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Epitaxic relations between coexisting pyroxenes

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SUMMARY. Many examples of epitaxic relations between coexisting pyroxenes are found in the picritic dykes that cut the Lewisian gneisses of the Lochinver area, Sutherland. In some dykes augite forms oriented overgrowths on the (100) face of orthopyroxene; in others orthopyroxene forms similar overgrowths on augite. A number of unusual variants of the first case are described together with a complex 'induced' twinning found in the augite overgrowths. Analyses of the coexisting pyroxenes suggest that the overgrowth relation has been facilitated by increased solid solution between the pyroxenes in an ordered sequence rather than simultaneously also seems to have been an important factor.

MANY examples of epitaxis ('parallel growth' or 'oriented overgrowth') are known among artificially produced crystals (van der Merwe, 1949; Buckley, 1951), the seeding of crystals on micas being particularly well documented. The basic requirements for oriented overgrowth are not only that there must be a two-dimensional similarity between the two structures at the plane of contact, but also that the cell parameters of the two crystals in these planes must either be very similar or be of a convenient ratio to allow a minimum of misfit. Thus it is the ease of stacking of the actual atoms of one crystal upon those of another that is the important factor controlling epitaxis.

While many instances of epitaxic overgrowth are known among natural minerals, there are few records of epitaxic relations between coexisting igneous or metamorphic pyroxenes. Brown (1957, p. 532) noted that, in the Skaergaard intrusion, pigeonite may occasionally grow on an adjacent augite so as to retain the same crystallographic axes, even to the extent of reflecting the twinning of the augite host. Such relations are to be expected in view of the similarity of structure and cell-dimensions of the two clinopyroxenes. Some form of structural continuity is usually, but not always, retained during the inversion of pigeonite to orthopyroxene, and, of course, is also found in the relationship of various exsolution lamellae to host pyroxenes. For instance, augite and orthopyroxene lamellae always have their crystallographic *c*- and *b*-axes and (100) planes positioned in common with their respective hosts, but in spite of this common structural plane, overgrowth relationships between augite and orthopyroxene do not seem to have been described previously. They are, however, relatively well displayed in the 2200-Myr-old bronzite-picrite and olivine-gabbro dykes near Lochinver, Sutherland.

Overgrowths of augite on orthopyroxene. The picrite dykes of the Lochinver area, first described by Teall (in Peach *et al.*, 1907) are coarse-grained igneous rocks composed of olivine, orthopyroxene, augite, plagioclase, a phlogopitic biotite, and accessory chromite and generally have orthopyroxene present in excess of augite. From textural evidence the order of crystallization of the major minerals follows

broadly the order as listed. However, both textural and petrological evidence (Tarney, in preparation) indicates that much of the orthopyroxene crystallized before the precipitation of augite, even when the two pyroxenes are present in roughly equal amounts. This differs from the situation found in many basic magmas where the beginning of precipitation of one pyroxene is generally followed fairly closely by crystallization of the other (Yoder and Tilley, 1962, p. 382) and may well be connected with the emplacement of these dykes at considerable depth (O'Hara, 1961; Tarney, 1963; Evans and Tarney, 1964), which may have favoured early crystallization of orthopyroxene (cf. Green and Ringwood, 1964).

As a result of this the abundant orthopyroxene crystals present in the magma may have formed sites favourable for the nucleation of augite, thus giving rise to the variety of oriented overgrowths, examples of which are shown in fig. 1 (A-G) and figs. 2 and 3. The examples given demonstrate that oriented overgrowths of augite on orthopyroxene always take place through initial nucleation of the new phase on the (100) face of the host in such a way that the (100) planes, c-axes, and b-axes of host and overgrowth coincide. This is the only plane that is structurally similar in the two pyroxenes, and is the plane on which diopsidic lamellae develop in the host orthopyroxene. It is clear in some cases that orthopyroxene has continued growing after nucleation of the augite, thus giving rise to the irregular boundaries. Some of these irregularities may also have arisen through nucleation of augite at two or more points on the same (100) face of the orthopyroxene, the overgrowths eventually coalescing. The augite overgrowths may also extend over on to the (001) faces of the host, the terminations often being wedge-shaped owing to continued growth of orthopyroxene, but few overgrowths have been observed to extend over on to (010), presumably because the ortho- and clino-structures are less compatible on this plane than on any other.

The overgrowth relationship frequently gives rise to some unusual textural features in thin section, particularly in certain late-stage products of the picrites that form small lens-shaped bodies within the dykes. These bodies are enriched in augite and plagioclase relative to the main dykes and show second-generation precipitation of more iron-rich orthopyroxene. Here the second generation orthopyroxene may form wide pleochroic rims over the earlier augite overgrowths. Such a case is shown in fig. 1, F (section cut parallel to (100)), where a number of idiomorphic augite inclusions all have the same orientation to the enclosing second generation orthopyroxene. The example demonstrates how a number of augites may nucleate and grow independently on the same (100) face of an orthopyroxene. In rare cases (fig. 1, G) the second generation orthopyroxene may itself nucleate apparently independently on an augite overgrowth, thus giving rise to double overgrowth groups.

While the presence of abundant early orthopyroxenes in the magma has probably favoured development of epitaxic relationships, it is pertinent to inquire whether other factors have had any control. The respective cell constants of the magnesium end-members of the two pyroxene groups,

enstatite
$$a = 18.24$$
, $b = 8.81$, $c = 5.19$ Å
diopside $a = 9.73$, $b = 8.91$, $c = 5.25$ Å

show that the b and c dimensions, which largely control the facility by which the

transformation from ortho- to clino-structure on (100) can take place, are not very different. It is likely, however, that the spacing along the c-direction exerts a greater influence on the ease of nucleation since less accommodation would seem possible along the strongly bonded pyroxene chains than at right angles to them. Since cell



FIG. I. Pyroxene overgrowth relationships. The ornament is somewhat diagrammatic for clarity. Unstippled: orthopyroxene. Stippled: augite (light and heavy stipple indicates twinned relationships). Heavy outline: olivine with magnetite-filled cracks. Scale bar in each case is 1 mm. A. Simple augite overgrowth on orthopyroxene. Section $\perp c$ -axis. B. Same. Section $\perp b$ -axis. Slight further growth of orthopyroxene. c. Augite overgrowth enclosing orthopyroxene on (100) and (001). D. Induced twinning in augite overgrowth. Section $\perp b$ -axis. E. Same. F. Idiomorphic twinned augite overgrowths overgrowth by more iron-rich second generation orthopyroxene. Section parallel to (100). G. Small iron-rich orthopyroxene from dyke margin showing external twinned augite overgrowths, twinned overgrowths, and also twinned internal overgrowths in holes in crystals. Section $\perp c$ -axis. I. Orthopyroxene overgrowth on augite in olivine gabbro dyke. Section $\perp c$ -axis.

parameters are dependent on composition (and hence also in this case would be the degree of misfit) this is obviously a factor to be considered.

Analyses of the coexisting pyroxenes from the centre (L. 119) and margin (L. 489) of the picrite dyke 1 mile W. of Loch Feith an Leothaid are given in table I. The picrite pyroxenes are interesting in that, contrary to the usual case with coexisting pyroxenes, the augites have a higher Fe/Mg ratio than the associated orthopyroxene; this agrees with the petrographic evidence that much of the orthopyroxene crystallized before

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augite. The pyroxenes are also unusual in that there is considerable mutual solid solution, as indicated by the respective calcium contents, and considerable substitution of aluminium, thus suggesting crystallization at high temperatures and fairly high pressures. Now calcium substitution in orthopyroxene has the important effect of

	Orthopyroxenes		Augites	
	L. 119	L. 489	L. 119	L. 489
SiO ₂	54.42	n.d.	52.81	n.d.
TiO ₂	0.13	0.09	0.22	0.52
Al ₂ O ₂	2.50	2.61	4.04	4.31
Fe ₂ O ₃	1.01	1.04	1.38	1.40
FeO	7:43	6.71	4.98	5.35
MnO	0.11	0.16	0.11	n.d.
MgO	29.96	30.85	18.66	19.70
CaO	2.95	2.53	16·49	15.42
Na ₂ O	0.20	0.17	0.60	0.66
K₂Ō	_	0.05		0.09
Cr_2O_3	n.d.*	n.d.*	0.83	n.d.
H_2O+	_		_	—
Total	99.31		100.12	
Si Al ^{iv} Ti Fe ³⁺ Fe ²⁺ Mn Mg Ca Na K Cr Wo	1.915 0.085 0.019 0.003 0.043 0.219 0.003 1.571 0.111 0.014 	$ \begin{pmatrix} (\dagger I \cdot 924) \\ 0 \cdot 076 \\ 0 \cdot 032 \\ 0 \cdot 002 \\ 0 \cdot 027 \\ 0 \cdot 196 \\ 0 \cdot 005 \\ I \cdot 608 \\ 0 \cdot 095 \\ 0 \cdot 012 \\ 0 \cdot 002 \\ \dagger (0 \cdot 022) \end{pmatrix} 2 \cdot 00 $	$ \begin{array}{c} 1 \cdot 909 \\ 0 \cdot 091 \\ 0 \cdot 081 \\ 0 \cdot 007 \\ 0 \cdot 038 \\ 0 \cdot 151 \\ 0 \cdot 003 \\ 1 \cdot 005 \\ 0 \cdot 042 \\ - \\ 0 \cdot 024 \end{array} $	$ \begin{array}{c} \uparrow (1 \cdot 887) \\ 0 \cdot 113 \\ 0 \cdot 067 \\ 0 \cdot 007 \\ 0 \cdot 038 \\ 0 \cdot 162 \\ \\ 1 \cdot 064 \\ 0 \cdot 599 \\ 0 \cdot 046 \\ 0 \cdot 004 \\ \uparrow (0 \cdot 023) \end{array} $
wo En	57/	4'9 82-5	54°7 54°9	57.2
Fs	13.3	11.6	10·4	10.7

 TABLE I. Chemical analyses and structural formulae of coexisting pyroxenes from Loch
 Feith an Leothaid

* The Cr_2O_3 content of an orthopyroxene of the same composition from another dyke (where more pure material was available) was 0.84 %.

† Equivalent values for Cr_2O_3 have been assumed in the recalculation to a six oxygen cell; likewise, the values for silica for L. 489 pyroxenes have been assumed by difference from 100 %.

increasing c (as well as a), while tetrahedral substitution of aluminium also increases c (Hess, 1952; Kuno, 1954). The orthopyroxenes are sufficiently endowed with Ca and Al (each up to 0.10 ions per six oxygens) for these substitutions to have an important effect on the c-parameter. The high proportion of orthopyroxene in solid solution in the augite would tend to decrease the c-dimension of the augite, the over-all effect

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being to reduce the misfit between the structures on (100). Besides, the high temperatures of crystallization would tend to minimize the effects of any remaining misfit. Thus there are probably three factors that combine to give an explanation of the fairly widespread development of augite overgrowths in these dykes. However, there are further manifestations of the overgrowth relationship, which are briefly described below.



FIGS. 2-5. Overgrowths. FIG. 2 (top left). (×15) Augite overgrowths on orthopyroxene. Section ⊥b-axis. Exsolution lamellae visible in orthopyroxene. FIG. 3 (top right). (×20) Same, double group. Sections ⊥b-axis and c axis respectively. FIG. 4 (bottom left). (×6) Irregular overgrown augite overgrowths (light coloured) in large poikilitic orthopyroxene. FIG. 5 (bottom right). (×30) Orthopyroxene overgrowths on augite in olivine gabbro dyke. Section ⊥b-axis. All crossed nicols.

Induced augite twinning. An interesting feature of the augite overgrowths is that they are frequently twinned, while augites present as separate crystals rarely show twinning. Moreover, the composition plane tends to be rather variable and the twins rather intergrown (see fig. 1, D, E, and F).

The common association of augite twinning with epitaxis is suggestive of a genetic relationship whereby augite may nucleate with two possible orientations on the orthopyroxene host. It is worth recalling that orthopyroxene itself is twinned on a unit cell scale with (100) as twin and composition plane. Thus each developing epitaxic augite nucleus has two possible alternative orientations to the host depending upon which half of the unit cell it is related to. Different augite nuclei developing with these twinned orientations on different parts of the host orthopyroxene lattice will subsequently interfere on further growth giving rise to the complex twinned overgrowths with variable composition plane. The result is probably a unique situation of



FIG. 6. Diagrammatic representation of crystallographic relationship of twinned augite overgrowths on orthopyroxene.

a crystal that, though not twinned itself, can nevertheless induce twinning of a somewhat unusual kind in another crystal species.

Oriented augite inclusions. Inclusions of one mineral in another generally point to the earlier crystallization of the inclusions or, when the latter bear a definite crystallographic relation to the host, to exsolution or replacement. Such oriented inclusions of augite in orthopyroxene can be seen in certain textural varieties of the Assynt picrites, but are too large to be attributed to exsolution (indeed 'Bushveld-type' exsolution lamellae are also present) and there is no evidence in favour of a replacement origin. These inclusions have the same epitaxic relation to the host orthopyroxenes as do the overgrowths described above, and here again, two

(twinned) orientations are found. Small augite overgrowths are generally found at the margins of the orthopyroxene crystals also, but while the overgrowths appear in all varieties of picrite, the oriented augite inclusions seem to be restricted to two particular petrological types, notably the coarse-grained, orthopyroxene-rich margins of the dykes and the coarse-grained poikilitic orthopyroxene rocks of the Loch Beannach picrite dyke.

The augite inclusions present in the large (1-3 cm diam.) poikilitic orthopyroxenes are irregularly distributed throughout the crystals, are of widely varying size (but all less then 1 mm) and are generally of irregular shape (fig. 4). The prefix 'poikilitic' here refers to the abundant olivine inclusions rather than to the augites; the olivines do not have any particular preferred orientation. The orthopyroxenes appear to have grown in situ (cf. Poldervaart and Taubeneck, 1960, p. 242), gradually enveloping the surrounding olivines and at the same time progressively reducing the volume of the interstitial liquid with its largely augite and plagioclase components. Under these conditions it is fairly easy to envisage incipient precipitation of augite in epitaxic fashion as the relative augite concentration in the liquid increased locally. However, with further crystallization of the orthopyroxene host the augites were enveloped and became inclusions. Considered in simple terms they are in fact 'overgrown overgrowths'.

A similar type of oriented augite inclusion occurs in the large orthopyroxenes of the orthopyroxene-rich margins of some dykes, but seems to have a rather different origin. Here the augites are found in irregular 'holes' inside the orthopyroxene crystals along with plagioclase and small amounts of biotite, but in contrast to the inclusions described above they generally display crystal faces against biotite and plagioclase, though their contact with orthopyroxene is invariably irregular. An example is shown in fig. I, H and the same crystal shows marginal augite overgrowths and irregularly shaped 'overgrown overgrowths'.

This type of augite inclusion has only been found in the coarser grained varieties of the orthopyroxene-rich margins of the dykes, the larger size of the crystals here being attributed to favourable diffusion conditions and rapid growth while the magma was still streaming upwards through relatively hot country rock (cf. Tarney, 1963). The irregular 'holes' are thought to represent pockets of liquid magma trapped inside the rapidly growing crystals, the augite component of which has subsequently nucleated and developed epitaxially inside its orthopyroxene host. Such augites are, paradoxically, 'internal overgrowths' if the usual terminology be extended to cover such occurrences.

Overgrowths of orthopyroxene on augite. The rare occurrence, noted above, of a second generation orthopyroxene forming overgrowths on an early augite is more commonly seen in the olivine-gabbro dykes of the Lochinver area. These dykes are probably the more iron- and alkali-rich derivatives of the picrites, and have a similar mineralogical assemblage, but with titanomagnetite and a brown hornblende in addition. A major difference as far as epitaxis is concerned, however, is the reversal of the crystallization sequence of the pyroxenes; here most of the augite crystallized before orthopyroxene, and as a consequence of this the margins of these dykes are augite-rich in the same way as the picrite dykes have orthopyroxene-rich margins. All examples of epitaxis found in these dykes show orthopyroxene overgrowths on augite (fig. 1, I, fig. 5). The overgrowths are not so common, however, as in the picrite dykes. There may be a number of reasons for this, but the fact that the augites in the olivine-gabbro dykes are not well crystallized but frequently give rise to distorted forms and multiple aggregates may have prejudiced nucleation of potential overgrowths.

Pyroxene overgrowth relations in other Lewisian dykes. A number of other Lewisian dyke types contain two pyroxenes (in fact probably the majority), though in some orthopyroxene is little more than an accessory. Overgrowth relations exist, but they are by no means as common as in the Lochinver picrites.

Of those examined, the picrite dykes at Braesclete, Lewis (Dearnley, 1963, p. 273) and at Gruinard Bay, Ross-shire (Bowes, Wright, and Park, 1964, p. 168) both show augite overgrowths on orthopyroxene. A norite dyke (N1) described by O'Hara (1962) from Badcall, Sutherland, also has a number of examples of twinned augite overgrowths on orthopyroxene. The chilled margins of this dyke contain abundant microphenocrysts of orthopyroxene, but not augite, thus indicating that a considerable

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number of sites favourable for nucleation of augite were available in the magma as augite crystallized. However, two-pyroxene dolerites similar to this norite occur near Loch Assynt (Tarney, 1963) but there are no overgrowths; the chilled margins of these dykes show only rare phenocrysts of augite and no orthopyroxene.

The normal 'Scourie' dolerites usually contain orthopyroxene, but in variable quantities. Overgrowths of orthopyroxene on augite can be seen, and further, may themselves be overgrown by primary green hornblende, which has epitaxic relations to both pyroxenes. The overgrowths always take place so that the *b*- and *c*-axes of hornblende and pyroxene coincide, thus leaving the chain structure undisturbed.

Conclusions. Overgrowths of one pyroxene on another seem to be favoured where many crystals of one species are present in the magma before crystallization of the other begins. It is possible, of course, that the slow cooling of many of these Lewisian dykes as a result of their intrusion into hot country rock may have been a major contributory factor in inducing nucleation of the overgrowth on the host. In the case of the Lochinver picrites it is equally clear that the high degree of solid solution between the two pyroxenes was an important factor in reducing the crystallographic misfit between overgrowth and host, and hence encouraging epitaxic relations. No analyses are as yet available for the coexisting pyroxenes from other Lewisian dykes to enable a comparison to be made of the relative degree of miscibility here.

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