

# SCAN ELECTRON MICROGRAPHS OF KAOLINS COLLECTED FROM DIVERSE ENVIRONMENTS OF ORIGIN—V. KAOLINS COLLECTED IN AUSTRALIA AND JAPAN ON FIELD TRIPS OF THE SIXTH AND SEVENTH CLAY CONFERENCES

W. D. KELLER

Department of Geology, University of Missouri, Columbia, MO 65201, U.S.A.

(Received 18 March 1977; and in final form 5 April 1977)

**Abstract**—Scan electron micrographs are shown of the textures of flint clays, tonsteins, kaolin associated with combustion-metamorphism, sedimentary kaolin, and dickite from the Sydney Basin of Australia. The textures of the flint clays and tonsteins indicate those clays were derived largely from volcanic products. The clay samples were collected at stops on the Kaolin Excursion No. 4 of the August, 1976, International Geological Congress.

In like manner, kaolin samples were collected in Japan from stops made on the field excursions of the Seventh Conference of the Committee on Correlation of Age and Genesis of Kaolin which met in Tokyo, September, 1976. SEMs illustrate representative kaolins of Japan, including Gaerome and Kibushi types of kaolin, Roseki ('wax stone') pyrophyllite and dickite, hydrothermal kaolin at Itaya, and flint clay at Iwate. Varied morphologies of halloysite, including spherical halloysite, from the Yamaka open-pit at Naegi are micrographed.

Word descriptions of the textures are frustratingly inadequate in comparison to what may be seen at a glance in the micrographs—hence, the abstract becomes in reality a rapid view of the SEMs.

## INTRODUCTION

Kaolin, defined as an earthy rock containing significant amounts of the kaolin minerals, embraces in petrology a wide variety of lithologies, textures, processes of genesis and parent materials. Kaolin therefore includes, besides ceramic and paper-coating white clays, other rocks such as flint clay, kaolinitic flint-like clay, tonsteins, ball clays and others.

Flint clays have achieved important industrial, as well as geologic, recognition because of the high-quality refractories produced from them. In the U.S.A., Scotland and South Africa, parent materials for flint clay dominantly were derived as weathered residues from an earlier cycle of sedimentary rocks (Keller, 1968). The textures of these clays have a distinctive similarity.

In Australia and Japan, however, the parent materials for flint clays and flint-like clays were dominantly volcanic in origin. Because the textures of those clays typically retain some relics of their heritage, they are dissimilar to the textures of most flint clays in the U.S.A.

The objectives of this report will, therefore be four-fold: (1) to record the textures of flint clays and tonsteins from the Sydney Basin, Australia, and of important deposits in Japan; (2) to illustrate textures of kaolins having a volcanic heritage; (3) to recognize by micrography the major role of volcanic material in argillic sediments in certain parts of Australia and Japan; and (4) to contrast the flint-clay petrology of

those countries with that in the U.S.A., Scotland and probably South Africa (Vereeniging district).

The Australian examples were collected from the deposits and occurrences visited while on Kaolin Field Excursion No. 4 conducted by Prof. F. C. Loughnan (1976) before the 1976 International Geological Congress, and the Symposium of the 6th Conference of the Committee on Correlation of Age and Genesis of Kaolin, CCAGK (referenced under Patterson, *Summary of Proceedings, CCAGK and Australian Clay Minerals Society*, 1976). The Japanese samples were collected while participating in the field trips led by Prof. H. Minato, and colleagues N. Fujii, A. Iijima, K. Nagasawa, H. Imai, and the Nippon Mining Co. hosts (referenced under Guide to Field Investigations, Tokyo, led by H. Minato, 1976; and Iwate Mine, Post-Symposium Field Trip, Iijima, 1976), which were taken after the meeting of the 7th Symposium of CCAGK in Tokyo (referenced under Seventh Symposium on Genesis of Kaolin, Minato and Shimoda, 1976).

The textures micrographed are those present on a freshly broken surface of the clay with no further processing than thinly coating them with gold to carry away excess charge from the electron beam.

## TONSTEINS FROM AUSTRALIA

The term 'tonstein', because of literal translation as 'clay stone or rock', has prompted some relatively

flexible applications of the name to various clay rocks, but preferred and accepted current usage was comprehensively summarized orally by Loughnan, while documented by Bohor and Pillmore (1976), who quote, and accept, the definition by Williamson (1970, p. 119):

“Tonsteins... dense mudstones containing kaolinite aggregates and crystals within essentially similar matrices. Usually occurring as thin beds less than 6 cm, they are typically developed in the coal measures facies. Although rare, they are of widespread distribution and form isochronous correlative horizons which have been extensively utilized in the European coal fields.”

Bohor and Pillmore further state that, “Field and mineralogical evidence indicate that the kaolinitic partings in these coals are actually altered volcanic ash layers” (p. 177).

The first SEM in this paper is from a tonstein in the Permian Wongawilli Coal on Meryla Pass about 40 air-km south of Mittagong, in the southern part of the Sydney Basin, stop 15 in the Guidebook by Loughnan (1976), Figure 1 (3308, 5000×). In Figure 2 (3646, 5000×) is a SEM from a tonstein in the Jurassic Digilak Formation. This formation conformably overlies the clay exposed in the nearby Merrygoen Quarry which also will subsequently be described.

The texture in these two examples is typical of other tonsteins I have micrographed. The crystals are small, apparently nearly equi-dimensional, and closely packed; black coaly material may be included, and porosity appears to be low. Most definitive, however, are the ragged to scalloped edges of the crystals which give an overall pattern suggestive of micro-oak or maple-leaf texture, as is developed in smectite, a common product of weathered ash (Borst and Keller, 1969). To illustrate better what is meant by this pattern, an example will be shown of another clay possessing the same texture, and known from independent geologic evidence to be derived from volcanic ash.

Figure 3 (2622, 5000×) is from a kaolin which can be traced directly, within a few meters, into volcanic ash in Oregon, U.S.A. In Figure 4 (2407, 10,000×) is shown kaolin derived from volcanic ash at a quarry and mine at Berzdorff, GDR (East Germany). Demonstrating that volcanic-ash parent material is the control for the texture in Figure 4, the kaolin developed from granodiorite only a few meters away within the same quarry is observed to be distinctly different, Figure 5 (2389, 2000×). The last 3 SEMs are repeated from part III of this kaolin-texture series (Keller, 1976, part III).

#### FLINT CLAY AND FLINT-LIKE CLAY, AUSTRALIA

Important production of flint clay comes from the

Merrygoen Quarry (Loughnan's stop 7, 1976) opened in the Jurassic Ukebung Creek Claystone where it crops out on the northwest margin of the Sydney Basin, next to the Great Artesian Basin. Flint clay from the lower, pelletal bed is shown in SEM, Figure 6 (3642, 3000×), and from the top bed, highest refractory (PCE 34) flint clay, Figure 7 (3640, 5000×). The similarity in texture to that of the tonstein, Figure 2, is self-evident. The cleavage faces of several sub-hedral books of kaolinite are present in Figure 7. Such crystals are relatively common in high-PCE flint clay from the Pennsylvanian-age deposits in the U.S.A. and Europe. One can speculate that this clay was more highly matured, or 'digested-diagenized', toward recrystallized kaolinite than was the lower-PCE clay in the lower bed, Figure 6.

A thickness of about 10 m of flint clay occurs in the Permian Illawarra Coal Measures on Cambewarra Mountain in the southern part of the Sydney Basin, about 75 miles south of Sydney, Loughnan's Stop 16. Loughnan wrote of these clay rocks, “Many of the clay aggregates, particularly those forming the conglomeratic clay rocks, have residual volcanic textures...” (p. 46). Six samples were collected at nearly regular intervals, or at major changes in lithology, up the 10-m section. Although the clay is much the same, different magnifications in the SEMs bring out the variety of morphologies present.

From the lower member, Figure 8 (3289, 5000×) shows the dominant, 'ragged-edge' morphology which is accompanied by sporadic basal cleavages of kaolin books which are more common in flint clays from the U.S.A. In Figure 9 (3279, 5000×), taken from the next bed above in the stratigraphic section, very fine crystals are associated with large, compactly interlocking, euhedral kaolin particles. The latter types have been observed in some very compact flint or flint-like clays in Mexico.

In Figure 10 (3275, 10,000×), at higher magnification, the finely granular, irregularly edged pattern earlier referred to is brought out. The same pattern can be detected, even at lower magnification, however, as is seen in the two preceding and subsequent micrographs. The center layer in the flint-clay formation clearly exhibits the fringed-edge clay at 5000×, as in Figure 11 (3363). The SEMs of the clay in the upper beds show, at 3000×, the pattern of packing present in essentially all of this clay deposit [Figures 12 and 13 (3358, 3357)]. These six samples from the deposit on Cambewarra Mountain are typical of the flint clay derived from volcanic material in the Sydney Basin.

On Meryla Pass, Stop 15, some 25–30 km northwest of the Cambewarra Mountain locality, the thickness of the clay is reduced to 3.7 m. The texture in two prominent beds is similar in essentials, and likewise similar to that on Cambewarra Mountain [Figures 14 and 15 (3314, 3000×; 3316, 3000×)].

A Triassic claystone, the Garie Member of the Bald Hill Claystone, is exposed in the National Park about

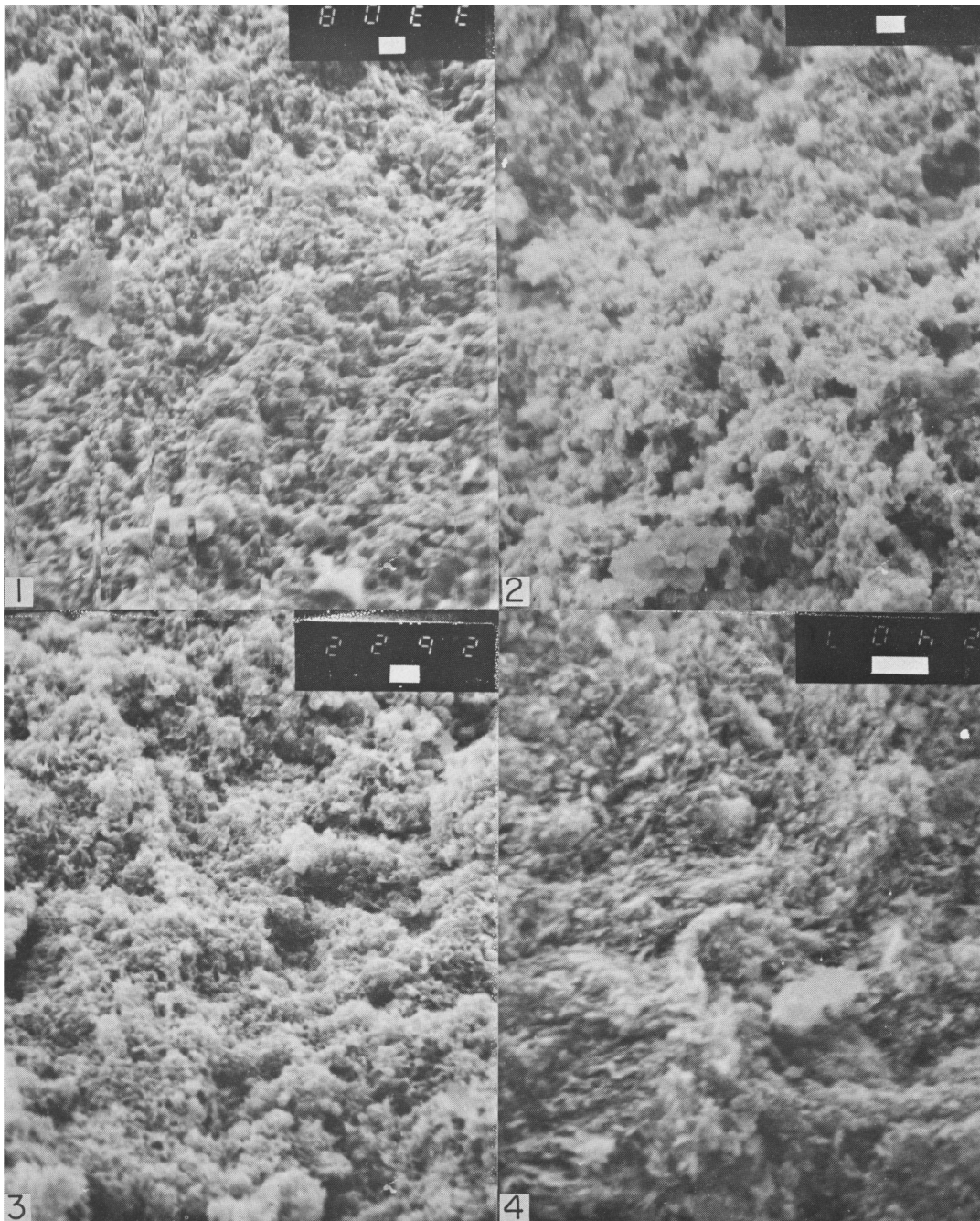


Figure 1. SEM of tonstein in Permian Wongawilli coal, Meryla Pass, Australia, 3308, 5000 $\times$ . The horizontal length of the white bar under the number of the negative represents 1  $\mu$ m unless otherwise designated by a number.

Figure 2. SEM of tonstein in Jurassic Digilak Formation, NW part of Sydney Basin, Australia, 3646, 5000 $\times$ .

Figure 3. SEM of flint clay, Kings locality no. 11, Sublimity, OR, U.S.A., 2622, 5000 $\times$ . This clay can be traced into volcanic ash within a few meters.

Figure 4. SEM of kaolin weathered from tuff, Berzdorff mine, as in Figure 5, GDR (East Germany), 2407, 10,000 $\times$ .

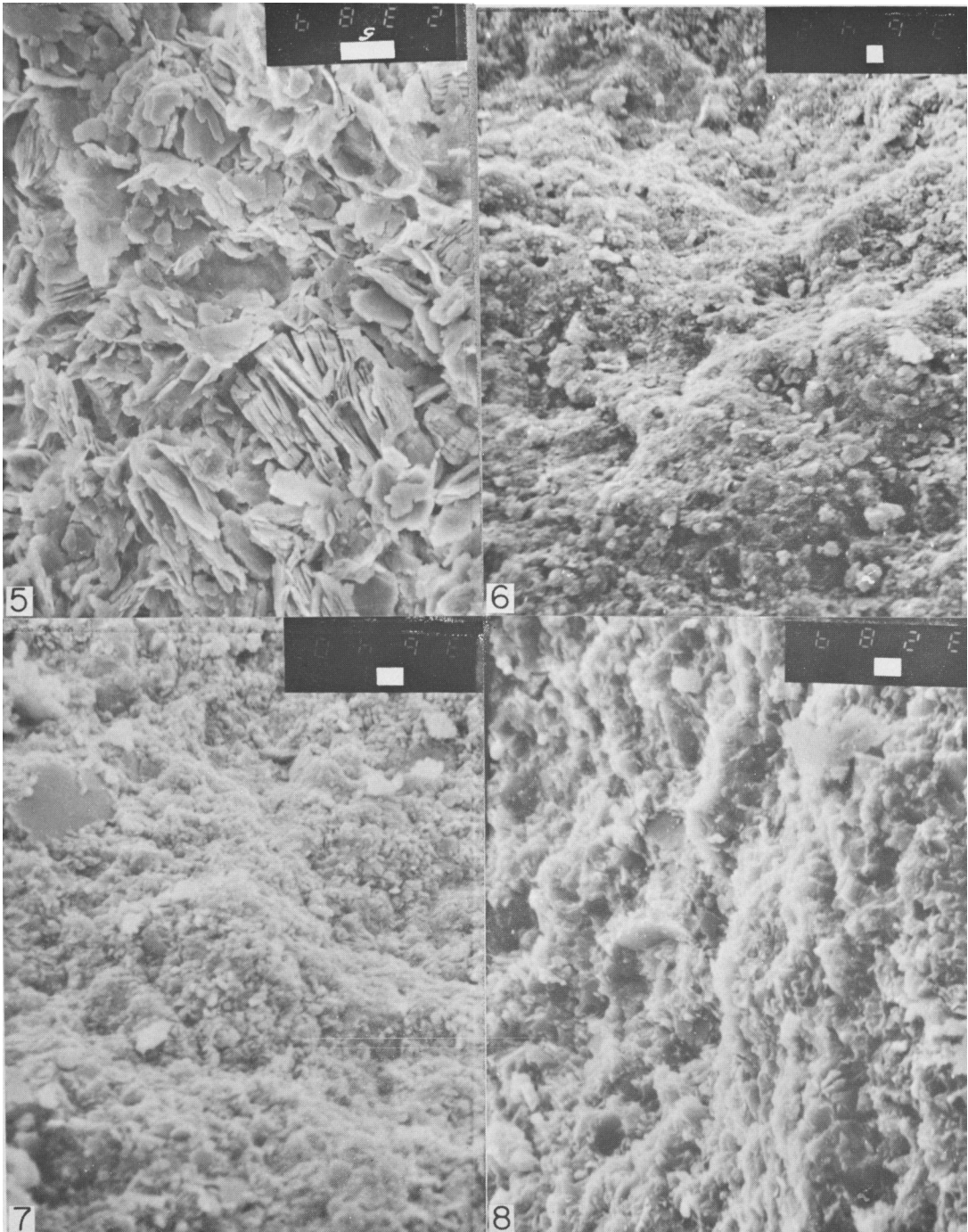


Figure 5. SEM of kaolin weathered from granodiorite, Berzdorff mine, as in Figure 4, GDR (East Germany), 2389, 2000  $\times$ .

Figure 6. SEM of pelletal flint clay, Merrygoen quarry, Sydney Basin, Australia, 3642, 3000  $\times$ .

Figure 7. SEM of high-refractory flint clay, Merrygoen quarry, as in Figure 6, 3640, 5000  $\times$ .

Figure 8. SEM of lower part of flint clay, Permian Illawarra Coal Measures, Cambewarra Mountain, southern part of Sydney Basin, Australia, 3289, 5000  $\times$ .

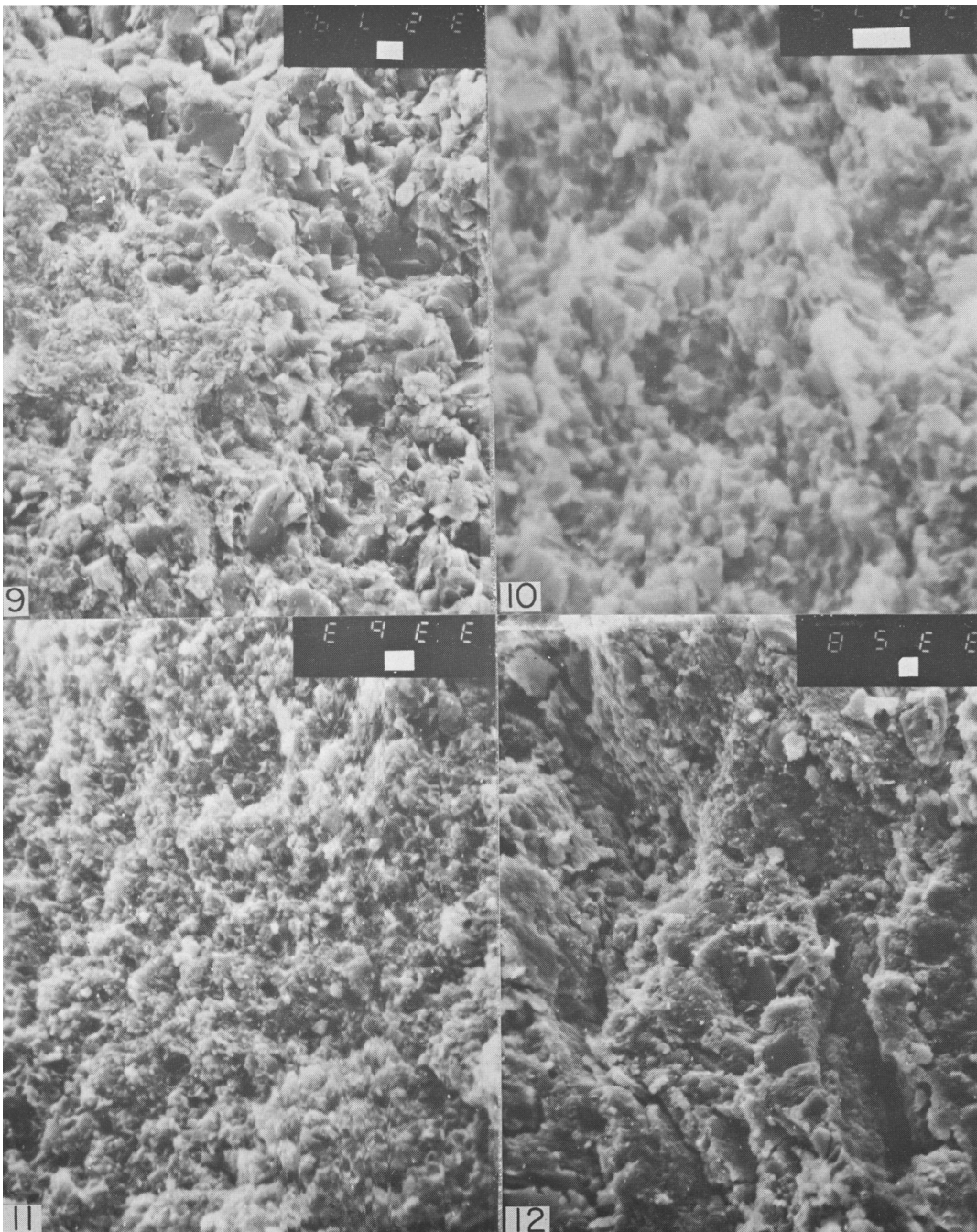


Figure 9. SEM of next higher layer of flint clay in deposit, as in Figure 8, 3279, 5000 $\times$ .

Figure 10. SEM of the finely crystalline clay in Figure 9 at 10,000 $\times$ , showing more detail of texture, 3275.

Figure 11. SEM of center layer of Cambewarra Mountain clay deposit, as of Figure 8, 3363, 5000 $\times$ .

Figure 12. SEM of upper beds, same clay deposit as in Figure 8, 3358, 3000 $\times$ .

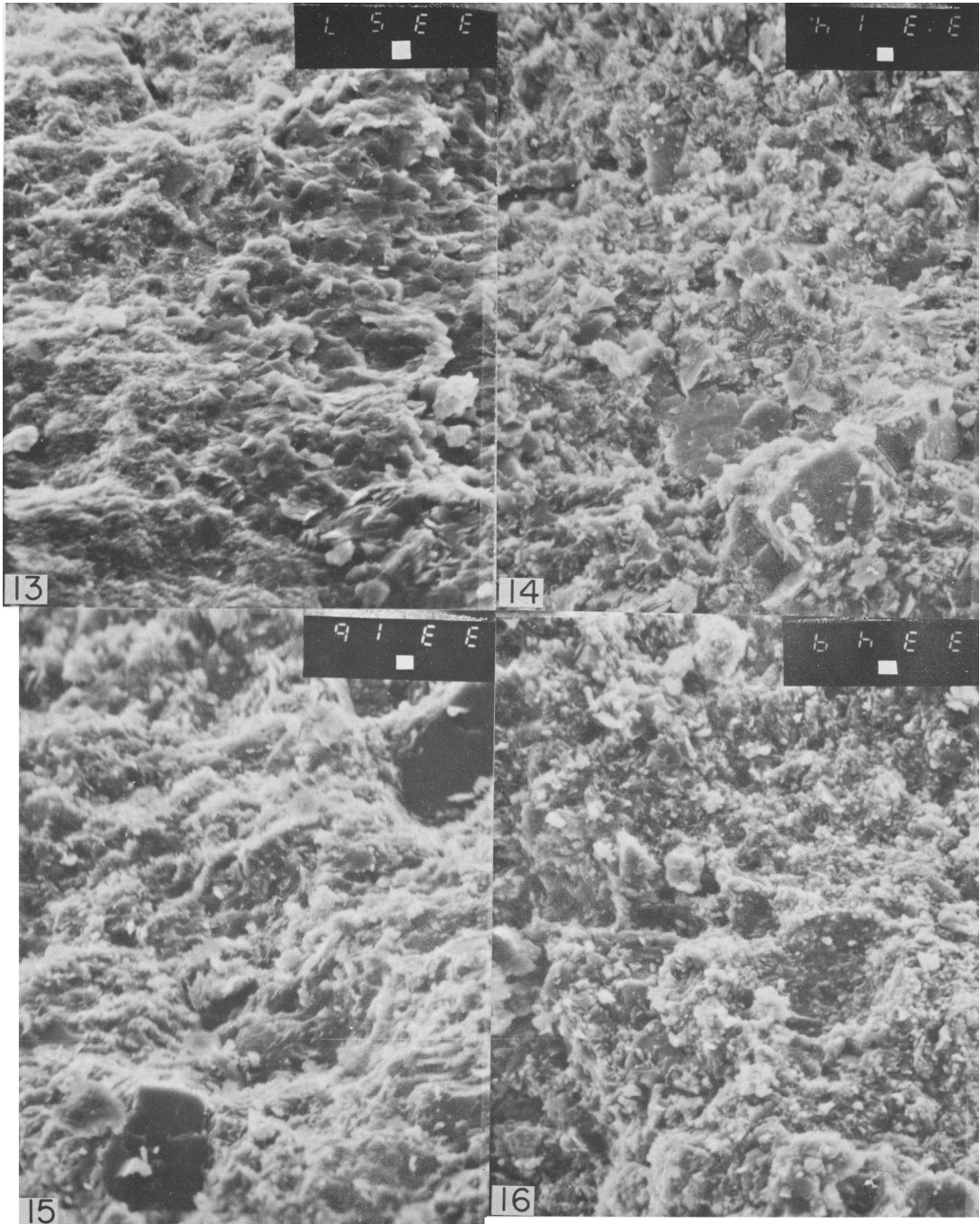


Figure 13. SEM of uppermost part of Cambewarra Mountain clay deposit, as of Figure 8, 3357, 3000 $\times$ .

Figure 14. SEM of clay in lower bed, in Permian Illawarra Coal Measures, Meryla Pass, Sydney Basin, Australia, 3314, 3000 $\times$ .

Figure 15. SEM of clay in upper bed, same deposit as Figure 14, 3316, 3000 $\times$ .

Figure 16. SEM of oolitic clay member of the Triassic Garie Member, National Park, south of Sydney, Australia, 3349, 3000 $\times$ .

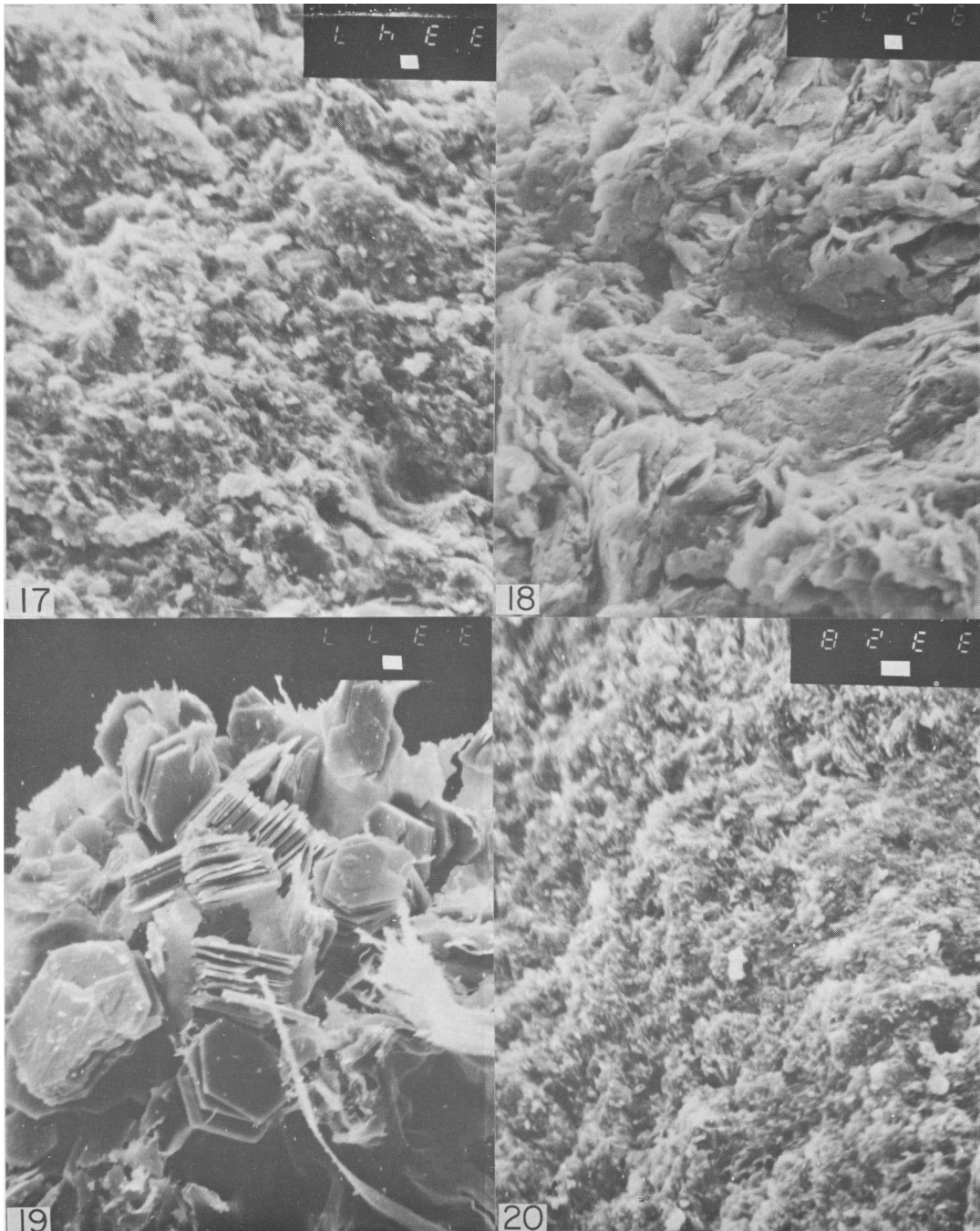


Figure 17. SEM of fine-grained clay member of the deposit as of Figure 16, 3347, 3000 $\times$ .

Figure 18. SEM of kaolinite from the Home Rule deposit, overlying the Late Carboniferous Gulgong Batholith, 3272, 3000 $\times$ .

Figure 19. SEM of dickite from pores in the Triassic Hawkesbury Sandstone, south of Sydney, Australia, 3377, 3000 $\times$ .

Figure 20. SEM of flint clay at 'The Pimple', Muswellbrook Anticline, about 100 km northwest of Sydney, Australia, 3328, 5000 $\times$ .

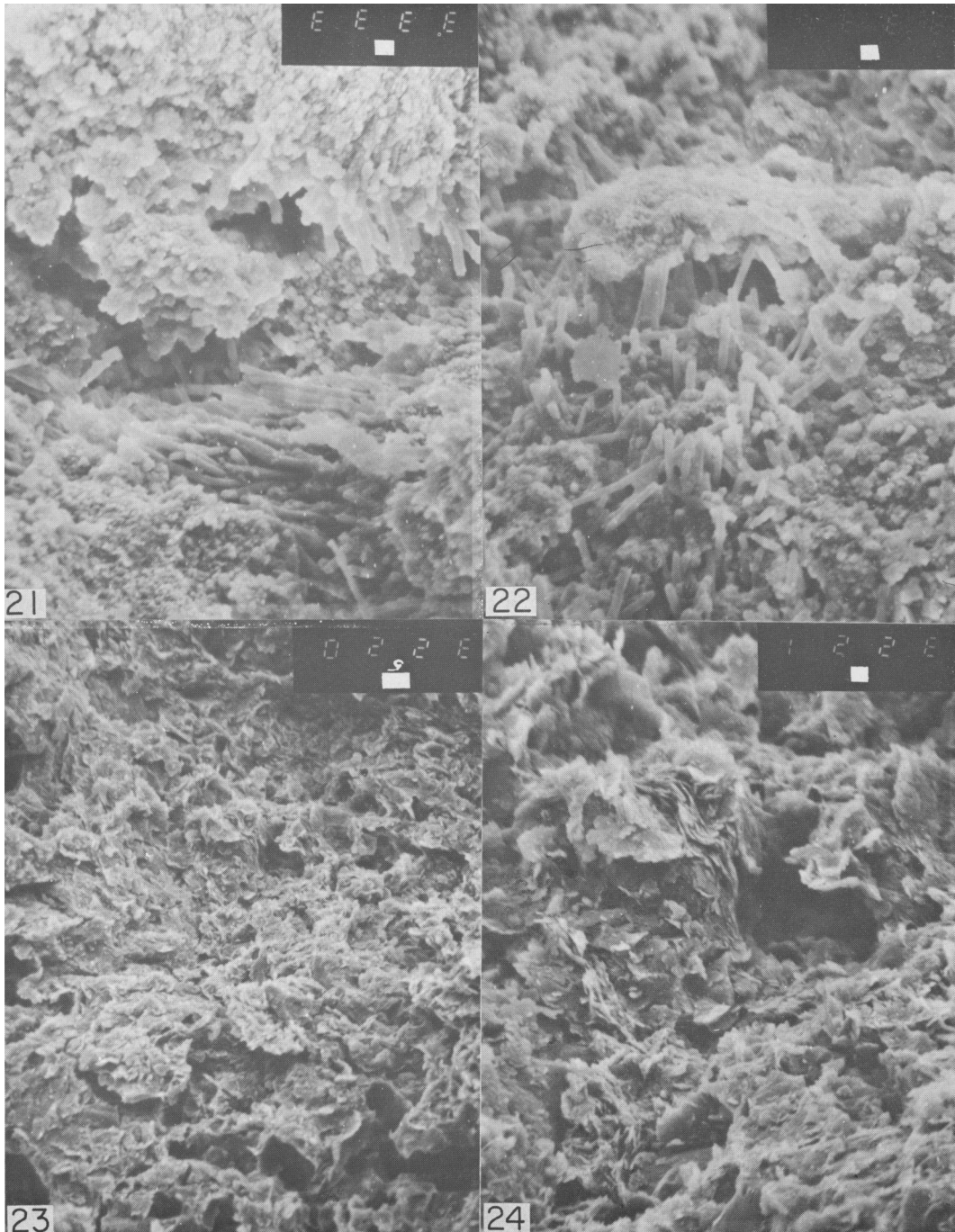


Figure 21. SEM, horizontal view, of halloysite elongates associated with flint clay and a burned out coal bed, at 'The Pimple', 3333, 3000 $\times$ .

Figure 22. SEM, vertical view, of halloysite elongates at 'The Pimple', 3336, 3000 $\times$ .

Figure 23. SEM of Gaerome kaolin, Maruhara Mine, near Nagoya, Japan, 3220, 1000 $\times$ .

Figure 24. SEM, portion of Figure 23 enlarged to 3000 $\times$ , to show relationship of kaolin flakes, 3221.



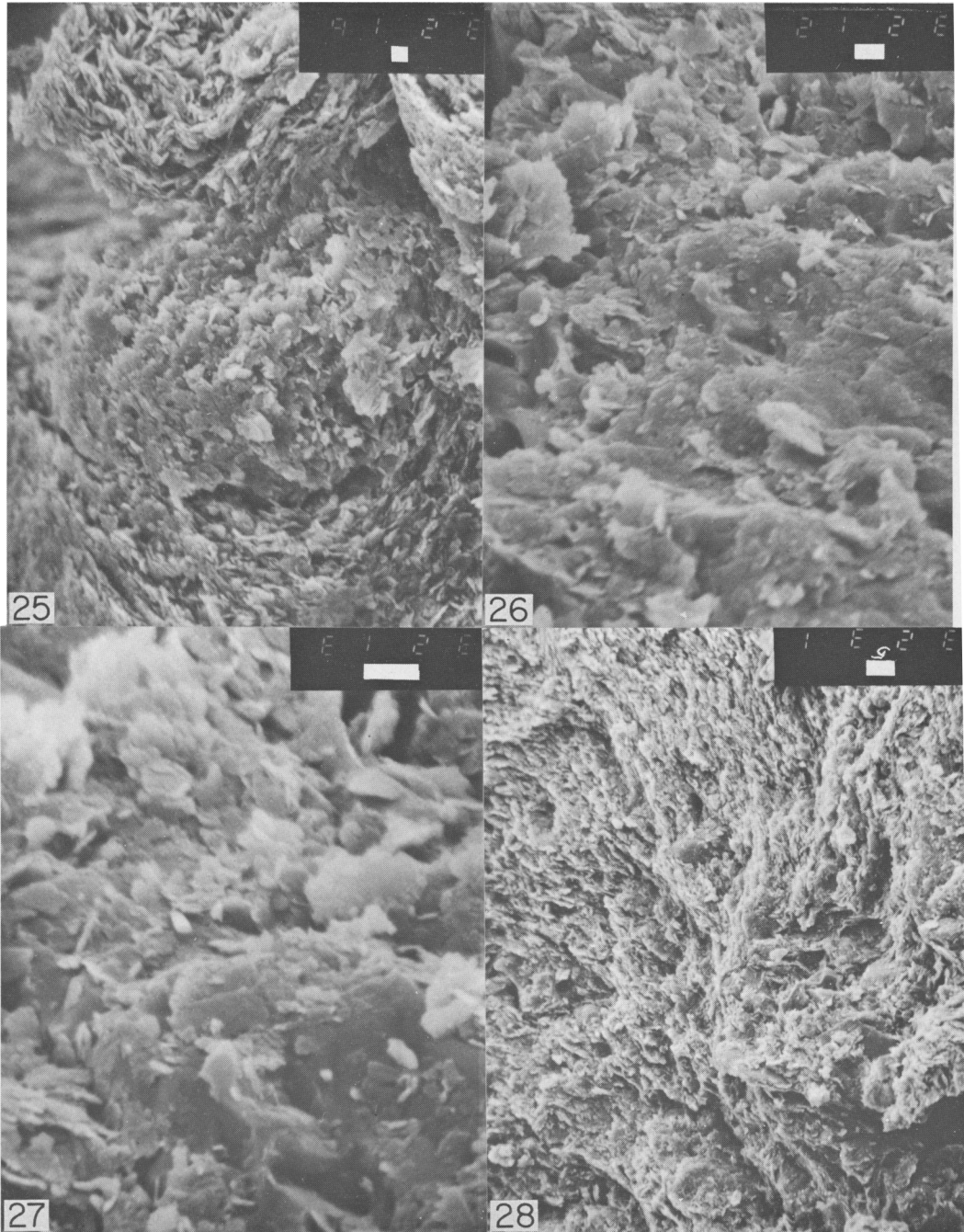


Figure 25. SEM of aggregates, possibly Kira (?), Maruhara Mine, 3216, 3000 $\times$ .

Figure 26. SEM of Gaerome kaolin, Maruhara Mine, 3212, 5000 $\times$ .

Figure 27. SEM, higher magnification, of Gaerome kaolin, Maruhara Mine, 3213, 10,000 $\times$ .

Figure 28. SEM of Kibushi kaolin clay, Maruhara Mine. Finely crystalline clay, thinly laminated, with some contorted or 'swirl' pattern as is typical of ball clay, 3231, 1000 $\times$ .

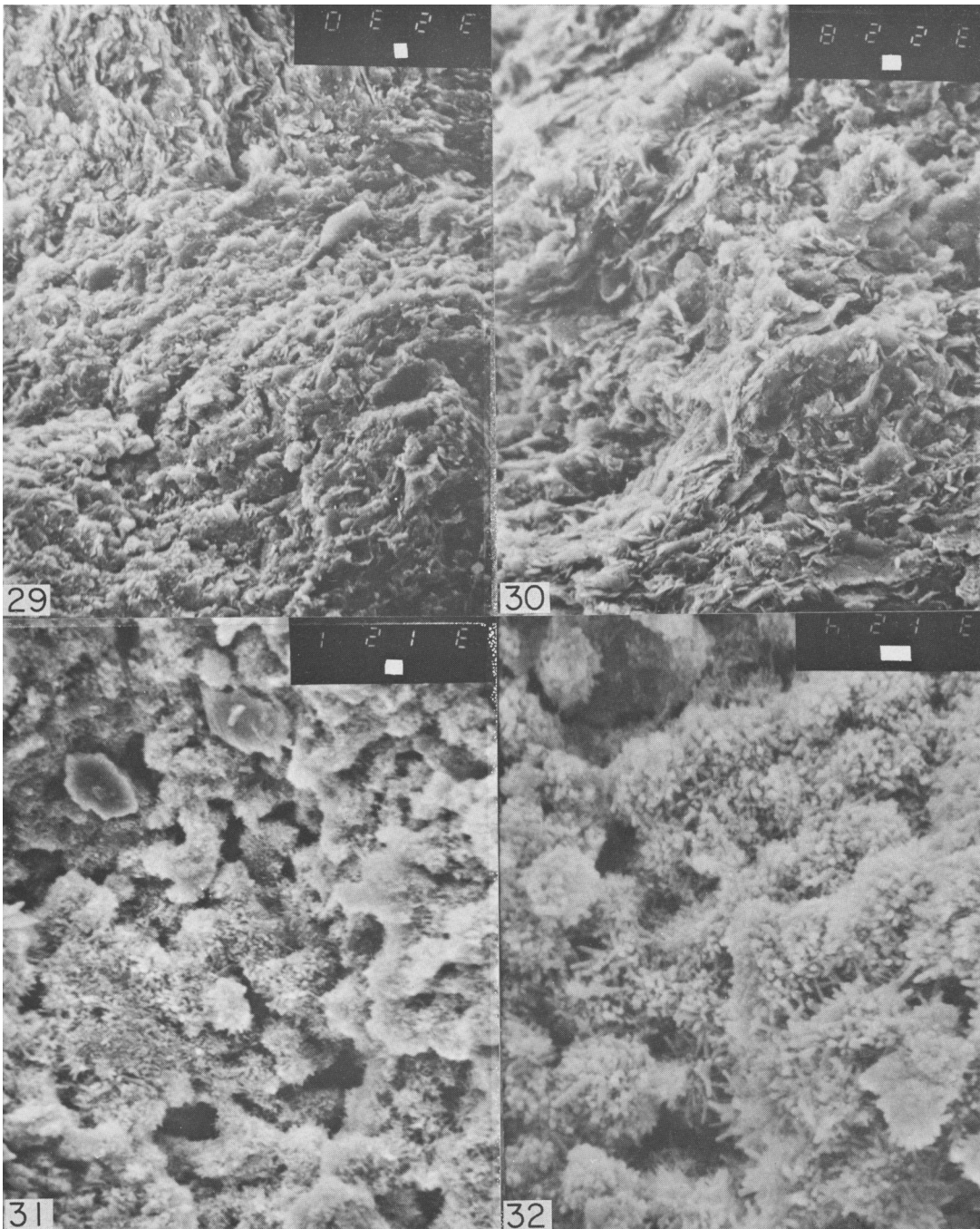


Figure 29. SEM of Kibushi kaolin clay, Maruhara Mine. Tight, face-to-face packing, change in orientation of clay flakes within a short distance, 3230, 2000 $\times$ .

Figure 30. SEM of Kibushi kaolin clay, small, thin flakes in face-to-face orientation, but not 'book-type' kaolin crystals, pattern of bend in crystals extends to micro dimensions, 3228, 3000 $\times$ .

Figure 31. SEM of shattered feldspar fragments (gruss) from which radiate elongate, halloysite crystals, a weathering product. From the Yamaka pit, near Naegi, Japan, 3121, 3000 $\times$ .

Figure 32. SEM of elongate kaolin (halloysite) crystals at 5000 $\times$  magnification. Most of the elongates are of the common stubby type, but a few approach the sharp-pointed 'spike' type which have been micrographed at Spruce Pine, NC, U.S.A. [Figure 24, Part I, Keller (1976)], 3124, 5000 $\times$ .

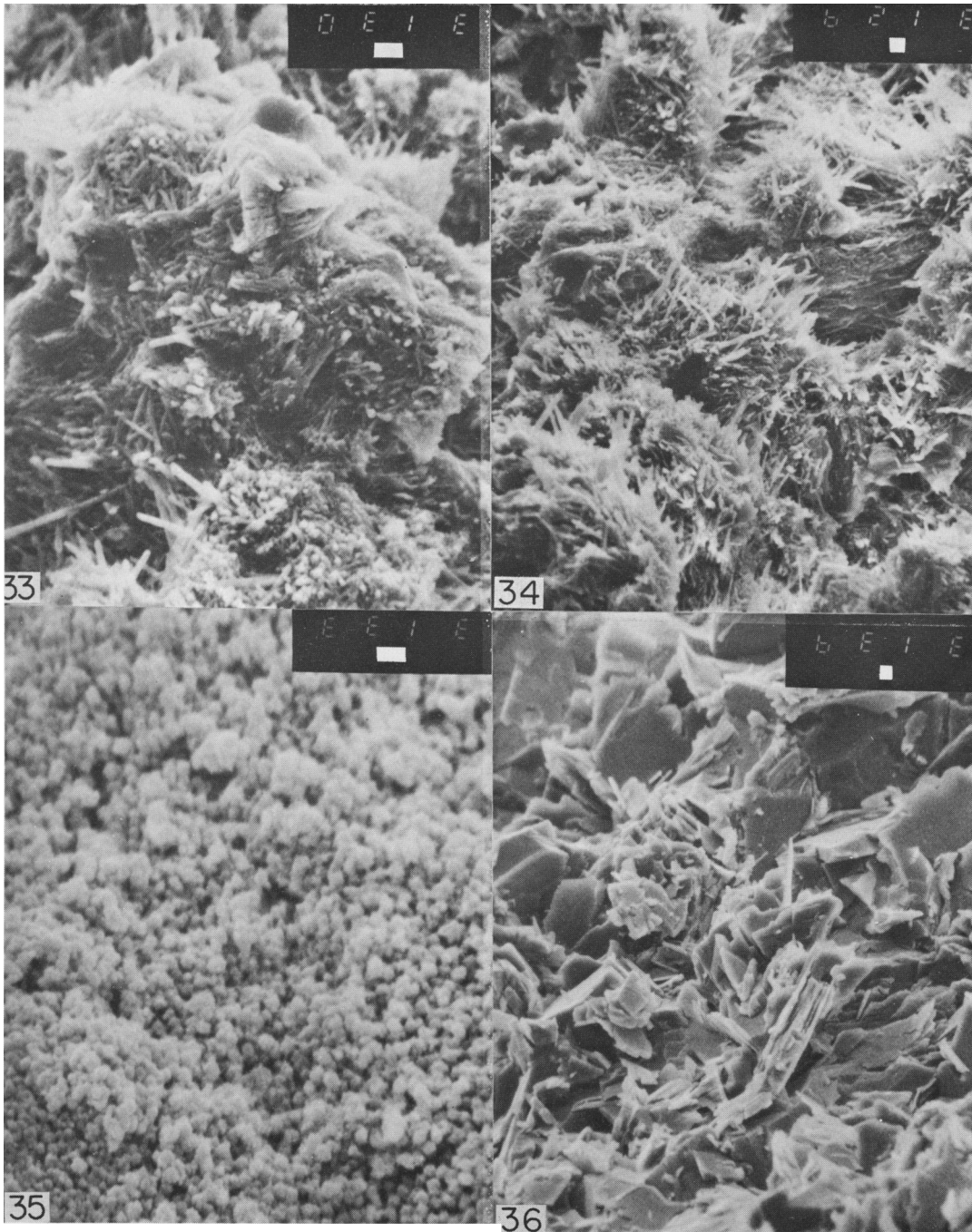


Figure 33. SEM of Gaerome kaolin clay, Yamaka pit, near Naegi, Japan. Note variety of kaolin morphology; subordinate cleaved plates and small kaolinite book, but predominant tubular halloysite, as reported by Nagasawa and Tuzuki, 3130, 5000 $\times$ .

Figure 34. SEM of Gaerome clay, Yamaka pit. Elongates are essentially filamentous, 3129, 3000 $\times$ .

Figure 35. SEM of 'spherical halloysite' (so described on the outcrop), weathered from a pumice bed, Yamaka pit, 3133, 5000 $\times$ .

Figure 36. SEM of first-quality Roseki stone, pyrophyllite, Shokozan, Japan, 3139, 2000 $\times$ .

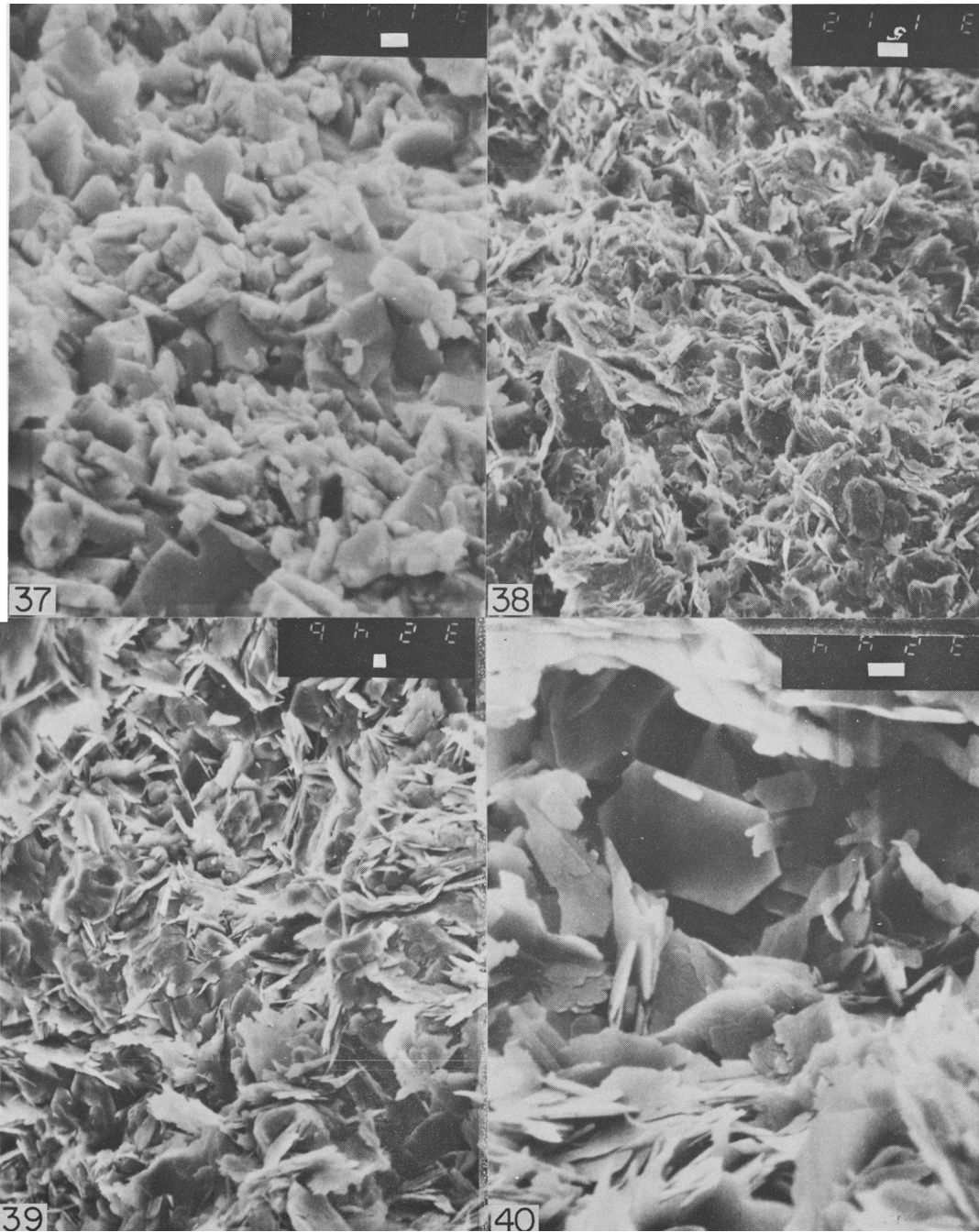


Figure 37. SEM of dickite in the Roseki, pyrophyllite, deposit, 3141, 5000 $\times$ .

Figure 38. SEM of ore-grade, hydrothermal kaolin from the Itaya open-pit mine, Yamagata Prefecture, Japan, 3112, 1000 $\times$ . Note pattern of random orientation of kaolin flakes, individual flakes or only a few in sheaves, but not books, hydrothermal quartz (?) crystal in lower left of micrograph.

Figure 39. SEM of Itaya clay at 2000 $\times$ . Again, note randomly oriented single or thin packet of flakes, 3246.

Figure 40. SEM of Itaya ore-grade kaolin showing angular crystal borders on flakes, and subhedral to euhedral crystals, 3244, 6000 $\times$ .

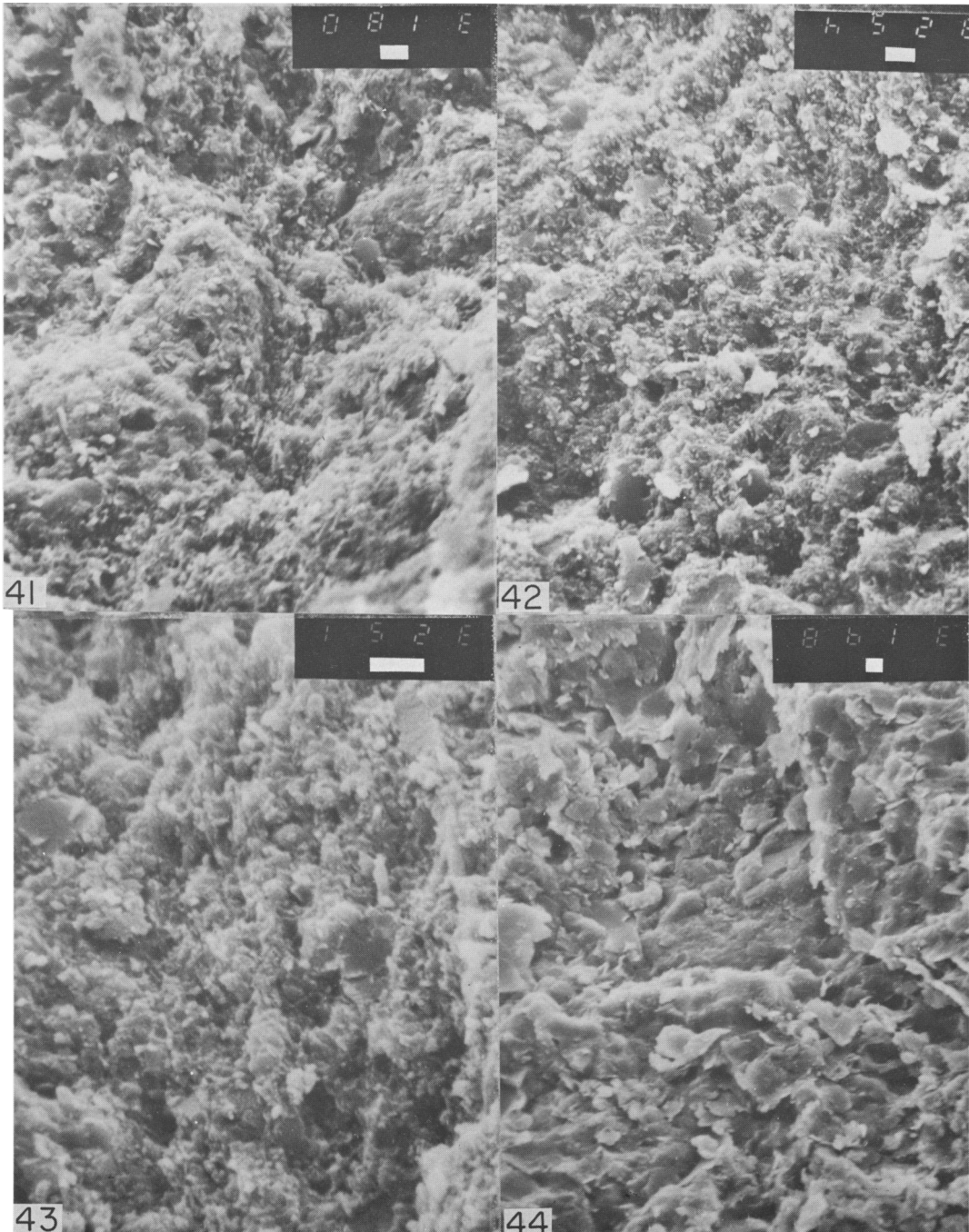


Figure 41. SEM of bentonitic, Cretaceous redbeds, Iwate mine, Japan, 3180, 5000 $\times$ .

Figure 42. SEM of first-quality flint clay (kaolin) stratigraphically above the bentonite beds, Iwate mine. Note similarity of inherited texture in this kaolin to that in the bentonite, 3254, 5000 $\times$ .

Figure 43. SEM of first-quality flint clay, as in Figure 42, at 10,000 $\times$ . At this magnification the kaolin flakes appear more conventional in morphology, but many crystal edges still retain a relic hint of smectite, 3251.

Figure 44. SEM of highest quality flint clay, underground mine at Iwate. Crystals are tightly packed and apparently recrystallized, 3198, 3000 $\times$ .



Figure 45. SEM, at 6000 $\times$ , of highest quality flint clay, underground mine at Iwate. Angularity of crystal edges and individual crystal plates attest to recrystallization of kaolin, presumably during structural tilting of the rocks, 3196.

50 km south of Sydney. Loughnan described it as having "a thickness of 0.3–2.0 m and varies in texture (hand specimen) from dense and ultra fine-grained to pelletal, brecciated and oolitic, with probably the most prevalent type being a clay arenite composed of angular to rounded clasts and scattered oolites" (Loughnan, 1976, p. 50). The oolitic variety is SE-micrographed in Figure 16 (3349, 3000×), and the dense, ultra fine-grained variety in Figure 17 (3347, 3000×). The micrographic textures of these two samples at 3000× are the same type, which also resembles that of the Permian clay.

Although heretofore the emphasis has been on kaolin derived from pyroclastics and its corresponding distinctive texture, where kaolinite of the Home Rule deposit was "apparently derived from granite of Late Carboniferous age" (Loughnan, 1976, p. 31), the texture is similar to that of 'Georgia' kaolin, Figure 18 (3272, 3000×). Loughnan further notes that, "The kaolin is similar to that from South Carolina . . . used as a semi-ball clay". Its texture in SEM likewise resembles that of the fine-grained kaolin in South Carolina, whose counterpart in the Kentucky–Tennessee deposits is a ball clay. For further variety in Australian kaolin minerals, dickite from the Triassic Hawkesbury Sandstone is shown in its typical euhedral morphology [Figure 19 (3377, 3000×)].

One of the most exciting assemblages of clay minerals and their products is located at 'The Pimple' and Burning Mountain on the Muswellbrook Anticline about 100 km northwest of Newcastle. Fully hydrated halloysite (endellite) is intimately associated with mullite, tridymite and cristobalite. The mullite was formed where claystone was heated by the burning of underlying coal beds, i.e. combustion metamorphism. Tridymite and cristobalite are easily accounted for by the high temperature of combustion but, as Loughnan (1976, p. 17) noted, their intimate association with "fully hydrated halloysite is somewhat more difficult to explain". Veins of endellite cutting mullite may have originated, in my opinion, as a deposit from hot fluids released during the dehydroxylation of underlying clay. Such fluids as could be generated as the kaolin was dehydroxylated, would probably be saturated with dissolved kaolin and, upon rising through rock that was cooler, would become over-saturated and deposit endellite. This is a parallel mechanism to that speculated for the deposition of endellite in veins in a hydrothermal kaolin deposit at Sombrerete, Mexico (Keller and Hanson, 1969). Likewise, the more massive deposits of Australian halloysite could be generated by hot fluids, either as heated meteoric water, or as dehydroxylation derivative, and preserve the textures observed by Loughnan as, "it has the same brecciated textures as the kaolinite clayrock and hence there can be little doubt it was derived from the latter presumably under the influence of heat from the fires" (p. 17).

Other flint clay, unchanged since its original lithification at this locality, is micrographed in Figure 20

(3328, 5000×). It has the same textural pattern observed in typical Australian flint clays. Halloysite, however, occurring below the burned-coal bed has an entirely different morphology, as in 2 orientations in Figures 21 and 22 (3333 and 3336, 3000×).

The repeated reference in this paper to volcanic ash parentage for the claystones, based primarily on texture by SEM, may arouse increasing skepticism of the reliability of SEM as a criterion of genesis—particularly among American readers whose geologic experience with mudstones in the U.S.A. less commonly involves volcanic detritus. This was also my first reaction, but two independent geological observations made in the field have dispelled any doubt in my mind about the voluminous volcanic sources for pelitic sediments in Australia.

Coal geologists from the U.S.A. who visited, on the 1976 Congress field trips, the Australian coal measures (with which the kaolins are commonly associated) were likewise impressed and convinced by a new and different geologic experience. At informal discussions and meal-time conversations, they repeatedly commented on the differences, with respect to the U.S.A., in coal geology, in the non-marine sedimentational environments, and the abundant volcanic ash in the Australian coal basins.

Secondly, at the Symposium on Correlation of Age and Genesis of Kaolin held jointly with the Sixth Conference of the Australian Clay Minerals Society, E. Slansky presented an illuminating paper that described the order of abundance of species of clay minerals in the Great Australian Basin of New South Wales which was atypical to most U.S.A. mudstones. To our amazement, Slansky documented in his talk that smectite (with some mixed layering) strongly dominates, and is followed by kaolin, whereas illite comes in a poor third in abundance in those mudstones. In his abstract he wrote, "In New South Wales, the massive occurrence of smectite is a characteristic feature of the Great Australian Basin . . . The occurrence of smectite is best explained in terms of alteration of volcanic and volcanoclastic material in or adjacent to the basin of deposition" (Slansky, in Patterson, *Proceedings 6th CCAGK*, 1976, p. 20).

#### SUMMARY AND INTERPRETATION

The interpretation made herein that the SEMs of flint clays and tonsteins of the Sydney Basin, Australia, indicate that the clays had a volcanic lineage, is supported by independent mineralogic data and geological field observations. Volcanic ash and other pyroclastic products contributed abundantly to the sedimentational process during Permian, Triassic, and also some other younger ages, in the Sydney Basin. Alteration of this volcanic material apparently first produced smectite, a very small part of it being pure enough to be commercial bentonite, but most of it being mixed with other rock-forming materials so as

to be only smectitic mudstone in general. The smectite clay minerals typically show a ragged- or scalloped-edge morphology in their plates.

Subsequently, the first-stage smectite underwent further leaching of silica and alkali and alkaline earth metals, with addition of hydrogen ions, and went to kaolin which retained part of the original smectite morphology. The alteration to kaolinite may have been a part of weathering by conventional leaching, or may have been driven by hydrogen ion-charged plant roots (coal) extracting nutrient ions, or by removal of metal ions and substitution with  $H^+$  from living and/or dead organic compounds.

#### KAOLINS COLLECTED IN JAPAN

Kaolins in Japan embrace a wide spectrum of genesis: saprolitic kaolinization of feldspathic phanerites, volcanic products weathered to flint clay, tonstein argillation, and diverse morphologic varieties of halloysite-endellite, hydrothermal argillation; high-alumina minerals formed by lateritization and hydrothermal action, and typical, transported sedimentary clay deposits.

Extensive deposits of weathered and transported kaolins occur in an important clay mining district east of the city of Nagoya. At the Maruhara mine, one of the largest in the district, the Seto Group of Pliocene Age produces 'Gaerome' and 'Kibushi' kaolin clays, along with quartz sand (*Guide to Field Investigations*, 1976).

Gaerome clay is a very poorly sorted, transported mixture of kaolin and quartz, resembling in composition a saprolite that had been moved without significant development of either sorting or bedding. Fujii stated that 'Gaerome' means 'frog eye' in Japanese, referring to the appearance of clays containing visible coarse quartz grains. 'Kibushi' means 'woody material'. . . "It is probable that Kibushi clay in Japan corresponds to 'Ball Clay' in other countries" (Fujii, p. 3, in Minato and Shimoda, 1976). The Kibushi clay, as observed at the Maruhara open pit, is a layered sedimentary clay that is darkened in color with lignitic and woody material, and has the typical lithology and bedding that characterize ball clay in the U.S.A.

The texture of the (kaolin) matrix between quartz grains in Gaerome clay from Maruhara is shown by SEM in Figure 23 (3220) at  $1000\times$ . The upper third of the field is enlarged to  $3000\times$  in Figure 24 (3221). In this sample, at least, the flakes of kaolin are small, and packed in face-to-face orientation, typical of ball clay. Moreover, the kaolin may be "associated with 'Kira', a silt-sized mineral aggregate largely composed of kaolinized mica" (Nagasawa and Tsuzuki, in *Geology of the Seto, Shokozan, and Itaya Kaolin Deposits*, 1976, p. 7). Possibly such aggregates are shown in Figure 25 (3216,  $3000\times$ ), but their identity as 'Kira' has not been confirmed. At higher magnifications, the orientation of clay flakes, both large and small, is

again clearly shown to be face-to-face [Figures 26 and 27 (3212,  $5000\times$ ; 3213,  $10,000\times$ )]. The Gaerome clay in this mine was derived from weathered hornblende granite present on the up-throw side of a nearby fault block.

The Kibushi clay from this mine is micrographed in Figures 28, 29, 30 (3231,  $1000\times$ ; 3230,  $2000\times$ ; 3228,  $3000\times$ ) to show by three magnifications the overall textural pattern of the clay, and details of packing and bending of individual clay flakes. The texture does, indeed, resemble that of typical ball clay in the U.S.A., although this particular occurrence of Kibushi clay is less tightly 'swirled' or contorted than are ball and plastic clays in Europe and the U.S.A. [Figures 1-5, Keller, Part II (1976)].

Whereas the Gaerome kaolin at the Maruhara pit was platy in morphology, at the Yamaka pit, Naegi, the easternmost producing area near Nagoya, the Gaerome kaolin is composed of elongates. Parent, basement granodiorite, where weathered to a crumbly, gruss-like state, yields tiny fragments to feldspar whose surfaces are covered with radiating, stubby 'whiskers' of elongates, identified by Nagasawa and Tuzuki as halloysite (*Geology of the Seto, Shokozan, and Itaya Kaolin Deposits*, 1976, p. 7); see Figures 31, 32 (3121,  $3000\times$ ; 3124,  $5000\times$ ). The clay of the superjacent Gaerome deposit likewise is composed of "long tubular halloysite and subordinate kaolinite" (Nagasawa and Tuzuki in the Guide); see Figure 33 (3130,  $5000\times$ ). In another specimen of the Gaerome clay, the elongates are so slender as to be essentially filamentous [Figure 34 (3129,  $3000\times$ )]. Although filamentous morphology of kaolin has not hitherto been commonly reported, another occurrence of even more slender, and relatively longer, hair-like filaments has been micrographed, also on weathering feldspar from both granite and gneiss, in the Georgia kaolin region (this SEM series, part IV, 1977).

In another unorthodox morphology, the lowermost bed 2 m thick, of five weathered pumice layers in the Yamaka deposit yields 'spherical halloysite' (as was orally described by the field-trip leader during the visit at the outcrop). Skeptics of a spherical variety of halloysite may be convinced by SEM Figure 35 (3133,  $5000\times$ ).

If I may be pardoned for asking a double rhetorical question, indeed more real than rhetorical, I would ask: "What are the definitive properties of halloysite, and how is halloysite differentiated from kaolinite?" At this one, relatively small deposit, kaolin clay occurs in plates, tubes, filaments and tiny spherules. "Is morphology a fundamental basis for differentiating halloysite from kaolinite? If so, what are the limits of halloysite morphology? On the other hand, if the distinguishing difference is ordering or quality of crystallization representable by X-ray powder diffraction, where is the line, arbitrary or natural, that separates kaolinite from halloysite? How is the quality of crystallization of kaolin minerals degraded—is it between layers (mixed layering), between sheets, by



anomalous articulation of tetrahedra and octahedra, rotations of tetrahedra, or other?" Endellite can, of course, be distinguished by  $4\text{H}_2\text{O}$  in its formula, a 10 Å spacing on 001, and distinctive DTA trace. Questioning need not stop on this end of the kaolin spectrum. If it is possible that kaolinite and dickite can or do crystallize as mixed-layer minerals [personal discussion with G. W. Brindley (1977)], the mineralogy and crystallography of the kaolin minerals would appear to now invite another fundamental and penetrating investigation to supplement the classic work by Ross and Kerr in 1930.

The famous Roseki (literal translation, 'wax stone') deposits of pyrophyllite occur at Shokozan and Shobara in the Hiroshima Prefecture (Takeshi, H., in *Geology of the Seto, Shokozan, and Itayo Kaolin Deposits*, pp. 10–15). In terms of the huge size and high quality of the deposits, it seems anti-climactic to show only 2 SEMs, but the first one is typical of the first-grade pyrophyllite [Figure 36 (3139, 2000×)], and the other one of the relatively massive dickite associated with the pyrophyllite [Figure 37 (3141, 5000×)]. The crystals are tightly and compactly interlocked, producing high-density, and strong, durable fragments in the freshly crushed ore. An adequate micrographic treatment of the Roseki deposit would include more extensive and detailed collecting to show the remarkable contact relationships between the pyrophyllite (major mineral) and associated dickite, nacrite, rarely kaolinite, boehmite, diasporite and, believe it or not, andalusite and corundum (blue crystals)—the mineral chemistry of which is exciting, and naively 'impossible' under simplistic geochemistry. It is hoped that our Japanese colleagues will soon prepare such a comprehensive and exhaustive micrographic report.

The Itaya kaolin open-pit mine in Yamagata Prefecture is one of the largest well-known hydrothermal kaolin deposits in Japan (Togashi, Y., in *Geology of the Seto, Shokozan, and Itaya Kaolin Deposits*, pp. 16–31). 'Acid sandy tuff and lapillic tuff' of late Miocene Age have been argillized into four zones by 'four episodes of hydrothermal alteration' (Togashi, p. 29). The first-quality kaolin ore is micrographed at three magnifications to show the over-all textural pattern and spatial relationships between plates [Figures 38–40 (3112, 1000×; 3246, 2000×; 3244, 6000×)]. Note the hexagonal, pyramidal crystal, presumably quartz, in Figure 38.

Compared to the texture of hydrothermal kaolins in Mexico, the Itaya plates are larger and less tightly packed than those in Mexico (Keller and Hanson, 1975). This textural difference reflects the difference in parent rock in the two occurrences. Whereas, in Mexico, the parent rock typically is compact lava, glassy rock, or hornblende schist (Keller and Hanson, 1969), at Itaya it is relatively porous tuff which provides a luxury of open space within which kaolin crystals can grow to larger size. Lump specific gravities for two typical kaolin ore clays from Itaya were found by Togashi to be 1.77 and 1.87, whereas in Mexico

bulk densities were found to be 1.83, 1.89, 2.20, 2.26, 2.47 and 2.50 (Baumann and Keller, 1975).

In northeast Japan, reminiscent of flint clays of volcanic lineage in Australia, is the large Iwate mine. Fujii wrote, "Particularly the Iwate mine is one of the most productive refractory clay mines and is noted for the only flint clay producing in Japan" (Fujii, pp. 1–10, in Minato and Shimoda, 1976). At Iwate, 85 m of Upper Cretaceous red-bed argillite (weathered welded tuff), is unconformably overlain by 95 m of Oligocene sediment. A bed of occasionally gibbsitic flint clay, 1–2 m thick, occurs at the base of the Oligocene Komatsu Formation (Nagasawa, pp. 32–45, in Minato and Shimoda, 1976). Fujii interpreted the flint clay as altered pyroclastic material, whereas "Iijima considered the red-bed as a weathering product from welded tuff, and the flint clay as sediment derived from the redbed" (Nagasawa, p. 35, in Minato and Shimoda, 1976; Iijima, 1976).

In Figure 41 (3180, 5000×) is shown the texture of the bentonitic Cretaceous red beds. Ragged- to scalloped-edged flakes, typical of the texture of smectite developed from volcanic ash, characterize the SEM. Texture of this clay accords completely with the interpretations by both Fujii and Iijima that volcanic material was its progenitor.

Flint clay of first quality, from a bed so designated on the field excursion, is shown in Figures 42 (3254) at 5000×, and 43 (3251) at 10,000×. The texture is of typical, fine-grained flint clay whose kaolin flakes 'remember' a smectitic-volcanic heritage. Cleavage faces of occasional larger crystals of kaolinite suggest they have matured and recrystallized diagenetically from prior, presumably smaller ragged-edged, crystals.

The bed of exposed first-quality flint clay increases in quality for refractory use down dip, where it is mined underground. Samples of this highest-quality flint clay are micrographed in Figures 44 (3198, 3000×) and 45 (3196, 6000×). The kaolin appears from texture to have so thoroughly recrystallized, presumably during structural tilting of the rocks, that it lost all smectite-relic texture. The new crystals were tightly compressed, compacted, and tightly interlocked. In hand specimen, the material is 'hard' (not on the Moh's scale, but non-friable), and 'heavy' (high bulk density). Lithologically it resembles the so-called 'hard, heavy, super-duty (refractory) flint clays' in North America.

Although texture by SEM indicates volcanic parentage for Iwate clay, not enough samples were collected near the critical contacts of the distinguishing formations to resolve the details of the alternative interpretations of Fujii and Iijima on its genesis.

From the same perspective, all of the other deposits in both Australia and Japan, should be studied in greater detail by SEM, and more comprehensive interpretations and conclusions be drawn. A pioneer report, such as this one, can only hope that its major contribution will be to stimulate and invite more

thorough investigations than were possible at this preliminary glimpse.

*Acknowledgements*—This research was supported by the National Science Foundation, NSF EAR 73-06648 A02 and DES 73-06648, for which I am grateful. Collection of specimens was facilitated by competent field guidance by Professor Loughnan in Australia, and Professors Minato, Fujii, Nagasawa, Iijima, Imai, and Staff Geologists at the Iwate mine, in Japan. Assistance from members of the SEM laboratory UMC, and secretaries in the Department of Geology, UMC, helped make this report possible.

#### REFERENCES

- Baumann, D. and Keller, W. D. (1975) Bulk densities of selected dried natural and fired kaolin clays: *Clays and Clay Minerals* **23**, 424–427.
- Bohor, B. F. and Pillmore, C. L. (1976) Tonstein occurrences in the Raton coal field, Colfax County, New Mexico: *New Mexico Geological Society Guidebook, 27th Field Conf., Vermejo Park*, 1976, pp. 177–183.
- Borst, R. L. and Keller, W. D. (1969) Scanning electron micrographs of API Reference Clay Minerals and other selected samples: *Proc. Int. Clay Conf., Tokyo*, Vol. 1, pp. 871–901.
- Geology of the Seto, Shokozan, and Itaya Kaolin Deposits, A Guide to the Field Investigations*, papers by Nagasawa, K., and Tsuzuki, Y.; Takeshi, H.; and Togashi, Y. (1976) at the 7th Symp. on Genesis of Kaolin: Mimeographed Field Guide distributed by H. Minato, University of Tokyo, 31 pp.
- Guide to Field Investigations, Geology of the Seto, Shokozan, and Itaya Kaolin Deposits* (1976) Mimeographed Field Guide, distributed by H. Minato, University of Tokyo, 16 pp.
- Iijima, A. (1976) *Upper Cretaceous and Oligocene Clay Ore Deposits in Iwate Clay Mine, Iwaizumi, Iwate Prefecture, Japan, 7th Kaolin Symposium, Japan*: Mimeographed Field Guide, A. Iijima, University of Tokyo, 16 pp.
- Keller, W. D. (1968) Flint clay and a flint-clay facies: *Clays and Clay Minerals* **16**, 113–128.
- Keller, W. D. (1976) Scan electron micrographs of kaolins collected from diverse origins—III. Influence of parent material on flint clays and flint-like clays: *Clays and Clay Minerals* **24**, 262–264.
- Keller, W. D. and Hanson, R. D. (1969) Hydrothermal argillification of volcanic pipes in limestone in Mexico: *Clays and Clay Minerals* **17**, 9–12.
- Keller, W. D. and Hanson, R. D. (1975) Dissimilar fabrics by scan electron microscopy of sedimentary versus hydrothermal kaolins in Mexico: *Clays and Clay Minerals* **23**, 201–204.
- Loughnan, F. C. (1976) *Kaolinite Clay Rocks of the Sydney Basin, Sydney, Australia: 6th Symp. Correlations Age Genesis of Kaolin, 25th Int. Geol. Cong.*, Mimeographed Field Trip Guide, 53 pp.
- Minato, H. and Shimoda, S., editors (1976) *7th Symp. on Genesis of Kaolin: Correlation of Age and Genesis of Kaolin*, H. Minato, Chairman, University of Tokyo, 150 pp.
- Patterson, J. H. (1976) *Summary of Proceedings, Symp. 6th Conf. CCAGK, and 6th Conf. Australian Clay Minerals Society*: Mimeographed copy distributed by J. H. Patterson, CSR Laboratories, Roseville, N.S.W., Aust., 20 pp.
- Ross, C. S. and Kerr, P. F. (1930) The kaolin minerals: *U.S. Geol. Survey Prof. Paper* **165E**, 151–176.
- Williamson, I. A. (1970) Tonsteins—their nature, origins, and uses: *Mining Mag. (London)* **122**, 119–125; **122**, 203–210.