

The production of chemically stratified and adcumulate plutonic igneous rocks

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Abstract

We review recent advances on the physical principles of crystallization in multicomponent systems, and use them to provide a framework for interpreting petrological and geochemical observations from igneous intrusions. The thermal structure of crystallizing boundary layers imposes strong constraints on the chemical and mineralogical compositions of the solid that can form from a given melt. The thermal problem is largely independent of the chemical composition of the melt, and sets the course of crystallization. A key problem to understand is the temperature of the solidification front (which we take to mean that point at which the last drop of liquid solidifies) particularly in the geologically relevant case in which the temperature at the cold boundary is below the eutectic temperature. Focussing on the solidification front rather than on the liquidus is a valuable perspective. Adcumulus growth requires specific conditions and much can be learned from trying to understand how these can develop from given starting conditions. We discuss the physical reasons and field evidence for the existence of mushy layers, where solid fraction and temperature vary by large amounts. In such regions of the magma chamber, thermodynamic equilibrium is nearly achieved locally and, for a given temperature, this specifies the composition of the interstitial melt. Thus, in a magma chamber, the whole liquid line of descent is present simultaneously. Compositional convection is likely to set in, and this exchange between the interior of the mushy layer and the main reservoir leads to a chemically stratified solid, and to adcumulus growth. The contribution of crystal settling to the floor cumulates is evaluated as a function of the magnitude of convective heat flux through the roof. It is shown that crystal settling is unlikely to overwhelm *in-situ* nucleation and growth at the floor.

KEYWORDS: Multi-component systems, crystallizing boundary layer, adcumulus growth.

Introduction

MANY large igneous intrusions exhibit a systematic stratigraphic progression of minerals from the floor upwards. Such a large-scale trend indicates that the igneous system, of which the presently exposed intrusion might only be a small part, was able to differentiate and produce a large volume of chemically evolved rocks. Determining the processes that led to such differentiation has been very difficult for two types of reasons. Firstly, magma chambers are unlikely to be closed to reinjection of new liquid, the volume, composition and temperature of which is hard to constrain (e.g. Brown, 1956; Irvine and Smith, 1967; Huppert and Sparks, 1980; Irvine and Sharpe, 1986). This introduces a potentially large degree of freedom into the problem. Secondly, the number of possible processes, even in a closed system, is embarrassing large, for example crystal

settling (Bowen, 1928), compositional convection (Hess, 1972; Morse, 1982; Sparks *et al.*, 1984; Tait *et al.*, 1984) and diffusive exchange ahead of a moving crystallization front (Cawthorn and McCarthy, 1980), among others. Each process requires specific conditions which may or may not be achieved in a real situation. Each should lead to a specific differentiation trend, but it is not easy to predict exactly the form that this will take as the phase equilibria of natural systems are very complex. It is also unlikely that these processes are mutually exclusive. Thus, for example, when a given closed-system model appears inconsistent with one observation, it is tempting to call for a new injection of liquid with the appropriate composition. In order that this procedure not be *ad-hoc*, we require considerable confidence in the predictions of a given model. Faced with such formidable difficulties, petrologists have multiplied the database, which now typically

includes major elements, trace elements and several isotopic systems of both minerals and whole-rocks. These different kinds of data do not have the same sensitivity to the different processes which operate in magma chambers. Major elements give the crystal record and define the solidus, whereas trace elements are more sensitive to the final interstitial liquid composition (Wager, 1963; Cawthorn and McCarthy, 1985). Isotopic systems are sensitive to contamination by crustal rocks. This wealth of data makes the testing of physical models difficult, as these are often too simply designed to simultaneously address all these various constraints.

Most petrological models have striven to explain the cumulate sequence using purely thermodynamic and chemical constraints often without considering explicitly the thermal aspects of the problem. The latter provides not only the driving mechanism for crystallization but also limits the number of mechanisms that may operate. Crystallization is achieved by cooling, and hence, ultimately, it is the thermal problem which determines how, where and how quickly crystals accumulate. A fruitful line of research has been the study of fluid dynamical phenomena which are caused by density changes due to cooling and differentiation. Such models have complex physics but, as yet, have only been quantitatively applied to chemically simple systems. Thus, they are unable to reproduce the mineral sequence and trace element compositions. However, they are able to outline large-scale chemical trends and make predictions about where in the cumulate sequence effects should be observed. The main interest of these models is that they introduce the thermal aspect of the problem. This ensures consistency with the cooling mechanisms and hence the cooling rates that may be achieved in geological situations.

In this paper, we evaluate two different mechanisms for igneous differentiation: *in-situ* growth in a thermal boundary layer and settling of crystals from above. This paper is not a general review, and, rather than attempting to discuss the many regimes that have been investigated, we concentrate on situations that are likely to develop from given starting conditions. We review evidence from the rock record with emphasis on adcumulate textures. We explain some recent general results on *in-situ* fractional crystallization in dynamic conditions. Three points are emphasized: the conditions required to produce a compositionally stratified solid, those required to produce adcumulate rocks, and consistency with the thermal budget. We evaluate the rates of the various processes and the key aspect of their stability, which in a system with multiple degrees of freedom will ultimately determine which one is likely to dominate over the others.

The cumulate sequence: inferences from observations

The floor cumulate sequence is usually much thicker than the roof sequence. An extreme case is that of the Bushveld intrusion where the roof sequence is missing altogether, and may well not have existed at any stage. This observation indicates that crystals accumulate mostly at the floor of an intrusion, and that a thick thermal boundary layer is prevented from growing at the top. Crystals that ultimately are found in the floor sequence can form *in-situ* in the boundary layers or in the interior. Important issues concerning how differentiation takes place are the thicknesses of the partially crystallized boundary layers and the extent of compositional gradients both in the chamber interior and through the boundary layers. In this section, we review field observations which bear on these issues.

Cumulate textures and crystal settling

Cumulate textures in the rocks of many intrusions have been likened to those of sedimentary rocks, and have been attributed to crystal settling from a large body of liquid. This creates a difficulty with respect to testing models concerned with large-scale magmatic differentiation as these have not been devised to predict specific aspects of rock texture. If we accept that the texture implies vertical sedimentation, nothing in the texture can tell us whether or not a grain has settled through a layer of liquid 1 km or 1 m thick. Furthermore, the textural descriptions of the Skaergaard Upper Border Group (i.e. the roof sequence) also identify cumulus texture with primocrysts and intercumulus material (Wager and Brown, 1968). This clearly implies that rocks with cumulate textures involving the same minerals were forming simultaneously at both the floor and the roof which is not possible by crystal settling. In this case, primocrysts were thought of as caught in the upper solidification front (see also McBirney and Noyes, 1979). In large magmatic intrusions, textural re-equilibration is likely to occur, which complicates the interpretation (e.g. Hunter, 1987). The idea of differentiation by crystal settling, i.e. a sedimentary process, may be supported by structures such as scours, slumps and graded bedding. However, such structures require the action of currents related to an inclined margin and, indeed, they are most often found near the side walls of intrusions, for example the trough bands of the Skaergaard.

Cumulus textures appear to occur once an intrusion is greater than a certain size, approximately of the order of 1 km in its smallest (generally vertical) dimension. Chemical and mineralogical differentiation of rocks occur, however, in smaller

bodies. The Palisades Sill, for example, shows marked chemical differentiation, though the rocks do not show well-developed cumulate textures anywhere. We conclude that cumulate textures do not necessarily imply that crystals settle from a large reservoir and that they are not a prerequisite for igneous differentiation.

Adcumulus growth

Adcumulate rocks are evidence that interstitial liquid solidifies *in-situ* whilst chemical exchange with the overlying reservoir is taking place, and that crystallization occurs with little decrease in temperature. Wager and colleagues interpreted adcumulates to mean that sedimentation was slow relative to solidification of the interstitial liquid. If fast chemical diffusion and slow accumulation were in fact to be the principal ingredients required for the formation of adcumulates, then the roof rocks should be most adcumulate. The accumulation rate is lower there, and there is no reason why chemical diffusion should be less efficient at the roof than at the floor. In fact the rocks of the Skaergaard Upper Border Group are described as orthocumulates (Wager and Brown, 1968). This point was appreciated by Hess in his Stillwater memoir (1960).

Adcumulates have conventionally been interpreted as implying a very thin partially crystallized layer — to all intents and purposes a flat interface. In this conceptual model there cannot be a large depth of material with interstitial melt because, in such conditions, chemical diffusion, the proposed agent bringing in the components required for adcumulus growth, would be inefficient. In the case of extreme adcumulates, interstitial melt remains only in very small quantities when chemical exchange with the overlying reservoir ceases, which presumably occurs at some depth in the cumulate pile. These are critical deductions which have received no satisfactory physical explanation. Wager (1963) gave a qualitative discussion of the mechanism by which heat might be removed from the cumulate pile during adcumulus growth at the floor. Recognizing the difficulty involved, he reached the conclusion that the melt in the chamber must be supercooled. Even this, however, does not resolve the apparent contradiction that orthocumulates form at the roof.

As this deduction of a flat interface is central to the conventional wisdom of igneous differentiation, attempts have been made to support it with field observations. For example, the dimensions of scour and slump structures have been used to constrain the thickness of unconsolidated material at the margins. Estimates are generally on the order of a few metres. However, the mechanical erosion of an unconsolidated layer will be restricted to its upper part, and

hence such estimates should be viewed as minimum bounds.

Conclusion: compatibility with cooling conditions

The theories concerning adcumulates and their formation at a flat interface imply a specific heat budget. In this model, temperature at any given time is essentially uniform, the difference between the temperature of the solidification front and that at which the cumulus phase nucleates being considered negligible. Thus, one essentially follows the solidification front by looking at the temperature and composition of the melt layer. Little attention is paid to the temperature profile in the solid below the front, and to whether or not this situation may in fact be achieved from given starting conditions. This point is highlighted by the observation that the rocks of the Skaergaard vary continuously from predominantly orthocumulates in the lower part of the Layered Series to predominantly adcumulates in the upper part (Wager and Brown, 1968). We may ask how the system adapts its thermal budget in order for the rocks formed to progressively change in this way. Thermodynamic constraints have often been used to infer what is occurring in the solid using information pertaining to the liquid part of the chamber. In contrast, cooling proceeds from the solid into the liquid, with a thermal boundary layer growing from the cold margins. We shall see that present models are able to reconcile both aspects, at the expense of a revision of the physical framework.

Crystallization in a thermal boundary layer

As far as the floor cumulate sequence is concerned, there are different possible origins for the crystals. Crystallization initially proceeds by local cooling because heat is extracted rapidly through the colder country rock. Field evidence for this includes a chilled margin and the fact that the country rock is a hornfels (eg., Page, 1979). Somewhat later, the effects of thermal convection can be felt at the floor. This convection is generated from a cold thermal boundary layer at the upper contact, transports cooled magma away from the roof, and crystals may be nucleated and grown in the reservoir interior. These crystals settle at the bottom, even in the presence of convection, because convective velocities vanish at the base of the chamber and hence cannot maintain crystals in suspension (Martin and Nokes, 1988). A final possibility is that crystals grow from undercooled melt coming from the roof or side-walls.

Although *in-situ* growth dominates initially at the floor, it decreases continuously as the cold boundary layer thickens, and after some time, other

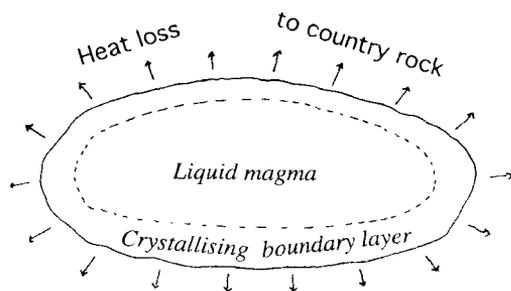


FIG. 1. Schematic diagram to emphasize how cooling proceeds in a magma chamber. The interior is, at all times, at the highest temperature. Cold boundary layers develop at the margins where steep gradients of temperature and melt composition exist.

mechanisms can contribute more crystals and even come to dominate the crystallization budget. One must therefore determine when the crossover occurs. The problem is complicated further by several effects that can be caused by compositional convection generated in the floor cumulate pile (Tait and Jaupart, 1992), and that change the evolution of both the floor sequence and reservoir interior. Compositional convection is associated with large density differences and issues from the very floor of the reservoir. Thus, the floor can no longer be treated as a solid boundary where the melt velocity drops to zero, which is specifically assumed in the settling model. Crystals could in fact be kept in suspension in the chamber interior by the strong upward currents. Another consequence is that the melt layer changes composition, which affects how crystallization proceeds at the roof, modifies the heat budget of the reservoir and could suppress crystallization in the chamber interior.

We now turn our attention to the *in-situ* crystallization regime. This problem may be well posed and is the key to our understanding of all processes in a magma chamber, in the sense that a solution of this problem implies an understanding of boundary layer structure. Cooling is effected at the margins, in boundary layers which may or may not become unstable. In a closed system, all magma motions are due to instabilities generated in them. We return to the thermal budget of a crystal settling regime later, and assess how this may interact with *in-situ* growth.

We summarize recent developments on how crystallization proceeds in a boundary layer, with emphasis on three specific points. One is that we must identify mechanisms that are able to generate a chemically stratified solid. The second point is that such mechanisms must be compatible with how

cooling proceeds. We shall see that this is a strong constraint on the compositional budget of crystallization, and that thermal and compositional effects are finely tuned. The third point is that one must pay attention to the contact temperature. A key fact is that the country rock contact is initially below the solidus of the magma. This leads to a chilled margin, which is commonly observed at the basal contact of many intrusions. Thus, there is a solid layer forming at the contacts of the intrusion, and the temperature profile which develops through the melt body spans the whole crystallization interval (Fig. 1). We focus on the temperature and average composition of the solidification front, which we take to be the point at which the last drop of liquid solidifies. We build this discussion by describing a series of crystallization models which are increasingly complex and progressively closer to geological reality. In each case, we first examine static conditions and evaluate the possibility of convection.

A flat solidification front

Consider a large flat crystal face growing at a constant growth rate into a liquid with trace-element partitioning between the phases. This situation has the merit of posing an initial problem in a simple and clear manner. It has been used by Cawthorn and McCarthy (1980, 1985) to discuss elemental profiles in the Bushveld complex. Starting with a liquid at composition C_0 , and taking a partition coefficient k , the initial solid which forms has composition kC_0 . This sets up a composition gradient ahead of the growing crystal/liquid interface (Fig. 2). With time, chemical diffusion acts to change the liquid composition at the crystal face, which, in turn, acts to change the solid composition. This leads to a compositionally zoned solid. The system evolves to a steady-state, such that the solid has the same composition as the far field liquid (Smith *et al.*, 1955). The solid thickness required to achieve this steady-state is determined by the time it takes to develop the steady-state diffusion profile in the melt ahead of the solidification front.

This model is geologically unrealistic by virtue of the assumptions of a constant growth rate and of a constant partition coefficient. It has value however, as it serves to demonstrate that by specifying the growth rate, in this particular case specifying that it remains constant with time, one actually specifies the composition of the solid grown. Furthermore, this model identifies two mechanisms for generating a chemically stratified solid: a change of compositional flux at the solid/liquid interface, and a change of growth rate. A general expectation is that the growth rate decreases with time following the injection of a magma batch.

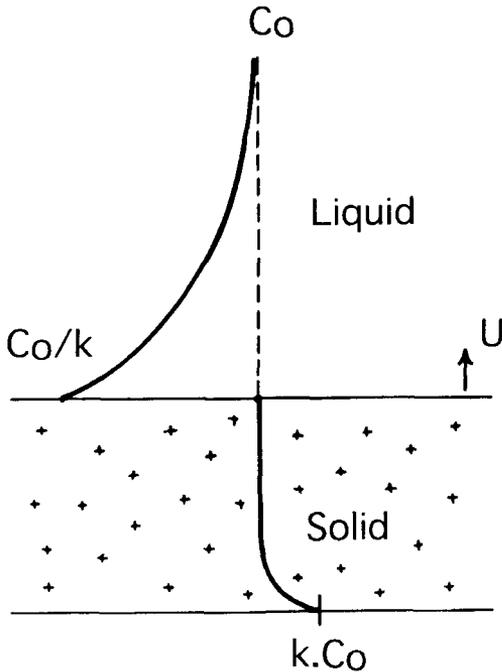


FIG. 2. Diagram showing the composition profile in both liquid and solid for a flat interface growing at constant velocity and with trace-element partitioning (Smith *et al.*, 1955). At steady-state, the solid grown has the same composition as the liquid in the far field outside the diffusive boundary layer that exists ahead of the solidification front.

We now turn to more realistic models, which all share the same features, namely that the composition of the solid grown cannot be specified *a priori* and must be solved for using chemical balance equations, and further that this composition is a function of growth rate (or accumulation rate).

Multicomponent melts

More realistic models have dealt with a binary solution with or without a eutectic (Roberts and Loper, 1983; Worster, 1983; Huppert and Worster, 1985; Worster, 1986). To solve the problem, one needs to specify the solid/liquid relationship. Solutions will be described, (a) in which all solidification takes place at one temperature, and (b) in which it takes place over a range of temperature. In the first case, a chemical boundary layer is present ahead of the solidification front, and in the second, a range of compositions is present in

the partially-solidified region. In neither case, therefore, is the whole solid forming in equilibrium with the whole body of liquid. The simplest discussion is again for the case of a constant growth rate.

It is instructive to observe conditions at the contact and then to proceed upwards into the thermal boundary layer. Right at the contact, a chill phase can form which has the composition of the liquid. This chill can, in certain circumstances, be reheated by latent heat released by the crystallization of melt (Brandeis *et al.*, 1984). At the solidification front, by definition, solid and liquid are coexisting and may plausibly be assumed to be at or close to thermodynamic equilibrium at least after some short initial phase. For a given chemical system, the lowest temperature at which liquid and solid can coexist is the eutectic temperature. One may set up the problem of having no solid phase grown away from the eutectic front, in other words with no crystallization occurring above the eutectic temperature such that the interface is essentially flat. At the eutectic temperature, the constraint of thermodynamic equilibrium imposes that the composition of both liquid and solid at the interface is eutectic. Chemical diffusion through the solid could, in principle modify its composition, but this is weak and hence the final solid is predicted to have eutectic composition. This situation is not likely to occur in geological conditions, but it is instructive to note that neither is it self-consistent for fundamental reasons. One is that the growth of a eutectic solid requires several phases to grow simultaneously, which implies that the solid/liquid interface has a complex local morphology and hence cannot be perfectly flat (Elliott, 1977). Another reason is that the chemical budget of crystallization is fixed, and hence requires a specific growth rate (Elliott, 1977; Worster, 1983; Woods and Huppert, 1989).

In reality, the solid/liquid interface is not perfectly flat. At a small distance from the completely solidified layer, the temperature is slightly above the eutectic value, which allows the growth of the high melting point phase. A solution thus requires an assumption about the detailed morphology of the interface. One approach that leads to a consistent solution is to average small scale details and assume that the interface is almost flat, with the solid having an average composition between the eutectic and the higher melting point component (Fig. 3). This average composition, although consistent with thermodynamics, is not set by the constraint of equilibrium and, instead, is solved for using chemical balance equations. There is a steady-state solution at constant growth rate, with a solid composition which differs from that of the initial melt (Elliott, 1977; Worster, 1983). For our present purposes, this implies that, save for a short transient analogous to

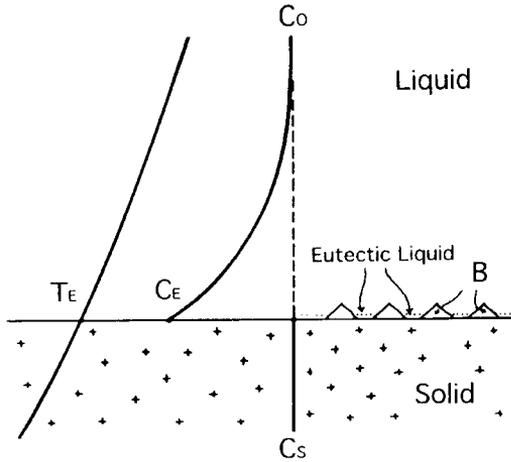


FIG. 3. Diagram showing the details of the solid/liquid interface for a binary eutectic system which is being cooled below the eutectic temperature. The interface cannot be perfectly flat. The solid grows with an average composition, that is enriched in the high melting point phase B with respect to the eutectic, and that must be solved for using mass balance equations. The thermal and chemical boundary layers are such that liquid just above the solidification front is undercooled.

that for the previous model, the solid grown is again homogeneous.

Cooling conditions

The crystallization models considered so far rely on the assumption of constant growth rate. In a realistic model, the growth rate must be solved for. This has been done in the specific case of a uniform initial temperature in the melt layer and a fixed boundary temperature. Solutions may be found for a basal temperature below or above the eutectic in static conditions, taking into account both chemical diffusion in the liquid and heat diffusion (Roberts and Loper, 1983; Worster, 1983; Huppert and Worster, 1985; Worster, 1986).

For a boundary temperature below the eutectic, it is possible to find a growth regime with a (rough) solid/liquid interface at the eutectic temperature, and whose position varies as the square root of time (Worster, 1983, 1986). What is remarkable is that the development of the diffusion profile, ahead of the solidification front, does not lead to transient conditions at the interface as in the two previous models, and conditions at the solidification front and in the solid remain steady. An homogeneous solid is again predicted. This is because the advance of the

solidification front is determined by the rate of heat diffusion which is proportional to that of chemical diffusion. A stratified solid may be produced in two different ways; by changing the growth rate with time in a way inconsistent with pure diffusive growth, or by changing the melt composition. Such effects could be due, for example, to non-equilibrium latent heat release (Brandeis *et al.*, 1984), or to some form of convection occurring.

Ahead of the solidification front, there are both a temperature gradient and a chemical gradient in the melt (Fig. 3), and two different kinds of instability may arise in this case. The first one is the so-called morphological instability, related to the geometry of the interface. Adding the phase diagram constraint, one can see that the melt can become undercooled, a situation referred to as "constitutional supercooling" (Mullins and Sekerka, 1964). This unstable situation occurs except at small growth rates. Instability takes the form of protuberances growing into the supercooled melt region. Thus, crystallization becomes distributed throughout some temperature interval, with the crystal fraction varying from 1 at the solidification front to 0 at the liquidus (Fig. 4). We distinguish between such a thick "mushy" layer, that spans the whole crystallization interval in terms of temperature, and a morphologically unstable interface that might just be considered rough and approximately isothermal. Another instability is of dynamical origin and related to the large composition gradient in the liquid, which for most systems will be associated with a density increase away from the solidification front. For the floor of an intrusion, this implies a gravitationally unstable situation, which leads to convection. According to Woods and Huppert (1989), some chemical systems evolve, with convection generated at an interface which is rough but remains essentially flat in the sense considered above and at the eutectic temperature. In this situation, convection prevents crystal growth at temperatures far above the eutectic. The compositional difference across the unstable boundary layer is equal to the interior composition minus the eutectic composition, which is large. This leads to a high compositional flux and to a stratified solid. This convective growth regime does not exist, however, if a thick mushy layer forms.

Mushy layers

Structure of a magmatic mushy layer: observational evidence

When the boundary temperature is below the eutectic temperature, the mushy layer spans the whole temperature interval between the liquidus and solidus. To specify the composition of the crystals,

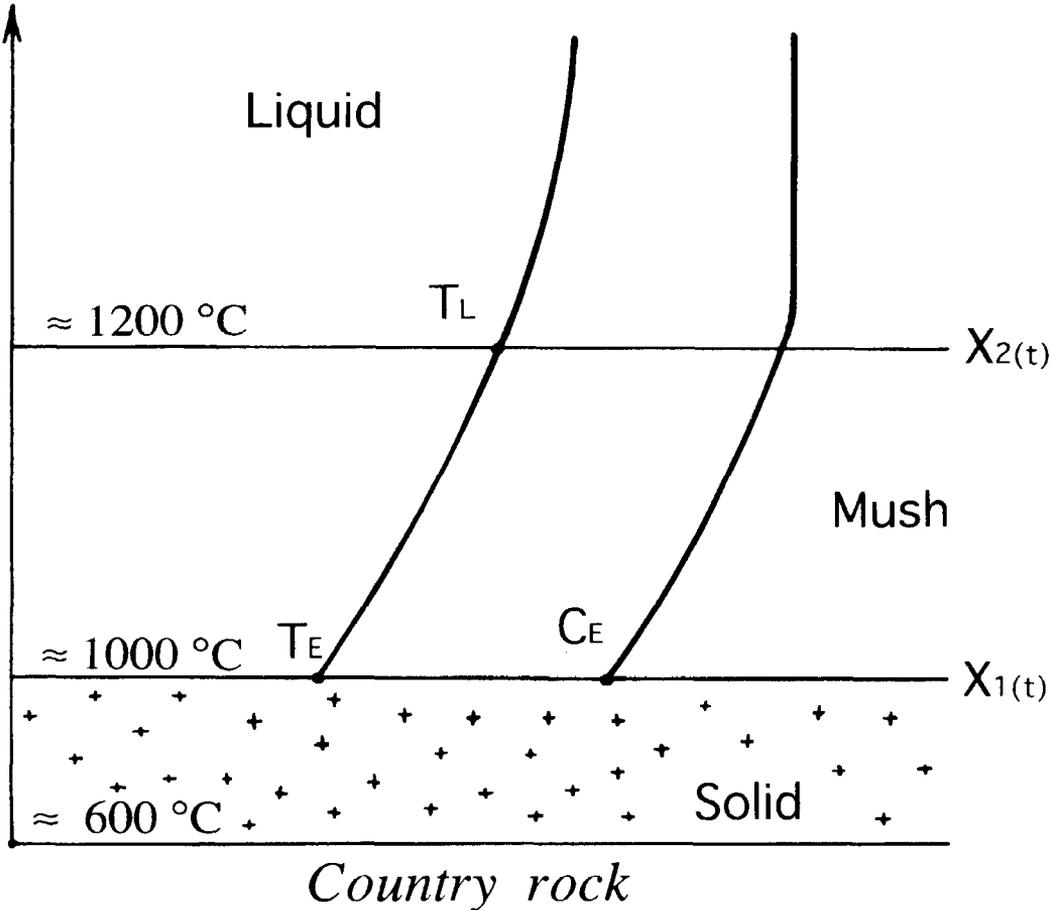


FIG. 4. Diagram showing temperature and composition profiles for a eutectic system cooled below the eutectic temperature when the flat interface is not stable and a mushy layer forms. The partially crystallized layer spans the whole crystallization interval between the liquidus and eutectic temperatures. In this layer, the interstitial liquid composition may be specified as a function of temperature using the constraint of local equilibrium. Approximate values of temperatures that might be expected in the case of a basaltic melt are indicated.

the simplest model is that of local equilibrium with the interstitial liquid at all points in the mush. This has been verified in laboratory experiments with various aqueous solutions both in conductive situations and in the presence of convective motions in the liquid. The whole liquid line of descent is represented in the mushy layer, which has far-ranging implications. A very important step is therefore to verify the validity of this model in a magmatic situation.

The successive drilling of the lava lakes of Kilauea Iki, Alae and Makaopuhi on the island of Hawaii provide access to the structure of a crystallizing boundary layer in a natural magmatic system (Richter

and Moore, 1966; Wright *et al.*, 1976; Helz, 1980; Helz and Thorber, 1987; Helz, 1987; Helz *et al.*, 1989). The observations are mainly concerned with the upper boundary layer. In these lava lakes, the proportions of mineral phases and interstitial liquid (recovered as glass) vary continuously through the upper boundary layers at any given instant in time. When it was studied, the thickness of the partially solidified boundary layer of Kilauea Iki, for example, was of the order of 10 m. The liquids of the three basaltic magmas that filled the different lava lakes varied somewhat, in all cases being above 1200°C, and the solids are all given as being approximately 980°C. Temperature and interstitial glass

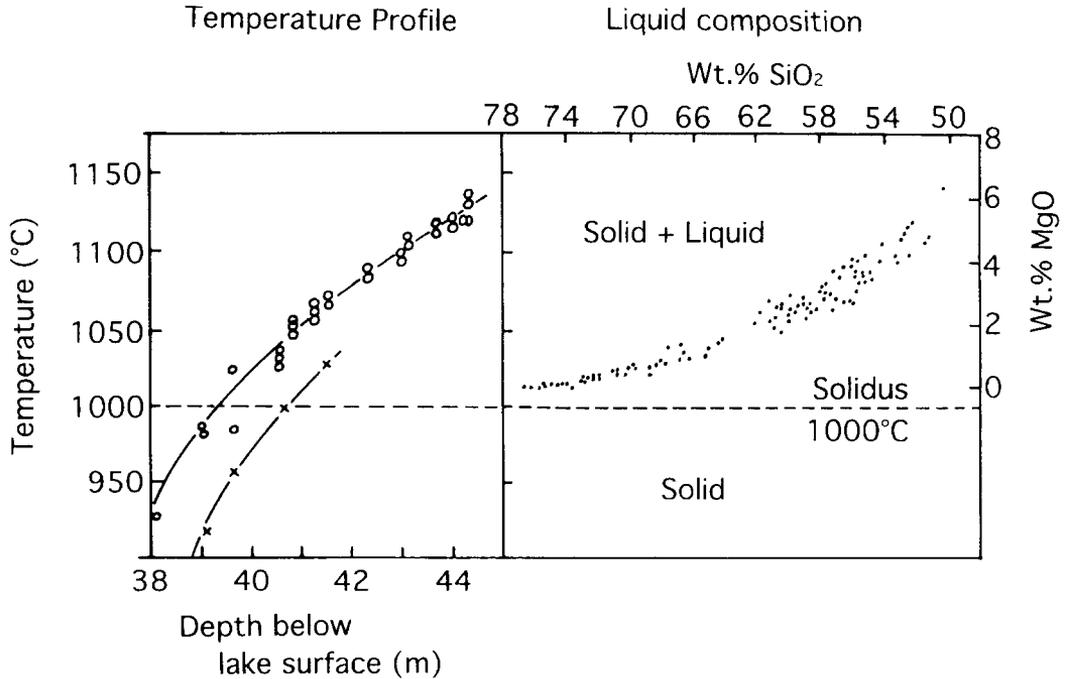


Fig. 5. Plots of temperature and interstitial melt composition in the upper boundary layer of Kilauea Iki basaltic lava lake (redrawn from Helz, 1980, 1987). Open circles represent temperatures calculated for the composition of interstitial melt and assuming local equilibrium. Crosses are thermocouple measurements.

compositions varied smoothly through the boundary layer at any given time (Fig. 5). The initial erupted magmas were olivine tholeiites with various proportions of olivine phenocrysts. The most evolved interstitial glasses analysed are reported by Helz (1987) as rhyolites containing 75–76 wt.% SiO_2 , and the electron microprobe data on these interstitial glasses clearly indicate that compositions representing the full liquid line of descent were present at any one time as a function of depth in the boundary layer (Fig. 5). Some relatively evolved liquid compositions (andesites with approximately 3–5 wt.% MgO) apparently had a tendency to collect in sub-horizontal bodies referred to as segregation veins.

Helz and Thornber (1987) carried out experiments on samples of Kilauean basalt in order to determine the equilibrium assemblages that coexist with residual melt for the case of the Kilauea Iki lava lake. In their experiments, they observed the same mineral assemblages as a function of temperature as were found in the Kilauea Iki samples. They also calibrated glass composition (in particular mass concentrations of CaO and MgO) as a geothermometer accurate to $\pm 10^\circ\text{C}$. Given the thermal and compositional stratification of the interstitial liquid,

one may deduce that it was stably stratified over at least most of the thickness of the mushy upper boundary layer. The corollary is that the interstitial liquid in a mushy lower boundary layer would be unstably stratified. At least some of the liquids towards the more evolved end of the compositional range observed are interpreted by Helz *et al.* (1989) as having risen buoyantly from the lower crystallizing boundary layer to be emplaced in the upper boundary layer. For the current purposes, the crucial point is that in the upper boundary layer of basaltic lava lakes the mineral assemblages and coexisting interstitial liquids spanning the whole interval between the solidus and the liquidus were simultaneously present in a mushy layer. Furthermore, experimental results confirm that to a good approximation local equilibrium was indeed established in this region.

The onset of compositional convection

The density gradient in the liquid at the floor that results from the combined effects of temperature and composition (Fig. 6) is unstable with respect to convective motions once the partially crystallized boundary layer has reached a certain critical

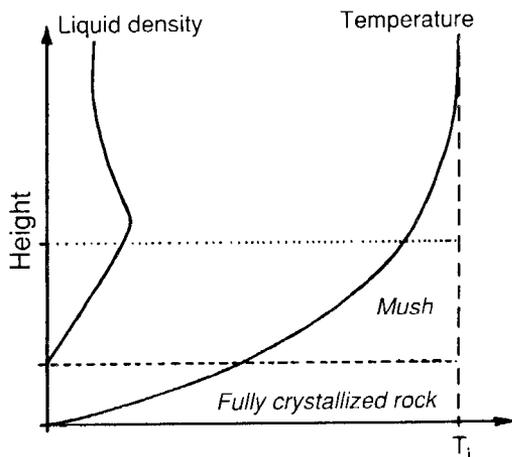


FIG. 6. Schematic plot of temperature and liquid density as a function of height in a mushy layer at the floor of an intrusion. The liquid density profile is unstable, and can lead to compositional convection. Two different instabilities can occur; the first is due to a thin boundary layer which develops at the top of the mush by chemical diffusion (Tait and Jaupart, 1989), the second occurs throughout the permeable mush and involves a larger composition difference. This second mode of convection generates the largest compositional flux and dominates the chemical evolution of the reservoir (Tait and Jaupart, 1992a).

thickness. Tait and Jaupart (1989, 1992a) have shown experimentally and Worster (1992) from a theoretical point of view that two independent convective instabilities are possible. The first is that of the thin diffusive chemical boundary layer that exists just above the mushy layer, and the second is that of the interstitial liquid within the mushy layer. The onset of both instabilities is found to be governed by different local criteria. The Rayleigh numbers for the chemical boundary layer and for the interstitial liquid increase as the thickness of the crystallizing zone grows until eventually the critical values are attained. Scaling arguments indicate that for the large values of the viscosity of common silicate melts, instability of the diffusive chemical boundary layer will tend to take place before that of the interstitial liquid.

These aspects concerning the onset of instability have now been experimentally verified (Tait and Jaupart, 1992b). It was also shown that the moments of onset of both instabilities can be expressed in terms of a certain crystallized thickness. Once the second instability (that of the interstitial liquid) has taken place and vigorous exchange with the overlying reservoir has begun, the thin chemical boundary layer at the top of the mush is destroyed, and hence,

only the mushy layer mode of compositional convection remains.

An important implication of the above is that it should often be possible to observe some record of the onset of these instabilities in the basal sequences of intrusions. It is expected that, adjacent to the basal contact, there will be a certain thickness of rock that has formed in a purely conductive regime. Such basal sequences have indeed been observed beneath the first accumulate rocks, and in some cases studied in detail (e.g. Page, 1979). We suggest that studies of basal sequences will be a valuable contribution in the future that can help to constrain models for the onset of chemical exchange and the rates at which it starts to affect the compositions of the rocks in the natural magmatic situation.

Convective processes of large-scale differentiation

In-situ crystallisation models, in which the solid grows in a thermal boundary layer at the chamber margins, predict the formation of a mushy layer in the case of multicomponent systems. The study of the physics of mushy layers has thus become the focus of attention in recent work. We indicate results of potential geological significance but will avoid going into great detail.

Scaling of differentiation processes

The impotence of the diffusive (i.e. without melt motion) *in-situ* crystallization models described earlier as a means of producing a chemically stratified solid, is mainly a consequence of two facts. The first is that the growth rate of the solid and the chemical diffusion profile in the liquid are not independent, being linked by the requirement of conservation of mass applied at the solidification front. In practice one can only impose changes in growth rate by changing the heat flux, and the chemical gradient will adjust so as to re-establish the steady state. The other point is that because chemical diffusivities in silicate melts are typically very low, chemical effects are always restricted to the vicinity of the solidification front.

Large scale differentiation of the liquid reservoir, requires large scale relative motions of solid and liquid such as occur by crystal settling or compositional convection. A crucial point is that models that are designed to describe these effects must be correctly scaled with respect to the natural situation. This requires, on the one hand, respecting geometrical similarity between the two situations and, on the other hand, assessing the relative magnitudes of the important forces acting. One consequence of this is that experiments carried out with natural magmas cannot be used to study the importance of convective

processes in a physically realistic manner because technical restrictions impose that these experiments are too small. Hence, a fundamental problem is posed by the characteristic length scales inherent in such phenomena. These determine, for example the magnitudes of the buoyancy and viscous forces acting in a convective flow and hence the dynamic regime. Whereas experiments with natural magmatic liquids establish equilibrium thermodynamic constraints where length scales do not have to be specified, they give no information on dynamic phenomena.

Experiments to study the dynamics of crystallizing systems have therefore employed liquids other than real magmas — very often aqueous solutions of simple salts as these can often be manipulated in order to attain a specific dynamic regime. Such solutions do not, of course, have the same range of chemical compositions and other thermodynamic variables as igneous melts and herein lies much of the difficulty in arriving at an integrated physical and

chemical approach to study igneous systems. It may be hoped that complex chemical thermodynamics can ultimately be integrated into models of convective processes, presumably by numerical calculations. At present we just wish to emphasize that there is no escaping constraints of correct scaling of physical processes that require the use of analogue materials rather than natural magmas.

Differentiation in the presence of a mush

In a boundary layer that is cooled from below, and that is such that the chemical gradient in the interstitial liquid leads to compositional convection, porosity fluctuations can develop because of the requirement of local thermodynamic equilibrium between crystals and liquid. Tait and Jaupart (1992a) give details of this process, but the principle, in brief, is that as fluid rises and sinks in the porous layer, it tends to adjust its temperature by horizontal diffusion of heat and therefore is required to adjust its

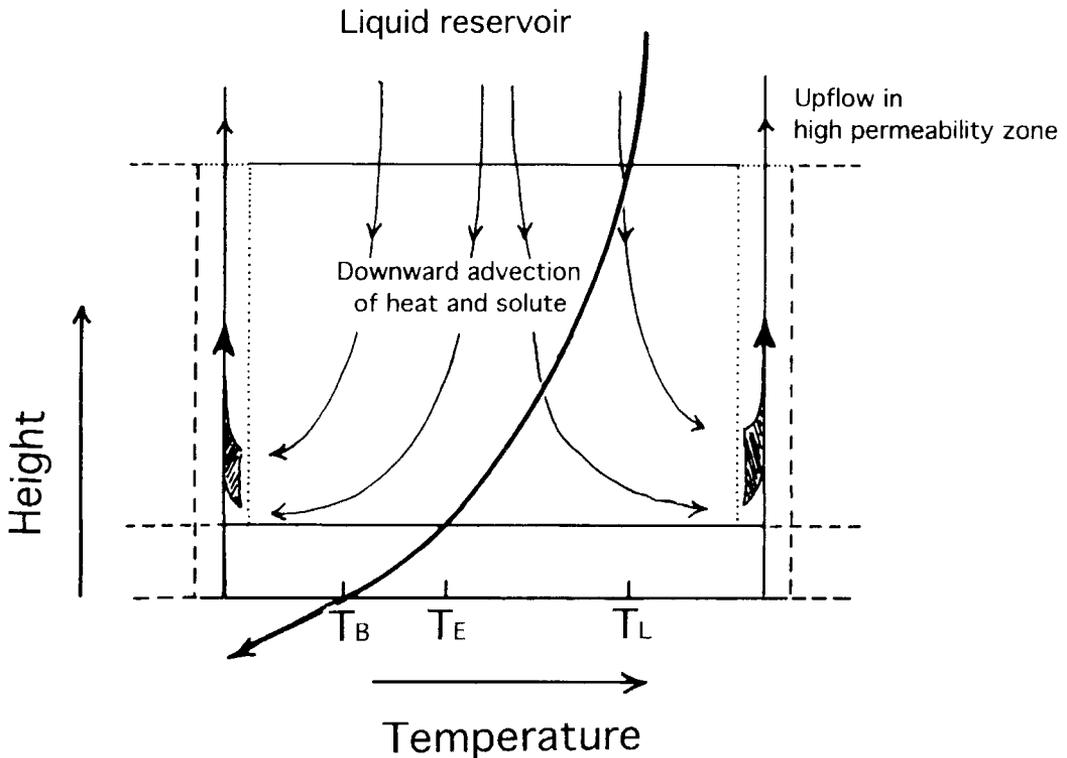


FIG. 7. A convection cell is shown in the mushy layer, that is delimited by the centre-lines of two high permeability regions in which upward flow of residual liquid is taking place. Heat is conducted out through the lower boundary. As hot, solute-rich fluid from the reservoir descends it cools and crystallizes. If the heat advected downwards by the motion balances the rate of downward conduction, crystallization takes place with the local temperature remaining constant.

composition in order to satisfy local equilibrium. This it does by dissolving crystals in upwellings and precipitating in downwellings. This leads to a change of permeability structure. Velocity increases in the upwellings, which implies a reduction of the area they occupy. Crystallization takes place in the broader downwellings in the porous layer between the upwellings. The system can ultimately form narrow, crystal-free 'chimneys' through which residual liquid rises into the overlying reservoir.

The occurrence of compositional convection has a number of geological implications. Firstly, it provides a mechanism of adcumulus growth, the heat budget of which can be prescribed in a consistent manner. The flow field brings magma down from the overlying reservoir into the crystallizing region and rejects residual liquid back up into the overlying reservoir. If the convective heat flux due to the downflow of hot magma from the reservoir into the porous medium balances the rate of heat extraction by conduction through the bottom boundary, crystallization takes place at constant temperature as is required to form adcumulates (Jahrling and Tait, in prep.) (Fig. 7). In this way, crystallization fundamentally modifies the heat budget of the lower boundary layer compared to the conductive regime. In the latter regime, crystallization only occurs if temperature is actually reduced, which does not produce an adcumulate. A variable that helps to visualize this process is the local chemical composition calculated as a weighted mean of solid plus liquid; this variable defines the solidus. The flux of chemical components from the overlying reservoir modifies this local chemical composition by adding refractory components (Fig. 8). For some phase diagrams, this can raise the local solidus of the system as crystallization is taking place, in contrast with the conductive regime.

Another interesting possibility is that the residual liquid rising into the reservoir is heated up by horizontal thermal diffusion and hence may become superheated. If the compositional flux is large enough, this effect can suppress crystallization in the interior of the liquid body. Some of the chemical consequences of such a mixing process associated with *in-situ* crystallization were explored by Langmuir (1989), without being specific about the physical mechanism causing the flow. We note that the chimney regime is the most favourable for this to take place because of the absence of crystals in the chimneys. A very important implication is that the fluid that is being mixed back into the reservoir will have seen some extraction of all of the mineral phases that exist on the liquid line of descent, in spite of the fact that these lower temperature phases are not yet on the liquidus of the melt making up the bulk of the overlying reservoir. The chances of measuring

such an effect will diminish with increasing size of the reservoir because of an effect of dilution. As an illustration, one could envisage a reservoir of melt with just olivine on the liquidus into which compositional convection was remixing melt that had seen some crystallization at lower temperatures of phases such as plagioclase, clinopyroxene, orthopyroxene and quartz. If the reservoir were to be sampled before the residual melt was perfectly mixed back in to form an homogeneous liquid, one might interpret such relationships as magma mixing. Indeed a process such as this has been observed by Helz *et al.* (1989) in the Kilauea Iki lava lake where mixing of two distinct magmas is not an acceptable explanation as it was a closed system.

The production of an adcumulate layer and a stratified solid.

We describe briefly an experiment with an aqueous solution of ammonium chloride, similar to those already reported by Tait and Jaupart (1992a), but done over a long time in a deep tank. The top and side-walls of the tank were thermally insulated and the initial temperature of the liquid was slightly above the liquidus. The temperature of the lower boundary was maintained at a value below the

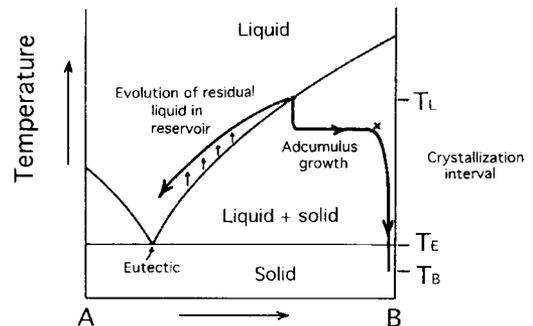


FIG. 8. Illustration of the adcumulus regime in a binary eutectic phase diagram. The temperature difference across the mush is ($T_L - T_E$). The heavy line indicates a possible evolution of a point fixed in space as crystallization progresses and the mush moves past it. In this example, when the point first enters the mush, compositional convection is weak and crystallization occurs by reduction of the local temperature. Compositional convection becomes established and solid B crystallizes as solute and heat are advected downwards. Once the solid fraction has greatly increased and the permeability is reduced such that convection is again weak at this level, crystallization of the remaining 'trapped liquid' occurs by the reduction of the local temperature until the eutectic value is reached.

eutectic, and hence we observed the growth of a totally solid layer at the base of the tank. This solid layer was made of ice and ammonium chloride crystals. There was also a mushy layer between the eutectic temperature and the liquidus. This layer grew to up to 10 cm. thick and was much thicker than the characteristic inter-crystal pore size, which was <1 mm. Compositional convection acted continuously to reduce porosity in the mushy layer; when the experiment was stopped after seven days, the mushy layer above the eutectic front was almost totally solid. The liquid fraction remaining was very small and hence we had produced an almost solid layer of ammonium chloride crystals. In geological terms, this upper layer could be called an ammonium chloride adcumulate. This experiment therefore resulted in a compositionally stratified solid and in an adcumulate layer. The extraction of ammonium chloride from the liquid circulating over the whole tank height led to chemical differentiation of the interior. The key result is that adcumulus growth can be achieved naturally by compositional convection generated in a relatively thick mushy layer at the base of the reservoir.

The overall differentiation trend of large igneous complexes

The overall differentiation trend that should be recorded in the final solid can be predicted in simple terms. If we isolate events in the lower boundary layer, in the contact zone, initially cooling will be rapid and solid rock will be formed before much in the way of chemical effects have been able to take place. Once compositional convection becomes important, the rocks that form will become enriched in refractory components relative to the starting liquid due to the flux down from the overlying reservoir. The reservoir will, at a rate depending on its overall size, eventually become depleted in refractory components and even though compositional convection may continue to act, there will be an overall large-scale trend towards more depleted compositions. This is shown schematically in Fig. 9 in which we identify three zones in the rocks that may ultimately be formed. First, there is a lower zone in which the rocks are enriched in refractory components with respect to the initial liquid, and in which this enrichment increases with stratigraphic height. The second zone is one in which the rocks are still enriched with respect to the initial liquid, but in which the enrichment decreases with height, and finally a third zone, in which the rocks are depleted in refractory components with respect to the initial liquid, and in which the concentrations of these components decreases progressively with height. One might argue that the incorporation of sedimentation

into the events will tend to smear out these trends but not change their qualitative features.

One might equally consider the profile for an incompatible trace element. As long as compositional convection is acting, the concentration of the element is likely to remain rather constant. When eventually the element builds up to such a level that a new phase appears in which it is compatible, the concentration of the element in the final rocks should rise abruptly. Such are the qualitative features, e.g. of the P_2O_5 profile determined by Wager (1963) in the layered series of the Skaergaard intrusion — the concentration remains low and the profile featureless until the appearance of apatite. Similar qualitative features are also shown by incompatible trace element profiles in the Bushveld rocks (Cawthorn and McCarthy, 1980, 1985) and the Palisades Sill (Shirley, 1987).

Cooling mechanisms and crystallization rates

The process of in-situ crystallization at the floor has been studied in detail and has been shown to produce adcumulates and a stratified solid. The physics of crystal settling has not yet been pursued in such a systematic manner, although a number of novel phenomena have been described (Koyaguchi *et al.*, 1993; Sparks *et al.*, 1993; Marsh and Maxey, 1985; Jarvis and Woods, 1994). It is nevertheless possible to evaluate its importance for the floor cumulate sequence. We note that crystal settling requires thermal convection, and that it is not favourable to adcumulus growth. As stated earlier, the rate of crystallization is determined by the cooling rate, and both *in-situ* growth and crystal settling are likely to occur at different rates. We evaluate both and

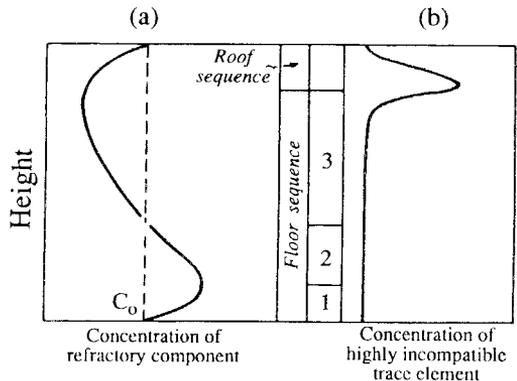


FIG. 9. The predictions of the differentiation model described in the text are shown for the stratigraphic profiles of chemical compositions of the rocks. (a) A refractory major element. (b) a highly incompatible trace element.

determine the conditions under which one may dominate over the other.

At the floor of an intrusion, we consider first the initial phase with no convection, during which the leading edge of the thermal boundary layer and solidification front are located at distances $X_1(t)$ and $X_2(t)$ from the margin, such that:

$$X_1(t) = 2 \lambda_1 \sqrt{\kappa t} \quad (1)$$

$$X_2(t) = 2 \lambda_2 \sqrt{\kappa t} \quad (2)$$

where κ is the thermal diffusivity of the partially crystallized layer and where constants λ_1 and λ_2 depend on the initial conditions and on latent heat release by crystallization (Worster, 1986; Brandeis and Jaupart, 1987). The form of these equations is insensitive to processes such as crystallization kinetics and melting of country rocks. For realistic geological conditions, constants λ_1 and λ_2 take values close to 1, but with λ_1 significantly greater than λ_2 . One important result is that the solidification front advances at a rapidly decreasing velocity. Also, note that a given temperature difference is achieved over a steadily increasing distance, which leads to a steadily decreasing heat flux.

At the roof contact, there is a large variation of viscosity across the boundary layer, and hence only its least viscous part goes unstable. Our present understanding of this process is limited to simple systems (Davaille and Jaupart, 1993a,b), and hence one must be cautious in estimating the heat flux of thermal convection. This heat flux, unlike that of conduction, remains high for an extended length of time because repeated instabilities act to maintain the thermal boundary layer at a small thickness. The heat flux decreases only over long times as the whole magma volume is being cooled. For a given heat flux, one may calculate a bulk crystallization rate that includes contributions from crystals held in suspension and from those which settle to the floor. A steady-state situation can be reached such that the amount of suspended crystals remains constant. In this case, the amount of crystals which settle out of the reservoir balances the amount of new crystals formed (Jarvis and Woods, 1994). The bulk heat balance for the convecting magma layer reads:

$$\rho C_p h \frac{dT}{dt} = -Q + \rho LR \quad (3)$$

where T is the average temperature of the chamber, h the thickness of melt (initially the chamber thickness), Q the convective heat flux, L latent heat of crystallization and R the accumulation rate of crystals at the floor per unit area. To calculate R , one needs to specify the heat flux as well as the cooling rate, which in turn depends on the crystallization rate. As shown in the Appendix, one may obtain bounds on the accumulation rate R , which may be written as:

$$R \approx \lambda \frac{Q}{\rho L} \quad (4)$$

where λ is a constant which takes a typical value of $\frac{1}{2}$ (Appendix).

The floor sequence grows by in-situ growth and by the addition of crystals settling from above. The former dominates initially, but decreases continuously, and settling may dominate accumulation at the floor after some time. We calculate the thickness at which this crossover should occur, and ask whether it occurs before or after the onset of compositional convection. If compositional convection starts first, it modifies the heat budget of the reservoir and the mechanisms of crystallization. The rate of advance of the floor crystallization front may be obtained directly from eqn. (1):

$$\frac{dX}{dt} = \lambda_1 \sqrt{\frac{\kappa}{t}} \quad (5)$$

This may be compared with the accumulation rate due to crystals settling from the convecting interior, and the thickness at which the crossover occurs calculated as a function of the heat flux (Fig. 10). Current estimates of the convective heat flux are in the range of 1 to 100 $\text{W}\cdot\text{m}^{-2}$. Thus, settling could dominate the floor sequence after a thickness of about 100 m. At the lower end of the range for the convective heat flux, this crossover occurs after more than 1000 m of cumulates. According to estimates by Tait and Jaupart (1992a,b), compositional convection starts when the thickness of the cumulate pile is a few hundred meters. Therefore, the question raised at the beginning of this section cannot be answered with

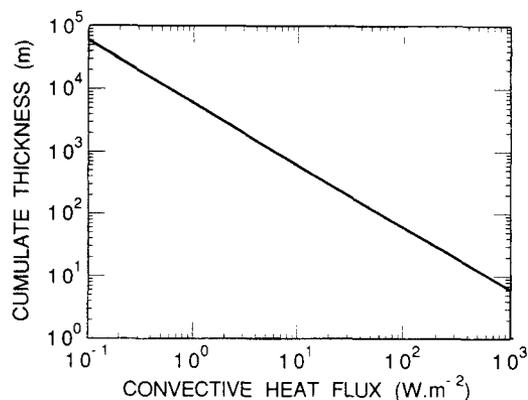


FIG. 10. Thickness of cumulates at the floor of an intrusion at which crystal settling from above begins to contribute more crystals than in-situ nucleation and growth. This thickness may be calculated as a function of convective heat flux through the roof, for which a typical value lies in a range of 1 to 100 $\text{W}\cdot\text{m}^{-2}$. Values of the variables used in the calculation are as follows: $k = 2 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$, $L = 8 \times 10^5 \text{ Jkg}^{-1}$, $C_p = 10^3 \text{ Jkg}^{-1}\text{K}^{-1}$.

confidence. Nevertheless, *in-situ* crystallization is expected to remain important, if not dominant. With regard to the thickness of the mushy layer, we note that crystal settling is unlikely to reduce it.

One point illustrated by this rough calculation is that, although there is a large volume of melt, cooling is limited by heat transfer in boundary layers which are much smaller. Thus, the crystallization rate is small, and in fact independent of the thickness of melt, as shown by eqn. (4). This emphasizes why understanding of boundary layer structure can be the key to understanding to what can happen in the interior.

Conclusion

Physical models of *in-situ* crystallization in transient thermal conditions have been developed which help identify the various regimes of solidification and macrosegregation. They are limited to binary and ternary systems, and hence are too simple to reproduce true rock sequences. However, they predict adcumulus growth and large-scale differentiation, i.e. the most significant large-scale features of igneous intrusions. In comparison, the study of crystal settling has not reached the same level of understanding. A rough calculation demonstrates that it is unlikely to overwhelm *in situ* crystallization at the floor. The most general result is that relatively thick layers of partially crystallized rocks must be present in magma chambers.

Interpretations of layered intrusions rely on petrological constraints, such as the order of crystallization of different phases as found on the outcrop. The recognition that mushy layers spanning the whole crystallization interval probably exist in such magma chambers should lead to a reevaluation of these arguments. The powerful implication is that the whole liquid line of descent is present in the reservoir, with large quantities available if the mushy layer is thick. Mixing of differentiated liquid with undifferentiated liquid may therefore occur in a closed system and lead to geochemical and petrological effects that have not been fully explored.

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Appendix

The crystallization rate due to heat loss at the roof

One may determine bounds on the rate of crystallization by considering different nucleation and growth regimes. In a kinetic regime, the temperature drops by some amount to promote nucleation and is then maintained at a constant value. Latent heat release balances the rate of heat loss in eqn. (3). Thus,

$$R_1 \approx \frac{Q}{\rho L} \quad (\text{A1})$$

This corresponds to an upper bound because it relies on the assumption that the temperature of the interior does not decrease. This is clearly an extreme case. In the other extreme, crystals are kept in equilibrium with the melt. Thus, temperature T and the fraction crystallized, χ , are given by the fractional crystallization relationship:

$$\chi = f(T) \text{ for } T_S < T < T_L \quad (\text{A2})$$

where f is some function which is known for a given chemical system. The function f is zero at the liquidus and 1 at the solidus. With this equation, we may calculate R as follows:

$$R = h \frac{d\chi}{dt} = h \frac{df}{dT} \frac{dT}{dt} = -hA \frac{dT}{dt} \quad (\text{A3})$$

where A is positive. Substituting for this in the heat balance equation, we find a lower bound to the accumulation rate:

$$R_2 = \frac{Q}{\rho \left(L + \frac{C_p}{A} \right)} \quad (\text{A4})$$

A reasonable estimate for both L and quantity $\frac{C_p}{A}$ at temperatures close to the liquidus is $5 \times 10^5 \text{ J kg}^{-1}$. Thus, the two estimates for the accumulation rate are within a factor of two.

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