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# The Fluid Dynamics of Evolving Magma Chambers [and Discussion]

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## The fluid dynamics of evolving magma chambers

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[Plates 1 and 2]

Recent developments in petrology indicate that fluid dynamic effects are of fundamental importance in controlling magma genesis. The forms of convection in magma chambers arise from compositional variations caused by processes such as fractional crystallization, partial melting and contamination, as well as from thermal effects. These processes, together with phase changes such as volatile exsolution, generally cause much larger density changes in magmas than the thermal effects arising from associated temperature changes. Magmas exhibit a wide range of convective phenomena not encountered in one-component fluids that are due to these compositional changes and to the differences between the diffusivities of chemical components and heat. When crystallization occurs in such multi-component systems, fluid immediately adjacent to the growing crystals is generally either depleted or enriched in heavy components and can convect away from its point of origin. Experimental studies of convection in crystallizing systems together with theoretical analyses suggest that convective separation of liquid from crystals is the dominant process of fractionation in magmas. This paper provides a synopsis of these new ideas on convection in magmas and their application to the interpretation of igneous rocks.

Crystal settling is shown to be an inadequate and, in many situations, improbable mechanism for fractional crystallization. The convective motions in chambers are usually sufficiently vigorous to keep crystals in suspension, although settling can occur from thin fluid layers and within the boundary layers at the margins of a magma chamber. We propose that *convective fractionation*, a term introduced to embrace a wide variety of convective phenomena caused by crystallization, is the dominant mechanism for crystal fractionation. The process enables compositional and thermal gradients to be formed in magma chambers both by closed-system crystallization and by repeated replenishment in open systems. During crystallization along the margins of a chamber, highly fractionated magmas can be generated without requiring large amounts of crystallization, because the removal and concentration of chemical components affects only a small fraction of the total magma. These convective effects also give insights into many features observed in layered intrusions, including the various types of layering and the formation of different kinds of cumulate rock.

### 1. INTRODUCTION

The last few years have seen considerable changes in ideas on the mechanisms of magmatic differentiation and on the processes that occur in magma chambers. In particular, geological and fluid dynamical studies suggest that the processes of crystallization, cooling and convection in magma chambers are likely to be much more complex than hitherto supposed. This article provides a synopsis of these new concepts that are largely based on recent developments in the study of convective phenomena in both multi-component fluids and crystallizing fluid systems.

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We believe that the emergence of these ideas will require a fundamental re-examination of many igneous processes.

Much of petrological research in the last half century has been concerned with investigations of chemical equilibria in silicate melt systems. The general approach was strongly influenced by N. L. Bowen's ideas (Bowen 1928) of the dominant role of fractional crystallization in the generation of the wide diversity of igneous rock compositions. This view was given strong support by field and petrological studies of igneous rocks, particularly from layered igneous intrusions (Wager & Brown 1967; Jackson 1971). In more recent times, partial melting has also come to be regarded as a major process in magma generation. Nevertheless, as techniques for geochemical and petrological analysis have become more sophisticated, evidence has accumulated to convince the majority of petrologists that most igneous rocks have been strongly influenced by fractional crystallization during their evolution. Detailed geochemical and petrological studies have also made it apparent that magma mixing and contamination can be important processes, often in modifying the details of fractionation.

The physical process that allowed fractional crystallization to take place has conventionally been based on a very simple concept. Crystals were considered to nucleate and grow within the magma and then to settle out to form cumulate rocks, or were erupted to form phenocrystal-bearing lava. Although many other processes were recognized, such as *in situ* crystallization on the margins of the magma chamber, magma mixing, contamination, immiscibility and liquid-state diffusion, they were widely regarded as subordinate to crystal settling in importance.

The important elements often neglected in conventional models of igneous petrogenesis were the dynamics of magma chamber processes. In the case of fractional crystallization, we shall demonstrate the overriding importance of convection in determining the nature of fractionation. Crystal settling will be shown to be an inadequate mechanism for causing fractionation, both from a theoretical viewpoint and from an appraisal of the geological evidence. From recent theoretical and experimental studies on fluid motions in both multi-component fluids and crystallizing fluid systems, we will demonstrate that the convective removal of liquid, as crystals form, is an attractive mechanism for fractionation of magmas. This mechanism is given the general name *convective fractionation*, which is used as a term to embrace a wide variety of convective phenomena in magma chambers. We will discuss a number of important characteristics of igneous rocks that are well explained by the new concepts.

## 2. CONVECTION IN MULTICOMPONENT SYSTEMS

Until the last few years, nearly all the applications of convection theory to magma chambers assumed that the relevant fluids could be described as simple one-component systems, in which the major cause of density variations was attributable to temperature differences. Shaw (1965) established that thermal convection would be important even in high viscosity granitic magma chambers. Shaw (1965, 1974) also recognized that compositional effects and phase changes, particularly gradients of volatile components and bubble content, could be important, and could lead to compositional stratification in a magma chamber.

The major problem with the application of thermal convection theory to magma chambers, however, is that magmas are multi-component systems and such systems can exhibit a variety of novel convective phenomena not encountered in one-component fluids (Turner 1979; McBirney & Noyes 1979; Huppert & Turner 1981*a*). The difference between single-component

and multi-component fluids can be illustrated conveniently by comparing two simple systems: one in which pure water, and the other in which water with a stable salinity gradient, is heated from below. In the case of pure water, small parcels of fluid heated at the base of a container expand and, finding themselves lighter than the surrounding fluid, ascend to drive convection throughout the whole system. In the case of a salinity gradient heated from below (Turner 1968; Huppert & Linden 1979), parcels of fluid at the base are heated and rise. However, they ascend into overlying fluid of decreasing temperature and salinity. Such parcels lose their heat faster than their salt content because of the larger diffusivity of heat. Consequently, the fluid parcels reach a level where they become equal in density to the surrounding fluid. They can rise no further and on losing more heat sink back, so that a well-mixed convecting layer is formed beneath the overlying gradient region. This layer heats the fluid above and another layer forms. This process is repeated until the whole system consists of a series of convecting layers each internally homogeneous with respect to composition and temperature. For aqueous solutions the scale of the layers can be predicted from the physical properties of the system (Huppert & Linden 1979). The convecting fluid layers are separated from one another by sharp diffusive interfaces across which heat and salt are transported by molecular diffusion. A crucial feature of such a system is that heat is transferred convectively through a fluid which has a stable density distribution.

The case of a salinity gradient heated from below is just one example of many novel convective phenomena that can occur in fluids with components of different diffusivities. Such phenomena are often referred to under the term double-diffusive convection. Turner (1979) and Huppert & Turner (1981*a*) provide more detailed treatments and discussion of the wide variety of effects attributable to double-diffusive convection. A major thesis of this paper is that double-diffusive effects are inevitable in magma chambers.

Thermal convection can be described in terms of two dimensionless parameters which characterize the motion. These parameters are the thermal Rayleigh number ( $Ra_T$ ) and Prandtl number ( $Pr$ ) defined as

$$Ra_T = g\alpha\Delta Td^3/\kappa_T\nu, \quad (1)$$

$$Pr = \nu/\kappa_T, \quad (2)$$

where  $g$  is the gravitational acceleration,  $\alpha$  the thermal expansion coefficient,  $\Delta T$  the temperature difference across the layer,  $d$  the fluid layer thickness,  $\kappa_T$  the thermal diffusivity and  $\nu$  the kinematic viscosity. For fluids with large Prandtl number (including all magmas) convection at Rayleigh numbers greater than around  $10^6$  is fully turbulent, taking the form of chaotic, unsteady motions. Steady, two-dimensional Rayleigh–Bernard cells are sometimes shown in sketches of magma chambers, with a central upwelling region and marginal downwelling regions. Such steady, two dimensional patterns of convection, however, require rather low Rayleigh numbers ( $1760 < Ra_T < 22600$ ) and are unlikely to be the normal convective pattern in magma chambers. In the vast majority of cases, for which  $Ra_T > 10^6$  (Shaw 1965; Bartlett 1969), a fully turbulent character should be regarded as the commonest style of convection.

In double-diffusive convection, two additional dimensionless parameters are required to characterize the motion: the compositional Rayleigh number ( $Ra_s$ ) and diffusivity ratio ( $\tau$ ) given by

$$Ra_s = g\beta\Delta Sd^3/\kappa_T\nu, \quad (3)$$

$$\tau = \kappa_s/\kappa_T, \quad (4)$$

where  $\beta$  is the coefficient of expansion due to compositional changes,  $\beta\Delta S$  the fractional density change across the fluid layer due to composition and  $\kappa_s$  the compositional diffusivity. The behaviour of a system cannot be specified by just  $Ra_T$ . Valid conclusions would only result if it is certain that there are no compositional gradients in a magmatic system. This will seldom be the case.

In double-diffusive systems, the characteristic length scale  $d$  will not be the container (magma chamber) depth, but the depth of the fluid layers that form. For example, in the case of the salinity gradient heated from below the appropriate length-scale  $d$  will be the thickness of an individual layer in calculating Rayleigh numbers.

For magmatic systems, the most important convective situation is the 'diffusive' régime (Turner 1979) in which compositional gradients are stabilizing, but temperature gradients are destabilizing. This is so because the more differentiated the magma, the lower its temperature and density (with the important exception of some basalts). The lightest coolest magmas tend, by a wide variety of processes to be described later, to accumulate at the top of a chamber, implying that the compositional variations have a larger effect on density than do the temperature gradients.

An important feature of double-diffusive layers was dramatically illustrated in a laboratory experiment described by Turner *et al.* (1983). A layer of hot  $KNO_3$  solution containing nitric acid was emplaced beneath a layer of  $K_2CO_3$  solution. While the lower  $KNO_3$  layer was hotter than the upper,  $K_2CO_3$  layer it was denser due to the  $KNO_3$  concentration and thus ponded at the base of the container. The two layers convected vigorously as heat was exchanged across the diffusive interface. Negligible chemical transfer occurred between the two layers, since the diffusivity of chemical components was so very much less than the diffusivity of heat ( $\tau \ll 1$ ). No chemical reaction took place between the nitric acid and  $K_2CO_3$  until the densities of the two fluids became equal so that overturning and mixing occurred leading to the vigorous production of  $CO_2$  bubbles. The experiments demonstrate that before overturning occurs fluids of different composition can coexist and exchange heat, but will evolve as chemically independent systems.

The most important convective effects, which have not been taken into account in conventional models, are due to crystallization. The essential principle is that light or heavy components are selectively removed from the liquid next to the growing crystals. This changes the melt density locally and enables compositional convection to occur. This process is believed to be of great importance in understanding magmatic evolution. Detailed discussion of convection in crystallizing systems is presented in § 5. Convection induced by crystallization can occur in a completely stable temperature gradient. For example, if a saturated solution is cooled from below, crystallization (removing heavy components) can release light fluid which drives purely compositional convection.

Another mechanism for causing density changes in magma chambers is related to phase changes. When crystals grow internally within a magma or are concentrated by some process, the bulk density of the magma will change. Little work has been done concerning the effects of suspended crystals on convection, but the density changes involved are considerably larger than those due to purely thermal effects. Gas exsolution provides another mechanism for causing convection or alternatively stabilizing systems. Large density changes can occur when gas exsolves and this can cause convection and mixing. Recent studies have suggested that mixing phenomena in calc-alkaline magmas can be attributed to volatile exsolution (Eichelberger 1980; Huppert *et al.* 1982*a*; Turner *et al.* 1983).



## 3. DENSITY VARIATIONS IN MAGMAS

Sparks & Huppert (1984) have considered the density changes that occur during the fractional crystallization of basaltic magma. Figure 1 shows schematically the variations in density of mafic magmas related to one another by crystal fractionation. Density is plotted against a convenient index of differentiation,  $Mg/(Mg + Fe)$ , which can, for a particular system, be related to liquidus temperatures. On the left side a high temperature primitive magma, such as a picritic basalt, fractionates mafic phases (olivine with or without pyroxenes) and generates successive residual liquids of decreasing density. The decrease in density occurs because the components removed from the fluid during crystallization are denser than the melt (Sparks & Huppert 1984). Although the residual melts are cooler, they are lighter because compositional effects dominate the density variations.

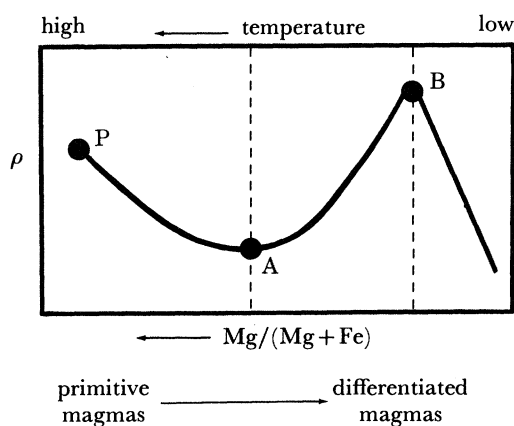


FIGURE 1. Schematic relation between magma density  $\rho$  and  $Mg/(Mg + Fe)$  for basaltic melts related to one another by fractional crystallization.

When co-crystallization of plagioclase with mafic phases occurs, the fluid density of the components being removed by crystallization often becomes lower than that of basaltic magma. Consequently, dry basaltic magmas fractionating cotectic assemblages containing plagioclase will generally increase in density, resulting in the formation of a density minimum (marked A in figure 1). Eventually, in highly differentiated basaltic magmas, dense phases such as titanomagnetite and ilmenite will join the crystallizing assemblage and olivines and pyroxenes will become iron-rich and dense. A density maximum can occur marked (B in figure 1) due to the removal of such dense phases. In more differentiated magmas (andesite–dacite–rhyolite) residual melt density decreases further during fractionation.

The case of density variations in evolving basaltic magmas (discussed in much greater detail by Sparks *et al.* 1980; Stolper & Walker 1980; Sparks & Huppert 1984) illustrates two fundamental principles that will be true of all magmas.

(i) Compositional changes will usually be larger than temperature changes in their effects on melt density during fractionation. In many systems, magma density *decreases* during fractionation despite the decrease in temperature.

(ii) Changes in slope of density–composition curves like figure 1 can be expected when new phases join old phases during fractionation. Maxima and minima can also be expected.

These principles will apply in any magmatic system. Furthermore, the remarks not only apply to liquids related by fractionation, but also to those linked by other processes. For example, fractional partial melting will produce successive melts which differ from one another in density largely because of compositional differences. Dynamic models of partial melting (Oxburgh 1980) imply that there will be spatial and temporal variations in magma compositions. If successive partial melts are fed into a chamber, compositional gradients can arise. Likewise, contamination of magma leads to density differences between contaminated and uncontaminated magmas which will generally be dominated by compositional effects rather than by the temperature changes associated with contamination.

#### 4. CRYSTAL SETTLING

Fractional crystallization implies separation of solid and residual liquid. Crystal settling provides an attractively simple and intuitively obvious mechanism to segregate crystals and liquid, but there are a number of inadequacies of this mechanism as a general process capable of explaining chemical variations of evolving magmas.

An important point, sometimes not clearly made in the literature, is that crystal settling is not synonymous with crystal fractionation. It is merely a physical mechanism that enables fractionation to occur. There is a major alternative to settling, which is the convective separation of residual liquid from crystals. At first sight, the difference may appear somewhat semantic. Does it really matter how solid or liquid move with respect to one another? We shall establish later that both the dynamics and the subsequent effects of these two alternatives are profoundly different. Interpretation of many features of igneous rocks will be strongly influenced by which process is considered as dominant. We review geological evidence which has been cited to support crystal settling and comment on some important physical constraints on the process.

##### (a) *Geological evidence*

The best evidence for crystal settling comes from relatively high temperature mafic and ultramafic lavas. In some picritic pillow lavas from Iceland, olivine crystals are observed to increase in size and abundance towards the base. In many komatiite lava flows, the lower half of the lava is highly enriched in olivine phenocrysts (Arndt *et al.* 1977). Geochemical evidence also points to some porphyritic basalt types being accumulative. For example, in m.o.r.b. lavas, plagioclase-rich varieties have bulk compositions which are never found as glass compositions in pillow margins (Flower 1980). A convincing case can be made for their compositions being controlled by concentration of plagioclase phenocrysts. Similar persuasive arguments can be made for some olivine-phyric lavas having been controlled by concentration of olivine, the lavas on Hawaii being a pertinent example (Wright 1971).

Although the above examples provide reasonably good evidence for crystal settling, even these cases are not without ambiguity and complications. Some phenocryst-rich lavas represent original liquids that have undergone extensive crystallization. Considerable detailed knowledge of the petrology and volcanological circumstances is required to demonstrate an accumulative origin (as is the case for Hawaiian and the m.o.r.b. lavas). Other processes of concentration can generate accumulative lavas (for example, flow differentiation and erosion and incorporation of pre-existing cumulate crystals within the chamber and conduits).

The most widely cited evidence for crystal settling comes from layered intrusions, following on from the model for the Skaergaard intrusions, summarized by Wager & Brown (1967). Paradoxically, such intrusions now provide perhaps some of the best evidence *against* settling. The crux of the argument is related to the plagioclase problem. Plagioclase is the major crystal phase of the Skaergaard and many other layered intrusions, and is demonstrably lower in density than Skaergaard liquids (McBirney & Noyes 1979; Campbell *et al.* 1978). The only physically reasonable explanation so far proposed to account for the occurrence of feldspar in the cumulates on the floor is to invoke *in situ* crystallization (McBirney & Noyes 1979; Morse 1979; see also Irvine 1980*b*). Similar problems arise with a settling model for mafic crystals found in the Upper Border Zone and Marginal Border Group of Skaergaard. The reader is referred to McBirney & Noyes (1979) and Campbell (1978) for more comprehensive discussions of the problem. The best evidence for crystal settling in layered intrusions comes from ultra-mafic bodies. For example, normally graded layers of olivine crystals in the Duke Island intrusion are attributed to settling from a density current (Irvine 1980*b*). Thus the available evidence does suggest that crystal settling has a role in basaltic magma evolution, but it certainly cannot be the only process or necessarily the most important.

In silicic igneous rocks convincing evidence for crystal settling is less apparent. In plutonic rocks the best evidence for compositional zonation due to fractional crystallization comes from the horizontal zoning from mafic margins to felsic interior found in many granitic plutons (see, for example, Bateman & Chappell 1979; Atherton 1981). The field evidence suggests that side-wall crystallization has occurred. Although crystal settling has been invoked, no field features have yet been found that are unambiguously the consequence of settling.

Zoned silicic ignimbrites often show a progression with time from crystal-poor ejecta to crystal-rich, more mafic ejecta during the course of an eruption (Smith 1979; Hildreth 1981). Such evidence could be interpreted as due to settling of crystals, resulting in their concentration in the lower parts of the chamber. However, the comprehensive geochemical study of the Bishop Tuff by Hildreth (1979) has shown that crystal settling cannot account for the geochemical gradients. Hildreth argues that the phenocrysts grew essentially *in situ* and that the compositional gradient existed in the magma *before* the phenocrysts formed. The increasing crystal content with depth in silicic systems is probably related to the suppression of the liquidus temperature and crystallization by roofward concentration of volatiles, and the upward transfer of heat from the higher temperature, more mafic magma, promoting crystallization in the lower levels of the chamber.

In summary, there is reasonably convincing evidence that crystal settling has occurred in nature, particularly in low viscosity lavas. However, there is little evidence to support its operation on a major scale.

(*b*) *Physical constraints*

There is no doubt that in many silicate melts crystals will sink or float in a static liquid dependent only on the sign of the density difference. If they sink and the Reynolds number is sufficiently small the velocity can be calculated from Stokes's law. Evidence from experimental studies (Bowen 1928; Campbell *et al.* 1978; Herzberg *et al.* 1982) and lavas (cited before) show that settling does occur in a static situation. McBirney & Noyes (1979) have suggested that some crystallizing liquids can possess a yield strength such that crystals cannot settle, although this has yet to be verified experimentally.



The main problem with crystal settling, however, lies in the unlikelihood of static conditions in most magma chambers. Most magma chambers will display strong turbulent convective motions with large thermal and compositional Rayleigh numbers. The effect of convective motions is to maintain crystals in suspension, as previously recognized by Bartlett (1969). Consequently, calculations using only the Stokes law to estimate accumulation rates probably have little relevance except in some lava flows or thin sills where static conditions can be expected.

A guide to the likely effects of convection can be assessed from work on the typical fluid velocities attained during thermal convection between two parallel plates (Deardorff & Willis 1967). The time-averaged vertical convective velocities  $w_m$  in the centre of the fluid layer are found to be related to the heat flux and other physical parameters for  $Ra > 10^6$  by

$$w_m = B[(g\alpha\Delta T)^4 \kappa_T^2 d^3 / \nu]^{1/3}, \quad (5)$$

where  $\Delta T$  is the temperature difference across the fluid layer,  $d$  is the layer thickness and  $B$  is a constant with an experimentally determined value of 0.44. Equation (5) is explicitly applicable to a thermally convecting fluid layer. In the double-diffusive situation, where layering is present, the heat flux across the interface is decreased by compositional effects which must be allowed for in evaluating the vertical turbulent velocities. Huppert & Sparks (1980*b*) have made such calculations for a simple two-layer system and the reader is referred to that paper for a more detailed background to the derivation of (5). It is reasonable to apply this relation to double-diffusive layers provided it is remembered that the length scale  $d$  is the thickness of an individual fluid layer and that (5) should be modified for particular cases to take account of compositional effects.

The Stokes free-fall terminal velocity is given by

$$V_t = 2(\Delta\rho)gl^2/9\mu, \quad (6)$$

where  $\Delta\rho$  is the density difference between the crystal and liquid and  $l$  is a typical length scale (radius for spheres) of the crystal. We draw attention to the observation that settling velocities of crystals are strongly dependent on viscosity (equation (6)) whereas convective velocities are very weakly dependent on viscosity (equation (5)). In figure 2, convective velocity is plotted against magma viscosity for a 500 m thick convecting layer. Two lines are shown for values of  $\alpha\Delta T = 10^{-2}$  and  $5 \times 10^{-5}$  respectively to cover a range of plausible conditions. The important feature to notice is that convective velocities are typically of the order centimetres per second for basaltic magmas and only reduce by a small factor in rhyolitic magmas under the same conditions. Also plotted on figure 2 are the variations of settling velocity with magma viscosity for crystals 0.1 and 0.5 cm radius and density excess  $\Delta\rho$  of  $0.5 \text{ g cm}^{-3}$ . In figure 3, convective velocities are plotted against fluid layer thickness for a low viscosity magma ( $10^2 \text{ P}\dagger$ ) and a high viscosity magma ( $10^6 \text{ P}$ ). Settling velocities of crystals are also shown for comparison.

Some important deductions can be made from figures 2 and 3. In the majority of magma chambers convective velocities are orders of magnitude larger than the settling velocities of individual crystals. Thus, unless considerable aggregation occurs, crystals will be kept in suspension. In high viscosity magmas (rhyolites) crystal settling appears almost impossible. Even a 10 cm radius object with  $\Delta\rho = 0.5 \text{ g cm}^{-3}$  has a settling velocity over two orders of magnitude lower than convective velocities in a magma chamber of viscosity  $10^6 \text{ P}$  and height 500 m. Even large mafic xenoliths can be kept in suspension in rhyolitic magma.

$\dagger 1 \text{ P} = 10^{-1} \text{ Pa s}$ .

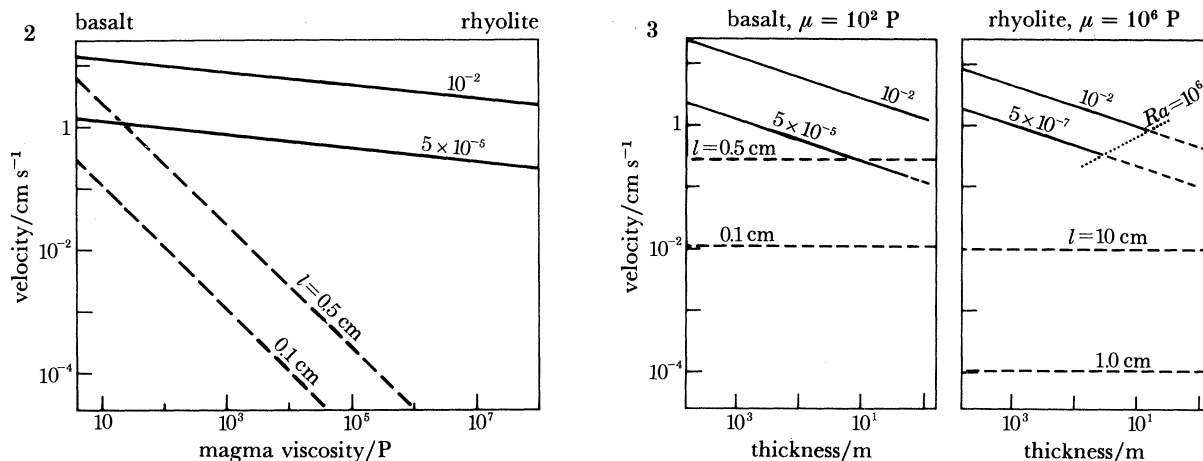


FIGURE 2. Convective velocities of magma in a 500 m thick layer plotted against magma viscosity. The two solid lines are shown for values of  $\alpha\Delta T = 10^{-2}$  and  $5 \times 10^{-5}$ . The settling velocities of spheres with a density excess  $\Delta\rho$  of  $0.5 \text{ g cm}^{-3}$  and radii of 0.5 and 0.1 cm are also shown with dashed lines as a function of magma viscosity. ( $1 \text{ P} = 0.1 \text{ Pa s}$ .)

FIGURE 3. Convective velocities of magma are plotted against layer thickness for magmas of viscosity  $10^2 \text{ P}$  (left side) and  $10^6 \text{ P}$  (right side) for values of  $\alpha\Delta T = 10^{-2}$  and  $5 \times 10^{-5}$ . On the right side a dotted line shows conditions when  $Ra_t = 10^6$ . The settling velocities of spheres with a density excess  $\Delta\rho$  of  $0.5 \text{ g cm}^{-3}$  and radii 10.0, 1.0, 0.5 and 0.1 cm are also shown by dashed lines.

Inspection of the figures suggests that crystal settling is only likely to become a dominant factor in thin fluid layers of low viscosity magma. Thus one would expect to find that crystal settling becomes important in lava flows, in thin double-diffusive layers or density currents within magma chambers of ultramafic, and less commonly, mafic magma (Irvine 1980*b*).

The convective velocities will decrease towards the boundaries of the parallel plates where, in thin boundary layers, velocities become a small fraction of the central velocities; thus crystals that enter the boundary layers might settle out. However, their rate of accumulation will not be governed by the Stokes law, but will be determined by the detailed pattern of convection and the exact structure of the boundary layer. Many convective régimes may be sufficiently vigorous that no deposition can occur. No studies have yet been made on sedimentation from convectively driven fluid layers. We have noticed, however, in several laboratory experiments that small particles incorporated accidentally into convecting fluid layers are circulated around and are never observed to fall out. As in sediment-laden turbulent flows (rivers and currents), calculations of static settling velocities will prove of little value in understanding how and when sedimentation can occur. There is also a clear need for experimental and theoretical investigations of sedimentation from convecting fluids.

A final comment of caution is required with regard to the use of (5). In constructing figures 2 and 3 we have not taken compositional effects into account. When such effects are considered, the convective velocities are reduced somewhat. For example, in the calculations of Huppert & Sparks (1980*a, b*), where compositional effects are included, the velocities are reduced by about a factor of two. Since it is apparent that the settling velocities are often orders of magnitude less than the convective velocities, such modifications make no difference to our general conclusions. However, in particular cases, especially in circumstances where the settling velocities are comparable with convective velocities, we recommend carrying out an exact assessment of compositional effects rather than using (5).

In summary, the majority of magmas convect and consequently crystal settling will be severely inhibited. The most likely situations in which settling can become important is where crystals are swept into the thin boundary layers close to the margins of the chamber and in thin fluid layers of low viscosity magma.

(c) *Kinetic constraints*

Another significant crystallization process is suggested by laboratory experiments on crystallization. In the case of aqueous solutions, it is difficult to nucleate crystals homogeneously within a cooling, supersaturated liquid. In most cases crystals nucleate and grow at solid boundaries where the maximum undercoolings are attained. Campbell (1978) and Berkebile & Dowty (1982) have shown that the same problem arises in experiments on silicate melts and they contend that crystals always nucleate on solid boundaries (capsule walls or pre-existing crystals). Thus the margins of a chamber are a much more probable site for nucleation in a magma chamber than internally within homogeneous magma.

It is possible, however, for crystals to nucleate internally away from solid boundaries, as we have observed in a number of laboratory situations. The two main cases are crystal nucleation at double-diffusive interfaces and during mixing of fluids. Nucleation can occur because the crystallizing compound is undersaturated in the two separated fluids, but becomes supersaturated in a mixture of the fluids. In one experiment, for example, a layer of hot  $\text{KNO}_3$  solution was emplaced beneath a layer of cooler  $\text{K}_2\text{CO}_3$  solution. At the double-diffusive interface  $\text{KNO}_3$  became highly supersaturated because of the decrease in its solubility in the presence of  $\text{K}_2\text{CO}_3$ . Crystals were continually generated at the interface and rained down into the lower layer. Another cause of nucleation can be large under-coolings caused by steep thermal gradients across an interface. In some experiments crystals were formed in the cold thermal boundary layer at the top of hot  $\text{KNO}_3$  solution emplaced beneath cold  $\text{NaNO}_3$  solution. Rapid mixing of fluids can also produce supersaturation effects and internal nucleation.

These observations call into question some of the conventional views on the nature of phenocrysts, which are generally viewed as crystals which have nucleated internally within homogeneous magma. From a physico-chemical standpoint such a mechanism is not the only way of forming crystals in magma. Rather phenocrysts could originate in a variety of other circumstances:

- (i) by erosion of crystals nucleated and grown on the margins of a chamber;
- (ii) by mixing between magmas which results in a supersaturated melt;
- (iii) by nucleation at double-diffusive interfaces where the mixed interfacial region is supersaturated or strongly undercooled;
- (iv) by transport of residue (restite) crystals from the source of partial melting (White & Chappell 1977);
- (v) by further growth of crystals formed by mechanisms (i)–(iv).

The complex and variable zoning patterns often observed in phenocrysts from single lavas may represent crystals formed by several mechanisms in the chamber. Such complex patterns are often not observed in cognate cumulate nodules where, for example, feldspars can often be unzoned or display much simpler zoning. The phenocrysts suspended by convection in magma may be formed under conditions different from those for crystals in associated cumulate rocks.

If phenocrysts settled out of, for example, basaltic andesite magmas one would commonly expect to find nodules containing feldspars with very complex zoning patterns. This is not observed, as can be ascertained from associated cumulate nodules in calc-alkaline volcanic rocks (see, for example, Nicholls 1971; Arculus & Wills 1980). Discrepancies between phenocryst and cumulate assemblages have also been noted along the mid-ocean ridges (Walker & Delong 1982). Settling of phenocrysts is perhaps not the dominant process in differentiation or in the formation of cumulates.

## 5. CONVECTIVE FRACTIONATION

### (a) *Introduction*

Having considered the problems associated with crystal settling as an all embracing model for fractionation, we now examine the case that can be made for the alternative mechanism: separation of liquid from solid by convection. The basic driving force for convection comes from the selective removal of heavy or light components into the crystal, locally resulting in a lighter or denser fluid. We propose that any fractionation process involving convection of fluid away from crystals should be called *convective fractionation*, a term used by Rice (1981).

Many of the basic features of convection in crystallizing systems have only been investigated in the last few years. Some of the convective phenomena are sufficiently novel that an important new field of fluid mechanics is emerging relevant not just to geological situations but also to many engineering, metallurgical and industrial processes (Huppert & Turner 1981*a*; Hurlle & Jakeman 1981).

Much recent progress has been made by laboratory experiments on crystallization in aqueous solutions of simple compounds (for example  $\text{KNO}_3$ ,  $\text{Na}_2\text{CO}_3$ ,  $\text{CuSO}_4$ ). These experiments have been designed to look at the effects of different container and cooling geometries, different physical properties and different compositional and thermal gradients on convective phenomena in crystallizing systems. Much of the work has been qualitative, except for the simple case of a two-layer system (Huppert & Sparks 1980*a, b*; Huppert & Turner 1981*b*).

### (b) *Experimental evidence*

The following section summarizes the variety of convective phenomena due to crystallization observed in saturated aqueous solutions of compounds such as  $\text{Na}_2\text{CO}_3$ ,  $\text{KNO}_3$ ,  $\text{CuSO}_4$  and other salts. The reader is referred to the following studies for more detailed descriptions (Turner & Gustafson 1978; Turner 1980; Chen & Turner 1980; McBirney 1980; Huppert & Turner 1981*b*; Huppert *et al.* 1982*b*; Kerr & Turner 1982; Turner *et al.* 1983). These experiments were designed to investigate two basic kinds of systems: closed systems, where compositional and thermal gradients are set up due to crystallization; and open systems, where new fluid of different composition and temperature is introduced into the container.

#### (i) *Closed systems*

In the simplest closed system a saturated solution is cooled from different boundaries which causes crystals to nucleate and grow. A fundamental feature of all these systems is that as crystals grow they locally deplete adjacent fluid in its denser components. The resulting fluid is lower in density than neighbouring fluid and so rises to cause convection. The convecting fluid can then effectively retain its composition due to the low diffusivity of chemical components.



The effects of release of light fluid vary considerably, depending on the geometry of the container and where cooling takes place. Figure 4, plate 1, shows several examples of experiments to illustrate some of the principles which have been identified. When cooling occurs from the roof (figure 4*a*), the light residual fluid rises to form a less dense stagnant zone beneath the roof in between the growing crystals. When cooling and crystallization occur along a vertical boundary, such as a wall or central pipe, light fluid is released and forms a stably stratified layer of light fluid at the top of the tank (figures 4*b*, *c*). As this layer forms, the destabilizing temperature gradient within it causes double-diffusive layering to develop (figure 4*c*). In this type of experiment (Chen & Turner 1980; McBirney 1980; Turner & Gustafson 1981) the initial

TABLE 1

(The table lists the total amount of crystals formed in the container as a mass percentage of the whole container system at different times. The parameter  $X$  represents the mass percentage of crystals required to be removed to generate the fluid in the stratified region from the initial fluid composition.)

time/h	depth of stratified fluid region/cm	total crystals (% by mass)	$X$ (% by mass)
7	2	1.0	11.5
12	4.5	1.7	9.0
23	9.0	2.5	8.5
72	23.0	12.0	14.1

solution, which is homogeneous with respect to both temperature and composition, becomes compositionally and thermally zoned. When crystallization takes place on the floor (figure 4*d*), thin plumes of fluid rise into the overlying fluid and drive compositional convection, which maintains a well-mixed solution. Our unpublished experiments have shown how crystallization on a tilted floor leads to stratification because the light fluid released from different levels along an inclined floor has to rise through different thicknesses of overlying fluid. Consequently the buoyant fluid mixes with different amounts of overlying fluid to produce horizontal and vertical compositional gradients.

Similar experiments have been conducted in a porous medium in which a layer of glass balls at the bottom of a tank provide a porous bed. These experiments (Tait *et al.* 1984) have been designed to investigate compositional convection in partially molten rocks and cumulus piles. In these experiments crystallization has been induced in a saturated solution of  $\text{KNO}_3$  by cooling from both above and below the layer of balls. Convective motions are observed as crystallization occurs between the balls. In the case of a porous bed cooled from below crystallization liberates light fluid from the pore space which rises and is continually replaced by warmer solute-rich fluid from above. These experiments illustrate the wide range of circumstances in which convective fractionation can occur.

A significant result of the laboratory experiments is that highly fractionated fluids can be produced by these processes without having to invoke large amounts of crystallization. This can be illustrated by measurements of composition made on layers formed during cooling of  $\text{Na}_2\text{CO}_3$  solution from a central pipe in an experiment reported by Turner & Gustafson (1981). Figures 4*b* and *c* show two photographs from these experiments with a layer of light fluid after 7 h (figure 4*b*) and a fully stratified and layered tank after 3 days (figure 4*c*). Table 1 lists the



depth of the interface at the bottom of the stratified fluid region and the total mass of crystals formed at different times in the experiment. At each stage the average composition of the stratified region was determined and the mass percentage of crystals  $X$  necessary to form this composition from the initial solution was calculated. These values are listed in table 1. After 7 h of the experiment, fluid had accumulated at the top of the container which required the removal of 11.5 %  $\text{Na}_2\text{CO}_3$  crystals from the affected fluid, yet only 1 % of crystals had formed in the container considered as a whole. After 3 days of the experiment, the composition of the fluid right at the top of the stratified tank indicated removal of 20 % crystals.

Many other experiments have confirmed that segregation of strongly solute-depleted fluid to the top of the container is a characteristic feature during sidewall crystallization. This occurs because only a small (but increasing) proportion of the total fluid in the box is involved in fractionation. Crystallization at the side wall reduces the concentration of solute *locally* in the surrounding fluid which convects upwards as a boundary layer. The bulk of the fluid in the interior of the container is unaffected by the crystallization. In the geological context it is possible to generate highly differentiated melts at an early stage in the evolution of a magma chamber without requiring large amounts of crystallization.

(ii) *Open systems*

The view that many magmatic systems are open and continually or periodically receive influxes of new primitive fluid from depth has been widely accepted by most petrologists. This idea has prompted a number of experimental and theoretical studies on the dynamics of replenishment and the influence of crystallization on the interactions between fluids of different composition and density.

Huppert & Turner (1981*b*) studied the case of a hot dense layer of  $\text{KNO}_3$  solution emplaced beneath a cold light layer of  $\text{NaNO}_3$  solution. The study was motivated by a theoretical analysis of the emplacement of a hot layer of picritic magma beneath a cold, lower density layer of basaltic magma in a chamber (Huppert & Sparks 1980*a, b*). In the experiment, the two layers of fluid exchanged heat, which resulted in crystallization of  $\text{KNO}_3$  in the lower layer and vigorous convection, driven partly by thermal effects and partly by release of light residual fluid from crystals growing on the floor (figure 5*a*, plate 2). Eventually crystallization reduced the density of residual  $\text{KNO}_3$  solution to the same value as the overlying  $\text{NaNO}_3$  solution and a rapid overturn took place (figure 5*b*). The experiments confirmed the theoretical predictions of temperature changes with time in the two layers.

Another series of experiments (Huppert *et al.* 1982*b*) investigated the effects of replenishment on a compositionally stratified fluid layer. The new fluid formed a separate layer (figure 5*c*) which heated the overlying stratified fluid to form a series of double-diffusive layers. Crystallization of the lower layer resulted in eventual mixing of the residual fluid with the lower parts of the stratified layer.

These experiments illustrate two principles of convection relevant to magma chambers. First, replenishment of new fluid which is denser than the resident magma can lead to a period of independent crystallization in which the new fluid evolves separately. Crystallization leads to changes in density of residual fluids which can eventually allow mixing. In the two-layer experiment (figure 5*a, b*) no crystals would form if the two fluids were immediately mixed together.

In a series of experiments (Huppert *et al.* 1983) the effects of changes in the viscosity of the two

fluid layers has also been studied. The steep viscosity gradient across an interface between glycerine and  $\text{KNO}_3$ , for example, significantly modifies the convective behaviour. Crystallization at the interface releases light residual fluid which is immediately swept up into the glycerine (figure 5*d*). No abrupt overturn occurs, but residual fluid is continually transferred across the interface.

(c) *Theoretical considerations*

In a quiescent fluid, rejection or incorporation of solute results in an exponential concentration profile in a thin film adjacent to the growing crystal (Coriell & Sekerka 1981). This film has a characteristic thickness,  $\delta$ , of  $\kappa_s/V$ , where  $V$  is the growth velocity of the crystal face and  $\kappa_s$  is the chemical diffusion coefficient. An approximate approach to determining the stability of a horizontal film above a growing crystal face is to define a Rayleigh number for the film

$$Ra_{\text{film}} = g\Delta\rho\kappa_s^2/V^3\mu. \quad (7)$$

For instability to occur, this Rayleigh number must exceed *ca.*  $10^3$ . For laboratory experiments, crystal growth rates are in the range  $10^{-3}$  to  $10^{-5}$   $\text{cm s}^{-1}$ , viscosities are of order  $10^{-2}$  P and diffusivities are of order  $10^{-5}$   $\text{cm}^2 \text{s}^{-1}$ . Thus Rayleigh numbers are substantially greater than  $10^3$  and horizontal crystal faces should become unstable and produce boundary layers much thinner than  $\kappa_s/V$ . Observations of plumes in the experiments with widths less than 1 mm are thus consistent with theory.

In magma chambers both crystal growth velocities (typically  $0.1$ – $10$   $\text{cm a}^{-1}$ ) and diffusivities are much smaller. However, even in magmas of rhyolitic compositions, values of Rayleigh

#### DESCRIPTION OF PLATES 1 AND 2

FIGURE 4. (a) Shadowgraph showing the results of cooling a constant concentration gradient of  $\text{Na}_2\text{CO}_3$  solution from above. The crystals grown on the roof caused a pool of light residual fluid to accumulate beneath the roof. The double-diffusive layering in the container is formed by cooling of the concentration gradient from above. Details of the experiment are given by Chen and Turner (1980).

(b), (c) Shadowgraph showing the results of cooling along a vertical central pipe immersed in a container of homogeneous  $\text{Na}_2\text{CO}_3$ . After 7 h (b) crystals growing along the pipe have generated boundary layer flows of light fluid which has formed a pool of residual fluid at the top of the container. The dye streak shows the presence of weak stratification in the main part of the container. After 3 days (c) sidewall crystallization has produced compositional stratification throughout the container. Further details are given in Turner & Gustafson (1981).

(d) Shadowgraph showing the results of crystallization from the floor of  $\text{Na}_2\text{CO}_3$  solution. After 4 h 55 min thin plumes of light residual fluid are seen rising from the floor. Further details of the experiment are given by Chen & Turner (1980).

FIGURE 5. (a) Shadowgraph showing a layer of hot  $\text{KNO}_3$  solution beneath a layer of cold  $\text{NaNO}_3$  solution. Thermal convection is visible in the upper and lower layers as they exchange heat across the double-diffusive interface. Crystals of  $\text{KNO}_3$  are growing on the floor of the container.

(b) Shadowgraph showing the same experiment as in (a) in which the lower layer of  $\text{KNO}_3$  solution has overturned and is mixing with the upper  $\text{NaNO}_3$  solution. Further details are given in Huppert & Turner (1981*b*).

(c) Shadowgraph showing the overturn of a layer of  $\text{KNO}_3$  solution into a gradient region of  $\text{K}_2\text{CO}_3$  solution. Note that the mixing is confined to the lower part of the stratified solution. The double-diffusive layering is formed by heating of a concentration gradient from below and by side-wall heating. Further details are given in Huppert *et al.* (1982*b*).

(d) Photograph of an experiment in which  $\text{KNO}_3$  solution at  $60^\circ\text{C}$  has been emplaced beneath glycerine at  $11^\circ$ . Blobs and plumes of residual  $\text{KNO}_3$  solution are generated at the interface and rise continuously into the glycerine. Crystals of  $\text{KNO}_3$  form in the plumes as they cool and the crystals, often in the form of a long chain, fall back. Residual  $\text{KNO}_3$  solution forms a layer above the glycerine. Further details are given in Huppert *et al.* (1983).



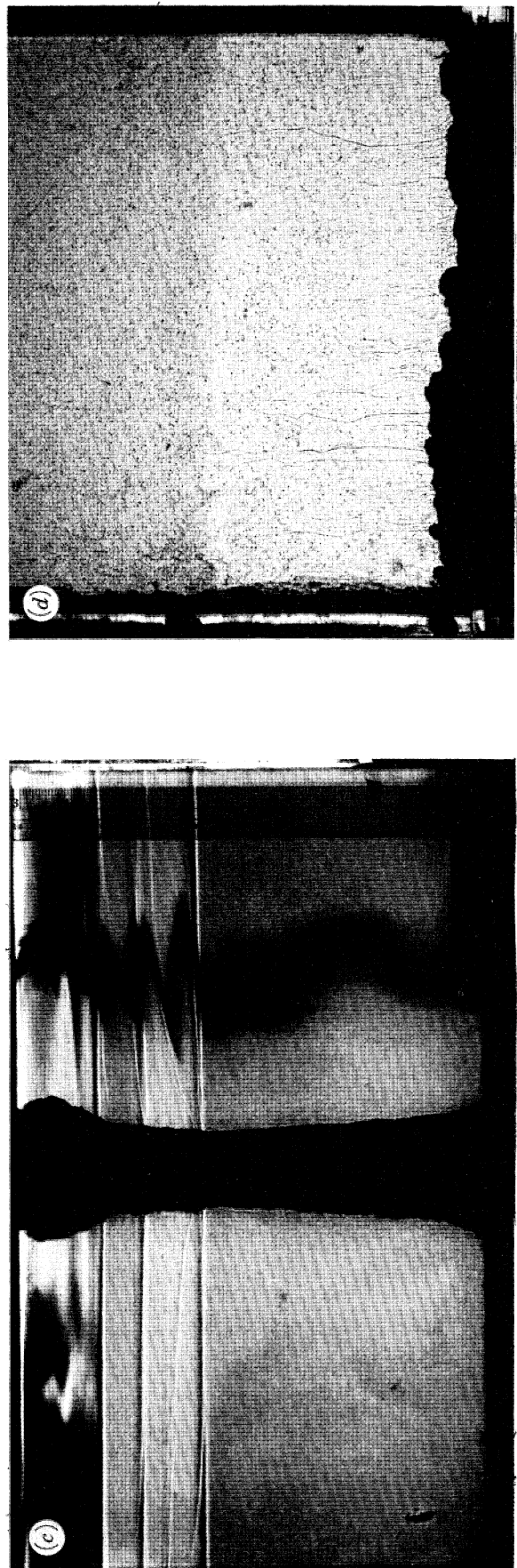
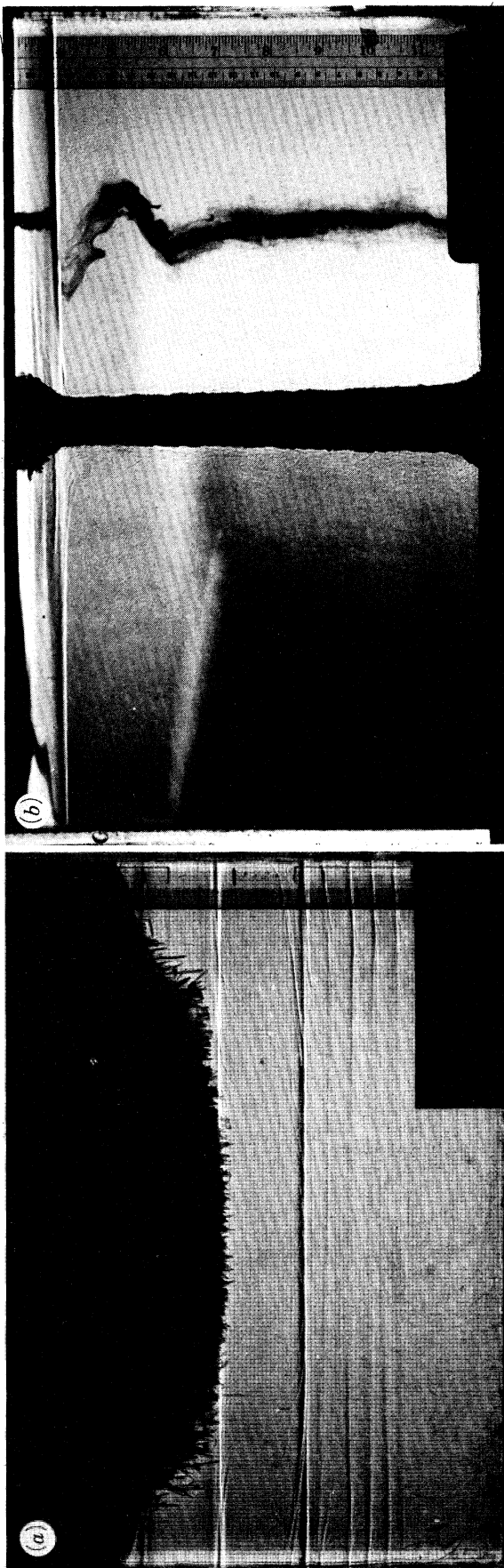


FIGURE 4. For description see opposite.



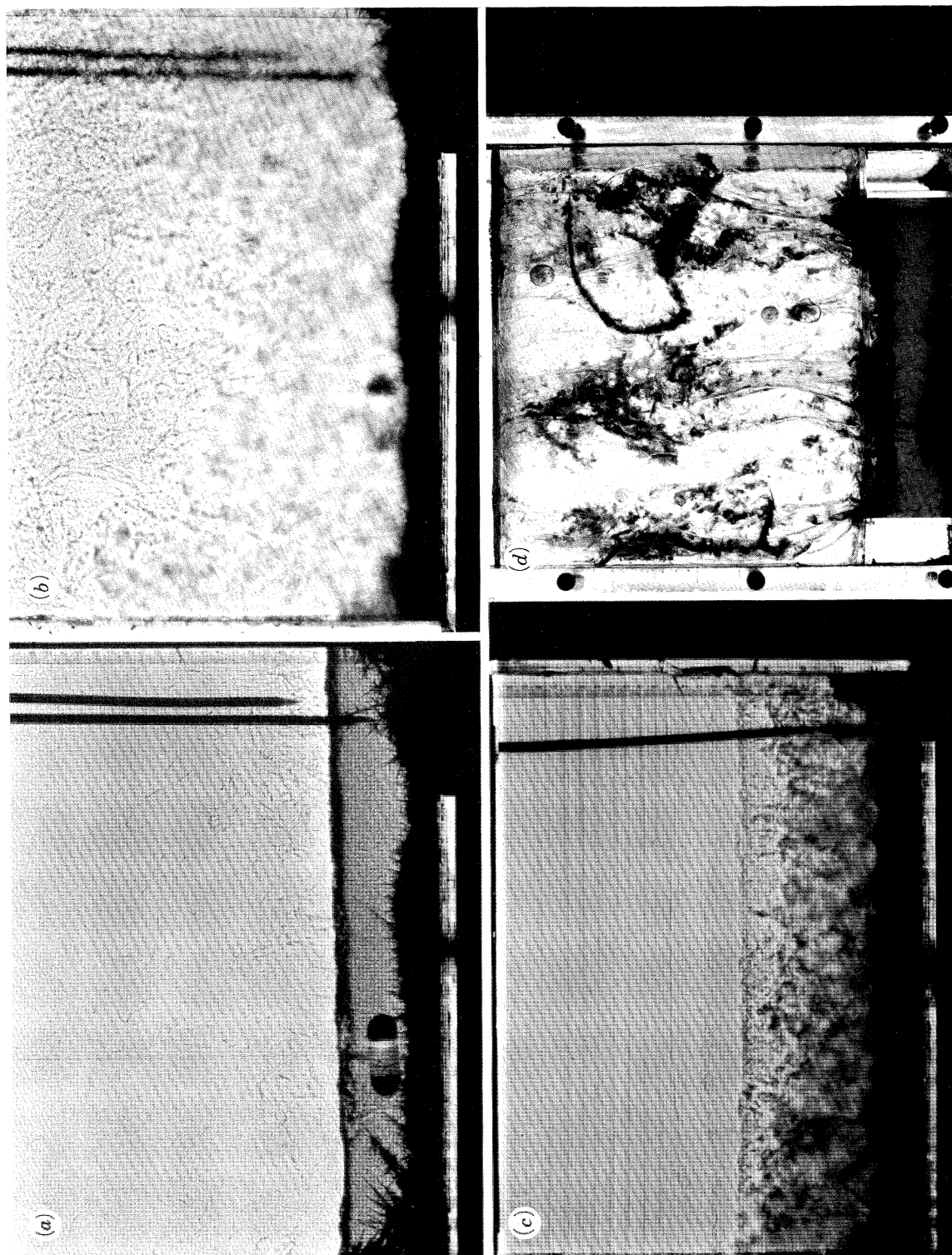


FIGURE 5. For description see p. 524.

number, as defined in (7), are generally much greater than  $10^3$  suggesting that compositional convection should occur (Huppert & Sparks 1984).

(d) *Conclusions*

In all the laboratory experiments, residual light fluid is convectively removed from growing crystals. The consequences of this convection vary greatly according to the physical and chemical properties of the fluids and the geometry of the container. Many of the effects observed in the laboratory experiments are strikingly different to what would have happened had the crystals nucleated within the fluid and fallen to the floor without any convection. Dimensional and theoretical arguments have shown that such convective effects should also happen in magmatic systems. Convective fractionation must therefore be viewed as a process of considerable importance in the genesis of different magmas.

Before proceeding to describe the geological implications of convective fractionation, some discussion is required of the geological applicability of investigations carried out in small laboratory tanks. Some scepticism has been expressed regarding the relevance to magma chamber dynamics of results obtained from experimental laboratory tanks because of the great differences in the scale of the reservoirs and in some of the physical parameters describing the fluids. It has been stated by Rice (1982), for example, that conclusions reached from the experiments are incorrect because of the lack of dynamic similitude. The theoretical analysis of convection caused by growth of a crystal face, however, indicates that the experimental studies are comparable, in dynamical terms, with geological situations. Further, such criticism fails to take into account the major conceptual justification of the experimental method employed.

In some of the work, quantitative relations for the dependent variables, such as temperature and concentration, have been obtained in terms of the physical parameters of the system using fundamental physical principles. These relations and the physical phenomena, such as fluid mixing, that accompany them, can then be tested by a series of experiments at known, but different, values of the parameters. Because explicit quantitative formulae have been obtained, it is not necessary to scale each variable in a definite proportion. Such a situation occurs in the simple two-layer replenishment system, investigated theoretically by Huppert & Sparks (1980*a, b*) and experimentally by Huppert & Turner (1981*b*).

Experiments have also been performed investigating phenomena for which an explicit quantitative relation governing the variables has not yet been obtained. Such experiments cannot yield *quantitative* results that can be taken over to magma chambers. However, the art of the experimental investigation is to replicate in the laboratory the dominant physical and chemical effects governing the evolution in the magma chamber. The experimental results will then qualitatively describe the effects that will occur in magma chambers. Further, quantitative analysis of the experimental results may suggest how the development of the related theoretical analysis should proceed.

Thus, for example, a cooling and crystallizing  $\text{KNO}_3$  solution releases light liquid deficient in  $\text{KNO}_3$  as it crystallizes. Being less dense than its surroundings, the released liquid rises. If there is a vertical density gradient in the solution, the released liquid will be constrained in its rise and a horizontal layer will result. This will occur in all multi-component crystallizing systems for which the released liquid is less dense, independent of whether the original fluid is an aqueous chemical solution, a magma or a molten metal. It has been said that investigation with relatively low viscosity aqueous solutions cannot model effects in much more



viscous magmas. However, consideration of the value of viscosity by itself is not helpful. It is not the coefficient of viscosity that governs the form of motion but the appropriate Rayleigh number. For example, in the case when buoyancy is introduced uniformly from below, turbulent motions occur for Rayleigh numbers greater than a value of *ca.*  $10^6$ . It is thus essential, and sufficient, that if the motion in a magma chamber whose Rayleigh number exceeds  $10^6$  is being investigated, then the Rayleigh numbers of the laboratory experiments do also. Likewise our earlier theoretical section demonstrated that both the laboratory fluids and magmas will generally be convectively unstable during the growth of crystals because the relevant compositional Rayleigh number greatly exceeds the critical value.

## 6. SOME GEOLOGICAL IMPLICATIONS

Convective fractionation and other dynamic effects arising from the density variations identified in previous sections have some fundamental implications for the origins of compositional and textural diversity in igneous rocks. These concepts are likely to lead to new insights and some changes in perspective on the processes of magma genesis. We have selected a number of diverse topics of current interest where these dynamic processes might play an important role.

### (a) *Compositional zoning in silicic magma chambers*

A large amount of evidence has accumulated from volcanic rocks in recent years to support the idea that many silicic magma chambers are both compositionally and thermally zoned (Smith 1979; Hildreth 1981). In the main, the evidence points to low density and often volatile-rich silicic magma overlying higher temperature more mafic magma. The gradations in magma composition vary from continuous to cases where there are sharp discontinuities in composition (Hildreth 1981). The general occurrence of stable compositional gradients and unstable temperature gradients implies that double-diffusive effects will be important. The discontinuities in composition represent major boundaries between double-diffusive layers.

The origin of these compositional gradients has attracted considerable recent attention, particularly with regard to the dramatic trace element gradients observed in high silica rhyolitic rocks (Hildreth 1979, 1981). Some of the compositional gradients can be explained with little difficulty by fractional crystallization and McBirney (1980) has attributed compositional zoning in such cases to side-wall crystallization as discussed earlier. In view of the great difficulties associated with invoking crystal settling, such a mechanism must now count as an attractive way of generating compositional zoning by fractionation. The mechanism is given added support by observations from zoned granitic plutons in which cumulate-like rocks (grading from mafic to felsic inwards) are found accreted to the margins of many plutons. Fractional crystallization has been invoked in such cases (see, for example, Karner 1968; Bateman & Chappell 1979; McCarthy & Groves 1979; Atherton 1981) with side-wall crystallization and crystal settling being regarded as the causative mechanisms. There is no field evidence to support crystal settling, but clear evidence for side-wall crystallization.

The greatest difficulty in advocating a fractional crystallization mechanism comes from considering the origin of striking trace element gradients in high-silica rhyolite magmas. Most trace elements in such systems show large variations from the top to the bottom of the magma chamber, whereas major elements show little change. The kinds of variations observed have been discussed at length by Hildreth (1979, 1981). He and others (see, for example, Shaw *et al.*

1976) have argued that such extreme gradients cannot be caused by any conventional differentiation mechanism such as crystal fractionation, partial melting, assimilation and immiscibility. They therefore involve a rather complex model (termed convection-driven thermogravitational diffusion) to explain the gradients. This model essentially involves boundary layer flows of H<sub>2</sub>O-rich magma along the side and then liquid-state Soret diffusion across a compositionally and thermally zoned region of high silica rhyolite at the top of the chamber. The gradients are generated by a 'diffusive' mechanism and perhaps volatile-fluxing.

We accept that Hildreth (1979, 1981) has made an excellent case to eliminate all conventional mechanisms *except* fractional crystallization. The detailed discussion of the gradients in the Bishop Tuff (Hildreth 1979) provides a well argued and completely convincing case against crystal settling. However, these arguments do not eliminate crystal fractionation, because there is the alternative mechanism of convective fractionation.

On close inspection the principal difficulty with fractionation as a means to produce the observed gradients is simply the large amounts of fractional crystallization required. For example, to generate the extreme depletion in Sr, Ba and Eu observed in the Bishop Tuff involves well in excess of 50 % of feldspar (Hildreth 1979). In a model of fractionation as a cause of the gradients in such systems, Michael (1983) has proposed that 65–70 % fractionation of the observed mineral phases can generate many of the variations observed.

Side-wall crystallization provides a straightforward mechanism to account for the Bishop Tuff gradients. In the laboratory, side-wall crystallization influences only a small fraction of the fluid in a container and consequently highly fractionated fluids can be generated at an early stage with little crystallization (as measured by the total mass fraction of crystals formed in the whole container). We do not yet know enough about the fluid dynamics of the chemical boundary layers to quantify the process in a magma chamber and demonstrate conclusively that it can produce large volumes of very differentiated fluid in a high silica rhyolite system. Nevertheless, it is perhaps premature to abandon fractional crystallization as a major process causing these gradients. The role of liquid-state Soret diffusion is equivocal. There is little known about Soret coefficients in geological systems except for the study by Walker & Delong (1982) and there is little compelling evidence to support its operation over large length scales. On the other hand, there are some factors in silicic systems that favour convective fractionation by side-wall crystallization. Crystallization increases the H<sub>2</sub>O content of silicic magma locally. The increased concentration of H<sub>2</sub>O in the compositional boundary layer lowers density and viscosity substantially (Shaw 1963) and also lowers the diffusivity of other components (Watson 1981). All these factors will promote the rates of production of buoyant magma in the boundary layer.

If side-wall crystallization is the correct mechanism for generating steep trace element gradients in high-silica rhyolite, then an additional degree of freedom enters into the modelling of crystal fractionation. It is the crystals accreted to the margins, not the phenocrysts in the erupted magma, which have controlled the chemical evolution. These crystals may differ somewhat in properties and compositions from phenocrysts. There are two great attractions for side-wall crystallization in explaining some old petrological enigmas. It can produce highly evolved magmas rapidly and does not require large amounts of crystallization from phenocrysts.

Side-wall crystallization is not the only plausible mechanism to generate compositional and thermal gradients. Suppose, for example, that progressive fractional melting of a crustal rock generated a series of silicic magmas which were successively fed into a chamber. The earliest melts would have the lowest temperature, and highest silica content and probably the lowest

density. Later melts would become hotter, more mafic and denser. A stable compositional and unstable temperature gradient could be set up. Repeated replenishment of a chamber by mantle-derived basalt could also generate zoned systems with complex interactions between fractionated products of the basalt and silicic magmas generated by crustal melting. Contamination must commonly lead to compositional gradients.

(b) *Layered intrusions*

The origin of layering in ultramafic and mafic intrusions has long attracted attention. There are many different varieties and scales of layering (Wager & Brown 1967; Irvine 1980a) to be found in nature. Thus there are undoubtedly several different mechanisms causing it. The application of the new fluid dynamical ideas to layered intrusions, as well as ideas based on the kinetics of *in situ* crystallization, have been given considerable attention in the recent petrological literature. Here we present a brief synopsis of these ideas and a few additional thoughts based on laboratory experiments.

(i) *Layering due to replenishment*

The large-scale (metres to tens of metres) cyclic layering found in the ultramafic zones and some parts of the mafic zones of layered intrusions has been widely attributed to replenishment by new primitive magma (Brown 1956; Irvine 1980a). A new aspect of the origin of this type of layering relates to the observation that high temperature primitive magma from the mantle will often be denser than differentiated basaltic magma. During replenishment such magma will form a separate layer at the base of the chamber and will cool and crystallize by exchanging heat across a double-diffusive interface until the residual liquid has the same density as the overlying magma. The theoretical and experimental analysis of this process and the geological evidence to support it has been discussed in detail elsewhere (Huppert & Sparks 1980a, b; Huppert & Turner 1981 b; Huppert *et al.* 1982 b; Sparks & Huppert 1984).

(ii) *Layering due to in situ crystallization*

A number of studies (Campbell 1978; McBirney & Noyes 1979; Morse 1979) have drawn attention to the difficulties in explaining many features of cumulate rocks by crystal settling. McBirney & Noyes (1979) propose that much of the crystallization takes place in boundary layers along the margin of a chamber and that small-scale (millimetre–centimetre) rhythmic layering can be formed by periodic oscillations in the nucleation and growth of different minerals. Morse (1979) draws attention to the possibility that rhythmic layering can be formed by oscillation around a cotectic boundary. One phase becomes supersaturated and grows making the surrounding fluid supersaturated in another phase, which then nucleates and grows.

The dynamically interesting point about *in situ* crystallization processes is that locally the fluid will be changed in density and consequently may be able to convect away from its point of origin. The effects of this convection, as already discussed, will depend very much on the chamber geometry and the nature of the density changes. In many cases *in situ* crystallization should lead to compositional and thermal gradients being set up in the chamber. The various possibilities of fluid layering due to *in situ* crystallization in mafic magma chambers are discussed by Sparks & Huppert (1984).

(iii) *Layering due to compositional gradients and double-diffusive fluid layers*

We have already inferred that compositional and thermal gradients should commonly be set up in magma chambers by replenishment or convective fractionation, and double-diffusive layers can form in these gradients. Several investigators have proposed that double-diffusive layers could be frozen in as rock layers (Turner & Gustafson 1979; McBirney & Noyes 1979). Recent laboratory investigations (Chen & Turner 1980; Kerr & Turner 1982) have shown how fluid layering can cause and influence layering in crystal layers, although it is important to note that the crystal layers differ in thickness and composition from the original fluid layers.

An interesting feature of some layered intrusions is that there can be a marked angular discordance between rhythmic layering and phase layering (Wager & Brown 1967; Casey & Karsen 1981; Wilson & Larsen 1982). In these cases the rhythmic layering is generally regarded or observed to be approximately parallel to the margins of the intrusions. Such discordance can come about in a system which develops compositional gradients and undergoes *in situ* crystallization along the margins. If convective fractionation or occasional replenishment of the starting composition leads to compositional gradients being set up, the horizontal layering in the gravitationally stratified fluid can be frozen in as discordant phase layering in the rock. These effects have been observed in preliminary experiments in which we have cooled compositionally zoned solutions along tilted boundaries.

(iv) *Layering due to sedimentation*

Our earlier critique of crystal settling should not be construed as implying that settling and sedimentation processes have no role at all in the development of structures in layered intrusions. There are many structures which are still best attributed to sedimentation phenomena, as was recognized by many other workers. Current-bedding, some varieties of graded layers, and slump structures, for example, are examples of structures attributable to sedimentation. The most compelling circumstances where sedimentation could become important are when slumping occurs to form magmatic density currents (Irvine 1980*b*) and where chamber replenishment occurs. In the case of the emplacement of a thin layer of dense, hot, low viscosity magma (Huppert & Sparks 1980*b*), settling velocities can be comparable with turbulent velocities allowing sedimentation. In addition, replenishment could lead to increasing vigour of convection allowing crystals to be eroded and stirred up from the floor and margins of a chamber.

(v) *Post-cumulus crystallization*

Tait *et al.* (1984) have shown that when crystallization takes place in a porous medium convective motions are still observed. This leads to the suggestion that convection plays an important role in post-cumulus processes in magma chambers. Wager *et al.* (1960) identify two end-member styles of post-cumulus growth: orthocumulates and adcumulates. They attribute the origin of these end-members to the interplay between diffusional exchange and crystal accumulation rates. When settling rates are high, diffusional exchange is inhibited and consequently orthocumulates formed. When settling rates are low, diffusional exchange can take place allowing adcumulus growth.

Convective fractionation provides an alternative mechanism which is more compelling from a physico-chemical viewpoint. The essential mechanism can be illustrated by considering



crystallization on the floor of an intrusion. Crystals (primocrysts) form and enclose pore space containing intercumulus liquid. If further crystallization in the pore space generates light residual liquids, continuous convective exchange occurs with overlying dense magma from the chamber, so keeping the composition of the pore fluid nearly constant and allowing adcumulus growth to occur. If the residual fluids are denser (as for example is the case with many assemblages involving plagioclase) they do not convect, but form a static, stable intercumulus melt. As crystallization proceeds in the crystal pile the residual fluid becomes denser. In these circumstances orthocumulate rocks might form. Orthocumulates may also form when low permeability or high viscosity inhibit convection of the pore fluid. The essential differences between this model and the conventional model is that exchange between the pore fluid and magma chamber occurs by convection rather than static diffusion and that the critical factor is not the crystal accumulation rate, but whether residual liquids become denser or lighter as crystallization takes place in the pore space.

## 8. CONCLUSIONS

Magmatic processes that involve changes of melt composition include fractional crystallization, partial melting and contamination. Such processes cause changes in melt density that are generally larger than density changes attributable to associated thermal effects. Experimental and theoretical experience with convection in crystallizing systems strongly suggests that these compositional effects on density will be fundamental in controlling the dynamic evolution of magmas.

The investigation of convection in multicomponent and in crystallizing systems is a comparatively new field. Theoretical and experimental studies can give new perspectives on some fundamental petrological problems such as the compositions of basaltic lavas and the origin of layering in ultramafic intrusions (Huppert & Sparks 1980*a, b*; Huppert & Turner 1981*b*). In this paper and that of Sparks & Huppert (1984), we have applied the new ideas to a broader range of problems, including the origin of zoning in plutonic and volcanic silicic rocks, the formation of layered intrusions and the mechanism of post-cumulus crystallization. We have emphasized, in particular, that there is an alternative to crystal settling as a mechanism for crystal fractionation, which is separation of liquid from solid by convection. We have argued that convective fractionation is in fact an important mechanism and that many geological features of igneous rocks are best explained by its occurrence.

Although this paper and previous studies have concentrated on fractional crystallization, the same kinds of principles will be relevant to other major processes of magma evolution, notably partial melting and contamination. Our present understanding of magma chamber evolution due to fractional crystallization and replenishment indicate that the kinds of magma that can be erupted and the kinds of plutonic rock that can form are strongly dependent on physical properties and dynamic effects. Little has yet been done on the dynamics of partial melting or contamination, but it is certain that in these processes, too, physical constraints will be as important. Magmas generated by melting, crystallization or contamination will owe their characteristics as much to physical and dynamic effects as to chemical conditions.

This field is so young that nearly every experiment yields novel and interesting phenomena. Thus although quantitative understanding is clearly desirable, the immediate development of the subject is likely to be dominated by identification of new phenomena and their qualitative documentation. There is also a need to design criteria which the field geologist and geochemist



can use to recognize the occurrence of the various effects caused by convection. We are only at the beginning of understanding the complex dynamic processes that affect magmas as they are generated and evolve within the mantle and crust.

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## REFERENCES

- Arculus, R. J. & Wills, K. J. 1980 *J. Petrol.* **21**, 743–799.
- Arndt, N. T., Naldrett, A. J. & Pyke, D. R. 1977 *J. Petrol.* **18**, 319–369.
- Atherton, M. P. 1981 *J. geol. Soc. Lond.* **138**, 343–350.
- Bartlett, R. W. 1969 *Am. J. Sci.* **267**, 1067–1082.
- Bateman, P. C. & Chappell, B. W. 1979 *Bull. geol. Soc. Am.* **90**, 465–482.
- Berkebile, C. A. & Dowty, E. 1982 *Am. Miner.* **67**, 886–889.
- Bowen, N. L. 1928 *The evolution of igneous rocks*. New Jersey: Princeton University Press.
- Brown, G. M. 1956 *Phil. Trans. R. Soc. Lond.* **240**, 1–53.
- Campbell, I. H. 1978 *Lithos* **11**, 311–321.
- Campbell, I. H., Roeder, P. L. & Dixon, J. M. 1978 *Contr. Miner. Petrol.* **67**, 369–377.
- Casey, J. F. & Karsen, J. A. 1981 *Nature, Lond.* **292**, 295–301.
- Chen, C. F. & Turner, J. S. 1980 *J. geophys. Res.* **85**, 2573–2593.
- Coriell, S. R. & Sekerka, R. F. 1981 *Phys. chem. Hydrodyn.* **2**, 281–293.
- Deardorff, J. W. & Willis, G. E. 1967 *J. Fluid Mech.* **28**, 675–704.
- Eichelberger, J. C. 1980 *Nature, Lond.* **288**, 446–450.
- Flower, M. F. J. 1980 *Nature, Lond.* **287**, 530–532.
- Herzberg, C. T., Baker, M. B. & Wendlandt, R. F. 1982 *Contr. Miner. Petrol.* **80**, 319–323.
- Hildreth, W. 1978 *Geol. Soc. Am. Spec. paper* 180, 43–72.
- Hildreth, W. 1981 *J. geophys. Res.* **86**, 10153–10192.
- Huppert, H. E. & Linden, P. F. 1979 *J. Fluid Mech.* **95**, 431–464.
- Huppert, H. E. & Sparks, R. S. J. 1980a *Nature, Lond.* **286**, 46–48.
- Huppert, H. E. & Sparks, R. S. J. 1980b *Contr. Miner. Petrol.* **75**, 279–289.
- Huppert, H. E. & Sparks, R. S. J. 1984 *A. Rev. Earth planet. Sci.* **12**, 11–37.
- Huppert, H. E., Sparks, R. S. J. & Turner, J. S. 1982a *Nature, Lond.* **297**, 554–557.
- Huppert, H. E., Sparks, R. S. J. & Turner, J. S. 1983 *Earth planet. Sci. Lett.* **65**, 377–381.
- Huppert, H. E. & Turner, J. S. 1981a *J. Fluid Mech.* **106**, 299–329.
- Huppert, H. E. & Turner, J. S. 1981b *Earth planet. Sci. Lett.* **54**, 144–152.
- Huppert, H. E., Turner, J. S. & Sparks, R. S. J. 1982b *Earth planet. Sci. Lett.* **57**, 345–357.
- Hurle, D. T. J. & Jakeman, E. 1981 *Phys. chem. Hydrodyn.* **2**, 237–244.
- Irvine, T. N. 1980a In *Physics of magmatic processes*, pp. 245–306. Princeton University Press.
- Irvine, T. N. 1980b *Am. J. Sci.* **A 280**, 1–58.
- Jackson, E. D. 1971 *Fortschr. Miner.* **48**, 128–174.
- Karner, F. R. 1968 *Bull. geol. Soc. Am.* **79**, 193–222.
- Kerr, R. & Turner, J. S. 1982 *Nature, Lond.* **298**, 731–733.
- McBirney, A. R. 1980 *J. Volcanol. geotherm. Res.* **7**, 357–371.
- McBirney, A. R. & Noyes, R. M. 1979 *J. Petrol.* **20**, 487–554.
- McCarthy, T. S. & Groves, D. I. 1979 *Contr. Miner. Petrol.* **71**, 193–209.
- Michael, P. J. 1983 *Geology* **11**, 31–34.
- Morse, S. A. 1979 *J. Petrol.* **20**, 555–590.
- Nicholls, I. A. 1971 *J. Petrol.* **12**, 67–119.
- Oxburgh, E. R. 1980 In *Physics of magmatic processes*, ch. 5, pp. 161–194. Princeton University Press.
- Rice, A. 1981 *J. geophys. Res.* **86**, 405–417.
- Rice, A. 1982 *Eos, Wash.* **63**, 1130.
- Shaw, H. R. 1963 *J. geophys. Res.* **68**, 6337–6343.
- Shaw, H. R. 1965 *Am. J. Sci.* **263**, 120–152.
- Shaw, H. R. 1974 *Carnegie Instn Wash. Yb.* **634**, 139–170.
- Shaw, H. R., Smith, R. L. & Hildreth, W. 1976 *Bull. geol. Soc. Am.* Abstracts with Program p. 1102.
- Smith, R. L. 1979 *Geol. Soc. Am. Spec. paper* 180, 5–28.
- Sparks, R. S. J. & Huppert, H. E. 1984 *Contr. Miner. Petrol.* (In the press.)
- Sparks, R. S. J., Meyer, P. & Sigurdsson, H. 1980 *Earth planet. Sci. Lett.* **46**, 419–430.
- Stolper, E. & Walker, D. 1980 *Contr. Miner. Petrol.* **74**, 7–12.
- Tait, S. R., Huppert, H. E. & Sparks, R. S. J. 1984 *Lithos* (In the press.)
- Turner, J. S. 1968 *J. Fluid Mech.* **33**, 183–200.
- Turner, J. S. 1979 *Buoyancy effects in fluids*, p. 368. Cambridge University Press.

- Turner, J. S. 1980 *Nature, Lond.* **285**, 213–215.  
 Turner, J. S. & Gustafson, L. B. 1978 *Econ. Geol.* **73**, 1082–1100.  
 Turner, J. S. & Gustafson, L. B. 1981 *J. Volcanol. geotherm. Res.* **11**, 93–125.  
 Turner, J. S., Huppert, H. E. & Sparks, R. S. J. 1983 *J. Volcanol. geotherm. Res.* **16**, 263–277.  
 Wager, L. R. & Brown, G. M. 1967 *Layered igneous rocks*. San Francisco, California: W. H. Freeman.  
 Wager, L. R., Brown, G. M., Wadsworth, W. J. 1960 *J. Petr.* **1**, 73–85.  
 Walker, D. & Delong, S. E. 1982 *Contr. Miner. Petr.* **79**, 231–240.  
 Watson, E. B. 1981 *Earth planet. Sci. Lett.* **52**, 291–301.  
 White, A. J. R. & Chappell, B. W. 1977 *Tectonophysics* **43**, 7–22.  
 Wilson, T. R. & Larsen, S. B. 1982 *Nature, Lond.* **299**, 625–626.  
 Wright, T. L. 1971 *U.S. Geol. Survey Prof. paper* **735**, 1–40.

### Discussion

M. SAKUYAMA (*Department of Geology, University of Southampton, SO9 5NH, U.K.*). Magma mixing between basaltic and dacitic magmas commonly occurs in calc-alkalic magma. However, density decreases monotonically with crystallization in calc-alkalic magma, so that the compositional stratification made by basaltic magma introduced into the bottom of silicic magma body cannot be destroyed by simple density change due to cooling at the boundary between them.

Would Dr Sparks suggest another possible mechanism to cause the breakage of the stratification and resultant magma mixing?

R. S. J. SPARKS. There are several alternative options for explaining the occurrence of mixing between dacitic and basaltic magmas in calc-alkaline systems.

(i) If the basaltic magma contains significant quantities of dissolved water and the dacitic magma were water-poor then basalt can be lower in density than dacite. Replenishment of a conventional magma chamber containing dacite by wet basalt in this circumstance could lead to mixing as the buoyant basalt rises through the dacite.

(ii) At low pressures, exsolution of a vapour phase can substantially reduce the density of basalt magma. Vesiculated mafic magma entering a high-level chamber may be lighter than the dacite leading to mixing. Alternatively, the mafic magma may be initially denser and pond beneath the dacite. However, heat transfer between the two magma layers can lead to crystallization and further volatile exsolution. The mafic magma can then mix when its bulk density reaches the same density as overlying dacite. The mixing could occur suddenly as envisaged by Huppert *et al.* (1982) or continuously from the interfacial region as envisaged by Eichelberger (1980). The experimental studies of Huppert *et al.* (1983) on two-layer fluid systems with large viscosity differences lead us to favour continuous release of vesiculated blobs of basalt into dacite, because these two magmas have viscosities differing by at least two orders of magnitude.

(iii) Dacitic magma could be formed at depth and be fed into the bottom of a basaltic magma chamber. Mixing could occur as the dacite rises into overlying basalt.

(iv) Wallrocks of dacitic composition could partly or completely melt when in contact with basalt magma. Under suitable circumstances such melts could hybridize with the basalt.

Which of these options is preferred will depend on detailed geological and petrological constraints. The greatest physical difficulty is how fluids of very different viscosity can be efficiently mixed together.

### References

- Eichelberger, J. C. 1980 *Nature, Lond.* **288**, 446–450.  
 Huppert, H. E., Sparks, R. S. J. & Turner, J. S. 1982 *Nature, Lond.* **297**, 554–557.  
 Huppert, H. E., Sparks, R. S. J. & Turner, J. S. 1983 *Earth planet. Sci. Lett.* **65**, 377–381..

M. J. O'HARA (*Department of Geology, U.C.W., Aberystwyth, Dyfed, U.K.*). Dr Sparks's results are obviously very bad news to those of us who seek to interpret the chemistry of erupted lavas in terms of simple fractional crystallization, with or without complications due to contamination, magma mixing and the variable proportions of trapped and escaping melts. Compositions of lavas erupted from the top of the magma chamber may be related to those of cumulates forming at the base of the same chamber only by complex and ill-understood processes.

These results are even worse news to those who seek to strip off the effects of shallow-seated processes and see through to the chemistry and processes of the underlying source regions of the magmas. I have little confidence in the uniqueness of most published interpretations which claim to see a *dominant* influence of partial melting processes and mantle heterogeneity upon erupted lava chemistry.

If progress is to be made, it is clearly very important to try to establish the scale, frequency and nature of double-diffusive layering that exist in different types of magma chambers. One way to approach the problem is through observations on consolidated chamber contents. Has Dr Sparks any information concerning the way in which double-diffusive systems might evolve during progressive consolidation? Can he suggest any observations on consolidated cumulates which would infallibly demonstrate the existence of double-diffusive layering in the magma chamber?

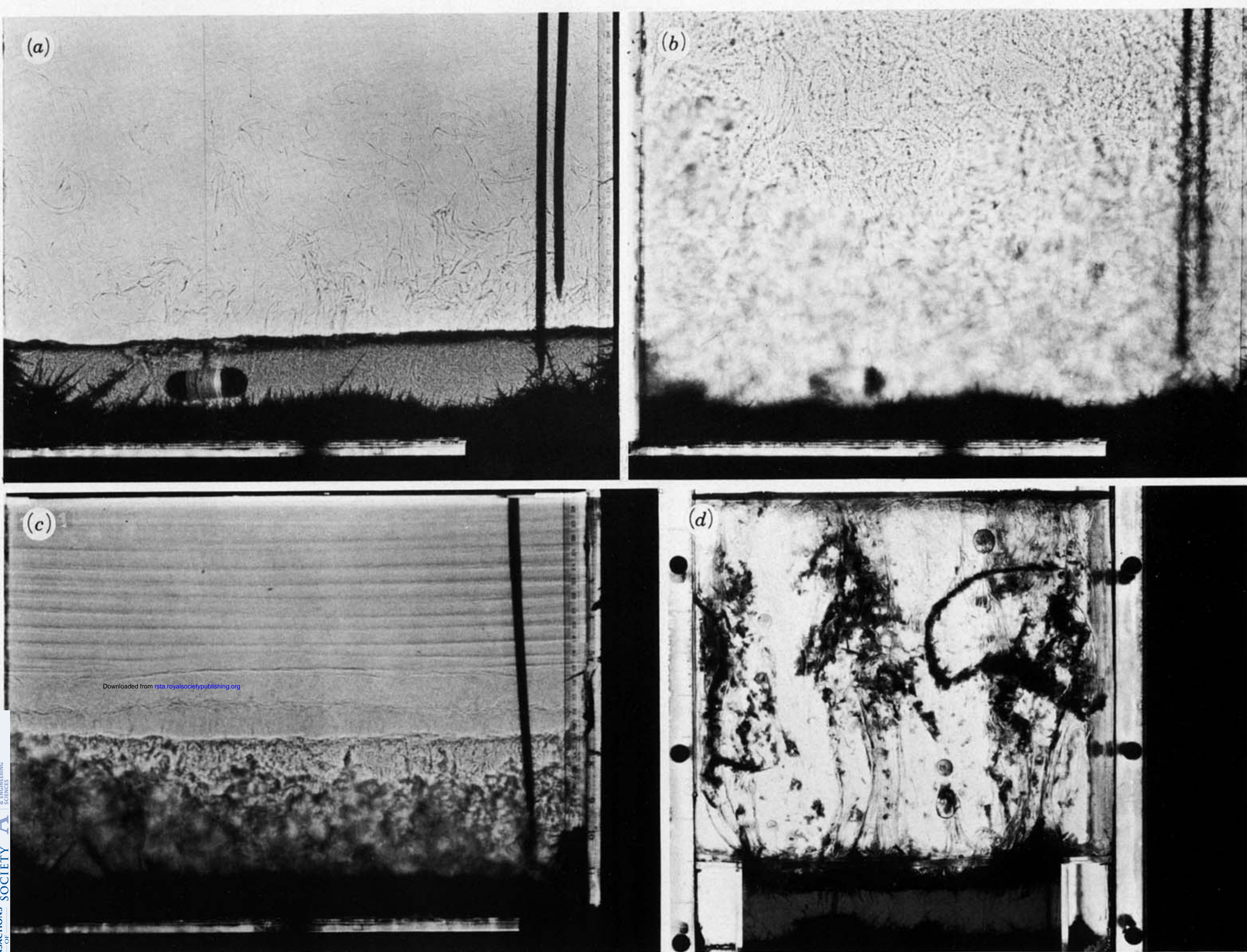
Viscosity and gravitational acceleration are important variables in the relations. How might the marked differences in these factors affect the behaviour of melts in, for example, lunar magma chambers relative to those in terrestrial magma chambers?

R. S. J. SPARKS. We agree with Professor O'Hara that the chemical evolution of magmas is governed by processes that are more complex than had once been envisaged. Perhaps the most important result of the tank experiments described in our paper is that crystallizing fluid systems, though much simpler than natural magmas, already show a wide range of novel and interesting fluid dynamic phenomena that have a major influence on the chemical evolution of the fluids. We take this opportunity, however, to emphasize that a great deal of experimental and theoretical research is still required to understand how these processes operate in magma chambers. To answer Professor O'Hara's questions with confidence, a number of steps are required. The scale of double-diffusive layering in magma chambers cannot be predicted until a wider range of fluid systems has been studied to establish the effects of different viscosities and chemical diffusivities on convection. Experiments on silicate melts must also be carried out to examine compositional convection due to crystallization. Another urgent requirement is to produce a good theory of compositional boundary-layer flow that can be verified experimentally.

We are certain that criteria will emerge to distinguish these processes from conventional ones. There is already a considerable amount of data on igneous rocks that are hard to explain without invoking new processes. Perhaps the best situation at present concerns the origin of compositional zonation in silicic magma systems. In some pyroclastic sequences the last explosions involve disruption of the chamber wall, resulting in eruption of partly consolidated plutonic nodules. There is a good chance that these nodules represent crystallization along the chamber margins that was contemporaneous with the evolution of the magma. In some of these systems it is already apparent that the phenocrysts are present neither in the appropriate proportions

nor with the right compositions to account for the extreme differentiated liquids. Detailed studies of co-genetic nodules, phenocrysts and liquids from such volcanic sequences could well provide clear evidence for convective fractionation and could constrain the time-scales involved. For example, it might emerge that the variations in liquid compositions are better explained by fractionation of nodule assemblages rather than by phenocrysts.





**FIGURE 4.** (a) Shadowgraph showing the results of cooling a constant concentration gradient of  $\text{Na}_2\text{CO}_3$  solution from above. The crystals grown on the roof caused a pool of light residual fluid to accumulate beneath the roof. The double-diffusive layering in the container is formed by cooling of the concentration gradient from above. Details of the experiment are given by Chen and Turner (1980).

(b), (c) Shadowgraph showing the results of cooling along a vertical central pipe immersed in a container of homogeneous  $\text{Na}_2\text{CO}_3$ . After 7 h (b) crystals growing along the pipe have generated boundary layer flows of light fluid which has formed a pool of residual fluid at the top of the container. The dye streak shows the presence of weak stratification in the main part of the container. After 3 days (c) sidewall crystallization has produced compositional stratification throughout the container. Further details are given in Turner & Gustafson (1981).

(d) Shadowgraph showing the results of crystallization from the floor of  $\text{Na}_2\text{CO}_3$  solution. After 4 h 55 min thin plumes of light residual fluid are seen rising from the floor. Further details of the experiment are given by Chen & Turner (1980).



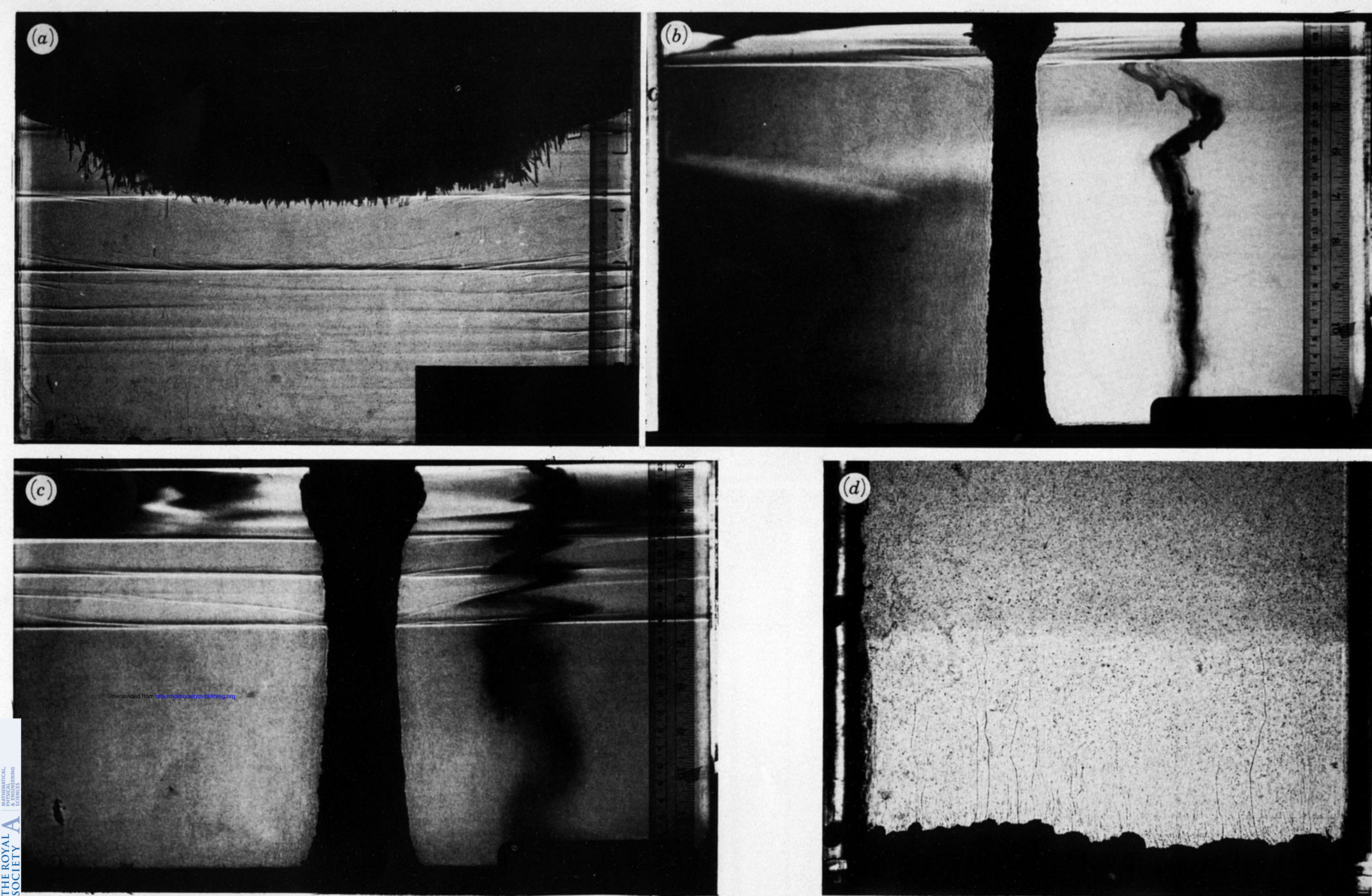


FIGURE 5. (a) Shadowgraph showing a layer of hot  $\text{KNO}_3$  solution beneath a layer of cold  $\text{NaNO}_3$  solution. Thermal convection is visible in the upper and lower layers as they exchange heat across the double-diffusive interface. Crystals of  $\text{KNO}_3$  are growing on the floor of the container.

(b) Shadowgraph showing the same experiment as in (a) in which the lower layer of  $\text{KNO}_3$  solution has overturned and is mixing with the upper  $\text{NaNO}_3$  solution. Further details are given in Huppert & Turner (1981b).

(c) Shadowgraph showing the overturn of a layer of  $\text{KNO}_3$  solution into a gradient region of  $\text{K}_2\text{CO}_3$  solution. Note that the mixing is confined to the lower part of the stratified solution. The double-diffusive layering is formed by heating of a concentration gradient from below and by side-wall heating. Further details are given in Huppert *et al.* (1982b).

(d) Photograph of an experiment in which  $\text{KNO}_3$  solution at  $60^\circ\text{C}$  has been emplaced beneath glycerine at  $11^\circ$ . Blobs and plumes of residual  $\text{KNO}_3$  solution are generated at the interface and rise continuously into the glycerine. Crystals of  $\text{KNO}_3$  form in the plumes as they cool and the crystals, often in the form of a long chain, fall back. Residual  $\text{KNO}_3$  solution forms a layer above the glycerine. Further details are given in Huppert *et al.* (1983).