

produce sub-micron globules. It would seem probable that some natural occurrences owe their origin to nucleation of a second liquid in the supercooled state, possibly with some subsequent coarsening of the texture. Criteria to distinguish stable immiscibility, which might lead to a petrologically important separation of two magma types, from metastable (supercooled) immiscibility can perhaps be found by a combination of experimental and petrographic techniques.

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Immiscibility in tholeiites

BIGGAR (1979) proposes that the two-liquid textures encountered in many lunar and terrestrial tholeiites may not have formed stably, but that the phase separation occurred in homogeneous liquids that had supercooled below their equilibrium crystallization temperatures. This possibility requires careful consideration, for if the immiscibility phenomena in the lavas, described by Roedder and Weiblen (1970, 1971, 1972) and Philpotts (1978), formed metastably, then any extrapolation to magmatic processes on a large scale is unwarranted. Certainly the mechanism proposed by Biggar (op. cit.) is technically feasible, for not only is metastable immiscibility commonly used in industry to develop special glasses and glass-ceramics (Charles 1973), but also metastable separation of supercooled liquids on the scale of the lunar glass-in-glass textures has been achieved in the closely related system K_2O -FeO- Al_2O_3 - SiO_2 (Irvine 1976). However, in these instances, the liquids are commonly first superheated to remove any crystal nuclei, whereas the tholeiites contain abundant crystals. In support of his proposal, Biggar (op. cit.) cites a disparity in size between his experimentally-produced stable immiscible globules, up to 80 μm diameter, and the natural textures, in general 2-10 μm (Roedder and Weiblen, op. cit., Philpotts op. cit.). I wish to discuss this disparity with refer-

ence to my own experimental studies in parts of the system K_2O -CaO-MgO-FeO- Al_2O_3 - SiO_2 - TiO_2 - P_2O_5 (Freestone 1978a, b and in progress).

Experimentally-produced textural features. The morphology and size of the two-liquid textures achieved in the experiments has been found to be extremely variable, depending on the relative proportions, viscosities and surface tensions of the immiscible melts and the presence and amount of crystalline phases. Above the liquidus, the formation of globules several hundreds of microns in diameter is readily achievable, and the separation of two layers in the charge may occur. However, the presence of a large proportion of crystalline phases inhibits the coarsening of the textures. In particular, where % crystals > % Si-rich melt > % Fe-rich melt, globules of Fe-rich melt in Si-rich melt are small, and range from 2 μm to 20 or 30 μm diameter. Globules greater than c. 10 μm are rare in these charges and, where they occur, often contain crystals (ore minerals, olivines, pyroxenes) which would nucleate further crystals on a slow quench, leading to their obliteration. Thus my own experiments suggest that the glassy globules in tholeiitic lavas which are 56-98% crystalline would in general be only 2-10 μm diameter if stable immiscibility had occurred. Furthermore, in their figures, Roedder and Weiblen show a few globules

of the order of 20 μm diameter (1970, fig. 35; 1972, figs. 9, 10, 11) and even *c.* 50 μm (1971, fig. 23).

The generally small size of globules developed in melts which are substantially crystalline probably stems from three effects:

(1) The presence of crystals isolated in the melts will obstruct the coalescence of globules.

(2) Bowen (1919) and Wright and Fiske (1971, appendix 3) have noted that only 50% crystallization of a magma is required to allow segregation of a melt into pockets—i.e. to form a three-dimensional crystalline framework. Such a framework is likely to exist in tholeiites by the onset of liquid immiscibility and will prevent movement and coalescence of globules.

(3) Changes in the rheological properties of the melt due to the presence of crystals (e.g. Sparks *et al.*, 1977) may also inhibit movement of globules.

Biggar's (1978) experiments on the lunar compositions involved a system which was largely liquid rather than crystalline as in the natural lavas. Thus coalescence and growth of the dispersed liquid was facilitated in the experimental relative to the natural situation. Furthermore, examination of the published photographs suggests that, in general, there is a much lower proportion of Fe-rich liquid than Si-rich liquid, whereas in the experiments, an equal weight proportion was used (Biggar 1978). The more dense distribution of globules in the experimental melts will have facilitated collision and coalescence.

The fine scale of the two-liquid textures observed in tholeiites is therefore consistent with an origin by stable liquid immiscibility. However, it has not been shown that these features did not originate by metastable processes, as proposed by Biggar (1979).

Role of metastable immiscibility. A number of experimental studies have been published which demonstrate immiscibility in late stage tholeiitic melts (e.g. Rutherford *et al.*, 1974, 1976, Hess *et al.*, 1975, Ryerson and Hess, 1978, Roedder and Weiblen, 1970, McBirney, 1975) and I interpret these to indicate that, at least in some cases, stable immiscibility may occur in the lavas. Even so, there is usually a metastable or quench separation associated with stable immiscibility (e.g. Freestone and Hamilton 1978), and this quench separation is also to be expected to have occurred in the tholeiites, where it may be responsible for the discrepancies in composition between natural and synthetic immiscible glasses noted by Biggar (1978) in an earlier paper. These discrepancies are illustrated in fig. 1, where it may be seen that the natural glasses are of more extreme composition than those produced experimentally, and that there is an apparent rotation of tie-lines of the natural glasses compared to the synthetic ones. These differences are probably

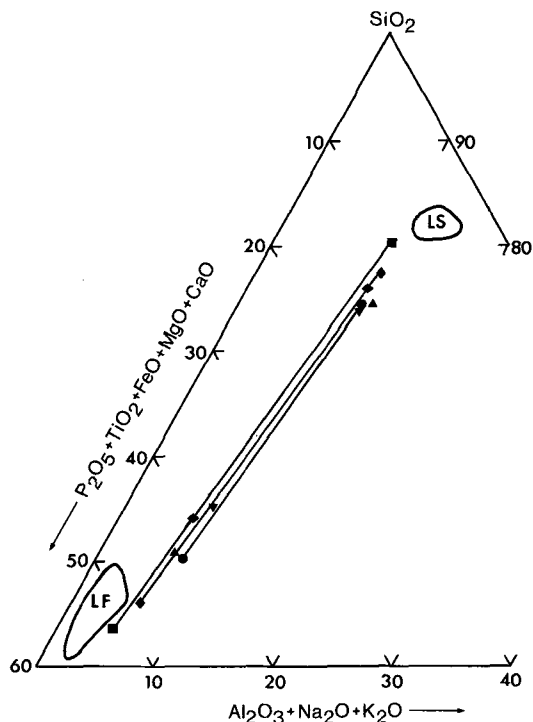


FIG. 1. Comparison of synthetic lunar immiscible melts produced experimentally by Rutherford *et al.* (1974) and Hess *et al.* (1975) with fields of lunar immiscible glasses, based on data of Roedder and Weiblen. Diagram is in mole %. Symbols indicate synthetic melt compositions, some of which are joined by tie lines. LS, LF signify fields of naturally occurring silica-rich and iron-rich glasses, respectively.

best explained as the result of further separation of globules which initially separated stably, down the metastable extension of the solvus. Quench crystallization may also have been responsible for some modification of the composition of the liquids. Thus the fact that the melting behaviour of synthetic compositions based on mixtures of the analysed immiscible glasses in tholeiites does not correspond precisely to that expected (e.g. Biggar 1978) does not negate the hypothesis that stable immiscibility occurred, because the analysed glasses may not represent the compositions of the stable immiscible melts.

Conclusion. The scales and morphologies of the textural features observed by Roedder and Weiblen (1970, 1971, 1972) and Philpotts (1978) in tholeiitic lavas are consistent with the occurrence of stable liquid immiscibility. This view is supported by the published experimental work. However, discrepancies in composition between naturally occurring

and synthetically produced immiscible glasses may be due to metastable or quench processes. A single phase melt which lies close to a stable two-liquid field will almost certainly separate a second liquid phase metastably during a slow quench, as in the interior of a lava flow. Therefore while there is apparently good evidence for the occurrence of stable immiscibility in some cases, it is probable that at least some reported examples of two-liquid textures in tholeiites formed metastably, as proposed by Biggar (op. cit.). As there is no discontinuity in the physical properties of a liquid as it moves from the stable to the metastable or supercooled state, a continuity is to be expected between those textures formed stably and those formed metastably, so that distinction between the two modes of formation on a morphological basis is unlikely. Assessment of the true extent of stable immiscibility in tholeiitic flows will require carefully integrated experimental, petrographical and analytical work.

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