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Formation of laths in fine-grained micas and its relationship to stacking mechanism

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SUMMARY. Transmission electron images of fine-grained micas from various clay deposits exhibit lath-shaped units arranged in modes similar to the stacking of layers in different mica polymorphs. Micas in a Georgia bentonite display two sets of parallel laths intersecting at an angle of 120°. Selected area electron diffraction (SAD) of these micas gives a sharp spot pattern indicating a perfect registry between the lattices of individual laths. Furthermore, the SAD pattern has an intensity distribution similar to that of a $2M_1$ dioctahedral mica. Another mica in an India bentonite shows one set of parallel laths with an intensity distribution expected from a 1M biotite. Some of the micas in the Marblehead illite exhibit three sets of parallel laths in a trigonal arrangement similar to the stacking in 3T mica.

THE mechanism of formation of different stacking sequences in micas is one of the most intriguing problems in mineralogy. Crystallographic and petrologic aspects of the problem were well defined by Smith and Yoder (1956). One of their conclusions concerned the effect of structural factors on the formation of different stacking sequences. In subsequent years many investigators have searched for these structural factors by refining crystal structures of a large number of micas; Güven (1971) gave a recent account of the structural factors by considering the results of these crystal structure determinations. Another approach to the problem was developed from direct observations on crystal growth features. Following the advancement of the screw dislocation theory of crystal growth by Frank (1949), and by Burton *et al.* (1949), the surface topography of micas was examined by Amelinckx (1952), and by Amelinckx and Dekeyser (1953); they proposed the screw dislocation mechanism for the growth of mica polymorphs. Recently, a variety of growth spiral patterns have been beautifully demonstrated by Sunagawa (1964) on a natural phlogopite and by Baronnet (1972) on synthetic phlogopites. There is no doubt that a screw dislocation mechanism is operative during the crystal growth of many micas. The question is whether the same mechanism is also responsible for the formation of different stacking sequences in these minerals. For this purpose one needs to show that growth spirals have step heights equal to or a submultiple of the unit-cell heights. Thus, both spiral step heights and the *c*-parameters of mica unit-cells must be determined unambiguously and accurately. However, this has not yet been done. Sunagawa (1962) and Baronnet (1972) attempt to identify different mica polymorphs by the morphology of growth spirals; this method for determining mica polymorphs seems to be rather hazardous.

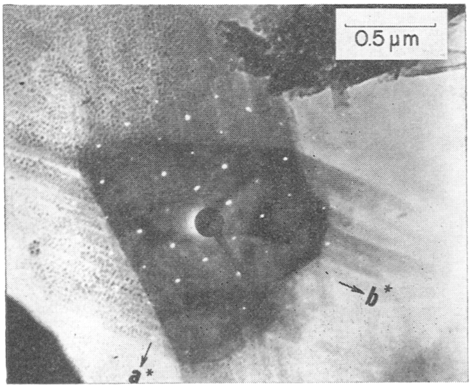
Fine-grained natural micas were not previously considered for extensive studies of growth mechanisms. Generally, they do not suffer from imperfections caused by sample preparation. The cleaving process of large micas may cause the screw dislocations to migrate out of the mica flake. It is, therefore, a very delicate matter to observe growth spirals, as was pointed out by Amelinckx and Dekeyser (1953). Fine-grained micas, frequently found as impurities in clays, may represent the initial stages of the growth process. Morphological features of these micas, reported below, may therefore add another perspective to the mechanism of stacking sequences in micas.

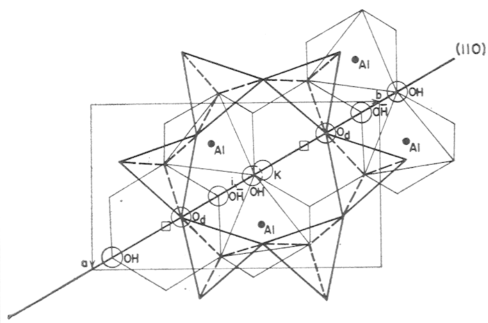
Presence of laths and their modes of arrangement in fine-grained micas

Three clays shown by X-ray powder diffraction patterns to contain micas were studied: bentonite from Akly deposit, Rajasthan, India; Wrens fuller's earth from Georgia, U.S.A.; Marblehead illite from Fond du Lac County, Wisconsin, U.S.A.

The < 2 μm fractions of these clays were separated and a drop of suspension was added to a mixture of water and tertiary butylamine so that the final concentration of clay was a few ppm. Electron-microscope grids were prepared by drying a drop of this suspension on a formvar film. They were subsequently shadowed with gold or a gold-palladium alloy at an angle of 26°. The grids were examined using a JEM-7 electron microscope operated at 80 kV and equipped with a completely variable field limiting aperture.

Fine-grained micas in Wrens fuller's earth show two sets of parallel laths (fig. 1), which are oriented at an angle of 120° to each other. The superimposed SAD pattern shows sharp spots, indicating that the laths have strict crystallographic orientation with respect to each other. The intensity distribution of the pattern is very similar to that of $2M_1$ dioctahedral micas. The observed intensity relationship, $I_{02} \ll I_{11} \approx I_{1\bar{1}}$ forms a diagnostic criterion for $2M_1$ dioctahedral micas as shown with experimental and theoretical data by Güven (1974). The widths of these laths vary between 0·03 and 0·2 μm. The thickness and length of the laths were measured on separate single laths occurring in the sample, as these dimensions are difficult to follow when the laths are aggregated. The laths are mainly visible at the edges of the mica flake. The thickness of the individual laths, as measured by Au-Pd shadowing, varies between 10 and 50 Å and their lengths between 1 and 3 μm. The inner portion of the mica (fig. 1) resembles an ordinary mica with no discontinuities, and displays the usual extinction contours arising from bending and thickness variations. It displays two well-developed forms at an angle of 60°, but these forms are not parallel to the laths. The crystallographic orientation of the laths can be determined from the superimposed selected area electron diffraction pattern. After a correction for the relative rotation between image and diffraction pattern it was found that one set of laths makes an angle of 30° with the **b*** direction of fig. 1. The laths therefore lie parallel to the [$\bar{1}$10] direction as referred to the $2M_1$ mica cell (fig. 2). This set of laths becomes parallel to the **a**-axis if it is referred to the 1M mica cell. As seen on fig. 2 the laths are bounded by (110) planes of the $2M_1$ cell passing through hydroxyl groups, oxygens, and vacant sites in dioctahedral micas. The (110) plane then forms the contact plane between the laths. If the association between laths proceeds over the OHs and oxygens in fig. 2, this will provide exact registry between the individual lattices of the laths. The boundary between the laths in this case will not cause any phase difference between the electrons passing through either side. Therefore, no contrast will be created at these boundaries and they will be invisible. This is the case in the inner portion of the mica. If, however, some slip occurs in the (110) plane this will cause phase difference for the transmitted electrons on either side of the boundaries and the resulting contrast may make the boundary visible. This may be the case for the laths at the periphery of the mica.

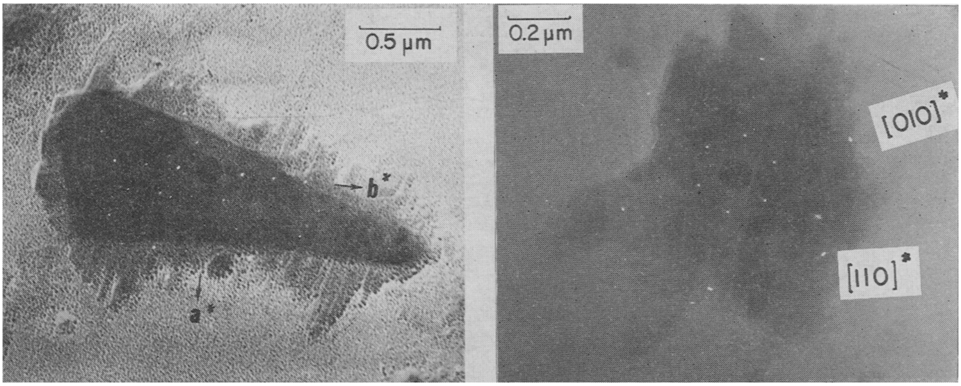




FIGS. 1 and 2: FIG. 1 (left). A typical mica in a Georgia bentonite, displaying reticulated laths with superimposed SAD spot pattern. (**a*** and **b*** are the reciprocal lattice directions.) FIG. 2 (right). The projection of $2M_1$ muscovite structure on the (001) plane. The contact plane between two laths is (110) and it passes through the ions designated in the figure.

Whether these laths are to be considered as twin individuals or as ribbons of unit-cell thickness is another question. This may be resolved by the SAD pattern since we can only measure the thickness of the superimposed laths in the mica. For a twinned association the SAD pattern would be expected to have hexagonal symmetry only if both twin sets are of equal volume. In the second case, the laths of unit-cell thickness will form a truly $2M_1$ sequence and the SAD pattern will show the characteristic intensity relationship: $I_{02} \ll I_{11} \approx I_{1\bar{1}}$. The observed SAD pattern for the above mica indicates that the laths are of unit-cell thickness.

The fine-grained micas in the India bentonite sample show a different arrangement of laths (fig. 3). There is one prominent set of parallel laths oriented nearly parallel to the **a*** direction of the superimposed SAD pattern after rotational correction of the image. Again, the central portion of the figure gives the appearance of an ordinary mica, and the laths are only visible at the edges. The SAD pattern was taken from the central portion of the flake containing the prominent set of laths. The intensities of the (02), (11), and (1$\bar{1}$) reflections have the relationship: $I_{02} \gg I_{11} > I_{1\bar{1}}$, which is very similar to the SAD pattern of 1M biotites. It is interesting to note that there is also a less conspicuous set of laths visible near the left edge of the mica flake in fig. 3, making an angle of 120° with the prominent set of laths. At this region of the mica a few growth spirals are also barely visible. Since these spirals appear on the thicker section of the mica flake, they seem to develop at a later stage of growth. In fact, if



FIGS. 3 and 4: FIG. 3 (left). A typical mica in an India bentonite with superimposed SAD spot pattern. The **b***-direction refers to 1M mica cell. FIG. 4 (right). An hexagonal arrangement of laths in micas from Marblehead. The reciprocal lattice directions [010]* and [110]* are tentatively assigned with respect to the $2M_1$ cell of dioctahedral micas. Note that the SAD pattern is rotated about 28° counter-clockwise.

laths with unequal thickness come in contact with each other, this may readily give rise to steps, which can wind up into spirals during the subsequent stage of crystal growth.

Micas with a strict geometrical arrangement of laths were previously described from the Marblehead illite (Güven, 1972). Some of these display a trigonal arrangement of three sets of laths, giving a hexagonal SAD pattern (fig. 4). A similar image from another illite aggregate was previously interpreted (Güven, 1972) as the disintegration mechanism of micas, and this possibility can also be considered for the formation of lath-shaped units in Wrens fuller's earth and in India bentonite. There is, of course, a close and often reciprocal relationship between growth and disintegration mechanisms. At least one may shed light on the other.

In conclusion, transmission electron images of the above fine-grained micas in clay deposits display geometrical arrangements of laths, which indicate that:

Lath-shaped units can associate in such a way that they form a perfect registry between their individual lattices. By doing so, they may laterally grow into layers. Thus, laths may be

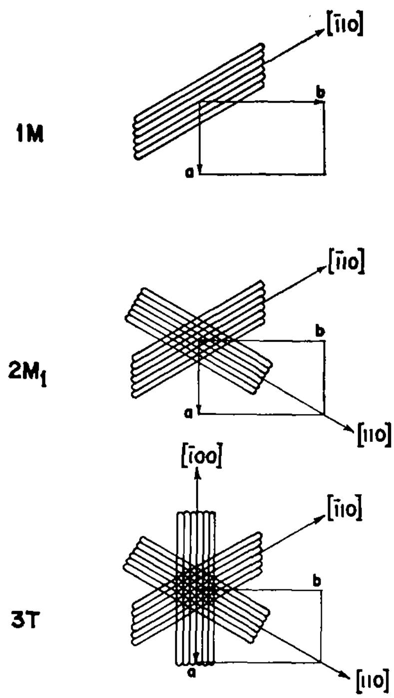


FIG. 5. Arrangement of laths similar to the stacking modes in 1M, $2M_1$, and 3T micas (in reference to the $2M_1$ muscovite cell).

considered as 'building blocks' for one mode of crystal growth in these layer silicates.

Parallel sets of these laths can form arrangements in a third dimension similar to the stacking of layers in mica polymorphs. This has been schematically represented in (fig. 5). The mica in Wrens fuller's earth has two sets of laths arranged at 120° to each other, similar to the stacking of layers in $2M_1$ micas. Fine-grained micas in the India bentonite display one set of parallel laths with no rotation between them, similar to the arrangement of layers in 1M micas. Finally, some of the Marblehead illite exhibits a trigonal arrangement of laths similar to that of single layers in a 3T mica.

The strict geometrical alignment of laths, resulting in new lattices, may be referred to as a 'reticulated arrangement'.

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