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GROWTH SPIRALS ON PHLOGOPITE CRYSTALS

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ABSTRACT

Five-sided growth spirals are observed on phlogopite crystals occurring in druses of trachybasalt lavas. From an analysis of the morphology of the growth spirals, it is concluded that the phlogopite belongs to the 1M polymorph. Polymorphs consisting of many layers such as 8Tc or 12M were not observed on the crystals investigated. From the orientations of five-sided growth features, it is shown that most of the crystals consist of many domains which are in twin relationship to each other. Twins by rotation around c' axis with rotation angles $\omega=60°$, 120° and 180° have been observed. It is considered that these twins are not polysynthetic twins but are of the ordinary rotation type.

INTRODUCTION

Since Verma observed interlaced growth spirals on SiC (Verma, 1951) and Frank put forward on the basis of Verma's observations a theory that different polytypes of SiC are generated from screw dislocations having different Burgers' vectors through a spiral growth mechanism (Frank, 1951), observations and theoretical considerations supporting Frank's theory have been reported on several substances (CdI_2 by Mitchell, 1956; SiC by Verma, 1951, Mitchell, 1957, and Sunagawa, 1963; hematite by Sunagawa, 1958, 1961; etc.).

Since mica polymorphs are derived from different ways of stacking of the fundamental mica layer, Frank's theory offered a new problem on the origin of mica polymorphs. In this sense, it has been realized that studies of surface topographies of mica crystals would give fundamentally important information concerning the origin of various polymorphs in micas (Smith and Yoder, 1956). However not much work has been done along this line, except that by Tolansky and his co-workers (1945, 1946a, b, 1960) and by Amelinckx and Dekeyser (1953). Tolansky's works deal with the surface topography and etch features of the cleavage faces of muscovite, biotite and phlogopite. Although Tolansky and his co-workers have shown that the flatness of the cleaved surfaces is different on different micas, their observations are not directly related to the mechanism of crystal growth, nor to the formation of the various polymorphs. Amelinckx and Dekeyser observed interlaced growth spirals on a biotite crystal from Monte Somma and interpreted the pattern as having been generated from polytypes of many layers, which were formed through a spiral growth mechanism from screw dislocations having large Burgers' vectors. This work is perhaps the only one that has been done on the relation between growth mechanism and the origin of various polymorphs or polytypes of micas and therefore it has been hoped that the work along this line would be done on the various mica species, as suggested in the paper by Smith and Yoder (1956).

In view of this, the writer has made detailed observations on the surface structures of natural basal planes of phlogopite crystals from several localities in Japan, using a sensitive phase contrast microscope and an interferometric microscope. The phlogopite crystals observed in this study were collected from Mutsure-zima, Shimonoseki-shi; Mutsumi-mura, Yamaguchi Pref.; and Nokono-shima, Fukuoka Pref., in Japan. They occur in druses of trachybasalt lavas of the Pleiocene age, as thin flaky crystals of 1 to 10 mm in diameter, together with hornblende crystals, and are considered to be formed at the latest stage of solidification of the trachybasalt magma under the condition of high vapor pressure. The phlogopite from Mutsure-zima has been studied by several workers (Kozu and Yoshiki, 1929; Kozu and Tsurumi, 1931; Tomisaka, 1958; Sadanaga and Takeuchi, 1961) and was identified by *x*-ray methods as a biotitic phlogopite with the structure of the 1M polymorph (Tomisaka, 1962). Crystals from the other localities show similar modes of occurrence as those from Mutsure-zima and are considered to have similar composition, structure and origin.

OBSERVATIONS

Crystals were mounted with Canada balsam on glass plates and silvered in a vacuum evaporation plant to secure high reflectivity necessary for observation under a reflection microscope. Since the surfaces of the crystals are somewhat wavy, phase contrast effect can be obtained only in a small area on the surface. However, because of the existence of impurity crystals preferentially adsorbed along growth steps, in addition to the high reflectivity obtained by silvering, visibility was high enough to detect surface features having very small step heights even under an ordinary reflection microscope.

The following three characteristic surface features have been observed on the basal planes:

1. Straight lines: There are several directions, which are inclined to each other at the angles of 30°(=150°), 60°(=120°) and 90°. Many are parallel. Most of the lines do not run across the entire surface, but extend only for short distances.
2. Five-sided patterns with straight sides: These are formed with the straight lines described above. In an extreme case, they take on equilateral triangular form.
3. Five-sided growth spirals with curved corners. Some spirals take on more deformed forms but are deviations from the five-sided spiral.

Figure 1 is a phase contrast photomicrograph showing the simplest growth spirals observed. Two spirals, right and left handed, are seen.

Both spirals exhibit a regular five-sided pattern (hexagon with one side truncated) in the center, but the spiral layers become more deformed as they spread outerwards. They are neither hexagonal nor trigonal and have only one symmetry plane, which is in accord with the symmetry of the basal plane of a monoclinic crystal. Investigation of the optical properties of the phlogopite revealed that the longer sides of the five-sided spiral are parallel to the *a* axis of the crystal.

As the writer has previously reported several times, the morphology of growth spirals strictly follows the symmetry elements of the face on which the spirals occur, provided that the step height of the spiral layer is small and that the spiral takes on a polygonal form (Sunagawa 1960c, 1961, 1963). In other words, it is possible to decide the symmetry of a crystal from the morphology of growth spirals. From this, it can be concluded that the phlogopite under investigation is a monoclinic polymorph and neither triclinic, orthorhombic nor hexagonal polymorphs are present. It is also noteworthy that the growth spirals take on five-sided form, instead of hexagonal form, in spite of the fact that the single sheet of phlogopite structure has a nearly hexagonal symmetry. This suggests that the morphology of growth spirals is controlled not by the symmetry of a single sheet but by the symmetry of the mica layer or crystal as a whole. Malformation of spiral morphology at the outer part of the spiral can be accounted for in the following way. In Fig. 1, two spirals appear on one surface of a layer, the edges of which are seen to be nearly parallel straight lines. The layer is stepped down from right to left in the photograph. It is expected that re-entrant corners will be formed between the right-side straight step and spiral growth fronts, as well as between the growth fronts starting from two screw dislocation points. Since such re-

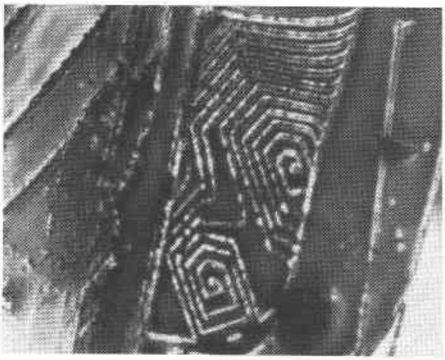

FIG. 1. Phase contrast photomicrograph, ×500.

FIG. 2. Phase contrast photomicrograph, ×415.

entrant corners are the places where atoms or molecules will preferentially be adsorbed, this will result in more rapid advance of growth fronts at the re-entrant corners than along steps. As a result, the morphology of the spiral will be malformed at places where such re-entrant corners exist.

Another point to be discussed here is the high visibility of the spirals. That the steps of the spirals are seen as white lines in the phase contrast photomicrograph shows that there are peaks along the steps of the spiral layers. These peaks are much higher than the step height of the spiral layers, and are considered to be impurity crystals preferentially adsorbed along the steps. They are also seen in Figs. 2 and 3 which are phase contrast and ordinary photomicrographs, respectively. That no white diffraction band is seen at the step of spiral layers on the phase contrast photomicrographs shows that the step height of spiral layers is very small and probably of the order of one unit cell. Because of the existence of impurity crystals along the step, growth spirals having such a small step height are easily seen under the microscope.

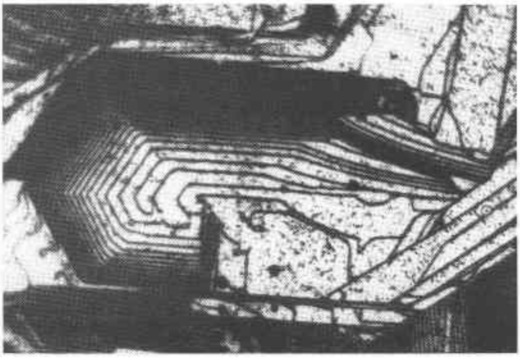

FIG. 3. Ordinary photomicrograph. Impurity crystals appear as dark lines and dots, ×105.

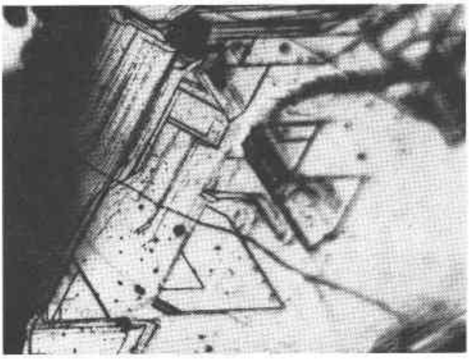

FIG. 4. Ordinary photomicrograph showing five-sided patterns and domain structure, ×105.

In Figs. 2 and 3 are shown more complicated spirals. In these spirals too, five-sided morphology is strictly maintained, at least in the central part. It can be noticed that the spiral growth layers originate from several or many screw dislocation points, which results in a somewhat complicated appearance of these spirals. Deformation from the five-sided form, however, can be accounted for in most cases in terms of preferential growth at re-entrant corners, even in the case of spirals having a hexagonal format such as that shown in Fig. 3. All of the growth spirals observed in this study have five-sided morphology, and spirals having hexagonal or triclinic symmetry have not been observed at all. Interlacing patterns such as those observed on growth spirals on biotite by Amelinckx and Dekeyser (1953) or on SiC by Verma (1951) have not been observed in this study. However, it is observed that the spacings of spiral layers in most cases are very narrow on one side and wide on the other four sides, as seen in the spiral shown in Fig. 2. Spirals such as those in Fig. 1, which have similar spacings on five sides, are exceptional and can be observed only in the case of spirals having a small number of turns. Since the side of close spacings is parallel to the *b* axis and does not show a single sharp cliff but closely spaced steps, it is considered that this phenomenon is the result of the nature of the stacking of the mica layers.

In Fig. 4 an example of five-sided patterns with straight sides is shown. This sort of pattern has been observed on all crystals studied. On the surface of a few of such patterns, growth spirals appear. Five-sided patterns have always the same orientation as those occurring on the surface of the pattern or near it. Equilateral triangular patterns with straight sides are rarely observed, and are definitely a deviation from the five-sided patterns. The straight lines constituting the five-sided patterns are the same as the straight lines widely developed on the surface. It is not certain if these lines are the edges of thick growth layers or the result of slips or faulting during crystal growth. Some of the lines certainly intersect each other, and so must be fault or slip lines, but other lines do not show such intersections. There is always a level difference at the step of a line, which is much bigger than the height of spiral layers. In addition growth spirals always occur either on the surface of five-sided patterns or of layers having straight edges. Therefore it is possible that some of these straight lines are edges of growth layers, which were formed by the bunching of thinner spiral layers.

It is observed on most crystals that the five-sided features (both spirals and patterns) are oriented in several different ways on the surface of one crystal, although they have a definite orientation within one domain. In other words, a crystal consists of many domains which are differently oriented with respect to each other. Figure 4 is an example of such a domain structure, and in Fig. 5 an example of domain structure on a crystal and the orientations of five-sided growth features on each domain are schematically shown. As the writer discussed in his papers on hematite (Sunagawa 1960a, b, c, 1963) and on SiC (Sunagawa 1963), if a crystal is single, the orientation of growth features is the same on the whole surface of a crystal face, whereas in the case of twinned crystals growth features have opposite orientations on different portions or domains. Therefore it is concluded that the domain structures observed on the phlogopites are the result of twinning. Observed rotation angles around the *c′* axis, which is perpendicular to (001), are 60°, 120° and 180°, which correspond to the twinning by rotation around twin axes of [110] (or[$1\bar{1}0$]), [310] (or [$3\bar{1}0$]) and [100], respectively. The composition plane of these twins is (001).

DISCUSSION

From these observations, it can be safely concluded that the phlogopite crystals were grown by a spiral growth mechanism, a mechanism which provides a memory of stacking, which in some substances may at times lead to the formation of different polytypes. Therefore, if growth starts from a screw dislocation having large Burgers' vector, a polytype consisting of many mica layers will be formed. In such a case, an interlacing pattern is expected to appear on the growth spiral, like those observed on a biotite crystal by Amelinckx and Dekeyser (1953). However, no interlaced spiral has been observed on any of the phlogopite crystals investigated, which shows that such polytypes do not occur in these crystals.

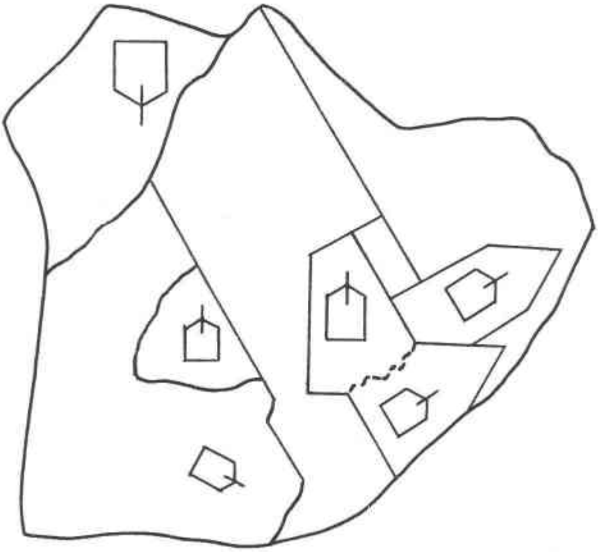

FIG. 5. Schematic figure showing orientations of five-sided growth features and domain structures observed on a crystal.

Smith and Yoder (1956) derived theoretically possible polymorphs from the manner of stacking mica layers in an ordered sequence, and showed six fundamental polymorphs of mica minerals, *i.e.*, 1M, $2M_1$, $2M_2$, 2O, 3T and 6H. Among these six types, only the 1M polymorph has one directional way of stacking, and the other polymorphs have zigzag stacking. Since a single mica layer has monoclinic symmetry, the advancing rate of growth of such layers will be different in a certain direction between one layer and the neighboring layer, in the case of polymorphs having zigzag stackings. As a result, an interlaced spiral is expected to appear in the case of polymorphs of $2M_1$, $2M_2$, 2O, 3T and 6H, but not in the case of the 1M polymorph. Observed growth spirals are not interlaced, and so it is considered that the phlogopite crystals investigated belong to the 1M polymorph, which agrees with the results obtained by x-ray investigations (Tomisaka 1962).

Takano and Takano (1958) discussed the possibility of mistaking x-ray diffraction patterns of twinned crystals of mica minerals as a new polytype. Sadanaga and Takeuchi (1961) theoretically derived possible types of polysynthetic twins of mica minerals (spiral twins with rotation angles $\omega=60°$, $120°$, $180°$; alternating twin with rotation angle $\omega=180°$; and complex twins) and experimentally showed a method of discriminating polysynthetic twins and polytypes. According to them it is possible to distinguish polysynthetic twins from polytypes on x-ray diffraction patterns, even in the case in which the former consists of individuals as small as of the order of a unit cell. They observed spiral twins with rotation angles of $\omega=60°$, $180°$ and alternating twins of $\omega=180°$ on phlogopite crystals from Mutsure-zima. The present study also revealed, through a different way of approach, that most of the phlogopite crystals from Mutsure-zima were twinned and that the observed rotation angles around the c' axis were 60°, 120° and 180°.

Now, here arises a problem whether these twins are of the polysynthetic type as Sadanaga and Takeuchi stated or whether they are twins of the ordinary rotation type. If they are polysynthetic twins consisting of individuals as small as of the order of a unit cell, interlacing patterns are expected to occur on growth patterns, since successive layers will have a different advancing rate in a certain direction. Even in the case in which each individual has a thickness of some hundreds or a thousand Å, which is of the order of the height of thick growth layers, interlacing patterns should be observed on the surface. However, if each individual is much thicker than this value, interlacing patterns will not be expected, since growth layers are not as thick as this. In such a case, however, it will not be appropriate to call such crystals polysynthetic twins, since the thickness of the phlogopite crystal is as small as several microns, and therefore such crystal will consist of only a small number of individuals. The present study showed that no interlacing patterns were observed on the phlogopite crystals, which suggests that the type of twinning observed is not of the polysynthetic but of the ordinary rotation type. The different orientations of the five-sided growth features add further evidence that the twinning is not polysynthetic.

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