Regularly Interstratified Clay Minerals of Chlorite and Saponite ("Corrensite") in the Miocene Green Tuff Formation in Japan

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Abstract

Regularly interstratified clay mineral of chlorite and saponite ("corrensite") is widely found in the Miocene basic pyroclastic sediments in Japan. X-ray analysis reveals that the mode of piling of elementary layer lattices is nearly regular and the probability of finding chlorite layer varies between 0.70 and 0.50. X-ray powder diffraction pattern between 35° and 55° ($2\theta FeK\alpha$) of the interstratified mineral can be divided into two types. Chemically the chlorite layer is close to diabantite whereas the saponite layer resembles a high aluminous saponite ("Al-saponite"). In view of the mode of occurrence, the interstratified mineral may have been formed progressively from iron-rich saponite in basic pyroclastic rocks. On the other hand, it is possible it was formed primarily by chemical reaction of interstitial water with volcanic materials in basic tuffaceous sandstone. Regional occurrence of the interstratified mineral suggests that the mineral is a stable alteration product in the Green Tuff area.

1. Introduction

Regularly interstratified clay mineral of chlorite and smectite (saponite) is commonly found in the basic pyroclastic sediments from the Miocene Green Tuff formation in Japan. Generally speaking, the interstratified mineral occurs on the border between two rock alteration zones, which are chlorite and montmorillonite (saponite) zones, and it is considered to be very significant for rock alteration zoning in the Green Tuff region.

The purpose of the present paper is to describe the mode of occurrence and some mineralogical data of the interstratified minerals of chlorite and saponite ("corrensite") from the selected localities.

2. Specimens

Five selected specimens were prepared for this study. In addition, the known Noto-specimen was investigated to compare with them.

(1) Specimen Y-20, (2) Specimen Y-27 (KIMBARA and Sudo, 1973)

The present both specimens were collected from the Miocene andesitic tuffaceous sandstone of the Kawanami formation, Yamanaka district, Ishikawa Prefecture. Specimen Y-20 fills interspace of green-coloured tuffaceous sandstone, and is closely associated with laumontite replacing plagioclase or filling the interspaces. A small amount of calcite is sporadically found in the interspace. Plagioclase crystals are suffered from weak albitization, but fresh pyroxene still remains. Specimen Y-27 accompanied by a small amount of prehnite also fills matrix of brown-coloured andesitic tuffaceous sandstone. The brown-coloured appearance of the specimen is attributed to oxidation of ferrous iron caused by surface weathering.

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(3) Specimen 53013 (KIMBARA, 1975)

This specimen was found in amygdaloidal basalt belonging to the Miocene Sunakobuchi formation in the northeastern part of Akita city, Akita Prefecture. It shows a typical basaltic texture and the interstratified mineral in question occurs as aggregate of fine radial crystals filling spheroidal amygdales and replacing basic glass in the groundmass. Mafic minerals such as pyroxene and olivine had been completely replaced by the interstratified mineral. Plagioclase laths are suffered from weak albitization. Leucoxene is scattered in the groundmass.

(4) Specimen H-45 (KIMBARA, 1973)

The specimen H-45 was found in basaltic tuff breccia of the Miocene Karasawagawa formation exposed along the Hayato River in the eastern part of the Tanzawa Mountains, Kanagawa Prefecture. Under the microscope, it shows dusty green-colour, and it replaces basic glass and fills small cavities. It is associated with a large amount of prehnite. In places, it shows brown-coloured appearance due to oxidation of ferrous iron.

(5) Specimen 72041 (KIMBARA et al., 1971)

The present specimen was collected from the Miocene Kanazawa formation, Yamakata district, Ibaragi Prefecture. It replaces ferromagnesian minerals, probably pyroxene, and in places fills amygdales of andesitic tuff breccia. A small amount of calcite and magnetite occurs in the groundmass.

(6) Noto-specimen (collected by K. Shimosaka, Geological Survey of Japan)

The Noto-specimen was found in the hydrothemal altered aureole of Miocene gypsum deposits, Noto mine, Ishikawa Prefecture. Sugiura (1961) had reported in detail the mineralogical data of specimen from the same mine and identified it is an interstratified chlorite-vermiculite. But the writer presents some additional mineralogical data on the Noto-specimen.

The photomicrographs of these specimens are shown in Plate 21.

3. X-ray analysis

X-ray diffraction data of natural specimen show clearly a reflection at 30Å and its higher orders. To clarify the thermal properties, the specimens were heated at 300°, 450°, 600°, 700° and 800°C for one hour and were sealed in a desicator for half one hour after cooling. After that, all of them were scanned using CuKα radiation. After heated at 300°C, the 30Å reflection disappeared and a reflection at 15Å shifted to 12–13Å, but the Noto-specimen showed strong ability of rehydration up to 450°C. A reflection of 24–23Å appeared after heated at 300–450°C and disappeared at 800°C. The 30Å reflection expanded to 32Å after treatment with ethylene glycol. After boiled with ammonium nitrate solution (1N) for ten minutes, the 30Å reflection was replaced by a 28Å one. Such experimental results as summarized in Table 1 strongly suggest that an expandable layer of the interstratified mineral is closer to montmorillonite-like mineral rather than vermiculite-like one. As given in X-ray powder diffraction data (Table 2), the (060) reflection at 1.54Å indicates a trioctahedral nature. Hence we can call the present all specimens including the Noto one as regularly interstratified mineral of chlorite and saponite.

However as seen in X-ray powder diffraction curves between 35°-55° ($2\theta \text{FeK}\alpha$) (Fig. 1), the diffraction pattern of specimen 53013 is closely related to the 20l reflections of IIb($\beta = 97^{\circ}$) chlorite, whereas that of specimen 72041 is similar to the 20l reflection of Ib($\beta = 90^{\circ}$) chlorite.

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Table 1 Average basal spacings (Å) before and after various treatments.

Condition Specimen	U.T.	300°C	450°C	600°C	700°C	800°C	E.G.	G. NH ₄ NO ₃	Mg ²⁺ (saturated) K ⁺ (satura U.T. E.G. G. U.T. E.G.	<u></u> '
Y-20	29.50	23.74	23.80	23.59	23.47	22.56	31.02	27.19		
Y-27	29.38	24.01	23.80	23.61	23.53	21.34	30.83	27.29		
53013	29.58	24.22	24.07	23.93			31.24	27.27		
H-45	29.38	24.28	24.15	24.17			30.95	27.37		
72041	29.55	24.18	23.81	23.85	_	_	31.42	27.37		
Noto	29.65	_	23.96	23.77			31.11	32.26	29.60 31.62 32.35 27.32 31.44	32.20
OF-1884A	29.32	23.96	23.48 (500°C)	23.74	23.73		31.11		27.18	

U.T.: natural state, 300°-800°C: after heated at each temperature for one hour,

E.G.: after treated with ethylene glycol, G.: after treated with glycerol,

NH₄NO₃: after treated with ammonium nitrate

OF-1884A (Yoshimura, 1971)

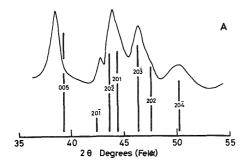
Table 2 X-ray powder diffraction data of regularly interstratified chlorite-saponite.

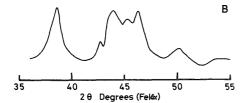
	Y-20		Y-27		53013		H-45		72041		Noto	
hkl	d(Å)	I	$d(\text{\AA})$	I	d(Å)	I	d(A)	Ι	$d(\text{\AA})$	I	d(Å)	I
001	31.7	53	30.8	49	31.7	53	30.8	52	32.6	47	30.4	53
002	15.00	100	14.80	100	14.80	100	14.80	100	15.00	100	14.72	100
003	9.746	12	9.812	12	9.832	14	9.832	16	9.832	14	9.817	7
004	7.319	34	7.367	35	7.367	29	7.367	30	7.367	39	7.369	16
005	5.896	5	5.896	7	5.896	5	5.927	7	5.927	7	5.901	3
006	4.897	11	4.876	16	4.897	17	4.897	18	4.876	17	4.924	15
02,11	4.599	11	4.618	14	4.599	17	4.618	21	4.581	24	4.572	13
007	4.239	5	4.239	5	4.239	5	4.087	4	_		4.247	5
800	3.668	17	3.657	20	3.668	22	3.668	19	3.657	24	3.690	12
009	3.264	9	3.246	14	3.255	9	3.264	11	3.219	17	3.288	9
0010	2.936	7	2.922	12	2.936	15	2.936	12	2.936	13	2.947	9
	2.659	3	2.653	5	2.653	6			2.653	9		
	2.584	7	2.584	13	2.578	16	2.584	15			2.562	13
	2.508	6							2.508	20		
	2.462	7	2.442	8	2.457	13	2.452	10			2.447	9
	2.278	2	2.282	4	2.282	5	2.273	5	2.295	4	2.279	5
	2.072	2	2.072	3	2.096	2	2.076	3	2.132	7	2.039	4
	2.000	2	2.013	3	2.025	3	2.013	4			1.913	3
	1.733	2	1.740	2			1.745	3	1.731	4	1.732	2
			1.705	2								
060	1.543	12	1.540	13	1.543	17	1.542	15	1.543	24	1.536	19
	1.512	2	1.511	4	1.512	4	1.515	3	1.507	5	1.507	3

Specimen Y-20 clearly gives an intermediate pattern between specimens 53013 and 72041. A structure study using a single crystal is actually required for further discussion.

4. Chemical analysis

Chemical compositions of the present six specimens together with the related minerals are





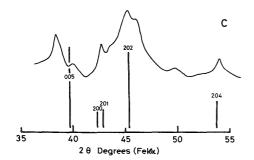


Fig. 1 X-ray powder diffraction pattern between 35° and 55° ($20FeK\alpha$)

- A. Specimen 53013 (Taiheizan, Akita Pref.) Indices are given as $IIb(\beta = 97^{\circ})$ chlorite.
- B. Specimen Y-20 (Yamanaka, Ishikawa Pref.)
- C. Specimen 72041 (Daigo, Ibaragi Pref.) Indices are given as $Ib(\beta = 90^{\circ})$ chlorite.

given in Table 3. Their structural formulae calculated on the basis of $O_{20}(OH)_{10}$ tentatively assuming to be 1:1 regular structure also are shown in the same table. The high contents of magnesium and iron suggest the presence of Fe-Mg or Mg-Fe chlorite and saponite as an elementary layer of the interstratified mineral. The total number of the cations in octahedral sites is less than 9. Sudo (1967) indicated that the cause of the deficiency of octahedral cations would be explained as an interstratified mineral having partly dioctahedral nature rather than completely trioctahedral one. However in the present case, it is also possible that the deficiency is caused by oxidation of ferrous iron. When the all ferric iron converts into ferrous one, the total number of octahedral cations becomes close to ideal number 9 though the deficiency still remains. The ratio of Si/Al in the tetrahedral sites is close to that of diabantite of chlorite group as well as saponite of smectite group. The cation exchange capacities determined by the Schollenberger's method are about 40–60 me/100g dried clay and their total exchangeable cations are about 30–60 me/100g dried clay for all specimens (Table 3). This values are close to that of the interstratified chlorite-vermiculite reported by Bradley and Weaver (1956). Calcium is predominant in the exchangeable cations.

Table 3 Chemical composition of interstratified minerals of chlorite and saponite.

	Y-20*	Y-27	53013	H-45	72041*	Noto	OF-1884A*	Furutobe
SiO ₂	33.57	31.91	35.73	34.72	35.59	34.65	37.16	35.26
TiO_2	0.59	0.77	0.11	0.57	0.30	0.34	0.34	0.96
Al_2O_3	13.60	14.63	14.05	13.94	15.13	16.09	14.98	16.02
$\mathrm{Fe_2O_3}$	8.38	16.45	7.17	11.45	6.65	1.61	12.01	6.66
FeO	11.31	4.41	7.22	4.14	9.15	1.30	4.11	8.19
MgO	12.93	11.56	18.84	16.61	14.76	20.97	14.79	14.61
MnO	0.26	0.25	0.11	0.20	0.38	0.07	0.13	
CaO	2.20	2.74	3.49	3.37	1.86	0.77	1.80	2.37
Na_2O	1.00	1.65	1.20	1.43	1.13	0.52	0.27	1.58
K_2O	0.80	0.65	0.15	0.30	1.13	0.10	0.88	0.21
$H_2O(+)$	9.68	10.47	7.80	8.34	7.30	10.88	5.12	8.48
$H_2O(-)$	5.68	4.90	4.09	5.69	6.62	6.58	8.41	4.92
Total (%)	100.00	100.39	99.56	100.76	100.00	99.88	100.00	99.21
CEC(me/100g)	53	45	39	84	56	51		
Ca	24	29	29	28	32	14		
Mg	4	3	5	5	1	1		
Na	3	3	4	3	5	15		
K	5	8	4	4	6	2		
Total (me/100g)	36	33	42	40	44	32		

 $[\]text{Y-20} \\ \hspace*{0.5in} : (Mg_{3.587}Fe^{3+}_{1.174}Fe^{2+}_{1.760}Al_{1.232}Mn_{0.041}) \\ (\text{Si}_{6.249}Al_{1.751}) \\ \hspace*{0.5in} (\text{Ca}_{0.438}\text{Na}_{0.361}K_{0.190}) \\ O_{20}(\text{OH})_{10}\text{nH}_2\text{O}(\text{OH})_{10} \\ \text{O}_{20}(\text{OH})_{10}\text{nH}_2\text{O}(\text{OH})_{10} \\ \text{O}_{20}(\text{OH})_{10} \\ \text{O}_{20}(\text{OH})_{10}$

5. Differential thermal analysis

DTA curves (Fig. 2) were recorded under heating rate of 5° or 10°C per minute. Thermal curves of the specimens resemble each other and accord well with those of interstratified minerals reported by Bradley and Weaver (1956) and Peterson (1961). Double endothermic reactions between 100° and 200°C are due to loss of interlayer water, and 5–6% loss of weight was detected in this temperature region. A clear endothermic reaction at 550°C is attributed to the dehydroxylation of brucite layer and this reaction is accompanied by about 6% weight loss. An endothermic reaction which is followed by an exothermic one between 800°–900°C is due to loss of hydroxyl water of silicate layer, and the weight loss is about 2%. This peak system is quite similar to that of ferromagnesian chlorite.

6. Discussion

The present five specimens from the Miocene Green Tuff formation are identified as regularly interstratification of chlorite and saponite. Chemically the chlorite layer is close to diabantite whereas the saponite layer resembles a high aluminous iron-rich saponite ("Al-saponite") which

 $Y-27 \qquad : (Mg_{2.180}Fe^{3+}_{2.284}Fe^{2+}_{0.681}Al_{1.069}Mn_{0.039}) (Si_{5.888}Al_{2.112}) \ (Ca_{0.542}Na_{0.590}K_{0.153})O_{20}(OH)_{10}nH_2OH_{20} + OH_{20}(OH)_{10}nH_2OH_{20} + OH_{20}(OH)_{10}nH_{20} + OH_{20}(OH)_{10}nH_2OH_{20} +$

 $[\]begin{array}{lll} & & & & & & \\ & & & & \\ & & & & \\ & & & \\ & & & \\ & & & \\ & & & \\ & & & \\ & & & \\ & & & \\ & & & \\ & & & \\ & & & \\ & & & \\ &$

 $[\]text{H-45} \qquad : \\ (\text{Mg}_{4.376}\text{Fe}^{3+}_{1.523}\text{Fe}^{2+}_{0.612}\text{Al}_{1.041}\text{Mn}_{0.030}) \\ (\text{Si}_{6.137}\text{Al}_{1.863}) \\ (\text{Ca}_{0.638}\text{Na}_{0.490}\text{K}_{0.068}) \\ \text{O}_{20}(\text{OH})_{10}\text{nH}_{2}\text{O}_{10} \\ \text{O}_{20}(\text{OH})_{10}\text{nH}_{20} \\ \text{O}_{20}(\text{OH})_{20}\text{nH}_{20} \\ \text{O}_{20}(\text{OH})_{20} \\ \text{O}_{20}(\text{OH})_{20}\text{nH}_{20} \\ \text{O}_{2$

 $[\]begin{array}{lll} 72041 & : (Mg_{3.893}Fe^{3+}_{0.895}Fe^{3+}_{1.868}Al_{1.563}Mn_{0.059})(Si_{6.371}Al_{1.629}) & (Ca_{6.857}Na_{6.391}K_{0.258})O_{26}(OH)_{10}nH_2O \\ Noto & : (Mg_{7.037}Fe^{3+}_{0.212}Fe^{2+}_{0.196}Al_{1.386}Mn_{0.010})(Si_{6.066}Al_{1.934}) & (Ca_{6.144}Na_{6.176}K_{0.022})O_{26}(OH)_{10}nH_2O \\ \end{array}$

 $OF-1884A \quad : (Mg_{3.79}Fe^{3+}_{1.58}Fe^{2+}_{0.62}Al_{1.61})(Si_{6.53}Al_{1.47})(Ca_{0.34}Na_{0.09}K_{0.19})O_{20}(OH)_{10}nH_2O \ \ (Yoshimura, 1971)$

Furutobe : $(Mg_{3.870}Fe^{2+}_{0.891}Fe^{2+}_{1.217}Al_{1.628})(Si_{0.270}Al_{1.730})(Ca_{0.452}Na_{0.545}K_{0.047})O_{20}(OH)_{10}nH_2O(Sudo, 1967)$

^{*} Recalculated value

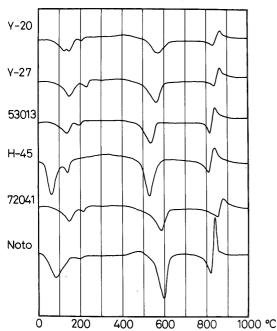


Fig. 2 DTA curves of interstratified minerals of chlorite and saponite

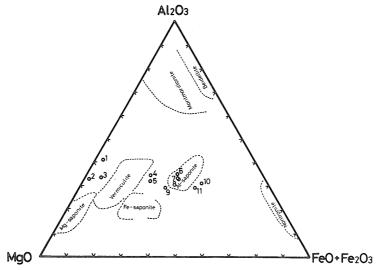


Fig. 3 A-F-M diagram showing the chemical composition of interstratified chlorite-saponite together with smectite and vermiculite

- 1: chlorite-vermiculite (Sugiura, 1962),
- 2: chlorite-montmorillonite (TAKAHASHI, 1959),
- 3: chlorite-vermiculite (BRADLEY and WEAVER, 1956),
- 4, 5: chlorite-vermiculite ("swelling chlorite") (Gallitelli, 1956),
- 6: 72041, 7: Furutobe (Sudo, 1967), 8: OF-1884A (Yoshimura, 1971),
- 9: 53013, 10: Y-27, 11: Y-20

is commonly found in the weakly altered pyroclastic sediments of Green Tuff formation (Fig. 3).

In order to obtain the mixing ratio of chlorite layer (14Å) and dehydrated saponite layer (10Å), a X-ray determinative curve was computed by Sato's method (1965). According to the

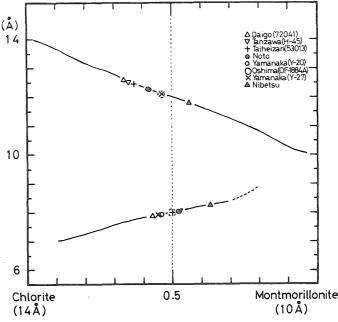


Fig. 4 X-ray determinative curve for mixing ratio of chlorite layer (14Å) and dehydrated montmorillonite (saponite) layer (10Å) of interstratified mineral

The specimen Nibetsu is a randomly interstratified mineral of chlorite and saponite (Kimbara and Shimoda, 1972)

Table 4 Mode of occurrence and associate minerals of interstratified chlorite-saponite in the Green Tuff formation.

1		Ze	olites			Clay minera		Others				
	anal.	heul.	laum.	wair.	chl.	mont.(sap.)	mica	preh.	pump.	epi.	cal.	
1	0		0		0	0	0				0	
2			0		0			0		0	0	
3	0		0		0	0	0	0	0			
4	0		0		0	0		0	0		0	
5	0		0	0	0		0		0			
6	0		0		0		0	0			0	
7					0						0	
8	0	0	0		0							

anal.: analcime, heul.: heulandite, laum.: laumontite, wair.: wairakite, chl.: chlorite, mont. (sap.): montmorillonite (saponite), preh.: prehnite, pump.: pumpellyite, epi.: epidote, cal.: calcite, \bigcirc : common, \circ : rare

A) in andesitic tuffaceous sandstone.

- 1: Oshima-Fukushima, Hokkaido (Yoshimura, 1971).
- 2: Yamanaka, Ishikawa Prefecture (KIMBARA and Sudo, 1973).
- B) in basaltic pyroclastic rocks.
 - 3: Ainai-Jimba, northern Akita (SHIMAZU and SATO, 1966).
 - 4: Tanzawa Mountains, Kanagawa Prefecture (Seki et al., 1969; Shimazu et al., 1971; Kimbara, 1973).
 - 5: Koma Mountains, Yamanashi Prefecture (Sugiyama, 1971).
- C) in andesitic pyroclastic rocks.
 - 6: Motojuku, Gumma Prefecture (Motojuku Research Group, 1970; Kimbara, 1970a, b).
 - 7: Daigo, Ibaragi Prefecture (SATO, 1970; KIMBARA et al., 1971).
 - 8: Taiheizan, Akita Prefecture (KIMBARA, 1970a).

curve (Fig. 4), the probability of finding chlorite layer of the interstratified minerals in question varies between 0.70 and 0.50. Both Noto-specimen and OF-1884A are considered to be nearly 1:1 regular interstratification.

The Noto-specimen has been reported as combination of chlorite and vermiculite by Sugiura (1961). However, it is very difficult to discriminate vermiculite from montmorillonite by treatment with ethylene glycol, for many vermiculite varieties, whose interlayer spaces are occupied with Ca, Rb and Sr, swell with ethylene glycol (Walker, 1957). When the basal spacing of magnesium saturated sample does not change after treatment with ethylene glycol, we can confirm a vermiculite character. The Noto-specimen after saturated Mg²⁺ expands after treatment with ethylene glycol (Table 1). The Noto-specimen, therefore, should be called as regularly interstratified mineral of chlorite and saponite.

Regional occurrence of interstratified chlorite-saponite in the Miocene Green Tuff formation reported by many geologists and petrologists and their mode of occurrences are summarized in Table 4. This table reveals that most of the interstratified minerals are commonly associated with laumontite. In basic pyroclastic rocks, they are associated with chlorite, saponite, prehnite and pumpellyite as well as laumontite. In the Tanzawa Mountains, the amount of iron-rich saponite accompanied by interstratified chlorite-saponite decreases with increasing alteration grade (Kimbara, 1973). This fact indicates that the interstratified chlorite-saponite seems to have been formed progressively from iron-rich saponite (probably "Al-saponite"). Thus in diagenetic condition, saponite would be gradually transformed into chlorite through interstratified chlorite-saponite with increasing alteration grade (or burial depth) as reported by Eckhardt (1958). This reaction may be expressed by the following chemical equation:

But in hydrothermal condition such as geothermal area, saponite or montmorillonite seems to be rapidly transformed into chlorite not through interstratified mineral. In tuffaceous sandstone, it is very rare that the interstratified chlorite-saponite occurs in close association with saponite or chlorite (Kimbara and Sudo, 1973). This fact shows that the interstratified mineral occurring in matrix of the sandstone seems to have been formed primarily by chemical reaction of interstitial water with volcanic materials. Wide occurrence of the interstratified mineral in the Green Tuff region may show that the mineral is a stable alteration product.

7. Occurrence of randomly interstratified chlorite-saponite

A randomly interstratified chlorite-saponite was also found in amygdales of pillow lava in the Miocene Sunakobuchi formation at Nibetsu, Akita Prefecture (Kimbara and Shimoda, 1972). It occurs in close association with iron-rich saponite. In the Green Tuff area, iron-rich saponite

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having an endothermic peak between 500°-600°C in its DTA curve occurs widely in basaltic sediments. Though further studies are actually required, it would be interpreted to be randomly interstratification with a small amount of chlorite.

Acknowledgment

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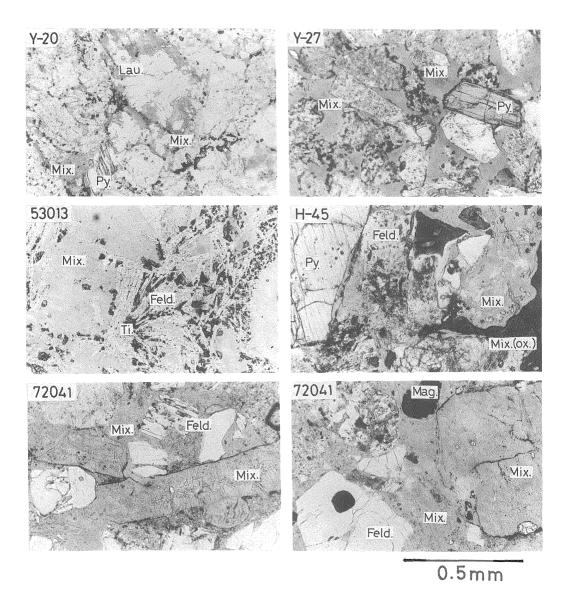
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新第三紀中新世グリンタフ層中の緑泥石-サポナイト規則型混合層鉱物

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グリンタフ層中より産出したいくつかの緑泥石一サポナイト混合層鉱物の鉱物学的性質を検討した. 定方位X線分析によれば混合層鉱物の緑泥石層の存在比は $0.5\sim0.7$ であり,いずれも規則型に近いものである.しかし $2\theta(\text{FeK}\alpha)=35°\sim55°$ のX線粉末回折パターンを検討すると,緑泥石の $\text{IIb}(\beta=97°)$ 型に近いタイプ, $\text{Ib}(\beta=90°)$ 型に近いタイプおよび両者が混合したタイプが認められる. 化学的にはいずれも鉄一マグネシウム質であり,構造式より推定される緑泥石層はダイアバンタイトに,またサポナイト層はアルミニウムに富む鉄サポナイトに近いことが予想される. グリンタフ層中では,この種の混合層鉱物は濁沸石などを伴って広域的に生じており,このことはこの鉱物が一つの安定な変質産物であることを示している.

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Photomicrographs showing the mode of occurrence of interstratified minerals

Py.: pyroxene, Feld.: feldspar, Mag.: magnetite, Lau.: laumontite, Mix.: interstratified mineral of chlorite and saponite, Mix.(ox.): oxidized interstratified mineral, Ti.: leucoxene