Gas mobility in rheologically-layered volcanic conduits:

the role of decompression rate and crystal content

on the ascent dynamics of magmas

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Abstract:

Unravelling the rheological behaviour of magmas is fundamental for hazard assessment. At shallow depth the combined effects of degassing, vesiculation and crystallization are likely to produce dramatic changes in the rheology, hence modulating flow dynamics and eruptive style. The rheological evolution from a low viscosity crystal-poor, bubble-free, water-rich melt to a highly viscous crystal-rich, vesicular magma containing a water-poor melt often occurs in the conduit. To clarify the viscous flow dynamics of rheologically-layered volcanic conduits, we performed decompression experiments using a magma analogue system characterized by a low-viscous Layer L (10 Pa s) at the bottom and a high-viscous particle-bearing Layer H (≥1000 Pas) at the top. Silicone oils and spherical glass beads are employed as magma and crystal analogues, respectively. Three sets of experiments address the effects of: 1) decompression rate (ca. 10⁻² and 10⁴ MPa/s); 2) crystal content in the high viscosity magma (0, 10, 30 and 70 vol.%); and 3) volume ratio of the two rheological layers (0.6 or 0.3). Our results indicate that decompression rate exerts the most dramatic role, yielding changes in time-scale of outgassing up to two orders of magnitude, and affecting the style of decompression response (permeable outgassing or fragmentation). The solid fraction 1) strongly modulates gas mobility, 2) influences the pervasiveness of fragmentation and 3) affects the extent of mingling in the experimental conduit. These results demonstrate that the properties of a shallow, partially-crystallized portion of the magmatic column and its response to varying ascent rate are primary controls on eruptive style.

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Highlights

- 1. First decompression experiments on a rheologically-layered analogue system
- 2. Decompression history is the most relevant parameter affecting gas mobility
- 39 3. At low decompression rate, solid loading affects permeability and mingling efficiency
- 40 4. At high decompression rate, solid fraction influences the spatial pervasiveness of fragmentation
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- **Keywords:** Decompression experiments, degassing, fragmentation, analogue magma, viscosity
- 43 contrast, mingling

1. Introduction

The rheological properties of magmas during their ascent within a feeding system are a crucial determinant of the state and distribution of magmatic fluids. Thus understanding how rheology affects magma flow dynamics and eruptive behaviour is of primary importance. Due to the sensitivity of rheology to crystallinity, liquid composition and vesicularity, rheological contrasts at shallow depth within the feeding systems of volcanoes are likely to be ubiquitous. As magma ascends to the surface, several processes occur which are capable of causing dramatic changes in the physical properties, and in particular in the rheological behaviour and permeability of the magma. Shallow exsolution of H₂O by magma degassing and resultant magma crystallization are amongst the most common processes inducing rheological stiffening at the top of the magmatic column, thus generating a rheological stratification [e.g. Cashman et al., 2004]. If the gradient of viscosity along the magmatic column is large, the rheological transition can be parameterised as a two-layer flow consisting of a more evolved viscous magma (likely crystal-rich) in the upper region and a less viscous crystal-poor fluid in the lower part [e.g. Barmin et al., 2002].

The resultant heterogeneity in the physical properties of magma within a volcanic conduit has been well-documented in the literature [e.g. Polacci et al., 2009]. Such rheological contrasts are, in turn, anticipated to modulate the style of eruptive activity and hence the range and nature of the volcanic hazards resulting [e.g. Lautze and Houghton, 2005; 2006; Del Bello et al., 2015]. Lava dome eruptions in particular, represent a good example of the control of rheological stratification of the magmatic column in the volcanic conduit on eruptive behaviour, dominated by crystallization kinetics [Melnik and Sparks, 1999; 2005] or thermal effects [Costa and Macedonio, 2002]. Such processes can operate on different time scales [Melnik et al., 2008 and references therein]. Extrusion of lava from active domes is commonly characterized by pulsations of the discharge rate, with large periodic fluctuations often corresponding to cycles observed in ground deformation and/or seismic data from active silicic volcanoes such as Soufriere Hills volcano (Montserrat), Mount Pinatubo (Philippines)

and Mt. St. Helens (U.S.A.) [e.g. Melnik et al., 2008 and references therein]. For long-term fluctuations (years), Barmin et al. [2002] proposed that periodic and non-pulsatory behaviour of magma discharge rate in extrusive eruptions is a consequence of non-linear dynamics and related to rheological stiffening of degassing and crystallizing magma, whereby the period of oscillation is controlled, amongst other factors, by the viscosity ratio between a crystallized region and an underlying crystal-poor magma. For short-term fluctuations (hours), Costa et al. [2012] modelled such periodicity in lava extrusion behaviour of Soufriere Hills Volcano (Montserrat) assuming a stick-slip condition at the shallow conduit wall and a deeper elastic dyke whereby a change in the periodicity can be due to an increase of overpressure in the system (as observed at Soufriere-Hills Volcano when the duration of dome cycles decreased from ca. 12 to ca. 7 hours in response to gravitational unloading of 2 MPa corresponding to ca. 100 m of the dome).

Here, we investigate the influence of the state of the physical system (i.e. crystal content, viscosity ratio, relative proportion of the heterogeneous magmas) at the rheological transition on eruptive style using decompression experiments. We conducted a series of analogue experiments, to mimic a scenario whereby two magmas with different viscosities (liquid viscosities: 10 and 1000 Pa s), are superimposed, one on the other, in a conduit. To account for the contribution to the rheological contrast resulting from shallow degassing, experiments were performed on a series of samples having fixed percentages of analogue crystals (0, 10, 30, and 70 vol. %) in the highly viscous magma. The viscosity ratios obtained at the interface span the range of 10²-10⁶, similar to theoretical estimates for volcanoes [e.g. Barmin et al., 2002; Capponi et al., 2016]. The experiments were performed at average decompression rates of 10⁻² MPa/s and 10⁴ MPa/s, to constrain the role of overpressure in the system. Finally, we investigated the effect of the relative proportions of the highly viscous phase versus the low viscosity magma.

2. Methods

2.1 Experimental apparatus

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The experimental setup centres on a cylindrical Plexiglass high-pressure autoclave (height = 20 cm and internal radius r = 2.0 cm; Figure 1), sealed at its top by a set of copper diaphragms, and attached to an expansion chamber operating at ambient pressure. The sample is located at the bottom of the autoclave, and saturated under Argon-gas at a pressure of 10 MPa for at least 3 days (6 days for Exp10, in accordance to the increased sample volume; **Table 1**); this time is chosen to allow for a complete saturation of the gas phase [Spina et al., 2016a,b]. Once complete saturation is achieved, the sample is then decompressed either fast or slowly. "Slow" decompression (average decompression rate ca. 10⁻² MPa/s; Spina et al., 2016a) is performed by opening a dedicated manual valve in the manometer and allowing gas to evacuate the system. Fast decompression is achieved by induced failure of the set of copper diaphragms sealing the autoclave at its top [Spina et al., 2016a,b] for further information on the setup]. Fast decompression rate is characterized by an exponential decay, approximated by two linear segments of the decompression curve: an initial decompression rate on the order of 10⁴ MPa/s (from decompression initiation up to ca. 70% of pressure decay) followed by a slower decompression of $\approx 10^3$ MPa/s. Like in Spina et al. [2016a], we refer to an average decompression rate of 10⁴ MPa/s as most representative of the depressurization process. Sample degassing is monitored by tracking the temporal evolution of sample expansion and outgassing periodicity using a high-speed camera with sampling rates of 5000 and 50 fps for the instantaneous and slow decompression experiments, respectively (Section 2.2 for more details). The pressure state within the system is recorded by a pressure sensor located at the top of the autoclave. Silicone oils with viscosities of ca. 10 and 1000 Pa s and density of 970 kg/m³ were chosen as proxies for melts with different rheological properties and juxtaposed vertically, with the high viscosity liquid (hereafter Layer H) placed at the top of the low viscosity liquid (here Layer L; **Figure 1b**). To evaluate the role of solid loading in the uppermost section of the conduit (here Layer H) we added in variable percentages (ϕ_1 ; **Table 1**) to the 1000 Pa s silicone oil spherical glass beads, with diameter and aspect ratio (estimated on a population of more than 500 particles) equal to 83±11um and 1.09±0.12.

respectively, and density of 790 kg/m³. A density of the analogue particles similar to that of the silicone oil is a fundamental prerequisite to avoid crystal settling. Note that it is our goal here to address the short-term interaction between two layers at a specific time rather than the long-term evolution of the system, which would involve crystal settling. The particle-bearing Layer H was prepared following Cimarelli et al. [2011]: silicone oil and particles were mixed according to their weight, then centrifuged for at least 1 hour to remove bubbles entrained in the mixture and finally stirred carefully to establish a homogeneous particle distribution. Next, we first poured Layer L into the apparatus, and then we slowly superimposed particle-bearing Layer H on the top. The maximum packing fraction for spheres of equal diameter is about 74 vol.%, whereas random loose packing is about 60 vol.% [Costa, 2005 and references therein]. Keeping in mind that the particle size distribution of analogue crystals, in our experiments, is not completely uniform (random packing of polydisperse spheres is about 87 vol.%; Costa, 2005 and references therein], the value of packing fraction, estimated for our experiments around 70 vol.% (evaluated according to Cimarelli et al., 2011), is within this range. The viscosity of Layer H after the addition of particles was estimated with the model of Costa et al. [2009; Table 1] using the best fit parameters obtained by Spina et al. [2016a]. The addition of particles causes Layer H to appear greyish and more opaque than Layer L. Therefore, it can be readily detected by image analysis. In order to distinguish the two layers in those experiments without particles (Exp01, Exp02) we added a small amount (0.025 g/cm³) of yellow food colorant (E102, E172) to the 10 Pa s oil (Layer L).

Different sets of experiments were performed to identify the role of 1) solid loading; 2) decompression rate, and 3) volume ratio, by changing the proportions of the two layers. A complete description of the experimental conditions is given in **Table 1**.

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2.2. Analytical methods

During decompression, the concentration of dissolved volatiles in the saturated samples eventually exceeds the equilibrium solubility at the lowered pressure and bubbles nucleate. The saturated samples expand under the dominant force of the growing bubble swarm. Then, the mixture enters the outgassing regime and periodic oscillations of the sample surface are observed in response to the degassing dynamics of the foamed sample. A detailed description of the slow and fast decompression of the different systems is given in **Section 3**. To quantify and compare the different behaviour of the investigated systems upon decompression, we measured the time-dependent length of expansion of the flow front F(t), i.e. the total height of the sample at a given time t, measured at the centre of the flow front surface (**Supplementary Table S_table01**). The measured expansion of the sample is a function of bubble nucleation and growth in response to decompression. This parameter has been used previously to characterize gas mobility in two phase systems [e.g. Taddeucci et al., 2006; Oppenheimer et al., 2015; Spina et al., 2016a;b] and provides information on the average velocity v of the flow front of the expanding mixture at a given time t, i.e. $v = (F(t) - F_0)/\Delta t$, as well as the average gas volume ratio $G(t) = F(t)/[F(t) + F_0]$, where F_0 is equal to the initial sample length.

To quantify the timescale of outgassing in the experiments, we automatically counted the number of peaks (i.e. local maxima) and the time elapsed between them in the flow front expansion curve, each representing an outgassing episode followed by the abrupt collapse of the sample surface (for this purpose we used the Matlab© function *findpeaks.m*). The expansive ascent of the fluid is a multi-scale phenomenon, i.e. characterized by the coexistence of timescales related to degassing at different scales, from the slow bulk mixture volume change due to exsolution to the small-scale surface height variations, related to the foam dynamics, that mark outgassing episodes. Hence, we have isolated the latter by picking only local maxima in the flow front expansion curve higher than 2.5 mm and spaced 15 seconds for slow decompression experiments and 0.5 seconds for fast decompression experiments. These parameters have been broadly tested to maximize the detection capability. An example of the obtained results is given in **Figure 2**.

3. Results

3.1 Visual observation at different decompression rates

The results of slow and fast decompression experiments are shown in **Figures 3** and **4**, respectively. The two sets of experiments are characterized by very different expansion timescales and degassing styles.

In the slow decompression experiments gas exsolution in Layer H is observed both in the experiment performed using only the liquid phase (Exp01) and in particle-bearing runs (Exp03, Ep05, Exp07, Exp09 and Exp10). While the expansion of Layer H is proceeding, the nucleation process initiates in Layer L; the generated bubbles ascend to the Layer L-Layer H interface and interact with the solid network. Bubble nucleation in Layer L occurs first homogeneously within the sample; following which bubbles are also nucleated heterogeneously from the bottom of the autoclave. At a later stage, the system is characterized by vigorous oscillations of the surface, marking cyclic outgassing of the analogue foam (Figure 3; Supplementary movies S01, S03, S05, S07, S09, S10), as previously observed in decompressed homogeneous single-layer samples with liquid viscosities above 100 Pa s by Spina et al. [2016a, b]. In all the cases shown in Figure 3 the absence of rough surfaces in the foam is consistent with the inference that no fragile rupture has occurred along the film between bubbles [e.g. Namiki and Manga, 2008].

In experiments Exp01, Exp03, and Exp05 bubbles from Layer L are easily entrained in the crystal- and/or bubble-rich Layer H. In contrast, in experiments performed at 70 vol.% of crystals (Exp07, Exp09, Exp10; **Supplementary movies S07, S09, S10**) bubbles were observed to be decelerating and gathering at the interface. In all experiments, we noticed the development of a region of mingling between the two layers.

In fast decompression experiments all the particle-bearing samples (Exp04, Exp06, Exp08) experienced fragmentation of Layer H, followed by the ejection of sample fragments. For the same system configuration (i.e. same viscosity contrast), the increased shear-rate induces a fragile-like

response of viscous Layer H (**Figures 4, 5; Supplementary movie S04, S06, S08**). In our case, foam expansion precedes the fragmentation by 2.8, 4.8, 5.4 ms in Exp04, Exp06 and Exp08, respectively (**Figure 5**), implying that rapid deformation due to bubble expansion is likely to play an active role in the fragmentation process. Successively, fragmentation occurs; in Exp04 (crystal content equal to 10 vol.%), fragmentation appears along a single surface, that cuts the shallowest portion of the Layer H (**Figure 4b**). The fragment is ejected and falls back above the non-fragmented part of Layer H. At higher crystal content (Exp06, 30 vol.%) sample cracking is more pervasive and generate different fragments that partially fall back on the sample (**Figure 4c**). At the maximum crystal content (Exp08; 70 vol.%) the disruption is completely pervasive, fragmented sample portions are ejected in the low pressure tank or entrained in the ascending Layer L (**Figure 4d**). In all cases, after fragmentation of the Layer H, Layer L, partially or completely unloaded, rapidly degasses and expands, permeating Layer H and reaching the sample surface.

Note that whereas for slow decompression experiments it is evident that Layer H and Layer L are well-coupled, the same is not true for particle-bearing fast decompression experiments, the partially-fragmented Layer H being permeated and finally overlain by bubble-bearing low viscosity Layer L. This distinct response (dominated by bubble expansion for Layer L, characterized by fragmentation for Layer H) prevents efficient coupling and mingling of the two liquids because the interaction between the two layers is occurring for a very short time during the experiment.

3.2 Measurements of sample expansion velocity and outgassing periodicity

In **Figure 6** we report the average velocity v (i.e. ascent speed mm/s) of the flow front of the expanding mixture measured from initial sample expansion until the beginning of the outgassing regime (**Section 2.2**).

For slow decompression experiments (**Figure 6a**), the average ascent velocity measured during the first episode of sample expansion ranges between ca. 0.03 and 0.10 mm/s, as a function of solid loading. A maximum value in the ascent speed is observed at solid loading corresponding to 10 vol.%, followed by a rapid decrease in ascent velocity with particle loading (**Section 4.2**). An increase

in the ascent speed of up to 2 orders of magnitude is observed upon increasing the decompression rate by orders of 10⁶. Decompression experiments performed at an average rate of 10⁴ MPa/s exhibited average initial velocities of 14.2 to 30.9 mm/s. Additionally, in fast decompression experiments, the increase in solid loading of Layer H is accompanied by a clear decrease in the ascent velocity (**Figure 6b**) of the liquid front.

In **Figure 7** we show the height of the sample flow front reached during each outgassing episode (an important indication of the amount of gas in the mixture at that stage; **Figure 7a**) together with the relative outgassing periodicity (**Figure 7b**), measured as the time interval between consecutive outgassing peaks (**Section 2.2**), for different experiments with the same system geometry (Exp09 and Exp10 are reproduced in **Figure 8**). For crystal-bearing experiments at crystal content up to 30 vol.%, the peak height (corresponding to the maximum sample expansion before outgassing) is higher than the experiments performed without glass beads. In general, the height of the peaks decreases as the crystal content increases. The same is observed for the fast decompression runs (**Figure 7a**); however, maximum peak height here is twice that observed during slow decompression experiments. The outgassing periodicity is ca. 1-1.7 seconds when pressure is instantaneously released; a much wider range, spanning in between ca. 15-150/200 seconds, characterizes slow decompression experiments.

The range of peak heights for Exp01 is notably wider than in the other experiments and reflects longer outgassing periods (**Figure 7a, b**). Exp01 was additionally accompanied by a decrease of the peak amplitude from average values of 2.6 mm (maxima: 14 mm) in the first half of the experiment toward average values of 1.28 mm (maxima: 5.37 mm) at the end. This decrease in peak amplitude marks the progressive mingling of the two layers, accompanied by the disruption of the high-viscous foam of Layer H as can be seen from **Figure 3a** and **Supplementary movie S01**.

3.3 Effect of volume ratio of different components

To investigate how the relative volumes of each layer (high viscosity Layer H and low viscosity Layer L) influence the outcomes of experiments, we performed experimental runs featuring the same average decompression rate (10⁻² MPa/s) and viscosity contrast (2.1*10⁶), but 1) with different volume ratios of the two layers (Exp09) and 2) doubling the volume of each layer (Exp10). Results are shown in **Figure 3**, **Figure 6a** and **Figure 8**.

No significant difference in the average velocity of the flow front of the various system configurations was detected: in **Figure 6a** all the experiments performed at 70 vol.% of particles fall within the same area. Similarly, outgassing periodicity (**Figure 8b**) does not differ significantly when accounting for different volume ratios of the Layer H and Layer L.

Conversely, a larger ratio of Layer H/Layer L (Exp07, Exp10) has been linked to a higher height of the outgassing peaks (**Figure 8a**), suggesting an efficient role of the solid loading of Layer H in promoting heterogeneous nucleation in slow decompression experiments. As a result, Exp10, featuring the maximum absolute volume of Layer H, shows the highest peak heights. This likely indicates an increasing amount of degassing volume available in this experimental run.

4. Discussion

4.1 High decompression rate: the fragmentation regime

The effect of decompression rate on gas mobility has been previously investigated both on analogue [e.g. Ichihara et al., 2002; Namiki and Manga, 2006; Kameda et al., 2008, 2013] and natural [e.g. Aldibirov and Dingwell, 2000; Kremers et al., 2010] samples. Two characteristic time-scales have been defined in literature to describe the fragmentation process. The first is the relaxation time, which quantifies the essential condition for reaching the critical stress value for the onset of solid-like brittle failure, defined as:

$$\tau_r = \frac{\eta}{G} \tag{1}$$

where η is the zero-shear viscosity and G represents the unrelaxed rigidity of the material [Barnes et al., 1989; Dingwell and Webb, 1989]. On the basis of ultrasound measurements, Ichihara et al. [2004] estimated a value of G for pure silicone oil (i.e. silicone oil without crystals) with a viscosity ca. 1000 Pa s in the range 0.22-3.9*10⁶ Pa. Equation (1) cannot be simply applied to particle-bearing fluids. that are rather characterized by complex shear modulus; however, considering the case of the crystalfree Exp02 and setting the liquid viscosity equal to 1000 Pas, the relaxation time τ_r for Layer H ranges from 2*10⁻⁴ to 5*10⁻³ s. Decompression curves in **Figure 5a** suggest that the unloading process lasted ca. 2-3*10⁻³ s. As a consequence, for Exp 02, there could have been enough time for the viscous Layer H to relax; coherently, Exp02 featured no fragmentation. The relaxation time τ_r and the decompression time t_{dec} can be combined in a non-dimensional parameter, the Deborah number for decompression processes (De^*_{dt}), i.e. the ratio between relaxation time τ_r and decompression time t_{dec} [e.g. Kameda et al., 2013]. This "operational" Deborah number De^*_{dt} is very convenient for shocktube experiments where decompression time is monitored and should not be directly compared with Deborah number computed using strain rate. Within the above-defined range for G, De^*_{dt} for Exp02 spans the values 0.1-2.5. Kameda et al. [2013] related De^*_{dt} of ca. 1 to potential delayed, partial and intermittent fragmentation, whereas $De^*_{dt} \ll 1$ marked experiments where no fragmentation was observed.

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Addressing the relaxation time of particle- and bubble- bearing fluids requires to account for several decompression rates [e.g. Sumita and Manga, 2008], and to consider the effect of crystals and bubbles loading on the physical properties of the system (e.g. increasing viscosity). At particle fractions above 40 vol.% solid-like behaviour was observed through oscillatory rheology measurements by Namiki and Tanaka [2017], particularly at low interstitial liquid viscosity. In our experiments, at the same decompression rate, the addition of crystals induced sample fragmentation; the amount of crystals in Layer H is positively related to the pervasiveness of fragmentation process (**Figure 4b-d, section 3.1**). In fact, our decompression experiments spanned from the almost monotone single detachment observed in Exp04 (10 vol.%) to the quasi-complete disruption observed

in Exp08 (70 vol.%). Cordonnier et al., [2012] observed that the critical applied stress scales with connected liquid fraction (1- ϕ *), being ϕ * equal to the ratio between crystal fraction ϕ and maximum packing fraction ϕ_{max} . In other words, the critical stress of the liquid has to be corrected for stress localization due to the geometrical effect of particles. The relative decrease of relaxation timescale to trigger fragmentation with increasing solid fraction is likely to be responsible for the observed increase in fragmentation efficiency at high solid loading. It follows that the rapid decompression of microlite-dense shallow regions of the conduit, which originate as a consequence of crystallization driven by decompression and cooling within the shallow conduit, might result in increased fragmentation efficiency. Earlier and deeper fragmentation associated to the presence of crystals was postulated also by Mourtada-Bonnefoi and Mader [2004]. In fact, Mourtada-Bonnefoi and Mader [2004] performed instantaneous decompression experiments on bubble- or particle-bearing GRA solutions, and found that pre-existing bubbles and heterogeneous nucleation due to the presence of particles enhance flow dynamics significantly. Additionally, they investigated the spatial effect of crystal distribution by using either sinking (i.e. silicon carbide, glass, mustard seeds) or floating particles (i.e. coriander seeds) as analogue for crystals. While sinking particles show little effect on flow dynamics, floating particles provided a high concentration of surface bubbles that lead to an earlier fragmentation pulse.

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However, foam acceleration prior to fragmentation has been observed in all the particle-bearing experiments, i.e. the experiments exhibit "liquid-like" fragmentation behaviour, as expected for low viscosity liquids [Kameda et al., 2013]. Hence, we accounted also for the second characteristic time-scale for bubble bearing magmas, that controls viscous expansion of the bubbles [e.g. Thomas et al., 1994, Ichihara et al., 2008]:

$$\tau_{v} = (1 - \psi) \frac{4\eta}{3p_{0}} \tag{2}$$

with p_0 and ψ are the initial pressure and porosity, respectively. For a liquid viscosity of Layer H of 1000 Pa s, an initial pressure of 10 MPa, and investigating a range of initial porosity of 0.01-0.9 the

characteristic timescale τ_v is on the order of 10^{-5} - 10^{-4} s, shorter than decompression time τ_{dec} , slightly above the range estimated for relaxation time scale τ_r . The condition $\tau_v << \tau_{dec}$ is in agreement with the evidence of no fragmentation in Exp02. For particle-bearing experiments, the concurrent increase in mixture viscosity is likely to increase the timescale for viscous expansion, especially for Exp08 (70 vol.%), making bubble viscous expansion a less likely primary mechanism for fragmentation.

Kameda et al. [2017] highlighted the importance of inhomogeneous bubble distribution within the sample inducing stress concentration, causing fragmentation along the surface defined by alignment of bubbles. In our experiments, in principle, we do not have relevant heterogeneities in bubble distribution, as the samples were carefully centrifuged to eliminate entrained bubbles during sample preparation, although in particle-rich experiments (e.g. Exp07, Exp08, Exp09 and Exp10) the very high viscosity of Layer H might prevent the removal of pre-existing bubbles even under centrifugal force. However, heterogeneous bubble nucleation might be responsible for inhomogeneity in bubbles distribution. Additionally, the latter changes as a function of solid loading, shifting toward connected tortuous bubble network as the percentage of solids increases [Polacci et al., 2008; Spina et al., 2016a]; the increasing gas channelling in the solid network might contribute to the pervasive disruption of Layer H observed at larger solid content (e.g. Exp08).

Lane et al. (2008) investigated the post-fragmentation dynamics of rapidly decompressed gum rosin and diethyl mixtures, and reported slow expansive ascent of the samples, accompanied by the occurrence of pressure cycles. According to the authors, bubbles pressure increases due to the confining yield strength and low permeability of the degassed flow head cap of the sample, until bubbles coalesce and collapse, forming a preferential pathway for gas escape. Pressure decrease acts then to self-seal the foam by closing the pathway, leading to the onset of a new cycle. Although there are some remarkable differences in that 1) they do not observe any physical motion of the sample surface coincident with the cycles (likely, as they claim, due to the concurrence of impulsive events at the surface); 2) the samples are characterized by a degassed plug, we believe the source of outgassing periodicity in our experiments to be similar. In fact, as suggested by Spina et al. [2016b]

for decompression experiments performed in the diluted regime for single-layer systems, the repeated foam collapse (as observed here for Layer L in fast decompression experiments and for the Layer H/Layer L system during slow decompression) is likely to be generated by the repetition of local foam collapse events (i.e. opening of gas escape pathways), facilitated by contextual bubble growth and foam shearing.

4.2 Slow decompression experiments: the permeability regime

Slow decompression experiments do not show fragmentation, in accordance with their very low Deborah number for decompression (De*_{dt}<2*10⁻⁵). As detailed below, degassing occurs by permeable outgassing; the extent and style of degassing depend on the crystal loading. This is in accordance with Spina et al. [2016a], that performed slow decompression experiments on single-layer homogeneous particle-free samples with different liquid viscosities (1, 10, 100, and 1000 Pa s) and particle-bearing samples with liquid viscosity of 100 Pa s. Additionally, the authors found no significant sample expansion nor foam build-up and outgassing in particle-free samples with liquid viscosity of 10 Pa s, whereas significant sample expansion, and sample flow front height increase up to 40-50 vol.% was observed in 1000 Pa s runs [Spina et al. 2016a]. In our experimental observations of two-layer systems, clear sample expansion and foam build-up and outgassing was noticed (Section 3.1). This evidence suggests a primary control of the shallower portion of the sample (Layer H) – although volumetrically inferior – in the flow dynamics at the surface.

The addition of solid particles to Layer H increases gas volume ratio within the system due to heterogeneous bubble nucleation, as seen from the higher flow front expansion peaks of Exp03 and Exp05 compared to Exp01 (**Figure 7a**). Further increase in the solid loading causes the level of the mixture to descend. Similarly, the average velocity of the flow front increases due to the addition of solid particles, and decays exponentially with increasing solid content (**Figure 6a**). This is likely to occur in response to the competition between the enhanced heterogeneous bubble nucleation, driving the gas volume increase, and the compelling force exerted by the solid network with increasing solid

fraction, hindering bubble expansion, as previously noticed in homogeneous samples [Spina et al., 2016a]. Additionally, the decrease in the bulk gas content due to the diminished liquid fraction available for saturation at 70 vol.% contributed to suppression of the expansive response of the samples. These observations are consistent with those from experiments performed at high temperature on vesiculating crystal-bearing (50-80 vol.%) hydrous dacite by Pistone et al. [2017]. These authors found that at crystal contents below 60 vol.%, bubble coalescence generates permeability within shear bands; at crystal content of 60-70 vol.% bubbles might become trapped within the solid network and gas expansion is limited, whereas above 80 vol.% large overpressurization triggers brittle fragmentation of the sample. Similarly, we observed limited gas expansion, linked to bubble trapping/coalescence leading to an overall decrease of the gas volume ratio in experiments Exp07, Exp09, and Exp10; on the contrary, lower crystal contents (Exp03 and Exp05) are characterized by larger gas/volume ratio. The positive effect of crystal confinement in favouring gas migration along channels and gas fingering (i.e. bubbles forced into more viscous fluids tend to form finger-like branches) has been proved by numerical and experimental studies [Oppenheimer et al., 2015; Parmigiani et al. 2017]. Parmigiani et al., [2017] found that the favourable crystallinity window for crystal confinement and limited capillary resistance allowing optimal volatile migration is in the range 40-70 vol.%. Above this threshold, an increase of pore pressure by compaction, development of bubble overpressure and capillary fracturing are to be expected [Holtzman et al., 2012; Parmigiani et al., 2017]. Therefore, both our study and previous investigations point to the evidence that the extent of crystallization (hence the growth rate) is tuning the eruptive style by determining optimal windows for gas confinement/migration. Therefore, variation in the crystal growth rate at the shallow portion of the magmatic column likely results in relevant variation of gas permeability. Enhanced gas loss, for instance, may either reduce the potential for explosive eruptions or further promote the formation of dense plugs in the conduit, increasing the probability of Vulcanian eruptions [Lindoo et al., 2017].

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409 Despite a slight tendency of outgassing periodicity to gather in a narrow range at larger crystal 410 volume fraction, no marked effects of crystal content in Layer H on outgassing timescales has been 411 detected in our experiments, suggesting that crystal content is not the primary factor in controlling 412 the frequency of outgassing. Slowly decompressed crystal-rich samples (Exp07, Exp09, and Exp10, 70 vol.%) experienced 413 414 effective mingling between Layer H and Layer L. Indeed, before being entrained in the crystal-rich 415 layer, bubbles slow down at the interface, and are occasionally trapped for few seconds 416 (Supplementary movie S07, S09, S10). When the buoyant bubbles are finally entrained in Layer H, 417 displacement, due to bubble ascent, causes some filaments from Layer H to be dripped down, likely 418 in response to the onset of Rayleigh-Taylor instability [Rayleigh, 1900; Taylor, 1950]. Rayleigh-419 Taylor instability is an almost ubiquitous process in nature that originates when a density gradient 420 due to gravitational force opposite to the vertical pressure gradient occurs. Descending plumes are 421 additionally folded by vortex generated by floating bubbles and stretched by the eddies, due to local 422 Kelvin-Helmholtz instabilities [Kelvin, 1871; Helmholtz, 1868] associated to the relative movement 423 between the upward-moving light gas bubbles from Layer L and the heavier crystal-rich Layer H. By 424 the end of the experiment, several plumes of Layer H had already reached the bottom of the sample 425 and crystals originally absent from Layer L are now distributed throughout the experimental system. 426 Notably, also the Saffman-Taylor instability [Saffman and Taylor, 1958], i.e. the formation of 427 morphological patterns at the unstable interface among two fluids due to the displacement of the more 428 viscous fluid (here Layer H) by the less viscous one (here Layer L), can be invoked to explain the 429 observed plumes. Saffman-Taylor instability has been observed in natural samples and experiments 430 with a notable morphological variability and complexity that depends on the viscosity ratio between 431 the two fluids [e.g. Perugini and Poli, 2005]. 432 Conversely, in fast decompression experiments, the fragile response of Layer H determined a 433 rheological decoupling that prevented effective mingling between the two phases (Figure 4 and

Supplementary movie S04, S06, S08). As clearly visible in Exp08, after the pervasive fragmentation

of Layer H, a relevant number of fragments from Layer H are entrained in the ascending Layer L as isolated crystal-rich blobs (Supplementary movie S08).

The crystal-rich experiments Exp07, Exp09, and Exp10 show analogies with lava dome periodicity. In fact, while experiments performed with lower crystal content are dominated by the dynamics of the foam, at higher crystal content the behaviour of the Layer H is more likely attributable to stickslip behaviour, as previously noticed for homogeneous mixture with more than 40 vol.% in Spina et al. [2016a]. A stick—slip mechanism was invoked also for cyclic behaviour observed during extrusive flow from silicic volcanoes [e.g. Costa et al., 2012]. Moreover, experiments with very high crystal content highlighted the relevance of bubble motion in promoting mingling between the two layers. The relevance of bubble advection in magma mixing for systems with viscosity contrast up to 4000 has also been demonstrated from experimental studies performed on basaltic-rhyolite systems [Wiesmaier et al., 2015]. Furthermore, Del Bello et al. [2015] experimentally observed that gas passing through stratified magma might promote mingling. At shallow level, exsolved volatiles are expected to interact with a partially solid plug, likely promoting mingling among the least evolved and the most crystallized layer. Here we suggest that very high decompression rates, leading to mechanical decoupling between the top layer and the crystal-free fresh magma as observed in Exp08, are more likely to produce enclave morphologies in the volcanic products.

5. Implication for natural systems

The presence of a rheological stratification within the volcanic conduit has been broadly documented in literature [e.g. Denlinger and Hoblitt, 1999; Lautze and Houghton, 2006; Lensky et al., 2008]. The intense processes capable of severely influencing the physical properties of the magma, such as gas-exsolution and crystallization, occur routinely at shallow depth. The extent of crystallization (i.e. crystal nucleation and growth rates) depends on several factors such as the degree of undercooling and the viscosity of the nucleating medium [e.g. Gibb, 1974; Vetere et al., 2013].

We investigated a wide range of viscosity contrasts (10^2 - 10^6) to encompass different degrees of crystallization of the shallow magmatic column. An increase in the absolute viscosity (liquid + crystals) of up to two orders of magnitude has been measured by Kolzenburg et al. [2016] upon cooling and crystallization of a primitive basaltic melt (from 1250 to 1150 °C), at a cooling rate of 3 K/min, similar to that expected in shallow dykes. Capponi et al. [2016] provided a viscosity contrast for Stromboli volcano between the degassed and crystallized plug and the underlying fresh magma in the range 33-3300. Lava dome eruptions are characterized by a viscosity contrast between the upper conduit and the underlying magma on the order of 10-10² [Barmin et al. 2002]. Similarly, in our study, experiments Exp01 to Exp06 have viscosity contrast between Layer H and Layer L on the order of 10². Higher viscosity contrasts (up to more than 10⁶) have been reported in literature in various volcanological settings such as lava flows and explosive eruptions [e.g. Freund and Tait, 1986; Vetere et al., 2015; Morgavi et al., 2016].

The experiments performed in this work mimic the following decompression history of a volcanic system, characterized by two main stages: (i) time zero, representing the condition where two magmas with different physical properties are superimposed within the conduit; such vertical heterogeneity can result from a sharp gradient in the physical properties due to crystallization upon cooling of the shallowest section of the magmatic column or the progressive ascent of different batches of magmas in time; (ii) time t_i outlines the eruptive phase: the system is decompressed or driven to surface by conduit replenishment; as a consequence, bubble nucleation and/or fragmentation occurs.

In volcanic systems, decompression-induced nucleation might occur both as a consequence of an external trigger, as for instance unroofing due to flank collapse [e.g. Belousov et al., 2007; Manconi et al., 2009], or resulting from the unloading of the magmatic column by static decompression [e.g. Poland et al., 2009] or by lithostatic release following the removal of temporal barrier [e.g. Johnson, 1998; Johnson and Lees, 2000; Massaro et al., 2018] or finally, be driven by magma ascent following conduit replenishment. According to Aldibirov and Dingwell [2000],

rock cracking and gravitational driven landslides, respectively. In our fast decompression experiments, we reproduced an average decompression rate of 10⁴ MPa/s, with a total pressure drop of 10 MPa. Remarkably, the hydrostatic pressure drop due to the 18 May 1980 landslide at Mt. St. Helens has been estimated to be up to 20 MPa. According to Namiki and Manga [2006] decompression rates above 2*10⁴ Pa/s are typical of explosive eruption in volcanoes with higher decompression rates (>10⁶ Pa/s) being linked to fragmentation processes. More generally speaking, decompression rates in natural systems spans several log units [Cassidy et al., 2018], hence magmatic systems might be characterized by different decompression timescales. According to Deborah numbers for decompression De*_{dt} estimated for crystal-free analogue magmas (Sections 4.1 and 4.2), the decompression rates explored in this work help constraining the behaviour of layered systems in the "no fragmentation" regime of Layer H (slow decompression experiments: De*_{dt}<<1) and brittle-like to brittle regime of Layer H (fast decompression experiments: De*_{dt}≈1 and De*_{dt}>>1) [Kameda et al., 2013]. Lower bulk strain rates are required to induce brittle transition in crystal bearing magmas [Wadsworth et al. 2017].

Notably, in volcanic systems, subsurface crystallization is generally accompanied by gas exsolution, culminating in gas depletion, if permeable degassing is effective. Alternatively, overpressure is likely to develop when outgassing is not allowed. For instance, with the exception of the very outermost degassed region, the shallowest region of the conduit feeding lava domes bears volatiles and crystals: rapid crystallization leads to intense diffusion of volatiles and growth of bubbles [e.g. Melnik and Sparks, 2005; Costa et al., 2013]. The high bubble volume fraction observed in our experiments in Layer H is coherent with such a process, although experiments exhibit no gas depletion or enrichment in the liquid component of Layer H before decompression.

6. Conclusions

Magmas with the same pre-eruptive composition and volatile content may generate different eruptive styles depending on the degassing modes during conduit ascent. To constrain the role of different parameters, i.e. physical properties and decompression rate, on the degassing styles of nonhomogeneous system we performed decompression experiments on analogue samples. Our experimental data illustrate that decompression rate (hence ascent rate) plays the most influential role on eruptive style, controlling outgassing periodicity and eruptive history. In fact, changing different experimental parameters of similar order of magnitudes for example viscosity ratio and decompression rate the second one shows the higher effect on the eruptive style. Decompression rate, modified over 6 orders of magnitude, showed the most dramatic effect on eruptive style compared to other experimental variables such as viscosity contrast at the interface, that was modified in a similar range (4 orders of magnitude). Thus, for a given volcanic system, very different scenarios can be envisaged when accounting for possible external triggers (such as un-roofing due to flank collapse) that can lead to a significant variation of the decompression rate. In fast decompression experiments, the solid fraction affects the spatial pervasiveness of fragmentation of Layer H, ranging from a complete absence of fragmentation (0 vol.%) to complete disruption (70 vol.%) at the same decompression rate, directly influencing the fragmentation efficiency of the upper layer. In slow decompression experiments, crystal content tunes gas volume ratio and determines the optimal window for gas confinement/migration: this evidence suggests that rate of crystal growth at the shallow portion of the magmatic column severely affects permeability; and that the control exerted by crystallinity on gas loss has direct implications for hazard assessment.

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Figure captions

Figure 1: a) Sketch of the experimental apparatus: the sample is inserted in the High Pressure Autoclave. A manometer, connected to an Argon tank, provides gas for sample saturation. Decompression is performed by blowing the set of diaphragms at the top of the High Pressure Autoclave (average decompression rate 10⁴ MPa/s) or by opening a dedicated valve located in the manometer and evacuating the system (average decompression rate 10⁻² MPa). An oscilloscope allows for the collection of data from the pressure sensor; b) details on the high pressure autoclave and sample configuration.

Figure 2: Flow front expansion curves (blue solid line, left side y-axis) and decompression curves (red solid line, right side y-axis) of (a) Exp06 (average decompression rate 10⁴ MPa/s) and (b) Exp05 (average decompression rate 10⁻² MPa/s). Flow front expansion curves were measured by picking the position of the flow front at its centre using high-speed-videos (sampling rate of 5000 and 50 fps for a) and b), respectively). Red circles represent detections of local maxima corresponding to outgassing episodes. Green dotted lines mark the first expansion phase of the sample.

Figure 3: Snapshots of slow decompression experiments, with increasing solid content in Layer H from the top to the bottom of the picture corresponding to (a) 0 vol.% (Exp01); (b) 10 vol.% (Exp03); (c) 30 vol.% (Exp05); (d-f) 70 vol.% (Exp07, Exp09, Exp10). In (a-d) the volume ratio between Layer H and Layer L is equal to 0.6, whereas in (e) it corresponds to 0.3 and in (f) Exp10 features the same volume ratio as in a-d, but the double absolute volume for each component. Snapshots I to X were taken each 200 s from video trigger (ca. 70s from decompression, at a pressure of 6 MPa). Spatial scale is provided by the diameter of the experimental conduit equal to 2 cm. The yellow line in frames IV and V in (b) marks the top of the sample holder; please note the vertical shift in the relative position between the camera and the autoclave in frames V-X compared to frames I-IV. In (e) frames IX and X are missing, due to a slightly shorter duration of the movie compared to the other experiments.

Figure 4: Snapshots of fast decompression experiments, with increasing solid content in Layer H from the top to the bottom of the picture corresponding to (a) 0 vol% (Exp02); (b) 10 vol% (Exp04); (c) 30 vol% (Exp06); (d-f) 70 vol% (Exp08). Snapshots were taken before decompression (I), at time zero (II) and at 0.2 (III), 0.4 (IV), 2 (V), 4 (VI), 6 (VII), 8 (VIII) and 10 seconds (IX) after decompression. Spatial scale is provided by the diameter of the experimental conduit.

Figure 5: Details of fragmentation process in particle-bearing experiments. a) Pressure curves for fast decompression experiments without (Exp02) and with (Exp04-Exp06-Exp08) particles. b, c, d) Snapshots of Exp04 (10vol.%), Exp06 (30vol.%) and Exp08 (70vol.%), respectively. Spatial scale is provided by the diameter of the experimental conduit, which is equal to 2 cm.

Figure 6: Initial average velocity of the flow front expanding mixture measured from high-speed-videos for slow (a) and fast (b) decompression experiments at different viscosity ratios (i.e. different solid loading in Layer H). The error in the measurement is of the same order of the symbol size.

Figure 7: (a) Height of detected peaks in the flow front expansion curves during outgassing episodes plotted against viscosity ratio; (b) elapsed times between two consecutive outgassing episodes, defined as local maxima in the flow front expansion curve (**Section 2.2**). In (a) and (b) slow decompression experiments are marked by full dots, whereas fast decompressions are indicated by crosses. Pink and azure chart areas mark experiments performed with pure silicone oils and crystal-bearing samples, respectively.

Figure 8: (a) Height of detected peaks in the flow front expansion curves during outgassing episodes and (b) elapsed time between two consecutive outgassing episodes versus volume ratio of Layer H to Layer L of the investigated system configuration.

Table captions

Table 1: Summary of experimental conditions.

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Supplementary files captions

Supplementary movie S01: Accelerated movie (32x) of Exp01, featuring 10 Pa s silicone oil bearing a small amount (0.025 g/cm3) of yellow food colorant (E102, E172) in Layer L, 1000 Pa s silicone oil in Layer H. Average decompression rate is equivalent to ca. 10⁻² MPa/s. The volume ratio between Layer H and Layer L is equal to 0.6.

Supplementary movie S02: Decelerated movie (0.19x) of Exp02 featuring 10 Pa s silicone oil bearing a small amount (0.025 g/cm3) of yellow food colorant (E102, E172) in Layer L, 1000 Pa s silicone oil in Layer H. Average decompression rate is equivalent to ca. 10⁴ MPa/s. The volume ratio between Layer H and Layer L is equal to 0.6.

Supplementary movie S03: Accelerated movie (32x) of Exp03, featuring 10 Pa s silicone oil in Layer L, 1000 Pa s silicone oil bearing 10 vol.% of crystals in Layer H. Average decompression rate is equivalent to ca. 10⁻² MPa/s. The volume ratio between Layer H and Layer L is equal to 0.6.

Supplementary movie S04: Decelerated movie (0.19x) of Exp04 featuring 10 Pa s silicone oil in Layer L, 1000 Pa s silicone oil bearing 10 vol.% of crystals in Layer H. Average decompression rate is equivalent to ca. 10⁴ MPa/s. The volume ratio between Layer H and Layer L is equal to 0.6.

Supplementary movie S05: Accelerated movie (32x) of Exp05, featuring 10 Pa s silicone oil in Layer L, 1000 Pa s silicone oil bearing 30 vol.% of crystals in Layer H. Average decompression rate is equivalent to ca. 10⁻² MPa/s. The volume ratio between Layer H and Layer L is equal to 0.6.

Supplementary movie S06: Decelerated movie (0.19x) of Exp06 featuring 10 Pa s silicone oil in Layer L, 1000 Pa s silicone oil bearing 30 vol.% of crystals in Layer H. Average decompression rate is equivalent to ca. 10⁴ MPa/s. The volume ratio between Layer H and Layer L is equal to 0.6.

Supplementary movie S07: Accelerated movie (32x) of Exp07, featuring 10 Pa s silicone oil in Layer L, 1000 Pa s silicone oil bearing 70 vol.% of crystals in Layer H. Average decompression rate is equivalent to ca. 10⁻² MPa/s. The volume ratio between Layer H and Layer L is equal to 0.6.

Supplementary movie S08: Decelerated movie (0.19x) of Exp08 featuring 10 Pa s silicone oil in Layer L, 1000 Pa s silicone oil bearing 70 vol.% of crystals in Layer H. Average decompression rate is equivalent to ca. 10⁴ MPa/s. The volume ratio between Layer H and Layer L is equal to 0.6. Supplementary movie S09: Accelerated movie (32x) of Exp09, featuring 10 Pa s silicone oil in Layer L, 1000 Pa s silicone oil bearing 70 vol.% of crystals in Layer H. Average decompression rate is equivalent to ca. 10^{-2} MPa/s. The volume ratio between Layer H and Layer L is equal to 0.3. **Supplementary movie S10:** Accelerated movie (32x) of Exp10, featuring 10 Pa s silicone oil in Layer L, 1000 Pa s silicone oil bearing 70 vol.% of crystals in Layer H. Average decompression rate is equivalent to ca. 10^{-2} MPa/s. The volume ratio between Layer H and Layer L is equal to 0.6; the absolute volume of each phase is doubled than Exp01-08. References Aldibirov, M., & Dingwell, D. B. (2000). Three fragmentation mechanisms for highly viscous

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