1 2	Analogue experiments on the rise of large bubbles through a solids-rich suspension: a "weak plug" model for Strombolian eruptions
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10 Abstract

11 Physical interactions between bubbles and crystals affect gas migration and may play a major role in 12 eruption dynamics of crystal-rich magmas. Strombolian eruptions represent an end member for 13 bubble-crystal interactions, in which large bubbles (significantly larger than the crystal size) rise 14 through a crystal-rich near-surface magma. Indeed, volcanoes that produce Strombolian eruptions 15 often generate ejecta with > 30 vol% (often > 45 vol%) average crystallinity. At Stromboli Volcano, 16 Italy, average crystallinity can reach 55 vol%, which is approaching the eruptibility limit for magmas. 17 At such high crystallinities the solids interact mechanically with each other and with bubbles. This 18 complex rheology complicates the two-phase (liquid-gas) slug flow model often applied to 19 Strombolian eruptions. To examine the effect of crystals on bubble rise, we performed analogue 20 experiments in which large bubbles rise in a vertical tube filled with silicone oil and polypropylene 21 particles. The particles have a slightly lower density than the oil, and therefore form a layer of oil + 22 particles at the upper surface. We varied surface pressure, particle volume fraction, length of the 23 particle-bearing cap, and bubble size to examine the ways in which these parameters influence 24 Strombolian-type eruptions. We show that in experiments, suspended solids begin to affect bubble 25 rise dynamics at particle volume fractions as low as 30 vol% (or, when divided by the random close 26 packing value, a normalized particle fraction $\varphi = 0.64$). Bubbles in experiments with higher particle 27 contents deform as they rise and burst through a small aperture, generating surface fountains that 28 begin abruptly and decay slowly, and longer-lasting acoustic signals of lower amplitude than in 29 crystal-poor experiments. In the experiments, particle fractions > 37 vol% (φ > 0.80) generated 30 strong deformations on fast-expanding bubbles that applied a high stress on the cap, but they 31 trapped bubbles that were less overpressured. Qualitatively, the gas release behavior observed in 32 particle-rich experiments is consistent with observations of Strombolian eruptions. Moreover, we 33 estimate that the observed crystallinity of pyroclasts at Stromboli volcano represents $\varphi > 0.8$. From 34 this we suggest a "weak plug" model for Strombolian eruptions that evolves towards a low-viscosity equivalent of Vulcanian-style plug failure with a more crystalline, stronger, and less permeable plug. Importantly, this model allows the rise of several bubbles in the conduit at the same time and suggests that longer-lasting, more pulsatory and complex eruptions may reveal an increase in nearsurface crystallinity, shedding some light on changing conduit conditions that could help determine the different gas rise regimes involved in passive degassing, puffing, and different expressions of Strombolian explosions.

41 <u>Keywords</u>: Eruption dynamics; Stromboli; Source mechanism; three-phase magma; Conduit
 42 processes; Analogue experiments

43 1. Introduction

44 Strombolian eruptions are generally small and frequent, but can be associated with dangerous 45 paroxysms, as demonstrated by recent explosions at Stromboli volcano, Italy (2019), and Fuego, 46 Guatemala (2018). In this way, Strombolian eruptions provide direct (and often long-term) insight 47 into changes in conduit conditions that precede and follow hazardous activity. Nevertheless, while 48 Strombolian eruptions are intensively studied (primarily at Stromboli volcano), our interpretation of 49 their source dynamics is hindered by a major flaw: the traditional two-phase model for Strombolian 50 eruptions neglects the effect of crystals on bubble rise, despite the crystal-rich nature of pyroclasts 51 associated with this activity.

Most models for Strombolian eruptions consider Taylor bubbles (slugs) ascending in a Newtonian liquid (e.g. Seyfried and Freundt, 2000; Blackburn et al., 1976; Vergniolle, 1998; Del Bello et al., 2012; Hasan et al., 2019). As slugs expand during ascent, their internal pressure exceeds that in the magma column above; this self-sustained bubble expansion accelerates the overlying liquid to accommodate the gas volume change and increases bubble overpressure at burst (James et al., 2008, 2009; Del Bello et al., 2012; Lane et al., 2013).

However, pyroclasts ejected by many Strombolian eruptions have moderate to high crystallinities.
For example, pyroclasts contain >55 vol% crystals at Etna, Italy (Polacci et al., 2006; Giordano et al.,
2010; Edwards et al., 2018); 30-40 vol% at Yasur, Vanuatu (Metrich et al., 2011); 45 – 55 vol% at

Stromboli, Italy (Metrich et al., 2010; Landi et al. 2011). This evidence has motivated models
involving more complex near-surface processes including magma mingling (Lautze and Houghton,
2005), conduit convection (Landi et al., 2004, 2011; Beckett et al., 2014), and near-surface
rheological changes (e.g. Gurioli et al., 2014; Del Bello et al., 2015; Gaudin et al., 2017).

Scaled analogue experiments with gas slugs ascending from a low to high viscosity Newtonian liquid have explored the effects of a near-surface viscous transition on slug flow in a cylindrical conduit (Del Bello et al., 2015; Capponi et al., 2016, 2017). These investigations proposed three geometrical configurations of the slug-cap interactions based on the viscosity of the upper layer (the viscous "cap") and the size of the cap relative to the bubble volume (Figure 1). Longer and more viscous caps impede bubble expansion (hence compress bubbles) below the cap, which affects pressure signals recorded at the base of the experiment, burst processes, and acoustic signals at the surface.



Figure 1: Flow configurations in Capponi et al. (2016, reprinted with permission). In configuration 1 (I), the slug is small enough to be fully accommodated within the cap and bursts at the cap surface. In configuration 2 (II), the bubble nose bursts from the cap into the air above while its tail is still in the lower liquid. In configuration 3 (III), bubble expansion during rise causes the lower viscosity liquid to intrude through the upper more viscous liquid, such that bubble burst occurs in the low-viscosity liquid that is emplaced above the cap.

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73 High magma crystallinity will not only increase viscosity but also create a non-Newtonian rheology. 74 Rheometry of synthetic and analogue magmas demonstrates that high crystallinities (> ~20-40 vol% 75 in basalts) produce both yield strengths and shear-thinning behavior (e.g. Hoover et al., 2001; Jerram 76 et al., 2003; Caricchi et al., 2007; Mueller et al., 2010; Picard et al., 2013; Moitra and Gonnermann, 77 2015). Consequently, the slug model has been modified to include both shear-thinning rheology and 78 an increasing viscosity towards the surface, which affect bubble overpressures at burst (Von der 79 Lieth and Hort, 2016). In contrast, Suckale et al. (2016) depart from the slug model, suggesting that 80 Strombolian eruptions occur by tensile failure of a porous, rigid plug, generated by bubble 81 overpressure below the plug and aided by regional tectonic stress.

82 Solid particles also directly affect bubbles by trapping, deforming, and/or splitting them (Belien et 83 al., 2010; Tran et al., 2015; Oppenheimer et al., 2015; Lindoo et al., 2017; Barth et al., 2019). 84 Migration regimes of growing bubbles depend primarily on particle concentrations (Oppenheimer et 85 al., 2015). At low particle fractions, the liquid-particle suspension is approximately Newtonian and 86 growing bubbles are round. At random loose packing (RLP, "the loosest possible random packing 87 that is mechanically stable"; Onoda and Liniger, 1990), the bubbles deform as they grow. At random 88 close packing (RCP, the maximum random packing), gas propagates in a fracture-like pattern. 89 Therefore, interaction with solids in a crystal-rich upper magma must also affect bubble morphology 90 and rise dynamics during Strombolian eruptions. In this context, we will refer to Taylor bubbles in 91 Newtonian liquids as "slugs" but use the term "bubble" for all geometries of bubbles in non-92 Newtonian suspensions.

Here, we use analogue experiments to model gas bubbles rising below and into a particle-laden
layer, investigating how particles in suspension affect the slug rise model for Strombolian eruptions.
We show that particle-rich suspensions affect bubble morphology and bubble rise and generate
burst dynamics consistent with observations at Stromboli Volcano, Italy. Our data suggest a weak

97 plug model as a source mechanism for Strombolian eruptions at hydrous open-system mafic98 volcanoes.

99 2. Activity at Stromboli Volcano, Italy

100 Stromboli Volcano is persistently active with eruption recurrence times of $10 - 10^3$ seconds (e.g. 101 Barberi et al., 1993; Rosi et al., 2013). Eruptions during "normal activity" last up to tens of seconds 102 and include several pulses with durations of 0.05 - 2 seconds (Patrick et al., 2007; Taddeucci et al., 103 2013; Gaudin et al., 2014). The frequency and intensity of normal activity at Stromboli varies with 104 time, sometimes within days (Harris and Ripepe, 2007a). Strombolian eruptions are classified into 105 four sub-categories: type 0 emit very few small particles at high velocity; type 1 eject mainly coarse 106 ballistics; type 2 generate ash-rich plumes with (2a) or without (2b) ballistics (Patrick et al., 2007; 107 Harris et al., 2013; Leduc et al., 2015). Eruptions derive from the North-East (NE), central, and South-108 West (SW) vents, with shorter, louder explosions in the NE (typically type 1), and longer, pulsatory 109 events in the SW (typically type 2; Ripepe and Marchetti, 2002; Chouet et al., 2003; Harris and 110 Ripepe, 2007a).

111 These eruptions produce pyroclasts with an average of 45 – 55 vol% phenocrysts during normal 112 activity (Landi et al., 2004; Metrich et al., 2010). Plagioclase begins to crystallize at 2 – 4 km depth 113 (Métrich et al., 2010), but plagioclase crystallinity rapidly increases ~800 m below the crater (Landi 114 et al., 2004; Agostini et al., 2013). Crystal-poor basalt resides at depths > 7 km and only erupts 115 during paroxysms (Metrich et al., 2010; LaFelice and Landi, 2011). This volatile-rich lower magma 116 provides the gas source for normal Strombolian eruptions. Therefore, a rheological transition from a 117 crystal-poor basalt to a crystal-rich cap begins at 2 - 4 km depth and accelerates at ~0.8 km depth. 118 These depths correspond to estimated slug source depths of 0.8 and 2.7 km (Burton et al., 2007), 119 estimates which assume, however, that bubbles form at chemical equilibrium and ascend quickly 120 without substantial exchange of volatiles with the melt.

121 Estimates of gas flux, however, show that Strombolian eruptions account for less than 10% of the 122 total mass of gas erupted; most outgassing occurs via puffing or quiescent degassing (Francis et al., 123 1993; Allard et al., 1994; Harris and Ripepe, 2007b). Gas compositions indicate that non-explosive 124 outgassing has a shallow source (Burton et al., 2007). Pyroclasts ejected during puffing are less 125 crystalline than in normal explosions (Landi et al., 2011). Puffing generates lower amplitude acoustic 126 signals than Strombolian eruptions, and is generally active in one vent at a time, but migrates 127 between vents, and is therefore interpreted as a train of small weakly overpressured bubbles of 128 shallower origin than those that generate Strombolian eruptions (Ripepe et al., 2007; Landi et al., 129 2011; Lane et al., 2013).

130 3. Methods

131 3.1. Experimental set-up

132 We performed experiments using the same apparatus as Del Bello et al. (2015) and Capponi et al. 133 (2016), although with a modified bubble injection system (Figure 2), and same low-viscosity liquid as 134 Capponi et al. (2016). A vertical 3-m-high tube of internal diameter $D = 0.0257 \pm 0.0001$ m was 135 connected to a bubble trap at its base and to a vacuum pump at the top to reduce ambient pressure 136 (P_{surf} = 0.3 ± 0.1 kPa, 1 ± 0.2 kPa, 50 ± 1 kPa, and 101 ± 1 kPa) and scale for near-surface gas 137 expansion at Stromboli (James et al., 2008; Lane et al., 2013). The tube was filled to 1.43 ± 0.01 m 138 above the bubble trap with silicone oil (Wacker Chemie AG – AS 100; viscosity 0.1 ± 0.05 Pa s; 139 surface tension 0.021 N/m) and polypropylene particles (A. Schulman – Icorene N4420-1200, sieved 140 by hand). Microscope analysis of 200 particles revealed angular morphologies with an average 141 equivalent sphere diameter of 0.8 mm (standard deviation of 0.48 mm; Supplementary Material). 142 The particle density (900 kg/m³) was slightly lower than the silicone oil (990 kg/m³); therefore, the 143 particles rose to the top of the column and, at the start of each experiment, formed a layer of oil + 144 particles (the "cap") above the pure oil. The caps contained particle masses of 5.4, 14, 23, 46, and 92

grams (± 10 %), which correspond to "packed" cap lengths of approximately 3.5, 8, 13, 25.4, and

146 53.5 cm respectively (Table 1).



147 To avoid pre-formed pathways within the cap, it was disrupted between each experiment with large 148 gas slugs that generated turbulence. This introduced small bubbles that foamed when ambient 149 pressure was reduced to experiment pressure, and dispersed particles throughout the tube. Particle 150 concentrations were then controlled by allowing the particles to rise until the cap reached the 151 desired length, so that for the same mass of particles, longer caps had lower particle concentrations. 152 For each experiment a bubble of volume 4, 10, 17, 32 or 49 ml (± 10 %) was injected into the bubble 153 trap and prevented from rising by a closed butterfly valve. When released from the trap, the bubble 154 rose as a gas slug in the tube. Cap lengths were non-dimensionalized as a function of internal tube 155 diameter (D). Particle fractions were divided by RCP to generate a normalized particle fraction, φ , for 156 comparison with other work involving particle-rich suspensions. RCP was measured as 47 ± 2 vol% by

pouring dry particles in a graduated cylinder (D = 28 mm) and tapping/shaking the container for minutes until the bulk volume was constant. We estimated φ by dividing the calculated cap length at RCP by the measured cap length in each experiment. Particle fractions were overestimated when particle-bearing clusters ejected during experiments remained attached to the tube walls. This error was larger for shorter caps because a small loss of particles caused a larger relative change in cap length. Additionally, as particle contents could vary within the cap, all reported particle fractions were averages of the full cap.

Simply allowing the particles to rise should have created a random *loose* packing because of the small density difference between particles and liquid (Onoda and Liniger, 1990). However, closer packings occurred ($\varphi < 0.92$) because residual small bubbles shook the suspension as they ascended through the particle layer during compaction. For our particles, RLP was likely near the median value for packed caps (~40 vol% particles). RCP was not achieved in experiments.

Two differential pressure transducers (Honeywell 163PC01D36; sampling frequency 5 kHz) recorded pressure changes in the air above the liquid surface with respect to the pressure in the vacuum chamber (Lane et al., 2013). Two cameras recorded the experiments: a Basler acA2000-340km filmed the entire column at 300 \pm 0.1 fps, and a Canon Powershot G15 recorded only the particle

Number of exps.	Initial bubble size (mL)	Surface pressure (kPa)	γ (plug-free)	Mass of particles (g)	Cap length (cm)	Dimensionless cap length (D)	Particle content (vol%)	ф
9	4, 10, 17, 32	0.3	8.8 - 70	0	0	0	0	0
15	4, 10, 17, 32, 49	1	0.83 - 10	0	0	0	0	0
8	4, 10, 32, 49	1	0.83 - 10	5.4	3 - 10	1.1 - 4.0	11 – 39	0.24 - 0.84
10	4, 10, 32, 49	50	(1.4 - 17) x 10 ⁻³	5.4	3 – 6	1.2 – 2.5	18 - 36	0.39 - 0.77
4	10, 49	100	(1.6 - 7.8) x 10 ⁻³	5.4	3 – 6	1.3 – 2.2	20 - 35	0.43 - 0.75
8	4, 10, 32, 49	1	0.83 - 10	14	7 – 12	2.8 - 4.8	24 - 42	0.52 - 0.90
12	4, 10, 32, 49	50	(1.4 - 17) x 10 ⁻³	14	8-14	3.2 - 5.6	21 - 37	0.45 - 0.79
8	10, 17, 32, 49	100	(1.6 - 7.8) x 10 ⁻³	14	7 - 13	2.8 - 5.1	23 - 42	0.49 - 0.90
4	10, 32	0.3	22 - 70	22 - 23.5	11 – 13	4.4 - 5.1	38 - 42	0.82 - 0.90
15	4, 10, 32, 49	1	0.83 - 10	22 - 23.5	12 – 28	4.6 - 11.0	17 – 42	0.37 - 0.90
10	4, 10, 32, 49	50	(1.4 - 17) x 10 ⁻³	23	13 – 20	4.9 - 7.9	24 - 39	0.52 - 0.84
12	2, 4, 10, 32, 49	100	(0.32 - 7.8) x 10 ⁻³	23	12 - 14	4.5 - 5.4	36 - 43	0.76 - 0.92
4	17	1	3.5	37.2	20 - 53	8.0 - 20.7	15 – 39	0.32 - 0.84
4	4, 10, 32	0.3	8.8 - 70	46	25	9.6 - 9.9	39 – 40	0.83 - 0.86
4	4, 10, 32, 49	1	0.83 - 10	46	25 – 27	9.6 - 10.5	37 – 40	0.79 - 0.85
1	32	0.3	70	92	53	20.7	38	0.8
2	10, 32	1	2.1 - 6.6	92	52 - 55	20.2 - 21.5	36 - 38	0.77 - 0.81
130	2 - 49	0.3 - 100	0.00032 - 70	0 - 92	0-55	0-21.5	0-43	0-0.92

Table 1: Summary of experimental conditions. A more detailed list is available in Supplementary Material.

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173 layer at 120 ± 1 fps. The images allowed estimates of surface rise velocities, bubble shapes, and 174 whether fragments were ejected during burst. Transducer data and the Basler videos were 175 synchronised through LabVIEW 2014 software.

176 3.2. Suspension rheology

We measured suspension rheology at 20°C using a HAAKE RheoStress1 with a modified concentric cylinder geometry and gap of 11.7 ± 0.1 mm. We ran the rheometer in controlled stress mode at 10 - 50 Pa with increments of 10 Pa, and for 15 – 50 seconds until the strain rate response to the



Figure 3: Rheology of the particle-bearing suspensions. Solid lines are calculated from the empirical model by Mueller et al. (2009, 2011), and dashed lines show how the error on RCP (of \pm 2) modify the model. Black crosses are rheology measurements with a concentric cylinder (this study), and grey crosses are measurements by Mueller (2009) for crushed particles. Together, these three datasets give an overall idea of cap rheology. A: The relative consistency is the consistency (*K*) divided by the viscosity of the Newtonian liquid (0.1 Pa s here). *K* has the dimensions Pa sⁿ and is equivalent to viscosity when n = 1. B: Yield strength. Experiment yield strengths determined through curve fitting were relatively consistent with the experimental data for crushed particles in Mueller et al. (2009). Our first experimentally measured yield strength occurred at 40 vol% particles (Supplementary Material). C: Flow index indicates whether the suspension is shear-thinning (n < 1), shear-thickening (n > 1), or Newtonian (n = 1). Here, our data falls between the crushed particles and model in Mueller et al. (2009), possibly indicating large errors. The apparent shear-thickening in experiments with low φ may also have been caused by particle rise.

induced stress equalized; we recorded the strain rate over the last three seconds. Instrumental errors on viscosity are ± 0.02 Pa s, although greater errors were caused by bubbles in suspension

182 and particle rise. We fitted our measurements to a Herschel-Bulkley (1926) model:

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$$\tau = \tau_y + K \dot{\gamma}^n, \tag{1}$$

184 where τ is applied stress, τ_{y} is yield strength, *n* is flow index, and *K* is consistency.

185 Due to the possibility of large experimental errors, we compared our data to models by Mueller et 186 al. (2009, 2011) by determining K_r (relative consistency), τ_{ν} , and *n* using equations 2.4, 4.1, and 5.2 187 from Mueller et al. (2009), which requires a fitting parameter, φ_m , that represents the particle 188 fraction at which suspensions can no longer be sheared. We use our measured RCP as a best 189 approximation of φ_m . Indeed, although sheared suspensions typically generate $\varphi_m > RCP$, Mueller et 190 al. (2009) generate values close to RCP. The aspect ratio used to determine n was also generated 191 from RCP, using equation 4 in Mueller et al. (2011). Additional details are available in Supplementary 192 Material.

193 3.3. Scaling considerations

We modelled our experiments assuming that rapid (non-equilibrium) bubble expansion near the surface at the volcano scale can be approximated at laboratory scale by varying surface pressure (P_{surf} ; James et al., 2008). James et al. (2009) and Del Bello et al. (2012) define a dimensionless expansion index (y) for slug flow in a Newtonian liquid as

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$$\gamma = \frac{\rho_l g A' V_a}{P_{surf} \pi r^2 (1 - A')},$$
 (2)

with p_l the liquid density, g the gravitational acceleration, A' the dimensionless cross-sectional area of the falling liquid film (equation (28) in Del Bello et al., 2012), V_a the slug volume at P_{surf} if it were at equilibrium, and r the tube radius. Rapid bubble expansion occurs when $\gamma > 1$. Therefore, γ provides an estimate of bubble expansion conditions in the Newtonian liquid below the cap, but ignores the viscous resistance from the cap. Our range of P_{surf} covers the full range of slow (equilibrium) and rapid (non-equilibrium) bubble expansion.

205 The density and viscosity of the lower liquid allowed bubble rise parameters, such as falling film 206 thickness, to be scaled to bubble rise in the lower (basaltic and crystal-free) magma at Stromboli 207 (Supplementary Table 1; Capponi et al., 2016). To account for uncertainty on cap thickness and 208 crystallinity at Stromboli, we covered a range of thicknesses and particle contents in experiments. 209 We did not attempt to scale particle sizes to crystal sizes at Stromboli. Instead, particle sizes were 210 chosen to avoid wall effects and preserve flow properties. We chose a conservative particle size that 211 fit >30 particles in the inner tube diameter to allow for the counter-flow of liquid + particles during 212 bubble rise in the suspension to approach a 10-particle thickness. Smaller particles could have 213 introduced non-negligible capillary and cohesion forces and increased the particle rise (cap "reset") 214 time between experiments.

215 Other simplifications included: smooth, cylindrical walls, no density-driven convection, no bubbles in 216 the solids-rich cap, little variation in particle shape/size, a single liquid phase, no diffusion between 217 phases, and an abrupt change from a particle-absent to particle-rich suspension. We therefore 218 ignored the effects of near-surface crystallization, vesiculation, and outgassing on magma 219 convection (Palma et al., 2011; Beckett et al., 2014; Gurioli et al., 2014) and melt viscosity, and did 220 not account for the decrease in overpressure from non-adiabatic bubble expansion (Bagdassarov, 221 1994). Furthermore, we neglected the effect of crystal-scale bubbles and variations in crystal shape 222 and size on cap rheology. Additional details are available in Supplementary Material.

223 4. Results

After injection, gas slugs rose through the column of silicone oil until they reached the layer of oil + particles (the cap). Subsequent gas migration behaviors depended on both the particle fraction in the cap and the ratio of bubble size to cap length.

4.1. Slug rise in the particle-free liquid

In a tube filled with a Newtonian liquid, the slug base rises at a constant velocity, while the slug nose accelerates until it bursts at the surface (e.g. Seyfried and Freundt, 2000; James et al., 2008, 2013; Lane et al., 2013; Del Bello et al., 2015). In our experiments, the rise velocity of the slug base in the Newtonian liquid below the particle-bearing cap was constant, regardless of cap parameters and surface pressure. On average, slug base velocity U_s = 0.149 m/s (standard deviation 0.005 m/s). This velocity is within error of the calculated velocity of 0.157 \pm 0.009 m/s following Wallis (1969; Supplementary Material).

4.2. Geometrical flow configurations

Slug flow in a liquid overlain by a higher viscosity liquid can be categorized by flow configurations that depend on the relative sizes of the bubble and the cap (Del Bello et al., 2015; Capponi et al., 2016): (1) the bubble is fully encased in the cap at burst, (2) the bubble base is still in the lower liquid at burst, and (3) bubble expansion is such that the lower liquid breaks through the cap (Figure 1). To compare with previous work with Newtonian caps, this section focuses on experiments with surface pressures of 1 kPa because they complement data from Capponi et al. (2016).



Figure 4: A: Flow configurations in Capponi et al. (2016; smaller filled symbols) and in experiments with Newtonian caps in this study (< ~30 vol% particles; large empty symbols). The "boundary 2/3" category describes experiments that were difficult to categorize due to the opacity of the particle layer: the lower liquid may or may not have reached the surface shortly ahead of the bubble. B: Flow configurations in experiments with non-Newtonian caps (> ~30 vol% particles). Since all experiments with > ~30 vol% particles are shown here, viscosities vary over ~1 order of magnitude. The dashed lines guide the eye to the transitions between regimes. All experiments in this figure have surface pressure = 1 kPa.

Our rheology measurements suggested that caps with < 30 vol% particles behaved as a Newtonian fluid ($n \approx 1$; Fig. 3) with effective viscosities between 0.1 Pa s (particle-free) and ~0.8 Pa s (30 vol% particles), which approached those used by Capponi et al. (2016; 1 Pa s). The presence of particles in these experiments did not significantly affect flow configurations (Figure 4A).

At particle contents > 30 vol%, the cap rheology was non-Newtonian (n<1 with a low yield strength, Figure 3). Under these conditions, transitions between configurations occurred at shorter cap lengths (Figure 4B) compared to experiments with particle-poor caps, consistent with the effect of a greater cap viscosity (Capponi et al., 2016). The experiments shown in Figure 4B contained 30 – 43 vol% particles, corresponding to a variation in consistency of slightly over one order of magnitude (Figure 3).

4.3. Rheological flow regimes

Bubble morphology and rise dynamics in the cap depended on cap rheology. Flow regimes were determined through visual characteristics of bubble rise (Figures 5 and 6). In some cases, gradients in particle fraction caused "hybrid" regimes, such that bubbles behaved differently at the bottom and top of the cap.

257 When the cap comprised a dilute particle suspension, bubble expansion in the lower liquid caused 258 the particle-free oil to intrude into the center of the cap ahead of the bubble (Figure 7A). In the cap, 259 the bubble remained axisymmetric but its diameter decreased and it developed a rounded base. This 260 sequence matches observations of slug flow into a Newtonian liquid of higher viscosity (Del Bello et 261 al., 2015; Capponi et al., 2016), and thus we classify this regime as a *slug flow regime* (Figure 5A). At 262 low P_{surf}, the film at the bubble nose broke in several places and discharged a few liquid-particle 263 fragments, in contrast to the particle-free scenario, where bubble burst generated one accelerated 264 droplet.

The *side flow regime* occurred with higher particle fractions. Here, bubble expansion in the lower liquid caused a finger of liquid to intrude the side of the cap and, although the lower portion of the 267 cap was diluted by the intruded liquid, most of the cap rose upwards. When it reached the cap, the 268 bubble nose filled the finger and then slowed as it pushed the cap upwards, while the tail of the 269 bubble continued rising as liquid drained from the falling film at the bubble walls. The bubble thus 270 became shorter and wider, and slowly migrated upwards. The duration of coupled rise of the bubble 271 and cap was greater for smaller bubbles and for longer caps with higher particle concentrations. In 272 the cap, the bubble flowed against the tube wall, usually as a linear continuation of the original 273 finger although in some cases it also spiraled slightly (Figure 5B & 7B). The particle suspension 274 remained in one piece, deforming around the bubble. Bubble burst occurred at the side of the tube 275 and was often accompanied by ejection of clasts that fountained above the suspension surface.



Figure 5: Comparison of the different regimes discussed in this research. These illustrative examples were produced by different experimental conditions.

A further increase in particle fraction induced a transition to the *deformed regime*. Here, bubble expansion in the lower liquid caused the cap to slide upwards as a plug, presumably with a film of oil at the wall; we did not explore whether the dynamics would be significantly different with a rough wall. Again, the bubble paused before entering the cap; the duration of this pause increased with particle fraction. In the cap, the bubble advanced intermittently, the particle suspension often separated into several pieces, and clusters of liquid + particles slid down the side of the tube as the bubble made its way through the cap (Figure 5C & 7B,C). Accelerations and decelerations of the cap surface reflected variations in bubble expansion rate. The bubble burst at the side of the tube and generated a fountain of small clasts (supplementary video 2).

At high particle fractions and low bubble expansion, some bubbles remained trapped indefinitely below (Figure 5D) or within the cap, thus defining the *trapped bubble regime*. Bubbles trapped within the cap initially rose in the side flow or deformed regime, then decelerated and stopped. The trapped bubbles could often be mobilized by injecting another slug at the base of the apparatus, when the sudden displacement of the entire column caused by slug injection and expansion was sufficient to push the trapped bubble upwards and break the cap. This process sometimes had to be repeated several times before the trapped gas was mobilized sufficiently to escape.



Figure 7: Bubble rise in the rheologically layered experiments can be subdivided into stages of ascent, as shown in (A) for a Newtonian cap and (B) for a non-Newtonian cap. (C) Shows tracks of the bubble, the cap, and the surface of an experiment in the deformed regime, for a bubble of initial volume 10 ml and γ > 1. First, the slug rose in the lower liquid (1). When the slug reached the cap (2), bubble behavior depended on flow regime. In the slug flow regime, bubbles flowed seamlessly from the lower liquid into the upper liquid. In the side and deformed regimes, bubbles were temporarily trapped below the cap. As the bubble pierced the cap (3), a portion of the liquid + particle suspension flowed down the side of the tube, forming a thick bubble film that is viscously supported by the tube wall. In stage (4), the bubble was fully encased in the cap, and in (5) the bubble burst at the surface. Note that in stage (4) the shear-thinning rheology is visible in the overall curvature of the bubble base position in (C). Base velocities of conduit-filling bubbles are constant in Newtonian liquids (White and Beardmore, 1962; Viana et al., 2003). Here, the bubble base accelerates upward with time as its increasing overpressure interacts with the shear-thinning cap.

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293 4.4. Regime diagrams

Of the four parameters varied in our experiments (particle fraction, initial bubble volume, surface pressure, and cap length), particle fraction provided the dominant control on regime transitions. Bubble expansion, controlled mainly by surface pressure but also by initial bubble size, had a moderate effect. Cap length, while affecting other flow characteristics, did not affect regimes (Figure 6).

299 Slug flow occurred only at particle fractions <30 vol%, or φ < 0.64, regardless of other parameters 300 (Figure 6, orange dashed line). The departure from the slug flow regime at $\varphi \approx 0.64$ corresponds 301 approximately to the departure from Newtonian rheology (Figure 3C). Immediately above $\varphi \approx 0.64$, 302 bubbles travelled through the cap in either the side flow or deformed regime. The transition 303 between these regimes depended on both the expansion index γ (an indicator of stress on the cap) 304 and particle fraction. In experiments with $\gamma < 1$ (low bubble expansion), the deformed bubble regime 305 was observed in experiments with particle contents >35 vol% (φ > 0.75). At γ > 1, the deformed 306 regime began at ~37 vol% particles ($\varphi \approx 0.80$; Figure 6A).

Bubbles were trapped at $\gamma < 1$ and high particle volume fractions. Notably, all bubbles with $\gamma < 1$ that interacted with a cap containing >37 vol% particles ($\varphi > 0.80$) were trapped, even for thin caps (Figure 6A,B). Bubble entrapment below, rather than within, the cap (Figure 5D) also occurred primarily at $\varphi > 0.80$, although bubble entrapment within the cap occurred at $\varphi > 0.7$, plausibly recording a hybrid regime between trapped bubbles and bubbles that break the surface. All trapped bubbles applied buoyancy forces < 0.6 N on the cap (Figure 6C).



Figure 6: Diagrams of rheological regimes. (A) The dimensionless expansion index γ (James et al., 2009; Del Bello et al., 2012), indicates whether the bubbles could theoretically expand to equilibrium pressure ($\gamma < 1$) or not ($\gamma > 1$) in the absence of a cap. γ gives an indication of stress on the cap from bubble overpressure and expansion for given initial bubble volumes and surface pressures. Dotted lines represent approximate regime transitions, while the dotted grey circle represents an overall area occupied by the trapped regimes. (B) Deconstruction of the effect of cap length and surface pressure on regime transitions. Each box contains experiments at a different surface pressure. Cap length does not affect regime transitions, but surface pressure does have a minor role. Symbols and dotted lines as in (A), with symbol size indicating configurations: large symbols are configuration 1; medium symbols are configuration 2; smallest are configuration 3 and were only observed at $P_{surf} = 1$ kPa due to experimental conditions. (C) Bubble buoyancy force applied on the cap when the bubble reaches the bottom of the cap. (Therefore, values depend on both bubble size and cap length.)

314 4.5. Flow regimes affect surface processes

315 The particle-rich caps also affected surface level rise, bubble burst dynamics, and acoustic signals 316 emitted during burst. Figure 8 compares maximum surface height variations in experiments 317 containing the same mass of particles (with varying cap lengths) for experiment suites with a given 318 bubble size. An increasing surface height indicates bubble expansion. In the particle-free scenario, 319 rapid bubble expansion accelerated the liquid ahead of the bubble, so that surface height increased 320 substantially. At φ < 0.64 (Newtonian cap), the total surface rise was only slightly reduced, as 321 exemplified by experiments with 17 ml bubbles. Maximum heights in experiments in the side flow 322 and deformed regimes were significantly reduced (by about half) compared to the particle-free 323 scenarios, indicating that the gas was more compressed when it reached the surface.



Figure 8: Total surface rise for experiments containing 23 g particles (and 37 g for the 17 mL bubble series) and at $P_{surf} = 1$ kPa. A surface height of 0 indicates the position at t0, which is 1.43 m above the bubble trap. Since particle mass is constant within each series, an increase in particle fraction causes a decrease in cap length. Colors indicate initial bubble size, and shapes indicate flow regime.

Figure 9 shows examples of acoustic signals for bubble burst in a particle-free (single viscosity) experiment and in two experiments with particle-rich caps. In the side flow and deformed regimes, the acoustic signals were more complex and smaller in amplitude than in the slug flow regime. The decrease in amplitude was complemented by longer signal duration, and thus an increase in the time during which expanding gas was emitted from the ruptured surface. Experiments within the deformed regime that had bigger bubbles and longer caps were more likely to generate several acoustic pulses, further increasing the total signal duration. This pulsatory style was occasionally captured in the camera images as impulsive variations in clast ejection velocities.



Figure 9: Example acoustic signals for bubbles of initial volume 32 mL, and at surface pressures of 1 kPa. The signal generated by a particle-free experiment (orange) is significantly louder and shorter than the particle-rich examples, which are in the side flow regime (red), and a side/deformed hybrid regime (purple).

332 5. Discussion

5.1. Flow configurations: comparison with Newtonian experiments

Flow configurations (Fig. 1) are a geometrical consequence of varying bubble and cap sizes; in Newtonian experiments, they control bubble rise beneath the cap, particularly bubble overpressure (Del Bello et al., 2015; Capponi et al., 2017). In our experiments with sufficiently low particle fractions for the cap to be Newtonian ($\varphi < 0.64$), flow configurations were not affected by particles (Fig. 4). In fact, the low φ experiments differed from those of Del Bello et al. (2015) and Capponi et 339 al. (2016) only in the style of slug burst in configurations 1 and 2, where the particle-bearing film 340 above the slug nose broke in several places; the result was emission of several particle-liquid clots 341 rather than one accelerated drop. This multi-source rupture was likely caused by particle-generated 342 discontinuities in the rapidly thinning film. For non-Newtonian caps ($\varphi > 0.64$), our experiments 343 showed that transitions in flow configurations shifted toward shorter caps, consistent with the 344 effects of increased viscosity (Capponi et al., 2016). These observations suggest that flow 345 configurations can be estimated using simplified rheological descriptions of the cap, regardless of 346 bubble behavior within that cap.

347 5.2. Rheological flow regimes

Particle fraction in the cap exerted a major control on regime transitions between slug flow, side flow, deformed bubbles, and trapped bubbles, consistent with Oppenheimer et al. (2015). We compare these transitions to the effects of particles on cap rheology (Figure 3).

351 The first regime transition, from slug flow to side flow, occurred at ~30 vol% particles ($\varphi \approx 0.64$; Fig. 352 6). At this value, shear-thinning and consistency increased dramatically, indicating the onset of non-353 Newtonian rheology and weak interactions between particles. The occurrence of particle clusters 354 with a weak yield strength may have contributed to the irregular shape of the bubble as it rose 355 through a heterogeneous cap. Inefficient packing near the tube walls, where particle concentrations 356 and suspension consistency were lower than in the middle of the tube, may have aided bubble rise, 357 while a small yield strength may have suppressed thin falling films, such that a larger physical 358 dimension was required for the exchange down-flow to overcome yield strength.

The next regime transition, from side flow to deformed bubbles, depended weakly upon stress on the cap (generated through bubble expansion, overpressure, and buoyancy; Fig. 6). γ is an imperfect measure of bubble expansion regimes since it ignores the cap, but it approximates stress on the cap from bubble expansion and overpressure. At low stress ($\gamma < 1$), the transition occurred at $\varphi \approx 0.75$. At higher stress ($\gamma > 1$) the transition occurred at $\varphi \approx 0.80$, a value that corresponds with a significant increase in yield strength in particle-bearing suspensions (e.g. Mueller et al., 2009; Brown et al.,
2011; Moitra and Gonnermann, 2015). Yield strength is generally associated with pervasive networks
of interacting particles (Caricchi et al., 2007; Mueller et al., 2009; Picard et al., 2013).

367 The transition to deformed bubbles at $\varphi \approx 0.75 - 0.80$ occurs at slightly lower φ than the random 368 loose packing (RLP) for spheres at φ = 0.86 (Jerkins et al., 2008). RLP is also associated with pervasive 369 particle networks (Onoda and Liniger, 1990) and deformed bubbles (Oppenheimer et al., 2015). Our 370 experiments show an added constraint of applied stress, however, with the regime transition 371 increasing from $\varphi \approx 0.75$ to 0.80 with increased γ . The high bubble overpressure (≤ 100 kPa) in 372 Oppenheimer et al. (2015) further suggests that RLP may provide the upper bound for transition to 373 deformed bubbles at high stress. Since bubble deformation is controlled primarily by networks of 374 interacting particles, this stress-dependence may suggest that particle networks begin at moderate 375 φ (\approx 0.75) before becoming pervasive at $\varphi \approx$ 0.86. However, other factors may affect regime 376 transitions, such as variable particle shape and size, whether the suspension is static or pre-sheared, 377 and experimental uncertainties.

378

5.3.

Bubble trapping mechanism

379 Bubble rise through the cap requires shearing of the liquid-particle mixture because surface tension 380 prevents bubble rise in the liquid network between particles. As surface tension is negligible on the 381 tube (slug) scale (Bo >> 1), the indefinite trapping of bubbles in or below the cap indicates that 382 buoyancy was insufficient to overcome the strength of the cap in those experiments. Figure 6C 383 shows that buoyancy force and cap rheology (φ) alone do not determine whether a bubble is 384 trapped, and that stresses from bubble overpressure and bubble nose acceleration (reflected in y; 385 Fig. 6A) are also important. Trapped bubbles occurred exclusively for $\gamma < 1$ and $\phi > 0.7$ (Fig. 6A); at ϕ 386 > 0.8, strong particle networks gave the suspension sufficient strength to arrest all bubbles with γ < 387 1. Therefore, bubble trapping may have been enhanced by the smooth experiment walls, which 388 allowed bubble expansion to be accommodated by the cap sliding upwards. However, bubble

389 buoyancy force (< 0.6 N, Fig. 6C) generated a stress on the base of the cap of up to 1200 Pa, which 390 was much greater than the suspension yield strengths (<< 100 Pa based on rotational rheometry; 391 e.g. Mueller et al., 2009, 2011; Tran et al. 2015; Fig. 3), suggesting that bubbles should not have 392 been trapped. The lateral spreading of the bubble may explain this discrepancy. Lateral spreading 393 was driven by deceleration of the bubble nose as it reached the more resistant (and slightly less 394 dense) cap while its base continued to rise in the less viscous pure liquid below. The resulting 395 thinning of the down-flowing annulus of fluid around the bubble reduces the yield stress (relative to 396 the buoyancy stress) required to trap a bubble indefinitely (Dubash and Frigaard, 2004). 397 Furthermore, complete spreading of the bubble to fill the tube below the cap generated horizontal 398 layers; thus gas ascent through the denser cap requires a Rayleigh-Taylor instability of a wavelength 399 limited by the tube diameter (Seropian et al, 2018). Therefore, bubble rise may depend more on the 400 deformation of the cap under its own weight.

401 Yield strength, consistency, and shear-thinning increase with φ (Figure 3). In some experiments, a 402 vertical increase in particle fraction within the cap caused rising bubbles to decelerate and change 403 regime; we classified these as hybrid regimes. Bubble entrapment within the cap occurred over a 404 wide range of average φ (Figure 6), which suggests that either (1) the top of these caps achieved φ > 405 0.8 whereupon bubbles became trapped regardless of cap thickness, or (2) the feedback between 406 energy loss during bubble rise and increase in φ was sufficient to trap bubbles even if $\varphi < 0.8$. In this 407 scenario, bubble expansion was insufficient to maintain stress on the cap, such that the apparent 408 viscosity increased and bubbles decelerated until the stress applied by the bubbles fell below the 409 cap's yield strength and/or the bubbles spread laterally to fill the tube. Therefore, the coupled effect 410 of a shear-dependent viscous response and increasing φ near the surface may be an effective 411 mechanism to trap bubbles in the cap.

412 6. Significance for Stromboli

413 6.1. Flow regimes in near-surface magma

The near-surface basaltic magma at Stromboli contains 45-55 vol% phenocrysts (Landi et al., 2004; Metrich et al., 2010). Rheological investigations of synthetic basalts observe the onset of yield strength at 20-40 vol% crystals (e.g. Philpotts et al., 1999; Jerram et al., 2003; Picard et al., 2013); experiments on andesites show that bubbles begin to deform and connect at ~20 vol% crystallinity (Lindoo et al., 2017). These values suggest that Strombolian magma is likely within or above the crystallinity range for the deformed bubble regime.

420 Our experiments in the deformed regime had pulsatory bubble burst events that lasted longer and 421 decayed more slowly than in the slug regime (Figure 9) and produced small clasts that fountained 422 above the surface (supplementary video 2). These characteristics may reflect smaller burst apertures 423 that vibrate as gas is released. With increasing φ , bubble deformation increases (Oppenheimer et al., 424 2015), hence the bubble aperture is likely to decrease and generate bubble bursts with higher 425 frequency and more gradual gas release (Fig. 9). By comparison, eruptions during normal activity at 426 Stromboli are pulsatory (Taddeucci et al., 2013; Gaudin et al., 2014), last up to tens of seconds 427 (Houghton et al., 2016), have an abrupt start and slow decay (Gaudin et al., 2014), and are 428 accompanied by pyroclast fountains hundreds of meters high. These characteristics are consistent 429 with a highly deformed bubble regime.

430 6.2. Variability of normal activity

The experimental regime transitions occur over a small range of particle fractions (~17 vol% from the onset of side flow to RCP). Hence small variations in solid fraction may cause significant transitions in gas migration regimes. At Stromboli, the NE vent typically has short, loud and ballisticgenerating eruptions (type 1 events) relative to the long, pulsatory, complex ash-generating eruptions from the SW vent (type 2 events; Ripepe and Marchetti, 2002; Harris and Ripepe, 2007a). In our experiments, increasing the particle fraction produced longer burst durations and more 437 complex pulsatory acoustic signals of lower amplitude (Figure 9). This resembles the description for 438 eruptions from the SW vent, consistent with observations that magma in the SW vent is more 439 crystalline (Landi et al., 2011). In contrast, puffing occurs at hotter (lower crystallinity) vents (Landi 440 et al., 2011). We therefore suggest that variations in crystallinity may cause spatial and temporal 441 variations in activity.

442 6.3. Slug model vs Plug model

There is growing evidence that the slug model for Strombolian eruptions requires modification to account for crystal-rich near-surface magma. Alternative models suggest rupture of a crystal-rich plug as a source mechanism (Gurioli et al., 2014; Suckale et al., 2016). Here bubbles accumulate under the plug until (1) the bubble pressure causes the plug to rupture or (2) the plug becomes gravitationally unstable, and the bubble pressure is suddenly released to the surface.

448 The near-surface crystal-bearing magma at Stromboli is likely deeper (0.8 km and 2-4 km depth; 449 Landi et al., 2004; Metrich et al., 2010; Agostini et al., 2013) than the onset of rapid (non-450 equilibrium) bubble expansion ($\gamma > 1$; James et al., 2008; Del Bello et al., 2012), which is estimated to 451 begin at ≤ 100 m below the crater (Del Bello et al., 2012). In such a case, bubbles reach the cap with y 452 < 1, and rapid expansion begins when the bubble is already in the crystal-rich layer. This condition 453 means that bubble rise occurs in a deformed (or other crystal-rich) regime and models predicting 454 overpressure must consider irregular bubble rise in non-Newtonian suspensions. Additionally, if $\gamma < 1$ 455 when the bubble reaches the crystal-rich layer, bubble entrapment may occur either beneath or 456 within that layer. In our experiments with $\gamma < 1$, bubble entrapment is related to yield strength. Since 457 basalts can develop yield strengths at 20 – 40 vol% crystals (e.g. Philpotts et al., 1999; Picard et al., 458 2013), even moderate crystallinities at depth can trap bubbles. Therefore, breaching the cap may 459 require external modifiers such as tectonic stresses and/or modified rheology and compressibility 460 caused by a reservoir of small bubbles in the cap (Suckale et al., 2016).

461 The crystal-rich layer can also enable the accumulation and coalescence of smaller bubbles into 462 conduit-sized bubbles that eventually rise through the plug (Belien et al., 2010; Suckale et al., 2016; 463 Barth et al., 2019). Furthermore, trapped bubbles can occur at any level in the crystal-rich magma. 464 Therefore, while the slug model depends on slow slug formation and rapid slug rise such that only 465 one slug can rise in the conduit at any time, a model where bubbles can be trapped or delayed no 466 longer precludes simultaneous rise from depth of several bubbles. Indeed, magma levels in the 467 conduit appear to increase with gas flux (Ripepe et al., 2002), which may indicate increased bubble 468 entrapment.

469 When analyzed from an equilibrium perspective, gas compositions from normal Strombolian activity 470 suggest source depths of 0.8 and 2.7 km below the craters (Burton et al., 2007). While both source 471 depths may be within crystal-bearing ("high porphyritic") magma, the shallower source corresponds 472 to the estimated depth of the crystal-rich cap (Landi et al., 2004; Agostini et al., 2013), and is 473 associated with weaker eruptions (Burton et al., 2007). Our data suggest that these bubbles could 474 have been trapped or delayed in a crystal-rich plug. Importantly, bubbles change chemistry by 475 volatile exchange with the surrounding melt. Estimates of bubble source depths (e.g., Burton et al., 476 2007) assume equilibrium degassing at depth followed by sufficiently fast ascent to preserve the 477 deep gas signature. This leads to erroneous depth estimates if gas chemistry changed without 478 equilibrating during bubble entrapment or hindered ascent (Pichavant et al., 2013).

The likelihood of bubble entrapment in the Stromboli conduit suggests that a weak plug model is reasonable. Importantly, many volcanoes that have Strombolian activity also have moderate-to-high crystallinities. Investigating eruption behavior at other volcanoes alongside crystal textures may help to constrain the role of crystals in Strombolian eruptions. For example, the 2000 eruption at Etna volcano, Italy, alternated between lava fountains (~35 vol% crystals) and Strombolian eruptions (> 55 vol%; Polacci et al., 2006; Giordano et al., 2010), as did activity during the 1943-1952 eruption of Paricutin (with 40-50% crystals; Erlund et al., 2010). At Tungurahua Volcano, Ecuador, which has 486 intermediate magma composition, Strombolian eruptions have lower crystallinity than (and 487 alternate with) Vulcanian eruptions (Wright et al., 2012). Cashman and Sparks (2013) show that 488 Strombolian and Vulcanian eruptions have similar ranges in crystallinity but differ mainly in melt 489 viscosity. These data support the soft plug model for Strombolian eruptions where near-surface 490 magmas have sufficiently high crystallinity to temporarily impede bubble rise, with stronger, more 491 crystalline, less permeable plugs leading to a low-melt-viscosity equivalent of Vulcanian-style plug 492 failure. At the other extreme are Strombolian eruptions formed by rise of individual large bubbles in 493 crystal-poor lava lakes, such as Kilauea and Erebus (e.g. Gerst et al. 2013; Qin et al. 2018).

494 7. Conclusions

495 Most models for Strombolian eruptions ignore the effect of crystals on bubble rise. Here, we show 496 that particles modulate the rise of conduit-filling bubbles, generating flow characteristics that 497 depend on particle fraction and stress applied by the bubble on the cap.

If the gas bubbles expand slowly (low stress on the cap), particle-rich suspensions can efficiently trap large bubbles, allowing gas to accumulate below or within the cap before rising through the cap. This is essentially a "weak plug" model. At Stromboli volcano, the crystal-bearing magma is deeper or of similar depth to the onset of rapid (high stress) gas expansion. Therefore, Stromboli may have a weak plug.

503 During rapid gas expansion in a particle-rich suspension, bubbles are highly deformed, and therefore 504 burst at the surface through a small aperture. Gradual gas release through a vibrating aperture leads 505 to pulsatory fountains of clasts and longer acoustic signals of lower amplitude, which start abruptly 506 and decay slowly. Since burst aperture decreases at higher particle fraction, these features likely 507 become more pronounced as particle fraction increases. These observations fit with observations at 508 Stromboli volcano, suggesting that variations in explosion duration, pulsations, and fountaining may 509 provide insight into near-surface magma crystallinity. This research suggests that Strombolian eruptions are linked to near-surface crystallization, and that local/temporal variations in crystallinity
 or crystal-bubble interactions may explain variations in degassing style.

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520 9. Supplementary videos

521 **Supplementary video 1** shows bubble rise and burst in an experiment in the slug flow regime (φ = 522 0.36; initial bubble volume = 17 mL). The video is slowed 5x. The start of the video corresponds with 523 bubble injection in the experiment.

Supplementary video 2a shows bubble rise in an experiment in the side flow regime, where bubble burst at the surface was more complex ($\varphi = 0.82$; initial bubble volume = 49 mL). The video is slowed 526 5x and starts at bubble injection. **Supplementary video 2b** shows a close-up of the surface at burst. 527 Video is slowed 10x.

528 10.Bibliography

Agostini, C., A. Fortunati, F. Arzilli, P. Landi, and M. R. Carroll (2013), Kinetics of crystal evolution
 as a probe to magmatism at Stromboli (Aeolian Archipelago, Italy), *Geochim. Cosmochim. Acta*,
 110, 135 – 151, doi:10.1016/j.gca.2013.02.027.

Allard, P., Carbonnelle, J., Metrich, N., Loyer, H., and Zettwoog, P. (1994). Sulfur output and
 magma degassing budget of Stromboli volcano. *Nature*, 368(6469):326–330.

- Bagdassarov, N. (1994). Pressure and volume changes in magmatic systems due to the vertical
 displacement of compressible materials. *Journal of Volcanology and Geothermal Research*, 63(1 2):95–100.
- 537 4. Barberi, F., Rosi, M., and Sodi, A. (1993). Volcanic hazard assessment at Stromboli based on
 538 review of historical data. *Acta Vulcanologica*. 3, 173 187.
- 5. Barth, A., Edmonds, M., and Woods, A. (2019). Valve-like dynamics of gas flow through a packed
 crystal mush and cyclic strombolian explosions. *Nature Scientific Reports*, 9, 821. DOI:
 10.1038/s41598-018-37013-8
- Beckett, F. M., Burton, M., Mader, H. M., Phillips, J. C., Polacci, M., Rust, A. C., and Witham, F.
 (2014). Conduit convection driving persistent degassing at basaltic volcanoes. *Journal of Volcanology and Geothermal Research*, 283:19–35.
- 545 7. Belien, I. B., Cashman, K. V., and Rempel, A. W. (2010). Gas accumulation in particle-rich
 546 suspensions and implications for bubble populations in crystal-rich magma. *Earth and Planetary*547 *Science Letters*, 297(1-2):133–140.
- 8. Blackburn, E. A., Wilson, L., and Sparks, R. S. J. (1976). Mechanisms and dynamics of strombolian
 activity. *Journal of the Geological Society*, 132(4):429–440.
- 9. Brown, E., Zhang, H. J., Forman, N. A., Maynor, B. W., Betts, D. E., DeSimone, J. M., and Jaeger, H.
 M. (2011). Shear thickening and jamming in densely packed suspensions of different particle
 shapes. *Physical Review E*, 84(3):11.
- 10.Burton, M., Allard, P., Mure, F., and La Spina, A. (2007). Magmatic gas composition reveals the
 source depth of slug-driven strombolian explosive activity. *Science*, 317(5835):227–230.
- 11.Capponi, A., James, M. R., and Lane, S. J. (2016). Gas slug ascent in a stratified magma:
 Implications of flow organisation and instability for strombolian eruption dynamics. *Earth and Planetary Science Letters*, 435:159–170.
- 12.Capponi, A., Lane, S., and James, M. (2017). The implications of gas slug ascent in a stratified
 magma for acoustic and seismic source mechanisms in strombolian eruptions. *Earth and Planetary Science Letters, 468: 101-111, https://doi.org/10.1016/j.epsl.2017.04.008.*
- 13.Caricchi, L., Burlini, L., Ulmer, P., Gerya, T., Vassalli, M., and Papale, P. (2007). Non-Newtonian
 rheology of crystal-bearing magmas and implications for magma ascent dynamics. *Earth and Planetary Science Letters*, 264(3-4):402–419.
- 14.Cashman, K. V. and Sparks, R. S. J. (2013). How volcanoes work: A 25 year perspective. *Geological Society of America Bulletin*, 125(5-6):664–690.
- 15.Castruccio, A., Rust, A. C., and Sparks, R. S. J. (2010). Rheology and flow of crystal-bearing lavas:
 Insights from analogue gravity currents. *Earth and Planetary Science Letters*, 297(3-4):471–480.
- 16.Chouet, B., Dawson, P., Ohminato, T., Martini, M., Saccorotti, G., Giudicepietro, F., De Luca, G.,
 Milana, G., and Scarpa, R. (2003). Source mechanisms of explosions at Stromboli volcano, Italy,
 determined from moment-tensor inversions of very-long-period data. *Journal of Geophysical Research Solid Earth*, 108(B1).

- 572 17.Conte, A. M., C. Perinelli, and R. Trigila (2006), Cooling kinetics experiments on different
 573 Stromboli lavas: Effects on crystal morphologies and phases composition, *J. Volcanol. Geotherm.*574 *Res.*, 155(3 4), 179 200, doi:10.1016/j.jvolgeores.2006.03.025.
- 18.Del Bello, E., Lane, S. J., James, M. R., Llewellin, E. W., Taddeucci, J., Scarlato, P., and Capponi, A.
 (2015). Viscous plugging can enhance and modulate explosivity of strombolian eruptions. *Earth and Planetary Science Letters*, 423:210–218.
- 578 19.Del Bello, E., Llewellin, E. W., Taddeucci, J., Scarlato, P., and Lane, S. J. (2012). An analytical model
 579 for gas overpressure in slug-driven explosions: Insights into strombolian volcanic eruptions.
 580 *Journal of Geophysical Research-Solid Earth*, 117.
- 20.Edwards, M. J., Pioli, L., Andronico, D., Scollo, S., Ferrari, F., Cristaldi, A. (2018) Shallow
 controlling factors on the explosivity of basaltic magmas: The May 17–25 eruption of Etna
 volcano (Italy). *JVGR*, doi:10.1016/j.jvolgeores.2018.05.015
- 21.Erlund, E.J., Cashman K.V., Wallace, P.J., Pioli, L., Rosi, M., Johnson E., Delgado Granados H.
 (2010). Compositional evolution of magma from Parícutin Volcano, Mexico: The tephra record.
 Journal of Volcanology and Geothermal Research, 167-187. DOI:
 10.1016/j.jvolgeores.2009.09.015
- 588 22.Francis, P., Oppenheimer, C., and Stevenson, D. (1993). Endogenous growth of persistently active
 589 volcanoes. *Nature*, 366(6455):554–557.
- 23.Gaudin, D., Taddeucci, J., Scarlato, P., Moroni, M., Freda, C., Gaeta, M., and Palladino, D. M.
 (2014). Pyroclast tracking velocimetry illuminates bomb ejection and explosion dynamics at
 Stromboli (Italy) and Yasur (Vanuatu) volcanoes. *Journal of Geophysical Research-Solid Earth*,
 119(7):5384–5397.
- 24.Gaudin, D., J. Taddeucci, P. Scarlato, E. del Bello, T. Ricci, T. Orr, B. Houghton, A. Harris, S. Rao,
 and A. Bucci (2017). Integrating puf fi ng and explosions in a general scheme for Strombolianstyle activity, J. Geophys. Res. Solid Earth, 122, 1860 1875, doi:10.1002/2016JB013707.
- 597 25.Gerst, A., M. Hort, R. C. Aster, J. B. Johnson, and P. R. Kyle (2013), The first second of volcanic
 598 eruptions from the Erebus volcano lava lake, Antarctica—Energies, pressures, seismology, and
 599 infrasound, J. Geophys. Res. Solid Earth, 118, 3318–3340, doi:10.1002/jgrb.50234
- 600 26.Giordano, D., Polacci, M., Papale P., and Caricchi, L. (2010). Rheological control on the dynamics
 601 of explosive activity in the 2000 summit eruption of Mt. Etna, *Solid Earth*, 1: 61-69.
- 602 27.Gurioli, L., Colo, L., Bollasina, A. J., Harris, A. J. L., Whittington, A., and Ripepe, M. (2014).
- Dynamics of strombolian explosions: Inferences from field and laboratory studies of erupted
 bombs from Stromboli volcano. *Journal of Geophysical Research-Solid Earth*, 119(1):319–345.
- 28. Harris, A. and Ripepe, M. (2007a). Synergy of multiple geophysical approaches to unravel
 explosive eruption conduit and source dynamics a case study from Stromboli. *Chemie Der Erde- Geochemistry*, 67(1):1–35.
- 608 29.Harris, A., and M. Ripepe (2007b), Temperature and dynamics of degassing at Stromboli, J.
 609 *Geophys. Res.*, 112, B03205, doi:10.1029/2006JB004393
- 610 30.Harris, A. J. L., Delle Donne, D., Dehn, J., Ripepe, M., and Worden, A. K. (2013). Volcanic plume
- and bomb field masses from thermal infrared camera imagery. *Earth and Planetary Science Letters*, 365:77–85.

- 31.Hasan, A. H., Mohammed, S. K., Pioli, L., Hewakandamby, B. H., Azzopardi, B. J. (2019). Gas rising
 through a large diameter column of very viscous liquid: Flow patterns and their dynamics
 characteristics. *International Journal of Multiphase Flow*. 116, 1 14.
- 616 32.Herschel, W. H. and Bulkley, R. (1926). Konsistenzmessungen von gummi-benzoll osungen.
 617 *Kolloid-Zeitschrift*, 39(4):291–300.
- 33.Hoover, S. R., Cashman, K. V., and Manga, M. (2001). The yield strength of subliquidus basalts experimental results. *Journal of Volcanology and Geothermal Research*, 107(1-3):1–18.
- 34.Houghton, B. F., Taddeucci, J., Andronico, D., Gonnermann, H. M., Pistolesi, M., Patrick, M. R.,
 Orr, T. R., Swanson, D. A., Edmonds, M., Gaudin, D., Carey, R. J., and Scarlato, P. (2016). Stronger
 or longer: Discriminating between hawaiian and strombolian eruption styles. *Geology*, 44(2):163–
 166.
- 35.James, M. R., Lane, S. J., and Chouet, B. A. (2006). Gas slug ascent through changes in conduit
 diameter: Laboratory insights into a volcano-seismic source process in low-viscosity magmas. *Journal of Geophysical Research-Solid Earth*, 111(B5).
- 36.James, M. R., Lane, S. J., and Corder, S. B. (2008). Modelling the rapid near-surface expansion of
 gas slugs in low-viscosity magmas. *Geological Society*, London, Special Publications, 307(1):147–
 167.
- 37.James M.R., Lane S.J., Wilson L., and Corder S.B. (2009). Degassing at low magma-viscosity
 volcanoes: Quantifying the transition between passive bubble-burst and Strombolian eruption.
 Journal of Volcanology and Geothermal Research, 180. 81 88.
- 38.James, M. R., Lane, S. J., and Houghton, B. F. (2013). Unsteady explosive activity. In Fagents, S. A.,
 Gregg, T. K. P., and Lopes, R. M. C., editors, *Modeling Volcanic Processes*, pages 107–128.
 Cambridge University Press.
- 39.Jerkins, M., Schroter, M., Swinney, H. L., Senden, T. J., Saadatfar, M., and Aste, T. (2008). Onset of
 mechanical stability in random packings of frictional spheres. *Physical Review Letters*, 101(1).
- 40.Jerram, D. A., Cheadle, M. J., and Philpotts, A. R. (2003). Quantifying the building blocks of
 igneous rocks: Are clustered crystal frameworks the foundation? *Journal of Petrology*,
 44(11):2033–2051.
- 41.La Felice, S., and Landi, P. (2011). A spatter-forming, large-scale paroxysm at Stromboli Volcano
 (Aeolian Islands, Italy): insight into magma evolution and eruption dynamics. *Bulletin of Volcanology*, 73(9), 1393–1406. Doi: 10.1007/s00445-011-0476-x
- 42.Landi, P., Marchetti, E., La Felice, S., Ripepe, M., and Rosi, M. (2011). Integrated petrochemical
 and geophysical data reveals thermal distribution of the feeding conduits at Stromboli volcano,
 italy. *Geophysical Research Letters*, 38.
- 43.Landi, P., Metrich, N., Bertagnini, A., and Rosi, M. (2004). Dynamics of magma mixing and
 degassing recorded in plagioclase at Stromboli (aeolian archipelago, Italy). *Contributions to Mineralogy and Petrology*, 147(2):213–227.
- 44.Lane, S. J., James, M. R., and Corder, S. B. (2013). Volcano infrasonic signals and magma
 degassing: First-order experimental insights and application to Stromboli. *Earth and Planetary Science Letters*, 377:169–179.

- 45.Lautze, N. C., Houghton, B. F. (2005). Physical mingling of magma and complex eruption dynamics
 in the shallow conduit at Stromboli volcano, Italy. *Geology*, 33(5), 425-428. Doi:
 10.1130/G21325.1
- 46.Leduc, L., Gurioli, L., Harris, A., Colo, L., and Rose-Koga, E. F. (2015). Types and mechanisms of
 strombolian explosions: characterization of a gas-dominated explosion at Stromboli. *Bulletin of Volcanology*, 77(1).
- 47.Lindoo A., Larsen J. F., Cashman K. V., Oppenheimer J. (2017) Crystal controls on permeability
 development and degassing in basaltic andesite magma. *Geology*, 45 (9), 831 834.
- 48.Mader, H. M., Llewellin, E. W., and Mueller, S. P. (2013). The rheology of two-phase magmas: A
 review and analysis. *Journal of Volcanology and Geothermal Research*, 257:135–158.
- 49.Metrich, N., Allard, P., Aiuppa, A., Bani P., Bertagnini A., Shinohara H., Parello F., Di Muro A.,
 Garaebiti E., Belhadj O., Massare D (2011). Magma and Volatile Supply to Post-collapse Volcanism
 and Block Resurgence in Siwi Caldera (Tanna Island, Vanuatu Arc). Journal of petrology 52(6),
 1077-1105.
- 50.Metrich, N., Bertagnini, A., and Di Muro, A. (2010). Conditions of magma storage, degassing and
 ascent at Stromboli: New insights into the volcano plumbing system with inferences on the
 eruptive dynamics. *Journal of Petrology*, 51(3):603–626.
- 51.Moitra, P. and Gonnermann, H. M. (2015). Effects of crystal shape- and size-modality on magma
 rheology. *Geochemistry Geophysics Geosystems*, 16(1):1–26.
- 52.Mueller, S., Llewellin, E. W., and Mader, H. M. (2009). The rheology of suspensions of solid
 particles. *Proceedings of the Royal Society a-Mathematical Physical and Engineering Sciences*,
 466(2116):1201–1228.
- 53.Mueller, S., Llewellin, E. W., and Mader, H. M. (2011). The effect of particle shape on suspension
 viscosity and implications for magmatic flows. *Geophysical Research Letters*, 38:5.
- 54.Onoda, G. Y. and Liniger, E. G. (1990). Random loose packings of uniform spheres and the
 dilatancy onset. *Physical Review Letters*, 64(22):2727–2730.
- 55.Oppenheimer, J., Rust, A., Cashman, K., and Sandnes, B. (2015). Gas migration regimes and
 outgassing in particle-rich suspensions. *Frontiers in Physics*, 3:60.
- 56.Palma, J. L., Blake, S., and Calder, E. S. (2011). Constraints on the rates of degassing and
 convection in basaltic open-vent volcanoes. *Geochemistry Geophysics Geosystems*, 12.
- 57.Patrick, M. R., Harris, A. J. L., Ripepe, M., Dehn, J., Rothery, D. A., and Calvari, S. (2007).
 Strombolian explosive styles and source conditions: insights from thermal (FLIR) video. *Bulletin of Volcanology*, 69(7):769–784.
- Parfitt, E. A. (2004) A discussion of the mechanisms of explosive basaltic eruptions. Journal of
 Volcanology and Geothermal Research, 134 (102), 77-107.
- 58. Philpotts, A. R., Brustman, C. M., Shi, J. Y., Carlson, W. D., and Denison, C. (1999). Plagioclasechain networks in slowly cooled basaltic magma. *American Mineralogist*, 84(11-12):1819–1829.

- 59.Picard, D., Arbaret, L., Pichavant, M., Champallier, R., and Launeau, P. (2013). The rheological
 transition in plagioclase-bearing magmas. *Journal of Geophysical Research-Solid Earth*,
 118(4):1363–1377.
- 60.Pichavant, M., Di Carlo, I., Rotolo, S. G., Scaillet, B., Burgisser, A., Le Gall, N., and Martel, C.
 (2013). Generation of co2-rich melts during basalt magma ascent and degassing. *Contributions to Mineralogy and Petrology*, 166(2):545–561.
- 696 61.Polacci, M., Corsaro, R. A., and Andronico, D. (2006) Coupled textural and compositional
 697 characterization of basaltic scoria: Insights into the transition from Strombolian to fire fountain
 698 activity at Mount Etna, Italy. *Geology*. 34 (3): 201-204. doi: 10.1130/G22318.1
- 699 62.Qin, Z., Soldati, A., Santana, L. C. V., Rust, A. C., Suckale, J., & Cashman, K. V. (2018). Slug stability
 700 in flaring geometries and ramifications for lava-lake degassing. Journal of Geophysical Research:
 701 Solid Earth.doi:10.1029/2018jb016113
- 63.Ripepe, M. and Marchetti, E. (2002). Array tracking of infrasonic sources at Stromboli volcano.
 Geophysical Research Letters, 29(22).
- 64. Ripepe, M., Harris A. J. L., Carniel R. (2002). Thermal, seismic and infrasonic evidences of variable
 degassing rates at Stromboli volcano. *Journal of Volcanology and Geothermal Research*, 118, 285297.
- 65.Ripepe, M., Marchetti, E., and Ulivieri, G. (2007). Infrasonic monitoring at stromboli volcano
 during the 2003 effusive eruption: Insights on the explosive and degassing process of an open
 conduit system. *Journal of Geophysical Research-Solid Earth*, 112(B9).
- 66.Rosi, M., Pistolesi, M., Bertagnini, A., Landi, P., Pompilio, M., & Di Roberto, A. (2013). Chapter 14
 Stromboli volcano, Aeolian Islands (Italy): present eruptive activity and hazards. *Geological Society*, London, Memoirs, 37(1), 473–490. doi:10.1144/m37.14
- 67.Seyfried, R., and Freundt, A., 2000, Experiments on conduit flow and eruption behaviour of
 basaltic volcanic eruptions. J. Geophys. Res., v. 105, no. B10, p. 727-740,
 doi:10.1029/2000JB900096
- 68.Suckale, J., Keller T., Cashman K. V., and Persson P.-O. (2016) Flow-to-fracture transition in a
 volcanic mush plug may govern normal eruptions at Stromboli, *Geophys. Res. Lett.*, 43, 12071 –
 12081. doi:10.1002/2016GL071501.
- 69.Taddeucci, J., Palladino, D. M., Sottili, G., Bernini, D., Andronico, D., and Cristaldi, A. (2013).
 Linked frequency and intensity of persistent volcanic activity at Stromboli (Italy). *Geophysical Research Letters*, 40(13):3384–3388.
- 70. Tran, A., Rudolph, M. L., and Manga, M. (2015). Bubble mobility in mud and magmatic volcanoes.
 Journal of Volcanology and Geothermal Research, 294:11–24.
- 724 71.Vergniolle, S. (1998). Modeling two-phase flow in a volcano. 13th Australasian Fluid Mechanics
 725 Conference.
- 726 72.Viana, F., Pardo, R., Yanez, R., Trallero, J. L., and Joseph, D. D. (2003). Universal correlation for the 727 rise velocity of long gas bubbles in round pipes. *Journal of Fluid Mechanics*, 494:379–398.

- 728 73.von der Lieth, J. and Hort, M. (2016). Slug ascent and associated stresses during strombolian
 729 activity with non-Newtonian rheology. *Journal of Geophysical Research: Solid Earth*, 121(7):4923–
 730 4942.
- 731 74.White, E.T. and Beardmore R.H. (1962). The velocity of rise of single cylindrical air bubbles
 732 through liquids contained in vertical tubes. *Chem. Eng. Sci.* 17, 351-361.
- 733 75. Wright, H. M. N., Cashman, K. V., Mothes, P. A., Hall, M. L., Ruiz, A. G., and Le Pennec, J. L. (2012).
- Estimating rates of decompression from textures of erupted ash particles produced by 1999-2006
 eruptions of Tungurahua volcano, Ecuador. *Geology*, 40(7):619–622.