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Early nucleation and crystal growth in tabular mafic intrusions: insights from analogue solidifying aqueous ammonium chloride experiments

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Deciphering the early crystallisation history of magmatic intrusion from the geological record remains a major challenge. To model processes occurring in early stage crystallisation, we conduct analogue solidification experiments, providing us with the opportunity to quantify the rheological and thermodynamic behaviour of crystal rich magmas shortly after emplacement.

The aqueous ammonium chloride (NH4Cl) system has been used previously to study the crystallization of multicomponent systems that result in the formation of a crystal mush and the rise of compositional convection. The results of these experiments have been used to develop an understanding of microstructural evolution and the formation of stratified igneous intrusions. However, most of these published studies focus on eutectic or mushy modes of solidification, regimes where crystal kinetics facilitate the formation of crystallization in-situ (i.e. equiaxed regime) have been less well studied.

In this study, we focus on the early burst of crystal nucleation and growth in a super-cooled binary system to explore the importance of this stage in the evolution of magmatic intrusions. We performed analogue experiments using a solution of NH4Cl-H_2O at 30 wt.% concentration in a rectangular tank with Perspex walls and an aluminium thermally-controlled base plate. The solution was poured into the tank at 34°C, just higher than the liquidus temperature (33.5°C). Though cooled from below, the liquid convected strongly during the initial phase of crystal nucleation and growth. Nucleation occurred both at the cooled base plate and throughout the liquid. We recorded the thickness of the fully solidified layer and the overlying crystal mush, as well as the size of the crystals suspended in the convecting liquid and the temperature and composition of that liquid during each experiment.

The early evolution of the binary NH4Cl-H_2O system can be divided into three stages:
(1) Initially, rapid nucleation occurs in the bulk liquid and on the cooled base plate as the fluid near the base is supercooled to the eutectic temperature (Tc = -15 °C). This crystallization drives vigorous compositional convection throughout the fluid, despite the stabilising thermal forcing (cooling from below).
(2) Subsequently, the temperature rapidly approaches the liquidus temperature due to the latent heat released on the initial burse of crystallisation. As equilibrium is approached, nucleation rates significantly decrease and the size of the crystals in suspension increases. Settling of these crystals on the base plate creates a coarsening-upwards sequence. It is likely that the increase in particle size may be due to growth and synneusis in the convecting solution.
(3) Finally, the settling and growth of a significant crystal layer on the cold base plate thermally buffers the liquid from the cold base plate, and the systems returns to a mushy mode of crystallisation characterised by local thermodynamic equilibrium.

We conclude that in natural tabular intrusions, supercooling is responsible for early nucleation, creating an abundance of crystals. An approach to equilibrium would be then achieved by continuous crystal growth and clustering during the first two stages. These early-formed crystals will then settle to form the accumulation sequence found in preserved mafic sills.