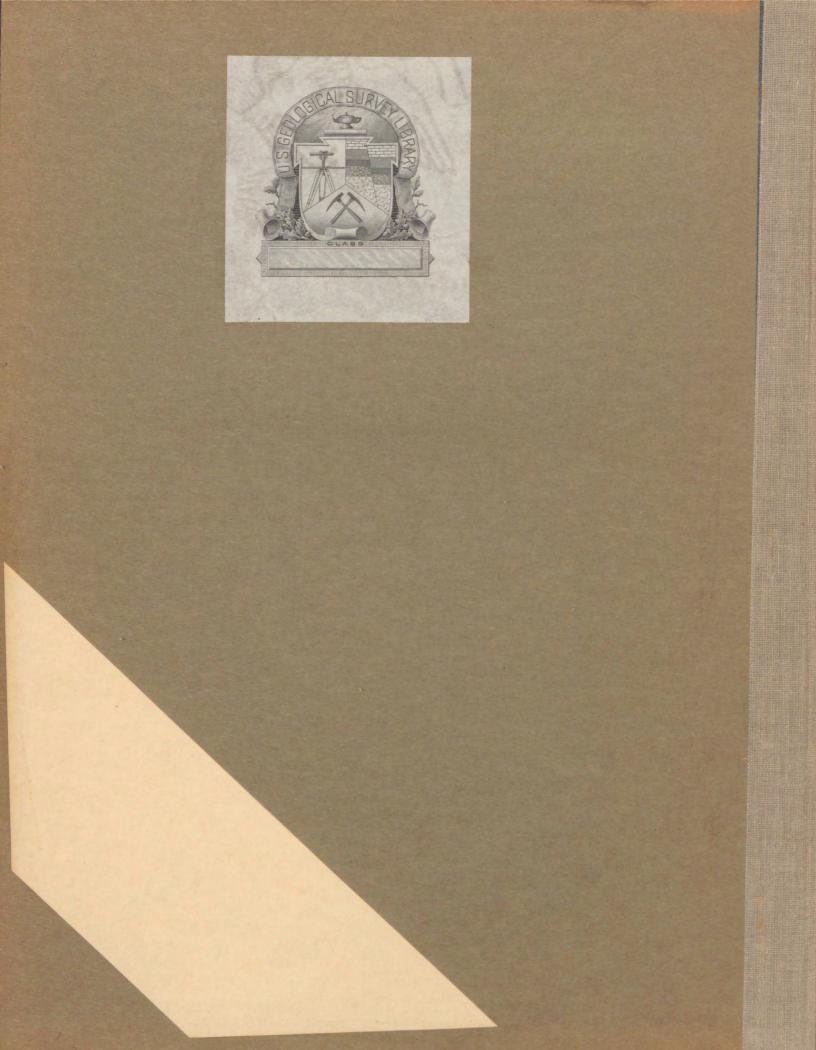
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VARIOUS ASPECTS OF URANIUM ORE ROLLS IN THE

UNITED STATES

By Larshman 1900 prepared for

(Text of a talk prepared for presentation at International Atomic Energy Agency Panel on Uranium Exploration Geology, Vienna, Austria, April 13-17, 1970)

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VARIOUS ASPECTS OF URANIUM ORE ROLLS IN THE UNITED STATES

By E. N. Harshman

U.S. Geological Survey, Denver, Colo.

ABSTRACT

About 40 percent of the uranium ore reserves in the United States, minable at \$8 per pound of contained U₂O₈, are in roll-type deposits in the State of Wyoming. The host rocks are arkosic sandstones, deposited in intermontane basins under fluvial conditions, and derived from the granitic cores of mountain ranges that flank the basins. The host rocks are Eocene and possibly Paleocene in age and are, or were, overlain by a sequence of continental tuffaceous siltstones, sandstones, and conglomerates 400-700 meters thick.

Most of the ore is unoxidized and lies below the water table. It contains pyrite, uraninite, coffinite, marcasite, hematite, ferroselite, and native selenium(?). The ore bodies range in size from a few hundred to several hundred thousand tons of material containing 0.10-5 percent U_3O_8 . As mined, the ore averages about 0.25 percent U_3O_8 .

The ore bodies are genetically related to and lie at the margins of large tongues of altered sandstone. The character of the alteration and the distribution of several elements within and near the altered sandstone suggest that the ore-bearing fluid was ground water, neutral to slightly alkaline and oxidizing with respect to the elements being transported.

Roll-type deposits in littoral and fluvial sandstones have recently been discovered in the Gulf Coastal Plain area of Texas. These deposits appear to be similar to the Wyoming deposits in form, distribution of elements, and genesis.

INTRODUCTION

Three principal areas account for about 95 percent of the United States reserves of uranium ore minable at \$8 per pound of contained U308. They are the Colorado Plateau, the Wyoming basins, and the Texas Coastal Plain (fig. 1). Prior to 1953 most of the known uranium deposits of commercial size and grade were in the Colorado Plateau area. of these deposits are in sandstones of Jurassic age and of continental origin. They are predominantly tabular approximately parallel to the bedding of the host rock and they have been called tabular deposits. About 1953, exploration in Wyoming uncovered large uranium deposits in continental sandstones of early Eocene age. Ore bodies in these deposits cut sharply across the bedding of the host rocks and they are called roll-type deposits. They appear to be similar to the infiltration-type deposits described by geologists in the U.S.S.R. Within the last few years, exploration in the Texas Coastal Plain area has shown the presence of roll-type deposits in littoral and fluvial sandstones of Eocene and Miocene age.

Ore rolls were first described from the Colorado Plateau, and the tabular deposits commonly contain minor roll features. Conversely, the roll-type deposits of Wyoming contain some tabular ore.

The tabular deposits have been known for 70 years; they have been studied extensively and described at length in the geologic literature. The roll-type deposits are of relatively recent discovery; they have been less well studied and have been described in only a few papers. They are, however, of major importance as a source of uranium in the United States, and in this paper I describe briefly their physical aspects and the distribution of some elements in them, and speculate on possible modes of origin.

GEOLOGIC SETTING

The principal and best known roll-type uranium deposits in the United States are in the basins of Wyoming, a semiarid region characterized by mountain ranges trending northwesterly separated by intermontane basins (fig. 2). The mountain ranges have Precambrian granitic cores and they are flanked by rocks of Paleozoic and Mesozoic age. The basins are downwarps and downfaulted blocks in Paleozoic and Mesozoic rocks, modified to some extent by erosion of soft Cretaceous shales and filled with Tertiary sediments. The fill consists of clastic material eroded from the mountain ranges and tuffaceous material originating from volcanic vents in northwestern Wyoming.

The Wyoming deposits are in beds of arkosic sandstone and conglomerate of fluvial origin and of early Eocene and possibly Paleocene age. The beds range in thickness from a few meters to as much as 100 meters. They are medium to coarse grained, poorly cemented or not cemented, pyritic, and carbonaceous. They contain almost no tuffaceous material. The sandstones are interbedded with siltstones and shales in most places and in some places with lignitic shales. The shale and siltstone beds within the sandstones are generally less than 1 meter thick. The major sandstone units are almost everywhere separated by beds of siltstone or silty claystone as much as 30 meters thick. In some exposures, sandfilled channels cut part way through the thick siltstone beds and dewatering operations have shown that the major sandstone units are connected hydraulically. Dips of beds in areas that contain large high-grade uranium deposits are generally low (1°-3°) but ore has been found in sandstones with dips as great as 20°.

A sequence of fluvial and lacustrine siltstones, sandstones, and conglomerates overlies, or once overlay, the ore-bearing formations. These rocks range in age from middle Eocene to Pliocene and at one time they covered the western two-thirds of Wyoming to a depth of 400-700 meters. Post-Pliocene erosion has reduced the thickness of these rocks over many of the mining areas and in some places it has completely removed them. All these rocks contain considerable tuffaceous material.

URANIUM DEPOSITS

General description

Roll-type uranium deposits range in size from a few hundred tons to a few hundred thousand tons of material that contain from 0.10 to 5.0 percent U₃08. The ore now being mined contains 0.10 to about 0.5 percent U₃08, depending on mining methods and costs; during the period 1960-70 the mined ore contained an average of about 0.25 percent U₃08.

Nearly all the ore deposits are below the water table and, therefore, they have been little affected by recent oxidation and redistribution. The principal ore mineral is uraninite; it is black, fine grained, and sooty. In some deposits coffinite is present in significant amounts. Also in the ore are pyrite, marcasite, jordisite, hematite, ferroselite, native selenium(?), and calcite. The ore and gangue minerals coat the sandstone grains and fill the openings between grains. Limited studies of paragenesis of the minerals suggest that ferroselite, Pyrite commonuraninite, and pyrite were the earliest minerals to form. ly replaces feldapar along cleavage planes; uraninite rarely does 30, Calcite is present in most of the Wyoming deposits either as large concretionary masses in a halo at the outer limits of ore or as small connecting masses in high-grade ore. Vanadium, plentiful in the tabular deposits of the Colorado Plateau, is present in trivial amounts in most of the roll-type deposits of Wyoming.

Relation to alteration

Roll-type uranium deposits are everywhere associated with large tabular masses or tongues of altered sandstone. The alteration was almost certainly produced by the ore-bearing solutions. The character of alteration differs from basin to basin but the overall effect was one of oxidation. The altered zones range considerably in size and shape; they may be as much as several hundred square kilometers in area and several tens of meters thick. In some places the altered sandstone tongues fill the entire sandstone interval, in other places only the most permeable parts of the interval are altered. The upper and lower surfaces of the tongues generally are conformable with the bedding of the enclosing rocks.

Most of the ore-bearing basins in Wyoming contain several altered sandstone tongues that have formed in sandstone intervals separated by fine-grained impermeable rocks. The tongues partly overlap, but their edges are superimposed in only a very few places. Two altered sandstone tongues in the Shirley Basin, separated by about 75 feet of siltstone, are shown on figure 3.

The largest ore bodies are at the edges of the altered sandstone tongues in what have been called terminal rolls. Smaller ore bodies are present on the top and bottom surfaces as subsidiary rolls and as tabular pods. The ore extends outward (away) from the altered sandstone for distances ranging from less than 1 meter to as much as 80 meters. Figure 4, a section across the edge of an altered sandstone tongue in the Shirley Basin of Wyoming, shows a terminal roll, a subsidiary roll, and small pods of ore along the top and bottom surfaces of the altered sandstone tongue. On the concave or trailing edge of the roll the contact of ore and altered sandstone is very sharp, but on the convex or leading edge, the ore gradually decreases in uranium content until it merges with unaltered, unmineralized sandstone.

The edges of the altered tongues are almost everywhere mineralized but they are not continuously ore bearing. Small ore bodies extend only a few meters along the tongues; large ones extend for a kilometer or more.

The positions of the ore-bearing segments in the Shirley Basin appear to be related to major changes in the strike of the edges of the tongues.

Distribution of elements

Analytical data from suites of samples of the Wyoming deposits show that there is a systematic distribution of some elements with respect to the edges of the altered sandstone tongues. Of particular interest, because of genetic implications, is the distribution of uranium, uranium daughter products, ferrous and ferric iron, sulfidesulfur, selenium, and mineral and organic carbon.

Uranium is distributed unevenly in the ore bodies, being most abundant at or near the edges of the altered sandstone tongues and decreasing gradually in amount away from them. In some deposits the highest grade ore is at the edges of the tongues but in others it is as much as a mater away from the edges. The uranium content of unaltered sandstone ranges from about 0.0002 to 0.0006 percent, or slightly less than the average uranium content of the granites from which the sediments were derived. Altered sandstone in the interior of the tongues contains from 0.0006 to about 0.0015 percent uranium—considerably more than unaltered sandstone contains.

Uranium is not in equilibrium with its daughter products in most of the Wyoming deposits. Radiometric analyses (beta-gamma) for equivalent uranium exceed chemical analyses (fluorimetric) for uranium on samples of poorly mineralized sandstone, both altered and unaltered, and chemical analyses exceed radiometric analyses on samples of ore and of ore-grade altered sandstone. Samples of poorly mineralized altered sandstone generally have higher eU/U ratios than do samples of poorly mineralized unaltered sandstone.

Iron is present in both the ferrous and ferric states and the distributions of the two are quite different. Unaltered sandstone contains 0.5-1 percent iron, but two-thirds of the iron is in the ferrous state--predominantly as pyrite. Altered sandstones contain similar amounts of iron, but about two-thirds of it is in the ferric state as limonite, goethite, and probably as iron associated with clay minerals. The ore contains 1-3 percent iron, almost all of which is present as ferrous iron in pyrite and(or) marcasite.

The distribution of sulfide sulfur somewhat follows that of ferrous iron and reflects the amount of pyrite in the various parts of the deposit. The sulfide sulfur content of unaltered sandstone is generally less than 1 percent, that of ore is 1-3 percent, and that of altered sandscone in the interior of the tongue is less than 0.002 percent. The low percentage of sulfide sulfur in the altered sandstone reflects the almost total absence of pyrite--a characteristic of nearly all of the altered sandstone tongues in Wyomng.

Selenium, present in amounts of 0.5-1 ppm in unaltered sandstone, has been concentrated in the ore and in altered sandstone. Almost all of the Wyoming deposits have a narrow band of high selenium content that is astride the contact of ore and altered sandstone. This band is rarely more than 5 meters wide and it may contain as much as 0.3 percent selenium. The selenium content drops rapidly in ore at increasing distances from this band and eventually reaches a background amount of 1± ppm. There is a similar rapid drop in the selenium content of altered sandstone at increasing distances from the band, but the selenium background of the entire altered tongue is in the range from 20 to 40 ppm.

Carbon is present in the deposits as mineral carbon in calcium carbonate and as organic carbon in fossil plant debris. Both forms show characteristic distribution patterns. Calcium carbonate, principally as calcite, is widely distributed in the unaltered host rocks in amounts ranging from 1 to 3 percent. Ore is generally not high in calcium carbonate but in most places it contains a few large calcitecemented sandstone concretions in a halo near the outer limits of ore and similar but smaller concretions in ore. Altered sandstone generally contains only a few hundredths percent carbonate except near the margins of the tongues, where a few small corroded concretions are present.

Organic carbon is present in the unaltered sandstone and the ore but has been almost completely destroyed in the altered sandstone. In some deposits high uranium correlates with high organic carbon, in others there is a negative correlation.

Molybdenum is present in some deposits, virtually absent in others. The molybdenum mineral is probably jordisite, a molybdenum disulfide. Where present, molybdenum is in a halo at the outer edge of and beyond the uranium ore.

Figure 5 is a graphical presentation of the analytical data from a suite of 20 samples taken across an ore body in the Shirley Basin, Wyoming. It illustrates some of the distribution patterns just discussed.

Origin

An acceptable concept of origin for the Wyoming roll-type deposits must explain their restricted stratigraphic distribution, their spatial relations to large bodies of altered sandstone, the systematic distribution of their component minerals, and their physical parameters. It must encompass the source of the uranium and associated elements, the nature and hydrodynamics of the ore-bearing fluid, and the method of precipitation of the ore minerals. Any discussion of origin will of

necessity be speculative and interpretative inasmuch as the deposits were formed eons ago. There is, however, considerable data on the Wyoming deposits that can be used to keep speculation within bounds and, it is hoped, to help interpret correctly some of the many complex genetic factors responsible for the deposits.

Classification of deposits

The Wyoming deposits are epigenetic as shown by the fact that, although some of the ore minerals pseudomorph crossbeds and other sedimentary structures, most of the ore cuts across the sandstone beds at sharp angles, clearly showing the ore minerals to be younger than the sandstone.

Source of the uranium

Unfortunately, evidence is much less clear as to the source of the uranium and other elements associated with it in the ore. Possible sources include: (1) magmatic hydrothermal solutions; (2) the arkosic sandstones that are the host rocks for the deposits; (3) the granitic cores of the mountain ranges that flank the basins; and (4) the tuffaceous rocks, and more specifically the shards in them, that overlie or once overlay the host formations.

Magmatic hydrothermal solutions seem to be the least likely source of the elements in the ore. There is no evidence of hydrothermal activity near any of the deposits and no known igneous activity of Tertiary age within 100 kilometers of many of them. The mountains that flank the basins, except for iron deposits of Precambrian age, are as little mineralized as any in the United States, and it seems incongruous to propose a genetic relationship between such poorly mineralized ranges and such well mineralized basins.

The altered arkose associated with the uranium deposits in the basins of Wyoming seems not to be the source of the uranium and other elements in the ore. Data presented earlier in this paper show that the altered sandstone contains slightly more uranium and 10 to 20 times more selenium than does the unaltered sandstone, clear evidence that during alteration, uranium and selenium were added to the arkose, not extracted from it.

The granitic rocks in the cores of the mountains that flank the Wyoming basins contain 3-10 ppm uranium, and laboratory tests show that some of this uranium is readily soluble in a leach solution that approximates normal groundwater in composition—high sulfate, high bicarbonate, and a pH of about 8. This uranium would become available for solution as the granitic rocks weathered, and it may have found its way to the basins and the uranium deposits in them.

Volcanic ash has long been considered as a possible source of various elements found in sedimentary rocks and soils. The rocks that overlie or once overlay the Wyoming deposits contain considerable ash and some relatively pure tuff beds, and it seems probable that some of the uranium and elements associated with it in the ore may have come from this source. The tuffs contain 4-7 ppm uranium, about the same as the granite, and small amounts of other elements found in the ore. Vadose water issuing from tuff beds in the Shirley Basin of Wyoming, water that has not been in contact with other rocks, contains 8-52 ppb uranium and lesser amounts of other elements. If present-day vadose water contains uranium and other elements, almost certainly derived from tuffaceous rocks overlying the ore-bearing sandstones, it seems only reasonable to assume that such rocks contributed uranium to ground water at the time the Wyoming deposits were being formed.

Although the available evidence on the source of uranium and associated elements in the Wyoming deposits is circumstantial, it indicates that the source was multiple and included the granite in the cores of the mountain ranges that flank the basins and the tuffaceous rocks that occupy or once occupied the eastern two-thirds of Wyoming.

Solution, transportation, and deposition of elements

Hostetler and Garrels (1962) have shown that uranium is soluble in the presence of CO₂ and at low temperatures and pressures, either in the 4- or 6-valent state, depending on the pH and Eh of the solution. In the 4-valent state and under slightly oxidizing conditions, it can be transported in solutions with pH values of less than 3. Transportation in the 6-valent state is possible in slightly acidic or slightly alkaline solutions at Eh values ranging from +0.3 to -0.4 volts. This wide range in conditions under which uranium is soluble makes this element a poor indicator of the character of the transporting medium and the genetic processes responsible for the roll-type deposits in Wyoming. However, other factors such as the geochemical environment of the basins, the character of the alteration produced by the ore-bearing solutions, and the geochemistry of the elements associated with uranium greatly restrict the conditions under which the deposits could have formed.

The permeable parts of the Tertiary rocks through which the orebearing solutions migrated are arkoses containing considerable feldspar and quartz and several percent acid soluble material, principally calcium carbonate. The formations that overlie or once overlay the ore-bearing rocks have clastic components similar to those in the host rocks, but in addition they contain considerable volcanic ash in relatively pure tuff bads or dispersed through the siltstones. Both the arkosic and the tuffaceous rocks are alkaline environments and they would tend to buffer the pH of water flowing through them. Garrels and Christ (1965) have shown that water in contact with CaCO3 generally has a pH of about 8, and Hemley and Jones (1964) have discussed at length the increase in alkalinity caused by hydrolysis of silicate minerals such as feldspar. Laboratory and theoretical data published by Hay (1963) show that alteration of silicic glass releases alkalic ions to ground water by reactions that produce pH values of 8 or more. These theoretical considerations are confirmed by the pH measurements of actual ground waters in the Wyoming basins, almost all of which were greater than 7.0.

Ground water flowing through the basins today is alkaline, the environment through which it flows is conducive to alkalinity, and if the present is a key to the past, it is only reasonable to conclude that the ore-bearing solutions were alkaline.

Transportation in oxidizing solutions is suggested by the character of the alteration produced as the solutions passed through the orebearing sandstones, and by the geochemistry of the elements in the ore. Data presented earlier in this paper show that during the alteration process, pyrite, carbonized plant material, and calcium carbonate were destroyed, the ferric/ferrous iron ratio in the sand was increased, and goethite and limonite were deposited. All of these changes require oxidation reactions.

All the elements in the ore can be transported in oxidizing, neutral to slightly alkaline solutions and some of them must be transported in such solutions. Lakin (1961) has shown that selenium cannot be taken into solution under natural conditions, as nobile selenate ion, at pH values of less than 7. Goleva and Lushnikov (1967), Granger and Warren (1969), and Warren (1968) suggest that selenium may be carried as a complex with sulfur rather than as selenate ion. These complexes are stable only in alkaline solutions that are weakly oxidizing.

In addition to the positive, although indirect evidence that the ore-bearing solutions were alkaline and oxidizing, there is reason to believe that they were not acid. Acid solutions of sufficient strength to transport uranium in the 4-valent state, and in sufficient volume to account for the uranium deposits in the Wyoming basins, would have produced considerable alteration of the feldspars, particularly plagioclase. Alteration of the feldspars in the altered sandstone host rocks is not intense and can be accounted for by a temporary drop in pH due to oxidation of pyrite in the cre body, a process to be described later in this paper. A second objection to acid solutions is the difficulty of generating them in rocks containing only small amounts of plant material and more than enough calcium carbonate to neutralize all the acid produced by oxidation of the pyrite in the unaltered sandstone.

Although there is no conclusive proof of the chemical character of the ore-bearing solution, the available data suggest that they were neutral to slightly alkaline and oxidizing with respect to the materials being transported.

The ore deposits appear to result from a reduction in the Eh of the transporting fluid, although a drop in pH may have been a contributing factor in their formation. The large amount of ferrous iron and the small amount of ferric iron in the ore, the presence of ferroselite in ore and in altered sandstone near ore, the reduced chemical state of the ore minerals, and the general nature of the environment in which ore is found, topics discussed earlier in this paper, all suggest that the zone of deposition was strongly reducing with respect to the ore The nature of the reductant is not known. Lindgren, Graton, and Gordon (1910) postulated that the red-bed copper deposits of New Mexico might have originated through reduction of copper by H2S of biogenic origin. More recently Jensen (1958), Cheney and Jensen (1966), and others have shown the probability that HoS of biogenic origin accounts for the sulfur in the pyrite intimately associated with uranium in the Wyoming deposits. Research now under way in the U.S. Geological Survey (Granger and Warren, 1969) has shown that pyrite associated with uranium ore may form in the absence of bacteria, by inorganic reactions between iron and sulfur species formed by oxidation of disseminated pyrite of biogenic origin. Bacterial reduction of sulfate is a necessary part of this concept, but the final reduction of the elements to form ore is independent of bacterial activity.

Grutt (1957) proposed that natural gas may have been the reductant for the uranium deposits in the Gas Hills area of Wyoming, but most of the ore bodies are not spatially related to the paleo-outcrops of the gas- and oil-producing beds as one would expect if gas were the precipitant. An excellent case can be made for gas as the precipitant for the recently discovered roll-type uranium deposits in the Coastal Plain area of Texas.

The poor correlation of uranium and carbon, discussed earlier in this paper, suggests that carbon itself has not played an important role in the formation of the deposits, although organic matter and redistributed organic compounds may have been the precipitant for some uraninite and pyrite.

Although available data do not define the reductant responsible for the uranium deposits in the Wyoming basins, they are compatible with the concept that H₂S of bicgenic origin was an important factor in ore genesis, either directly or through intermediate sulfur compounds formed by oxidation of an early biogenic pyrite.

The age of the Wyoming deposits is not known with any degree of certainty. Crude lead/uranium ratios determined on several samples of ore from the Shirley Basin, Wyoming, suggest an age of about 17 million years, and similar ratios on samples of ore from the Powder River Basin of Wyoming suggest ages of from 4.5 to 13 million years. Geologically, an age of 10 to 15 million years is reasonable, for in late Miocene time there was uplift of the mountains bounding the Wyoming basins, accompanied by vigorous renewed erosion. This uplift is thought to have been the event that spawned the hydrodynamic cycle responsible for the ore deposits.

INTEGRATED GENETIC MODEL

Conditions favorable for solution, transportation, and deposition of uranium probably came into being in late Miocene or early Pliocene time when the host rocks in the Wyoming basins were buried under 300-600 meters of clastic and tuffaceous rocks. Soon after uplift, oxidizing, alkaline ground water that carried uranium and other elements entered the permeable members of the host rocks and moved down the The oxygenated ground water was at first out flanks of the basins. of equilibrium with the reduced rocks through which it moved, but oxidation of the rock components, particularly pyrite and fossil wood, quickly restored conditions approximating equilibrium in an oxidized zone at and slightly downstream from the area of ground-water ingress. This zone, at first very small, gradually expanded in the direction of ground-water flow and a reaction zone was established between reducing conditions in the downdip part of the aquifer and oxidizing conditions in the updip part. Several interfaces were established in this zone, and the position of each was determined by the combined effect of Eh and pH changes on the solubility of various elements. The interface most commonly discussed is one between oxidized and reduced forms of iron, i.e., pyrite and goethite or limonite, but there are easily recognized and separate interfaces for iron, uranium, selenium, and molybdenum. Deposition of an element carried by ground water occurred most rapidly at the redox interface for that element and with decreasing rapidity as the sclution moved through the general reaction zone. was an overlap in the deposition of the several ore elements.

The mineralized zone established early in the cycle described above was small and low grade. It migrated in the direction of ground-vater flow, generally downdip, by oxidation and solution on the updip side of the zone and reduction and redeposition on the downdip side. With a continuous external supply of uranium and associated elements in the ground water passing through the zone of deposition, the mineralized zone grew in both size and grade, eventually reaching the

magnitude of present-day ore bodies. The process just described continued until such time as the supply of uranium and associated elements was exhausted, the environment causing deposition was destroyed, or the hydrodynamic system was disrupted. Figure 6 summarizes graphically the probable Eh and pH conditions during transportation and deposition of uranium and other elements.

Exploration in the last few years has shown the presence of rolltype uranium deposits in the Gulf Coastal Plain area of Texas. These deposits are in littoral and fluvial sandstones that contain considerable tuffaceous material. The host rocks range from Eocene to Miocene in age. The rolls are similar to the Wyoming rolls in form, mineralogy, and distribution of elements; their genesis also may be similar to the deposits in Wyoming.

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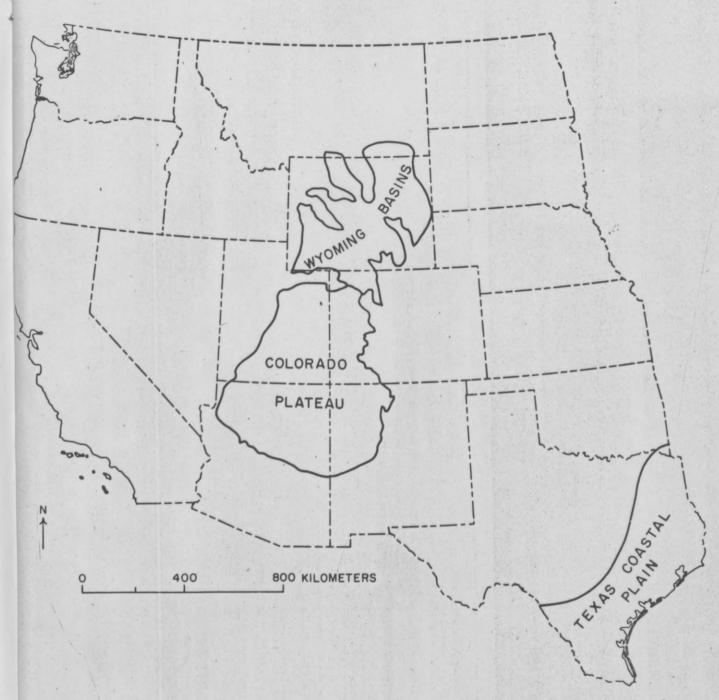


FIGURE 1. -- Map of the Western United States showing the principal uranium-bearing areas.

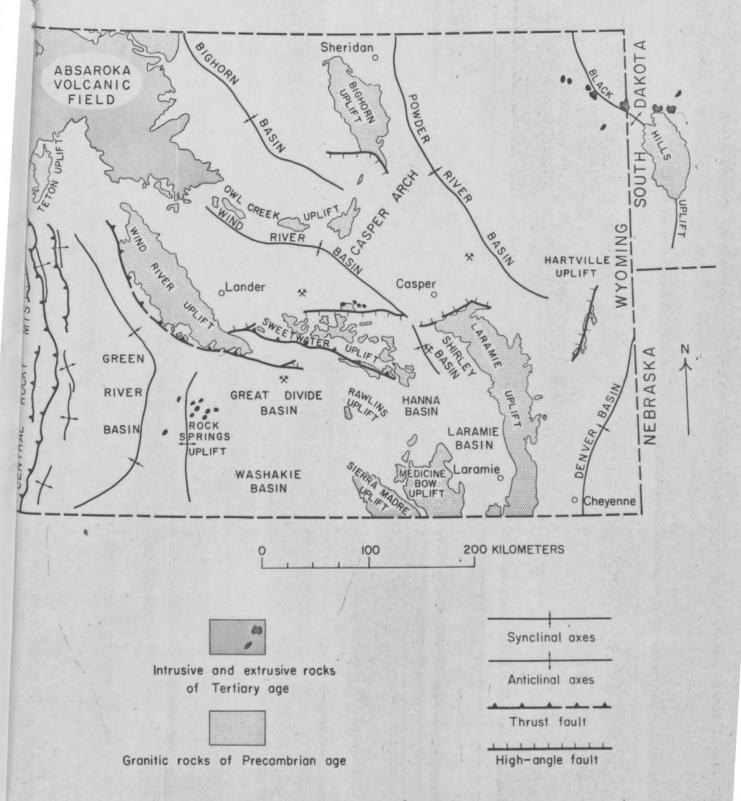


FIGURE 2. Tectonic map of Wyoming showing major uranium mining areas (父).

Modified from Cohee and others (1961).

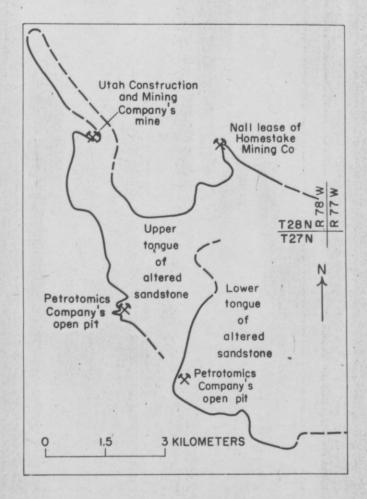


FIGURE 3.--Plan map showing outline of upper and lower altered sandstone tongues. Shirley Basin, Wyoming. Edges of tongues dashed where approximately located.

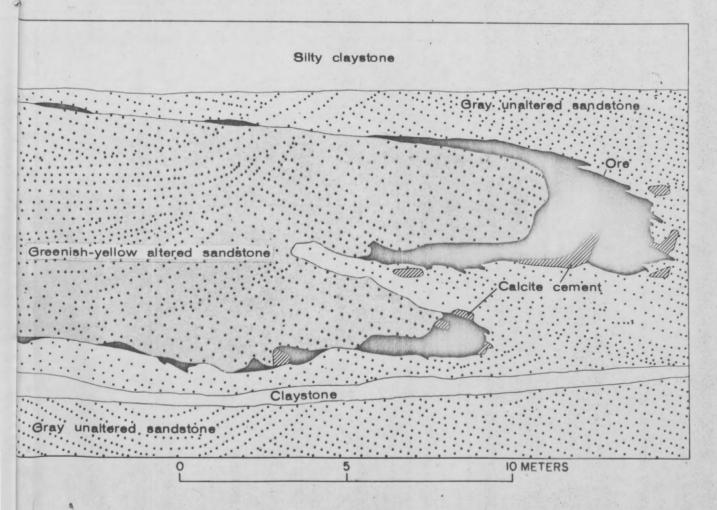


FIGURE 4. -- Section normal to edge of altered sandstone tongue showing relation of ore and calcite cement to altered sandstone.

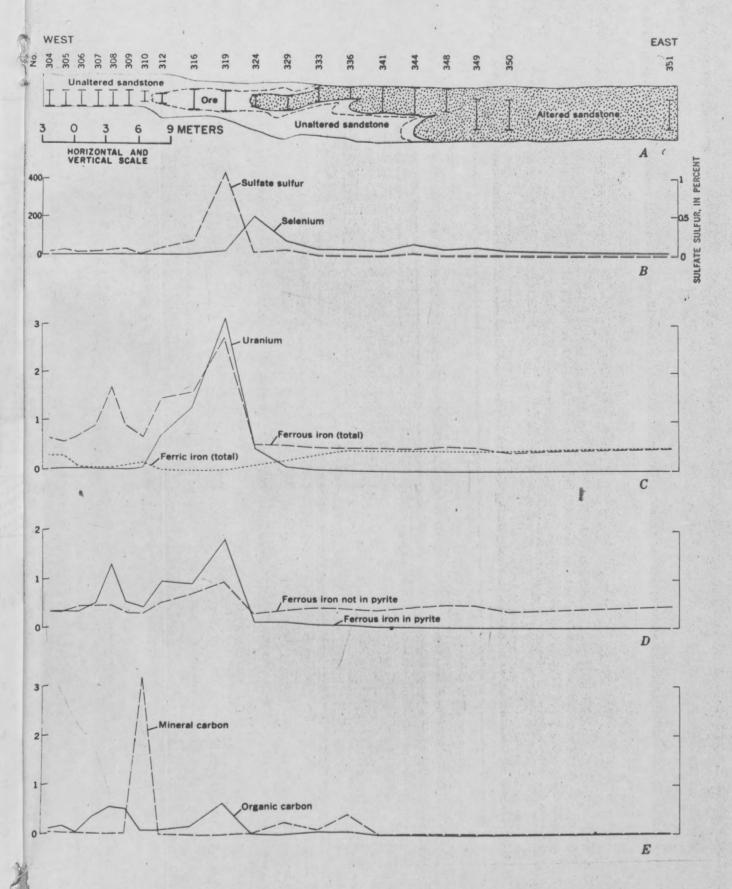
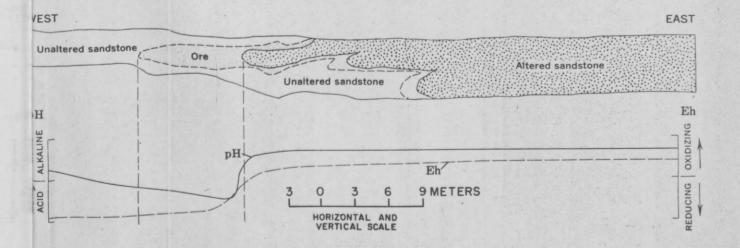


FIGURE 5. -- Vertical section (A) through uranium ore body showing sample locations and analytical date (B-E) from Petrotomics Co. pit, Shirley Basin, Wyoming.



IGURE 6.--Postulated Eh and pH conditions during transportation and deposition of uranium and other elements in the Wyoming basins.

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