STRUCTURE, COMPOSITION AND GENESIS OF SOME LONG-SPACING, MICA-LIKE MINERALS[1]

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Abstract

Long-spacing, mica-like minerals from Goto Island, Japan, are shown to resemble allevardite in structure, swelling and shrinking behavior, and chemical composition. The structure consists of a nearly regular alternation of swelling and non-swelling layers, the latter containing principally K and Ca as interlayer ions and the former Na and probably $H(H_2O)$, *i.e.*, hydronium ions. Comparison is made with the work of Brown and Norrish on hydrous micas. Two hypotheses are considered for the formation of long-spacing minerals of this type, namely (1) an unmixing of interlayer cations during a process of mild leaching and hydration, (2) Sudo's hypothesis which relates the alternating character to features previously existing in the parental mica.

Introduction

The present paper is concerned with the structure, composition, and genetic significance of a long-spacing, mica-like mineral from Goto Island, Japan, which exhibits reversible swelling and shrinking properties. The materials studied were made available by Mr. Shohei Niwa of the Toshiba Refractories Co., Kariya, Aichi-ken, Japan and came from the East and West Valleys of the island. The general geology of the region has been described by Iwao *et al.* (1953). Similar materials from Goto Mine and from other localities have been described by Takeshi (1958), by Shimoda and Sudo (1960), and by Sudo *et al.* (1962); a sample of material from Goto Mine was made available to the writers by Dr. Sudo. As Sudo *et al.* (1962) have pointed out, these mica-like materials represent micas in various stages of alteration and therefore they show many minor variations within short distances. The materials studied by the writers are sufficiently similar to those discussed by Sudo *et al.* (1962) that the experimental results can be presented very briefly and attention given mainly to questions of interpretation.

Other minerals of a similar kind include rectorite studied by Bradley (1950), and allevardite studied by Caillère, Mathieu-Sicaud and Hénin (1950) and by Brindley (1956). In many respects the long-spacing minerals from Goto Island are similar to allevardite and with the information available it seems unnecessary at present to suggest a new name.

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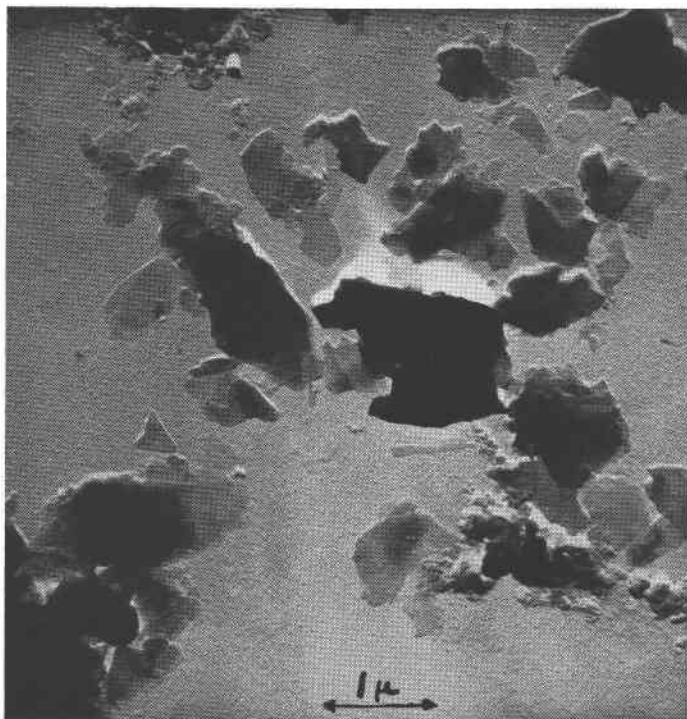[2] On leave of absence from the Physics Department, University of Athens, Greece.

FIG. 1. Electron micrograph of material from East Valley, Goto Island, Japan, containing mica and a long-spacing layer-lattice mineral.

EXPERIMENTAL RESULTS

Hand specimen appearance and morphology. Hand specimens are hard and brittle, almost pure white in color and with a slightly greasy feel. Electron micrographs of samples prepared by gentle crushing under water show irregular shaped, platy particles about 1 micron in size with no obvious distinction between the long-spacing mineral and a mica impurity (Fig. 1). Attempts at separating the two components by simple sedimentation were not successful.

Mineral associations. Of the materials available for study, the sample from the West Valley of Goto Island contained appreciable pyrophyllite and mica in addition to the long-spacing mineral, the sample from the East Valley contained only mica as a major impurity. The sample from Goto Mine, supplied by Dr. Sudo, contained pyrophyllite as a major impurity.

The present work has been concerned mainly with the East Valley sample.

Chemical analysis. A chemical analysis of a 10 g sample of the East Valley material is reported in Table 1. Because the material contained considerable mica, little can be said immediately of this analysis, beyond a statement that the mineral of interest is an alumino-silicate, probably hydrated, and containing an uncertain number of Na, K and Ca ions.

X-ray data; lattice spacings. Results obtained using thin, oriented aggregates under various experimental conditions are summarized in Table 2 together with corresponding data by Sudo and his collaborators (1962). The thermal stability and swelling characteristics observed by Sudo and in the present study are very similar. The "infinite" swelling in excess water observed by the present writers may indicate a minor difference between the East Valley material and that studied by Sudo *et al.*

TABLE 1. CHEMICAL ANALYSES OF A ROCK SAMPLE FROM EAST VALLEY, GOTO ISLAND, JAPAN, CONTAINING A LONG-SPACING, LAYER-LATTICE MINERAL, AND OF TWO RELATED MINERALS

	(1) Rock sample from Goto Island	(2) Mineral discussed by Sudo *et al.* (1962)	(3) Allevardite
SiO_2	42.11	44.80	45.20
Al_2O_3	40.96	33.88	32.15
TiO_2	0.26	—	0.30
Fe_2O_3	0.17	—	0.65
FeO	—	0.39	0.55
CaO	3.58	0.97	1.00
MgO	0.25	1.24	0.35
K_2O	3.98	1.13	1.10
Na_2O	0.88	1.88	2.25
BaO	0.25	—	—
P_2O_5	—	none	0.15
$H_2O(+110°\ C.)$	5.29	6.91	6.04
H_2O^-	2.07	8.13	9.72
Total	99.80	99.33	99.46

(1) Analysis by C. O. Ingamells, College of Mineral Industries, The Pennsylvania State University.

(2) T. Sudo *et al.* (1962).

(3) S. Caillère *et al.* (1950).

TABLE 2. X-RAY DATA FOR LONG-SPACING, MICA-LIKE MINERALS FROM GOTO
ISLAND, JAPAN

Condition of measurement	Basal spacings, d(001), in Å	
	Present work	Sudo *et al.* (1962)
With excess water	∞	28.8
Saturated with ethylene glycol	26.5±0.1	26.5
Sample stored over water	≈23.0±0.5	25.3
Open air conditions; no humidity control	≈22.5±0.5	
430° C. for 1 hr.; examined in dry air flow	19.4±0.05	19.6
700° C. for 1 day; dry air flow	19.4±0.05	19.6
Stored over water for 3 days after 700° C. heat treatment	22.7±0.3	rehydration occurs
900° C. for 3 hr.; dry air flow	19.5±0.05	19.6
Stored over water after 900° C. heat-treatment	19.5±0.05	
960° C. for 1.5 hr.	(mullite reflections)	decomposed at 1000° C.

Differential thermal analysis. The results of two runs taken with a tem-
perature rise of 8° C./min are given in Fig. 2. The dashed line (a) corre-
ponds to a finely ground sample in equilibrium with normal atmosphere
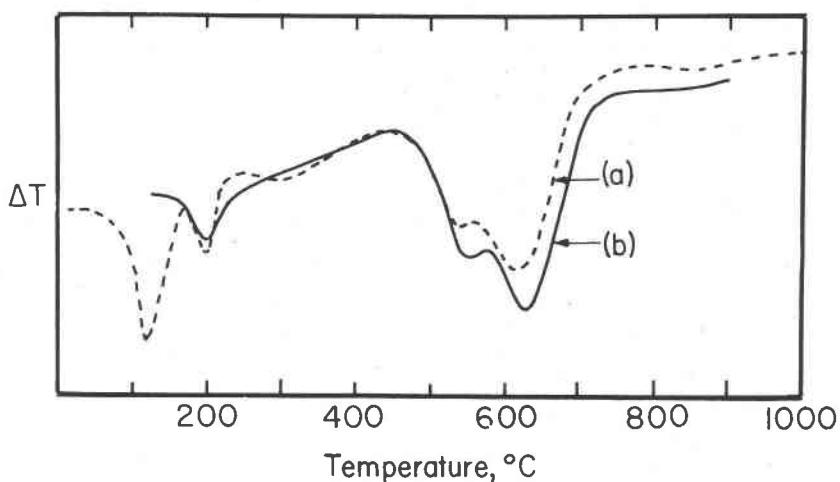conditions, with relative humidity ∼70%. The full line (b) is for a

FIG. 2. Differential thermal analyses of material containing a long-spacing, mica-like
mineral from Goto Island, Japan. (a) Material stored under atmospheric conditions with-
out special drying. (b) Material heated to constant weight at 110° C.

sample brought to constant weight at 110° C. and then held for about 1 hr in the DTA furnace at this temperature prior to the run.

There are four clearly defined endotherms with peak temperatures of 120° C., 200–205° C., 540–550° C. and 620–630° C.

The water loss associated with the first endotherm was determined by taking a similar sample stored under atmospheric conditions and heating it to constant weight at 110° C. A weight loss of 3.50% was obtained which is larger than the value 2.07% given for H_2O- Table 1. The difference arises almost certainly from the higher summer-time humidity when the DTA. and weight-loss measurements were made than the winter-time humidity when the chemical analysis was made.

The second endotherm at 200–205° C. corresponds to a much smaller weight loss determined by taking material dried to constant weight at 100–110° C., and then heating for 16 hr at 300° C.; this weight loss amounted to 0.50%. The ratio of the areas of the low-temperature peaks of curve (a), Fig. 2, is 4.6/1, whereas the ratio of the weight losses (*i.e.*, essentially water losses) is 3.5/0.5 or about 7/1.

THE LAYER SEQUENCE IN THE SWELLING MINERAL

The swelling characteristics and the thermal analysis data suggest that part of the structure consists of hydrated montmorillonite-like layers. Thus ethylene glycol expands the structure by $26.5 - 19.4 = 7.1$ Å. Under open-air conditions, the expansion is $(22.5 \pm 0.5) - 19.4 = 3.1 \pm 0.5$ Å which is comparable with that of a Na-montmorillonite. The large dispersion in excess water also agrees with the behavior of Na-montmorillonite.

By taking differences between the spacings of the swelling mineral and those of Na-montmorillonite, the spacing of the non-expanding component is obtained. For the ethylene glycol complexes, the difference is $26.5 - 16.85 = 9.65$ Å. For the fully collapsed state, the difference is $19.4 - 9.6 = 9.8$ Å. The spacing of the non-expanding part of the structure is therefore about 9.7 ± 0.1 Å, which comes much nearer to the basal spacings of micas $(9.8 - 10.0$ Å) than of pyrophyllite (9.2 Å).

The swelling mineral thus appears to be a regular or nearly regular alternation of non-expanding mica-like layers and expanding montmorillonite-like layers.

INTERPRETATION OF THE CHEMICAL ANALYSIS

Because of the large mica impurity, care is needed in the interpretation of the overall chemical analysis. However, the fact that *all* the material is mica-like in a broad sense makes an interpretation possible. The K_2O and CaO contents can be associated respectively with musco-

vite and margarite compositions, namely $KAl_2(Si_3Al)O_{10}(OH)_2$ and $CaAl_2(Si_2Al_2)O_{10}(OH)_2$. The remaining composition associated with Na_2O will probably represent the swelling part of the structure.

The treatment of the water content is important. From the H_2O $(+110°)$, 5.29%, given in Table 1, there has been subtracted H_2O $(+110°, -300°$ C.), 0.50%, which can be taken as water of hydration and not lattice "hydroxyl water." The allowance for this type of water, though a small correction, is quite important in the analysis of the data.

The main steps in the calculation are given in Table 3 and are nearly self-evident. The analysis in weight percent in column (2) is converted to relative moles in column (3) and relative ions in column (4). Columns

TABLE 3. CALCULATIONS RELATING TO THE COMPOSITION OF THE GOTO ISLAND MATERIAL

(1)	(2) Weight per cent	(3) Moles (relative)	(4) Ions (relative)		(5) 0.84 Mu[1]	(6) 0.64 Ma[2]	(7) (4)-(5)- (6)	(8) $(7) \times \dfrac{12.0}{12.27}$
SiO_2	42.11	7.01	Si	7.01	2.52	1.28	3.21	3.14
Al_2O_3	40.96	4.01	Al	8.03	2.52	2.56	2.95	2.88
Fe_2O_3	0.17	0.01	Fe^{3+}	0.02	—	—	0.02	0.02
CaO	3.58	0.64	Ca	0.64	—	0.64	—	—
MgO	0.25	0.06	Mg	0.06	—	—	0.06	0.06
K_2O	3.98	0.42	K	0.84	0.84	—	—	—
Na_2O	0.88	0.14	Na	0.28	—	—	0.28	0.27
BaO	0.25	0.02	Ba	0.02	—	—	0.02	0.02
$H_2O(+300)$	4.79	2.66	OH	5.32	1.68	1.28	2.36	2.31
$H_2O(+110, -300)$	0.50	0.28	H_2O	0.28	—	—	0.28	0.27
			O	24.71	8.40	6.40	9.91	9.69

[1] Mu = muscovite composition, $KAl_2(Si_3Al)O_{10}(OH)_2$.

[2] Ma = margarite composition, $CaAl_2(Si_2Al_2)O_{10}(OH)_2$.

(5) and (6) give respectively $0.84 \times$ (muscovite formula) and $0.64 \times$ (margarite formula). Then $(5)+(6)$ is subtracted from column (4) to give the remaining ions which are listed in column (7). The corresponding formula is obtained by scaling the $(O+OH)$ ions to 12.00, *i.e.*, by multiplying column (7) by 12.0/12.27 and these results are listed in column (8). The figures in column (7) are therefore $1.02 \times$ (swelling layer composition).

The results of these calculations will be discussed from several standpoints.

The swelling layer formula. The ionic distribution in column (8) of Table 3, obtained by scaling $(O+OH)$ ions to 12.00, gives a total for $(Si+Al +Fe)$ ions $= 6.04$, which is almost exactly the ideal value of 6.0 for a dioctahedral mica-type layer.

The layer formula can be written:

$$(Na_{0.27}Mg_{0.06})(Al_{2.02}Fe_{0.02})(Si_{3.14}Al_{0.86})O_{9.69}(OH)_{2.31}(H_2O)_{0.27} \qquad (1)$$
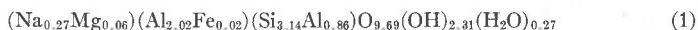
As in all calculations of this type, the result depends to some extent on the basis of normalization, in this case (O+OH) ions = 12, but since the same result is achieved on the basis of (total layer cations) = 6, the formula can be accepted as being reasonably reliable.

Formula (1) is not a montmorillonite composition nor even a mont-morillonite-type composition. In beidellites, where the layer charge arises from tetrahedral substitution of Al for Si, the amount of substitution is of the order of $(Si_{3.67}Al_{0.33})$. Formula (1) is essentially that of a mica with tetrahedral cations approximately (Si_3Al).

One sees, therefore, that the entire composition is that of a mica with some layers which show swelling and shrinking behavior, and others which are non-swelling. The artifice of associating the K_2O and CaO with muscovite and margarite formulae is now seen to be quite justified. Some interchange of the ions Na, K, and Ca could be made in the layer formulae without affecting the essential character of the final result, namely that some of the mica layers must have a marked deficiency in interlayer cations and it is reasonable to associate these layers with the swelling characteristics.

It is important next to consider the charge balance in formula (1). The cations in octahedral and tetrahedral positions have a net charge deficiency of 0.74 units, which is balanced partly by the interlayer ions (0.27 Na+0.06 Mg) providing a charge of 0.39 and partly by the additional 0.31 H ions, that is to say, additional over the ideal layer composition $O_{10}(OH)_2$.

It is a basic question where the additional H ions are accommodated, since the mica layer structure provides positions for only 2(OH). In the first place, the result may arise from the way in which the various kinds of water in the structure are assessed. If we neglect to make any allowance for H_2O (+110, −300) and treat this as "hydroxyl water," then the amount of (OH) in the lattice is increased. In order to arrive exactly at $O_{10}(OH)_2$ in formula (1), it would be necessary *almost to double* the correction for H_2O(+110, −300) and there is no experimental evidence pointing in this direction. At the same time, it must be recalled that it is not uncommon to find analyses yielding $O_{<10}(OH)_{>2}$, so that the present problem is not in the least uncommon.

When the O, OH composition given in formula (1) is written $O_{10}(OH)_2H_{0.31}$, one sees that the basic problem is to find a location for the additional 0.31 H ions. These must be placed *either* in the tetrahedral layers of the sheet structure, *or* in interlayer positions. The near equal-
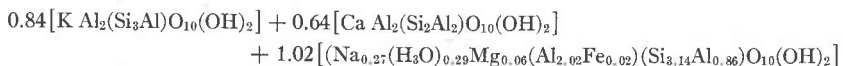
ity of 0.31 H ions and 0.27 H_2O molecules associated with the second endotherm suggests that the additional H ions exist as hydronium ions, $H(H_2O)^+$ or $(H_3O)^+$ ions. This was the conclusion reached by Brown and Norrish (1952) in their interpretation of the chemical analyses of some very carefully analyzed illites, but it should be observed that they had less information on which to base their conclusions in that the percentage of $H_2O(+105)$ was not broken down into two groups as in the present study, and d.t.a. and weight-loss data were not available. However, their conclusions seemed reasonable in that the interlayer cations came very close to unity on the basis $O_{10}(OH)_2$ when hydronium ions were taken into account.

If these conclusions are accepted, then formula (1) can be written:

$$(Na_{0.27}Mg_{0.06}(H_3O)_{0.29})(Al_{2.02}Fe^{3+}_{0.02})(Si_{3.14}Al_{0.86})O_{10}(OH)_2 \qquad (2)$$
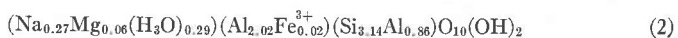
The interlayer cations now number 0.62 with a total charge of $+0.68$. The layer can be described as a *sodium-hydronium mica*.

The overall composition. The overall composition of the material under study, corresponding to columns (5), (6) and (7) of Table 3, is as follows:-

$$0.84[K\,Al_2(Si_3Al)O_{10}(OH)_2] + 0.64[Ca\,Al_2(Si_2Al_2)O_{10}(OH)_2]$$
$$+ 1.02[(Na_{0.27}(H_3O)_{0.29})Mg_{0.06}(Al_{2.02}Fe_{0.02})(Si_{3.14}Al_{0.86})O_{10}(OH)_2]$$

Exactly how these compositional units are distributed between the mica impurity and the long-spacing swelling mineral, and between the swelling and non-swelling components of the latter, is indeterminate. The following is a reasonable interpretation. Of the total 1.48 (K-mica +Ca-mica) units, 1.02 units can be taken to represent the non-swelling components of the long-spacing mineral and 0.46 (K-mica+Ca-mica) units the mica impurity.

The whole composition is then 0.46 units of K, Ca-mica and 1.02 units of the long spacing mineral comprising a 1:1 alternation of K, Ca non-swelling layers and Na, H swelling layers. On this basis the proportion of mica impurity is $0.46/(0.46+2.04)=18\%$. It is difficult to get an exact check on this figure from the x-ray data but rough estimates are of the order of 20–30%.

Comparison with allevardite. As pointed out by Sudo *et al.* (1962), the long-spacing mica-like minerals from Goto Island show very similar swelling and shrinking behavior to the mineral allevardite, of which a detailed x-ray study was made by Brindley (1956). It was shown that the alumino-silicate layers are mica-like in structure and composition with $(Si_{3.20}Al_{0.80})$ in tetrahedral positions and $(Al_{1.89}Fe_{0.07}Mg_{0.04})$ in

octahedral positions. The distribution of interlayer cations was uncertain, but from a Fourier synthesis on a fully-collapsed specimen it was evident that the interlayer cations in the non-swelling layer contained about 7 electrons per formula unit[3] and no detectable interlayer electrons in the swelling part of the structure. The available cations for interlayer positions can be divided into $(Na_{0.38}K_{0.20}Ca_{0.15})$ with 10 electrons, and $Na_{0.25}$ with 2.5 electrons, which agrees satisfactorily with 7 and zero electrons respectively from the Fourier analysis.

The O, (OH) content of allevardite was given as $O_{19.40}$ $(OH)_{4.60}$ for the two layers, which would correspond to $O_{10}(OH)_2$ for the non-swelling layer and $O_{9.40}(OH)_{2.60}$ for the swelling layer. The latter ratio can be written $O_{10}(OH)_2H_{0.60}$ and the 0.6 H ions can be expected to form hydronium ions with interlayer water molecules.

THE GENESIS OF LONG-SPACING, MICA-LIKE MINERALS

The genesis of these minerals has been discussed at length by Sudo, *et al.* (1962). Their work shows that considerable variability exists in the degree of regularity of the alternating layer sequence from one mineral to another. Allevardite probably has the most regular alternation yet observed in these mica-like minerals. The East Valley, Goto Island, material discussed in this paper has considerable regularity which, however, is much inferior to that of allevardite. The sample from Goto Mine described by Sudo *et al.* may be slightly less regular than the East Valley sample. At the opposite extreme, Sudo and his colleagues find many examples where the expandable and non-expandable layers form a nearly random sequence.

Interpretation given by Sudo et al. The origin of these minerals presents difficult questions, particularly for those minerals which have a regular or nearly regular alternation of layers of two kinds. Sudo *et al.* (1962, p. 391) attributed their formation to differential leaching and hydration of the parent minerals arising from structural and/or compositional variations from layer to layer within individual crystals; variations in the numbers and kinds of isomorphous ions are considered. To explain *regular* mixed-layer structures on this basis, they suppose that a regular alternation of some kind was already present in the parent material which became enhanced by the alteration process. For example, one may visualize a partial separation into K-rich and Na-rich layers which weather differentially and thus give an enhanced alternating character to the resulting mineral.

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[3] In the previous paper (Brindley 1956, p. 95) the double formula was used with 14 electrons in the interlayer non-swelling position. The number must be halved to conform with the formulae used here.

This kind of explanation, however, shifts the problem one stage further back. It would be necessary to consider that the partial separation would be a function of time, temperature, and pressure conditions when the original crystallization occurred.

Alternative interpretation based on unmixing of cations during the weathering process. An explanation along different lines has been sought. It may be accepted that leaching and hydration of a parent mica structure starts at particular points on the boundary surface where the layers terminate; edges rather than faces are likely to be attacked. Whether the points of attack are determined by chance or for energetic reasons is probably of secondary importance in relation to the alteration process because when the chemical change has commenced, it is likely to continue along the same structural plane or planes. The maintenance of many structural layers, apparently with little or no change of properties, points to a very mild chemical process which modifies mainly the interlayer ion population with little influence on the layer structure itself. Parenthetically it may be stated that if the layer composition itself is modified, then octahedral layers are more likely to be changed than tetrahedral layers, as Osthaus (1954, 1956) has shown in experiments on the acid extraction of ions from montmorillonites.

It is now suggested that the origin of the long-spacing minerals with layer sequences showing some approach to a regular alternation may arise from an unmixing of the interlayer cations. This idea is not entirely new though it has not been suggested previously as the origin of the long-spacing mica-like minerals. Méring and Glaeser (1954), Glaeser and Méring (1954, 1958) and Glaeser (1958) have shown that when the ratio of Ca to Na ions in mixed Ca, Na-montmorillonites exceeds a certain value, the two kinds of ions tend to unmix giving Na and Ca layers.

Another example of the formation of a long-spacing periodicity in a partially dehydrated layer mineral has been given by Walker (1956), who has shown that when a vermiculite containing one water layer between silicate sheets, with $d(001) = 11.59$ Å, is carefully dehydrated, a mixed-layer phase develops with $d(001) = 20.6$ Å. The fully dehydrated mineral has $d(001) = 9.02$ Å. The intermediate stage corresponds to a regular alternation of layers with spacings of 11.59 and 9.02 Å. The alternating sequence must have a high degree of regularity because many odd-order reflections up to (00, 13) are clearly observed, and even (00, 23) is recorded as a weak but diffuse reflection.

It is now suggested that when a mica is subjected to a mild leaching process in which only interlayer cations are affected, the process may take place randomly in the initial stages, with charge balance being maintained by hydration, *i.e.*, addition of H ions or hydronium ions. At

a later stage, an unmixing process may occur among the remaining cations with some layers becoming more or less filled with cations (these would be non-swelling layers) and others largely depleted, apart from $(H_3O)^+$ ions, (these would be the swelling layers).

Comparison of the two hypotheses. Both hypotheses regard the formation of these swelling mica minerals as being due to mild leaching and hydration which leaves the alumino-silicate structures unaffected. Sudo's hypothesis shifts the problem of the alternating structures back to the parent minerals where some kind of compositional alternation is considered to have existed. The "unmixing hypothesis" makes use of independent evidence obtained on montmorillonites but it still remains to be proved that a similar process may take place during the alteration of micas.

Some General Considerations

The subject under discussion represents a particular aspect of the broad subject of mica alternation, which includes the nature and origin of sericites and illites, Without any prior bias towards the Brown-Norrish hypothesis of hydronium ions in illites, the present analysis has led to a similar conclusion. However, one may well ask why the unmixing process suggested here as the origin of the alternating swelling and non-swelling layer sequence does not take place more generally in partially altered micas of the illite type. These questions are too extensive to be treated as an addendum to the present inquiry but a re-evaluation of illites and sericites seems very desirable.

Acknowledgments

References

BRADLEY, W. F. (1950) The alternating layer sequence of rectorite. *Am. Mineral.* **35,** 590–595.

BRINDLEY, G. W. (1956) Allevardite, a swelling double-layer mica mineral. *Am. Mineral.* **41,** 91–103.

BROWN, G. AND K. NORRISH, (1952) Hydrous micas. *Mineral. Mag.* **29,** 929–932.

CAILLÈRE, S., A. MATHIEU-SICAUD, AND S. HÉNIN, (1950) Nouvel essai d'identification du minéral de La Table près Allevard, l'allevardite. *Bull. Soc. Franç. Miner. Crist.* **73,** 193–201.

GLAESER, R. (1958) Détection de la démixtion des cations Na, Ca dans une hectorite bi-ionique. *C. R. Acad. Sci., Paris,* **246,** 2909–2912.

—— AND J. MÉRING, (1954) Isothermes d'hydratation des montmorillonites bi-ioniques (Na, Ca). *Clay Minerals Bull.* **2,** 188–193.

—— AND J. MÉRING, (1958) Le role de la valence des cations échangeables dans l'hectorite. *C. R. Acad. Sci., Paris,* **246,** 1569–1572.

IWAO, S., T. HAMACHI, M. YAMADA, AND H. INOUE, (1953) Pyrophyllite and diaspore deposits in Goto Islands, Kyushu. *Bull. Geol. Surv. Japan,* **4,** 81–97.

KODAMA, H. (1958) Mineralogical Study of some pyrophyllites in Japan. *Mineral. Jour. (Japan),* **2,** 236–244.

MÉRING, J. AND R. GLAESER, (1954) Sur le role de la valence des cations échangeables dans la montmorillonite, *Bull. Soc. Franç. Miner. Crist.* **77,** 519–530.

OSTHAUS, B. B. (1954) Chemical determination of tetrahedral ions in nontronite and montmorillonite. *Clays and Clay Minerals, Second National Conference,* pp. 404–417.

—— (1956) Kinetic studies on montmorillonites and nontronite by the acid dissolution technique. *Clays and Clay Minerals, Fourth National Conference,* pp. 301–321.

SHIMODA, S. AND T. SUDO (1960) An interstratified mixture of mica clay minerals. *Am. Mineral.* **45,** 1069–1077.

SUDO, T., H. HAYASHI, AND S. SHIMODA, (1962), Mineralogical problems of intermediate clay minerals. *Clays and Clay Minerals, Ninth National Conference,* pp. 378–392.

TAKESHI, H. (1958) Kaolin minerals in "roseki", (in Japanese). *Mineral. Jour. Japan,* **3,** 388–405.

WALKER, G. F. (1956) Mechanism of dehydration of Mg-vermiculite. *Clays and Clay Minerals, Fourth National Conference,* pp. 101–115.