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Textural evidence for liquid immiscibility in tholeiites

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SUMMARY. The residual liquids of many tholeiitic basalts and andesites, on cooling, split into iron-rich and silica-rich fractions, which may quench to brown glassy globules and clear glass respectively. More commonly, however, cooling is sufficiently slow for the iron-rich liquid to crystallize to globular single crystals of pyroxene. Depending on the cooling rate, these crystallized globules range in shape from spheres to elongated globules bounded by crystal faces. The fine grain size of the mesostasis of most tholeiites is partly due to these small crystallized globules. The silica-rich fraction, on the other hand, is more commonly quenched to a glass, and when preserved as globules in the crystallized iron-rich fraction, it may be bounded by negative crystal faces of the surrounding pyroxene. Globules of the iron-rich liquid commonly nucleate on the surface of the plagioclase crystals where they can become trapped, later crystallizing to spherical pyroxene grains that mostly contain a minor opaque phase. In contrast, iron-rich globules that form next to pyroxene grains commonly become attached to these crystals, giving them lobate boundaries. The immiscible silica-rich liquid becomes trapped between these lobes and, with sufficiently slow cooling, results in finger-like quartzo-feldspathic inclusions extending in from the margins of the pyroxene grains. These textures can provide evidence of immiscibility in a wide range of volcanic and hypabyssal rocks, even when no glass is present.

THE existence of silicate liquid immiscibility in magmas has been a source of controversy for over fifty years (Bowen, 1928; Holgate, 1954) but incontrovertible evidence of such magmatic splitting has recently been found in certain basalts, the mesostases of which consist of pairs of chemically distinct glasses, which form globules in each other (Roedder and Weiblen, 1971; De, 1974; Philpotts, 1978). Because these basalts are common tholeiites, it is surprising that the evidence for liquid immiscibility went undetected for so long and, indeed, was found only after similar evidence was discovered in lunar rocks (Roedder and Weiblen, 1970). The scarcity of completely unaltered glass-bearing terrestrial rocks and a general conceptual inertia opposed to immiscibility were both probably responsible for this delay. Today, however, there is little doubt that many tholeiitic magmas do pass through a two-liquid stage during crystallization.

Previous studies reveal little of the extent to which this immiscibility may have affected the differentiation of igneous rocks. Although some rocks develop immiscibility in only the final few per cent of residual liquid, others exhibit it when at least 50% liquid remains, a fraction that is large enough to permit the development of significant amounts of differentiation products through this process. Also, McBirney (1975) has shown, by experiment, that immiscibility may have played a role in producing granophyres during the final stages of crystallization of the Skaergaard magma. Furthermore, experimental studies suggest that silicate immiscibility may have been involved in the differentiation of some alkaline rocks (Philpotts, 1971, 1976; Ferguson and Currie, 1971).

The significance of liquid immiscibility cannot be properly evaluated if only fresh glassy rocks are considered, and yet completely crystalline ones cannot contain the definitive evidence of immiscibility, that is, two glasses. Crystalline rocks do, however, contain features that have been interpreted as products of immiscibility. For instance, the ocelli of many fine-grained mafic alkaline rocks, particularly those of camptonitic composition, exhibit many features that are consistent with their having formed as immiscible globules (Philpotts, 1976). Variolites in some basaltic rocks have also been interpreted as relict immiscible globules (Carstens, 1964; Gelinas *et al.*, 1976). But these features have been interpreted in other ways, and certainly do not provide conclusive evidence of immiscibility.

During an experimental investigation of immiscibility in a tholeiitic basalt from Connecticut (Philpotts, 1978), certain textures were found in the rock and in the experimental run products that, while resulting from immiscibility, were of such a nature that they clearly could be preserved, even in rocks that are completely crystalline. These textures have been found in other basalts and in some coarser-grained crystalline rocks, such as the standard diabase, W-1. This paper, then, describes these textures in the hope that they may be useful in providing evidence of immiscibility in rocks that have cooled too slowly to contain glasses.

Formation of immiscible globules. Some magmas may have compositions that would allow the separation of immiscible globules to occur on first cooling, but most magmas must initially undergo a degree of crystallization before the residual liquid has a composition that will permit the development of immiscible fractions. For example, in a tholeiitic basalt from the North Shore volcanic group of Lake Superior, immiscible liquids developed in the residual 44% liquid (Roedder and Weiblen, 1971), while in a basalt from Connecticut, they developed in the residual 32% liquid (Philpotts, 1978), and in a Deccan trap, in the residual 20% liquid (De, 1974). Of course, the degree of crystallization required to bring about immiscibility depends largely on the initial magma composition, but experiments indicate that higher oxygen fugacities can increase the extent of immiscibility (Naslund, 1976; Philpotts, 1978).

The initial magma composition and the path of fractional crystallization determine where the residual liquid will first intersect the immiscibility field, which, in turn, determines the proportions of the two liquids. For example, the residual liquid in the Connecticut basalt split into approximately equal proportions (fig. 1), whereas in the tholeiitic dykes of northern England (Holmes and Harwood, 1929), relatively early crystallization of magnetite led to a silica-rich residue rather than an iron-enriched one and, thus, only small numbers of iron-rich immiscible globules were formed (fig. 2). In contrast, strong iron-enrichment with little silica-enrichment in the residues of some Kilauean lavas led to a preponderance of the iron-rich immiscible phase (fig. 3). Regardless of the actual proportions of the two liquids, the iron-rich one almost always forms globules in the silica-rich one, except where the liquids are trapped as thin sheets between plagioclase laths (fig. 3).

Before considering the textures produced by immiscible liquids, it is worth reviewing the possible stages involved in the formation of immiscible

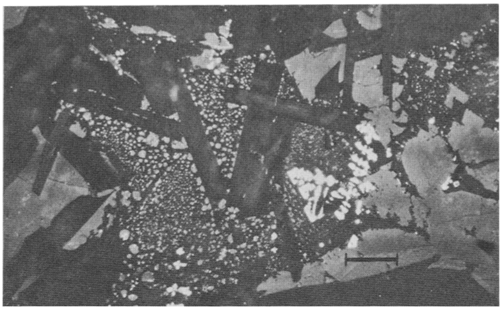

FIG. 1. Patch of mesostasis consisting of skeletal magnetite grains (white) and iron-rich glassy and crystallized globules (small light-grey bodies) in a silica-rich glassy host, surrounded by subophitically intergrown laths of plagioclase (dark) and pyroxene (grey). Basalt from Southbury, Connecticut. Reflected light, oil immersion. Scale bar is 50 μm.

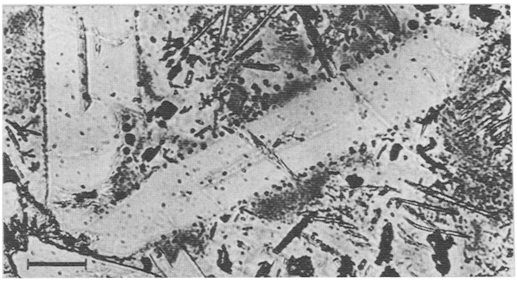

FIG. 2. Small glassy globules of iron-rich immiscible liquid on the surface of plagioclase laths and in the clear silica-rich glassy mesostasis of a tholeiitic dyke from Great Cumbrae, Firth of Clyde, Scotland. Scale bar is 50 μm.

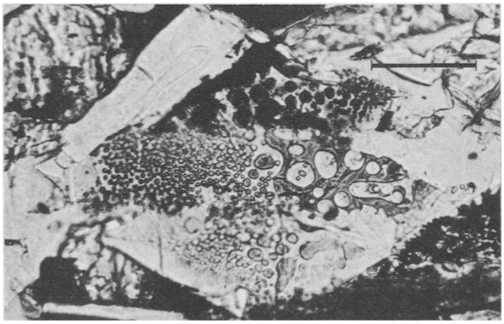

FIG. 3. Mesostasis of the prehistoric flow in the quarry north-east of the Volcano Observatory, Kilauea. Brown and clear glasses form globules in each other, and the dark mesostasis consists largely of immiscible globules that have crystallized to pyroxene and an opaque oxide. Cambridge thin section No. 57358 (Yoder and Tilley, 1962). Scale bar is 50 μm.

globules. This will be done for a basaltic magma, based on the textures and experimental investigations of the tholeiitic basalt from the Triassic-Jurassic basin of Southbury, Connecticut (Philpotts, 1978). This basalt forms a massive 50 to 100 m thick flow, which is rather fine-grained throughout. It consists of approximately 68% subophitically intergrown plagioclase and pyroxene clusters and 32% mesostasis, which contains iron-rich and silica-rich glasses, both of which form globules in each other.

In the experiments on the Connecticut basalt, the first immiscible globules to appear on cooling are of the iron-rich liquid. These nucleate on the surface of the plagioclase laths, most likely because the crystallization of plagioclase enriches the surrounding liquid in the components necessary to form the iron-rich liquid (McBirney and Nakamura, 1974). With further cooling, other globules nucleate in the remainder of the residual liquid, but the first-formed ones continue to grow and remain the largest. In the natural rock, most of the largest immiscible globules also occur on the faces of the plagioclase laths where they form regularly spaced hemispherical bodies projecting from the crystal faces into the patches of mesostasis (fig. 1).

The size and number of immiscible globules within residual patches of liquid vary considerably, even within those patches where the proportions of the two liquids remain approximately the same throughout. In fig. 1 this variation can be seen to occur over distances of as little as a few microns. Some of the variation, of course, is attributable to continued nucleation of globules during cooling and further exsolution and growth on the earlier ones. But the main cause seems to be variations in the rate of globule nucleation from place to place within the residual liquid, a phenomenon that must be sensitive to minor compositional variations within this liquid. There is a general tendency for those globules of iron-rich liquid that are in patches of liquid close to, or entirely surrounded by plagioclase crystals to be fewer in number but larger in size than those that occur in the vicinity of pyroxene crystals (fig. 1).

Many of the plagioclase crystals in the Connecticut basalt contain sheet-like inclusions of a brown glass, which invariably contains clear glassy globules of an immiscible silica-rich glass (fig. 5A). These inclusions never contain pyroxene crystals, despite this mineral's abundance in the surrounding rock, and zoning in the plagioclase of the walls of the inclusions suggests that reaction occurred between the plagioclase and the liquid following entrapment. This particular immiscibility, therefore, is believed to have developed metastably in a liquid that was supersaturated with pyroxene (Philpotts, 1978). Although these inclusions may not indicate stable immiscibility, they are particularly common in basalts that do, at a later stage of crystallization, develop stable immiscible liquids Hawaiian tholeiites and Deccan traps, for example.

Textures resulting from immiscibility

The mesostasis. Prior to the development of immiscible globules in the Connecticut basalt, pyroxene and plagioclase grew in relatively coarse subophitic clusters; however, with the onset of immiscibility, further nucleation and growth of pyroxene was restricted almost entirely to individual globules of the iron-rich liquid. This resulted in a sudden increase in the number of small pyroxene crystals and produced a mesostasis with a distinctly finer grain size than that of the rest of the rock (fig. 1). This texture is typical of tholeiites, as for example, in those from the type locality in the Saar-Nahe district of Germany, which are described by Rosenbusch (1887, p. 504) as having a fine-grained mesostasis containing glass and 'globulitic glass'. Similarly, the Hawaiian tholeiites studied by Yoder and Tilley (1962), and figured in their Plate 1, figures B and C, have a fine-grained mesostasis containing abundant immiscible globules of iron-rich glass in addition to small pyroxene and magnetite grains (fig. 3).

Textures within the Connecticut basalt suggest that the contrasting grain size between early subophitic clusters and the later mesostasis may extend to more slowly cooled rocks. Throughout the 50 to 100 m thickness of this basalt, there is very little variation in grain size (except at the very top and bottom of the flow), and no variation in the amount of mesostasis. Cooling rates, which must vary considerably through a flow of this thickness, can therefore have little to do with the textural change from a subophitic intergrowth to the finer multigrained aggregate of a mesostasis. Thus, a finer-grained late-crystallizing fraction could still be expected to result from immiscibility in a more slowly cooled rock.

Of course, in very slowly cooled magmas, only small numbers of immiscible globules might form, and these could well nucleate heterogeneously on the surface of those minerals they most closely approach in structure. In such a case, there need be no new nucleation of crystals at all, and thus no record of immiscibility would be left in the form of grain size variations.

Relict globule textures. Globules of immiscible liquid will crystallize on being cooled slowly, but this need not destroy their shapes. Different rates of cooling within the thick Connecticut basalt flow produced all gradations from completely glassy to crystalline globules, making it possible to interpret, with a considerable degree of certainty, those textures in the crystalline rock that owe their origin to immiscibility.

The most common type of immiscible globule, that of an iron-rich liquid in a silica-rich host, crystallizes easily, even in rapidly cooled rocks, and forms sphere-like single crystals of pyroxene. This probably results from the liquid having a composition and 'structure' approximating that of a pyroxene, and thus little, if any, diffusion is required for crystal nucleation and growth. Those constituents that cannot be accommodated by the pyroxene structure crystallize and form opaque grains, or separate as globules of silica-rich liquid (figs. 3 and 4). The opacity of much of the mesostasis in many tholeiites is due to the presence of these small opaque grains within the pyroxene spheres, and they are commonly so abundant as to completely obscure the spheres in normal thickness sections. Very thin sections or polished sections may be required to clearly reveal the spheres.

There is a marked difference in colour between the iron-rich globules that are preserved as glass and those that have crystallized to pyroxene, the glassy ones being dark brown and the crystalline ones a transparent green (fig. 4). Crystallization of the globules may, in addition, be accompanied by the development of crystal faces, but this is determined largely by the proximity of globules to earlier-formed pyroxene crystals. For example, in fig. 4, several skeletal pyroxene needles are shown in a patch of mesostasis that contains both brown glassy globules and green crystalline pyroxene globules. While the glassy ones are truly spherical, the crystalline ones are slightly elongated parallel to the lengths of the pyroxene needles with which they are in contact, and with which they crystallized in optical continuity. The resulting skeletal crystals consist largely of linked series of crystallized immiscible globules, with the original site of each globule marked by an opaque grain, which formed as an accessory from the iron-rich liquid.

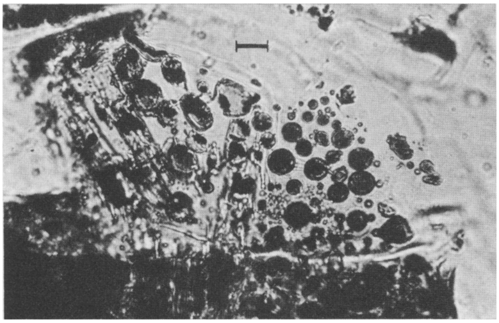

FIG. 4. Patch of mesostasis, in the same basalt as that of fig. 3, containing immiscible globules of an iron-rich liquid, which are preserved both as dark-brown glassy spheres (right) and as green ellipsoidal grains of pyroxene containing an opaque phase (left). The crystallized droplets all make contact (and are in optical continuity) with a few pyroxene needles that grew into the patch of mesostasis from earlier-formed pyroxene grains in the lower part of the figure. Scale bar is 10 μm.

With very slow cooling, iron-rich globules could have their original shapes completely destroyed during crystallization. None the less, the small opaque grains formed from that part of the immiscible liquid that cannot be accommodated in the pyroxene structure may serve to identify these crystalline bodies as relict immiscible globules. Although opaque grains could form by epitaxial growth on the surface of pyroxene crystals, such grains would not necessarily be present on every crystal. With immiscible liquids, however, all globules formed at similar temperatures should have similar compositions and thus, on crystallizing, all globules should contain the same proportions of magnetite and other accessory constituents.

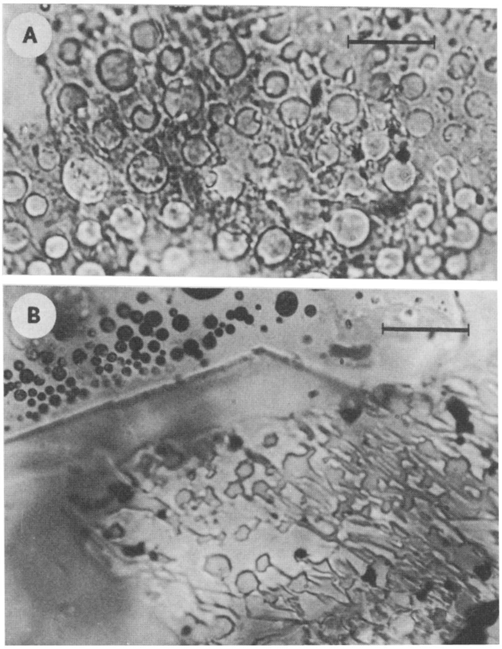

FIG. 5. Clear glassy globules preserved in a brown glassy host (A) and in a sheet of pyroxene formed from the crystallization of an iron-rich immiscible liquid (B) in the Connecticut basalt. The clear glassy globules in B are bounded by negative crystal faces of the surrounding pyroxene. This pyroxene also contains opaque grains, which distinguish it from the earlier-formed pyroxene in the subophitic clusters (see fig. 1). Small globules of the iron-rich liquid are still preserved in a glassy state in the upper left of the figure. Scale bar in A and B is 10 μm.

Where immiscible globules are composed of the high-silica liquid (fig. 5A), slow cooling allows the surrounding iron-rich liquid to crystallize, forming crystallographically continuous bodies of pyroxene enclosing the silica-rich globules and accessory opaque grains (fig. 5B). The globules may be spherical but most are bounded by negative crystal faces of the surrounding pyroxene. The growth of these faces distorts the globules first into equidimensional polyhedra and then into elongated negative crystals, which preserve little of the original form of the immiscible globules. Fig. 5B shows this entire range of shapes within a single sheet of pyroxene formed from the iron-rich melt. The orientations of the negative faces on each of the silica-rich globules can be seen to be similar, and in the upper right corner of the figure one of these directions predominates and causes the silica-rich phase to form elongated patches between needles of pyroxene. Thin sheets of liquid trapped within plagioclase crystals commonly develop this texture but, as indicated earlier, this immiscibility may form metastably with respect to pyroxene.

A not uncommon texture in many tholeiites results from the coalescence of globules of the iron-rich liquid around larger drops of the silica-rich liquid. Later crystallization or alteration of the iron-rich phase results in a dark annular ring or crescent surrounding a clear spherical or ellipsoidal core (fig. 6). This texture is most clearly seen on the surface of plagioclase crystals.

The Holyoke basalt, the thickest volcanic unit in the Triassic–Jurassic basin of the Connecticut river valley, has a composition and over-all texture identical to that of the basalt from Southbury, Connecticut. Alteration, however, has obscured the details of its mesostasis and has certainly destroyed any glass that might originally have been present. Its mesostasis, which has a cloudy, poorly developed granophyric texture, contains small dark-brown spherical bodies containing large proportions of sphene. These spheres have the same range of sizes and distribution within the mesostasis as the brown iron-rich glassy globules have in the unaltered Southbury basalt, and there is no doubt that they were originally globules of the iron-rich (also Ti-rich) immiscible liquid. Since many volcanic rocks are somewhat altered, it is important to recognize that such sphene-bearing globules may possibly indicate immiscibility.

All of these relict globule textures, which provide direct and unambiguous evidence of immiscibility, are rather common in crystalline basaltic rocks. Unfortunately they are not likely to be preserved in more slowly cooled rocks, and in these it is necessary to look for more permanent records of immiscibility.

Immiscible globule inclusions in plagioclase. The nucleation of immiscible globules of iron-rich liquid on the surface of plagioclase crystals in tholeiitic basalts and andesites can lead to the formation of regularly spaced iron-rich inclusions in this mineral. If plagioclase is in equilibrium with two immiscible liquids, it must crystallize with the same composition from both liquids, but this does not mean that it crystallizes in the same quantities from both. Indeed, the nutrients for plagioclase growth are more abundant in the silica-rich liquid and, thus, the growth rate of plagioclase is slower at the attachment sites of globules of the iron-rich liquid than at other parts of the crystal in contact with the silica-rich liquid. As the plagioclase continues to grow, therefore, the immiscible globules become trapped and form planar sets of regularly spaced inclusions marking growth surfaces in the plagioclase (fig. 7).

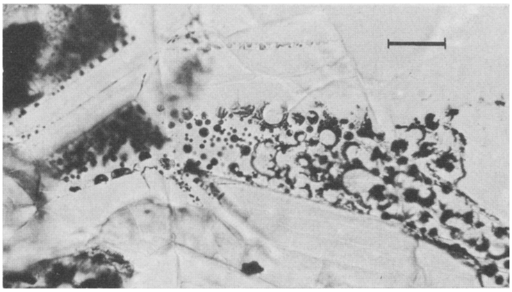

FIG. 6. Crescent and ring-like structures formed from the coalescence of iron-rich liquid around immiscible globules of silica-rich liquid in the mesostasis of an 1859 lava of Mauna Loa. Most of the larger crescents have crystallized to pyroxene and dendritic magnetite, but the small globules are still glassy. Note the regular array of immiscible iron-rich globules trapped in the margin of the plagioclase in the upper left. Scale bar is 20 μm.

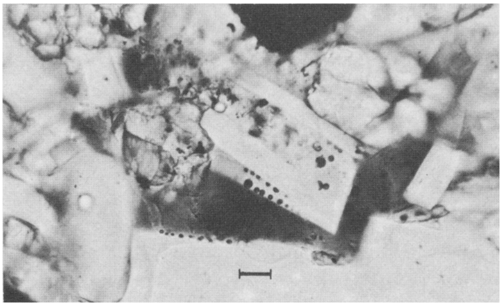

FIG. 7. Immiscible globules of iron-rich liquid in various stages of entrapment in the plagioclase of an andesite from the Usu volcano, Hokkaido. Note that the immiscible droplets nucleate only on the surface of the plagioclase laths and, only then, in areas remote from pyroxene grains; the remainder of the mesostasis consists of a homogeneous pale-brown glass. Scale bar is 10 μm.

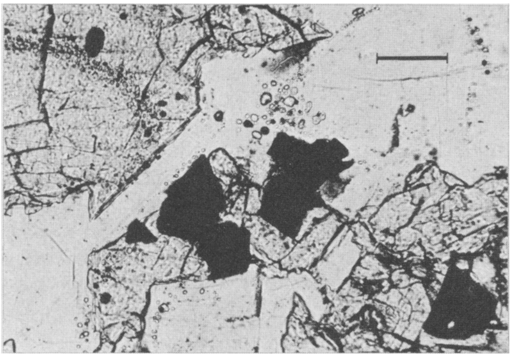

FIG. 8. Small pyroxene spheres containing opaque grains, interpreted as crystallized immiscible iron-rich liquid globules, are included in the margin of plagioclase crystals in this Deccan trap. Also, low refractive index colourless inclusions form prominent zones in many of the large pyroxene crystals and they may have formed from entrapped globules of the conjugate silica-rich immiscible liquid. Scale bar is 50 μm.

With slow cooling, the trapped globules crystallize to form inclusions of pyroxene and minor opaque grains (fig. 8), which must be carefully distinguished from inclusions of accidentally trapped minerals. Firstly, the spacing of immiscible globules on the surface of plagioclase crystals tends to be strikingly regular (figs. 1, 6, and 7) as a result of the nucleation process, whereas accidental inclusions need show no such regularity. Secondly, since any set of globules formed at a particular time on the surface of a growing plagioclase crystal would be formed at a single temperature from a liquid of a particular composition, each globule should have the same composition and, thus, on crystallizing, would have the same proportions of pyroxene, magnetite, and other minor constituents. Globules trapped on successive growth surfaces of the plagioclase, however, could have different compositions, reflecting changing temperatures and magma compositions during the growth of the plagioclase.

These crystallized globules can be preserved in the plagioclase of some quite slowly cooled rocks. For example, fig. 9 shows a sample of a tholeiitic dyke on the Island of Mull in which a plagioclase crystal contains regularly spaced iron-rich inclusions arranged along an early growth surface of the plagioclase. In this example the inclusions clearly originated as trapped immiscible globules, for they occur in that sector of the plagioclase crystal that grew into what is now a patch of granophyre containing abundant immiscible iron-rich globules. Furthermore, the inclusions have a similar spacing and size as those in the mesostasis directly in contact with the plagioclase. Even if this rock had not contained the immiscible globules in the mesostasis, the regularly spaced inclusions alone could have been interpreted as indicating the prior existence of immiscible liquids in this rock.

Pyroxene morphology. Experiments performed on the Connecticut basalt indicate that plagioclase and pyroxene crystallized together over a 100 °C interval before the residual liquid split into two immiscible fractions (Philpotts, 1978). During this interval, many of the pyroxene and plagioclase crystals became intergrown, and those parts of pyroxene grains in contact with the residual liquid are bounded by smooth crystal faces—this period of crystallization undoubtedly corresponds to that during which the subophitic texture was developed in the natural rock. Following the onset of immiscibility in the experiments, the pyroxene grains are no longer bounded by smooth planar faces but, instead, are distinctly lobate in outline. This results from the adherence and ready crystallization of globules of the iron-rich immiscible liquid on the surface of the pyroxene.

Pyroxene crystals in the natural basalt have identical lobate surfaces (fig. 10). The faces of crystals that project into patches of mesostasis containing immiscible globules are covered with lobes of approximately the same dimensions and spacings as those of the iron-rich immiscible globules in the mesostasis in the vicinity of that face. Compare, for example, the left and right sides of fig. 1.

Prior to the onset of immiscibility, crystallization of pyroxene would have involved diffusion of the necessary components to the growing crystal face, followed by organization of the pyroxene com-

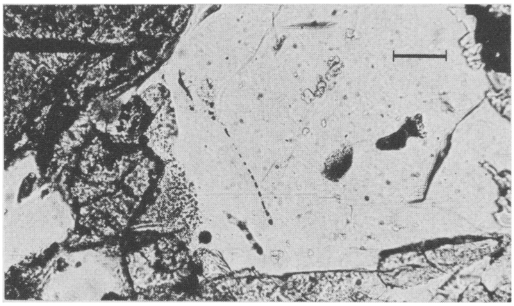

FIG. 9. Regularly spaced iron-rich inclusions along an early growth surface in that sector of a plagioclase crystal that extends out into a granophyric patch of mesostasis containing small iron-rich immiscible globules. A second row of three larger droplets occurs at the left end of the plagioclase grain and immediately above it the mesostasis contains one large immiscible sulfide globule. Tholeiitic dyke-rock, Island of Mull. Scale bar is 50 μm.

ponents into appropriate building units and, finally, precipitation of pyroxene on the surface of the crystal. Once the magma split into silica-rich and iron-rich fractions, however, the growth process must have drastically changed. The immiscible globules of iron-rich liquid are discrete bodies with pyroxene-like compositions and, though lacking the long-range order of a crystal, they probably have pyroxene-like structures. In the experiments, therefore, it is not surprising that, while some immiscible globules attached to the surface of pyroxene crystals are quenched to a glassy state, others exhibit weak birefringence, and still others are crystallized in optical continuity with the host grain.

There is little doubt from the experiments and the textures in the rock that, following the onset of immiscibility, growth of pyroxene is marked by the attachment and subsequent crystallization of globules of iron-rich liquid on the surface of pyroxene grains. The spacing of the resultant lobes on a crystal face is, then, determined by the density of nucleation sites of immiscible globules in the residual liquid (fig. 10A). Further crystallization of pyroxene can then proceed in two ways: material may diffuse from the next set of iron-rich globules through the silica-rich liquid to the lobate crystal face, or alternatively, more globules may simply become attached to the surface of the crystal. Both mechanisms probably occur, but in either case the initial surface irregularities on the crystal tend to remain.

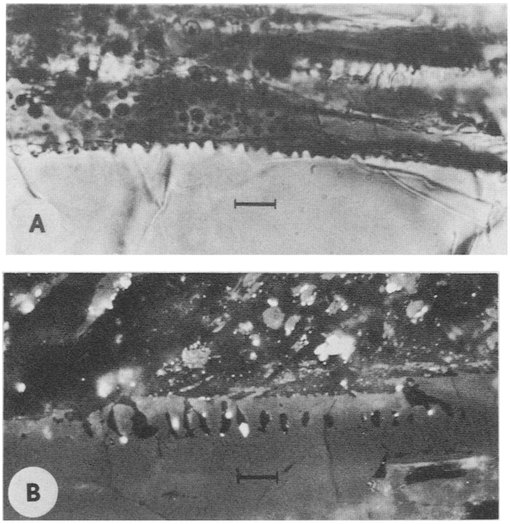

FIG. 10. (A) Initial stage of development of a lobate crystal face on a pyroxene grain in contact with a glassy mesostasis containing immiscible iron-rich globules in the Connecticut basalt. Note the similar dimensions and spacing of the immiscible globules and the lobes. Transmitted light. Scale bar is 10 μm. (B) Cellular zone on the margin of a pyroxene grain showing magnetite grains and chalcopyrite spheres associated with the inclusions of the silica-rich glass. Immiscible spheres of the iron-rich phase occur in the silica-rich glass next to the pyroxene crystal. Reflected light, oil immersion. Scale bar is 10 μm.

The silica-rich liquid trapped between the lobes slows the growth of pyroxene in these areas, in the same way that the iron-rich globules on the surface of plagioclase grains slow the growth of that mineral. As a consequence, the margin of a pyroxene crystal can develop a cellular structure with long finger-like inclusions of the silica-rich liquid extending into the crystal (fig. 10B). Similar features have been found on the margins of pyroxferroite crystals in lunar rocks (Roedder and Weiblen, 1971). At any stage in the crystallization, the shape of the inclusions can be affected by changes in the rate of crystal growth, the size of immiscible globules, or the disappearance of the second liquid. For example, in the Connecticut basalt some inclusions terminate before reaching the margin of the crystal (fig. 10B), whereas others never grew into elongate cells but, instead, form simple globular inclusions.

The composition of the iron-rich immiscible liquid is close to but not precisely that of a pyroxene and, thus, its crystallization on to the surface of pyroxene grains is accompanied by the exsolution and growth of other minerals. The most common of these is an opaque oxide, which is present as small grains in the pyroxene of the cellular zone (figs. 10B and 8).

In the Connecticut basalt, the cellular inclusions are composed largely of silica-rich glass, but morphologically identical inclusions in the marginal zones of pyroxenes in many coarse-grained diabases are composed of low-birefringent quartzofeldspathic material most of which is accompanied by an opaque accessory grain. While it is risky to extrapolate from textures observed in experimental run products or even in fine-grained basalts to those in coarse-grained rocks, the similarities of these inclusions are most striking and thus might indicate immiscibility in these rocks.

One particular crystalline rock, the standard diabase W-1, whose pyroxene grains contain irregular and cellular inclusions of quartzofeldspathic material in those parts that come in contact with late-crystallizing material, has been investigated experimentally (Philpotts, 1978). This rock was totally fused, homogenized, and then slowly cooled at one atmosphere total pressure with oxygen fugacities controlled by the Ni-NiO buffer. Following considerable crystallization, immiscibility appeared in the liquid residue at 1035 °C. The textural consequence of this immiscibility may well be recorded by the quartzofeldspathic inclusions in the margins of pyroxene grains.

Summary and conclusions

The residual liquids of many tholeiitic basalts and andesites split into iron-rich and silica-rich fractions on cooling. These can be preserved as globules of brown glass in a clear glassy host, but nature does not commonly provide the appropriate conditions for such preservation and, instead, most globules become obscured by later crystallization or alteration. The presence of two liquids, however, drastically alters the mechanism of crystal growth and nucleation and thus influences the textures developed, even in rocks that are completely crystalline. The resultant textures that may indicate immiscibility include:

The mesostasis. The fine-grain size of the mesostasis of many tholeiites is attributable to the growth of individual pyroxene grains in the large numbers of small immiscible globules of the iron-rich liquid that abound in the mesostasis of most tholeiites. While rapid quenching during the final stages of crystallization could produce a fine-grained mesostasis, it does not normally appear to be the cause, for in thick flows the amount of mesostasis is independent of position within the flow and therefore of cooling rate. Immiscibility, on the other hand, is dependent only on the degree of crystallization and fractionation of the residual liquid, and thus is, to a large extent, independent of cooling rate. It, therefore, would appear to be the most likely cause for the development of a fine-grained mesostasis in tholeiites.

Crystallized globules. The iron-rich immiscible liquid crystallizes to pyroxene and accessory opaque grains, whereas the silica-rich liquid crystallizes to a granophyric intergrowth. Depending on the cooling rate, crystallized immiscible globules of the iron-rich liquid can range in shape from true spheres to elongated globules bounded by crystal faces. The silica-rich globules are normally bounded by negative crystal faces formed when the surrounding iron-rich liquid crystallizes to pyroxene.

Iron-rich inclusions in plagioclase. Iron-rich immiscible globules are normally the first to form, nucleating and growing as rows of regularly spaced hemispherical globules on the surface of plagioclase crystals. With continued growth of the plagioclase these globules can become trapped, later crystallizing to form spherical pyroxene grains within the plagioclase.

Lobate pyroxene crystal faces. Globules of the iron-rich immiscible liquid, having pyroxene-like compositions, readily crystallize on to any pyroxene grain with which they come in contact. The resulting lobate crystal face can be preserved, even in totally crystallized rocks, with the silica-rich liquid trapped between the 'pyroxene' lobes eventually crystallizing to quartzofeldspathic inclusions.

With these textures, the range of rock types in which evidence suggesting immiscibility can be found is extended from glass-bearing rocks to completely crystalline volcanic ones, and even some hypabyssal rocks. But since these textures result from diffusion-controlled processes that are dependent on relatively rapid cooling rates, they are not likely to form in more slowly cooled plutonic rocks. With rapid cooling, immiscible globules mostly nucleate on the surface of crystals from which they differ most in composition, for it is here that, with slow diffusion rates, the liquid residue first becomes supersaturated in the immiscible phase. At sufficiently slow cooling rates, however, significant compositional gradients do not develop and then immiscible globules are more likely to nucleate epitaxially on the minerals which they most closely approach in composition and 'structure', that is, iron-rich globules on pyroxene and silica-rich globules on feldspar. Since these globules would eventually crystallize on to their substrate, it is not clear, at present, what textural evidence of immiscibility would remain in such rocks, and it is possible that other petrologic or geochemical data may provide the only evidence of immiscibility in slowly cooled plutonic rocks.

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