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MORPHOLOGY OF CHIOLITE TWINS FROM THE MOREFIELD MINE, AMELIA COUNTY, VIRGINIA

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ABSTRACT

Well-formed twinned crystals of chiolite to 3 mm long have recently been found in replacement bodies in pegmatitic granite at the Morefield mine, Amelia County, Virginia. Crystals are composed primarily of the forms {011} and {017}(?), and are invariably twinned on {011}. The twins range from subequant to elongate, and from relatively simple contact twins to complex penetration twins with hourglass- or S- or L-shaped cross-sections. Untwinned crystals have not been seen.

Keywords: chiolite, twin, growth dynamics, Morefield mine, Virginia.

SOMMAIRE

Nous décrivons des cristaux maclés de chiolite atteignant une longueur de 3 mm, récemment découverts dans les zones de remplacement d'une pegmatite granitique à la mine Morefield, comté d'Amelia, en Virginie. Les cristaux montrent surtout les formes {011} et {017}(?), et sont invariablement maclés sur {011}. Les macles sont à peu près équidimensionnelles ou bien allongées, et les individus sont maclés de façon relativement simple (macle de contact) ou en inter-pénétration complexe, avec une section en forme de sablier, en S ou en L. Nous n'avons pas trouvé de cristaux non maclés.

(Traduit par la Rédaction)

Mots-clés: chiolite, macle, cinétique de croissance, mine Morefield, Virginie.

INTRODUCTION

The Morefield pegmatite in the Amelia district of Virginia, discovered by Silas V. Morefield in 1929, has had a long and varied history of mining activity (*e.g.*, Kearns 1993, Sweet & Penick 1986, Brown 1962); it produced commercial quantities of mica and feldspar, and, in addition, gem materials (amazonitic microcline, topaz, spessartine, moonstone), members of the columbite – tantalite series, and rare-earth minerals such as microlite and monazite (Kearns 1993). In 1992, several small pods of aluminum fluoride replacement-type mineralization were encountered, one of which produced cryolite, prosopite, and well-formed crystals of chiolite, elpasolite, thomsenolite, pachnolite, and ralstonite. An account of the mineralogy of these replacement bodies is given in Kearns (1995). The chiolite crystals represent the first North American occurrence of this rare mineral; they are certainly among the largest and best ever found anywhere, and are invariably twinned, generally in a complex manner. The purpose of this paper is to describe these twinned crystals.

MINERALOGY AND CRYSTALLOGRAPHY

Chiolite is an aluminofluoride of sodium, $Na_5Al_3F_{14}$ (Clausen 1936). It is tetragonal, space group *P4/mnc*. A unit-cell refinement using the average *d*-values of four independent patterns of the Morefield material, and assuming *P4/mnc*, gave an *a* of 7.030(3) and a *c* of

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10.392(3) Å. These values differ only slightly from the published values, *a* 7.0138(8) and *c* 10.402(2) Å (Jacoboni *et al.* 1981).

The chiolite crystals are usually bipyramidal {011} (Palache *et al.* 1951), but distinct crystals are rare and very small at the other reported localities for chiolite. Untwinned crystals and twinned crystals that reflect undistorted tetragonal morphology have not been found at the Morefield mine.

MATERIALS

Whereas several aluminum fluoride replacement pods have been encountered at the Morefield mine, only one has produced euhedral crystals of chiolite. This 15 × 28 cm pocket, encountered in August 1992, produced several hundred well-formed, euhedral twinned crystals as isolated individuals and open-textured groups of crystals, associated with the other minerals mentioned above. These twinned crystals include 1) uncommon subequant pyramidal crystals (Figs. 1A–C), similar to twins from Miask in the Ilmen Mountains of Russia (Goldschmidt 1913, Pl. 200, Fig. 5), 2) elongate crystals with large curved faces (Figs. 1D–F, 2A), and 3) complex elongate crystals with hourglass- or S- or L-shaped cross-sections (Figs. 1G–I, 2B–D). These more complex twins range from contact twins with asymmetrical extensions through symmetrical penetration twins to asymmetrical penetration twins in which one individual is dominant over the other. Some elongate crystals show a different habit at each end.

Habit 2 is the most common, and is generally a constituent of the most complex crystals of habit 3. About 50 selected crystals and crystal groups were available for morphological study.

METHODS

The suite of crystals available was examined with a binocular microscope to determine the habit and evalu-

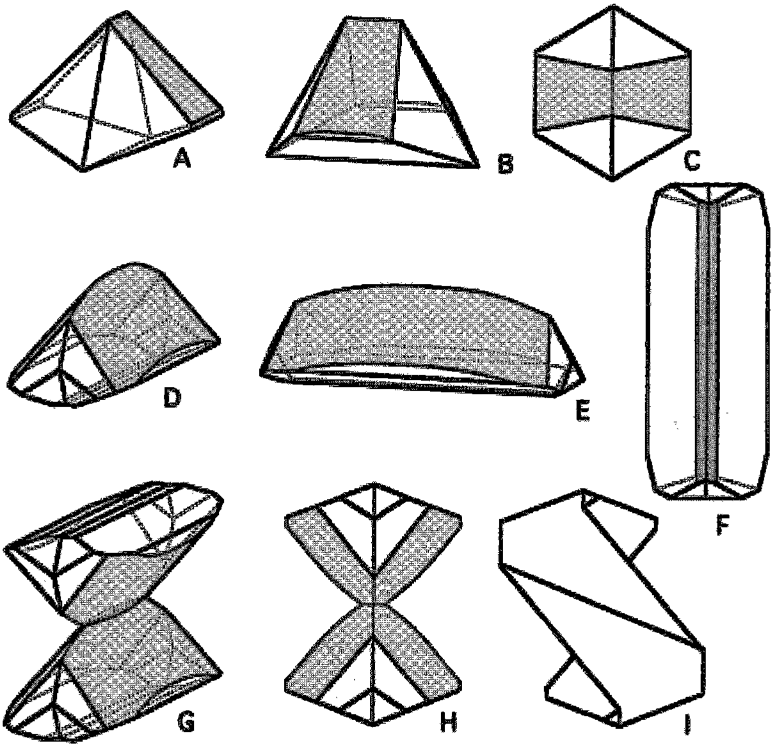

FIG. 1. Habits of chiolite twins. A–C. End, side, and top view of equant twin involving the least observed distortion relative to a hypothetical untwinned crystal. D–F. End, side, and bottom view of typical contact twins with elongation parallel to the *a* axis, which lies in the twin plane, and rounded lateral faces approximated by {017}. G–H. "Hourglass" habit produced by balanced development of penetration morphology. I. S-shaped habit produced by unbalanced development of penetration morphology. Front faces of crystals are shaded to indicate the forms to which they belong: {011} unshaded, {017} lightly shaded, {0 5 11} heavily shaded.

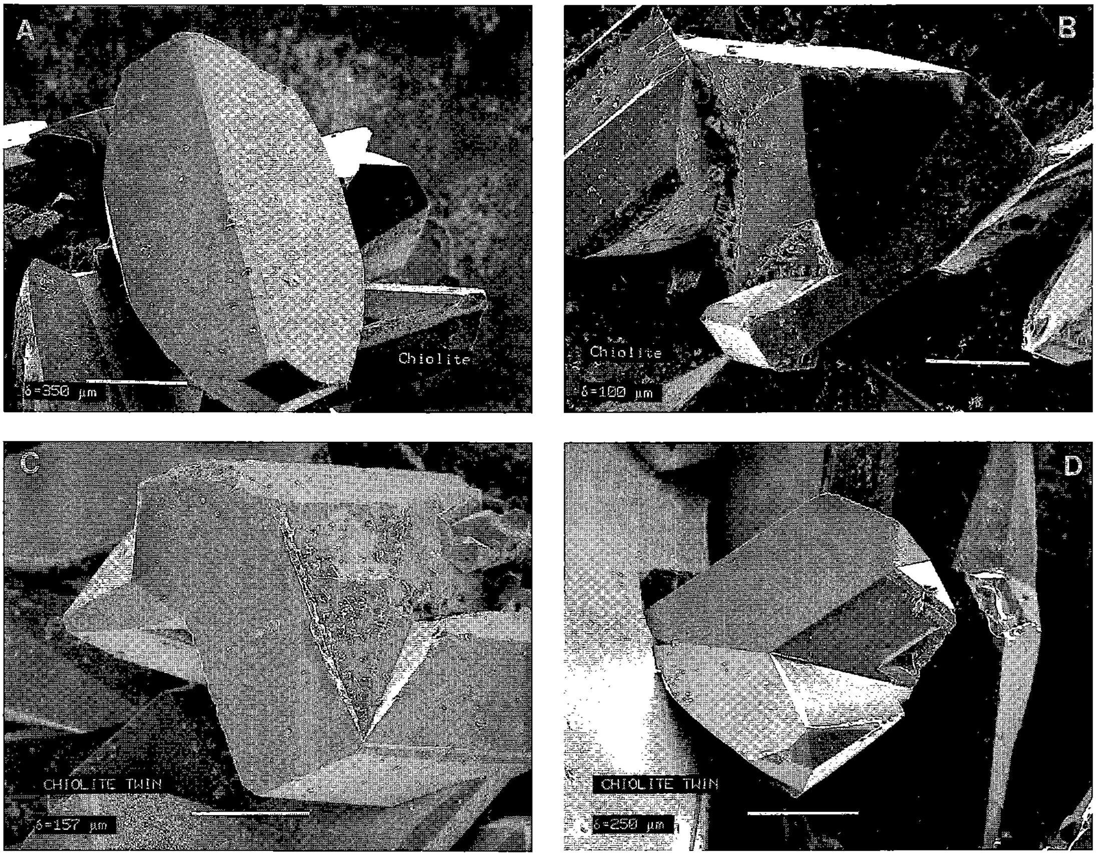

FIG. 2. Scanning electron microscopy (SEM) images of representative twins in chiolite. A. Typical contact twin, viewed onto bottom faces as drawn in Figure 1F. B. Contact twin with prominent flange projecting from one individual. C. S-shaped penetration habit. D. L-shaped habit involving twinning on at least three non-parallel {011} planes. Bar scales: A 350 μm, B 100 μm, C 157 μm, D 250 μm.

ate variations in the habit from crystal to crystal. Observation of twins of types 1 and 2 indicated an overall twin symmetry *mm*2 (Fig. 3). The twin law was determined by measurements of extinction directions in thin sections cut parallel to plane B of Figure 3, and confirmed by the X-ray method described below.

Representative twinned crystals were measured using a Stoe two-circle optical goniometer, and the measured faces were indexed as if the measurements were of an untwinned isometric crystal. The resulting indices were entered into the crystal drawing program SHAPE (Dowty 1980), which was used to rotate a stereographic projection of the faces to bring the twin plane into coincidence with the x–z cartesian plane. Hypothetical crystals of chiolite were drawn using low-index forms, and their stereographic projections were compared with those for the measured crystals, in order to determine what forms are present on the actual crystals. Indices for a number of faces on selected crystals were confirmed by X-ray examination.

Crystals chosen for X-ray study were mounted with wax on a sample spinner in a Norelco X-ray diffractometer. The configuration is usually used (without the wax) for diffraction studies of powder mounts. When used with a single crystal, it yields only peaks corresponding to the d-value for the face of interest and multiples of this spacing. Data from these peaks were refined using an extrapolation against $\sin^2(\theta)$ to provide a more accurate determination of the d-value than that obtained from any single measurement (Cook 1963). The resulting d-value was compared with standard X-ray-diffraction data to identify each face studied.

RESULTS

Crystal morphology

Observations of numerous crystals showed that, in spite of a wide range of habit, they are all composed of three kinds of faces. One set of faces is flat and highly

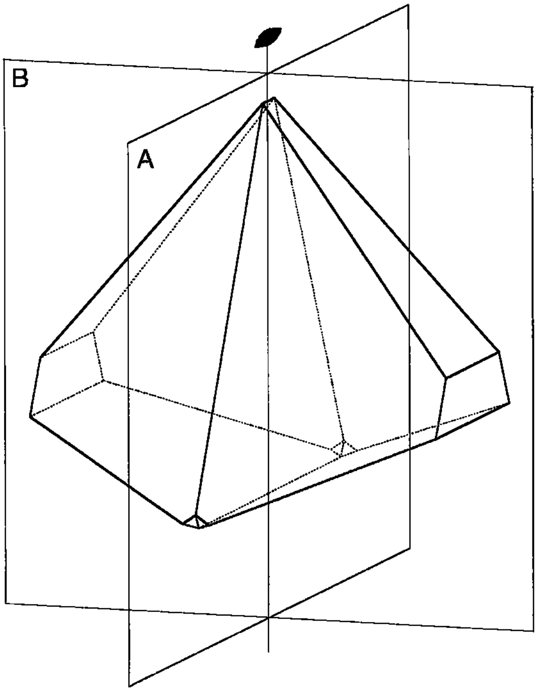

FIG. 3. Conventions for orientation used in this paper. Plane A is the twin plane. Plane B is a mirror plane common to the symmetry of each twinned individual, and thus also a mirror plane of the twin. It also contains the *c* axis and one *a* axis of each twinned individual.

reflective, with rare growth-steps on some faces, and gives excellent signals in the goniometer. Multiple or clouded signals are seen in some cases, indicating a mosaic texture with angles of misfit less than 0°20' between blocks. A second set of faces are convex to irregular, with limited flat areas of relatively low luster; signals are either non-existent or smeared in one direction over 5° or more, indicating reflection from a unidirectionally curved portion of the face. The third set occurs only as a narrow, parallel-edged band adjacent to the twin plane (*e.g.*, Figs. 1D, 1F, and 2A) on the bottom of the crystal. This band is generally located mostly or entirely on one crystal, but may span both individuals with only slight deflection across the twin plane, demonstrating that it is nearly perpendicular to the twin plane. It is commonly slightly depressed into the crystal, and bounded by a narrow face on each crystal with indices near $(01\bar{3})$.

Goniometric measurements and morphological modeling using SHAPE demonstrated that the flat faces all belong to {011}. The curved faces are bounded approximately by {013} and {001} in the zone parallel to [100], and by faces in the vicinity of {116} along their length. Many of them are not simple convex forms, but have hillocks and dimples and other irregular features on the surface. Given the poor quality and curved nature of these faces, and the inherent difficulty of measuring such faces by any technique, the indices assigned to them must be considered approximate.

X-ray determination of *d*-values for five flat faces and one curved face on one crystal, and three flat faces and one curved face on a second crystal, confirmed that all of the flat faces belong to {011}, and indicated that both curved faces are {013}. Measurements of *d* on the first crystal were replicated at a later date.

Faces in the narrow band adjacent to the twin plane could not be measured on the goniometer. However, if they are taken to be perpendicular to the twin plane, their indices can be calculated, and are approximately {0 5 11}.

Twinning

Parallel extinction was observed in thin sections oriented perpendicular to the two-fold axis of the twinned crystals (horizontal sections in the orientation of Fig. 3). One twinned crystal serendipitously shifted during mounting, to an orientation subparallel to the bottom face (in the orientation of Fig. 3) of one individual. The section of the other individual of this twin showed first-order gray birefringence color, and gave a slightly off-center uniaxial cross (with the Bertrand lens), indicating that the *c* axes of the twinned individuals were steeply inclined to the two-fold axis of the twin.

Measurements were made of the angle between extinction directions corresponding to the *c* axes of twinned individuals, in thin sections oriented parallel to plane B of Figure 3. Measurements on nine crystals yielded a mean angle of 111.78° and a median of 111.6°, with a standard deviation of 1.02°. Given the orientation of these sections, this angle is twice the ρ angle of the twin plane. The overall symmetry of the twin dictates that the twin law must be of the form (0*kl*) or (*hhl*). Using the unit-cell parameters listed above and standard crystallographic formulas (Palache *et al.* 1944), the Miller indices of the twin plane are calculated to be either (0.0999 0.0999 1) or (0.709 0.709 1). Since the smallest integral indices corresponding to the second alternative are {557}, and since {011} twinning is known for chiolite from other localities, the twin plane was taken to be {011}. This assumption was verified by X-ray measurements of *d*-values for a pair of faces, one on each half of the twin and both parallel to the twin plane.

DISCUSSION

The lack of etch pits indicates that all faces formed by growth, not dissolution. The characteristics of the faces of the two forms present on these crystals suggests that the crystals grew by layered growth on {011}, but that the lateral propagation of the layers was impeded by impurities, leading to the development of

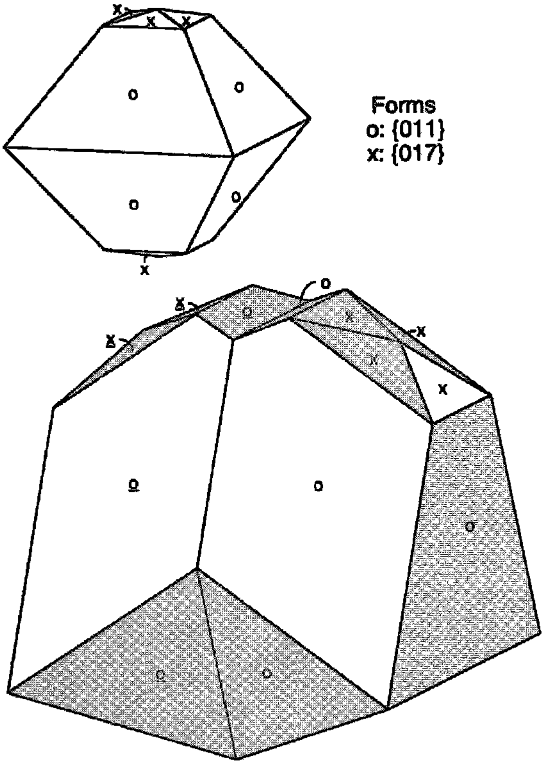

FIG. 4. Idealized chiolite with {011} and {017}, untwinned, and twinned on (011) with no distortions due to twinning, for comparison with observed twin-habits. To produce the habit shown in Figures 1A–C, the dark-shaded faces must be eliminated, and the light-shaded face must be greatly reduced in size.

the irregular curved faces of the second form. The elongation of typical twins indicates that the re-entrant notches were important sites of nucleation for new layers of growth.

The form {017}, intermediate in orientation between {013} and {001}, was chosen for drawing the observed morphology of these crystals. This form deviates from {013} by about 14° and from {011} by 12°, and was also tentatively identified on crystals from Miask by Koksharov (1862), as quoted in Palache *et al.* 1951).

The occurrence on many crystals of the narrow band of faces of {0 5 11}, always in the same position, and the consistent absence of other faces of this form, indicates that this band is a direct consequence of the growth dynamics adjacent to the twin plane, but no further explanation for it can be given.

The universal presence of twinning among these crystals deserves notice. Hartman (1955) explained the larger size of twinned crystals in many assemblages as a consequence of more frequent nucleation of growth layers in the re-entrant zone along the twin plane than elsewhere on the twin. Growth of isometric crystals as elongate whiskers (*e.g.*, pyrite and cuprite, var. chalcotrichite) through the influence of screw defects has a similar explanation. Apparently the presence of twinning in chiolite in this assemblage conferred a sufficient advantage to make the difference between growth and non-growth; the growth environment was apparently supersaturated with respect to twinned chiolite, but undersaturated with respect to untwinned chiolite.

The most striking aspect of these twinned crystals is the limited degree to which their morphology reflects the tetragonal structure of the individual crystals. A hypothetical crystal of chiolite bearing the forms seen on the actual twins is shown as a single crystal and an undistorted twin in Figure 4. The undistorted twin most closely resembles the actual twin shown in Figures 1A–C, a habit that is quite rare at the Morefield mine. To convert the hypothetical crystal to this nearest actual equivalent, the darkly shaded faces belonging to {011} and {017} must be eliminated, the lighter shaded face and its equivalent on the back side of the crystal must be drastically reduced in size, and the remaining {017} faces must be curved.

To develop a form like the more typical crystal shown in Figures 1D–F, the re-entrant faces of {011} in Figure 4 must be retained. However, their growth rates, and those of the {011} faces above them, must be 2 to 2.5 times as great as those of the small side faces of {011} that are parallel to the twin plane. In addition, the curvature of the curved {017} faces must be exaggerated.

Crystals like those in Figures 2B and 5A–B develop where one or both individuals "spill across" the twin plane (at different points along the contact if spillover goes in both directions). Once cross-over occurs, there is a new {011} growth face on a finger projecting into a region otherwise occupied only by a resultant face of the other individual. This may give the invading "finger" a competitive advantage, leading to its further growth. Crystals like those in Figures 5C–D can be understood as interpenetration equivalents of those in Figures 1D–F. Thin sections of crystals like those in Figure 5C document this cross-over phenomenon, with crossing in both directions present at different depths in the same section. Diagonally opposite sectors are crystallographically continuous.

This growth model is based on the idea of geometric competition (Grigor'ev 1961) applied to twinned aggregates. Similar interpretations have been advanced for other minerals (Senechal 1976, Sheftal' 1971, 1972, 1980, Donnelly 1967, Richards 1996). If the habit resulting from simple contact twinning produces a region in which the crystal is inefficient at accepting growth materials, then modifications of the crystal's configuration that increase the local ability of the crystal to grow may be favored. The upper surface of typical simple twins, composed of two large {017} faces, is such an area. Crossing or penetration removes this barrier to growth.

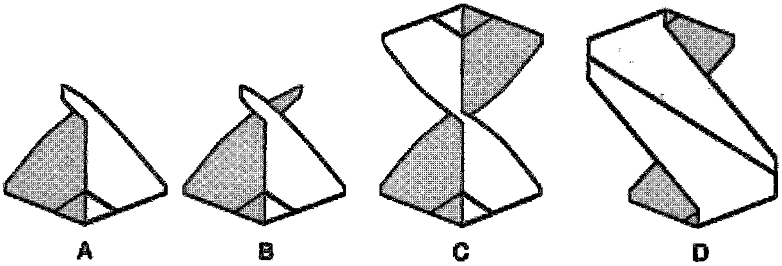

FIG. 5. Schematic representations of twinning of other than simple contact type, shaded to show optical properties. This sequence, taken from left to right, also represents a sequence of morphological development that unifies all of the observed habits with single composition planes. Sector development near the "waist" of the hourglass form is schematic, and varies from twin to twin, and from section to section within twins.

Some L-shaped aggregates result from twinning on two adjacent {011} twin planes, which form an angle of about 70° with each other. In these twins, one central crystal is simultaneously twinned with two other individuals, which bear no special crystallographic orientation to each other.

The L-shaped aggregate shown in Figure 2D is even more complex. Each leg is itself a contact twin. The crystallographic relationship between the two contact twins has not been established, but it most likely involves twinning on another {011} plane. Probably only two of the four individuals in the aggregate have a rational crystallographic relationship to each other across the angle; the other two have a non-rational relationship determined by the interaction of the relationships among the twinned members.

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