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Notes

Capturing crystal growth

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The processes by which silicate melts transform from liquid to solid are central to igneous petrology, influencing the chemical diversity and spatial distribution of magmas in the Earth's crust. Growing appreciation for the rheological feedbacks controlling magma transport in subsurface conduits has led to an increasingly prominent role for solidification processes in volcanology as well. Advances in analytical instrumentation are sharpening the picture of crystallization as it occurs in nature, while high-temperature laboratory experiments are providing tighter constraints on the timing of solidification processes. Schiavi et al. (2009; p. 963 in this issue of *Geology*) study crystal growth in a silicate melt similar to natural basaltic trachy-andesite using a novel experimental apparatus that enables in situ observation of solidification. Critical to realizing the interpretive power of igneous rock texture is a firm understanding of the incipient stages of solidification, including nucleation and early crystal growth, a sequence that is typically obscured or temporally unconstrained in natural sample suites. Equally vital, and usually inaccessible from natural samples, is knowledge of how populations of crystals evolve in size and shape with time to create the final rock texture. Both objectives are well suited for experimental study using the techniques developed by Schiavi et al.

Dynamic crystallization experiments offer the opportunity to examine the response of a system to changing environment, an important aspect of crystallization in the mush zones of reservoirs, sills, dikes, volcanic conduits, and extruded lavas. The rate at which a thermodynamic driving force for solidification is applied determines both the sequence in which minerals precipitate and their proclivities to accommodate elements in non-stoichiometric or non-equilibrium proportions, and thus cooling rate exerts a fundamental control over the chemical compositions of derivative magmas. Equally profound are the consequences of cooling rate for rock texture. Contextualized observations of natural rocks have long indicated that rapid cooling leads to fine-grained crystal textures, whereas slower cooling produces larger crystals. However, the controlled environments afforded by laboratory experiments permit not only quantification of crystal nucleation and growth rates in response to specific cooling rates, but also exploration of kinetic effects such as delays in nucleation, the relationship between crystal growth mechanism and morphology, and interactions among minerals arising from boundary layers and epitaxy. Dynamic experiments have been used to constrain thermobaric histories of materials as diverse as granitic pegmatites (London, 1992), tholeiite lavas (Leshner et al., 1999; Lofgren, 1983), komatiites (Faure et al., 2006), lunar basalts (Grove and Walker, 1977), Martian meteorites (Hammer, 2009; McCoy and Lofgren, 1999), chondrules (Libourel, 2000; Zieg and Lofgren, 2006), and presolar grains (Mendybaev et al., 2002).

Particularly illuminating is the "snapshot" technique, in which a set of identical dynamic experiments is performed in parallel, varying only in duration and thus representing progression along a particular cooling path. In concept, each experiment captures the state of the system at a given point in time, allowing study of the evolution of crystal populations during a thermal history of interest. The approach has been fruitfully applied to one-atmosphere cooling of basalts (Pupier et al., 2008; Walker et al., 1978) and at elevated pressure in isothermal H₂O-saturated decompression of felsic magmas (Brugger and Hammer, 2010). However, the method is not without drawbacks. The fidelity of run products in recording

the high-temperature state of the system is contingent upon quenching, the rapid cessation of chemical transport. If the rate of reactions exceeds the rate at which the experiment is terminated, the run product represents an ambiguous amalgam of high- and low-temperature processes.

Experiments involving magma are technically challenging, with one problem being a limited range of non-reactive materials capable of containing silicate melts near their melting points (typically >1000 °C). Thus, most experiments at one atmosphere, and essentially all elevated-pressure experiments, are performed inside sealed capsules of silver, gold, platinum, or other (optically opaque) noble metals or alloys. Schiavi et al. introduce an instrumental innovation that departs from traditional capsule materials and enables in situ observation of magma crystallization at ambient pressure. The sample is enclosed by a glassy carbon gasket and sandwiched between anvils of moissanite, a synthetic hexagonal silicon carbide with physical properties similar to diamond (Xu and Mao, 2000). Continuous observation of the sample is made possible by transmitted light microscopy, as the entire apparatus fits inside an optical microscope. The advantages of this arrangement over snapshot-style experiments are manifold. Continuous observation obviates concern about quench effects because the experiment isn't terminated until the entire thermal history has been observed; the requirement of multiple identical runs is lifted; and the assumption that parallel experiments proxy the textural evolution of a given cooling regime need not be invoked. Most importantly, in situ observation allows observation of individual crystals through time.

The methodology of Schiavi et al. consists of three stages: glass fusion above the liquidus temperature of the bulk composition, a nucleation stage at 360 °C below the liquidus, and an observation stage of crystal growth at 260 °C below the liquidus. This methodology does not invite direct comparison of results with those of traditional dynamic experiments, which are typically initiated near the liquidus and cooled monotonically through time. Comparison with natural rock textures is similarly tenuous. However, because nucleation of the target phase (plagioclase feldspar) is negligible during the observation stage, this approach is ideal for studying various processes that increase the sizes of crystals, any of which could be important in natural settings.

Key observations reported by Schiavi et al. are made uniquely possible by the in situ technique. (1) Crystal growth is crystallographically anisotropic, giving rise to prismatic habit. The lengths of four unimpeded crystals increase at rates that are not only unvarying through time (≤ 120 min interval), but are also apparently uniform among grains. (2) Neighboring crystals of arbitrary orientation impinge and subsequently continue growing, resulting in crystallographically distinct subgrains mantled by an overgrowth rim. (3) Perhaps most intriguingly, a special case of impingement termed "coalescence" is documented (Schiavi et al., their figure 4) in which neighboring crystals aligned end-to-end merge into one crystallographically uniform grain. Natural magmatic phases frequently display evidence of impingement and overgrowth. Further analysis of in situ experimental run products, using electron backscatter diffraction analysis for example, may be helpful in determining whether this process is even more important than is currently recognized; evidence of very early impingement may be erased by subsolidus grain boundary migration. Because crystals appear to be randomly oriented following homogeneous nucleation during the first thermal stage, Schiavi et al. postulate that coalescence occurs after neighboring crystals rotate

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into alignment. Direct evidence for such rotation preceding coalescence is lacking in the data set; moreover, it seems unlikely that the surface free energy providing the driving force for coalescence would be sufficient to surmount the forces resisting prism rotation through a relatively viscous silicate melt. Nevertheless, if either simple impingement or coalescence is as widespread as the authors infer, the implications for evolution of crystal size distributions would be indeed profound. Declining frequency in small size classes and increasing frequency in large classes causes log-linear crystal size distributions to pivot counterclockwise, a progression commonly observed in suites of comagmatic rocks (e.g., Hammer et al., 1999; Higgins, 2002) but infrequently attributed to impingement and never previously ascribed to coalescence.

Means and Park (1994) demonstrated the potential of in situ observation of crystallization processes using saturated ammonium solutions, vividly demonstrating that textures are dramatically modified during crystallization by dissolution-reprecipitation, intra-phase grain boundary migration, and even phase boundary migration. What remained unproven was the degree of suitability of these materials as analogs for magma. Fifteen years later, we may be poised to discover whether individual crystals grow at constant rates (Marsh, 1998), rates governed by thermal and chemical fluctuations (Eberl et al., 1998), or rates that depend on size and/or density of surface defects (Eberl et al., 2002). Are rates highly sensitive to the appearance of new crystals in the system, and thus constrained in a mass-balance sense by the tendency to distribute solute evenly over all appropriate substrates (Larsen, 2005)? How critical is the degree of undercooling (Muncill and Lasaga, 1987), compared with advection and melt stirring (Kile and Eberl, 2003) in controlling the rate and mechanism of crystal growth? The apparatus reported by Schiavi et al. is so promising because silicate melts can be studied over the temperature range relevant to incipient crystallization processes, and over time scales relevant to geologic, particularly volcanic, environments. Continued investigation of growth processes over longer durations, including more crystals and with additional undercooling regimes, will be crucial for addressing the outstanding problems and continuing to develop kinetic theory of crystallization tailored to natural silicate magmas.

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