris and the formation of biogenic pyrite during the evolution of the reef were described in the section, Bacterial Sulfur Cycle. In the presence of dissolved gold and uranium it is expectable that these elements should be present in the authigenic reef minerals. Gold is present both in the phyllosilicates and in pyrite (Liebenberg, 1955; Ramdohr, 1958; de Kock, 1964), but the time of its deposition is debatable. Recently P. R. Simpson, by fission-track measurements, has found more than 40 ppm of uranium disseminated in the phyllosilicates of the Rand reefs. He believes this to have been absorbed at the time of deposition (Simpson, oral commun., October 14, 1975). Similarly, the sericitic matrix of Elliot Lake conglomerates has been shown to contain disseminated uranium (Roscoe, oral commun., October 14, 1975). These findings independently suggest that uranium was mobile during deposition of the reefs.

The relation of uranium solution to gold precipitation and accumulation may be visualized more readily on figure 9. The assumption made earlier is restated here: The curve traced by average ore grades on figure 6 may be used to describe the maturation of the uranium-gold reefs. In figure 9 the abscissa has been transformed to a linear time scale by assuming that the uranium loss with time maintained a constant half life (this is further explained in the section, Competency of Radiolysis). The gold content is assumed to have increased exponentially with time owing to reworking, which concentrates the continually precipitating gold into an ever-thinning reef. The ratio of uranium loss to gold gain dropped continuously, rapidly at first and ever slower in the later stages of reef development. Figure 9 treats the initial uranium grade as constant, showing only a single curve, in a comprehensive treatment differing grade would generate a nested family of curves. Note that in addition to indicating the effects of the chemical reactions that take place during reef development, figure 9 is concerned with the grade of the metals as they become concentrated in the thinning reef.

Cousins (1956, p. 106-108) recognized that large samples of reefs from a single mine have a characteristic  $U_3O_8/Au$  ratio (see points 6, 8, and 9 on fig. 6). However, for several reasons adjacent small samples show wide and erratic variations in the ratio. As will be explained in a later section, in immature reefs the ratio is dependent on the uraninite concentration and thus changes from point to point. Ratio fluctuations are also caused by the sealing of small patches of reef against chemical in-

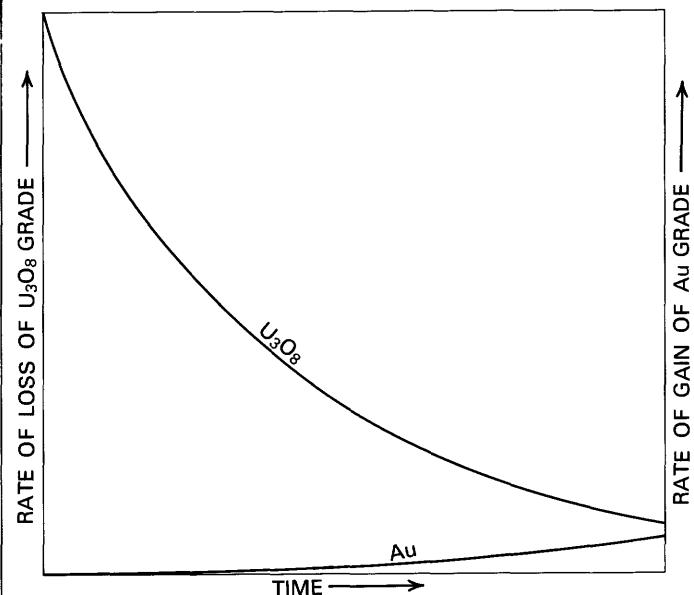


FIGURE 9. - Rate of uranium loss and gold gain of reefs in South Africa, Canada, and Brazil. Rates are plotted against time.

teraction (see the section, Thucholite), leaving uraninite-rich immature reef residual against more gold-rich areas. These immature patches skew the curve of figure 6, reflecting residual uranium on the gold-rich arm, from the ideal hyperbolic form. Lastly, the contents of the maturing reefs were mixed and concentrated by repeated reworking.

Pyrite appears to decrease in abundance with increasing maturity, seemingly along a curve like that of figure 6. In the Elliot Lake uranium reefs pyrite averages perhaps 10 weight percent (Griffith, 1967; Roscoe, 1969), dropping to 3-5 weight percent in the Serra de Jacobina gold ore (Bateman, 1958) and to about 3 weight percent in the average Rand conglomerate gold reef (Pretorius, 1964). In some of the thinnest, most mature reefs pyrite is scarce (Pretorius, 1964) or virtually absent (fig. 7). Some thin gold-rich reefs present a special case. At the Free State Geduld Mine (32, 36, 37 on table 2) the reef, a few centimeters thick, is rich in pyrite (Saager and Esselaar, 1969) and is overlain by barren quartzite very lean in pyrite. If one assumes that this reef is a reworked remnant of a thicker, less mature reef, then the pyrite of the remnant, calculated back to about a meter thickness, turns out to be about one-third that of the average Rand conglomerate gold reef.

Pyrite was depleted by the reaction that dispersed uraninite; buffering by pyrite moderated the oxidative dispersal of uranium. It follows that where pyrite was scanty or absent, uraninite was completely dissolved by the oxidizing effects of radiolysis.

Paradoxically, uraninite is locally more abundant than pyrite in immature ore at the Dominion Reefs Mine (Hiemstra, 1968a, p. 5). However, the high thorium content of the Dominion Reef uraninite (Grandstaff, this volume, chap. J; Feather, this volume, chap. Q) renders the mineral resistant to oxidation and solution, so that the association is not in conflict with the thesis advanced here.

Thucholite composition varies with maturity. Eleven Rand thucholites (de Kock, 1964) show a direct relation between weight-percent volatiles and the logarithm of the gold content (fig. 10). Organic analyses of 23 uraniferous petroleum asphaltites and one Rand thucholite (Pierce and others, 1958) show a strong direct correlation between volatile content and oxygen content (fig. 11). It follows that the gold-rich thucholites are more oxygenated than the gold-poor ones. The reactions that depleted uraninite and pyrite and precipitated gold in the Rand reefs also progressively oxygenated thucholite.

The positive correlation coefficients between  $U_3O_8$  and gold in the Rand ores (Saager and Esselaar, 1969) thus reflect not the detrital nature of both components, as these authors (p. 450) have postulated, but rather the dependence of the precipitated gold on the presence of detrital uraninite. In a like manner, the many demonstrations that gold in the Rand reefs responds to sedimentological factors primarily reflect such control of placer uraninite ( $d \sim 9$ ) rather than placer gold ( $d \sim 17\frac{1}{2}$ ).

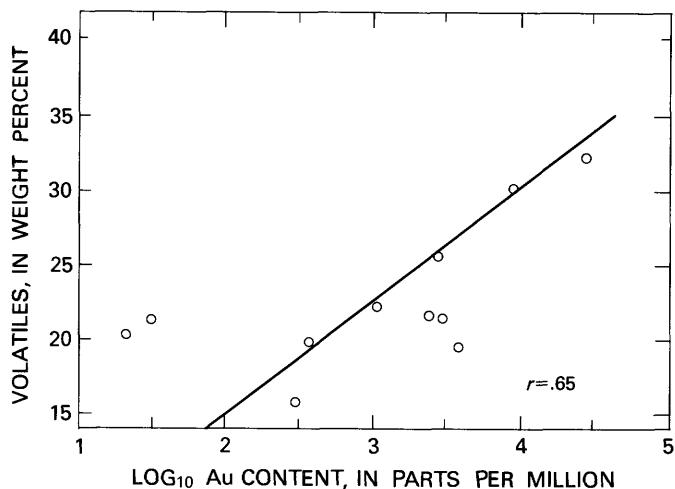


FIGURE 10. - Volatile content and logarithm of the gold content of 11 thucholites from the Rand reefs. Data from de Kock (1964) recalculated to an ash- and sulfur-free basis. The line is the best-fit linear regression calculated by the method of least squares.

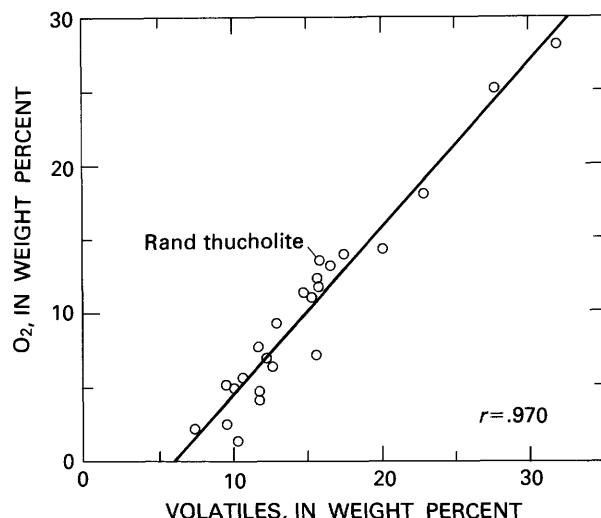


FIGURE 11. - Volatile content and oxygen content of 23 uraniferous petroleum asphaltites and one Rand thucholite. Data from Pierce and others (1958, table 1). Analyses presented on ash- and sulfur-included basis. The line is the best-fit linear regression calculated by the method of least squares.

### COMPETENCY OF RADIOLYSIS

The competency of the decay energy of uraninite as a driving force of the oxidizing reactions is indicated in the following sections. For each uranium nucleus decayed,  $3.8 \times 10^6$  hydroxyl free radicals are supplied by the radiolysis of water, whereas  $6.9 \times 10^6$  are needed to support the oxidation reactions. Evidently the calculated supply of oxidant falls short of a quantitative confirmation of competency, but in view of the limited data the fit is considered satisfactory, as supply and demand vary by less than a factor of two. (The gap between supply and demand of oxidant would be reduced or erased if an estimate could be made for the mechanical abrasion of pyrite, which is not allowed for in the following discussion but which surely must have taken place.)

In the Witwatersrand Basin the productive reefs are scattered through a stratigraphic section with a maximum thickness of about 9 km, spanning a time interval of half billion years. In the Central Rand a stratigraphic thickness of 8.55 km (Pretorius, 1964) separates the volcanics of the Dominion Reef Series ( $2.8 \times 10^9$  yr (Niekerk and Burger, 1964)) from the quartz porphyry in the Ventersdorp andesites ( $2.3 \times 10^9$  yr (Niekerk and Burger, 1964)).

Total time	$500 \times 10^6$ yr
Estimate of time lost by unconformity	$100 \times 10^6$ yr
Effective time	$400 \times 10^6$ yr

Therefore,

$$(8.55 \text{ km}) / (400 \times 10^6 \text{ yr}) = 21.38 \text{ m (70 ft) per million years.}$$

At the Loraine Mine, Orange Free State, the Basal Reef truncates a maximum of 140 ft (43 m) of footwall beds. Therefore,

$$140/70 =$$

2-million-year duration of Basal Reef unconformity.

Adoption of the Central Rand section, the thickest in the basin, is a conservative procedure, for it results in the minimum duration for an unconformity.

With an estimate of the length of time taken to form the Basal Reef at Loraine, it is possible to estimate the chemical half-life, or intensity of solution of detrital uraninite, during reef formation. In figure 6 it is seen that the Loraine point (28) projects along a maturity ray to intersect the normalized curve at a  $\text{U}_3\text{O}_8$  concentration of 165 ppm. The  $\text{U}_3\text{O}_8$  concentration at zero gold concentration is indeterminate, for beyond the upper limit of figure 6 the extrapolated curve becomes asymptotic to the zero gold ordinate. It is approximated here at about 2,640 ppm  $\text{U}_3\text{O}_8$ , 16 times the concentration of the normalized Loraine point. The Loraine point then lies 4 half-lives ( $2^4 = 16$ ) below the zero gold point. The apparent length of the chemical half-life of uraninite during reef formation is thus  $2,000,000 \text{ yrs}/4 = 500,000 \text{ yrs}$ .

However, this estimate does not take into account the effect of thinning the reef during reworking (see paragraph 2b in the section, Maturation of Reefs). It can be shown that by assuming a concentration of 2, due to thinning of the reef in 500,000 years to one-half its former thickness, which seems reasonable, the true chemical half-life of uraninite is halved from the net or apparent rate:  $500,000 \text{ yr}/2 = 250,000 \text{ yr}$ .

By comparing the chemical half-life of uraninite on the reef with the radioactive half-life of  $^{238}\text{U}$  and  $^{235}\text{U}$ , we can calculate the number of atoms of uranium chemically dissolved for each atom of uranium undergoing nuclear decay:

$$\frac{\text{radioactive half-life } ^{238}\text{U}}{\text{chemical half-life } \text{UO}_2 \text{ and reef}} = \frac{\text{number of molecules } \text{UO}_2 \text{ dissolved on reef}}{\text{}} = \frac{\text{}}{\text{}} = \frac{\text{}}{\text{}}$$

$$\frac{\text{}}{\text{}} = \frac{\text{}}{\text{}}$$

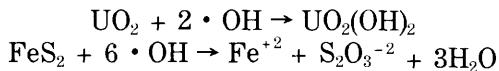
$$\frac{14.51 \times 10^9}{0.25 \times 10^6} = 18.04 \times 10^3 \text{ molecules } \text{UO}_2 \text{ dissolved per } ^{238}\text{U nucleus decayed, and}$$

for  $^{235}\text{U}$ ,

$$\frac{7.13 \times 10^8}{0.25 \times 10^6} = 28.52 \times 10^2 \text{ molecules } \text{UO}_2 \text{ dissolved per } ^{235}\text{U nucleus decayed.}$$

A large excess of pyrite was oxidized with the uraninite. The function of the pyrite as an oxidation buf-

fer indicates that it would only have been oxidized to the minimum necessary to dissolve the sulfide:



From the relative amounts of pyrite and uraninite in an immature and a highly mature reef (see section, Maturation of Reefs), the presumably steady-state relation varies from an  $\text{FeS}_2/\text{U}_3\text{O}_8$  ratio of 71, to one of 95, respectively. Because the amount of uraninite present  $2.5 \times 10^9 \text{ yr}$  ago was about 155 percent of that remaining today (see Appendix), the  $\text{FeS}_2/\text{UO}_2$  ratios originally prevailing were only 46.1 and 61.3, respectively. Assume that a pyrite-uraninite mix at an  $\text{FeS}_2/\text{UO}_2$  ratio of 61.3 was continually dissolved from the reef throughout its history. In the earliest stage assume this to be balanced by an increment of biogenic pyrite amounting to one-third that retained in the reef. This increment would diminish to zero in the most mature reef. (This is a simplifying assumption purely for ease of calculation.)

The oxidizing reactions presented and the  $\text{FeS}_2/\text{U}_3\text{O}_8$  ratio assumed may be combined (if the " $\text{UO}_2$ " of the chemical formula is equated with the  $\text{U}_3\text{O}_8$  of mining statistics) to estimate the number of hydroxyl free radicals needed to oxidize each molecule of uraninite and the accompanying pyrite:

Molecules oxidized	Hydroxyl free radicals consumed
1 $\text{UO}_2$	2
61.3 $\text{FeS}_2$	368
Total	370

For each  $^{238}\text{U}$  and  $^{235}\text{U}$  nucleus decayed, the minimum number of hydroxyl free radicals that must have been produced is as follows:

$$^{238}\text{U}: 18.04 \times 10^3 \times 370 = 6,674,800,$$

$$^{235}\text{U}: 28.52 \times 10^2 \times 370 = 1,055,240.$$

At  $2.5 \times 10^9$  years ago the  $^{235}\text{U}$  isotope, with shorter radioactive half-life than that of  $^{238}\text{U}$ , would have been more abundant in a sample of uranium. It can be shown (see Appendix) that  $^{235}\text{U}$  would then have constituted five percent of natural uranium:

$$\begin{array}{rcl} ^{238}\text{U}: 6,674,800 \times 94.786 \text{ percent} & = & 6,326,776 \\ ^{235}\text{U}: 1,055,240 \times 5.214 \text{ percent} & = & 550,202 \\ \hline \text{Total} & = & 6,876,978 \end{array}$$

Total for natural uranium =  $6.9 \times 10^6$  hydroxyl free radicals produced for each nucleus decayed.

Dragonić and Dragojić (1971, p. 125) report a mean value of  $4.1 \pm 0.5$  water molecules decomposed per 100 eV absorbed in the radiolysis of water. This value was averaged from "several hundred papers reporting data obtained under different experimental conditions."

Birch (1954, p. 151-152) lists the effective radioactive-decay energy of uranium as follows:

Isotope	MeV per atom	Cal per g-year
$^{238}\text{U}$	47.4	0.71
$^{235}\text{U}$	45.2	4.3

At  $2.5 \times 10^9$  years ago, the mass of a sample of uranium would have been more than 1 1/2 times its present mass, which has been depleted due to its continuing radioactive decay to lead (see Appendix). The radioheat production of  $^{238}\text{U}$  then would have been 146.84 percent of its present value; that of  $^{235}\text{U}$ , 48.93 percent of the present heat production of  $^{238}\text{U}$ . The decomposition of water molecules can now be calculated as follows:

$$\begin{aligned}
 ^{238}\text{U}: 4.1 \times 47.4 \times 10^4 \times 146.84 \text{ percent} &= 2,853,689 \\
 ^{235}\text{U}: 4.1 \times 45.2 \times 10^4 \times 48.93 \text{ percent} &= 906,771 \\
 \text{Total} &= 3,760,460 \\
 \text{Total} &= 3.8 \times 10^6 \text{ H}_2\text{O molecules decomposed per uranium nucleus decayed.}
 \end{aligned}$$

#### ORANGE FREE STATE GOLDFIELD

In the Orange Free State Goldfield, uranium is relatively high, and gold low, on the margins of the field, where the unconformity beneath the productive Basal Reef is minimal (Winter, 1964a, b; McKinney and others, 1964; Minter, this volume, chap. K). Gold increases and uranium diminishes as the unconformity becomes more pronounced: the reef increased in maturity where the unconformity was of longer duration. Detrital grains of platinoids correspondingly are more abundant—0.008 ppm—and less mature—Os + Ir = 66-71 percent (Cousins, 1973)—on the margins of the goldfield and less abundant—0.0025-0.004 ppm—and more mature—Os + Ir = 76-79 percent—where the unconformity is longest lived. The decrease in abundance is in part due to the electromotive replacement of less-noble platinum-group alloys by complexed gold. Some of the few platinoid grains met with in polished sections are partially replaced by gold (Liebenberg, 1955; Ramdohr, 1958). The rare near-spherical gold grains adduced by Liebenberg (1955) as evidence for the detrital origin of the Rand gold are here interpreted as complete replacements. (Figure 3 shows that thin flakes rather than near-spherical grains of placer gold would be expectable in the reefs. In contrast, the hard, brittle platinoids were not deformed into flakes.) The increase in osmium and iridium content with maturity in the anoxicogenic environment is due to chemical stability and superior hardness; oxidizable osmium and iridium both diminish with maturity in modern placers accumulating in an oxidizing atmosphere, and platinum becomes increasingly dominant (Mertie, 1939, p. 135-137).

#### WHITE REEF

A series of as many as a dozen generally uranium-rich reefs scattered through 80 m of quartzite (Toens and Griffiths, 1964) occur locally in the West Rand but are not as numerous elsewhere in the Witwatersrand Basin. The local proliferation of the Bird Reefs suggests strongly that they were, for the most part, short-lived phenomena. One of these is the White Reef (fig. 12). Small patches of mature White Reef ( $\text{U}_3\text{O}_8/\text{Au}$ , 10-15) have been mined for gold in the northeastern part of the field, but the greatest production has come from Randfontein, where much less mature reef (minimum  $\text{U}_3\text{O}_8/\text{Au}$ , range 61-261) was mined in large quantity for both uranium and gold (Toens and Griffiths, 1964). Figure 12 shows that the  $\text{U}_3\text{O}_8/\text{Au}$  ratio varies progressively from point to point across the mined extent. The small amount of mature gold reef mined, in contrast to the extensive mining of less mature reef, suggests that maturation may have been degenerative. Figure 13 shows the range of uranium-gold ratios for the White Reef and for the Basal Reef of the Orange Free State. The combined range of the two reefs, independently documented by figure 12 and by Minter (this volume, chap. K), is the greater part of the range of all pyritic conglomerate reefs for which uranium-gold ratios are available (fig. 6; table 2). The range of White Reef uranium-gold ratios crosses the gradation between immature and mature reefs, as sketched in tables 2 and 4 and figure 15, and suggests that no fundamental difference exists between the two types of reef.

#### MIDDLE ELSBURG REEFS

In the country lying between the West Rand and the West Wits Line a zone of conglomerates called the Middle Elsburg Reefs has recently been brought into production (Mining Magazine, 1976; Papenfus, 1964, p. 390). The mined reefs lie in the upper half of a sequence of three conglomeratic subzones spanning a stratigraphic thickness of as much as 120 m. The upper conglomerate of the top group of reefs is the most persistently auriferous, whereas the conglomerates of the middle group have the most consistent uranium content. Table 3 shows that in going from the immature to the mature end of the Cooke Section the gold content of 66 million metric tons of ore nearly doubles as the uranium content drops by more than half.

TABLE 3. - *Indicated ore, Cooke Section, Randfontein Mine*

[Mining Magazine, 1976]

Area	Metric tons (millions)	Au (ppm)	$\text{U}_3\text{O}_8$ (ppm)	$\text{U}_3\text{O}_8/\text{Au}$ (ratio)
No. 1 Shaft Area (North)	19	11	200	18
No. 2 Shaft Area (Central)	18	7	400	57
No. 3 Shaft Area (South)	29	6	450	75

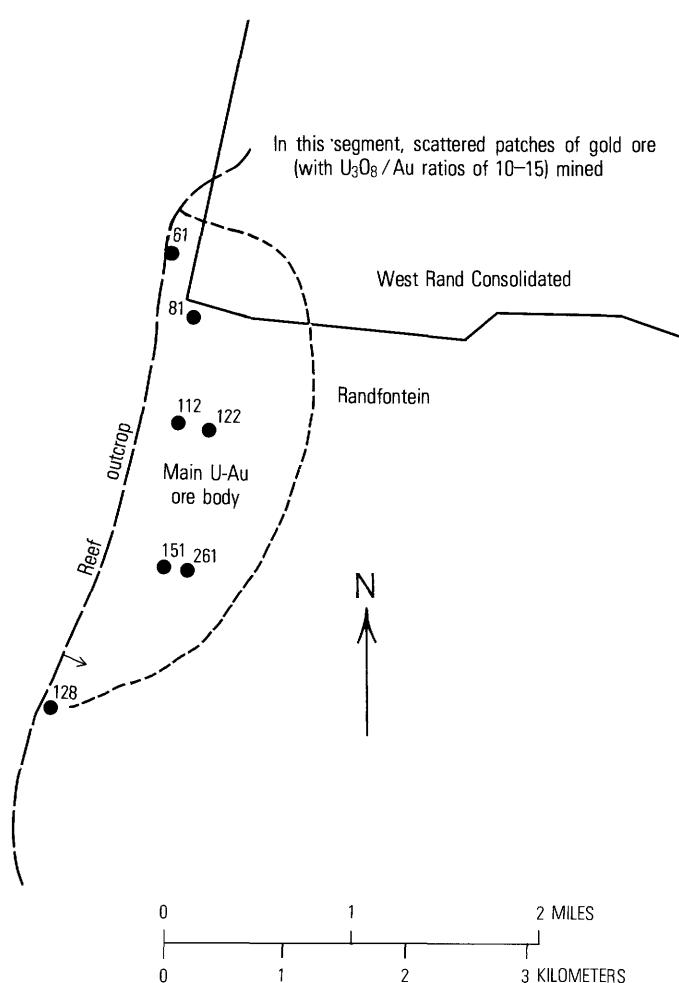


FIGURE 12.—Sketch map showing distribution of  $U_3O_8/Au$  ratios in the White Reef, West Rand. Randfontein  $U_3O_8/Au$  data based on 125 assays provided by Johannesburg Consolidated Investment Group. Other data are from Toens and Griffiths (1964).

#### DEGREES OF ASSOCIATION OF URANIUM AND GOLD

In the early stages of reef formation (immature reef) uraninite was present in excess of the amount needed to precipitate the amount of gold available in complexed solution. In the later stages (mature reef) uraninite was so diminished in amount that the effects of its radioactive decay did not precipitate all available dissolved gold. The amount of gold precipitated was thus controlled by the limited amount of uraninite present. Under the latter conditions a fixed amount of gold would have been precipitated for each unit of uraninite present. Thus in a mature reef a perfect association between uranium and gold would be expectable. In immature reefs the degree of association or correlation would have been less than perfect.

Discussions of the reef maturation process have until now made the simplifying assumption that the reef had

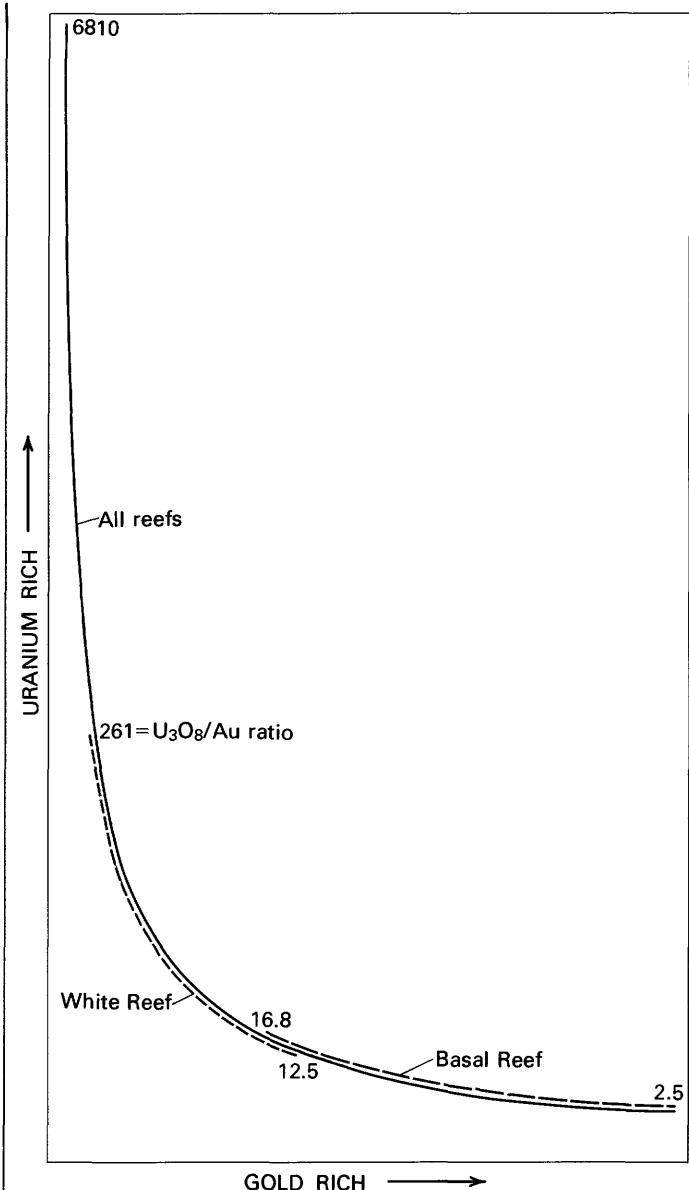


FIGURE 13.— $U_3O_8/Au$  range of the White Reef, West Rand, and the Basal Reef, Orange Free State, compared with all reefs. (Curve from fig. 6; specific values from tables 2 and 4 and fig. 12.)

a uniform grade. In any real situation the original concentration of the detrital uraninite would have been highly variable from point to point. Figure 14 shows the effect of different original concentrations of detrital uraninite in a maturing reef. Each curve represents the decrease with time of an arbitrary concentration of detrital uraninite, the highest concentration being represented by the steepest curve. The horizontal line connecting the bottoms of the curves represents the constant ratio ( $U_3O_8$  present per unit of gold precipitated) achieved as each successively higher concentration of detrital uraninite diminished to the level where it deter-

mined the rate of gold precipitation. This constant ratio defines maturity for a particular original concentration of detrital uraninite. At time  $t_0$  the ratios ( $U_3O_8$  present per unit of gold precipitated) would be markedly different still. At time  $t_0$  the degree of correlation between uranium and gold in a suite of samples would be poorer than in samples taken at time  $t_1$  or  $t_2$ . uranium and gold in a suite of samples would be poorer than in samples taken at time  $t_1$  or  $t_2$ .

Let us assume that the uppermost curve of figure 14 represents the highest original concentration of detrital uraninite occurring with significant frequency in the reef. Then the point just off the right-hand side of the figure, where this curve intersects the maturity line, represents the time at which the reef as a whole would have achieved maturity and thus a perfect correlation between uranium and gold.

As will be shown below, this condition does not seem to have been achieved in the Witwatersrand, for another factor intervened that progressively diminished the degree of association of uranium and gold. This factor is

the sealing-off of local patches of reef against chemical interaction (see the section, Maturation of Reefs), and it would have become more widespread with time, halting the maturation of the patches at intermediate levels. By juxtaposing various  $U_3O_8/Au$  ratios the process would have progressively reduced the degree of correlation between the two metals.

This analysis predicts that the degree of correlation between uranium and gold should rise from low values in a very immature reef to a maximum at intermediate maturity and then fall to lower values at higher maturities.

In this treatment the degree of correlation between the two elements will be expressed by the linear coefficient of correlation  $r$  (also termed the product-moment correlation coefficient). The linear relation between two variables  $X$  and  $Y$  is calculated by the formula

$$r = \frac{\sum (X_i - \bar{X})(Y_i - \bar{Y})}{\sqrt{\sum (X_i - \bar{X})^2 \sum (Y_i - \bar{Y})^2}}$$

where  $\bar{X}$  and  $\bar{Y}$  represent (Wonnacott and Wonnacott, 1970, p. 103-108) the arithmetic means of  $X$  and  $Y$ . The quantity  $r$  varies from +1 (perfect positive correlation) through 0 (no correlation) to -1 (perfect inverse correlation).

In figure 15,  $r$  for uranium and gold is plotted against the logarithm of the  $U_3O_8/Au$  ratio for thirteen samples of pyritic conglomerate of the widest range of maturity (table 4).

TABLE 4.— $U_3O_8/Au$  ratios and the linear coefficient of correlation  $r$  between uranium and gold for sample suites from reefs in South Africa and Canada

Sample suite	Number of samples (locality)	$U_3O_8/Au$ ratio	$r$	Source
1, Elliot Lake district	34	6,779	0.13	Roscoe (1969, Appendix table C).
Dominion Reef:				
2 Dominion Reefs Mine	20	477.2	.55	Hiemstra (1968b, table V).
White Reef:				
Randfontein Mine:				
3 F	24 (14 level)	261.1	.38	B.D. Stewart, Johannesburg Consolidated Investment Co., Ltd., written commun., Sept. 18, 1975.
4 E	15 (8 level)	150.7	.89	Do.
5 G	25 (6 level)	128.4	.36	Do.
6 D	13 (14 level)	122.1	.63	Do.
7 C	20 (8 level)	112.3	.91	Do.
8 B	17 (8 level)	81.30	.80	Do.
9 A	11 (3 level)	61.44	.85	Do.
Vaal Reef:				
10 Zandpan Mine	44	23.31	.88	Hiemstra and Rahden (1969).
11 Hartebeestfontein Mine	43	17.36	.86	Do.
Ventersdorp Contact Reef:				
12 Venterspost Mine	56	6.62	.70	Do.
Basal Reef:				
13 Free State Geduld Mine	109	2.78	.605	Saager and Esselaar (1969).

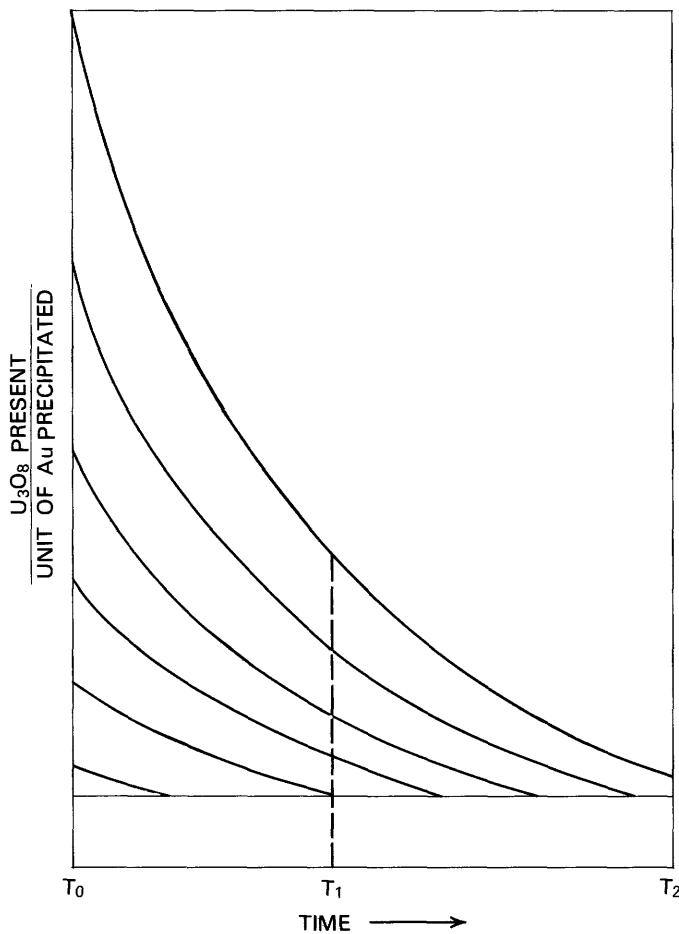


FIGURE 14.—Changes in the ratio  $U_3O_8$  present per unit of gold precipitated for different detrital uraninite concentrations in a reef.

<sup>1</sup> Numbered points on figure 15.

Immature reefs

Mature reefs

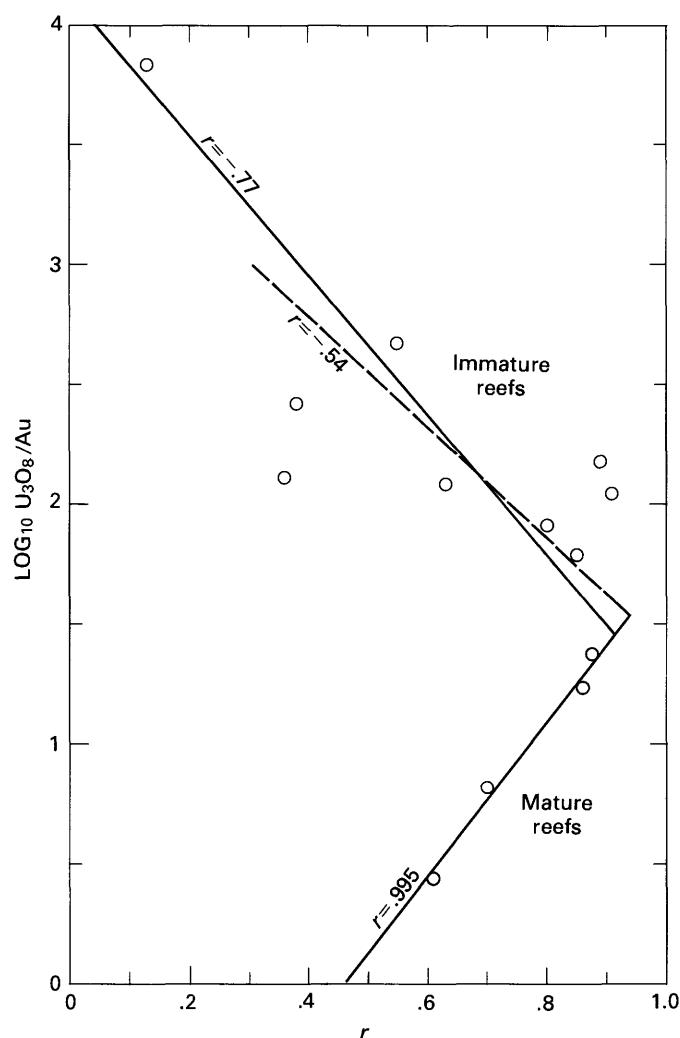


FIGURE 15. - The linear coefficient of correlation  $r$  between uranium and gold compared with the logarithm of the  $\text{U}_3\text{O}_8/\text{Au}$  ratio for individual reefs in the Witwatersrand and for the composited Blind River-Elliot Lake District. The dashed-line slope excludes the Canadian sample. (Data from table 4). The lines are the best-fit linear regressions calculated by the method of least squares.

The samples fall into an immature group, for which  $r$  increases with increasing maturity, and a mature group, for which  $r$  decreases with further increases in maturity. The variation of  $r$  is in accord with the foregoing analysis and clearly supports the outlined hypothesis of origin of the reefs. The relatively wide scatter of points from the immature reefs probably derived from the small sample sizes, although all but the Canadian sample are well above the limit for a five-percent level of significance. Because the Elliot Lake district sample is composited from several productive conglomerates rather than a single reef, it has been excluded from an alternate treatment of the immature reefs. Theoretical

considerations, however, and figure 15 combine to suggest that the linear correlation coefficient  $r$  of a sample suite of ore grade should not depend on whether the samples came from one or many reefs but only on the  $\text{U}_3\text{O}_8/\text{Au}$  ratio of the suite.

#### VOLCANIC INFLUENCE BLIND RIVER-ELLIOT LAKE

The Blind River-Elliot Lake uranium-bearing conglomerates occur at or near the base of a Proterozoic sedimentary sequence, which in the vicinity of the mines lies with angular unconformity upon Archaean greenstone (Roscoe, 1969), locally pyritized along its upper contact. The protecting pyrite in these reefs is abundant and coarsely angular. It may have been partly derived from sulfidization of iron-rich Archaean greenstone weathering nearby, for basal Proterozoic volcanism within the district itself (Roscoe, this volume, chap. W), and extending 30 km to the south (Roscoe, 1969), must have released abundant  $\text{H}_2\text{S}$  into the anoxic environment (in modern environments volcanic  $\text{H}_2\text{S}$  is quickly oxidized to  $\text{SO}_4^{2-}$ ). The Blind River reefs, in contrast to those of the Rand, characteristically contain tightly packed, subangular clastic grains of uraninite in unthucholitized streaks and cross laminae. Thucholite is generally rare but locally abundant (Roscoe, 1969; Ruzicka, this volume, chap. V); the low content of gold is attributed here to the brevity of periods of reef development. Uraniferous pyritic conglomerates extend eastward from Elliot Lake, and at Lake Timagami, 200 km easterly, a more mature basal conglomerate of similar age locally contains ten times more gold ( $\geq 2.7$  ppm Au,  $\leq 180$  ppm  $\text{U}_3\text{O}_8$ ; Thomson, 1960). Here, instead of the iron-rich greenstone of the Elliot Lake area, Archaean iron-formation lies unconformably below the mineralized conglomerate and presumably contributed the iron for the pyrite.

The Blind River-Elliot Lake ores are commonly regarded as containing insignificant amounts of gold. Yet the average gold content for the 34 samples for which both gold and uranium assays are available (Roscoe, 1969, Appendix table C) is 0.26 ppm, twice the average yield of gold dredging (see the section, Gold, Placer Origin Unlikely).

#### VENTERSDORP CONTACT REEF

In the Rand, the contact between Witwatersrand strata and overlying Ventersdorp andesites is commonly an angular unconformity marked by the Ventersdorp Contact Reef, which, though not widely productive and unique in its explicit volcanic association, is a typical Rand reef (Haughton, 1969; de Kock, 1964). In and near

the Western Areas Mine, however, no unconformity is detectable, and a tuffaceous gradational zone as much as 30 m thick, here believed the marine equivalent of early lavas, lies beneath andesite flows. Within the zone and as much as 100 m below, several thick, relatively poorly sorted, coarse conglomerates (Papenfus, 1964), polymictic in part, are atypically rich in gold. A high  $H_2S$  pressure during volcanism would have increased the solubility of aurous thiocomplexes and, with unconformably older reefs as additional local gold sources (de Kock, 1964; Papenfus, 1964), would have increased their concentration as well and so increased the rate of gold precipitation in the reefs.

## DISCUSSION

The traditional view that the gold of the pyritic conglomerates is placer has been generally refuted in the section, Gold. It remains to comment on the detailed mode of transportation of the allegedly placer gold. Minter (this volume, chap. E) has advanced the latest version of the fluvial model of sedimentation of the reefs. He has emphasized sedimentation on the branching distributaries of a large deltaic fan. Minter's model is silent as to the significance of the unconformities upon which the productive reefs lie. Deltas and fans, taken as a whole, are areas of uninterrupted deposition. Yet by contrast the Rand reefs, of delta-wide dimensions, are developed on through-going unconformities marked by the reworking of material eroded from the underlying beds.

Minter (this volume, chap. K) would explain the inverse relation between uranium and gold in the Orange Free State Goldfield by the preferential transport of less competent fine-grained detrital uraninite to the distal end of the fan. Gold would be preferentially deposited near the head of the fan. Such a mechanism is appealing but does not seem to fit field descriptions. Winter (1964a, p. 435-436, 446; 1964b, p. 530-532, 541, 545-546), describing this goldfield, built up a picture of oligomicitic, cleanly washed reefs forming on unconformities from underlying polymictic subgraywacke. He wrote (p. 436): "Generally, the  $U_3O_8$ :Au ratio is considerably higher in the somewhat polymictic conglomerates developed in the subgraywacke, even though the absolute gold contents do not nearly approach those of the other type" (oligomicitic cleanly washed conglomerate). This initial formation of relatively uraninite-rich, gold-poor conglomerate is in conflict with the mechanism of selectively bypassing uraninite down current, whereas the variable maturity model advanced here is in complete harmony with Winter's observations.

In the section on the Orange Free State Goldfield, an inverse relation between gold and detrital platinoid grains is described. If the Rand gold were detrital, a

direct relation between the two metallic particles would be expectable, for they have similar densities ( $d \sim 17.5$ ). This would follow even though the malleable gold would expectably have been flaky and thus in particles much heavier than the near-spherical brittle platinoid particles (fig. 3). The average median diameter of the latter (58  $\mu m$ ), as reported by Cousins (1973), is just that computed for hypothetical spherical particles with  $d \sim 17.5$  in hydraulic equivalence with the average Rand reef clasts (fig. 3). The hydraulic equivalence of platinoid grains and reef clasts, coupled with the inverse relation between platinoids and gold, strongly suggests that gold was not of placer origin but was transported in solution.

The thesis developed here has important implications in assessing the uranium reserves of the pyritic conglomerates, particularly those of the Witwatersrand, the mined representatives of which are very largely gold rich. If the gold-rich reefs are the products of long-continued maturation on unconformities, then they should both grade into and be apposed by less mature uranium-rich reefs. On the ground that unconformities of lesser duration should be more numerous than those of greater duration, less-mature uranium reefs should be the more abundant. Indeed, as much is stated by Cousins (1956, p. 106-108; quoted in this report in the section, Inverse Relation of Uranium and Gold), who noted that high ratios of  $U_3O_8$ :Au "indicate a more widespread distribution of uranium than gold." Nevertheless, the tonnage of uranium-rich reefs so far mined constitutes only 1.25 percent of that of gold-rich reefs. This paradox is held to be due simply to the historical pattern of an industry devoted first and foremost to the mining of gold.

Recognition of the presence of a significant amount of uranium in the reefs did not come until World War II (Bourret, this volume, chap. A), and wide knowledge of the abundance was delayed until about a decade later.

The West Rand Goldfield leads in production from uranium-rich Witwatersrand reefs. Yet the recently published reserves of a new section of a single mine (see table 3) contain more uranium than the total mined through 1975 from the West Rand. It is significant that the uranium-rich reefs of the Cooke Section of the Randfontein Mine exploit a stratigraphic zone not yet mined elsewhere. It seems likely that the Witwatersrand will emerge as an even more massive reserve of uranium than has been heretofore recognized.

## APPENDIX: DETERMINATION OF ORIGINAL URANIUM

The original amount of natural uranium and its isotopic distribution at  $2.5 \times 10^9$  yr ago may be determined from that remaining at present by use of tables of Steiff and others (1959). The tables, which were

computed for the calculation of lead-isotope ages, list the atomic ratios of radiogenic lead isotopes to the parent uranium isotopes. The original amount of uranium isotope is simply that remaining at present plus that decayed to the equivalent lead isotope.

*Atomic ratios of indicated isotopes at  $2.5 \times 10^9$  yr*

[Stieff and others (1959, p. 34)]

$N_{206}/N_{238}$	$N_{207}/N_{235}$
0.468478	10.36340

If the present amount of the parent uranium isotope is taken as unity, the original amounts are

$^{238}\text{U}$	$^{235}\text{U}$
1.468478	11.36340

The present  $^{235}\text{U}$  content of natural uranium = 0.711 percent.

$$11.363 \times 0.711 \text{ percent} = 8.079 \text{ percent},$$

$$^{238}\text{U} = 146.8478 \text{ percent present natural uranium},$$

$$^{235}\text{U} = 8.0794 \text{ percent present natural uranium},$$

Natural uranium at  $2.5 \times 10^9$  yr = 154.9272 percent present natural uranium.

The isotopic composition of the natural uranium would have been:

$$^{238}\text{U}: 146.8478/154.9272 = 94.79 \text{ percent},$$

$$^{235}\text{U}: 8.0794/154.9272 = 5.21 \text{ percent}.$$

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## DISCUSSION FOLLOWING TALKS BY DRS. STAATZ, CANNON, AND MYERS

**Skinner:** Brad [Myers], I'll confine myself to three points, three points that I don't think are stable points on which to build your hypothesis. The first is settling velocity. Dealing with the Witwatersrand, I think you are dealing entirely with bedload and settling velocities have little to do with it. So I would maintain that the Russian information on settling velocities is incorrectly applied by you to the grain size of the Witwatersrand gold. The second point concerns Eh-pH diagrams. How you draw those fields depends on the molalities of the various species in solution. You didn't really give any justification for those you chose, and I

think you could draw those diagrams somewhat differently.

*Myers:* If you would like, I can give you the details. I have a manuscript if you care to look at it.

*Skinner:* Even if we leave them the way you've drawn them, I would maintain that the diagram should be correctly interpreted as the reason why waters have not flushed out the uranium and gold, not why they brought gold in. That is, you have shown a field where you have uraninite, gold, and pyrite stable, and the pyrite controls the system in part and doesn't let the gold and uranium be flushed out. The third point is one I don't know very much about and I would hope somebody in the audience might know a great deal more about it, but I would very much doubt that the radiation flux in these waters is anywhere near high enough for the radiolysis reaction to go on. I would imagine that it is many orders of magnitude too low.

*Myers:* On the intensity of the radiolysis, it certainly would go on. The point here is whether it would go on fast enough. I did some preliminary calculations and thought that it would go on fast enough. I have had criticism on this point before, and at this point I do not want to tie myself down on it.

*Grandstaff:* I'd suggest that the presence of uraninite in other old Precambrian rocks, pegmatites for example, would suggest that this process is not important in destroying the uraninite.

*Myers:* In the original description of thucholite, the author stressed that the thucholite replaced coarse uraninite in the pegmatite. For the reaction that I described to take place, near-surface low-pressure conditions are needed. Uraninite in other Precambrian rocks may not have been subjected to the proper conditions for a sufficient length of time.

*B. Nagy:* Just a very brief comment. In your paper you have shown a normal alkane transformed by irradiation into what you call thucholite. In other words, the reaction says that thucholite is a polymerized hydrocarbon, obviously, basically aliphatic. Who analyzed the organic composition of the thucholite?

*Myers:* These are old analyses; they are in Liebenberg.

*B. Nagy:* How have the data been analyzed?

*Myers:* They have been analyzed by a relatively primitive method, but they are oxygen containing. Other thucholites have been analyzed by Pierce; these are oxygen containing.

*B. Nagy:* Then the hydrocarbon transformed into thucholite is not the parent material. In other words, you are making the thucholite or whatever (I hate to use thucholite because this is not the classical definition) from other materials also.

*Myers:* Wait, you say that the Rand carbonaceous material is not thucholite?

*B. Nagy:* Well, I don't want to get embroiled; I talked about this yesterday. This is just terminology. But what I am saying is this, that I would imagine that if people would accept your hypothesis that the starting material, which I call "solid organic substance" and what you call "thucholite," is something other than normal alkane hydrocarbons.

*Myers:* Of course it is. It's oxygenated, as my reaction shows.

*B. Nagy:* But you didn't show the steps.

*Myers:* I can't show the steps. However, Charlesby has irradiated polyethylene in a nuclear reactor, and he got very long chain hydrocarbons plus hydrogen plus methane, but this is done anhydrously. When you do it with water then you have radiolysis of water to contend with, and this is an oxygenating reaction. There have been experiments on crude oil irradiated with water, and you get an increase in the percentage of asphaltenes, and the reason this is oxidizing is because you form reducing gases in a bubble in a vial, and these do not equilibrate. These are very high radiation rates, of course.

*B. Nagy:* You brought in asphaltenes. I talked yesterday about asphaltenes, and all these things are aromatic heteroatomic compounds. They are not aliphatic compounds.

*Myers:* Which ones are you talking about?

*B. Nagy:* I'm talking about what you call thucholite. I'm talking about asphaltenes.

*Myers:* What you've found was as the result of pyrolysis?

*B. Nagy:* Yes.

*Armstrong:* I believe that you gentlemen should get together. Dr. Hallbauer? Dr. Hallbauer, you're not going to talk about thucholite, are you?

*Hallbauer:* I would like to make a comment on one diagram you used to support your hypothesis where you plotted the gold in ppm against the uranium oxide. In that diagram you used the milled ore, assuming that the milled ore is the same as reef.

*Myers:* I'm aware of this and I explained it.

*Hallbauer:* Your scale at the bottom went to about 50 ppm, but if you use reef you get values of 1,000, 2,000, sometimes 10,000 ppm, so these values would be way out, they would be somewhere up here. So your whole graph would be invalid. Say, for instance, if you had used the average stoping width of, say, 1 meter. Some mines might have all this as reef; other mines might have only 10 centimeters of reef, and in mining it is diluted with base rock. If you draw this as milled ore, you assume that it is all the same material. That is, it is all reef. But it's not all reef, and these thin conglomerates contain up to a few thousand ppm of gold. I would say, therefore, that your gold values would be somewhere up here, destroying the whole diagram.

*Myers:* The objection was made that I have used average values instead of individual values, and I can't see that that's a very serious objection. I used millions of tons of milled ore rather than individual reef samples, and it seems to me that this is the proper way of doing it. There are better figures, but I don't have them.

*Hallbauer:* Well, the only difference is if you would have used individual values for conglomerates then you couldn't have drawn this diagram. The diagram is invalid. You can't use it to support your hypothesis.

*Myers:* Why is it invalid?

*Hallbauer:* (Reply not recorded. Tape blank.)

*Myers:* Larry Minter supplied us with some very interesting curves, and he used 930,000 assays to make those curves, and he used assays of reef. There certainly has to be a pretty good correlation between the reef and the milled ore; it is not perfect, but there has to be a pretty good correlation.

*Armstrong:* I don't think this is going to be settled.

Larry [Minter], do you want to say something?

*Minter:* I think you have some terrible data. For instance, if you take the milled ore from Randfontein or the West Rand Consolidated, they happen to be mining twenty different reefs at the same time, and the whole lot are going through the mill, and every one of those reefs represents a different type of environment. That's just one point. The other point is that on FSG, for instance, the gold-uranium ratio happens to vary from seven to over forty. Now, which one do you take?

*Myers:* What I took were reserve values given to me by Anglo-American for the average grade of  $U_3O_8$  and gold for the eastern, central, and western sections of Free State Geduld.

*Minter:* I don't think that is really representative.

*Myers:* It's representative. It's the reserve data.

*Minter:* Yes, but it's mixed data.

*Myers:* It's all from Basal Reef.

*Minter:* No, it actually isn't.

*Myers:* It must be very close to all. As far as I know, Randfontein uranium production is very largely from White Reef.

*Minter:* There happen to be two reefs there.

*Myers:* And remember we had three rather straight plots; those were year-by-year plots with the average. West Rand Consolidated is not on there. It's another spot, and it's another spot because they do mine many [reefs], and if you plot yearly figures, you don't get a straight line. You get a Brownian movement because of what you just said.

*Minter:* The White Reef doesn't carry very high uranium content; it's the Monarch Reef that carries the high uranium content.

*Myers:* Would you care to see the data sent me by JCI?

*Minter:* Sure. One other point is that we still haven't really said why this unique process happened only once and where all the gold came from and why we happen to have got it all in the Witwatersrand? Has this happened anywhere else?

*Myers:* It's not happened to the same extent. However, there are other gold-uranium conglomerates—the Denny Dalton is one, in the Pongola Series southeast of the Witwatersrand. There are pyritic conglomerates in the Ukraine; we don't know how much uranium they contain, but they contain detrital uraninite, and they contain up to a gram or so of gold. In terms of crustal abundance, there's a fair amount of gold in Elliot Lake, and if we go down east of Sudbury to Lake Timagami you get a couple of parts per million of gold.

## **SUMMATION**



# Thoughts about Uranium-Bearing Quartz-Pebble Conglomerates: A Summary of Ideas Presented at the Workshop

*By* BRIAN J. SKINNER

GENESIS OF URANIUM- AND GOLD-BEARING PRECAMBRIAN  
QUARTZ-PEBBLE CONGLOMERATES

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GEOLOGICAL SURVEY PROFESSIONAL PAPER 1161-BB





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## THOUGHTS ABOUT URANIUM-BEARING QUARTZ-PEBBLE CONGLOMERATES: A SUMMARY OF IDEAS PRESENTED AT THE WORKSHOP

By BRIAN J. SKINNER <sup>1</sup>

### INTRODUCTION

The reasons for my selection as the summarizer and final speaker at this intriguing conference are just as obscure to me as they are to you. I suspect the main reason is that I know little or nothing about quartz-pebble conglomerates and their uranium contents, so I can be expected to approach my task in an unbiased way. Whatever the reason, let me remark that I found the talks to be extraordinarily provocative and my attendance at the conference a great privilege.

I cannot hope to summarize in a few minutes all of the fascinating discussions of the past three days. Therefore, I propose to comment on six or seven of the most interesting, and to my mind most pertinent, topics discussed. I will not include the debate of detrital versus nondetrital origin for the uranium, gold, and other constituents among my topics because it seems clear to me that even if some of the ore constituents were carried in solution, most of them were moved as detrital grains. Some of the points I make will probably be self-evident to those of you who have long acquaintances with uraniferous quartz-pebble conglomerates, but I believe the importance of the points makes them worth repeating.

### UNIQUENESS OF THE DEPOSITS

The first point concerns the uniqueness of quartz-pebble conglomerates. It struck me, as I listened to the presentations, that I had heard similar words 30 years ago, right after the Second World War, when one of the periodic debates about Precambrian iron-formations erupted. Similar debates about iron-formations had occurred at earlier times, but the discussions were necessarily restricted because the formations seemed to be

few in number and to have formed because of special, local geological circumstances. But easier travel and faster communication made it apparent by the second half of the 20th century that iron-formations of the Lake Superior type are really quite widespread. Surprisingly, large Precambrian iron-formations are still being discovered today—for example, the recently discovered Nabberu Basin deposits in Western Australia. Furthermore, it now is apparent that these iron-formations were normal sediments in their day. Local and special circumstances are not needed.

Our understanding of uranium-bearing quartz-pebble conglomerates today is like our understanding of iron-formations 30 years ago. It is biased by the preponderance of information from the Witwatersrand and Blind River deposits. But at this conference you have discussed a number of other quartz-pebble conglomerates, and you have convinced me that Precambrian conglomerates suitable to be uranium hosts are probably much more common than now accepted. They are not highly unique and probably not due to a special set of local circumstances. This suggests there are many more Precambrian quartz-pebble conglomerates yet to be found. Whether they will prove to be mineralized, and if mineralized, worth mining, remains to be seen. But as far as uniqueness or nonuniqueness is concerned, I am led to a conclusion of nonuniqueness. If we could return for another conference 30 years hence, I suggest we would be discussing three or more times the number of deposits we are discussing today.

### TIME OF FORMATION

My second point concerns time of formation. It is a question that many of the speakers mentioned and many of you questioned from the floor. Quartz-pebble conglomerates can, of course, be found in geological sections of any age. The ones we are interested in have

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special contents of uranium, pyrite, and gold, suggesting either that a special style of sedimentation must have prevailed for a limited time or that some special conditions of surface waters and atmosphere existed for a period of Earth's history. Regardless of the reasons, there does indeed seem to be a specific time during which conditions around the globe were generally conducive to formation of uraniferous quartz-pebble conglomerates. Those few small deposits that fall outside the specific time range (for example, those in the eastern Urals) seem to be due to special, local conditions and to have somewhat different and distinct characteristics.

There are no known deposits older than 2.8, or at a maximum, 3.0 b.y. (billion years). This may or may not be a real time barrier. One could argue that it is simply a problem of sampling. I maintain that this is not the case. I am persuaded by the observations of Stuart Roscoe that what we see in the record at 2.8–3.0 b.y. is a change of tectonic patterns, which is distinguished by a new style of sedimentation occurring in cratonic basins. If similar basins and suitable source areas had existed at earlier times, still older quartz-pebble conglomerates could have formed. But they did not form because the basins and adjacent sediment areas did not have the right geometries.

What of the other end of the time bracket? The youngest deposits that seem to fit the features of uraniferous quartz-pebble conglomerates are those at the base of the Transvaal System, as discussed by Dr. Whiteside. They were formed by reworking of older sediments but must still have involved transport of detrital grains of uraninite. The base of the Transvaal is about 2.0 b.y. old, which suggests that uraniferous quartz-pebble conglomerates were limited to a 1-b.y. time span.

The reason for a cutoff at 2 b.y. ago, if it was a distinct cutoff, was not settled at this conference. I do not feel that the argument for atmospheric control—a simple atmosphere composition control—was proven. I am not sure we have even been thinking correctly about the atmosphere. There was a lot of talk about an oxidizing atmosphere versus a reducing atmosphere. But an atmosphere with a low partial pressure of oxygen might be erroneously interpreted to be a reducing atmosphere if a given oxidation reaction did not proceed. The lack of reaction may simply mean that reaction kinetics are too slow for oxidation to occur in the time available. David Grandstaff demonstrated that atmospheres with partial pressures as high as  $10^{-4}$  atm of oxygen are compatible with extensive transport of detrital uraninite and pyrite. Temperature, he pointed out, played an even more important role in reaction rates. That point was strongly reinforced by Mort Staatz' report of modern detrital

uraninite [thorianite] in a glacial stream in Alaska. Thus, climate, and in particular the average temperatures, may be a vitally important control in the formation of uraniferous quartz-pebble conglomerates.

This leads me to an obvious question: Where were the cratons relative to the poles when the basins formed and were filled by sediments? Was the appropriate style of sedimentation a feature of high latitudes and hence low temperatures? Until that question is answered I believe the question of a cutoff time must remain open. Present evidence suggests we could very well have had an oxygenated atmosphere with as much as  $10^{-4}$  atm of oxygen as far back as 3 b.y., and still had transport of detrital grains of pyrite and uraninite for long distances. We cannot, therefore, conclude that the atmosphere did exert a control, and this in turn means we cannot conclude that the 2 b.y. is a real time limit.

## SITES OF DEPOSITION

The third point concerns the sedimentary environment in which the uraniferous sediments were deposited. Were they deposited in streams, in lakes, or in the ocean? The answer seems clear from the evidence at Blind River, from Brazil, and above all from the Witwatersrand that we are dealing with sediments deposited by surface-flowing streams and that the streams flowed into lakes or at least into bodies of water that were shallow and possibly intermittently wet and dry. We are obviously dealing with fluvial systems.

Think back about the geology of the different deposits that were discussed. I suggest that a genetic linkage can be discerned and that the different deposits simply represent different portions of large drainage systems. When Larry Minter spoke he used a simple sketch to demonstrate where, in a long drainage system, the Witwatersrand sediments formed. He drew a stream system which rose in highlands, meandered across a flood plain, then cut across fans and formed a delta. In figure 1, I have indicated the approximate depositional environment for the Witwatersrand conglomerates. The Blind River deposits are similar but with differences. There are no big distal fans and the deposits sit in places on regolith. I suggest that their depositional environment, relative to the Witwatersrand, is as shown in figure 1. The Brazilian deposits seem to be proximal facies. What the basement relations are remains to be seen, and of course the entire section of the Brazilian sediments remains to be exposed. However, I suggest that the Brazilian sediments fit in as shown in figure 1, the Moeda overlapping with the Elliot Lake environment and the Jacobina overlapping with the Witwatersrand fans. For most of the other deposits mentioned the geology was too sketchy to decide where they fit, but

one that probably can be placed is Montgomery Lake, a Canadian deposit with a lot of very well sorted material. It seems to include the distal ends of the fans and to reach beyond the ends of the fans.

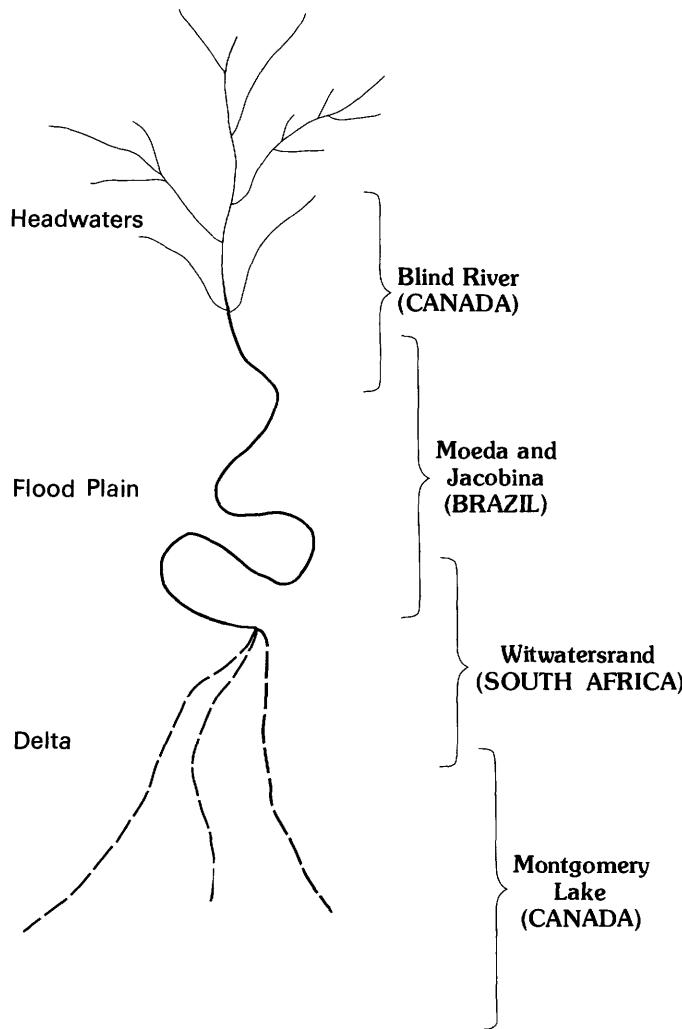


FIGURE 1.—Possible placement in an idealized fluvial system of five uraniferous detrital sediments from Canada, South Africa, and Brazil.

We might ask whether there are modern equivalents of the ancient drainage systems. I would draw an analogy with the drainage system that dispersed the Indonesian alluvial cassiterite during the Pleistocene. The drainage system, now largely drowned because of rising sea level, was at least as large as the systems that must have been operating in the Witwatersrand and Blind River cases. The dispersal of cassiterite in the alluvial deposits can be observed down through the stream system, and a steady decrease in grain size is found. Now marine reworking of the sediments is said to be occurring in places. I suggest there is a clue here for the

uranium explorationist. The specific gravity of uraninite is about 10.5; that of cassiterite, about 7; and that of ilmenite, about 4.7. Both cassiterite and ilmenite are relatively resistant to chemical attack by modern seawater and can be concentrated by marine currents. If uraninite was stable in sea and lake water between 2 and 3 b.y. ago, it, too, could have been concentrated in offshore sands by marine currents. Perhaps we should not focus all of our attention on fluvial systems..

### SOURCE AREAS

My fourth point is that, although the sediment source areas seem to share many things in common, the areas themselves are not unusual. The rock types common in the 3-b.y.-old source areas are greenstones, high-grade metamorphic rocks, and granites. The metamorphic rocks and granites all contain small amounts of uraninite. But there is an unresolved question about grain sizes. The observed sizes of detrital uraninite grains are larger than the grain sizes of primary uraninite grains in most gneisses and granites. So, we have to seek regions where metamorphism has sweated pegmatites and quartz veins out of host rocks and in the process grown relatively coarse grained uraninite. This leads to the conclusion that source areas must all be deeply eroded. Because most quartz-pebble conglomerates are near the bottom of the sediment piles, we must conclude that the tectonic forces that produced the sediment basins on the old cratonic blocks also formed highlands by elevating formerly deeply eroded terrains.

Considerable discussion was devoted to the importance of chemical versus mechanical weathering of the source areas. It seemed to me that the major role played by mechanical weathering was clearly established. The conclusion leads us back to the question of climate and the possibility that all of the uraniferous quartz-pebble conglomerates were influenced by glacial action. That is, we should consider the possibility that all of the source areas were high and that at least the initial rock breakage was mechanical and was due to frost action.

The possible significance of carbon from the sediment source area seemed to me to be a red herring. There is no compelling evidence to suggest that any carbon arrived from the sediment source areas. At least a third of the conference was spent discussing carbonaceous matter, and I have to admit that I remained unconvinced of its importance. The carbonaceous matter is present in the sediments because the primitive plants from which it was derived were happy in the shallow-water depositional environment. It is preserved because the environment is one in which organic matter was rapidly buried. I do not think the case for trapping detrital gold or uraninite in algal mats is very convincing. I was in-

trigued by the discussions that surrounded the importance of carbonaceous matter in diagenetic processes, however, and this leads to my fifth point.

### DIAGENESIS

Once we start dealing with questions of diagenesis we are dealing with a new set of chemical questions, because subsurface waters have different compositions from surface waters. The surface waters could, as we have heard, have been oxygenated, and the transport and deposition of unstable uraninite and pyrite could have been artifacts of slowed-down reaction kinetics. The subsurface waters must have had many sinks for oxygen and as a result were probably reducing relative to the surface waters.

I was impressed with data from the Witwatersrand suggesting that 30 percent of the sulfides were deposited after detrital deposition and that a significant fraction of the uranium has also been similarly deposited or redeposited. I suggest that the most likely time for this to happen was during diagenesis when the relatively porous sediments were bathed in waters that were at least slightly reducing and when primitive bacteria were, in all probability, living off the buried carbonaceous matter. The chemistry of the subsurface waters deserves further discussion and investigation. I suspect they were not too different from the subsurface waters we would find in similar environments today. By this I mean the water would allow the presence of both oxidation states of uranium and, depending on the water composition, variable amounts of  $U^{+4}$  and  $U^{+6}$  would be present. This in turn implies that uranium could move around in subsurface waters, dissolving here, depositing there. There could be other driving forces besides state of oxidation of the water, of course. One important driving force, for example, would be the tendency of small grains to dissolve and be redeposited on larger grains. Diagenetic dissolution and redistribution may possibly be the reason that some detrital minerals seem to be out of hydraulic equilibrium with their enclosing sediments.

### METAMORPHISM AND RECRYSTALLIZATION

All of the uraniferous quartz-pebble conglomerates we see today have been metamorphosed to some extent, and, of course, all of the materials in the deposits can be altered by chemical reactions and are susceptible to recrystallization, which causes changes in grain structure. The mineral on which most of us seem to have underestimated the influence of metamorphism is pyrite. From the information presented at the conference I believe that pyrite is the mineral to study insofar as unravelling metamorphic effects is concerned. Dr. Saager, in particular, seems to be on the right track

in his work on the Witwatersrand. Similar work is needed on other deposits because we do need to understand how materials have been changed and moved around.

What happened to the carbon during metamorphism and also during the earlier phase of diagenesis? Considering the location of thucholite relative to uraninite, it seems clear that some carbon compounds were moving during diagenesis. It is not clear just what it was that moved, however, because the possibility of methane being polymerized to form the compounds we see today seems remote, despite suggestions to the contrary. But even if carbon compounds did move during diagenesis, they have been baked during metamorphism, and we must understand the baking before we can be sure about the ingredients.

The scanning electron microscope images presented by Dr. Hallbauer are spectacular. But I was not convinced he was observing primary biogenic structures. Rather, I suspect he is seeing the products of surface reactions that occurred during diagenesis, or, more likely, during metamorphism. The internal carbon structures he discovered are very likely biogenic in origin, but my bet is that the extraordinary filamentous structures are artifacts of metamorphism.

### DISCOVERING NEW DEPOSITS

My seventh and final point is an answer to that fervent plea, "Where do I find another uraniferous quartz-pebble conglomerate?" If I knew, I would not be standing here, I would be out staking it. But I do think we can pick areas in which probabilities of finding such deposits are greater than in other areas.

I would concentrate on two features in choosing the broad areas to search. First, you obviously have to choose cratonic areas, preferably cratons that have large sedimentary basins formed between 2 and 3 b.y. ago. The easy way to approach this is to consider the sequence of sediments in the basins. A thick clastic section is followed, usually unconformably, by a distinctive chemical sediment we commonly call a banded iron-formation. Use this as a guide. Look at the sediment sections beneath the Lake Superior-type iron-formations. In Brazil the Caué Itabirite overlies the conglomerates; in Witwatersrand, the Transvaal System. But what about the Labrador Trough, Venezuela, Mauretania, Liberia, and Hamersley and Nabberu Basins in Australia, Krivoy Rog and the Kursk regions in Russia, the Indian iron-formations and many others? They all deserve study.

The second obvious question to ask concerns paleogeography. If there is any reality to the involvement of climate, we should be thinking about paleogeography. If cold climate and mechanical weathering are important, then we should look at

cratonic basins, of any age, formed within 30 degrees of the pole. I would, of course, concentrate first on basins existing between 2 and 3 b.y. ago, but I would not limit the search to this specific time. Why? Simply because the apparent younger age cutoff is so uncertain. Mortimer Staatz' observations suggest that, given the right conditions of source area and weathering, detrital uraninite can be of any age. Perhaps we should look at areas affected by extensive glaciations of all ages, even the Permian, provided a reasonable source area and suitable sedimentary basin exists.

#### DISCUSSION FOLLOWING DR. SKINNER'S SUMMATION

*Armstrong:* Incidentally, there are quartz-pebble conglomerates in Australia in the Hamersley area.

*Skinner:* That's the earliest prediction I've ever made that has paid off.

*Breger:* I'm delighted that you mentioned temperature, because we have found in the laboratory that both uraninite and pyrite are completely stable at room temperature even to atomic oxygen. But if you just raise the temperature a little bit, they will decompose. So apparently there is a limitation in temperature range and stability range even in the presence of oxygen that hasn't even been mentioned here.

*Skinner:* Maybe that's why there is so little organic matter left in these things; it was too cold for the organisms to grow anywhere else, except in those infrequent little puddles that were left to the open sun.

*Roscoe:* May I add a note of caution to this concept of temperature being so important, since Dr. Little attributed comments about cold temperature in the Elliot Lake area to me. In the Montgomery Lake area, where there is permafrost to great depth, the oxidation is to great depth, too. There is considerable oxidation to the deepest depth that has been drilled there, whereas at Elliot lake it's an eighth of an inch or so, except along fractures.

*Skinner:* You mean the paleoregolith at Montgomery Lake was several hundred feet?

*Roscoe:* No, on the present surface. Under the present climatic conditions with permafrost, the rock is deeply weathered and leached apparently, as compared to more southerly zones in the Canadian Shield. It's quite surprising.

*Skinner:* I would think that the analogy is perhaps not quite right, that one would need an elevated terrain and the rapid erosion that might come with permanent ice and seasonal thawing. The region that you are talk-

ing about is a highly glaciated one that once had glaciation but doesn't now.

*Roscoe:* No. The point I was trying to make was that, in this case at least, a cold climate has not protected pyrite and other easily oxidizable and leached material, particularly.

*Myers:* I'd like to comment. I thought that Irv Breger said that experiments showed that uraninite and pyrite were stable at room temperature in the presence of oxygen. Was that with water? And I want to go ahead and say that in the Rand both pyrite and uraninite now are not stable at room temperature, and with uraninite they have to be careful in even washing the specimens off, if they are going to analyze them for uranium. Uraninite disappears in a matter of months. The other comment I would like to make, to Brian Skinner, is that you are quite right, it just happens that pyritic conglomerates with both gold and uranium happen to be already known in the Kursk Magnetic Anomaly Series and in the Krivoy Rog Series.

*Skinner:* Anybody else want to fulfill some other predictions?

*Paul Martz, Gulf Mineral Resources Co., Denver, Colo.:* Since you mentioned the Hamersley Basin I just thought that I'd point out an occurrence of quartz-pebble conglomerate there that I think may be of interest to you. It's on the Rocklea Dome, which is near Tom Price and Parrabardu. As I recall, the core of the dome is Archean Warrawoona Volcanics intruded by granites. The core is unconformably overlain by a sequence of Proterozoic quartz-pebble conglomerates and sands that look very much like the pictures on the screen. Stratigraphically above that is the Fortescue Group of volcanics. At the top of the Fortescue are the Pyradie pyroclastics and above that, the Hamersley Group iron-formations. Unconformably overlying the dome is a thick sequence of quartz-pebble conglomerates. However, I never did see any real coarse clasts like some shown in the illustrations; most of the material is much finer grained. The conglomerate unit may be 2,000 to 3,000 feet thick.

*Skinner:* Any uranium?

*Martz:* I wasn't looking for uranium at the time.

*Hallbauer:* I would just like to say that from paleomagnetic measurements a latitude of somewhere between 0 and 30 degrees was calculated for the Precambrian area for the southern part of Africa. That has been published.

*Skinner:* For the Witwatersrand period?

*Hallbauer:* Yes.