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CLAY MINERALS IN THE MUDSTONES OF THE ORE-BEARING FORMATIONS OF THE COLORADO PLATEAU*

By

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By W. D. Keller

ABSTRACT

Mudstone samples were collected from uranium-vanadium ore-bearing sedimentary formations on the Colorado Plateau to determine if differences could be detected in the gross clay mineralogy between red and green mudstones which are otherwise stratigraphically equivalent, between ore-bearing and barren mudstone, and between other relationships referred to in the conclusions below. Clay mineral identifications, which were made by the use of X-ray powder diffraction patterns, differential thermal analyses, and petrographic examinations, lead to the following conclusions concerning the gross clay mineralogy of the mudstones.

Red and green (also purple and white) counterparts of mudstone do not show a significant difference in gross clay mineralogy in the Rico, Cutler, Moenkopi, Shinarump, and Chinle formations, and the Salt Wash and Brushy Basin members of the Morrison formation. Clay minerals within all the common clay mineral groups may be constituents of red and green mudstone without regard to color. No difference was detected in the gross clay mineralogy of red-green counterparts whether the green color was developed along mineralized fault planes which transected the red beds long after lithification or in isolated spots within thick, relatively impermeable mudstones whose color change may have occurred during diagenesis.

Mudstones may be ore bearing or barren without necessary differences in their clay mineral components.

Alunitization of red Moenkopi mudstone at the Oyler mine in Capitol Reef National Monument was preceded by bleaching of the red ferric oxide pigment, mobilization of the iron as sulfate and the formation of jarosite, removal of calcite, and formation of some gypsum.

It is exceedingly difficult to find unequivocal evidence of effects on the rocks from the solutions which carried uranium and vanadium apart from the ore minerals themselves.

INTRODUCTION

Mudstones, of which clay minerals are major components, constitute an integral part of the sequence of sedimentary rocks which contain uranium and vanadium ore on the Colorado Plateau. It is probable that the mudstones have played an essential role, one way or another, in the precipitation, localization, and concentration of many of the ore deposits. For example, the bulk properties of mudstones, especially low permeability, have made them important as barriers, traps, and guides for ore-bearing solutions (Mullens and Freeman, 1955). The chemical and mineralogical properties of the clay minerals, on the other hand, are probably more important where replacements by ore minerals occur. The color of the mudstones also has been of special interest because it has been observed that a high ratio of green to red mudstone and sandstone can be successfully used as an indicator of favorable ground (Weir, 1952) on the Plateau.

In consequence of close association of mudstone with ore deposits, and of the red-green (similarly, purple-white) color relationship to ore-bearing ground, at least two major questions have arisen regarding their mineralogy. Does the clay mineralogy of mudstones in localities productive of ore differ from that in barren country, and does the clay mineralogy of the green mudstone differ from that of the red which is stratigraphically equivalent?

This report is concerned with the above questions. It does not include consideration of the clay minerals finely disseminated in the sandstones or of the vanadiferous clay minerals which are ore minerals.

ACKNOWLEDGMENTS

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THE RED-GREEN PROBLEM

With reference to the question whether the clay minerals in red and green, stratigraphically equivalent mudstones are different, it is relatively simple to collect specimens from red and green counterparts at one locality and analyze them mineralogically to arrive at an answer. An analysis from a single locality merely opens up the geologic problem, however, because green and red clay-bearing rocks occur in contact over a wide variety of geologic conditions which bear on the deposition and concentration of ore.

For example, although the ratio of green to red mudstones and sandstones is higher in favorable ground, it is also recognized that apparently similar green-red pairs, except for ratios of green to red, occur in red beds far removed from localities in which uranium-vanadium mineralization has occurred. The problem enlarges, thereby, to inquire if red-green counterparts from within ore deposits are different from those in barren ground. Next, where green mudstones are mineralized, were the ore-transporting solutions responsible for the green color within red beds, or were the chemical and mineral environments within the green rock the causes for deposition of the ore minerals? Hence, a single question is immediately doubled or tripled.

Green spots may occur isolated within the interior of relatively impermeable thick mudstone layers indicating color differentiation probably during diagenesis, or early in the history of the rock, but in contrast to this relationship, elsewhere, green zones occur alongside joints and fault planes which clearly transect the primary stratification structures in red beds. Some of the fault and joint planes have been mineralized; others have not. Was the mineralization the cause or the effect of the secondarily developed green color, and what clay minerals are present across the color contact in these red-green rocks?

It may also be asked if the clay minerals, and possibly other minerals except the iron-oxide minerals, differ in red-green counterparts in rocks of different geologic ages, in those deposited under different sedimentational environments, in those composed of different clay mineral groups, in occurrences where the uranium-vanadium ore minerals are dark and low in oxidation state as against those higher in oxidation state, and in various other combinations. With the introduction of each new variable to the problem the number of questions increases geometrically to the point where it becomes impossible practically to collect red-green pairs for analysis in such a way as to eliminate all variables except color in the many combinations referred to above. Specimens were collected and analyzed, however, from the geologic features seen most commonly in field work and these features will be described.

FIELD AND LABORATORY WORK

Approximately 400 samples of mudstone and clayey sandstone were collected from localities on the Colorado Plateau during parts of the summers of 1953 and 1954. Where red and green counterparts were taken, they were always obtained from the same stratigraphic level and within a few inches across the color contact, unless otherwise stated. The clay minerals were identified mainly from X-ray powder diffraction patterns, but differential thermal analysis and the petrographic microscope were also used.

The clay fraction was extracted from sandstones by gently disaggregating in distilled water by rolling (not grinding) in a ball mill, and then recovering from sedimentation the particles having settling velocities equivalent to and less than 2-micron spheres according to Stokes' law _/. Some muds rich in gypsum or other soluble flocculating salts required repeated stirring with distilled water, settling, and decantation before the clay remained in a dispersed state, but no dispersing chemical, other than very dilute NH_4OH, was added.

During the first stages of the laboratory work all samples were dispersed and only minus 2-micron fractions analyzed, some in random orientation and others as oriented aggregates (clay films). The oriented-aggregate technique greatly enhances the X-ray reflections from the basal planes of platy minerals, making it possible to detect small amounts of platy clay minerals which might otherwise be missed, particularly mixed-layer types which characteristically give weak reflections.

_/ $x = \frac{2}{9} g r^2 \frac{d-d_1}{v}$; x = velocity of sphere when motion is steady; g = acceleration of gravity; r = radius of sphere; d = density of sphere; d_1 = density of fluid; and v - viscosity.

After comparing the diffraction patterns of those minus 2-micron fractions from mudstones with patterns from the bulk or entire mudstone, it was concluded that some mineral segregation and concentration occurred during sedimentation. Minerals heavier than the clay minerals, like iron oxides, iron sulfides, and some uranium-vanadium ore minerals, were removed to a significant degree from the minus 2-micron fraction of clay minerals owing to the more rapid settling of the heavier minerals; and, therefore, the minus 2-micron fraction was not always representative of the original rock.

Geologic conclusions based on mineral identifications of nonrepresentative fractions of a rock may be dangerously erroneous. It was necessary to choose in this study between sacrificing either some sensitivity in clay mineral identification or of fidelity in representation of the outcrop by the specimen. Because the first objective of the problem was to look for possible differences between the red and green counterparts of mudstones which might be utilized in geologic interpretation or as a guide to ore, it seemed of paramount importance that the sample which was analyzed must represent the rock.

Therefore, the decision was made to identify primarily the dominant clay mineral constituents in the whole mudstones, i. e., the bulk samples, even though it is possible that small amounts of some clay minerals detectible only by more refined methods, may have remained unreported. Many samples were fractionated, however, to check observations on the bulk samples. A truly refined clay mineral analysis requires determinations of ion exchange and perhaps even chemical analysis, which become so costly in terms of both time and money that the limit of diminishing returns is quickly reached in a geologic study where many clay samples are involved, as in this one.

Physical methods of clay mineral analysis, as were used, rarely permit recognition, at our present stage of knowledge, of the nature of cationic population in the octahedral, tetrahedral, and interlayers of the clay minerals. It is possible that chemical differences of this type occur between clay minerals, which would escape detection by the physical analytical techniques employed. This reservation must be made on the conclusions to be reached later in this report.

For the purposes of identification by X-ray, a 001 interplanar spacing of approximately 7A in the clay mineral was taken to indicate a member of the kaolin group. The reflections of kaolin-type minerals were regularly more characteristic of the "fire-clay type of kaolinite," one with layers stacked partially at random, than of well-crystallized kaolinite (Grim, 1953). The fire-clay type of kaolin is commonly developed in argillaceous sedimentary rocks whose source material is not previously well crystallized kaolinite.

Hydrous mica, which has also been called illite, is identified by an interplanar spacing of approximately 10A. This is commonly a broader (platykurtic) peak than the sharper one provided by clastic muscovite. Some diffraction patterns showed broad 10A bases which abruptly narrowed to a sharp peak, as if pierced by a sharp 10A shaft. Such patterns were interpreted as coming from both hydrous mica clay and clastic muscovite flakes. The presence of muscovite in mudstones showing this type of pattern was confirmed by microscopic examination.

Chloritic minerals were identified by a spacing of approximately 14A. Because chlorite peaks persisted regularly after heating several of these clays to 500° C. for 6 hours, which destroys the structure of kaolinite, it was assumed that the ancient sediments under study did not contain chlorite whose structure is destroyed at a lower temperature than that which wrecks kaolinite. This assumption may be incorrect because chlorite which was destroyed below 500° C. has been found in ore clays (Hathaway, personal communication, 1955) from the Colorado Plateau. Even if this assumption should prove to be incorrect, it does not invalidate the results obtained by comparing red and green counterparts, because the samples of both colors were treated the same.

Montmorillonite minerals were identified by swelling along th *c* axis to approximately 17A upon solvating with ethylene glycol.

These criteria for clay mineral identification, using the 001 spacings are in general use currently, although, they are vulnerable to criticism (Weaver, 1953). The 060 peaks of clay minerals also were regularly checked. X-ray equipment used was at the University of Missouri. A wide-range Geiger-Muller goniometer and nickel-filtered copper Kα c radiation were employed.

CLAY MINERALS IN REPRESENTATIVE SAMPLES OF MUDSTONE

Mineral analyses of representative mudstone samples have been organized in terms of stratigraphic sequence, productive (uranium-vanadium ore) and nonproductive localities, occurrence along faults, and a deposit where bleaching and alteration to alunite (the Oyler mine) has occurred. Many more mineral analyses were run than reported on herein, but these described represent the data which have lead to the final conclusions.

Clay minerals in red-green mudstone samples from selected stratigraphic formations

A record of representative red and green stratigraphically equivalent samples of mudstones from red-bed ore-bearing formations on the Colorado Plateau is given in table 1.

Many samples of which only 2 are recorded in table 1 were collected and analyzed from the Moenkopi formation, especially near its contact with the Shinarump (or other overlying sandstone) primarily at mining localities from Monument Valley, Ariz. , to the north end of the San Rafael Swell in Utah. Hydrous mica and muscovite characterize this zone in the Moenkopi, and kaolinite may, or may not, be present in it.

In figure 1 are shown X-ray diffractograms of stratigraphically equivalent green (No. 234) and red (No. 235) Moenkopi from a mineralized zone near the Oyler mine, Capitol Reef National Monument, Utah. Hydrous mica, muscovite, kaolinite, quartz, and probably calcite are represented in the diffractograms. No significant difference is found between the patterns of the clay minerals in the red and green counterparts.

The Shinarump formation exposed on Dille Butte in the San Rafael Swell, which is the source of samples Nos. 519 and 520 described herein, exhibits a purple-white mudstone zone which is probably representative of other purple-white occurrences, presumably parallels in color relationship the red-green pairs of red beds.

Table 1.--Clay minerals in mudstone samples from selected formations.

Sample No.	Formation or member and locality	Red	Green	Clay-size minerals
451 R	Rico formation, Hercules mine, Kane Springs district, Utah	X	-	Hydrous mica, muscovite quartz, calcite, hematite
451 G	Rico formation, Hercules mine, Kane Springs district, Utah	-	X	Hydrous mica, muscovite, quartz, calcite
270 R	Moenkopi formation, Capitol Reef Nat. Monument	X	-	Kaolinite, hydrous mica, dolomite
270 G	Moenkopi formation, Capitol Reef Nat. Monument	-	X	Kaolinite, hydrous mica, dolomite
519	Shinarump formation, Dille Butte, N. end San Rafael Swell	Purple	-	Kaolinite, hematite, quartz
520	Shinarump formation, Dille Butte, N. end San Rafael Swell	-	White	Kaolinite, quartz
273 R	Chinle formation, Pick's Delta mine, San Rafael Swell	X	-	Hydrous mica, muscovite, kaolinite, quartz, hematite
273 G	Chinle formation, Pick's Delta mine, San Rafael Swell	-	X	Hydrous mica, muscovite, kaolinite, quartz
15 R	Salt Wash member of Morrison formation, Uravan, Colo.	X	-	Hydrous mica, quartz
15 G	Salt Wash member of Morrison formation, Uravan, Colo.	-	X	Hydrous mica, quartz
	See table 3, for other Salt Wash samples			
5 R	Brushy Basin member of Morrison formation, Uravan, Colo.	X	-	Montmorillonite, hydrous mica (?)
6 G	Brushy Basin member of Morrison formation Uravan, Colo.	-	X	Montmorillonite, hydrous mica (?)

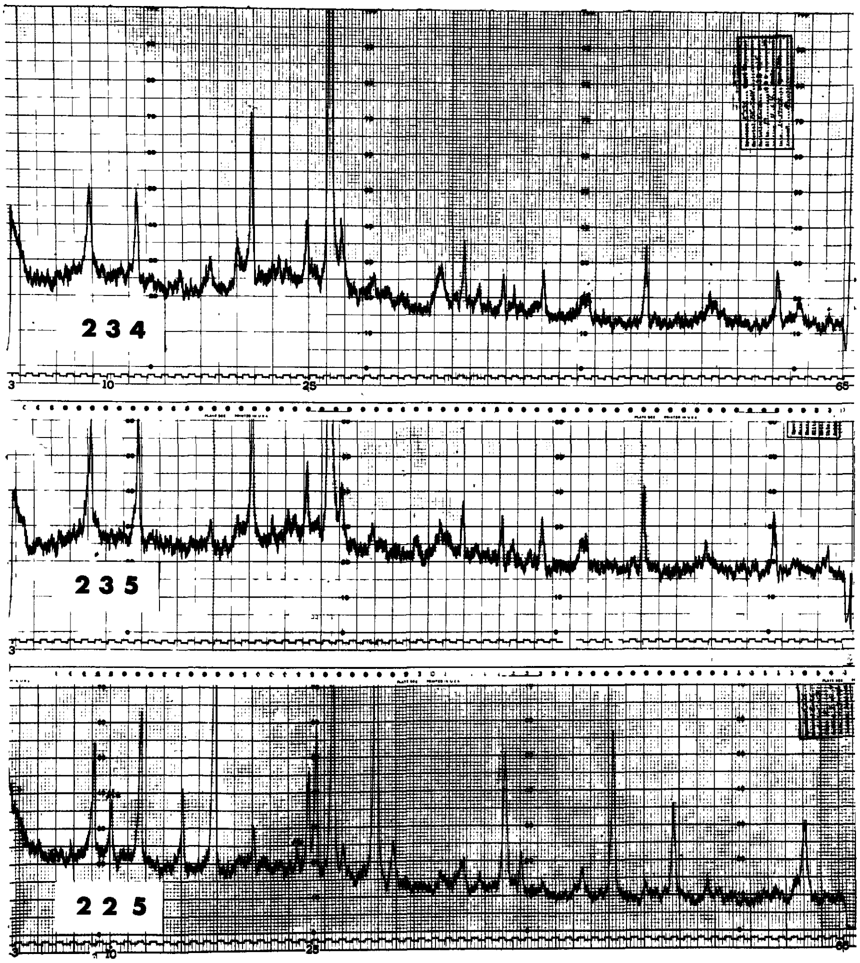

Figure 1. X-ray powder diffractograms of red mudstone (No. 235) from the Moenkopi formation collected about 600 feet west of the entrance to the Oyler mine in Capitol Reef National Monument, Utah, and a green counterpart (No. 234), stratigraphically equivalent 6 inches from No. 235. Diffractogram No. 225 is of an alunitized seam in bleached Moenkopi at what appears to be the same stratigraphic zone as that of Nos. 234 and 235.

The X-ray pattern (fig. 2) of the white Shinarump mudstone (No. 520) shows peaks of kaolinite and quartz while that of the purple counterpart (No. 519) shows hematite in addition to kaolinite and quartz. The hematite pattern of the purple sample is more nearly complete for hematite than are those of the hematite-red rocks of the red beds. The purple color (very dusky red purple 5RP2/2; Goddard, and others, 1948) is reminiscent of certain specular hematite, but when the rock is finely pulverized for analysis it becomes red (pale reddish brown, 10R5/4; Goddard, and others, 1948) that is typical of red-bed sedimentary hematite. Because the X-ray patterns of the purple and white counterparts of the Shinarump mudstone are practically superimposable, except for the additional hematite peaks in the purple sample, it is concluded that the clay mineral in the purple and white counterparts is the same.

The mineral composition of No. 520 was computed from its chemical composition (table 2) to be approximately, as follows:

Kaolinite	37 percent
Quartz	51
Hematite	10
TiO$_2$ etc. (Rutile, Anatase?)	2
	100

Extended studies with the microscope and differential thermal analysis of bulk and fine fractions of Nos. 519 and 520 support the clay mineral analysis as given above.

The Chinle samples contained hydrous mica and kaolinite, except that tuffaceous portions of the Chinle contained montmorillonite. The Brushy Basin member contains montmorillonitic zones (fig. 3), and zones in which hydrous mica is dominant. The diffractograms of the montmorillonite in the red (No. 5)- and green (No. 6) counterparts of the Brushy Basin samples are as nearly alike as are duplicate runs of the same sample. It is concluded, therefore, that the gross clay mineralogy of the two samples is the same.

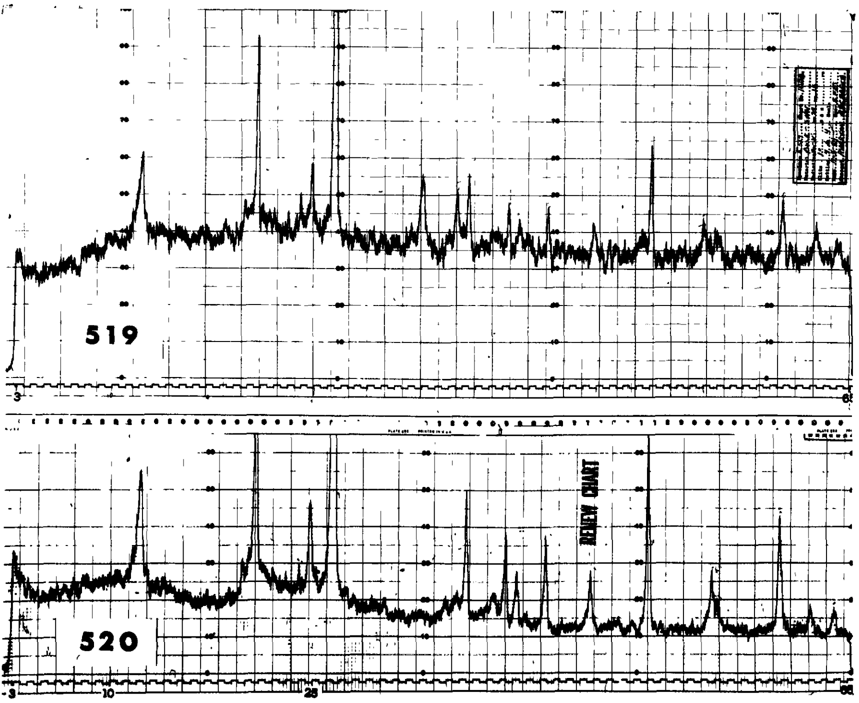

Figure 2. X-ray powder diffractograms of purple (No. 519) and white (No. 520) Shinarump from Dille Butte, north end of San Rafael Swell, Utah. Wide-range goniometer, Ni-filtered Cu-Kα radiation. Degrees 2θ are shown along base lines of diffractograms.

Table 2.--Chemical composition of purple (No. 519) and white (No. 520) silty mudstones from Dille Butte, north end of San Rafael Swell, Utah.

	Sample No. 519	Sample No. 520
SiO_2	68.03	78.75
Al_2O_3	14.31	13.42
Fe_2O_3	10.29	1.14
FeO	.10	.04
MgO	.04	.05
CaO	.06	.05
Na_2O	.06	.01
K_2O	.16	.15
H_2O-	.55	.44
H_2O+	5.09	4.62
TiO_2	1.06	1.03
CO_2	.02	.02
P_2O_5	.04	.03
MnO	.01	.00
	99.82	99.75

Analyst, Lois Trumbull, U. S. Geological Survey.

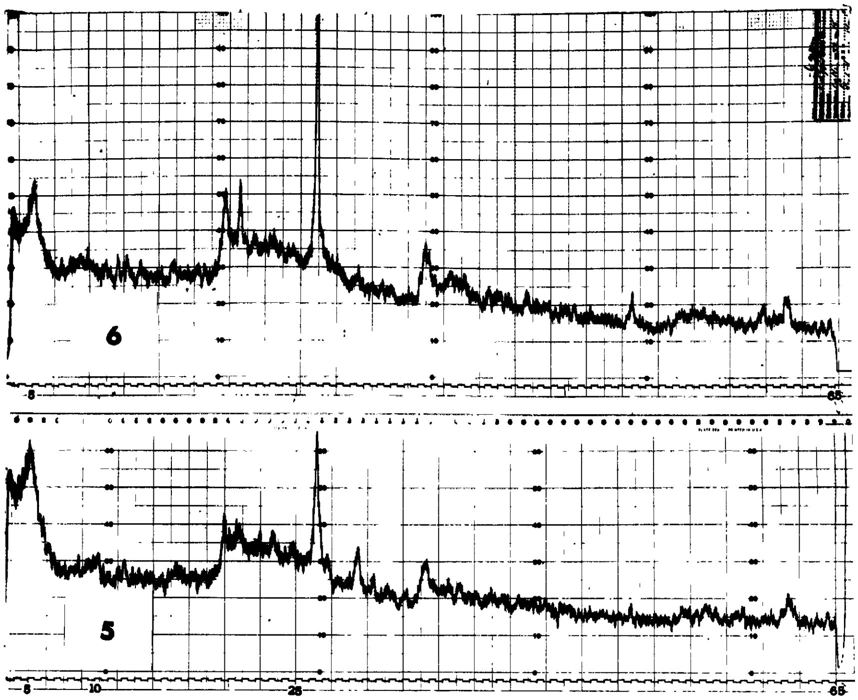

Figure 3. X-ray powder diffractograms of green (No. 6) and red (No. 5) stratigraphically equivalent mudstone from the Brushy Basin near Uravan, Colorado.

The data in table 1 may be summarized in the following conclusions. No significant difference was found between the clay minerals in stratigraphically equivalent red and green counterparts of mudstones collected from the Rico, Cutler, Moenkopi, Shinarump (purple and white), and Chinle formations, and the Salt Wash and Brushy Basin members of the Morrison formation, or within clay mineral groups including the kaolin group, the hydrous micas, the montmorillonites, and probably chlorite. The red color of mudstone red beds disappears with removal or absence of the ferric oxide; it need not, and ordinarily does not involve a change in the clay minerals.

Clay minerals in the Salt Wash member from productive and barren localities

Mudstone samples from the Salt Wash member of the Morrison formation were collected to study the following relationships of clay minerals to ore mineralization: mineralized mudstone, which was enriched with uranium-vanadium minerals of ore grade, taken from generally favorable districts (Club group, Uravan, Colo.; Lower group, Slick Rock, Colo.); nonmineralized mudstone from generally favorable districts (Uravan, Slick Rock, and Long Park, Colo.); mineralized mudstone from a less favorable to generally unfavorable district (Yellow Cat, Utah); and nonmineralized mudstone from a generally unfavorable district (Salt Wash member type section, Emery County, Utah). Red and green counterparts were collected from the nonproductive localities, but the mineralized mudstone was found in only green or gray colors. The results of the clay mineral analyses are shown in table 3.

It is concluded from this study that the clay minerals are not significantly different between the mineralized and nonmineralized mudstones, with one possible reservation. Mineralized mudstones are usually more likely to contain kaolinite and/or chlorite (?) in addition to hydrous mica, than are nonmineralized mudstone. It is doubted, however, that kaolinite per se, is responsible for deposition of ore minerals, because the same kaolinite-containing beds elsewhere along the strike may be nonmineralized. It appears, therefore, that kaolinite-containing mudstones represent a set of physical, chemical, or stratigraphic environments which are more likely to favor deposition of uranium-vanadium ore minerals than those in which hydrous mica alone, of the clay minerals was deposited.

Table 3. --Clay minerals in selected productive and nonproductive samples of the Salt Wash member of the Morrison formation.

Locality	Mineralized clay	Nonmineralized clay	Favorable district	Unfavorable district	Red	Green or gray	Clay minerals
Club group Uravan, Colo.	X	-	X	-	-	X	Hydrous mica, kaolinite
Club group, Uravan, Colo.	-	X	X	-	X	-	Hydrous mica
Club group, Uravan, Colo.	-	X	X	-	-	X	Hydrous mica
Lower group, Slick Rock, Colo.	X	-	X	-	-	X	Hydrous mica, kaolinite, chlorite
Lower group, Slick Rock, Colo.	-	X	X	-	-	X	Hydrous mica
Lower group, Slick Rock, Colo.	-	X	X	-	X	-	Hydrous mica
Long Park, Colo.	-	X	X	-	-	X	Hydrous mica
Long Park, Colo.	-	X	X	-	X	-	Hydrous mica, unidentified
Yellow Cat, Utah	X	-	-	X	-	X	Hydrous mica, chlorite (?)
Salt Wash member type section, Emery County, Utah	-	X	-	X	X	-	Hydrous mica
Salt Wash member type section, Emery County, Utah	-	X	-	X	-	X	Hydrous mica
Salt Wash member type section, Emery County, Utah	-	X	-	X	X	-	Hydrous mica
Salt Wash member type section, Emery County, Utah	-	X	-	X	-	X	Hydrous mica

No significant difference was noted between the clay minerals in green and red mudstone counterparts examined in the foregoing study. Hydrous mica is the common clay mineral in the Salt Wash member, as was found also by Weeks (1953).

Red-green mudstone along faults

Mudstone samples were collected from red-green counterparts along faults in the Kane Springs district, Utah, in the Cutler formation (Atomic King No. 2 mine), the Rico formation (Hercules mine), and the Moenkopi formation (supplied by Otis McRae); the faults are now mineralized by oxidized-type minerals. The green zone along the fault plane ranges from a few inches to 2 feet wide on each side. Clay mineral identifications are shown in table 4.

No significant differences were noted between the clay minerals in bleached mineralized zones along the fault planes, in their red counterparts, and in control samples collected nearby from barren rocks in the same stratigraphic beds as those containing ore minerals. From this lack of difference it is inferred that solutions which carried oxidized-type minerals did not necessarily bleach the red beds, and that they did not necessarily modify the gross clay mineralogy of the mudstones. It is possible, however, for solutions to react with and modify clay minerals in association with uranium-vanadium ore deposits.

Alunitization at the Oyler mine, Capitol Reef National Monument

A sequence of four samples of Moenkopi mudstone collected at the Oyler mine in Capitol Reef National Monument, Utah, illustrate the effect of sulfate reaction on the clay and carbonate minerals in the mudstone. (See table 5.) Red, barren Moenkopi (No. 235) occurring 600 feet west of the mine, contains hydrous mica, muscovite, kaolinite, quartz, calcite, and hematite. The same stratigraphic zone at the entrance to the mine is bleached of iron oxide but contains gypsum. A mineralized seam in the mine contains alunite and metatorbernite (fig. 1, diffractogram No. 225). The metatorbernite peak is marked "Me".

Table 4.--Clay minerals in mudstone samples from mineralized faults and nearby barren rocks in the Kane Springs district, Utah.

Sample No.	Locality	Red	Green or gray	Clay-size minerals
448 G	Hercules mine, fault in Rico formation mineralized	-	X	Hydrous mica, muscovite quartz, calcite, dolomite
449 R	Counterpart of 448 G	X	-	Hydrous mica, muscovite, quartz
451 G	Barren, control sample for 448 G	-	X	Hydrous mica, muscovite, quartz, calcite
451 R	Counterpart of 451 G	X	-	Hydrous mica, muscovite, quartz, calcite, hematite
447	Atomic King No. 2 mine, fault in Cutler formation, mineralized	-	X	Mixed layer hydrous mica and montmorillonite
446	Control sample for 447, collected 60 feet away	-	X	Mixed layer hydrous mica and montmorillonite
530-4	Altered, mineralized Moenkopi along fault	-	X	Hydrous mica, kaolinite quartz
530-1	Unaltered Monekopi, 1 foot from altered zone (530-4)	X	-	Hydrous mica, kaolinite quartz

It is inferred from this sequence that sulfate-bearing solutions, probably acidic, at the Oyler mine locality reacted with the ferric oxide in the red Moenkopi mudstone to bleach the pigment. Jarosite has been found at the mine and it is probable that the iron was moved, and partially removed, as iron sulfate. The sulfate reaction converted the calcite (and dolomite?) to gypsum. Further reaction of sulfate with aluminous clay minerals resulted in the development of alunite.

Table 5. Samples collected at the Oyler mine, Capitol Reef National Monument, Utah.

Sample No.	Description	Minerals present
225	White clay filling seams in bleached mudstone, Oyler mine entrance	Kaolinite, hydrous mica, quartz, alunite, metatorbernite
227	Bluish laminated mudstone, contains metatorbernite, Oyler mine entrance	Kaolinite, hydrous mica, quartz
229	Bleached Moenkopi, contains ore minerals, Oyler mine entrance	Kaolinite, hydrous mica, quartz, dolomite gypsum
235	Red Moenkopi, 600 feet west of Oyler mine entrance. Essentially equivalent stratigraphically to above samples, as nearly as can be observed	Hydrous mica, muscovite, kaolinite, quartz, calcite, hematite

In the presence of metatorbernite in the bleached portion of the Moenkopi and in the alunite-rich seams may be interpreted as follows: 1) that the sulfate solutions also carried in the metatorbernite, or alternatively, that 2) metatorbernite-bearing solutions followed the same avenues previously traversed by sulfate solutions. Spatial association of the minerals is permissive toward the first alternative, but the writer has not found compelling evidence that alunite and metatorbernite were genetically associated. It is exceedingly difficult to find unequivocal evidence of effects from the solutions which carried the ore elements apart from the ore minerals themselves.

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