

TUBULAR MORPHOLOGY OF SOME BRAZILIAN KAOLINS*

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DE ANDRADE. ELECTRON MICROGRAPHS BY A. C. VILLANOVA

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In the course of some studies on kaolins derived from pegmatites, from Minas Gerais, Brazil, striking differences were observed between these kaolins and those commonly described in the foreign literature, as regards the morphology of the particles. A planar and more or less hexagonal form has generally been ascribed to kaolin particles as revealed by the electron microscope. The Brazilian material, however, shows a completely different morphology, the particles often being of tubular form and having varying opacity and length (Fig. 1).

Initially these tubes were regarded as halloysite, but the frequency of their occurrence in the Brazilian material, compared with the general rarity of halloysite induced the authors to examine the material by *x*-rays. Diagrams made with a North American Philips Geiger diffractometer revealed kaolinite and not halloysite (see diagrams and electron micrographs, Figs. 2, 3 and 4).

Numerous samples of Brazilian kaolins from several different sources were examined, together with a few of foreign origin for comparison. The Brazilian samples showed great variation of shape among the particles. The samples from Minas Gerais were collected in places where the kaolin is mined, but those from other localities are probably a few chance specimens from among all that are available at each locality. Samples from the state of São Paulo as a rule show aggregates where great associations of tubular forms are not infrequent. On the other hand, those from the state of Paraná, sedimentary in origin, show large hexagonal plates. Of the samples from the state of Rio, a few show small plates while others show tubes.

The present description is limited mainly to the kaolins from Minas Gerais which display more frequently the tubular morphology and represent deposits over a large area.

Some samples show only tubular particles (Fig. 5) while others show tubes together with ill-shaped flakes (Fig. 6). The tubes when accompanied by flakes of this type present signs of decay which the authors believe usually occurs in three different ways:

(1) By a kind of concentric cleavage so that the tubes are loosened

* This investigation was first reported to the Brazilian Academy of Science on Aug. 12, 1953, under the title "Morfologia tubular de certos caulins brasileiros."

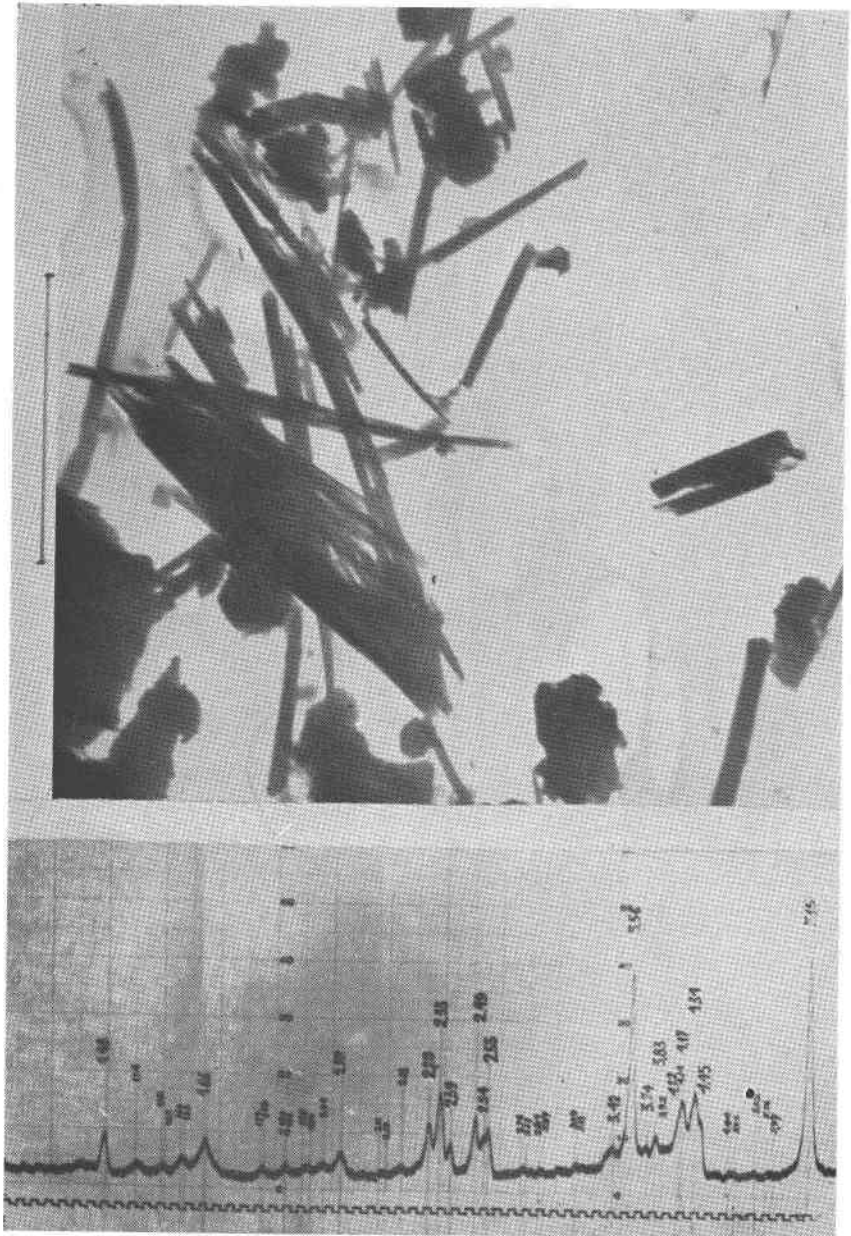


FIG. 1. Tubular kaolin particles from Minas Gerais, Brazil. Electron micrograph, $\times 40,000$. Philips diffractometer trace.

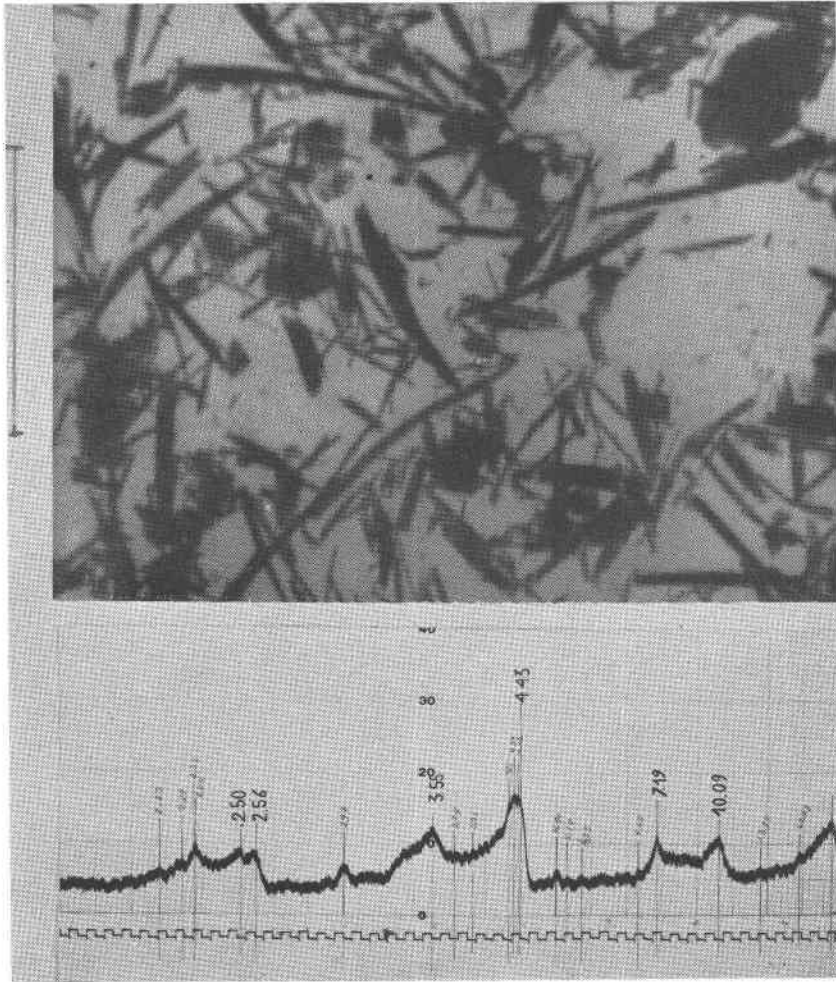


FIG. 2. Tubular halloysite from Djebel-Djebbar, Algeria. Electron micrograph, $\times 40,000$. Philips diffractometer trace.

and detached from the interior of the tubular formations leaving thinner walls and generating more thin tubes as a result of the cleavage.

(2) By cracking of the tube walls, thus giving rise to the "ill-shaped flakes."

(3) By an indiscriminate breakage of the tubes thereby producing more or less short tubes.

Generally, the three types of deterioration occur at the same time. The concentric cleavage is seen in electron micrographs showing one tube

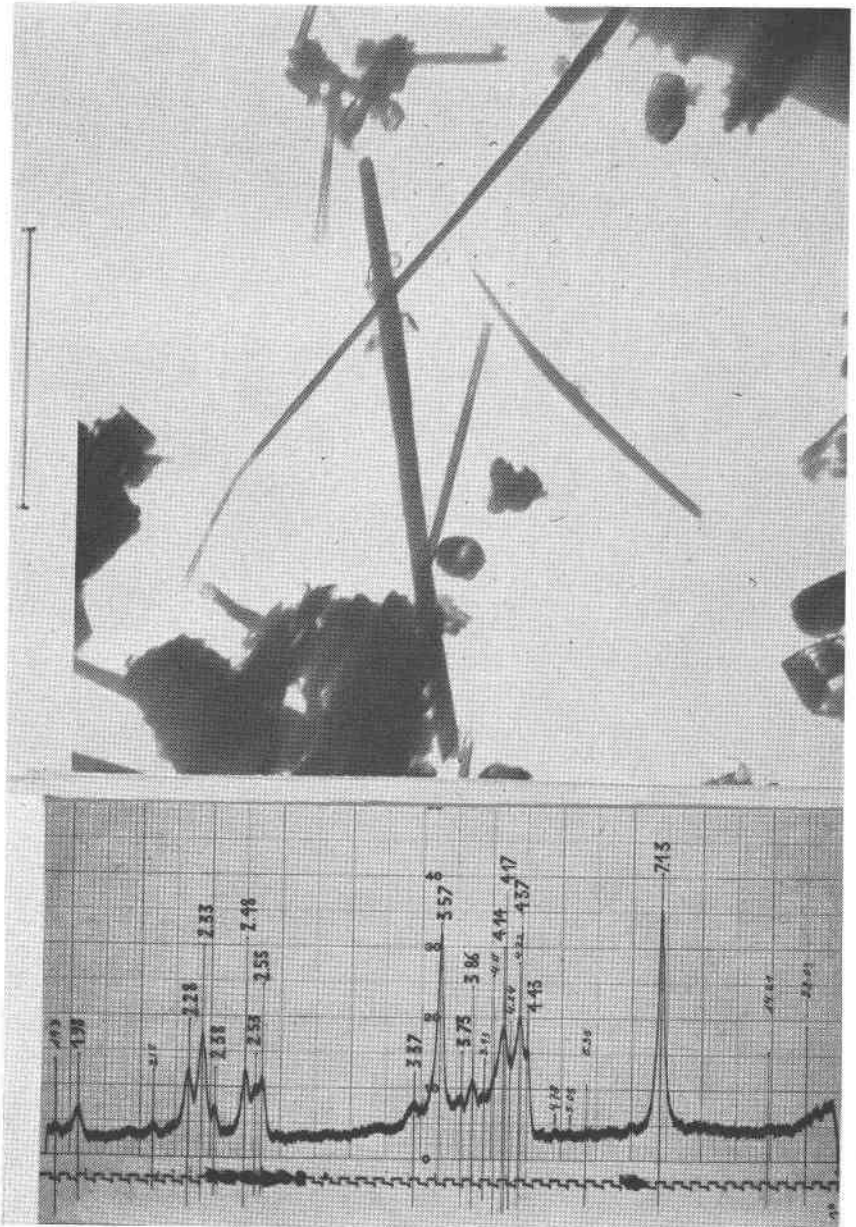


FIG. 3. A kaolinite from Minas Gerais, Brazil. Electron micrograph, $\times 40,000$. Philips diffractometer trace.

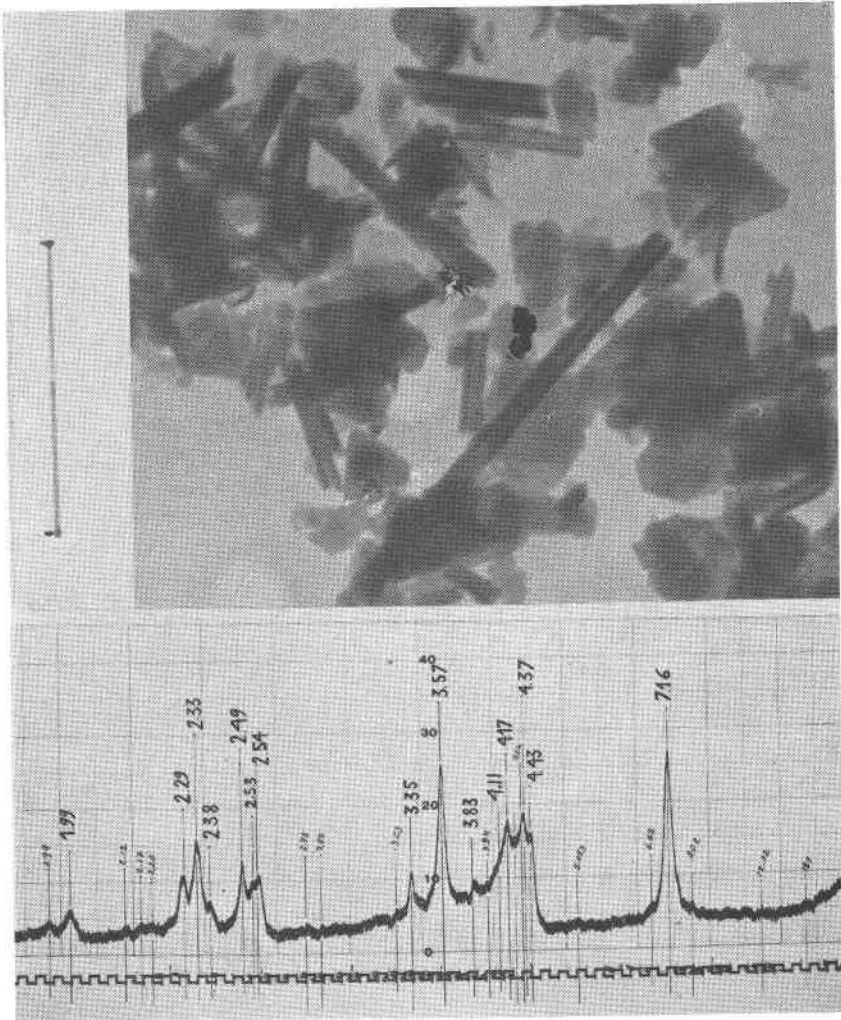


FIG. 4. Kaolinite from Les Eyzies, France. Electron micrograph, $\times 40,000$. Philips diffractometer trace.

emerging from another; it can be seen in Fig. 3. The destruction of the tubular form by cracking of the walls with the resulting production of ill-shaped particles is visible in Fig. 6 where tubes thick in the center and slender at both ends can be seen next to ill-shaped flakes. In other micrographs, tubes inside other tubes may be observed; in this case, light is noticeable along the walls and not along the central axis of the tube which is dark, meaning that tubes lie within outer tubes.

It is noticeable from the electron micrographs that the tubular formations become emptier and emptier as a result of the concentric cleavage. When the walls of the tubes remain unbroken, the tubes become thinner and thinner, until, when the wall is very thin, the tube may unroll more or less completely and ultimately elongated hexagons are left. The kaolin from Les Eyzies, France (Fig. 4) clearly illustrates this interpretation. Here, tubes and hexagonal, well developed but generally elongated plates are seen. The tubes are not dense nor well rolled, and most of them are almost completely unrolled. All the hexagonal plates seen are shorter than any one of the tubes. These are very long, but by careful observation it is possible to perceive by the degree of transparency that some of

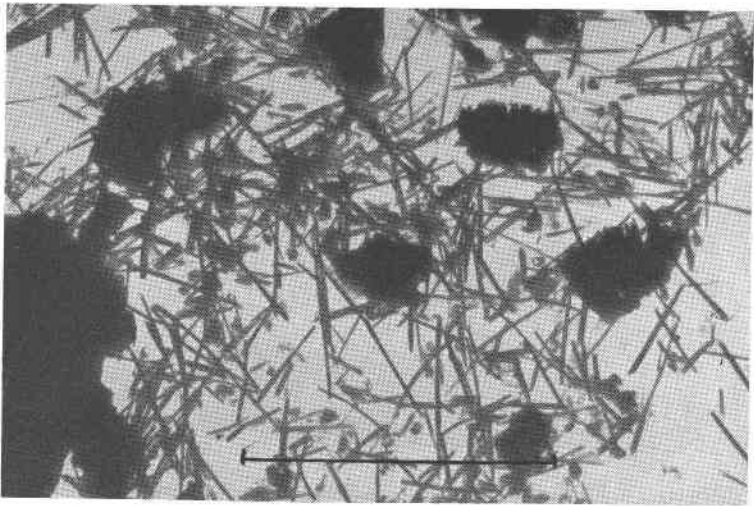


FIG. 5. A sample identified as kaolinite; from Minas Gerais, Brazil. $\times 7000$.

them are still attached to others by one of the extremities which means that they have not yet completely detached themselves from the others. When this occurs and when the unrolling has ended, hexagonal plates like those visible in the picture will remain. Apparently, before the tubes are completely unrolled, they have diminished in length, producing plates which are represented by more or less short and large hexagons. It is also possible that due to the cleavage, the elongated hexagons may be divided into many others of smaller size. The elongation of kaolin crystals observed by some authors may be explained in this way; such elongation may be reminiscent of their former tubular shape.

The above observations together with the fact that in sedimentary deposits the occurrence of tubes is non-existent or very rare instead of

frequent as in the residual kaolins from Minas Gerais, suggest an evolutionary process from the tube to the planar hexagonal form. Among the numerous residual and sedimentary samples examined, whenever the particles consisted of hexagonal plates, the percentage of tubes was practically none, but when the tubes predominated, the plates, if any, were ill-shaped.

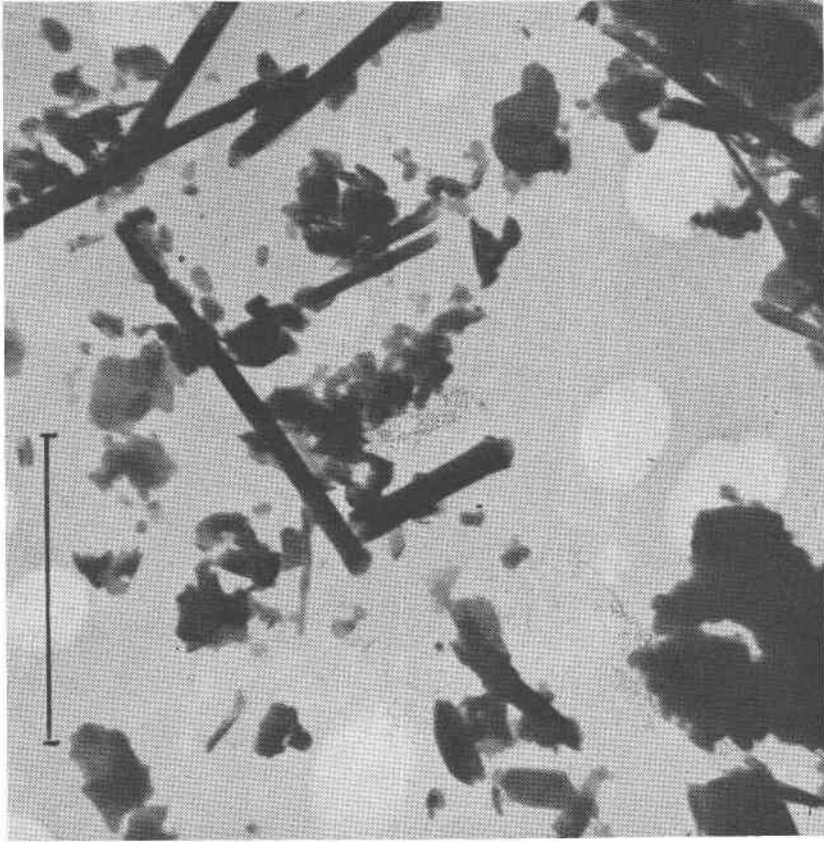


FIG. 6. Tubes next to ill-shaped particles; identified as kaolinite. From Minas Gerais, Brazil. $\times 40,000$.

It would be well to mention here that the authors do not think that all residual kaolins display a tubular morphology. The kaolin from St. Austell, England, for example, is of residual origin and shows hexagonal plates in the electron microscope; a hydrothermal action seems to be the principal factor in the genesis of this material. It is possible that, as

in this case, other factors may influence or modify the morphology of the kaolins.

Before examining the samples by the x -ray method, the various morphological types were selected in order to identify them one by one and to verify the existence of possible structural differences between them.

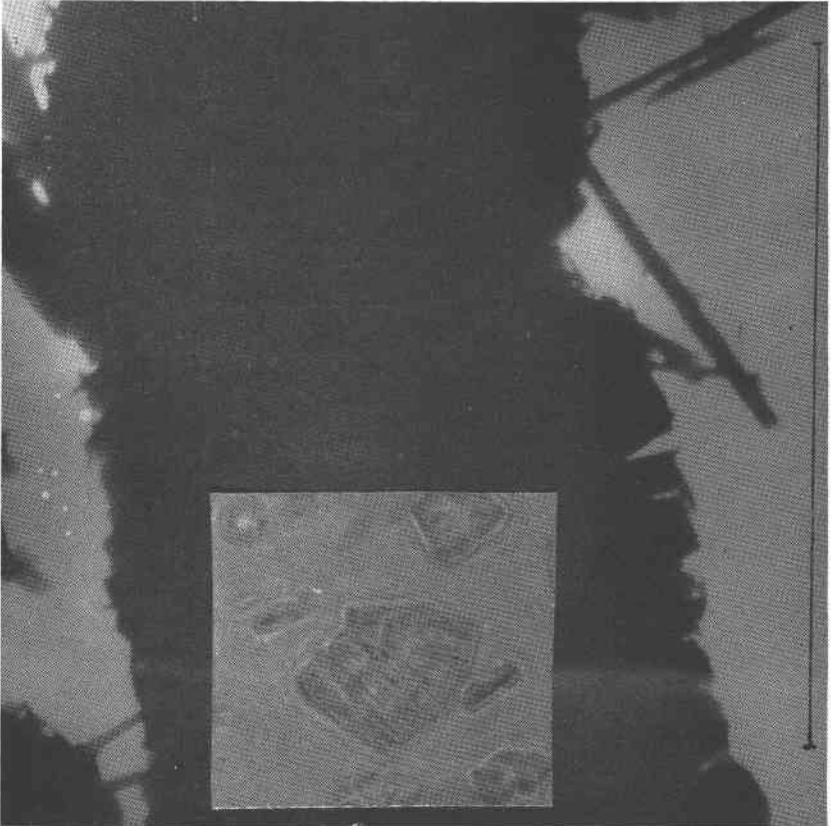


FIG. 7. Vermicular kaolin from Minas Gerais, Brazil. Electron micrograph showing large agglomeration of tubes, $\times 18,000$.

Inset shows micrograph taken with phase-contrast microscope, $\times 600$.

The results obtained with the Geiger counter diffractometer, together with the electron micrographs, have shown that:

- (1) Material which presented tubes only in the electron microscope was revealed as kaolinite by x -rays.
- (2) Material consisting of hexagonal plates only, gave x -ray diagrams apparently the same as those formerly obtained.

(3) Material consisting of tubes and ill-shaped particles, gave x -ray diagrams analogous to the former ones and also revealed kaolinite.

Furthermore, whenever the tubes are structurally the same as the hexagonal plates, this means that they consist of plates arranged in tubular formations which, when detached according to the natural planes of these formations, will produce hexagonal plates. It is not likely that each tubular formation should be the result of the rolling of a single membrane over itself because this would produce bands instead of peaks in the x -ray diagrams. Furthermore, the coexistence of tubes and plates structurally identified as kaolinite could hardly be explained if the tubes were not made of plates over plates.

The existence of kaolin in tubular form as described above, may help to clarify observations as yet not well explained, especially regarding the "vermicular kaolin." The existence of what is called vermicular kaolin has been mentioned for some time. The authors also have found kaolins of this type as revealed by microscope observation and still more clearly with the phase-contrast microscope (see inset to Fig. 7) but while studying this same material with the electron microscope, it was verified that the so-called vermicular kaolins consisted of large conglomerates or colonies of tubes placed side by side in close and more or less parallel orientation (see electron micrograph in Fig. 7).

An important question regarding the tubular morphology of kaolins, is its influence on the technical properties of this material. In spite of its great purity, the kaolin from Minas Gerais is not considered one of the best when compared with some foreign ones. It is possible that the "particle shape" factor may be partly responsible for its inferior quality as shown by many mechanical tests made by the authors.

The authors wish to take this occasion to express their deep appreciation to Professor G. W. Brindley of the Pennsylvania State University, University Park, Pa., for critically reading, revising and condensing the original manuscript in order to meet the Journal's requirements.

The editor regrets that it has not been possible to reproduce all the electron micrographs submitted by the authors. Certain observations in the paper, such as the large hexagonal flakes frequently shown by kaolinite, and the phenomenon of tubes emerging from tubes, which were well illustrated by the authors, have been frequently illustrated in other publications; these have been omitted for the sake of brevity.