

Forsterite dissolution in superheated basaltic, andesitic and rhyolitic melts

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Abstract

Dissolution rates of small forsterite spheres in superheated melts of basalt, andesite and rhyolite composition have been measured at 1300°C, atmospheric pressure. The rate is constant ($83 \mu\text{m hr}^{-1}$) in the basalt, regardless of run duration. In the andesite the initial dissolution rate is $200 \mu\text{m hr}^{-1}$, followed by a decrease to a constant value of $16 \mu\text{m hr}^{-1}$ in 2–3 hours. Dissolution rate in the rhyolite decreases from an initial value of 1.7 to $<0.1 \mu\text{m hr}^{-1}$ over 280 hours and never reaches a constant rate. Once the rate of dissolution has become constant, the film of contaminated melt that forms in melt about a crystal does not thicken with time, indicating attainment of a steady-state condition. Steady state is attributed to natural convection arising from the difference in density between the film of contaminated melt surrounding a crystal and that beyond. The density difference is approximately 2% of the density of the rock melt.

KEYWORDS: forsterite, dissolution rates, melts, basalt, rhyolite, andesite.

Introduction

RECENT contributions to the study of mineral dissolution in silicate melts have mostly used basic melts (see Donaldson, 1985, Table 1). Even in such fluid melts, dissolution rate is limited, not by the speed of the reactions occurring at the crystal–liquid interface, but rather by the speed at which the dissolved matter is removed from the interface into the melt. Characteristics of the dissolution kinetics are such that two mechanisms of transport are inferred to operate in the melt: diffusion of dissolved matter through the compositional boundary layer of melt that forms about the crystal, and natural convection tending to mix the boundary layer with the far-field melt (Kuo and Kirkpatrick, 1985; Donaldson, 1985, 1986; Brearly and Scarfe, 1986).

This paper reports results of further experiments on the dissolution of forsteritic olivine, designed to examine whether the mixed transport (diffusion and convection) mechanism also applies to melts more viscous than basic ones, and whether the length of incubation period for the onset of convection is related to melt viscosity.

The materials used as 'solvents' in the experiments are: a tholeiitic basalt from the Taos Plateau, New Mexico; an 'andesite' (or more accurately a 'boninite') prepared by addition of silica gel to the basalt; and a rhyolite from Glen

Table 1 Composition, superheating and viscosity of melt of each sample

| | Basalt ¹ | Andesite ¹ | Rhyolite ¹ |
|--------------------------------|---------------------|-----------------------|-----------------------|
| SiO ₂ | 52.2 | 57.7 | 75.1 |
| TiO ₂ | 1.2 | 1.1 | 0.2 |
| Al ₂ O ₃ | 15.8 | 14.3 | 14.3 |
| FeO | 9.8 | 8.8 | 0.9 |
| MnO | 0.2 | 0.2 | 0.1 |
| MgO | 7.3 | 6.4 | 0.2 |
| CaO | 8.9 | 7.6 | 0.6 |
| Na ₂ O | 3.1 | 2.6 | 3.9 |
| K ₂ O | 0.8 | 0.7 | 4.5 |
| | 99.8 | 99.6 | 99.8 |
| +ΔT, °C | 100 ± 4 | 150 ± 15 | 300 ± 25 |
| η, poise ² | 210 | 750 | 3.5 × 10 ⁵ |

¹ Electron probe analysis of glass bead melted at 1300 °C for 2 hours

² Computed by method of Shaw (1972).

Coe, Scotland. Homogeneous glasses of each composition were prepared by fusion and grinding at 1300°C and $p_{\text{O}_2} = 10^{-7}$ atm. (Table 1, in which all iron is reported as FeO). The olivines used are fresh phenocrysts from Igdlorsuit picrite intrusion, Ubekendt Ejland, west Greenland.

The experimental procedure involves determi-

nation of the change in radius of a small olivine sphere (approximately 1 mm diameter), following a period of dissolution. The experiments were conducted in a one-atmosphere, vertical, quenching furnace with p_{O_2} controlled by a CO-CO₂ gas mixture. An account of the methods employed has been given (Donaldson, 1985).

Pt wire-loop sample containers were used. During an experiment Pt reduces FeO in the melt and alloys with the liberated iron metal. In addition, some Na and K evaporate from the melt and are lost to the continuously flowing furnace atmosphere. At the flow rate, temperature and oxygen fugacity used ($30\text{ cm}^3\text{ min}^{-1}$, 1300°C , 10^{-7} atm., approx. the value of the QFM buffer) the losses of all three elements will be small (Donaldson, 1979; Corrigan and Gibb, 1979), particularly as most of the run durations were short (<0.5–16 hours). Very slow dissolution in the rhyolite, however, meant that some very much longer experiments were necessary, inevitably increasing the chemical changes in the melt. For example, electron probe analyses of the glass in a run of 280 hours duration reveal that approximately 10% of the iron and 30–40% of the Na and K were lost.

For a selection of the run products, the olivine-enriched film of glass adjacent to a crystal has been examined by individual 'spot' analyses ($\sim 1\ \mu\text{m}$ in diameter), obtained with a Jeol Superprobe, to investigate interface glass composition, film thickness, and composition variation in the film. All iron is reported as FeO in these analyses (Table 3). The form of these composition profiles (Fig. 2) indicates that sample quenching was fast enough to prevent regrowth of olivine on the forsterite.

Results

In thin section the sharp crystal-glass junctions are circular and smooth in outline. Crystals apparently remained spherical during dissolution, implying isotropic dissolution.

Experimental results are listed in Table 2 and plotted in graphs of change in radius versus time in Fig. 1 *a-c*. Some of the data on dissolution in the basalt were previously reported by Donaldson (1985).

The straight-line relationship between Δr and time for dissolution in the basalt (Fig. 1*a*) indicates that the rate ($83\ \mu\text{m hr}^{-1}$) is not a function of time, as would be expected if diffusion in the melt was the sole means of solute transport. Even in the shortest run conducted (9 minutes), the result is consistent with the constant rate; any initial transient condition must be shorter than this.

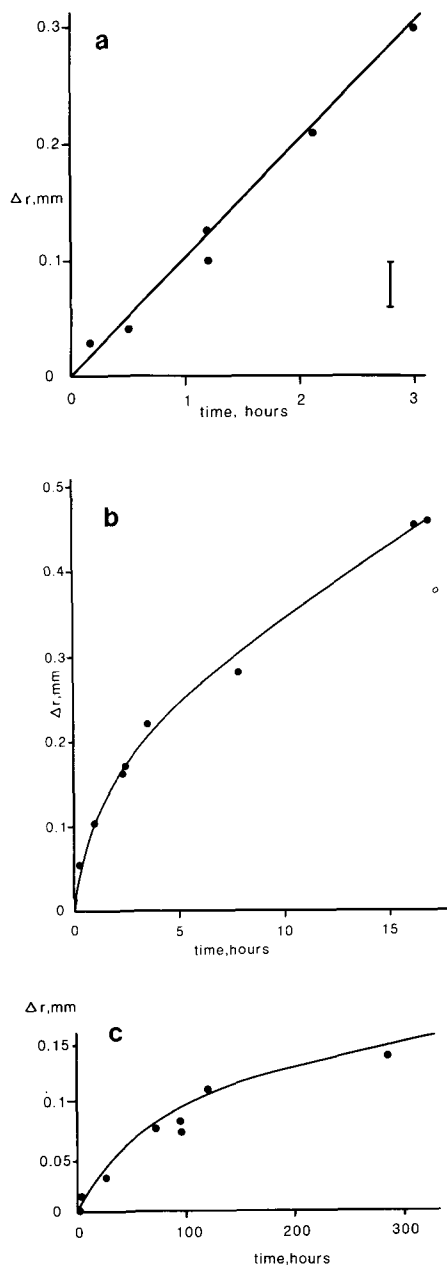


FIG. 1. Graphs of change in crystal radius versus run duration for (a) basalt, (b) andesite, and (c) rhyolite. Estimated error (2σ) in Δr is shown by bar. In (b) the circle indicates a minimum value of Δr . Lines drawn by eye.

Table 2 Data for individual runs

| Run no | Duration, hours | Initial diameter, mm | Final diameter, mm | Δ radius mm |
|-----------------|-----------------|----------------------|--------------------|--------------------|
| Basalt | | | | |
| 441 | 0.15 | 0.66 | 0.60 | 0.03 |
| 440 | 0.5 | 0.58 | 0.50 | 0.04 |
| 209 | 1.2 | 0.50 | 0.30 | 0.10 |
| 209a | 1.2 | 0.54 | 0.29 | 0.125 |
| 208 | 2.1 | 0.50 | 0.08 | 0.21 |
| 210 | 3.0 | 0.60 | 0 | >0.30 |
| Andesite | | | | |
| 372 | 0.3 | 0.65 | 0.54 | 0.055 |
| 371 | 1.0 | 0.66 | 0.45 | 0.105 |
| 375 | 2.25 | 0.95 | 0.63 | 0.16 |
| 373 | 2.4 | 0.36 | 0.02 | 0.17 |
| 366 | 3.5 | 0.74 | 0.30 | 0.22 |
| 368 | 7.9 | 0.82 | 0.26 | 0.28 |
| 374 | 16.25 | 0.92 | 0.01 | 0.455 |
| 365 | 17.25 | 0.75 | 0 | >0.375 |
| 378 | 17.3 | 1.90 | 0.98 | 0.46 |
| Rhyolite | | | | |
| 429 | 0.6 | 0.85 | 0.85 | 0 |
| 430 | 2.5 | 1.05 | 1.02 | 0.015 |
| 439 | 5.0 | 0.98 | 0.96 | 0.01 |
| 389 | 26.0 | 0.51 | 0.45 | 0.03 |
| 392 | 47.0 | 0.72 | 0.60 | 0.06 |
| 391 | 72.2 | 0.68 | 0.53 | 0.075 |
| 393 | 95.1 | 0.92 | 0.78 | 0.07 |
| 394 | 95.5 | 0.88 | 0.72 | 0.08 |
| 390 | 119.2 | 0.51 | 0.29 | 0.11 |
| 395 | 283 | 0.88 | 0.60 | 0.14 |

In contrast, the data for dissolution in the andesite melt describe a curve whose slope initially decreases with run duration and then becomes constant after 2–3 hours (Fig. 1*b*). The initial dissolution rate ($200\mu\text{m hr}^{-1}$) greatly exceeds that in the basalt melt, as would be expected from the larger driving force for dissolution, as represented by $+\Delta T$ (Table 1). The rate then diminishes to approximately one quarter of the value in the basalt, probably reflecting slower transport of dissolved olivine in the more viscous andesitic melt.

Dissolution rate in the rhyolitic melt also decreases with run duration (Fig. 1*c*), from an initial rate of $\sim 2\mu\text{m hr}^{-1}$, to approximately $0.7\mu\text{m hr}^{-1}$, but the pattern of data points shows that the rate continues to decrease up to 280 hours, i.e. constant rate was not established. Early dissolution is extremely slow compared with that in the basaltic and andesitic melts, despite the much larger $+\Delta T$ (Table 1), suggesting much slower kinetics for the reactions that detach ions from the crystal. Overall, the small dissolution rates in the rhyolite melt are to be expected as a result of slow material transport in a very viscous melt.

Fig. 2*a–e* illustrates the variations in MgO and

SiO₂ that exist in glass near to a crystal, for each of the three rock melts, and also indicate the effect on these composition profiles of run duration. Dissolution of a solid richer in Mg and poorer in Si than each of the melts, causes formation of a 'boundary layer' or 'contaminated film' of melt in which Mg decreases and Si increases outwards from the crystal. In the basalt, for example, glass at the crystal–liquid interface has *ca.* 3.5 wt.% more MgO and 2.5 wt.% less SiO₂ than that far from the interface (Fig. 2*a*). FeO is *ca.* 1 wt.% enriched near the interface and other constituents are depleted (Table 3). Regardless of run duration, the composition profiles for dissolution in basalt are virtually identical. The effective thickness of the composition boundary layer (δ_{cff}) is *ca.* 120–180 μm , the SiO₂ profile giving the larger figure. In the case of the andesite melt (Fig. 2*b, c*), the profiles change with time up to approximately 3 hours of dissolution, after which no change occurs. δ_{cff} ranges from $\sim 100\mu\text{m}$ in the shortest run illustrated, to 150–200 μm in the longest. As regards dissolution in the rhyolite (Fig. 2*d, e*), the compositional gradients evolve with time from steep to relatively shallow ones. δ_{cff} increases from *ca.* 50 to 150–200 μm over 120 hours.

Discussion

The limitation of dissolution rate. Dissolution requires that reactions occur on the crystal–liquid interface to remove ions from the lattice and incorporate them in the melt, and also that the dissolved matter is transported from the interface and dispersed through the melt. If the dissolution rate was limited solely by the speed of interface-detachment reactions, then the contaminated film of melt about a crystal would effectively be non-existent. For the present experiments this possibility can be excluded. Whether the dissolution rate is limited exclusively by transport in the melt or by a combination of transport and interface-detachment kinetics cannot be established for dissolution in rock melts. It can be established in a simple system for which the phase relations are known, because pure transport control of dissolution rate causes the melt in contact with the crystal to be in equilibrium with the crystal at that temperature (cf. Oishi *et al.*, 1965). Recent experiments by Kuo and Kirkpatrick (1985) dissolving crystals of quartz, olivine, diopside and enstatite in several melts in the system $\text{Mg}_2\text{SiO}_4\text{–CaMgSi}_2\text{O}_6\text{–SiO}_2$ (at 1 atm.) demonstrated a close approach to surface equilibrium in all runs, consistent with essentially pure transport control of dissolution rate. As illustrated in Fig. 3, several

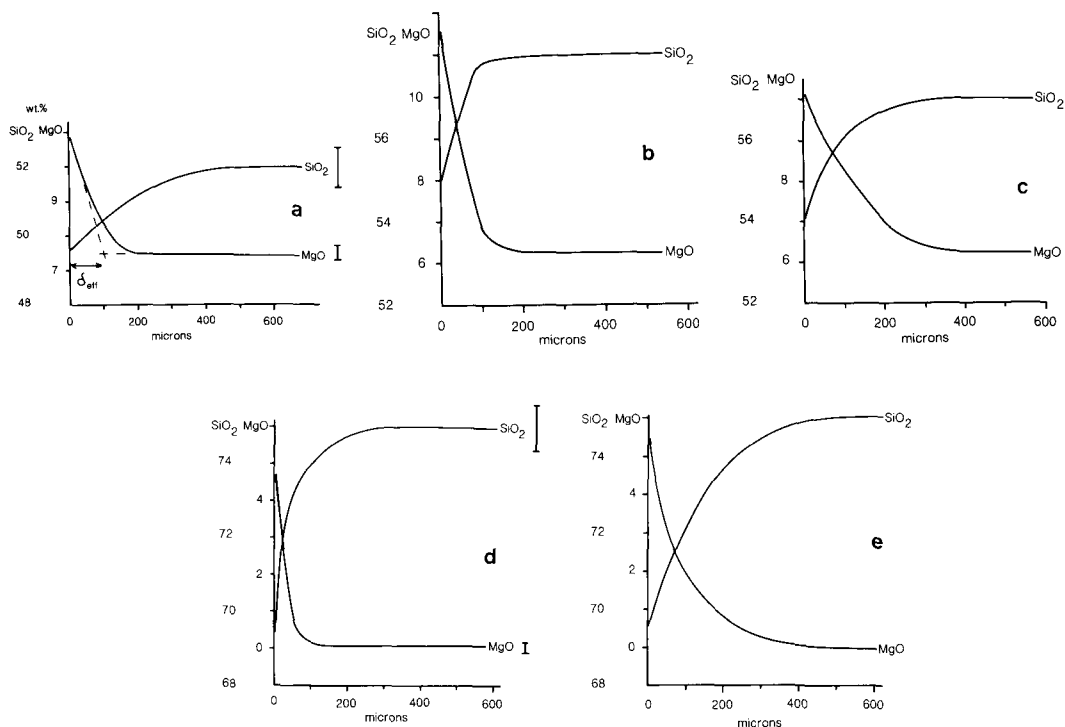


Fig. 2. MgO and SiO₂ variations in glass near partially dissolved crystals: (a) basalt, 1.2 hours, run 209 (b) and (c) andesite, 0.3 and 3.5 hours, runs 372 and 366 respectively; (d) and (e) rhyolite, 0.6 and 72 hours, runs 429 and 391 respectively. Estimated errors (2σ) shown by bars in (a) and (d). The lines have been drawn to smooth out the scatter associated with individual analyses. The means of estimating δ_{eff} is indicated in (a).

Table 3 Selected analyses of glass adjacent to partially dissolved forsterite (i.e. within 5 microns of crystal)

| | Basalt | | | Andesite | | | Rhyolite | | |
|--------------------------------|--------|------|------|----------|------|------|----------|------|------|
| Run | 480 | 209 | 210 | 372 | 375 | 378 | 438 | 392 | 394 |
| Duration hr | 0.5 | 1 | 3 | 0.3 | 2.5 | 17 | 0.6 | 47 | 95 |
| SiO ₂ | 49.6 | 49.4 | 49.6 | 55.1 | 55.2 | 55.7 | 69.5 | 69.3 | 69.8 |
| TiO ₂ | 1.1 | 1.0 | 1.1 | 0.8 | 0.9 | 0.8 | 0.1 | 0.1 | 0.1 |
| Al ₂ O ₃ | 13.2 | 13.5 | 13.4 | 11.7 | 11.3 | 11.8 | 13.2 | 13.3 | 12.9 |
| FeO | 10.8 | 11.0 | 11.0 | 9.1 | 8.9 | 8.3 | 2.5 | 2.6 | 2.7 |
| MgO | 11.2 | 11.2 | 10.9 | 11.8 | 12.5 | 12.2 | 5.2 | 6.3 | 6.3 |
| CaO | 9.0 | 9.1 | 8.8 | 7.7 | 6.4 | 7.1 | 1.3 | 1.6 | 1.4 |
| Na ₂ O | 2.6 | 2.7 | 2.7 | 1.9 | 2.3 | 2.0 | 2.4 | 2.7 | 2.6 |
| K ₂ O | 0.5 | 0.5 | 0.6 | 0.5 | 0.6 | 0.5 | 3.8 | 3.2 | 3.4 |
| | 98.0 | 98.4 | 98.1 | 98.6 | 98.1 | 98.4 | 98.0 | 99.1 | 99.2 |
| K_D^1 | 0.18 | 0.18 | 0.17 | 0.23 | 0.25 | 0.26 | 0.35 | 0.42 | 0.42 |

¹

$$\frac{X_{\text{FeO}}^{\text{ol}} \cdot X_{\text{MgO}}^{\text{liq}}}{X_{\text{FeO}}^{\text{liq}} \cdot X_{\text{MgO}}^{\text{ol}}}$$

(Roeder and Emslie, 1970)

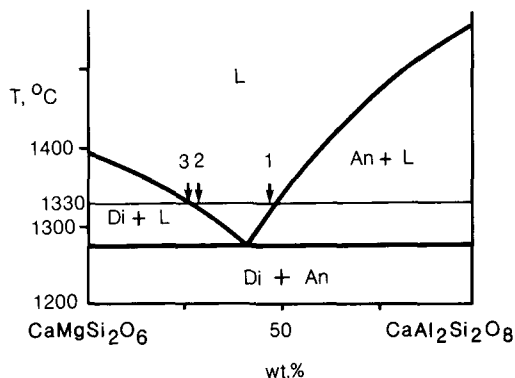


Fig. 3. Composition of glass (points 2 and 3 as obtained by electron probe analysis) adjacent to diopside crystal partially dissolved in melt of An₁₇Di₅₃ composition (1), at 1330 ± 5°C for 23 minutes (2) and 12 minutes (3), respectively. Phase relations from Bowen (1913).

experiments by the present author involving dissolution of diopside crystals in superheated melts in the system $\text{CaAl}_2\text{Si}_2\text{O}_8\text{-CaMgSi}_2\text{O}_6$ also attain surface equilibrium, within analytical error, even at very short run times. By analogy with these haplomagmatic systems, it is suggested that reactions on the surface of an olivine crystal are capable of releasing ions into rock melt more rapidly than they are transported away. In the simple systems mentioned, the melts used in the experiments have viscosities comparable to or less than those of basalts. As diffusion and viscous flow become more sluggish with increasing SiO_2 content of a melt, it is unlikely that detachment kinetics will replace transport as the dominant control of dissolution rate in the andesitic and rhyolitic melts.

Values computed for the well-known Roeder and Emslie (1970) distribution coefficient between olivine and liquid (Table 3) differ from the generally accepted figure of *ca.* 0.3 (cf. results of Zhang *et al.*, 1987, for dissolution of Fo_{90} crystals in basaltic andesite at 5 kbar and 1300°C). These calculations assume, unrealistically, that the analysed glasses contain no Fe_2O_3 ; corrections for this effect raise K_D by *ca.* 0.03 in the basalt, by *ca.* 0.04 for the andesite and by *ca.* 0.15 for the rhyolite (data of Sack *et al.*, 1980, were used to assess the likely $\text{Fe}_2\text{O}_3\text{:FeO}$ ratio of each melt at the approximate p_{O_2} of the quartz-fayalite-magnetite buffer). While $\text{Fe}^{2+}\text{-Mg}$ surface equilibrium may exist in the andesite, in the basalt there is a relative shortage of Mg and excess of Fe in the interfacial melt compared with that necessary for surface equilibrium; in the rhyolite the opposite obtains. Whether this departure from equilibrium is due only to multicomponent diffusive effects or additionally to interface-detachment kinetics is not clear, though the former is favoured by the fact that, at the end of an experiment, probe analyses reveal no marginal Mg-Fe zoning of olivine crystals.

Nature of the transport. Matter may diffuse, or convect, or both diffuse and convect in the melt during transport of dissolved olivine from the crystal. The existence of the composition gradients about crystals implies that if convection occurred it was never so vigorous as to reach to the crystal. On the other hand, if transport is solely by diffusion, Δr should be proportional to the square root of time (Christian, 1965) and the compositional boundary layer should thicken steadily with time. Olivine dissolution in the basalt does not conform to this behaviour, rather a steady state exists in which Δr is proportional to time (Fig. 1a) and δ is constant with time. These characteristics imply that there was convection in

this melt which prevented unrestricted growth of the contaminated film of melt about crystals (cf. Donaldson, 1985; Kuo and Kirkpatrick, 1985).

Dissolution in the andesite involves an early, transient stage during which Δr is proportional to the square root of time (Fig. 4a) and thereafter a stage in which Δr is proportional to time (Fig. 1b); apparently an early interval of purely diffusive transport gave way to one of mixed diffusive and convective transport. During the purely diffusive stage the composition gradient thickened

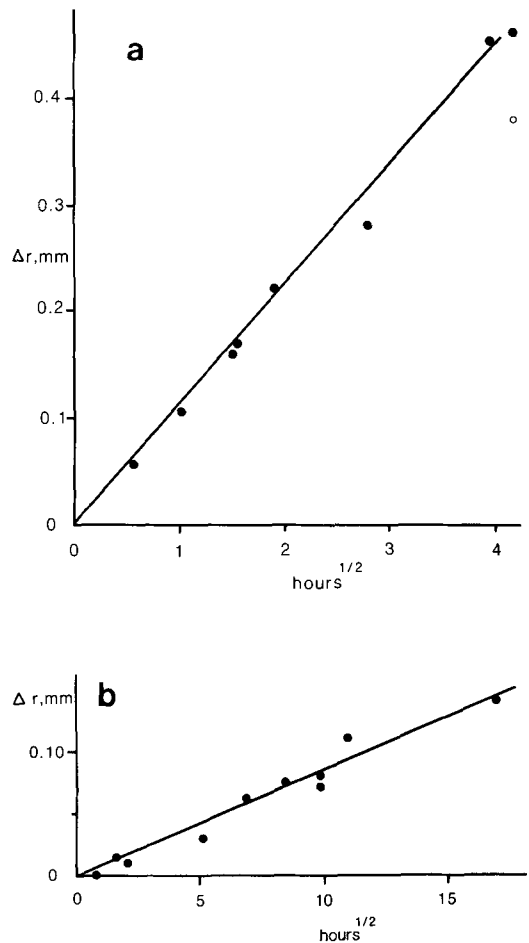


Fig. 4. Graphs of change in crystal radius versus square root of run duration for (a) andesite and (b) rhyolite.

until after 3 hours it had attained a thickness of 150–200 μm , convection must then have begun, preventing further thickening of the film.

Absence of a straight-line relationship between

Δr and time for dissolution in rhyolite, even after long duration, indicates that transport was only ever by diffusion, as might be expected in such a viscous melt. There is no means of assessing whether all the observations for this melt involve a transient condition evolving to a steady state of combined diffusive and convective solute transport, or whether the large viscosity will always prevent convection. Even if convection does set in at yet longer run times, it will undoubtedly be extremely slow and the dissolution rate will be no greater than $0.2 \mu\text{m hr}^{-1}$.

Cause of convection. The density variations in a fluid that cause convection are customarily assumed to result from temperature differences. Yet, as previously discussed by Donaldson (1985), even if a temperature gradient existed in the charges in the present nominally isothermal experiments, it could not possibly be large enough to initiate fluid motion.

Several other processes could be responsible for the inferred convection. 1. Motion of the crystal within the melt would cause shearing and mixing between the film of 'contaminated' melt and the bulk melt. However, it is found that olivine

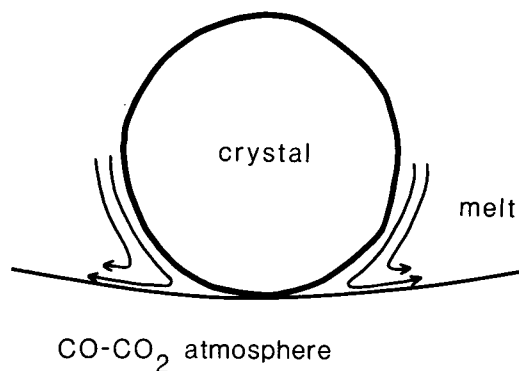


Fig. 5. Sketch of slice through experimental charge showing how convection might arise due to variation in surface tension of melt on the outside of the charge. It is assumed that the distance between the crystal and the bottom of the charge is less than δ_{eff} so that the composition of melt on the surface of the charge is not uniform. Melt would flow from an area of high surface tension to low; this is illustrated as being outwards but it could be the opposite.

crystals settle to the lower reaches of basalt charges within minutes of melting; for this means of convection to continue thereafter would require jiggling of the crystal which is unlikely in the absence of thermal convection. 2. Wilcox

(1983) notes that when crystals grow or dissolve in a solution, motion of the crystal-liquid interface itself can cause pushing or sucking of the melt; the same can arise due to differences in volumetric properties of the crystal and melt. Against the operation of these processes is the fact that there is a transient condition for dissolution in the andesite melt, and possibly in the rhyolite too; they cannot, however, be excluded as explanations of convection in the basalt for which no transient condition was detected. 3. Convection might also arise due to difference in surface tension between boundary layer and bulk melts, if a settled crystal is so near to the bottom of the charge that the film of contaminated melt locally contacts the atmosphere (Fig. 5; cf. Donaldson and Henderson, 1988). Typically, however, there is a layer of melt between the crystal and margin of the charge that is thicker than the contaminated film, and probe analyses do not reveal distension of the film near the bottom of a charge. 4. There are now many documented instances of fluid motion about crystals growing or dissolving in aqueous solutions and alloy metals, the motion being generated by the differences in density between the film of solute-depleted or -enriched solution around the crystal and that beyond (Wilcox, 1983; Hebditch and Hunt, 1973). This is one example of what has come to be known in petrology as 'compositional convection'. On balance, this seems the most likely cause of the inferred

Table 4 Difference in computed¹ density of interface and far-field melt (g cc^{-1})

| | Basalt | Andesite | Rhyolite |
|-------------------------------|--------|----------|----------|
| ρ interface | 2.68 | 2.61 | 2.41 |
| ρ far field ² | 2.62 | 2.56 | 2.31 |
| $\Delta\rho$ | 0.06 | 0.05 | 0.10 |
| $\Delta\rho \cdot 100$ | 2.3 | 1.9 | 4.3 |
| ρ far field | | | |

¹ Using partial molar volumes reported by Mo et al. (1982)

² Computed from analysis > 1000 μm from crystal.

fluid motion, but there is no evidence in the run products, or in the probe analyses of those products, for the existence of plumes or 'blobs' of glass of boundary layer composition isolated in glass distant from the crystal that would clinch this interpretation. Table 4 lists the computed densities of the basalt, andesite and rhyolite melts

and of the melts adjacent to the dissolved olivines. The differences in density assumed to drive convection are extremely small, ca. 2% of the bulk melt density. For comparison, note that the density difference in the (non-convecting) rhyolite melt exceeds this value.

Transport of dissolved matter by both diffusion within a boundary layer and compositional convection has previously been invoked by Kuo and Kirkpatrick (1985), Donaldson (1985) and Brearley and Scarfe (1986) to account for the kinetic characteristics of mineral dissolution in silicate melts, though see Thornber and Huebner (1985) for a diffusion-only interpretation. Compositional convection is also much favoured in recent petrologic literature as a means of speeding mass transport during crystal growth [e.g. in adcumulus growth (Morse, 1986)] and during wall-rock contamination of magma (e.g. Turner and Campbell, 1986; McBirney *et al.*, 1985). While the process has often been demonstrated in low-temperature aqueous solutions (see review of Turner and Campbell, 1986), there is just one record of its occurrence in silicate melts, in an experiment in which silica glass was dissolved in picrite melt (Donaldson and Hamilton, 1987). On the other hand, experiments by Zhang *et al.* (1987), in which horizontal plates of forsterite dissolved in basaltic andesites at 1300° and 5 kbar, did not develop convection after run times of up to 24 hours; during dissolution the contaminated film around crystals extended outwards to about 1 mm, according to $\sqrt{\text{time}}$. Such experiments are in their infancy and many more are needed in different melts and with different minerals to fully assess the possibility and importance of compositional convection in magmas.

Petrologic comment. Should a superheated magma of basalt or andesite or rhyolite composition become contaminated with olivine crystals (either from wall rocks or by mixing with mafic magma), the data presented here show that dissolution will be most rapid in the basalt melt and much slower as the melt becomes more siliceous, this despite the fact that forsterite-rich olivine should be most unstable in the rhyolite melt. In Nature, where any superheat in andesite and rhyolite would normally be much less than the values used in these experiments, the differences in dissolution rate between the basalt and the other two melts of course will be even more pronounced.

Given the very slow dissolution rates in intermediate and acid melts, it seems likely that any superheat possessed by such magmas would be eradicated by cooling before olivine was dissolved. Under this circumstance the olivine crystals

would be converted to pyroxene or amphibole by discontinuous reaction. In calc-alkaline suites, where evidence of magma mixing and assimilation is common, it is possible that some pyroxene, amphibole and biotite crystals are reacted olivine xenocrysts, rather than primocrysts from melt. The kinetics of this and other likely discontinuous reactions need investigation.

Summary

These dissolution experiments of a single mineral at a single temperature show clearly the key role played by transport, especially diffusion, in determining the dissolution rate. They also demonstrate how an early transient stage of diffusion-only transport can give way to a combined diffusion and convection stage, once a thick enough film of contaminated melt is established. Furthermore, the influence of melt composition and viscosity on the duration of the transient condition is shown by the very short duration in the basalt (<9 minutes), the 3-hour duration in the andesite, and the <280 hours transient condition in the rhyolite.

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