

THE RELATION BETWEEN "ILLITE," BEIDELLITE, AND MONTMORILLONITE¹

By

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ABSTRACT

Formulas calculated from analyses, written in the fractional notation used by Ross and Hendricks, and with a notation of tetrahedral and octahedral charge, reveal relationships between the hydrous micas called "illite," montmorillonites (using the term in the restricted sense), and beidellites not otherwise apparent. These relationships are shown graphically by means of histograms made up of five columns, representing respectively, total charge, tetrahedral charge, octahedral charge, potassium, and water.

"Illites" have a much higher total charge and a much higher tetrahedral charge than montmorillonites, of which the tetrahedral charge is usually insignificant. Consequently simple removal of fixed K and its replacement by exchangeable cations would not convert an "illite" to a montmorillonite nor would mere fixation of K between the layers of a montmorillonite result in the formation of an "illite" like any that have been described.

From the standpoint of octahedral-tetrahedral charge relationships, beidellites and "illites" should be more similar than montmorillonites and "illites." However, considerable doubt has been cast by Grim and Rowland on the purity of most of the materials that have been called beidellites; they interpreted the differential thermal curves of the beidellites examined by them as indicating mixtures of kaolinite or halloysite, "illite," and montmorillonite. Formulas based on calculated mixtures of these minerals in varying proportions were very similar to formulas calculated from analyses of the so-called beidellites. Thus the existence of a montmorillonite clay like that which would be formed if the fixed K in "illite" were removed and replaced by exchangeable cations has not been authenticated and the concept that montmorillonite and "illite" are analogous except for the presence or absence of nonexchangeable potassium is misleading and is an oversimplification of the relationship between the two groups of minerals.

INTRODUCTION

This paper is concerned with the layer composition and charge relationships between two members of the montmorillonite group—montmorillonite and beidellite—and the clay materials that have been called "illites." Structurally the montmorillonites and the "illites" are similar, both having 2-1 layer structure, and differentiation between them is based largely on the identity of the interlayer cation. If potassium is the predominant interlayer cation and the lattice is consequently unexpanded, the material is classified as an "illite"; if sodium, calcium, or magnesium is

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the predominant interlayer cation, and the lattice is expanded, the clay is characterized as a montmorillonite. This relationship between the two groups has led to the concept that a montmorillonite should be the result if potassium is removed from "illite" and replaced by sodium, calcium, or magnesium and that "illite" should be the result if potassium is fixed between the layers of a montmorillonite. Thus White (1950) has postulated, "If sufficient of the potassium ions could be removed from the illite without marked decomposition of the mineral, it should be structurally equivalent to a member of the montmorillonite series (beidellite)," and Nagelschmidt and Hicks (1943) state, "The replacement of all exchangeable bases by potash in minerals of the montmorillonite group should lead to the formation of illite." That "illite" might be formed from montmorillonite in marine sediments by adsorption of potassium from sea water has been suggested by Dietz (1942). However, a comparative study of the structural formulas of montmorillonites, beidellites, and "illites" shows that this concept is an oversimplification of the relations between the two groups of minerals.

STRUCTURAL FORMULAS

Structural formulas calculated from analytical data may, of course, be in error for such fine-grained minerals as these because of the possible presence of admixed, intergrown, or adsorbed impurities. However, if certain criteria are used in judging the calculations of the formula, they afford a convenient means of comparing the composition of the tetrahedral and octahedral layers and the charge relationships of minerals like these that have the same structure.

Kelley (1945) has shown that silica as an impurity is not necessarily reflected by an excess of calculated Si ions in the tetrahedral group; it may be reflected by a deficiency of octahedral ions. In montmorillonites that have little or no Al in tetrahedral coordination, even a very small amount of free silica may be reflected in an excess of Si ions, but in montmorillonites, beidellites, or "illites" that have considerable tetrahedral aluminum, Si present as an impurity is reflected by a calculated deficiency of octahedral cations. In Table 1 are shown calculations of the formula for a bentonitic montmorillonite from Santa Rita, New Mexico. In the computation shown on the left, in which the analytical values as reported are used, the presence of excess silica is reflected both in an excess of tetrahedral ions and in a deficiency of octahedral ions, and the sum of the layer charges is greatly in excess of the sum of the interlayer cations.

This clay shows no visible crystalline impurities under the microscope, but x-ray diffraction shows that it contains cristobalite. Analytical determination (Foster, 1953) showed the sample to contain 4.5 percent of excess or free silica. In the second computation (Table 1) allowance has been made for this amount of free silica, and a small amount of aluminum had to be allocated to the tetrahedral group giving it a charge of 0.04 for

TABLE 1.—MONTMORILLONITE (BENTONITE) FROM SANTA RITA, NEW MEXICO¹

No allowance for free SiO ₂						Allowing for 4.5 percent free SiO ₂						
Percent	G-equiv. of cations	G-equiv. per total of 44	Atoms per unit cell	Charge		Percent	G-equiv. of cations	G-equiv. per total of 44	Atoms per unit cell	Charge		
SiO ₂	54.15	3.6064	32.575	8.144	+1.144	SiO ₂	51.99	3.4624	31.829	7.957	Al ₂ O ₃ 0.043	
Al ₂ O ₃	15.99	.9411	8.506	2.835	-1.514	Al ₂ O ₃	16.74	.9852	9.057	8.000	2.976	
Fe ₂ O ₃	.66	.0248	.224	.075		Fe ₂ O ₃	.69	.0259	.238			.079
MgO	4.01	.1989	1.797	.898		MgO	4.20	.2083	1.915			.958
				3.808							4.013	
				Σ 1.904							Σ 2.006	
CaO	2.14	.0763	.689	.689		CaO	2.24	.0799	.735		.735	
MgO	.47	.0233	.210	.210		MgO	.49	.0243	.223		.223	
Na ₂ O	.00	.0000	.000	.000		Na ₂ O	.00	.0000	.000		.000	
K ₂ O	.02	.0004	.004	.004		K ₂ O	.02	.0004	.004		.004	
Determined Cation Exchange		4.8712		.903	-1.370	Determined Cation Exchange		4.7864		.962	-1.962	
Determined Cation Exchange		.0990		.894		Determined Cation Exchange		.1040		.956		

¹Formula (Al_{1.49} Fe_{.04} Mg_{.48}) (Si_{3.93} Al_{.02}) O₁₀ (OH)₂ x .48

the unit cell. The number of ions in the octahedral group is very close to 4.00 for the unit cell, or to 2.00 for the half cell; and the sum of the layer charges, the sum of the external cations, and the determined cation exchange all check very closely.

Kelley also showed that the effects of silica as an impurity might be largely cancelled by the presence of any of several other impurities, whether admixed or intergrown since the effect of free alumina, iron oxide, or alkaline earth carbonates, or of interlayered chlorite, talc, brucite, or certain other minerals on the calculation is the reverse of the effect produced by silica as an impurity, tending to produce an excess of octahedral ions. The fact that the total of the calculated octahedral ions is close to 2.00 is no guarantee that the sample is pure. However, the likelihood that the effect of one impurity would be cancelled by another is very remote. It is also improbable in such an eventuality that the sum of the calculated layer charges would be in agreement with the sum of the interlayer cations.

Calculation of all the magnesium content as being in the structure may also result in an excessive number of octahedral ions, the magnitude of the excess depending on the amount of exchangeable magnesium present. It may also lead to a discrepancy between the sum of the layer charges and the sum of the interlayer cations. In the calculations of the formula of the Santa Rita sample, the determined amount of exchangeable magnesium was included among the interlayer cations, and only nonexchangeable magnesium was considered as being in the octahedral group. It is apparent that, if exchangeable magnesium had been included with nonexchangeable magnesium, as is usually done when the amount of exchangeable magnesium is not known, the number of ions in the octahedral layer would have been considerably in excess of 4.00 (about 4.12) and that would have been a deficiency of interlayer cations as compared with the sum of the layer charges and the exchange capacity.

In the present study the reliability of structural formulas calculated from analyses was, therefore, judged by these two criteria: (1) the number of octahedral ions per half cell must be 2.00 (± 0.02), and (2) the sum of the layer charges and the sum of the interlayer cations must be in close agreement (-0.04). If the calculated formula failed to satisfy either one of these conditions, it was considered suspect and discarded. However, if the number of octahedral positions was only slightly more than 2.00, and if there was a deficiency of interlayer cations such that an allocation of some of the magnesium to interlayer positions resulted in a formula that did satisfy the conditions, it was considered justifiable to make such an allocation in view of the very common tendency of clays having exchangeable properties to contain exchangeable magnesium.

For comparative purposes use was made of the fractional notations introduced by Ross and Hendricks (1945) to represent substitutions of Fe and Mg for Al and of Al for Si in the general formula

Al₂Si₄O₁₀(OH)₂. Thus, the proportion of ions in different positions is directly shown in the formula, and the formulas of different 2-1 layer minerals as well as different samples of the same mineral are directly comparable. As a further aid in comparison the charge on each layer is noted above the notation of the layer composition.

CHARACTERISTICS OF "ILLITES" AND MONTMORILLONITES AS REVEALED BY STRUCTURAL FORMULAS

"Illites"

All the formulas for "illites" (Table 2) were calculated from analyses published by Grim, Bray, and Bradley (1937). These analyses were of carefully purified material from slightly weathered tills, clays, and shales from Illinois that had been subjected to x-ray and microscopic study and to differential thermal analysis.

TABLE 2.—STRUCTURAL FORMULAS OF ILLITES

Locality	Formula		Sum of external cations
Ford Co., Ill.	(Al ⁻²³ _{1.19} Fe ⁻⁵⁸ _{.58} Mg ⁻²⁴ _{.24})	(Si ⁻⁶² _{3.38} Al ⁻⁶² _{.62}) O ₁₀ (OH) ₂ x .85	.85 .53 K
Calhoun Co., Ill.	(Al ⁻³² _{1.40} Fe ⁻³⁵ _{.35} Mg ⁻²⁶ _{.26})	(Si ⁻⁶⁰ _{3.40} Al ⁻⁶⁰ _{.60}) O ₁₀ (OH) ₂ x .92	.92 .60 K
Alexander Co., Ill.	(Al ⁻²⁴ _{1.55} Fe ⁻²⁰ _{.20} Mg ⁻²⁵ _{.25})	(Si ⁻⁴⁹ _{3.50} Al ⁻⁵⁰ _{.50}) O ₁₀ (OH) ₂ x .73	.73 .56 K
Vermilion Co., Ill.	(Al ⁻²⁵ _{1.53} Fe ⁻³² _{.32} Mg ⁻¹⁴ _{.14})	(Si ⁻⁵⁸ _{3.42} Al ⁻⁵⁸ _{.58}) O ₁₀ (OH) ₂ x .83	.83 .51 K
Goose Lake, Ill.	(Al ⁻²² _{1.52} Fe ⁻³⁰ _{.30} Mg ⁻¹⁹ _{.19})	(Si ⁻⁴⁶ _{3.54} Al ⁻⁴⁶ _{.46}) O ₁₀ (OH) ₂ x .68	.68 .50 K

These formulas show a fairly wide range in the amount of ferric iron in the octahedral group—the range being from 0.20 to 0.58 positions with a reciprocal range in octahedral aluminum such that the total number of trivalent ions in the octahedral group is fairly constant, ranging only from 1.66 to 1.76 positions. From this it follows that the number of divalent ions present in, and consequently the charge on, the octahedral group is also fairly constant, the charge ranging from 0.22 to 0.32.

The tetrahedral layers show considerable similarity in the amount of aluminum substituted for silicon—the range being from 0.46 to 0.62 positions, which also represents the tetrahedral charge. Thus, the tetrahedral charge is much greater than the octahedral charge; in most instances it is about twice as great.

As represented by these analyses, therefore, the "illites" are characterized by a fairly high total charge, from 0.70 to 0.95, of which approximately two-thirds is on the tetrahedral layers.

Montmorillonites

The formulas of montmorillonites (Table 3) — using the term in the restricted sense — were calculated from analyses of bentonites and pegmatitic clays that had been separated as completely as possible from associated minerals. Included in the analyses were data on exchangeable cations, total exchange capacity, soluble salts, and free silica or alumina.

TABLE 3. — STRUCTURAL FORMULAS OF MONTMORILLONITES

Locality	Formula	
Belle Fourche, S. D.	$\left(\begin{array}{ccc} \text{Al} & \text{Fe} & \text{Mg} \\ 1.61 & .18 & .23 \end{array} \right)$	$\left(\begin{array}{cc} \text{Si} & \text{Al} \\ 3.87 & .13 \end{array} \right) \text{O}_{10} (\text{OH})_2 \times .30$
Nieder-Bayern, Germany	$\left(\begin{array}{ccc} \text{Al} & \text{Fe} & \text{Mg} \\ 1.46 & .21 & .35 \end{array} \right)$	$\left(\begin{array}{cc} \text{Si} & \text{Al} \\ 3.93 & .07 \end{array} \right) \text{O}_{10} (\text{OH})_2 \times .36$
Irish Creek, Va.	$\left(\begin{array}{ccc} \text{Al} & \text{Fe} & \text{Mg} \\ 1.47 & .14 & .40 \end{array} \right)$	$\left(\begin{array}{cc} \text{Si} & \text{Al} \\ 3.94 & .06 \end{array} \right) \text{O}_{10} (\text{OH})_2 \times .44$
Fort Steel, Wyo.	$\left(\begin{array}{ccc} \text{Al} & \text{Fe} & \text{Mg} \\ 1.49 & .10 & .43 \end{array} \right)$	$\left(\begin{array}{cc} \text{Si} & \text{Al} \\ 3.98 & .02 \end{array} \right) \text{O}_{10} (\text{OH})_2 \times .39$
Amargosa Valley, Calif.	$\left(\begin{array}{ccc} \text{Al} & \text{Fe} & \text{Mg} \\ 1.37 & .15 & .50 \end{array} \right)$	$\left(\begin{array}{cc} \text{Si} & \text{Al} \\ 3.97 & .03 \end{array} \right) \text{O}_{10} (\text{OH})_2 \times .48$

The most striking difference between these formulas of montmorillonites and those of "illites" is the very low degree of substitution of aluminum for silicon in the tetrahedral group of the montmorillonites and the much lower charge on the montmorillonite structure.

The total charge on the montmorillonites ranges from about 0.30 to 0.50, as compared with a charge ranging from 0.70 to 0.95 on the "illites." Thus, the highest charge found on the montmorillonites is considerably lower than the lowest charge found on the "illites." In the montmorillonites, there is little substitution of aluminum for silicon in the tetrahedral layers. The seat of the charge is, therefore, generally predominantly octahedral, as opposed to the predominantly tetrahedral charge of the "illites."

The octahedral layers of the montmorillonites tend to show a lower ferric iron and a higher magnesium content than those of the "illites," with the result that, although the total charge on the montmorillonites is much lower than that of the "illites," the octahedral charge is greater.

These relationships between the "illites" and the montmorillonites are brought out graphically in histograms of structural formulas (Fig. 1). In these histograms the first column represents the total charge, the second represents the tetrahedral charge, and the third represents the octahedral charge. The fourth column represents fixed and exchangeable cations and the fifth represents water. The water is the only value represented in terms of percent. All other values are in terms of units of charge per half cell.

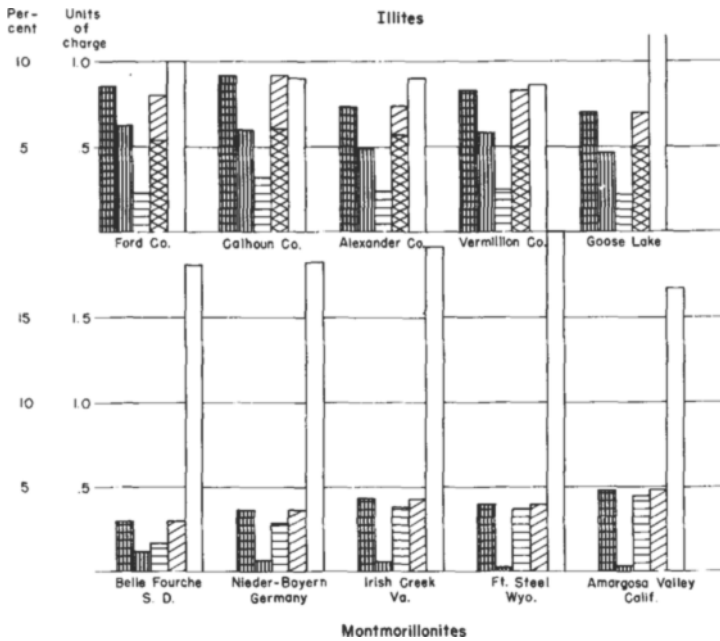


FIGURE 1. — Histograms of "illites" and montmorillonites.

These histograms bring out clearly the significant difference between the total amount of charge on the "illites" and that on the montmorillonites, and the equally significant difference in the location of the seat of the charge. In all the "illites" the top of the column representing total charge is well above the 0.5 line; in the montmorillonites it is below that line. In the "illites" the column representing tetrahedral charge is about twice as tall as the one representing octahedral charge. In the montmorillonites only one sample has a significant amount of tetrahedral charge; in all other samples the octahedral charge is greatly predominant. In the "illites" two-thirds or more of the charge is fixed by potassium and the area occupied by exchangeable bases is commonly smaller than the same area in the montmorillonites, although the highest amount of exchangeable bases

The calculated formulas indicate a fairly consistent type of material—whether it be a mixture or a single mineral—characterized by a total charge commonly lower than that found on the “illites” and higher than that on the montmorillonites. The charge is predominantly tetrahedral as on the “illites,” but the octahedral charge is lower than that on the “illites,” which in turn is lower than that on the montmorillonites. Hence the lower total charge cannot be attributed to a simple mixture of “illite” and montmorillonite, which would tend to produce a material having an octahedral charge of between about 0.25 and 0.42, whereas none of the so-called beidellites has an octahedral charge greater than 0.25. The K content of some of the so-called beidellites suggests the presence of some “illite,” but the highest K in any of the beidellites (?) is less than half the K usually found in the “illites.”

CALCULATED MIXTURES OF “ILLITE,” KAOLINITE, AND MONTMORILLONITE

Grim and Rowland (1942) interpreted the differential thermal curves of the beidellites examined by them as indicating mixtures of kaolinite or halloysite, “illite,” and montmorillonite. In order to determine whether such mixtures would have formulas similar to those calculated for the beidellites, mixtures in varying proportions of a kaolinite (Zettlitz, Czechoslovakia), an “illite” (Fithian, Ill.), and a montmorillonite (Nieder-Bayern, Germany) were calculated. The formulas derived from these mixtures are shown in Table 5.

TABLE 5.—FORMULAS FOR CALCULATED MIXTURES OF KAOLINITE,
ILLITE, AND MONTMORILLONITE

Mixture	Formula			
3/5 kaolinite 1/5 illite 1/5 montmorillonite	(Al	Fe	$\frac{+.04}{Mg}$) (Si $\frac{-.63}{Al}$) O (OH) ₂ x .59 <u>1.89 .13 .00</u> 3.37 .63 10 .16 K
1/3 kaolinite 1/3 illite 1/3 montmorillonite	(Al	Fe	$\frac{-.10}{Mg}$) (Si $\frac{-.56}{Al}$) O (OH) ₂ x .66 <u>1.71 .20 .10</u> 3.44 .56 10 .22 K
2/5 kaolinite 2/5 illite 1/5 montmorillonite	(Al	Fe	$\frac{-.08}{Mg}$) (Si $\frac{-.61}{Al}$) O (OH) ₂ x .69 <u>1.77 .19 .04</u> 3.39 .61 10 24. K
2/5 kaolinite 1/5 illite 2/5 montmorillonite	(Al	Fe	$\frac{-.09}{Mg}$) (Si $\frac{-.48}{Al}$) O (OH) ₂ x .57 <u>1.75 .16 .10</u> 3.52 .48 10 .14 K
2/5 illite 3/5 montmorillonite	(Al	Fe	$\frac{-.30}{Mg}$) (Si $\frac{-.27}{Al}$) O (OH) ₂ x .57 <u>1.47 .26 .28</u> 3.73 .27 10 .22 K
2/5 kaolinite 3/5 montmorillonite	(Al	Fe	$\frac{-.13}{Mg}$) (Si $\frac{-.39}{Al}$) O (OH) ₂ x .52 <u>1.74 .14 .12</u> 3.61 .39 10 .04 K

Two of the mixtures that were made up one-half or more of kaolinite produced a formula that had more than 2.00 positions in the octahedral group, a plus charge on the octahedral group, and a lack of agreement between the sum of the layer charges and the sum of the external cations, the sum of the external cations being considerably less than the sum of layer charges. These are also the characteristic features of the formula derived from the analysis of the so-called beidellite from Namiquipa, Chihuahua, Mexico, which Grim and Rowland (1942) interpret as being principally kaolinite or halloysite with perhaps a small amount of montmorillonite, although the K content of this sample suggests the presence of a little "illite." The amount of K reported in the analysis, 1.18 percent, suggests that the material was about one-fifth "illite."

The other mixtures produced formulas that were similar to those of the beidellites with respect to the charges on the octahedral and tetrahedral layers, the trivalent-bivalent ion composition of the octahedral layer, the aluminum content of the tetrahedral layers, and the K content. This similarity is shown graphically in the histograms (Fig. 2) of formulas derived from analyses of materials that have been called beidellites and from the calculated mixtures. Included for comparison are histograms of formulas of "illites."

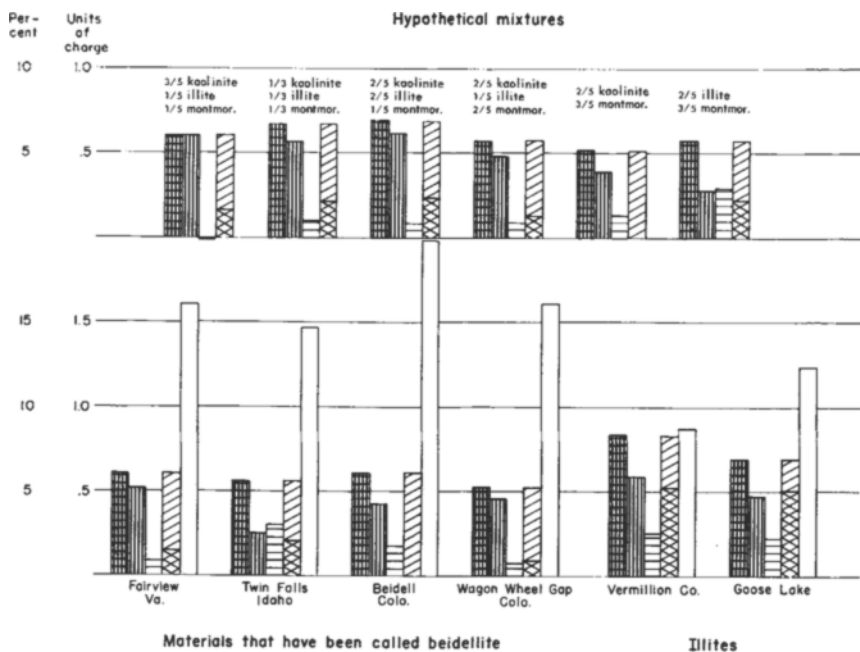


FIGURE 2. — Histograms of materials that have been called beidellite and "illite"; and hypothetical mixtures.

The mixture made up of two-fifths kaolinite, one-fifth "illite," and two-fifths montmorillonite is very similar in the amount of total charge and in tetrahedral-octahedral charge relationship to the materials from Fairview, Utah, and Wagon Wheel Gap, Colo. The K content and the relation between K and exchangeable cations are also comparable. The mixture made up of two-fifths "illite" and three-fifths montmorillonite is like the material from Twin Falls, Idaho, in these characteristics, and the material from Beidell, Colo., is very similar to a two-to-three mixture of kaolinite and montmorillonite. Grim and Rowland (1942) concluded, from the differential thermal curve of the crude material, that the Beidell, Colo., material is primarily a mixture of halloysite, "illite," and montmorillonite; but the presence of only a trace of K in the material indicates the absence of "illite." A differential thermal curve of hand-picked, apparently homogeneous material was the same as that of the crude material except for the size of the peak between 600°C and 700°C, and an x-ray analysis of the hand-picked material showed more pronounced montmorillonite diffractions than the crude sample.

Larsen and Wherry (1925) proposed the name beidellite for the material from Beidell, Colo., that had been previously described by them as leverrierite, and Ross and Shannon (1925) described the properties of beidellite. Ross and Hendricks (1945) included beidellite as a high-alumina member of the montmorillonite group on the basis of analyses that appeared to be similar to those of montmorillonite but with a higher alumina content. However, the fact that all the materials called beidellite, that have been studied carefully by x-rays and differential thermal analysis, seem to be mixtures of kaolinite or halloysite, "illite," and/or montmorillonite, casts considerable doubt on the validity of beidellite as a mineral species. Ross, himself, has been of the opinion for some time that the name beidellite should be discarded and that the name "alumian montmorillonite" be used for a montmorillonite having considerable aluminum in the tetrahedral group.

CONCLUSION

Beidellite, of all the montmorillonites, has a hypothetical composition most like the composition of the clay that would be formed if the K in "illite" were removed and replaced by exchangeable cations like sodium or calcium, but the existence of a natural montmorillonite clay of this type has not been authenticated. Conversion of "illite" to montmorillonite (using the term in the restricted sense), or vice versa, would require a much more deep-seated alteration than simple removal of potassium from "illite" and its replacement by sodium, calcium, or magnesium, or the fixation of potassium between the layers of a montmorillonite. Consequently, the concept that montmorillonite and "illite" are analogous except for the presence or absence of nonexchangeable potassium is misleading and an oversimplification of the relationship between these two groups of minerals.

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DISCUSSION

R. E. Grim. — Perhaps before the name "beidellite" is accepted we must find a natural mineral (single species and not mixed layer) that has the assigned beidellite formula.

C. E. Marshall. — I am strongly in favor of retaining the name beidellite. If it were not available, we should now find ourselves searching for a name to describe the end member of the montmorillonite group having an expanding lattice and a predominant charge on the silica layers. The fact that impurities were found in the type material is, in clay mineralogy, not surprising.

Rustum Roy. — I would agree with Dr. Marshall that even though the original material classified as beidellite may have been shown to be a mixture of various phases, other minerals do exist in nature which fall within the description usually associated with the term beidellite. And should this name be abandoned another will have to be found: for even if they do not occur in nature we can synthesize quite typical beidellites of known and controllable compositions. But here again the question must be raised as to whether or not these synthetic materials deserve mineral names. In this case, however, where it appears certain that the synthetic material would represent merely a pure end member of a particular species, it does emphasize the necessity for a name for the species.

Margaret D. Foster. — I agree that the name "beidellite" should be retained for the end member of the montmorillonite group having a predominant charge on the silica layers, even though no natural material having such a formula has, as yet, been described.