

# THE MINERALOGICAL MAGAZINE

AND

## JOURNAL OF THE MINERALOGICAL SOCIETY

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No. 233

June, 1956

Vol. XXXI

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*The characterization of serpentine minerals by  
X-ray diffraction.*

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[Read 3 November 1955.]

*Summary.*—X-ray diffraction data on serpentine minerals are discussed. There are three structural varieties of chrysotile: ortho-chrysotile, clinò-chrysotile, and para-chrysotile. Methods are described for the estimation of the proportions of the first two of these in mixed specimens, and their distribution in nature is reviewed. The variations observed in powder photographs of serpentine minerals are interpreted in accordance with this classification. The criteria proposed by Selfridge for distinguishing between chrysotile and antigorite are shown to be unreliable, but valid criteria are presented. The powder photographs also reveal the existence of a third serpentine mineral, distinct from chrysotile and antigorite, for which the name lizardite is proposed. Four samples of bastite are shown to consist of chrysotile or lizardite rather than antigorite. All the serpentine minerals examined consist either of antigorite or of one or more of the group comprising lizardite and the chrysotile varieties.

THIS paper presents a general discussion of the X-ray diffraction data from serpentine minerals from the point of view of classification, identification, and analysis. In Part I the grounds are presented for classifying normal fibrous chrysotile into two varieties, ortho-chrysotile ( $a = 5.34 \text{ \AA.}$ ,  $b = 9.2 \text{ \AA.}$ ,  $c = 14.63 \text{ \AA.}$ ) and clino-chrysotile ( $a = 5.34 \text{ \AA.}$ ,  $b = 9.2 \text{ \AA.}$ ,  $c = 14.65 \text{ \AA.}$ ,  $\beta = 93^\circ 16'$ ), rather than into four varieties with  $c = 7.3 \text{ \AA.}$  as proposed by Jagodzinski and Kunze (1954). Methods are described for quantitative determination of the

proportions of these varieties in a fibre specimen from X-ray fibre photographs, and the effects to be expected in powder photographs are calculated. The distribution in nature of ortho- and clino-chrysotile is discussed, and the name para-chrysotile is proposed for the further constituent of many chrysotile specimens which has a 9.2-Å. fibre axis. In Part II the variations observed in powder photographs of serpentine minerals are discussed in the light of the results presented in Part I, and the existence of a third distinct variety of serpentine mineral (approximate cell dimensions 5.3 Å., 9.2 Å., 7.3 Å.,  $\beta = 90^\circ$ ) is established. The name lizardite is proposed for this mineral. It is shown that the previously proposed X-ray method (Selfridge, 1936) for distinguishing between chrysotile and antigorite is unreliable, but that more accurate work permits the adoption of valid criteria for this distinction. Data from four samples of bastite are presented from which it is concluded that they are forms of chrysotile or lizardite rather than of antigorite. It is apparent from the powder data that all the serpentine minerals which have been examined consist of either antigorite, or of ortho- or clino-chrysotile, or lizardite, or of mixtures of two or all of the last three, but mixtures of antigorite with either chrysotile or lizardite have not been observed.

#### PART I (E. J. W. W.).

##### DESCRIPTION AND RECOGNITION OF CHRYSOTILE VARIETIES.

A complete interpretation of the diffraction pattern of chrysotile is complicated and can only be made in terms of the theory of diffraction by cylindrical lattices (Whittaker, 1954; 1955 *a, b, c, d*). The appearance of a fibre photograph of chrysotile is, however, unusually simple owing to the small number of reflections and their division into two easily distinguishable types, sharp and diffuse. The sharp reflections are confined to the even-order layer-lines. On the zero layer-line they form an invariable series which may be indexed as  $00l$  reflections from a structure with  $c \sin \beta = 7.31$  Å. or a multiple thereof.<sup>1</sup> On the other even-order layer-lines they form two series with constant relative intensities within each series, but the intensities of the two series relative to one another are infinitely variable, so that either one or the other may be entirely absent. These two series lie at the positions to be expected for  $h0l$  reflections from an orthorhombic structure (i) with  $c = 14.63$  Å. (ortho-chrysotile), and in pairs centred on these positions and separated by the

<sup>1</sup> In this paper the axial convention is that with  $a$  as the fibre axis and  $c$  as the inter-layer axis, in order to facilitate comparison with antigorite in Part II.

distance appropriate to  $h0l$  and  $h0\bar{l}$  reflections from a monoclinic structure (ii) with  $c \sin \beta = 14.63 \text{ \AA}$ . and  $c \cos \beta = -0.835 \text{ \AA}$ . (clino-chrysotile). On the 2nd layer-line the pattern is particularly simple because the reflections from structure (ii) with  $l$  odd are very weak, and very few of them can be detected even on a well-exposed photograph. Also, the spacing between the  $h0l$  and  $h0\bar{l}$  spots of a pair is small compared with the distance between such pairs. It is therefore easy to index the reflections and assign them to the appropriate lattice type by inspection.

Basing their argument on the appearance of the 2nd layer-line, Jagodzinski and Kunze (1954) have sought to interpret the pattern in terms of four structures each with a single-layer cell, i.e.  $c \sin \beta \simeq 7.3 \text{ \AA}$ . On this interpretation the reflections from the structures (i) and (ii) above, which have even values of  $l$ , are assigned to structures differing from those proposed here only by a halving of the  $c$ -axis. Those of series (i) with  $l$  odd are assigned to a monoclinic structure (iii) with  $c \cos \beta = a/4$  so that the reflection  $2.0.l-1$  coincides with  $20l$ ; and those of series (ii) with  $l$  odd are assigned to another monoclinic structure (iv) with  $c \cos \beta = a/6$ . However, these assignments are found to be impossible as soon as even layer-lines of order higher than the 2nd are examined. For example, on the 4th layer-line the reflections from structure (iii) would coincide with those from the single-layer structure of type (i) and those from structure (iv) would coincide with those from the single-layer structure of type (ii). In fact this does not occur, and the 4th and all the even layer-lines up to the 10th can be indexed on the basis of the two two-layer structures (i) and (ii).

*Determination of the ratio of ortho-chrysotile to clino-chrysotile.*

Specimens of substantially pure clino-chrysotile are of frequent occurrence, but only one specimen of substantially pure ortho-chrysotile fibre has so far been observed. This is a very small specimen of badly crimped fibre (1) from Silesia<sup>1</sup> (Bristol Museum specimen 324, locality unknown) and is not suitable for producing good fibre photographs. It is therefore impossible to prepare a calibrated series of fibre photographs of synthetic mixtures of the two varieties, apart from the experimental difficulty of preparing such mixtures in a well-oriented form. It is possible, however, to determine the ratio of the two varieties present in a specimen by using the intensities of the zero layer-line reflections as an internal standard. It is an empirical fact that the relative intensities of these reflections are

<sup>1</sup> The numerals in parentheses refer to the list of specimens studied, p. 125.

identical for both varieties, and it follows from their structures (Whittaker, 1953) that their absolute values must also be the same.

The method is as follows. The intensities of the zero and 2nd layer-line reflections from (2), a substantially pure specimen of clino-chrysotile (Bell Mine, Thetford, Quebec), were determined by micro-densitometry of fibre photographs taken by the semi-integrating method (Whittaker, 1952) with the multiple-film technique. Similar measurements were made of the reflections from (3), a specimen rich in ortho-chrysotile from Cuddapah, India. The intensities of the zero layer-line reflections from the two photographs were then put on to the same arbitrary scale. The ratio of the intensities of the 2nd layer-line clino-chrysotile reflections on the two photographs then gave the proportion of this variety in the Cuddapah specimen, and also permitted the intensities of the ortho-chrysotile reflections to be put on to the same scale.

The above method, which involves putting the intensities of reflections on at least two photographs on to the same scale, is necessary for the primary determination of the proportions of ortho- and clino-chrysotile. But because it permits the determination of the relative intensities of the reflections from the two varieties once and for all, it is possible subsequently to use these data in a much more convenient method for determining the proportions in other specimens, as the intensities of the reflections from each variety provide an internally calibrated scale against which those from the other may be determined. For suppose that on the photograph of a mixed specimen of clino- and ortho-chrysotile it is observed that some particular clino-chrysotile reflection is equal in intensity to some particular ortho-chrysotile reflection. Let the intensities of these reflections from specimens of the pure varieties (on the same arbitrary scale as discussed above) be  $I_{cl}$  and  $I_{or}$  respectively. Then the proportion of ortho-chrysotile in the mixed specimen is given by  $I_{cl}/(I_{or} + I_{cl})$ .

In practice there may not be two such reflections with equal intensities, but several inequalities may be established in the same way to give the proportion to within reasonably narrow limits. The method is particularly convenient in that the determination may be made by mere inspection of the photograph provided that the same radiation and orientation conditions are used each time so that the Lorentz and polarization factors are constant for each particular reflection. If the specimen is of small diameter and either cylindrical or rotated, the absorption factor varies so little among the reflections used that the specimen diameter is unimportant. Table I gives the appropriate values of  $I_{or}$  and  $I_{cl}$  for the

principal reflections on the 2nd layer-line of a normal beam fibre photograph taken with filtered Cu- $K\alpha$  radiation. Appropriate pairs of these reflections provide equalities for all proportions of ortho-chrysotile to within the nearest 3 % from 0 to 95 %. This accuracy is better than is justified by the data except at the extreme upper and lower ends of the range, where equalities corresponding to 1, 2, 3, 7, 10, 90, 91, 95, 96, and 97 % ortho-chrysotile can be found.

TABLE I. Uncorrected observed intensities of  $20l$  reflections as-observed on normal beam fibre photographs with Cu- $K\alpha$  radiation.

Ortho-chrysotile.		Clino-chrysotile.	
Reflection.	Intensity.	Reflection.	Intensity.
202	1910	202	1420
203	275	204	53
204	70	204	250
205	132	206	22
206	61	206	208
207	159	208	101
208	41	208	36
209	56		

Since it has been shown (Whittaker, 1953) that the difference between the two varieties lies in the different stacking arrangement of otherwise similar layers, it might be expected that stacking faults would occur leading to disordered intergrowths of the two varieties. This has been observed in three specimens from Kranskop, Natal (4, 5, and 6), and in a specimen from the Premier Mine, Transvaal (7), where it is evidenced by  $h0l$  reflections that are very much more diffuse than the  $00l$  reflections. In extreme cases the  $h0l$  reflections may almost disappear by merging together into a continuous layer-line. Under these circumstances the determination of the proportions of the two varieties present becomes very inaccurate. However, the specimens used for the primary determination, and the vast majority of other specimens examined, do not show this intergrowth phenomenon to an appreciable extent. In specimen (2) there is not more than 2 % ortho-chrysotile so that any considerable degree of intergrowth is obviously unlikely and the uncorrected integral breadths of the  $00l$  and  $h0l$  reflections are 0.011 and 0.020. In (3) the corresponding values are 0.013 and 0.019, so that the phenomenon is certainly no more extensive in this material. The difference between the breadths of the  $00l$  and  $h0l$  reflections is presumed to be due partly to instrumental factors, though it would be very sensitive to the presence of even a small number of layers in the 'wrong' stacking arrangement.

*Calculated powder patterns.*

The determination of the proportions of clino- and ortho-chrysotile in a powder specimen is more difficult than in an oriented fibre specimen owing to the overlapping of reflections of the various types  $00l$ ,  $h0l$ , and  $hk0$ . It would be possible to produce a series of calibration photographs from mixtures of powdered material prepared from specimens previously examined in the oriented state, but a microtome is necessary to produce satisfactory chrysotile 'powder' from long fibres, and the scarcity of good specimens of high ortho-content is a further difficulty. Theoretical microdensitometer curves for powder photographs taken with filtered  $\text{Cu-K}\alpha$  radiation have therefore been constructed from the data obtained from fibre photographs and are shown in fig. 1.

In this figure and throughout the remainder of the paper it is convenient to adopt a notation to distinguish between the indices of three types of reflections. Those which occur only if  $\beta = 90^\circ$  are followed by †, those which occur only if  $\beta \approx 93^\circ 16'$  are followed by \*, and those which are unaffected (or nearly so) by the value of  $\beta$  are unqualified.

In the construction of the curves of fig. 1 the values of  $|F|^2$  have been multiplied by the various factors involved in such a photograph, and the breadths of the reflections have been made to conform approximately to those commonly obtained. The curves are drawn for the range from  $\theta = 16^\circ$  to  $\theta = 36^\circ$  only. At lower angles the only reflections which occur are  $002$ ,  $020+110$ , and  $004$ , which are identical for the two varieties, and at higher angles the lines become very weak.

This region of the photograph is dominated by the  $130$  reflection with its intense tail, on which a number of the sharp reflections are superimposed. When such a photograph is examined visually, however, this feature is largely subtracted, subconsciously, as background. The resolution of the  $130$  peak from the following  $20\bar{2}^*$  reflection is critically dependent on the reflection breadths and on the detailed profile of the  $130$  tail immediately following the peak. As this differs appreciably from one specimen to another (probably as a function of fibril diameter distribution) another variable is introduced in addition to the variations in ortho-chrysotile content. This may result in the  $20\bar{2}^*$  reflection being resolved from  $130$ , merged with it into an unresolved intensity plateau, or even disappearing altogether, leaving the  $130$  reflection alone. These phenomena have all been observed in actual powder photographs.

As the region of the powder photograph involved in the method proposed by Selfridge (1936) for distinguishing between chrysotile and anti-

gorite is the same as the region that is sensitive to the proportion of ortho-chrysotile present, Selfridge's method is vitiated. A mixed ortho- and clino-chrysotile specimen can easily simulate an antigorite specimen according to this method, although the distinction can be made satisfactorily by the method described in Part II of this paper.

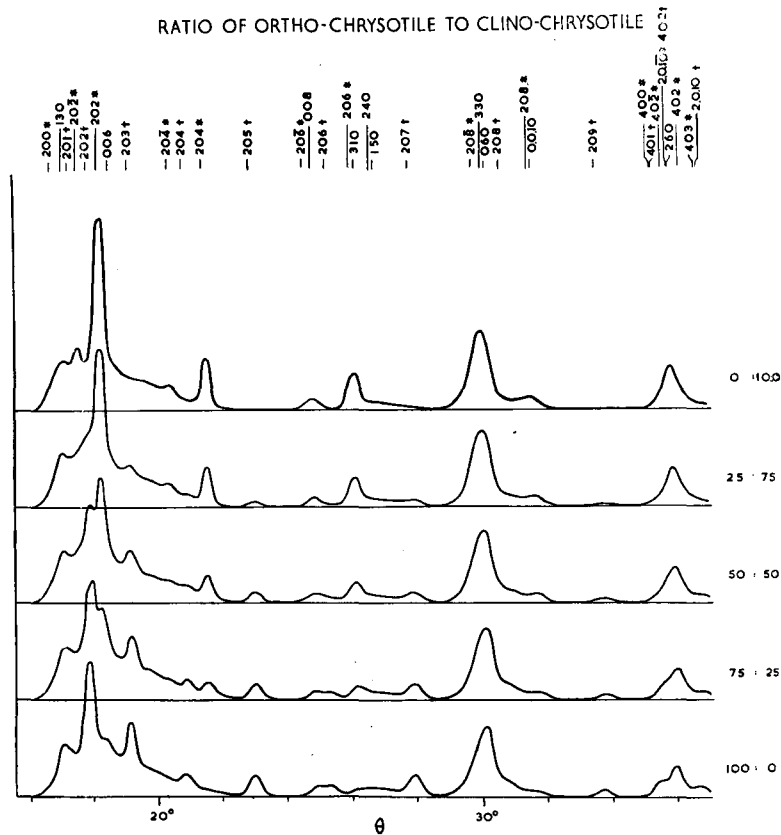


FIG. 1. Theoretical microdensitometer curves for powder photographs of mixtures of ortho- and clino-chrysotile taken with Cu- $K\alpha$  radiation.

*The distribution of ortho-chrysotile.*

Oriented fibre specimens containing a mixture of ortho- and clino-chrysotile have been shown to contain the same proportions of the two varieties at different points along their length and also in different parts of their cross-section. The latter result was obtained by taking a series of

fibre photographs after successively halving the cross-section of a specimen down to a diameter of 0.05 mm. This shows that if the fibrils of the two varieties have any tendency to segregate into homogeneous bundles, these bundles must be less than about  $10^{-3}$  cm. in diameter.

In an attempt to find some correlation between the proportion of orthochrysotile and the local geological or mineralogical conditions this proportion was determined in a series of specimens taken from known positions in the Shabani Mine, Southern Rhodesia, but no correlation

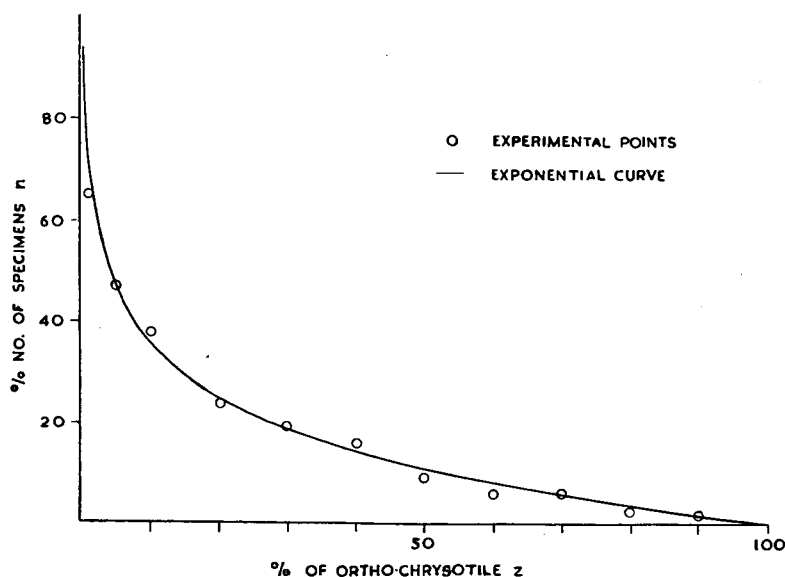


FIG. 2. Cumulative frequency curve for the occurrence of different percentages of ortho-chrysotile in admixture with clino-chrysotile.

with position was found. It was also observed that abrupt changes in proportion may occur. In one particular hand-specimen from Shabani the proportion of ortho-chrysotile was found to change abruptly from approximately zero to about 15% across an invisible boundary parallel to the fibres.

Rather more than a hundred oriented fibre specimens from many localities have been analysed for ortho-chrysotile content. From many localities only one specimen has been examined, but appreciable numbers have been examined from Thetford (Bell Mine), Shabani (170 Section), and Cuddapah. Substantially pure clino-chrysotile occurs in each of these localities, but the range of ortho-chrysotile contents varies markedly



from one locality to another. Thus the maximum values observed are Thetford 7 %, Shabani 35 %, and Cuddapah 81 %. It is also found that if all the specimens investigated are grouped together without distinction of locality, then the percentage number of specimens ( $n$ ) containing more than  $z$  % ortho-chrysotile is given by

$$z = 100e^{-0.065n}.$$

This is demonstrated in fig. 2. This procedure is certainly not beyond reproach, since there are very significant differences between the distributions from the different sources, and no attempt has been made to obtain a distribution representative of the world as a whole. However, as the relationship is so striking, and there is a tendency for the ortho-chrysotile contents of specimens from the individual sources to show similar distributions (albeit less perfectly and with different constants), it seems worth while to draw attention to it. No satisfactory explanation of the relationship has so far been found. It might presumably be related to energy differences between the two varieties during nucleation or during growth, or possibly to interconversion.

*The occurrence of a variety of chrysotile with a 9.2-Å. fibre axis.*

It has been reported by various workers (Aruja, 1943; Hargreaves and Taylor, 1946) that X-ray fibre photographs of chrysotile show weak extra layer-lines indicating a fibre axis of about 9.2 Å. Aruja showed that the reflections on the first three of these layer-lines correspond approximately to the stronger diffuse reflections of chrysotile if they are assumed to arise from material oriented with either the [010] or [310] axis parallel to the fibre direction. He also estimated the amount of such material in a specimen of Thetford chrysotile as about 3 %. Hargreaves and Taylor showed that this material is not present in all specimens of chrysotile, and may be removed by prolonged extraction with water.

These results have now been extended. The amount of the material has not been found to vary in the various Thetford specimens studied, but specimens from some Rhodesian and South African localities have been found to vary widely in this respect from zero to about 10 %. Specimens from Cuddapah on the other hand are found to be uniformly free from the material.

The availability of specimens giving stronger extra layer-lines has permitted the recording of a greater number of reflections, and the range of the reflections has been extended by obtaining photographs of the 2nd, 3rd, and 6th of these layer-lines in the equi-inclination position. It is

confirmed that the diffuse reflections may be indexed in accordance with Aruja's proposals, and it is shown that sharp reflections also occur, but only on the 6th layer-line. These sharp reflections characterize the material as having an orthorhombic cylindrical lattice structure (Whittaker, 1955c) with a cell dimension perpendicular to the layers of 14.6 Å., and disprove the suggestion of Hargreaves and Taylor that it might consist of layers stacked in a less orderly manner than in chrysotile. The detailed considerations leading to this structure assignment will be presented elsewhere, but it may be remarked here that the structure is very closely analogous to that of ortho-chrysotile apart from the orientation of the 9.2-Å. axis, instead of the 5.3-Å. axis, along the cylinder axis. This material must therefore clearly be regarded as chrysotile, not antigorite, notwithstanding the fact that Zussman (1953) has shown previously that fibrous serpentines commonly show an association between the 9.2-Å. fibre axis and antigoritic character. The classification of the serpentine minerals which is presented in Part II of this paper makes allowance for this exception to the general rule.

It is proposed that the variety of chrysotile with a 9.2-Å. fibre axis be described as para-chrysotile (para = alongside of; amiss).

#### PART II (J. Z.).

In this part powder data obtained from serpentine minerals are presented. Detail of the photographs is interpreted, as far as possible, in the light of the data given in Part I, and analogies and distinctions between different structural types are noted. All patterns were recorded with a 19-cm. camera using Ni-filtered radiation from a Cu target, and intensities were estimated by eye. The powder was in each case loaded into a thin-walled glass capillary of diameter 0.2 mm.

Since all except the antigorites (section 3 of the list of specimens, p. 125) possess closely related unit cells, their interplanar spacings may be listed conveniently in one table (table II). A few weak lines obtained from some specimens have been omitted since they were identified as deriving from an impurity (calcite, talc, magnesite, or magnetite). Specimens are referred to by number, and a key is provided which gives locality of origin and, where relevant, a collection number for each.

#### *Ortho- and clino-chrysotile.*

The powder photographs of the specimens in section 1 of the list of specimens (p. 125) show them to be most like those already dealt with in Part I, for they may be indexed on the basis of a cell with  $a = 5.32$ ,

TABLE II. Chrysotile and lizardite. Here are listed the  $d$  values in Å. and the intensities for individual, or average values for groups of similar, specimens. Column I gives the indices of the reflections, and the last column gives calculated spacings using the cell dimensions listed on page 119. The meanings of symbols following indices are explained on page 112.

Specn. <i>hkl</i> .	8 & 9.	10.	11.	14.	15 & 16.	17, 18, 19, and 20.	22.	23.	$d_{calc}$ .
002	7.36 vs	7.37 vs	7.36 vs	7.36 vs	7.35 vs	7.35 vs	7.36 vs	7.37 vs	7.32
020	4.38 m	4.58 m	4.56 m	4.62 ms	4.61 m	4.60 m	4.58 m	4.59 m	4.60
022	—	—	—	3.89 w	3.90 mw	—	—	—	3.89
004	3.66 vs	3.66 vs	3.66 s.	3.64 vs	3.66 vs	3.66 vs	3.66 vs	3.66 vs	3.66
024	—	—	—	—	2.85 w	—	—	—	2.86
130	2.66 mw	2.66 mw	2.66 w	2.65 mw	2.66 mw	2.66 w	—	2.66 mw	2.66
201†	—	2.604 mw	2.604 mw	—	—	—	2.617 w	2.614 vw	2.614
201°	2.594 mw	—	—	—	—	—	—	—	2.590
202°	2.549 m	2.548 mw	2.547 vw	—	—	—	—	—	2.548
202†	—	2.500 ms	2.500 m	2.495 s	2.499 s	2.500 s	2.498 s	2.500 s	2.497
202°	2.456 s	2.455 s	2.451 ms	—	2.446 m	—	—	2.450 m	2.454
006	—	—	—	—	—	2.437 mw	—	—	2.438
203†	—	2.338 vw	—	—	—	—	2.340 mw	2.340 vw	2.332
040	—	2.289 vw	2.285 vw	2.299 w	—	2.290 vvw	—	—	2.301
203°	2.282 w	—	—	—	—	—	—	—	2.280
204°	2.215 w	2.211 vw	2.212 vw	—	—	—	—	—	2.214
204†	—	2.152 mw	—	2.148 m	2.150 ms	2.151 ms	2.150 mw	2.147 m	2.148
204°	2.096 m	2.094 m	2.093 mw	—	2.092 w	—	—	2.096 vvw	2.094
205†	—	—	1.972 vw	—	—	—	1.968 w	—	1.966
008	1.829 w	1.830 vw	1.828 w	1.830 w	1.826 w	1.830 w	1.830 w	1.826 w	1.829
206†	—	1.798 w	—	1.794 mw	1.795 m	1.796 m	1.797 w	1.795 mw	1.796
206°	1.748 m	1.747 mw	1.746 mw	—	—	—	—	—	1.748
310	—	—	—	1.737 mw	1.740 w	1.740 w	1.739 mw	1.739 mw	1.739
028	—	—	—	1.695 vw	1.699 w	1.696 vw	—	—	1.698
207†	—	—	—	—	—	—	1.640 w	—	1.643
060	1.536 s	1.531 s	1.531 ms	1.534 ms	1.535 s	1.535 s	1.534 s	1.537s	1.534
208†	—	—	—	1.503 m	1.503 ms	1.504 ms	1.504 m	1.504 m	1.506
0.0.10	1.465 w	1.465 vw	1.464 w	1.460 vw	1.462 w	1.463 w	1.464 w	1.462 vw	1.463
064	—	—	—	1.415 w	1.415 w	1.416 w	—	—	1.414
400	—	—	—	1.328 w	1.328 w	—	1.326 w	—	1.328
402°	1.317 mw	1.321 mw	1.319 mw	—	1.319 vw	—	—	—	1.322
402†	—	1.309 mw	1.310 mw	1.307 m	1.308 m	1.308 m	1.308 m	1.310 m	1.307
2.0.10†	—	—	—	1.279 w	1.282 mw	1.281 vw	—	—	1.281
404†	—	—	—	1.252 vw	1.250 vw	1.247 vw	—	—	1.248
0.0.12	—	—	—	1.218 vw	1.217 vvw	1.218 vvw	—	—	1.218
406†	—	—	—	1.167 vw	1.168 vw	—	—	—	1.167
408†	—	—	—	1.073 vvw	1.074 vvw	—	—	—	1.074
510	—	—	—	1.056 vvw	1.058 vvw	—	—	—	1.056
531†	—	—	—	0.994 w	0.995 w	—	—	—	0.998

$b = 9.2$ ,  $c = 14.64$  Å., and  $\beta = 93^\circ 20'$  or  $\beta = 90^\circ$ , and are seen to be clino-chrysotile or mixtures of clino- and ortho-chrysotile.

Specimens (8) and (9) give the pattern of pure clino-chrysotile, while (11) is evidently a mixture of clino- and ortho-chrysotile. It is a very well-oriented silky asbestos similar to those examined by Whittaker and its fibre photograph shows approximately 16% of ortho-chrysotile to be present. (10) shows prominently the pattern of clino-chrysotile, but some

additional weak lines indicate the presence of another species (see next section). (12) gives rather diffuse and weaker lines on a slightly higher background but is recognizable as mainly clino-chrysotile. Specimen (13) has not been examined by the writer, but a diffractometer chart recording of its powder lines, made available by Dr. G. H. Francis, showed it to be a mixture of clino- and ortho-chrysotile.

For all specimens in this group reflections of the type  $Ok\bar{l}$  and  $hkl$  are absent (in (12) the  $hOl$  reflections are very weak). They may be presumed, therefore, to be structurally similar to chrysotile asbestos although their texture is generally splintery rather than silky. Many of them show at least a moderate degree of fibre orientation with  $a$  as fibre axis.  $201^*$  is the only clino-chrysotile line having  $l$  odd that is resolved from other lines. It may be seen in the pure clino- varieties but may be obscured by  $201^\dagger$  when there is some ortho-chrysotile present. A notable feature on all photographs is the long 'tail' of the 130 reflection which often appears to end in a sharp edge coinciding with the reflections  $202^*$  and 006. The shape of this tail and its effects on the pattern's appearance have been discussed in Part I. Furthermore, if the position of 060 is taken to define the  $b$  parameter it is seen that the 040 and 020 reflections are displaced to higher angles (Aruja, 1943; Whittaker, 1955a).

#### *Lizardite.*

The powder patterns of specimens in section 2 of the list of specimens (p. 125) may be indexed on the basis of a cell with  $a = 5.31$ ,  $b = 9.2$ ,  $c = 7.31 \text{ \AA}$ ,  $\beta = 90^\circ$ , but for ease of comparison with data from section 1, the indices given refer to a cell with  $c = 14.62 \text{ \AA}$ , so that reflections with  $l$  odd do not appear.

Specimens (14) and (15) show some reflections of the type  $Ok\bar{l}$ , and from (15) a few weak lines of clino-chrysotile are observed. (16) is similar to (15) but has a greater proportion of clino-chrysotile, while (17), (18), (19), and (20) show no 'clino-' lines and their  $Ok\bar{l}$  reflections are considerably weaker, in some cases being barely visible. These observations suggest that we are dealing with a new structural species of serpentine mineral composed of the  $\text{H}_4\text{Mg}_3\text{Si}_2\text{O}_9$  sheets stacked orthogonally over one another with varying degrees of three-dimensional order. It must be recorded, however, that the observed relative intensities are not consistent with those calculated for a simple model. In the case of ortho-chrysotile the two possible sets of coordinates for Mg and (OH) ions in the octahedral section are occupied alternately in successive layers (Whittaker, 1953), whereas a single-layer cell would require that the

same set of coordinates be occupied in every layer. The observed intensities from specimens of section 2 correspond more closely to those calculated from the former model, and it is probable that in these cases the two alternative sets of positions are occupied randomly in successive layers.

For specimens (22) and (23) the lines may all be indexed only if an ortho-cell with  $c = 14.6 \text{ \AA}$ . is assumed. They might appear, therefore, to be mainly ortho-chrysotile, but this is refuted by the relative intensities of the  $20l\ddagger$  lines. Table I shows that for ortho-chrysotile those with  $l = 3, 5,$  and  $7$  are stronger than those with  $l = 4, 6,$  and  $8,$  whereas here the even index lines are the stronger. This seems to indicate the presence of the single-layer serpentine mixed with a smaller amount of ortho-chrysotile. (22) would appear to have a greater proportion of ortho-chrysotile since its reflections with  $l$  odd are relatively stronger than corresponding ones from (23). From neither of these specimens are  $Ok\ddagger$  reflections observed. The intensities of 'ortho'-cell reflections with  $l$  odd as compared with those having  $l$  even were also significant for specimens (10) and (11). Both are predominantly clino-chrysotile, but for specimen (11)  $203\ddagger$  and  $205\ddagger$  are visible while  $204\ddagger, 206\ddagger,$  and  $208\ddagger$  are not. For specimen (10)  $204\ddagger, 206\ddagger,$  and  $208\ddagger$  are visible while  $203\ddagger$  and  $205\ddagger$  are not. Thus (10) is a mixture of clino-chrysotile with the single-layer serpentine and not with ortho-chrysotile.

It is of interest to note the consistency of  $d$  values shown in all photographs dealt with so far, particularly when specimens which have a cell with  $\beta = 90^\circ$  are considered separately from those which do not. This is so in spite of widely different sources of material, and suggests that at least some of the variation in corresponding spacings hitherto reported may be due to differences in experimental conditions. In the present work all specimens were examined under identical conditions and the variations in observed  $d$  values are generally within the limits of the estimated probable error ( $\pm 0.1 \%$  for lines at  $\theta \approx 30^\circ$ ).

The average of uncorrected  $d$  values for specimens in each of the above categories correspond to cell dimensions as follows:

$$\begin{array}{llll} \beta = 90^\circ & a = 5.31 & b = 9.20 & c = 7.31 \text{ (or } 14.62) \text{ \AA.} \\ \beta = 93^\circ 20' & a \sin \beta = 5.32 & b = 9.20 & c \sin \beta = 14.64 \text{ \AA.,} \end{array}$$

and these parameters were used to derive the ' $d_{\text{calc}}$ ' list shown in table II.

Specimens (15), (17), (18), and (22) can be described as 'massive serpentine and indeed are so fine-grained and unoriented that a stationary

specimen gives powder lines with no trace of spottiness. (16) and (23) show some orientation with fibre axis 5.3 Å. Specimen (14), however, is completely different in being white, not green, and separable into small platy crystals. It is the variety examined by Midgley (1951), who concluded that it was most like antigorite. This does not appear to be the case, and a more detailed investigation of the mineral is in progress.

#### *Antigorite.*

This section relates to those specimens whose lines cannot be indexed on the basis of the above cells or mixtures of them but which possess the large unit cell described below. Specimens (24) to (28) inclusive are practically monomineralic rock specimens from which small, platy single crystals may be obtained. Optical properties have been measured for three of these specimens (Hess, Smith, and Dengo, 1952; Zussman, 1954; Francis, 1956) and are seen to be closely similar. The data used for specimen (25) were presented by Hess *et al.*, while (26) (antigorite from Miconui, New Zealand) has already been studied in some detail and was seen to have cell dimensions:  $a = 43.5$ ,  $b = 9.26$ ,  $c = 7.28$  Å.,  $\beta = 91^\circ 24'$  (Aruja, 1944). Since the powder patterns of all five specimens are almost identical, it is safe to assume that this large cell is characteristic of all antigorites and not merely an exceptional variant, as may have been supposed. Table III contains average spacings, although some lines were not observed in all five patterns. The numerical intensity estimates are those measured by diffractometer for (25) by Hess, Smith, and Dengo, and these are approximately paralleled by the photographic visual estimates, with a few notable exceptions that may arise through orientation effects in the powder specimens.

Specimens (29) and (30) are best described as splintery with fibre axis 9.25 Å. Their powder photographs show fewer lines, which tend to be broader, but they correspond in each case with lines from the previous antigorite specimens, as can be seen in table III. It is assumed, therefore, that they possess the same basic cell and structure but are of smaller grain size and are less well ordered. (31) gives a photograph intermediate in quality between those from (28) and (30).

Indexing of the antigorite patterns was carried out with the aid of single-crystal Weissenberg and oscillation photographs, and the cell parameters corresponding to average uncorrected  $d$  values are as follows:  $a = 43.5$ ,  $b = 9.25$ ,  $c = 7.26$  Å.,  $\beta = 91^\circ 23'$  (cf. those by Aruja). Eight reflections of the type  $h0l$  and  $h\bar{0}l$  were used to determine  $\beta$ .

TABLE III. Antigorite. Column 1 gives intensities recorded by Hess *et al.* (1952, p. 70). Column 2 gives  $d$  values in Å. obtained by combining data from photographs of specimens (24), (26), (27), and (28) with those given by Hess *et al.* for (25). The average  $d$  value is recorded for each reflection, although some were not observed in all five cases. Those without a visually estimated intensity were observed only from (25). Column 3 lists  $d$  values calculated for a cell with dimensions shown on page 120, and column 4 the indices of the reflections. Where indices are given for more than one reflection ' $d_{\text{calc}}$ ' corresponds to the first mentioned. Columns 5 and 6 give observed  $d$  values and intensities for specimens (29) and (30).

$I$ .	$d_{\text{obs.}}$	$d_{\text{calc.}}$	$hkl$ .	$d_{\text{obs.}}$ (29).	$d_{\text{obs.}}$ (30).
400	7.30 vs	7.26	001	7.28 vs	7.30 vs
24	6.95 —	6.93	20 $\bar{1}$	—	—
16	6.52 w	6.56	30 $\bar{1}$	6.40 w	—
8	5.80 w	5.96	401	—	—
3	5.11 vvw	5.15	710	—	—
6	4.67 mw	4.68	810	—	—
7	4.64 mw	4.63	020	4.64 mw	4.66 m
4	4.27 w	4.28	910	4.24 w	4.25 w
6	3.95 mw	3.95	81 $\bar{1}$	—	—
300	3.63 s+	3.63	102, 10 $\bar{2}$	3.61 vs	—
24	3.52 vvw	3.50	302, 202	—	—
2	2.88 —	2.88	14.0. $\bar{1}$	—	—
3	2.66 vvw	2.66	15.0.1	—	2.67 vvw
4	2.60 vw	2.60	930	2.60 w	—
8	2.57 mw	2.55	17.0.0, 16.0. $\bar{1}$	—	—
70	2.53 vs	2.52	16.0.1	2.53 vs	2.53 ms
9	2.46 m	2.46	93 $\bar{1}$	2.45 w	—
38	2.42 mw	2.42	003, 18.0.0	2.42 w	2.42 w
9	2.39 w	2.39	17.0.1, 30 $\bar{3}$ , 10.3. $\bar{1}$	2.38 vw	—
5	2.35 mw	2.35	403?	—	—
6	2.24 mw	2.24	15.0.2	2.25 vw	—
7	2.21 mw	2.20	16.0. $\bar{2}$	2.21 w	—
22	2.169 ms	2.169	83 $\bar{2}$	2.163 m	—
20	2.153 m	2.150	16.0.2	—	—
4	2.127 w	2.126	93 $\bar{2}$	—	—
4	2.113 w	2.113	17.0. $\bar{2}$	—	—
4	2.035 vw	2.033	11.3. $\bar{2}$	—	2.03 mw
3	1.879 w	1.879	15.0. $\bar{3}$	—	—
12	1.832 m	1.836	15.0.3	1.835 w	—
23	1.813 w	1.815	004, 10 $\bar{4}$ , 833	1.813 w	1.819 mw
14	1.782 m	1.782	93 $\bar{3}$	1.779 m	—
4	1.755 w	1.756	10.3. $\bar{3}$	—	—
10	1.738 mw	1.737	17.0.3	1.732 mw	—
2	1.680 vw	1.680	21.3.1	—	—
2	1.638 vw	1.636	22.3.1	—	—
3	1.587 w	1.583	14.0. $\bar{4}$	—	—
12	1.563 m+	1.561	24.3.0	1.566 ms	1.561 m
9	1.541 m	1.541	060	1.541 ms	1.541 m
9	1.534 m	1.531	24.3. $\bar{1}$	—	—
13	1.523 m	1.522	15.0.4, 16.0. $\bar{4}$ , 22.3. $\bar{2}$	1.525 w	1.528 w
8	1.509 mw	1.508	061	1.509 w	1.508 mw
10	1.497 m—	1.495	17.0. $\bar{4}$ , 93 $\bar{4}$	1.494 w	—
7	1.480 w	1.479	934	1.477 vw	—
6	1.468 w	1.467	18.0. $\bar{4}$	—	—
6	1.462 —	1.462	10.3.4	—	—
10	1.452 w	1.451	20 $\bar{5}$	1.449 vw	1.453 w
9	1.448 mw	1.446	205	—	—

*Nomenclature.*

In view of X-ray studies in recent years it would be desirable to reconsider the nomenclature of the serpentine minerals. It is clear firstly that only those should be called antigorite that possess the large cell and give the specific pattern described above. If any other name is to be used, perhaps picrolite could be retained to describe those antigorites that are distinctly fibrous or splintery. Among other serpentine minerals the names ortho-, clino-, and para-chrysotile can be applied as defined by Whittaker. These have structures based upon cylindrical layers; they have  $c = 14.6 \text{ \AA}$ . and do not show three-dimensional order. There remains the orthogonal variety with  $c = 7.3 \text{ \AA}$ ., which can possess varying degrees of three-dimensional order. Unfortunately none of the previously used names (e.g. marmolite, metaxite, schweizerite) has been applied consistently to specimens of the latter type, but very many of them have been called 'serpentine'. Selfridge (1936) proposed that a third sub-group of the serpentine minerals should be referred to as 'serpentine', but use of this name for the whole and also for a particular part of the group has obvious disadvantages. It appears, therefore, that a new name is desirable by means of which specimens possessing the single-layer ortho-cell can be distinguished from chrysotile and from antigorite.<sup>1</sup> Since a well-known locality for serpentine minerals is the Lizard area of Cornwall, England, and since specimens of the sub-group in question are known to occur there (e.g. (14) and (22)), the name lizardite would seem appropriate.

Thus we have:

$$\text{serpentine} \left\{ \begin{array}{l} \text{chrysotile} \\ \text{lizardite} \\ \text{antigorite} \end{array} \right. \left\{ \begin{array}{l} \text{ortho-chrysotile} \\ \text{clino-chrysotile} \\ \text{para-chrysotile} \end{array} \right.$$

Most of the light green or yellowish-green massive rock specimens examined, which would commonly be called serpentine, have in fact turned out to belong to the lizardite sub-group according to the above classification. Most of the splintery specimens that have any degree of ordering about the  $a$ -axis belong to the chrysotile sub-group, a few to lizardite, and none to antigorite. All of the splintery ones with  $b$  as fibre axis are seen to be antigorite, with the exception of para-chrysotile,

<sup>1</sup> It does not seem wise to add to the already large number of 'modified chrysotile' names ortho-, clino-, para-,  $\alpha$ -,  $\beta$ -,  $\gamma$ -.



which has only been observed as intergrowths with ortho- and clinochrysotile. Generally, the non-fibrous antigorites are more coarsely grained than other varieties and are blue-green in colour.

*Bastite.*

'Bastite' is the name commonly applied to serpentine pseudomorphs after orthopyroxene in basic and ultra-basic rocks, and it has been used as a synonym of antigorite (e.g. Winchell, 1951; Hess, Smith, and Dengo, 1952), presumably because the relict structures exhibited resemble the platy character of antigorite rather than the fibrous nature of chrysotile.

In the present study three bastite specimens were examined, two of which are of material from the type locality. They are seen, however, to give patterns of chrysotile (12) or of lizardite ((19) and (20)) and not of antigorite. Another bastite (21), for which a diffractometer record was made available by G. H. Francis, gave broad peaks spread across the chrysotile and lizardite positions and did not give those characteristic of antigorite. It seems likely, therefore, that other specimens described as bastite are similarly not antigorite, if the latter mineral is defined in the strict sense suggested in this paper.

*Comparison between powder patterns of antigorite and other serpentine minerals.*

In much published work recognition of a serpentine mineral as either antigorite or chrysotile has been made on the basis of the powder data given by Selfridge (1936) and Gruner (1937). In the former case molybdenum radiation was used giving rather poor resolution, relatively few lines were recorded, and indexing was not attempted. In the latter work iron radiation was used, but with a camera of small radius, and so, in both, no clear indication of the various chrysotile and lizardite cells can be discerned. Distinction between antigorite and chrysotile is, however, possible, although deductions using the region of the pattern near  $202\ddagger$  may be misleading when resolution is poor, since, as can be seen from fig. 1, it is just here that the most marked variations within non-antigorites occur. Indexing of both antigorite and chrysotile using one set of cell dimensions has been attempted (Gruner, 1937) and led to some false 'correspondences'.

With the better resolution of a 19-cm. camera and the improved understanding of the atomic structure of serpentine minerals in the years since 1941,<sup>1</sup> the distinction between antigorite and the remainder

<sup>1</sup> Warren and Hering (1941) suggested a 'sheet' structure for chrysotile.

becomes possible in various parts of the powder diagram, but most reliably from the pattern of lines as a whole. Some antigorite lines do occur in positions close to those from chrysotile, but if the complete pattern is observed and accurately measured then the intensity relationships, the presence of many extra lines, and the small but significant differences between apparently corresponding spacings make correct identification certain. When the specimen is less well ordered (e.g. (29)) many weak lines are hardly, if at all, visible and the pattern may look very similar to one of lizardite, but distinction is still quite definite. The strong line from antigorite at  $d = 2.52 \text{ \AA}$ . may be confused with  $202\ddagger$  of lizardite but it should be noted that in the latter varieties  $d_{202\ddagger}$  has not been observed greater than  $2.50 \text{ \AA}$ . The strong line at  $d = 1.563 \text{ \AA}$ . in antigorite is important in that it is not 060 and has no counterpart in chrysotile or lizardite patterns. With the moderately strong 060 ( $d = 1.541 \text{ \AA}$ .) it forms a prominent pair of lines which are not strictly 'equivalent' to the strong pair at  $d = 1.535 \text{ \AA}$ . and  $1.503 \text{ \AA}$ . given by lizardite. Lines on antigorite photographs, even for the more poorly crystalline varieties, are generally sharper than those from other serpentine minerals.

Among other antigorites for which data are available are those from Antigorio, Hokitika, and Griffin Range (Aruja, 1943), and one referred to as bowenite (Francis, 1956). These all have patterns similar to those discussed above. One other from Unst, Shetlands, described by Brindley and von Knorring (1954) as ortho-antigorite, appears in the light of the present work to be predominantly ortho-chrysotile. Although it shows many closely spaced weak lines which suggest a large cell parameter of  $43.8 \text{ \AA}$ . it is certainly different from antigorite (see criteria above). The lines, moreover, are not the same as those given by the specimens here examined. Some discussion of the note by Brindley and von Knorring occurs elsewhere (Zussman, 1956).

While it is believed that the scheme of classification proposed here should form a sound basis for further work, it is not claimed to be exhaustive. A complete classification of the serpentine minerals may well involve further distinctions between structural varieties which have not so far been recognized.

*Acknowledgements.*—J. Z. wishes to acknowledge the co-operation of the many people involved in making available the specimens used in this investigation, and to thank Dr. G. H. Francis for helpful discussions. E. J. W. wishes to thank the Directors of Ferodo Ltd. for permission to publish the work described in Part I.

*List of specimens studied.*

<i>No.</i>	<i>Ref.</i> <sup>1</sup>	
		1. <i>Mainly chrysotile.</i>
1	B. 324	Chrysotile. Silesia.
2	—	„ Bell's Mine, Thetford, Quebec.
3	—	„ Cuddapah, Madras, India.
4	—	„ Kranskop, Natal.
5	—	„ „ „
6	—	„ „ „
7	—	„ Premier Mine, Transvaal.
8	O. 3930	'Schweizerite.' Findelen Glacier, Zermatt, Switzerland.
9	B.M. 27612	'Metaxite.' Reichenstein, Silesia.
10	B.M. 93645	'Marmolite.' Hoboken, New Jersey, U.S.A.
11	M. 14676	Chrysotile asbestos. Cross fibre vein in (17).
12	—	'Bastite.' Port Macquarie, New South Wales.
13	C. (H.) 76140	Chrysotile asbestos. Milton, Glen Urquhart, Scotland (Francis, 1956).
		2. <i>Mainly lizardite.</i>
14	—	'Serpentine mineral.' Kennack Cove, Lizard, Cornwall (Midgley, 1951).
15	O. 17788	'Serpentine mineral.' Snarum, Norway.
16	B.M. 47963	'Marmolite.' Hoboken, New Jersey, U.S.A.
17	M. 14676	Serpentine rock in dolomite. Transvaal.
18	—	'Serpentine mineral.' Hindubagh, Baluchistan.
19	B.M. 170 Ap.	Bastite. Baste, Harz, Germany.
20	B.M. 67807	„ „ „ „
21	C. (H.) 76113	„ Polmaily, Glen Urquhart, Scotland (Francis, 1956).
22	—	Serpentine. I.C.I. Quarry, Kennack, Lizard, Cornwall.
23	O. 1899	„ New York.
		3. <i>Antigorite.</i>
24	O. 16328 <sup>2</sup>	Serpentine. New York.
25	—	Antigorite. Caracas, Venezuela (Hess, Smith, and Dengo, 1952).
26	C. 11566	Antigorite. Cross River, Mikonui, New Zealand (Aruja, 1944; Zussman, 1954).
27	C. (H.) 76095	Antigorite. Milton, Glen Urquhart, Scotland (Francis, 1956).
28	O. 16327	Serpentine mineral. Val Antigorio, Switzerland.
29	O. 16336	Picrosmine, Tyrol.
30	C. T.W. 2915	Picrolite. Taberg, Sweden.
31	C. 7236	Serpentine. Asbestos Creek, Mikonui, New Zealand.

<sup>1</sup> B. Bristol Museum; O. Oxford University Museum; B.M. British Museum (Nat. Hist.); M. Manchester University, Dept. of Geology; C. Cambridge University, Dept. of Mineralogy and Petrology; (H.) Harker Collection.

<sup>2</sup> In the paper by Zussman (1954) two errors occur in table II. O. 16328 should not have been listed at all, and O. 1899 should refer to the specimen labelled 'Serpentine (New York)'.

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