SHORT COMMUNICATIONS

Immiscibility in tholeiites: a discussion

BIGGAR (1979) has pointed out that the immiscibility evident in the residual glasses of many tholeiites may have developed metastably in supercooled liquids. His main argument is that the immiscible globules are smaller ($<$ 10 μm) than those he has grown in synthetic systems (50–80 μm), which he has been able to coarsen by coalescence in short duration experiments. He hopes that experimental studies and further textural evidence can resolve the problem of distinguishing stable from metastable immiscible liquids in rocks.

Immiscibility in the residual liquid of a normal tholeiite has been investigated experimentally at one atmosphere pressure under controlled oxygen fugacities (Philpotts, 1979), and additional experiments on this rock are reported here. These results leave no doubt that, at least in this tholeiite, immiscibility was a stable phenomenon.

If, as Biggar suggests, immiscibility is developed in the supercooled liquid residue of tholeites, the rate of cooling of the lava, and consequently the position within a flow, might be expected to play a critical role in the development of this phenomenon. The two-phase glasses, however, are typically found in the slower cooled, coarser parts of flows and pillows (Philpotts, 1978), suggesting that crystal growth and fractionation of the residual liquid is more important to the development of immiscibility than rapid cooling rates that are likely to produce supercooling. In the case of the flood basalt from Southbury, Connecticut (Philpotts, 1979), which is over 50 m thick, very different cooling rates must have existed throughout the flow, and yet these have produced no variation whatsoever in the amount or size of the immiscible globules. Flows of such thickness provide the field geologists with the results of cooling over periods that vary from a rapid quench to the normal life expectancy of the average experimental petrologist, and for that reason are worthy of careful scrutiny.

The residual liquids of basalts and andesites that exhibit evidence of immiscibility (stable or metastable) first encounter the immiscibility field on its silica-rich side, as indicated by the occurrence of iron-rich globules in a more abundant silica-rich host. But the compositions used by Biggar in his experiments are considerably richer in iron and the immiscibility occurs at a higher temperature. As pointed out by Freestone (1979), both these factors will contribute to the synthetic immiscible globules being coarser than the natural ones.

The experiments on the Connecticut basalt (Philpotts, 1979), indicate that the immiscibility field is first encountered at a temperature of approximately 1035 °C. The viscosity of the silica-rich melt at this temperature is approximately 2.3×10^7 poise, as calculated by the method of Bottinga and Weill (1972), but it is an order of magnitude less at the temperatures encountered in the immiscibility field in the system $K_2O - FeO - Al_2O_3 - SiO_2$, investigated by Biggar. This difference will certainly result in slower rates of coalescence in the lower temperature natural liquids. Also, the viscosity of the iron-rich immiscible liquid is 25 000 times less than that of the silica-rich one, and hence, as the iron-rich liquid becomes more abundant, so the likelihood of coalescence increases.

Coarsening of globules may also occur by diffusion of material from small globules to larger ones. Diffusion rates in the iron-rich melts must be greater than those in the silica-rich ones, and hence, should coarsening take place by this method, its rate will be greatly enhanced by higher proportions of the iron-rich melt. Also, higher temperatures will increase diffusion rates and promote coarsening.

In summary, then, the size difference between immiscible globules obtained by Biggar in his experiments and those found in natural rocks can be explained by changes in the physical properties of the immiscible liquids with temperature and bulk composition.

The previous experiments on the Connecticut basalt (Philpotts, 1979) indicate that the temperature of onset of unmixing in the residual liquid is dependent on oxygen fugacity, rising from 1022 °C at a fugacity of $10^{-13.1}$ atm to 1047 °C at $10^{-9.7}$. This boundary was investigated with care to avoid possible confusion resulting from metastable immiscible liquids produced during quenching. First, the transition from homogeneous liquid to a pair of immiscible liquids was reversed, and second, experiments of several weeks duration were used to effect coalescence and coarsening of the globules so as to demonstrate that they were indeed present at the run temperatures and were not formed during quenching. These experiments, then,

do indicate, within ± 5 °C, the temperature of first appearance of immiscibility in the residual liquid produced in this basalt by the particular cooling and crystallization rates used in the experiments.

The experimental results do not, however, prove that this boundary marking the onset of immiscibility is a stable one; it is possible that the immiscible iron-rich globules were metastable with respect to pyroxene (+magnetite), and had there been sufficient time, pyroxene would have formed instead of the immiscible globules. This seems most unlikely, since pyroxene, plagioclase, and, in some experiments, magnetite were present with the two liquids. Similarly, in the centre of the 50 m-thick Connecticut basalt these same immiscible globules formed juxtaposed coarse ophitic clusters of pyroxene and plagioclase. But no matter how unlikely metastability seems, extreme prudence is warranted, especially in light of the rather discreditable reputation that immiscibility has had in the petrologic community for the last half century.

Throughout most of the thickness of the Connecticut basalt the textural evidence for immiscibility consists of silica-rich glass containing spheres of iron-rich pyroxene with inclusions of magnetite and other minor phases (Philpotts, 1979, fig. 6). These spheres are interpreted to have formed as immiscible globules of iron-rich melt that later crystallized to pyroxene and accessory phases. The question here is whether, under equilibrium conditions, iron-rich pyroxene would have crystallized prior to the formation of the immiscible iron-rich melt, and by so doing prevented unmixing from occurring. If this were the case, heating of the rock in the laboratory would result in the formation of just a single liquid during melting. On the other hand, if the pyroxene spheres do represent crystallized globules of stably formed immiscible liquid, then melting of the rock would produce immiscible liquids.

A sample of rock containing crystalline spheres was heated under a variety of controlled oxygen fugacities, for periods of from two days to two weeks, and immiscible liquids were indeed produced during partial melting. In these experiments, which will be reported fully elsewhere (Philpotts, Doyle, and Thomas, in preparation), uncrushed thin slabs of sample 10345 (Philpotts, 1979) were suspended on platinum wire loops (previously saturated with iron), and sealed in evacuated silica glass capsules, with powdered buffer separated from the wire at the base of the capsule. The sample weight, 0.01 to 0.02 g, was chosen so that the rock's inherent oxygen fugacity did not exceed the buffering capacity of the oxygen buffer used to control the experiments.
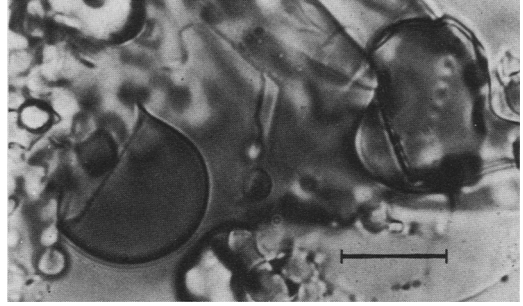

FIG. 1. Iron-rich immiscible globules in silica-rich glass produced after five days by the partial melting of pyroxene spheres in the mesostasis of a tholeiitic basalt at 1008 °C under an oxygen fugacity controlled by the magnetite-wustite buffer. The straight boundary in the globule on the left is the face of a plagioclase crystal. The amoeboid-shaped globule on the right contains a blocky crystal of pyroxene and several small magnetite grains (out of focus) which did not totally melt at this temperature. Other smaller immiscible globules occur throughout the field. Scale bar is 10 μm.

Fig. 1 illustrates the results of a five-day run at 1008 °C under an oxygen fugacity controlled by the magnetite-wustite buffer. Globules of iron-rich immiscible liquid, having approximately the same diameters as the initial pyroxene spheres (15 μm), are seen in a silica-rich liquid between larger crystals of plagioclase and pyroxene. The globule on the left has within itself a crystal of plagioclase, whereas the one on the right contains pyroxene and magnetite grains, which are the crystalline remains of the partially melted pyroxene sphere.

It is concluded, therefore, that stable immiscibility did develop in the residual liquid of this particular basalt, and because it is a common tholeiite, liquid immiscibility must be considered at least potentially important in low pressure, late stage differentiation of such magmas.

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