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CONVERSION OF MICA INTO AN INTERSTRATIFIED MINERAL

By

Katsutoshi TOMITA* and Toshio SUDO**

Abstract

Sericite from Gotō, Nagasaki Prefecture, Japan was used as a starting material. Powdered sample was heated in a platinum crucible for certain hours, quenched to room temperature and boiled with acids such as hydrochloric or sulphuric acids of known concentration. By the above procedure, the sample was easily changed into an interstratified mineral partly or entirely. In order to change the sample into an interstratified mineral, the sample should be heated up to the temperature range of dehydroxylation.

The X-ray reflection properties of the treated sample agree with an "alleverdite" structure having a single layer of water molecules reported by Brindley (1956). When this product was boiled in a magnesium chloride solution (1.2%) or calcium chloride solution (1.2%) for one hour, the properties of the sample have approached to a regular interstratified mineral of mica-montmorillonite.

Introduction

Interstratified or mixed-layer minerals were reported many years ago by Gruner (1934), Alexander, Hendricks and Nelson (1939), Sudo (1954a), Sudo et al. (1954b, c), Brindley et al. (1954), Heystek (1954), Brindley (1956) and others, but it is only recently with the development of adequate techniques for their identification that their great importance has been recognized. In an extensive investigation of sedimentary rocks, Weaver (1956) found such minerals to be among the commonest types present in the clay fraction.

Interstratified or mixed-layer minerals represent a special case of intergrowth. The simplest case is that of more or less hydrated layers. Most of the above-mentioned studies are concerned with pure description or with the statistics of mixing two or more types of layers in a sequence. Hendricks and Teller (1942), Méring (1949), and MacEwan et al. (1961) considered a possibility of nonrandom stacking sequences. The energetics of the interaction between layers was recently considered by Sato (1965) and Zen (1967).

The origin of long-spacing (mica-like 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) attributed their formation to differential leaching and

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hydration of the parent minerals arising from structure and/or compositional variations from layer to layer within individual crystal. Brindley and Sandalaki (1963) suggested that a regular alternation may arise from an unmixing of the interlayer cations. But these explanations are no more than a hypothesis.

Concerning synthesis of interstratified minerals only a few experiments have been reported. Iiyama and Roy (1963) synthesized mixed-layer minerals from pure chemical reagents. Synthesis of an interstratified mineral from micas was reported by Ueda and Sudo (1966) and recently by Tomita and Sudo (1968). Data of syntheses of mixed-layer minerals are considered to be important for interpretation of the mechanism of forming regular mixed-layer minerals. The present writers have succeeded in forming an interstratified mineral from mica. The procedure employed here is an easy method and its result seems to be pertinent to the discussion of the origin of interstratified clay minerals.

Experimental

A. Starting Material

Sericite from the Gotō mine, Nagasaki Prefecture, Japan was used as a starting material. It was ground in an agate mortar and fractions less than 2μ were collected by the sedimentation methods. The $<2\mu$ material was dried in air and was used in the experiments. The original sericite sample was investigated by *X*-ray analysis, differential thermal analysis, chemical analysis, infrared absorption analysis and by observation of electron micrograph. The *x*-ray powder pattern of the original sample is shown in Fig. 5, and its powder data is listed in Table 1. The differential thermal analysis curve is shown in Fig. 7 and the infrared absorption spectra of the original sericite is shown in Fig. 8. As a result, it was confirmed that the original sample is assigned to the $2M_1$ type in the polytype notation and does not contain any interstratified minerals.

B. Method

Powdered sample was heated in a platinum crucible for certain hours, quenched to room temperature and boiled with acids such as hydrochloric or sulphuric acids of known concentration. The experiments were carried out at various temperatures between 650°C and 950°C . Hydrochloric and sulphuric acids were used in the experiments. After boiling for a desired time, the sample was washed with distilled water by filtering until the filtrate showed no acid reaction. The washed sample was dried in air and investigated by *X*-ray analysis, differential thermal analysis, infrared absorption analysis and chemical analysis.

(A). Heating effects on sericite

Previous works and behaviour of the sericite from Gotō in heating

The original sericite should be heated above 650°C in order to change the sample into an interstratified structure. The behaviour of the sericite in heating was investigated in this experiment. It is a known fact that the dehydration of illite, no matter

Table 1. X-ray powder data for the original sericite(s) and the sericite heated at 800°C (specimen 1541).

hkl	(s)		(1541)	
	d (Å)	I	d (Å)	I
002	10.0	84	10.16	38
004	5.01	37	5.04	37
110	4.47	12	4.51	42
111				
111	4.29	3	4.33	5
022	4.11	4	4.19	5
112	3.95	2	3.98	6
113	3.90	6	3.92	16
023	3.74	9	3.82	20
113				
114	3.49	9	3.53	21
024	3.34	100	3.36	100
006				
114	3.20	11	3.23	19
115	3.12	1		
025	2.99	16	3.025	21
115	2.868	11	2.885	15
116	2.797	8	2.823	8
131	2.585	9	2.629	7
200				
202	2.560	16	2.592	21
131				
008	2.508	5	2.521	18
202	2.458	5		
133				
204	2.380	6	2.410	9
133				
204	2.238	2	2.259	10
135				
223	2.200	2		
206	2.146	3	2.171	3
135	2.127	7	2.156	7
136				
044	2.053	2	2.090	5
0, 0, 10	2.002	17	2.014	19
206	1.948	3	1.961	3
137				
138	1.731	3	1.752	4
2, 0, 10				
139	1.647	6	1.663	10
060	1.499	8	1.513	5
331				

whether it is di- or tri-octahedral, proceeds in a number of stages (Grim and Bradley 1940, 1948). Most of the water adsorbed on the surface of particles and a small amount which may be interlayered with illite sheets comes off rapidly below 110°C and the remainder more slowly between 110°C and 350°C . Water formed by expulsion of $(\text{OH})^{-}$ ions comes off rapidly at first between 300°C and 600°C , but a small amount remains to be given off by further heating.

In this experiment similar results were obtained. On the original sericite thermal dehydration studies were carried out. Some of the equilibrium experiments were carried out. The specimen was heated at various temperatures and was left at the respective temperatures until no more weight loss could be observed. Results thus obtained at 550°C and 650°C are shown in Fig. 1. At 550°C , approximately 85% of hydroxyl groups were actually expelled, but the liberation of the remainder did not take place at this stage. At 950°C , almost the whole quantity of hydroxyl groups was expelled as shown in Fig. 2.

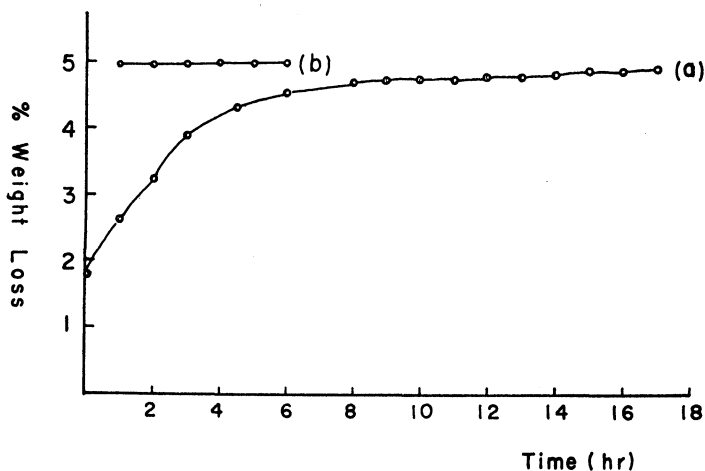


Fig. 1. Equilibrium curves of sericite.
(a). at 550°C , (b). at 650°C .

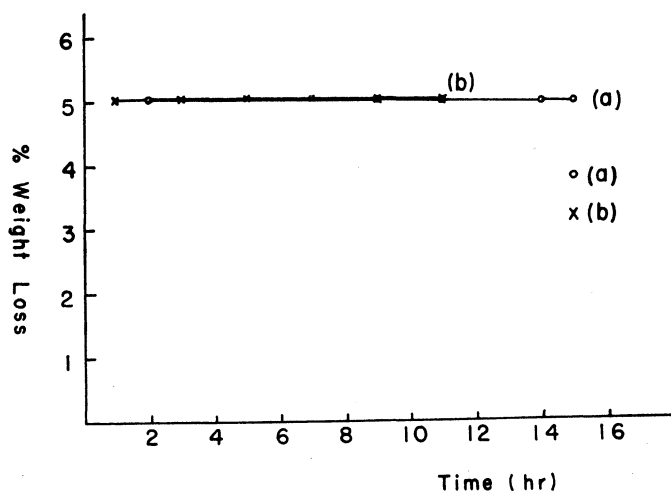


Fig. 2. Equilibrium curves of sericite.
(a). at 750°C , (b). at 950°C .

It is said that illite which has lost its constitutional water appears to be a stable modification having a slightly enlarged interlayer spacing, and gives basal reflections with somewhat changed relative intensities. Continuous x -ray diffraction records of positions and intensities of (002) and (006) reflections with increasing temperature were obtained in order to investigate the precise temperature at which the relative intensities of the basal reflections changed. The intensities of the (002) and (006) reflections obtained by continuous records are shown in Fig. 3. Above about 650°C the sericite transformed into an anhydrous modification and at 1050°C mullite was formed from the sample.

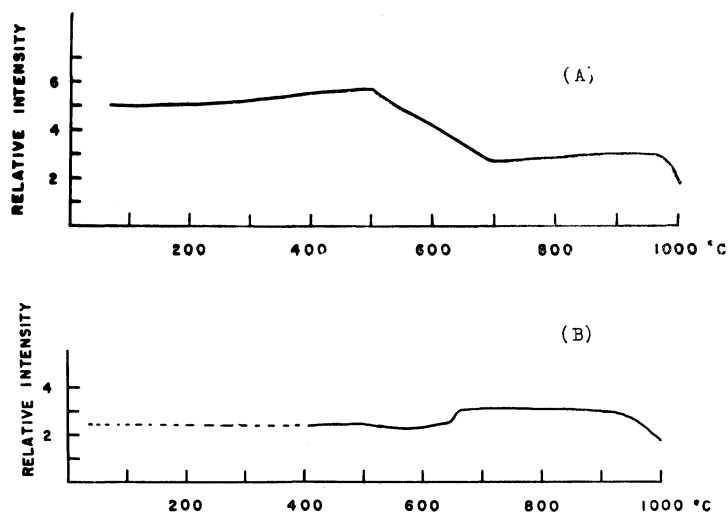


Fig. 3. Changes of intensities of (002) and (006) reflections of original sericite with increasing temperature.

(A). Intensity of (002) reflection.

(B). Intensity of (006) reflection.

Using a flake of muscovite from Ishikawa-yama, Fukushima Prefecture, Laue photographs were taken to investigate the structural change between the unheated mica and the heated mica at various temperatures. The same crystal was used throughout a series of heat treatments. The crystal was mounted, at first, on a thin glass fibre to obtain a diffraction pattern in the unheated state. Then, it was demounted from the specimen holder, heated at a certain temperature and mounted again to be examined for the pattern after that particular temperature; it was assumed that the structural state after heating was retained by the quench treatment. A Laue photograph radiated vertical to a cleavage plane of the unheated crystal show clear hexagonal spots and that of the sample heated at 800°C shows radial streaks as illustrated in Fig. 4. Such radial streaks are observed in a Laue photograph of a sample heated at temperature range between 650°C and 1000°C. The radial streaks indicate existence of stacking disorder of layers in the direction of c^* .

(B). Conversion of sericite into an interstratified mineral by treatment with 6N HCl solution

Specimens heated at different temperatures were placed in a boiling acid solution (6N

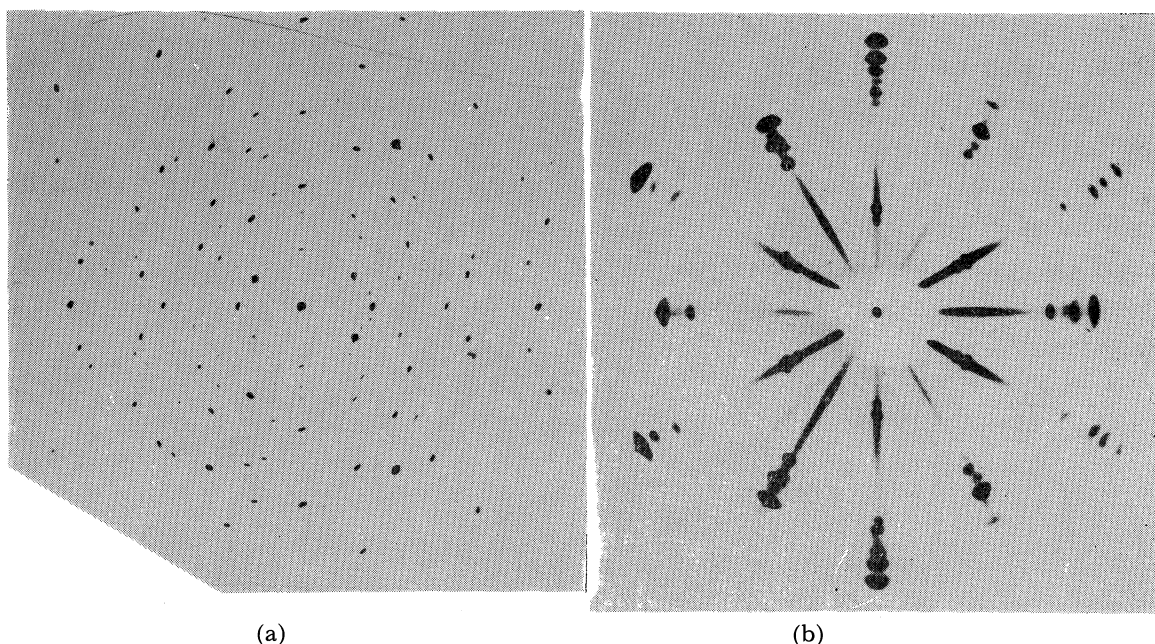


Fig. 4. Laue photographs of unheated sericite and sericite heated to 800°C.
(a). unheated sericite, (b). heated sericite.

HCl) and left for certain hours. Then the acid-treated samples were washed with distilled water by filtering. Unheated sample was also treated by the same method. The acid-treated samples of specimens heated above 650°C showed an interstratified structure, whereas acid-treated samples of unheated specimen and heated specimens at temperatures below 600°C showed no change in their x -ray powder patterns.

Effects of heating temperature

Acid-treated samples after heating at different temperatures were investigated with respect to the preparation of oriented aggregates and the observation of changes in the basal spacing of the minerals. All experiments were carried out under the same condition. The weight of each of the heated samples used was 0.1 g and quantity of solution was 50 cc. Figure 5 shows x -ray powder patterns of samples which were synthesized from heated sericites at various temperatures by boiling them in 6N HCl for 30 minutes. Specimen 1563 (heating temp.: 450°C) and specimen 1487 (heating temp.: 600°C) show no change in their x -ray powder patterns. The pattern of specimen 1497, which was boiled for 30 minutes in a 6N HCl solution after heated at 660°C for one hour, shows a 22.2 Å of (001) reflection. The 22.2 Å peak moved to 23.3 Å by treatment with ethylene glycol. The 10.5 Å peak was replaced by 11.6 Å by treatment with ethylene glycol. Specimens treated with 6N HCl after heating at 800°C and 900°C for one hour respectively showed the same behaviour as that of specimen 1497. The powder patterns of acid-treated samples after heating at 800°C and at 900°C are shown in Fig. 5, together with that of specimen 1497. The powder data of air-dried samples (1556 and 1582) and those after treatment with ethylene glycol are listed in Table 2. It is noteworthy that specimens heated at temperature range between 650°C and 950°C show long spacing by treatment with acid.

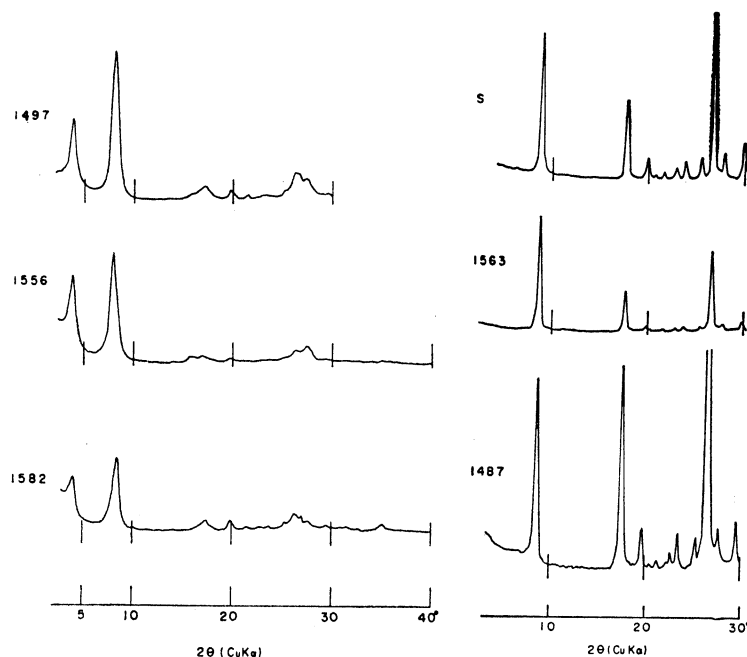


Fig. 5. X-ray diffraction patterns of sericite and specimens synthesized from sericite heated at various temperatures by treatment with 6N HCl. S: Sericite, 1563: from sericite heated to 450°C for 1 hr., 1487: from sericite heated to 600°C for 6 hrs., 1497: from sericite heated to 660°C for 1 hr., 1556: from sericite heated to 800°C for 1 hr., 1582: from sericite heated to 900°C for 1 hr.

Table 2. The powder data for the specimens 1556 and 1582. $\lambda(\text{CuK}\alpha)=1.5418\text{\AA}$

1556		E G		1582		E G	
d(Å)	I	d(Å)	I	d(Å)	I	d(Å)	I
23.2	57	24.5	89.5	22.1	45	23.9	90
11.05	100	11.9	100	10.65	100	11.9	100
5.6	7.5	5.79	4			5.83	45
4.51	4.7	4.67	7.5	4.46	14.5	4.65	9.5
3.24	14	3.30	30	3.75	6.5	3.88	7.5
				3.24	11	3.32	29

1556: sample treated with 6N HCl after heating at 800°C.

1582: sample treated with 6N HCl after heating at 900°C.

EG: treated with ethylene glycol.

Effects of heating period

As it became obvious that an interstratified mineral could be synthesized from sericite heated above 650°C by boiling it in 6N HCl, effects of different pre-heating periods were investigated. Figure 6 shows x-ray powder patterns of specimens which were treated with 6N HCl for 30 minutes after heating at constant temperature of 660°C for various periods and those of specimens treated with ethylene glycol. The patterns are of specimens synthesized from sericite heated for 20, 30 and 70 minutes respectively.

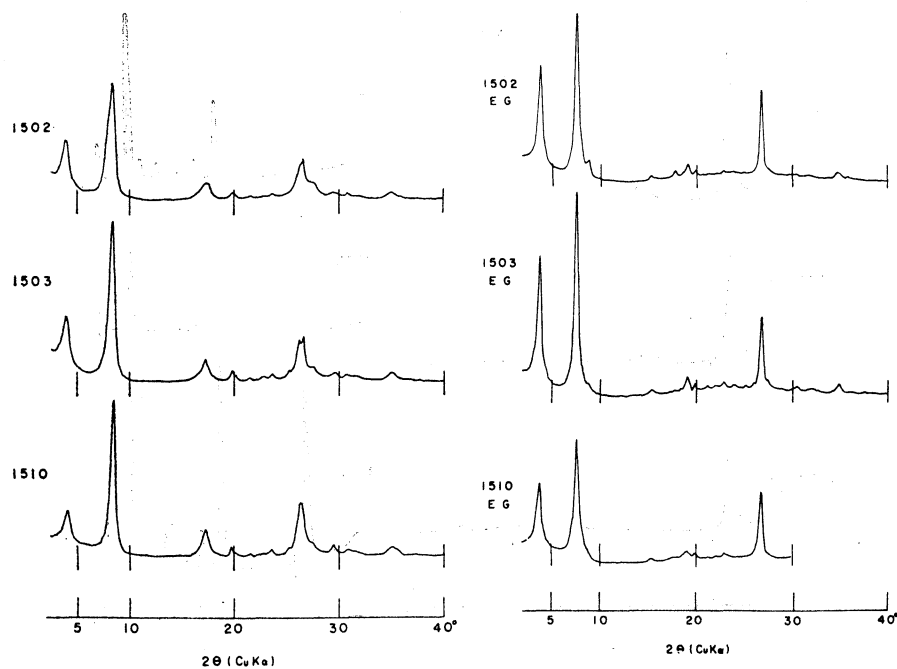


Fig. 6. X-ray diffraction patterns of specimens synthesized from sericite heated to 660°C for various periods of time. 1502: specimen synthesized from sericite heated for 20 minutes, 1503: from sericite heated for 30 minutes, 1510: from sericite heated for 70 minutes, EG: treated with ethylene glycol.

Table 3. The powder data of air-dried samples (1502, 1503 and 1510) and those after treatment with ethylene glycol. $\lambda(\text{CuK}\alpha)=1.5418\text{\AA}$

1502 (EG)		1503 (EG)		1510 (EG)	
d (Å)	I	d (Å)	I	d (Å)	I
23.8	60	23.8	62	23.9	53.8
11.8	100	11.8	100	11.7	100
5.83	3.5	5.79	3.3	5.79	4
4.67	7.5	4.67	7	4.67	6.8
3.33	52.3	3.326	36.4	3.339	56.2
2.578	4.6	2.575	5.2		
1502		1503		1510	
d (Å)	I	d (Å)	I	d (Å)	I
22.6	45.5	22.1	29	21.6	22
10.6	100	10.5	100	10.5	100
4.46	7	4.48	7.5	4.49	6.8
3.77	4	3.77	4.3	3.77	5
3.24	12.5	3.24	7.3	3.25	10
2.56	7	2.56	6	2.56	7

1502: specimen synthesized from sericite heated for 20 minutes.

1503: specimen synthesized from sericite heated for 30 minutes.

1510: specimen synthesized from sericite heated for 70 minutes.

Each of the x -ray powder patterns seems to be identical. Specimen 1502 synthesized from sericite heated for 20 minutes contains a small amount of sericite. This was confirmed by treatment with ethylene glycol. The 10 Å reflection of sericite overlapped a 10.6 Å peak of the interstratified mineral and the 10 Å peak was separated by treatment with ethylene glycol. The pre-heating period, so far as the duration was within 70 minutes, did not give any serious effects on the forming of the interstratified mineral. The powder data for the synthesized minerals and the specimens treated with ethylene glycol are listed in Table 3.

Effects of acid

A specimen heated at 800°C for one hour was boiled in distilled water for six hours. There was no change in x -ray diffraction pattern of the sample. The heated sericite was boiled in 1.2% MgCl₂ or 1.2% CaCl₂ solution for six hours, but any interstratified mineral could not be synthesized from heated sericite without boiling it in the presence of acids. By boiling in acid solution, a regularly mixed-layer mineral having one layer of water molecules in interlayer was always synthesized from specimens which had been heated at temperature range between 660°C and 950°C. By boiling for a few minutes, an interstratified mineral was formed from pre-heated mica.

Using a natural mixed-layer mineral reported by Shimoda et al. (1960), effect of acid solution was investigated. The natural mixed-layer mineral was boiled in a 6N HCl solution for 30 minutes. 27 Å of (001) reflection of the natural mixed-layer mineral moved to a 22 Å by treatment with acid.

Effects of cations

A mixed-layer mineral synthesized from heated sericite by boiling it in acid solution was always of the regular mixed-layer type with one layer of water molecules between silicate layers. In nature, many regular mixed-layer minerals having two layers of water molecules in interlayers are found. An experiment was carried out to synthesize such regularly interstratified minerals. The present authors have succeeded in synthesizing such an interstratified mineral. The procedure was as follows: Specimen 1497 was placed in an aqueous 1.2% MgCl₂ solution and was boiled for six hours. Then the boiled specimen was washed with distilled water. The x -ray powder pattern of the washed sample showed a 24 Å reflection, which was replaced by a 26.8 Å reflection by treatment with ethylene glycol. It was not possible to obtain from heated sericite such a regularly interstratified mineral with two layers of water molecules in interlayers by boiling it in water only, unless magnesium chloride or calcium chloride was used.

Properties of synthesized interstratified mineral

As an interstratified mineral was found to be formed from pre-heated sericite by boiling it in hydrochloric acid, the properties of a synthesized mineral was investigated. Specimen 1497 was selected for the investigation.

1. X-ray analysis

Specimen 1497 contains a small amount of sericite as a mixture. The x -ray powder

data for this specimen are listed in Table 4. Ethylene glycol caused the 22.2 Å basal reflection to shift to 23.3 Å and 10.5 Å reflection to 11.6 Å. After heating to 300°C, the 22.2 Å peak disappeared giving a 21.5 Å reflection which remained even at 500°C, but at 800°C the 21.5 Å peak was replaced by a 10.1 Å reflection. Judging from the powder data and the behaviour of basal reflections by treatment with ethylene glycol and by heating, this specimen is a nearly regularly interstratified mineral of mica and hydrated-mica having one layer of water molecules between silicate layers.

Table 4. X-ray diffraction data for the 1497 specimen after various treatments.
 $\lambda(\text{CuK}\alpha)=1.5418\text{\AA}$

unheated		E G		300°C		500°C		800°C	
d(Å)	I	d(Å)	I	d(Å)	I	d(Å)	I	d(Å)	I
22.2	65.0	23.3	71.0	21.5	13.5	21.5	5.2		
10.5	256.0	11.6	100.0	10.0	100.0	10.1	67.5	10.1	56.6
5.54	9.5	5.83	3.0	4.97	26.5	5.01	41.5	5.01	35.8
4.51	8.0	4.67	5.0						
3.23	28.0	3.33	36.5	3.30	55.8	3.33	100.0	3.34	100.0
2.528	12.5			2.57	1.2	2.52	4.0	2.50	5.9
2.280	1.5								
2.027	34.5			1.98	15.5	2.00	10.0	2.01	12.8
1.749	1.0							1.667	2.6
1.469	1.5								
1.370	5.5								
1.344	1.5								
1.259	3.0								

EG: treated with ethylene glycol.

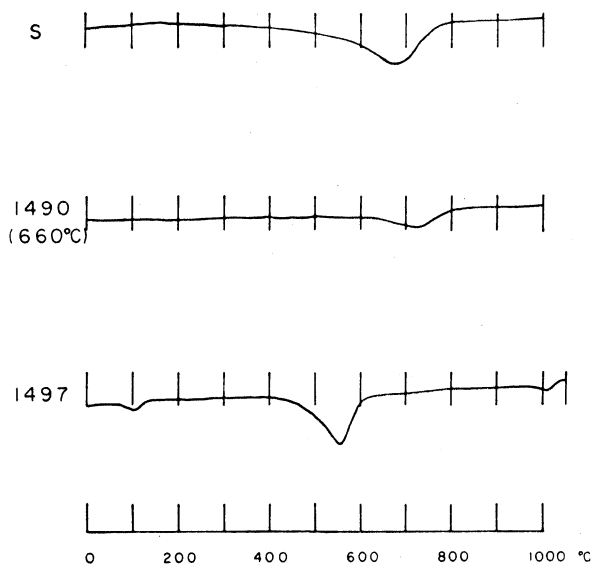


Fig. 7. Differential thermal analysis curves. s: original sericite, 1490: sericite heated to 660°C, 1497: specimen synthesized from specimen 1490 by treatment with 6N HCl.

2. Differential thermal analysis

Differential thermal analysis curve of specimen 1497 is shown in Fig. 7, together with that of the original sericite and the sericite heated at 660°C. The curve of the original sericite has an endothermic peak at 680°C. The curve of the sericite heated at 660°C (specimen 1490) shows a small endothermic peak at about 700°C and there is no endothermic peak below 600°C. The curve of specimen 1497 shows two endothermic peaks, one is at about 100°C and the other at 560°C. The first endothermic peak is due to the dehydration of adsorbed water and interlayer water, and the second one is due to the dehydroxylation. The endothermic peak at 560°C increased in intensity as compared with that of the heated sericite. This fact tells that rehydration and rehydroxylation occurred in the heated sericite when it was boiled in hydrochloride solution. The temperature of the endothermic peak is lower than that of the original sericite.

3. Infrared absorption spectra

As shown in Fig. 8, the original sericite has absorption bands at 3640 cm^{-1} , 1020 cm^{-1} , 920 cm^{-1} , 825 cm^{-1} and 800 cm^{-1} . The band at 3640 cm^{-1} is due to the OH stretching

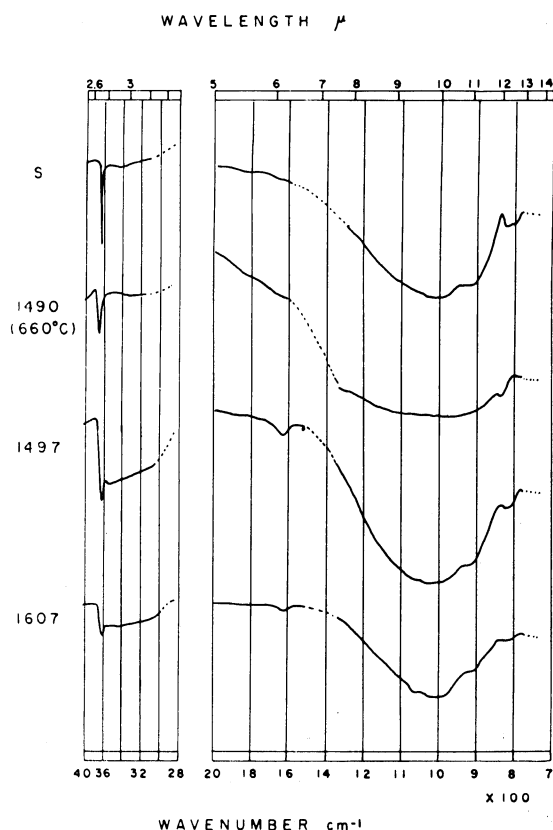


Fig. 8. Infrared absorption spectra of unheated, heated sericite and synthesized specimens. s: unheated sericite, 1490: sericite heated to 660°C, 1497: synthesized specimen from specimen 1490 by treatment with 6N HCl, 1607: synthesized specimen from specimen 1490 by treatment with 0.8N H_2SO_4 .

vibration and the band at 920 cm^{-1} is assigned to the O-H-Al^{3+} vibration. The absorption band at 920 cm^{-1} disappeared after heating to 660°C , but the band at 3640 cm^{-1} remained at the temperature. The spectra of specimen 1497 show a strong absorption band at 3640 cm^{-1} , and a new band at 1640 cm^{-1} which is due to adsorbed water. A new broad absorption band at 3400 cm^{-1} is also recognized distinctly. This absorption band is due to the adsorbed water in interlayers and is considered to be the same kind as that observed in absorption spectra of montmorillonite and hydrated halloysite. Judging from these results, rehydration and rehydroxylation in sericite heated at 660°C for one hour occurred when the specimen was boiled in hydrochloric acid. This fact agrees with the experimental results of differential thermal analysis.

4. Electron microscopic observation

Electron micrograph of specimen 1497 shows a hexagonal shape as shown in Fig. 9. The original shapes and sizes are still preserved well.



Fig. 9. Electron micrograph of specimen 1497.

5. Chemical analysis

Chemical analysis data for specimen 1497 are listed in Table 5 together with that of the original sericite. The chemical data of the two samples are very similar, but the original sample has a higher content of K_2O and a lower content of $\text{H}_2\text{O}(-)$ as compared with the synthesized sample. As the data of $\text{H}_2\text{O}(-)$ was obtained below 110°C , it is considered that the adsorbed water in interlayers of vermiculitic structure could not be completely expelled at this temperature. The value of $\text{H}_2\text{O}(+)$ of the interstratified mineral is a little larger for mica-like structure, for the reason mentioned above. Some

amounts of $\text{H}_2\text{O}(+)$ should be counted for $\text{H}_2\text{O}(-)$.

Table 5. Chemical compositions of the original sericite (a) and the synthetic mineral (b).

	(a)	(b)
SiO_2	47.24	48.45
TiO_2	0.38	tr.
Al_2O_3	35.04	33.02
Fe_2O_3)		
FeO)	0.59	0.58
MnO	tr.	tr.
MgO	0.21	0.18
CaO	0.16	0.08
K_2O	8.75	6.10
Na_2O	1.37	1.28
$\text{H}_2\text{O}(+)$	5.52	7.77
$\text{H}_2\text{O}(-)$	0.32	2.22
P_2O_5	none	none
Total	99.58%	99.68%

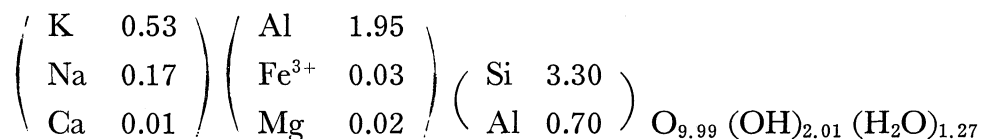
6. Calculation of structure formula

In the calculation of the structure formula of specimen 1497, the amount of sericite was neglected. Table 6 shows an example of the calculation for specimen 1497. The

Table 6. Calculation of chemical formulae for sample 1497.

(1)	(2) weight per cent	(3) Moles (relative)	(4) Ions (relative)	(5)
SiO_2	48.45	8.063	Si 8.063	3.30
Al_2O_3	33.02	3.239	Al 6.478	2.65
Fe_2O_3	0.58	0.036	Fe^{3+} 0.072	0.03
MgO	0.18	0.045	Mg 0.045	0.02
CaO	0.08	0.014	Ca 0.014	0.01
K_2O	6.10	0.648	K 1.296	0.53
Na_2O	1.28	0.207	Na 0.414	0.17
$\text{H}_2\text{O}(+)$	7.77	4.317	OH 8.634	3.54
$\text{H}_2\text{O}(-)$	2.22	1.233	H_2O 1.233	0.50
			O 22.548	9.23

analysis in weight % (col. 2) was converted into relative mols (col. 3) and relative ions (col. 4). Thus, the final figures in column 5 were obtained by scaling the total cationic charges to 22, which here is equal to the total anionic charges: $22 = 2x + y$, where x and y are subscripts as $\text{O}_x(\text{OH})_y$. The structure formula is as follows:



(C). Properties of products from pre-heated sericite by sulphuric acid treatment

As it was confirmed that pre-heated sericite was changed into an interstratified mineral by boiling it with hydrochloric acid, experiments using sulphuric acid instead of hydrochloric acid were carried out. In these experiments, the procedure was the same as in the case of hydrochloric acid treatment. Pre-heated sample of a constant amount (0.1g) was boiled in a sulphuric acid solution (0.8N) for various periods of time. 50cc of solution was used in all experiments. Boiled specimen was washed with distilled water many times. The properties of the air-dried samples were investigated. The samples thus treated showed an interstratified structure.

Effects of boiling period on properties of synthesized specimens

As it was confirmed that an interstratified mineral was formed from pre-heated sericite by boiling it with sulphuric acid, the effect of boiling period was investigated with respect to the observation of changes in the basal spacing, layer stackings and amount of interstratified mineral. In this experiment, sericite heated at 800°C for one hour was used. The pre-heated sericite was thrown into boiling sulphuric acid solution (0.8N) and was continuously boiled for proper periods of time. After boiling for an adequate period, synthesized sample was immediately washed with distilled water. Properties of the dried specimen in the air were investigated.

1. X-ray analysis

X-ray powder data of specimens synthesized under different periods of boiling are

Table 7. X-ray diffraction data for the specimens 1633, 1612, 1636 and 1649.
 $\lambda(\text{CuK}\alpha) = 1.5418\text{\AA}$

1633		1612		1636		1649	
d(Å)	I	d(Å)	I	d(Å)	I	d(Å)	I
23.25	184.5	23.1	4250	23.3	339	22.65	1083
11.05	271.0	11.05	7917	11.2	523	11.2	1825
		7.56	42	7.56	2.5		
5.6	15.7	5.64	1267	5.61	47	5.60	164
3.24	38.9	3.25	2553	3.24	46	3.24	781
		2.82	88				
		2.52	1			2.52	14
2.02	20.7	2.036	1036	2.036	39	2.036	168
		1.881	67	1.873	1		
1.439	1.0	1.417	1.5			1.439	19
1.258	4.8	1.258	3			1.257	1

listed in Table 7. Those of sericite heated at 800°C for one hour are listed in Table 1. Specimen 1633 which was synthesized by boiling for ten minutes shows a 23.25 Å reflection in its *x*-ray powder pattern, and it remained almost unchanged by treatment with ethylene glycol. The 23.25 Å peak contracted to 10 Å in heating at 300°C for one hour. The powder pattern of specimen after heating to 300°C for one hour showed the same pattern as that of the sericite heated at 800°C for one hour. A small amount of mullite was formed by heating at 1050°C from specimen 1633. In this specimen, 10 Å reflection of the sericite is observed. Powder pattern of specimen 1612 which was synthesized by boiling for 30 minutes shows a strong 23.1 Å reflection, and it remained almost unchanged by treatment with ethylene glycol. A strong 11.05 Å reflection is observed and it moved to a 11.5 Å by treatment with ethylene glycol. Powder pattern of specimen 1636 which was synthesized by boiling for four hours is similar to that of specimen 1612. 23.3 Å peak moved to 21 Å in heating at 300°C for one hour. The 21 Å peak still remained in heating at 500°C for one hour, but it contracted to 10 Å in heating at 800°C for one hour. Relative intensities of 23.3 Å and 11.2 Å reflections, corresponding respectively to the first and second order reflections for the 23.3 Å peak, increased as compared with those of specimen 1612. Intensity of 3.33 Å reflection of sericite decreased as compared with that of specimen 1612. These facts indicate that the amount of interstratified mineral increased and the amount of sericite decreased. Powder pattern of specimen 1649 synthesized by boiling for eight hours is similar to that of specimen 1636. Mullite was formed from specimen 1649 at 1050°C. The powder data for specimen 1649 treated with ethylene glycol and heated to 500°C are listed in Table 8.

Table 8. X-ray diffraction data for the specimen 1649 after various treatments.
 $\lambda(\text{CuK}\alpha)=1.5418\text{\AA}$

untreated		E G		500°C	
d(Å)	I	d(Å)	I	d(Å)	I
22.65	1083	23.3	88		
11.2	1825	11.6	147	10.16	249
5.60	164			5.04	153
		4.7	9		
3.24	781	3.34	108	3.35	356
2.52	14	2.57	3	2.515	32
2.036	168	2.10	3	2.01	35
		1.949	3	1.667	7
1.439	19	1.441	2	1.437	3
1.257	1	1.261	1	1.259	1

EG: treated with ethylene glycol.

2. Differential thermal analysis

Differential thermal analysis curves of interstratified minerals synthesized under different periods of boiling are shown in Fig. 10, together with those of the original sericite

and the sericite heated at 800°C for one hour. The endothermic peak at about 680°C in the sericite sample completely disappeared in the heated sericite and reappeared at a lower temperature, 575°C, in specimen 1633 and at about 550°C in specimen 1636.

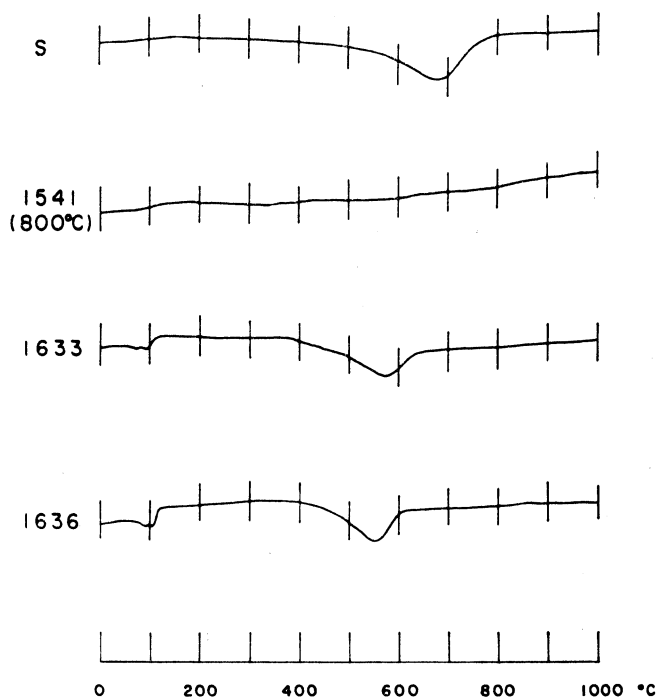


Fig. 10. Differential thermal analysis curves of unheated, heated sericite and synthesized specimens.

s: unheated sericite, 1541: sericite heated to 800°C for 1 hr., 1633: specimen synthesized from specimen 1541 by treatment with 0.8N H_2SO_4 for 10 minutes. 1636: specimen synthesized from specimen 1541 by treatment with 0.8N H_2SO_4 for 4 hrs.

As the endothermic peak at about 680°C in the sericite is due to the dehydroxylation, the reappearance of the endothermic peak in the interstratified minerals means that rehydroxylation occurred when the heated sericite was boiled with sulphuric acid. Endothermic peaks at about 100°C of specimens 1633 and 1636 show a doublet due to the dehydration of interlayer water and adsorbed water. The first endothermic peak is due to the dehydration of adsorbed water and the second one of the doublet is due to the dehydration of interlayer water accommodated to exchangeable cations.

3. Infrared absorption spectra

Infrared absorption spectra of interstratified minerals synthesized from pre-heated sericite by boiling for various periods of time are shown in Fig. 11, together with those of the original sericite and the sericite heated at 800°C for one hour. In the heated sample, absorption bands related to the hydroxyl groups disappeared. This fact agrees with the experimental results of Serratosa (1960), Grim and Kulbicki (1961) and Heller et al. (1962). Specimen 1633 showed absorption bands at about 3640 cm^{-1} and 1640 cm^{-1} , which are due to interlayer water and adsorbed water respectively. There

are no differences in the infrared absorption spectra of the interstratified minerals synthesized with different periods of boiling. Rehydration and rehydroxylation in heated sericite occurred when it was boiled in acid solution. These facts agree with the experimental result of differential thermal analysis.

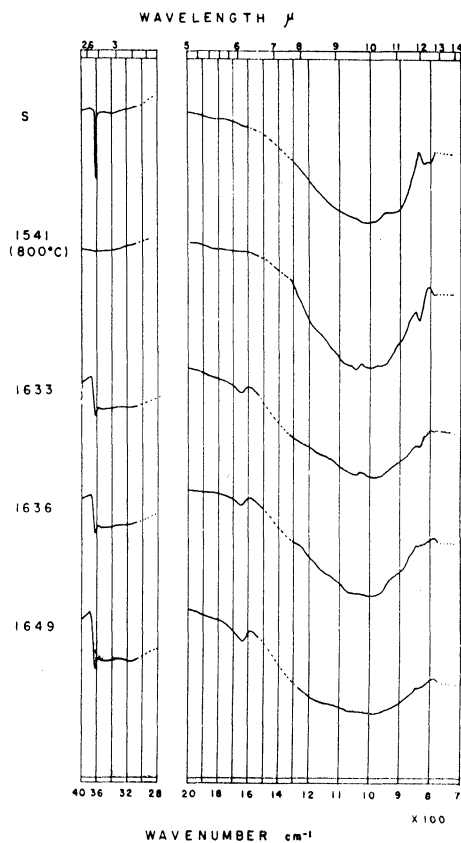


Fig. 11. Infrared absorption spectra of unheated, heated sericite and specimens synthesized from specimen 1541 by boiling it in 0.8N H_2SO_4 for various periods of time.

s: unheated sericite, 1541: sericite heated to 800°C, 1633: boiled for 10 minutes, 1636: boiled for 4 hours, 1649: boiled for 8 hours.

4. Electron microscopic observation

Electron micrographs of specimens 1612, 1633 and 1636 showed hexagonal shapes.

5. Fourier transform

Fourier transform method after MacEwan (1956) was employed to deduce the nature of the interstratification of the synthesized interstratified minerals. The equation employed in these calculations was formulated by MacEwan (1956) and can be written as

$$W_{(R)} = \sum_R \frac{I}{E |F_l|^2} \cos 2\pi \mu_R R$$

where E , $|F_l|^2$ and μ_R are values at the position of the intensity maximum, and I is

the integrated intensity. μ_R is the reciprocal spacing, and the values for $|F_l|^2$ were estimated from the tabular data of Cole and Lancucki (1966). The $|F_l|^2$ values of dioctahedral mica type layer with IK^+ and IH_2O in interlayers were used for the fourier transform. Such structure is considered to be similar to the interstratified minerals. $\frac{1 + \cos^2 2\theta}{\sin 2\theta}$ was used for the angular factor, E . Figure 12 shows the fourier transform

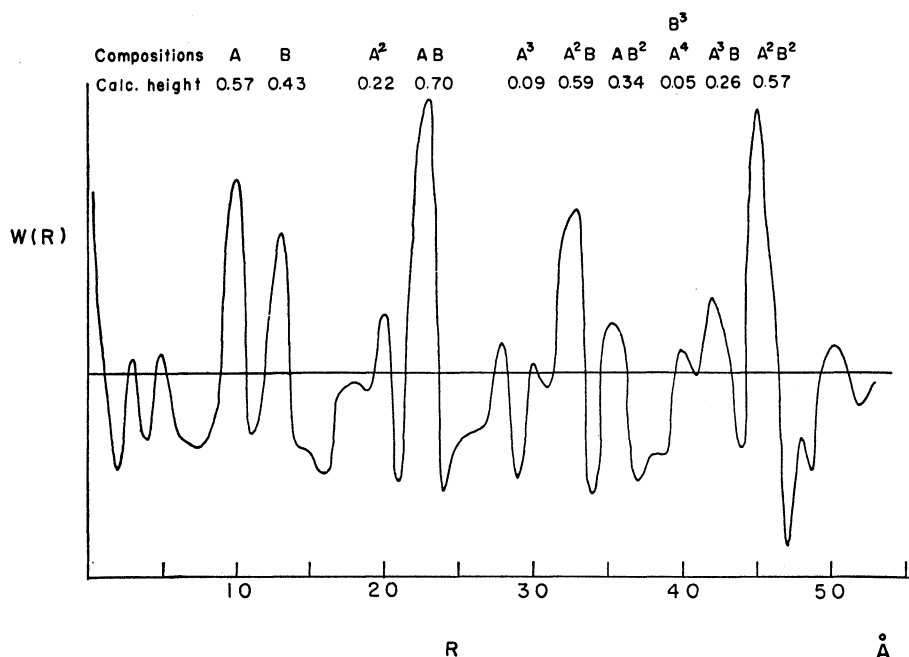


Fig. 12. Fourier transform of basal reflections of specimen 1612 which was synthesized from sericite heated to 800°C for 1 hour by boiling it in 0.8N H_2SO_4 for 30 minutes.

of basal reflections of specimen 1612. In the figure, A represents mica layer and B represents hydrated mica layer having a single layer of water molecules. The outstanding peaks of type AB, ABAB indicate a marked tendency for alternation of two different layers. Abbreviations $A^2 = AA$, $A^3 = AAA$, etc, are used in the figure. Numerals given above the curve are calculated peak heights for $P_A = 0.57$, $P_B = 0.43$, $P_{AA} = 0.39$, $P_{AB} = 0.61$, $P_{BA} = 0.81$, $P_{BB} = 0.19$, where A: mica layer, B: hydrated mica layer. Calculated and observed peak heights on the fourier transform from specimen 1612 are listed in Table 9. Calculated relative peak heights agree with the observed peak heights. P_A represents the frequency of occurrence of A, and P_B that of B. P_{AB} is the probability that B succeeds A, assuming that the first layer is A; P_{AA} , P_{BB} , P_{BA} are similarly defined. Figure 13 shows the fourier transform of basal reflections of specimen 1649. Peak heights above the curve were calculated from $P_A = 0.55$, $P_B = 0.45$, $P_{AA} = 0.27$, $P_{AB} = 0.73$, $P_{BA} = 0.89$, $P_{BB} = 0.11$, where A: mica layer, B: hydrated mica layer. Observed and calculated peak heights of the fourier transform from specimen 1649 are listed in Table 9. Results of specimens 1612, 1636 and 1649 are plotted in Fig. 14. The figure is based on a graph proposed by Sato (1965). In the figure, $P_{AA} = \alpha$, $P_{AB} = 1 - \alpha$, P_{BB}

$=\beta$, $P_{BA}=1-\beta$, $\beta=K\alpha+(1-K)$, $K=P_A/P_B$ were used.

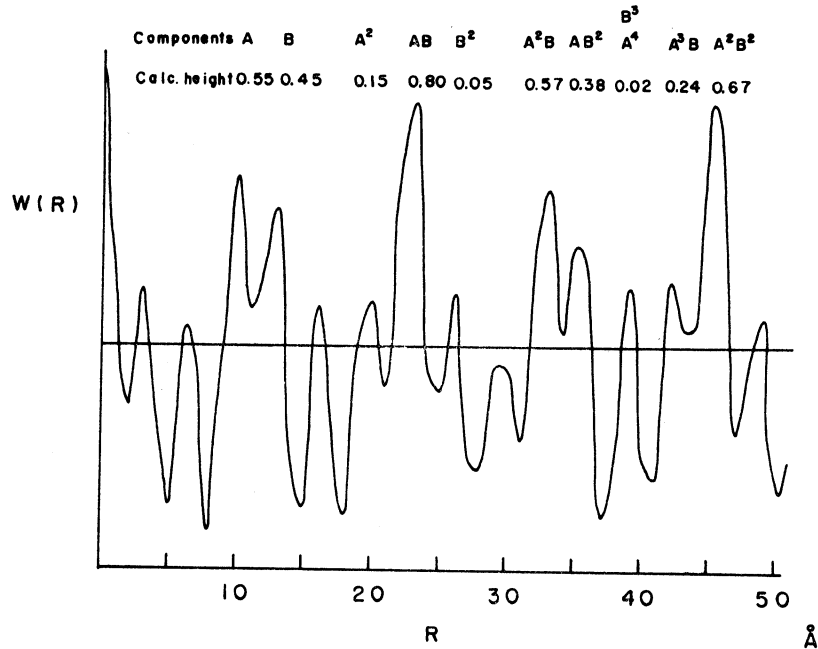


Fig. 13. Fourier transform of basal reflections of specimen 1649 which was synthesized from sericite heated to 800°C for 1 hour by boiling it in 0.8N H₂SO₄ for 8 hours.

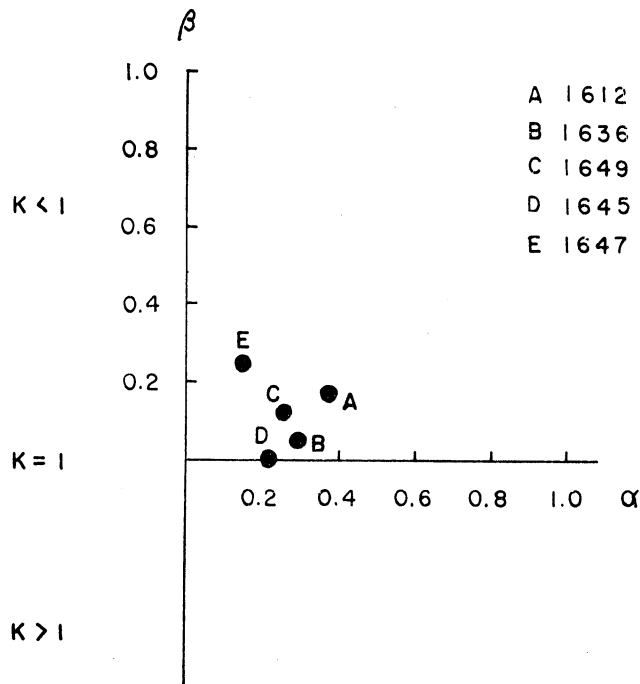


Fig. 14. Results of fourier transform for some synthesized specimens, where $P_{AA}=\alpha$, $P_{BB}=\beta$, $K=P_A/P_B$.

Table 9. Observed and calculated heights of peaks from fourier transform of the specimens 1612 and 1649.

Peak	1612		1649	
	Calc. rel. heights	Obs. rel. heights	Calc. rel. heights	Obs. rel. heights
A	0.57	0.57	0.55	0.55
B	0.43	0.43	0.45	0.45
AA	0.22	0.22	0.15	0.15
AB	0.70	0.77	0.80	0.80
AAA	0.09	0.10	0.04	0.00
BB	0.08	0.00	0.05	0.17
AAB	0.59	0.49	0.57	0.52
ABB	0.34	0.20	0.38	0.33
BBB	0.01	0.00		
AAAA	0.04	0.10	0.02	0.19
AAAB	0.26	0.32	0.24	0.21
ABAB	0.57	0.74	0.67	0.80

6. Calculation of mixing function

To obtain diffraction intensities from mixed-layer structure, mixing function Φ has to be multiplied by the squared modulus of the layer-structure factor F_l and by the appropriate angle-dependent factor E :

$$I = E |F_l|^2 \Phi$$

Φ and $|F_l|^2$ are functions of the reciprocal spacing $r^* = 2\sin\theta/\lambda$, where E is a function of θ . Methods of calculating Φ have been described by Hendricks and Teller (1942), by Mèring (1949), and by MacEwan (1958). MacEwan's method was employed in this paper. MacEwan's method consists of the direct calculation of the fourier transform for the given distribution, i. e. of the function

$$\Phi(r^*) = \text{Constant} \times \sum_{\theta^n}^N \sigma(R_n) \cos 2\pi r^* R_n$$

where $\sigma(R_n)$ is the frequency of the occurrence in the crystallites of the interlayer distance R_n ; N is the total number of layers per particle. Mixing function was calculated from observed data of the fourier transform, and the nature of mixing in the synthesized interstratified minerals was investigated. As the calculated intensity using a mixing function which was calculated using four as the number of layers per particle agreed with the observed intensity, this paper used four as the number of layers per particle.

Mixing function of specimen 1612 is given in Fig. 15. The mixing function was calculated from the following data; $A = 10 \text{ \AA}$, $B = 12.5 \text{ \AA}$, $P_A = 0.57$, $P_B = 0.43$, $P_{AA} = 0.39$, $P_{AB} = 0.61$, $P_{BA} = 0.81$, $P_{BB} = 0.19$, $N = 4$, where N is the number of layers per particle. The curve shows the effect of a tendency towards alternation of layers. Mixing functions of specimen 1636 and specimen 1649 are also shown in Fig. 15. The

mixing function of specimen 1636 is for $P_A=0.57$, $P_B=0.43$, $P_{AA}=0.30$, $P_{AB}=0.70$, $P_{BA}=0.93$, $P_{BB}=0.07$, $N=4$, where $A=10 \text{ \AA}$, $B=12.5 \text{ \AA}$. The mixing function of specimen 1649 is for $P_A=0.55$, $P_B=0.45$, $P_{AA}=0.27$, $P_{AB}=0.73$, $P_{BA}=0.89$, $P_{BB}=0.11$, $N=4$, where $A=10 \text{ \AA}$, $B=12.5 \text{ \AA}$. The three curves are similar and they show regular series of orders of a 22.5 \AA .

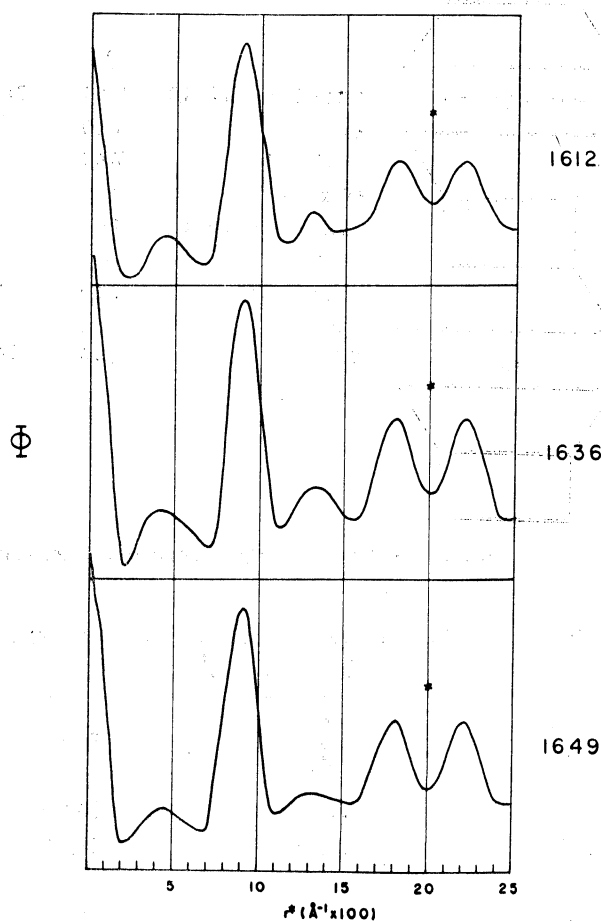


Fig. 15. Mixing functions of specimens 1612, 1636 and 1649.
* indicates center of symmetry.

7. Fourier syntheses of electron density normal to the basal plane

7-a. Fourier synthesis of air-dried sample

In this experiment, the authors could not succeed in forming a completely 1:1 regularly interstratified mineral from a sample heated at 800°C , but succeeded in forming such a mineral from a sericite heated at 650°C for one hour. Fourier synthesis of electron density normal to the basal plane for sample (specimen 1607) was carried out. Structure factors were derived from the integrated intensities by correcting for Lorentz-polarization factors and using the expression appropriate to x -ray reflection by an extended mosaic crystal. Their signs were determined by assuming that the structure consists of pairs of mica-type layers bound together by some of the cations K, Na, and

Ca, and with water layers separating the pairs of bound mica layers. The best layer parameters were then determined by different syntheses based on the chemical analysis data of specimen 1497. Calculated structure factors were obtained from a model shown in Fig. 16. Table 10 lists the basal spacings of the sample (specimen 1607) and the

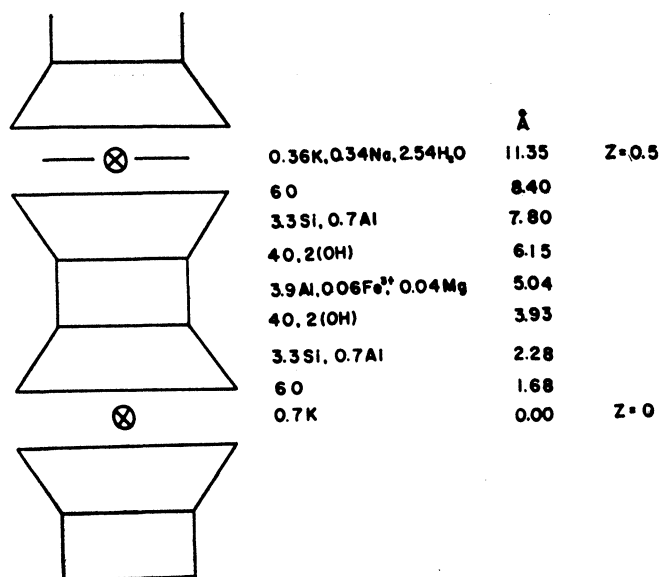


Fig. 16. z parameters of atomic layers in regularly interstratified mica-hydrated mica mineral.

Table 10. X-ray diffraction data for specimen 1607.
 $\lambda(\text{CuK}\alpha) = 1.5418 \text{ \AA}$

(00l)	d(00l) Å	d(001) Å	F _{obs}	F _{calc}
1	22.7	22.7	108.5	96.4
2	11.33	22.66	217.0	-152.7
3				9.2
4	5.64	22.56	90.4	-87.2
5	4.53	22.65	57.0	-69.5
6	3.754	22.53	58.8	-34.6
7	3.243	22.70	217.0	-223.5
8	2.823	22.58	48.0	65.0
9	2.522	22.70	46.1	-24.1
10				34.1
11	2.045	22.50	153.7	161.0
12	1.882	22.58	67.0	118.2
13				-18.1
14				-2.6
15	1.503	22.55	56.0	-73.1
16	1.410	22.56	67.0	-94.1
17	1.356	23.05	64.2	74.8
18	1.256	22.61	108.5	84.4

mean d(001) = 22.64 Å

observed and the calculated structure factors. Figure 17 shows the electron density distribution derived from the observed structure factors. The agreement between the observed and the calculated structure factors suggests the existence of potassium ion at $z=0$ and $z=0.5$. One layer of water molecules between the silicate layers was confirmed by fourier synthesis.

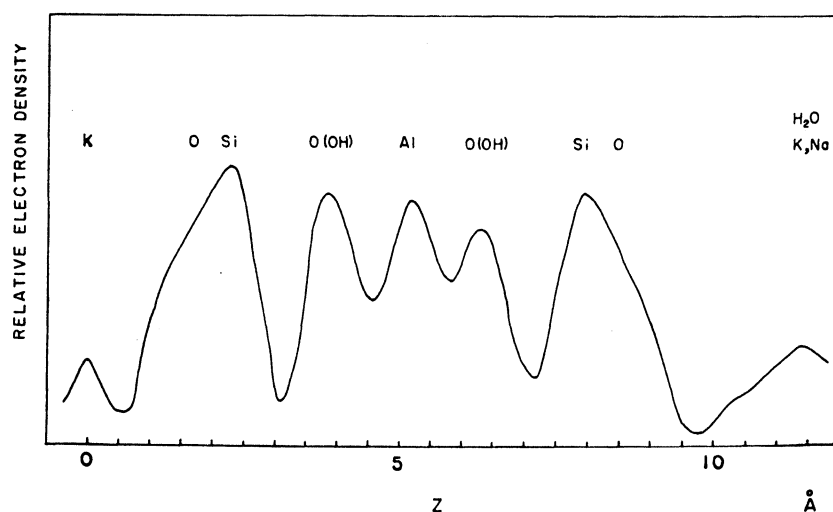


Fig. 17. One dimensional electron density distribution of specimen 1607.

7-b. Fourier syntheses of heated interstratified minerals

Specimens 1633, 1612 and 1649 were heated at 500°C for one hour, and structure factors for the heated specimens were derived from the integrated intensities by correcting for Lorentz-polarization factors. As their x -ray diffraction patterns show mica patterns, their signs were determined by assuming a mica structure. Lattice spacings and observed structure factors for the three heated samples are tabulated in Table 11.

Table 11. X -ray diffraction data for the specimens 1633, 1612 and 1649 after heating to 500°C.

$$\lambda(\text{CuK}\alpha) = 1.5418 \text{ \AA}$$

(00l)	1633		1612		1649	
	(500°C)		(500°C)		(500°C)	
	d (Å)	F _{obs}	d (Å)	F _{obs}	d (Å)	F _{obs}
2	10.05	11.83	10.16	11.11	10.16	11.38
4	5.01	10.62	5.04	11.52	5.04	12.76
6	3.34	21.83	3.35	20.60	3.35	24.40
8	2.51	7.16	2.515	8.48	2.515	8.74
10	2.01	10.92	2.01	10.40	2.01	10.51
12	1.664	6.53	1.667	7.93	1.661	5.46
14	1.439	5.06	1.441	5.15	1.437	4.22
16	1.259	6.07	1.261	4.74	1.259	2.47

The electron density distributions of the heated samples of specimens 1633 and 1649 are shown in Fig. 18. Electron density curve of specimen 1612 is similar to that of specimen 1633, but diminution of the O(OH) peak is observed in specimen 1649 as compared with that of specimen 1633. Their electron density curve are similar to that of a sericite heated at 500°C. This fact confirms the existence of potassium ion in every interlayer of the synthesized minerals.

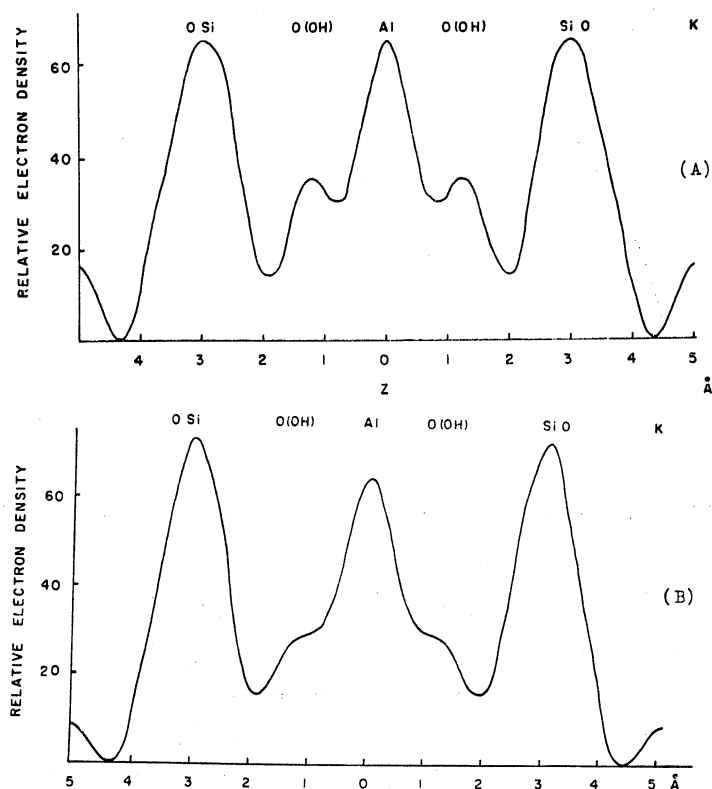


Fig. 18. One dimensional electron density distributions of heated samples of specimens 1633 and 1649.

(A). specimen 1633, (B). specimen 1649.

Effects of pre-heating period on properties of synthesized specimens

As it was confirmed that an interstratified mineral was formed from a pre-heated sericite by sulphuric acid treatment, effect of period of pre-heating time was investigated.

1. X-ray analysis

Figure 19 shows x -ray diffraction patterns of specimens which were synthesized from specimens heated at 800°C for various periods of heating time. The samples were synthesized by boiling in sulphuric acid solution (0.8N) for constant two hours. The x -ray powder pattern of specimen 1645 which was synthesized from a sericite heated at 800°C for 30 minutes shows a strong 23.3 Å reflection and a 11.25 Å reflection. A 3.23 Å peak corresponding to (007) reflection of the 23.3 Å reflection is strong. In this pattern, a peak corresponding to (004) reflection of the 23.3 Å reflection is clearly

visible. The 23.3 Å peak was replaced by a 24.5 Å peak by ethylene glycol treatment, and it contracted to 21.5 Å by heating at 300°C for one hour. The 21.5 Å peak was not changed by heating at 500°C for one hour, but it contracted to 10.16 Å by heating at 800°C for one hour. The diffraction pattern of the sample heated at 800°C showed

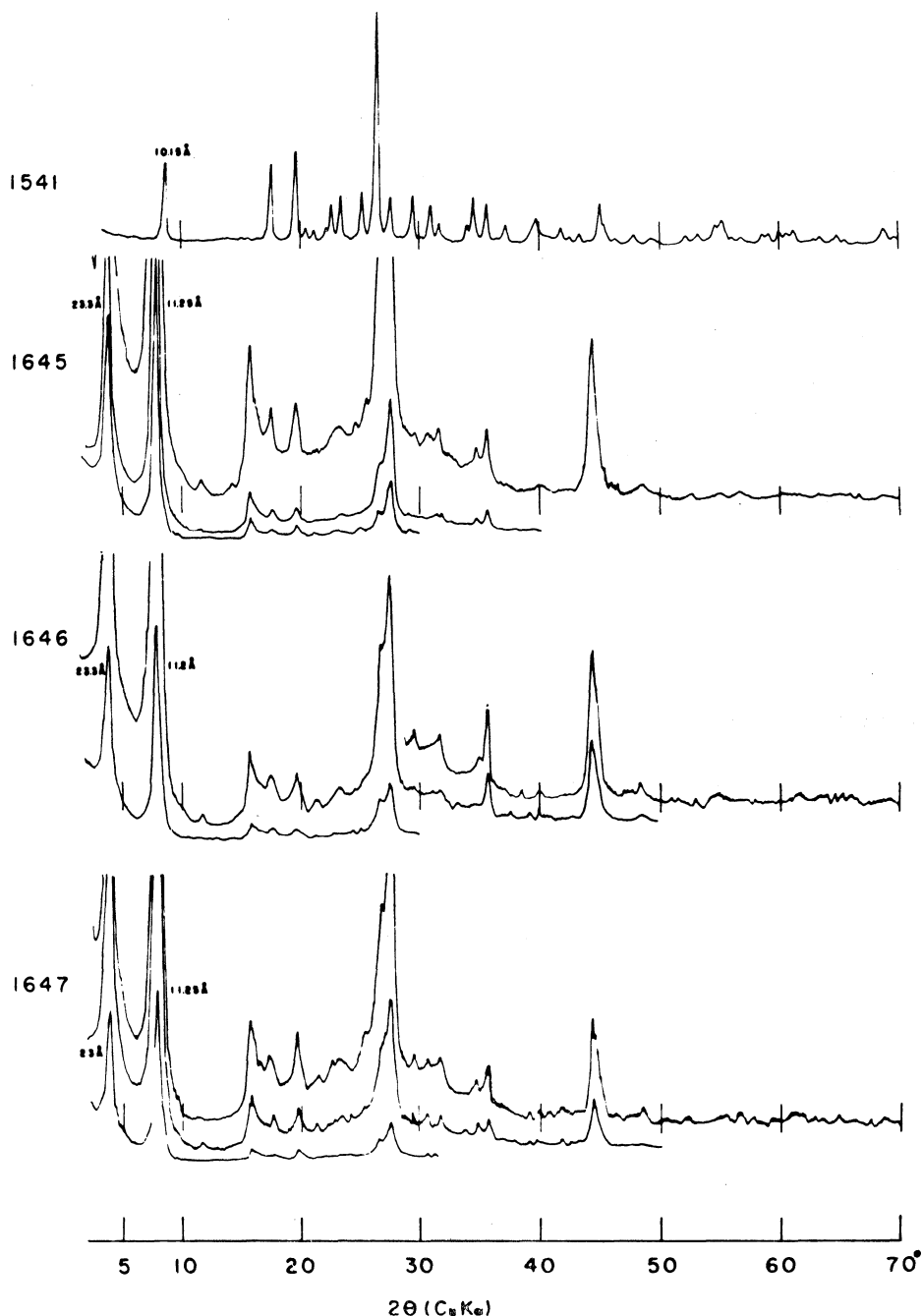


Fig. 19. X-ray diffraction patterns of sericite heated to 800°C and specimens synthesized by boiling specimens heated for various periods of time in 0.8N H_2SO_4 for 2 hours. 1541: sericite heated to 800°C, 1645: specimen synthesized from sericite by 2 hours' boiling after 30 minutes' heating, 1646: 3 hours' pre-heating time, 1647: 4 hours' pre-heating time.

a mica pattern. A small amount of mullite was formed from the specimen at 1050°C. The diffraction pattern of specimen 1646 which was synthesized from a sample heated for three hours shows also strong peaks of 23.3 Å and 11.2 Å. Powder pattern of specimen 1647, which was synthesized from a sericite heated for four hours, is similar to that of specimen 1646. A 23 Å peak moved to 24.5 Å by treatment with ethylene glycol and it contracted to 21 Å in heating at 300°C for one hour. The 21 Å peak disappeared in heating at 500°C for 1 hour. Mullite was formed at 1050°C from the specimen 1647. The powder data for specimens 1645 and 1647 are listed in Table 12.

Table 12. X-ray diffraction data for the specimens 1645 and 1647.
 $\lambda(\text{CuK}\alpha) = 1.5418 \text{ \AA}$

1645		E G		1647		E G	
d(Å)	I	d(Å)	I	d(Å)	I	d(Å)	I
23.3	679.0	24.5	18.9	23.0	157.5	24.5	21.0
11.25	1000.0	11.9	30.5	11.25	218.5	11.8	22.0
7.6	2.6						
5.61	57.0	5.83	1.0	5.6	10.0	5.87	1.0
4.53	6.5	4.70	3.0	4.5	3.5	4.72	3.0
3.23	258.0	3.35	21.8*	3.24	51.0	3.35	18.0*
2.51	5.0	2.58	1.7	2.51	1.5	2.58	3.5
2.04	71.5*			2.04	11.5*	2.113	1.5
1.88	1.0			1.877	1.0	1.949	1.5
1.25	7.5						

* reflections overlapped by mica reflections.

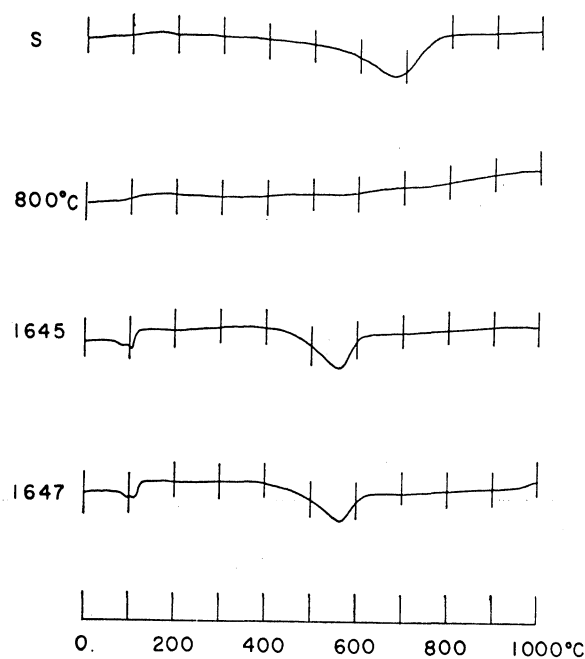


Fig. 20. Differential thermal analysis curves of unheated, heated sericite and synthesized specimens.

s: unheated sericite.

2. Differential thermal analysis

Differential thermal analysis curves of specimens 1645 and 1647 are shown in Fig. 20, together with those of the original sericite and the sericite heated at 800°C. No endothermic peaks are observed in the heated sericite. In the curve of specimen 1645, an endothermic peak at about 560°C and double endothermic peaks are observed. The double endothermic peaks are due to the dehydration of adsorbed free water and inter-layer water. The endothermic peak at about 560°C is due to the dehydroxylation of structure water. The curve of specimen 1647 shows a similar pattern to that of specimen 1645. Rehydration and rehydroxylation occurred in the heated specimen.

3. Infrared absorption spectra

Two specimens, 1645 and 1647, were studied by infrared spectroscopy. Their infrared absorption spectra are similar to those of specimens 1633 and 1636. In specimen 1645, double absorption bands in the range of 800–850 cm^{-1} , characteristic to regularly interstratified mineral (mica-montmorillonite type) as reported by Oinuma and Hayashi (1965), were clearly observed.

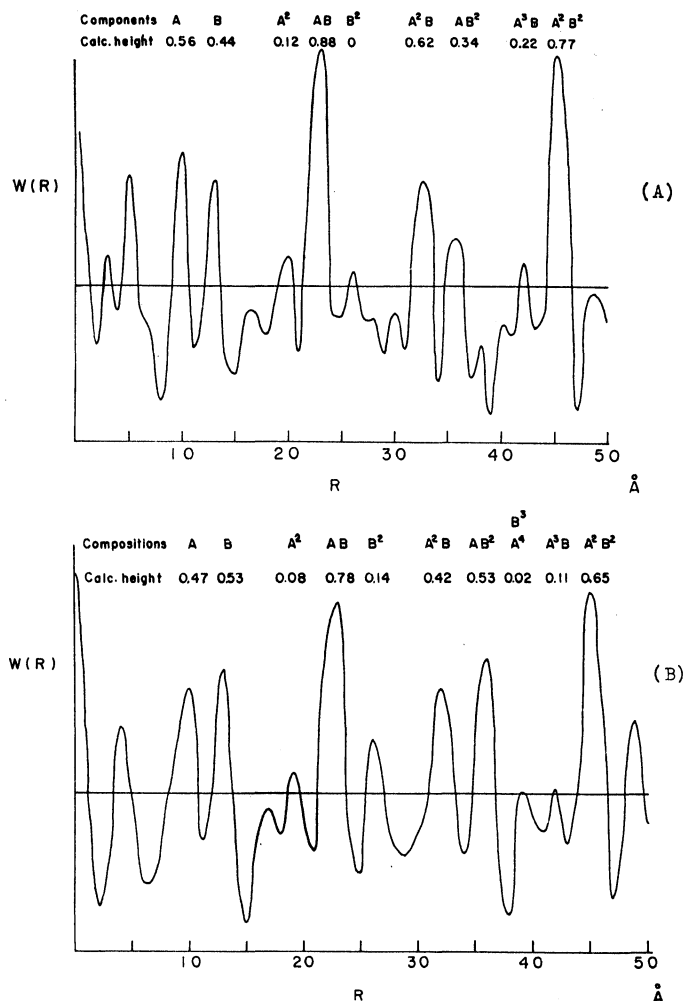


Fig. 21. Fourier transform of basal reflections of specimens 1645 and 1647. (A). specimen 1645, (B). specimen 1647.

4. Fourier transform

Fourier transform method was employed to investigate the nature of interstratification of specimens 1645 and 1647. The method is the same as mentioned in the preceding section. Figure 21 shows the fourier transform of basal reflections of specimens 1645 and 1647. Numerals given above the curve of the specimen 1645 are calculated peak heights for $P_A=0.56$, $P_B=0.44$, $P_{AA}=0.21$, $P_{AB}=0.79$, $P_{BA}=1$, $P_{BB}=0$, where A: mica layer and B: hydrated mica layer. Calculated peak heights given above the curve of specimen 1647 are for $P_A=0.47$, $P_B=0.53$, $P_{AA}=0.17$, $P_{AB}=0.83$, $P_{BA}=0.74$, $P_{BB}=0.26$, where A: mica layer, B: hydrated mica layer. Calculated and observed peak heights on the fourier transform from the both specimens are listed in Table 13. In specimen

Table 13. Observed and calculated heights of peaks from fourier transform of the specimens 1645 and 1647.

peak	1645		1647	
	Calc. rel. heights	Obs. rel. heights	Calc. rel. heights	Obs. rel. heights
A	0.56	0.56	0.47	0.47
B	0.44	0.44	0.53	0.53
AA	0.12	0.12	0.08	0.08
AB	0.88	0.98	0.78	0.83
BB	0.00	0.07	0.14	0.23
AAA	0.01	0.00	0.00	0.00
AAB	0.62	0.44	0.42	0.46
ABB	0.34	0.20	0.53	0.58
BBB	0.00	0.00	0.01	0.00
AAAA	0.00	0.00	0.00	0.00
AAAB	0.22	0.10	0.11	0.02
ABAB	0.77	0.96	0.65	0.88

1647, frequency of occurrence of hydrated mica layer increased as compared with that of specimen 1645. Results of the fourier transform for the two specimens are plotted in Fig. 14.

(D). Synthesis of an interstratified mineral having double layers of water molecules between silicate layers

Minerals synthesized by treatment with acids were always of the nearly regularly interstratified mineral of mica and hydrated mica having a single layer of water molecules between silicate layers. A specimen synthesized by sulphuric acid treatment was boiled in a solution of magnesium chloride or calcium chloride. In this experiment, 1.2% solution of magnesium chloride or calcium chloride was used. Table 14 shows diffraction data of specimen 1655 synthesized from specimen 1634, which was synthesized from a sericite heated at 800°C for 1 hour by boiling it in 0.8N H₂SO₄ for 1 hour, by boiling it in 1.2% solution of MgCl₂ for 6 hours. The diffraction pattern of specimen

1655 shows a 25.6 Å and a 12.5 Å reflections. The 25.6 Å peak expanded to 27.2 Å when treated with ethylene glycol, and the 12.5 Å peak to 13.6 Å. The 25.6 Å peak contracted to 20.5 Å after heating to 300°C for 1 hour, and the 20.5 Å peak contracted to 10.16 Å when heated at 500°C for 1 hour. Specimen 1656 which was synthesized from specimen 1634 by boiling it in 1.2% solution of CaCl₂ for 6 hours shows a 25 Å reflection in its powder pattern, and it expanded to 27.6 Å when treated with ethylene glycol. An interstratified mineral was formed from specimen 1634 by boiling it in 1.2% solution for 1 hour, but any interstratified minerals could not be formed from a sericite heated at 800°C by treatment with MgCl₂ or CaCl₂ solution.

Table 14. X-ray diffraction data for specimen 1655 after various treatments
λ(CuKα)=1.5418 Å

Untreated		E G		500°C	
d(Å)	I	d(Å)	I	d(Å)	I
25.6	115.0	27.2	108		
12.5	86.0	13.6	32	10.16	31.0
		9.1	4		
5.07	29.0	5.44	1	5.04	26.0
3.62	2.0				
3.20	1.3			3.35	84.0
2.823	1.0			2.52	9.0
		1.953	3	2.014	13.0
1.678	1.5			1.672	2.0
1.439	3.0			1.437	1.5
				1.259	1.0

EG: treated with ethylene glycol.

C. Factors for forming an interstratified mineral from sericite and some consideration of forming an interstratified mineral

In this study, an interstratified mineral was formed from a pre-heated mica by treatment with acids. So far as the pre-heating temperature is in the range between 650°C and 1000°C, an interstratified mineral could be formed from the pre-heated sericite by boiling it in an acid solution. Duration of pre-heating time did not give any serious effects on forming an interstratified mineral. In this experiment, sericite should be heated up to the dehydroxylation temperature in order to change it into an interstratified structure. It suggests that a pre-state of an interstratified structure was formed in the process of dehydroxylation. The present writers used a flake of muscovite (2M₁ type) for an experiment in order to investigate the structural change after heating at dehydroxylation temperature range. Laue photograph radiated vertical to a cleavage plane of mica flake heated at 800°C showed radial streaks, whereas that of the unheated specimen showed clear hexagonal spots. The radial streaks indicate some stacking disorder in the direction of *c**. Existence of stacking disorder in heated mica means that some

potassium ions in interlayers of heated mica came to be leached out in the chemical treatment more easily than those in unheated mica.

Any interstratified minerals could not be formed from a pre-heated sericite by boiling it in 1.2% $MgCl_2$ or 1.2% $CaCl_2$ solution, instead of an acid solution, for 6 hours. By boiling a pre-heated sericite in water, no interstratified minerals were formed. Pre-heated sericite should be boiled in acids in order to change it into an interstratified mineral. An interstratified mineral was formed even when the pre-heated sample was boiled in an acid solution for a few minutes. An interstratified mineral was formed from the pre-heated sericite by treatment with acids regardless of duration of boiling, and the longer the time of the treatment with acids, the larger amount of the interstratified mineral was formed. Acids play an important role in forming an interstratified structure from pre-heated sericite. Effects on the expanding properties of layer minerals will depend on the way the tetrahedral and octahedral substitutions occurred in the minerals. It is probable that silicate sheets are differences in composition. An asymmetric distribution of layer charge may account for various regular interstratifications. The general idea of a polar charge distribution to explain regular interstratification in clays appears to have been noted first by Sudo, Hayashi and Shimoda (1962). In most montmorillonite minerals, random interstratification was caused by treatment with ethylene glycol as was studied by Green-Kelly (1955), and Tettenhorst and Johns (1963). They suggested that it was due random distribution of layer charge. Formation of regularly interstratified structure is considered to be due to alternate leaching of potassium ions from heated mica by boiling it in acid solution. In this experiment, it is impossible to consider that an interstratified mineral was recrystallized from a decomposed state.

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