The metamorphism of amygdales at 'S Airde Beinn, northern Mull

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Summary. Amygdales found metamorphosed in the aureole of the volcanic plug at 'S Airde Beinn, northern Mull, may be divided into three classes by their behaviour on metamorphism. The first class, characterized by the original dominant presence of zeolites, forms feldspar readily. It is related to a type, composed to a greater degree or entirely of ferromagnesian minerals, which seems to have formed by the infilling of originally void amygdales during the recrystallisation of the basalts. The second class, originally containing dominant gyrolite, forms first reverite and then wollastonite on metamorphism, the wollastonite-filled amygdales being enveloped in a rim of aegirine-augite from the earliest stages owing to reaction between the wollastonite and the basalt. The third class originally contained calcite, now represented by the anhydrous calcium silicates larnite, rankinite and wollastonite. These minerals form concentric monomineralic zones decreasing in Ca/Si ratio outwards. Later reaction has led to the replacement of both basalt and calcium silicates by melilite. Thin aegirine-augite rims surround the amygdales

Introduction

THE hill of 'S Airde Beinn in northern Mull (G.R. NM 472540; $6^{\circ} 07' \text{ W.}, 56^{\circ} 37' \text{ N.}$) is composed of an oval volcanic plug elongate along a NNW.-SSE. axis, about 930 yd long by 480 yd wide. It intrudes basalts of the Mull lava plateau and metamorphoses them to a high grade within an aureole extending about 30 ft from the contact.

The plateau basalts surrounding the plug contain amygdales of several types in about the usual abundance for such basalts. These show progressive changes as the plug is approached, and the development of rims by interaction with the surrounding basalt. These changes are the subject of description in this paper.

The thermal metamorphism of amygdales is not a subject to which much attention has been paid. McLintock (1915) and Harker and Marr (1891 and 1893) described amygdales metamorphosed by a granophyre in Mull and the Shap adamellite respectively but the metamorphism in both cases took place in a hydrous environment at low and moderate temperatures, and is of a quite different character from that seen at 'S Airde Beinn. More comparable are the amygdales described from Skye by Harker (1908). In the aureole of the Cuillins gabbro, amygdales originally containing zeolites have been changed to aggregates of plagioclase, and some of these amgydales contain mafic rims and zones similar to those described in this paper. Bailey *et al.* (1924) and Richey and Thomas (1930) found plagioclase aggregates after zeolites in Mull and Ardnamurchan, one of the specimens described coming from the outer part of the aureole at 'S Airde Beinn.

More recently Le Bas (1955) has compared plagioclase phenocrysts in metamorphosed basalts with plagioclase formed in metamorphosed amygdales. His results are similar to those of Harker, though expressed in more detail.

Unmetamorphosed amygdales

The unmetamorphosed amygdales in the basalts surrounding the plug can be divided into three classes depending on their behaviour on metamorphism:

- (a) amygdales with dominant sodium and calcium aluminosilicates;
- (b) amygdales with dominant gyrolite;
- (c) amygdales with dominant calcite.

The amygdales of the first class are of several types, united by their containing minerals that give rise to plagioclase on metamorphism. At one extreme are amygdales containing large radiating bunches of clear thomsonite associated with radiating finely fibrous natrolite and with minor amounts of analcite (91851), and these grade through to amygdales almost completely composed of analcite with minor thomsonite and natrolite (91852). These are the most abundantly present amygdales of this class; amygdales containing heulandite or stilbite are occasionally found (91853).

Sometimes a rim of chlorophaeite is found separating the zeolites from the surrounding basalt, but commonly this zone is absent, and fresh basalt and the zeolites are in direct juxtaposition.

The second class of amygdale may contain, in addition to abundant gyrolite, smaller amounts of analcite, thomsonite, and natrolite. The gyrolite occurs, as is usual, as radiating aggregates of colourless plates, which are often seen growing on a layer of analcite lining the cavity in the basalt (91901). Amygdales of this type have only been found unmetamorphosed at one locality, at the north end of the plug at the foot of the crags 60 yd east of the wall, but the fairly common occurrence of their metamorphosed equivalents suggests that they are, in fact, widespread in the unmetamorphosed basalt. Walker (1960) comments that gyrolite is difficult to collect in Northern Ireland as it is only found in the massive centres of flows, and this may well be the case at 'S Airde Beinn.

Amygdales of the third class, which contain dominant calcite, are also rarely found in the unmetamorphosed basalts, but they are equally rare inside the aureole too. However, their complex development on metamorphism makes them perhaps the most interesting of the three classes.

A comparison of the minerals developed here with the zeolite zones delineated in Northern Ireland by Walker (1960) shows that the assemblages found at 'S Airde Beinn most closely parallel those of the stilbite-heulandite zone. All of the minerals recognized here are reported from the stilbite-heulandite zone, but the relative order of abundance of the minerals is very different. However, these differences probably reflect the different conditions prevailing in Mull, though there is a possibility that some at least of the difference is caused by there having been two stages of filling of the amygdales, one before and the other after the intrusion of the plug. This possibility will be discussed in a later section.

The metamorphism of the basalts, a summary

In order to correlate amygdales of different kinds from different parts of the aureole, it is necessary to consider the grade of metamorphism reached by the basalts in which they are found. It seems useful, then, to give a summary of the stages of metamorphism seen in the basalts as a guide to what follows.

The first signs of metamorphism in the basalts are an increase in interstitial chlorophaeite and a noticeable alteration of the ferromagnesian minerals. This is superseded by a rapid increase in the degree of oxidation of the basalt. Olivine is replaced by hypersthene and iron ore, and soon the basalt is a finely granular aggregate of hypersthene, iron ore, augite, and plagioclase. This is the stage of maximum oxidation: hypersthene is the dominant ferromagnesian mineral. At higher grades, corresponding to moderate to high grades of metamorphism, the basalt becomes more reduced and the grain size increases until the rock contains olivine, augite, plagioclase, and iron ore in about the same proportions as in the original basalt, though they are completely recrystallized. The hypersthene has by gradual degrees completely disappeared, and augite is the dominant ferromagnesian mineral. At very high grades of metamorphism, reaction takes place with the magma in the plug, the degree of oxidation is still further reduced, and augite is replaced by olivine as the dominant ferromagnesian mineral.

Thus there are three main stages in the metamorphism. The first, up to and somewhat beyond the stage of maximum oxidation, dominated by hyperstheme, the second, from this stage up to that at which reaction begins, by augite, and the third, in the reacted basalts, by olivine.

Metamorphism of amygdales containing dominant sodium and calcium aluminosilicates

The first stages of metamorphism of this type of amygdale are complicated by the fact that zeolites similar to those found in the metamorphosed basalts sometimes replace the products of metamorphism and often fill the interstices between them. As a result of this, thomsonite, for example, is found in an amygdale in a highly recrystallized and reacted basalt very near to the contact with the gabbro of the plug (91841), at a grade above that at which larnite forms on metamorphism of the calcite-bearing amygdales, and where it cannot have survived unaffected during metamorphism. Although such extreme instances are easy enough to distinguish, difficulties arise at the lower grades where it cannot be certain whether zeolites have remained unchanged, or whether they are secondary.

However, by relating the amygdale to the stage of metamorphism reached by the enclosing basalt, a consistent sequence of metamorphic change is seen to emerge. After the initial stages, two extreme types of product are seen side by side, though there is a complete gradation between them. There is the felsic type, characterized by an abundance of feldspar, and the mafic type, where ferromagnesian minerals are dominant.

The felsic type. The effects of metamorphism are first seen in these amygdales at a grade somewhat below that of maximum oxidation. Here analcite can be seen in the process of conversion to feldspar (91855), and at a somewhat higher grade rather irregular feldspar laths are seen distinctly pseudomorphing analcite. It is possible that natrolite may survive to a slightly higher grade before reacting to form feldspar, but nowhere is the relation clear enough to be certain that the natrolite was not introduced after the intrusion of the plug.

The feldspar formed from the zeolites is at first finely flaky around the margins of the amygdales, with somewhat larger ragged plates near the centre (cf. Le Bas, 1955), but at higher grades it takes on a more euhedral form at the margins, though it is still fine grained, while the plates at

the centre become very well formed and are often euhedral against patches of later zeolites (91856).

The chlorophaeite, which is sometimes seen to surround the amygdales at very low grades, becomes rapidly transformed to hyperstheme. This is replaced by augite and, sometimes, by olivine as these minerals become in turn the dominant ferromagnesian phase. Small stringers of hyperstheme often run through the amygdales and may again represent original chlorophaeite.

The mafic type. The amygdales of this type consist, in their most extreme form, of aggregates of crystals of the mafic minerals that are dominant in the rock at that particular grade, usually associated with a little feldspar. The genesis of this type of amygdale poses problems to which it is difficult to find a satisfactory answer, as will be seen.

At the lowest grade at which this type is seen, it is found as part of a composite amygdale (91902), most of which is of the felsic type containing feldspar typical of low grades of metamorphism and fringed by hypersthene. One lobe of this amygdale is filled with coarsely granular olivine associated with some fine-grained iron ore. The olivine is highly magnesian ($2V_{\gamma}$ is just less than 90°). The hypersthene fringing this lobe appears to pseudomorph olivine, as here it occurs as small rounded grains very different in habit from the normal granular of prismatic form.

In other amygdales at the same grade (e.g. 91857), olivine grains are found forming rims, and are in all stages of replacement by hypersthene, which, however, retains the typical olivine form. These rims enclose a central zone of granular hypersthene and spongy masses of brown hornblende $(2V_{\gamma} \simeq 85^{\circ}, \gamma \land c = 19^{\circ}, \text{ pleochroism: } \alpha = \text{pale yellow},$ $\beta = \text{reddish brown}, \gamma = \text{yellowish brown}$). Such amygdales seem comparable to those described by Bailey *et al.* (1924) (p. 154) from the aureole of the Loch Uisg granophyre.

By the time the stage of maximum oxidation has been reached, the olivine has disappeared, and has been replaced by hypersthene and iron ore. At about this stage it is common to find a thin zone of feldspar between the hypersthene rim and the hypersthene filling the core. The latter is sometimes seen to take the form of radiating prismatic crystals surrounding an area filled by later zeolites (fig. 1 and see later) (91859).

At grades higher than this, hypersthene becomes unstable and is replaced in the rim and core by augite, or, at very high grades, by olivine. In the extreme case, the amygdales remain mafic to the highest grade seen and these give the appearance of a mafic clot in the basalt (91860). The grain size of the mafic minerals in the amygdales is always greater than that of the basalt, and, as the grain size of the basalts increases with increasing grade of metamorphism, the grain size in the amygdales increases in step.

There are, as has been remarked above, a large number of amygdales transitional between the extreme felsic type and the extreme mafic type. Often a core of feldspar is found at the centre of an otherwise mafic amygdale, and is surrounded by euhedral ferromagnesian crystals (e.g. 91861).

It is difficult to decide on a plausible theory for the origin of these mafic amygdales. There seem to be three main possibilities; first that they are produced by metamorphism of chlorophaeite-filled amygdales, second that the mafic minerals replaced pre-existing zeolites, and third that they fill amygdales which were originally wholly or partly empty.

While the first hypothesis accounts satisfactorily for the thin mafic rims of felsic amygdales, as unmetamorphosed amygdales with rims of chlorophaeite are found, no unmetamorphosed amygdales were discovered at all that were totally filled with chlorophaeite, while the proportion of totally or highly mafic amygdales to felsic ones is not less than one to two. If this hypothesis is to account for all of the mafic amygdales it is strange that no unmetamorphosed equivalents are found.

The other two hypotheses are in essence similar to one another. They both involve large-scale mobility of ferromagnesian minerals, in one case accompanied by solution of pre-existing zeolites. In connexion with this problem it is perhaps useful to consider the amygdale type already referred to (91859), shown in fig. 1. Amygdales of this type, which occur at about the stage of maximum oxidation, show a thin rim of granular hypersthene and iron ore enveloping a thin zone of feldspar. This in turn encloses a broad band of radially oriented prisms of hypersthene, and finally a core of secondary zeolites.

The presence of the core of zeolites shows that after the metamorphism a void remained at the centre of the amygdale, and the radial arrangement of the hypersthene suggests that, rather than being produced by metamorphism of chlorophaeite, it was deposited on the walls of a cavity and grew inwards. The thin zone of feldspar, then, would represent an original thin coating of zeolites on the cavity wall, now metamorphosed, and the rim of hypersthene and iron ore would once have been chlorophaeite.

Thus for this type of amygdale an origin by infilling rather than replacement or metamorphism would seem to be fairly clear, and it is tempting to extend this to cover all the mafic amygdales, but there are many amygdales from this grade that contain granular rather than radiating hypersthene, and it is possible that replacement may have taken place to a certain extent.

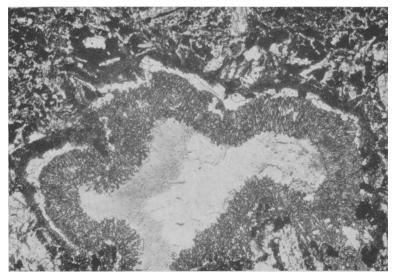


FIG. 1. Matic amygdale from the grade of maximum oxidation (see text for full description). Ordinary light, $\times 56$.

Whichever of the two more probable mechanisms is the more important, it is clear that in this case a remarkable degree of vapour transport of hypersthene and even of olivine has taken place.

The experiments of Bowen and Tuttle (1949) on the system $MgO-SiO_2$ - H_2O show that forsterite is not stable with vapour much below 400° C even at the lowest pressures investigated, and this is in a relatively silicapoor part of the system, while enstatite does not become stable with vapour until 600–700° C. The vapour transport observed could scarcely have started before these temperatures were reached, and this sets a limit for the stage of maximum oxidation at between 450° and 650° C, with a greater probability of its being nearer the upper limit.

Metamorphism of amygdales containing dominant gyrolite

The contents of the amygdales of this class undergo the simple and rapid change: $gyrolite \rightarrow reyerite \rightarrow wollastonite$ during progressive metamorphism. Gyrolite and reverite were identified by their optical properties, and specimens of each have been chemically analysed, and investigated by X-ray diffraction techniques. The reverite, pale green in bulk and with low birefringence, replaces the nearly colourless and highly birefringent gyrolite at a very early stage, when the basalts have scarcely recrystallized at all. Thomsonite, analcite, and natrolite are still stable (91817).

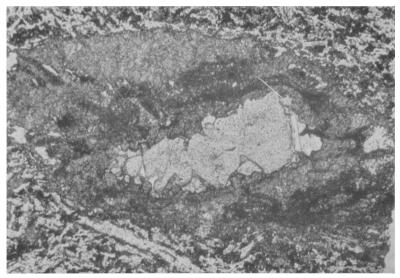


FIG. 2. Wollastonite-bearing amygdale from the grade of maximum oxidation. A rim of augite grading to aegirine-augite surrounds a central area of wollastonite. Ordinary light, $\times 45$.

Wollastonite replaces reyerite at a somewhat higher grade, but still before that of maximum oxidation of the basalts. Xonotlite, which has been reported as having been formed between reyerite and wollastonite experimentally (Meyer and Jaunarajs, 1961), was not found, despite an intensive search. Meyer and Jaunarajs report the formation of wollastonite from natural reyerite under hydrothermal conditions at 425° C, and this would, in general, fit with the placing of the stage of maximum oxidation nearer the upper limit of the range 450–650° C. Wollastonite is now the calcium silicate stable to the highest grades, though a considerable increase in its grain size takes place on further metamorphism (see fig. 2). Diffusion of Mg and Al into the amygdales sometimes gives rise to replacement of wollastonite by melilite, and rarely to replacement by monticellite. Nowhere is the wollastonite seen to take up iron as has been described by Tilley (1948).

As soon as the wollastonite has formed from the reyerite, a zone of aegirine-augite develops to surround the amygdale (or the part of the amygdale containing wollastonite). It is evident immediately the wollastonite is seen, and persists to the highest grades of metamorphism. In one or two amygdales at very high grades, aegirine-augite may rim an amygdale containing feldspar rather than wollastonite, but in such cases it seems more probable that the wollastonite has completely reacted away than that the rim has developed in its absence.

The rim is first seen as a thin zone of aegirine-augite inside the amygdale, surrounding newly formed wollastonite and itself surrounded by chlorophaeite (91864). At slightly higher grades the chlorophaeite has reacted to form hypersthene, while the aegirine-augite is now seen to grade into normal augite as it approaches it (91865). After this grade, replacement of the surrounding basalt becomes important, and hypersthene ceases to form the outside rim, though this is at about the stage of maximum oxidation when hypersthene is the dominant ferromagnesian mineral. Basalt near amygdales becomes enriched in augite by diffusion of lime outwards, and the sequence near an amygdale is characteristically:

basalt—augite-rich basalt—augite \pm plagioclase grading to aegirineaugite \pm plagioclase—wollastonite \pm aegirine-augite \pm plagioclase (e.g. 91867),

with minor modifications in different specimens. The aegirine-augite does not necessarily confine itself to the rims, but may grow into the wollastonite in large poikiloblastic crystals.

At the highest grades seen, equivalent to the grade at which larnite develops in the class of amygdales next to be described, the aegirineaugite becomes unstable at its contact with the surrounding basalt and breaks down at its outer edge to vermicular intergrowths of augite, iron ore, and plagioclase (91868) (see fig. 3). The basalt is now highly reduced, and it is clear that the aegirine-augite is not stable at the equivalent p_{0x} . The sequence is now:

basalt—augite+iron-ore+plagioclase—vermicular replacement aegirine-augite—wollastonite \pm aegirine-augite (91868).

By this stage the aegirine-augite, originally a pale soda-augite, has become very deeply coloured, and on its internal margins, especially where it abuts on feldspathic regions of the amygdale, approaches pure aegirine.

Apatite is commonly found in the felsic regions of these highly metamorphosed amygdales, and is presumably particularly abundant here because of the high concentration of CaO. Sphene is even more abundant, and is probably present for the same reason. It may be found in

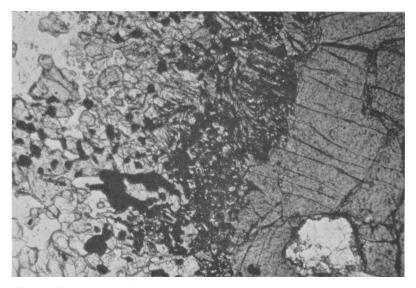


FIG. 3. Replacement of aegirine-augite (on right) by vermicular intergrowths of augite, iron ore, and plagioclase at high grades of metamorphism. Ordinary light, $\times 140$.

the lowest grade amygdales which contain wollastonite as granular strips within the pyroxene rim (91869). At higher grades it forms porphyroblasts within the aegirine-augite or in the felsic regions of the amygdales (91870).

Iron ore is stable in the outer part of the aegirine-augite rim, where it is probably a relic from reacted and replaced basalt. As it is displaced inwards towards more lime-rich parts of the amygdale by further outward growth of the aegirine-augite, it reacts to form a brown isotropic rim which may eventually completely replace the ore grains. This rim is probably of perovskite, or perhaps of garnet; it too can be found near the lowest grades at which wollastonite is stable.

The rim of aegirine-augite is clearly not formed by iso-chemical

metamorphism; there is no pre-existing material of this composition, nor could this account for the growth of the rim during metamorphism. It is apparently, then, a product of reaction.

On the basis of the phase equilibrium diagrams for the system $CaO-MgO-Al_2O_3-SiO_2$, the reaction between wollastonite and a diopside-forsterite-anorthite-enstatite assemblage would be expected to involve assemblages containing monticellite and melilite, while that between wollastonite and a diopside-anorthite-enstatite assemblage (corresponding to the oxidized basalts) would involve merely the disappearance of enstatite. Here the system is more complex, with the addition of Fe and Na notably, and it seems that in such a complex system the reactions predicted from the simple systems are suppressed in favour of the formation of aegirine-augite.

It is clear that a relatively high partial pressure of oxygen is at least partly necessary to stabilize the aegirine-augite, as at the highest grades, where the basalts have become very reduced and the oxidizing power of the vapour phase is considerably lower, the aegirine-augite is itself reduced to augite+iron ore+plagioclase, though this reaction is nowhere seen to be complete.

Metamorphism of amygdales containing dominant calcite

The class of amygdales originally containing calcite is very restricted in its occurrence round the plug. Two instances of unmetamorphosed amygdales containing calcite have been found, and one within the aureole with the assemblage wollastonite+calcite (91870), which represents an early stage in the metamorphism. The calcite in the latter case is found as euhedral crystals accompanied by euhedral wollastonite set in a base of later zeolites which partly corrode the euhedral minerals. Thus there can be little doubt that the association is primary. Some of the amygdales described in the previous section, which contain wollastonite alone, may have formed from calcite bearing amygdales at moderate grades of metamorphism, but as they are indistinguishable from those where the wollastonite has replaced gyrolte, the possibility is not an important one.

The amygdales of most interest in this class are of very restricted occurrence. They are found at a locality at the north end of the plug, near the foot of the crags and about 80 yd east of the wall. The locality is in basalts of a relatively high grade of metamorphism about 6 ft from the contact of the plug, and here, over an area 4 ft square, large amygdales up to 3 in. across may be found. Their contents, however, have almost always been altered by the percolating acid ground water (see later), and the following account is based on the few unaltered ones, which are all rather small (less than 1 in. across).

A typical amygdale is shown diagrammatically in fig. 4. The basalt outside the amygdales is abnormally rich in augite, which is accompanied

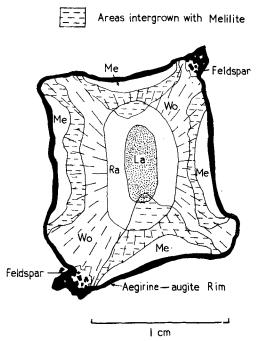


FIG. 4. Diagrammatic representation of a larnite-bearing amygdale. Me, melilite; Wo, wollastonite; Ra, rankinite; La, larnite.

by plagioclase, iron ore, and a little olivine. Hypersthene is quite absent, though little would in any case be expected at this grade.

The amygdales are surrounded by thin rims of aegirine-augite similar to those surrounding the amygdales of the class previously described. They are beginning to react at the outer margin to form the vermicular intergrowths seen outside aegirine-augite at high grades. The rims are, however, much paler than is usual at such a grade and are also abnormally thin.

Within the rim, and adjacent to it for most of its length, are elongate patches of anomalously birefringent equant melilite crystals which enclose iron-ore grains mantled with honey-yellow perovskite (see the previous section). Occasionally the rim bounds areas of large feldspar crystals associated with apatite and sphene; these are usually found at the corners of the amygdales, and between such regions and the melilite patches the zone of radiating wollastonite is usually in contact with the rim.

This zone, of radiating bladed crystals of wollastonite showing the normal polysynthetic twinning, runs continuously around the core of the amygdale. Where it lies inside a patch of melilite it is riddled with small rounded inclusions of melilite. Most often these inclusions do not persist to the inner margin of the wollastonite, leaving a clear zone of radiating crystals, but sometimes they continue to the very centre of the amygdale. The melilite changes character the further it penetrates into the core, becoming yellowish and losing its anomalous birefringence to acquire low grey interference colours. The colour suggests a melilite containing a certain amount of Na and Fe.

Inside the wollastonite is a zone of large equant crystals of rankinite $(Ca_3Si_2O_7)$ of an unusually large crystal size, and this in turn surrounds a core of granular larnite (Ca_2SiO_4) showing fine polysynthetic twinning.

Such assemblages can readily be derived by assuming that the amygdale originally contained calcite. The basic structure of the amygdale, that of concentric zones of calcium silicates, then takes the form of progressive decarbonation of the amygdale under a low partial pressure of CO_2 . The silica necessary must diffuse into the amygdale from the surrounding basalt, thus accounting for the concentric structure of the calcium silicates, decreasing in their Ca/Si ratio outwards.

It is interesting to note that as metamorphism must have taken place under a low partial pressure of CO_2 (tilleyite and spurrite are not seen to form), it must have taken place immediately to a grade at which larnite was stable, or the larnite could not have formed.

The inward diffusion of Si was accompanied by an outward diffusion of Ca. As a consequence of this, the basalt was enriched in augite, and, further, parts of the basalt near to the amygdales were totally replaced by melilite, and now form the melilite patches within the aegirineaugite rim. At the same time as this replacement of the basalt occurred, Mg and Al began to diffuse into the amygdales, with the formation of the round melilite crystals which riddle the calcium silicates.

The evidence for supposing that the areas of pure melilite represent replaced basalt is first through the mantled iron-ore grains and second through the shape of the areas. The presence of iron-ore grains mantled with perovskite within the areas of melilite, but not within the calcium silicates, seems to be best explained by supposing that they are the component of the basalt most resistant to metasomatic change, and have survived after the replacement of the rest of the basalt by being mantled with perovskite. The areas of melilite are crescent shaped, and join the edge of the amygdale generally on a line concave towards the core. Thus, as seems reasonable, the basalt was preferentially replaced where it approached closest to the centre of the amygdale. Similar evidence of calcium metasomatism leading to replacement by melilite has been seen at Ballycraigy in Northern Ireland (McConnell, personal communication).

The processes taking place here can thus be summarized as follows:

Stage	Amygdale		Basalt
0	calcite		olivine + augite + plagioclase + iron ore
1	larnite—	$Ca \rightarrow$	augite + iron ore + plagioclase
	rankinite— wollastonite	← Si	
2	above replaced by melilite	$\begin{array}{l} \mathrm{Ca} \rightarrow \\ \leftarrow \mathrm{Mg, \ Al} \end{array}$	melilite + iron-ore relicts

In the zoning of the calcium silicates, it is notable that bimineralic assemblages are rare. Instead of a zoning:

 $larnite_larnite+rankinite_rankinite_rankinite+wollastonite_wollastonite$

the zoning is the simple one:

larnite-rankinite-wollastonite

with pairs of minerals coexisting at the interfaces between zones.

This effect is one that would, in fact, be expected for simple metasomatism going to completion (Thompson, 1959), but it is found rarely enough for this example to be interesting.

Percolating ground water, which seems to enter the large amygdales very readily, produces an immediate alteration of the larnite to plombierite (McConnell, 1953). At first this preserves the habit and twinning of the larnite, though the birefringence and refractive index are considerably reduced, but later it breaks down to a cream-coloured gel, which is seen under the microscope to consist of minute flakes in all orientations. This is probably one of the low-temperature calcium silicate hydrates, with a Ca/Si atomic ratio near 1. Under the same conditions, the rankinite breaks down to a moderately birefringent fibrous mineral, while the wollastonite and melilite remain unchanged. Acknowledgements. I should like to acknowledge the help given to this work by discussions with Prof. C. E. Tilley, Dr. S. R. Nockolds, and Dr. J. D. C. McConnell. The research was carried out during the tenure of a D.S.I.R. research studentship and a research fellowship at St. John's College, Cambridge.

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