Crystallographic studies of some Norwegian aventurinised feldspars by optical, X-ray, and electron optical methods

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Four aventurinised oligoclases from SE Norway have non-feldspathic inclusions of which the majority are based on hematite-like structures. Detailed examination of these inclusions, however, shows considerable variability in diffraction data suggesting that they do not have a regular $\alpha - Fe_2O_3$ structural arrangement; this is interpreted in terms of unusual and sometimes irregular dispositions of Fe^{3+} (and possibly other cations) in the approximately hexagonal close-packed oxygen framework. There are minor amounts of other inclusions, but the only certain identification is that of alkali feldspar due to anti-perthitic exsolution. Data have been obtained on the mutual orientation of the feldspar and hematite-like inclusions, which, whilst demonstrating some accord with previous observations, indicate that such relations are variable and complex. Monotactic control exerted by 'close-packed' oxygen directions is examined, but the evidence is inconclusive.

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Petrographic literature contains sporadic reports of non-feldspathic inclusions in feldspar crystals; in some cases the inclusions appear to have a regular crystallographic orientation within the host. A recent comprehensive survey of such occurrences is provided by Smith (1974, vol. 2: 623 ff); this emphasises how difficult it is to correlate these occurrences with any systematic process or causal mechanism due to the paucity of experimental data which probably depend critically upon local geological conditions. This contrasts with the common and well-documented feldspar-feldspar intergrowths (as in perthites etc.) where the petrological origins are generally clear and for which the host-inclusion crystallographic relations are not only established but explicable in terms of strain energy minimization across the co-existing feldspar interface.

Our interest in non-feldspathic inclusions was stimulated during examination of lunar plagioclase where, like many others, we have observed such intergrowths, occasionally with textural indications of some systematic process of formation. However, like their terrestrial counterparts, the extremely erratic occurrence of many different kinds of inclusion make it presently impossible to use such examples to establish any host-inclusion crystallographic relations or to speculate upon their origin. For all specimens, terrestrial and lunar, data are very incomplete, and the complex structures both of the host feldspar and the included minerals would make it difficult to interpret any structural control exerted by the host feldspar.

In most cases where there has been any discussion of regular textural orientation of the inclusions the recurrent explanatory themes are of exsolution from a host feldspar and/or chemical alteration and replacement during or after feldspar formation. To shed any light on such problems, it is desirable that the non-feldspathic inclusions should have relatively simple structures and be of comparatively widespread occurrence; it was for this reason that our attention turned to aventurinised feldspars where the principal inclusions are of iron oxide (a relatively simple structure) and where some regular relationship of the inclusions to the host feldspar has already been recognised. In this first paper we are concerned with crystallographic aspects of such intergrowths, whilst a second paper will describe some heating experiments and discuss their relevance to the genesis of the textures.

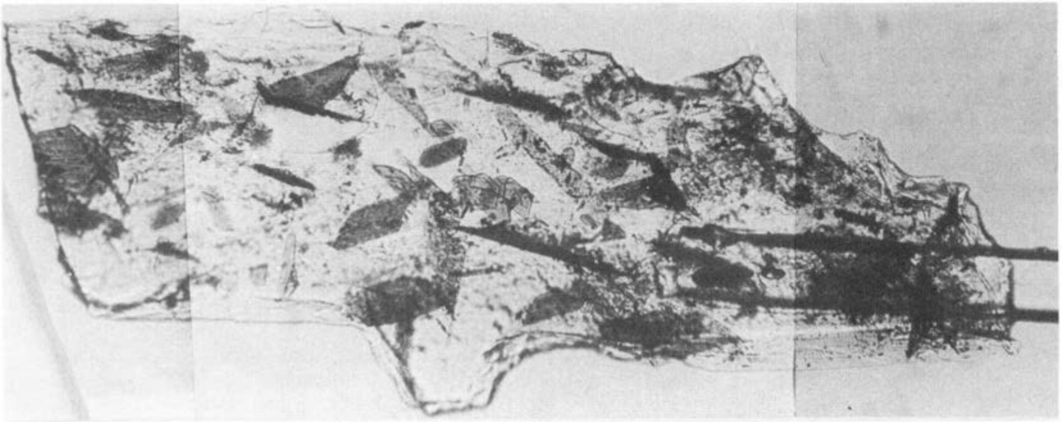

Fig. 1. An (001) cleavage flake from an aventurinised feldspar crystal from Arendal (specimen 3) mounted for single crystal X-ray studies, showing the dominant inclusions as well-developed platelets.

Summary of earlier work

The adjective 'aventurinised' refers strictly to an orange-red schiller effect produced by regularly arranged inclusions (often thin plates) in the host mineral; these inclusions can be of widely varying materials but in feldspars they are principally of an iron oxide, usually said to be hematite ($\alpha - Fe_2O_3$). In feldspars with the most marked schiller the oxide has recognisable morphological features in the form of thin lamellae of regular or distorted hexagonal shape (occasionally with more complex or irregular appearance) with apparently restricted orientation in the feldspar host; for some specimens with weaker schiller effects the platelets are less dominant but the oxide is still present, often as fine needles. The appearance of a typical cleavage flake showing well-developed platelets can be seen in Fig. 1; it should be stressed that although these lamellae (and needles) are numerous they are very thin so that the inclusion/host volume ratio is very low. Some details of the precipitate morphology for the present aventurinised feldspar specimens obtained by scanning electron microscopy have already been published (Copley & Gay 1978).

Such observations are based mainly on the comprehensive descriptions of aventurinised feldspars from a wide range of occurrences given by Andersen (1915). Using a reflecting goniometer to examine specimens with well-developed platelets he determined that the lamellae were mainly parallel to the (112), ($1\bar{1}2$), (150), and ($1\bar{5}0$) planes of the host feldspar; other orientations were occasionally present. He could not, however, determine whether there were any consistent orientations for the edges of the lamellae and was therefore unable to establish any mutual orientation(s) between the feldspar axes and those of the inclusions; later authors appear to have accepted or confirmed Andersen's observations. From the general appearance of the lamellae and chemical tests he concluded that they were hematite (rather than goethite as had been suggested earlier). It is a tribute to the quality of Andersen's work that over fifty years passed before Kraeft & Saalfeld (1967) recognised that the inclusions were not homogeneous. They distinguished three main types:

1. Red, orange, yellow transparent plates mainly on (112), (150) and ($1\bar{5}0$) feldspar planes, often showing the hexagonal outlines described by Andersen.

2. Dark red, opaque inclusions with transparent patches; these are often clustered as if they are relics of a reaction which has destroyed an original platelet, but occasionally occur as needles parallel to type 1 inclusion and rarely as rims to such inclusions. The authors infer a close relationship between types 1 and 2; there is some evidence of a transformation on heating, but both types show slightly anomalous electron diffraction patterns compared to basal sections of $\alpha - Fe_2O_3$ (these observations will be discussed more fully later):

3. Completely isolated transparent inclusions; these are not very common and, although clearly unrelated to the oxides of types 1 and 2, were not identified.

Specimens used in this work

Four specimens from aventurinised feldspar localities in SE Norway were chosen for investigation; preliminary examination was made to see which specimen(s) would be most suitable for detailed study. Preliminary data obtained for the host feldspar and the inclusions are given in Table 1. Only specimen 3 shows a preponderance of the well-developed lamellar precipitates described in earlier work; the other specimens have mainly acicular precipitates, and, when present, the sporadic orange platelets are difficult to examine optically (particularly for orientational study). At this stage we cannot offer any explanation for the relative development of the acicular or lamellar morphology of the inclusions except to note that it does not appear to be dependent on the composition or final structural state of the host plagioclase.

Since specimen 3 appears to be the most comparable to those described by Andersen (1915) and others, it was used for most of the detailed study. Apart from morphological aspects of the inclusions already described above, careful examination of thin sections provides other information about the lamellae and other precipitate forms. The platelets are very thin (often less than 0.5 μ) showing a considerable variation in colour from pale yellow to dark red. Examination on the universal stage demonstrated that the colour depended primarily on the optical path length in the precipitate; similarly oriented lamellae can display variation in colour due to change in thickness, and for an individual platelet the colour deepens as its orientation is changed to give a greater optical path in the inclusion. Whilst some of the lamellae are uniform in colour, others show variations, usually gradational but occasionally sharp; a marked colour boundary may be due to the interpenetration of platelets observed by scanning electron microscopy (Copley & Gay 1978). During the optical examination of this specimen it was difficult

Table 1

Specimen number / *Locality*		1 Bjordam	2 Havredal	3 Arendal	4 Havredal
Feldspars					
Cell constants					
	a (Å)	8.164 ± 0.003	8.142 ± 0.013	8.153 ± 0.005	8.194 ± 0.009
	b (Å)	12.843 ± 0.002	12.824 ± 0.011	12.843 ± 0.004	12.812 ± 0.008
	c (Å)	7.124 ± 0.002	7.143 ± 0.006	7.127 ± 0.003	7.143 ± 0.006
	α (°)	93.777 ± 0.023	93.885 ± 0.087	93.822 ± 0.037	94.023 ± 0.073
	β (°)	116.390 ± 0.017	116.458 ± 0.060	116.410 ± 0.033	116.534 ± 0.061
	γ (°)	89.359 ± 0.019	88.668 ± 0.122	89.327 ± 0.034	88.518 ± 0.079
2Vγ		92.5 ± 1°	84.0 ± 1°	96.0 ± 1°	82.0 ± 1°
An content		An_{24-25}	An_{10-12}	An_{23-26}	An_{10-12}
Structural state		Low	Low–intermediate	Low	Nearly low
Inclusions					
Shape		Mainly needles up to 0.05 mm length, diameter 3–5 μ; rare platelets up to 0.2 mm diameter with rather irregular shapes	Needles, 3–5 μ diameter often up to 0.15 mm length, occasionally up to twice this length	Mainly platelets up to 0.2 mm diameter (often smaller), generally of regular shape; rare colourless irregular blebs	Mainly needles up to 5 μ diameter and 0.05 mm length; rare platelets up to 0.1 mm diameter with rather irregular shapes
Colour		Pale orange	Yellow-orange	Variable, ranging from pale yellow to dark red, even opaque occasionally	Orange

Notes: All X-ray data on host feldspars were obtained from diffractometer records, and the optic axial angles by universal stage methods. The An content was assessed from these data, and checked by K-exchange (Viswanathan 1971, 1972); only for specimen 3 was confirmation sought by probe techniques. The structural state was determined from a knowledge of optical and X-ray parameters in combination with the An-content. More detailed description of the inclusions in specimen 3 is given in the text and by Copley & Gay (1978).

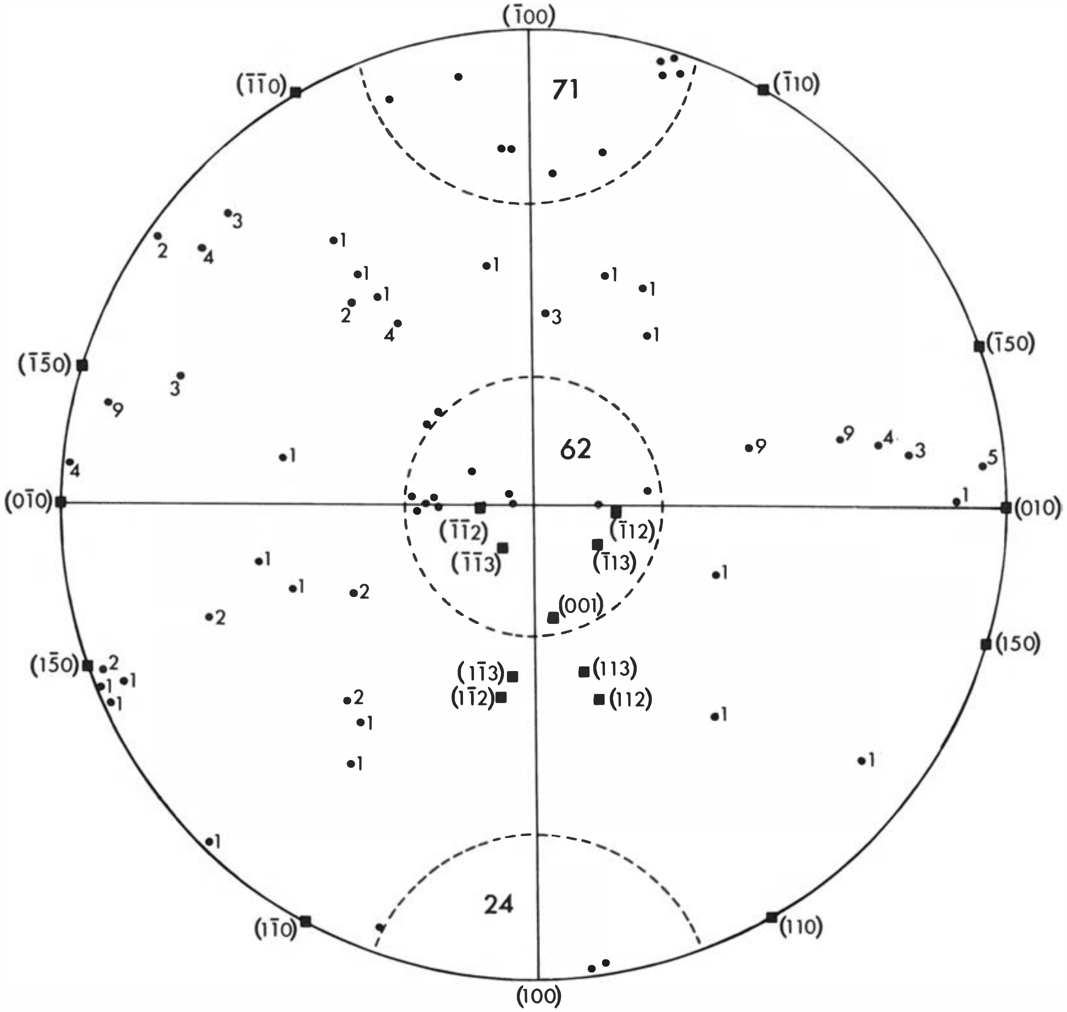

Fig. 2. A composite stereogram showing the poles of normals to plate-like inclusions relative to crystallographic directions for the host feldspar crystals. Dots show the location of poles with the adjacent figures representing the numbers of platelets in the corresponding orientations; within the areas defined by broken lines there are significant concentrations in orientations indicated by the numbers associated with each area. The squares show the location of feldspar poles previously reported as coincident with those of the platelets.

to recognise any clear distinction between types 1 and 2 precipitates defined by Kraeft & Saalfeld (1967), though some inclusions (of any colour or thickness) do show apparently colourless areas. There are also rare black rounded precipitates of small diameter (up to 0.05 mm) usually with a deep red centre, as well as occasional colourless blebs which could well be the type 3 inclusions of Kraeft & Saalfeld. From this qualitative work, we conclude that for specimen 3 the majority of the inclusions are lamellar platelets of non-feldspathic minerals of an identical or closely-related nature; however there are probably at least two other components within the host feldspar of minor sporadic occurrence.

For the platelets a large number of universal stage type measurements were made to determine the orientation of their normals with respect to host feldspar axes. The results obtained from three sections perpendicular to [001] and two sections parallel to (010) are plotted on the composite stereogram of Fig. 2. This shows a surprisingly wide scatter of possible orientations in this specimen, though there are some regions of relatively high density; in particular, there is a pronounced cluster around the [001] direction of the feldspar, with another marked concentration near (100). Of the orientations described by Andersen, the central cluster is near some of his specific feldspar planes (particularly (112) and ($1\bar{1}2$) which he describes as of common occurrence) and there is occasional correspondence with other feldspar planes mentioned in his work; on the optical evidence obtained from this specimen, however, it would be unwise to formulate particular and limited feldspar planes for the development of the lamellae. Turning to any correlation between directions within the plate-like inclusions and crystallographic directions within the host, the observations are even less promising, and, like Andersen, we can provide no evidence for any systematic relationship. At this point it seems that any host-intergrowth orientational controls in aventurinised feldspars are more variable than might appear on casual inspection, but their possible existence (and origin) will be discussed again later.

X-ray and electron-optical examination

Even in specimens in which there seems to be a high inclusion density the inclusion/host volume ratio is very low. No evidence for any non-feldspathic materials could be observed on any X-ray diffractometer powder trace but we have used standard range, heavy exposure single-crystal oscillation photographs for all four specimens, to see whether any inclusions can be detected.

The only previous electron-optical studies were those of Kraeft & Saalfeld (1967), who examined diffraction patterns from isolated basal platelet sections of types 1 and 2 obtained by transmission methods; their results will be discussed later. We have tried to prepare ion-beam thinned feldspar sections from all four specimens. The nature of the inclusions caused some difficulties in this method of preparation but satisfactory mounts for detailed study were obtained from specimens 1 and 3; such mounts were advantageous (by comparison with the basal flakes used by Kraeft & Saalfeld) in that a variety of orientations of both inclusions and host feldspars could be examined by 100 kV transmission electron micrography and diffraction.

Feldspars

The single crystal X-ray patterns revealed widespread sub-microscopic albite-twinning, but in general were characteristic of normal low-temperature plagioclase host of the appropriate composition. Some crystals (particularly from specimen 3) show anti-perthitic exsolution similar to that described by Bown & Gay (1971); in these anti-perthites the a* direction is common to all feldspar components. Structurally such a direction is one of the shortest between adjacent cations on planes parallel to (010) for a monoclinic feldspar; anti-perthitic exsolution could occur after segregation of potassium ions in this direction followed by an inversion to triclinic symmetry on cooling. This would lead to the two slightly different orientations of exsolved alkali feldspar component in a host plagioclase observed both in this and earlier work.

Electron-optical work on host feldspars adds little; plentiful albite twinning, minor anti-perthitic exsolution, and occasional dislocations can be seen. Specimens 2 and 4 show evidence of spinodal decomposition, whereas specimens 1 and 3 do not, in accord with the two compositional ranges of Table 1.

Non-feldspathic inclusions

Heavily-exposed X-ray single crystal photographs do show some maxima additional to those expected from the feldspar component(s); these occur as more or less sharp spots, as streaks along constant θ loci or sometimes as associated spots and weaker streaks. Superposition of a standard powder film of hematite ($\alpha - Fe_2O_3$) demonstrates that the majority of these maxima have d-values which fit this pattern; hand-picked coloured lamellae (from crushed aventurinised specimens) glued to a glass fibre also gave segments of powder arcs which apparently confirm this identification. The relatively few detectable maxima which do not accord with this interpretation are at variable θ values from crystal to crystal; some, occurring at lower θ values, do appear sufficiently regularly enough to imply possible inclusions of a magnetite type but without other confirmatory evidence any conclusion concerning these (or any other) non-hematite maxima is impossible. The standard range photographs from many single crystals, both from the same and different specimens, shows no obvious consistency in the position of $\alpha - Fe_2O_3$ maxima from crystal to crystal. All evidence confirms that there cannot be complete topotactic correspondence between the host feldspar and these inclusions but the X-ray data do not exclude monotactic control of particular linear directions at interfaces; such possibilities are best investigated in the electron diffraction studies which follow.

In the previous electron diffraction work by Kraeft & Saalfeld (1967), the few basal sections of coloured platelets that were examined gave rather inconclusive results. One red lamella showed a pattern similar to that expected from $\alpha - Fe_2O_3$ if the intensities of reflexions were disregarded; four other red lamellae with a similar a-axis repeat (5.03–5.05 Å) showed variable intensities from different regions in a way which suggested a primitive cell; finally two red-brown lamellae gave patterns which could be indexed in terms of an a-axis repeat of about 5.92 Å for a primitive hexagonal cell, though the axes of this larger cell are rotated relative to those of the other diffraction patterns and the platelet morphology. These unexpected complexities make it difficult to recognise the particular iron oxide of the inclusions, though the authors considered that both $\alpha - Fe_2O_3$ and $\beta - Fe_2O_3$ (an ill-characterised polymorph proposed by Finch & Sinha (1957) during oxidation studies) were present as well as an unidentifiable oxide with the larger a-cell side.

The crude X-ray techniques described above confirm a hematite-like structure for these inclusions, and elsewhere (Copley & Gay 1978) we have demonstrated that their composition is consistent with Fe_2O_3. In our electron-diffraction studies it has been possible to examine a wide variety of sections of the inclusions in the thinned specimens; we have used basal (a*–a*), prismatic (a*–c*), and general sections to try to resolve the difficulties encountered by Kraeft & Saalfeld, and our observations and conclusions are now summarised. There is considerable variability in detail in diffraction patterns from different inclusions and often from different areas of the same particle, though all maxima are in positions which can be related to the basic oxygen stacking sequences of $\alpha - Fe_2O_3$. In our specimens, patterns expected from normal hematite were not observed. Some patterns can be indexed on a primitive hexagonal cell with a~5.03. c~13.7 Å, but these are less common than more complex patterns which, although based upon a primitive hexagonal cell, have a~5.92 Å and a less well-defined c-repeat of approximately 23 Å. Fig. 3 illustrates two basal patterns in a similar orientation, and a simple rotation through 30° about c* causes some reflexions to become coincident. These patterns were obtained from different areas of the same precipitate particle; in this case there was no obvious discontinuity between the areas, though in some a boundary can be seen (e.g. a core with a~5.03 Å and a margin with a~5.92 Å) and others show only one type of pattern. Fig. 4 shows a prismatic pattern of the more complex type; there is diffuse streaking ∥ c* (particularly noticeable on row lines for which h ≠ 0) with sharper maxima developing alternately weak and strong at positions corresponding to about 10/6 times the normal hematite c-axis length (the weak maxima are apparently absent for h = 0).

From such evidence we conclude that whilst the majority of the inclusions have compositions approximating to Fe_2O_3, their structures in our specimens are ordered or partially ordered forms different from that of $\alpha - Fe_2O_3$. The normal hematite structure consists of layers of oxygens arranged in approximately hexagonal close packing; the octahedrally co-ordinated iron atoms are regularly situated in 2/3 of the available interstices in accordance with rhombohedral symmetry to build up a hexagonal cell repeating after 6 oxygen layers to give the c-repeat of 13.7 Å. Re-arrangement of the cations could lower the symmetry from rhombohedral to hexagonal whilst retaining dimensions close to those of hematite or produce related cells of different symmetry and dimensions. In this way, for example, the relatively common pattern with a=5.92 Å would develop from a different cation arrangement. Fig. 5 shows two adjacent close-packed oxygen layers with the conventional hexagonal hematite cell outlined; an alternative hexagonal cell with axes rotated through 30° and a~5.9 Å can be chosen. The existence of a structure with such a cell and its periodicity in the c-axis direction will depend only on the cation arrangement; disorder will be manifested by diffuse streaking and regularity by the development of sharper maxima at positions corresponding to multiples of the oxygen layer separation as observed (cf. Fig. 4). With such an interpretation microstructural features could be

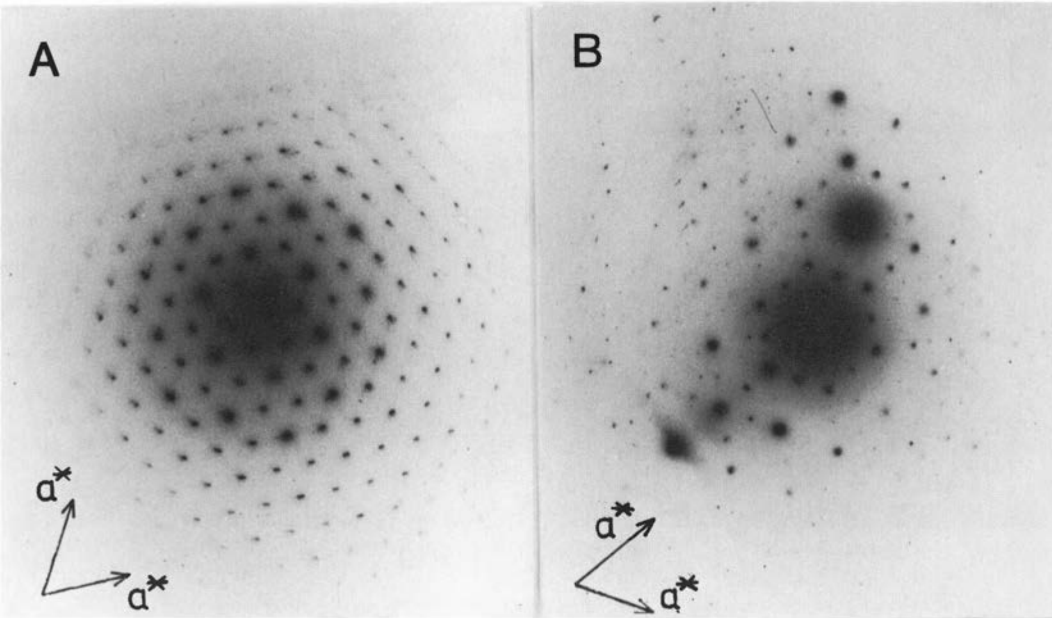

Fig. 3. Electron diffraction patterns from different areas of a basal section of the same inclusion; (A) can be interpreted on hexagonal axes with a~5.03 Å, whilst (B) can be indexed on similar axes rotated through 30° relative to those for (a) with a~5.92 Å.

expected in microscopic examinations; whilst some electron micrographs are featureless, some basal sections exhibit a domain microstructure and there is often a planar structure developed parallel to (0001) for prismatic sections (cf. Fig. 4).

Whilst our investigations accord with earlier work in confirming that the principal non-feldspathic component of aventurinised plagioclase is a predominantly ferric oxide, the structure of this oxide is unusual due to variations in the cation arrangements in an approximately hexagonal close-packed framework of oxygens. The origins of such structural modifications need further research. It is conceivable that they arise from the particular paragenesis of our specimens either directly due to the physical conditions of formation and/or indirectly in that the inclusions are not pure Fe_2O_3 but contain small amounts of other cations (e.g. Al^{3+}). In this context, it would be of interest to see if similar variations are found in 'hematite' inclusions observed in other minerals.

Feldspar-inclusion orientations

As described, X-ray data do not provide any conclusive evidence for structural control by the host feldspar crystal over the orientation(s) of the hematite-like inclusions. Using electron diffraction methods we have tried to establish whether there are at least any common directions in host-inclusion relations. Past experience shows that larger common anions are responsible for structural continuities in intergrowths and since the hematite-like inclusions are based on approximately close-packed oxygen planes, it is from these that any mutually compatible anion arrangements in host and inclusions might arise. The oxygen atoms of the feldspars are very irregularly arranged, but pseudo-cubic geometry in the morphology of crystals was known in the last century. Brunner (see Smith 1974, vol. 1: 39-43) used a topological approach to show how the T and O atom positions in feldspars could be related to distortions of simple close-packing; in particular the oxygen positions are derived from a c.c.p. pattern with layers normal to the feldspar c-axis and close-packed directions parallel to the feldspar b-axis. As Smith points out, this analysis does not imply that such features are immediately recognisable in any detailed examination of the feldspar structure, but they could provide a clue to any orientational control based on anion compatibility.

The platelet normal plot of Fig. 2, although

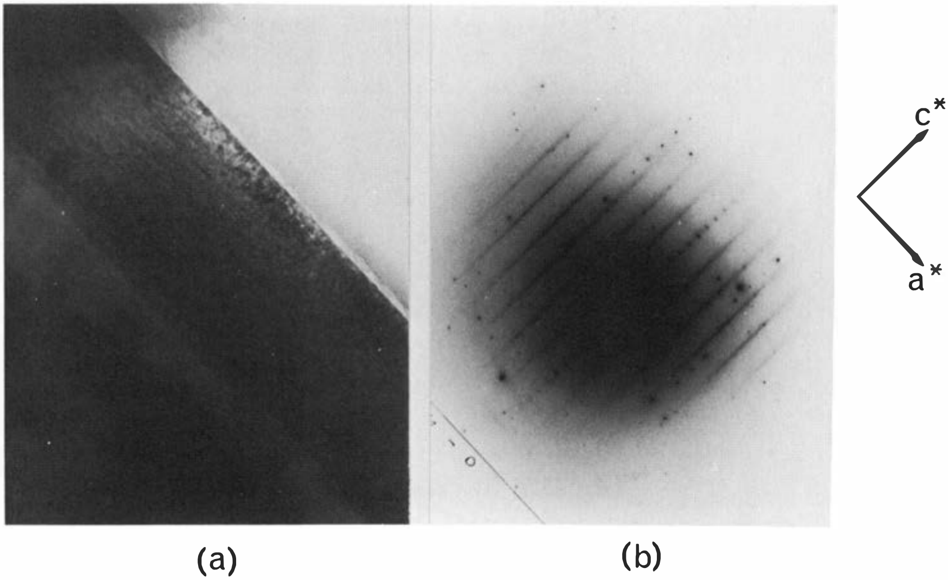

Fig. 4. Electron micrograph and diffraction pattern from a prismatic section of an inclusion; (a) shows the micrograph with a planar structure parallel to (0001), whilst (b) shows the diffraction pattern. Note the pronounced streaking and the maxima developed on row lines || c*.

showing a wide scatter, does show some encouraging features in that the common direction for the two greatest concentrations of lamellae normals is near the feldspar b-axis, a 'close-packed direction'. Unfortunately the limited electron diffraction data which we have been able to obtain do not significantly add to this

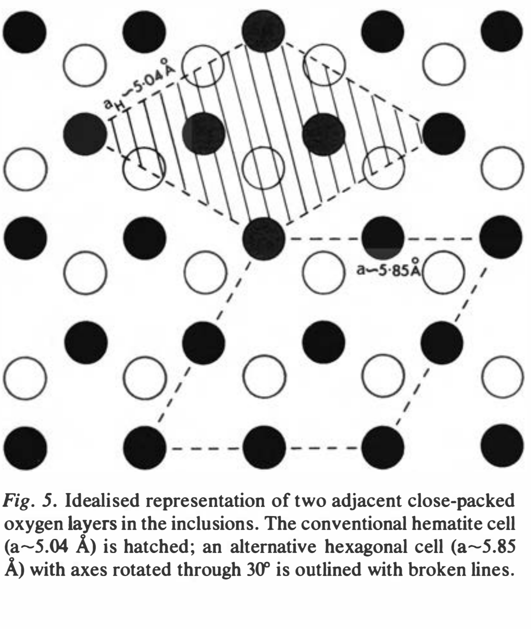

Fig. 5. Idealised representation of two adjacent close-packed oxygen layers in the inclusions. The conventional hematite cell (a~5.04 Å) is hatched; an alternative hexagonal cell (a~5.85 Å) with axes rotated through 30° is outlined with broken lines.

evidence. Often the c-axes of the feldspar and 'hematite' are parallel and there are sometimes other aspects (e.g. parallelism of host-inclusion a-axes) which could fit a hypothesis of matching 'close-packed' oxygen directions; however there are other observations which cannot apparently be accounted for by such simplistic interpretation. Any orientational control exercised by the oxygens of the feldspar framework must be regarded as unproven on the present evidence. Parallelism of the platelet inclusions is accepted as the cause of marked aventurinisation effects in feldspars (and other minerals), but in the present specimens, at least, the inability to recognise any regular structural origin may be significant in considering their paragenesis, a topic which will be discussed more fully after description of the effects of heat treatment in a forthcoming paper.

Conclusions

The majority of the non-feldspathic components in four aventurinised Norwegian plagioclases have hematite-like structures; although based on the $\alpha-Fe_2O_3$ structure, they exhibit a variety of ordered and disordered forms due to variable cation distributions in the oxygen framework.

Whilst their compositions approximate to Fe_2O_3, there may well be minor replacement of ferric ions by other cations leading to the different structural modification. Such inclusions are of variable colours in thin sections; this is due mainly to optical path lengths though minor changes could be caused by the different structural types. Although often occurring as thin lamellae of regular hexagonal or related shapes, they can often be found in acicular form. There may be very minor amounts of other non-plagioclase inclusions present, but only one of these, alkali feldspar arising from anti-perthitic exsolution, has been positively identified; such exsolved material could account for the occasional isolated colourless blebs seen in some specimens. Detailed study of the crystallographic relations between the plagioclase and the hematite-like inclusions shows orientations which are more variable than suggested in earlier work, and attempts to correlate the oxygen distributions in the two structures so as to account for the mutual orientation concentrations responsible for aventurinisation are inconclusive.

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