Terminations of multiple-chain lamellae in grunerite asbestos

E. J. W. WHITTAKER AND B. A. CRESSEY

Department of Geology and Mineralogy, Parks Road, Oxford OX1 3PR

AND J. L. HUTCHISON

Department of Metallurgy and Science of Materials, Parks Road, Oxford OX1 3PR

ABSTRACT. Grunerite asbestos (var. amosite) from Penge, Transvaal, contains lamellae consisting of multiple-chain *I*-beams which frequently terminate within the structure. Such terminations may be either coherent or incoherent, the latter involving disturbances of the structure akin to dislocations. These may be of the nature of edge dislocations or of dislocations having both edge and screw components.

Models are presented which have been used to elucidate the structural disturbances associated with the termination and side-stepping of multiple-chain lamellae. Odd-multiple-chains are shown to be of particular significance in giving rise to dislocations with a screw component along the *c*-axis.

IN recent years there have been a number of studies of amphiboles (or more generally, biopyriboles) by high-resolution electron microscopy which have revealed extensive defects involving multiple chains wider than the double silicate chains of amphibole itself. The earlier studies (Hutchison *et al.*, 1975; Veblen *et al.*, 1977; Jefferson *et al.*, 1978; Mallinson *et al.*, 1980) were confined to dispersed crystal fragments which could only be viewed parallel to the *a*- or *b*-axis. More recently, studies of ion thinned cross-sections perpendicular to the *c*-axis have been carried out (Alario Franco *et al.*, 1977; Veblen and Buseck, 1979, 1980) and have proved to be still more informative.

We have studied two specimens of grunerite asbestos (var. amosite) from the banded ironstones at Penge, Transvaal. Both were fibrous though in the optical microscope one looked coarser than the other. Both sections gave similar results. The specimens were embedded in Araldite CY212 and sections approximately 0.03-0.05 mm thick were cut approximately perpendicular to the fibre axis by normal sectioning techniques. Both sides of the section were polished. Great care had to be taken to prevent disintegration of the section because of the weak cohesion between the fibres. For this reason the section was kept undesirably thick at this stage, and the choice of embedding medium was crucial. The sections were ion-thinned from both sides using an Edwards IBT 200 machine with an accelerating voltage of 5.5 kV and the specimen inclined at 20° to the beam. This process was very prolonged, of the order of 150 hours, and failure of the specimen at this stage was also quite frequent. The micrographs were taken on a JEOL JEM100B at 100 kV at an under-focus of 800 Å.

Many multiple-chain lamellae have been observed in these specimens, varying in width from triple to 24-tuple. A substantial proportion of these lamellae have been observed to terminate within the structure, as has now also been observed by Veblen and Buseck (1980) for multiple-chain lamellae in anthophyllite, chesterite, jimthompsonite, and disordered biopyriboles from Chester, Vermont. In such terminations the multiple-chain lamellae give place either to double-chain lamellae of normal amphibole type or to multiple ones of lower multiplicity. This can occur in such a way that the structure is undisturbed except along a line (parallel to c) at the termination of a single multiple lamella, or along a plane or planes (parallel to c) joining the terminations of two (or more) multiple lamellae. For such 'coherent' terminations to occur certain criteria must be satisfied regarding the widths and numbers of lamellae before and after the termination, and these criteria have been formulated by Veblen and Buseck (1980) in two termination rules. Prior to the appearance of their paper we had also observed and formulated equivalent termination rules (Cressey et al., 1980). Rule 1 of Veblen and Buseck may be expressed by saying that the sum of the multiplicities of the *I*-beams passing through the region of the termination remains constant, and Rule 2 by saying that the number of *I*-beams may only change by an even number. However, whereas in their specimens only two cases of incoherent terminations (violating the rules) were observed, in grunerite we have found such incoherent terminations to be quite common, and we have accordingly analysed them in greater detail. The simpler structure of a clinoamphibole also makes it practicable to take account of the displacements in the cdirection that are involved at some kinds of termination. In ortho-amphibole, as Veblen and Buseck remark, 'the displacements parallel to c are complicated by the stacking sequence', but we do not accept that they are not necessary for an adequate discussion. We also introduce some additional discussion of co-operative terminations and sidestepping of lamellae of multiple *I*-beams.

Interpretation has been facilitated by the preparation of cardboard cutouts to represent the crosssections of amphibole I-beams, and also of I-beams of other widths, which can be arranged in interlocking patterns to represent the structures under consideration. However, when I-beams of a variety of widths are used (fig. 1) it is important to recognize that they only have mirror symmetry, with the mirror plane perpendicular to [010], if they are built from double silicate chains, as in amphibole itself, or from n-tuple chains where n is even. If they are built from single pyroxene chains, triple silicate chains, or n-tuple chains where n is odd, then the mirror plane is replaced by a *c*-glide plane, as may be seen from fig. 2. In these odd I-beams corresponding features at the two sides are at heights +c/4 and -c/4 relative to an appropriate datum level, and this is represented on the cut-out by + and – signs. In the stackings the *I*-beams are shown in black against a white background. This conforms with the contrast in electron micrographs taken at optimum underfocus of about 800 Å on the thinnest regions of the specimens (< 150 Å thick), where low projected charge density appears light, so that holes between the *I*-beams are white.

It is to be noted that the analogy between the real *I*-beam patterns and the patterns of cardboard cutouts is deficient in one respect. The gaps between the cutouts are elongated rectangles, but these rectangles are not uniformly 'empty' in the real structures. As may be seen in fig. 3, the number of nearby atoms in projection down [001] is least in the centre of the gap between two amphibole *I*beams at the position of the empty *A*-site. Thus the appearance of the gaps as round white blobs on the micrographs is not merely a result of lack of resolution, and the gaps between multiple chains in the best micrographs are resolved into a series of white blobs corresponding to the number of A-sites between the I-beams, there being n-1 such sites between two n-tuple I-beams. This permits the widths of the I-beams to be determined directly by inspection of the micrographs. When the resolution is not good enough for this the widths can always be determined from the spacing between the rows of



FIGS. 1 and 2. FIG. 1 (top). Drawings of the cut-out *I*beams used to produce model structures. The numbers denote the multiplicity of the corresponding silicate chains. Even multiplicities give *I*-beams having vertical mirror symmetry, and odd multiplicities give *I*-beams having vertical *c*-glide planes, as shown by the full and dotted median lines. FIG. 2 (bottom). Plan views of single, double, triple, and quadruple silicate chains demonstrate the mirror and glide symmetry introduced by even and odd multiplicity.



FIG. 3. Projection of the amphibole structure down the c-axis. The lowest density of atoms is around the A-site, so that this is also the region of lowest charge density.

white spots between the amphibole *I*-beams at each side; for an *n*-tuple *I*-beam this spacing is approximately (9 + 4.5n) Å.

Coherent termination of a single lamella of multiple I-beams. Within the context of amphibole subjected to retrograde alteration it seems unlikely that pyroxene I-beams will be found in conjunction with the double and higher multiple I-beams; we have seen none, and the one micrograph presented by Veblen and Buseck (1980) in which they invoke the presence of a single pyroxene I-beam column seems to be open to alternative interpretation. The following remarks are to be understood as presupposing absence of pyroxene I-beams; the discussion would have to be generalized in order to apply to cases of pyroxene subjected to corresponding alteration. We may also conveniently distinguish between total termination of a multiple lamella (i.e. with dissociation entirely into amphibole *I*-beams) and its reduction in width to form one or more multiple lamellae together with amphibole lamellae. In our material, at least, the former appears to be more common, though the latter also occurs to an appreciable extent.

With these restrictions, it follows from Rule 2 of Veblen and Buseck that coherent total termination of one *n*-tuple *I*-beam can only occur if n = 4N + 2.

Co-operative termination of multiple lamellae. If two multiple lamellae terminate jointly, the requirement that the increase in number of lamellae be even requires that if the two lamellae of multiple *I*-beams are *m*-tuple and *n*-tuple then m + n = 4N. However, the nature of the phenomenon depends on whether *m* and *n* are both even or both odd.

If the multiplicities are both even then they must both be multiples of 4 as otherwise the lamellae would each self-terminate in the way discussed above without any interaction. In the simplest case m = n = 4 as in fig. 4. The two terminating lamellae are linked by a row of holes which may lie along a (110) plane (fig. 4a) or may zigzag along a series of such planes so that, in the limit, they may approximate to a (100) plane (fig. 4b); but there is no fundamental difference between these two possibilities. Each hole is of the volume of half an amphibole I-beam, and the number of holes is independent of whether they lie on a single plane or a zigzag. It is given by r+2, where r is the number of amphibole lamellae between the multiples. If either m or n exceeds 4 then the effect is very similar and in general the number of holes is always r+N.



FIG. 4. Model of the joint coherent termination of two quadruple *I*-beam lamellae: (a, left) joined by a row of holes on (110); (b, right) joined by a row of holes on (100).



FIG. 5. Model of the joint coherent termination of a triple and a quintuple *I*-beam lamella: (a, left) with the lamella of type $4M_1 - 1$ terminating first; (b, right) with the lamella of type $4M_1 + 1$ terminating first. The region bounded by the row of holes and the negative signs is displaced by c/2 relative to the rest of the structure.

If m and n are odd they are necessarily unequal, as one must be of the form $4M_1 - 1$ and the other of form $4M_2 + 1$, where M_1 and M_2 may be either equal or unequal. The number of holes and broken



FIG. 6. Electron micrograph of grunerite asbestos parallel to the *c*-axis. An example corresponding to Fig. 5*a* occurs at the right of the photograph. Towards the left there is a coherent termination of a sextuple lamella. Scale bar = 50 Å.

bonds are given by the same expressions, but the volume of the holes depends on the relative lengths of the two lamellae. If the one of form $4M_1 - 1$ terminates first, and the holes lie on (110) as in fig. 5a, then there are r+1 holes with the volume of half a pyroxene *I*-beam and N-1 with the volume of half an amphibole I-beam. If the lamella of form $4M_2 + 1$ terminates first and the holes lie on (110) as in fig. 5b, then each of the r+1 holes increases in size to $1\frac{1}{2}$ times that of a pyroxene *I*-beam. If the lengths of the lamellae are more equal and the holes zigzag, then there is an intermediate number of enlarged holes. Also, the region of the structure bounded by the two terminating lamellae and the plane of holes is displaced by $\frac{1}{2}c$ relative to the rest of the structure, but is coherently bonded to it except across the plane of holes. This is evidenced in fig. 5 by the + and - signs on the triple and pentuple I-beams. An example of this type of mutual termination is shown at the right-hand side of fig. 6, where the two lamellae are triple and quintuple, with the latter extending further and the (minimal-sized) holes lying on (110).

The above discussion would require further generalization to deal with the possibility of cooperative termination involving more than two lamellae, which has been seen by Veblen and Buseck (1980) in anthophyllite, but which we have not observed in amosite.

Incoherent termination of one lamella of multiple I-beams. The effects here are of three types according as they involve termination of a lamella of *n*-tuple *I*-beams into an even number of narrower lamellae of the same total width (thereby violating



FIG. 7. Model of an incoherent termination of a quadruple lamella. There is structural disturbance having some of the characteristics of an edge dislocation, involving an extra half-row of *I*-beams entering from the left.

Rule 2), or into *I*-beams having a greater total width, or a lesser total width (thereby violating Rule 1).

The simplest case of the first type is the termination of a quadruple lamella into two amphibole lamellae. This is modelled in fig. 7, but exactly the same result would occur if any multiple were to dissociate into an odd number of amphibole lamellae together with a residue, e.g. $5 \rightarrow 2+3$, $10 \rightarrow$ 2+2+2+4, etc. Apart from the termination of the wide I-beams the other obvious feature on fig. 7 is an extra half-row of A-site holes lying on a (110) plane making an angle of 118° with the multiple lamella, and a close examination of the pattern of Ibeams reveals something very like an edge dislocation with an extra half-plane of I-beams on (100) entering from the left. There is, however, some difficulty in defining its Burgers vector because not all the 'unit cells' are identical. If we ignore this fact, then the Burgers vector is $\frac{1}{2}\mathbf{a} + \frac{1}{2}\mathbf{b}$, but if we treat a cell containing a quadruple I-beam as being of width $\frac{3}{2}\mathbf{b}$ (which is only approximate) then the Burgers vector is $\frac{1}{2}a^{1}$. The nearest available example is shown in fig. 8, where a quintuple lamella terminates into an amphibole lamella and a triple lamella, but the triple one simultaneously side-steps along (110). This is modelled in fig. 9.

¹ The 'dislocation' is along [001], so that a Burgers vector of $\frac{1}{2}a$ or $\frac{1}{2}a + \frac{1}{2}b$ implies a slight screw component in the monoclinic system. The photograph represents a projection on a plane perpendicular to [001], but the Burgers vector cannot lie in this plane without involving an irrational discontinuity in the *c*-direction.



FIGS. 8 and 9. FIG. 8 (left). Electron micrograph of grunerite asbestos parallel to the c-axis. A quintuple lamella terminates into an amphibole lamella and a triple lamella, but the latter immediately side-steps to the right. Scale bar = 20 Å. FIG. 9 (right). Model of the structure in Fig. 8. Note that the structure on opposite sides of the odd-multiple lamella is displaced by c/2.



FIG. 10. Electron micrograph of grunerite asbestos down the c-axis, showing a rectangular area containing 16-tuple *I*-beams tilted at an angle of 3° to the *b*-axis. This permits them to join on to the amphibole structure at each side without its being displaced parallel to the *a*-axis. Other multiple lamellae are present but are not sufficiently well resolved for comment. Scale bar = 80 Å.

A rather special case is shown in fig. 10. Here there is a lamella of 16-tuple I-beams which both begins and terminates within the structure, giving place at each end to 8 amphibole lamellae. Although the surrounding amphibole structure is not very well resolved it does not have to be dislocated in any way, because the short double-ended nature of the lamella permits the strain to be taken up entirely within the multiple lamella by a tilting about the c-axis. The required tilt involves a relative displacement of a/2 between opposite sides of the I-beams, so that for 16-tuple I-beams it amounts to 3.7° , and the measured value of 3° is in reasonable agreement. Take-up of the strain in this way is clearly only possible for very wide I-beams; a similar example has been shown by Veblen and Buseck (1980) for octuple I-beams in anthophyllite, and we have also observed it in the case of a 24tuple lamella.

If an odd multiple lamella terminates completely then there must be either an increase or decrease in over-all width along the *b*-axis by half an amphibole *I*-beam. These two cases are modelled in figs. 11 and 12 for a quintuple lamella, and both are illustrated in fig. 13 at opposite ends of a short lamella of seven quintuple *I*-beams, although there is some further disturbance of the structure just above the upper termination of this lamella. In fig. 11 the change in number of *I*-beams along the *b*-axis is even, so that Rule 2 is obeyed, and the



FIGS. 11 and 12. FIG. 11 (*left*). Model of the termination of a quintuple lamella into three double lamellae. There is an expansion in width of b/4 and the dislocation has a screw component of $\frac{1}{2}c$ in order to reconcile, at the top of the diagram, the relative displacement between the two sides of the odd-multiple lamella. FIG. 12 (*right*). Model of the termination of a quintuple lamella into two double lamellae. There is a contraction in width of b/4 and again a screw component of $\frac{1}{2}c$.

strain in the plane of the diagram is therefore purely dimensional, with no problems of misfit between the staggered rows of *I*-beams. There is, however, a misfit in the direction of the c-axis because of the glide symmetry of odd I-beams, so that the termination has some of the characteristics of a screw dislocation with Burgers vector $\frac{1}{2}c$. Within the plane of the diagram there is a Burgers vector of **b** in terms of unit cells, or of approximately $\frac{1}{4}\mathbf{b}$ in the same sense in terms of distances. In fig. 12 there is again a 'screw dislocation' component with Burgers vector $\frac{1}{2}c$, but the change in number of *I*-beams is odd, so that both rules are broken, and the 'edge component of the dislocation'² is either $\frac{1}{2}\mathbf{a} + \frac{1}{2}\mathbf{b}$ or $\frac{1}{2}\mathbf{a} - \frac{1}{4}\mathbf{b}$ in terms of unit cells or distances respectively.

It is to be noted that the two forms of the 'edge component of the dislocations' discussed above depend on whether the change in the number of *I*-beams at the termination is even or odd; whether

² Here again this Burgers vector is not perpendicular to the dislocation line but the small screw component arising in this way is not relevant to the implications of the $\frac{1}{2}c$ screw component for crystal growth. It is therefore preferable to keep the two separate and to associate $\frac{1}{2}a$ wholly with the 'edge component'.

they are to be associated with an increase or decrease in the total width of *I*-beams depends on the multiplicity of the terminating lamella. If this is *n*-tuple with *n* of the form 4N + 1 (as in fig. 13) then the above analysis applies in detail. If *n* were of the form 4N - 1 (e.g. a triple) then the opposite values of Burgers vectors would be associated with the increase and decrease in total width, and this would apply to the single case of this type observed by Veblen and Buseck (1980). However, as that was in anthophyllite the problem of the screw component requires more detailed analysis.

Termination of an even multiple lamella is also possible with a reduction in width and an even change in number of *I*-beams, as shown in fig. 14, though we have not yet observed a termination of this kind. In this case there is no screw component to the 'dislocation', and in the particular case shown the edge component is also zero in terms of unit cells, but it is $\frac{1}{2}$ b in terms of distances.

Sideways stepping of multiple I-beam lamellae. This phenomenon has been illustrated in anthophyllite by Veblen and Buseck (1980), although they did not discuss the nature of the associated holes. We have also found it in amosite. It is to be noted that two mechanisms for such sidestepping appear to be possible, at any rate for reasonably narrow multiple *I*-beams, although only the first of the mechanisms has so far been observed.



FIGS. 13 and 14. FIG. 13 (*left*). Electron micrograph of grunerite asbestos down the *c*-axis. The short lamella of seven quintuple *I*-beams terminates below as in Fig. 11 and above as in Fig. 12 (followed by some further ill-defined disturbance). Scale bar = 20 Å. FIG. 14 (*right*). Model of the termination of a quadruple lamella into one double lamella.



FIGS. 15 and 16. FIG. 15 (*left*). Model of the sideways stepping of a multiple lamella along (110). FIG. 16 (*right*). Electron micrograph of grunerite asbestos down the *c*-axis, showing side-stepping of a quadruple lamella along (110). Scale bar = 50 Å.

In the first mechanism the multiple I-beams can be off-set along a (110) plane as shown in fig. 15, and the diagonal line of multiple *I*-beams is then flanked on each side by a row of holes; each hole has the volume of half an amphibole I-beam, and there are two such holes for each sideways stepping by a vector **b** (total vectorial displacement $\mathbf{a} + \mathbf{b}$). This mechanism is applicable to all widths of I-beams, the only effect of the width being to move the flanking rows of holes further apart. An example (with quadruple I-beams) is shown in fig. 16. The second mechanism is shown in fig. 17 and leads to displacement along a (130) plane. It produces only a single line of holes, one per displaced I-beam instead of two, but the displacement is by $1.5\mathbf{a} + 0.5\mathbf{b}$, so that the number of holes is the same for a given total amount of lateral displacement. For triple I-beams the holes are also smaller than in the first mechanism, but they increase in size with the width of the *n*-tuple *I*-beams involved, their volume being $\frac{1}{4}(n-2)$ of an amphibole I-beam. This mechanism has not been observed, although for triple I-beams it might be expected to be the more stable one.

Screw dislocations and fibre growth. In terminations of the types shown in fig. 13 and modelled in figs. 11 and 12 we have seen that there is a screw component of $\frac{1}{2}c$ in the dislocation involved. Since in these structures there are two layers of both tetrahedra and octahedra per unit cell in the *c*direction it seems probable that this screw component could serve to propagate growth along the fibre direction. Moreover, it is to be noted that its



FIG. 17. Model of a mechanism for sideways stepping of a multiple lamella along (130).

presence does not lead to any diffraction contrast in the image. This fact throws doubt on the conclusion of Veblen and Buseck (1980) that there are no screw dislocations of ordinary type along [001] in their anthophyllite fibres. Such dislocations would only be made visible by diffraction contrast in these images if they involved some slight distortion in the direction of the *c*-axis. In absence of such distortion they would not affect the regularity of the projection of the structure down [001] and so would not affect the diffraction conditions in the [001] zone. The rigidity of the amphibole *I*-beams would tend to inhibit any such distortion, and the lack of diffraction effects in fig. 13 (where we know that a



FIG. 18. Electron micrograph of anthophyllite asbestos down the *c*-axis. A lamella of talc at the top of the photograph appears to be in process of extending downwards from the right by incorporating lamellae of amphibole theorem two at a time Seeh here $x \in 0$.

I-beams two at a time. Scale bar = 50 Å.

screw component must be present) provides experimental evidence for the invisibility of screw dislocations in this orientation.

The propagation of multiple-chain lamellae. Veblen and Buseck (1980) have pointed out that the existence of a hole at a coherent termination of a single lamella of multiple I-beams (such as that of sextuple I-beams shown in fig. 6) would provide a means of access to solutions which could effect the chemical alteration involved in the extension of the multiple lamella. Such a termination may therefore be an arrested stage in the growth of such a lamella within the amphibole structure, and this interpretation gains support from fig. 18. This shows a lamella of indefinite width (i.e. a talc-like alteration product) which is progressively widened by two I-beams at a time. Thus a simple step-wise mechanism is available for the formation of lamellae of n-tuple *I*-beams where *n* is of the form 4N + 2, or for the broadening of lamellae of triple, quadruple, and quintuple I-beams. It does not, however, provide a mechanism for the formation of lamellae of these latter three types, which are in fact among the commonest. Coherent terminations of lamellae of these types can only occur co-operatively, and in the case of odd *I*-beams involve a displacement of a substantial volume of the structure by c/2, as shown in fig. 5. It therefore seems probable that these lamellae are often growth features rather than alteration features.

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