

## DISSIMILAR FABRICS BY SCAN ELECTRON MICROSCOPY OF SEDIMENTARY VERSUS HYDROTHERMAL KAOLINS IN MEXICO

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(Received 20 August 1974)

**Abstract**—Dissimilar fabrics, observed by scan electron micrography (SEM) on freshly broken surfaces of kaolins from weathered, water-laid material vs hydrothermally altered material in Mexico are illustrated, contrasted, and tentatively related to their respective environments of genesis. Coarse, open-textured, well-preserved books and vermiforms of kaolinite characterize the kaolin from water-laid material at Jacal, Mexico. Hydrothermal kaolins tend to have more tightly compacted crystals of either kaolinite flakes (individuals or in packets) or elongates (halloysite morphology) or mixtures of both in the same deposit. Variations in porosity of parent rock whether sedimentary or igneous, and dissimilar permeation by meteoric water and by vapor and liquids of hydrothermal fluids are deemed to be significant in the processes producing distinctive fabrics. The question is raised as to the fundamental distinction between kaolinite and halloysite and their nomenclature.

### INTRODUCTION

The texture or in a broader sense, fabric, which may include some elements of structure observed in freshly broken surfaces of coarse-grained rocks, is an essential feature in classification and genetic interpretation of the rock types. For example the fabrics of granite, diabase, augen-gneiss, dunite, crinoidal limestone and others are distinctive and diagnostic of those rocks and their geneses.

Fine-grained rocks, such as clay mudstones, cherts, etc., have not been amenable to similar three-dimensional study of their fabric or texture until the advent of the scan electron microscope (SEM). Now, however, pictures of broken rock surfaces magnified, 1000–10,000 $\times$  (a useful range) by SEM, reveal fabrics of clays and cherts as discernibly as does a 10 $\times$  hand-lens for phanerites. Different families of clay minerals exhibit different and usually distinctive fabrics (Borst and Keller, 1969). Furthermore, within even a single clay-mineral family, e.g. the kaolin group, clays of diverse origins exhibit fabrics that are distinguishably different. Diversity in origin may arise at least from differences in parent material, genetic processes and/or physical and chemical environment during formation. For example, parent materials may be feldspar, mica, glass or aphanitic rocks. The process of formation may be weathering, hydrothermal alteration, direct precipitation or crystallization from solution, growth from a colloidal gel, resilication of an alumina mineral and others. The physical environment during crystallization may provide open space for free growth of euhedral crystals or it may

constrain space so that mutually competitive crystal boundaries grow in contact.

Will these several different genetic factors generate fabrics distinctive or diagnostic of them? That is a problem being investigated and on which this paper reports progress. Attention will be directed in this paper to fabrics of two dissimilar genetic types of kaolin in Mexico: One from a deposit of water-laid material and a group of others hydrothermally altered (literally 'thermal water', without specifying the sources of the water or heat).

Specimens were prepared for micrography by breaking them in hand, yielding a tensile fracture, which is therefore similar to breaking a hand specimen of rock with a hammer. The fresh surface was coated with a thin film of evaporated gold to conduct away excess charge when exposed to the electron beam. The topography of the fracture surface therefore is generated by the textural and cleavage-fracture properties of the minerals in the specimen. No additional processing, other than the very thin coating, minimizes (or eliminates) any possible modification or alteration of the natural surface or its micrograph by manipulative preparation of the samples.

Magnifications of 1000–5000 $\times$ , rarely to 20,000 $\times$  were most commonly used. Inasmuch as direct photomicrographs are 9  $\times$  11.5 cm, the dimensions of the field of view on the specimen observed in the photographs at 1000 $\times$  are 90  $\times$  115  $\mu$ m, and at 10,000 $\times$  are 9  $\times$  11.5  $\mu$ m. It may at first appear presumptuous to interpret the genetic environment of a thousand-ton kaolin deposit from the fabric

of a few areas of the order of  $0.01 \text{ mm}^2$  on a half-dozen small hand specimens but this approach is permissible on several grounds. Small areas of observation are proportional to the small crystal size, as are larger areas viewed on rocks of coarser grain. Consistent replication of fabric-type from randomly selected specimens (the unresolvable grain size by unaided eye precludes biased textural selection of specimens) assures confidence in the validity of the results. Extrapolation of small samples to larger populations has been valid in numerous other applications.

Photomicrography of otherwise unresolvable features has a certain subjective element to its execution. The eye may tend to be attracted unduly to the dramatic, the unusual or the more photogenic features, but be inclined to gloss over monotonously similar areas. A special effort has been made to document in published or exhibited micrographs the typical fabric or the variety of fabric in each specimen as it ranges from surfaces of relatively uniform texture to occasional 'vugs' in which individualistic patterns are developed—if such a range was observed. Fissility or layer-oriented fabrics or structures, although searched for in sedimented kaolins and lava flows, have not been significantly developed observable by SEM.

#### JACAL SEDIMENTARY KAOLIN

The kaolin derived from water-laid material is a bedded deposit, presumably in an interior lake, near Jacal, a small settlement in the State of Mexico, about 125 km west and north of Mexico City on National Highway 15. Scattered outcrops and prospect-drilled remnants from erosion outline a roughly elliptical deposit originally of the order of 3 km in length and 1–1.5 km in width. A thickness of about 4 m of commercially useable kaolin is underlain by gravel several meters thick. Horizontal bedding in the kaolin is exhibited by slight changes in color, oxidation bands, streaks of organic matter, and particle size. Beds range in thickness from 10 cm down to laminae a few millimeters thick. The parent material of this kaolin, as judged from partially altered sand and gravel margins of the deposit, was volcanic detritus eroded from surrounding heights, possibly augmented by some wind-carried original pyroclasts.

Typical SE Micrographs of Jacal kaolin are shown in Figs. 1–6. Figure 1 (389) is a view looking down onto the bedding-plane surface of one of the upper layers in the pit, and Fig. 2 (387) is at right angles to the same bed (facing the edge of the bed). Figure 3 (391) and 4 (394) are similarly oriented views of a bed about 1 m below 1 and 2. Figure 5 (194) and 6 (197) are of samples collected at random from a large stock-pile of reserve clay. Figure 5 resembles No. 1, whereas books in No. 6, at  $5000\times$ , are almost obscured by a heavy cover of tiny flakes and aggregates. Possibly the small particles represent a later (?) generation of kaolinite. A similar occurrence of

tiny particles on coarser kaolin has been observed in a kaolin (not reproduced here) from Cornwall, England.

Distinctive properties of the Jacal kaolin are well-preserved, expanded books, accordions, and vermiforms of relatively large-size kaolin flakes. The flakes typically are 5 to  $10 \mu\text{m}$  dia.; books and vermiforms may be up to at least  $70 \mu\text{m}$  (Fig. 1,  $1000\times$ ) in length. In Fig. 6 at  $5000\times$  the tinier flakes coating the large books range from about 0.2 to  $2 \mu\text{m}$  dia.; one book is  $10 \mu\text{m}$  long. In all specimens from Jacal the crystals are euhedral to subhedral since edges of hexagons may be ragged to ill-formed. Very high porosity in the clay is outstandingly developed.

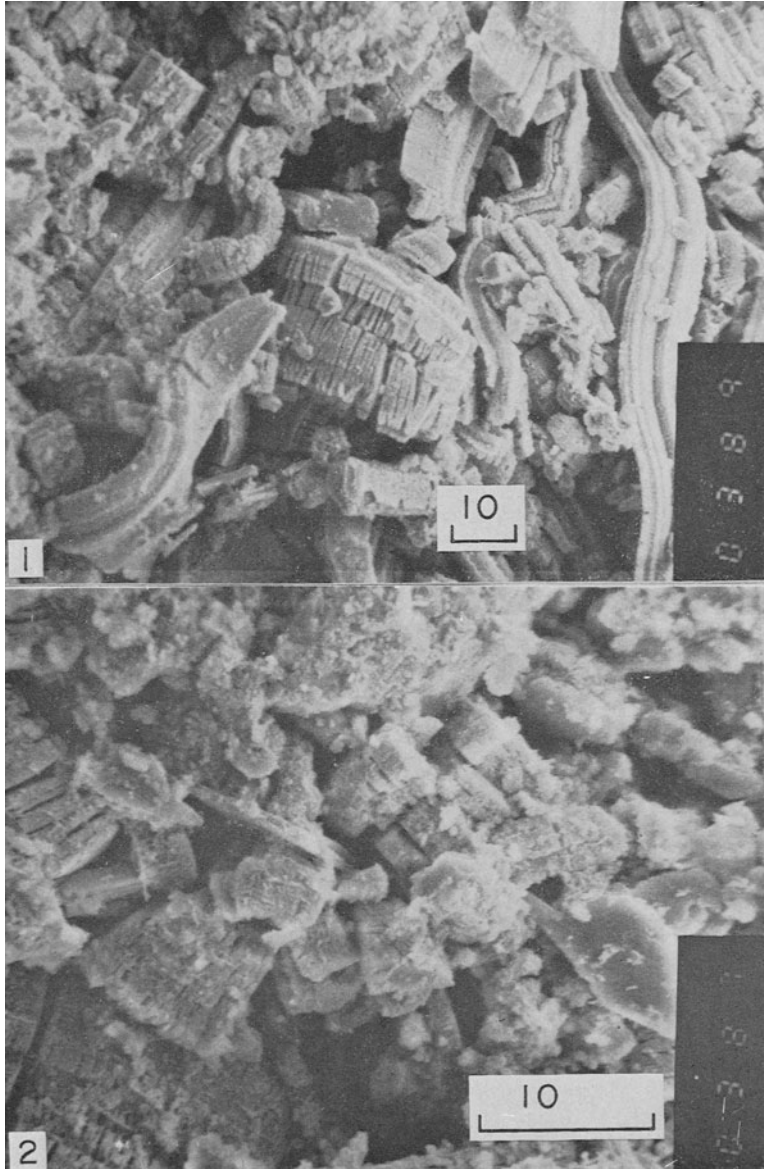
#### HYDROTHERMAL KAOLINS

Dissimilar fabrics from typical hydrothermal kaolins in Mexico are shown in Figs. 7–13. Figures 7 (212) and 8 (206) are from a mine at Sombrerete, Zacatecas, where the parent rock included volcanic ash and/or acidic (silicic) lava (Keller and Hanson, 1969). Note in Fig. 7,  $15,000\times$ , that the kaolin occurs in small ( $0.07$ – $0.5 \mu\text{m}$ ) well-formed hexagonal plates. They occur singly or in thin packets, closely and tightly consolidated. Inter-grain porosity is very low. In Fig. 8, at  $7,500\times$  the kaolin morphology is dominantly "elongates" (permissibly includes tubes, scrolls, fish-tails, or fiber-columns). They are up to  $3.5 \mu\text{m}$  in length and up to  $0.4 \mu\text{m}$  dia. and appear to be tightly packed, except for the few that are fluffed up at the broken top surface. Poorly resolved hexagonal plates (?)  $0.4$ – $0.5 \mu\text{m}$  dia. are visible in one corner of the micrograph. Porosity is low this clay.

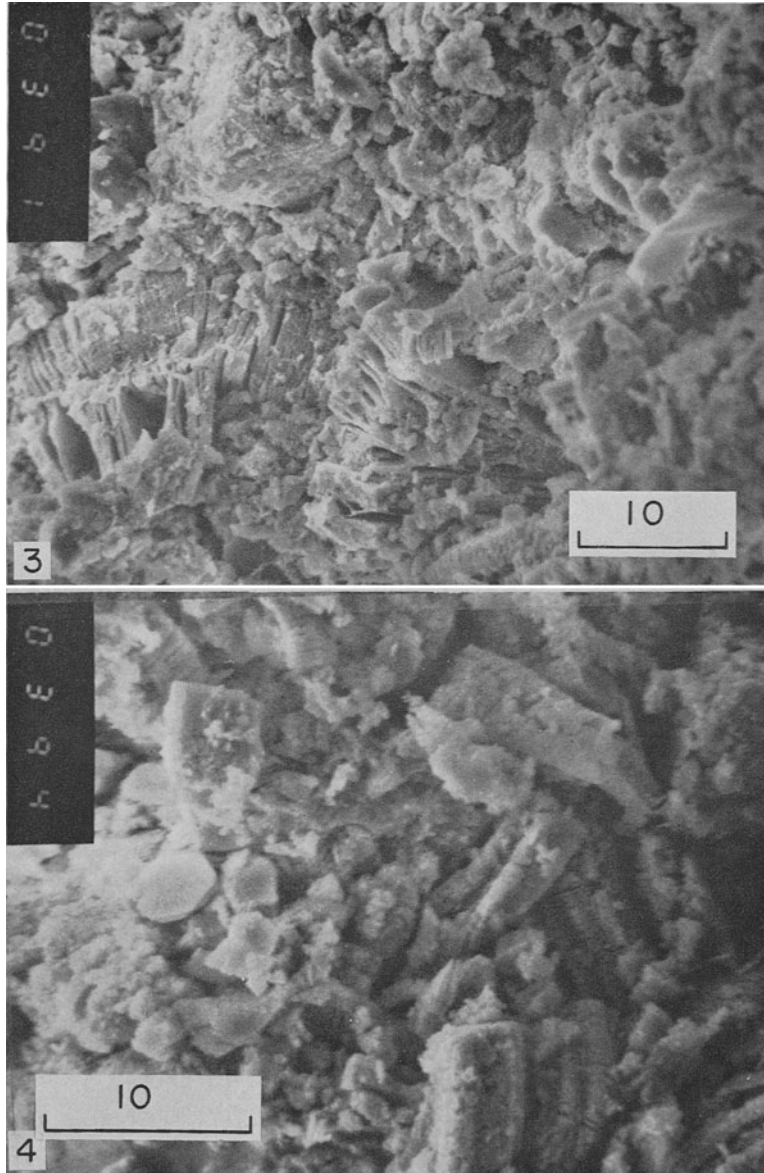
In Fig. 9 (218) the kaolin was developed from an amphibole chlorite schist near Guanajuato (Hanson and Keller, 1966). One kaolin packet,  $4 \mu\text{m}$  dia. is tightly embedded within a solid matrix of interlocking flakes from  $<0.1 \mu\text{m}$  up to 3 or  $4 \mu\text{m}$  dia. Porosity is low.

Kaolin from the large, General Zaragosa mine in San Luis Potosi, where the parent rock was a rhyolite flow and breccia (Keller and Hanson, 1968), is shown in Fig. 10 (198). The clay is a tightly packed, low-porosity mass of elongates, with possibly some plates. At  $7500\times$ , Fig. 11 (200) of the same specimen, both elongates ( $2 \mu\text{m}$  long and  $0.3 \mu\text{m}$  dia.) and plates ( $0.3$ – $0.5 \mu\text{m}$ ) are discernible. In the preceding 1968 publication cited, a transmission electron micrograph showed both hexagons and elongates of the same general dimensions as now micrographed by SEM.

In Fig. 12, at  $10,000\times$  (659) is a SE Micrograph of halloysite, known to be strongly disordered along both *a*- and *b*-axes from an X-ray diffractogram; derived from a perlite near Etzatlan, west of Guadalajara, in the State of Jalisco (Keller, 1963). Most of the clay in this micrograph, at about the limit of resolution of the clay-microscope pair, appears to be rudely hexagonal in outline. One might be tempted to call them kaolinite plates from their morphology but, as noted above, X-ray diffraction shows them

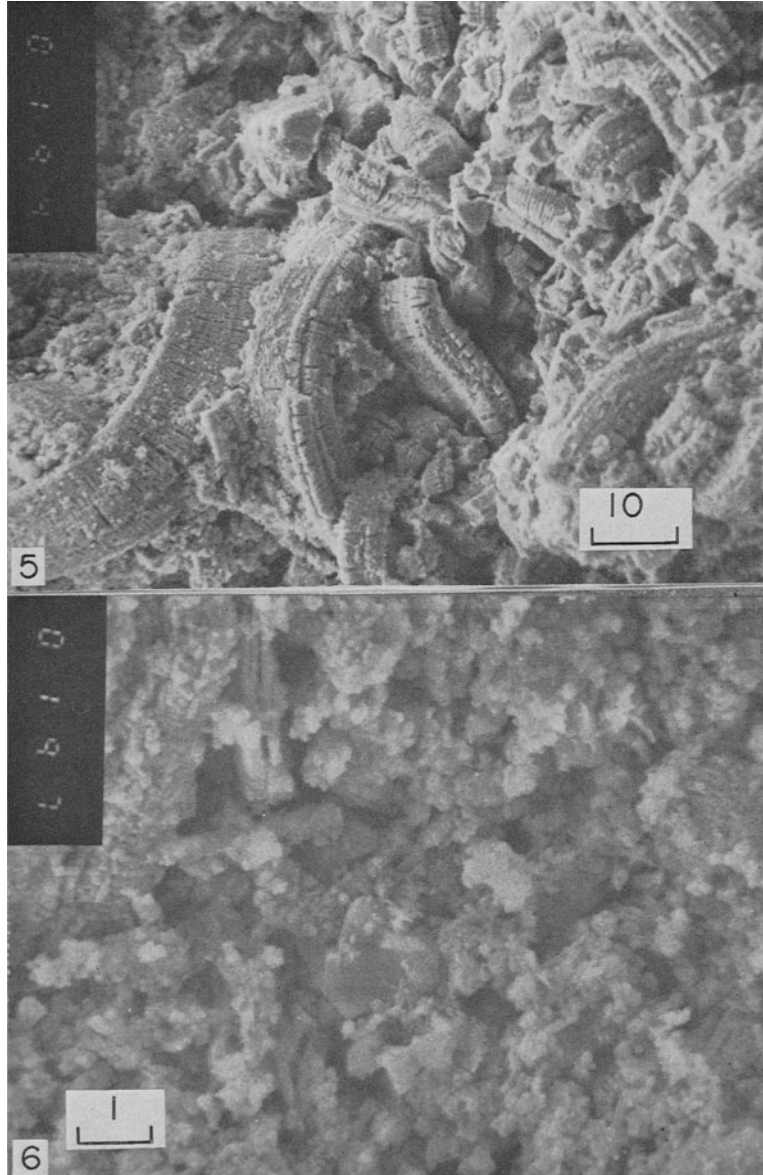


Figs. 1 and 2.

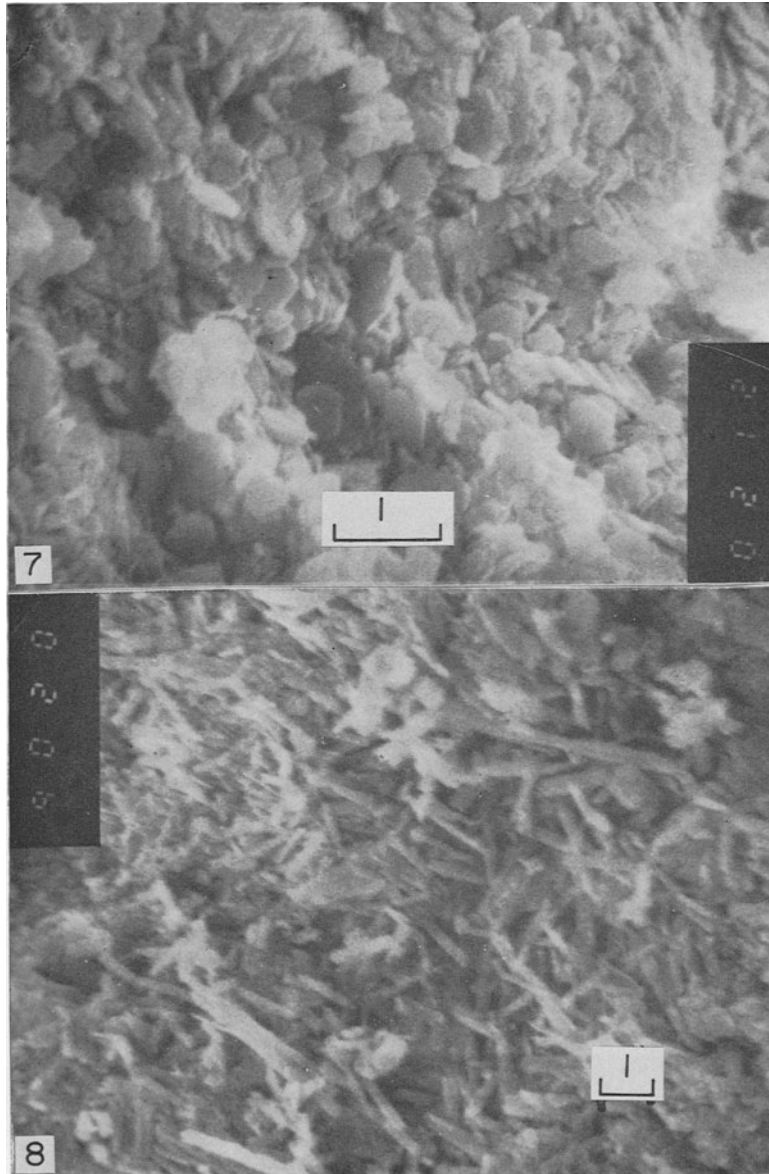


Figs. 3 and 4.

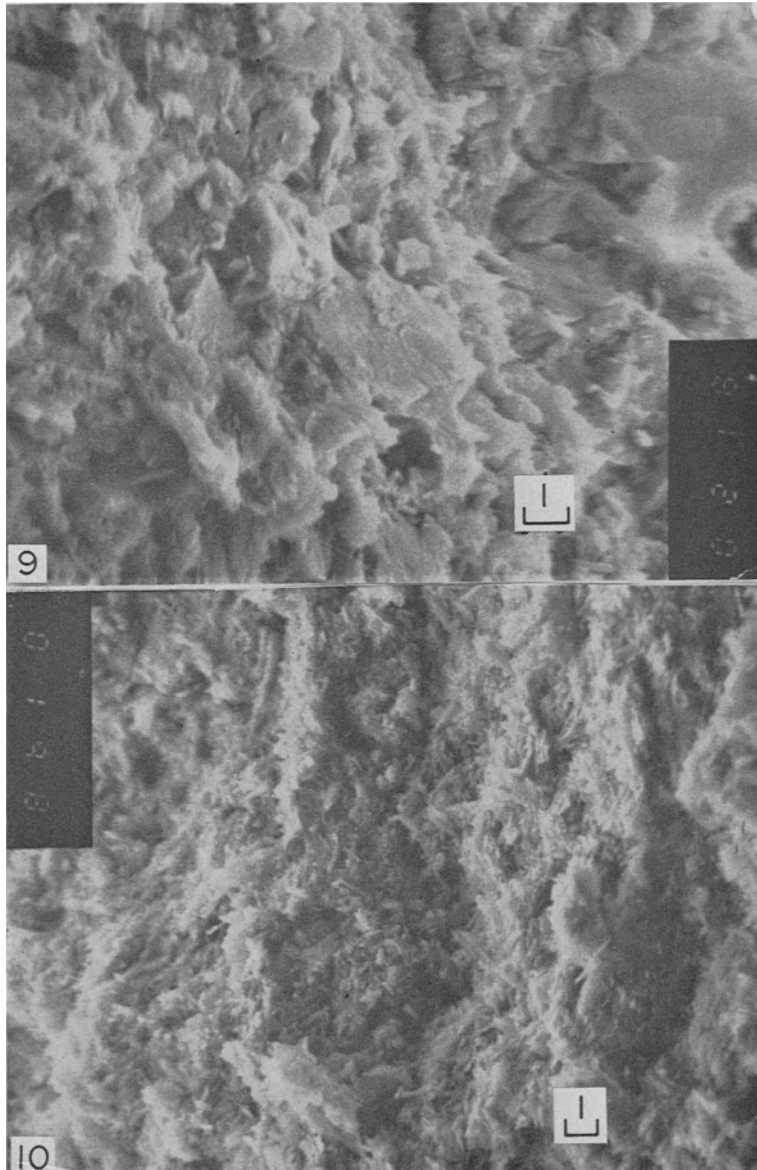




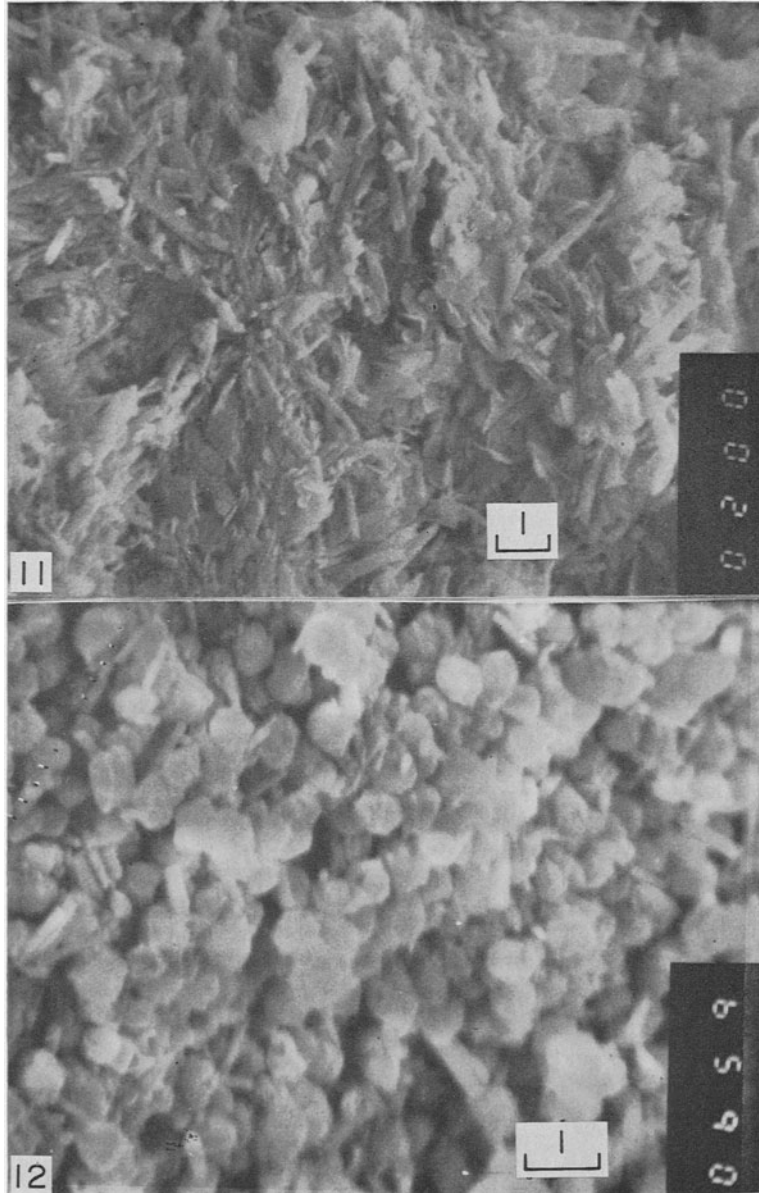
**Figs. 5 and 6.**



**Figs. 7 and 8.**

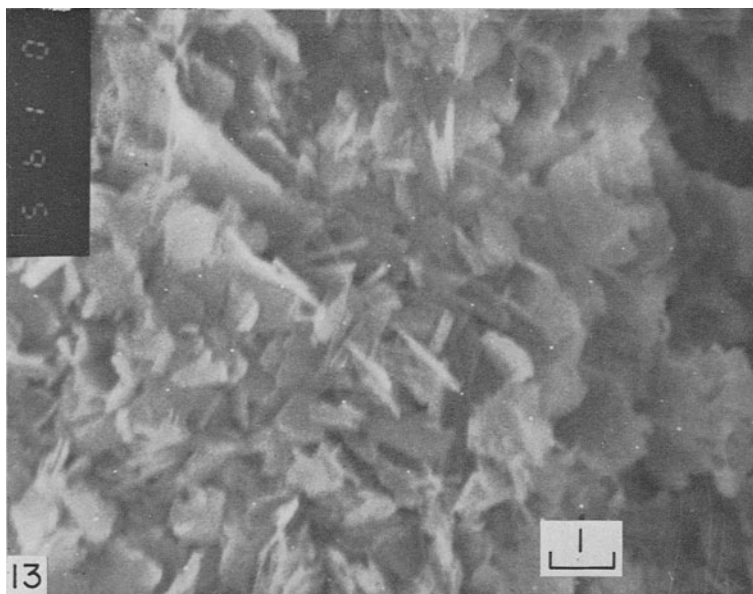


**Figs. 9 and 10.**



Figs. 11 and 12.





**Fig. 13.**

to be very poorly ordered. One wonders if this clay might be similar to the halloysite described by Chukhrov and Zvyagin (1966). At any rate the fabric of this clay is dissimilar to that of Jacal.

A platy kaolin, derived from silicic lava at Comonfort, State of Mexico, composed of essentially single, but sometimes curved, larger flakes of kaolin is shown in Fig. 13 (195). Most, if not all, of the elongate stripes in the micrograph are slightly curled edges of kaolin flakes that brighten, due apparently to "charging up" in the electron beam since the amount of conducting gold coating on the edges is very slight.

Generalizing for the fabric of hydrothermal kaolin in Mexico, it is characterized by relatively small size crystals, tight packing, and interlocking of flakes and packets, and relatively low porosity. Crystal morphology may be either plates or elongates. Obviously this is clearly dissimilar to the fabric of the material having a water-laid history.

#### FACTORS INFLUENCING THE FABRIC OF KAOLIN

Why are the fabrics of these two genetically different kaolins so dissimilar? To dismiss the question with an answer restricted solely to the process of alteration, i.e. weathering vs hydrothermal, would overlook at least seven other possible, important factors and a score of subordinate dependent variables. These will be formally tabulated and discussed in a forthcoming paper that more comprehensively considers world-wide occurrences of kaolins including all types known to the writer, such as sedimentary, primary, solution-deposited, flint-clay, resilitated and others. The factors that appear to apply positively to the two types from Mexico include the following. The weathering environment in which the Jacal kaolin (fabric) originated includes and implies certain characteristics of that environment (weathering), as follows:

(1) Porous and permeable parent rock providing open space (filled with gas or liquid) which permitted growth of euhedral-crystal morphology and expanded aggregates. This applies equally well whether the kaolin crystals formed *in situ* or if they formed on another weathering surface and were later washed in. It implies low pressure-weight of overburden.

(2) Alteration proceeded downward from open-air to solid interface, and the fragments altered from outside surfaces inward, at low pressure. Space for kaolin crystal growth was ample to copious.

(3) An available excess of relatively fresh rain water which potentially includes bacteria and other organic compounds which may 'catalyze' kaolinization (Hem and Lind, 1974; Linares and Huertas, 1971). Such a chemical environment may promote growth of coarse crystals of kaolinite.

(4) Parent material was feldspathic, although it is irrelevant whether in coarse crystals or an aphanitic

matrix. Feldspar may alter either to plates or elongates, by either weathering or hydrothermal action.

The environment of hydrothermal kaolinization in which that fabric type originated includes the following factors.

(1) (a) Low-porosity, low internal-permeability rock (except through structural channels) of lava flows; (b) intermediate to higher porosity-permeability of rocks such as volcanic ash and flow breccia.

(2) Thermal solutions rising through the body, or inundating it within a thermal water-table zone, would soak the rock body continuously and alter it, including its fragments, from inside out as well as outside in. One may visualize alteration in the interior of a rock or mineral body as being constrained by the volume, or space occupied, of the body undergoing alteration. Space for kaolin crystals to form must be generated by solution removal of parent material. Since kaolinization represents addition of water and formation of a lower bulk density material (clay) than the parent material, both of the two actions will tend to increase the volume of the clay over that of the parent. Simultaneously the removal of silica, alkali and alkaline earths will decrease the volume. Pseudomorphs of clay after feldspar, however, never exhibit a decrease in volume so the net volume change of feldspar to kaolin appears to be constant or an increase. Such an alteration change would produce a tightly packed mass of either plates or elongates, accompanied by very little intergranular porosity. Nuclei of kaolin crystals would be numerous because of numerous grains in the rock, i.e. in an igneous aphanite or ash, resulting in small crystals. Orientation of the newly-formed kaolin crystals would be random in a randomly crystallized aphanite whether or not influenced by the structural orientation of feldspar lattices. Packing of the small kaolin crystals, intergrown one with another, would be tight. Hence, if the process of hydrothermal kaolinization is correctly visualized (from what we have seen around hot springs in Mexico) the fabric of the kaolin so produced would logically be dissimilar to that formed by weathering during which a descending zone of alteration progressively converts a strong, tight, subjacent rock into a friable, loose regolith above (where relatively weak clay minerals can expand into space).

(3) Thermal water will possess higher vapor pressure, lower viscosity and additional hydro-pressure as it rises with vigor and will enhance chemical activity due to the higher temperature, than will descending rain water. Thermal water should be more capable of entering and penetrating, either as water or steam, the solid phases undergoing change. It will ordinarily lack an organic catalyst, but this is not needed in synthesis of kaolin at high temperature.

(4) As previously stated, either plates or elongates of kaolin minerals can be produced from a variety of parent material.

(5) What influences or determines whether plates or elongates, kaolinite or 'halloysite', will be produced

within and from the same parent material, as at Sombrerete?

Keller and Hanson speculated from the incomplete geologic evidence observed that in this deposit kaolinite formed when the crystallizing clay was nourished with Al and Si ions in solution derived from solid parent material located almost in immediate contact with the clay. Endellite, on the other hand, was deposited from solutions that had moved out too far to be continuously nourished in Al and Si by the parent material, but became oversaturated because of the changing physical environment.

In light of thermodynamic calculations by Huang (1974), the preceding interpretation based on geologic occurrence may be partly correct. Huang reported the  $\Delta G_r^\circ$  for halloysite and kaolinite to be  $-898.4$  and  $-902.9$  kcal/mole respectively. He further stated, "Halloysite may crystallize from a solution supersaturated with respect to halloysite, but kaolinite may crystallize under these same conditions or also from a solution saturated with respect to kaolinite. Moreover, any halloysite formed will tend to be spontaneously transformed to kaolinite ( $\Delta G_r^\circ$  being negative). Rate of transformation, however, is governed by the activation energy (not Gibbs free energy), the temperature, and the rate of kaolinite precipitation from such nutrient solutions as may be supplied from dissolution of halloysite through diagenetic processes".

#### WHAT IS THE FUNDAMENTAL BASIS DIFFERENTIATING KAOLINITE FROM HALLOYSITE?

By tradition, kaolinite has platy morphology whereas halloysite (which may be confused with meta-halloysite and endellite) has an elongate morphology (variously observed as tubes, scrolls, fish-tail ends and fiber-columns). By tradition, halloysite is more strongly disordered than is kaolinite—but how much more (?) disordered. The  $4\text{H}_2\text{O}$  variety is distinctive by its basal spacing of about  $10\text{Å}$ . Is morphology of crystal sufficiently fundamental to differentiate mineral species? If so, into how many mineral species should  $\text{CaCO}_3$  be sub-divided, although all give a diffractogram of calcite? A SEM of a clay from Idaho which shows in poor resolution what appears to be a mixture of plates, elongates, and—either curling plates or uncurling scrolls—which are they, and which way are they going? A SEM of elongate kaolin flakes whose ratio of length:width is up to 5:1 from a bore-hole in the Cornwall kaolin district (Penhale-Lower Ninestones Pit). If these flakes were to curl,

or fold even once, they would be classified as "elongates" crystals. Observations such as these give pause to otherwise hasty distinction on a basis of morphology between members of the kaolin family.

Should halloysite and kaolinite be delimited and defined, not on a basis of morphology, but by an empirically designated position in the spectrum of order-disorder of the kaolin minerals observed in X-ray diffractograms (Brindley and Robinson, 1946; Murray and Lyons, 1956)? Or, should halloysite be determined, or designated, only by electron diffraction as used by Chukhrov and Zvyagin (1966)?

The problem of nomenclature is larger than these two mineral names. Should the spectrum of kaolin minerals be defined using one of the three schemes of nomenclature proposed by Brindley and de Souza Santos in 1966? Indeed, until a fundamental basis of classification for kaolin minerals is accepted it seems preferable to use simple, descriptive 'plates and elongates'.

*Acknowledgement*—It is a pleasure to acknowledge support for research reported herein from the National Science Foundation Grant No. GA-38898.

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