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THE "LAYER CHARGE" OF REGULAR INTERSTRATIFIED 2:1 CLAY MINERALS

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Abstract—Alkylammonium ion exchange on mixed-layer minerals gives detailed information about the variation of cation density in succeeding interlayer spaces. Three mixed layer samples from Japan (supplied by Dr. H. Kodama) were investigated. Their nearly 1:1 interstratification is based on regularly alternating high- and low-charged interlayer spaces, which are caused by a regular sequence of polar layers. The cation density in the high-charged interlayer spaces is >0.8 eq/(Si, Al)$_4O_{10}$. The low-charged interlayer spaces have an average cation density of 0.4 eq/(Si, Al)$_4O_{10}$ and heterogeneous charge distribution. The kind of heterogeneity of the Goto Mine sample differs from that of the Yonago Mine and the Honami Mine samples.

The Goto Mine specimen has a rather regular sequence of the low- and high-charged interlayers in proportions close to 0.50:0.50. The other two samples contain interlayer spaces with pronounced unsymmetrical charge distribution. The Yonago Mine sample probably has in random distribution with the polar layers about 10% mica-like layers segregated to packets of three and more layers; the ratio of high-charged interlayers to the low-charged ones is increased to about 0.55:0.45. The Honami Mine sample probably contains isolated mica-like layers or pairs of them. The proportion of the high-charged interlayers is estimated to be about 0.53:0.47 and is lower than determined by Kodama from the glycerolated sample.

The samples investigated may be considered as end-members of a series of interstratified specimens which begins with smectites with mixed-layer like charge distribution.

Key Words—Exchange, Hydrous Mica, Interstratification, Mica, Mixed-Layers, Smectite.

INTRODUCTION

Almost all natural smectitic clay minerals have a heterogeneous charge distribution: the cationic charge density in the interlayer spaces varies within the interlayer spaces and from interlayer to interlayer. Indications of charge heterogeneity were reported by Jonas and Roberson (1966), Byrne (1954), McAtee (1958a, b), Sudo et al. (1962), Tettenhorst and Johns (1966), and Clementz and Mortland (1974). A more quantitative picture could be obtained from the X-ray investigation of organic derivatives, especially the alkylammonium derivatives (Stul and Mortier, 1974; Lagaly and Weiss, 1976; Lagaly et al., 1976; Rengasamy et al., 1976).

The interlayer cation density of smectites varies mostly between 0.28 and 0.42 eq/(Si,Al)$_4O_{10}$ (Lagaly and Weiss, 1976). Only about 10% of 300 smectites investigated in our laboratory present a uniform distribution of the interlayer cation density (Figure 1a). About 50% are characterized by the type of heterogeneity illustrated in Figure 1b. The crystals are composed of differently charged layers so that most of the interlayer spaces, for example, have either 0.24–0.26 or 0.28–0.31 cations per unit: the smectites are mixed-layer minerals of relatively similar layers which differ little by the charge density or charge distribution. The charge distribution of an ideal mixed-layer mineral is shown in Figure 1c. In real cases, the charge density of the interlayers should vary around two values as a consequence of the usual charge heterogeneity in smectites. Figure 2 illustrates different cases of smectitic interstratification with more or less overlapping distribution curves; Figure 2e shows an extreme case with interlayer spaces of about 0.9–1.0 eq/(Si,Al)$_4O_{10}$ alternating with low-charged smectitic interlayers with heterogeneous charge distribution.

Alkylammonium ion exchange provides a simple method for analysis of charge heterogeneity in smectites and vermiculites (Lagaly and Weiss, 1976). Random interstratification of smectitic interlayer spaces with cation densities varying between two limits and of mica-like interlayers probably also with varying cation densities makes the interpretation of the X-ray pattern of the alkylammonium derivatives difficult, but regular interstratification allows an extensive interpretation of the X-ray diagrams. We thank Dr. H. Kodama (Soil Research Institute, Ottawa, Ontario, Canada), who supplied us with three samples of regular interstratified hydrous micas from Japan for investigation.

EXPERIMENTAL

Description of the samples

The hydrous micas from Japan supplied by Dr. H. Kodama are hydrothermal alteration products, which were collected at the Goto Mine in the Nagasaki Prefecture, Yonago Mine, and Honami Mine in the Nagano Prefecture. They were described in detail by Kodama et al. (1969).

According to the X-ray patterns of the air-dried samples and their glycerol complexes they are rectorite-like minerals with mica-like and smectitic or vermiculitic

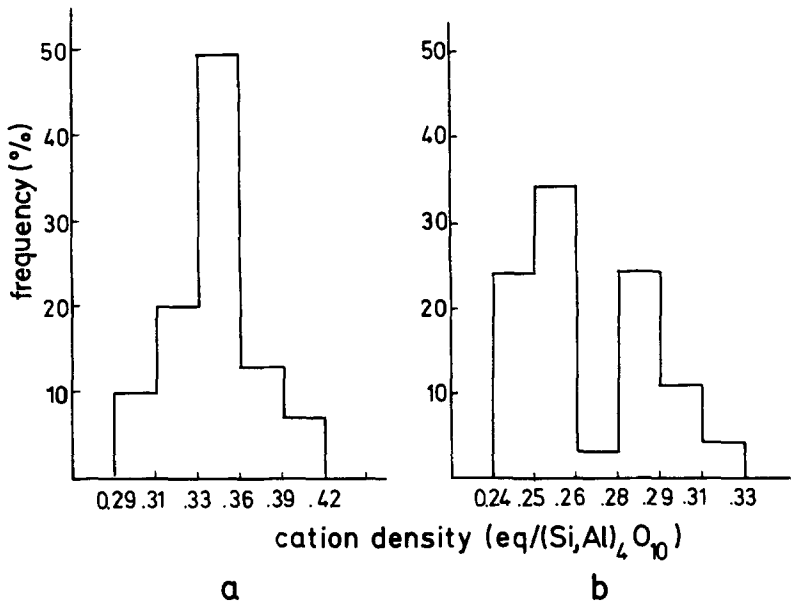

Fig. 1. Types of charge distribution in smectites. a. Uniform distribution (montmorillonite from Groschlattengrün D 3/1, Bavaria); and b. "mixed-layer" like charge distribution (montmorillonite from Italy, type Cinicola N, S. Cinicola, Mailand).

interlayers in a 1:1 ratio. From chemical analyses reported by Kodama the average of the layer charge is 0.65–0.70 eq/(Si,Al)$_4$O$_{10}$. Assuming a mixing ratio of about 1:1 and a cation density of ≈1 eq/(Si,Al)$_4$O$_{10}$ for the mica-like interlayer spaces one estimates the smectitic charge density ξ for the Goto sample from $(1 + \bar{\xi})/2 = 0.70$ to be $\bar{\xi} = 0.4$ eq/(Si,Al)$_4$O$_{10}$ and for Yonago and Honami $\bar{\xi} = 0.36$ and 0.4 eq/(Si,Al)$_4$O$_{10}$.

Data obtainable by the alkylammonium method

Smectites exchange quantitatively their interlayer cations for alkylammonium ions (n_C = 10–18[1]) within some hours. Micas with layer charges >0.8 eq/(Si,Al)$_4$O$_{10}$ react very slowly; within some days one cannot observe any change of the basal spacing due to K^+/alkylammonium ion exchange. A mixed-layer mineral with mica-like and smectitic interlayer spaces exchanges only the cations of the low-charged interlayer spaces if treated for a short period with alkylammonium chloride solutions. This allows exact determination of the cation density and charge heterogeneity of the smectitic interlayer spaces of regular or nearly regular mixed-layer minerals. The charge heterogeneity of the mica-like interlayers cannot be detected.

Preparation and investigation of alkylammonium derivatives

Small amounts (about 50 mg) of the three minerals were treated with alkylammonium chloride solutions for 2, 4, 30, and 100 days at 65°C. From the powder patterns (Debye-Scherrer cameras, 114.3 mm) of the washed and vacuum-dried alkylammonium derivatives the d-values of the (001)-reflections were determined. In a similar way the (001)-reflections of the alkylammonium derivatives swollen under alkanol were recorded and analyzed (for details see Lagaly and Weiss, 1976; Lagaly et al. 1976).

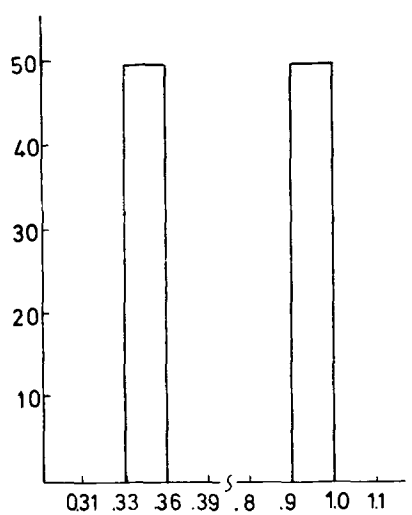

Fig. 1. c. Ideal mixed-layer mineral with smectitic and mica-like interlayer spaces.

[1] n_C: number of carbon atoms in the alkyl chain of the alkylammonium ions $C_{n_C}H_{2n_C+1}NH_3^+$.

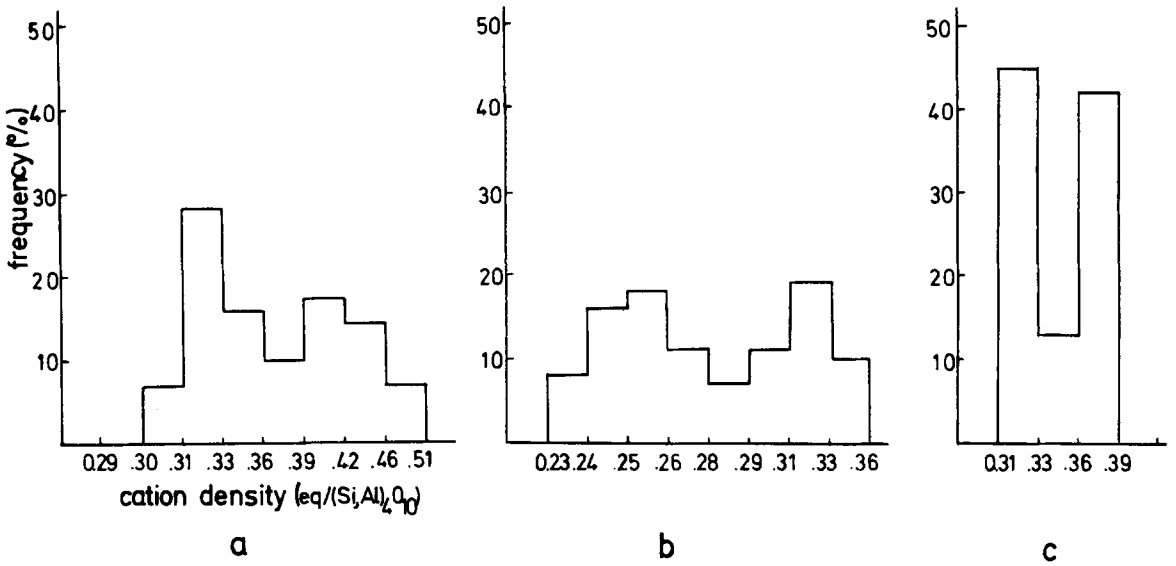

Fig. 2. Interstratification in smectitic mixed-layer minerals: a series of smectites with more or less overlapping charge distribution curves (a–c) ending in typical mixed-layer minerals (d, e). a. Beidellite B 13, Unterrupsroth/Rhön, Germany; b. Ponza bentonite, SC 34, Italy; and c. Bentonite, La Petatera, SC 43, Mexico.

RESULTS

Exchange of alkylammonium ions increases the basal spacing to 2.5–3.2 nm. There are no pronounced differences of the spacings if the reaction times are prolonged up to 4 weeks (Figure 3). The way in which the spacing increases with chain length is similar to that of a typical smectite (Figure 4). Since the spacings of the mixed-layer minerals are about 1 nm higher, it can be immediately concluded (Weiss et al., 1970) that smectitic interlayers alternate regularly with mica-like interlayers.

By subtracting 0.97 nm from the basal spacings of the Goto mineral one obtains a curve typical for a smectite with a monolayer plateau at 1.33 nm and a bilayer plateau at 1.77 nm. The rearrangement of the monolayers into bilayers starts at $n_C > 6$ and is completed at $n_C = 12$. From these points one calculates that the charge density of the smectitic interlayers varies between 0.31 (from $n_C = 12$) and 0.51 eq/$(Si,Al)_4O_{10}$ (from $n_C = 6$).

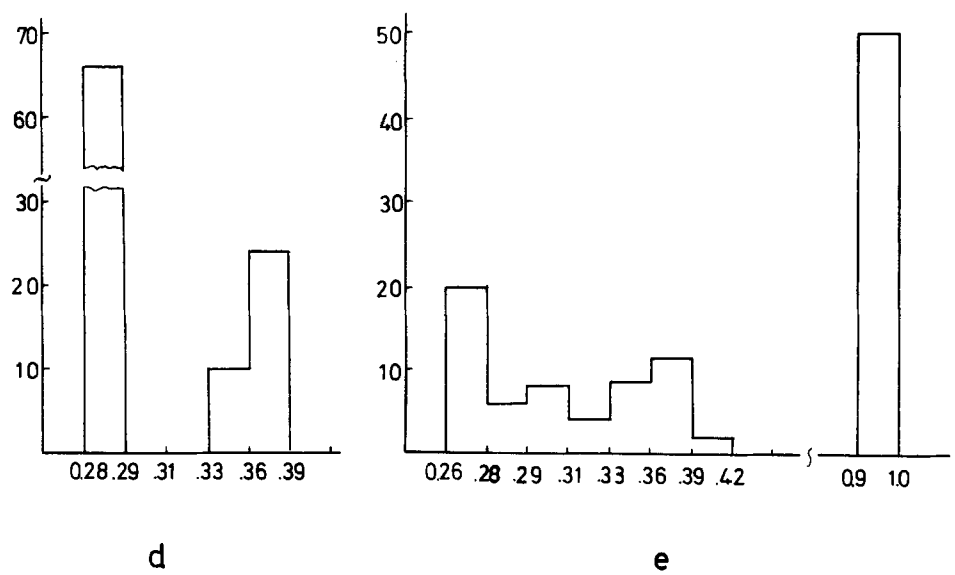

Fig. 2. d. Bentonite, Sardinia, SC 24, Italy; and e. Rectorite, Little Rock, Arkansas, USA.

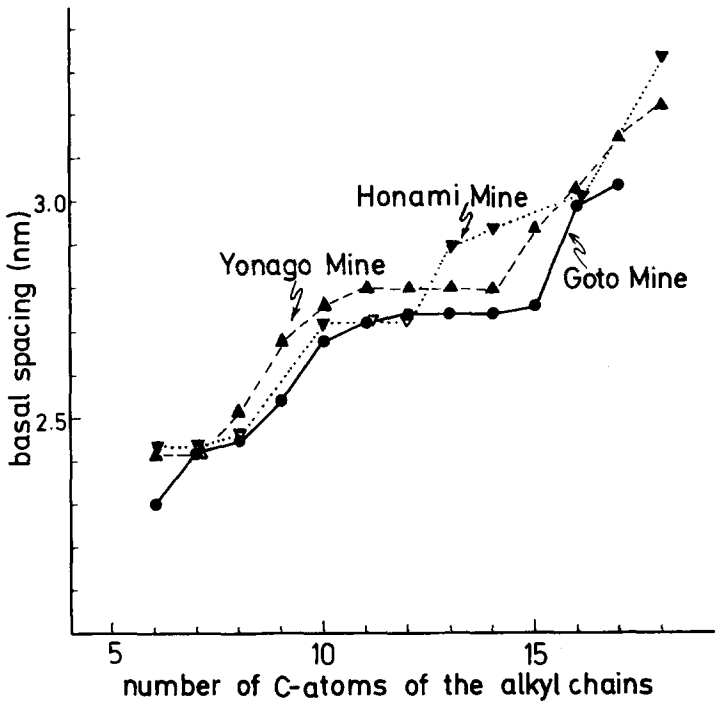

Fig. 3. Basal spacings d_1 of the alkylammonium derivatives of the Goto (●), Yonago (▲), and Honami (▼) Mine specimens.

At $n_C > 15$ the bilayer rearranges into the pseudo-trimolecular structure. This transition indicates an upper charge limit of 0.52 eq/$(Si,Al)_4O_{10}$ in good agreement with the monolayer/bilayer transition.

The charge distribution (Figure 5a) is calculated from the variation of d_{001} with n_C between $n_C = 6$ and $n_C = 12$ (Stul and Mortier, 1974; Lagaly and Weiss, 1976). There are two types of smectitic interlayers. About 76% of the smectitic interlayers have cation densities around 0.37 and 24% densities of 0.46–0.51 eq/$(Si,Al)_4O_{10}$.[2] The average cation density of 0.40 eq/$(Si,Al)_4O_{10}$ calculated from the distribution is in agreement with the value of 0.40 estimated from Kodama's analytical data.

The Yonago sample gives similar spacings (Figure 3) but a completely different charge distribution (Figure 5b). The frequency of the differently charged interlayers does not vary around a distinct value, but decreases with decreasing charge density. The upper limit of the cation density calculated from the transition: bilayer/pseudo-trimolecular layer is higher (0.55 eq/$(Si,Al)_4O_{10}$ from $n_C = 14$) than that from the monolayer/bilayer transition (0.46 eq/$(Si,Al)_4O_{10}$ from $n_C = 7$). This indicates interlayer spaces with unsymmetrical charge distribution (Lagaly et al., 1976). The average charge density calculated from the distribution curve is 0.40 eq/$(Si,Al)_4O_{10}$ in agreement with the value of 0.36 eq/$(Si,Al)_4O_{10}$ estimated from analytical data.

The sample from Honami has a similar charge distribution to that of the Yonago sample with interlayer charge densities between 0.33 and 0.46 eq/$(Si,Al)_4O_{10}$. The difference between the highest value calculated from the transition bilayer/pseudo-trimolecular layer (0.62 eq/$(Si,Al)_4O_{10}$ from $n_C = 12$) and that from the monolayer/bilayer transition at $n_C \geq 7$ (0.46 eq/$(Si,Al)_4O_{10}$) reveals pronounced unsymmetrical charge distributions in some interlayer spaces. The mean charge density of 0.40 is markedly higher than the value of about 0.30 estimated from the analytical data.

DISCUSSION

The term "layer charge" has been avoided in the preceding chapters. The alkylammonium method measures the density of the interlayer cations and not the layer charge; the layer charge has to be deduced from the interlayer cation density. Another reason is that interstratification can be detected only if the interlayers have different cation densities. An example is illustrated in Figure 6a. For a regular interstratification of high- and low-charged layers in ratios of about 1:1 all interlayer spaces have the same cation density. Since all tests for interstratification are based on differences of the interlayer cation density, the sample appears as a homogeneous mineral and the mixed-layer character cannot be detected. An indication of this type of inter-

[2] This proportion suggests some regularities of the charge density distribution in the smectitic interlayer spaces.

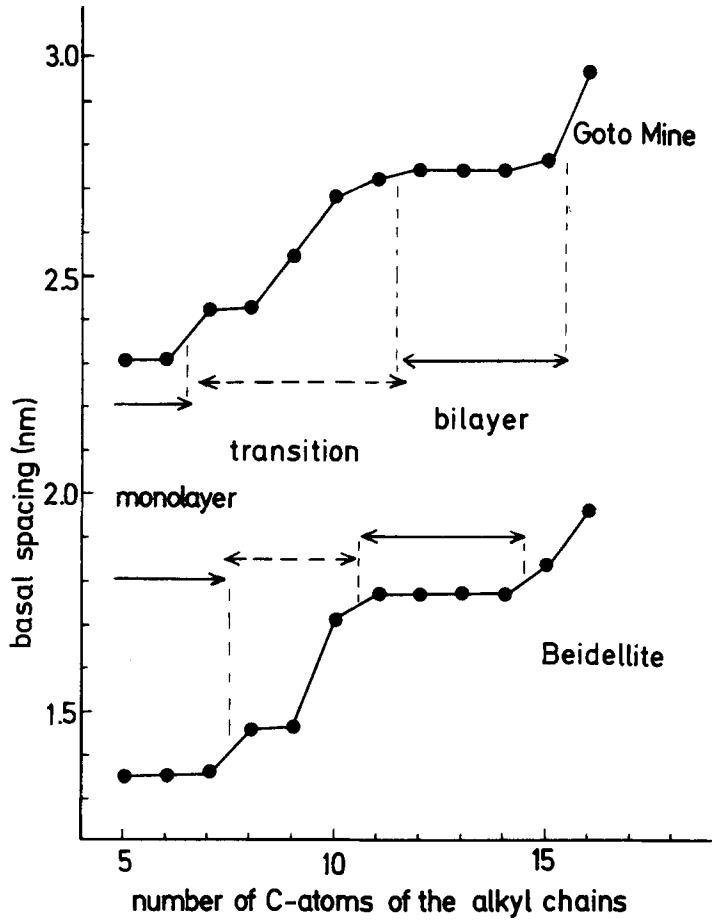

Fig. 4. Comparison of the basal spacings of the mixed-layer mineral from Goto Mine with a high-charged smectite. (Beidellite (B 15) of Unterrupsroth/Rhön, Germany; charge heterogeneity 0.33–0.46 eq/$(Si,Al)_4O_{10}$, average cation density (layer charge): 0.38 eq/$(Si,Al)_4O_{10}$.)

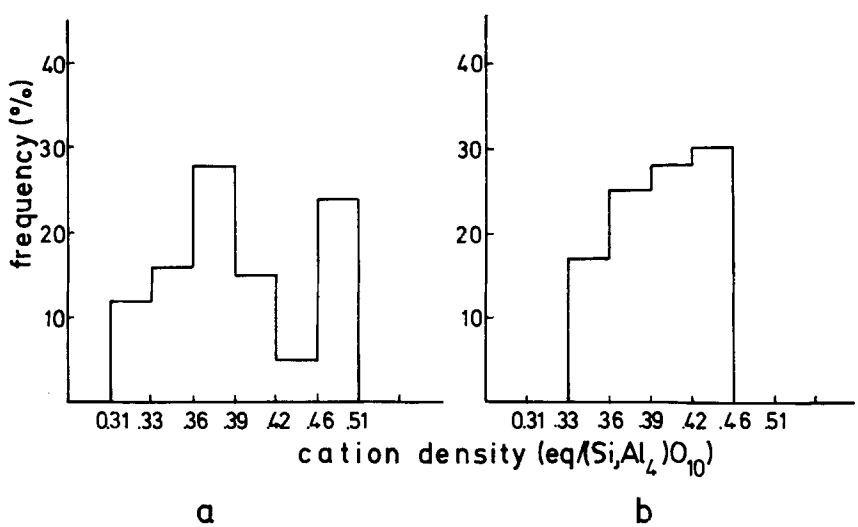

Fig. 5. Charge distribution of the samples from the Goto Mine (a) and the Yonago Mine (similar Honami Mine) (b).

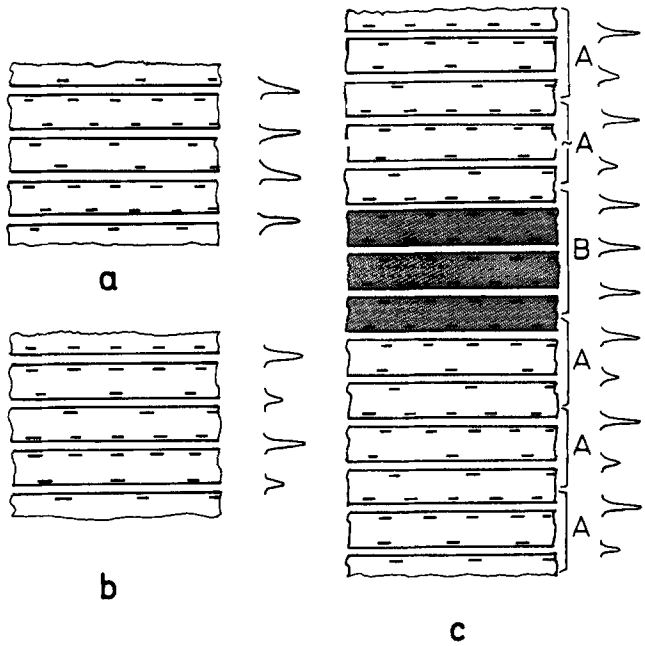

Fig. 6. a. Regular 1:1 interstratification of high- and low-charged silicate layers forming unsymmetrically charged interlayer spaces with identical cation densities (for clarity the interlayer cations are not shown; their density is illustrated by the peaks on the right side). b. Alternating high- and low-charged interlayer spaces formed by polar layers (Goto Mine sample). c. Random distribution of some units B of three or more mica layers in the sequence of polar layers (units A as in Figure 6b); model for the Yonago Mine sample.

stratification might be obtained by the alkylammonium exchange. As discussed above, differences between the upper limit of the cation density calculated from the transition bilayer/pseudo-trimolecular layer and that from the monolayer/bilayer transition reveal unsymmetrically charged interlayer spaces (Figure 6a). This implies that the "faces" of the layers forming the interlayer space are differently charged (Lagaly et al., 1976). If the proportion of the high- and low-charged layers differs from 1:1, two or three kinds of interlayer spaces occur and the interstratification can be recognized.

Experimental results which indicate a regular alternation of high- and low-charged interlayer spaces, can be interpreted best by the model in Figure 6b: the silicate layers are polar and their polar axes alternate regularly (Lippmann and Johns, 1969; Veniale, 1970; Weiss et al. 1970).

The same type of structure has to be postulated for the three samples investigated. The alkylammonium method gives an average charge density of the smectitic interlayers of 0.4 eq/$(Si,Al)_4O_{10}$; the charge density of the mica-like interlayers cannot be determined as explained above.

The proportions of the differently charged interlayer spaces cannot be directly obtained by the alkylammonium ion exchange. The procedure described above is based on the assumption that the proportion of high- and low-charged interlayer spaces is about 1:1. This seems to be especially true for the Goto sample. The conclusion is drawn from the fact that the difference between the bilayer plateau of the mixed-layer mineral (2.74 nm, Figures 3, 4) and the bilayer layer plateau of a typical smectite (1.77 nm) corresponds to the identity period in micas. In contrast, the Yonago sample has its bilayer plateau at 2.80 nm and, therefore, 1.03 nm above the bilayer plateau of smectites.

At first glance one might assume that in the Yonago sample the mica-like interlayers have reacted with alkylammonium ions to some extent or that the proportion of the smectitic interlayers is higher than 50%. The first assumption can be rejected since after exchange of alkylammonium ions the spacing of the mica-like interlayers would increase with chain length and prevent the formation of a bilayer plateau. The second assumption contradicts another experimental result. Swelling of the decylammonium derivative, for example, with decanol gives a spacing of 4.71 nm, which is 0.04 nm smaller than expected (4.75 nm = 3.78 nm for the smectitic + 0.97 nm for the mica-like interlayers). For the corresponding dodecylammonium derivative the spacing is again too low (0.04 nm). When the proportion of smectitic interlayers was increased, the swollen samples should have higher spacings! There are only certain models consistent with these experimental results. A simple model explains the experimental facts: Some of the units A (Figure 6c) including a high- and a low-charged interlayer space are randomly replaced by ag-

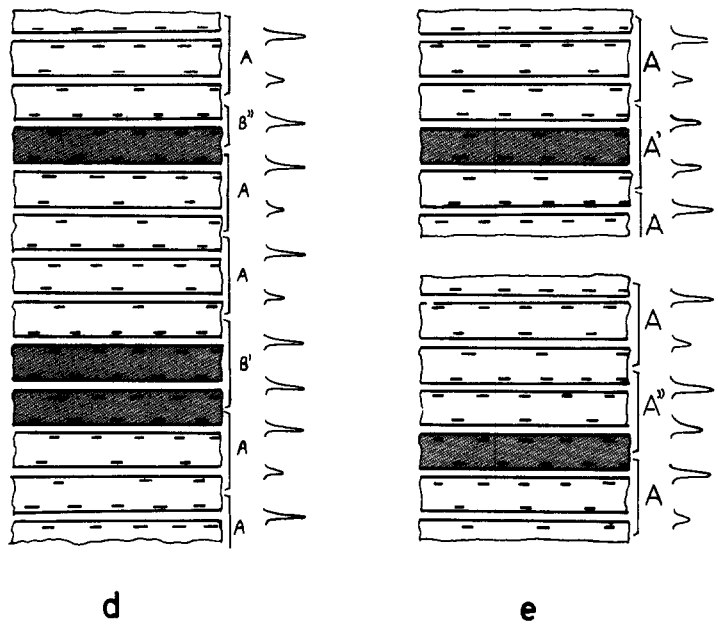

Fig. 6. d. Random distribution of some isolated mica layers or pairs of mica layers in the sequence of polar layers (units A); model for the Honami Mine sample. (In the three samples the charge density of the smectitic and probably also of the mica-like interlayer spaces are not exactly equal as shown in Figure 6 b–d for clarity but differ according to the observed heterogeneity.) e. Some special defects in the sequence of polar layers which contribute to the enhanced unsymmetrical charge distribution in the smectitic interlayer spaces.

gregates of three or more mica layers (units B). In the unswollen alkylammonium derivatives units B (with three mica-like interlayers about $3 \cdot 0.97 = 2.91$ nm or with four mica-like interlayers $4 \cdot 0.97 = 3.88$ nm thick) are larger than the units A (2.74 nm thick at the bilayer plateau); random interstratification of units A with units B, therefore, increases the spacing at the bilayer plateau. In the swollen samples units A become thicker ($3.78 + 0.97 = 4.75$ nm) than the units B (again 3.88 nm) and random interstratification decreases the spacing.

In a rough approximation the proportion x of units B is estimated from the observed spacing of 2.80 nm at the bilayer plateau by the relation:

$$(1 - x) \cdot 2.74 + 3.88x = 2.80; \; x = 0.05$$

(2.74 nm: thickness of units A at bilayer plateau; 3.88 nm: thickness of units B with four mica layers). For the monolayer plateau at 2.42 nm (units A: $1.36 + 0.97 = 2.33$ nm thick) one obtains:

$$(1 - x) \cdot 2.33 + 3.88x = 2.42; \; x = 0.06.$$

For the decylammonium derivative swollen under decanol (units A: $3.78 + 0.97 = 4.75$ nm thick; observed spacing: 4.71 nm):

$$(1 - x) \cdot 4.75 + 3.88x = 4.71; \; x = 0.05$$

(if the units B are considered to contain only three mica-like interlayers, the proportions of B become too high).

Interstratification of units A with 5% of units B (with four mica-like interlayers) shifts the proportion of mica-like interlayers to smectitic ones from 0.50:0.50 to 0.55:0.45 in agreement with Kodama's values (0.55:0.45) obtained from Fourier transforms of the glycerolated samples. The conclusions are also consistent with the probabilities reported: p_{AA} and p_{BA} high, p_{AB} lower and p_{BB} low. ("A" referring to mica-like interlayers, "B" to smectitic interlayers.)

The bilayer plateau of the Honami sample lies slightly (0.02 nm) below the bilayer plateau of the Goto Mine sample which might be considered as rather "ideal" regular 1:1 interstratification. The decreased spacing is produced by interstratification with isolated mica layers or pairs of them (Figure 6d) and not with larger aggregates as in the Yonago sample. An estimation shows that 3–5% of the layers are pairs of mica layers.

The Honami sample is characterized by a pronounced unsymmetrical charge distribution in the interlayers. The highest average cation density is about 0.46 eq/$(Si,Al)_4O_{10}$ (from transition: monolayer/bilayer) but the highest charge density of one of the two silicate surfaces forming an interlayer space is at least 0.6 eq/$(Si,Al)_4O_{10}$ (from transition: bilayer/pseudo-trimolecular layer).

Possibly the crystals contain, in addition to the usual charge heterogeneity, special defects such as contacts of the "smectitic face" of the polar layers with the "mica-face" or with isolated mica layers (Figure 6e).

The total number of mica layers may be, therefore, higher than 5% but there is no chance to determine the exact value.

According to the calculations of Kodama, 60% of the interlayer spaces behave as mica-like interlayer spaces by application of the glycerol test. Since the alkylammonium ion exchange reveals that there really are no more than 55% of true mica-like interlayers, some of the highly charged "smectitic" interlayer spaces do not completely expand with glycerol and behave as mica-like interlayers. In the presence of divalent cations the lattice expansion is restricted to 1.4–1.5 nm for charge densities ≥ 0.6 eq/$(Si,Al)_4O_{10}$.

CONCLUSION

The terms "interstratification" and "mixed-layering" are in general used for minerals which contain two (or more) kinds of layers in regular or random distribution. The presence of two (or more) kinds of layers can be observed only when the interstratification creates interlayer spaces with different cation density. Alternating interlayer spaces with high and low cation density do not imply different types of layers but are caused by polar layers (Figure 6b). Typical examples are the Goto, Yonago, and Honami minerals. These samples contain mica-like interlayers (cation density > 0.8 eq/$(Si,Al)_4O_{10}$) and smectitic interlayers (cation density about 0.4 eq/$(Si,Al)_4O_{10}$). Cation density and charge distribution can be exactly determined by the alkylammonium ion exchange for the smectitic interlayer spaces, but not for the mica-like ones; it can only be stated that their charge density must be higher than 0.8 eq/$(Si,Al)_4O_{10}$.

Some deviations from regular 1:1 interstratification are detected by careful inspection of the basal spacings of the alkylammonium derivatives. The Goto sample is a rather regular interstratification of mica-like and smectitic interlayer spaces in proportion 0.50:0.50 (Figure 6b). The Yonago sample contains in random interstratification about 5% of the layers as packets of about four mica-like layers (Figure 6c). In the Honami mineral (Figure 6d) pairs of two mica-like layers appear to be more frequent than in the other samples.

In all samples one can clearly distinguish between mica-like and smectitic interlayers. In other samples with decreased cation density in the mica-like interlayical for many smectites. These charge distributions might then be considered as "relicts" of interstratification. One cannot exclude the idea that in some cases the alteration of micas starts with forming interstratified samples as a consequence of cooperative processes (see Sawhney, 1972). The interstratified products are weathered to smectites which exhibit a charge distribution like that in Figure 2 which provides an indication of their origin.

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Резюме- Ионный обмен смешаннослойных минералов и алкиламмония позволяет получить детальную информацию о изменениях плотности катионов в последующих межслойных промежутках. Были исследованы три смешанно-слойных образца из Японии (предоставленных Др.Кодома). Их переслаивание,близкое 1:1 основано на регулярном чередовании сильно- и слабо-заряженных межслойных промежутков,что обусловлено регулярной последовательностью полярных слоев. Плотность катионов в сильно заряженных межслойных промежутках >0,8эк/$(Si,Al)_4O_{10}$. Слабо заряженные промежутки имеют среднюю плотность катионов 0,4эк$(Si,Al)_4O_{10}$ и гетерогенное распределение зарядов. Вид гетерогенности образцов Гото Майн отличается от видов гетерогенности образцов Янаго Майн и Гонами Майн. Образец Гото Майн имеет довольно регулярную последовательность слабо- и сильно-заряженных межслойных промежутков в пропорции,близкой к 0,50:0,50. Другие два образца содержат межслойные промежутки с явно несимметричным распределением зарядов. Образец Янаго Майн имеет около 10 процентов слюдо-подобных слоев,беспорядочно перемежающихся с полярными слоями и обособленных в пакеты из трех и более слоев; отношение сильно заряженных межслойных промежутков и слабо заряженных возрастает до примерно 0,55:0,45. Образец Гонами Майн возможно содержит изолированные слюдо-подобные слои или их пары. Пропорция сильно заряженных межслойных промежутков определяется примерно как 0,53:0,47 и ниже,чем было определено Кодома в обработанном глицерином образце.

Изученные образцы могут рассматриваться как конечные члены серии переслаивающихся образцов,которая начинается смектитами с подобным смешанно-слойному распределению зарядов.

RESUME - Der Alkylammoniumionenaustausch an 2:1 Schichtsilikaten mit Wechsellagerungsstruktur ermöglicht Aussagen über die Variationen der Kationendichte in aufeinanderfolgenden Zwischenschichträumen. Drei Proben aus Japan (zur Verfügung gestellt von Dr. H. Kodama) wurden untersucht. Ihre fast 1:1 Wechsellagerung beruht auf regelmäßig abwechselnden hoch und niedrig geladenen Zwischenschichträumen, welche durch eine regelmäßige Anordnung polarer Schichten verursacht wird. Die Kationendichte in den hoch geladenen Zwischenschichträumen ist >0,8 Äquivalente/$(Si,Al)_4O_{10}$. Die niedrig geladenen Zwischenschichträume haben eine mittlere Kationendichte von 0,4 Äquivalenten/$(Si,Al)_4O_{10}$ und die Ladungsverteilung ist heterogen. Die Proben von Goto Mine, Yonaga Mine und Honami Mine besitzen alle eine verschiedene Heterogenität. Die Goto Mine Probe hat eine recht regelmäßige Anordnung der hoch und niedrig geladenen Zwischenschichten im Verhältnis 0,50:0,50. Die anderen zwei Proben enthalten Zwischenschichträume mit ausgesprochen unsymmetrischer Ladungsverteilung. Wahrscheinlich enthält die Probe von Yonaga Mine zusätzlich Pakete aus mehreren Glimmerschichten. Das Verhältnis der hoch geladenen Schichten zu den niedrig geladenen nimmt bis etwa 0,55:0,45 zu. Die Honami-Probe enthält wahrscheinlich isolierte Glimmerschichten oder Paare davon. Der Anteil der hoch geladenen Zwischenschichten wird auf 0,53:0,47 geschätzt und ist niedriger als Kodamas Werte aus der Glycerinaufweitung. Die untersuchten Proben können als Entglieder einer Entwicklungsreihe von Wechsellagerungsverbindungen angesehen werden, an deren Anfang die Smektite mit wechsellagerungsartiger Ladungsverteilung stehen.

RESUME - L'échange d'ions alkylammonium sur des argiles interstratifiées donne une information détailée concernant la variation de densité de cations dans la succession d'espaces interfeuillets. Trois exemples du Japon (procurés par Dr.H.Kodama) ont été étudiés. Leur rapport d'interstratification, quasi 1:1, se base sur des espaces interfeuillets alternants régulièrement entre des charges hautes et basses, qui sont causés par une suite régulière de feuillets polaires. La densité de cations dans les espaces interfeuillets à haute charge est > 0,8 eq/$(Si,Al)_4O_{10}$. Les espaces interfeuillets à basses charges ont une densité de cation moyenne de 0,4 eq/$(Si,Al)_4O_{10}$ et une distribution de charge hétérogène. Le genre d'hétérogénéité des échantillons des mines de Goto est différant de celui des échantillons des mines de Yonago et de Honami.

L'échantillon de la mine de Goto a une séquence assez régulière d'espaces interfeuillets à haute et basse charge dans des proportions proches de 0,50:0,50. Les deux autres échantillons contiennent des espaces interfeuillets dont la distribution de charge est d'une assymétrie prononcée. L'échantillon de la mine de Yonago a probablement, en distribution fortuite avec des feuillets polaires, à peu près 10 pourcent de feuillets semblables au mica, rassemblés en paquets de trois feuillets ou plus; le rapport entre les espaces interfeuillets à haute charge et ceux de basse charge est augmenté à près de 0,55:0,45.

L'échantillon de la mine de Honami contient probablement des feuillets isolés semblables au mica, ou des paires de tels feuillets. La proportion des espaces interfeuillets à haute charge est estimée à près de 0,53:0,47 et est plus basse que celle déterminée par Kodama de l'échantillon glycérolé.

Les échantillons étudiés peuvent être considérés comme étant les termes extrêmes d'une série d'échantillons interstratifés qui commence avec les smectites à distribution de charge semblable à celle des argiles interstratifiées.