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Lacustrine-humate Model: Sedimentologic and Geochemical Model for Tabular Sandstone Uranium Deposits in the Morrison Formation, Utah,

and Application to Uranium Exploration

by

Fred Peterson and Christine E. Turner-Peterson

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UNIVERSITY OF UTAH RESEARCH INSTITUTE EARTH SCIENCE LAB.

This report is preliminary and has not been edited or reviewed for conformity with U.S. Geological Survey standards. Illustrations

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LACUSTRINE-HUMATE MODEL: SEDIMENTOLOGIC AND GEOCHEMICAL MODEL FOR TABULAR SANDSTONE URANIUM DEPOSITS IN THE MORRISON FORMATION, UTAH, AND APPLICATION TO URANIUM EXPLORATION

By Fred Peterson and Christine E. Turner-Peterson

Abstract

The lacustrine-humate model is derived from the observed occurrence of tabular uranium ore deposits in sandstone beds that consistently lie close to a distinct type of offshore-lacustrine gray mudstone. This report attempts to synthesize field observations, literature data, and preliminary laboratory work into an hypothesis or model of formation of this type of tabular uranium ore deposit. The basic premise of the model is that humic and fulvic acids generated in the offshore muddy sediments of humus-bearing lakes were expelled by compaction or seepage into nearby sandstone beds where the organic acids were fixed as tabular humate deposits. Subsequently, uranium-bearing ground water passed through the sandstone where the humate fixed and concentrated the uranium, forming tabular sandstone uranium deposits.

Introduction

The lacustrine-humate model was proposed by Turner-Peterson and Peterson (1978) to account for facies control of uranium mineralization in fluviallacustrine rocks of the Jurassic Salt Wash Member of the Morrison Formation, Utah, and the Triassic Stockton Formation of the Newark Group, New Jersey and Pennsylvania (fig. 1). The limitation of uranium to a particular facies association in both of these areas (figs. 2 and 3) implies that certain inherent features in the depositional environment were prerequisites for mineralization, and these features, in turn, place constraints on hypotheses concerning the processes that formed the ore. The most important constraints are that inferred pore-water and ground-water chemistry and ground-water flow patterns related to the mineralization must be consistent with those of the depositional environments (Peterson, 1977; Turner-Peterson, 1977, 1979, 1980).

The basic premise of the lacustrine-humate model is that structureless organic matter associated with uranium in the tabular sandstone ore deposits originated as soluble humic substances in the pore fluids of offshorelacustrine gray mudstones. The juxtaposition of these gray mudstones, hypothesized to be sources of soluble humic substances, with permeable sandstone beds (figs. 2, 3) appears to have been essential for uranium mineralization.

This report briefly presents the salient points of the model, with the Salt Wash Member of the Morrison Formation in southern Utah serving as an example. Features of other uranium districts are also included where applicable. The basic features of the model should prove helpful in exploration for tabular orebodies.





Figure 2.--Depositional environments in the lower and middle sequences of the Morrison Formation, Henry Mountains Mineral belt of southern Utah. Length of section about 40 km. Numbers indicate measured sections from Peterson (1978).



Figure 3: Interpretive cross section showing inferred structural and facies relationships within the Triassic-Jurassic Newark Basin of Pennsylvania and New Jersey; drawn parallel to the Delaware River

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Sedimentologic and Tectonic Setting

Discontinuous tabular uranium orebodies in the Salt Wash Member of the Morrison Formation in the Henry Mountains mineral belt, Utah, are present in fluvial and marginal-lacustrine sandstone beds that lie adjacent to a distinct type of offshore-lacustrine gray mudstone (fig. 2). The lacustrine mudstones associated with the ore deposits were deposited in the more distal facies of the lowermost alluvial-plain sequence of the Morrison, on the east side of the slowly subsiding Henry Basin. Here, fluvial energy or flow regimes and rates of clastic sedimentation by northeastward-flowing streams were low enough to allow lakes to form. Brief descriptions of most of the facies in this part of the Morrison are presented elsewhere (Peterson, 1980).

The distribution of favorable lacustrine environments was largely determined by structural movements at the time of deposition (Peterson, 1977, 1978, 1979). Formation of lakes in which the favorable gray mudstones were deposited largely depended on the interplay between tectonic subsidence and fluvial sedimentation. When the rate of subsidence in the synclines exceeded clastic sedimentation by fluvial processes, such as at times when fluvial sedimentation was by lower energy braided streams, lakes and ponds could form in the most sediment-starved parts of the synclines. The process was most

effective when the paleofold axes were oriented approximately perpendicular to the paleostream trends.

An isopach map of the lower sequence in the Henry Basin (fig. 4) shows three small synclines that existed during deposition of this part of the Morrison Formation. Braided streams that carried clastic sediment into the region flowed generally northeast and were predominately low-energy or lowflow regime types judging from the relatively high stratification ratios (Smith, 1970) of about 0.3 to 0.9. Scattered, discontinuous, favorable gray mudstone beds, which occur in the lowermost ore-bearing part of the lower sequence (section 3, fig. 2; fig. 5), coincide mainly with the middle syncline. The overall relationships indicate that the middle syncline was partly protected from fluvial sedimentation by the adjacent paleoanticlines to the southwest, allowing small lakes or ponds to form in the most sheltered and sediment-starved part of the syncline. Although not shown in this report, favorable offshore lacustrine gray mudstones associated with mineralization in the middle of the lower sequence occur in a smaller area in the middle syncline. It is important to note that the isopached areas on figure 5 as well as figure 6 show the areas where scattered, favorable gray mudstone lenses are present in or adjacent to the ore-bearing sandstone bed; no single, favorable gray mudstone bed occurs throughout the isopached area. In addition, the anticlines did not grow by upward movement; otherwise there would be evidence of erosion on their crests; instead, the entire region subsided and the anticlines merely subsided at a slower rate than the synclines.

Similarly, the relation of mineralization in the uppermost part of the lower sequence (sections 6 and 7, fig. 2) to growing folds is shown on figure 6. As on figure 5, the overall relationships indicate that the easternmost

EXPLANATION

- - •¹¹² Measured section ;thickness In feet
 - Oil and gas drill hole
- Crossbedding resultant in fluvial sandstones in lower sequence
 lsopach line sthickness in feet, dashed where uncertain; interval 50 feet

Figure 4: Isopach map of the lower sequence of the Morrison Formation in the southern part of the Henry basin. From Peterson, 1980.



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(modified from Hackman and Wyant. 1973). From Peterson (1980).





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ing total thickness of all favorable gray mudstone beds associated with ore zone 4. Inset shows known prospects and ore deposits with production or probable resources (modified from Williams and Hackman, 1971, and Hackman and Wyant, 1973). From Peterson, 1980.

syncline was partly protected from fluvial sedimentation by the adjacent paleoanticline to the west, allowing small lakes or ponds to form in the most sheltered or sediment-starved part of the syncline. Isopachs of the upper sequence do not show these folds although more data is needed to confirm this. One of the reasons the lakes did not form at this time was that the streams were of relatively higher energy types (stratification ratios of about 0.2 to 0.5) that carried large quantities of sediment and filled in the synclines with sediment as soon as subsidence occurred. Thus, the isopach maps (figs. 4, 5, 6) demonstrate that growing folds affected sedimentation by allowing lakes to form at times when clastic sedimentation by fluvial processes was not excessive.

Several small and subeconomic uranium deposits occur in the Salt Wash outside the mineral belt on the west and north sides of the Henry Basin. We suggest that the reason for this lack of significant quantities of mineral deposits outside the mineral belt is the lack of favorable gray mudstones. Sedimentologic studies and regional relationships suggest that these mudstones are not present outside the mineral belt because fluvial sedimentation was so rapid that ponding could not take place even though synclinal or basinal subsidence may have occurred.

Mudstones as Source Rocks of Humic Substances

Lacustrine mudstones associated with ore deposits may be considered as source rocks of soluble humic substances just as certain shales are considered source rocks of oil and gas. The principal differences between the two source rocks are the type of expelled organic material and the time of its expulsion. Humic substances dissolved in pore fluids are inferred to have been expelled from their source muds during or shortly after deposition, whereas oil and gas are expelled from their source rocks after a certain degree of thermal alteration. Humic substances differ from hydrocarbons by containing relatively more oxygen and generally less hydrogen, as shown in the accompanying Van Krevelen diagram (fig. 7). This diagram is commonly used in the petroleum and coal industries to show the relationships and differences between the various types of organic substances commonly found in ancient rocks and maturation pathways of the various organic materials.

The ability of humic substances to fix metals in large quantities, including uranium, apparently is largely, although not entirely, related to their relatively high content of oxygen-bearing functional groups (Schnitzer and Khan, 1978), especially the carboxyl (COOH) group (Borovec and others, 1979) and phenolic OH group. High uranium-enrichment factors for humic substances have been noted by several workers (Kochenov and others, 1965; Szalay, 1958; Schmidt-Collerus, 1969; Pauli, 1975; Walker and Leventhal, (1980).

Humic and fulvic acids are largely degradation products of plant tissues. Both acids are soluble in alkaline solutions (that is, solutions with pH greater than 7), but humic acids precipitate under acid conditions, whereas fulvic acids remain solubilized in acid conditions. Polyvalent cation reactions (especially with Ca, Mg, Fe, and Al) and surface interactions with



ATOMIC RATIO 0/C

Figure 7.--Van Krevelen diagram showing elemental ratios for some naturally occurring organic substances, kerogen evolution paths, and products of kerogen evolution. Sapropel field probably considerably larger than shown here by the only available analysis. Dots indicate analyses of humic acids obtained from lacustrine sediments by Ishiwatari (1967, 1973, 1975), Karavayev and Budyak (1960), Kemp (1973), Nissenbaum and others (1972), Otsuki and Hanya (1967), Povoledo and others (1975), Stevenson and Goh (1971), and Stuermer and others (1978). Analyses of lacustrine fulvic acids not available, but their field would overlap the right side of the humic acids field. Remainder compiled from Cooper and Murchison (1969), Dow (1977), Huc and Durand (1974, 1977), Hunt (1978), Tissot and others (1974), and Van Krevelen (1961). clays are other important means by which these acids are fixed in sedimentary rocks.

In general, gray muds deposited in reducing and alkaline conditions are considered potentially favorable source sediments for humic and fulvic acids because reducing conditions favor preservation of humic matter in the pore waters of lake-bottom sediments and because alkaline conditions favor solubilization of humic substances so they can be expelled along with the pore fluids.

Two types of organic-rich mudstones occur in the Morrison Formation, but only the favorable type is intimately related to the ore deposits. Favorable mudstones may be recognized on the basis of field criteria as well as by palynological, chemical, and vitrinite analysis.

Favorable gray mudstones, which occur in association with mineralized sandstones of the Salt Wash, commonly are finely laminated and contain carbonized plant debris, epidermal and cuticular tissue, scarce iron sulfides, and a palynomorph suite lacking the alga <u>Botryococcus</u>. These beds are noncalcareous to slightly calcareous, and they lack finely disseminated and powdery carbonaceous matter or microscopic carbon plates that are commonly found in oil shales (R. H. Tschudy, written commun., 1978, 1979). The type of organic material contained in these mudstones can be identified with a 10X hand lens in the field because they contain small, shredded fragments of carbonized wood in which the cellular structure can be recognized, and because they usually lack irregular, structureless, carbonaceous blebs and abundant powdery carbonaceous material. Humic and fulvic acids in lakes are degradation products commonly formed from plant materials and aquatic organisms. Thus, the fairly abundant plant material, well-preserved palynomorphs, and the scarce pyrite suggest that these mudstones were

deposited in lakes that may have had stratified waters and that most likely had humic and fulvic acids generated in oxygen-deficient bottom muds. Because these organic acids are toxic to <u>Botryococcus</u> (Dulhunty, 1944), the consistent lack of this aquatic alga in the favorable gray mudstones suggests that the acids may have been abundant in the lake waters.

Unfavorable gray, organic mudstones that occur in unmineralized parts of the Morrison are readily distinguished from the favorable gray mudstones. In fact, some of the unfavorable gray mudstones closely resemble oil shales. This similarity to oil shales may make them undesirable for fixing uranium because oil and gas hydrocarbons, by themselves, are known not to concentrate uranium in significant quantities (Erickson and others, 1954; Hyden, 1961; Moore, 1954; Andreyev and Chumachenko, 1964; Antropov and others, 1976).

Some unfavorable gray mudstones contain carbonized plant debris similar to that of favorable gray mudstones. However, unlike favorable mudstones, these unfavorable mudstones contain finely disseminated and structureless organic matter and microscopic carbon plates much like those found in oil shales (R. H. Tschudy, written commun., 1978, 1979). Examination with a 10X hand lens also reveals small irregular blebs of carbonaceous matter of unknown origin that lack cellular structure and appear to resemble dried oil. Locally abundant, fine, powdery carbonaceous material can also be seen with the aid of a hand lens, and, if present in sufficient abundance, it may be recognized by the sooty stain it leaves on one's hands and clothes. The absence of epidermal and cuticular tissue in the residue from palynological analysis (R. H. Tschudy, written commun., 1978, 1979) also distinguishes the unfavorable mudstones from favorable ones. Most of the unfavorable mudstones are calcareous or are closely associated with thin limestone beds.

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The common occurrence of the alga Botryococcus is one of the most interesting characteristics of the unfavorable gray mudstones. Botryococcus thrives only in humus-free lakes, apparently because high concentrations of humic and fulvic acids are toxic to it (Dulhunty, 1944). Because these lakes apparently do not provide the relatively large quantities of soluble and mobile humic substances which we postulate to be necessary for uranium mineralization, sandstone beds associated with oil-shale lake beds or Botryococcus-bearing mudstones, such as those of the Tertiary Green River Formation or the Lower Cretaceous Burro Canyon Formation, respectively, are considered poor exploration targets for uranium deposits. In addition, the relatively large amount of divalent cations (Ca, Mg) that must have been around, as indicated by chemical analyses and by the calcareous nature of these beds and their association with limestone beds, suggests that any humic substances that might have been produced were fixed in the bottom sediments by these cations and therefore could not have migrated out of the mudstones and into nearby sandstone beds.

The difference between organic materials in the two types of mudstone that contain carbonaceous material is shown in figure 8, which shows atomic hydrogen/carbon ratios versus vitrinite reflectance. The favorable gray mudstones contain humic kerogen, which is what would be expected if the kerogen in these rocks was derived from organic substances that contained appreciable quantities of humic and fulvic acids. On the other hand, the unfavorable <u>Botryococcus</u>-bearing gray mudstones contain liptinitic kerogen, indicating they have a somewhat different type of organic matter that has a closer affinity to oil shales or petroleum source rocks. The dissimilarity of the kerogens in the two different types of offshore lacustrine, gray mudstones is an important part of our model and may demonstrate that only certain types



Figure 8.--Diagram showing kerogen evolution pathways in terms of atomic H/C ratios and vitrinite reflectance (Dow, 1977). Kerogen isolated from nonradioactive gray mudstones of the Morrison Formation in the Henry Mountains mineral belt are indicated by triangles (favorable gray mudstones) and circles (Botryococcus-bearing unfavorable gray mudstones. Horizontal bars indicate range of reflectance measurements and vertical bars indicate one standard deviation on either side of the mean. Kerogen evolution pathways: A = algal kerogen; B = liptinitic kerogen; C = humic kerogen. Analyses by M. M. Page and W. G. Dow, written commun., 1979.

of organic matter participate in the ore-forming process, or that certain types of organic matter are more favorable for the mineralizing process than others.

Other types of mudstones that are not sources of soluble humic substances, and are therefore unfavorable, are red mudstones, bleached red mudstones, and calcareous gray mudstones commonly associated with limestone or gypsum. The oxidizing conditions that existed in red mudstones are not amenable to either the formation or preservation of humic substances. Mudstones that were red and were subsequently bleached are unfavorable for the same reason. Because they are now gray or green, bleached mudstones might be confused with favorable gray mudstones, but they can be distinguished by their lack of carbonized organic matter and by their close relationship to red mudstones. The abundant divalent cations in pore waters associated with the calcareous mudstones, especially Mg and Ca, would tend to fix within these mudstones any humic substances that might have been formed, rather than allow the humic substances to migrate out of these mudstones and into adjacent sandstone beds.

Precipitation of Humic Substances in Sandstone Beds,

and Associated Alteration Features

We postulate that the humic substances, which were dissolved in alkaline and reducing pore fluids of the lake bottom sediments, were expelled from the mudstones by compaction or seepage into nearby sandstones where they were fixed as tabular humate bodies. When the hydrologic conditions were such that the lakes lost water by seepage downward through the bottom sediments (fig. 9), pore fluids containing humic and fulvic acids and bisulfide (HS⁻) migrated out of the lacustrine bottom muds and were fixed in sands below the muds. If



Figure 9.--Lacustrine-humate model, "leaky" lake variation: the favorable gray mudstone lies directly above the ore-bearing sandstone bed. Pore waters in the offshore lacustrine muds bear dissolved humic substances (HA = humic acids, Fa = fulvic acids) and bisulfide (HS⁻). The waters seep downward into the underlying sandstone bed where the humic substances are fixed as a tabular humate mass. At a slightly later time, uranium carried by ground water moving laterally through the sandstone bed is fixed by the humate to form the ore deposit. uranium now marks the sites where the humic materials came to rest, then this set of paleohydrologic conditions applies to the numerous uranium deposits in the Salt Wash of the Henry Mountains region that lie in sandstones directly below favorable mudstone source rocks. In contrast, when the hydrologic conditions were such that no water was lost through the lake bottom sediments, pore waters containing the dissolved humic substances and bisulfide (HS⁻) were expelled by compaction (fig. 10) and moved laterally and(or) upward through, and were fixed within, the adjacent sandstone beds. This paleohydrologic configuration would apply to the Newark basin uranium occurrences as well as several occurrences in the Salt Wash of the Henry Basin in which the mineralization is in sandstones laterally adjacent to or above the favorable mudstones that were the source of the humic substances. Thus, pore fluids containing the dissolved humic substances moved either shortly before or during compaction and burial. The important point is that they did not move far from their source rocks because the one-to-one spatial relationship between the lacustrine mudstones and the uranium enrichment in nearby sandstones is still preserved.

Reaction of the organics with clay clasts and clay coatings on sand grains to form organo-clay complexes is the most likely mechanism by which the humic substances were fixed in the sandstone beds (fig. 11). Iron and aluminum hydroxides coating the surfaces of clay particles are abundant in near-surface sediments during early diagenesis. The hydroxides commonly form bridge linkages between the negatively charged clay films and the negatively charged organic-acid molecules, as hydroxides carry a positive charge below pH 8 (Greenland, 1971). Ground water entering the sands surrounding the Salt Wash and Newark lakes (figs. 9, 10) most likely would have had a neutral to slightly alkaline pH (about 7-7.5), in the range of normal ground water.



O URANIUM-RICH HUMATE ZONE

Figure 10--Lacustrine-humate model. compaction variation: The favorable gray mudstones are interbedded with, or lie laterally to the ore-bearing sandstone beds. Pore waters in the offshore lacustrine muds bear dissolved humic substances (HA = humic acids, FA = fulvic acids) and bisulfide (HS⁻). The waters are expelled by compaction and migrate laterally and (or) upward into nearby sandstones where the humic substances are fixed as a tabular humate mass. At a slightly later time, uranium carried by ground water moving laterally and upward through the sandstone bed is fixed by the humate to form the ore deposit. Ground water flow patterns near the margin of the lake after McBride and Pfannkuch (1975) and Winter (1978).



Figure 11.--Precipitation of humic substances delivered from pore fluids in lacustrine muds, and associated alteration features. Positively charged aluminum hydroxides $(Al(OH)_x^+)$ attract and fix organic anions $(HA^-; humic and fulvic acids)$ onto clay surfaces. Iron leached by humic substances combines with bisulfide (HS^-) derived from pore fluids of lake muds to form pyrite. The result is bleaching of clay clasts, pyrite formation, and humate precipitation. GW indicates ground water.

Thus, the hydroxides coating the clays in the sandstones would have had a positive surface charge, resulting in attraction for and precipitation of organic anions delivered by the pore fluids expelled from the more alkaline lake muds (fig. 11). Because of the inferred high alkalinity of the porewaters in the muds, the pH of the pore-waters immediately after expulsion may have been higher than 8, too high for the formation of organo-clay complexes by the bridge-linkage mechanism. Mixing of these initially alkaline humicbearing pore-waters with fresher ground water flowing through the sandstone beds, however, may have resulted in their dilution as they escaped through the permeable sandstone units. When the pH was lowered below 8, by mixing, the humic anions may then have been attracted to the hydroxides on the clay surfaces. Although hydroxides of iron and aluminum carry a positive charge below pH 8, they are most effective in fixing humic substances at around pH 7 (Evans and Russell, 1959; Martin, 1960; Martin and Reeve, 1960). In this manner, with only a slight dilution of the alkaline, humic-bearing pore waters, by mixing with normal ground water, organics may have been fixed by the hydroxides. With the hydroxides as bridge-linkages, precipitation of humate in clay clast zones as well as on clay coats on the sand grains may have been facilitated.

Iron and aluminum hydroxides coat sand grains in many environments (Hubert and Reed, 1978; Van Houten, 1972). Thus, even without clay surfaces, organics can be precipitated by iron and aluminum hydroxides, accounting for places in the tabular ores where humate impregnates the host rock by filling pore spaces. Curtis (1978) has pointed out that the most reactive phases during early diagenesis in sediments include iron and aluminum hydroxides, clays, and degraded organic matter. In addition, he notes that the presence of hydroxides in sediments is often underestimated because they are not

revealed by X-ray diffraction analyses. Proposed here is the idea that these reactive phases (hydroxides, clays, and organic matter) were important during early diagenetic stages of the Morrison Formation and Newark Group. Compaction is inferred to have caused movement of humic-rich pore fluids from the interbedded claystones and mudstones into nearby sandstones, and the interactions of the organics with hydroxides and hydroxide-coated clays in the sandstones may have caused the precipitation of the humate.

Common alteration features associated with tabular orebodies include bleaching of clay clasts and formation of pyrite in the zone of humate precipitation and uranium mineralization. These features can also be explained by the introduction of humic substances and bisulfide from the nearby lake muds. Pyrite formation probably consumed most of the iron hydroxides on clay particles around sand grains as well as in clay clasts because humic substances readily leach iron and would have made it available to join with the bisulfide to form pyrite (fig. 11). This suggests that the positively-charged aluminum hydroxides, rather than the iron hydroxides, were largely responsible for fixing the humic anions. Thus, bleaching of clay clasts and pyrite formation are viewed as alteration features that occurred simultaneously with the introduction and precipitation of organic acids expelled from the lake muds. The reaction can be summarized as follows: oxidized clay clasts and iron and aluminum hydroxides in the presence of soluble humic substances and bisulfide (HS⁻) yield bleached clay clasts, humate, and pyrite.

Because the mudstones were originally horizontal, most of the fluids migrating away from them during seepage or compaction would have had vertical chemical gradients. Therefore, precipitation of the humic substances contained in the pore fluids would have taken place in a given sandstone bed

at the same horizon above or below the mudstone source rock, resulting in the roughly tabular geometry of the precipitated humate. Preservation of the close spatial relationship between the ore deposits and the favorable gray mudstones suggests that the humic material did not move far from the source rocks. The discontinuous and irregular distribution of the tabular ore deposits is attributed to (1) the irregular and discontinuous distribution of the favorable gray mudstones, (2) movement of ground water in the sandstone beds during or slightly after expulsion of the humic substances from the mudstones, (3) local differences in porosity and permeability in the sandstone beds, and (4) Neogene leaching and redistribution of deposits that lie above the regional water table or of deposits that lie a short distance below the water table where ground water could have migrated through the sandstone beds containing the ore.

The humate-rich zones within the ore-bearing sandstones of the Salt Wash are medium gray to dark gray or brown from interstitial disseminated humic substances as well as carbonized plant debris. The pore spaces in the sandstone do not appear to be completely filled with humate, and thus the rock is not everywhere black as it is in the tabular ore zones of the Grants mineral belt in northwestern New Mexico. We feel that the mudstone beds in the Salt Wash were not sufficiently thick or efficient in generating humic substances, or that the lakes in which they were deposited did not exist long enough to provide sufficient humic material to completely fill the pore spaces in the sandstone. Of course, some of the organic substances were lost from the uranium deposits by maturation with time and temperature changes accompanying burial (Huc and Durand, 1977; Tissot and Welte, 1978) and from the effects of radiation (Breger, 1974; Leventhal and Threlkeld, 1978). To us, the difference in the quantity of humic material seems to account for the

difference in appearance between the gray to brown Salt Wash ore zones and the black ore zones of the Grants region.

A corollary of the model is that preservation of plant debris by carbonization as the so-called "carbon trash" in the orebodies is a consequence of the processes that formed the humate body rather than the chief cause of the mineralization. Most of the carbonized plant material in the sandstone beds does not occur in scours or trash pockets. Instead, carbonized plant fragments in and near the ore deposits lie scattered through the sandstone bed or are slightly concentrated along bedding laminae. The original wide distribution of plant fragments throughout the sandstone beds is indicated by the presence of silicified wood well away from the ore. deposits. This indicates that plant material originally had a far greater distribution in the sandstone beds than is apparent from the present-day distribution of the carbonized plant fragments; the plant material away from orebodies was either preserved by silicification or destroyed by oxidizing ground water or other degradation processes. For these reasons we believe that preservation by carbonization occurred only in those parts of the sandstone beds in which humate formed a protective chemical envelope that allowed carbonization to proceed. This concept is contrary to the commonly held idea that plant-trash zones occurred only in selected scours as trash piles or logjams within the sandstone beds.

Uranium Fixed within Humate Mass

During or subsequent to formation of the tabular humate body in the sandstone, ground water carrying dissolved uranium as uranyl dicarbonate or tricarbonate passed through the sandstone beds. We postulate that the uranium was fixed and concentrated largely, although not necessarily entirely, by the

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Figure 12.--Humate mass fixes uranium from ground water. Uranium carried in ground water as uranyl di- and tri-carbonate is fixed in +6 state by humate. The uranium may be reduced to the +4 state at a later time.

humate to form the ore deposit (fig. 12). Humic substances have the ability to remove and fix uranium in the oxidized (+6) state directly from ground water (Szalay, 1958, 1964; Schmidt-Collerus, 1969; Jennings and Leventhal, 1977). This ability is significant because mineralization can occur without the redox interface that is considered an essential part of other models, such as in roll-type deposits or where a stable fluid interface is hypothesized to lie between overlying uranium-bearing ground water and underlying reducing fluids. This also suggests that uranium can be carried in ground water that does not necessarily oxidize the pre-existing pyrite (Brookins, 1976; Granger and Warren, 1979), accounting for the location of tabular ores well within reduced ground without having to call upon rereduction. Alteration features, such as bleaching of clay clasts and pyrite formation, are attributed to the earlier introduction of organic-rich fluids; the later delivery of uranium in ground water apparently does not involve significant alteration of the host sandstone. Details of the processes in which uranium is fixed by humic substances are not well understood; however, the processes are believed to be a combination of chelation and complexing (Schmidt-Collerus, 1969) or of sorption followed by later reduction (Kochenov and others, 1977).

In addition to fixing at least some of the uranium, the humate probably formed a protected zone within the sandstone that may have allowed other processes that fix uranium to occur, such as adsorption by plant debris (Andreyev and others, 1962) or by processes involving sulfur. The role of sulfur is uncertain, but generation of appreciable quantities of H_2S by sulfate-reducing bacteria seems unlikely because vascular plant tissues are poor sources of nutrients for these organisms (Lyons and Gaudette, 1979). Also, small quantities of selenate probably were in the ore-bearing solutions or in waters surrounding the orebodies, judging from the presence of selenium

in the ore deposits, and the presence of selenate inhibits the process of H_2S generation by sulfate-reducing bacteria (Postgate, 1949). In addition, the presence of significant quantities of H_2S in sandstones would be expected to cause extensive bleaching of adjacent redbeds, but no appreciable amount of alteration has been found in red mudstones that lie directly above or below the ore-bearing sandstones.

No conclusive evidence has been found to indicate the source of the uranium, although most workers are of the opinion that it was derived from alteration of volcanic materials, especially ash, that were incorporated in sandstones of the Salt Wash (Waters and Granger, 1953; Cadigan, 1967).

> Application of the Model to Other Tabular Ores and Use as an Exploration Guide

As mentioned earlier, gray mudstones must have been deposited in reducing, alkaline, lake-bottom sediments to be considered favorable source rocks of humic substances because reducing conditions favor preservation of humic matter in the pore waters of the muds and because alkaline conditions favor solubilization of humic substances so they can be expelled along with the pore fluids. Also critical to the model is the type of organic matter incorporated in the sediments. Several continental sequences, both uraniumbearing and barren, were evaluated in light of these criteria, and the lacustrine-humate model apparently can explain why certain parts of the rock record, although favorable by conventional criteria, do not contain tabular sandstone uranium deposits.

An important balance between the amount of organic matter incorporated in the muds and the pH of the pore fluids appears to be an important factor in determining whether or not a particular formation is favorable or unfavorable

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for mineralization by processes involved in the lacustrine-humate model (fig. 13). Incorporation of abundant organic matter tends to lower the porewater pH because of the increased acidity associated with normal oxidation and fermentation of organic matter. This happens in peat bogs and coal swamps where the pH is commonly are as low as 3-6 (Baas Becking and others, 1960). This low pH may partly be why the Cretaceous Dakota Sandstone generally lacks significant uranium deposits. The acidity of the pore waters would have inhibited mobilization of humic acids, which are precipitated under acid conditions, and most of these acids would have remained trapped in the muds. Although fulvic acids as well as some of the humic acids may have migrated into sandstone beds and contributed to preservation of the plant debris, it is more likely that preservation of plant matter in some of the sandstone beds in the Dakota was due to widespread reducing conditions below a shallow regional water table, as suggested by the presence of carbonaceous organic matter in overbank mudstones of the Dakota. Thus, although the Dakota contains fluvial sandstones with locally abundant carbonaceous trash, and at first may appear favorable by conventional criteria, the lack of mobility of some of the humic substances decreases the likelihood of finding significant tabular uranium deposits in the formation.

Preservation of abundant plant debris in the Dakota also suggests an explanation for the apparent climatic control of many uranium deposits. The humid climate of the Late Cretaceous resulted in such an abundance of vegetation throughout the region that acid pore waters caused by decaying vegetation inhibited mobilization and expulsion of humic substances into nearby sandstone beds. Also, so much organic matter was present and reducing conditions were so widespread that any uranium that was brought into the region was trapped in a dispersed state wherever it met adsorbing organic substances or reducing conditions.



Percent organic matter

favorable for migration of humic substances

- Figure 13.--Schematic diagram showing inferred pH of pore waters in muds as a function of the amount of incorporated volcanic ash and organic matter. Alteration of ash tends to raise pH, whereas alteration of organic matter tends to depress pH. These two parameters may affect favorability of source rocks because alkaline conditions are conducive to solubilization and migration of humic substances.
- *, volcanic ash was not deposited in the Lockatong lake sediments, and the high pH for these mudstones is inferred from the presence of sedimentary analcime.

K+BB, "K" shales in Westwater Canyon Member and mudstones in Brushy Basin Member, Morrison Formation, Grants mineral belt, New Mexico.

L, Lockatong Formation, Newark basin, Pennsylvania and New Jersey.

- S, Salt wash Member, Morrison Formation, Henry Mountains mineral belt, Utah.
- C, Chinle Formation, Circle Cliffs and White Canyon districts, Utah.
- D, Dakota Formation, Colorado Plateau.

In contrast, semiarid paleoclimates appear to have been more favorable for the formation of tabular uranium deposits. In these climatic conditions, plant growth was more restricted, occurring only along water courses, in and near lakes, or at higher elevations where rainfall may have been more frequent. The greater aridity tended to discourage growth in other areas. Consequently, the amount of vegetation incorporated within the lake muds would probably have been less than under humid climatic conditions. The pH of the lake muds would therefore not have been lowered considerably by degradation of organic material incorporated in them. In addition, semiarid climates tend to favor development of alkaline lakes because of the concentration of ions by evaporation. These two factors combined would result in more alkaline pore waters in the anoxic lake-bottom sediments. The alkaline pore waters would promote solubilization of humic substances formed by degradation of the available plant material, so that these substances could be expelled into nearby sandstone beds where they could be fixed as tabular humate masses. Oxidizing conditions at and near the surface in sediments surrounding the lakes would tend to keep uranium in solution until it encountered the humate trap, rather than allow the uranium to be fixed in a dispersed state as it may have been in the Dakota.

Proposed here, therefore, is the idea that certain climates are conducive to the formation of tabular humate masses in sandstone and are therefore favorable for the formation of tabular uranium deposits. We feel that climate may be an important large scale factor in determining the favorability of finding tabular uranium deposits in different parts of the rock record.

In addition to the development of alkaline lakes in a semiarid climate, the incorporation of large amounts of volcanic debris in the lake muds at the time of deposition also favored mobilization of humic material. Alteration of

volcanic ash may have caused a rise in pH sufficient to solubilize humic acids and allow them to move out of the muds with the pore fluids. This probably happened in the Salt Wash of the Henry Mountains region (fig. 13) where mudstones associated with tabular orebodies are bentonitic, and it also applies to mudstones near ore-bearing sandstones in the Chinle Formation of the Colorado Plateau. In the Westwater Canyon Member of the Morrison Formation of the Grants region of New Mexico, the "K" shales, which interbed with the ore-bearing sandstones, would also be favorable source rocks for humic substances (fig. 13). Large amounts of volcanic debris incorporated in the muds of the "K" shales raised the pore-water pH high enough to effectively strip the muds of humic material and allow the pore waters to move the humic material into nearby sandstone beds, accounting for the world's largest tabular sandstone uranium orebodies. The increased pore-water pH in the "K" shales also resulted in complete destruction of plant fragments in the muds, so that the mudstones presently contain no organic matter at all. The "K" shales in the Grants region, which are lacustrine in origin (Turner-Peterson and others, 1980), are not only much thicker than the lacustrine mudstones in the Salt Wash of the Henry Mountains region but also contained more volcanic debris, which suggests that the primary difference between tabular Salt Wash ores and tabular Westwater Canyon ores is a matter of scale rather than origin.

Incorporation of volcanic debris in the lacustrine mudstones was probably the most effective way in which the pH of the pore fluids was raised, but alkaline pore fluids exist in some lakes in the absence of volcanic debris. In the Triassic Lockatong Formation in the Newark basin (fig. 13), for instance, sedimentary analcime occurs with no apparent tuffaceous precursor (Van Houten, 1962). A pore-water pH of 9 or greater probably existed in the

Lockatong pore fluids, based on recent studies of analcime in lake sediments devoid of volcanic material (Schreiber and others, 1972). Thus, even without volcanic debris, certain lake sediments contain pore waters that are sufficiently alkaline to solubilize humic substances and move them into adjacent sandstone beds.

No single criterion is sufficient to determine whether a particular mudstone is favorable or unfavorable as a source rock for humic substances, with the possible exception of <u>Botryococcus</u>, which appears to be a good indicator of the lack or scarcity of humic and fulvic acids in the lake waters. The type of organics and the early chemical nature of the pore waters probably are the critical factors. Field criteria, palynological analysis, elemental chemical analyses, and vitrinite reflectance are several techniques that are being used in discriminating favorable from unfavorable source rocks. As in oil and gas source-rock studies, however, a combination of criteria and tests is more reliable than any single one. At this point, using the lacustrine-humate model as a guide, favorable rocks (Morrison and Chinle Formations) can be readily distinguished from unfavorable rocks studies, the lacustrine-humate model could prove to be a significant exploration guide for tabular sandstone uranium deposits.

Summary and Conclusions

The lacustrine-humate model can be summarized as follows:

1. Formation of alkaline and reducing lakes in a semiarid climate, in areas of tectonic subsidence. Lake-sediment pore fluids contained solubilized humic substances.

2. Expulsion of the humic substances from the gray lacustrine mudstones, by seepage or compaction, into adjacent sandstone beds.

3. Formation of tabular humate bodies within the sandstone beds by reaction of humic-rich pore fluids with hydroxides on clay particles coating the sand grains.

4. Subsequent flow of uranium-bearing ground water around and through the humate bodies which extracted and concentrated uranium through time.

The lacustrine-humate model differs from other models in that the humic substances are inferred to have migrated only short distances from mudstone beds that lie near the ore-bearing sandstones. The model is also an attempt to work within the constraints of facies control: pore-water and ground-water chemistry and flow patterns are based on reconstruction of depositional environments and are consistent with what would be expected in a natural system containing those environments.

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