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Geology of Uranium in Coaly Carbonaceous Rocks

GEOLOGICAL SURVEY PROFESSIONAL PAPER 356-D

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By JAMES D. VINE

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UNITED STATES DEPARTMENT OF THE INTERIOR

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GEOLOGICAL SURVEY

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URANIUM IN CARBONACEOUS ROCKS

GEOLOGY OF URANIUM IN COALY CARBONACEOUS ROCKS

By JAMES D. VINE

ABSTRACT

Though the association of uranium with coaly rock near Denver, Colo., was first reported in 1875, very little was known about the relation of uranium to coaly rocks until the decade 1946-56. In 1954 and 1955 private prospecting in the Fort Union lignite region of western North and South Dakota resulted in the discovery of many thousands of tons of coaly carbonaceous rock containing more than 0.1 percent uranium.

Coaly carbonaceous rocks characteristically contain non-marine indigenous humic plant remains that accumulate in swamps. An arbitrary classification by percent ash is used in this report to distinguish coal, impure coal, coaly shale, and carbonaceous shale. Uranium is only one of many inorganic constituents of these rocks. Five general classes of inorganic constituents are: inherent, diagenetically fixed adventitious, detrital free adventitious, secondary free adventitious, and epigenetically fixed adventitious. The principal inherent elements include 15 essential to plant growth. The high percentage of silicon, aluminum, and iron in coal ash as compared to plant ash is due chiefly to the addition of acquired or adventitious mineral matter. Diagenetic alteration of coal results in a loss of manganese, calcium, magnesium, sodium, and potassium as compared to the original plant substance. Nearly every element except the inert gases and a few of the rare earths has been identified in coal. Though some of the minor elements may be inherent, some, such as germanium, are locally far too abundant and erratically distributed to be explained by other than epigenetic enrichment.

The average abundance of uranium in coaly carbonaceous rocks may normally be less than 0.0001 percent. Coaly carbonaceous rocks that contain at least 0.005 percent uranium are classed as uranium deposits. The known such deposits in the United States occur in five general rock types: unconsolidated peaty material, lignite and subbituminous coal, bituminous and anthracitic coal, impure lignite and lignitic shale, and carbonaceous shale.

The uranium in alpine-meadow peaty soils is evidently introduced continuously by spring waters and becomes concentrated in the living plants and especially in the decaying plant debris that accumulates in the meadow soil.

Lignite and subbituminous coal in the Rocky Mountain and Great Plains regions contains a number of large deposits of uranium, but the tenor for large tonnages rarely exceeds 0.015 percent. At least 10 deposits of this type having a total of more than 90 million tons of lignite are known in the Fort Union lignite region of western North and South Dakota. The Red Desert area of the Great Divide Basin, Wyoming, also

contains a very large reserve of uraniferous subbituminous and impure coal, but the average tenor of uranium is low.

Bituminous and anthracitic coal in the United States is not known to contain more than a few small, low-grade deposits of uranium. In the bituminous coal fields of the United States the highest concentrations of uranium are commonly found in the marine black shale that overlies coal beds or in the underclay directly below the coal beds.

At least 14 areas of impure lignite and lignitic shale in the United States contain economically significant deposits of uranium. In the Fort Union lignite region the principal deposits occur along the high eastern escarpment of the Little Missouri badlands in Billings County, N. Dak., and in the Cave Hills and Slim Buttes highlands in Harding County, S. Dak. Most of these deposits occur below thick sandstone beds in thin beds of lignitic rocks that have been weathered to soft earthy material and contain about 50 percent moisture. At least one deposit is in relatively fresh woody lignite. The deposits occur throughout a stratigraphic interval of nearly 1,000 feet. Rocks of Oligocene and Miocene age that contain abundant volcanic tuff unconformably overlie the deposits in South Dakota. Impure coal at La Ventana Mesa in Sandoval County, N. Mex., in the Gas Hills area, Fremont County, Wyo., and at the Old Leyden coal mine north of Golden, Colo., contains economically significant uranium deposits.

Carbonaceous-shale beds in the Goose Creek area, Cassia County, Idaho, in the Fall Creek area, Bonneville County, Idaho, and at the west end of the Zuni uplift near Gallup, N. Mex., contain uranium deposits.

Some types of coaly carbonaceous rocks are far more favorable host rocks for uranium than others. Unconsolidated peaty material is capable of being greatly enriched in uranium, but only rarely are deposits found in peat. Lignite and subbituminous coal have the capacity to take up abundant uranium, but the physical properties of these rocks are apparently less favorable for concentrating uranium than are those of impure lignite and of lignitic shale. Bituminous and anthracitic coals are generally unfavorable host rocks for uranium. Carbonaceous shale seems to be a chemically favorable but a physically unfavorable host rock.

The regional geology and tectonic setting can be related only vaguely to the distribution of uranium deposits. A regional unconformity may predispose an area as large as the Fort Union lignite region as a favorable setting, yet also by truncation of any single bed may cause selective enrichment of uranium in favorable host rocks. This relation exists throughout the Fort Union lignite region, which contains nearly half of

the deposits in the United States regarded as economically significant.

Uranium is concentrated locally in or adjacent to permeable formations and beds and in permeable zones within the host rock. Many deposits in the Fort Union lignite region are less than 300 feet below the restored base of Oligocene and Miocene rocks that contain abundant volcanic material. The volcanic material is regarded by some geologists as the probable source for uranium.

There is no evidence, other than the presence of the uranium deposits, that a uranium-rich province exists in the Fort Union lignite region. What has been called a province is probably merely a region where conditions and processes favorable for the concentration of uranium have been prevalent during the geologic past.

Many types of ground water may transport uranium. The structural position, size, shape, and type of an epigenetic uranium deposit is largely determined by whether the mineralizing ground water is thermal, artesian, unconfined in saturated ground, or vadose.

Other types of uranium deposits may be associated with those in coaly carbonaceous rocks. Because host-rock lithology is the basis for distinguishing uranium deposits in coaly carbonaceous rocks, genetically diverse types are included, each of which is associated with a different type of related deposit. Some uranium deposits in sandstone may consist of redeposited and hardened uraniferous humic acids that were extracted from coaly carbonaceous matter.

The physical and chemical form of uranium in coaly carbonaceous rocks is obscure because uranium minerals are rarely visible and never account for all the metal present. The amount of inherent uranium in these rocks is generally insignificant. Even where plants grow in a uraniferous environment the sorption of uranium on decaying plant materials far exceeds the contribution from living organisms. The contribution of uranium from detrital minerals is also insignificant. Locally, an appreciable amount of uranium is in the form of epigenetic uranium minerals, but most of the uranium in deposits in coaly carbonaceous rocks is in the form of fixed adventitious uranium.

A marked local variation in the abundance of uranium not clearly related to variable rock composition is characteristic of deposits in coaly carbonaceous rocks. Uranium is directly associated with carbonaceous matter but shows no pronounced preference for specific carbonaceous rock constituents except possibly for layers of translucent attritus containing abundant amorphous humic matter and yellow waxy matter.

Chemical tests show that, though three-fourths of the uranium in an unweathered coaly shale sample was in the hexavalent state, a large part of the uranium might occur as uraninite. The uranium in coaly carbonaceous rocks even in unweathered material is not volatile and is relatively insoluble in water and organic solvents but is readily soluble in strong acids or alkalis. Mild air oxidation of lignite may make possible the removal of some of the uranium by extraction of humic acids. The original sorption of uranium by peat, lignite, and humic acids apparently is an ion-exchange process that is effective even at the relatively low concentrations of uranium found in nature. Uranium is not lost during the early stages of the weathering of lignite. One study has shown a positive correlation between uranium and the concentration of calcium, magnesium, and iron, but there is reason to doubt the existence of a simple direct relation.

The role of coaly carbonaceous rocks in the geochemistry of uranium is known only in general terms and many questions remain imperfectly answered. Because coaly carbonaceous rocks are such effective agents for concentrating uranium, many other economically significant deposits in these rocks might be expected. Only those low-rank carbonaceous materials that have a chemical structure related to humic acids are commonly associated with uranium. Peat probably has the greatest capacity to take up uranium, but in the normal peat-bog environment uranium is rarely available. Because broad areas of coaly carbonaceous rocks are uniformly exposed to uranium-bearing waters in adjacent permeable units, epigenetic deposits are characteristically thinner and of lower grade than deposits in sandstone where mineral deposition can be localized. Epigenetic uranium in coaly carbonaceous rocks is probably directly related to the concentration of uranium available in solution, whereas the uranium contained in sandstone deposits is probably not related directly to the availability of uranium in solution as much as to the effectiveness of some mechanism for preprecipitation. The secondary uranium minerals found in weathered lignite may be deposited by evaporation of capillary pore moisture. Uranium is concentrated in the marine environment at the time of sediment accumulation, but the coaly carbonaceous rocks continue to concentrate uranium after burial of the sediment; thus, the role of uranium concentration in coaly carbonaceous rocks and marine black shale tends to be complementary.

The geochemical enrichment of uranium in coaly carbonaceous rocks is the result of cation exchange on humic or coaly carbonaceous matter, a unique geologic process which uranium does not share with any of the major elements in a sedimentary environment. The association of uranium-bearing solutions with low-rank humic or coaly carbonaceous matter required to form a deposit is a relatively rare geological phenomena that follows no easily predictable pattern.

INTRODUCTION

SCOPE OF THE REPORT

Much data has been collected in recent years regarding the host-rock characteristics, the distribution of deposits, the geologic relations, the physical and chemical form of occurrence, and the geochemistry of uranium in coaly carbonaceous rocks. This information is scattered through a number of reports, some published but others not publicly available. It is the purpose of this report to make a critical evaluation of the geologic data available to the U.S. Geological Survey as of June, 1958, that pertains to the occurrence of uranium in these rocks. This work was done on behalf of the Division of Raw Materials, U.S. Atomic Energy Commission.

This report on the geology of uranium in coaly carbonaceous rocks is one in a series of topical reports intended to provide a critical analysis of present knowledge about uranium deposits by types of deposit rather than by individual localities. Consequently, descriptions of individual deposits are scattered through many sections of the report. The

reader seeking reference to geographic areas of uranium-bearing coaly carbonaceous rocks or summary descriptions of these areas should refer to the index rather than to the table of contents. The reader desiring more comprehensive descriptions of localities will generally find references to other published reports.

The introduction defines the types of deposits to be discussed and provides a historical summary of their discovery and development. The next section deals with the general features of normal, nonuraniferous coaly carbonaceous rocks including the average uranium content of these rocks as a class. The discussion of uranium deposits summarizes the distribution of deposits according to five principal classes of coaly carbonaceous rocks and includes a discussion of these deposits as they are related to specific geologic features. The next two sections examine the information pertaining to the physical and chemical form of the uranium and to the role of coaly carbonaceous rocks in the geochemistry of uranium.

Brief reference is made to several prevailing hypotheses of origin of the uranium in the section on the relation of deposits to geologic features; the writer's preference is emphasized but no attempt is made to summarize all ideas. Similarly, no attempt is made to summarize the data pertaining to reserves and resources of uranium in coaly carbonaceous rock because no method of economic recovery of uranium from these rocks is yet known. Tabulations of reserves of coaly carbonaceous rock classified by percent of contained uranium are available in other published reports.

The writer is greatly indebted to many of his colleagues in the Geological Survey, for their countless suggestions regarding the organization and scope of material in this report and for their almost daily technical advice and criticisms.

DEFINITION OF URANIUM DEPOSITS IN COALY CARBONACEOUS ROCKS

As used in this report the expression "coaly carbonaceous rocks" refers to any sedimentary rock characterized by indigenous carbonaceous matter that consists predominantly of the various altered remains of vascular land plants. More specifically, the term "coaly carbonaceous rocks" includes the following four principal varieties listed in sequence of increasing proportion of carbonaceous to mineral matter: carbonaceous shale, coaly shale, impure coal, and coal. Additional varieties include lignitic shale, carbonaceous sandstone, and peat. The discussion of uranium deposits in peat is included in this report because of the genetic similarity between peat and coal

though, strictly speaking, peat is not a rock. More detailed descriptions and definitions of the rock varieties and related descriptive and petrographic terms as used in this report are given on pages 117-123.

Any natural concentration of uranium in a coaly carbonaceous rock may be regarded as a uranium deposit in the geologic (though not necessarily in the economic) sense. To simplify the discussion that follows, it was arbitrarily decided to use the value of 0.005 percent uranium, as determined by chemical analysis, as the lower limit for all deposits described in this report. As of July 1959, none of the deposits described in this report were being mined for uranium at a profit; thus, the term "deposit" is not used in the sense of an ore deposit, which by definition means a deposit that can be mined at a profit.

HISTORICAL SUMMARY

The association of uranium with coaly carbonaceous rock was first reported by Berthoud (1875) as a result of his examination of the Old Leyden coal mine a few miles northwest of Denver, Colo. His interpretation of a silicified bed of coal as a dike intruded into the sedimentary strata was a subject for debate at that time, but the identification of uranium as a major metallic constituent was little more than a geological curiosity. The mine was reexamined by Wilson (1923), who reported the occurrence of carnotite in the coal and considered it a possible source for radium.

Early investigations of the radium content of some coal beds and the carbonaceous rocks associated with the coal (table 1) are of historical interest in showing the unusually low average uranium content of these rocks.

The uranium contained in various ranks of coal, peat, and vegetable matter from central Europe were determined by direct chemical (fluorometric) analyses (Hoffman, 1943a). These data indicate an increase in uranium content from about 0.000003 per-

TABLE 1.—*Early estimates of the uranium content of coal and associated carbonaceous rocks*

Material	Number of samples	Locality	Percent uranium ¹ (average)	Source of data
Coal.....	11	Alabama.....	0.00005	Lloyd and Cunningham (1913).
Do.....	5	France.....	.00005	Moureu and Lepape (1914).
Do.....	5	England.....	.00001	Drakeley and Smith (1922).
Do.....	9	Donetz basin, Russia.	.00002	Burkser, Shapiro, and Bronshtein (1929).
Shale and underclay.	8	do.....	.0004	Do.
Coal.....	35	Kuznetz basin, Russia.	.000003	Burkser, Kondorugi, Kapustin, and Potapov (1934).

¹ Calculated from reported values for radium content using the formula: Uranium = 2.84×10^6 radium (Keevil, 1944, p. 313).

cent in dry vegetable matter to about 0.0001 percent in peat, 0.0003 percent in lignite, and as high as 0.0007 percent in anthracite. The uranium content of coal from Río Turbio, Argentina, was reported by Lexow and Maneschi (1950) to be about 0.000004 percent. The precision of these data is subject to question, however, inasmuch as values of less than 0.0001 percent have not been duplicated in fluorometric analyses in the Geological Survey laboratory.

Coaly carbonaceous rocks generally contain less uranium than other sedimentary rocks, not, apparently, because of any unfavorable characteristic as a host rock for uranium, but probably because the uranium content of the carbonaceous matter is normally so low that the carbonaceous matter serves to dilute the uranium contained in the extraneous mineral matter. Where soluble uranium is available to coaly carbonaceous rocks, they may be greatly enriched in uranium. The possibility that these rocks may contain abundant uranium locally became evident in 1945 when A. L. Slaughter and J. M. Nelson (written communication, 1946) of the Geological Survey discovered a large deposit of radioactive coal and coaly shale of Eocene age in the Red Desert of the Great Divide Basin, Sweetwater County, Wyo. Information was received in 1947 by the British government that coal was being mined for uranium in the Freital district near Dresden, in the Russian-occupied zone of Germany (Davidson and Ponsford, 1954, p. 268). In 1948, D. G. Wyant and E. P. Beroni (written communication, 1949) of the Geological Survey discovered abnormally radioactive lignite in the upper part of the Fort Union formation of Paleocene age in western North Dakota. The limited distribution of uranium suggested some kind of syngenetic control. In 1949, Beroni and H. L. Bauer, Jr., (written communication, 1952) of the Geological Survey found uranium-bearing lignite in northwestern South Dakota in the middle and lower parts of the Fort Union formation. The following year, further detailed studies were made by N. M. Denson, G. O. Bachman, and H. D. Zeller (written communication, 1950), also of the Geological Survey. As more data accumulated it became evident the uranium was epigenetically deposited.

The results of these studies inspired a general reconnaissance search by the Geological Survey for uranium-bearing coaly carbonaceous rocks in 1951. Four new areas were found during this reconnaissance: (a) uranium-bearing coaly and carbonaceous shale of Early Cretaceous age at Fall Creek in southeastern Idaho (Vine and Moore, 1952b); (b) uranium-bearing lignitic and carbonaceous shale of Pliocene

age at Goose Creek in southern Idaho (Duncan, 1953, p. 9-10); (c) uranium-bearing impure coal and coaly shale of Late Cretaceous age at La Ventana in New Mexico (Bachman and Read, 1952, p. 10); and (d) uranium-bearing coal and impure coal associated with uranium deposits in sandstone of Eocene age near Pumpkin Buttes in northeastern Wyoming (Love, 1952). These discoveries showed that uranium-bearing coaly carbonaceous rock is widely distributed both geographically and stratigraphically. By 1958, more than 100 deposits in 18 States had been located.

Meanwhile uranium-bearing coal was reported by Foldvari and Szalay in Hungary in the vicinity of the Mecsek and Velence Mountains (Szalay, 1954). Though the exact date of their discovery is not mentioned, it is presumed to be prior to 1951.

Interest in coaly carbonaceous rocks as a potential source for uranium fluctuated widely. Following the Red Desert discovery in 1945, considerable interest was shown in the possibility that large tonnages of slightly uraniferous coal might be found from whose ash uranium might be extracted as a byproduct of some large-scale industrial utilization of the coal. However, interest in all low-grade resources of uranium began to diminish about 1952. Meanwhile, most geologists investigating uranium-bearing coaly carbonaceous rocks placed emphasis on determining reserves of uranium in commercial-quality coal and paid only passing attention to impure coal and coaly shale. Random samples whose ash contained more than 0.1 percent, and even as much as 1 percent, uranium were reported from the Old Leyden coal mine in Colorado, the Cave Hills and Slim Buttes areas in South Dakota, the Fall Creek and Goose Creek areas in Idaho, and the La Ventana Mesa area in New Mexico, but these reports attracted little attention. Few geologists seriously considered that coaly carbonaceous rocks might contain uranium of sufficiently high grade to constitute ore. As a result, there was little publicity and little private prospecting of regions known to contain uraniferous coaly carbonaceous rocks until the public release in May 1954 of Gill's (1953a) description of carnotite in the Slim Buttes area of South Dakota. Soon thereafter private aerial radioactivity surveys resulted in the discovery of several deposits containing more than 0.1 percent uranium in thin beds of impure lignite in the Cave Hills and Slim Buttes areas of Harding County, S. Dak. (Gill, 1954; Burton, 1955). Publicity resulting from these discoveries touched off a wave of prospecting and land acquisition throughout western North and South Dakota and eastern Montana.

By January 1957, several shipments as large as 500 tons each of impure lignite and lignitic shale containing more than 0.1 percent uranium were made for metallurgical testing. Exploration drilling by private interests demonstrated the existence of a large reserve of similar grade rock but as of July 1959 no further development had taken place.

The commercial extraction of uranium from coal, even from deposits containing more than 0.1 percent uranium, may be partially dependent on the byproduct recovery of other useful materials. Haught (1954, p. 21) suggested that germanium, gallium, beryllium, silver, and mercury are sufficiently valuable and abundant in the ash of some coal to be worth extracting. Molybdenum is commonly associated with uranium in impure lignite and lignitic shales from the Dakota area (Denson and Gill, 1956, p. 416) and should be considered as a possible byproduct. J. T. Burnett (written communication, 1956) in discussing the commercial possibilities for the extraction of uranium from coal suggested gas for heat, ammonia, and various organic chemicals as other byproducts; he concluded that the economic recovery of uranium may not be possible unless all or part of the available byproducts are recovered.

GENERAL CHARACTERISTICS OF COALY CARBONACEOUS ROCKS

ORIGIN AND CLASSIFICATION OF CARBONACEOUS MATTER

Broadly used, the term "carbonaceous" may include limestone and related rocks composed chiefly of carbonate minerals; however, these are more commonly and precisely classified as carbonate rocks, and the term "carbonaceous" is used for rocks containing organically derived carbon in the elemental form or in chemical combination with hydrogen and other elements, such as oxygen, nitrogen, sulfur, and phosphorus, normally present in living organisms. Consequently, the expression "carbonaceous sedimentary rocks" generally denotes rocks characterized by their content of original organic tissues, their decomposition products and residues, or their fluid derivatives and extracts. It is in this restricted sense of organic chemical composition that the term "carbonaceous" is used throughout this report. Varieties of carbonaceous rock may be distinguished on the basis of (a) the proportions of mineral and carbonaceous matter, (b) the time of emplacement of the carbonaceous matter relative to the surrounding mineral matter (that is, whether the carbonaceous matter is indigenous or was secondarily emplaced), (c) the origin and type of organic tissues (whether plant or animal, land or marine organisms) which produced the car-

bonaceous matter, and (d) the degree of alteration of the organic matter, a quality generally determined by specific physical and chemical tests.

The preservation of carbonaceous matter in sedimentary rocks requires special environments of deposition. One such environment (the so-called paludal environment) is where plant growth is relatively rapid and the availability of oxygen to fallen plants is restricted by a cover of stagnant water as in a peat bog or swamp. Peat, coal, and carbonaceous shale generally form in a paludal environment. A second such environment is where plants and other organisms that live in poorly aerated marine water die and settle to the bottom where they may be preserved under a thin layer of stagnant water too toxic for life for other than anaerobic microorganisms. Black marine mud and black shale generally form in this environment. Rapid burial by younger sediment is also an important factor in the preservation of the carbonaceous material in both these environments.

Because carbonaceous matter is of variable composition, color, hardness, and texture, and is not crystalline in structure, it does not fit the usual concept of a mineral. It is therefore convenient to regard the terms "carbonaceous" and "mineral" in the complementary sense that for any given rock the sum of the carbonaceous and mineral matter is equal to the whole (moisture-free) rock.

In describing carbonaceous rocks, "inorganic" is a term related to but more inclusive than mineral matter. The inorganic constituents of a carbonaceous rock are the sum of the elements present in the discrete mineral matter plus the noncombustible (chiefly metallic) elements chemically bound to the carbonaceous matter. The term "inorganic" is most useful in describing the elemental composition of a carbonaceous rock as a whole when lacking knowledge of how the elements are distributed between the carbonaceous matter and the mineral matter. It is important to remember, however, that the terms "organic" and "inorganic" are not used in the complementary sense in which the terms "carbonaceous" and "mineral" are used. For example, many carbonate rocks have an organic origin even though the rock is composed of inorganic matter.

The percent ash residue, or ash, is the percent by weight of the rock that remains as an inorganic residue after ignition of the combustible substances, as determined by definite, prescribed methods. Thus, ash is closely related to both mineral matter and inorganic constituents but is not identical with either. During the process of ashing, volatile elements appropriately considered as part of the inorganic con-

stituents, such as selenium and germanium, may be lost and discrete minerals such as carbonates and sulfides may lose volatile products.

The scheme of classification for carbonaceous matter devised for this report may be helpful in the understanding of carbonaceous uranium deposits. It distinguishes coaly carbonaceous matter from other varieties of carbonaceous matter and forms the basis in this report for identifying and naming specific varieties within the family of coaly carbonaceous rocks.

By careful examination of the rock textures, it is generally possible to determine whether the carbonaceous matter accumulated with other rock-forming materials and is therefore indigenous or was introduced after lithification. This should be regarded as the first basis of subdivision of carbonaceous matter. If indigenous, the carbonaceous and mineral matter should have a clastic texture that is readily visible except perhaps in very fine grained rocks. If secondarily introduced, the carbonaceous matter will form a coating on mineral grains and fill interstices between grains. In figure 28, a diagram that shows the generalized relation among types of carbonaceous materials, coaly matter falls within the triangle depicting indigenous carbonaceous matter.

If the carbonaceous matter is indigenous, it is distinguished by composition and origin of identifiable plant tissues into two general types, humic and sapropelic. Carbonaceous materials are generally a mixture of these two types; however, carbonaceous material is described as humic if composed predominantly of vascular (woody) plant tissues, in contrast to spores, algae, resins, and waxes, which are dominant in sapropelic carbonaceous material. Though this distinction is commonly made by visual examination of the rock,

the fundamental difference is based on the chemical composition, which is a function both of the original plant types and the environment (pH and Eh) during decomposition of the plants. Humic matter is characterized by abundant oxygen, whereas sapropelic matter is characterized by a low oxygen and high hydrogen content. Both types grade imperceptibly into each other as well as into a third type which is high in carbon and low in both hydrogen and oxygen. These three chemical types are represented as forming the end members of the triangular diagram (fig. 28) depicting varieties of indigenous carbonaceous matter. Though the carbonaceous material of coaly carbonaceous rocks generally falls on the right half of the triangle and that of marine black shale generally falls on the left half, they overlap, for some coal is sapropelic and much marine mud is humic. Thus, paleontologic evidence as to the marine or non-marine environment of deposition of the plant material is also helpful in distinguishing coaly carbonaceous rocks of paludal origin from similar marine rocks.

Gradations and mixtures of different types of carbonaceous matter are common. Coaly carbonaceous matter differs from sapropelic derivatives including bitumen by having a clastic texture and abundant oxygen. Because of chemical similarity, coaly carbonaceous matter is distinguished from humic extracts only on the basis of clastic texture. Coaly carbonaceous material is distinguished from marine carbonaceous material partly on the basis of the higher carbon to hydrogen ratio of the former and partly on the geologic interpretation of the environment of deposition.

SUBDIVISIONS

Once it has been established that the carbonaceous matter in a rock is indigenous and of paludal (in contrast to marine) origin and thus belongs to the family of coaly carbonaceous rocks, a rock name may be assigned on the basis of the proportion of carbonaceous to mineral matter and on the degree of compaction, of biochemical alteration, and of dynamochemical alteration or metamorphism of the carbonaceous matter. These changes follow a pattern that for coal is termed "changes in rank." The standard system used in the United States for the classification of coal by rank (table 2) published by the American Society for Testing Materials (1955) is intended primarily for commercial coal but provides a useful classification for the whole range of coaly carbonaceous rocks. The four main classes of coal distinguished according to this system are: anthracitic, bituminous, subbituminous, and lignitic. No criteria

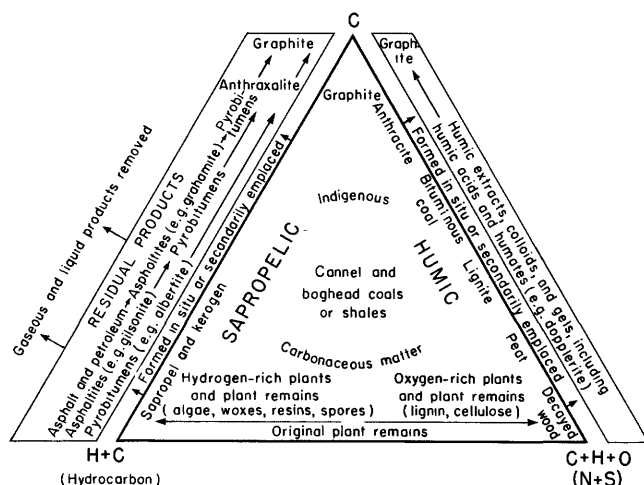


FIGURE 28.—Diagram showing generalized relations among the types of carbonaceous materials. After Vine, Swanson, and Bell (1958).

TABLE 2.—*Classification of coals by rank*

Explanation: FC, fixed carbon; VM, volatile matter; Btu, British thermal units.
This classification does not include a few coals which have unusual physical and chemical properties and which come within the limits of fixed carbon or Btu of the high-volatile bituminous and subbituminous ranks. All of these coals either contain less than 48 percent dry, mineral-matter-free fixed carbon or have more than 15,500 moist, mineral-matter-free Btu.

[After American Society for Testing Materials (1955)]

Class	Group	Limits of fixed carbon or Btu mineral-matter-free basis	Requisite physical properties
I. Anthracitic.....	1. Meta-anthracite. 2. Anthracite..... 3. Semianthracite.	Dry FC, 98 percent or more (dry VM, 2 percent or less). Dry FC, 92 percent or more and less than 98 percent (dry VM, 8 percent or less and more than 2 percent). Dry FC, 86 percent or more and less than 92 percent (dry VM, 14 percent or less and more than 8 percent).	Nonagglomerating. ¹
II. Bituminous ² ...	1. Low-volatile bituminous coal. 2. Medium-volatile bituminous coal. 3. High-volatile A bituminous coal. 4. High-volatile B bituminous coal. 5. High-volatile C bituminous coal.	Dry FC, 78 percent or more and less than 86 percent (dry VM, 22 percent or less and more than 14 percent). Dry FC, 69 percent or more and less than 78 percent (dry VM, 31 percent or less and more than 22 percent). Dry FC, less than 69 percent (dry VM, more than 31 percent); and moist ³ Btu, 14,000 ⁴ or more. Moist ³ Btu, 13,000 or more and less than 14,000. ⁴ Moist Btu, 11,000 or more and less than 13,000. ⁴	Either agglomerating or nonweathering. ⁵ Both weathering and nonagglomerating.
III. Subbituminous.	1. Subbituminous A coal. 2. Subbituminous B coal. 3. Subbituminous C coal.	Moist Btu, 11,000 or more and less than 13,000. ⁴ Moist Btu, 9,500 or more and less than 11,000. ⁴ Moist Btu, 8,300 or more and less than 9,500. ⁴	
IV. Lignitic.....	1. Lignite..... 2. Brown coal.....	Moist Btu, less than 8,300. Moist Btu, less than 8,300.	Consolidated. Unconsolidated.

¹ If agglomerating, classify in low-volatile group of the bituminous class.

² It is recognized that there may be noncaking varieties in each group of the bituminous class.

³ Moist Btu refers to coal containing its natural bed moisture but not including visible water on the surface of the coal.

⁴ Coals having 69 percent or more fixed carbon on the dry, mineral-matter-free basis shall be classified according to fixed carbon, regardless of Btu.

⁵ There are three varieties of coal in the high-volatile C bituminous coal group, namely, variety 1, agglomerating and nonweathering; variety 2, agglomerating and weathering; variety 3, nonagglomerating and nonweathering.

are established for the identification of peat according to this system, though peat is generally recognized as the initial stage in the process of coalification.

An arbitrary system of classification of coaly carbonaceous rock providing for specific names was adopted for use in this report. Three general categories, based on the degree of consolidation or the rank, are recognized: peat, lignite, and coal. Additional categories corresponding to the higher rank coals recognized by the classification of coals in table

2 generally have not been required. Because the percent ash has been determined for many of the samples involved in this study, ash is arbitrarily used as a basis of further classification in lieu of a more precise measure of mineral matter. Four categories of ash content are recognized:

1. 68.1 percent or more ash.
2. At least 31.6 but less than 68.1 percent ash.
3. At least 14.7 but less than 31.6 percent ash.
4. Less than 14.7 percent ash.

These four categories of ash content are selected because the percentages 14.7, 31.6, and 68.1 represent the midpoint values (reduced to three significant figures) for the bracketed intervals in which certain semiquantitative spectrochemical analyses are reported. The numbers are based on a logarithmic progression that has special application to the spectrochemical analyses of ashed coaly carbonaceous rocks. Furthermore, the categories are convenient in a general classification because the percentages are sufficiently close to $\frac{1}{10}$, $\frac{1}{3}$, and $\frac{2}{3}$ that these fractions may be used for a field classification. The classification and names for specific rock types are shown on figure 29.

The definition of coal given in figure 29 is not intended to supplant the more general definitions such as those by Tomkeiff (1954, p. 38) and Schopf (1956, p. 527). Schopf's definition is quoted here for its contribution to an understanding of coaly carbonaceous rocks: "Coal is a readily combustible rock containing more than 50 percent by weight and more than 70 percent by volume of carbonaceous material, formed from compaction or induration of variously altered plant remains similar to those of peaty deposits. Differences in the kinds of plant materials (type), in degree of metamorphism (rank), and range of impurity (grade), are characteristic of the varieties of coal." In a diagram that accompanies this

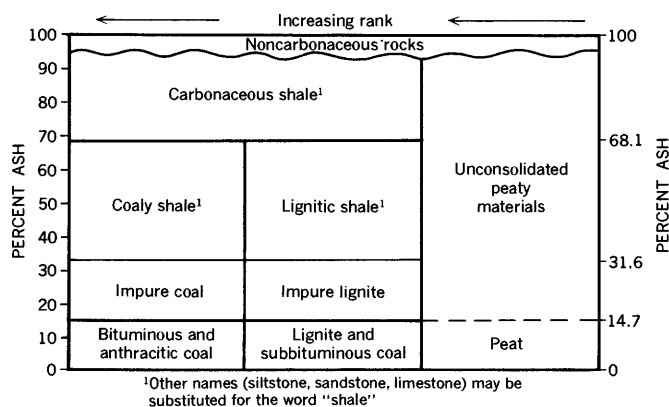


FIGURE 29.—A classification of coaly carbonaceous rocks.

definition Schopf indicated a range of commercial coal and a range of impure coal but did not define the boundary between them. Thus, if an arbitrary boundary is placed at 14.7 percent ash, there is no conflict between his definition and the one used in this report. The only real conflict is in restricting the range of impure coal to rocks containing less than 31.6 percent ash, which is well below the 50 percent by weight or 70 percent by volume of carbonaceous matter suggested in Schopf's definition. This restriction seems justified for the purposes of this report because rocks covering the whole range in composition of carbonaceous and mineral matter are considered.

PETROGRAPHIC FEATURES

Because coaly carbonaceous rocks are characterized by petrographic features not found in the descriptions of mineral rocks, mention will be made of some of the more frequently used special terms. The definitions of terms are modified after Schopf's instructions (written communication, 1951) for describing coal, but are useful also for describing any megascopically visible indigenous carbonaceous matter. The terms included are taken from both the European and American nomenclature (Stopes, 1919; Thiessen, 1920, p. 185).

Vitrain.—The ingredient in banded coal that has a vitreous or brilliant luster, a conchoidal fracture, and (in bituminous coal) a close cleating is called vitrain. Isolated lenses of vitrain are common in shale and sandstone. Single lenses of vitrain usually represent individual fragments of woody trunk, root, or bark. Morphologic features of the original plants are commonly visible in thin section and may be visible to the unaided eye in low-rank carbonaceous material. Vitrain usually contains less mineral matter and yields less ash than other carbonaceous constituents. It is generally the most chemically reactive of the major carbonaceous constituents. Vitrain is translucent red or orange in thin section. By definition, vitrain is composed of megascopically visible fragments, in contrast to anthraxylon which includes much material of smaller size. The dense woody lenses in lignite that are clearly equivalent to the vitrain of higher rank carbonaceous material are called previtrain. It is distinguished from vitrain by having less luster and less tendency to be cleated. It is tough and woody in texture.

Attritus.—The microfragmental matrix of carbonaceous material that occupies the interstices between coarser carbonaceous ingredients is called attritus.

Attritus forms the dull or satiny-lustered bands that alternate with vitrain in banded carbonaceous material. It is formed from a wide variety of finely divided plant debris, some of which may be identifiable as spores or pollen in thin section, but it consists mostly of unidentifiable plant fragments. The luster of attritus is intermediate between vitrain and fusain. The chemical and physical properties of attritus vary greatly depending on the types of original plant debris and the degree of their alteration. Translucent and opaque varieties of attritus are readily distinguished in thin section.

Fusain.—The carbonaceous material that forms dull black bands so soft and friable that it readily soils the fingers is called fusain. It is most conspicuous in bituminous coal but is occasionally abundant in lower rank carbonaceous materials. Both the appearance and chemical composition of fusain are similar to charcoal. In banded coal, fusain generally forms less than 10 percent of the total carbonaceous material, but as isolated fragments in shale or sandstone fusain may form the entire lens. Fusain generally has a minute fibrous or porous structure derived from the empty cellular cavities of the original plant materials. Because of this porous structure, fusain may contain considerable secondary mineral matter but is in itself relatively inert. Fusain is opaque in thin section.

Banded and nonbanded carbonaceous matter.—The alternating laminae of dull attritus and lustrous vitrain commonly visible in commercial coal is called banding. Banded carbonaceous matter is generally an indication of humic composition. Nonbanded carbonaceous matter, such as cannel and boghead coal, is generally attrital in origin, has a dull or satiny luster, and is of sapropelic composition.

INORGANIC CONSTITUENTS

An understanding of the distribution, the physical and chemical form of occurrence, the relation of uranium to other trace elements, and the geochemistry of uranium in coaly carbonaceous rocks requires an understanding of the ways uranium and other inorganic constituents may occur in these rocks. Uranium is only one of many elements in carbonaceous rocks and is generally only a minor inorganic constituent. To understand the complex interrelations between the carbonaceous matter and the inorganic constituents and between different types of inorganic constituents some knowledge of plant nutrition, botany, geochemistry, mineralogy, and organic chemistry is desirable. It is the purpose of this section to bring together enough background information on the gen-

eral subject of inorganic constituents in coaly carbonaceous rocks to place in proper perspective the occurrence of uranium in these rocks.

Coaly carbonaceous rocks grade from rocks that are predominantly carbonaceous with almost no visible mineral matter to rocks with only enough carbonaceous matter to impart a characteristic dark-brown or black color. Even seemingly pure coal, however, contains enough inorganic matter to yield ash on ignition, as might be expected because living plants similar to those that form coal contain from 0.1 to 3 percent inorganic matter (U.S. Bureau of Mines, 1954, p. 50). It is desirable, therefore, to distinguish between the inorganic matter that is chemically fixed or combined with the carbonaceous matter and the free mineral matter that was introduced into the deposit by sedimentary processes.

CLASSIFICATION

The free mineral matter and the inorganic constituents of carbonaceous rocks that are chemically fixed to the carbonaceous matter are commonly referred to as the extraneous and the intrinsic inorganic components, respectively. It is not necessarily true, however, that all the elements that are chemically fixed to the carbonaceous matter are derived from the mineral nutrients of the parent plants. Reactions between ground-water solutions and the decomposing plant material in a peat bog or after burial of the peat deposit may introduce new elements that become chemically fixed to the carbonaceous matter. Similarly, not all the extraneous mineral matter is derived from clastic detritus washed or blown into the peat bog. Other mineral matter may precipitate

TABLE 3.—*Classification of inorganic constituents in coaly carbonaceous rocks*

Syngenetic constituents

1. Inherent inorganic matter derived from the mineral nutrients of the parent plants.
2. Diagenetically fixed adventitious inorganic matter derived from solutions from which certain elements are chemically combined with the carbonaceous matter during the process of plant decay and accumulation.
3. Detrital free adventitious mineral matter derived from silt and mud introduced into the peat bog at the time of plant decay and accumulation.

Epigenetic constituents

4. Secondary free adventitious mineral matter derived from solutions that deposited mineral matter along joints and in pore spaces after consolidation.
5. Epigenetically fixed adventitious inorganic matter derived from solutions from which certain elements are chemically combined with the mineral or carbonaceous matter after consolidation.

from solutions either during peat accumulations or after burial. For purposes of geochemical interpretation, therefore, at least five classes of inorganic constituents in coaly carbonaceous rocks are distinguished according to the manner of origin and time of deposition. The classification of inorganic constituents (table 3), modified after Francis (1954, p. 483-509), is particularly useful in discussing the mode of occurrence, the geochemistry and the origin of uranium in carbonaceous rocks.

PRINCIPAL INHERENT ELEMENTS

In addition to the carbon, hydrogen, and oxygen that plants derive from air and water, they also derive seven essential elements from the soil: nitrogen, phosphorus, potassium, calcium, magnesium, sulfur, and iron. Five other elements known as micronutrients are also essential, though in much lower concentration, to the growth of most plants. These are boron, zinc, copper, manganese, and molybdenum (Arnon, 1953, p. 318-324). Five additional elements, aluminum, sodium, titanium, silicon, and chlorine, are present in most plants but are not necessarily essential to their metabolic processes. Many other elements may also be present in plants, dependent partly on the availability of various elements in the soil and partly on the particular species of plant, for some plants are capable of concentrating certain elements in excess of that in the soil. The distribution of some plants is largely dependent upon the distribution and availability of certain elements in the soil. For example, the growth of some species of *Astragalus* is dependent on the availability of selenium in the soil and the plants are capable of concentrating selenium in the tissue (Trelease and Beath, 1949, p. 121-164). These selenium-indicator plants, as they are known, have been the subject of considerable study because they are sometimes poisonous to livestock and because of their usefulness in geobotanical prospecting for uranium (Cannon, 1954). Similar highly selective absorption and concentration of other rare elements by living plants is uncommon.

MAJOR INORGANIC CONSTITUENTS

Coal ash contains many of the same elements that are absorbed by plants in far different proportions. From a study of many analyses of coal ash, Selvig and Gibson (1956), p. 32) suggested typical limits for the principal constituents of the ash of bituminous coal in the United States (see table 4).

Of the principal elements of coal ash, silicon, aluminum, and titanium are not essential to the growth of plants. Phosphorous is generally less concentrated

TABLE 4.—*Typical limits of the principal constituents of coal ash compared with those of plant ash*

Constituent	Suggested typical limits (percent) in—	
	Coal ash ¹	Plant ash ²
SiO ₂	20-60	0.7-70
Al ₂ O ₃	10-35	-----
Fe ₂ O ₃	5-35	.6-1.7
CaO.....	1-20	8-36.9
MgO.....	3-4	1.8-16.4
TiO ₂5-2.5	-----
Na ₂ O.....	1-4	.6-4.1
K ₂ O.....		8.5-34.8
SO ₃		3.5-7.9

¹ After Selvig and Gibson (1956, p. 32).² As indicated by four analyses reprinted by Haught (1954, p. 21).

in coal ash than in plant ash. Thiessen (1945, p. 486) reported that the oxides of aluminum, silicon, iron, calcium, and magnesium constitute more than 95 percent of most coal ash. These elements are also major constituents of common sedimentary rocks and their occurrence in coal ash is probably attributable chiefly to the presence of free mineral matter. Thus, from the abundance of major rock-forming elements in coal ash as well as the fact that most coal contains a far greater percentage of inorganic constituents than living or dead plants, it is clear that the adventitious mineral matter in coal generally exceeds the inherent inorganic matter derived from plant nutrients. Impure coal, coaly shale, and carbonaceous shale must therefore have a progressively smaller proportion of the total inorganic matter derived from plant nutrients.

DIAGENETIC ALTERATION

The process of plant accumulation and decay is accompanied by changes in the composition of the inherent inorganic matter as well as in the carbonaceous matter (Francis, 1954, p. 485). Katchenkov (1952) and Haught (1954, p. 21) show that the ash of coal of various ranks contains a smaller proportion of soluble elements than the ash of wood. Water-soluble constituents such as the chlorides of sodium, potassium, and magnesium are leached, and a concentration of some of the less soluble constituents has resulted. Thus, coal ash is comparatively richer than plant ash in silicon, aluminum, and iron and poorer in manganese, calcium, magnesium, sodium, and potassium. (The examples of lignite ash given by Haught (1954, p. 21) contain more calcium and sodium than either the higher rank coals or the plant ash.) The behavior of nitrogen and sulfur compounds from plant proteins is complex and depends on the environmental conditions of the deposit. Sulfur released during the hydrolysis and decomposition of plant proteins is commonly reduced to hydrogen sulfide which readily reacts with iron and other metallic

ions present to form nodules, lenses, or finely disseminated sulfides such as pyrite and marcasite.

MINOR INORGANIC CONSTITUENTS

In addition to the major constituents of coal ash, many rare elements are concentrated in the ash of some coals. V. M. Goldschmidt was one of the first to become interested in the minor-element composition of coal ash (Goldschmidt and Peters, 1933; Goldschmidt, 1935, 1937, 1950). In 1930 germanium that was in sufficient concentration to be of possible commercial value was discovered in British coal from the Hartley district (Goldschmidt, 1935). As a result of this discovery and the development of the spectrographic technique for identifying small amounts of the rare elements, investigations were made in many countries in a search for rare and trace elements in crude oil, asphalt, and coal. In Russia, for example, an extensive search resulted in the discovery of unusual quantities of vanadium (Zilbermintz, 1935) and germanium (Zilbermintz, Rusanov, and Kostrikin, 1936) in some coals. By 1944 the literature pertaining to the occurrence of uncommon chemical elements in coal was so extensive that Gibson and Selvig (1944) prepared a summary in which they described many occurrences of the following elements: arsenic, barium, boron, beryllium, chlorine, chromium, copper, fluorine, germanium, gallium, gold, iodine, lead, zinc, manganese, mercury, molybdenum, nickel, phosphorous, radium, selenium, silver, titanium, uranium, and vanadium. Since then nearly every element but the inert gases and a few of the rare earths has been identified in coal ash.

Though a biochemical source may be adequate to explain the origin of some rare elements in coal ash, Goldschmidt (1950, p. 244) recognized that it may not be adequate to explain the unusually high concentration of germanium in certain coal ashes. Particularly puzzling was his discovery that in different samples of coal from the same bed the germanium content varied inversely with the ash content, a relation to be expected of an element that occurs as part of the inherent ash but not for adventitious mineral matter such as germanium. However, he suggested three possibilities whereby germanium and other rare elements might be concentrated in coal; (a) concentration during the life of the plant; (b) concentration during decay of the organic substances; and (c) concentration after the plant debris has been buried under sediments, by reaction of the coal or associated minerals with circulating aqueous solutions containing rare elements. Other workers soon discovered that the germanium was concentrated particu-

larly in the top or bottom few inches of certain beds of coal. In order to explain this anomalous distribution of germanium, Headlee and Hunter (1951, p. 11, 12) suggested that the organic matter of coal may absorb germanium from the water or gaseous solutions that enter the coal during the later stages of metamorphism. Research on trace elements in British coal has shown (Reynolds, 1948; Aubrey, 1952) that not only germanium but also gallium, vanadium, chromium, titanium, nickel, copper, and barium are concentrated in the top and bottom few inches of some coal seams, particularly in narrow bands of vitrain that overlie the main seams. A study of germanium in United States coals by Stadnichenko and others (1953) confirmed the inverse relation of germanium to ash and the usual enrichment of germanium at the top or bottom of a bed of coal or adjacent to a parting; it also produced evidence of a much greater concentration of germanium in vitrain than in fusain.

Thiessen (1945, p. 495) noted that because of its colloidal nature coal is highly absorptive and would tend to absorb mineral matter from water that leached it from other beds. Katchenkov (1952) observed that where accumulation of some elements in coal ash shows an enrichment factor of hundreds or thousands of times more than average abundance in the earth's crust, such an accumulation must be primarily associated with the circulation of aqueous solutions. He further suggested that many elements are extracted from these solutions through absorption, by reducing action of coal, and by the precipitation of sulfides of heavy metals. The quantity of minor elements that Horton and Aubrey (1950, p. S 42) found associated with the pure coal substance of certain vitrains is greater than they were willing to designate simply as part of the composition of the original plants. They noted the pronounced base-exchange properties of brown coal and the probability that coal would absorb metallic ions from solution.

Table 5 suggests criteria for distinguishing the various classes of inorganic constituents in coaly carbonaceous rocks.

TABLE 5.—Criteria for classifying inorganic constituents in coaly carbonaceous rocks

Criteria	Suggested interpretation (see table 3)
1. Uniform lateral and vertical distribution of an element or constituent for any given lithology.	Syngenetic.
2. Inverse correlation between constituent and ash.	Chemical combination with carbonaceous matter.

TABLE 5.—Criteria for classifying inorganic constituents in coaly carbonaceous rocks—Continued

Criteria	Suggested interpretation (see table 3)
3. Concentration of constituent in light fraction of mechanically separated rock.	Do.
4. Constituent elements known to be essential plant nutrients (N, P, K, Ca, Mg, S, and Fe); plus small amounts of B, Zn, Cu, Mn, and Mo; plus variable amounts of elements commonly found in plants (Al, Na, Ti, Si, and Cl).	Inherent.
5. Higher concentration of constituent in attritus and fusain than in vitrain.	Deposition from solution during plant decay and accumulation.
6. Petrographically identifiable mineral matter.	Detrital sediment or deposited from solutions.
7. Concentration of constituent in heavy fraction of mechanically separated rock.	Do.
8. Constituent has a normal clastic textural relation with other rock constituents.	Detrital sediment.
9. Direct (positive) correlation between constituent and ash content.	Free adventitious mineral matter.
10. Concentration in attritus but not in fusain or vitrain.	Detrital sediment.
11. Erratic vertical and lateral distribution of constituents (for example, concentration at top or bottom of bed, or along structural features).	Epigenetic.
12. Mineral matter discordant to clastic texture (for example, interstitial pore fillings, joint fillings, replacement bodies, or discordant nodules or concretions).	Do.
13. Higher concentration of mineral matter in fusain than in less porous carbonaceous constituents.	Do.
14. Constituent is one of the commonly authigenic minerals (for example, the carbonates and sulfates of calcium and magnesium plus the oxides of iron, plus quartz and clay minerals).	Do.
15. Constituent is associated with vitrain and attritus but not with fusain.	Chemically combined with carbonaceous matter after consolidation.
16. Constituent occurs in much higher concentration than is normal for rocks of the same type.	Do.

AVERAGE ABUNDANCE OF URANIUM

Precise chemical analyses of random samples of coaly carbonaceous rocks are not available on which to base an estimate of the average or normal abundance of syngenetic uranium in the family of coaly carbonaceous rocks. The files of the Geological Survey contain several individual uranium analyses for carbonaceous rocks in which the results are reported to the nearest 0.0001 percent. Almost invariably, however, the samples analyzed were selected for some reason such as high radioactivity as compared with adjacent rocks so that the results do not give the average or normal abundance of uranium.

Studies of radioactivity in sedimentary rocks for use in interpretation of gamma-ray logs of bore holes has provided much additional information on the relative radioactivity of coal and other sedimentary rocks. Results of a study by Russell (1945) show that, whereas carbonaceous marine rocks such as black shale are among the most radioactive sedimentary rocks, the comparable fresh-water carbonaceous rocks such as coal are among the least radioactive sedimentary rocks. The correlation of gamma-ray logs with sample cuttings from bore holes led workers in Great Britain (Davidson and Ponsford, 1954, p. 266) to the same conclusion, that coal in general is characterized by very low radioactivity. Newmarch (1950, p. 143) found an increase in radioactivity with an increase in ash content of the coal from the Crowsnest coal basin of British Columbia, and suggested that the small quantity of radioactive elements are present as components of the adventitious mineral matter and not as inherent or fixed constituents of the carbonaceous matter.

A search from 1952 to 1954 for radioactive coals in the Eastern Interior and Appalachian coal provinces of the United States resulted in the collection of the

most nearly representative samples available for estimating the normal or average uranium content. The samples were collected for the most part without regard to radioactivity. This sampling contrasts sharply with that of coal in the Great Plains and Rocky Mountain States where coal beds are commonly exposed almost continuously for miles; anyone examining those beds with radiation detection equipment has tended to collect samples only where radioactivity anomalies are found.

A summary tabulation of radioactivity and uranium data for coal in the Eastern United States (tables 6 and 7) shows that of 442 coal localities examined, coal samples from 404 localities were reported to contain less than 0.001 percent equivalent uranium. Coal at only four localities, including two separated by only a few hundred feet, contains as much as 0.005 percent equivalent uranium. Coal at 34 additional localities contains as much as 0.001 but less than 0.005 percent equivalent uranium. Of a total of 38 localities where coal was found to contain 0.001 percent or more equivalent uranium, chemical uranium analyses have been reported for 15. Seven of these showed less than 0.001 percent uranium. From this data it is apparent that the normal or mean uranium content of coal from the 442 localities examined is probably considerably less than 0.001 percent and may be even less than 0.0001 percent.

Coal samples from other provinces may contain slightly more or less syngenetic uranium than the group described above, depending on the availability of uranium at the time of deposition. In the family of coaly carbonaceous rocks, those containing a higher ratio of mineral to carbonaceous matter than in coal probably have a proportionately higher uranium content because the normal quantity of uranium in mineral matter is higher than in organic remains.

TABLE 6.—Summary of localities in Eastern United States where coal has been examined for radioactivity and uranium

Area, field, region, or strata investigated	Localities examined				Source of data
	Total	Where coal contains 0.005 percent or more eU	Where coal contains 0.001-0.005 percent eU	Where coal contains <0.001 percent eU	
Bituminous coal region of Pennsylvania.....	108	1	2	105	Patterson (1954c).
Coal of Pennsylvanian and Permian age in northern West Virginia.....	16	0	4	12	Patterson (1954b).
Coal in Ohio.....	37	0	3	34	Snider (1953a).
Indiana coal field.....	47	0	5	42	Snider (1954).
Southern West Virginia and southwestern Virginia.....	30	0	1	29	Snider (1953b).
Eastern Pennsylvania anthracite field.....	46	0	1	42	Welch (1953b).
Eastern Kentucky coal field.....	60	0	5	55	Welch (1953a).
Beaver, Clearfield, and Jefferson Counties, Pa.....	57	2	3	52	Ferm (1955).
Southern Illinois.....	29	1	4	24	Patterson (1954a).
Western Kentucky.....	12	0	3	9	John Huddle and E. D. Patterson (written communication, 1954).
Total.....	442	4	34	404	

¹ Does not include 13 additional localities where coal was estimated from outcrop radioactivity to contain 0.001 percent equivalent uranium.

² The two localities are about 400 feet apart.

TABLE 7.—Analyses of radioactive coal samples from localities in the Eastern United States

Area, field, region, or strata investigated	Bed	Interval sampled		eU (per-cent)	Uranium (per-cent)	Ash (per-cent)	Locality description	Source of data
		Position in bed	Thickness (feet)					
Bituminous coal region of Pennsylvania.	Lower Freeport	Top	0.1	0.033	0.041	11.9	Krolick Coal Co. strip mine at LeContes Mills.	Patterson (1954c).
	Upper Freeport	Bottom	.5	.001			Abandoned strip mine at LeContes Mills.	Do.
	Lower Freeport	Top	1	.001			Gibson Coal Co. strip mine 3 mi east of Ashville.	Do.
Coal of Pennsylvanian and Permian age in northern West Virginia.	Dunkard	Top	.5	.002			Roadcut north of Mannington.	Patterson (1954b).
	do	do	.4	.001			Roadcut east of Hundred.	Do.
	Pittsburgh	Top	.3	.002			Keely Construction Co. strip mine north of West Milford.	Do.
Coal in Ohio	do	Bottom	1	.003			Scott Coal Co. strip mine east of McWhorter.	Do.
	Pittsburgh No. 8	Top	6	.003	.0001	10.7	Bradford Coal Co. mine at Crescent.	Snider (1953a).
	Brookville No. 4	Top	0.5	.001	.0010	33.3	Broken Aro Coal Co. mine east city limit of Latrobe.	Do.
Indiana coal field	Pittsburgh No. 8	do	.33	.001			Greasy Ridge Coal Co. mine south of Greasy Ridge.	Do.
	Minshall	do	3.7	.001			Mulzer Mines Co. abandoned mine 3.7 mi southeast of Gentryville.	Snider (1954).
	Gentryville	Top	1.2	.002	.0012	20.4	Roadcut 1.8 mi south of Gentryville.	Do.
Southern West Virginia and southwestern Virginia.	Coal V	Top	.5	.002			Tecumseh Coal Corp. mine 6 mi southeast of Lynnville.	Do.
	do	do	.7	.002	.002	11.1	A.B.C. Coal Co. mine 3 mi west of Linton.	Do.
	do	do	.7	.002			Tecumseh Coal Corp. mine 2.5 mi southeast of Scalesville.	Do.
Eastern Pennsylvania anthracite field.	Merrimac	Bottom	1.75	.001			Northside Coal Co. mine 1.5 mi west of Blacksburg.	Snider (1953b).
	Buck Mountain	do	2.33	.001			Carbon County strip mine 0.4 mi east of Tresckow.	Welch (1953b).
	Lykens Valley No. 2	Bottom	2	.001			Northumberland County abandoned mine 2.5 mi northwest of Mount Carmel.	Do.
Eastern Kentucky coal field	No. 6 coal	Top	2	.001			Schuykill County roadcut 1 mi northeast of Frackville.	Do.
	Buck Mountain	Bottom	1	.001			Schuykill County abandoned mine 0.2 mi south of Delano.	Do.
	Elkhorn coals (2 beds).	do		.001			Republic Coal Co. mine 5 mi southeast of Elkhorn City.	Welch (1953a).
Beaver, Clearfield, and Jefferson Counties, Pa.	"C" coal	do	4.67	.001(?)	<.001		International Harvester Mines 0.8 mi east of Benham.	Do.
	Creech	do	.83	.001(?)	<.001		Blue Diamond Coal Co. mine 5.5 mi southeast of Harlan.	Do.
	Blue Gem	do	1.67	.002(?)	<.001		B. R. Campbell & Son Coal Co. mine, McCreary County.	Do.
Southern Illinois	Lily (cannel)	Top	.75	.001			Laurel County truck mine.	Do.
	Lower Freeport	Bottom	.25	.005	.007	17.0	Roadcut on State highway 51 1.5 mi southeast of Darlington.	Ferm (1955).
	do	do	.5	.010	.010	16.9	Prospect on State highway 51 1.5 mi southeast of Darlington.	Do.
Western Kentucky	Mercer	do	1.17	.001			Roadcut on State highway 28 0.6 mi east of Summerville.	Do.
	C	Top	.75	.001	.0004	62.3	Strip pit 1.8 mi southeast of Keewaydin.	Do.
	Mercer	Middle	.5	.002	.0006	31.9	Strip pit 0.7 mi south of Chestnut.	Do.
Southern Illinois	do	Bottom	.58	.002	.0006	25.1	do	Do.
	Herrin (No. 6)	Top	1	.008	.0085	6.8	Sahara Coal Co. mine No. 6, sec. 31, T. 9 S., R. 5 E.	Patterson (1954a).
	Harrisburg (No. 5)	do	1	.002			B. & W. Coal Co. mine, sec. 12, T. 9 S., R. 8 E.	Do.
Western Kentucky	Herrin (No. 6)	do	1	.001			Republic Coal & Coke Co., sec. 21, T. 9 S., R. 4 E.	Do.
	Harrisburg (No. 5)	do	.7	.001			Shawneetown Coal Co. mine, sec. 8, T. 10 W., R. 9 E.	Do.
	Herrin (No. 6)	do	.8	.001			Reinheimer slope mine, sec. 32, T. 1 S., R. 7 W.	Do.
Western Kentucky	No. 9	Bottom	.8	.002	.001		Stoney Point Coal Co. mine 4.5 mi south of Providence.	John Huddle and E. D. Patterson (written communication, 1954).
	No. 14	do	.2	.001			Chandler Bros. mine 1 mi northwest of Walton Creek Church.	Do.
	No. 9	Top	.5	.0020			Drill core 3 mi southeast of Zion.	Do.

DISTRIBUTION OF URANIUM DEPOSITS IN COALY CARBONACEOUS ROCKS

It has been shown that the normal syngenetic uranium content of some coals is considerably less than 0.001 percent and may even be less than 0.0001 percent. Data concerning abnormal concentrations, but less than 0.005 percent or 50 ppm (parts per million) uranium, are few because it has been common laboratory practice in the Geological Survey to make chemical determinations for uranium only on samples having a radioactivity of 0.005 percent or more equivalent uranium. Concentrations of 0.005 percent or

more uranium are generally easy to detect by their anomalous radioactivity; therefore a content of 0.005 percent uranium is used as an arbitrary cut-off value for consideration of a deposit in the following discussion. Table 8 is a listing of 113 deposits, localities, or areas in the United States from which one or more samples have been collected that meet the following specifications:

1. The uranium-bearing rock named in the report, or its description as nearly as can be interpreted, indicates that it belongs to the family of coaly carbonaceous rocks.

TABLE 8.—*Deposits of uranium in coaly carbonaceous rocks in the United States*

[Deposit type: 1, unconsolidated peaty material; 2, lignite and subbituminous coal; 3, bituminous and anthracitic coal; 4, impure lignite and lignitic shale; 5, carbonaceous shale. Asterisk (*) indicates an estimated reserve of more than about 1,000 tons]

Locality			Stratigraphic unit	Age	Deposit type	Source of data
No. (pl. 1)	Name	Description				
Arkansas						
1	Owens lease.....	Sec. 12, T. 13 N., R. 15 W.....	Atoka.....	Pennsylvanian.....	5	E. P. Beroni, H. S. Stafford, and J. F. Foran, written communication, Nov. 1955.
California						
2	Hoffman Meadow area.....	Sec. 13, T. 6 S., R. 25 E.....	Unnamed.....	Quaternary.....	*1	W. E. Bales and G. M. Haselton, written communication, Nov. 1955.
3	Chiquito Creek area.....	Secs. 17, 19, 29, 30, 33, T. 6 S., R. 24 E.....	do.....	do.....	1	U.S. Atomic Energy Commission, written communication, Dec. 1956.
4	Fireflex (Monterey) mine.....	Sec. 21, T. 17 S., R. 10 E.....	Unknown.....	Tertiary.....	2	Moore and Stephens (1954, p. 5).
5	Pettit Ranch meadow.....	Sec. 26, T. 26 S., R. 31 E.....	Unnamed.....	Quaternary.....	*1	W. A. Bowes, W. E. Bales, and G. M. Haselton, written communication, 1957.
6	Caliente Creek area.....	Sec. 20, T. 30 S., R. 33 E.....	do.....	do.....	1	U.S. Atomic Energy Commission, written communication, Feb. 1957.
7	Good property.....	Sec. 36, T. 8 N., R. 18 W.....	Peace Valley.....	Pliocene.....	4	W. A. Bowes, and J. R. Tillman, written communication, 1954.
8	Beer Can No. 12 claim.....	Sec. 2, T. 4 N., R. 24 W.....	Sespe.....	Eocene and Oligocene.....	5	U.S. Atomic Energy Commission, written communication, Nov. 1955.
9	Newhall prospect.....	Sec. 9, T. 3 N., R. 16 W.....	Saugus.....	Pliocene and pleistocene.....	2	Moore and Stephens (1954, p. 5).
Colorado						
10	Hangover claims.....	Sec. 16, T. 3 N., R. 101 W.....	Mesaverde.....	Cretaceous.....	5	J. D. Vine, written communication, 1955.
11	Lay mine.....	Sec. 31, T. 7 N., R. 93 W.....	do.....	do.....	4	Vine and Moore (1952a, p. 10).
12	Bob Cat group.....	Sec. 3, T. 7 N., R. 93 W.....	Lance(?).....	do.....	4	M. J. Bergin, written communication, 1955.
13	Fred Brand ranch.....	Sec. 3, T. 9 N., R. 81 W.....	Unnamed.....	Quaternary.....	1	K. E. Baker, and L. E. Smith, written communication, Nov. 1953.
14	Coalmont area.....	Sec. 24, T. 7 N., R. 81 W.....	Coalmont.....	Eocene.....	2	Vine and Moore (1952a, p. 7).
15	Carr area.....	Sec. 26, T. 11 N., R. 68 W.....	Laramie.....	Cretaceous.....	5	Gill (1953b, p. 118).
16	Old Leyden coal mine.....	Sec. 28, T. 2 S., R. 70 W.....	do.....	do.....	*4	Gude and McKeown (1952).
17	Uncompahgre area.....	Sec. 14, T. 11 S., R. 102 W.....	Dakota.....	do.....	5	Baltz (1955, p. 25).
18	Almont area.....	Sec. 28, T. 15 S., R. 84 W.....	Belden.....	Pennsylvanian.....	5	Gill (1953b, p. 118).
19	Tuffy claims.....	Sec. 20, T. 26 S., R. 68 W.....	Dakota.....	Cretaceous.....	5	M. R. Simon, written communication, Nov. 1956.
Florida						
20	Venice beach.....	Venice.....	Unnamed.....	Recent.....	1	A. L. Slaughter, written communication, 1945.
21	Mud Lake.....	Sec. 2, T. 14 N., R. 24 E.....	do.....	Quaternary.....	1	Koppe and others (1955, p. 60).
Idaho						
22	Fall Creek area.....	Sec. 4, T. 1 S., R. 42 E.....	Bear River.....	Cretaceous.....	*5	Vine and Moore, (1952b).
23	Grays Lake area.....	Sec. 23, T. 3 S., R. 43 E.....	do.....	do.....	5	Vine (1959, p. 288).
24	Goose Creek area.....	T. 16 S., R. 21 E.....	Salt Lake.....	Pliocene.....	*5	Mapel and Hail (1959).
Illinois						
25	Sahara Coal Co. strip mine.....	Sec. 31, T. 9 S., R. 5 E.....	Carbondale.....	Pennsylvanian.....	3	Patterson (1954a, p. 13).
Kansas						
26	Fort Scott area.....	Sec. 27, T. 25 S., R. 25 E.....	Des Moines.....	Pennsylvanian.....	*3	Walter Danilchik, oral communication, 1956.
Missouri						
27	Bellamy area.....	Sec. 25, T. 34 N., R. 30 W.....	Cherokee.....	Pennsylvanian.....	3	W. J. Hail, Jr., written communication, 1954.
Montana						
28	Flint Creek valley area.....	Sec. 35, T. 11 N., R. 13 W.....	Tertiary basin deposits.....	Oligocene(?).....	4	Hail and Gill (1953, p. 6).
29	Prickly Pear Valley area.....	Sec. 28, T. 11 N., R. 2 W.....	do.....	do.....	4	Hail and Gill (1953, p. 5).
30	Winston area.....	Secs. 18 and 28, T. 9 N., R. 1 E.....	do.....	do.....	*5	Becraft (1958).
31	Leland Peterson farm.....	Secs. 2 and 10, T. 2 N., R. 1 W.....	do.....	do.....	4	R. G. Pruitt, Jr., written communication, 1955.
32	Stanolind Oil Co. drill hole.....	Sec. 21, T. 25 N., R. 55 E.....	Fort Union.....	Paleocene.....	2	U.S. Atomic Energy Commission, written communications, Nov. and Dec., 1954.
33	Blue Butte.....	Secs. 2 and 10, T. 17 N., R. 59 E.....	do.....	do.....	4	E. P. Beroni and H. L. Bauer, Jr., written communication, 1952.
34	T. C. Waldron property.....	Sec. 23, T. 10 N., R. 60 E.....	do.....	do.....	4	L. Y. Marks, written communication, 1955.
35	Ekalaka Hills lignite field.....	T. 1 N., Rs. 58 and 59 E.....	do.....	do.....	*2	Gill (1959).
36	Iong Pine Hills.....	Sec. 20, T. 1 S., R. 61 E.....	do.....	do.....	2	Denson, Bachman, and Zeller (1959, p. 46-47).
37	U.S. Government.....	Sec. 20, T. 2 S., R. 62 E.....	do.....	do.....	5	Gill (1954, p. 154).
38	Zelbert Portwine prospect.....	Sec. 5, T. 9 S., R. 50 E.....	do.....	do.....	2	D. L. Norton, written communication, 1955.

TABLE 8.—*Deposits of uranium in coaly carbonaceous rocks in the United States—Continued*

[Deposit type: 1, unconsolidated peaty material; 2, lignite and subbituminous coal; 3, bituminous and anthracitic coal; 4, impure lignite and lignitic shale; 5, carbonaceous shale. Asterisk (*) indicates an estimated reserve of more than about 1,000 tons]

Locality			Stratigraphic unit	Age	Deposit type	Source of data
No. (pl. 1)	Name	Description				
Nevada						
39	Gamma property	Sec. (?), T. 16 N., R. 37 E.	Unnamed	Tertiary	4	Staatz and Bauer (1954, p. 76-77).
New Mexico						
40	Beautiful Mountain	E½ T. 26 N., R. 20 W.	Gallup	Cretaceous	4	Bachman and Read (1952, p. 11).
41	Hogback No. 4 mine	Sec. 12, T. 15 N., R. 18 W.	Dakota	do	*4	Gabelman (1956, p. 307-308).
42	Dalton Pass area	Sec. 28, T. 17 N., R. 14 W.; sec. 2, T. 16 N., R. 14 W., and sec. 5, T. 16 N., R. 13 W.	Mesaverde	do	4	G. O. Bachman, E. H. Baltz, and R. B. O'Sullivan, written communication, 1953.
43	Hosta Butte area	Secs. 26 and 27, T. 16 N., R. 13 W., and sec. 32, T. 16 N., R. 12 W.	do	do	4	Do.
44	Canyon Mulatto	Secs. 14 and 24, T. 14 N., R. 9 W.	do	do	4	Do.
45	La Ventana Mesa area	Secs. 14, 28, 29, 32, and 33, T. 19 N., R. 1 W.	Mesaverde and Dakota	do	*4	Bachman and others (1959).
46	Butler claims	Secs. 12 and 13, T. 20 N., R. 1 W.	Morrison	Jurassic	*5	Gabelman (1956, p. 308-312).
47	Coyote area	Sec. 27(?), T. 22 N., R. 3 E.	Todilto	do	5	G. O. Bachman, and C. B. Read, written communication, 1951.
North Dakota						
48	Killdeer Mountain area	Sec. 14, T. 146 N., R. 99 W.	Sentinel Butte	Paleocene	4	Gill (1954, p. 154).
49	Gorham area	Sec. 7, T. 141 N., R. 99 W.; sec. 15, T. 141 N., R. 100 W.; and sec. 23, T. 142 N., R. 100 W.	do	do	*4	G. I. Haines, and R. C. Barkley, written communications, Sept.-Dec. 1955.
50	Klym School area	Secs. 23 and 26, T. 142 N., R. 99 W.	do	do	*4	U.S. Atomic Energy Commission, written communication, Nov. 1956.
51	Manthien School area	Secs. 3, 9, and 10, T. 140 N., R. 100 W.	do	do	*4	G. I. Haines, and R. C. Barkley, written communication, Aug. 1955.
52	North Belfield area	Secs. 3 and 6, T. 140 N., R. 99 W., and sec. 33, 34, 35, and 36, T. 141 N., R. 99 W.	do	do	*4	G. I. Haines, R. C. Barkley, and A. S. Florio, written communications, Nov. 1955-Feb. 1956.
53	Sentinel Butte area	Secs. 6 and 8, T. 139 N., R. 104 W.	do	do	*2	Moore, Melin, and Kepferle (1959, p. 154).
54	Flat Top Butte area	Secs. 8, 9, 16, and 17, T. 139 N., R. 103 W.	do	do	*2	E. P. Beroni, and H. L. Bauer, Jr., written communication, 1952.
55	Little Badlands area	Sec. 31, T. 139 N., R. 97 W.	Golden Valley	Eocene	2	Moore, Melin, and Kepferle (1959, p. 164).
56	Bullion Butte area	Secs. 18 and 19, T. 137 N., R. 102 W.	Sentinel Butte	Paleocene	*2	Do.
57	North Rocky Ridge area	N½ T. 137 N., R. 100 W.	do	do	*4	G. I. Haines, and R. C. Barkley, written communications, Aug. 1955-Feb. 1956.
58	South Rocky Ridge area	Secs. 26 and 34, T. 137 N., R. 100 W., and sec. 5, T. 136 N., R. 100 W.	do	do	*4	G. I. Haines, H. B. Young, and W. J. Weaver, written communications, May-Aug. 1955.
59	Martha Gabbert property	Sec. 12, T. 137 N., R. 95 W.	do	do	4	R. C. Barkley, written communication, Oct. 1955.
60	Chalky Buttes area	Tps. 133 and 134 N., Rs. 100, 101, and 102 W.	do	do	*2	Moore, Melin, and Kepferle (1959, p. 154-161).
61	Rhame area	Sec. 25, T. 132 N., R. 103 W.	do	do	4	Gill (1954, p. 154).
62	Whetstone Buttes area	Sec. 32, T. 132 N., R. 98 W.	do	do	2	D. L. Norton, written communication, 1955.
63	Frank Leuton property	Sec. 18, T. 131 N., R. 101 W.	do	do	2	Do.
64	Medicine Pole Hills area	Tps. 130 and 131 N., R. 104 W.	do	do	*2	Denson, Bachman, and Zeller (1959, p. 44-45).
Oklahoma						
65	Nowata area	Sec. 27, T. 26 N., R. 15 E.	Missouri	Pennsylvanian	3	Walter Danilchik, written communication, 1956.
Pennsylvania						
66	Darlington area	2 miles southeast of Darlington	Lower Freeport	Pennsylvanian	3	Ferm (1955, p. 36).
67	Krolick Coal Co. mine	LeContes Mills	do	do	3	Patterson (1954c, p. 13).
South Dakota						
68	Tepee Butte area	Sec. 8, T. 22 N., R. 7 E.	Tongue River	Paleocene	*5	Gill (1954, p. 152).
69	North Cave Hills area	S½ T. 22 N., R. 5 E.	do	do	*4	Gill (1955, p. 153-158); Kepferle and Chisholm (1956, p. 243-254); King and Young (1956).
70	Table Mountain area	Secs. 5 and 9, T. 22 N., R. 4 E.	do	do	*4	Denson, Bachman, and Zeller (1959, p. 43-44).
71	South Cave Hills area	SE¼ T. 21 N., R. 4 E.; SW¼ T. 21 N., R. 5 E.; and NW¼ T. 20 N., R. 5 E.	Ludlow and Tongue River	do	*4	King and Young (1956).
72	Falcon group	Sec. 17, T. 19 N., R. 3 E.	Ludlow	do	4	W. J. Weaver and H. H. Rabke, written communication, July 1955.
73	Lodgepole and Johnson outlier area	Sec. 19, T. 21 N., R. 12 E., and sec. 9 and 10, T. 21 N., R. 11 E.	do	do	*2	Denson, Bachman, and Zeller (1959, p. 45); Zeller and Schopf (1959, p. 63-65).
74	Calamity Jane No. 2 claim	Sec. 24, T. 19 N., R. 7 E.	do	do	2	Lynn Burton and H. B. Young, written communication, Dec. 1954.

TABLE 8.—*Deposits of uranium in coaly carbonaceous rocks in the United States—Continued*

[Deposit type: 1, unconsolidated peaty material; 2, lignite and subbituminous coal; 3, bituminous and anthracitic coal; 4, impure lignite and lignitic shale; 5, carbonaceous shale. Asterisk (*) indicates an estimated reserve of more than about 1,000 tons]

Locality			Stratigraphic unit	Age	Deposit type	Source of data
No. (pl. 1)	Name	Description				
South Dakota—Continued						
75	Northern Slim Buttes area....	Secs. 20, 21, 28, and 29, T. 19 N., R. 8 E., and Tps. 17 and 18 N., Rs. 7 and 8 E.	Ludlow.....	Paleocene.....	*2	Denson, Bachman, and Zeller (1959, p. 45-46).
76	Southern Slim Buttes area....	E½ T. 16 N., R. 8 E., and W½ T. 16 N., R. 9 E.	do.....	do.....	*4	King and Young (1956); Gill and Denson (1955; 1957).
77	Edgemont area.....	Sec. 14, T. 9 S., R. 3 E.	Lakota.....	Cretaceous.....	4?	U.S. Atomic Energy Commission, written communication, Aug. 1957.
Texas						
78	Rauch farm.....	Northern Fayette County.....	Jackson.....	Eocene.....	5	R. G. Blair, written communication, Nov. 1955.
79	Speed farm.....	Muldoon.....	do.....	do.....	5	Do.
Utah						
80	Pettit farm.....	Sec. 2, T. 4 N., R. 1 W.	Unnamed	Quaternary.....	1	Duncan (1953, p. 21).
81	Steinaker Draw area.....	Sec. 13, T. 3 S., R. 21 E.	Morrison.....	Jurassic.....	5	Vine and Moore (1952a, p. 18).
82	South Myton oil field area.....	Sec. 34, T. 8 S., R. 16 E.	Bridger.....	Eocene.....	2	Kenneth Bell, oral communication, 1954.
83	Kaiparowitz Plateau area.....	Sec. 35, T. 37 S., R. 4 E.	Dakota (?).....	Cretaceous.....	5	Zeller (1955, p. 2).
Wyoming						
84	Crooked Creek group.....	Secs. 27 and 28, T. 58 N., R. 95 W.	Morrison.....	Jurassic.....	5	O. M. Hart, written communication, May 1955.
85	Al Egbert property.....	Sec. 3, T. 54 N., R. 95 W.	do.....	do.....	5	Do.
86	Hyattville area.....	Sec. 31, T. 49 N., R. 89 W.	do.....	do.....	5	W. J. Hail, Jr., written communication, 1954.
87	Matteson and Sturdevant property.....	Secs. 14, 15, 22, and 23, T. 48 N., R. 90 W.	do.....	do.....	5	O. M. Hart, written communication, April 1955.
88	Christenson Brothers' Ranch.....	Secs. 5 and 8, T. 45 N., R. 76 W.	Wasatch.....	Eocene.....	4	C. P. Bromley and R. G. Lindlof, written communication, 1954; Troyer and others (1954, p. 7).
89	Cambria coal field.....	Secs. 20 and 29, T. 46 N., R. 61 W.	Lakota.....	Cretaceous.....	3	W. P. Horner and J. B. Ridlon, written communication, Sept. 1952.
90	Wind River Indian Reservation.....	Secs. 6, 7, and 8, T. 7 N., R. 5 W.	Aycross.....	Eocene.....	5	D. L. Norton, written communication, 1957.
91	Red Rose claims.....	Secs. 20 and 21, T. 43 N., R. 93 W.	Morrison.....	Jurassic.....	5	W. R. Peterson, written communication, 1954.
92	Great Pine Ridge area.....	Sec. 11, T. 42 N., R. 78 W.	Fort Union.....	Paleocene.....	4	Love (1952, p. 9).
93	Dry Fork Powder River area.....	Sec. 5, T. 42 N., R. 76 W.	Wasatch.....	Eocene.....	5	Troyer and others (1954, p. 8).
94	Pumpkin Buttes area.....	Sec. 22, T. 43 N., R. 76 W., and sec. 4, T. 43 N., R. 75 W.	do.....	do.....	2	Troyer and others (1954, p. 4, 7).
95	Kinsely and Peterson property.....	Sec. 6(?), T. 40 N., R. 75 W.	do.....	do.....	2	H. E. Geslin, written communication, July 1953.
96	Shoshoni area.....	Sec. 10(?), T. 38 N., R. 94 W.	Wind River.....	do.....	4	T. H. W. Loomis and D. P. King, written communication, Jan. 1955.
97	Lux No. 4 claim.....	Sec. 12, T. 37 N., R. 90 W.	do.....	do.....	4	D. L. Norton, written communication, June 1955.
98	Jenkins and Hand property.....	Sec. 32, T. 33 N., R. 90 W.	do.....	do.....	5	J. D. Love and N. M. Denson, written communication, Oct. 1953.
99	Gas Hills area.....	Sec. 27, T. 33 N., R. 89 W.	do.....	do.....	*4	H. D. Zeller, written communication, Dec. 1957.
100	Fulton C. Jameson Ranch.....	Sec. 3, T. 33 N., R. 88 W.	Morrison.....	Jurassic.....	5	W. J. Hail, Jr., written communication, 1954.
101	Pine Mountain area.....	Sec. 28, T. 34 N., R. 84 W.	Wind River.....	Eocene.....	5	H. D. Zeller and P. E. Soister, written communication, Nov. 1954.
102	Nine Mile Lake area.....	Secs. 29 and 33, T. 35 N., R. 79 E.	Mesaverde.....	Cretaceous.....	*5	H. E. Geslin, written communication, 1955.
103	J. W. Strook property.....	Sec. 27, T. 33 N., R. 72 W.	Fort Union.....	Paleocene.....	2	C. P. Bromley, written communication, 1955.
104	Bob Cat claims.....	Sec. 24, T. 33 N., R. 68 W.	do.....	do.....	2	Bromley (1955, p. 15).
105	New Hope claims.....	Sec. 24, T. 34 N., R. 67 W.	do.....	do.....	2	Do.
106	Ace High and Lillian claims.....	Sec. 19, T. 36 N., R. 64 W.	Fox Hills.....	Cretaceous.....	5	C. P. Bromley, written communication, 1954.
107	Shamrock Ranch.....	Sec. 12, T. 25 N., R. 79 W.	Morrison.....	Jurassic.....	5	W. J. Hail, Jr., written communication, 1954.
108	Red Desert area.....	Tps. 20-24, N., Rs. 92-95 W.	Wasatch.....	Eocene.....	*2	Masursky and Pipiringos (1959). Wyant, Sharp, and Sheridan (1956, p. 246-254).
109	Black Butte area.....	Sec. 2(?), T. 18 N., R. 101 W., and sec. 11, T. 18 N., R. 101 W.	Ericson.....	Cretaceous.....	5	C. P. Bromley and J. F. Whalen, written communication, July 1955.
110	Little Kay claims.....	Sec. 26, T. 17 N., R. 102 W.	Almond.....	do.....	5	J. F. Whalen and C. W. Holmquist, written communication, July 1955.
111	Pine Mountain, Red Creek Basin.....	Sec. 31, T. 13 N., R. 103 W., and sec. 25 and 26, T. 13 N., R. 104 W.	Wasatch.....	Eocene.....	5	E. C. Winterhalder, written communication, 1953.
112	Saratoga area.....	Sec. 12, T. 17 N., R. 84 W.	North Park(?).....	Pliocene(?).....	5	Stephens and Bergin (1959, p. 330).
113	Arbob claims.....	Sec. 2 and 3, T. 27 N., R. 78 W.	Wind River.....	Eocene.....	5	R. R. Guiling and J. J. Schulte, written communication, Dec. 1955.

2. Either (a) chemical analysis by the Geological Survey Laboratory or other reliable source indicates at least 0.005 percent uranium in the rock, or (b) a radioactivity measurement by the Geological Survey laboratory or other reliable source indicates a level of radioactivity for the rock equivalent to that of similar material containing at least 0.005 percent uranium in radioactive equilibrium with its natural products of radioactive decay that is equivalent uranium.
3. The description of the location and stratigraphic position where the sample was collected is adequate to locate the sample within 1 mile geographically and within a single formation stratigraphically.
4. An authoritative reference is available for citation.

The deposits, localities, and areas listed in table 8 are shown on plate 1. Five categories of host rocks and two categories of deposit sizes are shown. Where information is available, areas known to contain more than 1,000 tons of uraniferous rock are indicated. Where deposits are closely spaced several are generally grouped under an area name in order not to reveal the size of any individual privately owned property or deposit.

UNCONSOLIDATED PEATY MATERIALS

Uranium deposits in unconsolidated peaty materials including peat, impure peat, peaty and carbonaceous sediment (that is, mud, sand, and soil) are considered as a group because of the similarity in the deposits. Uranium deposits of this type are forming locally at the present time or were formed in Pleistocene or Recent swamps, peat bogs, mountain meadows, and similar places where plant debris accumulates.

Two deposits of uranium in unconsolidated peaty material were reported in alpine meadows in the Sierra Nevada region of California during 1955. The largest of the two is at Hoffman Meadow, an open grassy area surrounded by a conifer forest at an altitude of about 6,800 feet near Huntington Lake in northern Fresno County. Uranium is concentrated in a spring-fed water-saturated meadow soil composed of a thin turf of living grass and herbs whose roots are interwoven with dead and decaying roots and other plant debris. The turf grades downward into a mixture of decayed plant material and disintegrated rock and mineral fragments from underlying granodiorite. The meadow soil has an average thickness of about 2 feet and covers several acres. Samples of decomposed plant debris contain as much as

0.7 percent uranium (W. E. Bales and G. M. Haselton, written communication, 1955). Living grasses, herbs, and trees in and surrounding the meadow also contain abnormally high concentrations of uranium, but the highest concentrations of uranium are in the water-saturated decaying plant debris within the soil.

The second of the two deposits of uranium in unconsolidated peaty material discovered in the Sierra Nevada region of California during 1955 is in the Pettit Ranch area in the Greenhorn Mountains east of Bakersfield, Kern County. This deposit, which is very similar to the one at Hoffman Meadow, is in an open grassy meadow surrounded by conifer forest at an altitude of about 5,000 feet. Uranium is concentrated in the decaying plant material of the soil, which is as much as 8 feet thick locally but averages only about 2 feet thick (W. A. Bowes, W. E. Bales, and G. M. Haselton, written communication, 1957). Uranium is concentrated in an area of less than 2 acres which is kept moist by a line of seeps. Samples of water from the seeps contain from 40 to 300 ppb (parts per billion) uranium.

Other deposits of uranium in unconsolidated peaty material in mountain meadows of the Sierra Nevada area in California have been reported and are listed in table 8, but little detailed information is available.

Deposits in unconsolidated peaty materials of comparable size and grade to those at Hoffman Meadow and Pettit Ranch are not known in other parts of the country, though minor occurrences have been reported. At Fred Brand's ranch in the North Park area, Jackson County, Colo., a deposit of uranium-bearing unconsolidated peaty material is associated with ferruginous tufa at springs and seeps issuing from near the margin of a faulted block of Precambrian rock (K. E. Baker and L. E. Smith, written communication, 1953). A sample of peaty mud from a Pleistocene terrace deposit of the former Lake Bonneville at the E. W. Pettit farm, Davis County, Utah, contains 0.006 percent uranium, 58 percent ash, and 0.01 percent uranium in the ash (Duncan, 1953, p. 21).

A modern peat bog at Mud Lake, Marion County, Fla., contains 0.005 percent uranium in a layer of peat several inches thick, about 14 feet below the surface of the water (Koppe and others, 1955, p. 60). Muck along the modern beach at Venice, Fla., has been estimated to contain about 0.005 percent equivalent uranium (A. L. Slaughter, written communication, 1945), but there is no evidence to suggest whether the uranium is associated with the carbonaceous or the mineral matter.

LIGNITE AND SUBBITUMINOUS COAL

Deposits of uranium in lignite are found at a number of places or areas in the northern Great Plains, the intermontane basins of the Rocky Mountain region, and the far western States. A few deposits are in subbituminous coal that can be distinguished from lignite only by careful chemical analysis and are therefore grouped with the deposits in lignite.

That part of the Fort Union lignite region which includes southwestern North Dakota, northwestern South Dakota, and eastern Montana contains the largest number of uranium deposits in lignite of any region in the United States. The same region also contains the largest number of uranium deposits in impure lignite and lignitic shale, which grade imperceptibly into the deposits in lignite but which are here treated separately in accordance with the arbitrary definitions previously established for these rock types.

The deposits are characterized by a relatively low concentration of uranium but by relatively large areal extent. A few contain more than 0.1 percent uranium but these are generally small irregular parts of larger deposits that consist primarily of impure lignite or lignitic shale.

In the Fort Union lignite region, beds of lignite contain as much as 0.005 percent uranium in at least 10 relatively large areas, as follows: Sentinel Butte, Flat Top Butte, Bullion Butte, Chalky Buttes, and Medicine Pole Hills in North Dakota; the Ekalaka Hills in Montana; the Lodgepole area, the Johnson outlier, and two areas in the northern Slim Buttes, the Bar H area and the Mendenhall area, in South Dakota. Plate 1 and figure 30 show the relative position of these areas. In all these areas the uraniumiferous beds are woody lignites of the Fort Union formation of Paleocene age where these beds are exposed on the flanks of buttes and other topographic highs. Where the beds are more than about 2 feet thick, uranium is concentrated in the top or bottom few inches. From south to north the uranium occurs in progressively younger beds of the Fort Union formation through a stratigraphic interval of at least 1,000 feet. The Fort Union formation consists of three members, the Ludlow, Tongue River, and Sentinel Butte, from oldest to youngest. The uraniumiferous lignite beds occur in the Ludlow member in the Slim Buttes and Ekalaka areas, in the Tongue River member in the Cave Hills, Lodgepole, and Medicine Pole Hills areas, and in the Sentinel Butte member in the Chalky Buttes, Bullion Butte, Flat Top Butte, and Sentinel Butte areas. The general geologic setting,

the stratigraphy, and the results of the Geological Survey's search for uraniumiferous lignites in the Fort Union lignite region, including the results of exploratory drilling projects, are summarized in the following reports: Denson, Bachman, and Zeller (1959), Zeller and Schopf (1959), Gill, Zeller and Schopf (1959), Moore, Melin, and Kepferle (1959), and Gill (1959).

A summary of the individual estimates for each of the above areas indicates that more than 90 million tons of lignite in the Fort Union lignite region contain 0.006 to 0.015 percent uranium and 10 to 14 percent ash. This lignite therefore contains a large amount of uranium that may become a valuable by-product of future industrial utilization of the lignite for heat or chemical raw material.

The only area known to contain a large reserve of low-grade uranium in lignite or coal comparable to that of the Fort Union lignite region is the Red Desert of the Great Divide Basin, Sweetwater County, Wyo. There, uranium is contained in beds of subbituminous coal and impure coal in the Wasatch formation of Eocene age. The coal is similar in most respects to that of the Fort Union lignite region but has a higher heating value and a higher proportion of attritus to vitrain (J. M. Schopf and R. J. Gray, written communication, 1955; J. M. Schopf, R. J. Gray, and C. J. Felix, written communication, 1955). About 20 beds of coal, underlying several hundred square miles of relatively flat desert land, are known to contain uranium, but over large parts of the area the uranium content is less than 0.005 percent. Individual deposits containing 0.005 percent uranium or more, and as much as several square miles in size, are known in a belt that trends northwestward for about 25 miles across the central part of the area. These deposits were briefly described by Wyant, Sharp, and Sheridan (1956, p. 246-254) and were later mapped and sampled in detail (Masursky and Pipiringos, 1959; Masursky, 1961; Pipiringos, 1956b. See also Masursky, 1956; Pipiringos, 1956a). The Red Desert area probably contains more than 700 million tons of coal and impure coal with 0.003 percent uranium, but less than 30 million tons with as much as 0.005 percent uranium. The ash content of the host rock ranges from about 12 to 30 percent.

Several other deposits of uranium in lignite and subbituminous coal are listed in table 8 and shown on plate 3. These include two in California, one in Colorado, three in Montana, one in Nevada, three in North Dakota, one in Utah, and four in Wyoming. Most of these deposits appear to be small.

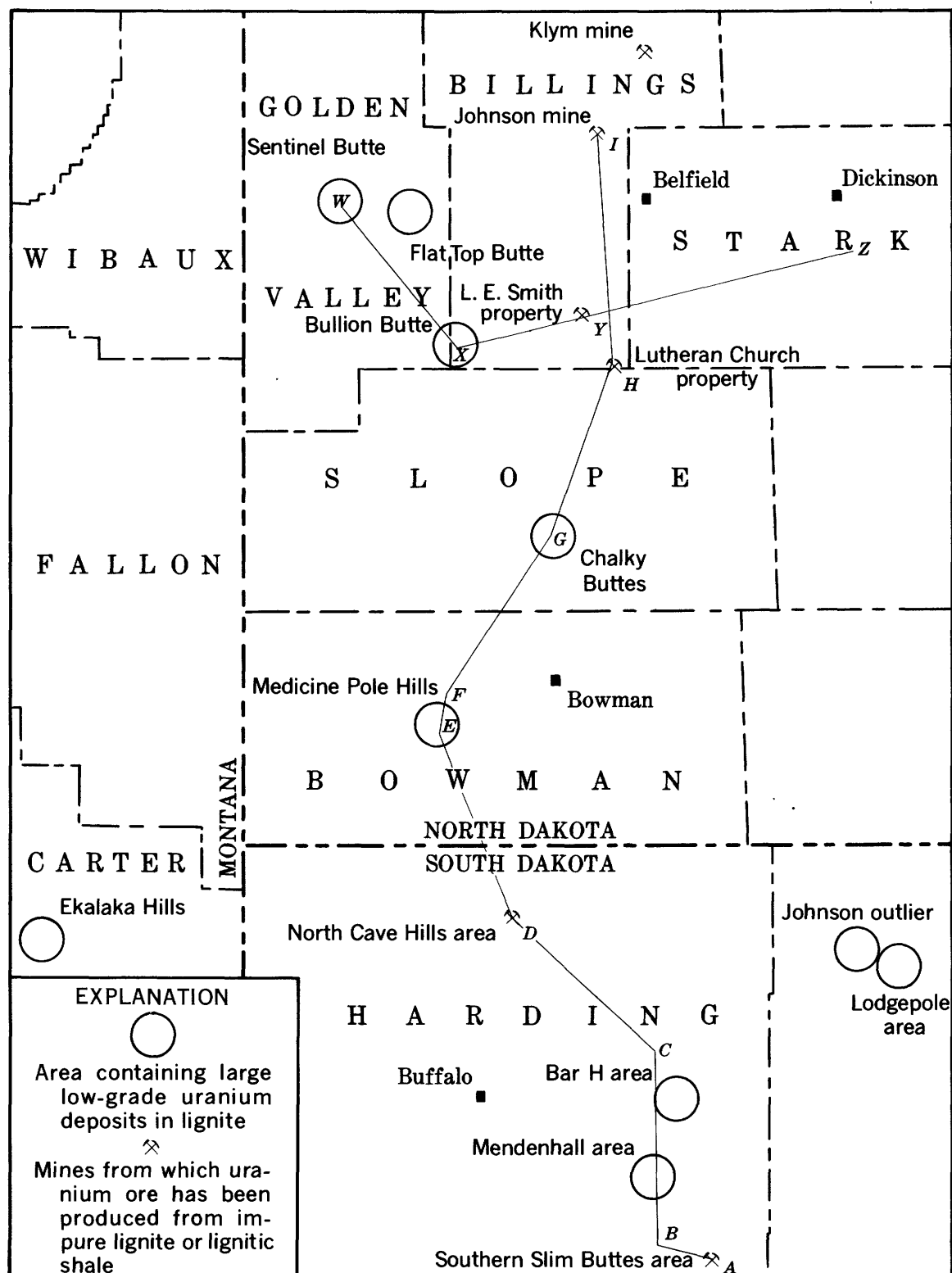


FIGURE 30.—Distribution of uranium deposits in lignite, impure lignite, and lignitic shale, North Dakota, South Dakota, and Montana. Line of sections shown on figure 36.

BITUMINOUS AND ANTHRACITIC COAL

No significant deposits of uranium in bituminous or anthracitic coals are known in the United States. Samples of bituminous coal containing 0.005 percent uranium or more have been collected from seven localities (pl. 3; table 8), one each in Illinois, Kansas, Missouri, Oklahoma, and Wyoming, and two in Pennsylvania (see also table 7). With the exception of the deposit in Wyoming, which is in a local bed of bituminous coal in the Lakota formation of Early Cretaceous age, all the deposits occur in bituminous coal of Pennsylvanian age. So far as known, all the deposits with the possible exception of the one in Kansas, are small erratic concentrations of uranium. The Kansas locality is a roadcut on U.S. Highway 59 northeast of Fort Scott which exposes the Mulky coal of the upper part of the Cabaniss group of Pennsylvanian age (Walter Danilchik, written communication, 1956). A sample through the 0.9-foot-thick bed of coal contains 0.007 percent uranium and 9.7 percent ash. The bed is said to be radioactive for at least a mile along the outcrop. Such continuity is suggestive of a syngenetic origin (criterion 1, table 5). So far as known none of the other deposits are continuous for more than a few inches.

A sample of bituminous coal of Pennsylvanian age from the Rio de Peixe coal field near Cambui in the northern Paraná, Brazil, contains more than 0.1 percent uranium (D. D. Haynes and C. T. Pierson, written communication, 1957). If similar high concentrations of uranium occur in the United States, they have not as yet been reported.

In the Interior coal province (Indiana, Illinois, Kansas, Missouri, and Oklahoma), the highest uranium concentration in a cyclic sequence of coal-bearing rocks is commonly in marine black shale that overlies the coal, and the continuity of uranium concentration in the shale seems to be much greater than in the coal. Patterson (1954a, p. 14) reported as much as 0.017 percent uranium and 61.4 percent ash in a grab sample of marine black shale that overlies the Harrisburg (No. 5) coal in Saline County, Ill. In Pennsylvania, where cyclic deposition is not so pronounced, the highest uranium concentration in the coal-bearing strata is generally in underclay directly below the coal. Ferm (1955, p. 37) reported as much as 0.016 percent uranium and 60.7 percent ash in the top 3 inches of underclay below the Lower Freeport coal near Darlington, Beaver County, Pa.

IMPURE LIGNITE AND LIGNITIC SHALE

Uranium deposits in impure lignite and lignitic shale (including some that are in impure coal and coaly shale) represent the most economically valuable deposits of uranium in coaly carbonaceous rocks. The lithology and occurrence of these deposits is transitional between large low-grade deposits of uranium in lignite and the uranium deposits in carbonaceous shale. It is a curious and not very well understood fact that most of the relatively high-grade substantial-sized deposits of uranium in coaly carbonaceous rocks (deposits containing 0.1 percent or more uranium) are in this category. At least 14 areas in the United States contain such deposits. Of the relatively high-grade deposits six are in North Dakota, five in South Dakota, and one each in Colorado, New Mexico, and Wyoming.

Substantial deposits containing more than 0.1 percent uranium in impure lignite and lignitic shale are known in at least 11 areas in the Fort Union lignite region (pl. 1; fig. 30). This same region contains several smaller or lower grade deposits of the same type. The geologic setting for these deposits is similar to that for the large low-grade deposits of uranium in lignite in the same region. The higher grade deposits in impure lignite and lignitic shale are smaller and more erratic and show a closer relation to small-scale features of the structure and the stratigraphy. With several notable exceptions they are in weathered rock with a moisture content of about 50 percent and an ash content of about 50 percent on a moisture-free dry-weight basis. Secondary enrichment related to the present cycle of erosion has probably been a factor in the localization of some of the deposits.

In Billings County, N. Dak., many of the highest grade uranium deposits in impure lignite and lignitic shale extend along a north-south belt for about 35 miles within a few miles east of the Little Missouri Badlands. Within this area, individual deposits ranging from a few acres to several hundred acres in size are clustered in groups, as shown on plate 1.

Two groups of ore deposits called the North and South Rocky Ridge areas are about 12 to 18 miles south of Belfield. In both groups uranium occurs chiefly in a 2- to 4-foot-thick bed of impure lignite, known locally as the Rocky Ridge lignite, which is about 350 feet above the base of the Sentinel Butte member of the Fort Union formation (R. E. Curtiss, written communication, 1957). In the South Rocky Ridge area, as much as 100 feet of sandstone overlies

the mineralized bed and protects it from weathering. The bed is exposed by mining along a very steep escarpment where it consists of dense, tough, woody lignite with an ash content averaging about 20 to 25 percent on the moisture-free dry-weight basis. The uranium content of the fresh impure lignite is very erratic, even in distances of only a few inches. In the North Rocky Ridge area, mining operations have exposed the mineralized bed where it was covered by only a few feet of overburden. There, the bed is weathered friable lignitic shale with a moisture content of nearly 50 percent and an ash content of nearly 40 percent on a moisture-free dry-weight basis. The uranium is more uniformly distributed than in the South Rocky Ridge area but is erratic when considered in relation to the whole deposit. The Rocky Ridge lignite in the north area is overlain by two additional carbonaceous beds, each less than 18 inches thick, about 55 and 70 feet stratigraphically higher than the main zone. The higher beds are discontinuous, but they also contain some of the relatively high grade deposits.

At least four areas of relatively high grade uranium deposits in impure lignite and lignitic shale, each consisting of a cluster of individual deposits, occur in Billings County north of Belfield. These are similar in nearly all respects to the deposits at Rocky Ridge south of Belfield except that the precise correlation of the carbonaceous beds between the areas is not certain. Only very minor structural irregularities interrupt a gentle regional dip of about 10 to 30 feet per mile toward the northeast. At least three beds of impure lignite and lignitic shale under less than 100 feet of overburden contain relatively high grade uranium deposits in this region. Information obtained by private drilling operations indicates that most of the high-grade uranium deposits are lenticular bodies in weathered impure lignite and lignitic shale and that they are not more than about 1 mile down dip from, and parallel to, the trace of the outcrop (Fred Hilpert, oral communication, 1957).

Two areas in Harding County, S. Dak., contain most of the known relatively high grade deposits of uranium in impure lignite and lignitic shale in that state: the North Cave Hills area (Pipiringos, 1957) and the southern part of the Slim Buttes area (Moore and Gill, 1955). Smaller high-grade deposits are also found in the Table Mountain and the South Cave Hills area.

Uranium is erratically distributed throughout parts of the North Cave Hills in a bed of attrital impure

lignite and lignitic shale that averages about 1 foot thick and lies directly above a cliff-forming basal sandstone unit of the Tongue River member of the Fort Union formation. This lignite bed, which is called the *E* bed, is weathered and friable at most outcrops. Secondary uranium minerals of the autunite type are visible locally in some parts of the Riley Pass area of the North Cave Hills in the lignite bed where it is overlain by a thin mantle of soil and is weathered to a soft, black earthy or sooty material. A thin bed of impure lignite and lignitic shale, the *F* bed, lies about 20 feet stratigraphically higher than the *E* bed in parts of the area. The *F* bed also contains some deposits of uranium but is thinner and less persistent than the *E* bed. On the northwest side of the North Cave Hills, at least one deposit of high-grade uranium is in the first lignite zone below the *E* bed, referred to as the *C* lignite zone. The geology and stratigraphic relations of lignite beds in the Cave Hills area are shown on figures 3 and 32. A detailed geologic map of the Cave Hills area is in Denson and others (1955).

The lower part of the Ludlow member of the Fort Union formation contains several high-grade uranium deposits in impure lignite and lignitic shale at the southern end of the Slim Buttes. Uranium is erratically distributed in four or five thin lenticular beds of impure lignite and lignitic shale that are exposed along the flanks of the Buttes. Rocks belonging to the Chadron and Brule formations of Oligocene age unconformably overlie the rocks of the Fort Union formation and these are in turn unconformably overlain by the Arikaree formation of Miocene age, which forms the resistant caprock of the Slim Buttes. The general geologic setting and stratigraphic relations of the uranium-bearing lignite beds in the southern part of the Slim Buttes area are shown on figures 33 and 34. Most of the deposits are near a gentle trough formed at the base of the Miocene rocks.

A deposit of uranium in impure coal and coaly shale at La Ventana Mesa in Sandoval County, N. Mex., is in nearly flat lying strata of the Mesa-verde group of Late Cretaceous age (Bachman and others, 1959) and is similar in many respects to the deposits in the Fort Union lignite region. The uranium-bearing bed of impure coal and coaly shale ranges from about 3 inches to 4 feet in thickness and lies directly below the La Ventana tongue of the Cliff House sandstone. The highest uranium concentrations are in small, erratically distributed areas that in part may be associated with zones of greater permeability and solution traps.

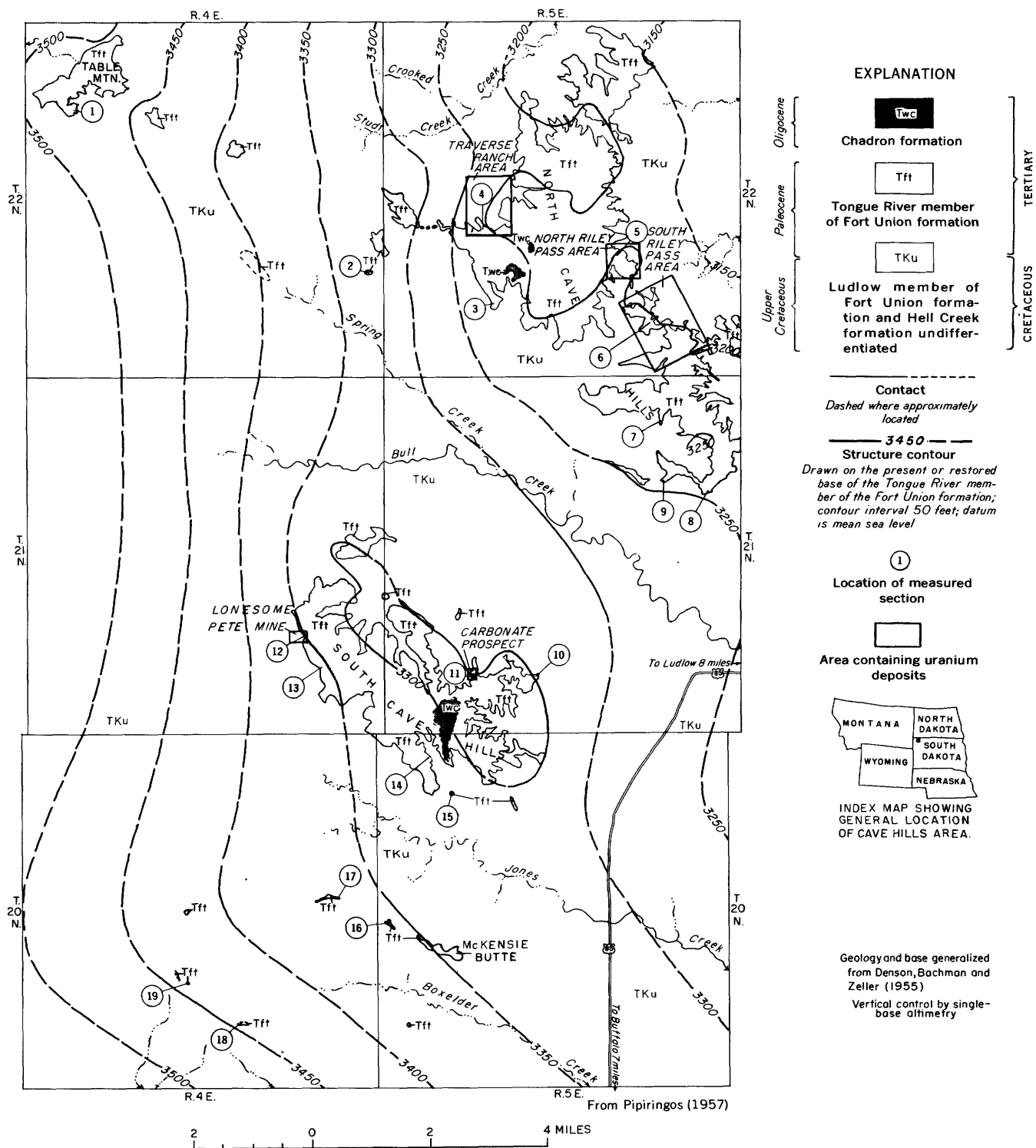


FIGURE 31.—Geologic map of Cave Hills area, Harding County, S. Dak.

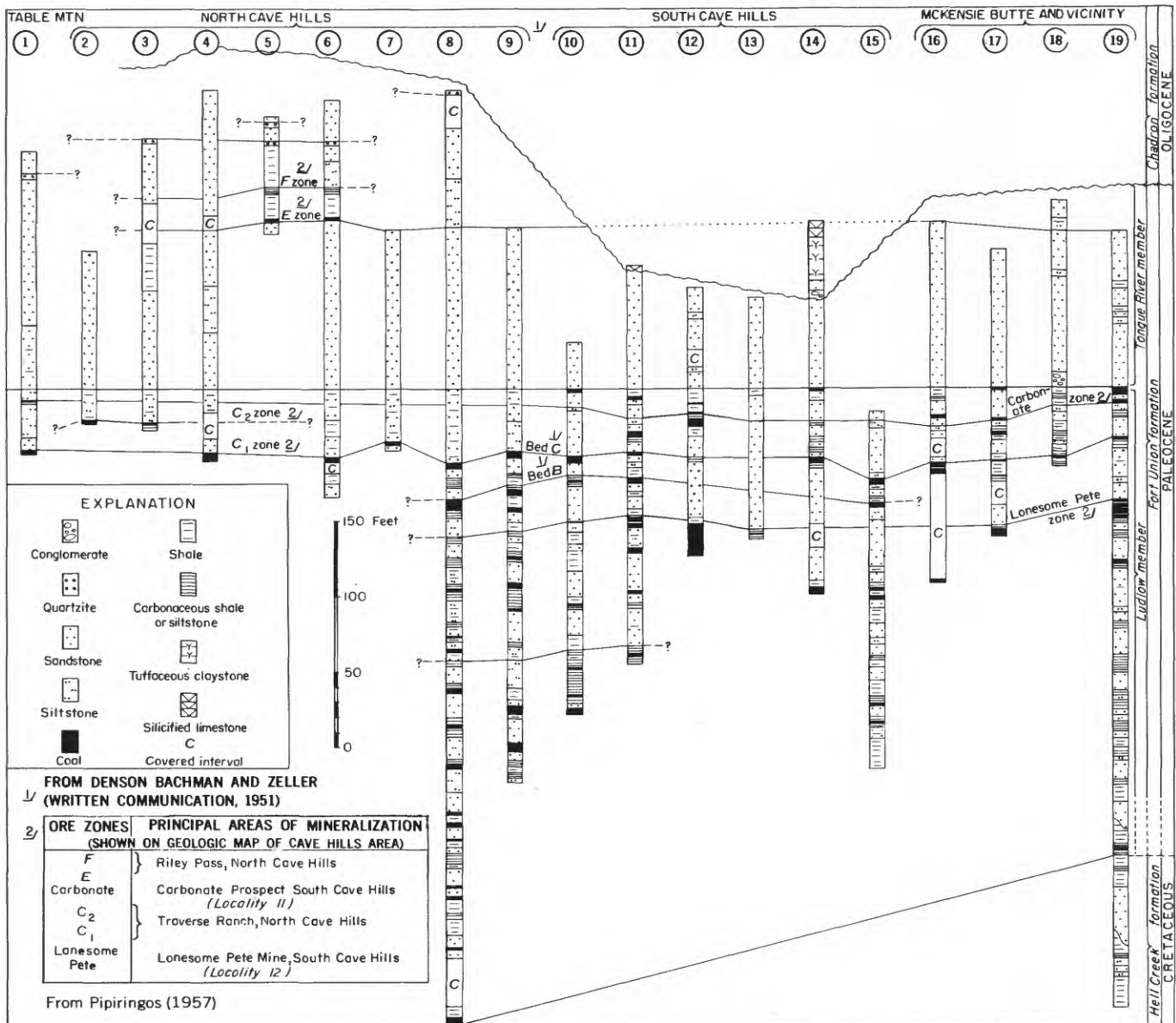


FIGURE 32.—Correlation of uranium ore zones and associated rocks in the Cave Hills area, Harding County, S. Dak.

A local bed of impure coal and coaly shale about 4 feet thick forming a part of a deposit of uranium in sandstone contains as much as 1 percent uranium in the Gas Hills area, Fremont County, Wyo. Uranium ore in sandstone of the Wind River formation of Eocene age intertongues with the bed of impure coal which is continuously exposed for about 100 feet in the Lucky Mc mine. The impure coal differs from the lenses of carbonaceous matter commonly associated with uranium deposits in sandstone in that it constitutes an entire bed of coal instead of being a coalified fragment of a single plant.

A bed of impure coal in the Laramie formation of Late Cretaceous age has been mined for uranium at the Old Leyden coal mine north of Golden, Colo. The rocks at the mine are nearly vertical, and incompetent strata including the impure coal are sheared and brecciated. The higher concentrations of uranium are quite local and may be associated with zones of greater shearing though information is too meager to be certain. Carnotite is in some silicified and brecciated parts of the impure coal, but most of the coal has no visible uranium mineral.

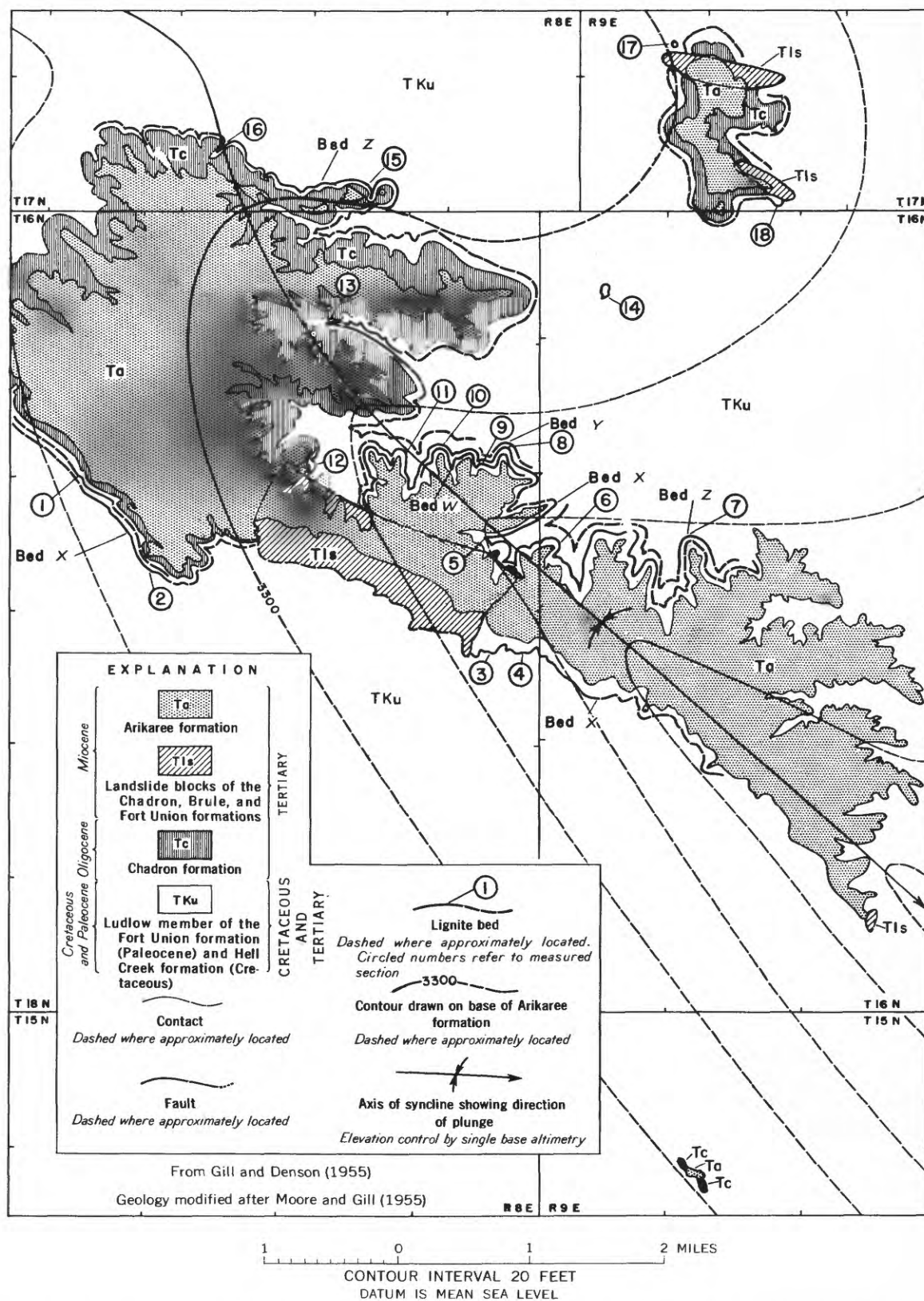


FIGURE 33.—Geologic map of the southern part of the Slim Buttes area, Harding County, S. Dak. Correlation of lignite beds shown on figure 34.

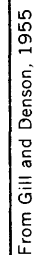


FIGURE 34.—Uranium content and correlation of lignite beds in the southern part of the Slim Buttes area, Harding County, S. Dak.

CARBONACEOUS SHALE

Uranium deposits in carbonaceous shale include deposits in nonmarine rocks that are characterized by coaly carbonaceous matter but are too high in ash content to be called coal, impure coal, or coaly shale. Some uranium deposits, such as those in the Goose Creek and the Fall Creek areas of Idaho, are in black carbonaceous shale beds that are transitional with and locally grade into beds of impure lignite or coal. Others are in beds of gray or brown carbonaceous shale or clay. Still others are in lenticular masses or zones of carbonaceous matter in rocks that otherwise are predominantly sandy.

Carbonaceous shale, lignitic shale, and impure lignite in the Goose Creek area, Cassia County, Idaho, contain as much as 0.1 percent uranium locally and perhaps as much as 0.01 percent uranium distributed throughout several million tons of rock (Duncan, 1953, p. 9; Mapel and Hail, 1959). The uranium occurs chiefly in two zones of coaly carbonaceous rock in the Salt Lake formation of Pliocene age in the Goose Creek basin, an intermontane basin near the northern margin of the Basin and Range province. The Salt Lake formation consists of more than 2,000 feet of gray to white volcanic ash and welded tuff interbedded with lenticular beds of conglomerate, sandstone, and in the lower part, carbonaceous shale and lignitic shale. The carbonaceous material was described by Schopf and Gray (1954, p. 7-9) as a highly attrital, nonbanded type, rich in humic matter derived from the aerobic decomposition of plant products. The distribution of uranium in these beds is erratic but there may be a slightly greater concentration of uranium in the central part of the Goose Creek basin.

Carbonaceous shale, coaly shale, and carbonaceous limestone in the upper part of the Bear River formation of Early Cretaceous age contain uranium in the Fall Creek area, Bonneville County, Idaho (Vine and Moore, 1952b; Vine, 1959). The area is intensely folded and faulted and the zone of carbonaceous rocks is sheared, the bedding partly obliterated, and the zone itself discontinuous. Where exposed in the 85-foot inclined shaft of an abandoned coal prospect, the carbonaceous rocks form a zone 7 feet thick. A bed of carbonaceous limestone, $1\frac{1}{2}$ feet thick with an undulating and cavernous lower surface, forms the roof of the inclined shaft. A zone about $1\frac{1}{2}$ feet thick below the limestone consists of clay gouge, carbonaceous shale, and lenses of limestone and coaly material. At the base is a 4-foot-thick zone of sheared carbonaceous and coaly shale with a lenticular fracture and lenses of clay and coaly material. The lime-

stone and the $1\frac{1}{2}$ -foot-thick zone below the limestone each contain about 0.02 percent uranium. Generally, the highest uranium concentration is in the top foot of the 4-foot-thick zone of carbonaceous and coaly shale, which contains an average of about 0.045 percent uranium and as much as 0.13 percent locally. The lower 3 feet of the 4-foot-thick zone of carbonaceous and coaly shale contains an average of only about 0.011 percent uranium. The carbonaceous limestone consists of fragments of fossil shells, cemented in a matrix of carbonaceous material, and is probably of brackish-water origin. The zone of carbonaceous rocks is underlain by at least 4,000 feet of interfingering marine and nonmarine Jurassic and Lower Cretaceous rocks and is overlain by at least 3,000 feet of nonmarine sandstone and red shale in the Wayan formation of Early(?) and Late Cretaceous age.

The zone of carbonaceous rocks in the Fall Creek area is thought to be about the same age as beds of impure coal and coaly shale about 45 miles to the south near Auburn, Wyo. (Vine, 1959, p. 267), and near Cokeville (Vine and Moore, 1952a, p. 19-20) and Sage (Beroni and McKeown, 1952, p. 8-9) that contain 0.002 to 0.0035 percent uranium. Exposures are rare, but this rarity suggests widespread distribution of uranium for a narrowly restricted stratigraphic zone of carbonaceous material and in this respect contrasts with the Red Desert area and Fort Union lignite region where single zones are only locally mineralized.

A deposit of uranium occurs in carbonaceous shale in the Dakota sandstone of Early Cretaceous age at the Hogback No. 4 mine on the flank of the Zuni uplift near Gallup, N. Mex. Gabelman (1956, p. 307-308) described the host rock as a fissile black shale 1 to 3 feet thick with "carbonized" plant fragments; his comparison of it to a bed of peat suggests abundant plant remains and impressions. Mining has exposed the uranium-bearing shale for about 700 feet along the strike. Uranium-bearing shale has been removed to a depth of 60 feet along the dip slope of a resistant sandstone at its base. Neither structural nor stratigraphic control is apparent for the localization of the deposit.

Several deposits of uranium in carbonaceous shale similar to that at the Hogback No. 4 mine are along the hogback of the Dakota along the west flank of the Sierra Nacimiento near La Ventana, N. Mex. (Gabelman, 1956, p. 310-312) described the occurrence of uranium in black shale or "peat" at the Butler deposit and indicated that mineralized rock terminates down-dip against a fault. The same bed can be traced beyond the fault but is not mineralized.

Uranium is erratically distributed in a number of small low-grade deposits in carbonaceous shale in the Morrison formation of Late Jurassic age and in the Cloverly formation of Early Cretaceous age in parts of Wyoming and adjacent States. Generally, the more carbonaceous material in the rock, the higher the uranium concentration in these deposits. None of these deposits is known to have commercial potential.

Carbonaceous shale and related carbonaceous siltstone, mudstone, and claystone contain local deposits of uranium at many other localities throughout the Western United States. Localization of the uranium at many of these deposits is clearly related to the local abundance of coalified plant fragments and the size of the deposit is controlled by the extent of the carbonaceous matter. At other deposits the entire bed or unit is carbonaceous, and the local control for the size of the deposit is not apparent. There is no sharp distinction between some of the deposits listed in this category and some that are classed as uranium deposits in sandstone.

URANIUM DISTRIBUTION AS RELATED TO GEOLOGIC FEATURES

Deposits of uranium in coaly carbonaceous rocks tend to be associated more commonly with some geologic features than with others. It is the purpose of this section to identify and evaluate the more significant geologic features that may cause any given geologic environment to be favorable for the deposition of uranium in coaly carbonaceous rocks.

FAVORABLE HOST-ROCK LITHOLOGIES

As host rocks for uranium, coaly carbonaceous rocks differ in one important respect from most other sedimentary rocks that contain uranium. A ground-water solution containing dissolved uranium may move for a distance of many yards or even miles within a sandstone aquifer before meeting a chemical or physical change in environment favorable for the deposition or concentration of uranium, but the same kind of solution will lose its uranium close to the original point of contact with coaly carbonaceous rocks. The passage of uranium-bearing solutions through even a few inches of coaly carbonaceous rock may be sufficient to remove most of the uranium from solution. Thus, where a deposit occurs in fresh homogeneous coaly rock, the direction and relative volume of flow of uranium-bearing solutions can be determined by an increase in the concentration of uranium toward the source.

The same characteristic is also important not only in determining the position of a deposit but also

in determining the size, shape, and symmetry of the deposit relative to the route traveled by the solutions that transport uranium. Coaly carbonaceous rocks are the most avid collectors of epigenetic uranium known among sedimentary rocks, but more economically important deposits are found in sandstone, probably because sandstone is a more common rock and more permeable.

The favorability of each of the five general classes of coaly carbonaceous rock as a host for uranium may be described as follows:

1. Unconsolidated peaty material is generally capable of being greatly enriched in uranium (as much as several percent) by the introduction of uranium-bearing waters into the bog or swamp where partly decomposed plant remains accumulate. Uranium deposits in peat are not more common because only rarely is uranium-bearing water available in the environment of peat deposition.
2. Beds of lignite and subbituminous coal contain as much as 0.01 percent uranium (on a dry-weight basis) over hundreds of acres in the Fort Union lignite region, but selected samples from outcrops and drill holes that contain more than 0.1 percent uranium show that these rocks rarely ever contain as much uranium as they are capable of holding. The commonly greater concentration of uranium at the top or bottom few inches of thick beds of lignite or subbituminous coal indicates that a physical or chemical restriction to the uniform dissemination of uranium is characteristic of lignite and subbituminous coal. Low permeability to the passage of uranium-bearing solutions plus the relatively rapid fixation of the uranium by the carbonaceous material may partly explain this characteristic.
3. Beds of bituminous and anthracitic coal in this country may contain as much as 0.005 percent uranium of syngenetic origin within areas of a few hundred acres, but deposits in excess of 0.005 percent rarely exceed a few tons in size. The seemingly unfavorable characteristic of bituminous and anthracitic coal as compared with lower rank coal, impure lignite, and lignitic shale as a host for epigenetic uranium may be due in part to the change in chemical composition that accompanies increase in rank of the carbonaceous material but is probably also caused by a lower permeability to the passage of mineralizing solutions.
4. Most of the deposits of uranium in coaly carbonaceous rocks in the United States that contain

more than 0.1 percent uranium are in thin beds of impure lignite and lignitic shale. Reasons for this group being the most favorable host rocks are not apparent. One possibly important, but unevaluated factor is that impure lignite and lignitic shale are probably more permeable than either relatively pure lignite or carbonaceous shale.

5. Beds of carbonaceous shale contain locally high concentrations of uranium at many places, but only a few of the deposits are large enough to have commercial value. In some areas, large tonnages of carbonaceous shale containing an average of 0.01 to 0.02 percent uranium are known. Carbonaceous shale varies greatly in its degree of compaction, permeability, and condition of preservation of the carbonaceous matter so that its favorability as a host rock for uranium might also be expected to vary. Many beds of carbonaceous shale are apparently less favorable host rocks for uranium than impure lignite and lignitic shale, but a few may be almost equally favorable.

REGIONAL GEOLOGIC AND TECTONIC SETTING

If one regards as significant only those known deposits of uranium in coaly carbonaceous rocks that contain at least 1,000 tons of uraniferous rock, there are only about 32 deposits, areas, or properties to be considered (pl. 1; table 8). Many of these deposits occur in the Fort Union lignite region and may be so grouped for the purpose of relating the deposits to the regional geologic and tectonic setting. Selected mines and areas representative of various types of uranium deposits in coaly carbonaceous rocks are listed in table 9 together with their regional geologic and

tectonic setting. The list shows that deposits of uranium in coaly carbonaceous rocks are found in a wide variety of settings. It follows, therefore, that no single setting is prerequisite to the formation of a deposit of uranium in coaly carbonaceous rocks.

Though local structural features are assumed to have local control on the size and shape or habit of individual uranium deposits, they are probably not decisive to the existence of the deposits. The relation of uranium distribution to some kinds of local structural features is mentioned briefly in other parts of the report.

The stratigraphic interval between the deposits in coaly carbonaceous rocks and Precambrian crystalline rocks or other igneous or metamorphic rocks ranges from zero to at least 15,000 feet, with the average being probably close to 10,000 feet. The intensity of deformation also varies widely, from none through slight, moderate, intense, to extremely intense. A diversity of regional geologic and tectonic settings may be said to characterize deposits of uranium in coaly carbonaceous rocks. Many other variables in addition to tectonic setting, interval from deposit to Precambrian crystalline rocks, and deformation intensity could be considered, but the relatively small number of deposits provides an inadequate basis for further speculation.

Most of the 32 significant uranium deposits are in the Fort Union lignite region; hence, their distribution is not random. Evidently some aspect of the regional geology is broad enough to predispose favorably the Fort Union lignite over many hundreds of square miles, yet specific enough to select only a relatively few beds to be enriched in uranium in local areas; for example, the *E* bed in the North Cave Hills area, Harding County, S. Dak., or the Rocky

TABLE 9.—Regional geologic and tectonic setting of selected deposits of uranium in coaly carbonaceous rocks

Host-rock type	Mine or area	Tectonic setting	Interval from deposit to Precambrian, igneous, or metamorphic rocks (feet)	Deformation intensity	Relation to ancient or modern erosion surface
Unconsolidated peaty material. Lignite and subbituminous coal.	Hoffman Meadow, Fresno County, Calif.	Sierra Nevada batholith	Direct contact	None	On present-day surface.
	Fort Union lignite region, western North and South Dakota.	Williston basin	12,000-15,000	Slight	Less than 200 ft below pre-Oligocene surface.
	Red Desert area, Sweetwater County, Wyo.	Great Divide Basin	10,000-15,000	do	Permissive evidence for a close relation to pre-Miocene surface.
Bituminous coal	Fort Scott area, Bourbon County, Kans.	Continental platform	<1,000	do	Near present-day surface.
Impure lignite and lignitic shale.	Old Leyden coal mine, Jefferson County, Colo.	Denver basin, steep flank	9,000	Intense	Directly below a late Cenozoic pediment surface.
	Gas Hills area, Fremont County, Wyo.	Wind River Basin, marginal.	>8,000	Moderate	Permissive evidence for a close relation to any one of several Cenozoic surfaces.
	Fort Union lignite region, western North and South Dakota.	Williston basin	12,000-15,000	Slight	Less than 300 ft below pre-Oligocene surface.
Carbonaceous shale	Fall Creek area, Bonneville County, Idaho.	Overthrust and isoclinally folded Caribou Mountains.	>10,000	Extremely intense	About 700-800 ft below pre-Miocene surface.
	Goose Creek area, Cassia County, Idaho.	Basin and Range province, north margin.	3,000+	Moderate	None apparent.
	Hogback No. 4 mine, McKinley County, N. Mex.	Nose of Zuni uplift, margin of San Juan Basin.	<3,000	do	Permissive evidence for a close relation to middle or late Tertiary surface.

Ridge bed in southern Billings County, N. Dak. One such aspect that may be considered because it fits the requirements stated above is proximity to an ancient surface of erosion or unconformity. A regional unconformity extends for many square miles and locally truncates beds of favorable host-rock lithology.

A reasonably clear relation can be demonstrated between the deposits of uranium and an ancient erosion surface only for the Fort Union lignite region. The principal evidence favoring such an association can be summarized as follows: In the Fort Union lignite region all 10 large low-grade deposits of uranium in lignite are less than 200 feet below the unconformity at the base of the Oligocene or Miocene rock as they are preserved today or of the projected or restored base as it existed before erosion (table 10). In addition, the six areas of high-grade deposits of uranium in impure lignite and lignitic shale that have produced more than 100 tons of uranium ore are within about 300 feet of this same surface (figs. 30, 36; table 10). Although this relation may not be characteristic of all the deposits of uranium in coaly carbonaceous rocks, it seems of critical importance for nearly half of the 32 deposits considered significant.

TABLE 10.—Interval between uranium deposits and the reconstructed surface of truncation at the base of Oligocene or Miocene rocks in the Fort Union lignite region

Locality	Altitude (feet)		Interval (feet)
	Base of truncated surface ¹	Uranium deposit ²	
Uranium deposits in lignite			
Sentinel Butte area, Golden Valley County, N. Dak	3,390	3,300	90
Flat Top Butte area, Golden Valley County, N. Dak	3,290	3,260	30
Bullion Butte area, Billings County, N. Dak	3,345	3,170	175
Chalky Buttes area, Slope County, N. Dak	3,200	3,150	50
Medicine Pole Hills, Bowman County, N. Dak	3,420	3,390	30
Johnson outlier, Perkins County, S. Dak	3,015	3,000	15
Lodgepole area, Perkins County, S. Dak	2,950	2,920	30
Bar H area, Harding County, S. Dak	3,250	3,145	105
Mendenhall area, Harding County, S. Dak	3,330	3,300	30
Ekalka Hills, Carter County, Mont	3,850	3,820	30
Average			58.5
Uranium deposits in impure lignite and lignitic shale			
Klym mine, Billings County, N. Dak	2,970	2,700	270
Johnson mine, Billings County, N. Dak	2,980	2,750	230
L. E. Smith property, Billings County, N. Dak	3,070	2,890	180
Lutheran Church property, Billings County, N. Dak	3,110	2,910	200
North Cave Hills area, Harding County, S. Dak	3,430	3,300	130
South Slim Buttes area, Harding County, S. Dak	3,270	3,235	35
Average			174

¹ Interpolated from contour map of Gill and Denson (1955, p. 234) and supplemented by their field notes.

² Interpolated from contour map of Gill and Denson (1955, p. 234) and supplemented by their field notes and by data from topographic maps.

SEDIMENTARY FEATURES

Studies of lithofacies from outcrop and bore-hole data in the Red Desert area have provided a detailed picture of the uranium distribution and its relation to facies changes of the enclosing rocks (Masursky and Pipiringos, 1959, p. 188–195; Masursky, 1961; Pipiringos, 1956b, p. 79).

During part of early Eocene time a cyclic sequence of fluvialite siltstone, sandstone, coal, and shale was deposited in the Wasatch formation of the Red Desert area marginal to the vast lake where the Green River formation was being deposited. Lacustrine shale, sandstone, and oil-shale beds of the Green River formation on the southwest intertongue with fluvialite rocks of the Wasatch formation on the northeast. Fluvialite sandstone and shale of the Wasatch formation, in turn, intertongue with coarse-grained arkosic sandstone and conglomerate of the Battle Spring formation (Pipiringos, 1955; 1956a, p. 434–436) which thickens and increases in coarseness toward the Green Mountains to the northeast. Coal beds in this area are thickest and contain the least ash along a northwest-trending zone called the Red Desert syncline. Northeast of this zone, the ash content of the same beds increases until they grade into carbonaceous shales that intertongue with the very permeable Battle Spring formation. To the southwest of this zone the thick coal beds thin out between beds of lacustrine shale. The highest concentration of uranium in raw coal (dry-weight basis) lies in a belt along the northeast side of the zone of thick coal beds where they are high in ash. Parallel to this belt, but to the southeast, there is another belt that represents the highest concentration of uranium in coal ash. The two belts exist because of the proportionately greater concentration of inorganic constituents effected during the ashing of low-ash coal as compared with high-ash coal.

Uranium distribution also appears to be related to variations in permeability within individual beds of coal. Thick beds of coal in the Red Desert area generally have an irregular vertical distribution of uranium. Commonly, the concentration of uranium is greatest adjacent to layers with a high percent ash and is least near the middle of thick layers of pure coal. Samples of uraniferous coaly rock studied by Masursky show that the aqueous permeability of impure coal layers is about 0.3 millidarcy, whereas the permeability of pure coal layers is less than 0.1 millidarcy. Thus, the distribution of uranium within a thick coal bed is probably controlled locally by the permeability of the coal itself.

The uranium distribution is also related to variations in the permeability of the enclosing strata. Coal beds that rest directly on coarse-grained, relatively permeable sandstone contain the highest concentration of uranium adjacent to the contact, and in general contain more uranium than beds underlain by relatively impermeable shale.

The distribution of uranium in coaly carbonaceous rocks in the Red Desert area is directly related to variations in permeability of the enclosing strata that is, of the intertonguing formations, of the directly adjacent beds, and of the coal itself. The uranium distribution is also related to facies changes in the lithology of the host rock, with the highest concentration in impure coal when figured on the dry-weight basis but in relatively pure coal when figured on the ashed-sample basis.

The occurrence of high uranium concentration in coaly carbonaceous rocks adjacent to zones or beds of permeable rock has been observed in several areas other than the Red Desert. One of the most obvious examples of this association was observed at a prospect pit in sec. 31, T. 21 N., R. 5 E., in the South Cave Hills area, Harding County, S. Dak. A 2-foot-thick bed of lignitic and carbonaceous shale is there intersected by a group of clastic dikes of lignitic sandstone as much as 2 inches wide (fig. 35). The concentration of uranium ranges from 0.9 percent in the immediate vicinity of the clastic dikes to less than 0.01 percent within a few inches or feet on either side of the dikes (Kepferle and Chisholm, 1956, p. 246-248). Even though the clastic dikes extend only 1 or 2 feet beyond the carbonaceous shale bed, the association of high uranium concentration with these zones of permeability is most impressive.



FIGURE 35.—A small deposit of uranium in lignitic and carbonaceous shale associated with a group of clastic dikes (indicated by arrows). The uranium concentration in the shale diminishes with increasing distance from the clastic dikes.

In many areas deposits of uranium in coaly carbonaceous rocks are closely associated with sedimentary strata containing moderately abundant quantities of volcanic ash and tuff. This association is emphasized by those who would link it to a theory that the volcanic material is the disseminated source for uranium later mobilized, transported, and concentrated in the coaly carbonaceous host rocks. The association is regarded by many as the most important evidence in favor of the theory. As examples they cite nearly every locality where major uranium deposits occur in coaly carbonaceous rocks. Others refuse to regard the association as critical because volcanic rocks are, or were formerly, so widely distributed geographically that it is almost impossible to find any area in the Western United States not now or previously associated with deposits of volcanic debris. From the opponents' viewpoint the theory can be neither proved nor disproved.

A genetic association between uranium deposits and rocks of volcanic origin is possible in the Fort Union lignite region where, as previously shown, all the known significant deposits are less than 300 feet below the base of the pre-Oligocene unconformity. Tuffaceous sandstone and mudstone, bentonite beds, calcareous mudstone, and fine- to coarse-grained arkosic sandstone and conglomerate as much as several hundred feet thick were deposited on this unconformity in Oligocene and Miocene time and have since been eroded away except for isolated remnants on high buttes. The ratio of pyroclastic to detrital material in these rocks probably varies from about 4:1 to 1:10. The nearness of these uranium deposits to the Oligocene and Miocene rocks is shown in figure 36.

The strength of an association between volcanic rocks and any given deposit of uranium is commonly evaluated by the interval or distance between them. Yet interval or distance alone is not geologically significant unless consideration is also given to differences in the permeability of the intervening strata, which is usually unknown. It may be noted that for the six areas of high-grade deposits of uranium in impure lignite, many of which are overlain by sandstone, the average interval between the uranium deposits and the base of the Oligocene or Miocene tuffaceous rocks is almost three times as great as for the 10 areas of large low-grade deposits of uranium in lignite, many of which are overlain by interbedded shale and sandstone. Other variables related to the theory of volcanic origin, and to the strength of the association, are largely unevaluated. These include the type, absolute quantity, and degree of alteration of the volcanic material, its quantitative relation to

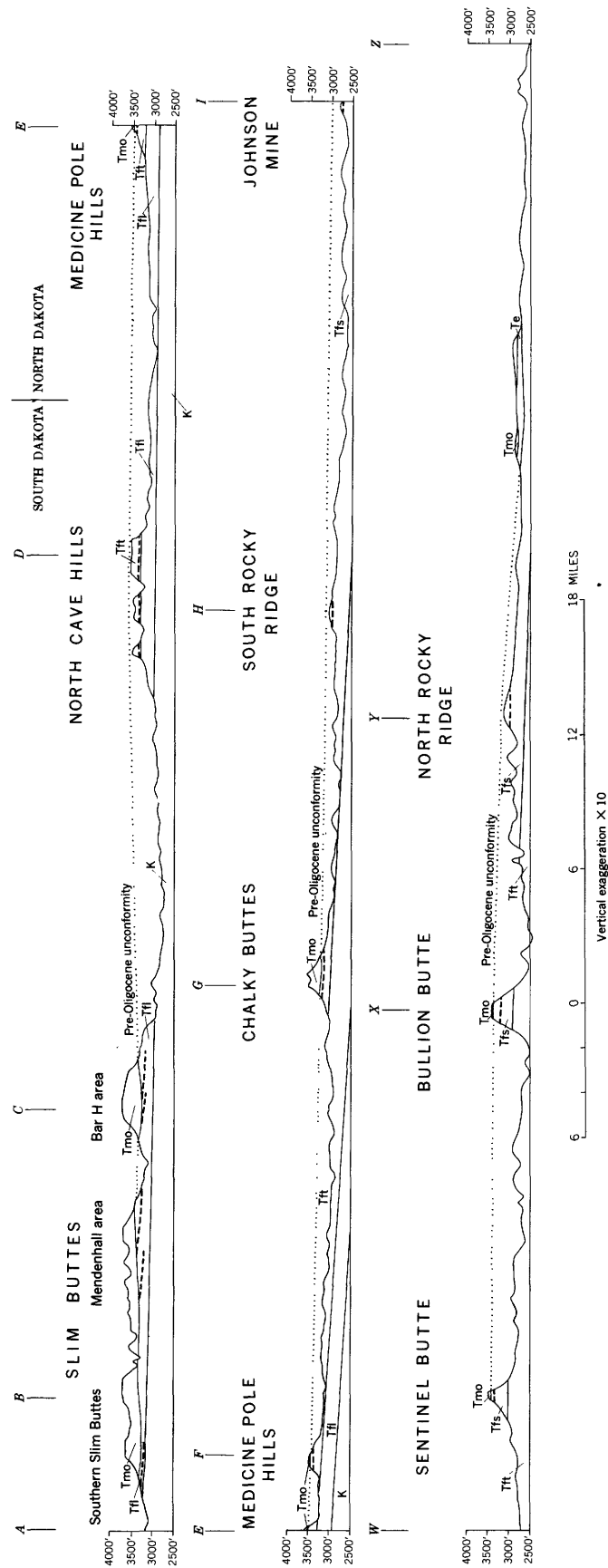


FIGURE 36.—Cross sections showing position of uranium deposits in relation to the pre-Oligocene unconformity in parts of North Dakota and South Dakota. Tmo, Miocene and Oligocene rocks; Tfs, Eocene rocks; Tft, Tft, and Tfs, Ludlow, Tongue River, and Sentinel Butte members of Fort Union formation; K, Cretaceous rocks. Uranium deposit in coaly carbonaceous rock shown by heavy dashed line. Line of cross sections shown on figure 30.

other rock constituents, and the possible relation these factors have to the amount and availability of uranium.

In most other localities the evidence favoring an association is less convincing than in the Fort Union lignite region because of fewer deposits and fewer remnants of volcanic rocks upon which to determine the relative position or distance between volcanic rocks and the uranium deposits. One other area, the Goose Creek area, Cassia County, Idaho, is especially worthy of mention because the uranium-bearing coaly carbonaceous rocks are interbedded with a thick sequence of predominantly volcanic material, thus providing an indisputable close association between the host rocks and volcanic material.

The significance of the observed association between volcanic rocks and uranium deposits in coaly carbonaceous rocks is very largely a matter of personal opinion because the data are too meager and variable to be treated statistically.

URANIUM PROVINCES

The concept of uranium provinces, like the concept of metallogenetic and other geologic provinces, is the natural outgrowth of an attempt to generalize or to place individual localities within the framework of a broader picture. The recognition of a province is a sophisticated extension of the simple prospecting guide that the easiest place to find a new deposit is to search in the vicinity of a known deposit. A possible definition of a uranium province is: an area known to contain deposits of uranium and in which there is reasonable geologic evidence to expect that additional similar deposits may be found. It is generally accepted that ore deposits do, in fact, occur in provinces and that the existence of several similar deposits in an area is evidence in itself for the existence of a province. Though the logic of this line of reasoning is somewhat tenuous, those who believe strongly in the concept make still further assumptions that are difficult to prove or disprove: (a) that there are original inhomogeneities, or element concentration differences, in large segments of the earth's crust; and (b) that through long periods of earth history geologic processes tend to preserve and even increase these differences. In partial defense of these assumptions the province concept is then interwoven with various ideas regarding the differentiation, emplacement, and crystallization of a magma and is further related to stages in the history of orogenic belts, processes of weathering, sedimentation, and eventual recycling of sedimentary material into new igneous rocks. Un-

fortunately, the concept is sufficiently vague that it means many different things to many different people, yet is largely taken for granted without adequate examination of the basic assumptions.

In a report describing uranium provinces Klepper and Wyant (1956, p. 17) assumed that the uranium deposits in the Fort Union lignite region are part of a uranium province that forms a broad belt in and adjacent to the Rocky Mountains from New Mexico and Arizona northward to the Dakotas and Montana. This area does indeed contain many diverse types of uranium deposits but no evidence was presented to suggest that this segment of the earth's crust contains a greater total quantity or greater average concentration of uranium than any other.

A circular line may be drawn on a map (see pl. 1) so as to enclose the area in western North and South Dakota and eastern Montana that includes the most important deposits of uranium in impure lignite and lignitic shale. The area so bounded may be described as the Fort Union uranium-bearing impure lignite and lignitic shale province. By qualifying the province name with the rock name it is hoped to avoid any implication of a genetic relation to other types of uranium deposits in adjacent areas. As thus defined, the Fort Union uranium-bearing impure lignite and lignitic shale province represents an area wherein certain geologic processes have operated to form similar deposits of uranium. Because the geologic conditions and processes that caused these deposits differed in certain details from the conditions and processes that caused uranium to be deposited in sandstone in other areas in South Dakota, Wyoming, and the Colorado Plateau it seems reasonable to regard them as separate provinces. Any apparent tendency for these separate provinces to be clustered should be sought in the similarity of geologic processes such as the chemistry of the ground water favorable for the concentration of uranium in different host rocks, rather than in the original composition of the earth's crust.

GROUND WATER

At least four general hydrodynamic classes of ground water have deposited uranium in coaly carbonaceous rocks: (a) thermal water, (b) artesian water, (c) unconfined water in saturated ground, and (d) vadose water. Thermal water is that activated by vapor pressure in excess of confining pressure as the result of elevated temperatures. Thermal water moves in the direction of decreasing pressure gradient, which is generally towards the earth's surface. Although high pressure may enable thermal water

TABLE 11.—*Hypothetical relation of class of ground water to epigenetic uranium deposits in coaly carbonaceous rocks*

Class of ground water	Position of deposit relative to structure	Symmetry of deposit	Mineralogy	Suggested locality
Thermal.....	Deposits occur adjacent to major vertical rock openings and show preference for structural highs.	Intensity of mineralization increases toward major vertical rock openings and toward the base of the host rock.	High-temperature wall-rock alteration plus primary ore and gangue minerals similar to hydrothermal vein deposits may be diagnostic.	Old Leyden coal mine, Colorado, (some respects only); impure coal from Paraná, Brazil.
Artesian.....	Deposits occur in or adjacent to porous rocks that may serve as artesian aquifers; no preference for structural highs or lows shown.	Intensity of mineralization increases in the direction of highest porosity in or adjacent to the host rock.	Clay minerals may be altered by base exchange. Local cementation by common rock minerals (carbonates, silica).	Red Desert area, Wyoming; possibly also the Cambria coal field, Wyoming, and the Edgemont area, South Dakota.
Unconfined water in saturated ground.	Local deposits may occur along structural lows. Blanketlike deposits may show local variations associated with small structural and permeability changes.	Intensity of mineralization increases toward the top of the host rock.	Common secondary rock minerals are characteristic but not diagnostic.	Most of the large low-grade deposits in the Fort Union lignite region.
Vadose.....	Deposits may occur at or near the ground surface.	Intensity of mineralization increases toward the top of the host rock or close to ground surface.	Oxidation has altered many minerals to forms with higher valence states; for example, pyrite to limonite, gypsum, and jarosite. Some uranium occurs as hexavalent phosphates, arsenates and vanadates, which are diagnostic.	Most of the higher grade deposits that have yellow uranium minerals in weathered impure lignite and lignitic shale in the Fort Union lignite region.

to move long distances through minute rock pores, most thermal water reaches the earth's surface along major rock openings such as faults, which concentrate the flow. Artesian water is activated by a hydrostatic pressure gradient within a confining system such as that provided by a porous sandstone unit underlain and overlain by nonporous shale. The pressures involved depend on the difference in altitude between the points of entrance and escape from the confining system. Small folds that do not interrupt the continuity of the aquifer within the limits of the confining system have little influence on the volume of artesian water flow, but faults of sufficient displacement to offset the entire aquifer have a pronounced effect on the direction and volume of flow. Unconfined water below the zone of water-saturated ground moves primarily in response to gravity, while vadose water in the zone of aerated ground moves in response both to gravity and to surface tension in capillary size openings. The movement of these waters is greatly influenced by changes in porosity and permeability as well as small-scale structural features.

Uranium-bearing ground water may belong to any of the classes described above. The structural position, size, shape, and type of epigenetic uranium deposits are largely determined by the class of ground water that is the transporting agent. The position of a deposit relative to structure, the symmetry of the deposit, and mineralogy all may provide clues as to the class of ground water from which the uranium was deposited. If the class can be determined, then further estimates can be made relative to the probable size, shape, and form of the deposit. Table 11 shows features of epigenetic uranium deposits in coaly carbonaceous rocks that might be related to various classes of ground water.

RELATION OF URANIUM DEPOSITS IN COALY CARBONACEOUS ROCKS TO THOSE IN OTHER HOST ROCKS

By the common system of classifying uranium deposits on the basis of a variety of structural, host-rock, and textural characteristics (for example, vein deposits; deposits in sandstone, limestone, igneous rock, coal, black shale, and phosphorite; deposits in pegmatites), some deposits may be in more than one class. In this report, the host-rock lithology is the only basis for distinguishing the uranium deposits in coaly carbonaceous rocks from other types of uranium deposits. For example, the deposit in the Old Leyden coal mine near Denver, Colo., is here classed as a deposit of uranium in impure coal, but its proximity to hydrothermal vein deposits of uranium in the Colorado Front Range suggests there may also be a genetic relation to vein deposits.

The deposits of uranium in unconsolidated peaty material in the mountain meadows of California are syngenetic deposits because uranium is constantly added from spring waters prior to burial and consolidation of the organic remains. The source of the uranium must be from the igneous intrusive rocks underlying the meadows, but it is not known whether the spring waters leach uranium from preexisting vein deposits in the igneous rock or from uranium disseminated throughout the igneous rock mass, or if the spring water is juvenile thermal water expelled from a cooling magma at depth.

The deposit of uranium in impure coal, coaly shale, carbonaceous shale, and carbonaceous limestone in the Fall Creek area, Bonneville County, Idaho, is restricted to a single stratigraphic zone. The fact that this same stratigraphic zone contains uranium at many widely scattered localities in a variety of structural positions suggests (criterion 1, table 5) that there may be at least some syngenetic uranium con-

concentrated in this zone. The erratic distribution of uranium (criterion 11, table 5) at the Fall Creek coal prospect is difficult to explain by a syngenetic origin, so there must be some epigenetic redistribution and probably some addition of uranium, at least in this one area.

Many deposits of uranium in coaly carbonaceous rocks are geologically similar to epigenetic deposits of uranium in nonmarine sandstone. Though the direct cause of uranium deposition or concentration may differ in the two types, both tend to parallel the stratification in general, although transecting the strata in detail. Chemical environment is probably the chief factor influencing concentration of uranium along the bedding in coaly rocks, whereas permeability may be the principal factor in sandstone. Local discordance with the bedding in both types of deposits indicates an epigenetic origin. Many deposits of uranium of both types have minor structural features that provide local control for the shape of the deposit in detail, and major structural features that control the relative position of the deposit as a whole. However, these features are of secondary importance to the chemical and physical environment provided by, or in, the host rock that determined the primary existence and general size of the deposit.

Important differences exist between epigenetic deposits of uranium in coaly carbonaceous rocks and those in other sedimentary rocks. Unweathered deposits in coaly rocks generally contain no visible uranium minerals even in rock containing as much as 1 percent uranium, whereas both weathered and unweathered deposits of uranium in sandstone generally contain visible uranium minerals. Uranium deposits in coaly carbonaceous rocks tend to be thin tabular bodies that cover more area per quantity of contained uranium than other epigenetic deposits. The thinness may be due in part to the fact that the coaly carbonaceous host rocks are commonly only 1 to 2 feet thick, but even where 5 to 10 feet thick the uranium is generally concentrated in a thin zone near the top or bottom of the bed. Thus, the thin tabular shape is as much a characteristic of uranium deposits in coaly carbonaceous rocks as the absence of visible uranium minerals. Both characteristics are probably due to the direct chemical reaction between the carbonaceous material of the host rock and the uranium-bearing ground water, without precipitation of crystalline mineral matter from solution.

The close association of uranium deposits of different types in the same general area indicates that there may be a common origin for the uranium that happens to become concentrated in different environ-

ments. The interfingering of sandstone and impure coal, both containing uranium deposits in the Gas Hills area, Wyoming, and the close occurrence of uranium deposits in both sandstone and coal in the Pumpkin Buttes and Red Desert areas of Wyoming, emphasize the geologic similarity between these two types of deposits. The similarity poses the problem of how to explain the relative sparsity of uranium deposits in sandstone in the Fort Union lignite region and in coals on the Colorado Plateau where deposits are abundant in sandstone. The answers to this problem must be sought in the local rather than in the provincial abundance of uranium, and in the comparative favorability for uranium where several potential host rocks exist together.

In the South Cave Hills area, Harding County, S. Dak., uranium ore occurs mostly in coaly carbonaceous rocks, but at the Lonesome Pete mine, sec. 24, T. 21 N., R. 4 E., uranium ore has also been mined from a gray phosphatic mudstone. The ore zone, about 6 inches thick, lies in the upper third of a bed of mudstone about 4 feet thick that is overlain by porous sandstone as much as 150 feet thick and underlain by 12 feet of lignite that contains 0.015 to 0.020 percent uranium in the upper 3 feet. The overlying sandstone includes several lenses of shale and lignite that contain less than 0.008 percent uranium. The genetic relation, if any, of the uranium in the lignite to the uranium in the phosphatic mudstone is not known.

In the Temple Mountain area on the southeast flank of the San Rafael uplift, Emery County, Utah, at least three varieties of carbonaceous material may be distinguished—all associated with the uranium deposits, but only one a major constituent of the ore. Liquid petroleum and individual carbonaceous fragments of vascular land plants are associated with the uranium deposits, but a hard massive structureless variety of carbonaceous material that fills pore spaces and locally replaces rock minerals is the principal uranium-bearing material of much of the ore. The uranium-bearing carbonaceous material is insoluble in both carbon disulfide and alkaline solutions. Samples of this uranium-bearing carbonaceous material were studied by Breger and Deul (1959) who found by ultimate, by differential thermal, and by infrared-absorption analyses that this material is similar to a low-rank coal in composition and chemical structure and is unlike bitumens and pyrobitumens such as gilsonite and wurtzilite, which it was formerly assumed to resemble (Kelley and Kerr, 1958). They further suggested that the present insoluble nature of the material may have been caused by cross linkage of

coal molecules as a result of alpha-particle irradiation. To have a composition similar to coal yet be secondarily emplaced as a fluid that is later hardened suggests that the carbonaceous material was probably originally related to humic acids extracted from coaly carbonaceous matter. These substances are avid collectors and transporters of uranium and differ from the other carbonaceous materials described in this report chiefly in being extracted and secondarily emplaced rather than being indigenous (Vine, Swanson, and Bell, 1958).

Different types of carbonaceous materials are associated with the uranium deposits on the Colorado Plateau. Most are related to coal although a few are possibly related to petroleum. Individual fragments of vascular land plants such as wood, bark, and roots may be preserved in several different forms. Commonly they are preserved as vitrain or fusain, and many are partly replaced by mineral matter such as silica, opal, limonite, pyrite, and various uranium and copper minerals. The form of preservation probably determines what role, if any, the carbonaceous material plays in the localization and deposition of uranium. Petroleum and the residual products of petroleum are less frequently in direct association with deposits of uranium than are carbonaceous materials related to coal.

PHYSICAL AND CHEMICAL FORM OF URANIUM IN COALY CARBONACEOUS ROCKS

URANIUM CONTENT OF THE TYPES OF INORGANIC CONSTITUENTS

INHERENT URANIUM

Uranium is not among the 10 major essential elements for plant nutrition or the 5 micronutrients, or even among the nonessential elements most commonly reported in plants as listed on page 121. Investigations by Hoffmann (1942) indicated, however, that uranium is a regular constituent of plants, ranging from 7×10^{-9} to 2.3×10^{-13} grams of uranium per gram of plant substance (0.007 to 0.00000023 ppm), depending on the plant species. Analyses of the ashes of about 30 plants (Hoffman, 1943b) showed a range in uranium content from about 0.000000X to about 0.X percent uranium in the ash. These data probably represent a reasonable estimate of the range and average uranium content of living plants and plant ashes.

Plants growing in a uraniferous environment may be expected to contain greater amounts of uranium. Table 12 lists the uranium contents of living plants collected from certain uranium-rich mountain meadows of the Sierra Nevada region in California. The living trees, shrubs, and grasses of the uraniferous

TABLE 12.—*Uranium content of live plants from uraniferous mountain meadows in California*

[Analyses by C. G. Angelo and G. T. Burrow, U.S. Geological Survey. Determinations made on the basis of dried sample as received in the laboratory]

Sample		Percent		
No.	Description	Ash	Uranium (calculated)	Uranium in ash
Pettit Ranch, Kern County, Calif.				
245811	Green apples.....	3.5	0.00021	0.006
245808	Branch tips and fronds of Giant Cedar (<i>Thuja plicata</i> Donn).....	4.0	.000048	.0012
245802	Common rush (<i>Juncus effusus</i> Linnaeus).....	4.0	.00012	.0029
245810	Leaves and twigs of willow, probably <i>Salix lasiolepis</i> Benth.....	4.7	.000075	.0016
245812	Apple leaves and twigs.....	7.7	.000054	.0007
245801	Stems and seeds of bluegrass (<i>Poa</i> sp.).....	9.0	.000018	.0002
Hoffman Meadow area, Fresno County, Calif.				
245817	Pine needles (<i>Pinus ponderosa</i> Lawson).....	2.5	0.00015	0.006
245822	Corn lily leaves (<i>Veratrum</i> sp.).....	13.0	.00043	.0033
245825	Mixed grasses and herbs.....	15.2	.0038	.025
245823	Algae from a pool of water.....	22.7	.066	.29

mountain meadows contain from 20 to 300 times as much uranium as the maximum reported by Hoffmann (1942). A comparison of 0.29 percent uranium in the ash of algae from the mountain meadow with the 0.00091 percent reported in the ash of algae by Hoffmann (1941) indicates about 300 times as much uranium in the sample from a uraniferous environment. While different plant species growing in the same environment may show different concentrations of uranium, the same species growing in different environments may show even more pronounced differences depending on the availability of uranium.

Spring waters from the uranium-rich mountain meadows contain from 40 to 300 ppb uranium (W. A. Bowes, W. E. Bales, and G. M. Haselton, written communication, 1957), or about 400 to 1,000 times the average uranium content of about 0.1 ppb for natural waters (Fix, 1956). This ratio suggests that living plants concentrate uranium in proportion to the availability of soluble uranium.

Cannon (1952, p. 738-739) cited the work of numerous investigators to show that in small quantities available uranium in the soil stimulates plant development, but the amount that is beneficial in this way is generally only a few parts per million; in higher concentrations uranium is toxic. She also stated that cell activity is inhibited and injury to plant roots results when uranium absorbed by the plant is stored as a yellow deposit in the cell nuclei of the roots. One percent uranium in the soil completely inhibits the germination of seeds.

It is apparent that the intake of uranium by plants varies greatly with species, the quantity available from the soil and water, and, where high concentra-

tions of uranium are found in the soil, with the tolerance of the various species to the poisonous effects. While trace amounts of inherent uranium are probably present in nearly every deposit of coaly carbonaceous material, the concentration even for plants growing in a uranium-rich environment, such as certain mountain meadows of California, rarely exceeds that commonly found in normal sedimentary rocks. Only in the unusual circumstance that the carbonaceous matter is derived chiefly from algae growing in highly uraniferous water is the concentration of inherent uranium likely to exceed 0.005 percent, which is the lower limit of uranium concentration for deposits described in this report. The author knows of no uranium deposits in coaly carbonaceous rocks in which the uranium is thought to be derived chiefly from inherent uranium in the carbonaceous material.

DIAGENETICALLY FIXED ADVENTITIOUS URANIUM

Metals derived from solutions and introduced into a peat bog during the stage of accumulation and decay of plant remains may combine with the carbonaceous matter. Porphyrin-type compounds of vanadium and nickel are known to be common constituents of petroleum but they have not been studied in coal. Uranium porphyrins have not been identified. The salts of organic acids such as humates are commonly associated with coaly rocks (Hawkes, 1957, p. 259). Considerable evidence indicates that uranium humates form readily from the decomposition of vascular plant substances (Szalay, 1954, 1957; Manskaya, Drozdova, and Emelyanova, 1956; Vine, Swanson, and Bell, 1958). Working with low concentrations of uranium in solution, Szalay (1957, p. 29) showed that peat at equilibrium will contain about 10,000 times greater concentration of uranium than associated water. The actual concentration of uranium in samples of dead and partly decayed plant remains collected from the uranium-rich mountain meadows in California is listed in table 13. The highest concentration of uranium on the basis of the dried samples is in the humus-rich soils at or directly below the mat of grass roots. This is rather remarkable in view of the large proportion of rock and mineral fragments in these samples as reflected in the ash contents, which range from 62.7 to as high as 84 percent. Evidently the finely divided and thoroughly decayed plant remains in this zone contain more uranium per weight of organic matter present than coarse woody stems that are not so thoroughly decayed. When calculated on the basis of the ash, some of the woody stems with a low ash content have the highest concentration of uranium. Although the samples listed

TABLE 13.—*Uranium content of dead and decaying plant remains from uraniferous mountain meadows of California*

[Analyses by C. G. Angelo and G. T. Burro. Determinations made on the basis of dried sample as received in the laboratory. Asterisk (*) indicates percent uranium in sample calculated from percent ash and percent uranium in ash]

Sample		Percent		
No.	Description	Ash	Uranium	Uranium in ash
Pettit Ranch, Kern County, Calif.				
245804	Conifer stem submerged in wet humus..	1.0	*0.0003	0.029
245806	Conifer stem submerged in spring.....	2.5	*.0007	.028
245803	Woody rootlets submerged in wet humus.....	5.9	*.0012	.021
245814	Decayed wood submerged in stream....	6.9	*.0076	.11
245809	Gopher droppings.....	20.5	*.0086	.042
245816	Willow stem submerged in wet humus..	30.0	*.0510	.17
245805	Mat of grass roots.....	62.7	.051	.094
245807	Humus directly below root mat.....	70.5	.070	.12
245813	Humus 6 in. below root mat.....	74.5	.058	.085
Hoffman Meadow area, Fresno County, Calif.				
245819	Conifer stem submerged in wet humus..	1.8	*0.0115	0.64
245820	Conifer stem submerged in wet humus..	5.7	*.0775	1.36
245826	Mixed dead pine needles, dry.....	7.7	*.0011	.014
245821	Top 2 in. wet humus soil.....	66.2	.054	.11
245824	Humus directly below root mat.....	76.5	.12	.28
245830	Top 2 in. wet humus soil.....	80.5	.14	.17
245818	Humus directly below root mat.....	84.0	.16	.20

in tables 12 and 13 were not intended to determine the ratio of uranium enrichment between the decaying plant remains and living plants, samples of willow stems and twigs from the Pettit Ranch locality indicate about 680 times more uranium in the decaying plant remains than in the living plant.

The data presented shows that decaying plant remains, where exposed to a replenishable supply of water-soluble uranium, will take up and retain a far greater concentration of uranium than living plants in the same environment. Thus, for any given deposit of uranium in a swamp or bog environment, inherent uranium is generally of small importance as compared to diagenetically fixed adventitious uranium.

DETRITAL URANIUM MINERALS

Fragments of rocks or minerals washed or blown into an environment where plant remains decay and accumulate commonly form the bulk of the mineral matter and ash-forming ingredients of coaly carbonaceous rocks. Such detrital minerals are not an important source for uranium in these rocks because uranium is rarely an important constituent of detrital minerals; if it were, noncarbonaceous detrital rocks would also contain far more syngenetic uranium than any now known. However, uranium in coaly carbonaceous rocks in characteristically small or trace quantities probably is a constituent of the detrital mineral fraction of these rocks. This interpretation is suggested by the direct variation of radioactivity and of percent ash in the coals of the Fernie area, British Columbia (Newmarch, 1950, p. 143).

EPIGENETIC URANIUM MINERALS

Free mineral matter deposited along joints and in pore spaces after coalification contains some of the uranium in highly uraniferous weathered coaly carbonaceous rocks. In the Cave Hills area, Harding County, S. Dak., for example, several hexavalent secondary uranium minerals have been identified in impure lignite and lignitic shale (table 14). However, these secondary minerals are found only in weathered rock that contains more than about 0.1 percent uranium; they probably explain only the high-grade uranium in very small pockets. Secondary uranium phosphate and arsenate minerals, including meta-autunite, metatorbernite, metazeunerite, novacekite, saléite, and abernathyite, occur in the pockets of high-grade ore as coatings on fracture and joint surfaces, or more rarely in the interstices or as impregnations. Carnotite has been reported only in the impure coal at the Old Leyden coal mine, northwest of Denver, Colo. An unusual occurrence of autunite in decayed

wood fragments in unconsolidated peaty material in the Chiquito Creek area of the Sierra Nevada region of California was reported. The locations and sources of data of these and other occurrences of uranium minerals in coaly carbonaceous rocks are listed in table 14. Visible uranium minerals have been reported from several localities in the Fort Union lignite region, and those listed in table 14 are probably typical of the varieties present. These secondary uranium minerals are doubtless the result of weathering processes that have redistributed the uranium originally disseminated in the unweathered carbonaceous matter.

Identification of the black quadrivalent uranium minerals uraninite and coffinite in coaly carbonaceous rocks is particularly difficult because their characteristic small grain size complicates the problem of mineral separation. Neither megascopic nor microscopic methods of visual examination have been adequate to identify these minerals in coaly carbonaceous rocks.

TABLE 14.—Uranium minerals in coaly carbonaceous rocks

Mineral	Locality		Source of data
	Name	Description	
Oxides			
Uraninite (UO ₂)-----	Tepee Butte area-----	S. Dak., sec. 8, T. 22 N., R. 7 E-----	Gruner (1956, p. 504; written communication, 1955).
Becquerelite (7UO ₃ ·11H ₂ O)-----	Long Pine Hills area-----	Mont., sec. 2, T. 3 S., R. 60 E-----	J.R. Gill (oral communication, 1957).
Silicate			
Coffinite (U(SiO ₄) ₁₋₂ (OH) ₁₋₂)-----	La Ventana Mesa area-----	N. Mex., sec. 32, T. 19 N., R. 1 W-----	Gruner and Smith (1955, p. 16-24).
Phosphates			
Autunite (Ca(UO ₂) ₂ (PO ₄) ₂ ·nH ₂ O)-----	South Rocky Ridge area----- Chiquito Creek area-----	N. Dak., sec. 34, T. 137 N., R. 100 W----- Calif., sec. 33, T. 6 S., R. 24 E-----	J. R. Gill (oral communication, 1957). U.S. Atomic Energy Commission (written communication, 1956). Gill (1955, p. 153).
Meta-autunite (Ca(UO ₂) ₂ (PO ₄) ₂ ·nH ₂ O)-----	North Cave Hills area-----	S. Dak., secs. 22, 27, 35, and 36, T. 22 N., R. 5 E-----	White (1958, p. 34-35). White (1958, p. 36-41).
Sodium-autunite (Na ₂ (UO ₂) ₂ (PO ₄) ₂ ·8H ₂ O)-----	Cave Hills area----- North Cave Hills area-----	S. Dak., sec. 27, T. 22 N., R. 6 E----- S. Dak., sec. 26, 35, and 36, T. 22 N., R. 5 E., and sec. 2, T. 21 N., R. 5 E-----	Do. White (1958, p. 47-50).
Hydrogen-autunite (H ₂ (UO ₂) ₂ (PO ₄) ₂ ·8H ₂ O)-----	Southern Slim Buttes area----- North Rocky Ridge area----- South Rocky Ridge area-----	S. Dak., sec. 13, T. 16 N., R. 8 E----- N. Dak., sec. 6, T. 137 N., R. 100 W----- N. Dak., sec. 34, T. 137 N., R. 100 W-----	Do. White (1958, p. 30-33).
Meta-uranocircite (Ba(UO ₂) ₂ (PO ₄) ₂ ·8H ₂ O)-----	Southern Slim Buttes area----- Lupion Campo 2 mine-----	S. Dak., sec. 12, T. 16 N., R. 8 E----- Paraná, Brazil-----	J. W. Adams (written communication, 1957).
Saléite (Mg(UO ₂) ₂ (PO ₄) ₂ ·8-10H ₂ O)-----	Gorham area----- do-----	N. Dak., sec. 7, T. 141 N., R. 99 W----- do-----	J. R. Gill (oral communication, 1957). White (1958, p. 42-43).
Metatorbernite (Cu(UO ₂) ₂ (PO ₄) ₂ ·12H ₂ O)-----	North Cave Hills area-----	S. Dak., secs. 22, 27, 35, and 36, T. 22 N., R. 5 E-----	Gill (1955, p. 153).
Bassetite (Fe(UO ₂) ₂ (PO ₄) ₂ ·8H ₂ O)-----	Gorham area-----	N. Dak., sec. 7, T. 141 N., R. 99 W-----	J. R. Gill (oral communication, 1957).
Sabugalite-saléite(?) ((HAl, Mg) ₁₋₂ (UO ₂) ₂ (PO ₄) ₂ ·16H ₂ O)-----	North Rocky Ridge area-----	N. Dak., sec. 6, T. 137 N., R. 100 W-----	White (1958, p. 43-46).
Arsenates			
Abernathyite (K ₂ (UO ₂) ₂ (AsO ₄) ₂ ·8H ₂ O)-----	Southern Slim Buttes area----- South Rocky Ridge area-----	S. Dak., sec. 27, T. 22 N., R. 6 E----- N. Dak., sec. 34, T. 137 N., R. 100 W-----	White (1958, p. 51-52). J. R. Gill (oral communication, 1957).
Metazeunerite (Cu(UO ₂) ₂ (AsO ₄) ₂ ·8H ₂ O)-----	North Cave Hills area-----	S. Dak., secs. 22, 27, 35, and 36, T. 22 N., R. 5 E-----	Gill (1955, p. 153).
Novacekite (Mg(UO ₂) ₂ (AsO ₄) ₂ ·8-10H ₂ O)-----	do-----	do-----	Kepferle and Chisholm (1956, p. 246).
Vanadates			
Carnotite (K ₂ (UO ₂) ₂ (VO ₄) ₂ ·3H ₂ O)-----	Old Leyden coal mine-----	Colo., sec. 28, T. 2 S., R. 70 W-----	Wilson (1923).

Gruner and Smith (1955, p. 16-24) first identified coffinite in a sample of uranium-rich coaly shale from the La Ventana Mesa area, New Mexico, by X-ray methods. Later, both uraninite and coffinite were reported from lignitic shale samples from South Dakota, though the broad, diffuse X-ray line patterns from which identification was made suggest that the particles are of colloidal dimensions (Gruner, 1956, p. 504).

EPIGENETICALLY FIXED ADVENTITIOUS URANIUM

Epigenetically introduced metals combine with carbonaceous matter in much the same way as diagenetically introduced metals. Such differences as may exist are chiefly tenuous differences in the distribution of the metal within the bed. Differences may exist both in the types and in the amounts of organic constituents favorable for reaction with solution, due largely to physical and chemical changes that accompany coalification. Woody carbonaceous fragments are generally less permeable than attrital plant debris during the peat stage, whereas the reverse seems to be true when these constituents are consolidated to form vitrain and attritus. Metal-bearing waters that flow into a peat deposit have relatively free access to the entire deposit, but after burial and consolidation both access and flow are highly restricted by the impermeability of the deposit and of the adjacent rocks. As a result, both the vertical and horizontal distribution of uranium is quite erratic in epigenetic deposits.

With the exception of the uranium deposits in present-day mountain meadows, most of the significant deposits of uranium in coaly carbonaceous rocks have an erratic distribution of uranium and therefore are thought to have been epigenetically fixed adventitious deposits.

VARIABILITY OF AMOUNT OF URANIUM

The amount of uranium contained in coaly carbonaceous rocks is closely dependent on the amount and relative proportions of the five classes (p. 121) of inorganic constituents in carbonaceous rocks, other factors being equal. Normally, the uranium content of coaly carbonaceous rocks is an insignificant trace amount approaching the lower limit of detection. That in normal coaly carbonaceous rocks is derived from the inherent, detrital, and diagenetically fixed adventitious uranium present at the time of burial. Relatively pure coal that has not been disturbed has a very low permeability which inhibits the later introduction of epigenetic uranium. Uranium that occurs as an insignificant trace metal constituent of

carbonaceous rocks may vary directly or indirectly with the amount of ash, depending on whether or not it is a part of the major ash-forming inorganic constituents. Commonly, the amount of uranium in detrital minerals exceeds the organically fixed uranium and hence varies directly with the percent ash, as indicated by the direct relation of ash to radioactivity noted by Newmarch (1950, p. 143).

Because coaly carbonaceous rocks owe their favorable host-rock characteristics to the carbonaceous matter, one might expect to find that uranium concentration would vary inversely with the ash content. Actually, the quantity of uranium in deposits that occur in coal, impure coal, and coaly shale locally may vary either directly or inversely with the ash content. The inconsistent relation of uranium to ash in these deposits is partly the result of different sampling scales and methods, but is partly also a function of variable permeability and other factors such as degree of mineralization, which may increase or decrease the ash-forming ingredients of a carbonaceous rock. Whereas an inverse relation may be found between uranium and ash in closely spaced samples collected along a single thin stratigraphic zone where permeability is not an important variable, a direct relation to ash may be found in closely spaced column samples collected across a thick bed where permeability to epigenetic solutions is a variable factor that is dependent on ash content.

It has been tacitly assumed by many investigators that if the uranium in coaly carbonaceous rocks is syngenetic it should have a fairly uniform distribution in which the variation could be related to regional changes in lithology and in which any local variation would be on the scale of, and related to, the variations in the major carbonaceous or mineral constituents that might represent the principal locus of deposition. In contrast to this, most of the deposits that have been studied in detail, with the exception of the several mountain-meadow deposits, show a pronounced local variation in uranium of about the same scale but almost independent of the variations in major carbonaceous and mineral constituents, and larger scale variations unrelated to regional lithologic changes. A single 25-pound sample of relatively unweathered lignite from the Evangelical Lutheran Church property, Billings County, N. Dak., was broken into 16 pieces, each for separate analysis. The uranium content ranged from 0.02 to 0.6 percent and had a mean value of 0.15 percent. Whether such an extreme variation in uranium content within a small sample is typical of unweathered, highly uraniferous lignite cannot be known without further investiga-

tion. However, it is suggestive of the range of uranium content that is commonly associated with epigenetic uranium deposits in other types of host rocks.

A vertical exposure of lignitic and carbonaceous shale at a prospect in the South Cave Hills area, sec. 31, T. 21 N., R. 5 E., (fig. 35) shows a range of nearly two orders of magnitude in uranium content within a distance of approximately 1 foot (Kepferle and Chisholm, 1956, p. 247), and a total range of 0.002 to 0.9 percent uranium. The uranium content is highest in several small areas 3 to 10 inches across, near and between sandstone dikes that cut the host rock but that are not directly in contact with the dikes. Visible secondary uranium minerals are confined to those areas of rock having the highest uranium content. Similar abrupt variations in uranium content were observed along the stripped top surface of lignitic shale in the Riley Pass area of the North Cave Hills, Harding County, S. Dak. The weathered outcrops of uranium deposits in coaly carbonaceous rocks containing more than 0.1 percent uranium are characterized by a variable content of uranium, but bore-hole drilling in areas of both weathered and unweathered uranium deposits in the North Dakota area indicates that the distribution of uranium in impure lignite and lignitic shale may be even more erratic in unweathered deposits than in weathered deposits. The large low-grade deposits of uranium in lignite and subbituminous coal also show a pronounced variation in uranium content but generally on a larger and less erratic scale than the high-grade deposits, probably because the total range is restricted.

RELATION OF URANIUM TO CARBONACEOUS AND MINERAL CONSTITUENTS

The occurrence of uranium in coaly carbonaceous rocks in excess of that due to the presence of minerals that contain uranium as a major element raises a question regarding the distribution of uranium among various carbonaceous and mineral constituents of the rock. Field investigations offer no consistent evidence of a direct relation between amounts of uranium and carbonaceous matter or between amounts of uranium and mineral matter of coaly rocks. In different areas investigated, relations have been observed to vary somewhat according to the scale of investigation and to the range in percent ash and uranium found. Pipiringos (1956a, p. 436) noted an increase in uranium content in a single coal bed from about 0.002 to 0.009 percent with a corresponding increase in ash content of about 17 to 42 percent over a distance of about $7\frac{1}{2}$ miles through the central part of the Great Divide Basin. Masursky (1961)

stated that in the eastern part of the Great Divide Basin the highest uranium content within a coal bed occurs in impure coal layers adjacent to partings or layers of high ash content, whereas the lowest content of uranium occurs in the central part of pure coal intervals. The uranium content of coaly rocks in the Fall Creek area ranging from about 44 to 80 percent ash is, in general, inversely proportional to the ash content (Vine, 1959, p. 273). Samples from the La Ventana Mesa area containing more than 0.1 percent uranium range from about 15 to 78 percent ash and average about 50 percent; this content, similar to that of samples containing little or no uranium, thus indicates no consistent relation of uranium to ash in that area. Petrographic studies of core samples of uraniferous lignite from various parts of the Dakota area (J. M. Schopf, R. J. Gray, and C. J. Felix, written communication, 1955) indicate an erratic relation of uranium concentration to percent ash. A similar study of high-grade deposits found the highest concentrations of uranium in coal with 10 to 18 percent ash (J. M. Schopf and R. J. Gray, written communication, 1956).

Laboratory investigations have generally been more successful than field investigations in defining the relation of uranium to the carbonaceous and mineral constituents of coaly rocks. Among the techniques tried, probably the most useful have been analyses of hand-picked samples, heavy liquid separations, flotation and air classification, and statistical analysis of chemical data.

Hand-picked samples of uraniferous lignite from the Slim Buttes area of South Dakota containing about 15 percent ash (as contrasted with 30 percent average for the entire bed) were analyzed for uranium by chemical means and for other metals by spectrochemical means to determine whether the uranium would show an enrichment in the low-ash samples and whether the uranium is concentrated with the carbonaceous material alone or went along with some other metallic constituent that is enriched in the organic matter (Brown, Schimmel, and Nelson, 1952, p. 21; Brown, Coleman, and Schimmel, written communication, 1953). The analyses showed an increase in uranium content such as would be expected if the uranium were associated with the carbonaceous matter. Some other elements also showed a slight increase in the selected low-ash samples, but the fact that the increase was not as great as for uranium probably indicates that the uranium was not associated with other metallic constituents. Handpicked samples from lignite exposed on Sentinel Butte, Golden Valley County, N. Dak., in which the predominant mineral

TABLE 15.—*Uranium and principal minerals of hand-picked samples from lignite exposed on Sentinel Butte, Golden Valley County, N. Dak.*

[Uranium analyses on air-dried samples; not corrected for surface moisture or water of hydration. After Ewing and others (1950, p. 21)]

Sample	Uranium (percent)	Estimated mineral composition	
		Constituent	Percent
Bulk lignite	0.0085	Quartz	35
A	.0013	Analcite	30
		Carbonaceous	25
		Others	10
B	.0017	Gypsum	60
		Carbonaceous	20
		Jarosite	15
		Analcite	5
C	.0031	Jarosite	45
		Carbonaceous	25
		Gypsum	20
		Analcite	10
D	.0042	Jarosite	65
		Gypsum	20
		Carbonaceous	15
E	.0048	Magnetite, hematite, goethite, and limonite	60
		Gypsum	40

was from 10 to 100 times more abundant than in the bulk lignite were analyzed for uranium (Ewing and others, 1950, p. 15–20). The results of this study, as shown in table 15, indicate that the uranium in the mineral constituents in the lignite is considerably depleted.

Several laboratories have separated carbonaceous and mineral-rich fractions of uraniferous coaly rock by the use of heavy liquids. Breger, Deul, and Rubinstein (1955, p. 208–210) obtained a concentrate of carbonaceous material from the uraniferous lignite at the Mendenhall strip mine, Harding County, S. Dak. (table 16). The original sample contained about 32 percent ash and 0.12 percent uranium in the ash, whereas the light (carbonaceous) separate contained only about 14 percent ash and 0.31 percent uranium in the ash. The carbonaceous fraction represented about 80 percent of the original sample but

contained 93 percent of the total uranium present in the original sample. A similar separation was obtained from a sample of coal from the Red Desert area, Wyo. (Breger, Deul, and Meyrowitz, 1955, p. 612–615). The original sample contained about 13 percent ash and 0.017 percent uranium in the ash, whereas the light (carbonaceous) separate contained only about 9.5 percent ash but 0.02 percent uranium in the ash. The carbonaceous fraction represented about 92 percent of the original sample but contained 99.7 percent of the total uranium present in the original sample. The greater concentration of uranium in the carbonaceous fraction in each sample indicates that the uranium is associated primarily with the carbonaceous rather than the mineral fraction of coal and lignite. Sink-float tests conducted by Brown, Schimmel, and Nelson (1952, p. 29) on unsized uraniferous lignite showed a slight enrichment of uranium in the float fraction. The weight-percent distribution of total uranium between the two fractions corresponds very closely to the distribution of total carbon.

Analyses of samples from four 500-ton shipments of ore-grade uraniferous lignitic shale obtained from the U.S. Atomic Energy Commission office, Grand Junction, Colo., are shown in table 17. Chemical determinations of the ash content, uranium, phosphorus, arsenic, and selenium were made on the original samples and also on light and heavy fractions separated in a mixture of carbon tetrachloride and bromoform. The specific gravity of the heavy liquid used for separation into light and heavy fractions was adjusted for each sample so as to provide an adequate size sample in both fractions to permit the desired analytical work. The specific gravity at which each fraction was separated was not determined. In order to make

TABLE 16.—*Percent uranium in carbonaceous and mineral-rich fractions separated by heavy liquid*

Original sample		Carbonaceous (light) fraction				Mineral-rich (heavy) fraction			
Ash	U in ash	Ash	U in ash	Weight relative to whole		Ash	U in ash	Weight relative to whole	
				Fraction	Uranium			Fraction	Uranium
Lignite from Mendenhall strip mine, Harding County, S. Dak.									
[Data from Breger, Deul, and Rubenstein (1955, p. 208-212)]									
32.4	0.12	13.8	0.31	80	93	155-75	10.014-0.017	20	7
Coal from Red Desert area, Sweetwater County, Wyo.									
[Data from Breger, Deul, and Meyrowitz (1955, p. 612-614)]									
13	0.017	9.5	0.02	92	99.7	89.6	0.0006	8	0.3

¹ Range for different sieve fractions.

the data comparable, with reference to the original sample, the percentage of total ash, uranium, phosphorus, arsenic, and selenium was calculated for the various light and heavy fractions. Comparison of these percentages with the percentages of the total dry weight represented by each fraction, gives the true proportion of each constituent in the light and the heavy fractions.

Uranium consistently shows a direct relation to the light (carbonaceous) fraction (enrichment ratio greater than 1); this relation provides further evidence that the uranium is related to the carbonaceous matter, even in bulk quantities of ore-grade rock. Of the three other elements investigated, phosphorus is consistently enriched in the high-ash fraction, whereas arsenic and selenium show no consistent enrichment in either fraction.

RELATION OF URANIUM TO THE TYPES OF CARBONACEOUS CONSTITUENTS

The association of uranium with the carbonaceous matter of coaly carbonaceous rocks is reasonably well established from the study of carbonaceous mineral fractions of uraniferous coaly rocks. Because coal is a heterogenous mixture of different types of carbonaceous constituents with widely differing chemical and

physical properties, it seems reasonable to expect that these various constituents may differ considerably in their capacity to hold uranium. Several laboratories have attempted to establish a relation between uranium and various coal components by examination of thin or polished sections of coal under high magnification.

A comparison of the petrographic composition of uraniferous lignite from the Slim Buttes area of South Dakota with that of uraniferous coaly shale from the Goose Creek area of Idaho led Schopf and Gray (1954, p. 9) to suggest that plant material that has been most subject to decay is the most receptive to uranium emplacement. This interpretation is based on the discovery that many of the more highly uraniferous layers in these deposits contain a very large percentage of translucent attritus, including amorphous humic matter, yellow waxy matter, and fungal remains, as compared with anthraxylon or fusain. More detailed petrographic studies of lignite from western North and South Dakota, including beds containing more than 0.1 percent uranium, indicate that woody layers contain less uranium than attrital layers (Schopf, Gray, and Felix, written communication, 1955; Schopf and Gray, written communication, 1956). The attrital layers, which generally contain more than

TABLE 17.—Distribution of ash, uranium, phosphorus, arsenic, and selenium in light and heavy fractions of uraniferous lignitic shale

[Analyses by W. W. Niles, E. J. Fennelly, H. H. Lipp, C. G. Angelo, L. F. Rader, Jr., Claude Huffman, G. T. Burrow, and J. P. Schuch]

Fraction	Percent of whole, by weight	Constituent										Ratio of percentage of total component in fraction to weight percentage of fraction				
		Percent of sample or fraction (dry weight)					Percent of total component, each fraction									
		Ash	U	P ₂ O ₅	As	Se	Ash	U	P ₂ O ₅	As	Se	Ash	U	P ₂ O ₅	As	Se
Klym mine, sec. 26, T. 142 N., R. 99 W., Billings County, N. Dak. ¹																
Whole sample.....	100	49.8	0.32	0.15	0.029	0.0010	-----	-----	-----	-----	-----	-----	-----	-----	-----	-----
Light.....	22.3	37.8	.36	.18	.018	.0008	16.3	33.7	17.1	14.3	10.3	0.73	1.51	0.77	0.64	0.46
Heavy.....	77.7	55.4	.20	.25	.031	.0020	83.7	66.3	82.9	85.7	89.7	1.07	.85	1.07	1.10	1.15
Smith No. 1 mine, sec. 6, T. 137 N., R. 100 W., Billings County, N. Dak. ¹																
Whole sample.....	100	38.4	0.095	0.06	0.036	0.0150	-----	-----	-----	-----	-----	-----	-----	-----	-----	-----
Light.....	74.8	29.7	.12	.02	.031	.0100	61.0	80.6	59.7	67.1	70.3	0.82	1.08	0.80	0.90	0.94
Heavy.....	25.2	56.4	.086	.04	.045	.0125	39.0	19.4	40.3	32.9	29.7	1.55	.77	1.60	1.30	1.18
Johnson mine, sec. 3, T. 140 N., R. 100 W., Billings County, N. Dak. ²																
Whole sample.....	100	48.5	0.30	0.16	0.13	0.0005	-----	-----	-----	-----	-----	-----	-----	-----	-----	-----
Light.....	64.1	40.7	.19	.13	.12	.0004	55.4	72.3	60.8	66.0	70.5	0.87	1.13	0.95	1.03	1.10
Heavy.....	35.9	58.5	.13	.15	.11	.0003	44.6	27.7	39.2	34.0	29.5	1.24	.77	1.09	.95	.82
Last Chance Nos. 1 and 2 mines, secs. 21 and 22, T. 22 N., R. 5 E., Harding County, S. Dak. ³																
Whole sample.....	100	42.7	0.094	0.10	0.069	0.0008	-----	-----	-----	-----	-----	-----	-----	-----	-----	-----
Light.....	60.5	27.8	.11	.06	.062	.0008	38.7	77.1	50.5	52.4	67.1	0.65	1.27	0.83	0.87	1.11
Heavy.....	39.5	67.6	.05	.09	.086	.0006	61.3	22.9	49.5	47.6	32.9	1.55	.58	1.25	1.20	.83

¹ The light and heavy fractions were separated in a solution of carbon tetrachloride and bromoform mixed in the following proportions: 300 ml carbon tetrachloride plus 110 ml bromoform.

² The light and heavy fractions were separated in a solution of carbon tetrachloride and bromoform mixed in the following proportions: 300 ml carbon tetrachloride plus 51 ml bromoform.

³ The light and heavy fractions were separated in a solution of carbon tetrachloride and bromoform mixed in the following proportions: 600 ml carbon tetrachloride plus 161 ml bromoform (a filtered mixture of the solutions listed in ¹ and ² above).

10 percent ash, are probably more permeable than the woody layers. However, all organic components of the lignite apparently contain uranium, and the relative proportions of the organic constituents are of secondary importance to the position of lignite layers with respect to zones of permeability within or adjacent to the beds of coal. Similar petrographic studies of uraniferous lignite from the Slim Buttes area of South Dakota (Koppe and others, 1955, p. 30-33) also suggest that none of the petrographically distinguishable constituents, either organic or inorganic, are intimately related to the concentration of uranium.

A detailed petrographic study and comparison was made of two cores from the impure coal and carbonaceous shale deposits of the Salt Lake formation of Pliocene age, Goose Creek area, Cassia County, Idaho (R. J. Gray, written communication, 1957). Though one core contained much highly uraniferous carbonaceous rock and the carbonaceous rock in the other was almost nonuraniferous, there was little difference in the gross composition of the carbonaceous materials present in the two cores. The core that contained the most uraniferous carbonaceous material was associated with much-altered volcanic material, whereas the nonuraniferous core was associated with unaltered volcanic material. Uranium content of the more uraniferous core, considered alone, shows a direct relation to the carbonaceous matter, which characteristically is minutely fragmented and contains abundant humic matter and yellow waxy amorphous matter.

The more highly uraniferous coal layers in a core from the Lumen coal zone of the Red Desert area (J. M. Schopf and R. J. Gray, written communication, 1955) contain about 59 percent translucent attritus, or 10 percent more translucent attritus than the less uraniferous layers; this translucent attritus in the more highly uraniferous layers has about 6 or 7 times as much yellow waxy matter as the average for this coal. The yellow waxy matter includes cuticle, spores, yellow attrital resins, amorphous waxy material, and wax in clay. Thus, coal layers with an abundance of yellow waxy matter in the Red Desert coal would seem to be a more favorable environment for deposition of uranium, provided other conditions of availability and access to the source of uranium are equal. Though direct evidence for the origin of the amorphous waxy material is lacking, J. M. Schopf and R. J. Gray (written communication, 1955) suggested the possibility that it is derived from the plant wax known as suberin, which is sometimes abundant in the corky tissues of angiospermous plants.

The fact that the vertical distribution of uranium within a thick unit of coaly rocks so commonly shows a markedly greater variation than does that of coal petrographic constituents of selected sections suggests that some factor other than the petrographic types of coal constituents is the primary control for uranium distribution. The evidence available certainly does not prove, however, that no relation exists between uranium and the types of organic constituents in coaly rocks. In order to define such a relation, care must be taken to compare only those samples with differing organic composition that have been uniformly exposed to mineralizing solutions. The examination preferably should be restricted to samples from a single thin layer or stratum of rock.

Autoradiographic studies of thin sections and polished blocks of uraniferous lignite from the Slim Buttes area of South Dakota (Koppe and others, 1955, p. 16-17) show a possible positive correlation between alpha radiation and small crystals of barite (White, 1958, p. 66) that fill the cell lumina of fusain fragments. The radioactivity associated with the barite is probably derived from radium.

Though the mineral forms of uranium certainly explain part of the uranium present in some coaly carbonaceous rocks, the physical and chemical form of much of the uranium is unknown. Its association with the carbonaceous matter is well established and there is some indication that it shows a preference for the more highly decayed matter that forms layers of translucent attritus in coal. Translucent attritus is a heterogeneous mixture of finely divided plant particles. Layers of translucent attritus that contain abundant yellow waxy matter, possibly derived from the plant wax known as suberin, seem to be the most favorable for the deposition of uranium in the Red Desert area. Further understanding of the form of the uranium requires a study of the chemical state, availability, and reactivity of the uranium associated with carbonaceous matter.

CHEMICAL STATE AND REACTIVITY OF URANIUM

Because the physical and chemical form of uranium bound by carbonaceous material cannot be identified by usual mineralogic and petrographic techniques, investigation of the chemical state and reactivity of the uranium is necessary to narrow the numerous possibilities.

VALENCE STATE OF URANIUM

In an attempt to determine the valence state of uranium in coaly carbonaceous rock, G. J. Neuerburg supervised separation of hexavalent uranium from quadrivalent uranium using the following procedure: A

piece of unweathered coaly shale from a depth of 102.9 feet from core hole 72 in the Red Desert area (Masursky, 1961) and containing 61 percent ash and about 40.6 ppm uranium was selected for the experiment. The sample was ground and then leached with ammonium carbonate, which should selectively dissolve the sexivalent uranium but not the quadrivalent uranium. Dissolved humic acids stained the leachate brown. The humic acids were flocculated and each fraction, the humic acids, the leachate, and the leached residue was analyzed separately for uranium. The results are given below, in parts per million of uranium.

Original sample	Humic acids	(NH ₄) ₂ CO ₃ leachate	Leached residue
40.6	0.20	30.4	10.0

A negligible quantity of uranium was retained in the humic acids. About three-fourths of the uranium was in the ammonium carbonate leachate and is presumed to be present in the sexivalent state. About one-fourth of the uranium remained in the leached residue and is presumed to be in the quadrivalent state. Uraninite discernable by X-ray powder photographs generally contains at least 30 percent and as much as 80 percent sexivalent uranium (Garrels and Christ, 1959). Thus, at least one-third and perhaps most of the uranium present in this unweathered sample may be present in the form of the mineral uraninite.

SOLUBILITY

The form of occurrence of uranium in coaly carbonaceous rocks is suggested by the solubility of the uranium under various natural and laboratory conditions. Laboratory experiments on the extraction of uranium from aqueous solution by peat and low-rank coal (Szalay, 1954, p. 304; Moore, 1954, p. 654) indicate that once the uranium is taken up by the carbonaceous matter it is fixed and cannot be removed or separated from the carbonaceous matter by leaching with distilled water. In other words, the chemical bond between uranium and carbonaceous matter is sufficient that the reaction cannot be reversed by using a more dilute solution. In contrast to coaly carbonaceous material, charcoal will extract uranium from solution but the uranium can then be removed by leaching with distilled water (Tolmachev, 1943; also Belcher and Lutz, 1950, p. 6-8). The fact that uranium is so readily leached from charcoal seems to indicate that it is held by physical rather than by chemical adsorption. The extraction of uranium from solution by coaly carbonaceous material must be more complex and it is therefore presumed to be held by a process other than physical adsorption.

Szalay (1954, p. 304) made further experiments that showed 1 percent acetic acid will remove part of the uranium from peat and 1 percent hydrochloric acid will remove nearly all of it. Elutriation experiments in which columns of coal were leached with 1*N* and 6*N* hydrochloric acid (Breger, Deul, and Rubinstein, 1955, p. 215-224) showed that the removal of uranium takes place at a pH of slightly less than 2.2. The data further show that the percent uranium in the ash of the dry solid residue remaining after evaporation of the solution collected from the base of the column varies inversely with the percent ash; in other words, the uranium content varies directly with the carbonaceous material content in the total solids.

In an attempt to determine whether the uranium in weathered lignite is soluble in several common reagents the following experiment was performed with the assistance of C. A. Horr: A sample of friable weathered impure lignite from the Pickpocket claim in the North Cave Hills, Harding County, S. Dak., was selected for the experiment. The original sample contained 1.96 percent uranium. The percentages of the original uranium soluble in hot and cold water, in several organic solvents, in dilute sulfuric acid, and in dilute sodium carbonate solution are shown in table 18.

It is noteworthy that uranium is more soluble in water than in common organic solvents. The unusually low solubility in organic solvents indicates that the uranium probably does not occur in combination with hydrocarbons normally soluble in organic solvents. Thus, the association of uranium in coaly rocks with organic matter probably is different from that of the small amount of uranium in hydrocarbons like petroleum and natural bitumens that are soluble in organic solvents.

Mild oxidation induced by weathering converts part of the coaly carbonaceous matter to water-soluble humic acids that can transport uranium in solution. As much as several thousand parts per billion uranium has been found in samples of dark-brown water collected from pools of rain water that have

TABLE 18.—*Solubility of uranium in weathered lignite*

Extractant	Percentage of original uranium extracted
Cold water.....	0.31
Hot water.....	.41
Acetone.....	.087
Benzene.....	.0008
Carbon disulfide.....	.003
5 percent sulfuric acid (cold).....	81.63
5 percent sodium carbonate (cold).....	93.88

washed a fresh exposure of uraniferous lignitic shale. The color of the water is probably due to soluble humic acids extracted from the weathered carbonaceous rock. The bulk of the carbonaceous matter may in time be dissolved, for as shown by Wheeler (1931, p. 344) the carbonaceous constituents of coal consist of a mixture of three groups of chemical compounds: (a) the extractable resins and hydrocarbons; (b) the resistant plant products, including skins, spore exines, cuticles, and fusainized cellular tissues; and (c) ulmins, or coalified humic material which is the brown matter derived through the decay of the cellulose, proteins, and carbohydrates that compose from 70 to 90 percent of the carbonaceous matter in coal and can be oxidized to form soluble humic acids.

An experiment was performed to determine how much uranium could be removed from coaly carbonaceous rock by the extraction of humic acids. Humic acids, constituting about 7 percent of the dry weight of the original sample, were extracted in a 0.5*N* solution of Na_2CO_3 (pH about 11) from a sample of weathered lignitic shale containing 0.23 percent uranium. The extracted humic acids were separated and found to contain about 0.32 percent uranium, which was about 10 percent by weight of the total uranium in the original sample. Sodium carbonate ground water is relatively common in certain areas including the Fort Union lignite region, and it evidently transports the soluble humic acids extracted from weathered lignite and at the same time removes some of the uranium.

Though intended primarily to test the maximum uranium-sorption capacity of a coaly carbonaceous rock, the following experiment described by Masursky (1955, p. 162-163) also raises some interesting questions that warrant further investigation regarding the solubility of uranium: A representative sample of subbituminous coal from the Red Desert area was ground to minus 100 mesh and immersed in a uranyl nitrate solution containing 990 ppm uranium. After a period of 27 days the coal had extracted 95 percent of the uranium from the solution, and the coal contained 8.6 percent uranium. After air drying, the sample was reimmersed in a fresh solution containing 550 ppm uranium for a period of 120 days. At the end of this time the coal had extracted 35 percent of the uranium from the solution, and the concentration of uranium in the coal had increased to nearly 13 percent. At this point the coal was oven dried at a temperature of about 100°C and then immersed in a final solution containing 980 ppm uranium. At the end of 11 days the solution contained about 18 percent more uranium than was present at

the start. The loss of uranium from the coal in the final step of the experiment suggests that the heat involved in drying the sample altered the capacity of the coal to extract and retain uranium. A polished section of the coal fragments that had been soaked in the uranyl solutions showed that the fragments were surrounded by black particles that may have been uraninite. X-ray examination of the material by E. J. Dwornik also indicated that uraninite was probably present. The effect of elevated temperature on the ability of coaly carbonaceous rocks to extract and retain uranium, the formation of uraninite, and the solubility of the uranium need further investigation.

URANIUM SORPTION AND ION-EXCHANGE CAPACITY OF CARBONACEOUS MATERIALS

The sorption capacity of coaly carbonaceous rocks for uranium has been noted (p. 155-156). Experiments by Moore (1954) indicated a high capacity of peat, lignite, and subbituminous coal to extract and retain uranium from solution. These materials extracted 98 percent or more of the uranium from a solution of uranyl sulfate containing 200 ppm uranium at a pH of 2.45. Under similar conditions bituminous coal extracted only 17 percent, anthracite 34 percent, and white pine wood 40 percent of the uranium from solution. The discovery of uranium-bearing coal in Hungary led Szalay (1954, p. 303-306) to conduct a series of uranium-sorption experiments with coal and other carbonaceous materials. Partly decayed plant debris that had been cleaned of extraneous mineral matter was powdered and then placed in a filter paper funnel and a solution of 1 percent uranyl nitrate poured over the material. No uranium was detected in the filtrate, the uranium having been extracted by the plant debris, and no uranium was released from the plant debris by washing with distilled water. A solution of 1 percent hydrochloric acid was required to remove most of the uranium from the plant debris. Szalay stated that the sorption was completed within a few minutes or seconds. Experiments with lignite and brown coal (unconsolidated lignite) yielded similar results. The effectiveness of the extraction was not altered by use of a solution of sodium uranyl carbonate in place of the uranyl nitrate.

In the same report Szalay described the results of an experiment to determine the maximum quantity of uranium that could be taken up by lignite and peat. Lignite became "saturated" with about 50 mg uranyl nitrate per gram (dry weight) of lignite, but peat took up as much as 250 mg per gram, about 2 percent by weight of uranium for lignite and about 8 percent by weight of uranium for peat. These con-

centrations compare with a maximum of nearly 13 percent uranium in subbituminous coal that Masursky (1955, p. 162-163) got by repeated immersions in a uranyl nitrate solution.

In order to isolate the chemical constituent of lignite responsible for holding the uranyl ion, Szalay also experimented with a commercial humic acid and with humic acid extracted from lignite. Both these substances showed a capacity to extract and retain the uranyl ion from solution that was equal to or greater than the capacity of the original lignite. These experiments led Szalay (1954, p. 309) to conclude: "The geochemical enrichment of uranium in carbonaceous rocks (bioliths) is caused by the adsorption of dissolved uranium by the humic acid content of the sediments. The adsorption is a cation exchange process."

In a later paper, Szalay (1957) showed that for low concentrations, the uranium in peat is about 10,000 times that in the water in which it is in equilibrium. Experiments are also described which show that the fixation of the uranyl ions is a cation-exchange process whereby H-ions are replaced by UO_2^{+2} ions and that the adsorbed uranyl ion can be easily exchanged by cations of higher valency and high atomic weight such as Th^{+4} and La^{+3} . The cation-exchange process is in contrast to the natural uranium contained in coal, which Szalay was unable to release by ion exchange. Probably the natural uranium in coaly carbonaceous rocks undergoes secondary alterations, for example, reduction to the quadrivalent state by H_2S . As an extension of the work on peat, Szalay (1957, p. 33) arranged a series of cations according to their enrichment factor, and in which each cation is readily replaced by the ones to the right in an exchange column filled with peat, as follows: $\text{Ag}^{+1} < \text{Ni}^{+2} < \text{Cu}^{+2} < \text{Ba}^{+2} < \text{Pb}^{+2} < (\text{UO}_2)^{+2} < \text{Fe}^{+3} < \text{Th}^{+4}$.

In nearly all these experiments to determine the sorption capacity of peat, lignite, coal, and humic acids, the solutions used ranged in uranium content from about 0.05 to 1 percent, a much higher concentration than that in natural solvents. Consequently, the writer, in collaboration with V. E. Swanson, conducted the following experiment to determine the effectiveness of uranium sorption from a solution of low concentration:

From a sample of peat from the Caribou peat bog, sec. 5, T. 1 S., R. 73 W., Boulder County, Colo., the humic acids were extracted in a solution of NH_4OH at a pH of about 9.1. To 4,088 ml of the dark-brown alkaline solution of humic acids, 250 gammas of uranium was added in the form of $\text{UO}_2(\text{NO}_3)_2$, which was sufficient to bring the concentration of the solu-

tion to 61 ppb. The humic acids and uranyl humates were flocculated by lowering the pH to about 2.5 by the addition of HNO_3 . About half of the humic acids remained in solution at this pH, but it was feared that further lowering of the pH might free any organically bound uranium. After drying at a temperature of about 95°C , the recovered humic acids weighed 3.5405 grams and contained 5.19 percent ash and 0.055 percent uranium in the ash, or about 29.4 ppm uranium in the dried sample. One hundred and one gammas of uranium, or about 40 percent of that added to the solution, was recovered from the dried sample and represents a concentration, by weight, about 800 times the equilibrium concentration of the solution. Undoubtedly a much larger proportion of the original uranium could have been recovered had a more efficient method been used that would have flocculated all the humic acids from the solution.

An experiment was designed by Breger, Deul, and Rubinstein (1955, p. 220-224) to test the capacity of lignite to hold uranium in an ion-exchange position. A solution of lanthanum nitrate ($\text{La}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$) was elutriated through a column of uraniferous lignite from the Slim Buttes area, Harding County, S. Dak., in a manner similar to that employed with ion-exchange resins. It was assumed that if the uranium were held in the lignite by ion exchange as the uranyl ion with a valence of two, the lanthanum ion because of its valence of three should displace the uranium and cause it to be carried down in solution and recovered from the base of the column. However, only about 1.2 percent of the total uranium in the lignite was released to the solution and the conclusion was drawn that most of the uranium must therefore be chemically fixed as an organic complex. However, this experiment does not prove that uranium is not originally taken up by coaly carbonaceous rocks by an ion-exchange process. There is the definite possibility that uranium originally fixed by ion exchange is later altered to colloidal-size particles of uraninite by the reducing action of hydrogen sulfide. Furthermore, in the experiment described above, if the lignite used had an ion-exchange capacity that is not saturated, as suggested by the authors, and even if the lanthanum will replace uranium, it seems possible that uranium ions displaced by lanthanum ions at the top of the column would be adsorbed again lower in the column.

VOLATILITY

The possibility that the uranium in coaly carbonaceous rock may be present in a volatile form has been the subject of research related to methods of

analyses and recovery of uranium. Breger, Meyrowitz, and Deul (1954) determined the quantity of uranium volatilized during the process of destructive distillation of four samples of carbonaceous substances including one sample of coal from the Red Desert area, Sweetwater County, Wyo. They concluded that the destructive distillation of the carbonaceous substances investigated resulted in no appreciable volatilization of uranium. The results of uranium recovery tests on roasted and ashed samples of lignite from the western Dakota region confirm the tests described above by showing that uranium can be recovered from the spent or ashed rock (Ewing and others, 1950, p. 23-40).

COMPARISON OF WEATHERED AND UNWEATHERED LIGNITIC ROCKS

Many of the natural exposures and prospect pits of uraniferous lignitic rock in the Fort Union lignite region of western North and South Dakota are considerably weathered and oxidized, and the process of weathering has affected the physical and chemical form as well as the content of uranium. During weathering, lignitic rock that is normally tough and contains abundant pyrite or marcasite becomes progressively more friable and the pyrite is altered to secondary minerals including limonite, gypsum, and jarosite. Visible secondary hexavalent uranium minerals have been observed only in the weathered rock. It is not known whether these secondary uranium minerals result from the oxidation of colloidal-size particles of uraninite and coffinite or are derived from uranium formerly fixed by the carbonaceous matter and made available during the oxidation that accompanies weathering. Where the process of oxidation results in the formation of humic acids that are soluble in water or in weak alkaline solutions, the humic acids could presumably serve as a carrier for the uranium and be redistributed and eventually leached from the outcrop. It might be anticipated therefore, that the weathered outcrop of lignite would be depleted of its uranium content. However, results of an investigation of two columns of lignite from the same bed, one weathered and one unweathered (Koppe and others, 1955, p. 25-29), indicate little or no loss of uranium. The weathered lignite is reduced in volume and contains a proportionately greater percent ash, moisture, and volatile matter but almost the same percent uranium in the ash as the unweathered lignite. Koppe and others (1955, p. 47) suggested the possibility that loss in carbon by conversion to gases during weathering may explain the changes observed.

There is no evidence that uranium in coaly rocks is lost during the early stages of weathering when pyrite is oxidized and the rock is reduced in volume. Normally, the weathering of pyrite produces sulfuric acid, which might be expected to leach uranium from the rock. If sulfuric acid is formed, it is probably quickly neutralized by reaction with other rock constituents or does not accumulate in sufficient concentration to lower the pH below 2, at which point the uranium should go into ionic solution.

Most of the uranium in lignitic rocks is apparently retained during alteration of the originally tough woody lignite to a soft friable earthy mass and probably begins to be lost from the outcrop only when oxidation of the organic substance is sufficiently advanced that organic acids are carried away by solution and take with them some of the uranium. This process apparently continues until nearly all the carbonaceous material is removed from the outcrop leaving only a bleached residue of inorganic mineral matter almost entirely free of uranium.

ASSOCIATION OF URANIUM WITH OTHER ELEMENTS OR CONSTITUENTS IN COALY CARBONACEOUS ROCKS

One approach to investigating the physical and chemical form of uranium in coaly carbonaceous rocks is to study the association of uranium with other elements or constituents of the rock that can be determined by chemical or spectrographic analysis. However, coaly carbonaceous rocks present certain special problems. In particular, the varying proportions of combustible and noncombustible material, moisture, and other volatile matter make it imperative that the condition of the sample or the basis of reporting the analysis be specifically stated and that only similar samples reported on a similar basis be used for comparison.

Standard methods of calculating and reporting proximate and ultimate analyses of coal by the U.S. Bureau of Mines were described by Fieldner and Selvig (1951). These data may be reported on the basis of four conditions: (a) as received, (b) air-dried, (c) moisture-free, (d) moisture- and ash-free. These four conditions are equally applicable to the impure lignite and the lignitic shale in which uranium is found.

Koppe and others (1955, p. 74-82) presented chemical analyses on a moisture-free basis for 18 samples, each from an interval 1 inch thick in an unweathered 18-inch thick lignite bed in the Slim Buttes area, Harding County, S. Dak. From these data they calculated that at the 95 percent probability level, uranium has significant positive correlation with cal-

cium, magnesium, and free iron, but not significant correlation with ash, moisture, carbon, hydrogen, total iron, sulfide iron, total sulfur, sulfide sulfur, organic sulfur, silicon, aluminum, or reflectance, though sulfate sulfur comes very close to having a significant positive correlation.

A diagram (fig. 37A) showing the relation of the amount of uranium to calcium in the 18 samples mentioned above reveals that 10 low values of uranium correspond to 10 low values of calcium and 8 high values of uranium correspond to 8 high values of calcium. However, there appear to be two separate

curves, such that low values indicate a slight positive correlation and the high values a stronger negative correlation. It is noteworthy that the 8 high values represent the top 8 inches of lignite and the 10 low values represent the lower 10 inches of lignite. Thus, although all samples taken together suggest a positive correlation between uranium and calcium, samples having high values of uranium have a negative correlation between uranium and calcium.

The single test of Koppe and others for the relation of uranium to calcium may be the result of an unevaluated variable or a local condition. However, a similar complication was observed from the U_3O_8 and $CaCO_3$ analyses furnished by the U.S. Atomic Energy Commission for 34 carload shipments of lignitic uranium ore. (The 34 carload shipments represent four bulk samples of 500 tons each from the same four mines for which additional analytical data are presented in table 16.) A diagram (fig. 37B) showing the relation of U_3O_8 to $CaCO_3$ in these samples is very similar to the diagram for uranium and calcium in 18 samples from a single bed. It is noteworthy that the cluster of eight samples that show high U_3O_8 and high $CaCO_3$ are all from a single mine. Considered separately the high U_3O_8 samples show an inverse relation with $CaCO_3$, and the remaining samples are random.

SUMMARY

Uranium in coaly carbonaceous rocks may have any of five possible forms of occurrence. The maximum likely to be concentrated in any given form is:

	Percent
1. Inherent uranium.....	0.00X
2. Diagenetically fixed adventitious uranium.....	.X
3. Detrital uranium minerals.....	.000X
4. Epigenetic uranium minerals.....	.X
5. Epigenetically fixed adventitious uranium.....	X

Uranium introduced epigenetically into coaly carbonaceous rocks may vary in concentration by as much as two orders of magnitude within a distance of several inches. The distribution of uranium in high-grade uranium deposits in unweathered lignite may be even more erratic than in weathered parts of the same bed.

Uranium in coaly carbonaceous rocks is nonvolatile and relatively insoluble under natural conditions, except where alkaline solutions react with oxidized coal to dissolve humic substances that carry uranium. Uranium may be retained by coaly rocks in either the quadrivalent or hexivalent state and very likely as a mixture of both. Discrete uranium minerals are seldom recognizable in uraniferous coaly rocks, though hexivalent uranium minerals occur locally on weath-

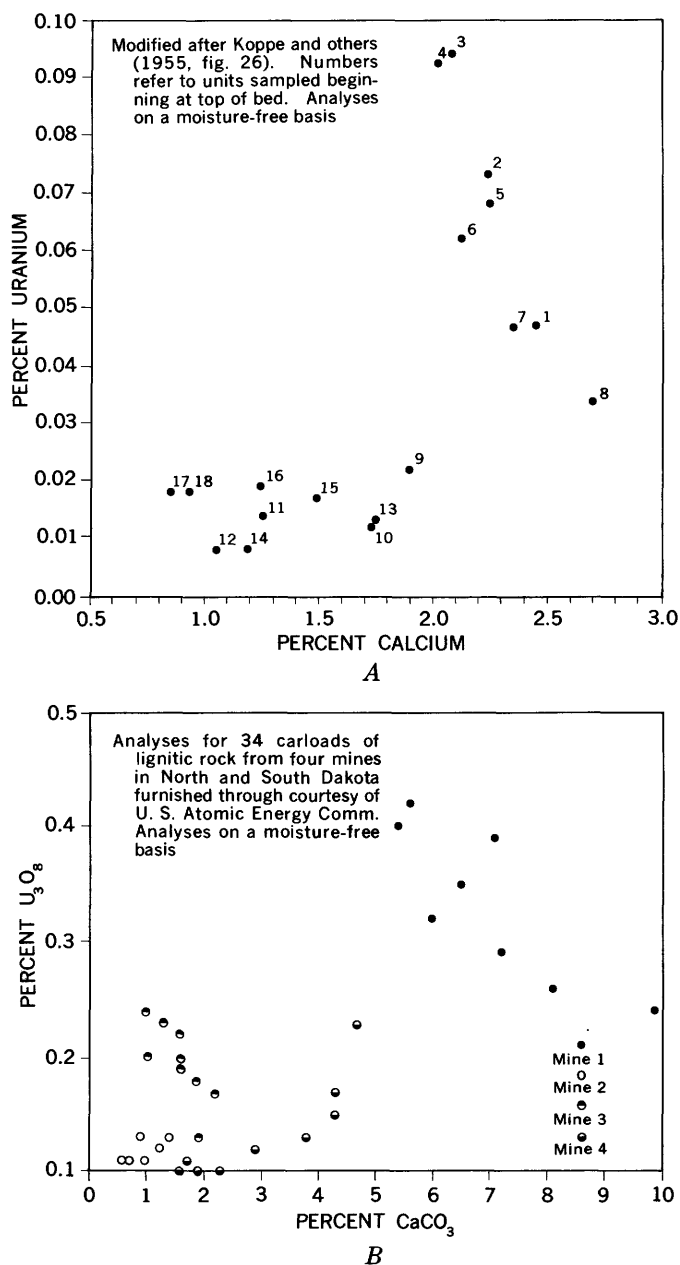


FIGURE 37.—Diagram showing variations among lignitic rock constituents; A, uranium vs. calcium; B, U_3O_8 vs. $CaCO_3$.

ered outcrops, and colloidal-size quadrivalent uranium minerals have been identified in unweathered material by X-ray techniques. Heavy liquid separations of uraniferous coaly rocks into carbonaceous-rich and mineral-rich fractions indicate the bulk of the uranium is associated with the carbonaceous constituents rather than with the mineral constituents of the rock. Peat, lignite, and subbituminous coal readily extract uranium from solution in amounts of as much as 8 percent by dry weight of the rock and will not release the uranium in distilled water. Opinion is divided as to whether the uranium is adsorbed by humic acids in the carbonaceous matter by a cation-exchange process or by some other as yet unidentified process.

Petrographic investigations of uraniferous coaly carbonaceous rocks indicate that all types of carbonaceous matter probably contain uranium but that uranium shows a slight preference for the more degraded attrital material, including amorphous humic matter, and in one group of deposits possibly for yellow waxy matter in the attritus. Permeability of the rocks and availability of uraniferous solutions seem to influence the distribution of the uranium far more than the proportions of different carbonaceous substances. During the early stages of the weathering of lignite, loss of total volume, probably due to loss in carbon, results in an increase in the percent ash, moisture, and volatile matter but in no appreciable change in the percent uranium in the ash.

One investigation indicated a positive correlation of uranium with calcium, magnesium, and free iron, but not with carbon or hydrogen. A diagram of the association between uranium and calcium, however, suggests that the relation may not be a simple direct one, but may actually be inverse in the samples containing the most uranium.

CONCLUSIONS

Uranium may be in several forms in coaly carbonaceous rocks. Deposits of high-grade uranium in the Fort Union region in impure lignite and lignitic shale contain both hexavalent and quadrivalent uranium minerals, depending on the state of uranium oxidation, but discrete minerals do not explain all the uranium present. Some uranium appears to be chemically bonded or physically attached to the carbonaceous substance with sufficient force that physical or chemical alteration of the carbonaceous substance is generally required to detach it. No single organic chemical chiefly responsible for fixing the uranium has been isolated, though it has been suggested that humic acids

or salts and their coalified equivalents (coal ulmins) are among the most effective uranium extractors.

No single explanation yet offered for the known physical and chemical relations of uranium associated with coaly carbonaceous rocks has been accepted by all investigators working with these materials. The explanations offered generally involve adsorption, ion exchange, formation of an organo-uranium compound, or reduction of uranyl ions and precipitation of colloidal size uraninite. Present data indicate that UO_2^{+2} ions may become attached to one of the available bonds on such carbonaceous substances as humic acids or possibly a salt formed from a humic acid. Hydrogen ions may be exchanged for the uranyl ions. Eventually some of the uranyl ions so bonded may be reduced to the quadrivalent form, UO_2 , by the reducing action of either the carbonaceous material or the almost universally associated sulfide ions. In the quadrivalent state uranium does not ionize readily at a pH of over 2 and is relatively insoluble at low temperatures and pressures. In this relatively immobile condition it remains dispersed in a colloidal form and does not form discrete uranium minerals.

PROBABLE ROLE OF COALY CARBONACEOUS ROCKS IN THE GEOCHEMISTRY OF URANIUM

Uranium, with a crustal abundance estimated to be about 4 ppm (Mason, 1952, p. 41), is a relatively minor element, yet perhaps is 40 times more abundant than silver and 800 times more abundant than gold. A concentration of 250 times the crustal abundance of uranium is approximately the minimum grade of uranium-bearing material considered commercial. Though the average abundance of uranium in normal coaly carbonaceous rocks is probably even less than one quarter the average crustal abundance, a number of epigenetic deposits of uranium in coaly rocks contain from 10 to 1000 times the average crustal abundance. These rocks are, therefore, considered to be quite effective agents for the concentration of uranium. Because of the large total quantity of uranium in coaly carbonaceous rocks, they represent a potential large resource of low-grade deposits of uranium that may some day be exploited when higher grade deposits of uranium in sandstone and veins are exhausted. There is, therefore, a need to understand the role of coaly carbonaceous rocks in the geochemistry of uranium.

In spite of the numerous publications dealing with the geology and geochemistry of uranium in coaly carbonaceous rocks, many questions remain unanswered. For example, if uranium is as readily con-

centrated by coaly carbonaceous matter as indicated by laboratory experiments, why are there not more deposits of uranium in carbonaceous rocks? It has been repeatedly observed that abundant coaly carbonaceous matter is associated with high-grade deposits of uranium in sandstone. Why do not coaly carbonaceous rocks contain even more and higher grade deposits of uranium than sandstone? Why are there apparently so few deposits of uranium in the coal of the Interior and Appalachian coal regions of the United States, or for that matter, in the lignite of the Gulf Coastal Plain or in the Cretaceous coal of the Rocky Mountain region? Though firm answers cannot now be given to these questions, the speculations in the following section may provide ideas for future investigations.

ENVIRONMENTAL FACTORS IN THE DEPOSITION OF URANIUM

Most of the known uranium deposits in coaly carbonaceous rocks in the United States are thought to represent epigenetically fixed adventitious uranium. It may seem strange that fossil deposits similar to those of the uraniferous mountain meadows of California are not more commonly preserved as syngenetic uranium deposits in coaly carbonaceous rocks. Why does the marine black shale that overlies coal in parts of southern Illinois and Indiana contain as much as 0.006 percent uranium over widespread areas (Snider, 1954, p. 20-22) and represent the highest concentration of uranium in the sequence of coaly rocks, while the coal, which represents more concentrated carbonaceous matter, is practically barren of uranium? Comparison of the environment of deposition of the coal with the California mountain meadows indicates that fundamental differences exist that may help to explain the absence of syngenetic deposits in coal.

Most of the large coal deposits in the United States were formed from peat bogs in poorly drained flatlands marginal to the sea. Unlike the California mountain meadows which derive uranium ions from spring waters that issue from the underlying granitic rocks, streams were the chief source of uranium ions available to the coastal plain peat bog. Although many streams originate chiefly in the bog, even those few that cross it are so heavily charged with soluble brown organic matter (Clark, 1924, p. 110) that any uranium ions would tend to remain fixed by the organic matter in solution, and thus be carried out to sea. This fact may explain why, in the transgressing continental and marine sequence that includes coal beds of Pennsylvanian age in the Interior and Eastern coal fields of the United States, uranium is com-

monly concentrated in the marine black shales that overlie coal beds. The uranyl humates carried in solution by streams are probably concentrated as a flocculant precipitate by the action of sea water.

Unusual circumstances must be invoked to explain the rare syngenetic concentration of uranium in plant debris such as is in the California mountain meadows, which derive uranium ions from spring water. Locally, sea water may be the source of uranium in plant debris that is deposited near the strand line.

GEOCHEMICAL AND PHYSICAL FACTORS IN THE DEPOSITION OF URANIUM

Not all carbonaceous matter is equally effective in removing uranium from solution. Figure 28 shows that indigenous carbonaceous matter may be divided into humic and sapropelic types on the basis of the kinds of original plant remains that predominate. Hydrogen-rich plant remains yield sapropelic matter, while oxygen-rich plant remains yield humic matter, the principal constituent of banded coals. Each of these in turn gives rise to mobile products that may be secondarily emplaced. Sapropelic matter yields bitumens including gaseous and liquid petroleum and a variety of residual products. Humic matter yields alkaline-soluble humic extracts including humic acids and humates. Of these various types of carbonaceous matter, only low-rank indigenous humic matter and the alkaline extracts therefrom are commonly associated with uranium. These materials are closely related in chemical structure to humic acids, the group of natural organic chemical compounds most effective in the sorption of uranium. Higher rank indigenous humic matter, including bituminous and anthracitic coals, contain progressively less oxygen, a chemical constituent of humic acids, and may thus represent one critical factor in determining the capacity of the carbonaceous matter to take uranium out of solution. The common loss in higher rank coals of nitrogen and sulfur, also essential to the chemical structure of humic acids, may be secondary factors in limiting the capacity of these coals to take up and retain uranium.

If a fundamental difference existed between the origin of high-rank coal and low-rank coal, then this difference could partially explain the scarcity of uranium deposits in high-rank coal. Such a difference was postulated by Fuchs (1942, p. 229); he suggested that bituminous coal forms from the reduction of plant remains without passing through an intermediate lignite stage. This suggestion, however, has not been accepted by most workers, who believe that high-rank coal is the metamorphic product of low-rank coal.

The epigenetic deposits of uranium in impure lignite and lignitic shale seem to be characteristically thin submarginal to marginal ore-grade deposits in contrast to deposits in sandstone. The reason for this is probably both physical and geochemical. Whereas uranium minerals in sandstone may be deposited from mineralizing laterally moving solutions in response to local chemical changes in environment, lateral movement of ground water is probably not common in lignitic rocks. Beds of sandstone adjacent to lignite probably transport most of the water, but a slight upward or downward direction of movement is indicated by the bottom- or top-preferential enrichment of uranium common in lignitic rocks. Thus, the solutions that transport uranium are exposed to a broad area of lignite all of which has approximately equal capacity and opportunity to take up uranium. In sandstone, the environmental conditions that favor deposition of uranium minerals are probably localized by the interstitial pore fluids and may tend to be dispersed equidimensionally through a local mass of rock.

GEOCHEMICAL ROLE OF CARBONACEOUS MATTER IN SANDSTONE AND IN COALY CARBONACEOUS ROCKS

Because uranium shows a direct relation to carbonaceous matter in sandstone, marine black shale, and coaly carbonaceous rocks, is there a geochemical or merely a physical explanation for the fact that deposits in sandstone yield more ore than coaly carbonaceous rocks, which may contain more abundant carbonaceous matter? Perhaps the role of the carbonaceous matter is somewhat different in the various types of deposits, and this difference may have geochemical significance. Carbonaceous matter in sandstone deposits apparently serves as the locus for precipitation of uranium minerals far in excess of the uranium that may be retained by sorption or chemical reaction with carbonaceous material. Where coaly carbonaceous matter takes uranyl ions directly out of solution by sorption or chemical reaction, the concentration of uranium in the resulting deposit tends to be a function of the concentration in solution. No such simple relation can be demonstrated for uranium minerals associated with carbonaceous matter in sandstone.

In addition to the sorption of uranyl ions by organic chemicals related to humic acids, secondary alteration of the uranium is apparently common in coaly rocks because the uranium cannot be removed by simple cation exchange. This may be interpreted to mean that in some deposits a very stable organic compound of uranium is formed, but the discovery of colloidal-size coffinite and uraninite (Gruner, 1956, p.

504) in lignite from the Fort Union lignite region suggests that once taken up by the carbonaceous material some of the uranyl ions may be reduced to the quadrivalent state. While some doubt remains regarding the ability of various types of carbonaceous matter to reduce uranium at ordinary temperatures and pressures, almost everywhere enough hydrogen sulfide is present in carbonaceous rocks to explain the reduction of uranyl ions to the quadrivalent state. The process of sorption and reduction, if repeated, might explain the large amount of uranium relative to carbonaceous matter in some sandstone deposits.

Though not clearly understood, the formation of uranyl phosphate and arsenate minerals at the weathered outcrop of uraniferous impure lignite beds is probably not related to the deposition of uranium minerals in sandstone. In weathered impure lignite, uranyl ions made available by the oxidation of the lignite are concentrated and deposited at the weathered outcrop by evaporation of capillary pore moisture. The original deposition of uranium minerals in sandstone, while possibly related to the reducing environment surrounding the carbonaceous matter, does not involve concentration by evaporation.

GEOCHEMICAL ROLE OF MARINE BLACK SHALE AND OF COALY CARBONACEOUS ROCKS

Just as carbonaceous marine black shale and phosphatic marine shale form important depositories for uranium in a marine environment, so also coaly carbonaceous rocks form an important depository for uranium in a nonmarine environment. However, an important difference exists. Whereas most of the uranium is concentrated from sea water at the time of deposition of marine shale, coaly carbonaceous rocks rarely collect uranium at the time of rock deposition. Most of the uranium is concentrated from groundwater solutions that penetrate the rock long after deposition and burial of the sediment. From the viewpoint of the geochemical cycle of uranium in sedimentary rocks, carbonaceous or phosphatic marine shale and coaly carbonaceous rocks complement each other. During periods of erosion, uranium that is made soluble and introduced into surface waters such as streams tends to be transported to sea and concentrated in carbonaceous marine mud, probably largely in that rich in humic as compared with sapropelic matter (Swanson, 1960) or in phosphate-rich sediment. Soluble uranium introduced to ground water either from thermal water with an igneous source or from alteration of the local sedimentary rocks (for example, devitrification of tuffaceous material) is generally deposited either in sandstone or coaly carbonaceous rocks. If the ground water that

carries uranium comes first into contact with coaly carbonaceous rocks the uranium will be deposited there, otherwise it will probably be transported through sandstone aquifers where sooner or later the solutions will probably meet reducing conditions or some other chemical environment that effects the precipitation of uranium minerals.

GEOCHEMICAL ENRICHMENT OF URANIUM IN COALY CARBONACEOUS ROCKS

Coaly carbonaceous rocks locally contain far greater concentrations of uranium than most marine black shale, even though the uranium in both may have been deposited by the same process of cation exchange on humic carbonaceous matter. The differences are due to differences in uranium concentration of waters normally present in the environment of deposition and to the relative accessibility of the rocks to mineralizing ground waters after burial.

Unlike the major elements in a sedimentary environment which separate into resistates, hydrolystates, oxides, carbonates, and evaporates, uranium tends to remain dispersed throughout the processes of weathering, erosion, transportation, sedimentation, burial, and diagenetic alteration of sediment until such time as uranyl ions come in contact with humic carbonaceous matter or phosphatic material. This contact may occur at any one of a number of different times and places, largely because the cause of enrichment is unlike that for any of the major elements. Uranium is probably not unique in this process, for many other metals, including certain rare metals, are also enriched in carbonaceous rocks (Krauskopf, 1955, p. 420). Uranium differs from those described by Krauskopf because it is normally deficient, not enriched, in coaly carbonaceous rocks. Because of the infrequent association of soluble uranium with coaly carbonaceous rocks, only rarely is a marked enrichment observed.

The principles governing the geochemical enrichment of uranium in coaly carbonaceous rocks may be summarized as follows:

1. Indigenous carbonaceous matter of the humic type that has not been altered beyond the rank of subbituminous coal will readily extract uranyl ions from solution by as much as several percent by dry weight of carbonaceous matter. The extraction probably results from cation exchange in chemical compounds similar to humic acids.
2. The concentration of uranium in coaly carbonaceous rocks is a function of the following factors:
 - a. The proportion of chemically reactive humic carbonaceous matter in the rock.
 - b. The concentration of uranyl ions in solutions that have access to the rock.
 - c. The number of times fresh uraniferous solution is brought into contact with the carbonaceous matter.
 - d. The porosity and permeability of the rock.
3. Indigenous carbonaceous matter of the humic type that contains little or no uranium was either never exposed to uranium-bearing solutions or was altered to the point that it lost its capacity to take up and retain uranium.
4. Once the uranium is "fixed" by the carbonaceous matter it is tenaciously held, or bound by the metallic-organic chemical structure, though in time it may be reduced to uraninite or coffinite—mineral forms that are relatively stable except in the zone of oxidation.
5. Under natural conditions uranium probably remains disseminated in coaly carbonaceous rocks as originally deposited until the rock undergoes weathering and oxidation, during which time uranium may be redistributed and hexavalent uranium minerals formed. Eventual loss of uranium from the outcrop of carbonaceous matter accompanies the final stage of complete oxidation and alteration of the humic acids.

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