

## Olivine Flotation and Settling Experiments on the Join $Mg_2SiO_4$ - $Fe_2SiO_4$

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**Abstract.** Some unusual density relations between olivine and coexisting liquid in the system fosterite-fayalite are reported. At 1 atmosphere pressure olivine floats on its coexisting liquid for intermediate compositions on this binary because of extreme partitioning of Fe into the melt phase. At 20 kilobars the usual behavior of olivine settling occurs because the partitioning of Fe in the melt is reduced, aided possibly by the dissolution of CO<sub>2</sub> in the melt from use of a graphite container. Olivine flotation and settling are rapid in a time period of only a few hours because viscosities are a little greater than that of paraffin oil at room temperature. Some adcumulate textures with good triple junction grain boundaries are developed. General observations of differentiated magmatic systems on a number of scales and experimental data indicate that the mechanisms by which magmas can differentiate vary considerably in the ultramafic to tholeiitic compositional range.

### Introduction

In a paper by McBirney and Noyes (1979) on the differentiation of the Skaergaard intrusion, a strong and convincing case was made for in situ crystallization (eg., Jackson 1961) based on non-Newtonian properties of the magma as well as field observations such as (i) layering on the roof, walls, and floor (ii) the densities of plagioclase and mafic minerals are less than and greater than that of magma respectively, yet these minerals are distributed throughout the intrusion and (iii) numerous textures can be attributed to crystal growth in situ including graded and rhythmic layering. Indeed, numerous petrologists have noted that in the best documented layered intrusions there are no clear accumulations of floated plagioclase near the roof contact (eg., McBirney and Noyes 1979; Irvine 1980); where anorthosites do occur they can be found in relatively low stratigraphic horizons such as those in the Bushveld and Stillwater intrusions (Hall 1932; McCallum et al. 1980).

Although some authors totally reject the importance of crystal settling in convecting and nonconvecting media (Rice 1981), an important alternative has crystal growth on vertical walls of convecting magma chambers, flotation or settling of the residual liquid along the upwelling or downwelling convection cell limb depending on its density, and ultimately the development of a magma chamber with

vertical composition gradients and horizontal double-diffusive layers (Turner 1980; Turner and Gustafson 1981). The end product of this model may be similar to simple phase settling and flotation; however, it may be a much more efficient way of affecting phase separation particularly in high viscosity magmas.

There are actually many examples of crystal settling, including filter pressing variants, and these have received a considerable amount of attention (eg., Daly 1914; Bowen 1919; Irvine 1980). However, the scale where this is best developed is characteristically larger than those which are readily accessible to direct observation. Although poorly understood, it is generally acknowledged that the earth has been differentiated by processes involving silicate liquids (eg., Safronov 1978). On another planetary scale, the fact that plagioclase can float (Bottinga and Weill 1970; Campbell et al. 1978) has been used extensively in models of the differentiation of the earth's moon which must explain a lunar stratigraphy consisting of an uppermost layer or unit of anorthosite (Wood et al. 1970; Longhi and Boudreau 1979; Herzberg and Baker 1980).

Although apparently self evident, the very existence of volcanoes is proof that magmas can separate from their source regions at depth in the earth and ascent because of buoyancy forces. Indeed, this occurs even where the proportion of liquid to solid is small (ie., low degrees of partial melting; see for example O'Hara 1968 and Stolper et al. 1981) and the crystal mush assumes the properties of viscoelastic solids; these conditions are considered necessary for the prevention of solid-liquid phase separation and segregation in a gravity field for many layered intrusions of a tholeiitic composition (McBirney and Noyes 1979). Clearly, strong rheological changes can develop from the point of magma segregation in the mantle to eventual crystallization in the crust, and this must be due in part to changes in the composition of the melt phase (eg., O'Hara 1968), although deviatoric stresses in the mantle may help to promote melt segregation (Spera 1980). Gravity-induced phase separation is likely to be a much more important process in picritic or ultramafic magmas generated in the mantle than their evolved tholeiitic counterparts in the crust. This can also explain rather well why we perceive many examples of in situ crystallization such as granite plutons, the Skaergaard intrusion, and many basic sills in the earth's crust, but few striking examples of crystal settling. Many picritic and ultramafic magmas do not intrude into or extrude on the earth's crust where crystal settling

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can operate for eventual inspection because the densities of these magmas are too high (Huppert and Sparks 1980; Stolper and Walker 1980); they probably remain in the mantle or pond below a low density boundary layer such as basaltic magma chambers along oceanic ridges and the base of the crust (i.e., noteworthy exceptions are the unusual Archean komatiite flows with large volumes of basal olivine cumulates; Arndt et al. 1977). Our scale of observation can thus determine the importance we place on mechanisms of differentiation.

Although the rheological properties of magmas need to be understood quantitatively (Spera 1980), this paper will only attempt to illustrate conceptually the importance of viscosity on phase separation in ultramafic compositions. This is shown by some unusual density relations between olivine and its coexisting liquid in the system forsterite-fayalite at low and high pressures and the extraordinarily high rate at which olivine settles and floats in these experiments.

### Experimental Observations

Figure 1 is a reproduction of the forsterite-fayalite solidus-liquidus loop determined experimentally by Bowen and Schairer (1935). The historical integrity of this diagram has been maintained although the reader should be reminded that in fact fayalite, and presumably other compositions on this join, melt incongruently to metallic Fe + liquid.

The densities of coexisting olivine and liquid pairs along the loop were calculated, and the results are given in Fig. 2. The densities of the solid olivine compositions were determined from their values at 25° C adjusted for the effects of thermal expansion using thermal expansivities of  $3.2$  to  $3.8 \times 10^{-5} \text{ } ^\circ\text{C}^{-1}$  [Fisher and Medaris 1969; Smyth 1975; Hazen 1976;  $\alpha = 1/V(\partial V/\partial T)_p$ ]. The densities of the liquid compositions were determined from the partial molar volumes and thermal expansivities of MgO and SiO<sub>2</sub> reported by Bottinga et al. (1982) and for FeO those given by Nelson and Carmichael (1979).

If the errors in these silicate liquid compositions are less than about  $\pm 1\%$  (Nelson and Carmichael 1979), olivine crystals should have lower densities than their coexisting liquid compositions along most intermediate compositions along this join at 1 atmosphere pressure. The possibility that olivine might float was tested experimentally and verified.

One experiment was run on the bulk composition  $(\text{Mg}_{0.3}\text{Fe}_{0.7})_2\text{SiO}_4$  where the density relations shown in Fig. 2 indicate that olivine is less dense than its coexisting liquid at 1,425° C and 1 atmosphere pressure. The experimental methods used were very similar to those adopted by Bowen and Schairer (1935) but with a number of exceptions. The amount of sample used was large (i.e., 200 mg), and the inner diameter of the spec pure Fe container was similarly large (i.e., 4 mm). This was required in order to minimize capillary action and concomitant draining of the capsule of its contents during the course of the experiment. The sample was run at 1,425° C for 2.5 h with the oxygen fugacity crudely controlled by an argon atmosphere.

The results of this experiment is shown in Fig. 3. Cumulate olivine crystals had floated to the top as anticipated by the calculated densities, and the extent of the solid-liquid phase separation was strong in only 2.5 h. Using the lever rule and Fig. 1, the bulk olivine to liquid proportion is

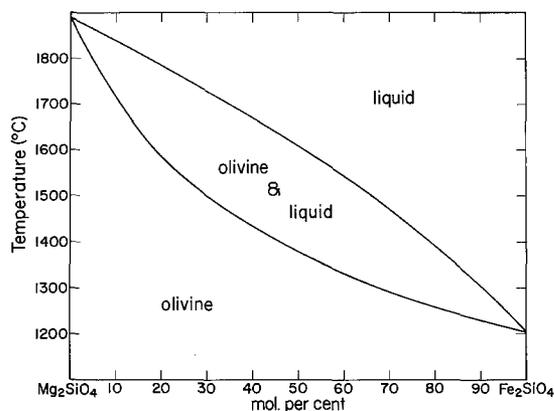


Fig. 1. The forsterite ( $\text{Mg}_2\text{SiO}_4$ ) - fayalite ( $\text{Fe}_2\text{SiO}_4$ ) solidus-liquidus loop at 1 atmosphere pressure determined by Bowen and Schairer (1935)

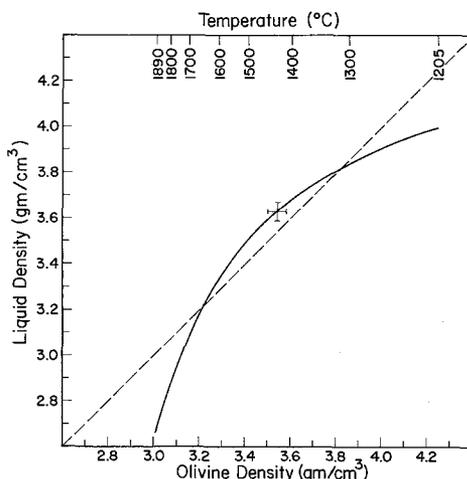
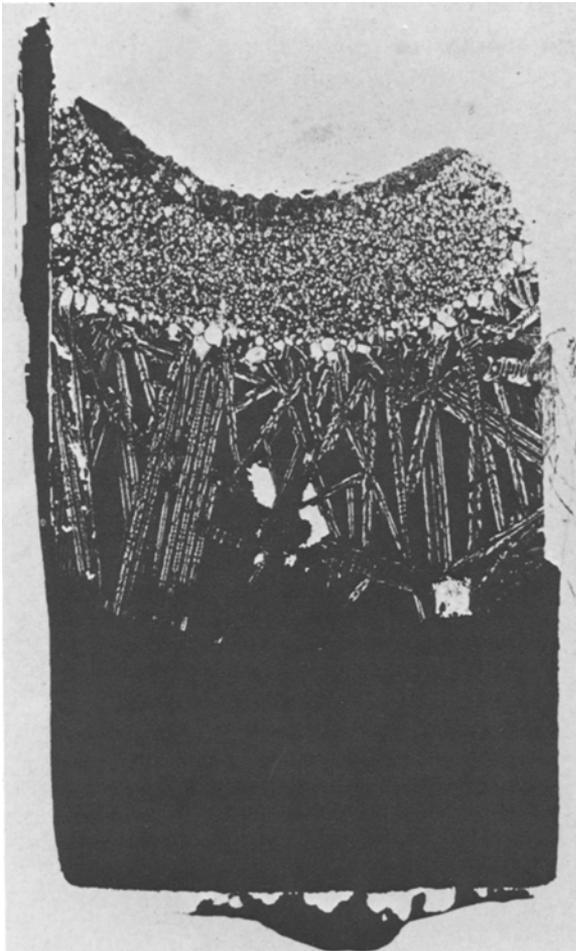


Fig. 2. Densities of olivine and coexisting liquid along the loop of Fig. 1. Error bars at 1,425° C are  $\pm 1\%$ . Note the region above the broken line where the density of liquid is greater than that of coexisting olivine crystals

about 1 to 4. Most of the olivine is contained in the uppermost cumulate layer, and is about 60% by volume; the remaining 40% is intercumulus liquid. Note also that the meniscus is occupied by this cumulate crystal mush.

Another experiment was made on the same bulk composition, but at 1,500° C and 20 kilobars and for only 1.5 h in duration. The composition was placed in a graphite capsule which in turn was placed into a platinum sheath. The sample was dried in a partial vacuum and the platinum welded in order to keep the composition water-free. Pressure was imposed on the sample by means of a piston-cylinder solid media machine. The results are shown in Fig. 4a and b. In this experiment the cumulus olivine crystals had largely settled to the bottom of the container and the coexisting liquid dispersed both as an intercumulus phase as well as a segregated layer of 100% liquid at the top of the capsule. Even with a bulk olivine to liquid ratio of about 44 to 56 determined by point counting, the extent of the phase separation was strong during the time period of only 1.5 h. Figure 4b shows the nature of the cumulate texture approximately half way up the cumulate pile. Although this would be described as a mesocumulate (Wager and Brown 1968), adcumulus features with good triple junc-



**Fig. 3.** Photomicrograph of an experimental result on the bulk composition Fo<sub>30</sub> run at 1,425° C for 2.5 h in an argon atmosphere. The calculated olivine and liquid densities are shown by the error bars in Fig. 2. Note the pervasive concentration of liquidus olivine crystals at the top of the charge. Parts of the iron container wall at the right as well as some of the meniscus were lost during thin section preparation. The white patches near the center are bits of the thin section which were plucked. The interface between the cumulus olivine layer and quench liquid contains large olivine crystals as well as some round gas bubbles which appear entrained. Inner diameter of charge = 4 mm

tion grain boundaries are beginning to develop. Most grain boundaries appear to be wetted with a melt phase as indicated by the distribution of Fe metal particles and visible grain boundaries in Fig. 4b. However, clusters of olivine crystals with adcumulate textures seem to have grain boundaries which are free of a melt phase. The base of the charge (Fig. 4a) is entirely adcumulate, the complementary liquid fraction having been transposed to the top of the charge. Note also the adcumulates along the left hand side of the capsule. This is due to a temperature gradient in the charge which either affects a marginal density contrast between olivine and liquid via differential thermal expansion or imparts some convective movement to the crystal mush. If the latter explanation is correct, the sense of circulation would have been counterclockwise since the right hand side appears to have been at a higher temperature. The overall distribution of the solid and liquid phases

appear to be somewhat similar to those of the 'filling box' effect described by Turner and Gustafson (1981) on crystallization-convection experiments in carbonate solutions. Electron microprobe determinations of 16 randomly selected olivine grains shows them to be a very homogeneous forsterite  $49.8 \pm 0.6$ , fayalite  $50.2 \pm 0.5$ . Consequently, any temperature variations along the loop of Fig. 1 could not have been very large.

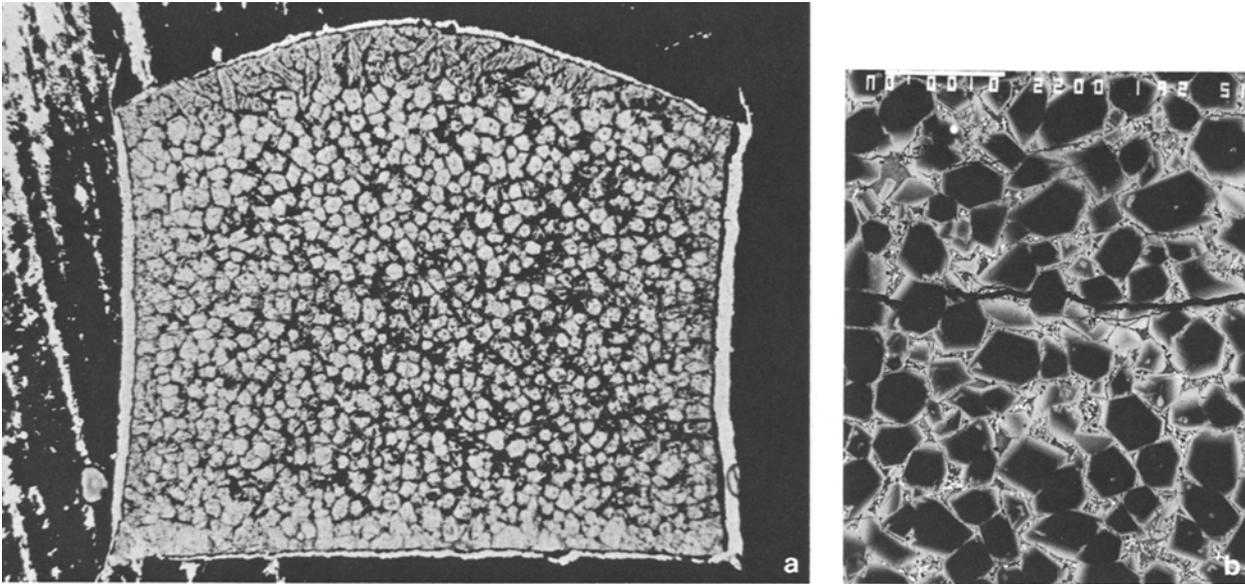
The very high liquid densities shown in Fig. 2 are in part the result of low concentrations of Si and O compared to those normally found in natural basaltic magmas, and the absence of other elements like Ca, Na, and K which tend to lower the intrinsic density of the liquid. Additionally, liquids at 1 atmosphere pressure can be more dense than coexisting olivine because of the extreme partitioning of the heavy element Fe into the liquid phase. This partitioning is usually expressed:

$$K_D = \frac{X_{ol}^{FeO} X_l^{MgO}}{X^{FeO} X_{ol}^{MgO}}$$

and takes the value of 0.30–0.33 for many terrestrial basalts (Roeder and Emslie 1970). However, over the temperature and composition range experimentally investigated by Bowen and Schairer (1935) in Fig. 1,  $K_D$  is only about 0.22. Together with other experimental observations, this demonstrates the profound effect that SiO<sub>2</sub> content of a magma can have on  $K_D$  (Longhi et al. 1978; lower values of  $K_D$  corresponds to increased partitioning of Fe into the liquid phase). Additions of basalt-like concentrations of SiO<sub>2</sub> to the join forsterite-fayalite results in the usual case of olivine settling at 1 atmosphere because it not only lowers the intrinsic density of the liquid but it also reduces the amount of Fe that becomes incorporated in the melt.

The imposition of pressure on the Fe–Mg exchange equilibrium between olivine and silicate liquid reduces the extent to which Fe partitions into the melt by favoring the smaller Mg cation. Our tentative interpretation is that  $K_D$  is increased sufficiently within 20 kilobars that an olivine-liquid density inversion is established. Although we have not quantified these 20 kilobar density relations rigorously, we expect that olivine is only marginally more dense than its coexisting liquid at these pressures based on the effect of pressure on  $K_D$  (Longhi et al. 1978) and the distribution of adcumulates along the left hand side of the charge in Fig. 4b as well as the bottom. However, the reader is cautioned about a possible ambiguity concerning these 20 kilobar results. Although the use of a graphite container eliminates the problem of Fe loss from the composition, it may have introduced some CO<sub>2</sub> into the liquid phase. If correct, the exact nature of the density inversion is rendered ambiguous.

The high rate at which phase separation and segregation are established in these experiments is due largely to the low viscosity of the liquids. Viscosity calculations based on the method of Bottinga and Weill (1972) yield about 2 poises, which is only slightly larger than that of paraffin oil at room temperature. This compares with 4.6 poises for komatiite P9-118 of Arndt et al. (1977), 15 poises for picrite NT-23 of Elthon (1979), and 55 poises for average FAMOUS glass composition reported by Elthon (1979); these calculations apply to 1,400° C. Comparable examples of rapid melt segregation have been reported in melting experiments on garnet-lherzolite and komatiite compositions (Arndt 1977; Green et al. 1975).



**Fig. 4.** **a** Experiment run on the bulk composition  $Fe_{30}$  at  $1,500^{\circ}C$ , 20 kilobars for 1.5 h. The segregated liquid layer at the roof of the charge is represented by quenched olivine. Accumulate textures are most strongly developed at the base and the left hand side of the charge. Width of capsule is 1.5 mm. **b** Back scatter SEM image of acumulate and mesocumulate textures near the middle of the charge. Reflective intercumulus specks are metallic Fe

## Discussion

There are many examples of igneous intrusions which apparently failed to differentiate by phase separation in a gravity field. However, there is also no paucity of examples where gravity-induced phase separation was an important geological process. The extent to which it operates as opposed to other mechanisms of differentiation can depend strongly on the composition of the magmas involved, the products of which are manifest in different geological scales of observation. Whereas tholeiitic to granitic magmas in the continental crust commonly display characteristics of in situ crystallization as envisaged by McBirney and Noyes (1979) for the Skaergaard intrusion, less silicic magmas such as partial melts in the mantle and ultramafic magmas in differentiating planets can give rise to strong phase separation even when the amount of liquid is small. Experimental results in the system forsterite-fayalite demonstrate this in part by showing rapid flotation and settling of olivine at 1 atmosphere and 20 kilobars respectively within a time duration of several hours. This gives rise to rapid segregation of a melt phase and the development of complementary accumulates. Although this simple binary differs chemically from natural systems in the mantle, it appears to be a good analogue for exploring the mechanics of fluid flow in porous media of mantle-like silicate systems (eg., Spera 1980).

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