



Kinetics of cooling- and decompression-induced crystallization in hydrous mafic-intermediate magmas



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ABSTRACT

The crystallization of a hydrous basaltic-andesite magma was investigated using cooling and decompression experiments. These experiments were designed so that the driving force for crystallization of feldspar (the undercooling) is equal whether in the direction of cooling or decompression. After initial equilibration steps at conditions $T = 1025\text{ °C}$ and $P_{\text{H}_2\text{O}} = 150\text{ MPa}$, charges were either cooled or decompressed near-instantaneously to final conditions corresponding to undercoolings $\Delta T^* = 52\text{--}154\text{ °C}$, with dwell times of 12, 24 and 48 h at the final P – T . This ‘single-step’ experimental strategy (SSD for decompression experiments, SSC for cooling) allowed derivation of kinetic parameters such as nucleation and growth rates as a function of undercooling. Even though our setup was primarily designed to investigate feldspar crystallization, we report kinetic and geochemical data for all phases present (glass, feldspar, clinopyroxene, olivine and amphibole). At low to intermediate undercoolings, the solidification behavior of mafic-intermediate magmas is very similar texturally and compositionally whether the crystallization mechanism is cooling or decompression. At high degrees of undercooling, feldspar and clinopyroxene nucleation rates are nevertheless higher in SSD compared to SSC runs, perhaps due to slower diffusivities or larger nucleation barriers in the cooling runs. Minerals transition from interface-limited growth to diffusion-limited growth with increasing undercooling, while nucleation regimes shift from homogeneous to heterogeneous. Crystal morphologies are highly variable but can be used qualitatively to assess the extent of undercooling. Relative to more silicic melts with similar crystal contents, feldspar formed in andesitic melts nucleate at a lower rate but grow to larger sizes.

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1. Introduction

Magma crystallization under dynamic conditions is a key focus of volcanology because it largely controls their rheological behavior (e.g. Shaw, 1969; Lejeune and Richet, 1995), and because the crystals formed preserve petrological signatures of their storage and/or ascent histories (e.g. Blundy and Cashman, 2008; Chakraborty, 2008). Retrieving the solidification history of magmas from phase relations, their chemical traits, zoning patterns, or the textural characteristics of their mineral constituents requires a priori knowledge of the physics as well as the kinetics and energetics of crystallization (Hammer, 2008). In this respect, laboratory experiments carried out in controlled environments have allowed significant improvements in our understanding of magma solidification, and often provide the essential feed for predictive physical, numerical and thermodynamic models (e.g. Ghiorsso and Sack, 1995; Mastin, 2002; Toramaru, 2006). Early experimental studies consisted dominantly of dynamic cooling experiments, in which crystallization was stimulated by decreasing the temperature, thus applying a certain amount of undercooling ΔT ($\Delta T = T_{\text{liq}} - T$, the difference between the phase liquidus temperature and the ambient

run temperature). These early investigations dominantly employed mafic magma compositions, and shaped much of our current comprehension of crystal nucleation and growth kinetics through systematic examinations of the effects of varying melt composition (Kirkpatrick, 1974; Lofgren, 1974; Muncill and Lasaga, 1987), undercooling (Gibb, 1974; Lofgren, 1974; Nabelek et al., 1978; Tsuchiyama, 1985), cooling rates (Lofgren et al., 1974; Walker et al., 1976; Grove and Bence, 1979; Baker and Grove, 1985), superliquidus history (Gibb, 1974; Walker et al., 1978; Corrigan, 1982), pressure and water concentration in the melt (Baker and Grove, 1985), oxygen fugacity (Hill and Roeder, 1974), or the presence of a nucleation substrate (Berkebile and Dowty, 1982).

Studies focusing on crystal formation in more silicic compositions, while initially less numerous (Fenn, 1977; Swanson, 1977; Muncill and Lasaga, 1988), subsequently proliferated as decompression became recognized as one of the main driving forces behind crystallization during magmatic ascent (e.g. Lipman et al., 1985; Cashman, 1992; Geschwind and Rutherford, 1995; Métrich and Rutherford, 1998; Hammer et al., 1999). Crystallization induced by devolatilization of H_2O during decompression now has been studied experimentally in hydrous magmas of rhyodacitic to rhyolitic melt compositions (Hammer and Rutherford, 2002; Couch et al., 2003; Martel and Schmidt, 2003; Brugger and Hammer, 2010; Cichy et al., 2011; Martel, 2012; Mollard et al., 2012); fewer studies have focused on hydrous mafic or intermediate melts

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(Nicholis and Rutherford, 2004; Szramek et al., 2006), despite the growing interest of the community for groundmass textures investigations involving natural basaltic to andesitic samples (e.g. Taddeucci et al., 2004; Sable et al., 2006; Suzuki and Fujii, 2010). In contrast to cooling-induced crystallization, the driving force behind decompression-induced crystallization is the change in the mineral liquidus temperature caused by the loss of H₂O dissolved in the melt. At a given pressure $P_{\text{H}_2\text{O}}$, the difference between the hydrous mineral-in temperature and the ambient run temperature, termed the ‘effective undercooling’ ΔT_{eff} (Hammer and Rutherford, 2002), provided investigators with a means of direct comparison with the conventional undercooling ΔT . However, crystal nucleation and growth behavior driven by the two crystallization driving mechanisms, ΔT and ΔT_{eff} , were not investigated in a comparative sense. The comparison is fundamental to ascertaining whether crystallization kinetics are mechanism-dependent, but also potentially important in the interpretation of crystal textures in natural samples. Magmas may undergo both decompression and cooling during ascent, and a textural ‘fingerprint’ of one process or another would be helpful in piecing together magma ascent history.

By investigating crystal textures (e.g., abundance, number density, size and morphology) as a function of ΔT_{eff} or ΔT (noted ΔT^* if unspecified), experimental observations can be placed in the context of the classical analytical formulations of nucleation and growth (Kirkpatrick, 1974, 1981; Tsuchiyama, 1983; Muncill and Lasaga, 1987, 1988; Davis and Ihinger, 1998; Hammer, 2004, 2008; Shea et al., 2009). Among the key findings from these types of investigations is the notion that crystal textures are controlled by the interplay between nucleation and growth. That is, both crystal nucleation rate and growth rate exhibit asymmetrical bell-shaped curves with respect to thermodynamic undercooling, but the maxima occur at different values of ΔT^* (e.g. Kirkpatrick, 1981; Hammer and Rutherford, 2002). In effect, the degree of undercooling (and the rate at which undercooling is applied) defines the textural evolution of magmas: at small to moderate values of ΔT^* or $\Delta T^*/\Delta t$, growth typically dominates nucleation, leading to coarse crystallinity. At high values of ΔT^* or $\Delta T^*/\Delta t$, nucleation exerts the stronger control on texture and favors numerous small crystals (e.g. Lofgren et al., 1974; Swanson, 1977; Nabelek et al., 1978; Lofgren et al., 1979; Cashman, 1991). Hence, defining the nucleation and growth behaviors of crystal phases in magmas is a crucial step towards solving the inverse problem, i.e., what is the cooling or decompression history of a given volcanic rock collected at the surface (Cashman, 1991; Hammer, 2008 and references therein)? For a given mineral, however, substantial complications arise when considering that the amplitude, width, shape and position of nucleation and growth curves all depend on melt composition (e.g. Kirkpatrick, 1983; Davis et al., 1997). The characteristics of these curves have not yet been defined for hydrous magmas at the mafic and intermediate ends of the compositional spectrum. The occurrence of H₂O-rich magmas with basaltic-andesitic and andesitic compositions under arc-volcanoes (e.g. Ownby et al., 2011), and the potential for unveiling greater insights from rock microtextures motivate this work.

Through series of laboratory decompression and cooling experiments, we (1) determine rates of nucleation and growth of minerals (with a particular emphasis on feldspar) in a basaltic-andesite as a function of the temperature and pressure perturbations, (2) characterize qualitatively the evolution of crystal morphology with applied undercooling, (3) determine whether the kinetics of the two undercooling mechanisms (decompression and cooling) differ, and (4) compare the crystallization kinetics of hydrous mafic magmas with those of more evolved compositions.

2. Experimental approach

2.1. Choice of starting material

Natural crushed samples or synthetic compositions made from oxide reagents are the two types of starting material typically used

in magma crystallization experiments. Compared with natural samples, synthetic powders have the advantage that starting glasses/melts are completely homogeneous and that a state of total chemical equilibrium can theoretically be attained (Pichavant et al., 2007); synthetic melts are, however, not easily linked with natural magmas because they have undergone steps of high temperature superliquidus fusion, which are known to strongly modify the nucleation behavior of aluminosilicate melts (e.g. Walker et al., 1978; Sato, 1995; Davis and Ihinger, 2002; Pupier et al., 2008). In contrast, the use of natural starting materials often requires a careful step of coarse crushing to avoid exposing phenocryst cores that may be in disequilibrium with the groundmass (i.e. only a state of partial equilibrium can ever be achieved), but are more directly comparable to natural magmas in terms of crystallization kinetics (Pichavant et al., 2007; Hammer, 2008). As in natural magmas, experimental melts obtained from these starting materials contain pre-existing phases that may influence crystal nucleation in their direct vicinity (e.g. Fokin et al., 1999). Hence, even though synthetic starting materials provide a more homogeneous medium for nucleation, we follow Lesher et al. (1999), Hammer and Rutherford (2002) and Shea et al. (2009), and favor natural starting materials because they yield initial conditions more akin to those of natural magmas. For our experimental series, a natural sample from a Mascota volcanic field (Mexico) lava flow with a basaltic-andesite bulk composition (~55.7 wt.% SiO₂, ~5.2% Na₂O + K₂O, Table 1) and with an andesitic groundmass composition (~58.8 wt.% SiO₂, ~5.6% Na₂O + K₂O) was used as starting material (sample Mas22, kindly provided by the Smithsonian Department of Mineral Sciences). The bulk composition is germane to mafic-intermediate magmas erupted from arc volcanoes. This rock was characterized petrologically by Lange and Carmichael (1990), and was later the subject of a phase equilibria study by Moore and Carmichael (1998). The sample is homogeneous, contains microphenocrysts and phenocrysts of feldspar (13.1 vol.%), clinopyroxene (7.5%), and olivine (7.4%) and has a glass-poor groundmass (72 vol.%) consisting of feldspar, clinopyroxene, orthopyroxene (Carmichael et al., 1996). The initial conditions for the experiments were selected so that the groundmass of this starting material becomes the *reactive magma* (i.e. the portion of the melt-crystal system that is in equilibrium, as opposed to the phenocryst interiors, which are out of

Table 1
Chemical compositions of starting materials used in this study.

	Mas22 bulk composition (wt.%) ^a	Mas22 fused groundmass glass composition (wt.%) ^b
SiO ₂	55.65	58.78 (0.87)
TiO ₂	0.74	0.87 (0.05)
Al ₂ O ₃	17.53	18.66 (0.35)
FeO	6.02	5.6 (0.11)
MnO	0.12	0.11 (0.005)
MgO	6.73	3.32 (0.17)
CaO	7.33	6.77 (0.45)
Na ₂ O	4.00	4.08 (0.11)
K ₂ O	1.19	1.51 (0.15)
P ₂ O ₅	0.27	0.3 (0.02)
H ₂ O	0.61	4.34 ^c
Total ^d	99.28	
n		5 × 4 ^e
		Starting conditions
t (h)		> 30
T (°C)		1025
P (MPa)		150
fO ₂		ΔNNO + 0.5 to + 1

^a Analyses from Carmichael et al., 1996, recalculated on a dry basis.

^b Glass compositions analyzed in five different batches of annealed starting material with propagated standard deviations.

^c Calculated using H2OSOLvX1 (Moore et al. 1998).

^d Raw uncorrected totals.

^e Each of the five starting glasses was analyzed four times.

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