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MICROTOPOGRAPHY OF REGULARLY-INTERSTRATIFIED MICA AND SMECTITE

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Abstract—The gold decoration technique of electron microscopy was used to observe the microtopography of natural (001) surfaces of 1:1 regularly-interstratified mica/smectite minerals (expandable layer: 40–45%) collected from four different pyrophyllite deposits in Japan. The specimens are characterized by parallel growth steps of malformed, circular or polygonal forms with varying step separations. Many particles exhibit paired steps that seem to show spiral growth. Microtopographic observations suggest that the growth of regular interstratification (at least for the specimens investigated in this study) normally takes place by an interlacing of paired steps. If the height of a single step corresponds to that of a mica or a smectite layer, the particles are estimated to be normally 40–300 Å in thickness. If the particles on which a spiral center is observed are single crystals of interstratified mica and smectite, then some crystals investigated in this study are far thicker than fundamental particles. The results of this study are interpreted to suggest that these regularly-interstratified mica/smectites were formed by hydrothermal metasomatism from their respective host rocks.

Key Words—Au-decoration, Electron microscopy, Fundamental particles, Interstratified mica/smectite, Surface microtopography.

INTRODUCTION

The mode of occurrence and the mineralogical characteristics of interstratified mica/smectite minerals have been studied by many investigators. The interstratified or mixed-layer minerals may form in several ways: 1) as intermediate products when mica transforms to smectite during weathering or in a hydrothermal process (Keller *et al.,* 1986; Tomita, 1978), 2) as intermediate products when smectite transforms to mica during diagenesis or in a hydrothermal process (Bethke and Altaner, 1986; Ramseyer and Boles, 1986; Inoue *et al.,* 1988; Ahn and Peacor, 1986; Bethke *et al.,* 1986; Bell, 1986), 3) direct formation as mixed-layer minerals from amorphous material or hydrothermal alteration of rocks (Iiyama and Roy, 1963; Eberl and Hower, 1977).

The conversion of mica to smectite is easily explained by leaching of potassium ions and hydration (Sudo *et al.,* 1962; Norrish, 1972; Bassett, 1960; Giese, 1971, 1972; Gilkes *et al.,* 1972; Tomita, 1978). This process is a solid-state transformation mechanism. On the other hand, the mechanism for the conversion of smectite to illite is still controversial (Hower *et al.,* 1976; Nadeau *et al.,* 1985; Inoue *et al.,* 1987, 1988). In particular, Inoue *et al.* (1988) proposed that the mechanism of transformation from smectite to illite is controlled by the Ostwald ripening process. Tomura *et al.* (1979) discussed the Ostwald ripening growth mechanism with regard to clay minerals following transmission electron microscope (TEM) observations of mica crystal surfaces studied by the Au-decoration technique. Spiral growth patterns of clay minerals were observed in the TEM by Gritsaenko and Samotoyin (1966), Baronnet (1972), Sunagawa and Koshino (1975), Sunagawa *et al.* (1975), Sunagawa (1977), Tomura *et al.* (1979), Baronnet (1980), and Kitagawa *et al.* (1983). These workers observed circular or malformed, spiral or parallel steps under the transmission electron microscope using the Au-decoration technique. Kitagawa *et al.* (1983) critically analyzed surface microtopographies of mica clay minerals in relation to their modes of occurrence. The different microtopographies observed on crystals formed directly from hydrothermal solutions and those formed by hydrothermal metasomatism were explained based on the recent understanding of spiral growth morphology. This kind of microtopographic study has not previously been carried out for the interstratified minerals. The authors believe that the surface microtopography of the interstratified minerals may provide evidence of whether their formation was controlled by Ostwald ripening, by direct formation from solution, and/or by metasomatism.

Nadeau *et al.* (1984a, 1984b) proposed the fundamental particle theory as a new conceptual model for the interstratified mica/smectite minerals. They suggested that the material identified as regularly-interstratified mica/smectite (M/S) by X-ray diffraction (XRD) is composed primarily of elementary "mica" particles (fundamental particles) whose interfaces are capable of forming complexes with water and organic

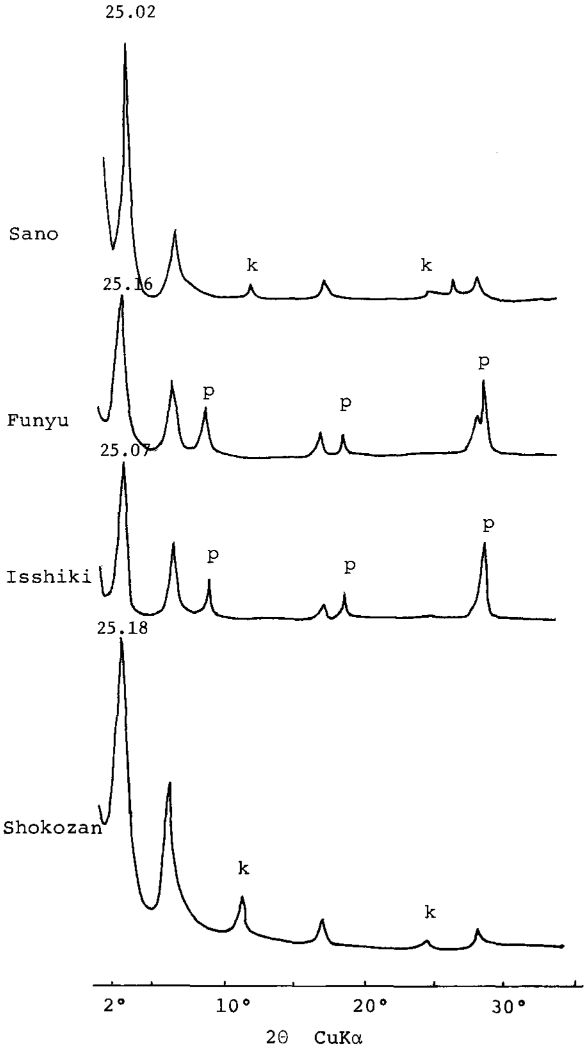

Figure 1. X-ray powder diffraction patterns of untreated specimens collected from the Sano, Funyu, Isshiki, and Shokozan mines. K: kaolinite, P: pyrophyllite.

molecules. Standard XRD data may not be able to distinguish between true interstratification and the interparticle effects of intimate physical mixtures. Therefore, materials yielding XRD patterns of M/S do not necessarily contain both mica and smectite interlayers. According to Nadeau *et al.* (1984a, 1984b) the individual fundamental particles (20–50 Å thick) measured by the Pt-shadowing method are primary crystallization products rather than secondary particles disaggregated from larger crystals during sample preparation.

On the other hand, Ahn and Peacor (1986) and Ahn and Buseck (1990) suggested, based on observation by high-resolution transmission electron microscopy (HRTEM), that thin, fundamental particles are secondary crystallites derived from larger crystals by cleaving at smectite interlayers during sample preparation.

In this study we have attempted to observe the surface microtopographies of the 1:1 regularly-interstratified mica/smectite minerals by means of the Au-decoration technique (Gritsaenko and Samotoyin, 1966; Sunagawa *et al.*, 1975). In addition, we attempted to measure the thickness of individual particles of the regularly-interstratified mica/smectite in order to judge the applicability of the fundamental particle theory of Nadeau *et al.* (1984a, 1984b).

MATERIALS AND METHODS

The interstratified mica/smectite specimens investigated in the present study come from four pyrophyllite deposits in Japan: the Sano mine (Kakuma, Nagano Prefecture), the Isshiki mine (Horai, Aichi Prefecture), the Funyu mine (Shioya, Tochigi Prefecture) and the Shokozan mine (Shobara, Hiroshima Prefecture). The specimen from the Sano mine was taken from a vein in a moderately-altered porphyry (Matsuda, in preparation). The specimen from the Isshiki mine was taken from a veinlet in the deposit that was formed by the alteration of mylonite (Matsuda, 1984). The specimen from the Funyu mine was taken from a sericite clay in the outer part of an alteration area, in a Miocene tuff-breccia (Matsuda *et al.*, 1981b). The specimen from the Shokozan mine was collected from a vein in a deposit that originated from the alteration of andesitic tuffs (Morita and Kakitani, 1977). Each of these specimens was clearly formed by hydrothermal activity (Matsuda *et al.*, 1981a; Matsuda, 1984; Morita and Kakitani, 1977).

Quartz impurities were removed by sedimentation. The four specimens in their natural states were examined by X-ray diffraction (Figure 1).

The percentage of expandable layers and the ordering type (Reichweite) of the interstratified mineral were determined using the diagram proposed by Watanabe (1988). The technique defines three peaks. First, (l_1) at 5.1°–7.6°2θ, second, (l_2) at 8.9°–10.2° 2θ, and third, (l_3) at 16.1°–17.2°2θ (Cu$k\alpha$ radiation) after ethylene glycol treatment. The angular differences represented as $2\theta_1 = l_2 - l_1$, and $2\theta_2 = l_3 - l_2$ are plotted on Watanabe's diagram.

Morphological features were studied in the transmission electron microscope (JEM-T7). Chemistry was determined by atomic absorption spectroscopy.

For the gold decoration technique, the samples were dispersed in distilled water and collected on a thin cover glass (less than 0.1 mm in thickness). After drying, the cover glass with samples was heated in a vacuum of 10^{-4} torr at about 400°–500°C for about two hours. Heating the specimens gives a cleaner surface and a higher mobility of gold, which enables selective nucleation of gold along the steps (Tomura *et al.*, 1979). Gold was flash-evaporated from a tungsten coil heater. After a carbon coating was applied, the specimens were removed and immersed in HF solution (<8 wt. %) for

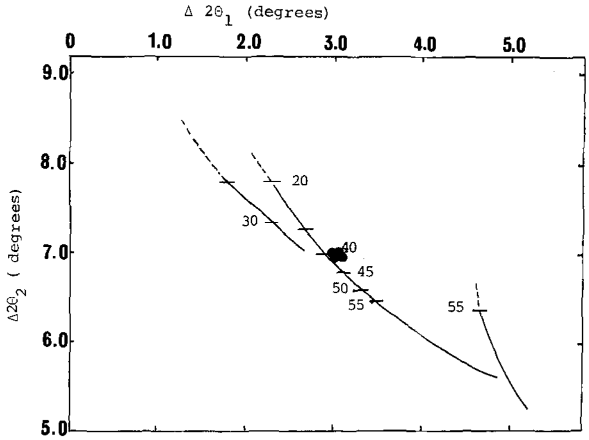

Figure 2. The interstratified minerals from four mines plotted onto the diagram of Watanabe (1988), with which it is possible to identify types of interstratification and to determine the ratio of the expandable layers to the mica-like layers. See text for definition of $2\theta_1$ and $2\theta_2$. The numbers on the diagram represent the percentage of expandable layers.

about three or four days to completely dissolve the silicate minerals. After soaking the remains in distilled water, the thin films were collected on copper mesh grids and ready for observation under the transmission electron microscope (JEM-T7).

RESULTS

As can be seen in Figure 1, specimens from Funyu and Isshiki are accompanied by a small amount of pyrophyllite, and those from Sano and Shokozan are composed of the interstratified M/S mineral with a small amount of kaolinite.

The results obtained from the Watanabe method are shown in Figure 2. This indicates that the percentages of expandable layers of these four specimens are in the range of about 40–45%, i.e., they are nearly ideal 1:1 regular mica/smectite.

The basic morphological features obtained by transmission electron microscopy reveal crystal outlines varying from rectangular, rhombic, and lath-shaped to irregular (Figure 3).

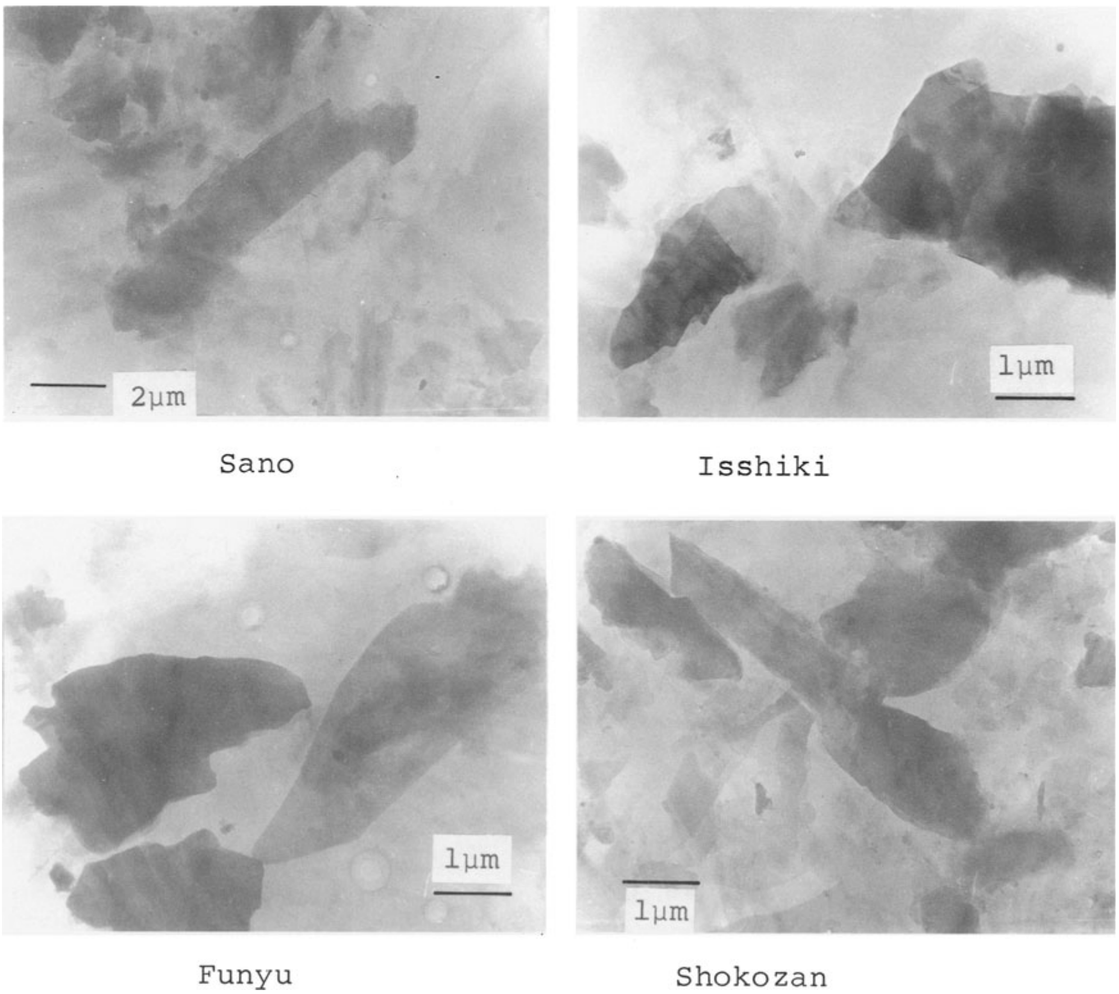

Figure 3. Transmission electron micrographs of the interstratified minerals investigated.

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Table 1. Chemical analysis data of specimens from four pyrophyllite deposits.

	Sa	Is	Fu	Sh
SiO_2	47.78	48.48	43.47	56.57
TiO_2	0.12	0.27	0.35	0.16
Al_2O_3	30.98	31.37	33.37	31.37
Fe_2O_3	0.51	0.11	0.78	0.10
MgO	1.11	0.34	0.36	0.20
CaO	1.53	1.75	1.90	0.33
Na_2O	2.01	2.61	2.33	2.30
K_2O	2.84	1.67	2.14	0.61
P_2O_5		0.02	0.01	
$H_2O(+)$	6.51	7.29	8.31	7.99
$H_2O(-)$	7.10	5.83	6.70	
Total	100.49	99.68	99.72	99.43

Sa: Sano, Is: Isshiki, Fu: Funyu, Sh: Shokozan.

Table 1 lists the results of atomic absorption analysis of our samples and others from the Funyu and Isshiki mines (Matsuda, 1984). According to Matsuda (1984), the exchangeable interlayer cations are composed of Ca, Mg, Na, and K, and those of the mica-like layers are composed of Na, K, and Ca. The K and Ca contents in the Shokozan specimen are remarkably lower than those in specimens of Sano, Isshiki and Funyu. On the other hand, the SiO_2 content in the Shokozan specimen is higher than in the other specimens.

The results of microtopographic observations on crystal surfaces of regular interstratified mica/smectite are summarized in Table 2. The electron photomicrographs in Figures 4 and 5 show that gold grains preferentially nucleate along parallel steps. These step patterns represent growth steps and/or crystal aggregates, not cleavage, and the heights of single steps are assumed to be of one-layer thickness (about 10 Å; refer to Sunagawa and Koshino, 1975; Tomura *et al.*, 1979; Sato, 1970).

The step patterns were observed on all the crystals investigated (Figures 4, 5). They are polygonal or malformed circular steps. As shown in the figures, paired steps are observed on many crystal surfaces. These paired steps are similar to those seen on the crystal surfaces of 2M mica minerals (Tomura *et al.*, 1979; Kitagawa *et al.*, 1983), and on dickite (Sunagawa and Koshino, 1975). The paired steps are due to a spiral growth mechanism similar to that for the formation of the interlacings on kaolin minerals and micas discussed by Sunagawa and Koshino (1975) and Tomura *et al.* (1979).

The spiral center seems to be situated at the central portion of the basal plane in specimens from Sano, Isshiki and Funyu as shown in Figure 5, though it is not always clear. The spiral center of most particles in the Shokozan specimen is unclear in general, although a small number of particles do have their spiral center as shown in Figure 5. These spiral centers may also be observed on the crystal surfaces of pyrophyllite impurities.

As mentioned above, the heights of single steps are assumed to be of unit cell size (10 Å for mica layers) in keeping with studies of Sunagawa and Koshino (1975), Tomura *et al.* (1979), and Sato (1970). Therefore, the thickness of crystals and/or their aggregates can be inferred from the number of steps as shown in Figure 6. On this basis, the thickness of particles in specimens from Sano, Isshiki and Funyu varies from 40–300 Å (Table 2). The thickness of particles in the Shokozan specimen is far thicker than that of other specimens, and reaches 500–800 Å (Figure 6).

DISCUSSION

Crystal growth of the 1:1 regularly-interstratified mineral

Each layer of the paired growth steps has the height of a single mica and smectite layer, as was inferred in studies of the growth mechanism of 2M mica and dickite (Sunagawa *et al.*, 1975; Kitagawa *et al.*, 1983). If these interstratified minerals formed from mica clay minerals by leaching of interlayer cations and subsequent hydration (Sudo *et al.*, 1962; Tomita, 1978), it may be possible that the paired steps are relics of the crystal surfaces of the mica clay mineral. However, according to the study of Matsuda (1984), there is a low probability that these specimens were formed from mica clay minerals. According to Sunagawa *et al.* (1975) and Kitagawa *et al.* (1983), the microtopographies on mica clay minerals show polygonal or circular spiral steps. The growth patterns on crystal surfaces of the present specimens are only slightly different from those on mica clay minerals.

According to Sunagawa (1984), sheet silicate minerals grown from the vapor phase exhibit step-separation versus step-height ratios ranging from 10^4 to 10^5, as compared with much smaller ratios (10^2–10^3) for minerals grown from hydrothermal solutions. This ratio for specimens in the present study is 10^1–10^3 in the

Table 2. Summary of microtopographic observations.

Specimen	Morphology	Particle size ϕ (μm)		Spiral form	Step separation (Å)	Thickness of particles (Å)
Sano	Irregular lath	0.1–3.0	M > P	Paired > single	100–2000	40–300
Isshiki	Irregular rhombic	0.1–3.0	M > P	Paired > single	100–2000	40–300
Funyu	Irregular rhombic	0.1–3.0	M > P	Paired > single	100–2000	40–300
Shokozan	Irregular rectangular	1.0–10.0	M > P	Paired > single	100–5000	40–800

M: Malformed circular spiral, P: polygonal spiral.

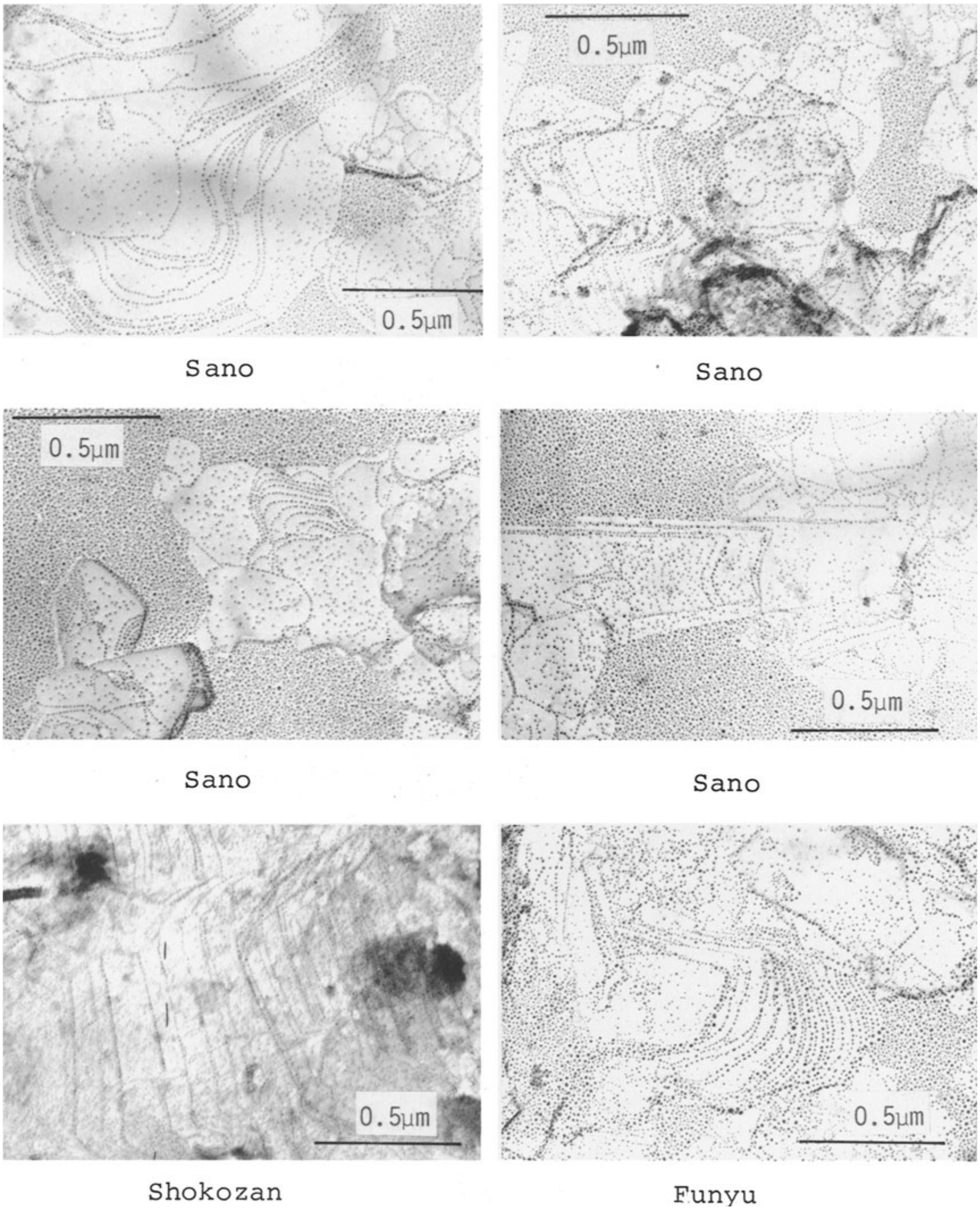

Figure 4. Transmission electron micrographs showing the surface microtopographies of interstratified minerals.

majority of cases. Thus it may be assumed that these regularly-interstratified mica/smectite minerals and impurities (kaolinite and pyrophyllite) grew from a hydrothermal solution rather than from a high temperature vapor phase.

As mentioned above, if microtopographies on the particle surfaces indicate the growth mechanism of the interstratified mineral, then the paired polygonal or malformed circular spirals and the interlacing patterns reflect the growth of mica and expandable layers, respectively.

Sunagawa *et al.* (1975) suggested that coalescence of crystals occurs more frequently in violently moving solutions in open fractures than in static solutions.

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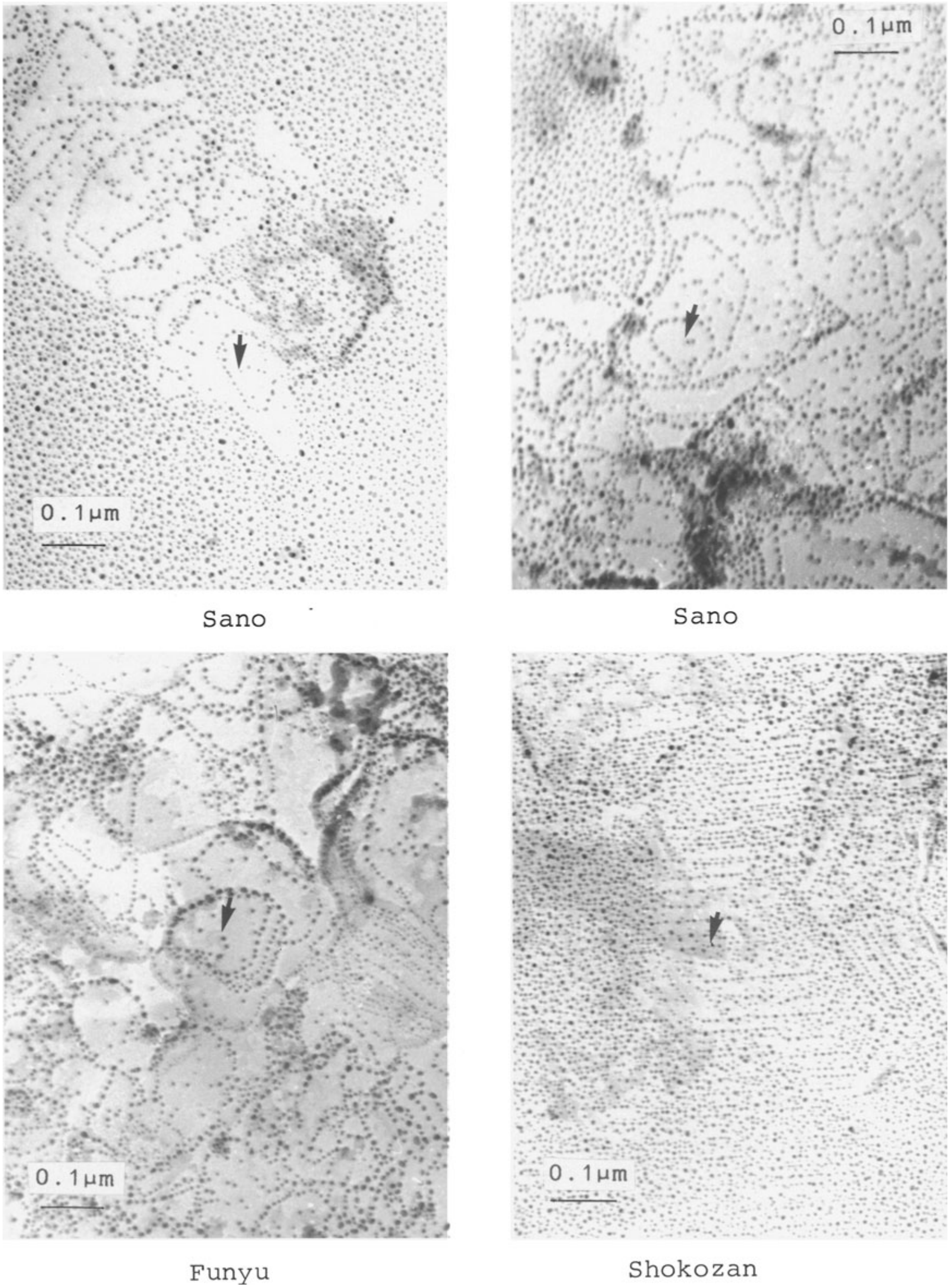

Figure 5. Transmission electron microtopographs showing spiral growth centers. The dislocation centers are marked by arrows.

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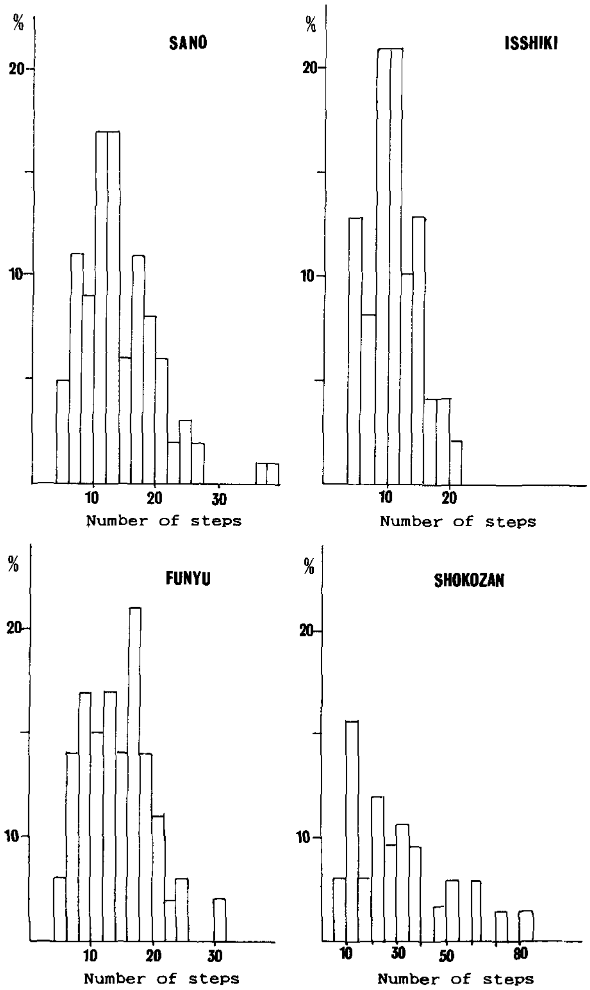

Figure 6. Histograms showing the number of steps observed on individual particles and their distribution.

However, the coalescence of crystals was not observed in these specimens. Regular spacing between growth steps is also important evidence for the lack of coalescence of these crystals.

The minerals may have grown primarily by hydrothermal metasomatism from their respective host rocks, even though some of them occur in veins or veinlets.

Implication for the fundamental particle theory

In this study, many particles seem to have formed by a spiral growth mechanism as shown in Figure 5. Particles gradually thicken from the edge toward the center. If the height of one step corresponds to a 2:1 silicate layer (mica or smectite layer), the thickness of the particles is 40–300 Å (Figure 6; Table 2). If each particle having a spiral center corresponds to the interstratified mica/smectite, the particles investigated in this study are clearly thicker than the 20 Å suggested by Nadeau *et al.* (1984a, 1984b). Also, if many particles in these specimens correspond to individual crystals, the thickness of the crystals measured by the Pt-shadowing method of Nadeau *et al.* (1984a, 1984b) may not result in accurate thickness values of individual crystals, but may be showing the thickness of the edge area. The thickness of particles obtained in this study by the Au-decoration method is consistent with HRTEM thickness data of the interstratified mica/smectite reported by Ahn and Peacor (1986a, 1986b) and Ahn and Buseck (1990).

CONCLUSIONS

The observations of microtopography by the Au-decoration technique have shown that polygonal and/or malformed circular steps are common on particle surfaces in specimens composed of regularly-interstratified mica and smectite, accompanied by a small amount of kaolinite and pyrophyllite. Many particles also show paired steps. If any of these particles correspond to individual crystals of the interstratified mica/smectite, the thickness of these individual crystals is estimated to be 40–300 Å, which is clearly thicker than that of fundamental particles. However, the observed steps on the surfaces may correspond not only to individual crystals of interstratified mica/smectite, but also to crystal aggregates and impurities.

Based on the results of the step-separation versus step-height ratios, and also on the lack of coalescence in these specimens, we conclude that all crystals were formed primarily by hydrothermal metasomatism from the respective host rocks.

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