Evaluation of Chemical and Soret Diffusion at Crystallizing Boundaries

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The effect of thermal or Soret diffusion on differentiation in layered intrusions has recently been reexamined for explaining marginal reversals (Latypov, 2003). Previously, Lesher (1986) and Lesher and Walker (1986) provided a detailed analysis of the potential role of Soret diffusion in magmatic systems and published experimentally determined Soret coefficients for different composition melts. In these papers and more directly in numerical studies done by Cygan and Carrigan (1992) the effect of Soret diffusion compared to chemical diffusion was shown to be relatively minor, because of the roughly three order of magnitude smaller Soret coefficient and the relatively small thermal gradients expected in magmatic systems. Although Cygan and Carrigan (1992) provided numerical solutions to the coupled Soret – chemical diffusion problem for binary systems in an above-liquidus magma, they suggested that the minor effect they showed would be even smaller in subliquidus magmas.

Given the coupled nature of heat and mass transfer at the margins of intrusions, complicated by crystallization and convection, it is not always evident what the evolution of mineral abundances, compositions, and the melt evolution would be at such interfaces. In this contribution, a numerical study is presented to provide some insight into the effects of these coupled processes on igneous differentiation. The starting point for this study is the coupling of heat transfer, chemical diffusion, and crystallization as described in Sonnenthal and McBirney (1998). The methodology has been extended to account for the full chemical diffusion tensor in a twelve-component system, including the Soret coefficients for the diagonal components. Fluxes owing to chemical and Soret diffusion in a partially crystallized magma can be written as follows (modified after Lasaga, 1998):

$$J_i = -D_i \phi \sigma_i X_i (1 - X_i) \nabla T - D_i \phi \nabla X_i$$

where J_i is the sum of the diffusive fluxes for the each component i, X_i is the mole fraction of component i, D_i is the chemical diffusion coefficient, σ_I is the Soret coefficient, ϕ is the melt fraction, and ∇ is the gradient operator. These diffusive fluxes are then included in the full equation for the evolution of chemical species over time, considering mass balance, mass transport, and crystal-liquid reactions for a multicomponent system.

Data for Soret diffusion coefficients in a basaltic liquid were obtained from Lasaga (1998), which are based on the experiments by Lesher (1986). The Soret coefficient for H_2O was given a value (2×10^{-2}) approximately three times that of Na and twice that of K, based on the observation that H_2O is observed to diffuse to the hot end in fluid inclusions in halite and in NaCl-rich brines (Cygan and Carrigan, 1992). Chemical

diffusion coefficients and cross coefficients for oxides and H_2O were based on experimental data from various sources or estimated.

The modifying effects of Soret diffusion on chemical diffusion are potentially greatest for H_2O, because of its much larger chemical diffusion coefficient compared to the nonvolatile oxides and which, in the absence of hydrous minerals and below saturation, would increase as a conservative species in the zone of crystallization. This large increase in concentration leads to large compositional gradients, resulting in strong chemical diffusion. The zone of crystallization is also characterized by the larger temperature gradients in the intrusion, which would be conducive to Soret diffusion into the hotter magma ahead of the crystallization front. The increased diffusive fluxes for water would also result in a reduction of the melt viscosity and density in the boundary layer, thus making upward migration of volatile-rich melt at the sidewalls more effective. Yet, the effects of these processes, while intriguing, require a more rigorous analysis in terms of their magnitude in a strongly coupled system.

Therefore, three scenarios are examined numerically that could potentially be conducive to the effects of Soret diffusion on crystal-liquid relations, melt transfer, and metasomatic processes. The scenarios include crystallization at the base of an intrusion, sidewall crystallization, and interactions of cooler inclusions, such as the large blocks observed in the Skaergaard intrusion with hotter magma.