THE CRYSTAL STRUCTURE OF A HEXAGONAL Al-SERPENTINE

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Abstract

A few porcelain-white, platy, hexagonal crystals, measuring up to 2 mm in diameter, were found in the vugs of a Keweenawan rhyolite pebble on the north shore of Lake Superior. The mineral's chemical formula of $Mg_{2.0}Al_{0.6}(Fe, Ca, K)_{0.1}(Si_{1.4}Al_{0.6})O_{4.6}(OH)_{4.4}$ was established by electron microprobe analysis and the hexagonal unit cell dimensions of $a = 5.295 \pm 0.004$ and $c = 63.99 \pm 0.06$ Å were obtained from single crystal and refined from powder diffraction data. Three-dimensional intensities were collected with a precession camera and an equi-inclination, single crystal diffractometer. Several models, consistent with the observed $\bar{3}m$ diffraction symmetry of the crystal and with $\bar{3}$ were tested and the one yielding acceptable intensities was refined with a least-squares technique. A weighting scheme to approximate the effects of stacking disorder and to account for the quality of reflections was applied and the final R factor of 10.2 percent was obtained. The substructure has a symmetry of $P3_1$ (or $P3_2$) and is composed of nine serpentine-like layers per unit cell. A superstructure of 18-layers or polysynthetic twinning is assumed to be responsible for the higher observed diffraction symmetry.

Introduction

A few years ago Mrs. Dorothy Hays brought to the authors a pebble she collected on the North Shore beaches of Lake Superior in Minnesota. The pebble was a specimen of the Keweenawan rhyolite and contained several vugs filled with quartz and with hexagonal, platy crystals of a porcelain-white mineral, resembling kaolinite and other clay minerals. Because the crystals were exceptionally well developed and appeared to be truly hexagonal their crystallographic investigation seemed to offer unusual potential.

A photomicrograph of the four largest crystals is shown in Figure 1.

The routine mineralogical investigation of these crystals gave a hardness of 2.5, a density of 2.54 gm/cm³ and indices of refraction between 1.560 and 1.566. Only one grain was found to display a satisfactory interference figure which showed an optic angle of 2–3 degrees. This deviation from hexagonal symmetry, however, could be attributed to stacking disorders practically always present in sheet silicates. The X-ray powder pattern of the mineral was identified by S. W. Bailey of the University of Wisconsin as that of an Al-serpentine, similar to the one he described earlier (Bailey and Tyler, 1960).

A few grains of the mineral were analyzed with the MAC 400 electron

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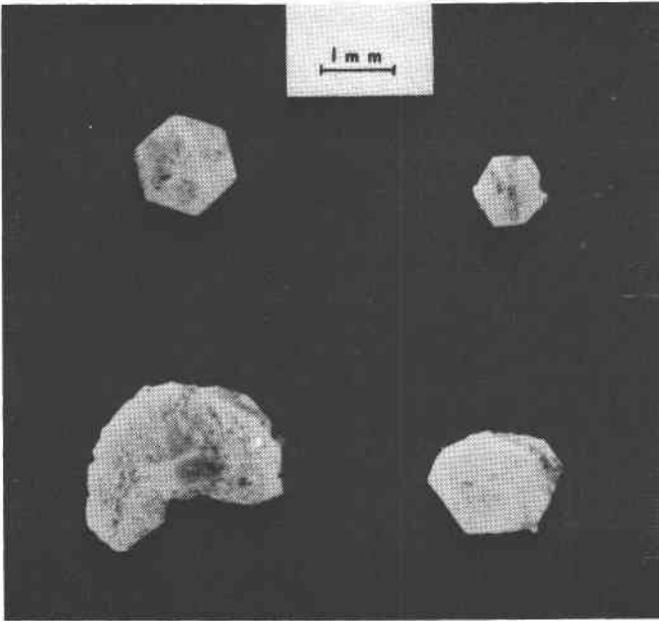
Fig. 1. Photomicrographs of Al-serpentine crystals.

microprobe of the University of Minnesota to establish a chemical formula. Analyzed samples of serpentine, kaolinite and phlogopite, from the former Rock Analysis Laboratory of the University of Minnesota, were used for standards. A reproducibility of 95 percent was obtained in the analysis. However, no reasonable values of error can be given due to the lack of stated error in the standards. The following results were obtained.

	Weight/percent
SiO_2	31.1
MgO	29.3
Al_2O_3	22.2
FeO	1.9
CaO	0.3
K_2O	1.0
Total	85.8

Due to shortage of material no (OH) determination could be undertaken, and the above composition could yield two possible formulae by assuming full tetrahedral occupation. If the remaining 14.2 weight percent in the analysis is assumed to be water the formula is:

$$(Mg_{2.02}Al_{0.64}Fe_{0.07}Ca_{0.01}K_{0.06})(Si_{1.43}Al_{0.57})O_{4.62}(OH)_{4.38} \qquad (1)$$

while if the ideal O:OH ratio is maintained and the method given by Faust and Fahey (1962) is applied the formula becomes:

$$(Mg_{2.07}Al_{0.72}Fe_{0.07}Ca_{0.01}K_{0.06})(Si_{1.47}Al_{0.53})O_5(OH)_4 \qquad (2)$$
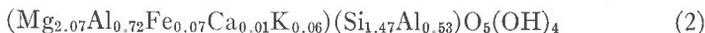
The former formula is more hydrous than the latter and a few grains of the mineral were sacrificed for differential thermal and infrared ab-
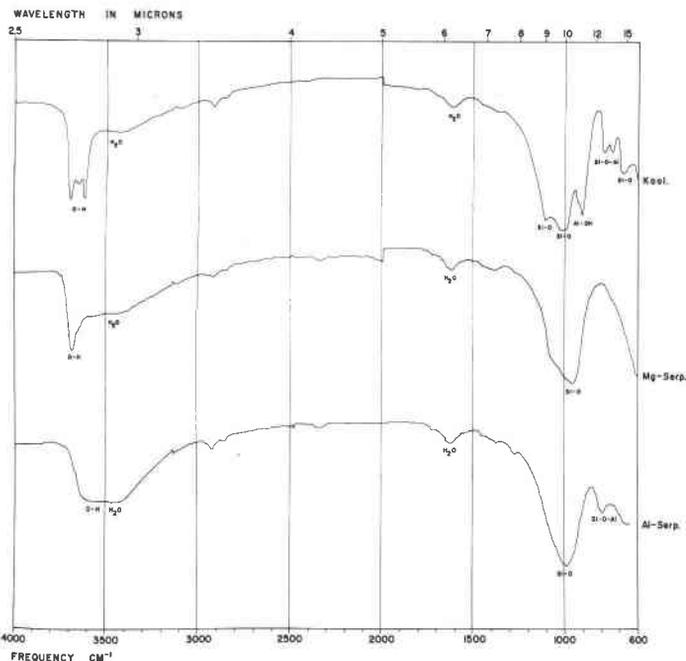
FIG. 2. Infrared spectra of a kaolinite, an Mg-serpentine (from former Rock Analysis Laboratory of the University of Minnesota) and the Al-serpentine of the present study at 22°C.

sorption analyses to obtain an indication of perference between the two formulae. Both analyses showed the presence of excessive water, and are supporting the more hydrous formula.

Figure 2 shows the infrared spectra of kaolinite, Mg- and Al-serpentines. The very broad and strong absorption band observed in Al-serpentine in the range of 3700–3200 cm⁻¹ may be interpreted as being due to the combination of free hydroxyl ions (3700–3500 cm⁻¹) and hydrogen-bonded water molecules (3550–3200 cm⁻¹). The band at 1620 cm⁻¹

represents the bending mode of the water in the structure. The magnitude of these bands and the fact that they persist at temperatures to over 420°C suggest that there is a considerable amount of strongly bonded water.

Both formulae would require, however, that some of the cation positions in the octahedral layers be vacant. Vacant sites would amount to 6.7 percent in the first and 2.3 percent in the second formula.

Several zero and higher level precession photographs were taken of over a half dozen apparently single crystals of this Al-serpentine. All photographs displayed the diffraction symmetry of $\bar{3}m$ and showed effects of mosaic structure and stacking faults. Some regularity in the pattern of stacking faults could be established from the diffusion of the $k \neq 3n$ diffraction spots in the $0kl$ photographs along the c-axis. Similar observations were made in some chlorites and amesites (G. W. Brindley *et al.*, 1950, 1951) indicating frequent displacement of layers by $b/3$.

TABLE 1. UNIT CELL PARAMETERS OF THE 9-LAYER STRUCTURE

Orthorhombic Cell	Hexagonal Cell
$a = 5.295 \pm 0.004$ Å $b = 9.171 \pm 0.007$ $c = 63.99 \pm 0.06$	$a = 5.295 \pm 0.004$ Å $c = 63.99 \pm 0.06$

Of the $00l$ reflections only those were observed where $l = 3n$. This implies that the structure is composed of $3n$ serpentine layers in the unit cell of the mineral. Later it was found that there are 9 layers in the unit cell.

Using the above information and a carefully processed and measured powder photograph, the unit-cell parameters were refined with D. J. Britton's[1] (1964) least-squares program. Although the hexagonal cell is accepted in this study the corresponding parameters of the centered orthorhombic cell are also given in Table 1, for the sake of its comparison with data for other serpentine minerals.

CRYSTAL STRUCTURE DETERMINATION

In order to determine the crystal structure of the mineral a reasonably good crystal $(0.23 \times 0.18 \times 0.08$ mm$)$ was mounted with its basal plane perpendicular to the rotation axis of the goniometer. Three-dimensional intensities for 149 reflections were obtained from (1) the single crystal

[1] Britton, D. (1964) A Fortran program for refining unit-cell parameters. Department of Chemistry, University of Minnesota.

precession photographs by visual comparison of the diffraction spots with calibrated intensity strips and from (2) an equi-inclination single crystal diffractometer. The radiation used was $MoK\alpha$ for the former and $CuK\alpha$ for the latter technique. After the usual corrections of the measurements, acceptable agreement was found between the two sets of the data for over 96 percent of the reflections. The intensities of five strongly-diffused reflections, however, differed in the two sets by more than the limits of the measurement errors. These reflections were omitted. Due to the diffused nature of the reflections the film intensities were assumed to be the more reliable and were used for the structure analysis.

To determine the space group of this Al-serpentine, all the space groups consistent with the diffraction symmetries of $\bar{3}m$ and $\bar{3}$ were examined. The intensities of the basal reflections could be explained only if all the serpentine layers were oriented with their tetrahedra pointing in the same direction. This conclusion eliminated the centric space groups and also those possessing reflection planes or two-fold rotation axes normal to the c-axis. Later it was found that the correct space group of the mineral is $P3_1$ or $P3_2$.

Using the assumption that the basic unit in the crystal structure of this Al-serpentine is similar to the tetrahedral-octahedral layers of the serpentines and that the structure is composed of a multiple of three serpentine layers the structure factors of possible polytypes having space groups of $P3_1$ (or $P3_2$) were calculated. Only one 9-layer model gave intensities comparable to the observed values.

The stacking sequence of this 9-layer per unit-cell model bears similarities to the 3-layer chlorite of Brindley (1950) and to the polytypes of amesite and cronstedtite described by Steadman and Nuttall (1962 and 1963). The nine layers are built up by the respective 120° rotation (3_1 or 3_2 screw axis rotation) of identical 3-layer units of an AAB stacking order, where the third layer is displaced by $a_1/3 + a_2/3$ and rotated by 60° (or 180°) with respect to the identically oriented first two layers. Figure 3 illustrates this relationship between these 3 layers and the accumulation of the 9-layer $AABAACAAD$ unit cell.

The diffraction symmetry consistent with the space group of this model, however, is $\bar{3}$ while the observed diffraction symmetry is $\bar{3}m$. This can be explained by regarding the 9-layer unit as a subcell and an 18-layer unit (two 9-layer units), approximating the space group symmetry of $P31c$, as its supercell. This model of the supercell is supported by the presence of the additional, but hardly resolvable, diffused diffraction spots in the $10l$ and $01l$ reciprocal lattice rows observed in $CuK\alpha$ precession photographs. The higher diffraction symmetry could also be explained by the alternating twinning of approximately equal quantities of right- and left-handed 9-layer crystals.
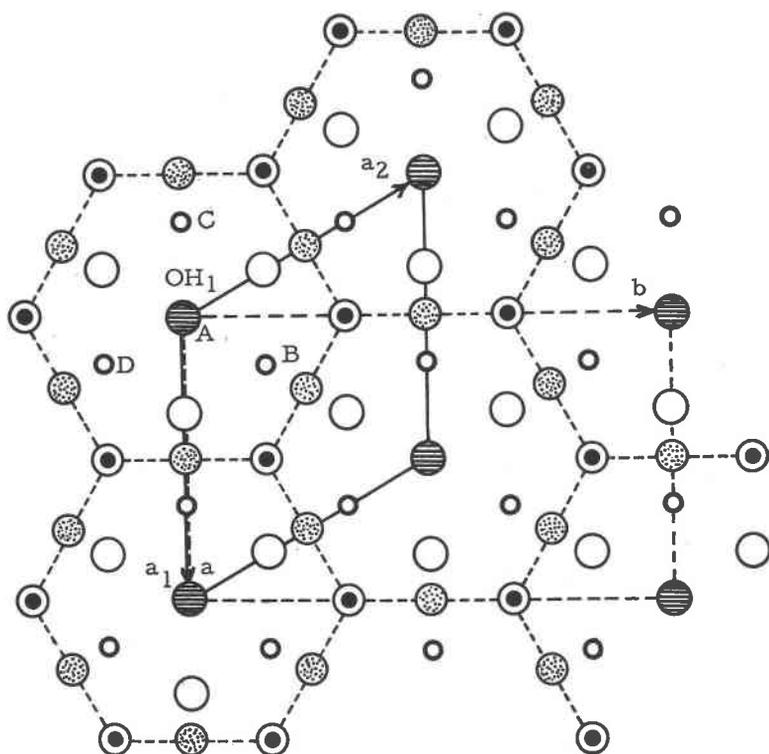
Fig. 3. c-axis projection of an idealized serpentine layer. A, B, C, D refer to the respective location of the origin of layers in the 9-layer unit cell. (Symbols: dot Si, Al; small circle Mg, Al; large circle OH_2; dashed circle OH_1; dotted circle O_{basal}; circle over dot O_{apical}).

The apparent $\bar{3}m$ diffraction symmetry of the crystal could further be explained by the polysynthetic twinning of the 3-layer (AAB) units by 120° rotation about the c-axis. This was observed by Sadanaga and Takeuchi (1961) in micas. This model, however, would fail to account for the weak and diffused extra diffraction spots observed, for example, in the $10l$ and $20l$ reciprocal lattice rows.

REFINEMENT

The refinement of the 9-layer structure model was done with the CDC 1604 computer of the University of Minnesota, using Finger's[1] least-squares program. Because of the difficulties of estimating the intensities of the diffused and weak diffraction spots, only those 144 reflections were

[1] Finger, L. W. (1965) A least-squares and Fourier program for crystallographic problems, Department of Geology and Geophysics, University of Minnesota.

TABLE 2. THE REFINED ATOMIC COORDINATES AND TEMPERATURE FACTORS OF THE
FIRST LAYER (REFERRED TO THE HEXAGONAL UNIT CELL OF TABLE 1)

Atom	Equipoint	x	y	z	$B(\text{Å}^2)$
O_b	c	0.46 ± 0.02	0	0	2.35
(Si, Al)	b	1/3	2/3	0.0090 ± 0.0006	1.25
O_a	b	1/3	2/3	0.0353 ± 0.0008	2.65
OH(1)	a	0	0	0.0353 ± 0.0008	2.65
(Mg, Al)	c	2/3	0	0.0513 ± 0.0009	1.55
OH(2)	c	1/3	0	0.0670 ± 0.0010	2.74

utilized in the refinement which had $l = 3n$. Half ionized scattering factors were used (International Tables for X-Ray Crystallography, 1962, V. III) and the intensities were weighted in accordance with the quality of the reflections and inversely proportional with l. The latter was empirically estimated to counterbalance the effect of stacking disorder.

The refinement program was carried out in four stages:

In the first stage the x coordinate of the basal oxygen was refined from $hk0$ reflections. In the second stage all the reflections were used and in

TABLE 3. OBSERVED AND CALCULATED STRUCTURE FACTORS

h	k	l	F_{obs}	F_{cal}	h	k	l	F_{obs}	F_{cal}	h	k	l	F_{obs}	F_{cal}	h	k	l	F_{obs}	F_{cal}
1	0	0	26.5	24.7	3	1	6	6.3	5.7	1	1	12	77.5	72.6	2	2	-18	16.0	14.0
2	0	0	8.7	7.8	-1	4	6	6.3	6.4	1	1	-12	21.2	13.7	1	1	21	44.4	37.8
3	0	0	98.8	104.9	-3	4	6	6.3	6.4	2	2	12	25.6	23.2	1	1	-21	20.4	24.4
1	1	0	11.9	11.4	1	1	-6	34.3	23.7	2	2	-12	48.7	41.1	2	2	21	15.4	20.0
2	2	0	20.5	19.7	2	2	6	18.2	20.6	1	0	15	12.3	11.4	2	2	-21	24.5	21.3
1	2	0	16.2	15.3	2	2	-6	38.5	42.7	0	1	15	12.3	11.4	1	1	-24	22.4	22.2
2	1	0	16.2	15.3	1	0	9	22.9	24.6	2	0	15	5.6	6.2	2	2	24	10.0	10.0
3	1	0	11.0	12.1	0	1	9	22.9	24.6	0	2	15	5.6	6.2	2	2	-24	25.0	33.7
1	3	0	11.0	12.1	2	0	9	8.9	8.7	1	2	15	8.1	8.1	0	0	27	58.5	51.4
1	4	0	11.4	10.4	0	2	9	8.9	8.7	2	1	15	8.1	8.1	3	0	27	33.0	31.7
4	1	0	11.4	10.4	2	1	9	15.1	16.3	-1	3	15	8.1	6.6	0	3	27	33.0	31.7
1	0	3	12.8	12.4	1	2	9	15.1	16.3	-2	3	15	8.1	6.6	1	1	27	55.3	57.1
0	1	3	12.8	12.4	1	3	9	12.0	11.0	1	3	15	5.6	4.8	1	1	-27	18.9	14.4
2	0	3	4.6	3.9	-1	3	9	15.1	14.6	3	1	15	5.6	4.8	2	2	27	5.1	3.9
0	2	3	4.6	3.9	-2	3	9	15.1	14.6	-1	4	15	5.6	6.7	2	2	-27	35.9	39.5
1	2	3	7.6	7.8	3	1	9	12.0	11.0	-3	4	15	5.6	6.7	1	1	30	54.0	60.1
2	1	3	7.6	7.8	-1	4	9	12.0	13.2	1	1	-15	20.7	11.0	1	1	-30	23.7	18.6
-1	3	3	7.6	7.5	-3	4	9	12.0	13.2	2	2	15	29.1	24.6	2	2	30	15.5	17.7
-2	3	3	7.6	7.5	3	0	9	51.9	47.2	2	2	-15	25.5	27.3	2	2	-30	34.2	37.3
1	3	3	6.1	5.9	0	3	9	51.9	47.2	1	0	18	20.4	21.0	1	1	-33	43.8	35.4
3	1	3	6.1	5.9	1	1	9	67.8	66.8	0	1	18	20.4	21.0	2	2	33	32.3	28.4
-1	4	3	6.1	6.2	1	1	-9	26.2	21.4	2	0	18	13.6	14.4	0	0	36	35.8	31.0
-3	4	3	6.1	6.2	2	2	9	25.2	22.5	0	2	18	13.6	14.4	3	0	36	21.9	18.1
1	1	3	39.0	27.0	2	2	-9	49.5	46.8	1	2	18	15.3	15.7	0	3	36	21.9	18.1
1	1	-3	16.8	18.1	1	0	12	12.8	12.0	2	1	18	15.3	15.7	1	1	36	34.0	37.0
2	2	3	14.1	17.0	0	1	12	12.8	12.0	-1	3	18	15.3	12.2	1	1	-36	50.8	49.8
2	2	-3	37.4	31.4	2	0	12	4.7	5.2	-2	3	18	15.3	12.2	1	1	39	23.0	20.1
1	0	6	12.8	12.4	0	2	12	4.7	5.2	1	3	18	11.9	8.7	1	1	42	14.5	10.2
0	1	6	12.8	12.4	1	2	12	7.6	8.2	3	1	18	11.9	8.7	1	1	-42	55.2	52.6
2	0	6	4.9	4.0	2	1	12	7.6	8.2	-1	4	18	11.9	13.1	0	0	45	52.6	48.4
0	2	6	4.9	4.0	-1	3	12	7.6	7.0	-3	4	18	11.9	13.1	3	0	45	43.5	41.1
1	2	6	7.6	8.0	-2	3	12	7.6	7.0	3	0	18	42.5	39.1	0	3	45	43.5	41.1
2	1	6	7.6	8.0	1	3	12	5.8	5.2	0	3	18	42.5	39.1	1	1	45	17.1	13.2
-1	3	6	7.6	7.4	3	1	12	5.8	5.2	1	1	18	51.5	48.4	1	1	-45	40.5	40.5
-2	3	6	7.6	7.4	-1	4	12	5.8	6.7	1	1	-18	19.0	19.0	0	0	54	48.8	43.6
1	3	6	6.3	5.7	-3	4	12	5.8	6.7	2	2	18	25.3	24.5	0	0	63	36.0	32.0

Scale Factor = F_{obs} / F_{cal} = .252

c-axis projection   a-axis projection
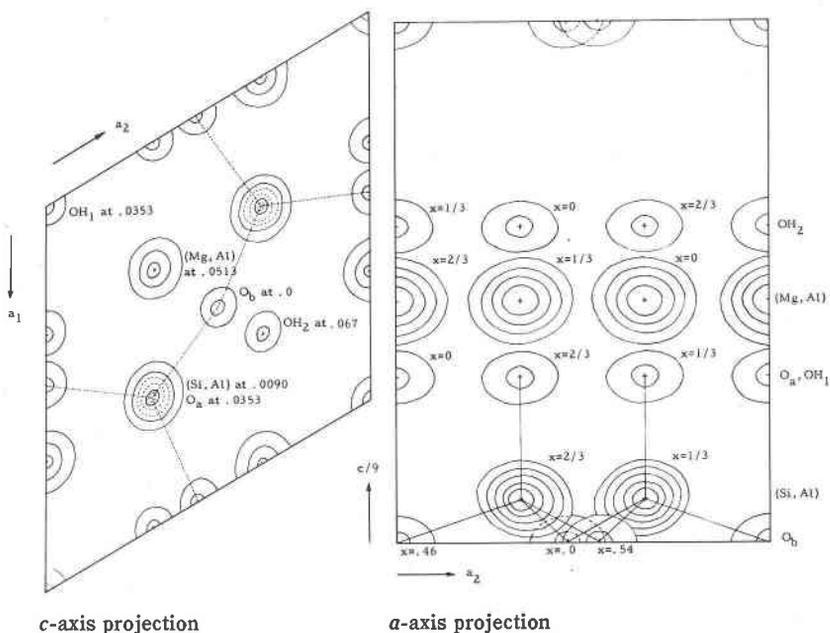
FIG. 4. c-axis projection of three-dimensional electron density sections of the first layers of the Al-serpentine structure.

FIG. 5. a-axis projection of three-dimensional electron density sections of the first layers of the Al-serpentine structure.

addition to the above, the z coordinates of all atoms and the scale factor were allowed to vary. In the third stage isotropic temperature factors were also refined.

At this point difficulties were encountered in getting some unreasonably large and one negative temperature factor. Forty-five reflections with $(|F_O| - |F_C|)/|F_O|$ larger than 15 percent were rejected and the refinement of stage three was repeated by varying only the temperature and scale factors. In the final stage of refinement the temperature factor was fixed and the other parameters were refined.

The parameters thus obtained from a total of 17 cycles of refinement with the corresponding standard deviations are given in Table 2. The final R factor for all 144 reflections was 10.2 percent. The list of the observed and calculated structure factors is given in Table 3.

Three dimensional electron densities and interatomic distances and angles were calculated using L. Finger's[1] program. Two composite pro-

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[1] Finger, L. W. (1966) A Fortran program for calculating bond distances and angles, thermal ellipsoid of vibrations and errors, Department of Geology and Geophysics, University of Minnesota.

jections of electron density sections passing through atoms are shown in Figures 4 and 5 and selected interatomic distances and angles with the corresponding standard deviations are given in Table 4.

DISCUSSION OF THE STRUCTURE

In the final structure the symmetry of the tetrahedral sheets is ditrigonal as in many other sheet silicates. With respect to the ideal hex-

TABLE 4. IMPORTANT INTERATOMIC DISTANCES AND ANGLES FOR AL-SERPENTINE

Interatomic Distances in Ångstroms	
T-T	3.057 ± 0.002
O_b-O_b	2.67 ± 0.02
O_b-O_a	2.74 ± 0.04
T-O_b	1.65 ± 0.01
T-O_a	1.68 ± 0.06
T-O (average)	1.66 ± 0.02
M-M	3.057 ± 0.002
M-OH_1	2.04 ± 0.04
M-OH_2	2.03 ± 0.04
OH-OH (shared edges)	2.69 ± 0.06
OH-OH (non-shared edges)	3.057 ± 0.002
O_b-OH_2	2.90 ± 0.06
Octahedral volume in $Å^3$	11.2 ± 0.2

Bond Angles in Degrees	
O_b-T-O_b	108 ± 1
O_b-T-O_a	110 ± 1
T-O_b-T	136 ± 3
α (obs.)	8 ± 3
α (calc.)	8 ± 3

T = tetrahedral = (Si, Al)
M = octahedral = (Mg, Al)
b = basal   a = apical
α = angle of tetrahedral rotation

agonal network, the tetrahedra are rotated in opposite directions by 8 degrees. As a consequence of this rotation the basal oxygen is moved by 0.21 Å from its hexagonal position toward the (OH) in the octahedral sheet.

The O-O (2.67–2.74 Å) and (Si, Al)-O (1.65–1.68 Å) distances are shorter in this hexagonal Al-serpentine than in amesite (2.72–2.78 Å; 1.67–1.71 Å) and longer than in clinoenstatite (2.62–2.63 Å; 1.57–1.63 Å). These differences are in agreement with the degree of Al substitution in these three minerals and the corresponding and expected changes in interatomic distances. On the other hand the (Mg, Al)-O distances are

almost the same in Al-serpentine and in amesite implying similar Mg contents in the octahedral sites.

The percentage of Al in the tetrahedral site can be estimated from the average (Si, Al)-O distance using the charts of Smith and Bailey (1963) and from the basal spacing using the charts of Nelson and Roy (1958). The former technique indicates an Al content of 0.54 and the latter of 0.59 in the chemical formula. This is in good agreement with the value of 0.57 obtained from the microprobe analysis.

The amount of aluminum in this Al-serpentine can also be confirmed
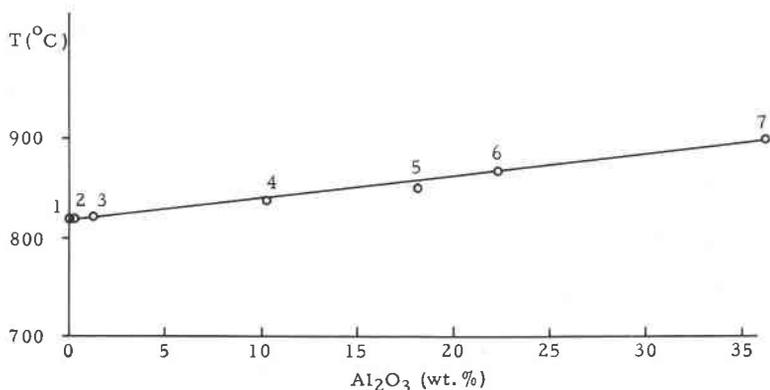
FIG. 6. Chart of the decomposition temperature and Al content of seven serpentines. (1) Orthoantigorite (Brindley and Knorring, 1954) (2) (3) Analyzed Mg-serpentines from the former Rock Analyses Laboratory of the University of Minnesota (4) Synthetic Al-serpentine (Roy and Roy, 1952) (5) Synthetic Al-serpentine (Yoder, 1952) (6) Hexagonal Al-serpentine of this study (7) Amensite (Brindley et al., (1951).

from its decomposition temperature observed in the D.T.A. curve and the graph of Figure 6. This graph is a plot of the decomposition temperatures of several serpentines, an orthoantigorite and an amesite against their Al_2O_3 content. It reveals the linear relationship between these two quantities and is in agreement with the observations of Roy and Roy (1952) concerning the increase of the thermal stability of serpentine with increasing Al-content.

The relationship between the crystal habit and the amount of aluminum in serpentines was investigated by Nelson and Roy (1954) and Gillery (1959). They synthesized a number of Al-serpentines with general formulae of $(Mg_{6-x}Al_x)(Si_{4-x}Al_x)O_{10}(OH)_8$ with x ranging from 0 to 2.5. For $x=0-0.25$ they obtained a fibrous, for $x=0.75$ a platy 1-layer, and for $x=1.5$ a platy 6-layer orthohexagonal polytype. Radoslovich (1963) suggested that high Al-content favors the formation of $3n$-layered polytypes. The corresponding value of x in our Al-serpentine is in the order

of 1.2. Thus its good hexagonal platy crystals and its $3n$ stacking sequence confirm the above observations and predictions.

Radoslovich (1963) in a study of layered silicates concluded that most of the dimensional adjustment is achieved by changes in bond angles. According to him, in serpentines the angle between the apical and basal oxygen through the tetrahedral cation is a function of tetrahedral Al/Si ratio and expresses the stress in the tetrahedral and octahedral layers. When this angle is lower than 107°, the stress is high and there is a tendency for development of fibrous crystals. For angles considerably greater than 107°, the stress is low and better platy crystals can grow. In our Al-serpentine this angle is 110°, consistent with the development of good platy crystals.

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