Felsic Melt and Gas Mobilization During Magma Solidification: An Experimental Study at 1.1 kbar

Mattia Pistone1,2*, Lukas P. Baumgartner1, Florence Bégué1, Paul A. Jarvis3, Elias Bloch1, Martin Robyr1, Othmar Müntener1, Thomas W. Sisson4 and Jon D. Blundy5

1 Institute of Earth Sciences, University of Lausanne, Lausanne, Switzerland, 2 Department of Geology, University of Georgia, Athens, GA, United States, 3 Section of Earth and Environmental Sciences, University of Geneva, Geneva, Switzerland, 4 US Geological Survey, Menlo Park, CA, United States, 5 School of Earth Sciences, University of Bristol, Bristol, United Kingdom

Melt and gas transfer processes are essential to the formation and growth of the Earth’s crust and for sustaining volcanic activity. These processes also play a major role in magma fractionation at shallow depths (<10 km) where magmas stall rheologically and solidify. In this scenario, the conditions of melt and gas mobilization during progressive cooling of crystal mushes down to their solidus remain poorly understood. We present experimental data (at 1.1 kbar) showing how a combination of temperature and crystal content control the ability of melt and gas to escape from cooling and solidifying hydrous silicic magmas with initial crystal volume fractions (Φ) of 0.6, 0.7, and 0.8, and for temperature snapshots of 850, 800, and 750°C. Microstructural observations and chemical data show that the amount of extracted melt increases by 70% from 850 to 750°C and by 40% from Φ = 0.6 to 0.8 at 750°C, due to the formation of interconnected crystal frameworks, gas expansion in constricted pore space, and filter pressing during cooling. As a result, our experiments suggest that melt and gas extraction from cooling mushes increases in proximity to their solidus and can operate efficiently at 0.6 < Φ < 0.93. These observations shed light on maximum estimates of the segregation of gas-rich, crystal-poor magmas (0.02 m/year at 850°C to 9 m/year at 750°C) to form felsic dykes or eruptible systems feeding volcanoes.

Keywords: melt, gas, magma, cooling, filter pressing

INTRODUCTION

Crystal fractionation does not depend solely upon gravitational separation of crystals from melt due to density differences (Wager et al., 1960; McKenzie, 1984; Spiegelman et al., 2001; Connolly et al., 2009; Schmidt et al., 2012); in crystal-rich magmas, other processes may operate. Bowen (1928), for example, avers that “the squeezing out of silicate melt residues by forces external or internal to crystal mushes may be the most important of all.” In the Earth’s shallow crust (<10 km; <2.5 kbar) where most of the erupting silicic magma reservoirs are located (Huber et al., 2019; Zhang et al., 2020), cooling magmas tend to be volatile-saturated (6–8 wt% H2O; Hui et al., 2009) and become crystal-rich (crystal fraction Φ > 0.4; Marsh, 1981; Dufek and Bachmann, 2010; Huber et al., 2010a), becoming extremely viscous over a very small temperature (T) range as they approach their solidus (Holtz and Johannes, 1994; Holtz et al., 2001; Pistone et al., 2013).
These factors impede any gravitational segregation of crystals from melt, yet there is clear field evidence for such segregation in natural rocks (Holness, 2018). Instead, these near-solidus systems are capable of releasing high-silica melt (formed during crystallization at low-pressure; Gualda and Ghiroso, 2013) through porous flow (Olsen et al., 2004) to form crystal-poor, evolved magmas of rhyolitic composition that are capable of eruption at the surface (Hildreth, 1981, 2004; Marsh, 1981; Bachmann and Bergantz, 2004; Hildreth and Wilson, 2007; Dufek and Bachmann, 2010; Waters and Lange, 2017).

Using phase equilibria constraints in $P$–$T$–$H_2O$ space, most felsic magmas have initial $H_2O$ contents ranging from 5–7 wt% at 4 kbar to 4–5 wt% at 3 kbar and 2–4 wt% at 1 kbar at $T = 750–900°C$ (Clemens and Wall, 1981; Clemens et al., 1986; Webster et al., 1987; Whitney, 1988; Johnson and Rutherford, 1989; Dall’Agno et al., 1994; Scaillet and Pichavant, 1994; Scaillet et al., 1995a,b). During magma cooling, extraction of hydrous silicic magmas is most likely at crystal fractions ranging between 0.5 and 0.7 (Dufek and Bachmann, 2010). This physical condition is then enhanced by the time spent by crystallizing magmas at conditions optimized for melt extraction: the longer magma spends at such conditions, the greater the amount of extracted residual melt (Huber et al., 2009; Dufek and Bachmann, 2010; Hartung et al., 2019). The competition between the rate of heat loss and the rate of latent heat release due to crystallization during progressive cooling and solidification of magmas exerts an important control on the extraction of interstitial melts (Huber et al., 2009; Caricchi and Blundy, 2015; Lee et al., 2015).

The process of melt transfer from large-volume melt domains to nearly solidified ones is similar to models of filter pressing (Robin, 1979; Sawyer and Robin, 1986; Brown et al., 1995). However, in a rheologically locked magma undergoing solidification, the diminished volume fraction of residual melt concentrates dissolved volatiles and favors exsolution of gaseous bubbles (Bacon, 1986; Sisson and Bacon, 1999; Masotta et al., 2010) and supercritical fluids or brines (Driesner, 2007; Driesner and Heinrich, 2007; Blundy et al., 2015; Afanasiev et al., 2018), a process known as (isobaric) second or resurgent boiling (e.g., Candela and Blevin, 1995; Candela, 1997; Su et al., 2016). Bubble growth is a function of many factors (Gonnermann and Manga, 2007, and references therein). At constant pressure, bubble growth is controlled by the balance between the rate of diffusion of $H_2O$ molecules toward the bubble-melt interface and the rate of viscous deformation of the surrounding melt (Proussevitch et al., 1993; Proussevitch and Sahagian, 1998; Liu and Zhang, 2000). Where the rate of deformation exceeds the rate of diffusion, growth is described as diffusion-limited, whereas the inverse is viscosity-limited (Navon et al., 1998; Colucci et al., 2015). Gas exsolution at low pressures ($P < 4$ kbar) causes the mush to expand, whereas its high viscosity ($10^5$ to $10^{18}$ Pa s; Sparks et al., 2019) resists inflation, along with the contribution of rigid reservoir/conduit walls that play in the background. In this scenario, gradients in $\Phi$ and vesiculation drive the melt toward regions of lower $\Phi$, $T$, and $P$ (Huber et al., 2010b; Parmigiani et al., 2016; Huber and Parmigiani, 2018). Under these conditions, felsic magmas become $H_2O$-saturated and, thus, the presence of exsolved gas within the interstitial melt is likely. Indeed, field examples of plutonic felsic rocks with vuggy porosity, such as spherical vesicles, angular dikttytaxitic voids, and miarolitic cavities, indicate the migration of intergranular melt and gas between interlocking crystals that act as a filter in solidifying mushes (Figure 1; Candela and Piccoli, 1995; Candela, 1997). Melt and supercritical fluid percolation generates out-of-equilibrium textures (e.g., Grosse et al., 2010; Vigneresse, 2015; Lissenberg and MacLeod, 2016; McCarthy and Müntener, 2016; Pistone et al., 2020). In cooling magmas emplaced in the shallow crust, melt and gas segregation can produce internal geochemical heterogeneities with macrorhythmic sequences of inter- and cross-connected melanocratic, mesocratic, and leucocratic units with vuggy porosity (Figures 1A–C), “foamy” rocks produced as a result of magma decomposition following dyking and brecciation of pluton peripheries in contact with brittle country rocks (Figures 1D,E), and coarse-grained comagmatic melt-percolated pegmatite pods (Figure 1F) produced by “swinging eutectic point” (Balashov et al., 2000), adiabatic drop in pressure (Shannon et al., 1982), and chemical undercooling (Pistone et al., 2016) occurring rapidly with no achievement of chemical and textural equilibrium (e.g., Fenn, 1977; Webber et al., 1997; Nabelek et al., 2010).

One relatively rapid melt extraction mechanism in the upper crust that has been proposed over the last decades is gas filter pressing (Anderson et al., 1984; Bacon, 1986; Sanders, 1986; Sisson and Bacon, 1999; Pistone et al., 2015), which can produce overlying pegmatites and aplites from extracted gas-rich melts (Sisson and Bacon, 1999; Hartung et al., 2017). Gas filter pressing is probably the dominant process by which highly evolved melts segregate from crystal mush to form aplitic dikes in granitic plutons and accounts for the production of voluminous, crystal-poor rhyolites (Sisson and Bacon, 1999).

Gas filter pressing operates in crystal-rich systems ($\Phi \geq 0.6$) below the maximum packing fraction of bubbles and crystals (~0.74; Pistone et al., 2015), in agreement with experimental observations of melt percolation down a pressure gradient along grain boundaries at maximum crystal volume fractions of 0.85–0.9 (Dell’Angelo and Tulis, 1988). Above the maximum packing fraction of solid minerals and expanding bubbles, the mush tends to fracture; gas escapes through open cracks (Parmigiani et al., 2014; Oppenheimer et al., 2015; Pistone et al., 2015). The residual melt remains trapped within the crystal framework and is forced to crystallize and solidify.

To date, field observations offer only the terminal snapshot of the sequence of the dynamic processes that lead to melt extraction. Conversely, the most recent models (Degruyter et al., 2019, and references therein) shed light on the change of the local pore pressure during gas exsolution in cooling magmas, without simulating the actual transport of melt and exsolved fluids during second boiling. What remains unclear is how melt and gas are extracted at the microscale during cooling of hydrous magmas. To address this, our study aims to quantify how $\Phi$ and $T$ control the mobilization of volatile-rich melts during mush solidification. We achieve this through high-temperature and -pressure experiments, observing how microstructural textures of crystal-rich magmas control melt segregation, supported by petrological modeling based on Rhyolite-MELTS (Gualda et al., 2012).
FIGURE 1 | Examples of melt and gas segregation within cooling magmas. (A–C) Outcrops of the Beaucette Marina battery of the gabbro-dioritic Bordeaux Group (North-Quercy Intrusive Complex, Armorican Massif; Caroff et al., 2011) show a cumulative macrorhythmic sequence of meladiorite and diorite rocks. The meladiorite unit consists of alternating dark and few cm-thick mesocratic layers and is veined by leucocratic diorite with vuggy porosity, forming an inter- and cross-connected network of cm-thick sheets. (D) Image of the ca. 900 m high N-W cliff of the Cerro Nido el Condor (Patagonia, Chile) showing outcrop of the frontal part of the Torres del Paine igneous complex. The “foamy” granite from Unit I (Michel et al., 2008) is accumulated at the front and is enriched in miarolitic cavities. Note ubiquitous radial diking, and brecciation at the bottom of the laccolith. (E) Close-up on one of the dikes from the bottom of the Unit I, where both sedimentary xenoliths and magma solidification act as a barrier to gas and melt movements following dike emplacement. (F) Coarse pegmatite pods with hornblende and feldspar minerals arranged in a comb layer (a variant of unidirectional solidification texture or UST) with respect to the equigranular host gabbro from the Val Fredda Complex (Blundy and Sparks, 1992) of the Tertiary Adamello Batholith (Alps, Italy).
MATERIALS AND METHODS

Material Preparation and Synthesis

The preparation of the materials followed the approach of Pistone et al. (2016). We use samples of F dacite, which is equivalent to the natural sample JM101 (dacite/tonalite from Adamello batholith, Italy; Blundy and Sparks, 1992). Samples were prepared by mixing dried, fine-grained (~1 µm) oxides and hydroxides in appropriate proportions before the addition of a known volume of quartz crystals that had been fired for 5 h in a one-atmosphere muffle furnace at 1,000°C. Quartz crystals (DORSILIT 2500: SiO$_2$ = 98.90 wt%; Al$_2$O$_3$ = 0.41 wt%; Fe$_2$O$_3$ = 0.02 wt%; TiO$_2$ = 0.04 wt%; minor trace elements; Alberto Luisoni AG, Switzerland) had aspect ratios between 1.0 and 3.5, and three-dimensional grain size distributions with a mean diameter of 68 µm and positive skewness of 0.37, checked using the Malvern Laser Diffraction Grain Sizer at the Limno-Geology Laboratory, ETH-Zurich. Quartz particles were selected to ensure no reaction with the melt in which they resided (Pistone et al., 2012). Aliquots of F dacite glass were mixed with quartz particles to create additional starting materials of different crystallinity: F50 = 0.5 crystal fraction (Φ), F60 = 0.6, F70 = 0.7, and F80 = 0.8. The crystal-glass mixtures were pressed into cylindrical stainless steel canisters (110 mm long, 35 mm inner diameter, 3 mm wall thickness) lined with a thin (25 µm) molybdenum foil to avoid chemical contamination, using a uniaxial cold press. The canisters were stored at 110°C for 24 h, then arc-welded shut. The tightness of the seal was checked by weight after immersion in a water bath under vacuum. The canisters were then hot isostatically pressed (HIP) for 24 h in a large capacity (170 mm in diameter; 500 mm high), internally heated pressure vessel (Sinter-HIP-Kompaktanlage, ABRA Fluid AG: Rock Deformation Laboratory of ETH-Zurich). The chosen P and T conditions ensured the generation of chemically homogeneous glass and complete dissolution of H2O in the melt phase (i.e., water-undersaturated melt). The maximum temperature difference across the sample was 18°C as monitored by three C-type thermocouples placed at the bottom and top and in the middle of the pressure vessel. Samples were then cooled at 60°C/min to a temperature of 100°C above the glass transition temperature (i.e., viscosity equivalent to 10$^{12}$ Pa s) to prevent crystallization; followed by slower cooling (0.6°C/min) to room temperature to obtain thermally relaxed glasses. The confining pressure was decreased by 37.6 bar/min during rapid cooling, and by 1.3 bar/min during slow cooling. Comparison of canister weights showed less than 0.5% change in mass, suggesting H$_2$O loss during synthesis is negligible. Cores of 2-mm diameter and 2- to 4-mm length were then drilled from each synthesized sample, and the top and bottom faces were ground flat and parallel. The compositions of the starting materials are reported in Pistone et al. (2016) and are presented in the Supplementary Material.

High-Pressure and High-Temperature Experiments

To simulate melt and gas mobilization upon magma cooling, three high-temperature (T = 750, 800, and 850°C), 1.1-kbar pressure experiments were conducted on a suite of pre-synthesized, H$_2$O- and crystal-rich glasses (dacite with 4.2 wt% H$_2$O in the glass, and Φ = 0.6, 0.7, 0.8). The experimental pressure was chosen within the range of emplacement depth of the plutonic systems reported in Figure 1 (0.75 ± 0.25 kbar for the laccolith of Torres del Paine; Putlitz et al., 2001; Baumgartner et al., 2007, 2014; Leuthold et al., 2014; 3–4 kbar for the Plutonic Complex of Guernsey; D’Lemos, 1987; de Bremond, d’Ars et al., 1992) and consistent with some natural cases of erupting systems with very shallow pre-eruptive residence (~1 kbar in Taupo Volcanic Zone; Bégué et al., 2014; Gualda et al., 2018). Also, to generate the initial gas phase within the cooling system, the chosen pressure (0.7 kbar lower than the pressure for material synthesis) allowed the achievement of water-saturation conditions during experiments (0.1–0.2 wt% H$_2$O in excess in the starting melt composition prior to crystallization, calculated using the model of Papale et al., 2006). The different Φ values bracketed the initial crystallinities of natural plutonic gas-saturated mushes; each experiment at specific T represents a snapshot of the state of the mush along its cooling path. Three sandwiched samples were inserted into 4-mm diameter Au capsule, crimped and welded before experiments. Using vertical, hydrothermal cold-seal pressure vessels (University of Lausanne), samples were first pressurized to 1.1 kbar at room temperature, with pressure monitored using both an IMS digital pressure sensor and a Heise 3-kbar bourdon-tube gauge (±5 bar). Then, T was increased at a rate of 30°C/min up to the target temperature. Experiments were run for 8 days at constant P–T and H$_2$O-saturated conditions (~4.1 wt% H$_2$O in the starting melt prior to crystallization at all studied temperatures; Papale et al., 2006) above the wet granite solidus (~710°C at 1.1 kbar; Tuttle and Bowen, 1958; Holtz et al., 2001). Experiments run between 750 and 850°C at 1.1 kbar allowed equivalent conditions of initial melt viscosity (Δlog$_{10}$melt < 0.5 and, thus, allowed comparison of processes simulated in the experimental runs at different T (Holtz et al., 2001; see also Supplementary Material). We did not simulate magma cooling by changing T with a specific cooling rate; rather the different temperatures used span those occurring in natural gas-saturated mushes approaching their solidus. Temperature was monitored with an N-type thermocouple (±2°C) connected to a Eurotherm 815/831 controller. Based on temperature calibration at room pressure prior to the high-pressure and high-temperature experiments, no T gradients were found in the sample placed in the center of an isothermal 30-mm hot zone. Experiments were terminated by turning off the furnace power and using conventional compressed air quenches (quench rate of ~4°C/s). Quenching to the solidus (at which the sample microstructure freezes) was thus completed in 15, 30, and 50 s from 750, 800, and 850°C, respectively. Cooling to room T was then followed by slow depressurisation and sample recovery.

Microstructural Analysis

Both starting materials and recovered samples were explored using X-ray computed tomography (CT) using a spatial resolution of 6 µm/pixel at 40 kV, 200 A, and 800 ms exposure time with a SkyScan 1173 (University of Lausanne). Under these acquisition conditions, quartz and glass were easily resolvable.
phases. The higher X-ray attenuation of the glass was largely due to its Fe content. Tomographic 3D renderings were inspected using the IDL-based Blob3D software (Ketcham, 2005a,b) to quantify the spatial distribution and first-order volume fraction of each phase, with an uncertainty of 0.005 in volume fraction. Three-dimensional tomographic analysis is used to capture the bulk microstructural features of the samples, including glass, pre-existing quartz, and vesicle spatial distribution.

In order to image microstructural details at higher spatial resolution (<6 µm/pixel), samples were also inspected using a FEG-EPMA. Samples were cut in half to form a peripheral and a central portion (Figures 2, 3); both portions were impregnated in epoxy, polished, and carbon-coated for analytical inspection. Backscatter electron (BSE) images were acquired at 15-kV accelerating voltage and 1.5-nA beam current. BSE images with a spatial resolution of 1-µm were used to estimate the volume fractions of glass, vesicles, and minerals in the different portions of run products using the image processing software JMicroVision® v1.2.7 (Roduit, 2011), with an uncertainty of 0.002 in volume fraction based on inspections of three representative 200 × 200 µm areas for each sample. The vesicle size distribution (VSD), based on Feret’s radius, was carried out using the Java 8 Fiji software (Schindelin et al., 2012). The Feret’s radius is the half of the maximum distance or caliper between two parallel lines tangential to the particle outline and is used to particles of low aspect ratio (<3) and as long as particle morphology is not largely irregular (Liu et al., 2015). Study of the VSD allowed exploration of the different processes that lead to bubble growth: Ostwald ripening (Stoffregen, 1996; Lautze et al., 2011), H2O diffusion (Zhang et al., 2000) or physical gas coalescence (Castro et al., 2012). The BSE analysis was used to capture the differences in glass volume fraction between peripheral and central sections of the run products and, thus, to evaluate the movement of residual melt in the synthetic crystal mush at high P–T conditions.

Chemical Analysis of Glass

A major challenge with glass analyses is alkali loss (Morgan and London, 1996), a very common problem that can be minimized by using a large beam diameter and low beam current on the electron probe micro-analyser (EPMA). However, the available surface of inclusion- and bubble-free glass precluded the use of a defocused beam almost everywhere throughout the experimental run products. Hence, we had to adjust our analytical conditions using a low accelerating voltage and 3–5-µm beam size for the chemical analyses of the experimental glasses. We used two EPMA instruments: a JEOL JXA-8350F HyperProbe (FEG-EPMA; University of Lausanne) and a CAMECA SX Five EPMA (University of Massachusetts in Amherst, MA) at 15 kV, 2 nA, and ∼5- to 10-µm beam diameter. The counting time on the peak and background positions was 30 and 15 s for Al, Ti, Ca, Mg, and Mn and 10 and 5 s for Si, Fe, Na, K, and P, respectively. Natural and synthetic standards were used and data were ZAF-corrected (Armstrong, 1988). The FEG-EPMA standardization using a small beam diameter (<5-µm) at 7 kV included the analysis of hydrous glasses with known water content (H2O = 5.7 wt%; Müntener et al., 2001) to monitor any alkali loss. Using these secondary standard glasses let us assume that no alkali loss occurred during FEG-EPMA analysis with a 5-µm beam. At 7 kV, neither Mn or P were analyzed (<0.05 wt% total). Analyses at 15 kV targeted only large glass pools located in the vesicle- and glass-rich regions away from the solidification fronts. The analyses in the run product at 750°C were almost exclusively located in the “glass coronae” around the vesicles and using 10-µm beam size did not yield reliable data, which were discarded (Supplementary Material). The analyses at 7 kV targeted the “glass channels” where melt percolation occurred within the areas affected by crystallization fronts during experiments. The two sets of analyses display a major difference in alkali content (especially Na), which is high in the glass-rich areas devoid of plagioclase and remote (>10-µm) from vesicles (Supplementary Material). Glass major element compositions and related uncertainties are in the Supplementary Material. Newly formed minerals (quartz, plagioclase, hornblende, and oxide) were qualitatively analyzed using an SDD EDS detector (spectral resolution of 129 eV on Mn) at 15 kV, 1.5 nA, and ∼1-µm beam diameter.

Further glass analyses in representative “glass channels” in samples with Φ = 0.6 and Φ = 0.7 at 750°C were carried out using the CAMECA SX Five EPMA (University of Massachusetts in Amherst, MA) at 15 kV, 2 nA, and ∼5- to 10-µm beam diameter. The counting time on the peak and background positions was 30 and 15 s for Al, Ti, Ca, Mg, and Mn, and 10 and 5 s for Si, Fe, Na, K, and P, respectively. We used two glass standards of rhyolitic (USNM 72854 VG-568 from the Smithsonian Institution), haplogranitic (HGGB-3 glasses from Pistone et al., 2012), and dacitic composition (F0 from Pistone et al., 2016) and data were ZAF-corrected. This EPMA analysis helped to show that the FEG-EPMA analysis of the experimental glasses in F60, F70, and F80 at 7 kV were affected by alkali migration due to electron charge with an overall alkali loss of up to 50%. This alkali loss at low voltage and reduced beam size modified the number of counts for Si during analysis.

Estimation of H2O Content and Calculation of Melt Viscosity

H2O content in the glass phase was estimated using the volatile-by-difference method (Devine et al., 1995) for both the 7- and 15-kV sets of analyses, with limited uncertainty (<1 wt%) for H2O contents between 4.5 and 7.2 wt%. This method was applied by calibrating the analytical reproducibility at the chosen FEG-EPMA (7 and 15 kV, respectively) and EPMA analytical conditions (15 kV) with hydrous felsic glasses of similar H2O content (samples B659 and B736 from Müntener et al., 2001; F0 from Pistone et al., 2017; samples HGGB-3...
FIGURE 2 | FEG-probe-based BSE images of peripheral sections of all crystal-bearing samples from experiments at: (A) 750, (B) 800, and (C) 850°C. Acronyms are: Qz₁ = pre-existing quartz (dark gray); Ves = vesicle (black objects, sometimes filled with whitish material that produces electron charging); Glass = residual glass (least dark gray phase) that can be populated with microlites.

from Pistone et al., 2012). Most of the results agree with H₂O-saturation concentrations calculated using Papale et al. (2006) for the run pressure and temperatures. However, apparent H₂O contents appreciably exceeding H₂O-saturation values were determined for most analyses of 750°C glasses, even for analyses at 15 kV (summation deficits to as great as 9.7 wt%). These high apparent concentrations were determined in 10-µm thick glass halos around vesicles and in widespread glass domains populated by potential cavitation nuclei or “nanobubbles”; the latter were not observable at the analytical spatial resolution of the BSE images, but inferred from the “foggy” aspect of the residual glass using optical microscopy. Glass adjacent to bubbles can hydrate during quench-cooling to room temperature, and the electron beam’s X-ray excitation volume may intersect nanobubbles in “foggy” glass domains. Analyses near bubble walls and in “foggy” glass domains of 750°C runs were therefore excluded. Hydration during quenching, alkali-loss during analysis, and overlap with sub-surface bubbles cannot be ruled out entirely for the accepted analyses of glasses synthesized at 750°C, so the values calculated for their viscosities and saturation overpressures are minima and maxima, respectively.

Melt viscosities at P–T conditions were estimated using the model of Hui et al. (2009) with log₁₀(η_melt) variations of maximum 0.2 with H₂O contents ranging between 4.5 and 7.2 wt%, as estimated with the volatile-by-difference method applied to 15-kV data (marked in yellow in the analytical data summary table of the Supplementary Material), in line with calculations from solubility modeling (Papale et al., 2006).

Rhyolite-MELTS Crystallization Simulations

Crystallization paths of hydrous dacite from 850 to 750°C at 1.1 kbar were simulated using Rhyolite-MELTS (Gualda et al., 2012) to reconstruct the sequence of crystallization of the major phases (hornblende, plagioclase, and quartz) and volume percentage of excess H₂O-rich bubbles at the different temperatures in the experimental samples. Rhyolite-MELTS software currently lacks
a thermodynamic model for simulating amphibole crystallization in silicate melts, and phases such as pyroxenes are calculated. However, this did not affect the purpose of our simulations here, as melt compositions calculated by Rhyolite-MELTS were not directly considered. Calculations of melt crystallization were run using starting compositions in a near-fractional crystallization style; i.e., the chemical composition of the starting material for the simulation is the measured composition of the glass from the previous crystallization stage at the specified temperature. Starting conditions and results of each simulation are reported in the Supplementary Material.

**H$_2$O Diffusion and Bubble Growth**

The characteristic timescales for diffusion ($\tau_d$) and viscous deformation or relaxation ($\tau_v$) are:

$$\tau_d = \frac{r^2}{D_{H_2O}}$$

(1)

$$\tau_v = \frac{\eta_{melt}}{\Delta P_g}$$

(2)

where $r$ is the bubble radius, $D_{H_2O}$ the H$_2$O diffusion coefficient or diffusivity (Zhang et al., 2000), $\eta_{melt}$ the melt viscosity, and $\Delta P_g$ the gas overpressure. $D_{H_2O}$ in rhyolitic melts with H$_2$O content <7.7 wt% in the $T$ range 403–1629$^\circ$C and at $P < 19$ kbar is estimated using the model of Ni and Zhang (2008):

$$\ln \left( \frac{D_{H_2O}}{X} \right) = 13.47 - 49.996X + 7.0827\sqrt{X} + 1.8875P - 9532.3 - 91933 + 13403\sqrt{X} + 3625.6P$$

(3)

where $X$ is the mole fraction of H$_2$O on a single oxygen basis, $T$ temperature in K, and $P$ pressure in GPa. $\Delta P_g$ is estimated using the equation of state for H$_2$O reported in Pitzer and Sterner (1994) in the JavaScript-based Fugacity Calculator: https://www.esci.umn.edu/people/researchers/withe012/fugacity.htm.
To determine whether diffusion- or viscosity-limited, the non-dimensional Péclet number ($Pe$), combining Eqs 1 and 2, must be determined (Navon and Lyakhovsky, 1998; Navon et al., 1998):

$$Pe = \frac{\chi_d}{\tau_v} = \frac{\Delta P r^2}{\eta_melt D_{H_2O}}$$

In our experiments gas exsolution occurs under conditions of $Pe < 1$, therefore the rate of expansion is unimportant relative to that of diffusive mass transfer. The $H_2O$ diffusion length scale ($l$) from melt to gas bubble was determined using $l = \sqrt{T \times D_{H_2O}}$, with $t$ as the experiment timescale.

**RESULTS**

Collectively, the experiments display a sequential record of microstructural changes toward the solidus in a $T$-$\Phi$ space (Figures 2, 3). Visual inspection of the central sections of the samples shows that, as also observed in nature (Figure 1), none of the samples achieved textural equilibrium due to rapid crystallization and vesiculation (Anderson et al., 1984; Sanders, 1986; Sisson and Bacon, 1999). Degassing occurred in all experiments with many vesicles found trapped between mineral clusters and in direct contact with surfaces of newly formed minerals. Using three-dimensional image analysis and two-dimensional analysis of vesicle modal abundance, we observe how the distribution of both glass (including hornblende and oxides, which are not resolvable at the spatial resolution of the microtomography) and vesicles changes as the experimental system approaches the solidus (Figure 4).

Here we report the observations of the different phase (glass, minerals, and vesicles) proportions and changes as a function of initial crystallinity, sample portion (central versus peripheral section), and temperature (Figure 4). The vesicle volume fraction in sample F80 slightly increases from central to peripheral sections in the $T$ range from 800 and 750°C. At 850°C the vesicle content from central to peripheral sections of the cylindrical materials is more variable, with F70 displaying the largest increase in vesicle abundance in the peripheral section (Figures 4B,C) where large (up to 500-$\mu$m wide) tubular and connected vesicles are found (Figure 2C). In both central and peripheral sections of the cylindrical materials we observe the vesicle content decrease from 850 to 800°C and then its increase from 800 to 750°C, except for the case of sample F70 whose vesicle content decreases at 850°C (Figures 4B,C). This non-linear change of the vesicle content is a function of both SiO$_2$ increase in the residual melt during moderate crystallization from 850 to 800°C and during extensive crystallization from 800 to 750°C, and non-equilibrium processes, which include relatively rapid crystallization at the chosen temperatures and melt and vesicle movement during the experiments. At constant $P$, both SiO$_2$ increase in melt composition and $T$ decrease concomitantly increase $H_2O$ solubility in the residual melt due to retrograde or inverse volatile solubility with lowering $T$ (Fogel and Rutherford, 1990; Paullat et al., 1992; Blank et al., 1993; Lowenstern, 2001).

At 850 and 800°C, the vesicle content tends to remain unchanged in both central and peripheral sections of each sample, whereas, at 750°C, vesicle abundance increases by up to 50% at largest crystallinity ($\Phi = 0.8$; Figures 4B,C). Initial crystallinity increases from 2 to 20% in run products at $T = 850°C$, whereas it increases by 45% in F60, by 11% in F70, and by 5% in F80 at 750°C, based on the comparison between the 3D renderings of the run products at 850 and 750°C, respectively (Figure 4A). At 800°C the initial crystallinity does not vary (<2%) by either partial mineral resorption as observed at 850°C or the formation of new minerals as observed at 750°C (Figure 4). The total glass abundance decreases by 10% (in F70 and F80) to 70% (in F60) from 850 to 750°C (Figure 4A). Glass modal proportion increases from 20% (in F60 and F70) to 50% (in F80) from central and peripheral sections of samples at 750°C (Figures 4B,C). Limited glass volume fraction increase is observed between central and peripheral section of samples at 850°C (<5%) or no increase at all at 800°C (Figures 4B,C). Analysis of the VSD shows a narrow distribution of Feret’s radius centered around 5- to 10-$\mu$m (Supplementary Material) unlike that expected for Ostwald ripening, which would exhibit a smaller, broader peak (Stoffregen, 1996).

Detailed microstructural inspection of representative sample regions at microscopic scale reveals key features at specific $T$ and initial $\Phi$ conditions (Figures 5–7). Fractures seen in all samples are produced at the end of the experiment during either sample cooling, recovery, or preparation prior to analysis.

At 850°C (Figure 5), vesicles are sub-rounded and surrounded by newly-formed quartz grains (Qz$_2$) that are 20-$\mu$m across and roundish and, thus, devoid of asperities such as those of the pre-existing quartz (70-$\mu$m across, Qz$_1$). Quartz of round shape is typically formed in H$_2$O-saturated felsic melts (Cabane et al., 2001). Hornblende needles (10-$\mu$m long) and tabular plagioclase (40-$\mu$m long) are primarily found in vesicle-free regions. Glass-rich regions are free of vesicles and newly-formed minerals (Figure 5). Melt viscosity is homogeneously distributed in the sample, with $log_{n_melt} = 3.5 \pm 0.2$ (Figure 5).

At 800°C (Figure 6), new quartz (Qz$_2$), plagioclase, and microscopic oxides form on pre-existing quartz (Qz$_1$). The presence of oxides “coating” quartz and plagioclase favors round vesicles (10-$\mu$m diameter) to adhere to the mineral surface, in agreement with previous experimental observations (Hurwitz and Navon, 1994; Laporte, 1994; Navon and Lyakhovsky, 1998; Navon et al., 1998). Glass is populated by oxides (1-$\mu$m) and hornblende needles that, at $\Phi = 0.6$, form an alignment texture within glass regions of 10- to 30-$\mu$m size (Figure 6A). At $\Phi = 0.8$, plagioclase is abundant (Figure 6C). Both vesicles and glass are found within the glass-rich portions of the starting materials, which were sealed by the progression of crystallization and gas exsolution (Figure 6C). Glass could not be analyzed due to a lack of crystal-free spots; thus, no $log_{n_melt}$ estimations are presented.

At 750°C (Figure 7), new quartz (Qz$_2$), plagioclase, oxide, and hornblende adhering to pre-existing quartz (Qz$_1$) are widespread. Quartz and plagioclase are arranged in unidirectional solidification textures (USTs; Shannon et al., 1982) that also entrap 3-$\mu$m diameter vesicles (Figure 7C). In places, these textures end with new 50-$\mu$m quartz grown from the USTs (Figure 7B). The glass region is constricted by large vesicles (20 $\mu$m) and hornblende needles, aligned
FIGURE 4 | Total volume fractions of phases: (A) in each tomographic 3D rendering, (B) in each BSE central section, and (C) in each BSE peripheral section of the experimental specimens. Volume fraction uncertainties (2σ) are 0.005, 0.002, and 0.002, respectively. Horizontal pink bars indicate initial level of crystallinity in the run products analyzed using X-ray tomographic microscopy.
FIGURE 5 | FEG-probe-based BSE images of selected portions within the central polished sections of: (A) F60, (B) F70, and (C) F80 at 850°C. Error bars corresponding to $2\sigma$ are smaller than symbols. Acronyms are for: Qz$_1$ = pre-existing quartz (dark gray); Plg = plagioclase (light gray); Hbl = hornblende (white elongated phase); Ves = vesicle (black objects, sometimes filled with whitish material that produces electron charging); Glass = residual glass (least dark gray phase). Sketches of cylindrical samples (light gray) show the location of the sample portion (black rectangles) inspected in the sample central section (orange plane).
perpendicularly to the USTs (Figure 7B). Pre-existing quartz is surrounded by a micrographic texture of ∼1-µm oxides and hornblende (Figures 7B,C). Log 10[melt] decreases by ∼0.5 from crystal-rich and bubble-poor regions to low crystallinity bubble-rich regions within the same sample, especially in specimen F60 (Figure 7A). Moreover, samples with high crystallinity (F70 and F80) display calculated log 10[melt] of 3.5–4.0, which is 0.5 orders of magnitude (Figure 7) lower than the value obtained for the low crystallinity samples (F60).

At each T, bubble growth was driven by H2O diffusion, with an initially high D H2O (4·10−11 m2/s at 4.2 wt% H2O) that increased with increasing H2O in the residual melt (10−10 m2/s at 5–7 wt% H2O) during crystallization. In agreement with previous studies (Lyakhovsky et al., 1996), high D H2O leads to large H2O diffusion distances (∼8 mm > sample length and diameter) in the range of T and run duration of our experiments. There is no evidence of physical interconnectivity of vesicles (Figures 5–7), which suggests that bubbles formed by H2O diffusion only. Locally, H2O in the glass exceeds H2O-saturation conditions (>5–7 wt% H2O at experimental P-T; Papale et al., 2006; Supplementary Material), either in the 10-µm thick glass halos around vesicles due to H2O resorption from bubbles during cooling (McIntosh et al., 2014) or in glass portions suggested to be populated by “nanobubbles” (Davydov et al., 2005). The volume fraction of new minerals increases with decreasing initial ø at all T. This is due to the fact that samples with low crystallinity (F60) display larger amount of residual melt that degas and crystallize with respect to the high-crystallinity systems (F70 and F80).

Rhyolite-MELTS calculations predict that the starting melt would crystallize plagioclase at 850°C, followed by quartz at 800°C (Figure 8A). Solidification fronts from 850 to 750°C would thus be driven by: (i) initial homogenous nucleation of hornblende, plagioclase, and limited quartz, (ii) heterogeneous growth of hornblende, quartz, and plagioclase upon pre-existing quartz, and (iii) increase of vesicle content (Figure 8A). These calculations provide minimum mineral fractions in the absence of amphibole, which increases H2O in the melt and, thus, leads to less extensive crystallization, as observed in our experiments. Overall, the volume fraction of vesicles tends to decrease from the sample center toward its periphery at highest crystallinity (Figures 4B,C).

**DISCUSSION**

The microstructures produced experimentally are the consequence of undercooled partial crystallization of magma that lead to enhanced differentiation of the interstitial melt (from dacite to rhyolite) and disequilibrium rock textures. Both microstructural and chemical analyses demonstrate that in situ melt differentiation is driven by vapor-saturated magma crystallization and physical migration of the residual melt, as a result of pressure gradient caused by gas exsolution.

ø and T govern melt and gas expulsion from the magma during solidification. The reduction of pore space by “crystal zipping” (Figures 7B,C), driven by the advancement of solidification fronts (Marsh, 1995), forces both melt and bubbles to migrate toward the crystal-poor regions and to be segregated toward the sample periphery (Figure 8B). We quantify the outward movement of melt by measuring the final volume fraction of glass/melt in both the central and peripheral sections of the sample, Xf(center) and Xf(peripheral), respectively, where the
FIGURE 7 | FEG-probe-based BSE images of selected portions within the central polished sections of: (A) F60, (B) F70, and (C) F80 at 750°C. Error bars corresponding to 2σ are smaller than symbols. See acronyms from Figures 5, 6, plus USTs = unidirectional solidification textures, consisting of plagioclase and quartz (Qz₂) aligned perpendicular to the surface of the pre-existing quartz minerals (Qz₁). Sketches of cylindrical samples (light gray) show the location of the sample portion (black rectangles) inspected in the sample central section (orange plane).
peripheral section is located at 1 mm radial distance. We then define the quantity $X_{ex}$ to be

$$X_{ex} = X_f(\text{peripheral}) - X_f(\text{central})$$

and note that the larger the value of $X_{ex}$, the greater the volume of outward melt transport. We observe that $X_{ex} \leq 0.03 \pm 0.01$ at 850 and 800°C, and $X_{ex} \leq 0.10 \pm 0.01$ at 750°C. At 750°C, crystallization produces a continuous network of minerals upon the pre-existing quartz grains and enhances the $P$ increase during gas expansion in the constricted channels (Figure 7B). Concurrent pore restriction by crystallization and volume expansion upon $H_2O$ exsolution by diffusion into bubbles leads to highly efficient extraction of large melt fraction by gas filter pressing. Conversely, in the runs at 800 and 850°C, appreciable melt volume extraction occurs at $\Phi \geq 0.8$ due to: gas expansion in absence of melt pore restriction caused by crystallization at 850°C (Figure 5C), and limited crystallization without extensive gas exsolution at 800°C (Figure 6C). Both cases make evident the limited effectiveness of melt extraction driven by exsolved gas (Figure 8B).

The volumetric estimates of extracted melt in our experiments are in line with similar estimates determined using mass balance calculations and textural observations of rinds and cores of glass-bearing andesitic enclaves in volcanic rocks (Bacon, 1986). Specifically, Bacon (1986) showed that the extraction of 0.02–0.04 volume fraction of rhyolitic melt is required to produce the chemical discrepancies between cores and rinds of andesitic inclusions. A few millimeters thick glass linings in vesicles and the rhyolitic glass-filled vesicles found in rinds testify to the migration of interstitial melt from core to margin of the enclaves by gas filter pressing (Anderson et al., 1984), similar to what observed in this study (Figure 8B).

In previous experiments simulating gas filter pressing in the absence of concurrent crystallization (Pistone et al., 2015), melt extraction was efficient for $0.6 \leq \Phi \leq 0.74$. Our new study shows how melt and gas extraction proceeds in more complex systems undergoing crystallization. Very evolved, $H_2O$-rich (5–7 wt% $H_2O$) melts with $\eta_{melt} \leq 10^4$ Pa s (Figures 5, 7) are highly mobile and their migration is efficient even at $\Phi > 0.8$, below the maximum crystallinity preserving melt connectivity in volatile-rich systems ($\Phi = 0.89–0.93$; Faul, 2001; Cheadle et al., 2004; Rosenberg and Handy, 2005). The presence of continuous chains of minerals formed at the solidification fronts (Philpotts et al., 1998, 1999; Philpotts and Dickson, 2000) builds a strong network that is key to promoting melt extraction by gas filter pressing.

**IMPLICATIONS**

Microstructural and chemical results can be extrapolated to melt and gas movements during magma cooling, for example of a granitic pluton in the upper crust. We can constrain the operating timescale for gas and melt extraction prior to complete
solidification of the host mush. Below the melt connectivity transition (Rosenberg and Handy, 2005), we first estimate melt permeability in our quartz-rich magmas ($\kappa$) with melt fraction ($X_f$; Figure 8) and average mineral size ($R$) at the specific $T$ (Richter and McKenzie, 1984):

$$\kappa = \frac{X_f^2 R^2}{A(1 - X_f)} \quad (6)$$

where the permeability coefficient $A = 50$ (McKenzie, 1984; Jackson et al., 2003; Bachmann and Bergantz, 2004; Pistone et al., 2015) for $0.07 < X_f < 0.1$ (in the presence of gas bubbles) and $R$ ranging from 70 $\mu$m (initial quartz size) at 850$^\circ$C to 150 $\mu$m (due to overgrowth of new quartz and plagioclase) at 750$^\circ$C. $\kappa$ ranges from $3 \times 10^{-13}$ m$^2$ at 850$^\circ$C to $8 \times 10^{-12}$ m$^2$ at 750$^\circ$C. Here we assume magmatic suspensions containing a monodisperse distribution of crystal size. As magmas contain crystals of different size and shape (i.e., polydisperse suspensions), which both tend to reduce the permeability with respect to monodisperse systems (Bachmann and Bergantz, 2004; Rust and Cashman, 2011), the volumetric rates of melt extraction calculated here for each mineral size (70–150 $\mu$m) are maxima.

The velocity ($v$) of melt and gas migration out of the crystal network can be estimated using Darcy’s law:

$$v = \frac{\kappa}{\eta_{\text{melt}}} \times \frac{1}{X_f} \frac{\Delta P_g}{x} \quad (7)$$

$\Delta P_g/x$ is the gas pressure gradient to move melt from the sample center to its periphery ($x = 1$-mm radius distance) and is calculated based on the difference between experimental $P$ and gas saturation $P$. From 850 to 750$^\circ$C, $\Delta P_g/x$ changes from 0.001 to 0.12 kbar/m. Hence, at 1.1 kbar, the extraction of the same amount of H$_2$O-rich (4–7 wt%) melt ($X_f = 0.1$) occurs at $\sim 1$ to $\sim 3$ $\mu$m/day at 850$^\circ$C (with $\eta_{\text{melt}} = 10^4$ to $10^5$ Pa-s) and $\sim 80$ to $\sim 800$ $\mu$m/day at 750$^\circ$C (with $\eta_{\text{melt}} = 10^5$ to $10^6$ Pa-s), respectively.

Assuming natural magmas have a phenocryst size of $\sim 3$ to 10 mm (Figure 1), Eqs 6 and 7 predict expulsion velocities of melt and gas to increase from $\sim 1$ to $\sim 8$ m/year. These are, however, likely to be maximum estimates, as real magmas could attain, for equivalent $\Phi$ timescales that are several orders of magnitude slower at slower cooling rates, and bulk expansion of melt and crystals could minimize melt migration (Sisson and Bacon, 1999). The timescales of extraction of viscous felsic melt from highly crystallized magmas are broadly constrained from months to thousands of years (Huber et al., 2012; Wilson and Charlier, 2016; Gualda et al., 2018). Viable processes that can speed up melt extraction are: (i) volume contraction of magma of up to 20% upon cooling and solidification, which leads to constrictional volume that drives expulsion of residual melts (John and Blundy, 1993; John and Stüinütz, 1997), (ii) dike into brittle country rocks or pre-existing magmas (Mahood and Cornejo, 1992) that triggers pluton decompression at its margins leading to magma brecciation and removal or “unzipping” of the melt (Figures 1D,E), and (iii) eruptions that rapidly evacuate magma reservoirs, similar to that observed during drilling into magma, where the melt (from a high-$P$ region) invades the drill hole (low-$P$) within few minutes (Hardee et al., 1981; Helz, 1984). The cooling of gas-saturated magmas in proximity to their solidus has the potential to account for the extraction of interstitial melts by gas filter pressing that form pegmatitic caps or become eruptible magmas feeding volcanoes.

CONCLUSION

We present the first experimental study to show melt segregation driven by gas filter pressing from crystal mushes at conditions relevant to the Earth’s shallow crust. Through the use of conventional high-temperature and high-pressure experiments combined with high-spatial resolution techniques and Rhyolite-MELTS simulations, we have illustrated the physical conditions that allow gas-driven filter pressing to be an efficient process for melt extraction from near-solidus systems. Specifically, we have determined the effectiveness of gas-driven filter pressing to expel the silicic melt out of the crystal framework as a function of crystallinity and temperature conditions that bracket solidification of natural mushes. The results from our study are of prime importance for understanding the modality of the generation and extraction of volatile-rich crystal-poor magmas. At a more fundamental level, we document and explain how second boiling at the microscopic level affects the macroscopic dynamics of melt and gas mobilization. The cooling of volatile-saturated magmas in proximity to their solidus could represent a critical stage for silica-rich interstitial melts driven by gas filter pressing to be extracted and form pegmatitic caps or become eruptible magmas feeding volcanoes.

Large-scale magmatic reservoirs should be characterised by slow kinetics of crystallization and rate of gas exsolution (Sisson and Bacon, 1999), which were not simulated in our experiments. There is still need to investigate which short timescale process (years to decades) would speed up extraction and accumulation of large melt lenses (10–1000 km$^3$; Bachmann and Bergantz, 2004; Bachmann and Huber, 2019) of the size required for large volcanic eruptions (Volcanic Explosivity Index > 4; Newhall and Self, 1982). Using the present-day high-resolution local seismic tomography (cell volume of 125 km$^3$; e.g., Miller and Smith, 1999), there is little geophysical evidence for the presence of crystal-poor bodies of large volume. Their “absence” could be related to long-term storage of melt-rich lenses within large long-lived mush zones with massive thermal inertia that favor slow cooling (Huber et al., 2009; Morse, 2011), as well as to sufficiently fast extraction and eruption of these melt lenses before complete solidification. Further effort is therefore required for scaling up the experimental results to magmatic reservoirs in the Earth’s crust.

DATA AVAILABILITY STATEMENT

All datasets presented in this study are included in the article/Supplementary Material.
AUTHOR CONTRIBUTIONS

MP and EB conceived and carried out the high-temperature and high-pressure experiments (with technical guidance and assistance from LB and TS). MP performed all the chemical and microstructural analyses (with technical guidance and assistance from MR) and computational calculations and prepared the manuscript. The quantitative information was finalized by MP and PJ. MP and FB explored the link between experiments and natural scenarios. LB, OM, TS, and JB provided critical guidance during the preparation of the manuscript, and all authors were instrumental in the resulting manuscript.

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SUPPLEMENTARY MATERIAL

The Supplementary Material for this article can be found online at: https://www.frontiersin.org/articles/10.3389/feart.2020.00175/full#supplementary-material

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Conflict of Interest: The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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