Since the structure is the complex *APD* structure of the type ($[M] \mid [\bar{M}]$), we should have

$$\omega_{(S)}^{(1)} = \omega_{(S)}^{(2)} \quad \text{and} \quad \omega_{(S)}^{(2)} = \omega_{(S)}^{(1)} . \tag{A33}$$

As a result, we have

$$\omega_{(S)} = \omega_{(S)}^{(1)} + \omega_{(S)}^{(1)} = \omega_{\overline{(S)}} \tag{A34}$$

and hence

$$\omega_{(S)} + \omega_{\overline{(S)}} = 2\{\omega_{(S)}^{(1)} + \omega_{(S)}^{(1)}\} , \tag{A35}$$

this meaning that the following relation holds in general:

$$\{\Omega_r\} = 2\{\Omega_r^*\} . \tag{A36}$$

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Study of Microstructure of Chrysotile Asbestos by High Resolution Electron Microscopy

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(*Received* 3 *February* 1971 *and in revised form* 20 *July* 1971)

(*Dedicated to Professor Tadatosi Hibi in honour o, his* 61*st birthday*)

Several samples of chrysotile asbestos from different localities, including a synthetic sample, were electron-microscopically observed by the lattice imaging method along two directions parallel and perpendicular to the fibre axis. The results are as follows: (*a*) The lattice fringes of 4·5 Å corresponding to 020 were often tilted to the edge of the fibrils with an angular distribution ranging up to about 10° with a peak value at a few degrees, depending on the sample. (*b*) Most of the fibrils examined were hollow cylinders and their circumferential lattice layers form spiral or multi-spiral layers. The perfectly concentric cylindrical layers were also found with a frequency depending on the sample. (*c*) Unusual growth patterns which cannot be explained by Jagodzinski and Kunze's model were observed. (*d*) The lattice images of the conical fibrils (cone-in-cone shape) were observed in the synthetic sample. (*e*) Most fibrils greater than about 350 Å in diameter showed traces of discontinuous growth in two or three steps, depending on the growth conditions, and this gave rise to various distributions of the fibril diameters.

Introduction

From studies using the methods of X-ray diffraction, electron microscopy and electron diffraction, it has been pointed out that there are morphological and structural variations in chrysotile asbestos (Whittaker, 1951; Jagodzinski & Kunze, 1954; Whittaker, 1955, 1956*a*,*b*,*c*, 1957; Whittaker & Zussman; 1956). In earlier X-ray studies, however, single crystals were not available, while in subsequent studies made on single crystals (individual fibrils), by means of electron microscopy combined with selected area electron diffraction, the instrumental resolution was not high enough to resolve the fine structures (Honjo & Mihama, 1954; Zussman, Brindley & Comer, 1957; Bates & Comer, 1957).

Recently, it has become possible to observe lattice planes in the individual fibrils of chrysotile (Fernández-Morán, 1966; Yada, 1967), and it has been found, for example, that the circumferential lattice images observed in the cross-section of a fibril show a spiral or multi-spiral structure. The previous work by the present author, however, was done for a sample from only one source (Quebec, Canada,) and, moreover, the observation of the detailed structure at the inter- and intra-fibril sites was hindered by the damage due to irradiation by the electron beam required for high magnification electron microscopy. Therefore, in order to obtain a comprehensive understanding of the microstructures and growth mechanism of chrysotile, it seemed desirable to study samples from different localities by the use of a improved technique by which the radiation damage was minimized.

The specimens and experimental technique

Table 1 shows a list of the samples examined. Most of these samples are chrysotile ores, except the last one which is powder Mg-chrysotile synthesized under controlled conditions (Noll, Kircher & Sybertz, 1958, 1960).

A small quantity of chrysotile was torn off with tweezers, and as in the previous work (Yada, 1967) observations were made from two directions, parallel and perpendicular to the fibre axis, employing the sectioning technique by ultramicrotomy as well as the

usual suspension technique. The sectioned specimens were cut with a diamond knife and mounted on a thin carbon supporting film. The electron microscope observations were made without dissolving the embedding plastic of the sectioned specimen in order to preserve the original state of the specimen as far as possible.

The contamination and radiation damage due to the electron beam was minimized by using an anti-contamination device to cool the vicinity of the specimen holder, and by keeping the beam intensity as low as possible. The electron optical magnification was 110000–150000 times, with a small illuminating beam of about 1μ in diameter formed by a pointed cathode. The exposure time was about 10 seconds.

Experimental results

Observation from the direction perpendicular to the fibre axis.

(*a*) *Behaviour of the* 4·5 Å *fringes corresponding to* 020.

The three kinds of lattice fringe system previously reported (Yada, 1967) were observed in most cases. They are the 4·5 Å fringes corresponding to the 020 reflexion, the 7·3 Å fringes (or those with a half-spacing depending on the diffraction condition) corresponding to 001, and the diagonally crossed two sets of 4·6 Å fringes corresponding to 110. When the incident beam was nearly perpendicular to the fibre axis, these three kinds of fringes were consistently observed in fibrils from all sources, including the synthetic one. For the sample from Coalinga, California, however, it was found that the 4·5 Å fringes along the fibre axis were very often tilted to the fibril edge by a small angle or even up to several degrees. Fig. 1 shows such examples, with drawings illustrating the configuration of the fibrils. The 4·5 Å fringes are tilted by the angles inscribed in the micrograph, in which it is noted that the fringe contrast disappears or becomes very weak periodically, at the places indicated by the arrows. This anomalous contrast is attributed to a moiré effect due to two sets of Bragg reflexions 020 from the segmental lattice planes which are radially disposed with respect to the fibre axis and at the same time nearly parallel to the incident beam. The period D of the fringe disappearance is given by $D = d/2 \sin \alpha$, where d is the fringe spacing concerned, being 4·5 Å in this case, and α is the tilt angle of the fringes to the edge of the fibrils. An estimated value of D is about 50 Å for $d = 4{\cdot}5$ Å and $\alpha = 2{\cdot}5°$, and nearly agrees with the observed value.

It is seen that the image contrast of the side walls of each fibril is nearly uniform and almost continous along the fibre axis, while that of the central void shows variations from place to place. This suggests that the central voids of fibrils are sometimes filled with amorphous material as reported by Martinez & Comer (1964). This phenomenon will be shown later more clearly in Fig. 5.

Fig. 2 shows two electron diffraction patterns from the Coalinga sample, (*a*) one obtained for a fibril showing the parallel fringes, and (*b*) one for a fibril showing the tilted fringes, where the terms 'parallel' and 'tilted' are used to mean the 4·5 Å fringe systems parallel and tilted to the fibril edge respectively. It is seen that the 020 reflexion in (*b*) is split into two spots with a separation of about 5°, which is twice the tilt angle, 2·5°. The *hk*0 spots are also split, while the 00*l* and *h*0*l* spots do not differ appreciably from those in (*a*).

The tilted 4·5 Å fringes and the related splitting of *hk*0 reflexions were subsequently found in all the other samples studied, including the synthetic one. Fig. 3 is a micrograph of a synthetic fibril in which the tilted 4·5 Å fringes are clearly seen with a relatively large tilt angle of 6°. Fig. 4 is an electron diffraction pattern from a synthetic fibril showing the tilted 4·5 Å fringes. It is seen that the *h*0*l* spots are similar to those from the fibril showing parallel 4·5 Å fringes [*e.g.* Fig. 2(*a*)], although the 0*k*0 spots are clearly split, subtending 5° in this case. In Fig. 5 is shown a low magnification image of two adjoining fibrils from the Transvaal and the corresponding selected area diffraction pattern. This diffraction pattern indicates that the 4·5 Å fringes of one of these two fibrils must be tilted and those of the other must be parallel. It is clearly seen in the micrograph that the intra-fibril voids are not always empty, but often filled with amorphous material at relatively regular intervals as mentioned above.

An examination was made of the distribution of the tilt angle of the 4·5 Å fringes of individual fibrils in several samples. The tilt angles were grouped at intervals of 0·5°, which was the lowest value of the tilt angle measurable on the electron micrographs. The histogram of fibrils showing parallel fringes differs

Table 1. *Chrysotile samples from different localities*

(in the order of observation)

Locality	State	Remarks
Quebec, Canada	pulverized, crude No. 1	Jeffrey mine, ref. Yada (1967)
Coalinga, California	ore, leathery sheet	ref. Mumpton & Thompson (1967)
Globe, Arizona	ore, crude No. 1	
Transvaal, S. Africa	ore, crude No. 1	
Tasmania, Australia	ore, crude No. 3	Rennison Bell mine
synthetic Mg-chrysotile	powder	ref. Noll *et al.* (1958,1960)

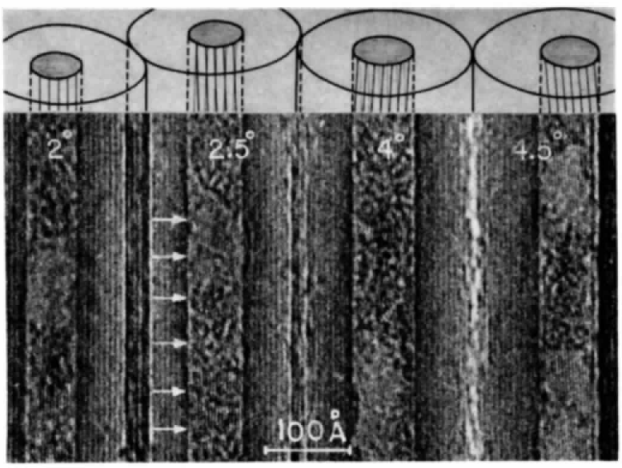

Fig. 1. Four fibrils showing the tilted 4·5 Å fringes in the middle of each fibril (Coalinga sample). The arrows indicate places where the contrast of the 4·5 Å fringes disappears by the moiré effect.

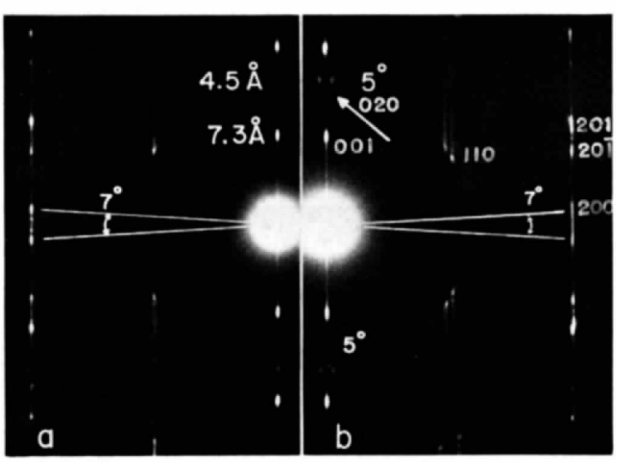

Fig. 2. Electron diffraction patterns from the fibrils showing (*a*) the parallel fringes and (*b*) the tilted fringes (Coalinga sample).

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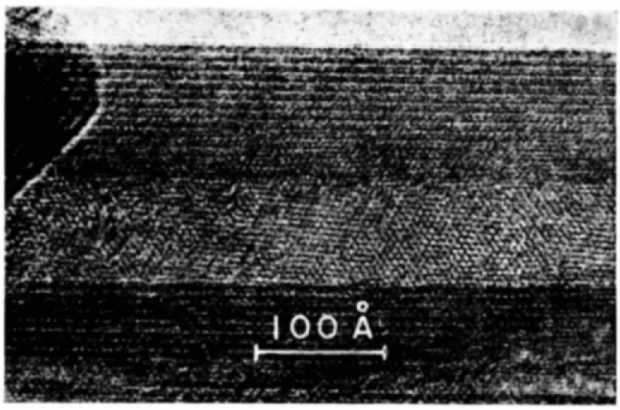

Fig. 3. A synthetic fibril showing the tilted 4·5 Å fringes with a relatively large tilt angle of 6° in the middle.

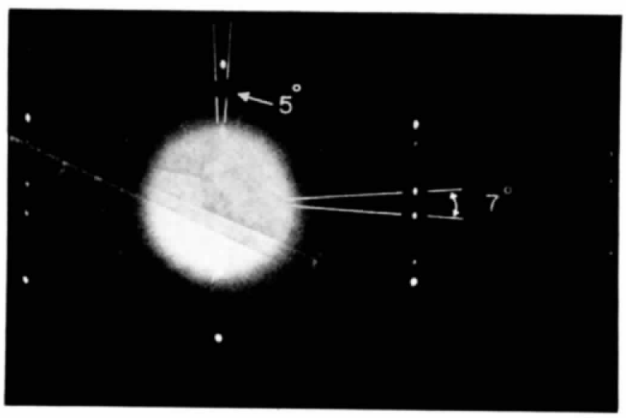

Fig. 4. Electron diffraction pattern from a synthetic fibril showing the tilted 4·5 Å fringes.

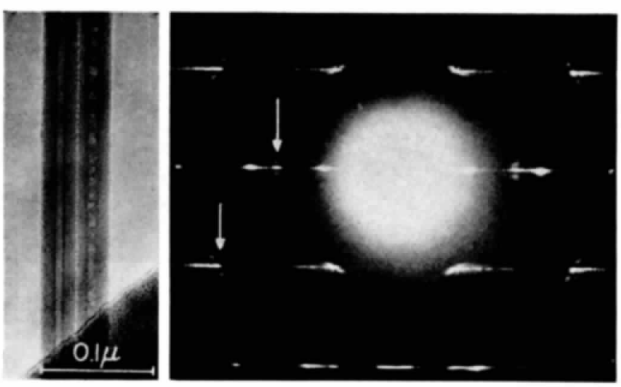

Fig. 5. Low magnification image of two adjoining fibrils having the vacant and the periodically filled intrafibril voids with the corresponding electron diffraction pattern. Diffraction spots indicated by arrows suggest that the 4·5 Å fringes of one of these fibrils are tilted and those of other are parallel (Transvaal sample).

appreciably from sample to sample as shown in Fig. 6. This is one of the new facts found by the lattice image method. In the case of the Coalinga sample, for instance, the percentage of the fibrils showing parallel fringes is about 20%. The synthetic sample showed a similar feature, as can be seen in Fig. 6. On the other hand, the sample from Quebec which was studied in the previous work (Yada, 1967) gave a fairly high percentage of 94% for the fibrils showing the parallel fringes, though its histogram is not included here. The samples from other sources gave intermediate values. It is seen that the angular distribution of the tilted 4·5 Å fringes shows a peak value at 2–3° and ranges up to about 10° depending on the source of the samples.

(*b*) *Para-chrysotile and ortho-chrysotile*

The existence of a small number of para-chrysotile fibrils (Whittaker, 1956*c*) was confirmed in certain samples (Globe and Tasmania) by observing the 4·5 Å fringes nearly perpendicular to the fibre axis, as was reported in the previous paper (Yada, 1967).

In principle, identification of ortho-chrysotile (Whittaker, 1956; Zussman, Brindley & Comer, 1957) would be possible by observing the 2·6 Å transverse

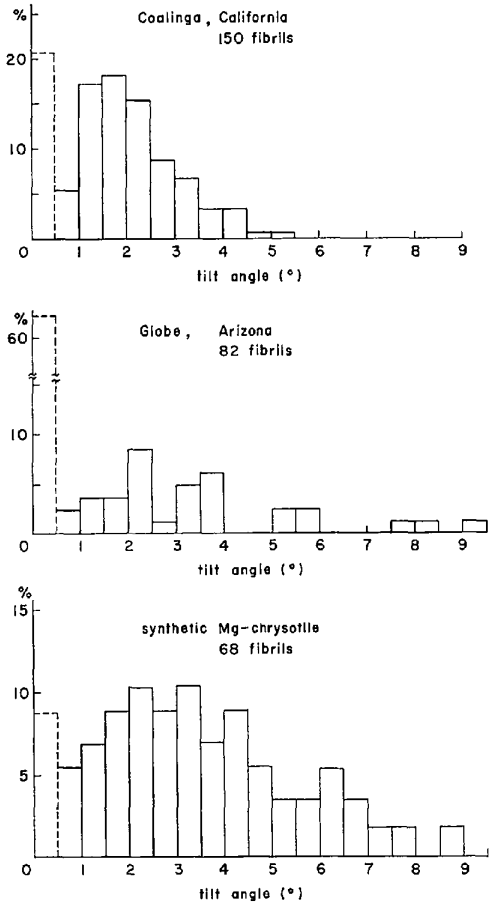

Fig. 6. Angular distribution of the frequency of the tilted 4·5 Å fringes.

fringes corresponding to 200, but, in practice, this was not possible because of the very poor contrast of this fringe system compared with the much higher contrast of the 7·3 Å fringes, crossing them. Diffraction patterns corresponding to ortho-chrysotile were rarely obtained. The results obtained on these para- and ortho-chrysotile fibrils will be reported in detail elsewhere.

(*c*) *Growth patterns*

Lattice images suggesting some new information about the growth mechanism were found at the growth end of the fibrils. Fig. 7 shows an example obtained from the Coalinga sample in which (*a*) is a low magnification image and (*b*) its enlargement. It is seen that the surface of the fibrils is quite clean on the molecular scale, without any amorphous material, and that the end of the fibril has a conical shape formed by steps of two or three sheets rolled up together. It is also seen that this conical fibril is considerably thicker at the bottom than at the top [the lower darkened area in (*a*)], which suggests a discontinuous step-growth as described later. Such a conically stepped structure was also observed inside a fibril as seen in Fig. 8 These observations seem to support the coupled dislocation mechanism proposed by Jagodzinski & Kunze (1954).

In the case of synthetic chrysotile, another type of cone-shaped fibrils was found in which the 7·3 Å fringes on opposite sides of a fibril are not parallel, as shown in Fig. 9. These two sets of 7·3 Å fringes make an angle of 3° for fibril *A* and of 5·5° for *B*. The 4·5 Å fringes in the middle of these fibrils are also tilted at a fairly large angle, to the fibre axis, though their fringe contrast is poor. This is illustrated in the drawing (*b*) which traces the fringe systems of *A* and *B* in (*a*). It is noted that the tilting of the two sets of the 4·5 Å fringes are asymmetrical about the fibre axis with an angular difference of a few degrees. This type of growth pattern seems to correspond to the cone-in-cone pattern which was reported by Bates & Comer (1957).

Observation from the direction parallel to the fibre axis

The general feature that most fibrils are hollow but a few are solid, was seen in the cross section of the samples from all the sources examined. Further observations revealed the following.

(*a*) *Step-growth*

Fig. 10 shows the cross section of a rather thick fibril from Coalinga indicating a two-step growth. The two or three-step growth was also observed in relatively thick fibrils in the samples from other sources. The frequency of such two- or three-step growth is different from sample to sample, and is particularly high in the specimen from Tasmania as shown in Fig. 11. It is seen that fibrils thicker than about 350 Å show traces of the stepwise growth. In the analysis of the distribution of the fibril diameter, therefore, such a stepwise growth should be taken into consideration.

(*b*) *Growth patterns*

Fig. 12 shows the typical cross-section of the sample from Globe, Arizona. Besides a normal cylinder at *A*, there are many anomalous ones, including a multi-layer spiral at *B*, a non-hollowed cylinder at *C*, two-step growth at *D*, and an inter-fibril segment at *E*. Anomalous black and white contrast indicated by the arrows seems to be due to higher order reflexions which are excited simultaneously. It is seen that inter- and intra-fibril parts are well preserved without serious radiation damage, as a result of the precautions referred to in the preceding section. We can see that the inter-fibril sites are generally vacant.

These anomalous growth patterns were rarely seen in the samples from Quebec, Coalinga and Transvaal, while they were often observed in the samples from Globe and Tasmania.

(*c*) *Behaviour of the* 7·3 Å *fringes*

It was reported in the previous paper (Yada, 1967) that all the circumferential 7·3 Å fringes observed were spiral or multi-spiral. Although such spiral structures were found in all the samples studied, it was shown for the first time that a perfectly concentric structure also exists in some samples. In Fig. 13, (*a*) shows the spiral structure and (*b*) the multi-spiral one, seen in the sample from Transvaal. Fig. 14 shows the concentric structure in the same Transvaal sample, in which two pairs of through-focused images are shown, and forming the concentricity. It is noted that the upper fibril clearly has concentric structure, though it is a little elliptical in shape. The 4·5 Å fringes corresponding to 020, in the radial direction, are clearly visible in the upper images, while they are scarcely seen in the lower ones. The disappearance of these fringes seems to be caused not by a lack of resolution but by a deviation from the Bragg condition due to the tilting of the 010 layers. Table 2 shows the distribution of the concentric and spiral structures for three samples. It is seen that the percentage of fibrils showing the concentric structure is fairly high, being nearly a half in the Transvaal sample, while it is low for the Coalinga and Tasmania samples. Although the total numbers of cross sections examined are not sufficiently large for statistical analysis, the observed differences seem to be significant and are presumably related to the different growth conditions.

(*d*) *Distribution of diameters*

From the histograms for the distributions of the inner and outer diameters shown in Fig. 15, the following points are noted: (1) The distribution of the diameters is considerably different from sample to sample, presumably depending on the growth conditions in each locality. (2) In samples for which the outer diameters are distributed over wide ranges, there is a tendency for the peak position in the histogram to shift to a larger value. In such samples, fibrils both with a very small inner diameter and without the central void are often observed. The samples from Quebec, Globe and Tasmania, which contain a number of fibrils consisting of a multi-spiral lattice, are classified into this group. (3) The most frequent values of the outer and inner diameters of the samples showing a sharp distribution (Coalinga and Transvaal) are 220–270 Å and 70–80 Å, respectively. The value of 260 Å for the outer diameter obtained by Whittaker (1957), (partly theoretically and partly from X-ray diffraction measurements on a sample from Bell Mine, Quebec), is in good agreement with the present value, but his value of 110 Å for the inner diameter is not. (4) Most fibrils thicker than about 350 Å seem to contain a discontinous step-growth.

Discussion

(*a*) *Crystal structure*

Although Padurow (1950) suggested a triclinic structure for chrysotile, it is thought at present that the principal form of chrysotile is clino-chrysotile, while ortho- and para-chrysotile are less common (Whittaker & Zussman, 1956). The result found in the present experiment that the 4·5 Å fringes are not always parallel to the fibril edge seems to be important in connexion with the crystal structure of chrysotile. The tilting of these fringes is equally compatible with either of the following structures. (1) a helical roll with a rectangular lattice, retaining the monoclinic structure (a clino-chrysotile in which the *a* axis is not exactly parallel to the fibre axis and the *b* axis not exactly perpendicular to the fibre axis) and (2) a straight roll with a non-rectangular lattice (an anorthic structure in which the *b* axis is perpendicular to the fibre axis but the *a* axis is not parallel and the *c* axis not perpendicular to the fibre axis). A choice between these two possibilities could be made if the 2·6 Å transverse fringes corresponding to 200 in the central region of the fibrils were clearly observed. Unfortunately, however, it was difficult to resolve these fringes, as mentioned in the preceding section. However, by a dark field method in which a 200 reflexion is selected by the use of a small objective aperture and the corresponding dark field images compared with a normal bright field image, it is possible to decide which of the helical roll and the straight roll is the true structure.

According to a preliminary test using such a method it appears that the former is the case.* With a model of the helical roll it is quite easy to understand the experimental fact that the tilt angle of the 4·5 Å fringes varies over a range of up to about ten degrees from fibril to fibril, though no periodicity of the angular distribution was observed in the present experiment.

There is still the possibility of a polytype structure. The non-parallel 7·3 Å fringes seen in the synthetic chrysotile (Fig. 9) are the result of such a polytype. This type of structure, however, does not necessarily

* The details will be reported elsewhere.

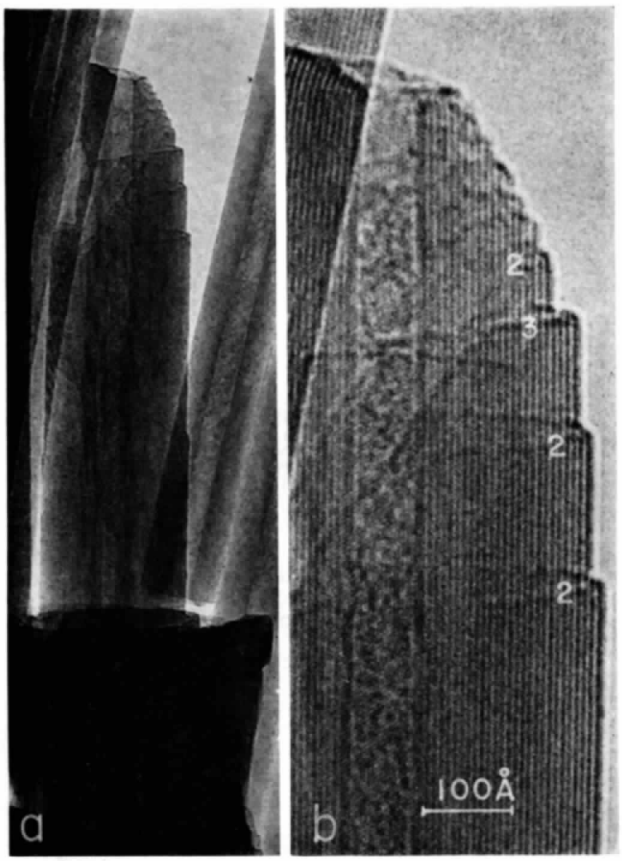

Fig. 7. Conically stepped growth end rolled with two or three sheets (Coalinga sample). A very thick root is seen in (*a*) the low magnification image and (*b*) the surface of the enlarged image is quite clean.

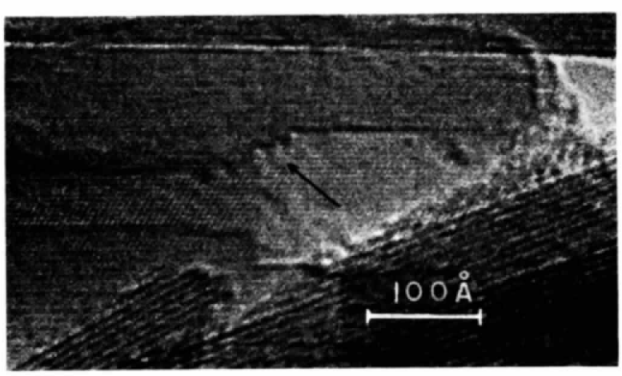

Fig. 8. Conically stepped structure inside a fibril (Globe sample).

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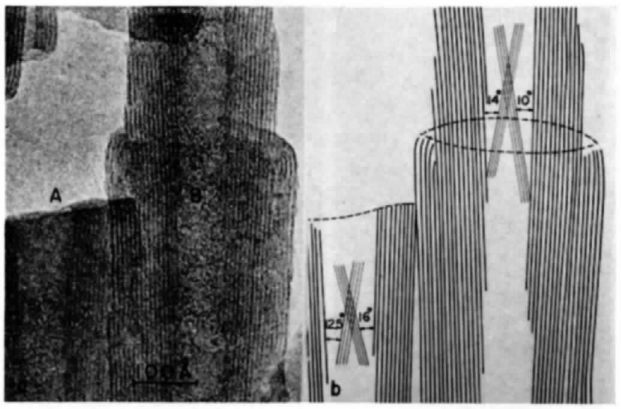

Fig. 9. Conical fibril in which the 7·3 Å lattice fringes on opposite sides are not parallel to each other (synthetic sample). The conically tapered angles are 3° (A) and 5·5° (B). (*b*) is a drawing of the fringe systems.

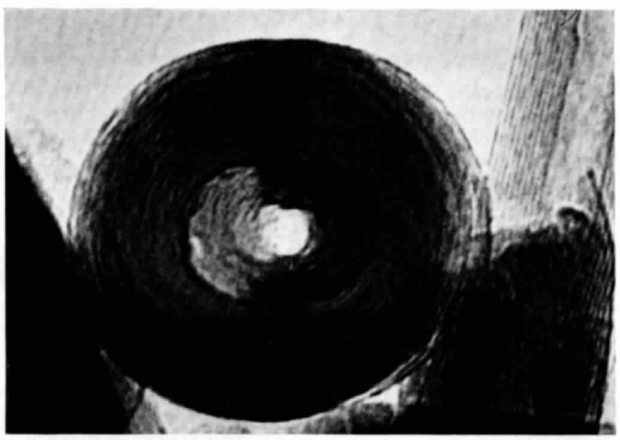

Fig. 10. Cross section of a thick fibril showing two-step growth (Coalinga sample).

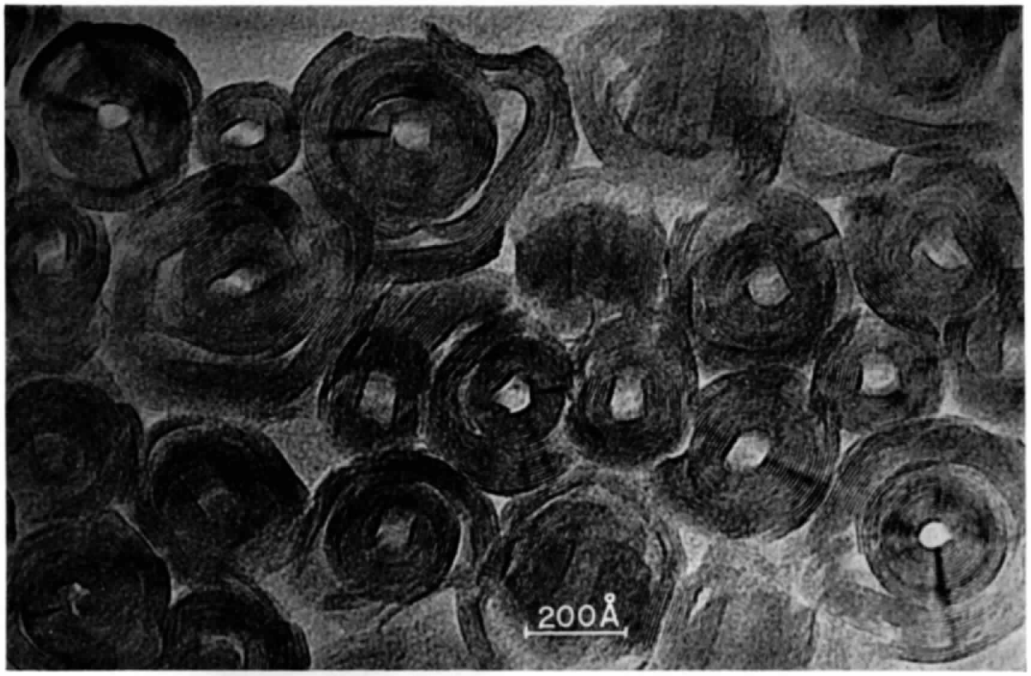

Fig. 11. A remarkable example of the step-wise growth seen in the Tasmania sample.

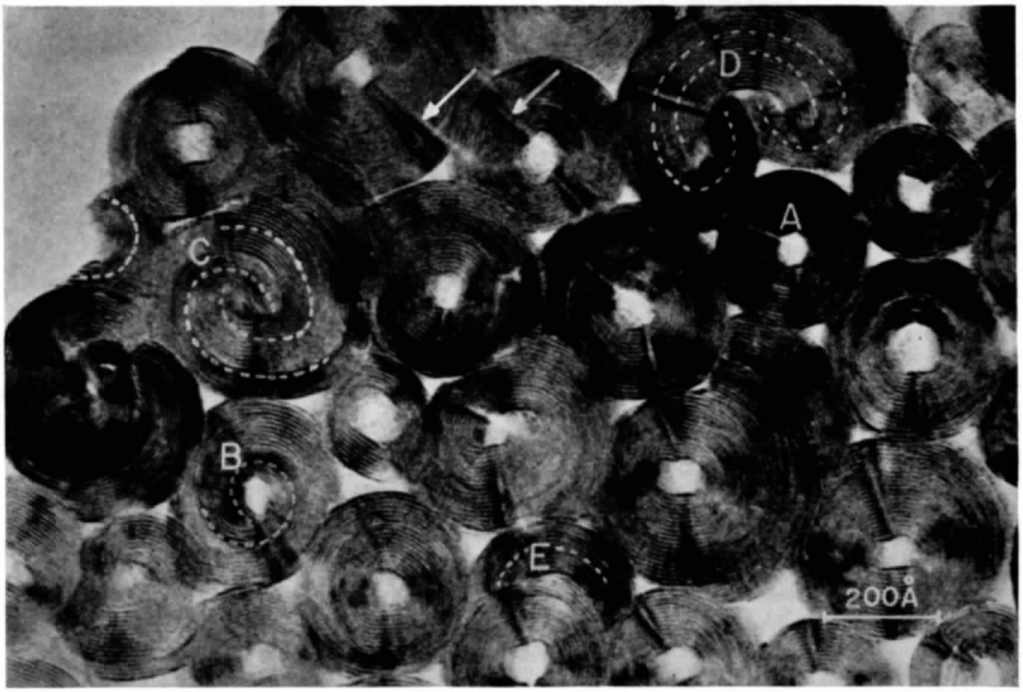

Fig. 12. An example of the Globe sample showing the coexistence of many types of growth patterns.

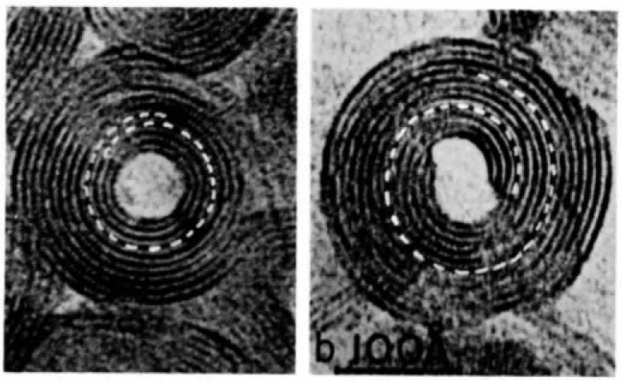

Fig. 13. Spiral structure in the cross section of the Transvaal sample. (*a*) is a single spiral and (*b*) a multi-spiral structure.

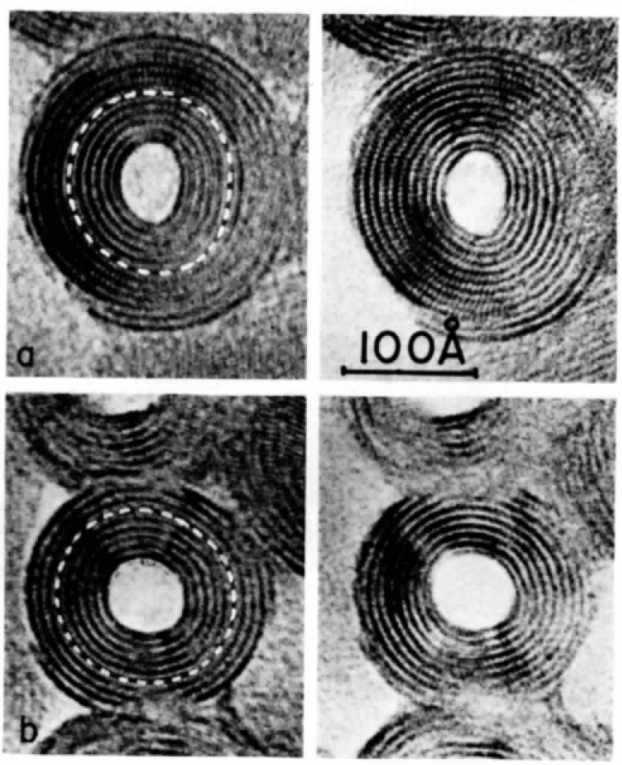

Fig. 14. Two pairs of through-focused images showing the concentric structure in the cross section of the Transvaal sample.

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Table 2. *Distribution of concentric and spiral structures in the circumferential lattice layers*

Sample	Concentric structure (%)	Spiral structure (%)				Total number examined
		Single	Double	Triple	Quadruple	
Coalinga, California	28	42	16	11	3	36
Transvaal, S. Africa	54	25	19	2	0	43
Tasmania, Australia	13	33	13	13	27	15

conflict with the clino-chrysotile, because such a structure may occur by a component of the conical roll additional to the original helical roll. In this case, the stacking of the 200 layers becomes imperfect and is accompanied by a component of the conical roll. Accordingly, the contrast of the 4·5 Å fringes would become poor. The fact that the tilt angles of two sets of the 4·5 Å fringes are asymmetrical seems to support this idea.

(*b*) *Growth mechanism*

The possibility of both concentric and spiral structures for the circumferential lattice layer, suggested by Jagodzinski & Kunze (1954) in terms of the dislocation theory is confirmed by the present observations. The conically stepped structure of the growth end (Figs. 7 and 8) can also be explained by this model. It was found at the same time that the ratio of the concentric structure to the spiral one varied from sample to sample, presumably depending on their growth conditions. From these results, the concentric lattice layers seem to exist in the Quebec sample studied in the previous work (Yada, 1967) though its frequency of appearance must be very small.

On the other hand, exceptional growth patterns such as in fibrils *D* and *E* in Fig. 12 were found to exist,

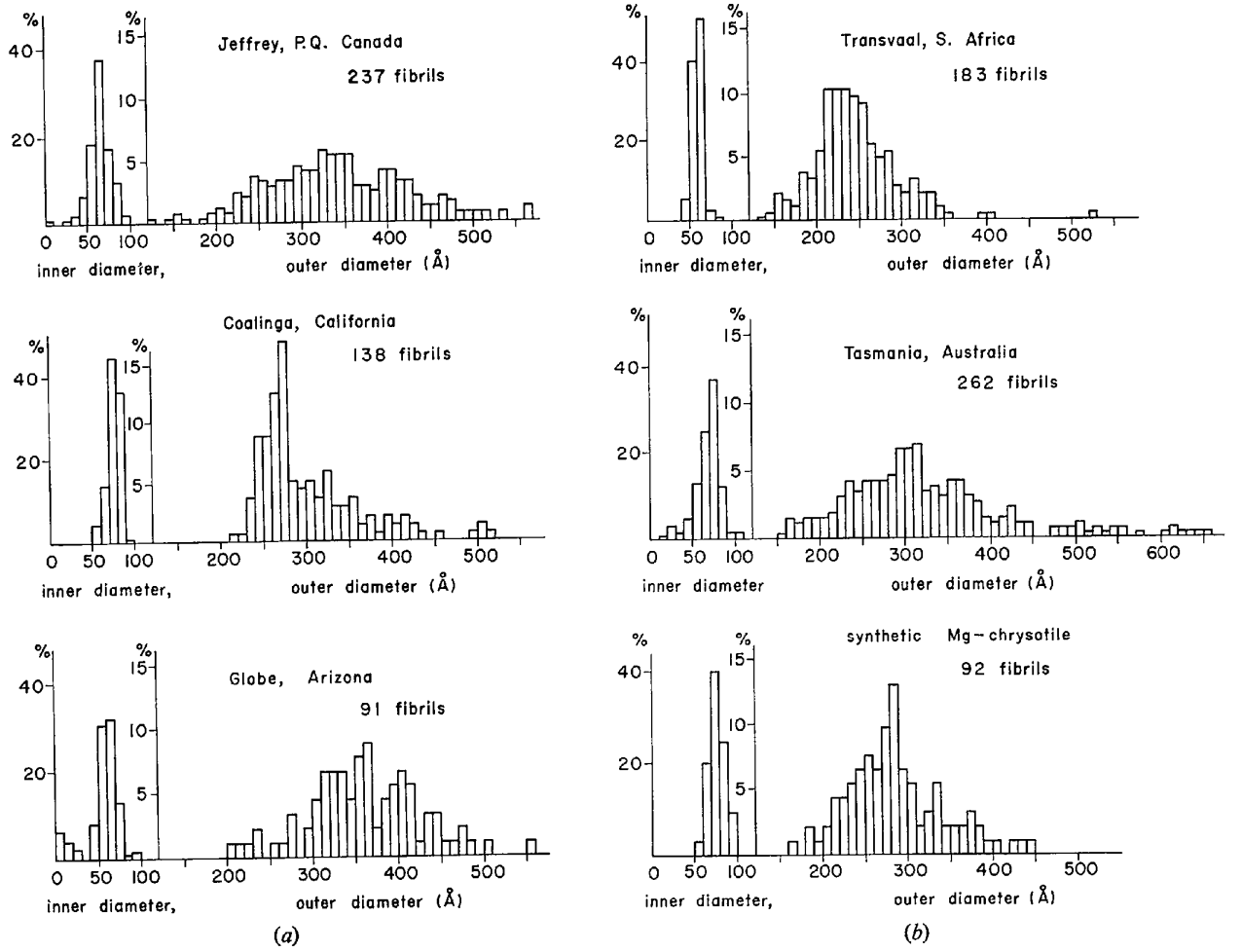

Fig. 15. Frequency distribution of inner and outer diameters for the samples from different localities whose names are inscribed with the number of fibrils examined.

even frequently, in a particular sample. To understand the reason for the variety of growth patterns, it seems to be necessary to study the effect of impurities, lattice defect concentration *etc.* For synthetic chrysotile it was found that most of the fibrils had the form of hollow cylinders, and the distribution of the tilt angle of the 4·5 Å fringes was similar to that of natural chrysotile. The concentration of lattice defects for the direction perpendicular to the fibre axis was not found to be particularly different from that of natural chrysotile. Therefore, the role of impurities does not seem to be important in controlling the polytype during the growth process.

It was found that most of fibrils thicker than about 350 Å in diameter were discontinuously grown in two or three steps (Figs. 10 and 11) perhaps because of changes in the growth conditions. The diameter of the core part of these thick fibrils was of nearly the same order as the peak value of the sample whose outer diameter was sharply distributed and is in good agreement with the theoretically expected value, 260 Å (Whittaker, 1957). The Tasmania sample, consisting of very short fibres, is typical of this step-growth and also shows a low frequency of appearance of the concentric lattice layer, while the Transvaal sample consisting of long and uniform fibres, has a considerably high frequency of appearance of concentricity. It may be said on the basis of the theory of Jagodzinski & Kunze (1954) that the former sample was mainly grown by the radial or coupled dislocation mechanism under conditions of high supersaturation, while for the latter sample the axial dislocation mechanism prevailed under relatively low supersaturation conditions.

The outer surfaces of fibrils were generally found to be very smooth and clean. This fact suggests that the inter-fibril sites of the aggregated fibrils are generally vacant and free from any amorphous material. On the other hand, the central voids are frequently filled with amorphous material. This phenomenon may be correlated with the mechanism of the transfer of amorphous material through the central voids by capillary action.

The author is indebted to Prof. T. Hibi for continuous encouragement, to Dr E. J. W. Whittaker for valuable discussions and for critical reading of the manuscript, and also to Mr K. Shibata for experimental assistance. The author is grateful to Prof. H. Fernández-Morán, The University of Chicago, Mr T. Otouma, Japan Asbestos Co., Dr F. A. Mumpton, Union Carbide Corporation, Mr A. F. Wilson, Applied Mineralogy CSIRO and Prof. N. W. Noll, Bayerwerk, for supplying the samples of chrysotile.

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