Clays and Clay Minerals, Vol. 28, No. 2, pp. 97–104, 1980.

KAOLIN FROM THE ORIGINAL KAULING (GAOLING) MINE LOCALITY, KIANGSI PROVINCE, CHINA

W. D. KELLER,[1] HSIA CHENG,[2] W. D. JOHNS,[1] AND CHI-SHENG MENG[3]

Abstract—Although the specific outcrop from which the original kaolin at Kauling (Gaoling) Mine, China, was collected cannot now be relocated, samples were collected and studied from the mine tunnel, country rock, and pegmatite which constitute the sources of kaolin in this region. The kaolin is a residual product of weathering. Where the parent rock was a granite the clay is a mixture of platy and elongate kaolin-group minerals, whereas from the pegmatite portion of the parent rock it is halloysite(10Å) with elongate morphology. These mineral identifications are based on X-ray powder diffractograms, scanning electron micrographs, differential thermograms, and an infrared spectrum hitherto not documented for material from this area.

Although the Kauling locality is the region for which kaolin is named, the mineral kaolinite is a defined species without a specific type locality. The 11th century Chinese locality was not mentioned in the two classic research papers defining kaolinite. The data on the Chinese kaolin, therefore, cannot be used directly to provide criteria for sharply differentiating the otherwise vague boundary between the minerals kaolinite and halloysite(7Å). Pertinent questions on these kaolin-group mineral relationships are brought into clearer focus.

Key Words—China, Endellite, Halloysite, Kaolinite, Morphology, Nomenclature.

INTRODUCTION

This report documents data recently obtained from kaolin-group minerals collected in the region from which kaolin was named. As discussed below, kaolinite is a defined clay-mineral species, not identifiable with a specific source locality. The original locality for kaolin (rock), however, has long been reported as Kauling (Grim, 1968), alternatively as Kao-ling (or Gaoling in Pinyin spelling), Kiangsi Province (also spelled Jiangxi), China. Kaolin was produced, according to Chinese lore, from the Kauling Mine, near Kaulin Village, in a large development as early as the 11th Century, but the mine was depleted and closed in 1964. Thus, the original location, if there ever was one assignable to a particular outcrop or mine face, cannot now be recovered. Samples collected, however, from the tunnel of Kauling mine and other nearby samples from the parent muscovite-granite and pegmatite at or near Kaulin Village are representative of the region.

MATERIALS AND GEOLOGY

A schematic map showing the kaolin bodies, the associated varieties of granite (known as "Fulian" granite) and pegmatite, Kaulin Village, and collecting locations is reproduced in Figure 1.

Samples from locations 1 and 2 are from a partially weathered, medium-grained muscovite granite. Sample 3 is from weathered residuum of a pegmatite about 1.0 km from the main deposit. Sample 4 is a weathered residue of kaolinized muscovite granite, near Kaulin Village. These samples are believed to represent the kaolin at the original locality. Sample 5 is kaolin from an altered acid vein rock in Suzhou, Jiangsu (Kiangsu) Province, a neighboring province to Jiangxi (Kiangsi).

All samples are white, or slightly off-white; samples 1 and 2 are relatively harsh, i.e., only partly kaolinized feldspar. The small size of the samples received yielded less kaolin material than ideally desired. Sample 4 is typically partly kaolinized granite, i.e., it consists of quartz, decomposed feldspar, mica flakes, and white powdery kaolin. Samples 3 and 5 are white, compact, small lumps of relatively pure kaolin.

Presumably, samples 1, 2, and 4 represent the original kaolin if this material was derived from weathered granite. On the other hand, if the original kaolin was from the pegmatite, which produces a quartz-free, relatively pure white clay, it would be represented by sample 3. Sample 5 is mineralogically similar to sample 3, but geographically it was collected in the neighboring province.

LABORATORY RESULTS

The sieved, clay-size portion of sample 4 yields the X-ray powder diffractogram in Figure 2A. Kaolin-group mineral(s) and a small amount of mica, quartz, and probably feldspar are indicated. The kaolin-group mineral(s) apparently is not well ordered.

Scanning electron micrographs yield visual evidence of the mineralogy and course of weathering of the residual material. In Figures 3 and 11 feldspar that was pitted and corroded during weathering is speckled with

[1] University of Missouri–Columbia, Columbia, Missouri, USA 65211.
[2] Institute of Geology, Academia Sinica, Peking, PRC.
[3] Geological Society of China, Peking, PRC.

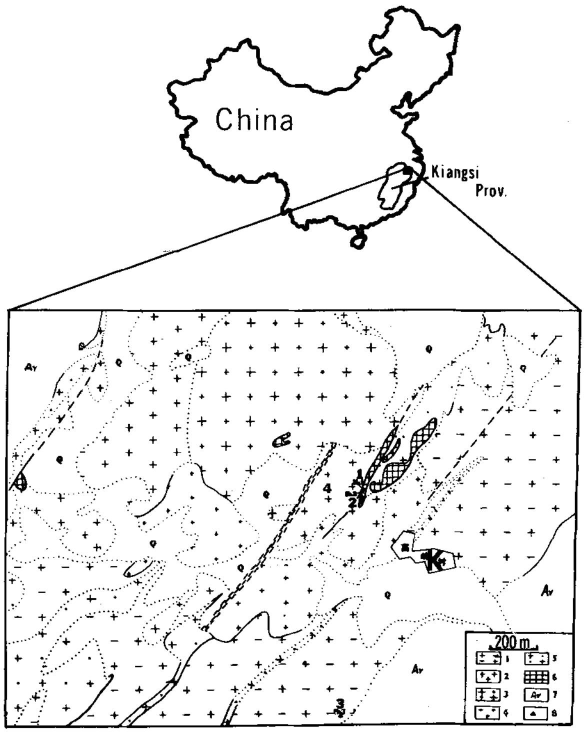


Figure 1. Schematic map of kaolin bodies, granites (Fulian granite group) and pegmatite, Kaulin Village, collecting localities, Kiangsi (Jiangxi) Province, and location in the PRC. Sample locations are designated in bold face numbers; Kaulin Village is marked by "K"; rock legends are: 1 = medium-grained biotite granite; 2 = medium-grained muscovite granite; 3 = medium-grained porphyritic granite; 4 = fine-grained granite; 5 = granite porphyry; 6 = kaolin; 7 = pre-Sinian metamorphic rock; 8 = sample localities, over-printed in bold-faced numbers, 1–4.

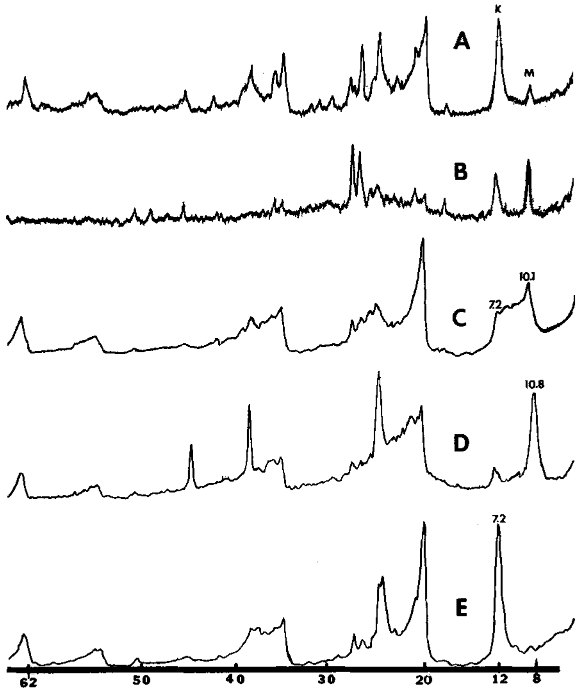


Figure 2. X-ray powder diffractograms, Cu Kα radiation, °2θ indicated along base. (A) Sample 4. Poorly ordered kaolin (K) and mica (M). (B) Samples 1 and 2. Typical, partially kaolinized feldspathic (granitic) rock; the primary silicates are largely decomposed while the kaolin is incompletely developed, yielding a poorly developed diffractogram characteristic of intermediate products. (C) Sample 3, halloysite(10Å) from pegmatite. The basal reflections are continuous from 10.1 to 7.2 Å showing the halloysite(10Å) to be in progress of collapse. (D) Sample 3 solvated in ethylene glycol yielding a basal reflection at 10.8 Å. (E) Sample 3 heated to 105°C, collapsed, and yielding a basal reflection at 7.2 Å.

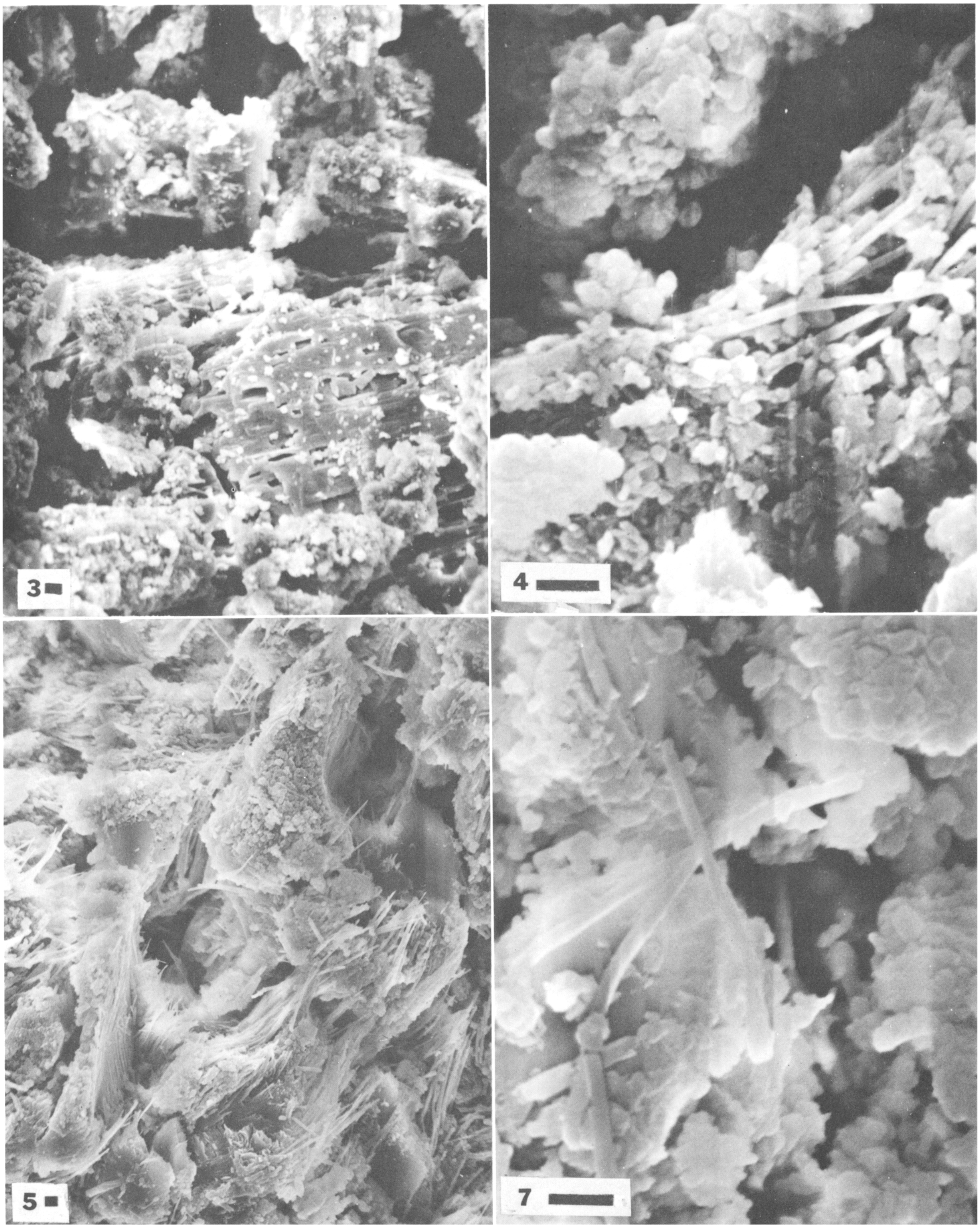
kaolin. This effect is typical of early-stage alteration of feldspar in granite, as has been observed in Georgia and elsewhere (Keller, 1977, Figures 28–40, 70–72). More completely kaolinized, smaller particles of the Kauling sample show at high magnification both elongate and platy morphologies of kaolin (Figure 4).

The low degree of ordering indicated by the X-ray powder diffractogram of sample 4 may be due to the combined effects of two intimately mixed kaolin-group minerals, i.e., elongate particles and plates, as has been demonstrated to result from artificial mixtures (Keller and Haenni, 1978). Alternatively, it may be due to immature crystallization accompanying weathering, or to whatever type of genetic environment that yields a low degree of ordering.

A typical differential thermal analysis (DTA) curve of kaolinite was produced from sample 4 heated in a Robert Stone instrument (Figure 6A). If the elongate crystals shown in the SEMs were originally halloysite(10Å), they must have converted to the 7-Å dehydrated variety which did not produce an endothermic reaction below 200°C.

Samples 1 and 2 were collected from the tunnel leading to Kauling (Gaoling) mine; both represent essentially the same material. The X-ray powder diffractogram, Figure 2B, taken from an unavoidably small sample of mulled suspension on a glass slide shows only weak reflections of kaolin-group minerals, mica, quartz, and feldspar. The diffractogram is illustrative of only partially kaolinized feldspathic (granitic) rock, typical of flux-containing "china clay" commonly used in the manufacture of ceramic white wares. Weathering has degraded the crystallinity of the feldspar and mica, but the kaolin-group mineral is not well enough developed and crystallized to yield a good diffractogram. The 7-Å reflection is skewed toward the wider spacing side.

Shattered feldspar and mica fragments in sample 2 have been altered to both clusters of plates and relatively long, curving elongate particles (Figure 5). Those from sample 1 show essentially the same type of ka-



Figures 3, 4, 5, and 7. (3) Scanning electron micrograph of sample 4. The length of the bar indicates 1 μm. (4) Scanning electron micrograph of sample 4 at higher magnification than Figure 3. The length of the bar indicates 1 μm. (5) Scanning electron micrograph of sample 2. The length of the bar indicates 1 μm. (7) Scanning electron micrograph of sample 1. The length of the bar indicates 1 μm.

https://doi.org/10.1346/CCMN.1980.0280204 Published online by Cambridge University Press

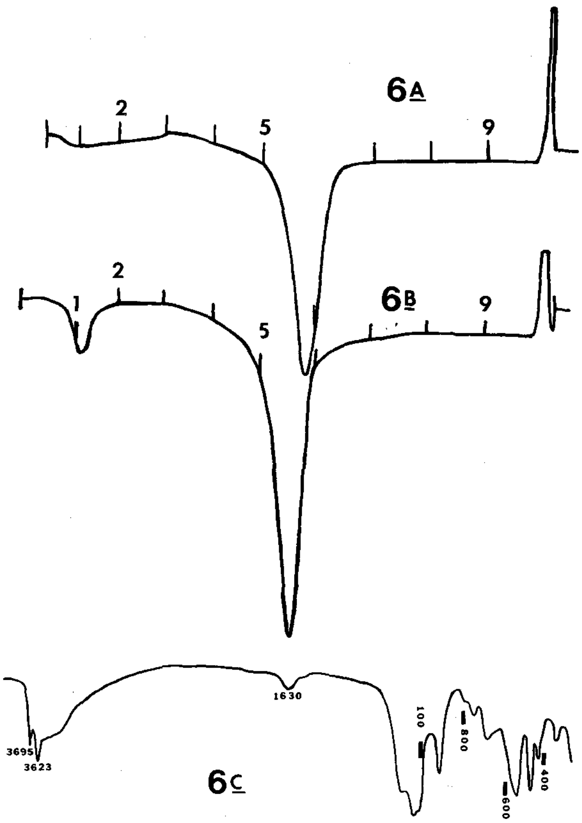


Figure 6. Differential thermal analysis curves (6A and 6B) run on a Robert Stone apparatus. Temperature is indicated in hundreds of degrees C. Curve 6A is of sample 4 and is typical of kaolinite. Curve 6B is of sample 3 and suggests halloysite(10Å), partly dehydrated, as indicated by the relatively low amplitude of the low-temperature endotherm, and corroborated by the partially collapsed basal spacing in the X-ray powder diffractogram. Curve 6C is an IR curve of sample 3 and is typical of halloysite(10Å). Wave numbers are indicated as cm^{-1}.

olinization (see Figure 7). As in sample 4, the kaolin in samples 1 and 2, from the tunnel to the old, original Kauling mine, is a polymineralic mixture of plates and elongate particles. The amount of the samples available was too small for DTA.

Sample 3, from the pegmatite about 1 km south of the Kauling Mine, is unequivocally halloysite(10Å). The small lump of clay, tightly wrapped, retained enough chemically combined water despite shipment from China to yield a broad, basal X-ray diffraction reflection spreading from 10.1 to 7.2 Å (see Figure 2C). Solvation in ethylene glycol expanded the spacing to a sharper peak at 10.8 Å (Figure 2D), which when heated to 100°–105°C collapsed to 7.2 Å (Figure 2E). Halloysite(10Å) from Gardner Ridge, Indiana, examined as a control sample yielded a 10.8-Å peak with ethylene glycol and a 7.3-Å peak when heated to 100°–105°C. In the DTA of the Chinese clay (Figure 6B), an endothermic peak at 110°C supports the identification of this material as halloysite(10Å). The relatively small amplitude of the dehydration peak supports the X-ray diffraction data which indicate partial collapse of the halloysite(10Å) structure.

The infrared spectrogram (Figure 6C) is typical of halloysite(10Å), i.e., it suggests the presence of water in addition to the $2H_2O$ in the formula (Van der Marel and Beutelspacher, 1976).

Scanning electron micrographs illustrate both the elongate and tubular morphology of the halloysite(10Å) and the course of its origin. Shattered feldspar (and mica?) particles are coated with "wool" or "whiskers" of elongate kaolinite (Figure 8) similar to those from Georgia occurrences (Keller, 1977, Figures 30–34, 69–71). In Figure 9 a mat apparently of elongate grains forms a vermicular "book" of needles instead of typical platy crystals. In this pegmatite, 1 km from the Gaoling mine, the kaolin mineral is dominately halloysite(10Å), not classic platy kaolinite.

Sample 5 is from an altered acid vein rock in Suzhou and is a mixture of plates and elongate grains, as shown in Figure 10.

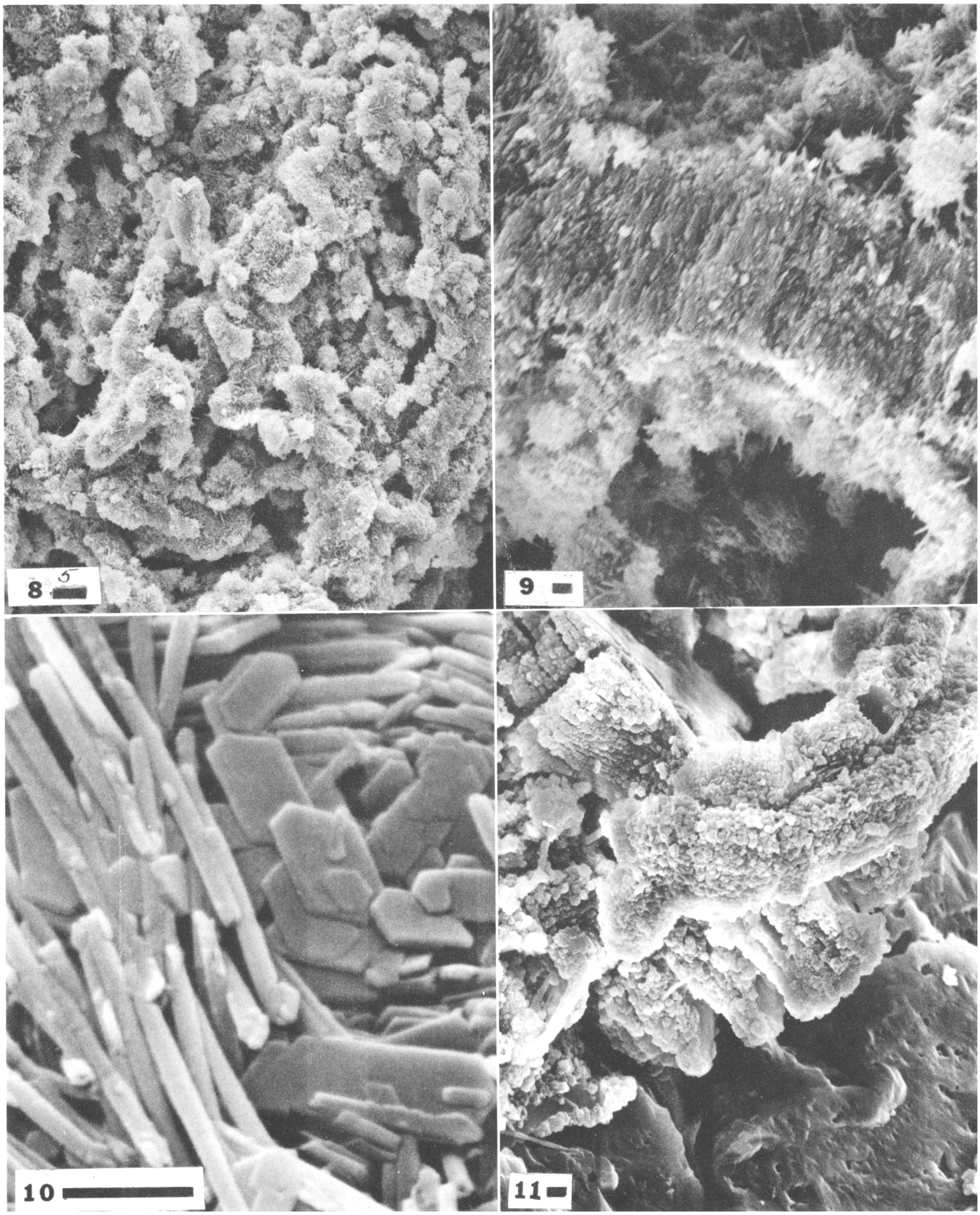
DISCUSSION AND CONCLUSIONS

The clay from Kauling, China, holds interest for at least two reasons: (1) it has a long historical record as a good ceramic material, and (2) being from the region for which kaolin (rock) was named, the kaolin mineral(s) contained in it should represent the clay mineral(s) of kaolin.

Kaolinite is a defined mineral which has no type locality. Johnson and Blake (1867) first proposed "Kaolinite, in allusion to the material [kaolin] which furnishes it most abundantly." Ross and Kerr (1931) then defined kaolinite "In common usage today, as well as when proposed, 'kaolinite' is taken to mean the clay mineral of kaolin." It follows, therefore, that kaolinite should be present in the type kaolin, although the Chinese locality was not mentioned by either Ross and Kerr, or Johnson and Blake. Parenthetically, however, it should also be realized that if "kaolin is defined as an earthy rock characterized by a significant (or dominant) content of a kaolin mineral"—a tentative definition by the International Committee on Correlation of Age and Genesis of Kaolin—the definitions are mutually circular.

Conclusions resulting from the examination of the Chinese kaolin are listed below and considered further in terms of a larger problem of identification, genesis, and stability of the kaolin-group minerals.

1. Kaolin produced by residual weathering of granite in the region for which kaolin was named is an intimate mixture of two morphologic varieties of kaolin-group minerals: plates and elongate grains.
2. From the weathered pegmatite in the region, the



Figures 8–11. (8) Scanning electron micrograph of halloysite(10Å) from sample 3 (pegmatite). The length of the bar indicates 5 μm. (9) Scanning electron micrograph of sample 3 (pegmatite) showing elongate crystals packed and matted in a book-like structure. The length of the bar indicates 1 μm. (10) Scanning electron micrograph of sample 5 (Suzhou, Jiangsi Province), showing elongate crystals and plates in sharp, immediate micro-contact. Note the two distinct morphologies. The length of the bar indicates 1 μm. (11) Scanning electron micrograph of sample of granite from the Kauling mine tunnel showing pitted feldspar and contemporaneous crystallization of platy kaolinite on mica during weathering. The length of the bar indicates 1 μm.

only kaolin-group mineral observed was identified as halloysite(10Å) collapsing to halloysite(7Å).

3. The platy crystals in the kaolin samples yield X-ray powder diffractograms and DTA curves that fall within the range of examples of kaolinite given by Ross and Kerr (1931).
4. The elongate crystals in the residuum appear similar to those from the pegmatite. Both (a) appear to meet the requisites of halloysite formally described by Chukhrov and Zvyagin (1966) "It is important to realize that halloysite does not, in fact, crystallize in the form of single crystals with a single lattice, but as elongated particles representing a complex combination of several radial zones diverging from one axis"; and (b) do not conflict in properties with most of the halloysite examples described by Ross and Kerr (1934). It should be noted, however, that not all of their examples possess the mineralogic properties typically distinctive of halloysite. The flint clay from Rolla, Missouri, cited as halloysite by them, has been found by one of the present authors (W.D.K.) to be composed of tiny intergrown packets of platy kaolinite and to yield an X-ray powder diffractogram and DTA curve comparable to those of kaolinite from the Chinese clay described in conclusion 3.
5. Thus far, the cited distinction between kaolinite and halloysite in the Chinese kaolin has been based on morphologic differences such as are visible by electron microscopy. However, is morphology a valid criterion for differentiating between kaolinite and halloysite? Whereas morphology, *per se*, can hardly be so fundamental as to serve as a basis for differentiation of minerals in general, Chukhrov and Zvyagin (1966), offered a basic reason why plates and elongate grains of kaolin-group minerals can represent two distinct mineral species. To examine this question more closely one must raise three more specific questions:
   a. Are all halloysite crystals necessarily elongate?
      (1) It would appear that Chukhrov and Zvyagin (1966) thought that halloysite is elongate when they wrote ". . . halloysite . . . crystallize(s) . . . as elongate particles . . . ."
      (2) Brindley and de Souza Santos (1966), however, cast doubt upon elongate morphology as a necessary criterion for halloysite: "The present writers, with Helena de Souza Santos, have studied a platy kaolin-group mineral from Pocos de Caldas, Minas Gerais, Brazil, which conforms much more strikingly with the behavior of halloysite. In its normal wet state (which must be preserved at all time) the mineral has a basal spacing of 10.05 Å, which expands readily in ethylene glycol to 10.90 Å, and in dry atmosphere collapses largely irreversibly to a spacing of about 7.25 Å. All the diffraction patterns very closely resemble those of halloysite, but exhibit a small orientation effect consistent with the platy morphology. If it is called 'halloysite,' then a rolled and fibrous morphology is not an essential characteristic of halloysite."
   b. Are all elongate kaolin-group minerals halloysite?
      (1) While it might be presumed from the description of halloysite by Chukhrov and Zvyagin (1966) that they considered all elongate kaolin-group minerals to be halloysite (or later discovered imogolite), this question is not specifically addressed in their paper.
      (2) Brindley and de Souza Santos (1966) left doubt that all elongate crystals are halloysite "A more striking example of structural order in a fibrous kaolin-group mineral has been obtained by the present writers and H. de Souza Santos. A macroscopically fibrous mineral, from Piedade, Sao Paulo, Brazil is seen under the electron microscope to consist of fine fibers, probably tubes, many microns in length. Macroscopic bundles give X-ray diffraction patterns showing evidence for considerable stacking order . . . although the diffraction data have not yet been fully evaluated, the evidence for an appreciable degree of order in some fibrous forms seems beyond question."
   c. Are criteria available, either theoretical in scope, experimentally observable, or arbitrarily defined, that can be used to differentiate unequivocally between highly disordered kaolinite and halloysite(7Å)?
      (1) As matters stand at present, there appears to be a realm in which it is impossible to differentiate between kaolinite and halloysite(7Å). Chukhrov and Zvyagin (1966) stated "At the extreme degree of randomness, when it is impossible to establish the details of any model, the two series converge." Such is their conclusion despite their statement that well developed kaolinite and halloysite are distinct mineral species!
      (2) Halloysite(10Å), however, is clearly distinguishable from kaolinite by X-ray powder diffraction, DTA, IR, chemical composition, and indices of refraction. This phase is probably more clearly defined with limiting measurable properties than either kaolinite or halloysite(7Å). This is the kaolin-group mineral named endellite by Alexander *et al.* (1943) and endorsed by Fleischer, Frondel, Hendricks, Kulp, Ross, and Schaller, as

quoted by Faust (1955) (see also Keller and Johns, 1976), but not approved by the AIPEA Committee on Nomenclature (written communication from S. W. Bailey, Department of Geology, University of Wisconsin, Madison, Wisconsin).

6. Obviously, rigorous characterization criteria for kaolinite and halloysite(7Å) do not exist today—and scant help was furnished by the few data obtained from the kaolin material from the Kauling locality. Adequate definition of limiting properties would lead to a tidier nomenclature, unambiguous name tags, and more precise crystallography and evaluation of crystal bond energies. It could also help to clarify procedures used to investigate genesis, thermodynamic relationships, geologic occurrences, and paragenesis of the individual kaolin-group minerals. For example, in the occurrence illustrated by Figure 10, did the two morphological forms crystallize simultaneously—if so, were they both stable, and how can differences in free energies of formation between them be reconciled? If one form crystallized before the other, which one developed first, and chemically why? If they are not both stable, which mineral is transforming into the other and can ground-water environments change so as to reverse the transformation?

ACKNOWLEDGMENTS

Two of us (WDK and WDJ) are grateful to Drs. P. Y. Chen and W. H. Huang for their Chinese-English translations. We are indebted to Prof. E. E. Pickett who prepared the IR spectrogram. Scanning electron micrographs were prepared with assistance from National Science Foundation Grant EAR 76-18804 to W.D.K.

REFERENCES

Alexander, L. T., Faust, G. T., and Hendricks, S. B. (1943) Relationship of the clay minerals halloysite and endellite: *Amer. Mineral.* **28,** 1–18.

Brindley, G. W. and de Souza Santos, P. (1966) New varieties of kaolin-group minerals and the problem of finding a suitable nomenclature: *Proc. Int. Clay Conf. Jerusalem* **1,** 3–9.

Chukhrov, F. V. and Zvyagin, B. B. (1966) Halloysite, a crystallochemically and mineralogically distinct species: *Proc. Int. Clay Conf. Jerusalem* **1,** 11–26.

Faust, G. T. (1955) The endellite–halloysite nomenclature: *Amer. Mineral.* **40,** 1110–1118.

Grim, R. E. (1968) *Clay Mineralogy:* McGraw-Hill, New York, 596 pp.

Johnson, S. W. and Blake, J. M. (1867) On kaolinite and pholerite: *Amer. J. Sci.* 2d ser. **43,** 351–361.

Keller, W. D. (1977) Scan electron micrographs of kaolins collected from diverse environments of origin—IV. Georgia kaolin and kaolinizing source rocks: *Clays & Clay Minerals* **25,** 311–345.

Keller, W. D. and Haenni, R. P. (1978) Effects of micro-sized mixtures of kaolin minerals on properties of kaolinites: *Clays & Clay Minerals* **26,** 384–396.

Keller, W. D. and Johns, W. D. (1976) "Endellite" will reduce ambiguity and confusion in nomenclature of "halloysite": *Clays & Clay Minerals* **24,** 149.

Ross, C. S. and Kerr, P. F. (1931) The kaolin minerals: *U.S. Geol. Surv. Prof. Pap.* **165E,** 151–175.

Ross, C. S. and Kerr, P. F. (1934) Halloysite and allophane: *U.S. Geol. Surv. Prof. Pap.* **185G,** 135–148.

Van der Marel, H. W. and Beutelspacher, H. (1976) *Atlas of infrared spectroscopy of clay minerals and their admixtures:* Elsevier, New York, 396 pp.

(Received 21 May 1979; accepted 17 November 1979)

Резюме—Хотя нельзя точно определить обнажение пород, из которого был отобран первоначальный каолин в районе шахты Каулинг (Гаолинг), Китай, были собраны и изучены образцы из шахтного туннеля, местных пород и пегматита, которые являются источниками каолина в этом районе. Каолин представляет собой остаточный продукт выветривания. Если материнская порода была гранитом, глина является смесью пластинчатых и удлиненных минералов каолиновой группы, тогда как из пегматитовой части материнской погоды образуется галлуазит(10Å) с удлиненной морфологией кристаллов. Эти минералогические определения основаны на рентгеновских порошковчх диффрактограммах, сканирующих электронных микроснимках, дифференциальных термограммах и инфракрасном спектре до этого не использовавшемся для материала из этого района.

Хотя местность Каулинг является районом, от которого каолин получил свое название, минерал каолинит является определенной породой без определенного типа местонахождения. Китайское местонахождение 11 века не было упомянуто в двух классических исследованиях, определяющих каолинит. Следовательно, сведения о Китайском каолините не могут быть использованы непосредственно, чтобы обеспечить критерий для резкого разграничения в других отношениях не ясной границы между минералами каолинит и галлуазит(7Å). Освещаются существенные вопросы этих отношений каолиновых минеоалов. [N. R.]

Resümee—Obwohl der spezifische Aufschluß, von dem der ursprüngliche Kaolin in der Kauling (Gaoling) Mine, China, gesammelt wurde, jetzt nicht mehr festgestellt werden kann, wurden Proben aus dem Minen-Tunnel vom anstehenden Gestein und vom Pegmatit, der das Ausgangsmaterial für den Kaolin in dieser Gegend darstellt, gesammelt und untersucht. Der Kaolin ist ein Restprodukt der Verwitterung. Wo das Ausgangsmaterial ein Granit war, besteht der Ton aus einer Mischung von plattigen und länglichen Kaolinmineralen, während aus dem pegmatitischen Anteil des Ausgangsmaterials Halloysit(10Å) mit faseriger Ausbildung entstand. Diese Mineralbestimmungen erfolgten mittels Röntgenpulverdiffraktometrie, Rasterelektronenmikroskopie, Differentialthermoanalyse und einem Infrarotspektrum, die bisher von dem Material dieses Gebietes noch nicht vorliegen.

Obwohl die Kauling Lokalität das Gebiet ist, nach dem Kaolin benannt wurde, ist das Mineral Kaolinit eine definierte Mineralart ohne eine spezifische Typlokalität. Dieses chinesische Vorkommen, bekannt seit dem 11. Jahrhundert, wurde in den beiden klassischen Schriften, in denen Kaolinit definiert ist, nicht erwähnt. Die Ergebnisse über den chinesischen Kaolin können daher nicht direkt verwendet werden, um Kriterien für eine scharfe Unterscheidung zwischen den sonst ungenau begrenzten Mineralen Kaolinit und Halloysit(7Å) zu liefern. Sachgemäße Fragen zu diesen Kaolinmineralbeziehungen werden näher diskutiert. [U.W.]

Résumé—Malgré que l'affleurement exact d'où a été ramassée la kaoline originale à Kauling (Gaoling), en Chine, n'a pas pû être retrouvé, des échantillons du tunnel de la mine, de la roche régionale, et de la pegmatite qui sont des sources de kaoline dans cette région, ont été rassemblés et étudiés. La kaoline est un produit résiduel de l'altération. Lorsque la roche mère est un granite, l'argile est un mélange de minéraux plats et allongés du groupe kaoline, tandis que de la portion pegmatite de la roche mère, elle est une halloysite(10Å) ayant une morphologie allongée. Les identifications de ces minéraux qui jusqu'à présent n'étaient pas documentés pour des matériaux de cette région sont basées sur des diffractogrammes aux rayons-X, des micrographes balayants d'électrons, des thermogrammes différentiels, et un spectre infrarouge.

Quoique la localité de Kauling est la région qui a donné son nom à la kaoline, le minéral kaoline est une espèce définie sans localité type. La localité chinoise du onzième siècle n'était pas mentionée dans les deux rapports scientifiques classiques définissant la kaolinite. Les données provenant de la kaoline chinoise ne peuvent par conséquent pas être utilisées directement pour fournir des critères distinguant précisément les limites autrement vagues entre les minéraux kaolinite et halloysite(7Å). Des questions pertinentes concernant les relations kaoline-minéraux deviennent plus claires. [D.J.]