DOUBLE-DIFFUSIVE CONVECTION DUE TO CRYSTALLIZATION IN MAGMAS

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1. INTRODUCTION

An important development in the understanding of the fluid dynamics of convection has been the recognition that heat- and mass-transfer processes in multicomponent systems are often fundamentally different to those in the more familiar one-component systems. In a system containing two or more properties that have different molecular diffusivities and opposing effects on the vertical density gradient, a wide range of novel and complex convective phenomena can occur. In the last few years fluid dynamicists and geologists have recognized that such convection, known by the general term of *doublediffusive convection*, can occur in magmas and in other fluids of geophysical interest. The implications of this form of convection are far-reaching and are likely to revolutionize our perceptions of many geophysical processes.

Theoretical and experimental analyses of double-diffusive convection were first developed in relation to the oceans, specifically with a view to explaining several different kinds of layering (Huppert & Turner 1981a). It is also apparent that double-diffusive convection is likely to occur in many other systems of geophysical interest. Much attention has been given in particular to the notion that double-diffusive convection can play an important role in the differentiation of magmas. Some of the most important effects in magmas are due to the crystallization induced by cooling.

This article reviews the theoretical and experimental developments in the study of convective phenomena in both multicomponent fluids and crystallizing fluid systems. We concentrate on convection in magmas because of the recent blossoming of research in this area. We also indicate other types of fluids where double-diffusive and compositional effects are likely to be important.

2. CONVECTION IN MULTICOMPONENT FLUIDS

Most of the early studies of the fluid dynamics of double-diffusive convection were developed with oceanographic applications in mind (Huppert & Turner 1981a), for which the two relevant properties are heat and salt. These two properties have different molecular diffusivities $(D_{\text{HEAT}} \approx 10^{-3} \text{ cm}^2 \text{ s}^{-1}; D_{\text{SALT}} \approx 10^{-5} \text{ cm}^2 \text{ s}^{-1})$. In many natural situations the gradients of temperature and salinity are such that one of the properties has a destabilizing effect (i.e. leads to convection), although the system has an overall apparently stable density stratification. In these circumstances, horizontal layering inevitably occurs. Layering developed in this way has attracted the attention of petrologists because of the resemblance of these structures to layering observed in igneous intrusions (McBirney & Noyes 1979). This is arguably a superficial resemblance, but the physical and chemical properties of silicate melts are such that we consider double-diffusive convection certain to occur. Recent detailed studies of volcanic rocks have also demonstrated that many magma chambers contain stable compositional gradients but unstable temperature gradients (Hildreth 1981), which is a situation where double-diffusive effects can be expected.

Before illustrating the principles of double-diffusive convection with some examples, we briefly review the physics of thermal convection in a one-component system so that the differences will be more apparent. If a layer of uniform fluid is heated from below, the lowermost parts of the layer heat up, expand, and become buoyant. The buoyant fluid parcels rise. If the heating is sufficiently vigorous, these parcels cause circulation throughout the fluid layer, which is recognized as convection. The pattern of circulation and the heat transfer across the fluid can generally be predicted given the physical properties of the fluid and the applied temperature field. The dimensionless parameters characterizing the motion are the thermal Rayleigh number and the Prandtl number, defined as

$$Ra_{T} = g\alpha\Delta T d^{3}/\kappa_{T} v$$

(1)

and

$$Pr = \nu/\kappa_{\rm T},\tag{2}$$

where g is the gravitational acceleration, α the thermal expansion coefficient, ΔT the temperature difference across the layer, d the fluid layer thickness, κ_T the thermal diffusivity, and v the kinematic viscosity. The Rayleigh number is the ratio of the buoyancy force driving convection to the viscous force resisting fluid movement, and the Prandtl number is the ratio of the diffusivities of momentum to heat. Convective motions are initiated when Ra_T is of the order 10³. The exact value depends on the specific boundary conditions but is independent of the Prandtl number. The convective motion becomes turbulent when Ra_T exceeds a value of approximately 10⁶ for the large Prandtl numbers that characterize magmas.

Several authors have calculated typical thermal Rayleigh numbers in magma chambers (Shaw 1965, Bartlett 1969, Rice 1981). These calculations yield Rayleigh numbers generally well in excess of 10^6 and range up to 10^{23} for large basaltic chambers. Even in a 1-km-deep magma chamber containing a relatively high-viscosity rhyolitic melt, the calculated Rayleigh number can still be 10^9 . Such high values imply that the vast majority of magma chambers are in vigorous turbulent convection. This application of purely thermal convection theory has two implications. First, crystal settling would be inhibited because the convective velocities in the interior of the chamber are orders of magnitude larger than the settling velocities of crystals (Sparks et al 1984). Second, because of the convection, no vertical property gradients could survive in the interior of a magma chamber. Of course, these problems do not necessarily arise, because in many cases the application of purely thermal convection is inappropriate, as is described below.

Two specific concepts of double-diffusive convection are illustrated in Figure 1, which shows part of a laboratory experiment using aqueous solutions of K_2CO_3 and KNO_3 . In the experiment, described in detail by Huppert et al (1982b), a tank measuring $40 \times 20 \times 30$ cm high was filled with aqueous K_2CO_3 in such a way that there was a linear density gradient with depth, resulting in a stable stratification. The temperature of the K_2CO_3 solution was initially uniform at 11°C. An intrusion of a hot (65°C), relatively heavy, dyed KNO_3 solution was rapidly introduced at the base of the tank. Because of its density, the KNO_3 solution ponded at the base of the tank to form a separatelayer, as marked by the dye. We return in a later section to the behavior of the lower layer; in this section, we are concerned only with the early behavior of the K_2CO_3 solution.

Figure 1 shows the experiment 20 minutes after the introduction of the KNO_3 layer. We observe that the stably stratified K_2CO_3 layer has been transformed into two different series of layers. These are a consequence of double-diffusive convection. There are two scales of lavers produced, and we consider their origin in turn. The finer-scale layers in the upper part of the K₂CO₃ solution are the result of sidewall heating and were produced, before the introduction of the hot KNO₃ solution, by the following mechanism. The laboratory temperature was approximately 20°C, and heat was transferred into the tank. At any given level, fluid close to the sidewall becomes hotter and therefore lighter. As heating proceeds, the thermal boundary layer thickens and produces upward flow. However, because of the small diffusivity of K₂CO₃, the upward-flowing fluid retains its K₂CO₃ content from its level of origin, while moving upward into a decreasing K₂CO₃ concentration and therefore a decreasing density. The upward flow is thus constrained by the density stratification and eventually reaches a level of equal density, where it is diverted sideways. This creates a local circulation that progressively drives a layer of circulating fluid into the



Figure 1 Shadowgraph view of an experiment in which an upper layer of compositionally stratified K_2CO_3 solution is heated from below by a hot layer of KNO_3 solution. Double-diffusive layering occurs in the K_2CO_3 solution, seen here 20 minutes after the experiment began. Details are given in Huppert et al (1982b).

interior of the tank. Such a series of layers cannot occur in a one-component system.

The formation of the layers depends critically on the contrast in diffusivities of K_2CO_3 and heat. It is because the diffusivity of K_2CO_3 is much less than the diffusivity of heat that the hot fluid rising along the sidewall retains almost all its original content of K_2CO_3 . The heat transferred from the room is only capable of changing the density slightly. In contrast, the background vertical density variations due to compositional effects are relatively large. Consequently, the fluid can only rise a short distance before coming to a level where the densities are equal. The spacing of layers formed in this way is dependent on the temperature difference across the walls of the tank, the vertical density gradient, and the physical properties of the fluid. The same type of layering would form if the exterior were cold and the compositionally graded fluid in the tank were hot. The only difference between the two cases would be the sense of circulation (see Figures 3 and 5 in Huppert & Turner 1980).

In Figure 1, layering of a larger scale is observed in the lower regions of the K₂CO₃ solution immediately above the hot KNO₃ layer. These layers are the consequence of the heat transfer to the compositional gradient from the KNO₃ layer below. The temperature at the base of the K₂CO₃ solution increases, forming a thermal boundary layer that grows with time. Eventually, the base of the K_2CO_3 solution is heated sufficiently to cause parcels of buoyant fluid to rise and initiate convection. However, in contrast to the one-component case, a parcel of fluid retains its K₂CO₃ content as it rises (due to its low diffusivity). In an ambient density gradient, a parcel of fluid can only rise a short distance before it reaches a level where the ambient density has the same value as that of the parcel. A layer of circulation is formed up to this level. This convecting layer grows while transferring some heat to the fluid above. With time this causes another convecting layer to form, and then another and another. In this way the potential energy available from the thermal input drives convection, despite the stable stratification in the K₂CO₃ solution. The spacing of these layers is greater than that of the layers formed from sidewall heating because the thermal energy is greater from the hot KNO₃ layer, thus providing more potential energy to create thicker layers.

The phenomena illustrated in Figure 1 are just two of many different examples of double-diffusive structures (Huppert & Turner 1981a), some of which are considered later. However, the principles discussed in this section are general: variations in properties with different diffusivities have a profound influence on convection. In the case of silicate melts, quite small changes in composition can produce substantial density changes. Furthermore, the major chemical components in silicate melts have very small diffusivities (typically in the range 10^{-6} to 10^{-10} cm² s⁻¹), whereas the thermal diffusivity ranges approximately from 10^{-2} to 10^{-3} cm² s⁻¹. Magmas are thus suitable fluids for the formation of double-diffusive structures.

The theoretical and experimental understanding of double-diffusive convection has been recently reviewed by Turner (1979, Chapter 8) and Huppert & Turner (1981a). The reader is referred to these articles for more detailed and comprehensive treatments. We now briefly review aspects of this work that are particularly relevant to geological systems. In a twocomponent system containing two properties with different molecular diffusivities, four different situations can be envisaged. In practice the two properties of interest are temperature (T) and composition (S). The four different possibilities are shown in Figure 2. One of these situations is absolutely stable (Figure 2*a*); in this case the temperature increases upward and the heavy chemical components are concentrated toward the base of the system. Another situation is unstable (Figure 2*b*); in this case the temperature decreases upward and the heavy chemical components are



Figure 2 Sketches of the four different kinds of temperature and chemical composition that can occur through a fluid layer. Temperature (T) variation with depth in the layer is shown as a solid line, and the variation in the distribution of heavy chemical components (S) is shown as a broken line. (a) Both properties have stable distributions; (b) both properties have unstable distributions; (c) temperature distribution is destabilizing, and compositional distribution is stabilizing; (d) compositional distribution is destabilizing, and temperature distribution is stabilizing.

concentrated toward the top. The resulting convection in this situation is similar to that in a one-component system, with the compositional distribution enhancing the convection. The Rayleigh-number criterion can be used to predict when convection can begin. Equation (1) is modified to include the influence of compositional variations in density, such that

$$Ra_{e} = Ra_{T} + g\beta\Delta Sd^{3}/\kappa_{s}\nu, \qquad (3)$$

where β is the coefficient of expansion due to compositional change, $\rho_0\beta\Delta S$ is the (positive) difference in density due to compositional changes across a fluid layer of mean density ρ_0 and thickness *d*, and κ_s is the compositional diffusivity.

The two other possibilities of temperature and compositional gradients (Figures 2c, d) constitute the conditions for double-diffusive convection. In both cases the overall density is stable. In Figure 2c the system is unstable with respect to temperature (the temperature increases downward), but stable with respect to composition, since the heavy components are concentrated downward. This situation was illustrated in the experiment described earlier (Figure 1). It is known as the "diffusive" regime, and a series of horizontal layers can readily form in this situation. We believe it is the geologically most important situation, because there are many circumstances where hot magma can lie beneath colder magma of lower density. Some of these circumstances are discussed in the sections that follow.

The fourth situation (Figure 2d) involves a stable temperature distribution (temperature increasing with height) with an unstable compositional profile in which heavy components are concentrated upward. In this situation the phenomenon of "fingering" occurs, in which fluids from different levels of the system interpenetrate in long, thin convection cells (see Figures 1 and 3 in Huppert & Turner 1981a). Adjacent cells transfer their heat in the manner of a heat exchanger, with the energy for the motion coming from the unstable composition field. While such motions definitely occur in the ocean, where they are called "salt-fingers" and have been photographed at a depth of 1500 m in the North Atlantic (Williams 1974), so far this situation does not appear to be geologically important. This is because rather unusual circumstances have to be invoked to get hot, compositionally dense magma to overlie colder, compositionally less-dense magma. However, Kantha (1980) has suggested that some columnar jointing in lavas can be produced in this way, but there is little evidence to favor this explanation over contraction jointing.

At this point we outline some of the theoretical considerations governing double-diffusive convection. In a one-component system the motion can be specified by the thermal Rayleigh number (Equation 1) and the Prandtl number (Equation 2). In a two-component system two additional parameters are required, such as the compositional Rayleigh number¹

$$Ra_{\rm s} = g\beta\Delta S d^3 / \kappa_{\rm T} v \tag{4}$$

and the ratio of the diffusivities

$$t = \kappa_{\rm S} / \kappa_{\rm T}, \tag{5}$$

which lies between 0 and 1. Frequently, it is convenient to use the ratio of the compositional to the thermal Rayleigh number

$$Q = \beta \Delta S / \alpha \Delta T \tag{6}$$

in place of the compositional Rayleigh number.

Linear stability theory applied to a single layer of fluid indicates that infinitesimal motions are initiated for $Pr \gg 1$ and $\tau \ll 1$ when $Ra_T - Ra_s$ exceeds a critical value of order 10³. However, nonlinear convection is known to be possible at lower values of $Ra_T - Ra_s$ (Huppert & Moore 1976), and Proctor (1981) has shown that in the limit of very small τ , nonlinear convection is possible if Ra_T alone exceeds a critical value of order 10³. The minimum Rayleigh number needed to maintain a series of layers still remains to be found; but all laboratory experiments indicate that convecting layers easily form in double-diffusive systems.

Because much of the development of double-diffusive convection has been related to the oceans, many of the intuitive and quantitative results obtained are for parameters appropriate for salty water. Considerable caution is required in applying results in a quantitative way to geological systems. The Prandtl number and ratio of the diffusivities are different in the two cases, and the quantitative influences of these two parameters have not been evaluated for all possible situations. For example, the thickness of the thinner layers in Figure 1 is given by (Huppert & Turner 1980)

$$h = 0.65\Delta_{\rm h}\rho \left| \frac{d\rho}{dz} \right|,\tag{7}$$

where $\Delta_{\rm h}\rho$ is the horizontal density difference between fluid at the wall and in the far interior, and $d\rho/dz$ is the vertical density gradient. The constant of proportionality should vary little, if at all, with variations in Pr and τ . However, the mean thickness \bar{h} of the lowest layers in Figure 1 is given by (Huppert & Linden 1979)

$$\bar{h} \propto \kappa_{\rm T}^{1/2} (g \ d\rho/dz)^{-1/4},$$
 (8)

¹ We use κ_T , rather than κ_S , in the denominator for convenience and historical reasons; one could use the single parameter Ra_S/ τ , which would have κ_S in the denominator.

where the constant of proportionality for NaCl in water is 51. This value is likely to be strongly dependent on Pr and τ , and its value for magmatic values of Pr and τ is as yet undetermined.

3. CONVECTION IN CRYSTALLIZING FLUIDS

In the previous section we showed that once compositional and thermal gradients are established, double-diffusive effects can occur in any multicomponent fluid, but we left aside the question of how suitable gradients can be generated in geophysically interesting fluids. An important mechanism in the case of magmas is crystallization, which generally causes much greater changes in melt density than are caused by associated temperature changes (McBirney 1980, Sparks & Huppert 1984). Following studies by Chen & Turner (1980) and McBirney (1980), it has become evident that crystallization in a fluid can cause a wide range of novel and interesting convective effects, including the generation of compositional and thermal gradients in initially homogeneous fluids. The convective effects of crystallization are of fundamental importance to the understanding of magma genesis and the behavior of the Earth's core (Gubbins et al 1979, Loper & Roberts 1983), as well as being important in many industrial processes [see Vol. 2, No. 4 of Physicochemical Hydrodynamics, edited by Hurle & Jakeman (1981)].

The basic principles of convection due to crystallization are simple. Crystals growing from a multicomponent, saturated fluid selectively deplete fluid adjacent to the growing interface in either light or heavy components. The composition, and therefore the density of the fluid, is changed and convection can occur.

The effects of crystallization on convection have been studied using aqueous solutions of simple salts (Chen & Turner 1980, McBirney 1980, Turner & Gustafson 1981, Huppert & Turner, 1981b, Huppert et al 1982b, Kerr & Turner 1982). The solubility of most salts in water increases with temperature. Thus, cooling a saturated solution (for example, KNO_3) will cause crystallization and a decrease in the density of the residual solution (Huppert & Turner 1981b). The decrease in fluid density is a consequence of the removal of heavy components to form crystals. Purely thermal contributions to density, which would tend to increase the density of a cooling solution, are here dominated by the compositional contributions. In some systems, crystallization can cause density to increase in residual fluids as a result of selective removal of light components. Examples include the formation of ice from some aqueous solutions and the geologically important case of plagioclase feldspar crystallizing from basaltic magma (Sparks & Huppert 1984).

Laboratory Experiments

The study of convective phenomena in crystallizing solutions is a new field of fluid mechanics. Some of the physical and chemical effects are currently being elucidated by laboratory experiments on aqueous solutions. One type of experiment has involved crystallization of an initially homogeneous solution by cooling at a boundary. In the case of cooling along a vertical wall, a number of experimental studies have shown that when crystallization takes place on the vertical wall, so that heavy components are removed from the fluid, a boundary-layer flow of light solute-depleted fluid is generated (McBirney 1980, Turner 1980, Turner & Gustafson 1981). Light fluid accumulates at the top of the container to form a layer that is thermally and compositionally stratified. This stratified region grows at the expense of a convecting, well-mixed lower layer (Figure 3). Layering is formed in the stratified upper fluid by various double-diffusive processes, such as internally generated instabilities, cooling across a sidewall, and heating by the underlying homogeneous fluid. Eventually the whole container can become stratified and layered. The structure of the boundary layer is strongly influenced by double-diffusive effects because the low



Figure 3 Shadowgraph view of the cooling of a container of homogeneous Na_2CO_3 solution at a vertical central pipe. After 23 hours 19 minutes, crystals have grown around the pipe, and a layer of compositionally stratified and depleted solution occupies the upper part of the tank. The temperature gradient imposed across the stratified region has resulted in double-diffusive layers forming. Details are given in Turner & Gustafson (1981).

diffusivity of chemical components enables the light, solute-depleted fluid generated by crystallization to retain its composition.

When crystallization occurs by cooling the floor of a container, convection can also occur, even though the temperature gradient is stable. Once crystals form they are observed to generate thin streams of uprising depleted fluid that drive convective circulation (Chen & Turner 1980). Such experiments reveal some interesting features of compositional convection driven by crystallization. The vertical streamers are typically very thin (<0.1 cm), and the flow is usually laminar. Consequently, such convection produces rapid vertical exchange but rather slow lateral mixing.

A variety of experiments have been performed to investigate additional interesting effects. For example, Kerr & Turner (1982) showed that the crystal layers in the solidified product resulted from fluid layers in a container, although the compositions and thicknesses of the fluid and crystal layers were different from one another. Tait et al (1984) have shown that compositional convection can occur during crystallization in a porous bed of glass balls. We have also carried out as yet unpublished experiments (with J. S. Turner) on crystallization along tilted boundaries. Compositional gradients are formed as a consequence, because light fluid rising from different positions along the boundary mixes with different amounts of overlying fluid.

A different kind of experiment involves replenishment of a container with fluid of another, dissimilar composition and temperature. These experiments were designed to investigate fluid-dynamic aspects of open-system magma chambers, for which there is an influx of primitive magma into a chamber containing more-differentiated magma (see Huppert & Sparks 1980a,b). In the experiments (Huppert & Turner 1981b), a layer of hot KNO₃ solution was emplaced beneath a layer of cold and lower-density NaNO₃ solution. A sharp double-diffusive interface was formed between the layers, across which heat was transferred. Convective cooling of the lower layer resulted in crystals of KNO₃ growing in the interior and on the bottom of the container. Compositional convection augmented the effects of thermal convection, maintaining a well-mixed lower layer. The density of the residual fluid decreased with time and eventually reached the same density as the overlying NaNO₃ solution. Thereafter, the residual fluid ascended rapidly into the overlying layer and mixed thoroughly with it.

This experiment illustrates some important principles, which are relevant to a number of geological situations. Hot fluid can lie stably beneath cold fluid if its composition is such that it is denser. Such a situation can occur in nature when primitive dense magma from the mantle is emplaced into a chamber containing more-differentiated magma (Sparks et al 1980, Huppert & Sparks 1980a,b), when hot brines or sulfide-bearing hydro-

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thermal solutions are discharged onto the seafloor (Turner & Gustafson 1978, Solomon & Walshe 1979), or when a layer of higher-temperature mantle of one composition underlies a mantle layer of another composition. In the case of magmas and brines, the cooling of such hot fluid layers can lead to crystallization or precipitation, respectively, and allow instabilities to develop, causing the fluid layers to mix. An important principle related to igneous processes is that fluid layers of different composition can coexist in the same container and evolve as chemically independent systems. We note that no crystals could have formed in the experiment described had the fluids been initially mixed together.

Huppert et al (1982b) modified this experiment in two ways. In the first variation, the hot KNO_3 solution was introduced beneath compositionally stratified K_2CO_3 solution, as already described (Figure 1). The lower layer of KNO_3 solution (see Figure 1) will eventually overturn owing to crystallization, but because of the vertical density gradient, it only mixes with the lower parts of the stratified fluid. In the second variation, hot KNO_3 solution. Quench crystallization occurred around the inlet orifice. This released light residual fluid into the container, and a mound of interlocking crystals was formed.

A further modification of the replenishment studies has been to investigate the effects of viscosity variations (Huppert et al 1983). In these experiments the viscosity and density of the upper layer in the two-layer system were systematically varied by using combinations of glycerin, sugar, and water. When the viscosity ratio was large (upper:lower viscosity = 2700:1), crystallization took place along the interface, and light residual KNO₃ solution was continuously swept up into overlying viscous fluid (Figure 4). No abrupt overturn took place, and release of residual fluid began immediately. This behavior is believed to be due to viscous coupling at the interface, which enables the residual fluid from the lower layer to be incorporated into thermal plumes in the viscous fluid.

As plumes of light fluid ascended into the residual fluid, further crystallization of KNO_3 occurred. These crystals were initially carried up in the plumes but eventually fell out. The plumes ascended to the top of the tank and deposited a layer of light KNO_3 solution on top of the glycerin. The experiments indicate that viscosity differences could play an important role in the fluid dynamics of crystallizing systems, with interfacial effects becoming significant. They also illustrate the difficulty of mixing together fluids of strongly contrasted viscosity.

Theoretical Aspects

Much of the research on convection in crystallizing systems has been concerned with identifying new phenomena, and many of the processes

observed in the experiments have yet to be characterized quantitatively. There have been only limited developments in the theoretical understanding of convection associated with crystal growth. Loper & Roberts (1983) have developed a theoretical model, which has yet to be tested by experiment. In industrial contexts, where the main interest is in eliminating convective effects (see Hurle & Jakeman 1981, Coriell et al 1980, Coriell & Sekerka 1981, Schaefer & Coriell 1982), some theoretical work has been completed concerning instabilities along single faces of crystals.

Major differences between compositional convection associated with growing crystals and thermal convection are the length scales of the flows and the critical conditions for instability. Some insight into these differences can be ascertained by first considering the development of a compositional boundary layer next to a growing crystal face. In a quiescent liquid, rejection or incorporation of solute results in an exponential concentration profile in a thin film adjacent to the growing crystal (Coriell et al 1980, Coriell & Sekerka 1981). This film has a characteristic thickness $\delta = \kappa_s/V$, where κ_s is the diffusion coefficient, and V is the growth velocity of the crystal face. An approximate approach to determining the stability of a



Figure 4 Photograph of an experiment in which KNO_3 solution at 60°C has been emplaced beneath glycerin at 11°C. Blobs and plumes of residual KNO_3 solution are generated at the interface and rise continuously into the glycerin. Crystals of KNO_3 form in the plumes as they cool, and the crystals, often in the form of a long chain, fall back. Residual KNO_3 solution forms a layer above the glycerin.

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compositional film is to substitute the film thickness into the compositional Rayleigh number, which can be re-expressed as

$$Ra_{s} = \frac{g\Delta\rho\kappa_{s}^{2}}{V^{3}\mu}.$$
(9)

For a horizontal film above a crystal face (g vertical), the classic Rayleigh criterion would be $Ra_s > 10^3$.

Figure 5 shows fields of stable films (Ra_s < 10³) and unstable films (Ra_s > 10³), for fluids of viscosities 10² and 10⁶ poise and a value of $\Delta \rho = 0.01$ g cm⁻³, on a plot of diffusion coefficient against crystal growth velocity. Growth velocities in the range 0.1–10 cm yr⁻¹ are typical of slowly cooled magma chambers. Also shown are lines of constant κ_s/V . The diagrams illustrate that in many magmatic situations compositional convection should occur as crystallization proceeds.

A similar diagram can be constructed for the laboratory experiments

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Figure 5 The solid lines in this diagram mark the value of $Ra_{(film)} = 10^3$ as a function of chemical diffusivity and crystal growth velocity for magma with low viscosity ($\mu = 10^2$ poise) and with high viscosity ($\mu = 10^6$ poise). A value of $\Delta \rho = 0.01$ g cm⁻³ in Equation (9) is assumed. These lines separate conditions of stability ($Ra < 10^3$) from conditions where compositional convection away from a horizontal crystal face is expected. The dashed lines are lines of constant film thickness (κ_s/V).

with aqueous solutions (Figure 6), where crystal growth rates are in the range 10^{-3} to 10^{-5} cm s⁻¹, viscosities are of the order 10^{-2} poise, and diffusivities are of the order 10^{-5} cm² s⁻¹. In all the experimental studies referred to previously, horizontal crystal faces should generate unstable compositional boundary layers. From this point of view, a KNO₃ crystal face growing at 10^{-4} cm s⁻¹ ($D = 10^{-5}$ cm² s⁻¹ and $\mu = 0.01$ poise) would be dynamically similar to an anhydrous crystal growing at 0.66 cm yr⁻¹ in a wet rhyolitic magma ($D \approx 10^{-7}$ cm² s⁻¹ and $\mu = 10^{6}$ poise). In the laboratory experiments the compositional film would become unstable at $\delta \approx 0.01$ cm. This is broadly consistent with the plumes that are observed to rise from growing crystals, which are certainly considerably thinner than a millimeter. We note that a horizontal crystal face represents the most stable configuration and thus the worst case for assessing whether convection will occur. For vertical crystal faces, no stable condition exists. Consequently, there will always be some tendency for the compositional film to detach from the crystal surface.

Unfortunately, an analysis of the dynamic development of a compositional boundary layer next to growing crystals has not yet been completed. There are already, however, some indications of the nature of such boundary layers and the problems likely to be encountered with the



Figure 6 The solid line marks the position of conditions where $Ra_{(film)} = 10^3$ on a plot of chemical diffusivity vs crystal growth velocity. The fluid in this case has a viscosity of 0.01 poise, a typical value for the aqueous solutions used in the laboratory experiments. The value of $\Delta\rho$ in Equation (9) is taken as 0.01 g cm⁻³. The laboratory experiments all fall in the field of Ra > 10³.

analysis. The stagnant-film model described above and experimental observations indicate that the flows will be thin. In both the laboratory experiments and in magmas, the flow will be laminar. One problem is that the boundary layers are often much thinner than the associated crystals. Consequently the flow will be complicated by the geometrical irregularities of the surrounding solid boundaries.

These features suggest some interesting possibilities for experimental studies of silicate systems. Because of the small length scales necessary for compositional convection, these effects should be noticeable in small capsules, including some currently used by experimental petrologists. Dowty & Berkebile (1982), for example, have reported strong gradients in melt composition during crystallization experiments in capsules of 1 cm diameter. Although the authors attribute the gradients to crystal settling, there is no evidence to favor this interpretation over one involving convection. Kirkpatrick et al (1981) have also identified compositional gradients adjacent to olivine crystals grown in a melt of diopside composition. The effects of compositional convection should be identifiable in silicate melts, and suitably designed experiments would provide a most interesting approach.

4. CONTROLS ON MAGMA DENSITY

From the previous discussions on convection it is clearly important to establish the relative contributions of composition and temperature to the density of magmas, particularly during crystallization and melting processes. There have been a large number of determinations of the densities of silicate melts, and it is now possible to predict the density of a melt of given composition and temperature with an accuracy generally better than 1% (Nelson & Carmichael 1979, Bottinga et al 1982). These studies reveal that small changes in melt composition can have large effects on density.

Recent studies (McBirney 1980, Sparks et al 1980, Stolper & Walker 1980, Sparks & Huppert 1984) have shown that during fractional crystallization of basaltic magmas the compositional changes during an increment of fractionation are generally much larger than associated thermal effects. The quantitative aspects were considered by Sparks & Huppert (1984), who introduced a parameter called the *fractionation density*, which is defined as the density of the fluid components being selectively removed by crystallization. The fractionation density is thus

$$\rho_{\rm c} = \frac{\hat{M}_{\rm c}}{\hat{V}_{\rm c}},\tag{10}$$

where \hat{M}_{c} is the gram formula weight, and \hat{V}_{c} is the partial molar volume of the components being removed from the melt into crystals. When ρ_{c} is greater than the melt density, the residual melt decreases in density. When ρ_{c} is less than the melt density, the residual melt increases in density.

Figure 7 plots the fractionation density against crystal composition of some common minerals formed during fractionation of basaltic magma. In the plot, the composition of the crystals is expressed in terms of the two main end members of the solid solution series (for example, A = forsterite and B = fayalite for olivine). The upper and lower density lines for each mineral represent temperatures of 1200 and 1400°C, respectively. The typical ranges of densities for basaltic melts are shown. The diagram demonstrates that in general the effects on density of fractionating minerals are much greater than are those of temperature.



Figure 7 Fractionation densities of olivine, orthopyroxene, clinopyroxene, and plagioclase plotted against composition. The components A and B are separately defined for each mineral: olivine, $A = Mg_2SiO_4$ and $B = Fe_2SiO_4$; orthopyroxene, $A = MgSiO_3$ and $B = FeSiO_3$; clinopyroxene, $A = CaMgSi_2O_6$ and $B = CaFeSi_3O_8$; plagioclase, $A = CaAl_2Si_2O_8$ and $B = NaAlSi_3O_8$. For each mineral, fractionation densities have been calculated at $1200^{\circ}C$ (upper line) and at $1400^{\circ}C$ (lower line). The density range of common basaltic magmas is indicated, and the 2σ error bar is shown in the top left-hand corner for typical density estimates.

Figure 8 shows a schematic plot of density vs a convenient compositional parameter (Mg/Mg + Fe) for basaltic magmas related to one another by fractional crystallization. The general shape of the curve for density vs composition is considered characteristic of dry basaltic systems and can be interpreted with the aid of Figure 7. For high values of (Mg/Mg + Fe) a high-temperature primitive basalt (marked P in Figure 8) fractionates olivine and/or pyroxenes. The fractionation density of mafic minerals is greater than basaltic melt, and $\beta \Delta S / \alpha \Delta T \gg 1$. Consequently, the residual melts decrease in density despite the temperature decrease. Once plagioclase joins other mafic phases in cotectic assemblages, the fractionation density can often become less than the basalt melt density. For example, olivine gabbro assemblages have fractionation densities in the range 2.62-2.64 g cm⁻³ (Sparks & Huppert 1984). Thus, the density of residual melts can increase, causing a density minimum marked A in Figure 8. Although both temperature and compositional changes contribute to the increase in density, compositional effects are usually dominant. When other dense phases join in (for example, magnetite), or the ferromagnesian phases become very iron-rich, the fractionation density can become larger than the melt density, causing a decrease in density and a density maximum B.

Some important principles are illustrated in these diagrams. First, compositional effects usually dominate over thermal effects in determining melt density. The same statement will be true of other processes involving crystal-melt interaction, such as partial melting and contamination. Thus melts can decrease in density during fractionation despite thermal effects. Second, when new phases enter during fractionation, there can be important changes in slope, including density maxima and minima. Although only basaltic systems have so far been considered in detail by this approach, we feel that these principles will apply to all magmatic systems. They imply that compositional effects dominate thermal effects on density during major igneous processes and that both double-diffusive and compositional effects will be important in convection.



Figure 8 Schematic relationship between magma density and (Mg/Mg + Fe) for basaltic melts related to one another by fractional crystallization.

5. APPLICATIONS TO MAGMA GENESIS

Much of the recent interest in double-diffusive convection has focused on its role in the evolution of igneous rocks. In only a few years an extensive literature has developed. We briefly summarize the main ideas and applications that have emerged. Double-diffusive effects probably play a role in all major igneous processes.

Crystal Fractionation

There is now a general consensus among petrologists that the bulk of magmas are affected by crystal fractionation during their evolution. This process requires solid and liquid phases to be separated. The major mechanism for fractionation was thought until recently to be crystal settling (Bowen 1928, Wager & Brown 1967). There is, however, a good deal of evidence to suggest that crystal settling is an inadequate explanation. Some of the geological situations cited to support settling have been found suspect (see Campbell 1978, McBirney & Noyes 1979, Hildreth 1979). Theoretical arguments indicate that settling is opposed by convection in large magma chambers and that the fluid motions can keep crystals in suspension (Bartlett 1969, Sparks et al 1983).

Separation of liquid from crystals by compositional convection provides an alternative mechanism for fractionation (McBirney 1980). We have proposed the name *convective fractionation* for the process (Sparks et al 1984). This term [also used by Rice (1981)] embraces a wide variety of convective phenomena caused by crystallization, as already described from laboratory experiments on aqueous solutions. Below we summarize the main concepts and some speculations, which provide an alternative to settling. We do not have the space here to provide a detailed case for the new views, but instead we recommend that the reader consult the appropriate references.

In closed-system fractionation of magma, much of the crystallization will occur along the margins of the chamber, where the steepest temperature gradients and largest undercoolings occur and nuclei already exist (see Campbell 1978, McBirney & Noyes 1979). Crystals grown on the margins or in thin viscous boundary layers change the composition and therefore the density of the magma *locally*, and this melt can be convected away from its point of origin. The effects of this convection will depend strongly on the physical properties of the magma, on the fractionation density of the crystallizing phases, and on the chamber geometry (McBirney 1980, Sparks & Huppert 1984, Sparks et al 1984). In many circumstances, compositional and thermal gradients can be generated that are appropriate for the development of double-diffusive layering.

Some consequences and implications of this model for closed-system evolution of magmas are as follows:

1. Compositional and thermal gradients can be set up in magma chambers containing initially homogeneous magma. Gradients can be such that double-diffusive layering can develop. Eruption of parts of a chamber can produce volcanic rock sequences that are zoned and contain compositional discontinuities.

2. Once compositional gradients are established in a chamber, crystallization along the margins will lead to cumulates varying in composition with position. In mafic intrusions, rhythmic layering parallel to the margins will be discordant to phase layering parallel to gravitational stratification in the fluid (see Casey & Karson 1981, Wilson & Larsen 1982, Sparks et al 1984). Double-diffusive layering in the magma could also influence crystal layering in mafic intrusions (Kerr & Turner 1982, Irvine et al 1982).

3. Sidewall crystallization influences immediately only a small proportion of the total magma body, because selective removal of components occurs locally and affects only the magma in the thin compositional boundary layer. This can result in highly fractionated magmas accumulating at the top, or bottom, of a chamber without requiring large amounts of crystallization. Crystal-settling models often require large fractions of the magma chamber to crystallize in order to form highly differentiated melts (see Sparks et al 1984). Highly differentiated melts can form at the top of a chamber at an early stage by sidewall crystallization.

4. Once compositional gradients are established, double-diffusive layers can be formed, and each layer can subsequently evolve as a chemically independent system.

5. Compositional convection can occur in porous media. Tait et al (1984) have proposed that convection will play a major role in postcumulus crystallization. For example, they suggest that adcumulates can form when intercumulus crystallization produces light residual fluid that rises through the crystal pile and is continuously replaced by heavy magma from the overlying magma chamber.

6. The crystals that form cumulates and those that form phenocrysts in magmas may have different origins. Phenocrysts can form internally in a magma chamber at double-diffusive interfaces, where undercoolings and supersaturations are greatest. They can also form during the mixing of magmas (see below), by erosion of cumulates, and as residual crystals from the source region (restite). Once crystals of diverse origins are suspended in the magma, further growth can occur. [Sparks et al (1984) provide a fuller discussion of the origins of phenocrysts.] Phenocrysts, therefore, do not

necessarily provide accurate information on the proportions and compositions of fractionating phases.

Magma-Chamber Replenishment

There is now a great deal of evidence to support the idea that many magma chambers are open systems that are periodically or continuously replenished by new magma from depth. This is now perceived by many petrologists as important in controlling many petrological and geochemical features of igneous rocks (see, for example, Walker et al 1979, Eichelberger 1980, Huppert & Sparks 1980b, O'Hara & Matthews 1981, Sakuyama 1981). Double-diffusive phenomena are certain to occur during replenishment, because the compositions, temperatures, and densities of the new magma and of the magma already resident in the chamber are generally different. These compositional differences arise because the resident magma is more differentiated than the new magma.

A number of interesting convective processes due to crystallization and double-diffusive effects can occur during magma-chamber replenishment. Two fundamental situations can be considered, depending on the sign of the density difference between the new magma and the resident magma. In nearly all cases the incoming magma will have a higher temperature than the resident magma. The density contrast will, however, generally be attributable to the compositional difference between the magmas.

When the new magma is denser than the resident magma, the new magma will pond at the base of the chamber and form a layer with a doublediffusive interface beneath the overlying magma. This situation has been analyzed in some detail for the case of picritic magma emplaced into a basaltic magma chamber (Sparks et al 1980, Smewing 1981, Huppert & Sparks 1980a,b). The important features of the analysis are that heat is exchanged across the double-diffusive interface and that crystallization occurs in the basal magma layer. When the density of the residual melt has become equal to the density of the overlying magma, rapid mixing will occur between the magmas. The basic features of the model were demonstrated in laboratory experiments (Huppert & Turner 1981b, Huppert et al 1982b).

Input of dense mafic magma from the mantle is undoubtedly a fundamental feature of many magmatic plumbing systems. Overturn will not necessarily occur, particularly if the resident magma has a low density; for example, if basalt is emplaced beneath rhyolite, overturn cannot take place unless some other factor such as exsolution of abundant volatiles in the basaltic magma occurs (Eichelberger 1980, Huppert et al 1982a). Rice (1981) has postulated that when basalt is emplaced beneath rhyolite, layering develops at the interface and eventually overturn occurs. There is no experimental or theoretical justification for this model. Overturn only happens when the densities of adjacent magma layers become equal, and it is not clear how this is accomplished in Rice's model.

When the new magma is lighter than the resident magma, mixing can take place immediately. This situation can arise in the important case of replenishment of basaltic magma chambers with primitive magma, which is lighter than highly fractionated basaltic magma (Sparks et al 1980). The light magma will rise to form a turbulent plume, provided the Reynolds number is sufficiently large, resulting in mixing between the magmas. The hybrid magma will then rise to the top of the chamber to produce compositional stratification. Alternatively, if the Reynolds number is small, the new magma will rise without significant mixing to form a layer at the top of the chamber.

From the foregoing discussion it is evident that both compositional stratification and magma mixing can result from replenishment. Repeated replenishment can cause compositional stratification and thermal gradients suitable for double-diffusive convection. At the same time, crystallization can cause magma layers to change and mix with one another. These ideas have been used to explain the compositional uniformity of mid-ocean ridge basalts (Huppert & Sparks 1980a), the origin of cyclic layering in ultramafic intrusions (Huppert & Sparks 1980b), and the origin of chemical diversity in composite intrusions (Marshall & Sparks 1983). They have also been used to interpret mixing phenomena in calcalkaline volcanic rocks (Eichelberger 1980, Huppert et al 1982a).

Compositional Zoning

The origin of compositional zonation in magma chambers has attracted considerable attention (Hildreth 1979, 1981). We have already indicated that crystallization along the margins of a chamber and replenishment are two mechanisms for generating zonation. Two other mechanisms could also be significant in nature. First, fractional partial melting can generate a succession of magmas of increasing density and temperature. If successive melts are fed into a chamber, zonation could result. Second, contamination of the magma along the walls of a chamber can produce zonation. For example, melting of the granitic walls can generate boundary-layer flows of low-density rhyolite that can rise to the top of the chamber. If the melting involves assimilation at the margins, the contaminated magma can differ in density from the uncontaminated magma and lead to compositional convection and stratification.

Much has been made in particular of the striking trace-element gradients to be found in some high-silica rhyolite magma chambers (Hildreth 1979,

1981). Diffusive effects have been invoked to explain these gradients, and a rather complicated model, termed *convection-driven thermogravitational diffusion*, has been invoked to explain these gradients (Shaw et al 1976, Hildreth 1981). The model involves ascent of water-rich boundary layers along the sides of the chamber to form a stratified region at the top. The compositional gradients are then considered to originate by Soret diffusion due to the temperature gradient imposed across the stratified region and to the volatile fluxing from beneath. The boundary layers are considered to form by diffusion of water into the magma from outside.

Although Soret diffusion should not be dismissed as a differentiation mechanism (Walker & De Long 1982, Schott 1983), there is no clear evidence to support its operation on a large scale. While it may be that Soret effects could become significant in a stratified magma chamber containing convecting double-diffusive layers (Schott 1983), experiments on Soret differentiation in rhyolitic melts produced gradients opposite in sign to those observed in nature (Lesher et al 1982). We suggest that fractional crystallization by sidewall accretion and convection coupled with volatile fluxing is a simpler explanation for the gradients in high-silica rhyolites. The principal objection to the role of crystal fractionation (Hildreth 1981) is that large amounts of crystallization are required to generate the rhyolitic magmas with extreme trace-element concentrations. This problem does not arise in a sidewall crystallization model, as highly fractionated fluids can be generated in the boundary-layer flows. A fuller discussion of these issues can be found in the articles by Hildreth (1981), Michael (1983), Schott (1983), and Sparks et al (1984).

Magmatic Ore Deposits

The formation of some types of magmatic ore deposits could be influenced by double-diffusive effects. Chromitites appear to form during replenishment of magma chambers, and mixing processes can be invoked to account for their formation (Irvine 1977, Huppert & Sparks 1980b, Sparks & Huppert 1984). Campbell et al (1983) have suggested that platinum sulfide horizons in layered intrusions are generated when primitive magma is emplaced into a magma chamber. Mixing of large volumes of the resident magma with the new magma leads to the precipitation of platinum sulfides.

In silicic systems, highly differentiated magma, enriched in volatiles and incompatible elements, can accumulate at the top of a chamber by sidewall crystallization. The evolution of a vapor phase from such highly evolved magma could lead to mineralization events. Tin mineralization in the roof zones of some granitoids (see, for example, Groves & McCarthy 1978) might provide an example of this process. A new aspect of the mineralization envisaged here is that "late-stage" fluids may be formed early in the evolution of some magma chambers. Mineralization of the roof zones could occur from an evolved stratified region at the top of the chamber while a substantial fraction of the chamber still contains undifferentiated magma.

6. OTHER GEOPHYSICAL APPLICATIONS

There are other important examples of natural multicomponent fluids in which double-diffusive effects can be anticipated. The solidification of the inner core of the Earth from the molten outer core involves growth of metallic iron-nickel crystals and rejection of light components such as silicon. It has been argued that compositional convection resulting from crystallization will be much more important than thermal convection in driving fluid motions in the outer core and generating the Earth's magnetic field (Gubbins et al 1979, Loper & Roberts 1983).

Many hydrothermal fluids cool and crystallize as they ascend to the Earth's surface, and double-diffusive effects should be expected. The discharge of hot brines onto the seafloor can be cited as one situation where double-diffusive phenomena occur (Turner & Gustafson 1978). Pore fluids in sedimentary rocks vary in both composition and temperature and could develop gradients appropriate for double-diffusive convection. Evaporites also involve crystallization from complex aqueous solutions, and doublediffusive phenomena should occur.

Perhaps the largest-scale example of double-diffusive convection is to be found in the dynamic behavior of the Earth's mantle. Richter (1979) has suggested that layering in the mantle may have originated by the heating of a chemical gradient from below. Compositional variations occur in the oceanic lithosphere, and when subduction occurs, cold dense parts of the lithosphere can become lighter than the surrounding mantle when they are heated (see Ringwood 1983). Partial melting could also play a role analogous to crystallization in magmas, because large-density changes occur when melting takes place.

Many of the early developments in the study of double-diffusive convection were motivated by oceanographical considerations. There is a large primary literature and a number of reviews devoted to this application, many of which are cited in Huppert & Turner (1981a). It would upset the balance of this article to discuss the oceanographic situation in detail. It suffices to say that double-diffusive effects of both the diffusive type (Figure 2c) and the fingering type (Figure 2d) are known to play a large role in many different parts of the world's oceans. The influence is felt over sizes ranging from the small scale of naturally occurring salt-fingers to the scale of fronts. In all cases, double-diffusive effects are undoubtedly important, but accurate quantitative determinations of the extent to which these phenomena affect oceanic mixing are not yet available. Once known, the influence of such mixing will have to be assessed in comparison with internal wave breaking, turbulence, and other effects.

7. CONCLUSIONS

Multicomponent fluid systems have the potential for developing doublediffusive convection. In the case of magmas and probably many other geophysically significant fluids, changes in chemical composition have much greater effects on density than associated temperature changes. In addition, natural fluids cool as they approach the Earth's surface. Thus, unstable temperature gradients and stable compositional gradients should be common. Crystallization of multicomponent fluids also leads to a variety of important convection effects. In particular, stable compositional gradients can often be formed in initially homogeneous fluid systems. Experimental and theoretical evidence suggests that double-diffusive convection should be important in magmas. The application of these principles to a number of geological situations has already led to new insights into the origin of igneous rocks.

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