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THE IDENTIFICATION OF ORIENTED INCLUSIONS IN PYROXENE CRYSTALS

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Abstract

Simple *x*-ray techniques enable the identification of oriented intergrowths of minor components in "single" crystals. The methods developed have been applied to pyroxene crystals. Results are presented for the orientation and identification of augite, pigeonite and orthopyroxene in poly-phase crystals. Inclusions of ore minerals and amphibole can also be characterised.

Introduction

Not infrequently a single crystal of a mineral contains many smaller crystals of a different substance, produced for example by exsolution, alteration or partial inversion. Irrespective of the method of formation such a crystal composed of host and inclusions may be called an intergrowth. A distinction can be made between oriented intergrowths, in which there is a definite, constant, and structurally significant relationship between the axial directions of the host and inclusions, and disoriented intergrowths which have no such relationship.

Microscopic examination of an intergrowth often allows the identification of the minor components as well as the host, the determination of any fixed orientation of the optical properties and of the shape of the crystallites relative to the host, and hence their mutual axial orientations. However, should the crystallites be very small identification using the microscope may be uncertain, and in the limit the very presence of inclusions may be unsuspected. In such cases the methods of *x*-ray diffraction may aid the petrologist.

For identification alone the *x*-ray powder method may suffice, though it can never give information about orientation. It will not be successful, however, should the volume of inclusions be less than about 10% of the total volume, or should the powder patterns of the two phases be complex; for in the first case the pattern of the minor component will scarcely be seen at all, and in the second case the overlapping of the two patterns will make any identification uncertain. If the composite crystals are large enough for single crystal *x*-ray photography to be practicable (greater than 0.05 mm.) the resolution can be improved, for the pattern of the host crystal will be a series of spots instead of the lines given by a powder. Should the intergrowth be a disoriented one the inclusions will still give their own powder pattern, but the problem of overlapping is

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removed, and identification is easier. The most favourable case occurs if the inclusions are all parallel, for then the diffraction patterns of the individual crystallites superimpose on one another, giving a second "single-crystal" pattern. This concentration of intensity enables very small amounts of inclusion to be detected. If the identification can be made then the axial orientation can also be found; indeed the orientation not infrequently is helpful in the identification. Of course it is impossible by a diffraction method to determine the shape and macroscopic orientation of the crystallites, except to a limited extent in very special cases.

Thus for the study of oriented intergrowths the *x*-ray method is a particularly useful tool. In the rest of this paper the application of the method to many of the possible intergrowths in pyroxenes of the diopside-hedenbergite-ferrosilite-enstatite system is described. The techniques are simple and demand only slight experience in *x*-ray crystallography: further, it would not be difficult to apply similar methods to other oriented intergrowths.

METHOD

As the work requires a survey of the whole of reciprocal space, an oscillation photograph showing the projection of a three dimensional sample is more valuable than a Weissenberg or precession photograph showing only a two dimensional layer. For pyroxenes *z*-axis oscillation photographs were taken with FeKα radiation in two 15° ranges, symmetrically across the normals to (100) and (010). (CuKα radiation can be used, but fluorescent scattering from iron atoms gives a dark background.) Oscillation ranges chosen so that the photographs show some symmetry are easy to locate accurately by Laue photographs. These are desirable in that they enable diffraction spots from non-oriented crystallites, whether inclusions or adhering fragments, to be separated from the rest, for such spots will not be related by the symmetry. It should be emphasised that oriented crystallites will give spots related in position by the host symmetry, whatever their own symmetry or orientation. For example, when monoclinic augite lamellae separate from a crystal of ortho-pyroxene it is found that the two phases have the *y* and *z* crystallographic directions in common. Since $\beta \neq 90°$ for augite, there are then two possible orientations for the augite, related as by twinning about the normal to (100), which are equivalent with respect to the orthopyroxene. In the absence of disturbing factors the intergrowth should contain as many lamellae in one orientation as in the other, on the average, so that the compound crystal will have the same orthorhombic symmetry as the host. If there are only a few lamellae in the crystal the numbers in the two orientations may be quite different. In the diffraction pattern the augite spot positions, but not their intensities would be related by the orthorhombic symmetry. The principle is a general one, and in the rest of this paper is referred to as "the intergrowth symmetry principle." (The symmetry of the minor phase may, of course, be greater than that of the host.)

By comparing such photographs of composite crystals with "standard" photographs of the pure host material, taken under the same experimental conditions, the diffraction spots given by the host may be most easily separated from those due to the inclusions. From the latter the nature and orientation of the inclusions have to be found. The practice of comparing photographs is to be preferred to calculation of spot positions and intensities, as being quicker, and possible even when the cell size and crystal structure are unknown. If allowance is made for slight changes in position and intensities for different compositions, only two standard photographs (one for each oscillation range) are needed for each of the pyroxene structure types.

If from optical observation or otherwise, the nature and orientation of the inclusions can be postulated, a photograph of the pure substance taken in the suspected orientation can be compared directly with the "inclusion spots" on the photograph of the intergrowth. A good match of positions and relative intensities provides confirmation, and the intergrowth photograph can itself be used as a "standard" in further investigations.

In a more difficult case it may not be possible by microscopic examination to identify the inclusions from among several substances, or to get any information about orientation. For example, an ore mineral exsolved from a pyroxene is often difficult to characterise optically. The identification may then be carried a stage further by comparing the inclusion spots on the intergrowth photograph with powder photographs, (taken in the single-crystal camera) of each of the possible materials. If for one of the powder photographs each inclusion spot lies on one of the powder rings a tentative identification is made (of course it is not necessary for there to be a spot on every ring). The orientation of the intergrowth must next be found. This may be done (1) by measuring the layer line spacing of the intergrowth spots, if these lie on layer lines; (2) by indexing the spots from the indexed powder photograph; (3) by arguments as to structural similarity between the two phases; (4) by a combination of these approaches suited to the particular example. The identification is only completed by a satisfactory correspondence between the inclusion spots and a "standard" photograph taken in the predicted orientation.

In the most difficult circumstances there is no previous clue to the identity of the inclusions. Then measurement of the *d*-spacings of the inclusion spots on a series of oscillation photographs and use of the A.S.T.M. index may lead to a definite identification, but this procedure should only be used as a last resort; fortunately it is rare that no suggestion as to the identity can be made.

By these methods the orientations of many of the common intergrowths in pyroxene crystals have been determined; the following sections describe the results which, it is hoped, may be of use to other investigators. Discussion of the structural significance of the results is held over for a subsequent paper. It is convenient to treat separately the cases of intergrowths of pyroxenes in pyroxenes, and other inclusions in pyroxenes.

Pyroxene Intergrowths in Pyroxene Crystals

The three main pyroxene structure types may be called augite (monoclinic, $C2/c$, a 9.7 Å, b 8.9 Å, c 5.25 Å, β 106°) pigeonite (monoclinic, $P2_1/c$, a, b, and c as for augite, β 109°) and hypersthene (orthorhombic, $Pbca$, a 18.2 Å, b 8.9 Å, c 5.2 Å). Ca-rich pyroxenes such as diopside, salite, hedenbergite and augite have the augite structure, Ca-poor clinopyroxenes the pigeonite structure, and all orthopyroxenes (except protoenstatite) the hypersthene structure.

We have examined five different combinations of host and lamellae.

(1) *Augite with (001) pigeonite lamellae*

In agreement with Morimoto (1956) it is found that the directions of the x and y axes of the two minerals are the same, while the z axes are inclined at about 3° to one another, by reason of the different β angles, (Fig. 1*a*). On oscillation photographs taken with the z-axis of augite as oscillation axis the layer lines through the pigeonite spots are twisted away from the straight augite layer lines, as shown in Figs. 2 and 3.

(2) *Augite with (100) hypersthene lamellae*

The two phases share the y and z axis directions, as shown in Fig. 1*b*, so that on the standard z-axis photographs the hypersthene spots occur on the augite layer lines. As the position of the spots are different, and those due to hypersthene show more symmetry, the two sets are easily distinguished.

(3) *Augite with both pigeonite and hypersthene lamellae*

The axial arrangement is a simple combination of the previous two, and so is the resulting photograph. Fig. 4 shows the three-phase combination: the spots from the different structures are readily identified.

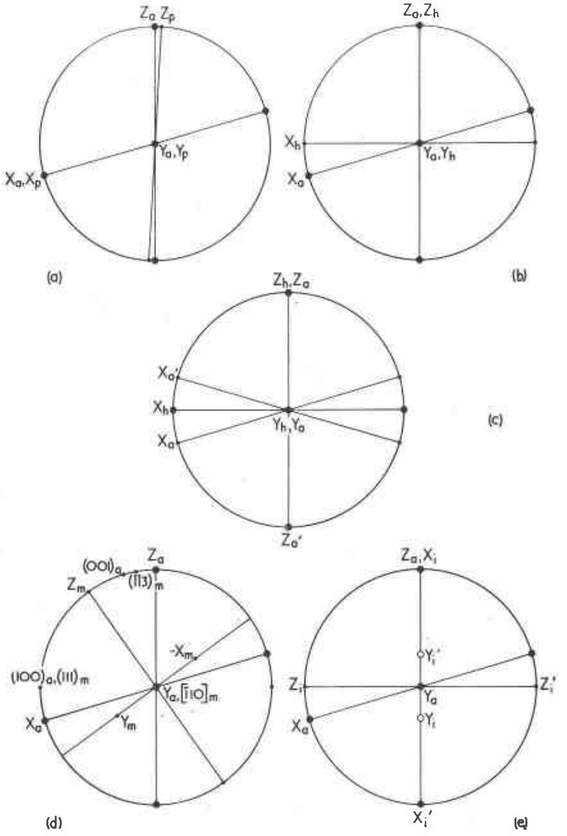

FIG. 1. Axial orientations in pyroxene intergrowths. (*a*) augite (*a*) with pigeonite lamellae (*p*). (*b*) augite (*a*) with hypersthene lamellae (*h*). (*c*) hypersthene (*h*) with "twinned" augite lamellae (*a*, *a'*). (*d*) augite (*a*) with exsolved magnetite (*m*) in the first orientation. (*e*) augite (*a*) with "twinned" ilmenite (*i*, *i'*).

(4) *Pigeonite with (001) augite lamellae*

The relative orientation is exactly the same as in (1) and the photographs are similar to Fig. 3, except that the pigeonite spots would be the stronger set, and as the pigeonite *z*-axis would have been taken as oscillation axis, the augite layer lines would be twisted and those of the pigeonite straight.

(5) *Hypersthene with (100) augite lamellae*

As in case (2) the *y* and *z* axial directions are in common, but now in accordance with the symmetry principle stated earlier, the augite lamel-

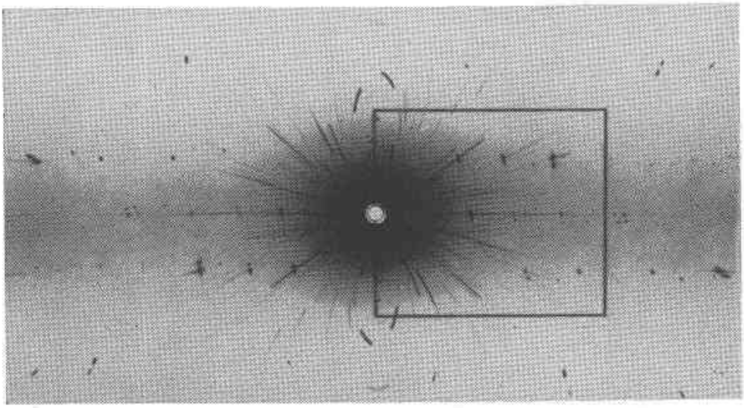

FIG. 2. Oscillation photograph about z-axis of an augite crystal containing pigeonite lamellae, taken with filtered Fe radiation. At the centre of the oscillation range the x-ray beam was parallel to the diad y-axis. The black lines indicate approximately the region illustrated for different specimens in Figs. 3 to 8. With the exception of Fig. 4, all diffraction patterns were obtained with filtered Fe radiation. [Specimen from Mt. Wellington, Tasmania.]

lae are in two orientations, as shown in Fig. 1*c*. Fig. 5 shows the type of photograph obtained: in this case there is less augite in one orientation than in the other.

Other combinations of phases and other orientations are sometimes found, but these five are the most common, and they persist even when additional phases such as ores and amphibole are present. No list of ξ, ζ coordinates for the spots of the various phases is given since it is defi-

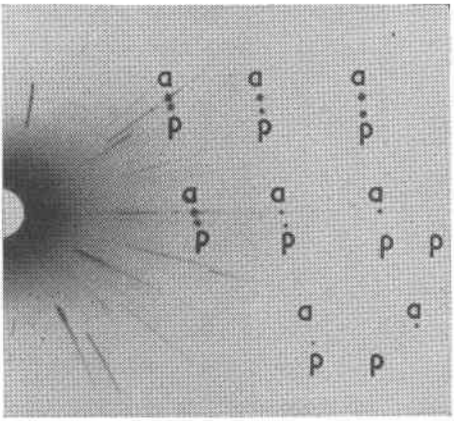

FIG. 3. Augite (a), with exsolved pigeonite (p). [Specimen from Skaergaard Intrusion, E. Greenland. No. 1691.]

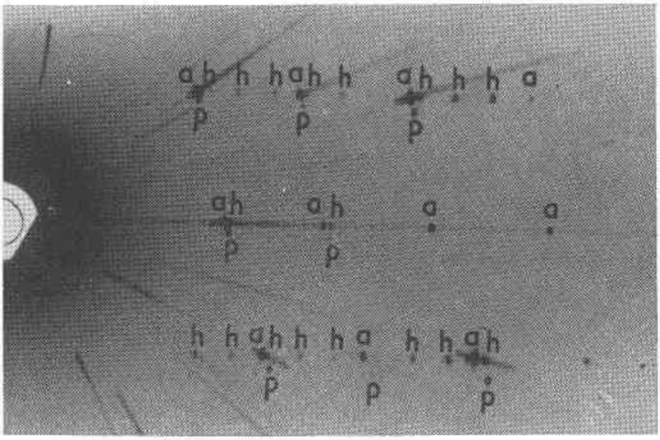

FIG. 4. Augite (*a*), with exsolved pigeonite (*p*) and hypersthene (*h*) (filtered Cu radiation). [Specimen from Skaergaard Intrusion, E. Greenland. No. 4087.]

nitely to be recommended that "standard" comparison photographs should be made by anyone using the method.

OTHER INTERGROWTHS IN PYROXENE CRYSTALS

The examples given in this section are those which the authors have encountered and examined; others are certainly to be found, and could be treated by the same methods.

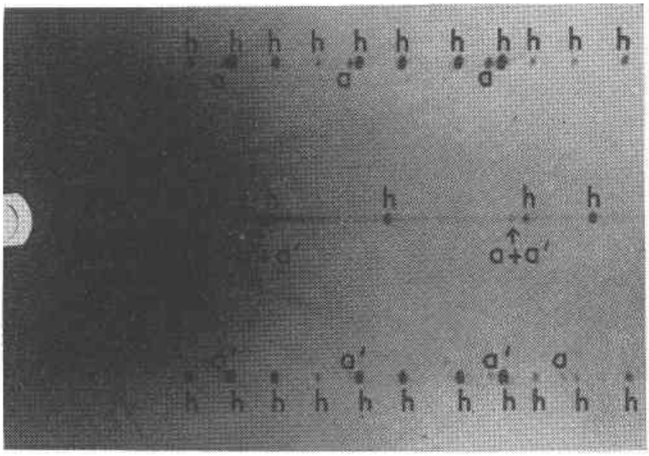

FIG. 5. Hypersthene (*h*) with unequal amounts of "twinned" augite (*a*, *a'*). [Specimen from Bushveldt Complex, S. Africa.]

(1) *Magnetite and spinel*

Magnetite may occur as thin plates or rods lying in the (100) and (001) planes of augite. Two axial orientations are found, which are thought to correspond to these two macroscopic dispositions. In the first of these the axes are arranged as in Fig. 1*d*, the (111) plane of magnetite being parallel to (100) of augite. The second orientation is the same but for a rotation of approximately 7° about the $[\bar{1}10]$ magnetite axis so as to bring $(\overline{11}3)$ of magnetite parallel to (001) of augite. The two orientations seem to be independent, for the relative proportions present can vary in different crystals from the same specimen. Magnetite has a spinel-type crystal structure, and it would be expected that any other spinel intergrowth with augite should show the same orientation as magnetite. (Indeed without careful intensity measurements it is impossible to prove from the *x*-ray evidence alone that the phase assumed to be magnetite is not another spinel of approximately the same cell size.) Fig. 6 shows an augite host with, among other phases, both magnetite and the iron-aluminium spinel hercynite in both orientations.

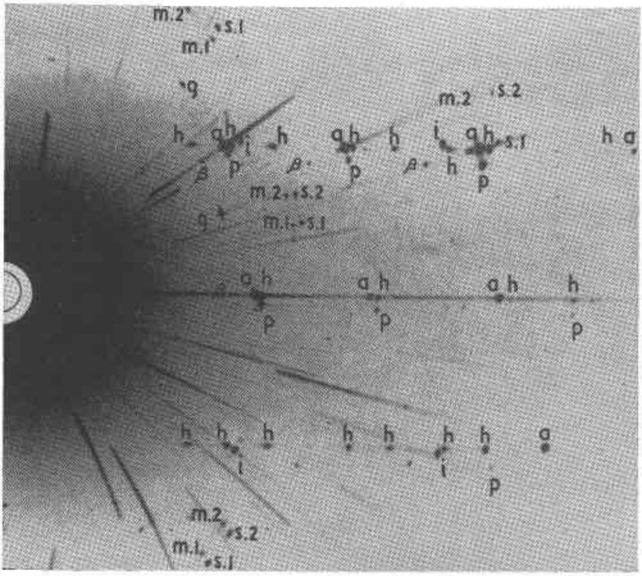

FIG. 6. Augite (*a*), with both pigeonite (*p*) and hypersthene (*h*). Other phases are ilmenite (*i*), magnetite and spinel (probably hercynite) each in two orientations (*m*.1, *m*.2, *s*.1, *s*.2). *g* indicates spots from a disoriented fragment, and *β* those produced by FeK*β* radiation. (A few spots due to clino-amphibole are also present but are unmarked.) [Specimen from Kragerö, Norway.]

Natural hypersthenes may also contain magnetite: this is found to have the first orientation described above for augite, together with the "twin" about the normal to (111) of magnetite demanded by the intergrowth symmetry principle. When iron-bearing orthopyroxenes are heated in air magnetite is formed in the same orientations as for the natural material; an example of this is shown in Fig. 7.

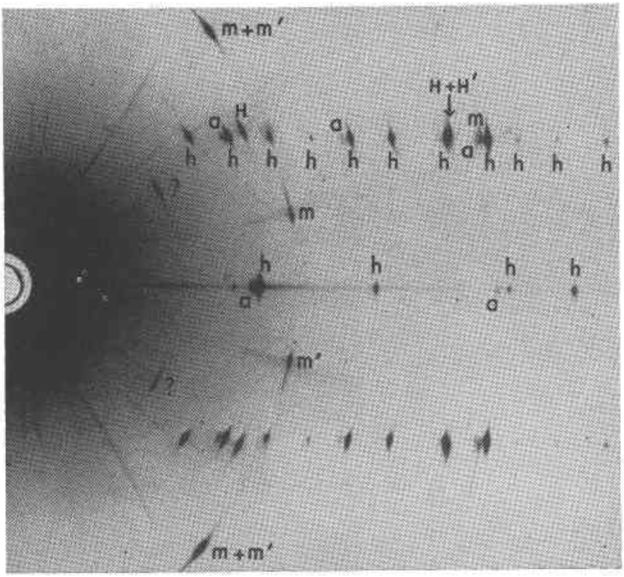

FIG. 7. Initially single-phase volcanic hypersthene (h), which on being heated at 1050° C. in air has exsolved "twinned" augite (a, a'), magnetite (m, m') and haematite (H, H'). The origin of the short streaks marked ? is uncertain: they may be from an oriented form of silica. [Specimen from Koolau, Oahu.]

No naturally occurring example of an intergrowth of magnetite and pigeonite has been examined, but one of the effects of heating single-phase pigeonite in air is to form magnetite in the two orientations as described for augite.

(2) *Ilmenite and haematite*

Associations of ilmenite and augite occur naturally, and the axes of the trigonal ilmenite are arranged as shown in Fig. 1*e*, "twinned" to conform with the intergrowth symmetry principle. Fig. 6 shows the positions of the ilmenite spots.

Ilmenite is sometimes found intergrown with hypersthene: the arrangement of ilmenite in the two positions of Fig. 1*e* has effectively orthorhombic symmetry,* and the same arrangement is found in hypersthene. When hypersthenes are heated in air some haematite is formed as well as magnetite. Haematite is structurally very similar to ilmenite, and it takes up the same orientations: because of the difference in cell sizes, however, the two phases can be distinguished. Fig. 7 shows haematite spots.

Heating single-phase pigeonite in air produces haematite as well as magnetite, and the orientations found are the same as those for ilmenite in augite. Natural intergrowth of ilmenite and pigeonite was not encountered.

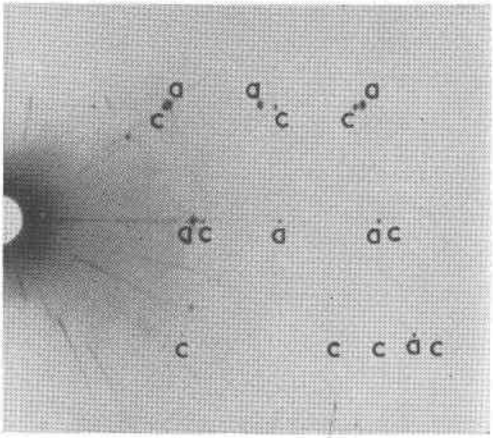

FIG. 8. Augite (*a*), with clino-amphibole (*c*). [Specimen from Ballachulish, Scotland, No. 343.]

(3) *Clino-amphibole*

Hornblende is a fairly common alteration product of augite, and is often orientated so that the x, y and z axial directions are all in common with that of the augite. (This is possible as the β angles of the two phases are virtually the same.) Fig. 8 illustrates this situation.

Hypersthene alters to twinned clino-amphibole rather than to orthorhombic amphibole. The y and z directions are common, and the amphibole is "twinned" about the normal to (100).

CONCLUSION

The fact that pyroxene crystals often contain other phases in a definite crystallographic relation enables the nature of the minor components

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* Strictly speaking ilmenite has point-group $\bar{3}$, so that four orientations are needed to give orthorhombic symmetry, but it has strong pseudo-symmetry $\bar{3}m$. Haematite has $\bar{3}m$ as point-group.

to be established by single-crystal methods even when they are present in very small amounts. A recommended procedure for such identification has been outlined, illustrated by typical photographs.

ACKNOWLEDGMENTS

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