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SURFACE MICROTOPOGRAPHY OF ILLITE CRYSTALS FROM DIFFERENT MODES OF OCCURRENCE

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Abstract

By means of transmission electron microscopy (TEM) and the gold-decoration technique, the surface microtopography of illite crystals from various environments was investigated. On the crystal surfaces, gold decoration successfully revealed growth steps of ~1 nm in height. The illite samples investigated exclusively exhibit growth spirals of either polygonal or circular form, with varying separation of steps. The spiral morphological characteristics were critically analyzed in relation to their mode of occurrence. Crystals from massive metasomatic specimens (aggregation texture with very low porosity) are characterized by circular growth-spirals with a narrow separation of steps, whereas those from veins and druses (aggregation texture with high porosity) are characterized by polygonal growth-spirals with a wide separation of steps. Hexagonal and lath-shaped crystals were controlled by polygonal spiral-growth; on the other hand, irregular platy crystals are characterized by circular or malformed circular spiral-growth. The morphology of illite crystals seems to reflect the spiral-growth process under the various conditions.

Keywords: illite, microtopography, spiral growth, Au-decoration method, morphology, hydrothermal environment, metasomatism.

Sommaire

A l'aide de la microscopie électronique en transmission et la technique de décoration des surfaces par déposition d'or, on a caractérisé la microtopographie de la surface des cristaux d'illite provenant de divers milieux de croissance. La technique de décorer les surfaces avec de l'or a révélé la présence de couches de croissance d'environ un nanomètre en hauteur. Les échantillons d'illite ne montrent que des spirales de croissance, mais celles-ci peuvent avoir une forme soit polygonale ou bien circulaire, et la forme dépendrait du mode de formation. Les cristaux provenant de spécimens métasomatiques massifs (texture d'agrégation à porosité très faible) possèdent des spirales de croissance circulaires, et une étroite séparation entre couches de croissance. Par contre, les cristaux provenant de veines et de cavités (texture d'agrégation à porosité élevée) ont des spirales de croissance polygonales et une plus grande épaisseur des couches de croissance. Les cristaux hexagonaux ou en plaquettes irrégulières possèdent des spirales de croissance circulaires ou plutôt difformes. La morphologie des cristaux d'illite semble résulter d'un processus de croissance en spirales dans les conditions considérées ici.

(Traduit par la Rédaction)

Mots-clés: illite, microtopographie, croissance en spirales, décoration avec de l'or, morphologie, milieu hydrothermal, métasomatose.

Introduction

The mineralogical properties of illite are very similar, regardless of mode of occurrence, whether it occurs in veins or as an alteration product of granite and rhyolite. Yet, it must have formed in different environmental conditions, judging from distinct modes of occurrence, and thus must differ in some subtle way. If such subtle differences could be elucidated, they should provide insight into the genesis of illite deposits.

The microtopography of the surfaces of layer silicates such as kaolinite, mica, pyrophyllite, chlorite, rectorite and mixed-layer illite–smectite has been studied by many investigators for the purpose of elucidating the growth mechanisms of the crystals (Baronnet 1972, 1980, Sunagawa *et al.* 1975, Tomura *et al.* 1979, Inoue & Kitagawa 1994, Kitagawa *et al.* 1994, Kitagawa 1997). The growth-induced step-patterns, a few nanometers in height, were revealed by the gold-decoration technique employed with transmission electron-

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microscopy (TEM), as developed by Bassett (1958) and Gritsaenko & Samotoyin (1966). These examinations revealed spiral and parallel-step patterns on the (001) crystal surfaces of various layer silicates, so that their growth mechanisms and conditions of growth could be inferred (Sunagawa *et al.* 1975, Tomura *et al.* 1979).

Using the Au-decoration technique, Sunagawa *et al.* (1975) and Tomura *et al.* (1979) demonstrated that 1) in hydrothermal solution (*e.g.*, chlorite that occurs as a vein-forming gangue mineral at the Ani mine, Japan), coalescence of crystals is common, 2) in a metasomatic environment (*e.g.*, hydrothermal metasomatic clay deposits of massive type, such as the Itaya kaolinite deposit and the Azuma illite deposit in Japan), host rocks (*e.g.*, volcanic rocks such as andesitic, dacitic and rhyolitic) are dissolved to form a weakly supersaturated hydrothermal solution, in which crystals grow by the spiral-growth mechanism, and 3) in regional metamorphic environments (*e.g.*, illite and porphyroblastic white mica in pelitic shists of the Sanbagawa metamorphic terrane in the Shiragayama area, central Shikoku, Japan), Ostwald ripening plays an essential role in the growth of layer silicates, and the spiral-growth mechanism is absent.

These results demonstrate that the observations on the surface microtopographs can provide important information as to the genesis of layer silicates. Thus I applied the decoration technique to the (001) surfaces of illite crystals collected from many localities, such as hydrothermal clay deposits, veins, faults and druses. In this paper, the results of these observations are described, and are analyzed in relation to growth environment of these samples.

Materials and Methods

Characteristics of the specimens

The samples of illite investigated in the present study come from more than 20 localities, as indicated in Table 1. Specimens were collected from hydrothermal clay deposits, veins, replacement products of vein form, faults, alteration zone of ore deposits and druses in rocks. The mode of occurrence, mineralogy and geochemistry of these specimens have been studied by many investigators, *e.g.*, Iwao (1963), Matsumoto (1965), Minato (1969), Kakitani & Kitagawa (1977), Ishihara *et al.* (1980), Kitagawa & Kakitani (1978a, b, 1979, 1981a, b), Kitagawa *et al.* (1981, 1982) and Kitagawa & Kameoka (1986).

According to the modes of occurrence, observations by optical microscopy (thin sections), X-ray analysis (dispersion in distilled water and then oriented mount on the thin cover glass), transmission electron microscopy (TEM) (samples dispersed in distilled water) and scanning electron microscopy (SEM), the samples can be considered of three types, respectively. Firstly, on the basis of mode of occurrence, there are massive metasomatic specimens (clay deposits), metasomatic specimens (clay deposits, replacement product in vein form, fault) and fissure-filling specimens (veins, druses). Secondly, on the basis of polytype present, 2*M*, 1*M* and 1M_d, are recognized. Thirdly, three kinds of morphology are recognized in TEM observations: hexagonal plate, lath-shaped plate and irregular plate (Fig. 1). Finally, on the basis of the state of aggregation, as revealed by the texture of particles in SEM observations, there are low-porosity, moderate-porosity and higher-porosity samples (Fig. 2). The mode of occurrence and mineralogical characteristics of each specimen in this study are summarized in Table 1, based on the three types grouped as mentioned above.

Method of observation

The decoration technique of electron microscopy, combined with Pt-shadowing technique, has been

TABLE 1. MODE OF OCCURRENCE AND MINERALOGICAL CHARACTERISTICS OF ILLITE

Specimen	Polytype	Morphology	Porosity*
Massive metasomatic specimens			
Oguni deposit	2*M*	Irregular	Very low
Naburi deposit	2*M*	Irregular	Very low
Kumano deposit	2*M*	Irregular	Very low
Goto deposit	2*M*	Irregular	Very low
Tsuchihashi deposit-1	2*M*	Irregular	Very low
Tsuchihashi deposit-2	1*M*	Irregular	Very low
Metasomatic specimens			
Nabeyama deposit-1	2*M* + 1*M*	Hexagonal	Low
Nabeyama deposit-2	1*M*	Irregular + Lath	Low
Igi deposit	2*M* + 1*M*	Irregular	Low
Mitoya deposit	2*M*	Irregular	Low
Toyosaka deposit	2*M* + 1*M*	Hexagonal	Low
Metasomatic specimens, Vein form			
Kumogi-1	2*M*	Hexagonal + Irregular	Low
Kumogi-2	2*M*	Hexagonal	High
Daiwa	2*M* + 1*M*	Hexagonal + Lath	High
Kure	1M_d	Irregular	High
Takaya	1*M* + 1M_d	Hexagonal + Lath	High
Iwakuni	2*M*	Hexagonal	High
Fissure filling, vein form			
Iwaya deposit	2*M*	Hexagonal	Low
Hinotani deposit	1*M*	Irregular	Low
Nishikouya deposit	1*M*	Hexagonal + Lath	High
Komaki deposit	2*M* + 1M_d	Hexagonal + Irregular	High
Kuroko deposit	2*M* + 1*M*	Hexagonal + Lath	High
Oshima	1*M*	Hexagonal + Lath	High
Hinotani deposit	1M_d	Irregular	High
Fracture filling, druse			
Kumano deposit	2*M*	Hexagonal + Irregular	High
Kumogi	2*M*	Hexagonal	High
Tsuchihashi deposit-1	2*M*	Hexagonal	High
Tsuchijashi deposit-2	2*M*	Lath	High

* Porosity refers to the aggregation texture.

applied to carry out observations of surface microtopography. The decoration technique was originally developed by Bassett (1958), and since then has been extensively applied to study processes of growth and evaporation of NaCl crystals by Bassett (1958), Bethge (1962, 1967) and Bethge *et al.* (1968), and to silicate minerals by Gritsaenko & Samotoyin (1966), Sunagawa & Koshino (1975), and Sunagawa *et al.* (1975). In the gold-decoration technique, gold is flash-evaporated in the vacuum-evaporation apparatus onto the crystal surfaces, so that minute grains of gold preferentially nucleate along the steps on the surface, thus revealing the

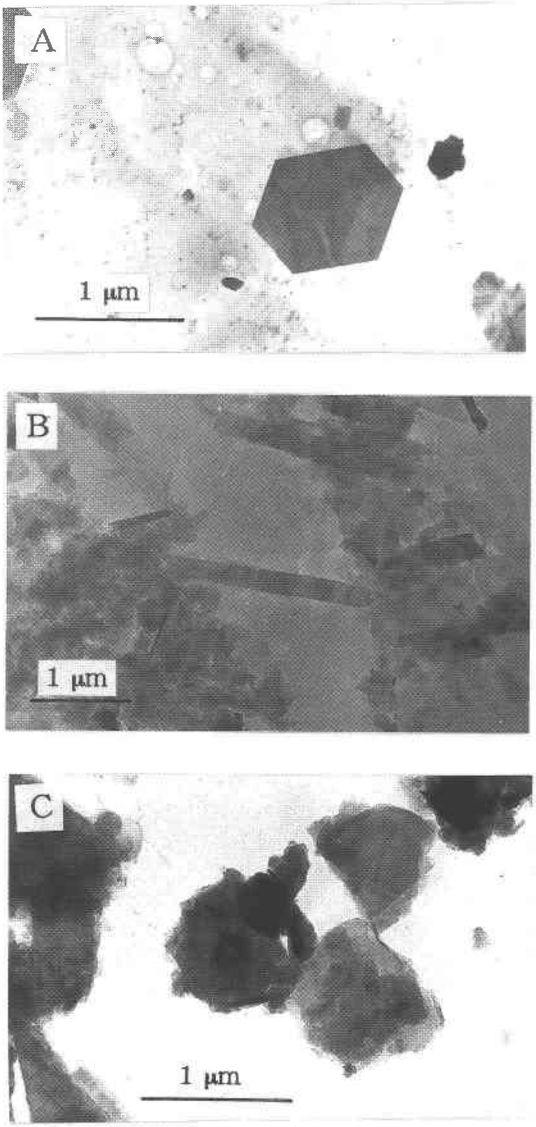

FIG. 1. Three different morphologies of illite crystals. A. Hexagonal-shaped crystal. B. Lath-shaped crystal. C. Irregularly shaped crystal.

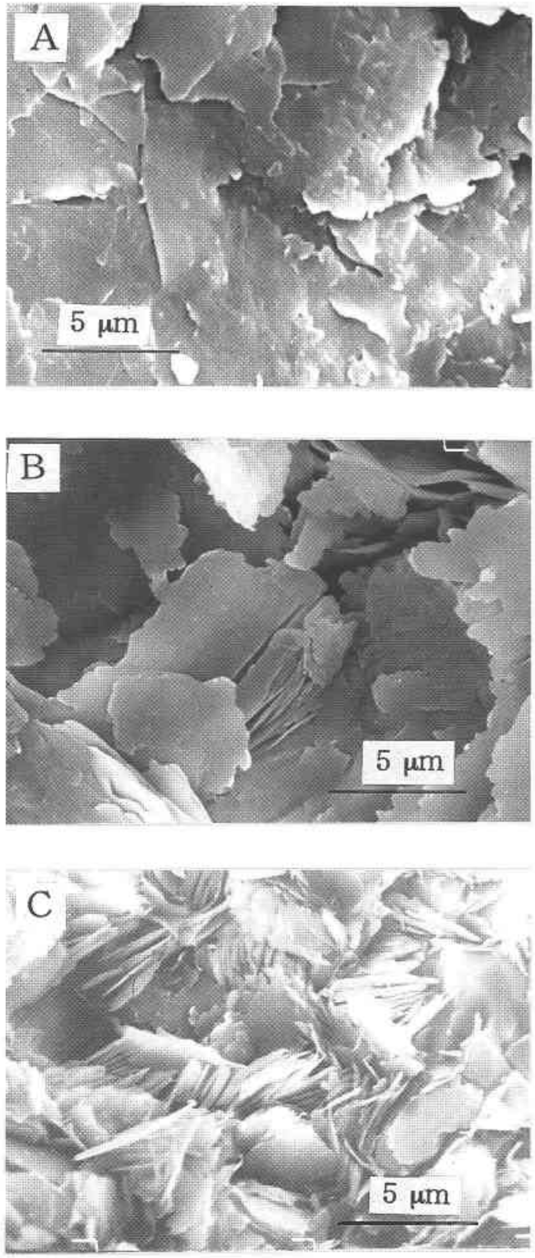

FIG. 2. Aggregation textures of illite crystals. A. Massive metasomatic specimen of low porosity (Oguni deposit in Shimane Prefecture, Japan). B. Metasomatic specimen of moderate porosity (Nabeyama deposit in Shimane Prefecture). C. Fissure-filling specimen of high porosity (vein at Kumogi, Shimane Prefecture).

surface microtopography of crystal faces. Steps of the height of a unit cell or less can be clearly revealed by this technique under the electron microscope, if the coating is adequately applied.

For the Au-decoration technique, the specimen was dispersed in distilled water and collected on a thin cover-glass. After drying, the cover-glass with the specimen was heated in a vacuum of 10^{-4} torr at ~400–500°C for 2–3 hours. Heating the specimen gives a clearer surface without impurity and a higher mobility of Au, enabling selective nucleation of Au along the growth steps (Bassett 1958, Tomura *et al.* 1979). Gold was flash-evaporated from a tungsten coil heater. After a carbon coating was applied, the specimen was removed and immersed in a solution of 10% HF for 3–4 days (depending on each specimen), in order to completely dissolve the specimen. After soaking the remains in distilled water, the thin carbon films with gold grains were collected on copper mesh grids and were then ready for observation with a TEM.

Results

According to Sato (1970) and Sunagawa *et al.* (1975), cleavage surfaces of mica-group minerals exhibit characteristic patterns of gold decoration in which steps may terminate at points on the surface, and many intersect each other. Grains of gold also precipitate more or less unevenly on the surfaces and not only along the steps. We used these criteria to distinguish as-grown surfaces from cleavage surfaces. The electron micrographs in Figures 3 and 4 show that the grains of gold preferentially nucleated along the steps of spirals, thus exhibiting the step patterns of crystal surfaces, but not cleavage patterns. The single steps are assumed to be the height of a unit-cell layer (1 nm; *cf.* Sato 1970, Sunagawa *et al.* 1975, Tomura *et al.* 1979). Kuwahara *et al.* (1998) confirmed by observation that the steps have a height of a unit layer (1 nm) using atomic force microscopy (AFM).

Spiral patterns were observed on a few crystals investigated, though some kinds of patterns are found on all the crystals. The other patterns correspond to features of the cleavage. Both polygonal and circular or malformed circular spirals have been observed (Figs. 3, 4). The separation between the neighboring steps is variable.

Mode of occurrence (*Table* 2)

Illite crystals collected from massive metasomatic specimens are characterized by growth spirals that are circular or malformed circular (Figs. 3, 4). Although metasomatic specimens exhibit circular, malformed circular or polygonal spirals, polygonal growth-patterns are dominant (Fig. 3D). Polygonal spiral patterns are observed on the surfaces of illite crystals formed by the filling of fissures (Fig. 3B).

Polytype (*Table* 3)

Illite crystals of the $2M$ polytype are characterized by the interlacing of polygonal spiral steps (Fig. 3A) or paired steps with circular or malformed spirals (Figs. 4A, B). The $1M$ polytype exhibits polygonal

TABLE 2. RELATIONSHIP BETWEEN MODE OF OCCURRENCE AND PATTERN OF GROWTH

Specimen	Pattern of spirals
Massive metasomatic specimens	
Oguni, Naburi, Kumano, Goto deposits Tsuchihashi deposit-1,2	Circular or Malformed circular
Metasomatic specimens	
Nabeyama deposit-1,2, Igi, Toyosaka deposits	Polygonal
Metasomatic specimens, Vein form	
Daiwa, Kure, Takaya, Iwakuni	Polygonal
Kumogi	Polygonal + Circular
Fissure filling, Vein form	
Iwaya, Hinotani, Nishikouya, Komaki, Kuroko deposits	Polygonal
Ohshima, Hinotani deposit	Circular or Malformed circular
Fracture filling, Druse	
Kumano, Tsuchihashi deposits, Kumogi	Polygonal

TABLE 3. RELATIONSHIP BETWEEN POLYTYPE AND GROWTH PATTERN

Specimen	Pattern of spirals
Polytype $2M$	
Oguni, Naburi, Kumano, Goto deposits Tsuchihashi deposit-1, Kumogi	Circular or Malformed circular
Tsuchihashi deposit-2, Nabeyama deposit-1 Igi, Mitoya, Toyosaka, Iwaya deposits Komaki deposit, Daiwa, Kuroko deposit	Polygonal
Polytype $1M$	
Nabeyama deposit-2, Igi, Toyosaka, Hinotani, Nishikouya deposits Daiwa, Takaya, Kuroko deposit	Polygonal
Ohshima	Circular or Malformed circular
Polytype $1M_d$	
Kure, Takaya	Polygonal
Hinotani deposit	Circular or Malformed circular

(Figs. 3C, D) or malformed circular spirals with a single step (Fig. 4C). However, $1M_d$ illite shows a complex step-pattern, as shown in Figure 4D.

Morphology (*Table* 4)

Crystals exhibit hexagonal, lath-shaped or irregular morphology (Fig. 1). The hexagonal or lath-shaped crystals are characterized by polygonal spirals (Figs. 3A, D), whereas irregular crystals show circular or malformed circular growth-spirals (Figs. 4A, C).

Aggregation texture (*Table* 5)

Based on SEM observation, crystals in massive metasomatic specimens are aggregated, with very low porosity, as shown in Figure 2A. On the other hand, crystals formed in metasomatic specimens, druses and

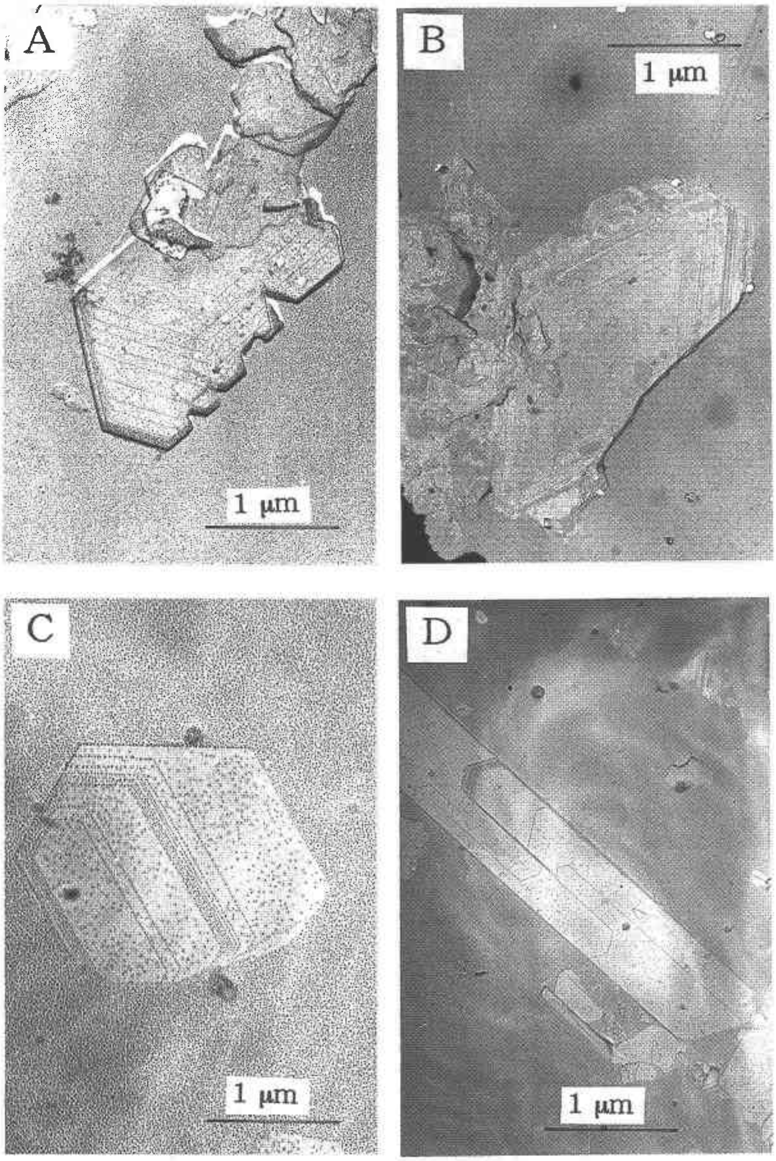

FIG. 3. Surface microtopography (polygonal patterns) of illite after gold decoration. A. Alteration zone in Kuroko deposit. B. Druse in granite (Shimane Prefecture, Japan). C and D. Tsuchihashi deposit in Okayama Prefecture, Japan.

fissures are aggregated with a higher porosity (Fig. 2B). Crystals in specimens with a higher porosity are characterized by polygonal spiral-growth patterns (Fig. 3), whereas specimens with a lower porosity exhibit circular or malformed circular spiral patterns (Figs. 4A, B).

Discussion

Morphologies of growth spirals are classified into polygonal and circular or malformed circular, with various ranges of step separation. According to the

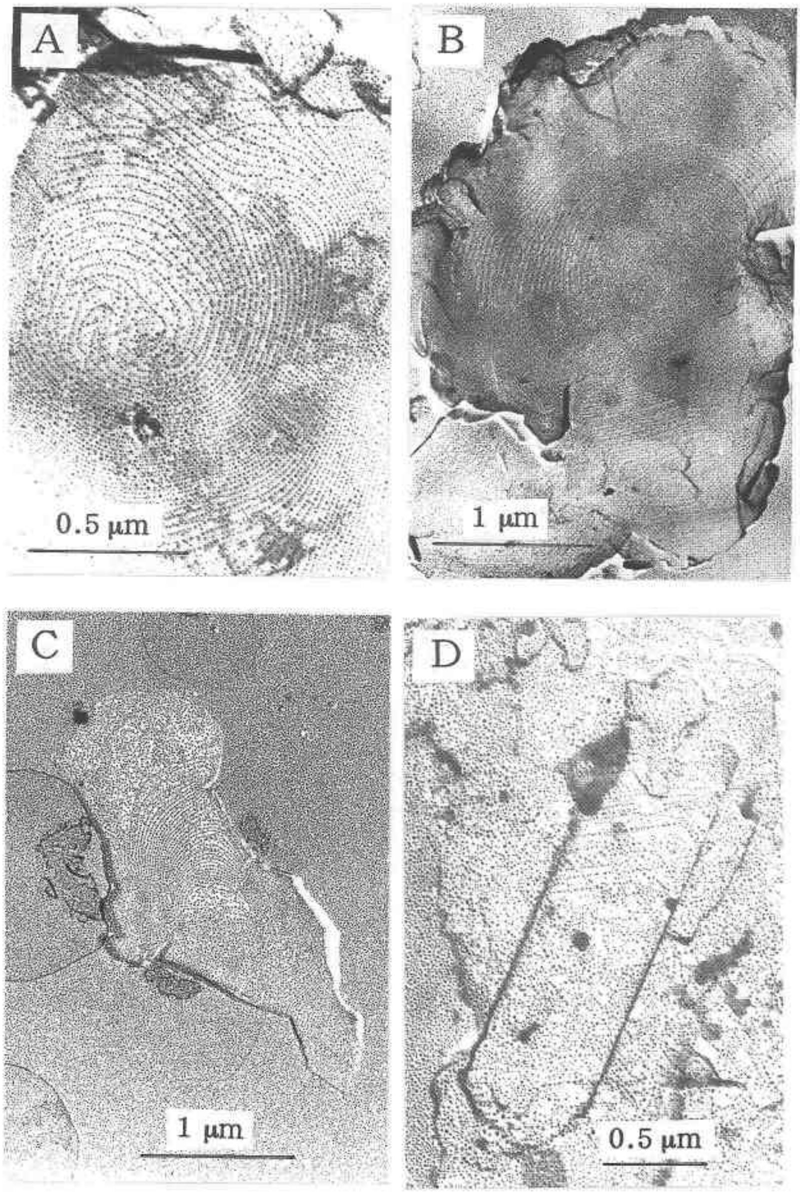

FIG. 4. Surface microtopography (circular and complex patterns) of illite after gold decoration. A and B. Oguni deposit in Shimane Prefecture, Japan. C. Hinotani deposit in Shimane Prefecture, Japan. D. Vein in Kure, Hiroshima Prefecture, Japan.

TABLE 4. RELATIONSHIP BETWEEN CRYSTAL MORPHOLOGY OF ILLITE AND GROWTH PATTERN

Specimen	Pattern of spirals
Hexagonal plate	
Nabeyama deposit-1, Toyosaka, Iwaya, Nishikouya, Komaki deposits, Daiwa, Kumano deposit, Kumogi, Tsuchihashi deposit-1, Kuroko deposit, Takaya	Polygonal
Lath-shaped plate	
Nabeyama deposit-2, Nishikouya deposit, Daiwa, Takaya, Ohshima, Takaya, Tsuchihashi deposit-2, Kuroko deposit	Polygonal
Irregular plate	
Oguni, Naburi, Kumano, Goto, Hinotani, Kumogi deposits	Circular or Malformed circular
Igi, Mitoya, Komaki deposits Kure	Polygonal

TABLE 5. RELATIONSHIP BETWEEN AGGREGATION TEXTURE OF ILLITE PARTICLES AND GROWTH PATTERN

Specimen	Pattern of spirals
High porosity	
Nishikouya, Komaki deposits, Daiwa, Kure, Takaya, Kumano deposit Tsuchihashi deposit-1,2, Kuroko deposit	Polygonal
Low porosity	
Nabeyama deposit-1,2, Igi, Toyosaka, Iwaya deposits	Polygonal
Hinotani, Kumogi deposits	Circular or Malformed circular
Very low porosity	
Oguni, Naburi, Kumano, Goto, Tsuchihashi deposits	Circular or Malformed circular

microtopographies, the morphology of growth patterns is closely related to the mode of occurrence (massive metasomatic, metasomatic and fissure filling), aggregation textures of crystals (lower and higher porosities in specimen), and morphology of crystals (hexagonal, lath and irregular shape).

According to Sunagawa *et al.* (1975) and Tomura *et al.* (1979), illite that is assumed to have been directly precipitated from hydrothermal solutions in open fractures such as druses (high-porosity environment) is characterized by the development of polygonal spirals. In contrast, crystals of illite formed in massive metasomatic rocks (lower porosity) by hydrothermal activity exhibit a circular or malformed circular outline. Their step separations vary from locality to locality. Circular or malformed circular spirals have, in general, a narrower separation of steps than in the case with polygonal spirals. It is natural to assume conditions of higher temperature or higher supersaturation or both for circular growth-spirals than polygonal ones.

Hexagonal and lath-shaped crystals are not observed in massive metasomatic samples, whereas they are commonly found in illite directly precipitated from solution. All hexagonal and lath-shaped crystals are characterized by polygonal spirals. In general, irregular plates are characterized by circular or malformed circular patterns.

Polytypes identified by X-ray powder method are indicated in Table 1 for all samples; $2M$, $1M$ and $1M_d$ have been identified. Crystals belonging to the $2M$ polytype invariably exhibit either interlaced patterns (Fig. 3A) or paired steps (Figs. 4A, B), whereas those belonging to $1M$ and $1M_d$ do not show such step patterns (Figs. 3C, 4C, D). As has been demonstrated with clay minerals by Sunagawa *et al.* (1975) and Sunagawa (1984), when a structure contains zigzag stacking, one can expect interlaced pattern or paired steps to appear by a spiral-growth process. Interlaced patterns and paired steps will appear, depending upon the symmetries of the elemental sheet, their mode of stacking, and the number of stacked sheets, owing to the difference in rate of advance in the same direction among the successive elemental sheets.

According to Sunagawa (1982) and Sunagawa & Bennema (1982), whether a spiral takes a polygonal or a circular form is determined by the roughness of the spiral step, and thus is related to the α-factor and the chemical potential difference, $\Delta\mu/kT$. Here, α is expressed, in general form, as $\alpha = \xi[\Phi sf - 1/2(\Phi ss + \Phi ff)]/kT$, where Φss is the bond energy in the solid, Φsf, the interaction energy between solid and fluid components, Φff, the interaction energy between the neighboring fluid–fluid components. ξ is an orientational factor, T is (absolute) temperature, and k is Boltzmann's constant. As the value of α becomes smaller, the step becomes rougher, as it contains a higher density of kinks. The step, then, advances isotropically, *i.e.*, with the same rate in all directions in that plane, resulting in a circular spiral. On the other hand, if the value of α becomes larger, the step becomes smoother, it contains a lower density of kinks, and the rate of advance is now strongly controlled by the crystallographic anisotropy, resulting in a polygonal spiral. Therefore, polygonal spirals are expected at a lower temperature at constant supersaturation or at lower supersaturation (lower difference in chemical potential) under isothermal conditions than for circular spirals. This generalization may be valid if crystals grow from the same phase (*e.g.*, hydrothermal solution) and if the influence of impurities is neglected.

According to the same authors, the step separation (λo) of a spiral is related to the radius r* of the two-dimensional critical nucleus, which is defined by the edge free energy per elemental sheet, γ, the chemical potential difference $\Delta\mu$ and the molar volume σ: $\lambda o = 19r^* = 19\gamma\ \sigma/\Delta\mu$. Since γ is assumed to be equal if crystals grow from a similar phase, if T is assumed constant, and if the effect of impurities is neglected, we may expect a narrower separation of steps for crystals grown from a solution at higher supersaturation.

The circular or malformed circular spiral patterns have a narrower separation of steps than polygonal spiral patterns. Illite in massive metasomatic specimens is considered to have formed under conditions of higher temperature or higher supersaturation of hydrothermal solutions (or both) than those in the other modes of occurrences, *i.e.*, metasomatic, druses and fissure fillings. Depending on the characterization of subtle differences in surface microtopography in illitic materials from each locality, details about their genesis can be elucidated.

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