CRYSTALLOGRAPHY OF SPANGOLITE*

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ABSTRACT

Spangolite is shown by x-ray Weissenberg and morphological study to be ditrigonal-pyramidal (3 m) in crystallization. Unit cell dimensions: a₀ = 8.245 Å, c₀ = 14.34 Å; a₀:c₀ = 1:1.739. Cell contents: Cu₉Al₂(SO₄)₂(OH)₄Cl·6H₂O. Transformation, morphological cell of Penfield (1890) to the structure cell, $\frac{1}{3}$[000]/$\frac{1}{3}$[000]/i/000; a:c = 1:1.741 (Tombstone; Penfield, 1890). Space group C3c. Crystals of a hemimorphic habit from Bisbee, Arizona, and from a new locality at Majuba Hill, Pershing County, Nevada, are described. The indices of refraction of crystals from different localities vary measureably, with O ranging from 1.680 to 1.687; the variation probably is due to substitution of OH for Cl.

MORPHOLOGY

The rare and interesting mineral spangolite was first described in 1890 by Penfield, who established the formula as Cu₉Al₂(SO₄)₂(OH)₄Cl·3H₂O and on the basis of the evidence then available assigned the mineral to the hexagonal-scalenohedral (calcite) crystal class, $\overline{3}$ 2/m. The locality was given only as within a radius of 200 miles of Tombstone, Arizona, and has not since been more clearly established. Penfield's material comprised

doubly terminated hexagonal tablets flattened on {0001} with large faces of a hexagonal prism and a series of inclined faces in a zone therewith. Etch figures produced by very dilute acids on {0001} exhibited rhombohedral symmetry, and the inclined faces, which seemingly conformed to six-fold symmetry about [0001], were set as pyramids together with the prism in the second order position. The crystals were specifically stated

* Contribution from the Department of Mineralogy and Petrography, Harvard University, No. 302.
to show no evidence of lower than hexagonal holohedral symmetry (Fig. 1). The spangolite crystals described by Pelloux from Arenas, Sardinia, also are hexagonal holohedral in appearance. Miers, however, described an occurrence of the mineral in Cornwall and proved by means of pyro-electric and etching tests that the [0001] axis was polar. Some of his crystals had a hemimorphic appearance, but the curved and tapering nature of the faces precluded goniometric measurements. Ford later mentioned but did not describe crystals of a hemimorphic habit from the Grand Central mine, Tintic, Utah.

A few months ago, Mr. Hatfield Goudey of Yerington, Nevada, found a new occurrence of spangolite at Majuba Hill, Pershing County, Nevada, and kindly offered specimens for study. The mineral forms thin crusts of minute crystals associated with azurite, chalcophyllite and cyanotrichite on altered rhyolite. The spangolite crystals are sharply developed and have a simple and strikingly hemimorphic habit. Examination under a high power binocular microscope showed that the crystals were terminated at one end—often the point of attachment to the matrix—by a basal pinacoid, with a hexagonal prism and a seeming hexagonal pyramid which tapered out to a sharp point or was terminated by a small basal pinacoid. A few crystals about 0.003 inch in size were mounted on a reflecting goniometer by picking them up under a microscope on the point of a needle that had been thinly coated with a tacky wax. Visible although inferior reflections were obtained which served to identify the forms listed in Table 2; the forms are here given in the new orientation required by the x-ray study described beyond. The habit is shown in Fig. 2, the acute termination being arbitrarily taken as the antilogous pole. Crystals of a different habit were found on another specimen from this locality. These comprised minute hexagonal prisms attached to the matrix by one end of the c-axis and terminated at the other end of this axis by a slightly concave basal pedion. A few of these crystals were noted to have a rounded three-sided cross section, resembling that of tourmaline, but the roughness of the prism faces prevented goniometric measurement.

Material better suited for goniometric study was obtained on a specimen of spangolite from the Czar mine, Bisbee, Arizona, purchased for the Harvard collection in 1941. These crystals were thin tabular on {0001} and are doubly terminated with a hemimorphic habit as shown in Fig. 3. The trigonal pyramids {1011} and {0111} [new orientation] were consistently observed on only one pole of [0001], here taken as the antilogous pole. The other forms occasionally occurred at both poles but usually were present only at the analogous pole. {0001} and {0001} always were different in size, with {0001} largest. Adjacent inclined faces on both the top and bottom poles of the crystals are commonly equal in size, and
when they are not equal the distortion is of a random rather than of a three-fold nature. The inclined faces and the prism faces ordinarily are deeply striated and grooved horizontally, but no correlation between these markings could be distinguished on either alternate or adjacent faces. The inclined faces on the antilogous termination of the crystals ordinarily are more deeply striated. These remarks apply also to the crystals from Majuba Hill and to the crystals from Tintic mentioned beyond. Morphologically, there is no evidence of trigonal symmetry in the arrangement of the inclined faces on spangolite although x-ray study proves that the seeming pyramids actually are sets of positive and negative trigonal pyramids. The \{10\overline{1}3\} and \{01\overline{1}3\} faces of the crystals are relatively smooth and large and afforded a number of measures of fair quality. The average of these gave \( \rho = 33^\circ 44' \), which is in satisfactory agreement with the value \( 33^\circ 50' \) calculated from the morphological ratio of Penfield. His ratio was based on the angle \( \rho = 63^\circ 33' \) obtained from faces of \{10\overline{1}1\} and \{01\overline{1}1\} of superior quality on an otherwise deeply striated crystal. The specific gravity of the Bisbee material was determined on the microbalance as \( 3.135 \pm 0.01 \).

Crystals from the Tintic locality mentioned by Ford also were available for study. These were dull in luster and strongly striated but measurements of sufficient accuracy to identify the forms present (Table 2) were obtained. The crystals have a decided hemimorphic appearance, but this is due to an oscillatory combination of \{10\overline{1}0\} and \{01\overline{1}0\} with \{10\overline{1}0\} at the antilogous pole of the crystals only, causing the crystals to taper at this pole in an irregular fashion, and is not due to asymmetry in the kind of faces developed at the top and bottom poles. The crystals from Cornwall described by Miers probably also were of this nature.

**X-Ray Crystallography**

Tiny doubly terminated crystals from the Tintic and the Bisbee localities were examined by the rotation and the Weissenberg methods. Relatively long exposures were taken in copper radiation and photographs of excellent quality resulted. The rotation period about the perpendicular to the morphological \{11\overline{2}0\} in Penfield's orientation proved to be \( \sqrt{3} \) times larger than that about the horizontal axis at \( 30^\circ \) thereto. This finding identifies the principal prism as of the first order, and the supposed pyramids become sets of trigonal pyramids in equal development. 0- and \( n \)-layer Weissenberg photographs about the \( c \)-axis showed the plane symmetries \( C_{6h} \) and \( C_{3h} \) and proved the lattice type to be hexagonal. 0- and \( n \)-layer photographs about the morphological \( a \)-axis (in the new orientation) showed only two-fold axes of symmetry, \( C_2 \). The 0-layer about the axis perpendicular thereto revealed a two-fold axis and plane of symmetry, \( C_{2h} \), and the 1-layer showed a plane of symmetry only, \( C_1 \).
These layer symmetries identify the centrosymmetrical point group of the crystal as $3 \overline{m}$, which includes the crystal classes $3 \overline{m}$, $3 2$ and $3 \overline{2}/m$. The polar nature of [0001] as proven by morphological and physical evidence establishes the crystal class as ditrigonal-pyramidal, $3 \overline{m}$ (tourmaline class). The space group is $C_{3v}^3 = C3c$. Spangolite was earlier placed in the tourmaline crystal class by Ford in the 4th edition of Dana’s *Textbook of Mineralogy* but without discussion of the basis for this classification. The unit cell dimensions (new wavelengths) and ratio obtained by calculation from the appropriate 0-layer Weissenberg films are:

$$a_0 = 8.245 \pm 0.01 \text{ Å}, \quad c_0 = 14.34 \pm 0.01, \quad a_0:c_0 = 1:1.739,$$

An angle table in the new orientation using the elements obtained by Penfield on the Tombstone material is given in Table 1. Penfield’s elements are very close to the x-ray values. A list of the forms observed on crystals from the known localities is given in Table 2. The transformation from the original morphological orientation to the structure cell is $\frac{1}{3}[000] / \frac{1}{3}[000] / i/000 \frac{1}{3}$. The x-ray powder spacing data obtained in filtered copper crystals are given in Table 3.

<table>
<thead>
<tr>
<th>Lower</th>
<th>Upper</th>
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<th>$\phi$</th>
<th>$\rho = C$</th>
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<th>$A_2$</th>
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<td>a 1120</td>
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<td>61 101</td>
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CRYSTALLOGRAPHY OF SPANGOLITE

Table 2. Forms Observed on Spangolite from Different Localities

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<tr>
<th>Forms</th>
<th>Tombstone†</th>
<th>Majuba Hill</th>
<th>Bisbee</th>
<th>Tintic</th>
<th>Sardinia</th>
<th>Cornwall</th>
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† Also the following trigonal pyramids with their corresponding negative upper and positive and negative lower geometrical equivalents: 1014, 3034, 3032, 2021, 3031.

Table 3. X-Ray Powder Diffraction Data for Spangolite from Tintic, Utah

Copper radiation, nickel filter

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<tr>
<th>(d)</th>
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<th>(d)</th>
<th>(I)</th>
<th>(d)</th>
<th>(I)</th>
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Chemistry and Indices of Refraction

The only chemical analyses reported of spangolite are those of Penfield on the original material from the region of Tombstone, Arizona. Table 4 gives the unit cell contents calculated from the average of these
analyses using the cell dimensions cited and the specific gravity, 3.141, obtained by Penfield on the analyzed sample and here confirmed on material from Bisbee. The simplest formula for the mineral is seen to be

$$\text{Cu}_2\text{Al}(<\text{SO}_4)\text{(OH)}_2\text{Cl} \cdot 3\text{H}_2\text{O},$$

which is identical with that originally derived by Penfield. There are two of these formula units in the unit cell.

**Table 4. Unit Cell Contents of Spangolite**

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<th></th>
<th>1</th>
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<th>3</th>
<th>4</th>
<th>5</th>
<th>6</th>
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<td>59.51</td>
<td>7479</td>
<td>(Cu)</td>
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1. Average of four analyses, three partial, by Penfield (1890) on material from the region of Tombstone, Arizona. Measured G = 3.141.
2. Theoretical weight percentages for Cu₂Al(<SO₄)OH₂Cl·3H₂O.
3. Molecular quotients.
5. Calculated unit cell contents. Molecular weight = 1597 (meas.).

The calculated atomic contents of the unit cell correspond very closely to whole numbers with the exception of Cl. The value for Cl is slightly low and presumably this element is substituted for in part by OH. A small but significant variation is found in the indices of refraction and

<table>
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<th>Locality</th>
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<tr>
<td>New Mexico</td>
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<tr>
<td>Majuba Hill, Ariz.</td>
<td>1.681 ± 0.002</td>
<td>1.627 ± 0.002</td>
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<tr>
<td>Sardinia</td>
<td>1.682</td>
<td></td>
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<tr>
<td>Sardinia</td>
<td>1.683 ± 0.002</td>
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</tr>
<tr>
<td>Bisbee, Ariz.</td>
<td>1.685 ± 0.002</td>
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</tr>
<tr>
<td>Tintic, Utah</td>
<td>1.686 ± 0.002</td>
<td>1.638 ± 0.002</td>
</tr>
<tr>
<td>Tombstone, Ariz.</td>
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<td></td>
</tr>
<tr>
<td>Tombstone, Ariz.</td>
<td>1.694</td>
<td>1.641</td>
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* Private communication, Miss Jewell Glass, U. S. Geol. Survey.
* Private communication, Dr. Waldemar T. Schaller, U. S. Geol. Survey.
* Penfield (1890).
birefringence of spangolite from different localities, as tabulated below. This variation doubtless is due to variation in the Cl:OH ratio or to some other type of compositional variation in the mineral. It may be noted that at least the index of refraction for the ordinary ray given by Penfield for the Tombstone material is in error according to the measurements of Miss Glass on the type material.

**SYSTEMATIC RELATIONS**

Spangolite does not appear to have any close relatives either crystallographically or chemically. A number of other hydrated basic sulfates of divalent and trivalent cations are known and are listed below. A comparison of the properties of these minerals with those of spangolite, including a comparison of the x-ray powder photographs, did not reveal any obvious relationships. The powder pattern of chalcophyllite, however, resembles that of spangolite in part, due to a near identity in position of intense reflections from 0001. The cell dimensions and cell contents of spangolite and chalcophyllite* are compared below.

<table>
<thead>
<tr>
<th>Cell contents</th>
<th>Space group</th>
<th>(a_0)</th>
<th>(c_0)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Spangolite (\text{Cu}_6\text{Al}_5(\text{SO}_4)_2(\text{OH})_8\cdot6\text{H}_2\text{O})</td>
<td>C3c</td>
<td>8.245</td>
<td>14.34</td>
</tr>
<tr>
<td>Chalcophyllite (\text{Cu}_6\text{Al}_5(\text{SO}_4)_2(\text{OH})_8(\text{AsO}_4)_3\cdot33\text{H}_2\text{O})</td>
<td>R 3 m (?)</td>
<td>10.75</td>
<td>57.40</td>
</tr>
</tbody>
</table>

* Data of L. G. Berry, private communication, 1948.

**ACKNOWLEDGMENT**

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**REFERENCES**


