

A STRUCTURAL STUDY OF THE THERMAL TRANSFORMATION OF SERPENTINE MINERALS TO FORSTERITE*

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ABSTRACT

X-ray single crystal and powder methods have been applied to a study of the thermal transformation of all the known crystal structure varieties of serpentine to forsterite. Directional and dimensional relations are established between the serpentine and forsterite structures. Evidence is also obtained for some degree of ordering during the transition process which appears to be related to the structural and chemical characteristics of the initial mineral.

INTRODUCTION

The transformation of serpentine minerals to forsterite (or olivine) by heating in air has been studied many times in the past when the main objectives were identification of the products formed (see for example Hargreaves and Taylor, 1946) and the determination of the thermal changes accompanying the process. More detailed structural studies by Aruja (1943) and by Hey and Bannister (1948) on the transformation of chrysotile fibres showed certain orientational relations between the initial and final materials. A similar study by Brindley and Ali (1949) of the transformation of chlorite to olivine established relations between the initial and final unit cells, and plausible suggestions were made regarding the mechanism of the transformation. Similar methods have now been applied to the transformations of all the known crystal-structural varieties of serpentines. The detailed work in recent years by Whittaker, Zussman, Jagodzinski and Kunze, Brindley and others on the structural varieties of the serpentines makes it opportune now to examine their transformation characteristics.

Although the present studies are concerned with dry heating conditions, the results obtained from hydrothermal methods may have some relevance, particularly if a water vapor atmosphere is trapped in the material at or near the transformation temperature. Bowen and Tuttle (1949) studied pure synthetic magnesian chrysotile and obtained forsterite and talc as reaction products at 500–530° C. and over a wide range of water vapor pressures. Nelson and Roy (1954 and private discussion)

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applied both hydrothermal and dry heat treatments at about 590° C. to natural chrysotile and antigorite, and to various synthetic materials ranging in composition from pure serpentine, through chlorite, to amesite. Among other results, they found that antigorite transforms hydrothermally to talc, forsterite and chlorite, but chrysotile in general gives only talc and forsterite. Under appropriate dry heating conditions, they found a broad reflection corresponding to a spacing of about 13.5–14 Å was developed which they considered "may be due to some intermediate metastable arrangement." The formation of chlorite from antigorite but not generally from chrysotile may be correlated with the somewhat greater content of aluminum and other trivalent ions in antigorite than in chrysotiles.

EXPERIMENTAL METHODS AND SPECIMENS USED

Specimens have been used which were previously studied by Zussman, Brindley and Comer (1957) by *x*-ray and electron diffraction methods and with the electron microscope. They are as follows:

Chrysotiles	{	(1) Silky chrysotile fibres (Transvaal) 85% clino-chrysotile, 15% ortho-chrysotile
		(2) Splintery clino-chrysotile (Zermatt)
6-layer ortho-serpentine	{	(3) Massive green serpentine (Unst, Shetland Isles)
		(4) Fibrous blue-green serpentine (Unst, Shetland Isles)
Lizardites 1-layer ortho-serpentine	{	(5) Massive green serpentine (Snarum, Norway)
		(6) White platy serpentine (Kennack Cove, Cornwall)
Antigorites (long <i>a</i> spacing)	{	(7) Fibrous antigorite (Shipton, Quebec)
		(8) Platy antigorite (Glen Urquhart, Scotland)

Chemical analyses of some of these minerals and of several other specimens examined in the previous work are listed in an appendix to this paper.

The materials were finely powdered, heated in air in a muffle furnace, and kept at successively higher temperatures for periods of 12 hrs. each. Before the initial heating and after cooling from each of the temperatures, 500, 550, 575, 600, 625, 650, 700, 750 and 800° C., diffractometer traces were obtained with standard Norelco *x*-ray equipment. Fibre and single crystal diagrams were recorded for specimens 1, 2, 4, 6 and 7 using a Unicam single-crystal goniometer. The specimens were maintained successively at various temperatures for periods between 6 and 12 hours.

RESULTS

(a) *X*-ray powder data

In view of previous descriptions by Whittaker and Zussman (1956) it suffices to state the results for the unheated specimens very briefly.

TABLE 1. SEQUENCE OF CHANGES IN X-RAY POWDER PATTERNS OF HEATED SERPENTINES
 Each column gives the approximate temperature, deg. C., at which a change in the pattern is clearly observed

Specimen	Weakening of $h0l$ reflections	Weakening of $00l$ reflections	Changes in hkl bands	Low angle reflection appears	Low angle reflection max. int.	Forsterite pattern appears	Serpentine pattern absent
1. Chrysotile, silky fibre	500	550-575	550	575	575-600	575-600	625
2. Chrysotile, spintery	550-575	575	575	550-575	575-600	600	625-650
3. 6-layer ortho, massive	550	550	550-575	550-575	575-600	575-600	575-600
4. 6-layer ortho, fibrous	575-600	575	550-575	575	625	650	625
5. Lizardite, massive	500-550	550	550	550	575	600	575-600
6. Lizardite, platy	500-550	575	575	<500	575-600	575	575-600
7. Antigorite, fibrous	575	625-650	—	625	650	625	650-700
8. Antigorite, platy	600	625	—	650	700	650	650-700

TABLE 2. "d" VALUES OF LOW-ANGLE PEAK IN POWDER TRACES OF HEATED SERPENTINES

No.	Specimen	No. of layers per cell	Temp. ° C.									
			500	550	575	600	625	650	700	750	800	
1	Silky chrysotile	2	—	—	15.2†	15.2	15.0	14.7	—	—	—	—
2	Spintery chrysotile	2	—	—	14.7†	14.7	14.7	14.2	—	—	—	—
3	6-layer ortho (massive)	6	—	—	15.0 _{zw} †	15.2 _{zw}	—	—	—	—	—	—
4	6-layer ortho (fibrous)	6	—	—	14.3	14.3	14.2†	14.2	—	—	—	—
5	Lizardite (massive)	1	—	—	14.7 _{zw} †	14.3 _{zw}	—	—	—	—	—	—
6	Lizardite (platy)	1	14.0	13.6	12.6†	11.8	11.6	11.2	10.8	10.5	—	—
7	Antigorite (fibrous)	1	—	—	—	—	13.6	13.2†	11.6	—	—	—
8	Antigorite (platy)	1	—	—	—	—	—	12.6	10.3†	9.7	—	—

† Indicates temperature at which reflection is of maximal intensity.

_{zw} Signifies weak indications of a peak.

The weakening of $h0l$, together with any hkl reflections there may be, corresponds to a disruption of stacking order. The weakening and broadening of hk bands indicate disintegration of the layer network into ever smaller ordered regions. The development of a broad reflection at low angles, about $2\theta = 6^\circ$ for $\text{CuK}\alpha$ radiation, corresponding to a spacing of about 14 Å, was previously noted by Nelson and Roy (1954). The subsequent behavior of this peak, see Table 2, varies for different specimens, remaining largely constant for specimens 1-4, and perhaps also for specimen 5, but diminishing down to about 10 Å for antigorites and platy lizardite, specimens 6, 7 and 8 (c.f. Fig. 1).

At about 575-600° C., forsterite peaks are observed and this is followed by complete disappearance of the serpentine pattern. For specimens Nos. 3 and 5, both of which are massive, the serpentine peaks vanished before forsterite appeared. This is probably the result of a very fine-grained texture, rather than of any structural features. In the final stage of the transformation, the low-angle reflection in the 10-14 Å spacing range and the forsterite pattern remain superimposed on the scattering band of an amorphous product.

(b) *Single crystal x-ray diagrams*

These are important for the evidence they provide of orientation relations between the initial and final stages of the transformation, and also for additional evidence bearing on the transitional stage itself.

For silky chrysotile, specimen No. 1, results similar to those of Aruja (1943) and of Hey and Bannister (1948) are obtained, confirming that after heating to 600° C. spots and short powder arcs are produced belonging to the patterns of forsterite crystals* which have $[010]_F$ and $[013]_F$ approximately parallel to the original fibre axis $[100]_S$. Layer lines from $[010]_F$ crystals were superimposed upon even-order layer lines from $[013]_F$, but spots belonging to the $[010]_F$ orientation were identified by measurement of ξ values. Also the following features additional to those previously reported, were noted:

- (i) A few weak spots were observed suggesting the presence of crystals with orientation $[001]_F$ parallel to $[100]_S$.
- (ii) hk streaks which extended along layer lines persisted later than other serpentine reflections as diffuse scattering areas, having lost their "tails" and spread in the direction normal to the layer lines.
- (iii) Near the center of the film is a region of scattering corresponding to a spacing greater than 14 Å which is confined to the zero layer line.

* Subscripts F and S are used henceforth for indices referring to forsterite and serpentine cells respectively.

This is related to the broad, low-angle peak seen in the powder traces and shows the latter to be $00l$ or an $0kl$ reflection.

The splintery and fibrous specimens, Nos. 2 and 4, also with $[100]_S$ along their length, yielded forsterite crystals with orientations as for specimen 1, but their diffraction patterns were less clear owing to the large range of misalignment present at all stages.

Fibrous antigorite (picrolite), specimen No. 7, is poorly oriented with fibre axis $[010]_S$ and on heating transforms to forsterite crystals of which most have $[001]_F$, but some have $[011]_F$, parallel to $[010]_S$. These orientations are approximately perpendicular to those produced from serpentines with $[100]_S$ as fibre axis.

Single crystals of white platy lizardite (specimen No. 6) yielded oscillation and rotation photographs which showed reflections of forsterite after heating to 580° C. After 600° C., the serpentine reflections had disappeared except for hk bands which are seen as diffuse streaks in Fig. 2(a). Spots and arcs in this figure are mainly from forsterite with $[001]_F$ parallel to $[010]_S$. Above 650° C. other diffuse areas appeared on layer lines corresponding to a repeat distance of about 18 \AA ; these are seen in Fig. 2(b). They became stronger on further heating and could be indexed using a cell similar to that of serpentine but with double its b -axis. Evidence for this diffuse scattering was also seen in the diffractometer traces of powdered lizardite (see Fig. 1, traces at 600° and 650° C.). In rotation photographs about $[100]_S$ some of the diffuse reflections were of the type which would be absent from serpentine through the face-centering of its lattice.

The rotation diagrams showed the forsterite to be strictly orientated with respect to the original serpentine in the following way:

$$\begin{aligned} [010]_F \text{ and } [013]_F &\text{ parallel to } [100]_S, \\ [001]_F \text{ and } [011]_F &\text{ parallel to } [010]_S, \end{aligned}$$

and by implication

$$[100]_F \text{ is parallel to } c^*_S.$$

These relations hold for all the serpentines examined in this work so far as they can be evaluated from the x -ray data; limitations are imposed when fibre diagrams only can be obtained.

In addition the following relations hold between the unit cell parameters:

$$2a_S \simeq b_F \text{ and } 2b_S \simeq 3c_F.$$

THE RELATIONS BETWEEN THE SERPENTINE AND FORSTERITE STRUCTURES

The serpentine minerals have layer structures, each layer comprising one sheet of linked Si-O tetrahedra and one sheet of Mg-O(OH) octa-

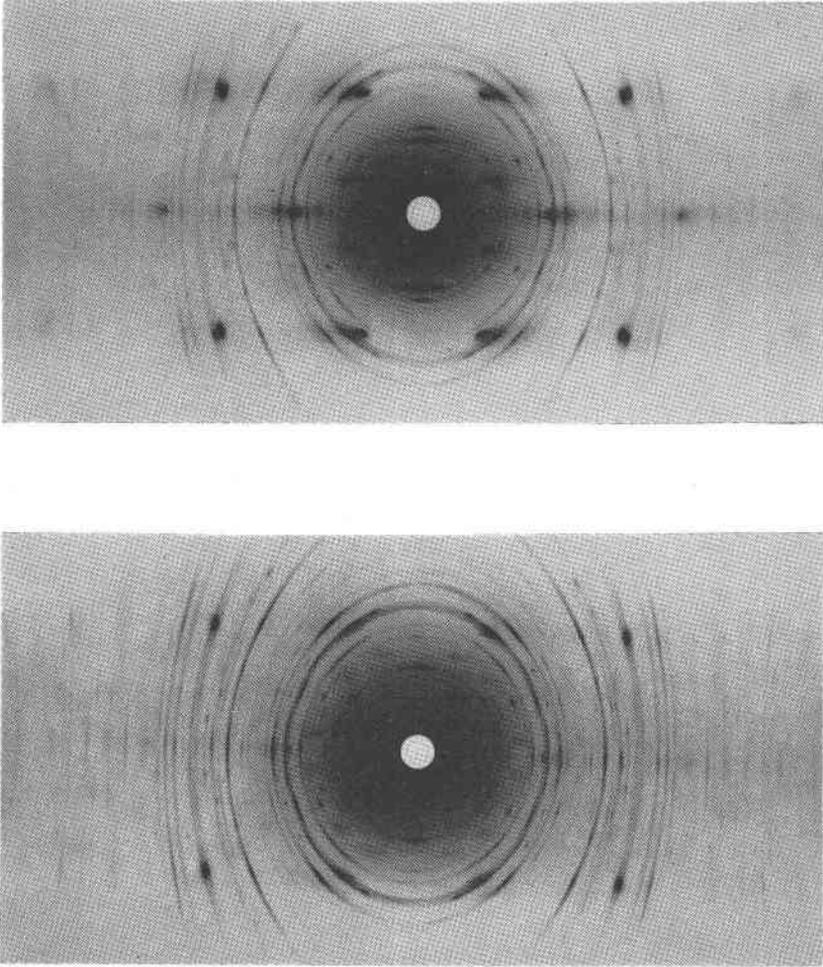


FIG. 2. Rotation diagrams about the *b*-axis of a single flake of specimen No. 6.
 (a) Above. After heat treatment at 580° C.
 (b) Below. After heat treatment at 800° C.

hedra; the overall composition is $Mg_3Si_2O_5(OH)_4$. Forsterite, Mg_2SiO_4 , contains discrete SiO_4 tetrahedral groups joined by Mg-O octahedral groups. Tilley (1948) suggested the following relation for the transformation of pure magnesian serpentine:



The unit cell relations, $2a_S = b_F$ and $2b_S = 3c_F$, show that four cells in one layer of serpentine transform to 3 cells in one layer of forsterite. The

transformation can be considered layer by layer with the following atoms involved:

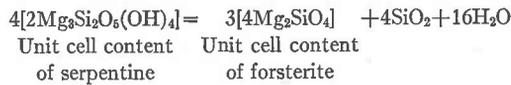


Fig. 3 suggests schematically the movement of atoms when the serpentine structure containing three oxygen-hydroxyl sheets per layer collapses to the forsterite structure containing two oxygen sheets per unit of

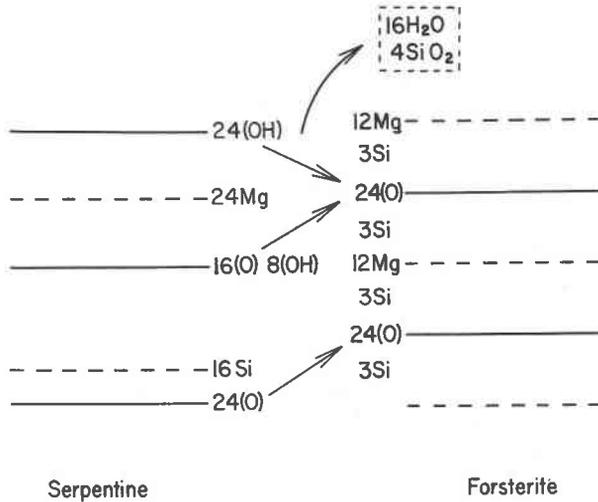


FIG. 3. Schematic diagram showing transition from serpentine to forsterite. Numbers of atoms given are contained in 4 unit cells of serpentine, and 3 unit cells of forsterite respectively.

structure. It is evident that there must be considerable re-organization of the Mg-O(OH) part of the serpentine layer when dehydration occurs, accompanied by a collapse of the layer structure to the three-dimensionally coordinated forsterite structure.

It is supposed that the Si-O bonds in the serpentine structure remain largely intact when the transformation takes place, although some must be broken because (a) the continuously linked tetrahedra of serpentine give place to separate tetrahedra in forsterite, and (b) some silica is discarded if the chemical relation given above is correct.

Figs. 4(a) and 4(b) show superpositions of the serpentine and forsterite structures with the correct unit cell relations. For clarity only the tetrahedrally coordinated part of the serpentine layer is indicated by means of broken lines; in any case, the Mg-O(OH) part suffers major reorganiza-

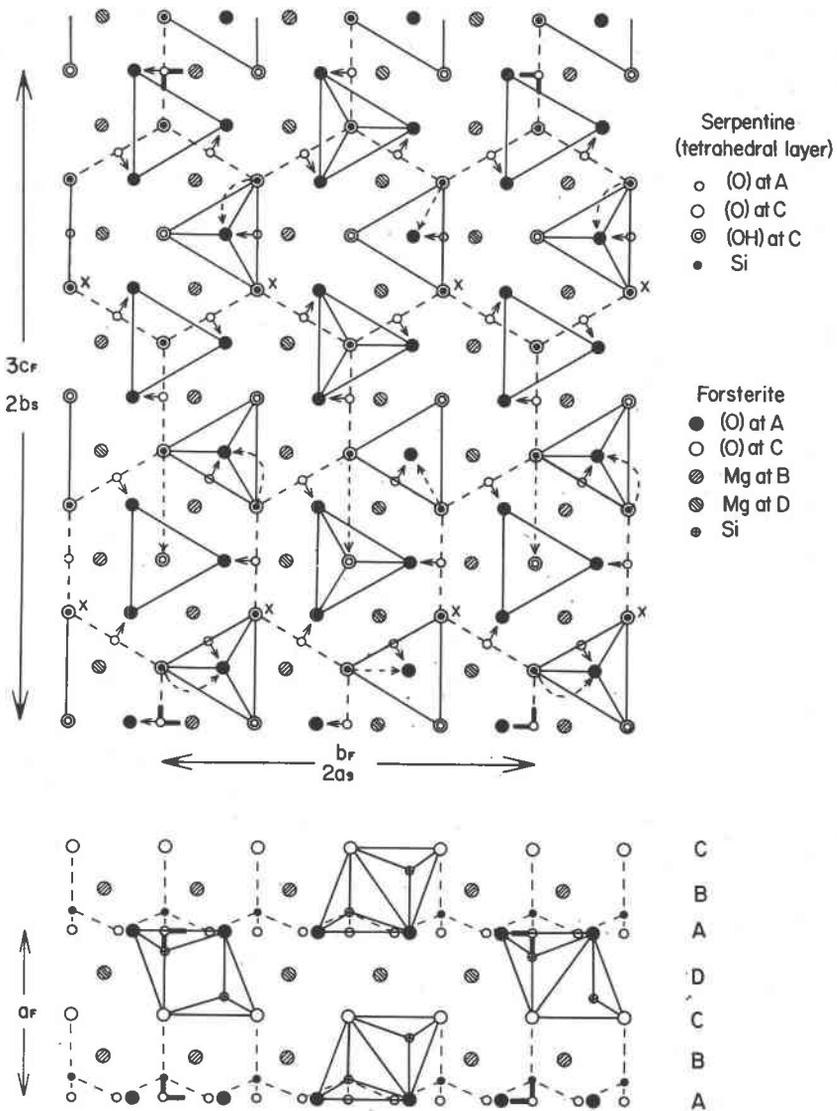


FIG. 4. (a) Above. Relation between the forsterite structure and the tetrahedral part of the serpentine structures as viewed along a_F . Solid lines: edges of tetrahedra in forsterite. Broken lines: hexagonal Si-O network in serpentine. Solid arrows: movements of oxygen atoms. Dashed arrows: movements of silicon atoms.

(b) Below. Relation between forsterite and collapsed serpentine as seen along c_F . Solid lines: tetrahedra and Si-O bonds in forsterite. Broken lines: tetrahedral part of serpentine structure.

tion as previously explained. The tetrahedra of the forsterite structure are heavily outlined, and the edges of the tetrahedra are shown in such a manner as to distinguish between those pointing up and those pointing down with respect to a_F . A comparison of Figs. 4(a) and 4(b) makes these orientations clear. In serpentine the tetrahedra all point in one direction and the Si atoms lie at one level. However, one can see by comparison of Figs. 3, 4(a) and 4(b), that some tetrahedra in serpentine require only a small rotation to pass to the arrangement in forsterite, and in other cases it is only necessary for the Si atoms to migrate to the other side of the oxygen network. The collapse of the serpentine layer structure indicated in Fig. 3 can be considered in relation to the structure shown in Fig. 4(b). The latter is essentially the structure of forsterite, and in consequence the initial tetrahedral networks indicated by broken lines are shown closer together than their true separation in serpentine.

THE TRANSITION STAGE

Additional evidence concerning the transition process is provided by the low-angle reflection recorded in powder diagrams (see Table 2). The salient features of this reflection are that it remains relatively stationary and fairly sharp at about 14–15 Å for specimens 1–4. These are essentially 2-layer structures, and chemically appear to be among the purer magnesian silicates. (The material showing the 6-layer characteristics may contain a large proportion of 2-layer type material, or the 6-layer structure may not be very different from a 2-layer structure; the reflections which indicate the 6-layer character are all weak and are few in number.) The low-angle reflection shows a progressive shift from about 14 Å down to about 10 Å, for specimens 6, 7 and 8; these are essentially 1-layer structures and chemically they are richer in R_2O_3 than specimens 1–4. These reflections are broader and would be interpreted generally as arising from a variable mixed-layer sequence. A priori, it is difficult to decide whether the structural or the chemical aspect or both of the original minerals is to be associated with the long spacing developed transitionally by the heat treatment.

In the first place it may be supposed that the two-layer structural character of specimens 1–4 imposes a corresponding regularity on the transformation product, giving rise to a $2 \times 7.3 = 14.6$ Å periodicity. The possibility of a chlorite-like material is also suggested by a 14 Å spacing but several arguments point against this. In the first place, chlorites seldom if ever give a spacing greater than 14.3 Å whereas the observed spacing ranges mainly from about 14.5–15.0 Å. Chlorites normally contain appreciable R_2O_3 components, but specimens 1–4 are among the purer magnesian serpentines. The hydrothermal experiments of Bowen and Tuttle (1949) showed no development of chlorite from pure magne-

sian serpentine. It is tempting but perhaps misleading to refer also to the observation by Hill (1955), examined in more detail by Roy and Brindley (1956), that a similar transitional 14 Å reflection can be obtained by heating the aluminian silicate, dickite, which also has a two-layer structure. Here, however, the dehydrated phase (metadickite) probably retains the whole of the silica and alumina of the original mineral. Hill (1956) has advanced certain hypotheses concerning the 14 Å reflection from dickite based on a new linkage of the type Al-O-Si between the layers, but it still remains difficult to see how this accounts for the 14 Å reflection, and it is equally difficult in the magnesian case.

While no certain proof can be founded on a single observation, we are inclined to the view that the nearly stationary 14 Å reflection from specimens 1-4 arises in some as yet unknown way from the 2-layer type of structure. It could perhaps arise from a concentration of the discarded silica in sheets 14 Å apart i.e., we may visualize two serpentine layers transforming to two forsterite layers plus one layer of silica. As heating progresses, silica migrates out from these interlayer positions allowing the forsterite sheets to come together; the powder diagrams do, in fact, suggest that the forsterite pattern grows in intensity as the 14 Å reflection diminishes. It is perhaps also significant that the transition to forsterite occurs at a somewhat lower temperature for the two-layer structures as compared with specimens 7 and 8, but evidence of this kind has to be accepted very cautiously because solid state reactivity depends on many factors. The observed wide band of scattering may arise from amorphous SiO₂.

The second case to be considered is that of the 1-layer serpentines which show the low-angle reflection moving from 14 Å to about 10 Å. The R₂O₃ content of these specimens is favorable to the formation of a considerable number of chlorite-like layers and moreover only very small amounts of Al₂O₃ are acceptable in the olivine structure. Roy and Roy (1955) have already discussed the formation of chlorite layers from aluminian serpentines while Nelson and Roy (1954) have shown the hydrothermal development of chlorites from such serpentines. The broad character of the observed reflection and its movement towards 10 Å at higher temperatures points strongly to a mixed-layer sequence of roughly 14 and 10 Å layers. If, with the expulsion of water, the chlorite-like 14 Å units break down into pairs of forsterite units of thickness $2 \times 4.73 = 9.56$ Å, then we have a simple mechanism by which 14 and 10 Å units will coexist in proportions varying with the heat-treatment. It may be recalled that Brindley and Ali (1950) showed that chlorite-like 14 Å layers persisted after the partial dehydration of magnesian chlorites.

Additional evidence concerning the transition product of a 1-layer serpentine is obtained from the diffuse reflections seen in Fig. 2(b) which

may be indexed using a cell with dimensions $3a_F \times b_F \times 3c_F$ ($14.28 \times 10.20 \times 17.97$). Possible indices, with ξ and ζ values are given in Table 3.

If this indexing is more than fortuitous it suggests that these repeat distances refer to the transformation product. Two of them are evident in Fig. 4(a), namely $b_F \simeq 2a_S$ and $3c_F \simeq 2b_S$.

Reflections 3, 6 and 7 are the strongest and are probably responsible for the broad peak at about $2\theta = 29^\circ$ seen in Fig. 1 (600° and 650°).

TABLE 3. DIFFUSE REFLECTIONS FROM A TRANSITION PRODUCT OBTAINED ON HEATING A ONE-LAYER SERPENTINE (LIZARDITE)

	<i>hkl</i>	ζ calc.	ζ obs. ($\pm .005$)	ξ calc.	ξ obs. ($\pm .005$)
1	530	0.000	0.000	.705	.710
2	240	0.000	0.000	.642	.640
3	231	0.086	0.085	.502	.500
4	022	0.171	0.170	.302	.300
5	062	0.171	0.170	.906	.905
6	304	0.343	0.340	.324	.330
7	115	0.429	0.430	.186	.200
8	316	0.514	0.510	.357	.360

CONCLUSIONS

Serpentines transform to forsterite in the same general way as do magnesian chlorites. Although the final transformation appears to be the same or closely similar for all structural varieties of serpentines, the transitional stage appears to depend on whether the initial minerals have a two-layer or a one-layer type of structure; the six-layer type appears to behave like the two-layer and it may have a predominantly two-layer character. The two-layer serpentines behave similarly to the two-layer aluminian silicate, dickite, giving a transitional product with a nearly constant 14.5 Å spacing, while the one-layer serpentines give a transitional product with a spacing which diminishes from about 14 down to 10 Å. A single crystal study of a one-layer serpentine showed a doubling of the a_S and b_S parameters during the transition stage.

ACKNOWLEDGMENTS

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APPENDIX

SPECTROGRAPHIC ANALYSES OF SOME SERPENTINE MINERALS
(Data supplied by Dr. F. L. Pundsack and Mr. Richard Wiley)

	Silky crysotile (1) (Transvaal)	Splintery crysotile (2) (Zermatt)	Massive Lizardite (5) (Snarum)	Fibrous Antigorite (7) (Quebec)	Fibrous Antigorite (Maryland)	Platy Antigorite (Antigorio)
SiO ₂	43 ± 3%	43 ± 3%	41 ± 3%	43 ± 3%	45 ± 3%	44 ± 3%
TiO ₂	0.001	0.001	0.003	0.009	0.001	0.003
Al ₂ O ₃	0.09	0.07	0.17	0.27	0.68	2.7 ± 0.2
Cr ₂ O ₃	nil	nil	0.009	nil	0.05	0.23
Fe ₂ O ₃	0.82	2.1 ± 0.2	1.8 ± 0.1	3.1 ± 0.2	6.3 ± 0.4	5.6 ± 0.3
Mn ₂ O ₃	0.04	0.05	0.009	0.09	0.07	0.11
B ₂ O ₃	0.04	0.04	0.11	<0.01	<0.01	<0.01
MgO	44 ± 3%	43 ± 3%	45 ± 3%	42 ± 3%	37 ± 3%	38 ± 3%
NiO	0.004	0.007	0.02	0.03	0.17	0.12
CaO	0.02	0.05	0.11	0.03	0.03	0.09
Na ₂ O	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05
K ₂ O	<0.10	<0.10	<0.10	<0.10	<0.10	<0.10
V ₂ O ₅	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Ignition loss 175°-1000°	13.16	12.83	13.35	11.88	10.73	10.33

WET CHEMICAL ANALYSES OF SOME SERPENTINE MINERALS

	(a)	(b)	(c)	(d)	(e)
	Silky chrysotile (1) (Transvaal)	Massive serpentine (3) (Unst)	Platy lizardite (6) (Kennack)	Platy antigorite (Caracas)	Platy antigorite (Mikonui)
SiO ₂	41.33	41.65	44.49	43.60	43.45
TiO ₂	0.02	nil	0.03	0.01	0.02
Al ₂ O ₃	0.80	0.10	2.26	1.03	0.81
Cr ₂ O ₃	—	—	—	0.02	n.d.
Fe ₂ O ₃	1.29	2.88	0.48	0.90	0.88
FeO	0.08	0.16	—	0.81	0.69
MnO	0.04	0.05	—	0.04	nil
NiO	—	—	—	0.16	n.d.
MgO	41.39	41.06	40.27	41.00	41.90
CaO	trace	nil	0.03	0.05	0.04
Na ₂ O	—	—	—	0.01	0.05
K ₂ O	—	—	—	0.03	0.02
H ₂ O+	13.66	13.10	12.80	12.18	12.29
H ₂ O-	1.57	1.12	—	0.08	0.04
	100.18	100.12	100.36	99.92	100.19

Sources of data

Analyst	Reference
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