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GEOLOGICAL SURVEY'S WORK ON ISOTOPE GEOLOGY OF URANIUM, THORIUM, AND THEIR DECAY PRODUCTS

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GEOLOGY AND MINERALOGY

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GEOLOGICAL SURVEY'S WORK ON ISOTOPE GEOLOGY OF URANIUM, THORIUM, AND THEIR DECAY PRODUCTS*

By R. S. Cannon, Jr.

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ABSTRACT

A program of research on the isotope geology of the uranium and thorium series is being carried on by the Geological Survey. Work is in progress on uranium-lead relationships in uranium ores of the Colorado Plateau region; on uranium-thorium-lead relationships in granite; on geologic variations in the isotopic composition of lead; and on radon and helium in natural gas. A continuing program of systematic studies will try to establish methods in this field on a surer footing and to apply the methods to the solution of important geologic and mineral-resource problems.

INTRODUCTION TO THE PROBLEM

I want to tell you about the Survey's research in a field that we are calling "the isotope geology of uranium, thorium, and their decay products". This field concerns the study and interpretation of the quantitative relations in geologic materials of the various nuclear species that belong to the uranium, actino-uranium, and thorium radioactive families. The Geological Survey has a natural interest in the whole field of isotope geology — but it has a compelling reason right now for special interest in this particular segment of isotope geology. Many practical problems that our geologists face daily in searching out and evaluating our mineral resources can be solved, we believe, by systematic research on

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the isotope geology of these radioelements. Probably you will all agree that the results of such research are of special importance to the search for potential sources of radioactive raw materials.

The scientific objectives of our work in this field are broadly two-fold. One major objective is to estimate the date of certain geologic events — as for example the date of formation (or geologic age) of uraniferous rocks or minerals. A second objective of equal interest and importance to us is to use the abundance relations of the several nuclear species of the uranium and thorium series as evidence of the nature of the various geologic processes in which they have been involved. For example, we hope to be able to determine from the isotopic composition of lead in an ore deposit whether this particular batch of lead came from some deep source below the earth's crust, or from average sialic material of the crust, or from sialic material with an unusually rich content of uranium, or from sea water by precipitation in the marine sediments of some earlier geologic period. Or, as a quite different example, we may hope to determine whether uranium has been leached from a certain uraniferous rock by alkali waters in the zone of weathering.

In theory we are interested in relationships between all the nuclear species of the 3 principal radioactive families, but in geological practice we are most concerned with those that have relatively long half-lives, that occur in significant abundance in geological materials, and for which adequate analytical techniques have been developed. These specifications focus our attention on the three radioactive parent isotopes; on a few radioactive daughters—such as thorium^{230}, radium^{226}, and radon^{222} and on the stable end-products—the stable isotopes of helium and lead.

The raw materials for this research include the rocks, minerals, and natural fluids that contain one or more of these isotopes. The essence of the problem is to interpret the ratios between daughter and parent isotopes in these materials. Three general types of daughter-parent relationships can be cited to illustrate the scope of the problem.

Two types of relationships concern the association of daughters with parents in materials like uranium minerals. Some uranium minerals have behaved almost like closed systems in which radioactive equilibrium has been continuously maintained and the stockpiling of decay products has proceeded without interruption. Such materials have traditionally been sought for lead-uranium age work. In the strictest sense closed systems of this sort do not exist in nature, yet many uranium and thorium minerals have been found in which parent and daughter isotopes are very nearly in balance.

Many uranium minerals, on the other hand, have not behaved as closed systems since they were formed. Commonly there has been some loss or escape of decay products. In other cases daughter isotopes may have been introduced from outside the system, or parent isotopes may have been either lost or introduced. Thus in radioactive materials the ratios of radiogenic lead isotopes to parental uranium and thorium isotopes are commonly not consistent, and radiogenic helium is generally deficient. These departures from a state of balance between parent and daughter isotopes complicate determinations of geologic age but they also serve to reveal what sort of geologic processes have affected the materials.

A third category includes geologic concentrations of daughter isotopes that are now disassociated from their parents — that is, accumulations of decay products outside the geologic bodies in which they were generated.

These can be accumulations of radioactive daughters—like accumulations of radon in natural gas, or like radium-bearing barite. Or they can be accumulations of stable end-products—such as helium-bearing natural gas, or the radiogenic isotopes of lead in base-metal ores.

To investigate the isotope geology of these materials successfully we need skills and special knowledge from many fields. As a geologist, I am especially aware of the contributions that geology must make toward planning a wise program of investigation; toward selecting, collecting, and preparing the most suitable of sample materials; and toward the geologic phases of interpreting and applying results, I am, however, keenly aware that the other physical sciences must contribute also to planning and interpretation and must likewise carry the brunt of laboratory investigations. Accurate quantitative measurements, by chemical or physical methods, are needed for at least a half dozen elements (U, Th, Ra, Rn, He, and Pb) in a wide variety of materials. Accurate mass-spectrum measurements are needed of the isotope ratios of lead, of uranium, and of helium. And the tools of mathematical analysis are needed to decipher some of the complexities of our data. Because so many unrelated skills are needed to solve each problem, coordination of effort in this field of investigation is of paramount importance. If we can succeed in integrating all the requisite skills into a coordinated attack, we shall obtain a fund of new knowledge on the date and mode of origin of radioactive materials; on the geologic history of such materials since their birth; on the earlier geologic adventures of certain of the elements; and on some of the more general problems of earth processes, earth structure, and early earth history.

PREVIOUS INVESTIGATIONS

Much work has already been done during the past 40 years or so in the field I have defined—most of it for purely scientific reasons. Perhaps best known to geologists are the lead-uranium age investigations of exceptionally pure and unaltered samples of uranium and thorium minerals. A considerable amount of work has also been done investigating helium-uranium relations in ordinary igneous rocks. With the rapid development of the mass-spectrometer in the 1930's studies were started of the isotopic composition of uranium, of helium, and of lead. In the latter case there have been studies both of "associated" radiogenic lead in uranium minerals, leading to better understanding of lead-uranium age problems, as well as studies of "disassociated" lead in base-metal ores.

The Survey made no systematic studies in this field prior to World War II but did make some contributions that were helpful to other workers. These contributions include high-quality analyses of uranium and thorium minerals, especially by Hillebrand; studies of the geology of helium, by Rubey and others; and contributions to the work of the Committee on the Measurement of Geologic Time of the National Research Council.

During the past decade progress in this field has not seemed commensurate with the growing importance of uranium, even when due allowance is made for unpublished results. When the Survey started intensive work on the geologic occurrence of uranium and thorium in the United States, during the war, the need for much better understanding of geologic relationships between uranium and thorium and their daughter products was soon apparent. It did not prove possible to undertake systematic research on these relationships to support our exploratory program until some years after the war.

when a first step was taken as one phase of a study of the mineralogy of the carnotite ores of the Colorado Plateau. Since then, several further projects in this field have been undertaken, and recently a start has been made toward coordinating these several investigations in a research program sponsored by the Atomic Energy Commission.

PRESENT SURVEY PROGRAM

The Survey's present program on the isotope geology of uranium, thorium, and decay products includes 4 projects on which planning or active work has been in progress for several years. In brief, these projects concern lead-uranium relationships in carnotite and other ores of the Colorado Plateau region; lead-uranium-thorium relationships in granites; variations in the isotopic composition of lead; and the occurrence of radon and helium in air and natural gas.

The investigation of lead-uranium relationships in uranium deposits in and adjacent to the Colorado Plateau by L. R. Stieff and T. W. Stern, and in primary uranium ores of the Colorado Front Range by George Phair, is an intensive application of these methods to our most productive uranium province. This work is yielding new evidence to help solve the riddle of the date and mode of formation of the carnotite ore bodies. Quantitative data have been obtained on the content of uranium and the isotopes of lead in about 40 samples of carnotite and several vein uraninites. About 40 more samples from carnotite ores and a dozen from other types of deposits are in process of analysis. This work is perhaps the most intensive study that has yet been made of lead-uranium data on uraniferous materials from a single province, but it is more noteworthy as the first major attempt to use materials that by all accepted criteria are decidedly inferior for such work. The data on carnotite ores reveal a lack of balance between the uranium and lead isotopes not unlike the anomalous data from the Swedish kolm. Pb^{206}/U^{238} ratio ages are around 75 million years, and Pb^{207}/Pb^{206} ratio ages around 400 million years. Evidence has not yet clearly shown whether this anomaly is related to loss of radon from the ores, or to introduction of old radiogenic lead at the time of ore formation. It is worthy of mention that ores from the Shinarump conglomerate of Triassic age are yielding lead-uranium data indistinguishable from those on ores from the Morrison formation of Upper Jurassic age; and that Pb^{206}/U^{238} ratio ages for the carnotite ores are not greatly different from those for Early Tertiary pitchblende of the Colorado Front Range. Though it would be premature to try to draw a firm conclusion at this stage, we are optimistic that the final results of this study will point to a specific answer.

Investigations of the isotope balance in radioactive minerals of granites are being made by E. S. Larsen, Jr, and J. B. Mertie, Jr. The determination of the geologic age of ordinary igneous rocks from their lead-uranium-thorium relationships promises to become a usable method within the near future. Professor Larsen is one of the pioneers primarily responsible for this achievement. His work on the geochemistry of the mineral components of the Southern California batholith led him head-on into this problem some years ago. Before he retired from the faculty at Harvard he collaborated with Keevil and others in attempting age determinations on his meticulously prepared monomineralic concentrates. More recently, collaborating with Harrison Brown and others, he split a pre-Cambrian granite from the Haliburton area, Ontario, into monomineralic fractions which Brown is analyzing by his isotope dilution methods. The preliminary results of this work, as reported by Brown several days ago, are most encouraging.

Professor Larsen is now continuing work on suites of igneous rocks from the Southern California batholith and is completing the preparation of mineral concentrates for analysis for lead, uranium, and thorium. J. B. Mertie has prepared concentrates of monazite and zircon from the monazite-bearing granites of the Southeastern United States for similar analysis.

The investigation of variations in the isotopic composition of lead in geologic materials is my own hobby. But Stieff and Stern have made the Survey's major contribution of new data in this field to date, with isotope investigations of a suite of about 50 samples of lead extracted from various materials from the Colorado Plateau region. Their data show an orderly range in the composition of lead from carnotite deposits, from nearly pure uranogenic lead of certain carnotite samples to common lead or ore-lead in samples of galena from carnotite deposits.

In regard to our more general investigation of the isotope geology of lead, planning of the program and collection of suitable sample materials have been in progress for 4 years, but our laboratory investigations are just beginning. The first few isotope analyses, made several months ago, are of rather general interest. These half dozen samples were cut from successive growth zones of a single cube of galena to determine whether the isotopic composition of lead is homogeneous throughout a single crystal of galena as has been tacitly assumed in prior work. The data show that the lead in this crystal is not homogeneous but varies rather systematically with larger proportions of the radiogenic isotopes in the outer or younger growth layers. The total variation in this single crystal is considerable, for it is roughly 10 percent as large as the total variation that Nier found in 25 samples of lead from lead minerals from widely different sources. This information has rather amazing geologic implications and it also serves to emphasize the complexity of the problems of geologic sampling of natural materials for isotope investigations. It is a pleasure to acknowledge the support of the Oak Ridge Office of Research and Medicine (AEC) and the cooperation of the Mass Assay Laboratory of the Y-12 Plant (ORNL) in providing these isotopic analyses.

A fourth active project, the investigation of radon and helium in natural gases, by Gott, Faul, Manger, Sakakura, and others will be discussed by Faul.

FUTURE PROGRAM

The Geological Survey's plans for future work on the isotope geology of the uranium and thorium series are concerned first of all with carrying out the projects now in progress. Some of the current projects have limited scope, like the work on Colorado carnotites, whereas others will be of longer duration. We view our work on isotope relationships in igneous rocks, on the isotopic composition of lead, and on radon-helium in natural gases as long-term investigations, though we may not always be able to formulate detailed work plans far in advance. As we conclude successive phases of this work, we hope to modify our program into a more concerted attack on the basic problems of this field.

We have outlined in a proposal to the Atomic Energy Commission and again in Trace Elements Investigations Report 201 many phases of research in isotope geology that we judge to be needed. Some of these are problems that we especially wish to attack with the diversified geologic talents of the Survey staff. There are many problems on which we could collaborate with other investigators to mutual advantage. And there are other important problems on which we would like to encourage research by other scientists or institutions that possess special skills or interests.

The nature of our own future program will depend in part on our success in achieving optimum coordination with other investigators in this field. But we can state two basic considerations that will greatly influence our plans in any event. One is our recognition of the need to establish lead-uranium isotope investigations on the firmest possible foundation as rapidly as possible. We need better analytical techniques, and we need to re-examine critically some of the physical and geologic assumptions on which lead-uranium age methods are now based. The ultimate goal can be figuratively stated as a really accurate lead-uranium time-scale and a reliable correlation of this time-scale with the stratigraphic and paleontologic record.

The second consideration is that we will try to direct our research toward investigations that appear to promise results of optimum usefulness to the Survey's varied geologic activities. As most of the Survey's geologic work is designed to implement — either directly or indirectly — the appraisal and discovery of the nation's mineral resources, we are going to show greatest interest in those problems of the isotope geology of the uranium and thorium series that are clearly important to the investigation of critical mineral raw materials. This objective requires both basic research to establish our methods on a firm scientific footing as well as specific investigations of geologic chronology and geologic processes with practical objectives in key situations.