

Implications of Magma Chamber Dynamics for Soret-Related Fractionation

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Convection of silicate melts in magma chambers is considered as a possible mechanism for producing significant or, at least, detectable chemical fractionation by the Soret effect. Thermal boundary layer analyses show that Soret fractionation would be, at best, an extremely weak process in evolving magmatic systems. For very large amplitude thermally driven convection at horizontal chamber margins, both the magnitude of the temperature gradient and the time scale for residence of magma in the unstable thermal boundary layer are entirely inappropriate for chemical fractionation of the magma at levels comparable to those obtained in laboratory experiments (5–60%). For a typical convecting body a relative concentration enhancement of 0.04% is obtained as an estimate of the upper limit of Soret fractionation. Even if exceedingly large temperature gradients (of the order of 10^4 °C/m) in a transient thermal regime are attained by compositionally driven convection, the magma residence time in the thermal boundary layer is so brief (100 s) that much less chemical fractionation results (0.002%). For convection near vertical margins a kinematic model of a countercurrent flow regime provides estimates of chemical separation produced by the thermogravitational fractionation mechanism. Incorporating a range of physical and chemical parameters that characterize magma chamber convection, steady state values of concentration enhancement are even smaller than for fractionation near horizontal boundaries.

1. INTRODUCTION

Intrusion of magma into cooler host rock gives rise to temperature variations capable of driving convective motions through the production of thermal and chemical buoyancy. Owing to the low thermal diffusivity of molten rock, even weak convection in magma is expected to be characterized by strong boundary layer behavior. (Here, the term thermal diffusivity refers to the flow of heat only and should not be confused with the transport of chemical species induced by temperature gradients which we refer to as Soret diffusion.) The parameters defining a thermal boundary layer such as the characteristic thickness and average temperature contrast determine the rate of heat loss from the magma body. In recent years the thermal boundary layer regime has also been linked to hypothesized magmatic processes involving Soret fractionation which counter the tendency of convection to homogenize magma [Shaw *et al.*, 1976; Hildreth, 1981; Schott, 1983; Walker *et al.*, 1981; Walker and DeLong, 1982, 1984; Walker, 1983]. Such unmixing mechanisms are potentially significant processes in both the chemical and dynamical evolution of magmatic systems. However, most discussions of these boundary layer-related processes are heuristic and depend a great deal on the presumed thermal and dynamical behavior of that regime.

The nature of buoyancy can strongly affect the amplitude and style of convection. Thermally driven flows, where buoyancy is derived from thermal expansion and contraction of the magma, will exist in chambers cooled from above and/or from the sides. Somewhat larger amplitudes than can be achieved by thermal buoyancy are possible if chemical changes produce buoyancy in the magma. Crystal fractionation processes are the most likely mechanisms for producing substantial chemical or compositional buoyancy. For example, plating of crystals on the cooler margin of a magma body can give rise to a compositionally distinct mobile layer of magma capable of driving circulations [McBirney and Noyes, 1979]. Recent studies of double diffusive convection have considered this source

of buoyancy in conjunction with thermal buoyancy [Huppert and Turner, 1981]. The relative magnitudes of chemical and thermal buoyancy as well as chemical and thermal diffusivities will affect both the style and amplitude of circulations within the magma.

Fortunately, uncertainties about the style and amplitude of convection in magma, while complicating the analysis, do not affect most of the fundamental models used to estimate the role of magma circulations for enhancing Soret fractionation. For simplicity, most of our discussions are presented in the context of thermal buoyancy, although we include comments about chemical buoyancy and double diffusive convection when their roles might be significant for Soret processes.

One important effect, which may involve crystal plating on the margin of an evolving magma chamber, is the layering or stratification of the intrusion that occurs during the evolution of the magma body [Irvine, 1980; Spera and Crisp, 1981]. Because of the smaller density contrasts associated with thermal buoyancy it is expected that thermally driven circulations will be mostly confined to individual layers in the chamber. Progressive compartmentalization of convection into compositionally distinct layers will eventually decrease the amplitude of circulations in the body. While the models presented here are general enough to be applied to this situation, we will not consider this case in which convection is "handicapped." Rather, we consider only cases where thermally or compositionally driven convection provides the thermal regime that is optimal for Soret processes.

An often overlooked but important fact about magma chamber modeling is that the character of the boundary layer regime not only depends on the internal nature of the system, that is, properties of magma related to buoyancy and viscosity, but also upon the thermal properties of the boundaries. The thermal conditions applied to the boundary of the body should model the series heat transfer relationship involving both magma and host rock. Constant temperature conditions imply that magma convection controls the rate of heat transfer from the body, while constant heat flux conditions simulate a host environment that controls heat transfer. Several models [Jaeger, 1964; Carrigan, 1983, 1986] indicate that constant flux conditions model this relationship more accurately than do constant temperature conditions. On the other hand, a

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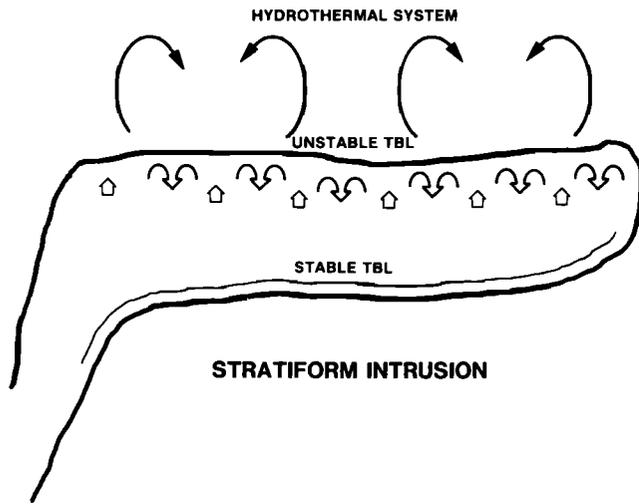


Fig. 1. In a stratiform intrusion, thermal layers will form on the top and bottom margins. Only the top margin will be unstable, giving rise to thermally driven flows. Magma will not remain indefinitely within this unstable thermal layer but will have a characteristic residence time ranging from hours to a thousand hours.

constant temperature condition permits estimates to be made of the maximum realizable temperature gradients occurring in a given magmatic system. This is useful for bounding the possible effect of convection for Sorét processes.

In this paper we focus on the relationship between boundary layer convection and the Sorét and thermogravitational mechanisms. We develop a dynamical model for convection near horizontal boundaries that is used to evaluate the chemical transport equations. For vertical margins a kinematic flow regime is used to assess the importance of the thermogravitational mechanism.

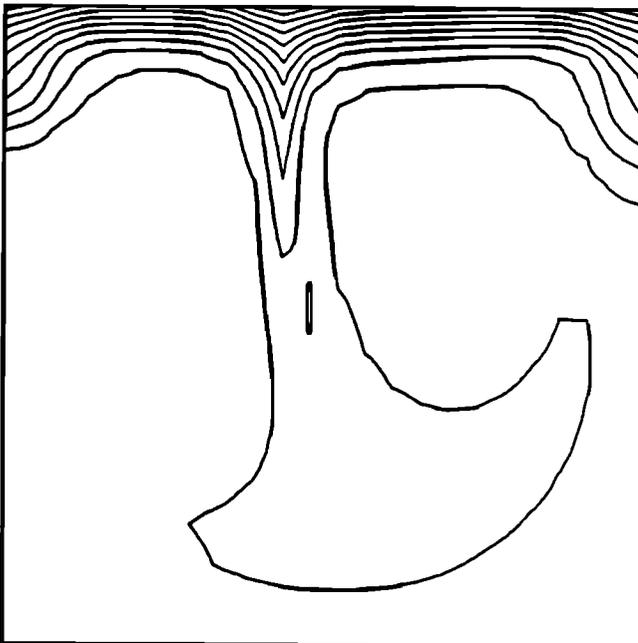


Fig. 2. The collapse of a gravitationally unstable thermal layer in a cooling reservoir experiment. Heat is lost through the top of the box simulating the unstable layer in the intrusion of Figure 1. In these kinds of numerical experiments the ratio of the viscosity at the top of thermal layer to the viscosity at its base may approach 10^5 .

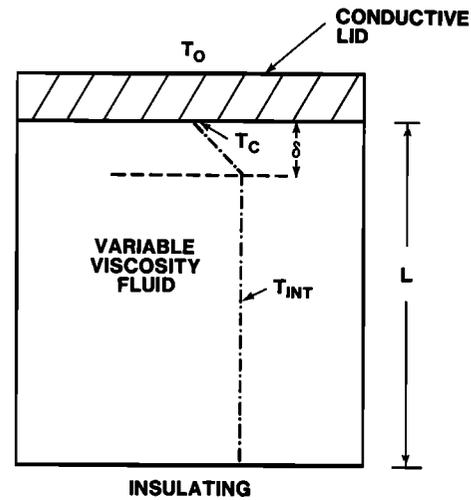


Fig. 3. The geometry, boundary conditions, and assumed shape of the temperature profile are shown schematically for the parameterized model. The thickness of the conductive lid may be varied to simulate a uniform temperature (lid thin compared to thermal layer) or uniform heat flux (lid thick relative to thermal layer width).

2. THERMAL LAYER FORMATION ON HORIZONTAL MARGINS

Initially, we will be concerned with the problem of thermal layer formation adjacent to horizontal margins. Broad sill-like magma chambers like the model suggested by Walker and DeLong [1982] as a source for mid-ocean ridge basalts might be characterized by a large ratio of horizontal to vertical surface area resulting in a dynamical and thermal regime dominated by the thermal boundary layers forming on the horizontal margins. The cooler margins give rise to a stably stratified layer on the bottom and an unstable layer on the top. Only the upper unstable layer is dynamically significant for thermal convection. The lower layer will behave like a thermal buffer between the hotter magma and the cooler margin [Jaupart *et al.*, 1985]. The Prandtl number for convecting magma, the ratio of the kinematic viscosity ν to the thermal diffusivity κ , will likely exceed 10^5 for any given system. Because of the relatively low diffusivity of heat, thermal features such as the boundary layer will tend to exist on a scale which is small compared to the scale for fluid flow. For high Rayleigh number, time dependent thermal convection characterizing magma chambers, a thermal layer will gradually form adjacent to the upper margin (see Figure 1). Progressive thickening of the layer occurs as heat is conducted from the magma across the margin. However, the thickening of this cooler region will not proceed indefinitely. Since the layer is cooler than the surrounding magma, it is also characterized by higher density than its surroundings owing to thermal contraction and possibly the crystallization of minerals which remain in suspension. This increase in density causes the growing layer to become gravitationally unstable. Finally, it breaks away locally from the margin and falls into the surrounding magma. The growth and detachment of the thermal layer occurs locally over the entire horizontal margin on a relatively short time scale (hours to hundreds of hours) driving a large-scale circulation in the magma. This time dependent boundary layer behavior has been observed in a variety of convection experiments, numerical and laboratory, which simulate the dynamics and boundary conditions considered here [Hewitt *et al.*, 1980; Carrigan, 1982, 1985]. Figure 2 illustrates

a scaled down numerical model of the collapse of the thermal layer into a reservoir containing a fluid with properties simulating those of magma. Numerical models show that the cooled boundary layer fluid may either mix in with the magma or settle out on the floor of the reservoir, giving rise to a thermal inversion there.

We quantify some aspects of this process using a one-dimensional parameterized model. This method has successfully simulated the horizontally averaged temperature field in a finite element model of a convectively cooling reservoir containing fluid with strongly temperature dependent viscosity [Carrigan, 1984] in which the viscosity varies by 5 orders of magnitude. Figure 3 illustrates the geometry, the boundary conditions, and the form of the temperature field assumed in the parameterized model. We reiterate that only the average characteristics of the temperature field are predicted by this method. The model relates the heat flux Q leaving the top through the conducting lid to the time rate of change of the average temperature T_A of the fluid:

$$Q = -\rho C_p \frac{d}{dt} T_A L \quad (1)$$

where ρ , C_p , and L are the magma density, the specific heat, and the depth of the chamber, usually taken to be 1 km. The heat flow through the lid may also be expressed in terms of a heat transfer coefficient H as

$$Q = H(T_c - T_0) \quad (2)$$

Assuming that heat flow through the lid balances that across the boundary layer at any given time yields

$$\frac{k}{\delta} (T_{int} - T_c) = H(T_c - T_0) \quad (3)$$

where the boundary layer thickness is calculated from

$$\delta = L \left(\frac{Ra_c}{Ra} \right)^\beta \quad (4)$$

with the flux-based Rayleigh number Ra having the form

$$Ra = \frac{g\alpha L^4 Q}{\kappa k \nu} \quad (5)$$

The quantities g , α , k , κ , and ν are gravitational acceleration, fluid thermal expansion coefficient, fluid thermal conductivity, fluid thermal diffusivity, and kinematic viscosity, respectively. Ra_c is the critical value of the Rayleigh number taken to be about 2700, and the exponent β is taken to have a value of 0.25; although *Morris and Canright* [1984] find 0.175 to be a better value assuming the Rayleigh number defined in equation (5). Our use of a larger value of β permits an upper bound for the temperature gradients in the thermal boundary layer to be predicted as a function of time. Thus we are able to estimate the maximum efficiency of convection in transporting heat to the margin. Finally, we may write the average temperature of the fluid T_A as the weighted average of the mean boundary layer temperature and the temperature of the isothermal interior:

$$T_A = \frac{T_c + T_{int}}{2} \frac{\delta}{L} + T_{int} \frac{L - \delta}{L} \quad (6)$$

These equations represent a nonlinear initial value problem for the thermal regime as a function of time. At time $t = 0$ it is

assumed that the entire body is at a uniform temperature. The solution procedure involves a stiff ordinary differential equation solver [Carrigan, 1984].

The thermal layer parameters will depend both on the dynamics of convection and the thermal boundary conditions. By selecting constant temperature boundary conditions we can eliminate the effect of the host rock in determining heat flow from the magma. The calculated temperature gradients will be overestimates of the actual ones, while average boundary layer thicknesses will tend to be underestimated. The constant temperature results presented here, then, may be interpreted as the best that convection can do in providing a high gradient thermal regime for chemical fractionation.

3. PREDICTIONS OF PARAMETRIC SCHEME

In applying the parameterized model we consider a kilometer-sized sill cooled mainly by heat transfer at the top. Two different viscosity ranges are considered in making the calculations. One corresponds to a magma having a more or less basaltic composition. In this case, a starting viscosity of 100 P is assumed increasing 2 orders of magnitude per 100°C of cooling. For a sill containing relatively dry rhyolitic magma, a starting viscosity of 10⁶ P is used, increasing again at the same rate. A temperature difference of 400°C is assumed to exist between the margin of the body and the magma at time of emplacement ($t = 0$). This intended overestimate of the temperature drop across the thermal boundary layer contributes to the extreme upper bound of the thermal gradient. For a 1-km-sized body losing heat to a constant temperature boundary, Figure 4 shows estimates of the temperature gradient as a function of time. For these calculations we assume that the magma chamber is a closed system with respect to mass to show how the temperature gradient might change with time. If the system was periodically recharged, a different time dependence might apply, although the maximum realizable gradients would be expected to fall within in the range plotted here. Very early in the cooling history, gradients in the

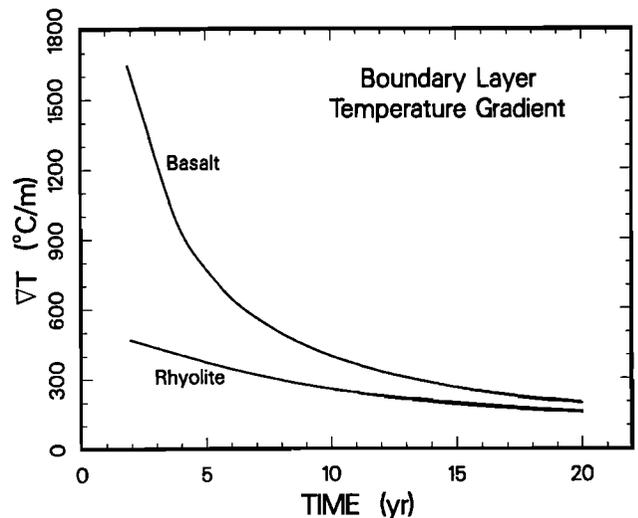


Fig. 4. Time dependent thermal boundary layer temperature gradients for basalt and rhyolite in a cooling, 1-km body with infinitely conducting boundary represented by constant temperature condition. In this limit, heat flow and temperature gradient are controlled by magma convection only. Initial temperature difference between magma and boundary is 400°C. Initial viscosities of basalt and rhyolite are taken to be 10² and 10⁶ P, respectively. Viscosities increase by a factor of 10² per 100°C of cooling.

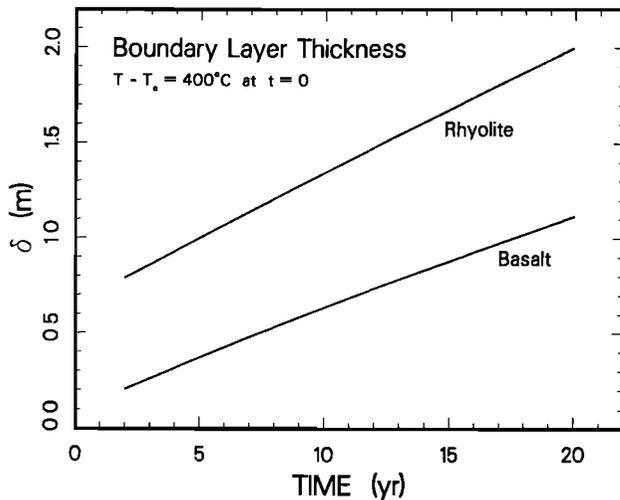


Fig. 5. A plot of the thermal boundary layer width for basalt and rhyolite in a 1-km intrusion as a function of time.

basalt may exceed $1000^{\circ}\text{C}/\text{m}$. But within 15 years after emplacement the gradients have fallen to about $300^{\circ}\text{C}/\text{m}$ being comparable to the value for the rhyolite case at the same time. Maximum gradients in rhyolite are about $500^{\circ}\text{C}/\text{m}$ and taper off more slowly than in the basaltic case.

Regarding the thermal layer thickness (Figure 5), we find that the basaltic thermal layer is under a meter thick, while the rhyolitic thermal layer is not much thicker in spite of the much higher viscosity of the rhyolite magma. The weak dependence of the boundary layer thickness follows from the power law relation (equation (4)) between the layer thickness and Rayleigh number which includes magma viscosity.

We wish to emphasize that the parameter values given here represent an extreme: the best that thermal convection is likely to do in producing large gradients at the margin of the body. Some idea of how heat transfer in the host rock will affect convection in the body can be provided by comparing the distances over which a significant temperature change occurs in the magma with the distance in the host environment over which a comparable drop in the temperature is likely to take place. Hardee [1982] has developed a two-phase model for hydrothermal heat transfer above magma heat sources. Besides the formation of two-phase permeable convection zones, he predicts the existence of conduction-dominated dryout zones which vary in thickness from about 10 m for a permeability of 0.3 darcy (Kilauea Iki Lava Lake) to upward of 1 km for permeabilities in the millidarcy range. The temperature range in the zone is defined at one extreme by the cracking temperature, the temperature at which thermal stresses in the cooling magma gives rise to brittle failure. Ryan and Sammis [1981] estimate this temperature to be about 725°C in the case of basalt. The cool boundary of the conductive dryout zone is defined by the vapor saturation temperature which will fall somewhere between 100°C and 375°C depending on the depth where the phase change occurs. Taking the maximum temperature difference ($725^{\circ}\text{C} - 100^{\circ}\text{C} = 625^{\circ}\text{C}$) as representative of the temperature drop and assuming that it occurs across a zone only 10 m thick, yields a conductive temperature gradient of only about $60^{\circ}\text{C}/\text{m}$. If the magma body heat transfer balances that in the host environment, then it must also be characterized by a similar gradient. If magma chamber convection cannot match this gradient, then infiltration will occur. Alternatively, the magma may melt

the host rock during part of the evolutionary period of the body. But the largest gradients which can occur in this case will still be bounded on the upper side by the gradients derived assuming the constant temperature, infinitely conducting boundaries.

We now calculate characteristic residence times within the boundary layer for a particular mass of fluid, although the parameterized model alone does not provide this information. To obtain residence time estimates, we consider Howard's [1966] statistical model for high Rayleigh number convection. Howard's calculations are based on the observation that near a boundary, a zone of fluid cooler than the rest gradually develops as a result of conductive heat loss. The growth stops when the fluid suddenly breaks away from the boundary falling into the interior in a manner analogous to the formation of thermals in the atmosphere. These thermals or sinking jets have also been observed in high Prandtl number experiments bearing some resemblance, in terms of the dynamics and boundary conditions, to magma chamber convection [Carrigan, 1982, 1985]. The process described here is governed primarily by a conductive time scale, that is, it is assumed that the time t_* required for conductive cooling to thicken the layer until it becomes unstable is long compared to the time for removal of the thermal from the boundary. This process then repeats on the time scale t_* . At any given time, thermals will be in different stages of development over the entire layer.

We calculate t_* , the characteristic residence time of magma in the boundary layer, by expressing it in terms of the critical layer thickness δ following Kenyon and Turcotte [1983]. The solution for the time dependent conductive heat flux resulting from the cooling of an isothermal half-space is

$$Q = \frac{k\Delta T}{(\pi\kappa t)^{1/2}} \quad (7)$$

where ΔT is the temperature drop across the cooling layer. If it is assumed that the temperature variation across the cooling layer is linear during the formation of the thermal, then we

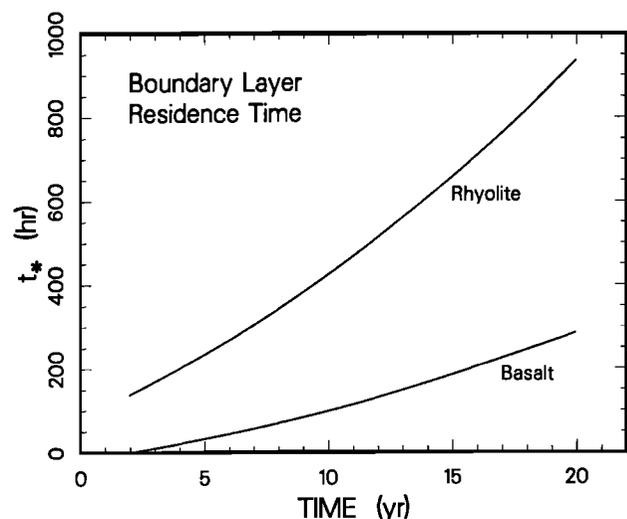


Fig. 6. The thermal boundary layer residence time is plotted as a function of time since emplacement for basalt and rhyolite in a 1-km cooling intrusion. The residence time may be thought of as the exposure time of a parcel of magma to a temperature gradient. Larger temperature gradients imply shorter residence times for magma within the boundary layer.

can also write the heat flux as

$$Q = \frac{k\Delta T}{\delta} \quad (8)$$

The layer thickness δ may then be identified with the denominator of the preceding equation, that is,

$$\delta = (\pi\kappa t)^{1/2} \quad (9)$$

Now, to obtain t_* , we must select the value of δ for which the thermal will break away from the boundary. In principle we could use the theory for the Rayleigh-Taylor instability to determine the value of δ , but no model of this mechanism has been developed for fluids with strong viscosity variations. For his calculations, Howard obtained empirical estimates of δ from experiments. Similarly, we can use estimates of the thermal layer thickness obtained from the parameterized results illustrated in Figure 5. Figure 6 shows our estimates of the fluid residence times obtained from equation (9) for the two magma compositions that we are considering. For the basalt, residence times vary from about 2 hours to 300 hours in the first 20 years after emplacement, while for the rhyolite the residence times range from 150 to almost 1000 hours during the same period. These values should be treated as order of magnitude estimates of the residence times for magma within the boundary layer. In this respect, they may also be thought of as representing estimates of the times during which significant fractionation can occur in the thermal boundary layer or at the margin subjected to the temperature gradient characterizing the layer.

4. POSSIBLE EFFECTS OF THERMOCHEMICAL PROCESSES

Experimental evaluations of the Soret effect as applied to magmatic processes have revealed that sizable chemical fractionations can be produced by imposing a thermal gradient upon a melt, albeit of extraordinary magnitude [Walker *et al.*, 1981; Lesher *et al.*, 1982; Walker and DeLong, 1982; Ponander and Mahood, 1984]. Recent studies of silicic volcanic regions [Hildreth, 1979, 1981; Smith, 1979; Mahood, 1981] have suggested that magmatic Soret processes are the dominant mechanisms for the chemical fractionation that is ultimately observed in the erupted ash flow tuffs. However, the observed chemical trends are not in agreement with the results of some recent Soret experiments for rhyolitic magma [Lesher *et al.*, 1982; Ponander and Mahood, 1984] and suggest a reevaluation of crystal fractionation processes for generation of compositional zoning in magma [see Michael, 1983]. Nevertheless, there has been no quantitative analysis of the significance of Soret-related processes in convective environments under a variety of thermal and compositional conditions.

4.1. Soret Diffusion in a Convection-Free Environment

Soret diffusion is defined as the mass transport response of a chemical system to a thermal gradient. (Note that Soret diffusion, thermal diffusion, and thermodiffusion are all names utilized in the literature for thermal-gradient-induced mass transport). Given an initially homogeneous multicomponent system which is nonisothermal, certain chemical species will migrate toward the higher-temperature region, while other species migrate toward the lower-temperature region. This process will continue until a steady state is reached when the opposing effect of chemical diffusion is sufficient to balance the flux of material induced by the Soret process. This Soret steady state (often misnamed Soret equilibrium) will persist as long as the temperature gradient is maintained in the system.

The discussion that follows will examine the possible degrees of chemical separation (fractionation) which can be obtained by Soret processes for end member magma compositions (basalt and rhyolite). A static or convection-free (true Soret diffusion) thermal regime is examined first.

The following one-dimensional differential equation provides us with a time dependent description of the Soret process in a static fluid [deGroot and Mazur, 1962; Powers, 1962; Grodzka and Facemire, 1977]:

$$\frac{\partial C}{\partial t} = -\frac{\partial J_x}{\partial x} \quad (10)$$

where the mass flux term J_x is defined as

$$J_x = -D\sigma C(1 - C)\frac{\partial T}{\partial x} - D\frac{\partial C}{\partial x} \quad (11)$$

D is the chemical diffusion coefficient of the fluid, σ is the Soret coefficient, C denotes the concentration of a particular chemical species (assume for simplicity a binary chemical system), and T is the temperature. Equation (10) can be solved subject to the condition that $J_x = 0$ at the boundaries of the domain, that is, the mass flux at the margins vanishes. The first term of equation (11) denotes the contribution of Soret diffusion induced by the thermal gradient, while the second term represents the opposing effect of Fickian diffusion. The Soret coefficient σ has the convention of being positive when mass transport occurs from a region of high temperature to one of low temperature.

For examining the extent of chemical separation in a magma, it is useful to define a nonlinear fractionation term q :

$$q = \frac{C_h(1 - C_c)}{C_c(1 - C_h)} \quad (12)$$

Again, C denotes the concentration with the subscripts h and c denoting either the hot or cold end of the system, respectively. For simplicity we deal with a binary system so that q is defined in terms of a single component. If there is an enrichment of the component in the hot zone of the system, q will be greater than unity (negative σ), whereas if the component becomes enriched in the cold zone, q will be less than unity (positive σ). Note that q reduces to a simple partition term ($q = C_h/C_c$) for the case of a minor or trace component of a pseudobinary chemical system.

Because of our interest in the greatest possible chemical fractionation that can be obtained from a Soret process, we initially consider the steady state solutions. In the steady state, at which time the net flux J_x is zero, the solution to equation (11) is simply

$$\ln q_{ss} = \sigma\Delta T \quad (13)$$

where ΔT is the temperature difference between the hot and cold ends of the system. We assume that σ and D are constant over the concentration range of interest and that a linear temperature gradient is achieved within a very short time relative to the rate of mass transfer. The time for the system to reach Soret steady state (t_{ss}) is given by

$$t_{ss} = \frac{4d^2}{\pi^2 D} \quad (14)$$

Here, the additional term d is the width of the Soret system which may be identified with the boundary layer thickness δ . The steady state time results from the time dependent solution of the mass transport equations with zero mass flux at the boundaries [see Powers, 1962] and in the case of equation (14)

TABLE 1. Values of Dynamical Properties and Chemical Transport Parameters Used in Model Calculations

	Value
Dynamical property values	
α	$4 \times 10^{-5} \text{ deg}^{-1}$
κ	$3.8 \times 10^{-7} \text{ m}^2/\text{s}$
μ_{basalt}	$10^2 \exp(0.04605\Delta T) \text{ P}$
μ_{rhyolite}	$10^6 \exp(0.04605\Delta T) \text{ P}$
ρ	$2.6 \times 10^3 \text{ kg/m}^3$
g	9.81 m/s^2
H	$2 \times 10^2 \text{ W/m}^2 \text{ deg}$
k	1.25 W/m deg
C_p	1.255 J/kg deg
Chemical transport values	
$D_{\text{basalt}} (1200^\circ\text{C})$	$8 \times 10^{-12} \text{ m}^2/\text{s}$ [Hofmann, 1980]
$D_{\text{rhyolite}} (950^\circ\text{C})$	$1 \times 10^{-12} \text{ m}^2/\text{s}$ [Hofmann, 1980]
σ	thermal diffusion coefficient
	isothermal diffusion coefficient
σ_{basalt}	$-3 \times 10^{-3} \text{ deg}^{-1}$ [Walker et al., 1981]
σ_{rhyolite}	$-1 \times 10^{-3} \text{ deg}^{-1}$ [Lesher et al., 1982]

represents the time required to each $(1 - e^{-4})$ or 98% of the steady state separation. Equation (14) is appropriate for temperature differences of less than 1000°C in magmatic applications (see below) otherwise a more complicated expression is required [see Grodzka and Facemire, 1977].

Using values listed in Table 1, equation (13) has been evaluated for a range of temperature differences. Figure 7 gives an idea of the extent of chemical fractionation that can be obtained for a simple nonconvecting Soret cell. The separation factor q_{ss} is plotted with respect to the temperature difference between two confining walls. We have taken the chemical species to be enriched at the hot wall using the experimental Soret coefficients derived by Walker et al. [1981] for basaltic magmas. The smaller Soret coefficient for the lower temperature rhyolite magma [Lesher et al., 1982] was utilized and follows due to the highly polymerized nature of the silicic melt relative to the basaltic data. It should be noted that the experimental values for σ were derived at high temperatures (1500°C – 1700°C) and therefore can be considered as overestimates of the Soret values required for the present applications. Note that the fractionation term at steady state is not a function of

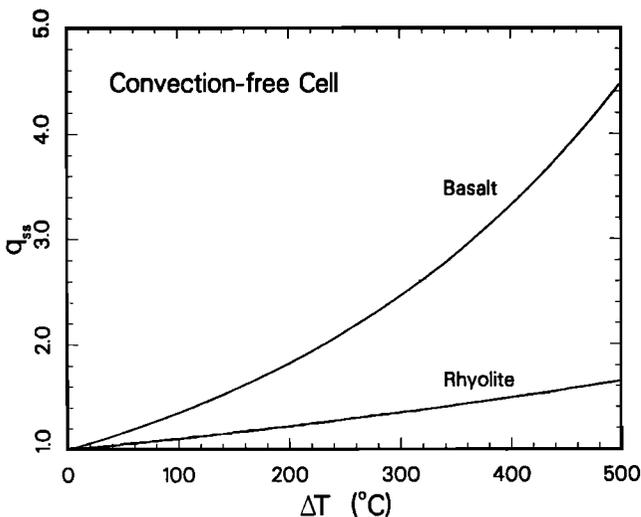


Fig. 7. The steady state Soret fractionation for magma in a convection-free cell as a function of the temperature difference between cell walls.

the width of the Soret cell (boundary layer thickness). However, equation (14) demonstrates that the cell width, and hence thermal gradient, will determine the time required to reach steady state and the attainment of q_{ss} . (It is noteworthy that the time to reach steady state is inversely proportional to the chemical diffusion coefficient and therefore suggests that any nonconvecting Soret fractionation process will be limited by the diffusive remixing.) From Figure 7 the final value of q would be less than 1.05 for either basalt or rhyolite assuming a 200°C/m temperature gradient and less than 1.2 for a 400°C/m gradient. (We reiterate that both gradients are considered to be extreme upper bounds for thermal convection.) Figure 8 provides the steady state times required to achieve the separations indicated in Figure 7. For diffusion lengths of only 0.1 m, 30 years would be required to reach steady state in the basalt, while 300 years would be required in the rhyolite. Clearly, the residence times of magma parcels in the horizontal thermal layers would obviously limit the importance of the fractionation process within the layer itself. From Figure 6 the residence time of a magma parcel in the boundary layer characterized by the smaller gradient is only 450 hours in rhyolite and 300 hours in basalt. For a 400°C/m gradient the residence times are about 180 hours for rhyolite and 100 hours for basalt. In the final analysis, thermal layer magma will not be subjected to a temperature gradient long enough for Soret steady state to be approached before it breaks away from the margin. Even if significant fractionation did occur within the layer, when it fell into the interior, remixing of the fractionated components would be expected. Furthermore, the occurrence of large thermal gradients in special situations as when a cooler mid-ocean ridge basalt (MORB) magma overlies a hotter picritic magma really does not improve the environment for Soret processes. Using the results of Huppert and Sparks [1980], Walker and DeLong [1982] have suggested that temperature contrasts of 100°C or more might persist for more than a month within a layer 0.01–0.02 m thick at the interface between the two layers. But such a layer formed by cooling from above would be gravitationally unstable and from equation (9) would be characterized by a residence time of about 2 min! Solution of the time dependent species transport equation (equation (10)) for a 0.01-m layer having a thermal gradient of 10^4°C/m yields a concentration enhancement of only 0.002% after 2 min (R. T. Cygan and C. R. Carrigan,

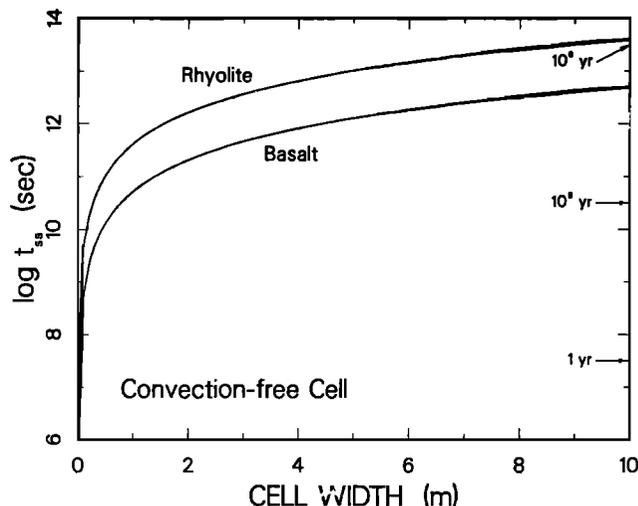


Fig. 8. The logarithm of the time to reach Soret steady state as a function of the cell width for magma in a convection-free cell.

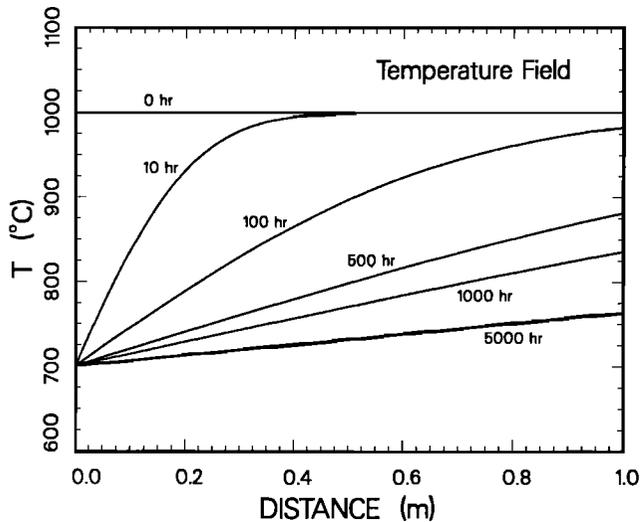


Fig. 9. Evolution of temperature field at a magma chamber margin with a fixed temperature boundary. The analytical results are derived using equation (4-58) of Eckert and Drake [1972].

unpublished manuscript, 1986). Even for other fractionation mechanisms, this period of time is likely to be too brief to be significant. In general, the existence of large boundary layer thermal gradients suggests very short magma residence times in the boundary layer, a fact that does not depend on the source of buoyancy but does depend on the thermal diffusivity.

Walker and DeLong [1982] have suggested that the Soret signature could be preserved in a magma body by solidification of Soret fractionates from the thermal boundary layer regime. We expect that this is likely to be the only way, if at all, that the effects of Soret diffusion could ever be preserved in a convecting system. It appears reasonable that solidification at an advancing margin could "freeze in" very thin zones exhibiting a concentration gradient before the associated magma parcel breaks away. But again, the thermal boundary layer would be very far from a Soret steady state. As noted above, the non-steady state solutions to equation (10) (R. T. Cygan and C. R. Carrigan, unpublished manuscript, 1986), subject to boundary conditions requiring zero mass flux through the system boundaries, permit an estimate of the maximum concentration enhancement that can occur prior to boundary layer disruption. For these calculations we include the time dependence of the thermal regime ($T = f(x, t)$) simulating the propagation of the thermal field through the hot magma parcel as it cools while in contact with a constant temperature margin. The results, obtained using an implicit finite difference solver, are presented in Figures 9 and 10. Figure 9 illustrates the evaluation of the temperature field in the magma parcel as a function of time since initial contact between the parcel and the constant temperature margin. For a pseudobinary system the resultant distribution of the oxide component in the boundary layer is shown for the equivalent times in Figure 10. The maximum concentration enhancement occurs at the margin but falls off to its initial value at only 0.02 m from the boundary. After a period characteristic of the boundary layer detachment time the maximum concentration enhancement is found to be less than 0.04%. This value must be interpreted as an upper limit on any possible enhancements of concentration owing to the particular model chosen for the thermal evolution of the magma parcel in the vicinity of the margin.

4.2. Chemical Separation by Thermogravitational Mechanisms

Another mechanism has been proposed for vertical thermal boundary layers which might, in principle, overcome some of the time scale problems associated with convection-free Soret fractionation in horizontal thermal layers. Unlike horizontal thermal layers, flow in vertical layers will not be characterized by such extreme time dependence [Spera *et al.*, 1982]. For a sufficiently long vertical margin one might expect that fluid within an associated vertical thermal layer could remain there for an arbitrarily long time depending on the velocity within the layer. It has been suggested, although not in detail, by Hildreth [1981] and Schott [1983] that some kind of interaction between wall rock and the vertical thermal layer could result in a fractionation process involving both convection and Soret diffusion. Presumably, because of either melting or crystallization at the vertical margin of the body, a thin, less dense layer might form in the thermal layer. Owing to its positive buoyancy, the thin chemically distinct layer would rise creating a counterflow relative to the downward thermally driven flow which itself results from heat loss through the margin. The counterflow occurring within a zone of significant temperature variation, that is, the thermal layer, is suggestive of conditions required for thermogravitational fractionation (TGF) [Clusius and Dickel, 1938; Furry *et al.*, 1939; Jones and Furry, 1946; Powers, 1962].

Thermogravitational fractionation is similar to Soret separation except that a convecting cell is utilized to prevent diffusive remixing of the Soret fractionate. The presence of a circulating current sweeps material away normal to the thermal gradient so that back diffusion is limited and a simple Soret steady state is prevented. The convective flow required by TGF or Clusius-Dickel (CD) cells is induced by thermal buoyancy resulting from the different temperatures of the cell walls. Figure 11a provides a schematic of a classic Clusius-Dickel cell of length L and width d . The wall temperatures are maintained constant, and a linear thermal gradient across the fluid

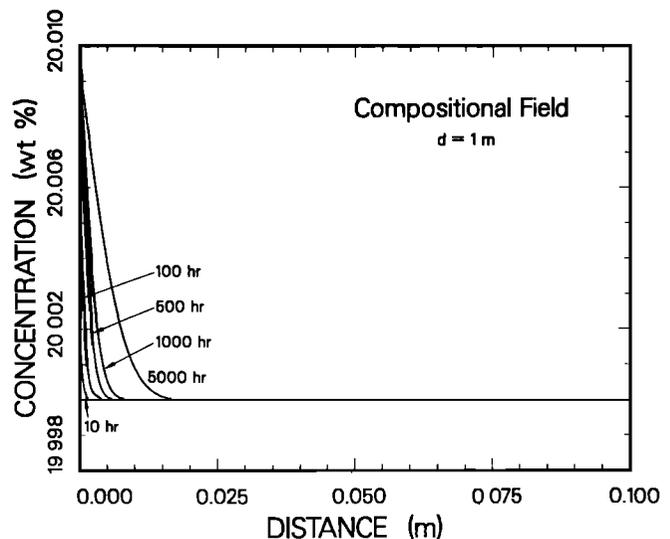


Fig. 10. Evolution of a compositional field near the extreme edge (first 0.1 m) of a magma chamber margin ($d = 1$ m) as induced by Soret diffusion and corresponding to the time dependent temperature field presented in Figure 9. The spatial distribution of the idealized component across the entire Soret cell as a function of time is determined by an implicit central difference numerical model (R. T. Cygan and C. R. Carrigan, unpublished manuscript, 1986).

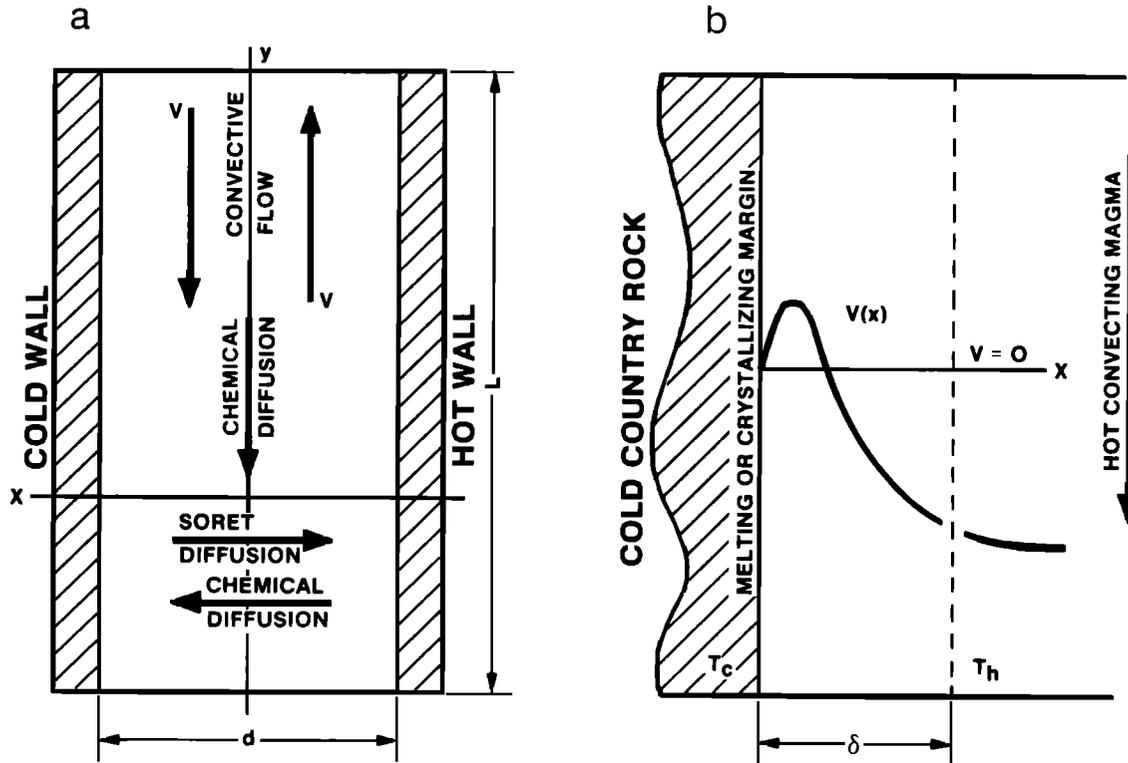


Fig. 11. (a) Schematic of a classical Clusius-Dickel or TGF cell denoting the major Soret diffusion, chemical diffusion, and convective fluxes. (b) Model of a magmatic thermal boundary layer showing the expected thermal gradient layer and principle convection flow. The velocity field required for proper CD separation is indicated in the boundary layer.

is assumed. Soret diffusion tends to enrich the hot rising boundary layer in one of the chemical components producing a chemical gradient. The enriched material is swept up the hot wall by the hotter and less dense fluid thereby limiting the chemical back diffusion. Similar behavior occurs for the depleted fluid at the cold wall except in the opposite direction.

The fundamental differential equation [Jones and Furry, 1946; Powers, 1962] is provided below and follows as an extension of equations (10) and (11) with the addition of an advection term:

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2} + \frac{\partial^2 C}{\partial y^2} + \sigma D \frac{dT}{dx} \frac{\partial[C(1-C)]}{\partial x} - v(x) \frac{\partial C}{\partial y} \quad (15)$$

Chemical diffusion along the vertical direction (y axis) of the CD cell has been introduced as well as a new term involving $v(x)$, which denotes the change in concentration resulting from the convection current. A mean convection velocity is used to obtain an analytical solution to equation (15). As in the convection-free case, with the boundary conditions of zero mass flux across the cell walls, a flux equation can be obtained which defines the net transport τ of one component through a plane perpendicular to the walls of the CD cell [Jones and Furry, 1946; Powers, 1962]:

$$\tau = MC(1-C) - K \frac{dC}{dy} \quad (16)$$

The new terms introduced in this solution are given below:

$$M \equiv \frac{d^3 \rho g \alpha B \Delta T^2}{6! v} \quad (17)$$

$$K = K_C + K_D \quad (18)$$

$$K_C \equiv \frac{d^7 \rho g^2 \alpha^2 B \Delta T^2}{9! v^2 D} \quad (19)$$

$$K_D \equiv d \rho D B \quad (20)$$

The M term represents the positive contributions of Soret diffusion and convective transport to the chemical separation, while K_C denotes the convective remixing and K_D represents the chemical diffusion back down the cell. The B term refers to the breadth of the CD cell and will eventually cancel out in the equations for q_{ss} and t_{ss} . Figure 11a illustrates the major fluxes that are incorporated in the model.

If we examine the case of chemical separation at steady state ($\tau = 0$), it is possible to obtain an exact solution for the concentration change by integrating equation (16). The steady state result can be presented in terms of the fractionation term q_{ss} :

$$\ln q_{ss} = 2AL \quad (21)$$

L is defined as the length of the CD cell for the present application. The A term represents a ratio of the separation forces to the remixing forces:

$$A \equiv M/2K \quad (22)$$

Application of CD theory to magma chambers is not straightforward since reservoirs of magma are likely to differ in many important respects from the ideal CD cell. Simple convective flow down a cool chamber margin does not resemble the closed cell circulation required for operation of a CD cell. In the cell, Soret-separated components tend to migrate to opposite ends of the cell but in the case of magma convection, the flow entering the thermal layer from the reservoir at the top is returned to the reservoir at the bottom. If, however,

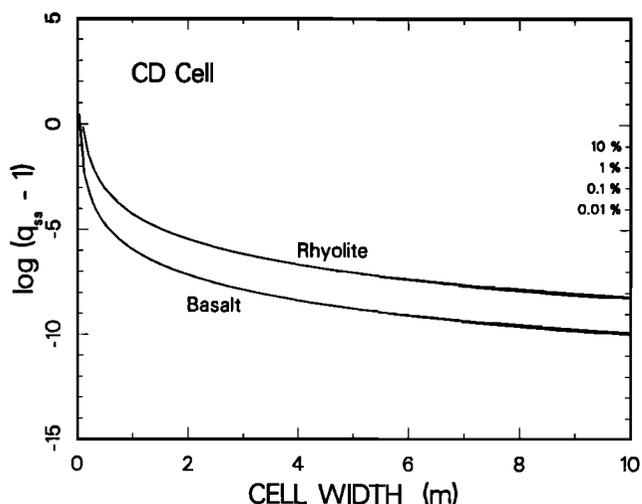


Fig. 12. The steady state fractionation of magma in a CD cell of 1-km length as a function of the cell width. The logarithm of $(q_{ss} - 1)$ has been utilized to express the wide range of q_{ss} values and the almost negligible separation that is expected for magmas.

a counterflow in the thermal layer is present as was described at the beginning of this section, the potential exists for removing the Soret fractionate from the thermal layer before remixing in the reservoir occurs. Whether a counterflow as shown in Figure 11b can exist for realistic thermal boundary conditions is not known. Certainly a buoyant, chemically distinct layer resulting from crystal plating [McBirney, 1980; Nilson *et al.*, 1985] or melting at the margin of a chamber can rise if it is sufficiently thick. However, Spera *et al.* [1984] argue that double diffusive counterbuoyant flow may be outside the parameter regime characterizing most magma chambers. Furthermore, there are some data to suggest that overlying layers, typically 500 m deep, form in silicic chambers on a time scale of 10^5 years [Spera and Crisp, 1981]. To form such a layer in a chamber having a horizontal to vertical aspect ratio of 2 would require assimilating wall rock at the rate of only 0.005 m/yr in the case of a counterbuoyant flow produced by melting. Such an extremely low production rate of silicic melt will

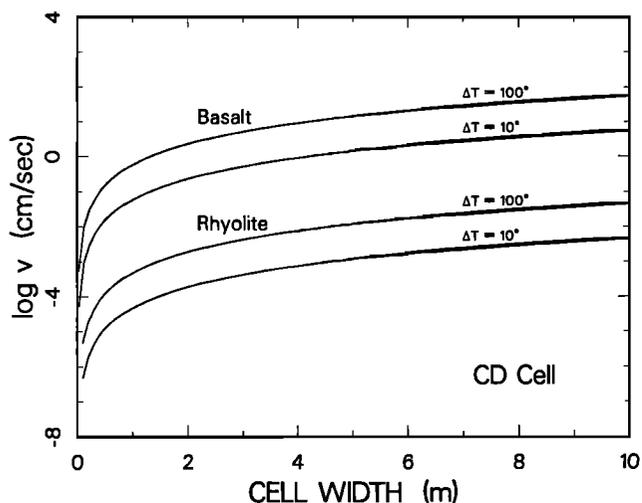


Fig. 13. The logarithm of the mean convection velocity for magma in a CD cell as a function of the cell width and for two wall temperature differences. Magma velocity calculations are based on the thermal buoyancy expected along the linear thermal gradient of the cell.

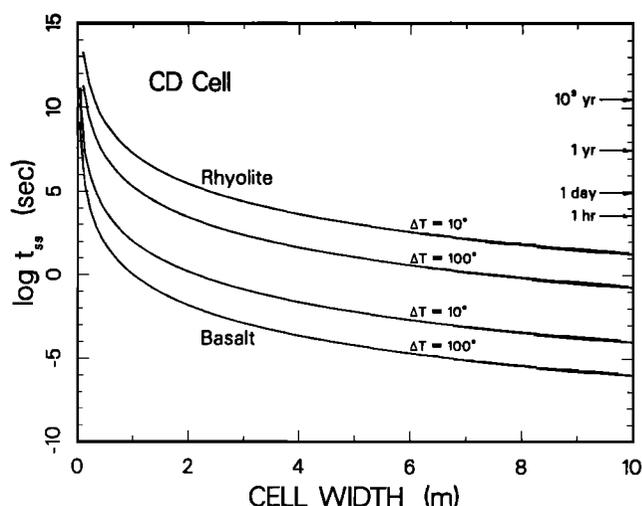


Fig. 14. The logarithm of the time to reach steady state for magma in a CD cell of 1-km length as a function of cell width for two wall temperature differences.

not likely give rise to a counterbuoyant flow in a system where convection is considered significant. It is much more likely that the downflow along the side of the chamber will scour the margin. The melt which is removed will then be carried into the interior of the chamber where it will begin to rise in little parcels toward the roof zone.

For the sake of discussion, we take the kinematic approach by assuming that a counterflow exists. Then we determine the possible effect on separation of a magma boundary layer treated as an ideal CD cell. In applying equation (21) to a magmatic system, we have chosen the physical parameters outlined in Table 1. The chemical separation of a component induced by Soret diffusion and enhanced by convection is presented in Figure 12 for the two different magmas. The temperature of the magma was taken to be approximately the liquidus value for the compositions considered. Figure 12 provides a plot of chemical fractionation at steady state as a function of the cell width, the latter being equated to the thickness of the thermal boundary layer. Because the scale of q_{ss} covers such a wide range, we have chosen the function $\log(q_{ss} - 1)$ to display the results. The plot shows that it is necessary to have cell widths less than 0.1 m to provide any sizable (or measurable) fractionation (10%) of a magma component. At these smaller cell widths it is significant to point out that there is almost a runaway effect in the magnitude of the fractionation. Factors as large as $q_{ss} = 10^5$ can be obtained in magmas, albeit under artificial thermal boundary conditions. For silicic magma chambers, Spera *et al.* [1982] determined that the characteristic boundary layer thickness at a vertical wall will be of the order of tens of meters. If we were to examine the TGF effect for even a 1-m-thick boundary layer, the chemical separation is insignificant. For a CD cell width of 1 m, Figure 12 shows that a rhyolite magma will have a q_{ss} value of 1.000056, while for a basaltic magma the steady state value is only 1.0000011. Also noteworthy is that q_{ss} is independent of ΔT except for very small cell widths (less than 0.03 m for a basalt or 0.1 m for a rhyolite magma). Negligible Soret diffusion occurs across the larger cell distances. Figure 13 provides the mean velocity values for these examples in both a basaltic and rhyolitic magma. For cell widths of about 1 m, these velocities are comparable to velocities expected in thermally driven magma convection models.

As for convection-free Soret diffusion, it is worthwhile to examine the time required to achieve steady state fractionation. The decay time for the solutions of equations (15) and (16) is [Jones and Furry, 1946]

$$t_{ss} = \frac{8\rho dB/AM}{1 + (\pi/AL)^2} \quad (23)$$

The steady state time represents the time to reach 98% of the steady state separation. The results are provided in Figure 14 for both magma types as a function of the cell width and for two expected temperature differences. Extremely small cell widths are characterized by considerably less convection and require more time to develop the steady state compositional profile, whereas the convective velocities are more significant at the greater widths (see Figure 13). The times required to reach steady state for a 1-m-thick boundary layer range from a few seconds for a basaltic magma at 100°C/m to 6 years for a rhyolite magma at 10°C/m. Unfortunately, the corresponding fractionation values presented in Figure 12 are much too insignificant to make these steady state times meaningful. Nevertheless, for significant fractionation to occur as associated with the very small cell widths, steady state times undoubtedly exceed likely residence times of magma in the boundary layer.

5. CONCLUSION

We have attempted to resolve the relationship between the convective thermal boundary layer regime in magma and chemical separation by Soret and thermogravitational processes. In the case of thermal layer formation on horizontal chamber margins, temperature gradients and magma residence times are not appropriate for significant Soret fractionation to occur. This conclusion is not affected by the type of buoyancy driving convection. Extreme upper limits of 0.04% fractionation of magma "frozen" into an advancing solidification front are obtained.

Analysis of the role of convection on a vertical margin as a mechanism permitting thermogravitational separation is more difficult owing to the significant differences existing between the thermal layer regime in magma chambers and classical Clusius-Dickel separation columns. Whether or not the required counterflow regime can exist remains an open question. However, a kinematic flow model for an ideal CD cell is developed using magma properties to obtain estimates of chemical fractionation. Values of fractionation smaller than those obtained in the horizontal case are predicted.

Our analyses of Soret processes have been limited to the dynamically significant parts of convecting magma chambers. These processes may also exist in the marginal zones that do not participate in convection. Magma in the stably stratified layer forming at the chamber bottom (Figure 1) has an indefinite residence time and would be subjected to a temperature gradient there. However, the temperature gradient in the stable layer will be smaller and its rate of decay greater than the gradient occurring in a magma body that is cooling by conduction only. At the top of the chamber, magma in the extremely viscous partial melt zone can also have a long residence time. Nonetheless, the temperature gradient there will not significantly differ from the purely conductive case because the host regime controls heat transfer and not convection in the magma body. We conclude that separation in these stagnant zones will be less than or, at most, comparable to separation occurring in a magma body cooled only by conduction.

Bowen [1928] dismissed Soret diffusion as a significant fractionation mechanism in magmas cooling by conduction only. We find that magma convection does not alter this conclusion. In fact, magma convection will tend to produce chemical separations smaller than those associated with a body cooling by conduction alone.

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