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# CONCEPTS OF GENESIS OF SANDSTONE-TYPE URANIUM ORE DEPOSITS

#### HANS H, ADLER

#### ABSTRACT

Although the sources of uranium in sandstone-type ores are in most cases obscure, the formation of the ore deposits can be attributed on the basis of sulfur isotopic data to the precipitating action of hydrogen sulfide of bacteriological origin. In some areas deposition has been related heretofore to natural gas, but the isotopic evidence does not support this origin. Certain fracture-controlled and breccia-pipe deposits also show isotopic evidence of having been formed under conditions similar to those existing for the sandstone-type ores.

Geochemical relations suggest that salt-dome structures as well as fractures transecting carbonaceous sediments may be favorable sites for uranium accumulation. Certain features provide a unique basis for appraising the uranium potential of salt domes.

The sulfur isotopic data reflecting bacteriological involvement in uranium deposition in no way refute the possibility that the uranium was initially derived as an igneous emanation, but the tuff-leach mechanism may provide a more favorable model on which to base exploration in some areas of the U.S.A.

The concepts discussed may aid greatly in developing models of uranium distribution which may be applied in future exploration.

#### INTRODUCTION

The geological and geochemical processes that fashioned the uranium deposits of the western United States are gradually becoming better understood as a result of the many studies that have been undertaken and are currently in progress. With the foundation that has been established, it is possible to construct conceptual models of uranium ore distribution to serve as a basis for future exploration. It is not the intent in this paper to demonstrate these models or their application but rather to present some of the important concepts and data that provide the background for their construction.

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Since geological criteria used in exploration for or evaluation of mineral deposits in some way involve assumptions of genesis of the ore metals, they are vulnerable to controversy. This is especially evident in the number of competing theories that exist for the formation of uranium ore deposits. Each observer has his own conception of the problem, and the choice of values to support his conclusions is generally biased to some degree. Nevertheless, the source of uranium, although controversial, is explicable.

We are considerably more fortunate, however, with respect to our knowledge of the factors specifically involved in uranium emplacement. Although certain environmental relations have been recognized for many years, several new lines of evidence have recently shed considerable additional light on the uranium ore-forming processes. Fortunately, the factors effecting ore deposition in sedimentary rocks are to a large degree independent of consideration of the uranium source and may be treated accordingly.

#### UNONHDIZED SANDSTONE-TYPE URANIUM ORES

The bulk of the uranium ore reserves of the United Stares are contained in continental sedimentary rocks and are commonly referred to as sandstonetype ores. For the most part, these ores are mineralogically simple, i.e., they contain few important metallic minerals other than uraninite or coffinite. Major deposits of this type are found in the Grants-Laguna district of New Mexico and in the Wind River and Shirley basins of Wyoming. Unoxidized copper-uranium and vanadium-uranium ores in which the accessory metals are relatively abundant also occur in sedimentary rocks, but they constitute a minor part of domestic uranium reserves. The vanadiferous ores are best developed in the central Colorado Plateau area and in the Powder River Basin and Black Hills regions in Wyoming and South Dakota, and their oxidized analogs are also encountered there. The more important copperuranium deposits are found in the southwestern Colorado Plateau.

It has been established from geological observation that the environment of deposition of all three types of black ores is characteristic of continental and marginal-marine clastic sediments deposited under fluviatile conditions. The ores are in virtually all cases associated with organic debris that has been assumed to play a dominant role in the precipitation of the uranium as well as accompanying sulfide minerals. The uranium deposits are commonly, although not in all cases, near mudstone-sandstone contacts and are commonly bottomed by impermeable strata. The presence of the mudstone is important because it (1) helps define the effective margins of paleostream channels by providing a permeability barrier to transgressing ore solutions, (2) establishes, by comparison with sandstone abundance, an indication of the regional transmissivity of the sediments, and (3) helps support bacterial viability by means of its organic-matter content, a factor that formerly has been neglected within the framework of theories on ore genesis.

Although the sources of the uranium are in most cases obscure, i.e., indiscernible from geological and chemical relations observed at the deposition site, many geologists have felt compelled to argue for a favored theory of GENES

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genesis. This has been particularly noticeable in the writings of Gruner (6), Kerr (12) and Page (18). There is in fact no good reason to doubt that uranium could have been derived from either of the most commonly suggested sources, i.e., magmatic hydrothermal or weathering solutions, and the lack of confirming geological indications which has been taken as a counterpoint acither points to nor disproves the specific source. For example, the argunent that a magnatic hydrothermal source is untenable because of the apparout absence of magmatic traits and obvious nearby feeder structures or igneous rocks would appear to be unjustified on geochemical grounds, because the likelihood of identifying a sandstone-type ore deposit with a magmatic source would become increasingly remote as magnatic solutions become mixed with copious ground waters and lose their magmatic characteristics. This premise is supported by geochemical studies that have demonstrated that uranium can migrate for considerable distances in ground water and that this capability is a function of the complexing of the uranyl ion, UO<sub>3</sub><sup>2\*</sup>, regardless of its source, with carbonate or sulfate radicals prevalent under various pH and Eh conditions characteristic of sedimentary environments. On the other hand, for the same reasons the possibility of a "rock-leach" derivation also cannot be denied.

It may appear to some geologists to be somewhat paradoxical, therefore, that although a magmatic origin is not necessarily disclaimed, the tuff-leach theory seems to provide a more favorable model on which to base exploration activities in some areas of the U.S.A. For instance, one should certainly not ignore the possibility of widespread influence of the tuffaceous White River formation on ore distribution in Wyoming and South Dakota, and this factor should be seriously considered in any attempt to define areas of discovery potential in that region of the country.

The persistent association of uranium with fossil-plant remains and the almost ubiquitous presence of pyrite presuppose a chemical relationship between the metallic constituents and the organic matter. Although this association is regarded as of prime importance in the development of uranium ore bodies, its genetic significance has only been charified during the past few years through studies of stable isotopes. Among the more interesting precedent explanations are the chemical absorption of the uranium by humates and other organic compounds, interaction of uranyl and sulfide ions accompanying organic material undergoing degradation, redox interaction of uranyl and ferrous ions, and replacement of calcite and detrital minerals by uranium oxide. The suggestion that the quantity of carbonaceous material was insufficient to constitute a control on uranium deposition has also been encountered.

The same relationship can also be considered on the basis of the isotopic studies of Jensen (8), who concluded from his data that sulfate-reducing anaerobic bacteria promoted deposition through evolution of the effective reductant hydrogen sulfide. Although no attempt will be made here to review all of the evidence supporting such a consideration, e.g., in the works of Kulp, Ault and Feely (14), Feely and Kulp (3), and Thode, Monster and

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Dunford (23), it should be pointed out that a considerable amount of investigation involving various natural environments has been undertaken, and it is presently possible in many cases to distinguish between certain conditions of formation of sulfide and sulfate minerals by measuring their  $S^{32}/S^{34}$  ratios and to recognize thereby sulfides formed through bacteriological agencies (9, 10).

The original specific indication that hydrogen sulfide produced by sulfatereducing bacteria may have been instrumental in precipitating uranium ores exists in the isotopic analyses made by Jensen (8) on sulfides from various sandstone-type uranium deposits of the Colorado Plateau and Wyoming. The conclusion is based primarily on the unusually high  $S^{32}/S^{34}$  ratios and comparatively broad spread in ratio values for sulfides associated with the uranium minerals comprising the ores and has been substantiated by a much more extensive study by Jensen and Field (11).

One need only understand the requirements for viability of sulfatereducing bacteria to appreciate their role in ore formation. The growth process of these bacteria involves the reduction of sulfate accompanying the consumption of organic matter which is their source of energy. The sulfate sulfur serves specifically as a hydrogen acceptor, and the H<sub>2</sub>S is evolved as a waste product. Experiments have shown that the amount of H<sub>2</sub>S produced is related directly to the concentration of both nutrient and sulfate ions, whereas the degree of enrichment of S<sup>32</sup> in the hydrogen sulfide is inversely related to the magnitude of H<sub>2</sub>S production or the rate of reduction of the sulfate. Thus, the more energetically and copiously the gas is produced, the lower will be its S<sup>32</sup> content (16).

The nutritional requirements of the bacteria are met by an abundance of organic debris in the sediments and by sulfate ions which are a widespread component of ground waters. The anaerobes are almost ubiquitous in nature and are known to persist far below the surface of the earth, e.g., they have been reported in petroleum deposits and salt-dome cap rocks. Their role in cap-rock carbonate and sulfur formation has been described by Feely and Kulp (3).

The unusual characteristic of bacteriologically produced sulfides in sandstone-type uranium ores is their extraordinarily high  $S^{32}$  content, or more specifically their high  $S^{32}/S^{34}$  ratios, with respect to ordinary sedimentary sulfate. Isotopic fractionation, of course, can be accomplished inorganically, but the reduction of sulfate apparently does not occur except at temperatures above 500° C. Hence, sulfate in ground waters is seemingly reduced only by bacteriological means, a factor which is of considerable geologic importance.

It is generally possible to make a distinction between bacteriogenic and magmatic hydrothermal or igneous sulfides (10) on the basis of the degree of  $S^{32}$  difference from the isotopic norm (meteoritic ratio of 22.220, which is most likely the same as primordial earth sulfur) and the heterogeneity or homogeneity of the isotopic composition. Moreover, a similar comparison can be made between sulfide formed bacteriologically and that in gases associated with petroleum deposits. Thode, et al. (23), have shown that for

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given oil fields petroliferous  $H_2S$  has a fairly homogeneous isotopic composition that is not much different from petroleum sulfur. This is to be expected, if the gas is derived by maturation processes and biogenically, possibly because of the depletion of sulfate in connate waters at the margins of oil pools and the opportunity for mixing during accumulation in gas pockets and traps.

The isotopic ratios observed for gaseous and crystalline sulfides from various sources are illustrated in Figures 1 and 2. It is particularly note-





worthy that although the ratio spectra differ for sulfur from different oil fields, e.g., for Wyoning and Cauadian pools, the spread is, nevertheless, rather narrow for any given deposit. This observation is of considerable significance in regard to its implication concerning the possibility of uranium having been precipitated in ore deposits by means of petroliferous gases.

Grutt (7) has proposed that the uranium deposits in the Tertiary basins of Wyoming are related to occurrences of natural gas associated with petroleum deposits. His contention is that gas from underlying Cretaceous and older formations presumably seeped into permeable Tertiary strata and co-

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except the unambiguously magnuatic hydrothermal ores have yielded isotopic evidence of having been derived bacteriogenically and not one has shown indication through isotopic evidence of a petroliferous association. Grutt's idea is, therefore, unsubstantiated by this line of evidence. It is nevertheless noteworthy that several areas containing both uranium and gas deposits, e.g., the Maybell and Baggs districts of north-central Colorado and south-central Wyoming and the Karnes County uranium district in Texas, are only now being studied from this standpoint, and a petroliferous origin for the  $H_3S$  cannot, therefore, be ruled out for these areas.

The fact that sulfide minerals from the Gas Hills, as well as many other uranium ore deposits, show widespread S<sup>32</sup>/S<sup>34</sup> ratios is significant from the standpoint of establishing certain geological conditions that may have prepared the ground for ore deposition. Since the formation of the H<sub>\*</sub>S requires an energy source for the bacteria, one should expect to find uranium deposits at or near accumulations of organic matter, as is indeed almost always the case. Although recognizable plant remains in the sandstones constitute the most important food reserve, adjacent mudstones may also have contributed to the supply of organic matter. Concentrations of uranium have been observed about mud galls, and there is ample indication of the accumulation of organic debris in muds. These geochemical and isotopic relations are repeated again and again in the ore-bearing regions of the United States and clearly indicate the nature of the geochemical control. All the important productive ores that have been analyzed isotopically, e.g., those in the Gas Hills, Ambrosia Lake, Jackpile and Big Indian Wash deposits, show evidence (Fig. 1) of having been formed by the reducing action of bacteriogenic hydrogen sulfide, and there are no known sandstone-type deposits that do not appear to conform.

#### CYCLIC ACCRETION OF URANIUM IN SEDIMENTS

The concept of cyclic accretion of uranium ores was launched with considerable impact by Gruner's (6) notable treatise on the multiple accretion of uranium ores and Grutt's (7) astute observations of leaching and enrichment in the Gas Hills. Although it may never be documented that application of the multiple accretion hypothesis has led directly to the discovery of ore, the concept is basic to all uranium exploration and has undoubtedly been applied gainfully by knowledgeable geologists.

Gruner's (6) hypothesis is essentially concerned with the culmination of all preceding events involved in the aggregation of uranium from various sources. It presumes that uranium accretion could have begun at any time under favorable geologic conditions, but that the final accumulation as we see it today may be the result of several cycles of deposition, oxidation, migration, reprecipitation and, probably, enrichment. That Gruner (6) chose to derive uranium initially from tuffaceous rock and granite for his purpose is of no great consequence and may be ascribed to the special appeal of weathering solutions as a sustaining source of uranium over a considerable span of geologic time. Therefore, one should gain from Gruner's (6) hypothesis not the suggestion of the initial source of uranium but the basic idea of accumu-

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lation by multiple accretion through reworking of pre-existing uranium deposits.

The observations of Gruner (6) and Grutt (7) were followed by the radiochemical studies of Rosholt (20, 21, 22) and Robinson and Rosholt (19) which yielded unquestionable indications of uranium accretion in recent geological time. Analyses of various radiogenic nuclides in ore samples from the Gas Hills and Black Hills of Wyoming gave proof of recent leaching of uranium and re-accumulation in deeper sediments. In the Black Hills area, oxidation, leaching and redeposition can be related to the position of the present ground-water table. Redistribution of the ore apparently did not begin before 180,000 years ago and has continued to the present. Initial deposition was ascertained to have taken place more than 250,000 years ago for deposits now lying at or above the present water table. Unfortunately, no age can be established for the initial accumulation of uranium in these deposits because of the limits imposed by the half-lives of the radionuclides used in establishing the age relations. In the Gas Hills, migration and accumulation have apparently taken place within the past few tens of thousands of vears (20).

The position of many uranium ore districts on the flanks of or adjacent to anticlinal uplifts testifies that such uplifts probably exerted a dominant influence on ground-water drainage peripheral to the uplifted areas and were effective in limiting ore distribution. It would be possible at this point to speculate about the structural control of paleodrainage patterns and groundwater flow, the influence of anticlinal uplifts on drainage trends for both dormant and active structures, and possible related effects on uranium migration and localization, but such subjects are beyond the immediate scope of this paper. These factors are, nevertheless, of primary importance in establishing patterns of ore distribution. They will undoubtedly assume increasing importance in future exploration and will present challenging problems to the exploration geologist.

It is sufficient for the purpose of this paper to emphasize the importance of the concepts that have evolved from geologic observations on migration and accretion, age relations among nuclides involved in these processes, and the geochemical relations prescribed by isotopic studies, for they constitute an invaluable framework for formulating models to guide future exploration for sandstone-type ores. Certainly when combined with a knowledge of the hydrologic setting, the regional structural and stratigraphic relations, and some intelligent notions concerning the sequence of geologic events, it should be possible to depict probable uranium distribution patterns in sediments. This may be a requisite for discovering new ore bodies and new ore districts which are not revealed by surface indications.

That uranium concentrates at or below the water table through re-accumulation and accretion has been substantiated radiochemically, but it was not too many years ago that the suggestion of supergene leaching and enrichment encountered serious doubt or at best cautious speculation. Currently, there is considerable reluctance in accepting isotopic evidence of bacteriogenic mechanisms in minerhas shown that it is when it requires the

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tively shallow regions of crustal rocks, hypogene solutions may become oxygenated and lose their capacity to precipitate uranium as well as sulfides. Because uranium is readily oxidized to the uranyl state, precipitation does not ordinarily take place again until reducing conditions are encountered. It is interesting to note in this respect that no sandstone-type uranium deposit has shown evidence from isotopic data of magmatic hydrothermal identity.

It may be argued on the basis of the examples cited that the absence of unique magnatic hydrothermal characteristics in ore bodies is no criterion for denying such an origin for the initial solutions inasunch as the influence of the sedimentary environment is demonstrable immediately adjacent to igneous areas. On the other hand, these observations cannot be used to deny other source concepts.

It is the writer's opinion that sulfur-isotopic data reflecting bacteriologic involvement in uranium deposition in no way refute the possibility that the uranium was initially derived as an igneous emanation. The only conclusion that can be drawn from such data is that bacteriogenic  $H_2S$  was the effective precipitant. This, in itself, is of considerable significance inasmuch as it directs attention to the locus and the condition of emplacement, not to the source of the uranium, and should encourage the exploration geologist to focus attention on geologic environments favorable for bacteriological activity, past or present, as a guide to uranium ore deposits.

#### BRECCIA-PIPE DEPOSITS

Although substantially less common and productive than other ore types, uranium deposits within breecia pipes in sediments are of considerable interest because of the unusual geological and geochemical control on mineralization. Gabelman and Boyer (4) have described several of these pipes in the vicinity of the Grand Canyon, Arizona, and others are known in the Grants district, New Mexico, and elsewhere. The Orphan pipe near Grand Canyon, which has been mined for uranium, is considered by Gabelman and Boyer (4) to be a collapse structure of cryptovolcanic origin. The mineral assemblage consists of uraninite and various sulfides of copper and other metals as well as gangue minerals. To date, only two sulfide samples have been analyzed isotopically (11) (Fig. 2). Although the geologic character of the deposit is indicative of magmatic hydrothermal emplacement, the isotopic data do not support this conviction, and additional investigation seems desirable.

Sulfides from the uranium-bearing Woodrow pipe in the Laguna district, New Mexico, yield  $S^{32}/S^{34}$  ratios that are distinctive (Fig. 2) in that they reflect bacterial fractionation by their inhomogeneity but are considerably lower than those normally found in sulfides from sandstone-type ore deposits. Jensen and Field (11) interpret the extended range of low isotopic values to be an indication of the availability of a finite quantity of sulfate that was gradually depleted with consequent progressive lowering of the S<sup>32</sup> content in the remaining sulfate and resultant hydrogen sulfide. The isotopic data are suggestive of microbial transformation of sulfate to sulfide but provide no information regarding the source of the uranium or the origin of the structure.

structures intersecting es for the accumulation where sediments with hyporation along such re either was deposited stone-type origin; howite criteria for location oroblem are brought to ta on deposits in and In this area uranium as and in the Dakota parallel to the contact mine and the adjacent tiles to the cast of the geologic setting of these in, apparently favoring rect, one may conclude and sulfides were pregenic H<sub>2</sub>S since petrove supported bacterial

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It is particularly intriguing that both pipes occur in sediments and show no isotopic indication of magnatic hydrothermal emplacement, although a magnatic hydrothermal origin probably would be found acceptable on the basis of a more conventional geological appraisal. In evaluating the geological potential of structures of this type for manium—and the same could probably be said for ground heavily fractured by intersecting fault systems such as in the Ambrosia Lake area—the basic idea of accumulation stemming from interaction with products of microbial activity ought to be invoked, and the source of the manium may be regarded to some extent as incidental. A digression from this approach is warranted, however, on the basis of the unique discovery by Jensen and Field (11) of S<sup>32</sup> enrichment in the Schwartzwalder deposit which was a chance byproduct of an endeavor to establish typical magnatic hydrothermal isotopic ratios for manium-bearing sulfide deposits.

Isotopic ratios obtained on sulfides from the Schwartzwalder mine near Golden, Colorado, reflect the superposition of bacteriogenic conditions on a magmatic hydrothermal milieu in crystalline gneisses and schists. Although the dense silicified veins yield sulfides having typically magmatic isotope ratios, samples from near-surface extensions of these veins, where mamimm oxide and iron sulfide have been reworked by supergene processes, yield very different ratios with much higher  $S^{32}$  values (Fig. 2). Jensen and Field (11) have interpreted these high  $S^{32}/S^{34}$  ratios as resulting from near-surface bacterial activity brought on by the formation of sulfate ions from oxidation of primary sulfides in the vicinity of organic matter flushed into the veins from the ground surface. This unusually interesting observation provokes consideration of a mechanism of sulfide enrichment unlike that involved in the classic examples of sulfide replacement and suggests the possibility of a complicated interplay of geologic processes under supergene conditions.

#### SALT-DOME DEPOSITS

The studies of Feely and Kulp (3) have given us considerable insight into the genetic and geochemical factors involved in uranium emplacement in salt-dome structures.

The uranium occurrence in the Palangana salt dome in Duval County. Texas, described by Weeks and Eargle (24), lies in a wet aquifer sand approximately 100 feet above the cap rock and at a depth of 325 feet from the ground surface. The sand is reported to contain pyrite, hydrogen sulfide, organic debris including fossilized animal and plant remains and is locally impregnated with a small amount of oil.

Three pyrite samples collected by the writer and analyzed by Jensen and Field (11) yielded remarkably consistent  $S^{32}/S^{34}$  ratios (Fig. 1) showing considerable enrichment in  $S^{32}$  in comparison to the sulfides from Gulf Coast salt-dome deposits studied by Feely and Kulp (3). The pyrite could conceivably have been formed from H<sub>2</sub>S emanating from the cap rock; however, its  $S^{32}$  content is higher than that of any bleedwater H<sub>2</sub>S analyzed by Feely and Kulp (3) (Fig. 1). The  $S^{32}/S^{34}$  ratios for cap-rock sulfur and anhydrite

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from the Palangana dome are normal. Insufficient work has been done to conclude whether the pyrite was formed from  $H_2S$  migrating from the cap rock or was produced by microbial activity within the ore sand. However, the meager isotopic evidence would lead one to believe that the gas came from a reservoir containing isotopically homogeneous  $H_2S$ .

Space does not permit an account of the genetic relations within the saltdome environment involving petroleum, sulfur, H2S, and cap-rock sulfate and carbonate, and for this the reader should refer to the notable work of Feely and Kulp (3). Although not specifically directed toward the problem of uranium emplacement, their concepts provide a most interesting basis for appraising the uranium potential of salt-dome structures. In brief, salt domes that have penetrated sediments containing circulating ground waters are exposed to bacterial action. Petroleum accumulations peripheral to the dome provide a source of nourishment for the microbes, and the sulfate is consequently reduced to H<sub>2</sub>S which is in turn oxidized to sulfur. Since sulfur accumulates only where H2S is produced, sulfur-bearing domes, by virtue of this evidence of past bacterial activity and H<sub>2</sub>S formation, are ostensibly more favorable for uranium accumulation than structures that do not contain sulfur. The sulfur generally occurs on the dome close to the petroleum reservoir, and from a knowledge of these spatial relationships and the overlying structure it should be possible to predict the locus of uranium deposition. Criteria of this kind, applied intelligently, may prove an invaluable aid to future exploration for uranium in the Gulf Coastal Plain region.

#### FURTHER CONSIDERATIONS

The practical value of recognizing the aforementioned criteria of genesis is that they aid greatly in developing models of uranium distribution. Furthermore, they will undoubtedly lend themselves exceptionally well to assessing the geologic potential of unexplored regions. The success of future exploration cannot be assured, however, without profound knowledge of tectonic factors, sedimentary controls and what may be referred to as provincial prediliction, which is one way of explaining why uranium often is not where it ought to be. Of equal importance is the need to recognize the direct influence of ground-water migration, past and present, on ore accumulation and redistribution. There does not appear to be a simple relationship between the water table and the redox zone, for many of our large ore bodies are significantly deeper than existing water tables, which are considered by some geologists to be currently in their lowest stratigraphic position. The problems that remain are compellingly attractive.

The writer is indebted to Dr. M. L. Jeusen for his valuable suggestions concerning the isotopic data.

Division of Raw Materials, U. S. Atomic Energy Commission, Washington, D. C., fanuary 17, 1963

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