THE SHAPE OF VOLCANIC CONDUITS INFERRED FROM BUBBLE SIZE DISTRIBUTIONS

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ABSTRACT

The most intense known explosive volcanic eruptions on Earth are Plinian eruptions of silicic magma. Geospeedometers indicate that Plinian magma erupts from high pressure within the magma chamber at average speeds of 0.001-1 MPa/s. At the same time, dissolved magmatic volatiles, predominantly water, nucleate about one quadrillion bubbles per cubic meter of melt, preserved as vesicles within tephra. Vesicles span several orders of magnitude in size, with power-law size distributions, and vesicularities of approximately 70±20 vol.% To date these combined observations of relatively modest decompression rates, a reflection of the fluid dynamics of magma ascent, as well as the exceedingly large bubble number densities with power-law size distributions, a consequence of bubble nucleation and growth kinetics, have never been explained in a quantitatively coherent manner. Here we demonstrate that the integration of these observations requires that bubble nucleation commences when magma starts ascending from within the chamber and continues until the magma fragments to produce tephra, contrary to the conventional view that nucleation occurs as a burst at high supersaturation. We substantiate experimentally that nucleation in rhyolitic melt can occur continuously over prolonged time intervals. We then use integrated modeling of bubble nucleation and fluid dynamics of magma ascent to demonstrate that bubble size distributions in Plinian pyroclasts are the product of continuous nucleation throughout magma ascent, and at average decompression rates that are consistent with geospeedometers. An important outcome of our results is that the transition from magma chamber to volcanic conduit is gradual, resembling a cupola that narrows upward into a conduit.

1 Introduction

The abundant bubbles that form during magma ascent in the conduit power explosive volcanic eruptions. The supercritical aqueous fluid within those bubbles is of lower density than the silicate melt from whence it exsolved [1, 2]. By virtue of conservation of mass the volume of magma, that is silicate melt plus bubbles, increases as it rises toward the Earth’s surface. This results in magma acceleration and expansion [3, 4]. During explosive eruptions the associated strains and strain rates will cause the magma to fragment, which in turn is a defining process of explosive silicic eruptions [5, 6, 7].

Bubbles form by exsolution of volatile elements dissolved in the magma, as a consequence of decreasing magma pressure en route to the surface [8]. Volatile exsolution involves the simultaneous nucleation of new bubbles and diffusion from the melt into the existing bubbles [9, 10]. Volatiles become supersaturated as magma pressure decreases, inducing bubble nucleation [8, 10, 11]. Diffusion of volatiles, in particular H2O, from the melt into existing bubbles causes bubbles to grow [9, 10], while at the same time decreasing supersaturation and increasing magma viscosity [12]. A dynamical feedback between decompression, which increases supersaturation, and diffusion, which decreases supersaturation, ensues. This feedback controls bubble nucleation and growth rates [10, 13]. A record of these intertwined processes is the bubble size distribution (BSD), preserved in pyroclasts as the vesicle size distribution (VSD) [14, 15, 16, 17, 18, 19].

Common characteristics of VSDs from Plinian eruptions are high number densities (~10^{15} m^{-3}), as well as vesicle sizes with a power-law distribution across at least three orders of magnitude. Although VSDs have been interpreted in terms of bubble nucleation and growth [14, 15, 16, 17, 18, 20, 19, 21, 22, 23], which ultimately are the consequence of eruptive magma ascent, they have never been reproduced congruently within the context of the fluid dynamics of magma ascent. Moreover, existing models that predict bubble number density tend to infer magma decompression rates that are at odds with geospeedometers, which yield orders of magnitude lower decompression rates [24, 25]. Here we resolve both discrepancies using numerical modeling of eruptive magma ascent and vesiculation, with the latter encompassing bubble nucleation and growth. Our simulated BSDs reproduce observed VSDs at average decompression rates that are consistent with geospeedometers.

2 Approach

2.1 Continuous nucleation

The continuous power-law vesicle size distributions in pyroclasts are thought to be the result of protracted bubble nucleation throughout magma ascent [20, 21]. Although such continuous nucleation is in principle consistent with nucleation theory [10, 27, 28], broad VSDs as observed in Plinian pyroclasts are usually not produced in nucleation experiments [29, 30, 26]. Moreover, because it is nearly impossible to directly observe bubble nucleation during these high pressure and temperature experiments, continuous nucleation cannot be substantiated through direct observation.

The objective of our experiments is to validate that bubble nucleation during decompression of hydrated rhyolitic melt can indeed be continuous. The experiments consisted of three steps

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Figure 1: Schematic diagram of the magma storage-conduit system envisaged during Plinian eruptions. Bubbles begin nucleating during magma withdrawal within the upper reaches (cupola) of the magma reservoir. Nucleation continues up to the point of magma fragmentation. Bubble size distributions, preserved in Plinian pyroclasts, are produced by a continuous and protracted nucleation process. Inset: scanning electron microscope image of air-fall pumice from the 7.7 ka Plinian eruption of Mt. Mazama, Oregon (courtesy T. Giachetti), and a comparison between VSD in Plinian pyroclasts and in nucleation experiments [26]. Data for eruptions are a composite VSD of Plinian fall deposits from 1912 Novarupta [15] and 7.7 ka Mt. Mazama [14] eruptions.

2.2 Modeling of magma ascent and bubble nucleation

We simulate the fluid dynamics of eruptive magma ascent, together with bubble nucleation and growth during Plinian eruptions, as illustrated conceptually in Figure 1 (see Methods for details). We assume a cylindrical conduit whose radius may vary with depth and solve the steady mass and momentum conservation equations for the change in magma velocity and pressure with depth. The latter provides the boundary condition for the integrated bubble nucleation and growth model. We only consider H₂O, the most abundant volatile phase [32] and driver of bubble nucleation [8, 10], and we assume heterogeneous nucleation [24, 25]. Our objective is to test the hypothesis that VSDs in Plinian pyroclasts are the product of continuous nucleation during magma ascent. Each simulation is therefore constrained to maintain sufficient supersaturation for nucleation. This is achieved by varying conduit size, such that at any given depth the magma decompression rate is slightly greater than the in-
verse characteristic diffusion time. Conceptually this approach is similar that employed in conduit models with a near-lithostatic magma pressure \([33]\). Thus, aside from predicting decompression rate, bubble number density and size distribution, the model also predicts conduit radius as a function of depth.

3 Results

3.1 Experimental verification of continuous bubble nucleation

Our experiments produced bubble number densities that vary over 5 orders of magnitude \((10^3 - 10^{13} \text{ bubbles per m}^3 \text{ of melt})\). Although supersaturation, defined as the difference between the initial and final pressures, drives nucleation, experimental BNDs are not correlated with supersaturation (Supplementary Figures 1 and 2). Instead, experiments within a suite produced BNDs that vary by up to 2 orders of magnitude and are correlated with the annealing time, \(t_{\text{post}}\).

Because BNDs among various suites vary significantly, a synthesis of all experiments requires non-dimensionalization of bubble number density and annealing time. \(t_{\text{post}}\) is scaled by the nucleation time scale, \(\tau\), which represents the time that nucleation rate drops by one e-folding time. Bubble number density \(N_{\text{post}}\) is non-dimensionalized by \(N_{\text{eq}}\), the final bubble number density that a sample could theoretically have attained. For each experimental suite \(N_{\text{eq}}\) and \(\tau\) are estimated by minimizing the difference between predicted (equation 33) and observed bubble number densities (Supplementary Figure 3). The resultant values are similar for samples within a given experimental suite and predominately depend on the initial and final pressures (Supplementary Table 2). Non-dimensional BND is predicted to increase linearly between 0 \(\leq t_{\text{post}} \leq \tau\), after which it reaches its maximum value \([27]\). Thus, non-dimensionalization should collapse all experiments during which nucleation was continuous onto the black line shown in Figure 2, which is the case within the expected experimental variability. Given that the maximum value of \(\tau\) in our experiments was \(\approx 1000s\), our experiments indicate that bubble nucleation in rhyolitic melt can be a continuous process for at least that long.

3.2 Key results for bubble nucleation during magma ascent

Figures 3 and 4 show simulation results for a typical Plinian discharge rate of \(Q = 5 \times 10^7 \text{ kg/s}\) and an initial \(\text{H}_2\text{O}\) saturation at a pressure of 145 MPa, typical values for Plinian eruptions \([3]\). We assume choked flow at the surface \([33, 34]\), and similar results are obtained for different discharge rates and/or saturation pressures. To examine the effect of bubbles that had formed prior to an eruption on bubble nucleation \([35, 36]\), we simulated a case with no initial bubbles (shown in blue) and a case with an initial volume fraction of 0.03 bubbles (shown in red). The simulations shown in thick blue and red lines were obtained by adjusting conduit radius such that the decompression rate is greater than the inverse characteristic \(\text{H}_2\text{O}\) diffusion time at all depths above where the first bubbles nucleate (for more details see equation (30) in Methods). In other words, the resultant conduit radius gives decompression rates at which supersaturation and nucleation are maintained throughout magma ascent. No other parameters were adjusted to obtain the simulation results shown. Figure 4 shows the modeled BSDs together with VSDs in pyroclasts from several Plinian eruptions. The close match is strictly a model outcome of continuous nucleation. It is not a consequence of ‘tuning’ model simulations to achieve a match. For all cases shown, the match between predicted bubble size distributions and those measured in Plinian pyroclasts is remarkable and suggests that bubble nucleation during eruptions is indeed continuous throughout magma ascent, presumably commencing shortly above the magma chamber or perhaps already during magma withdrawal from the chamber.

The thin blue and red simulations illustrate model sensitivity to variations in predicted conduit radii, relative to the two base cases for which conduit radius was constrained to assure continuous nucleation. Conduit radii were shifted to smaller and larger values relative to the base cases. The key result is an upward narrowing conduit is necessary for continuous nucleation. Variations in conduit radius primarily affect decompression rate, fragmentation depth, and bubble number density, but still yield results for all relevant parameters that are well within range of observed or inferred values for Plinian eruptions. At very low nucleation rates \((J < 10^9 \text{ m}^{-3} \text{ s}^{-1})\) nucleation is quite sensitive to small variations in conduit radius (decompression rate), resulting in oscillations in nucleation rate. These, however, quickly dampen as bubble number densities, \(N_{\text{in}}\) exceed values of \(10^{10}\) bubbles per \(\text{m}^3\) of melt.

3.3 Eruption dynamics

As bubbles nucleate and grow, the distance between bubbles decreases. Consequently the characteristic diffusion rate, which scales as the inverse squared distance between bubbles, increases.
Figure 3: Simulation results for a mass discharge rate of $5 \times 10^7$ kg/s and H$_2$O saturation pressure of 145 MPa. The blue and red curves represent simulations with three percent and without pre-existing bubbles, respectively. The results are shown from nucleation onset until magma reaches the surface. (a-b) Pressure decreases as magma ascends, due to hydrostatic and viscous pressure loss. (c) As a consequence H$_2$O becomes supersaturated, causing heterogeneous bubble nucleation. (d) Consequently bubble number density increases as magma ascends to shallower depths. (e) Nucleation, in conjunction with diffusion of H$_2$O into existing bubbles, as well as decompression of the fluid inside bubbles, results in an increase of volume fraction of exsolved H$_2$O. (a-e) At the depth of magma fragmentation there is an abrupt change in the trend of the aforementioned variables. (f) Conduit radius as a function of depth required to obtain continuous nucleation during magma ascent.

To maintain supersaturation and nucleation, decompression rate has to accelerate with distance above the magma reservoir. This acceleration is in part due to the upward narrowing conduit. Together with bubble nucleation and growth, it increases the
average magma velocity, which in turn increases the viscous pressure loss. At the same time and also due to bubble nucleation and growth, the average water concentration within the melt decreases, which increases magma viscosity and further enhances the viscous pressure loss. Eventually this results in a runaway feedback between decompression rate, water exsolution and viscosity. In Figures 3a and 3b this manifests itself as the pronounced curvature in $P_m$ and decompression rate prior to fragmentation. The rapidly accelerating decompression rate results in increasing supersaturation and therefore an increasing nucleation rate (Figure 3c). At the same time bubble number density and volume fraction of bubbles also increase (Figures 3d & e). Once the feedback between decompression rate, water exsolution and viscosity becomes dominant the conduit radius no longer needs to decrease in order to maintain supersaturation (Figure 3f). Instead, it is this runaway feedback that produces the high overall bubble number density. The shift in dynamics due to the feedback is associated with the small diameter tail in the bubble size distribution, which can also be found in Plinian VSDs, where it is associated with a decrease in the power-law exponent of the distribution (Figure 4).

Upon fragmentation gas pressure inside bubbles quickly equalizes with magma pressure. This is a consequence of permeable gas flow from pyroclasts into the surrounding fractures. It produces a rapid increase in gas volume fraction as the gas-pyroclast mixture expands (Figure 3e). Outgassing of the pyroclasts is simulated using a pore-pressure relaxation rate (equation (23) in Methods), using pyroclast permeabilities [37]. At the same time decompression rate decreases, because of the abrupt change in viscosity, which is much lower for the gas-pyroclast-mixture above fragmentation than for the bubbly magma (Figure 3b). As a consequence there is rapid water diffusion out of the melt and loss of supersaturation conditions. H$_2$O continues to exsolve from the melt inside the pyroclasts en route to the surface, contributing together with the expansion of the exsolved H$_2$O to the continued increase in gas volume fraction (Figure 3e). The mixture of gas-pyroclasts thus accelerates and reaches choked flow conditions at the conduit exit [3, 33, 38]. We assume that bubbles do not nucleate and grow within the pyroclasts after fragmentation. In other words, any remaining dissolved H$_2$O exsolves through diffusion into existing bubbles and then enters the gas phase that surrounds pyroclasts by permeable flow within the pyroclasts [39].

4 Discussion

Continuous nucleation reconciles multiple observations from Plinian eruptions. When such nucleation is incorporated into numerical models for eruptive magma ascent the predicted bubble number number densities and vesicularities are within the range of Plinian pyroclasts (Figure 3). Moreover, the predicted BSDs also mirror those measured in Plinian pyroclasts (Figure 4). They span several orders of magnitude in size. In addition, the observed change in power-law exponents from $d = 3-4$ to $d = 1-2$ with decreasing vesicle size [15, 19, 16, 14] is also present in the simulated BSDs. This change in slope of the size distribution can be attributed to the aforementioned accelerating feedback between viscous pressure loss and nucleation. Although magma decompression rate increases continuously as magma rises toward the level of fragmentation, the predicted

![Figure 4: Simulated bubble size distributions corresponding to the simulations of Figure 3 (red and blue curves), together with distributions from Plinian pyroclasts (gray curves). The latter are from the 1912 Novarupta eruption in Alaska[15], the 1875 Askja eruption in Iceland[16], the 2008 Chaiten eruption in Chile[19], and the 7.7 ka Mazama eruption, Oregon[14]. Plinian BSDs have been subdivided into two overlapping power law distributions with exponents of $d \approx 3-4$ and $d \approx 1-2$. In the simulations the latter size fraction of the distribution represents bubbles nucleated during the rapid increase in decompression rate in the depth range where the feedback between water exsolution and viscous pressure loss accelerates up to fragmentation.](image-url)
or not some bubbles already exist in the magma prior to eruption. A necessary condition is that conduits widen with depth and transition via a broad cupola into the upper reaches of magma reservoirs.

5 Methods

We model the one-dimensional fluid dynamics of steady magma flow in the volcanic conduit, coupled with the nucleation and growth of H$_2$O bubbles. Simulations start at H$_2$O saturation, corresponding to a pressure $P_{H_2O}$. Assuming a constant lithostatic pressure gradient of $ρ_{rock}g$, where $ρ_{rock} = 2400$ kg/m$^3$ is rock density and $g$ is gravity acceleration, the saturation pressure $P_{H_2O}$ corresponds to a depth, $Z_0 = P_{H_2O}/(ρ_{rock}g)$. Simulations end when magma reaches the surface at $Z = 0$. As magma flows up the volcanic conduit magma pressure, $P_m$, decreases due to hydrostatic and viscous pressure loss. The magma thus becomes supersaturated in H$_2$O, resulting in bubble nucleation. At the same time H$_2$O diffuses from the melt into existing bubbles, as they grow. The fluid dynamics of magma ascent are coupled with the nucleation and growth of bubbles through magma pressure $P_m$, which provides a depth-varying boundary condition for the bubble nucleation and growth calculations. In the subsequent sections we will detail the methodologies for employed to model magma flow in the conduit as well as bubble nucleation and growth within the ascending magma.

5.1 Magma flow in the conduit

We assume a vertical cylindrical conduit, whose radius can vary with depth, $Z$. We assume the flow is steady because the duration of magma ascent is much shorter than the duration of Plinian eruptions [43]. We assume flow is one dimensional and integrate flow properties over the cross-sectional area of the conduit. Below the level of fragmentation we define magma as the mixture of silicate melt and H$_2$O bubbles. Above the level of fragmentation magma is the mixture of continuous H$_2$O vapor with suspended fragments of vesicular magma, that is pyroclasts. Throughout the relative velocity between the two phases (melt and H$_2$O vapor) is negligible. Below fragmentation this is justified because the buoyant rise velocity of bubbles is negligible, given the large melt viscosity [33]. Above fragmentation it is a commonly employed approximation [3, 33] and one that does not affect the salient results of our simulations in any significant manner. The properties of the mixture, that is of the magma, are the volumetric average of the two phases. The flow is furthermore assumed to be isothermal, another common approximation that does not significantly impact bubble nucleation rate [38].

With these assumptions, conservation of mass and momentum are

\[
\frac{∂(ρAuA)}{∂z} = 0,
\]

and

\[
ρau \frac{∂u}{∂z} = -\frac{∂p_m}{∂z} - ρu^2g - F_{fic},
\]

respectively. Here $\rho$ is magma density, averaged over liquid and gas phases,

\[
ρ = φρ_u + (1 - φ)ρ_l.
\]

$φ$ is the volume fraction of bubbles, $u$ is magma ascent rate, $A = πa^2$ is the conduit cross sectional area, $a$ is conduit radius, and $ρ_u$ and $ρ_l = 2400$ kg/m$^3$ are gas and melt densities respectively. $F_{fic}$ is the frictional pressure loss estimated as $ρu^2f/α$ where $f$ is the friction factor. Before fragmentation $f = 16/Re + f_0$ and after fragmentation $f = f_0$. $Re = 2ρuμu/η$ is the Reynolds number, $η$ is the viscosity of the mixture, and $f_0$ is a factor related to conduit wall’s roughness and assumed to be 0.0025 [33].

6 Results

By substituting equation (1) into (2) one obtains

\[
\frac{∂p_m}{∂z} = ρu^2g + F_{fic} - \frac{ρu^2f}{A} \frac{∂A}{∂z} - u^2 \frac{∂p_m}{∂z}.
\]

Here

\[
\frac{∂p_m}{∂z} = \frac{∂p_m}{∂z} - \frac{∂p_m}{∂z} \frac{∂p_m}{∂z},
\]

and given that under constant entropy

\[
\frac{∂p_m}{∂z} = c^2,
\]

where $c$ is the speed of sound within the magma. Equation (4) can thus be simplified to

\[
\frac{∂p_m}{∂z} = \frac{ρu^2g + F_{fic} - \frac{c^2}{1 - M^2}}{1 - M^2},
\]

where $M = u/c$ is the Mach number of the mixture. The sound speed before fragmentation is estimated from $c^2 = K_m/ρ_m$ where $K_m$ is bulk modulus of the mixture

\[
\frac{1}{K_m} = \frac{ρ}{K_1} + \frac{1}{K_1} - \frac{1}{K_1}.
\]

The bulk modulus of the gas phase, $K_g$, is calculated from equation of state [2] and the liquid is assumed to be incompressible [38]. The sound speed of the gas-pyroclast mixture after fragmentation is assumed to be equivalent to the sound speed in the gas phase [34]. Alternate models for the sound speed of the gas-pyroclast mixture [33] do not affect the outcome of the model results in terms of bubble nucleation, which occurs prior to fragmentation.

5.2 Bubble nucleation and growth

We assume H$_2$O is the only volatile phase because it is most abundant and controls final bubble number density [32]. H$_2$O exsolves through bubble nucleation and diffusion into already nucleated bubbles. We assume nucleation is heterogeneous and facilitated by abundant pre-existing magnetite nanolites [24, 25]. We use the far field approximation to calculate H$_2$O diffusion from melt into bubbles [10]. Lastly, we assume that permeable escape of H$_2$O vapor from the bubbles is negligible before magma fragments, but account for it after fragmentation. The bubble nucleation and growth model is in the Langrangian frame of reference. It is integrated to the steady state equations for magma flow in the conduit, which is in Eulerian frame of reference, by considering $d/dt = u∂/∂z$.

The number of bubbles in volcanic systems is too high to track growth of each individual bubble. We therefore use the method of moments to calculate the evolution of the bubble population [10]. The corresponding moments of the population, $μ_k$, are defined as

\[
μ_k(t) = ∫_0^∞ R^kΛ (R, t) dR,
\]

where $R$ is the bubble radius, $Λ (R, t)$ is the bubble population per unit volume of melt within the interval of $R$ and $R + dR$, and subscript $k = 0$-3 refers to the order of the moment. Each moment refers to a measurable characteristic quantity [10]. $μ_0$ is the bubble number density, that is the number of bubbles per unit volume of melt, $μ_1$ is the sum of bubbles radius, $4πμ_2$ is the total surface area of bubbles, and $4/3σμ_3$ is the total volume of bubbles. The evolution of moments through time is given by

\[
\frac{dμ_k}{dt} = J,
\]

and for $k ≥ 1$, by

\[
\frac{dμ_k}{dt} = kG(μ)μ_{k-1} + JR^k_e,
\]

where $J$ is nucleation rate of bubbles, $G(μ)$ is the growth rate of bubbles assumed to be equal for all bubbles and equivalent to the growth rate of a bubble with mean bubble size, $R = μ_1/μ_0$, and $R_e$ is the critical size for nucleating bubbles.
5.2.1 Below fragmentation

We use classical nucleation theory to estimate nucleation rate of stable bubble nuclei as a function of supersaturation pressure. Nucleation rate of stable bubbles are [11]

\[ J = J_0 \exp \left( -\frac{W}{k_B T} \right), \]  
(12)

and bubbles are stable if they are larger than a critical size, \( R_c \), given by

\[ R_c = \frac{2\gamma}{p_m - p_a}. \]  
(13)

Here \( T \) is the absolute temperature, \( k_B \) is the Boltzmann constant, \( \gamma \) is the surface tension of bubble nuclei, \( P_b \) is the pressure inside a bubble nucleus, and \( P_m \) is pressure in the mixture. \( p_m \) is related to the saturation pressure of volatiles, \( p_{sat} \), through [44]

\[ f(p_{sat}, p_m) = f(p_{sat}, T) p_{sat} \exp(\Omega (p_v - p_{sat})/k_B T), \]  
(14)

where \( f(p, T) \) is the fugacity coefficient, and \( \Omega \) is the volume of volatile molecules. \( W \) is the change in free energy as a result of nucleation of a bubble and is given by

\[ W = \frac{16\gamma^3}{3(p_m - p_a)^2}, \]  
(15)

where \( \alpha \) is the heterogeneous nucleation factor and depends on the contact angle, \( \theta \), between bubble nuclei and pre-existing crystals. It is defined as

\[ \alpha = \frac{(2 - \cos \theta)(1 + \cos \theta)^2}{4}, \]  
(16)

and we assume nucleation is facilitated by magnetite crystals with \( \theta = 145^\circ \) [45, 46].

The pre-exponential factor, \( J_0 \), in equation (12) is defined as

\[ J_0 = \frac{2\Omega n_0^2 D}{a_0} \sqrt{\frac{\gamma}{k_B T}}, \]  
(17)

where \( n_0 \) is the concentration of volatiles molecules in the melt, \( D \) is the diffusion coefficient, \( a_0 \) is the average distance between volatiles molecules.

After nucleation bubble nuclei grow by H$_2$O diffusion, because the concentration of H$_2$O at the bubble-melt interface, \( C_b \), is lower than the concentration in the surrounding melt, \( C_m \). The H$_2$O flux into bubbles is given by

\[ q = D \frac{\partial c}{\partial r} \bigg|_{r=R}, \]  
(18)

where \( D \) is diffusion coefficient, \( r \) is the distance from bubble’s center, \( R \) is bubble radius, and \( c \) is the water concentration in the surrounding melt given by

\[ \frac{\partial c}{\partial t} + \frac{dR}{dt} \frac{\partial c}{\partial R} = \frac{1}{r^2} \frac{\partial}{\partial r} \left( r^2 \frac{\partial c}{\partial r} \right). \]  
(19)

At low supersaturation pressure the left hand side in equation (19) can be neglected [47] and the concentration gradient at the melt-vapor interface can be approximated as

\[ \left. \frac{\partial c}{\partial r} \right|_{r=R} = \frac{C_m - C_b}{R}. \]  
(20)

The mass of H$_2$O inside bubbles, \( m_b \), will increase due to diffusion at a rate

\[ \frac{dm_b}{dt} = (4\pi \mu c) \eta \rho \gamma q, \]  
(21)

whereas the bubble growth rate is given by [48]

\[ G(\tilde{R}) = \frac{R}{4\eta} \left( p_m - p_a - 2\gamma \right), \]  
(22)

where the inertia terms are neglected [10]. Here \( \eta \) is the viscosity of melt, \( p_m \) is the pressure the H$_2$O fluid (vapor) inside bubbles, estimated using the equation of state for H$_2$O.

5.2.2 Above fragmentation

After fragmentation we assume bubbles that bubbles no longer nucleate or grow [39]. In other words, \( J = 0 \) and \( G(\tilde{R}) = 0 \). H$_2$O, however, continues to exsolve into bubbles with a rate given by equation (18). Exsolved H$_2$O escapes by permeable flow from within the pyroclasts into the gas phase surrounding the pyroclasts. Consequently, the gas pressure within pyroclasts decreases at a rate of

\[ \frac{dp}{dt} = \frac{p_m - p_a}{\tau_k}, \]  
(23)

where \( \tau_k \) is the characteristic time scale for permeable outgassing estimated from Darcy’s law as

\[ \tau_k = \frac{\bar{L}^2}{k/(\eta \beta \phi)}. \]  
(24)

Here \( l \approx 10 \text{cm} \) is the characteristics length scale [39], \( k \approx 10^{-12} \text{m}^2 \) is permeability [37], \( \eta_\phi = 10^{-5} \text{Pa.s} \) is viscosity of the gas phase, and \( \beta \) is the compressibility of the gas phase, and \( \phi \) is porosity of bubbles in the pyroclasts.

Lastly, the concentration of dissolved H$_2$O and thus the saturation pressure, both below and above fragmentation, decrease as a result of the diffusion of water into bubbles. The resultant conservation of H$_2$O requires that

\[ \frac{dp_{sat}}{dt} + \frac{dC_m}{dt} = -\frac{1}{\rho_m} \left( M_0 \frac{dm_b}{dt} + J_m \right), \]  
(25)

where \( \rho_m \) is the melt density, assumed to be constant throughout magma decompression, and \( m_b \) is the mass of a bubble nuclei estimated from equation of state.

5.3 Model simulation

The parameters in the governing system of equations are either specified or calculated from existing formulations: H$_2$O solubility [49], diffusion coefficient [50], equation of state [2], fugacity coefficient [2], surface tension [28], melt viscosity [12], and the molecular volume of H$_2$O [1]. For a given simulation we integrated equations (7), (10), (11) for \( k = 1 \) through 3, as well as equations (23) and (25) using the ode15s function of MATLAB®. The boundary condition at the initial depth, \( z_0 \), are

\[ p_m(z_0) = \rho_i \gamma g z_0, \]
\[ C_m(z_0) = C_{eq}(p_m(z_0)), \]
\[ \mu(z_0) = N_0 R_0^2, \]

Here \( \rho_i \approx 2400 \text{ kg/m}^3 \) is the rock density, \( C_{eq} \) are the equilibrium water concentration, and \( N_0 \) and \( R_0 \) are the number density and radius of pre-existing bubbles. The boundary conditions at the surface are

\[ p_m(z = 0) = p_{atm}, \]

or

\[ M(z = 0) = 1. \]

We assume magma fragments at a critical porosity, \( \phi_c \), for each simulation run, we vary \( \phi_c \) to meet the boundary condition at the surface. We estimate bubble size distribution for each simulation by post-processing the simulation results. We discretize \( z \) into multiple bins and estimate number of bubbles nucleated at each bin as

\[ N_m(z) = \int_z^{z+d} \frac{d\tilde{r}}{u}dz, \]  
(28)

and the final size of bubbles nucleated at each bin as [38]

\[ L_m(z) = 2 \left( R_0(z) + \int_z^0 G(\tilde{R}) \, dz \right). \]  
(29)
The objective of our model is to find conditions at which bubble nucleation is continuous from nucleation onset until magma fragmentation. Nucleation is driven by supersaturation pressure, $P_{\text{sat}} - P_m$, and is thus controlled by the competition between decomposition rate, $dP_{\text{sat}}/dt$ and $dP_m/dt$. The latter is proportional to the diffusion rate and is estimated from equation (25). To maintain sufficient supersaturation for nucleation, we assume that the decomposition rate at any given depth is greater than diffusion rate, that is

$$\left(\frac{dP_m}{dt}\right)\left(\frac{dP_{\text{sat}}}{dt}\right)^{-1} = \lambda, \quad (30)$$

where $\lambda > 1$ is a constant. This approach requires that $dP_{\text{sat}}/dt > 0$, that is sufficient number of bubbles are nucleated, to be able to estimate decomposition rate. We use equation (30) at minimum bubble number density of 1 mm$^{-3}$. From decomposition rate we estimate conduit cross sectional area, $dA/dz$ from equation (7) analogous to the approach in Mastin and Ghiroso [33]. After finding an initial conduit radius we used the MATLAB® pchip function to smooth the obtained function and assess the sensitivity of model predictions to variations in conduit radius.

5.4 Nucleation time scale in experiments

Bubble nucleation is driven by supersaturation pressure, $\Delta P_{\text{sat}} = P_{\text{sat}} - P$, defined as the difference between pressure at which H$_2$O would be saturated and the sample’s pressure, $P$. $\Delta P_{\text{sat}}$ increases as $P$ decreases. Considering that diffusion is ineffective during decompression [28], $P_{\text{sat}}$ is expected to remain the same as the initial pressure, $P_i$, throughout decompression. $\Delta P_{\text{sat}}$ thus reaches the maximum potential supersaturation pressure when sample pressure reaches the final pressure, $\Delta P_{\text{sat}} = \Delta P_{\text{sat}}$. Supplementary Figure 1. Consequently, the nucleation rate in the experiments is expected to increase as sample pressure decreases toward $P_i$, because $\Delta P_{\text{sat}} \approx \Delta P$ and nucleation rate scales as $\exp\left(-1/\Delta P_{\text{sat}}^2\right)$ [11]. Nucleation rate reaches a maximum value at $P_i$ and nucleation continues at this rate until the sample is quenched, unless at some point diffusion becomes non-negligible. Thus, the majority of bubbles are expected to nucleate once $\Delta P$ has reached its maximum $\Delta P_{\text{sat}} = P_i - P_t$. If (and once) enough bubbles have nucleated to decrease the characteristic diffusion time to the point where water diffusion into existing bubbles becomes non-negligible, then $\Delta P_{\text{sat}} < \Delta P_{\text{sat}}$ and nucleation rate decreases.

We used the nucleation model described in Yamada et al. [27] to quantify nucleation rate during samples hold time from observed bubble number densities. The model was obtained by analytical solution of bubble nucleation and growth formulations described in Toramaru [10]. Nucleation rate, $J(t)$, as a function of time is given by

$$J(t) = J_i \exp\left(-t/\tau\right)^{1/2}, \quad (31)$$

Here $t$ is time at the final pressure, $J_i$ is the steady nucleation rate and $\tau$ is the nucleation time scale, which is the $\epsilon$-folding time of nucleation. $\tau$ scales inversely with $J_i$ as [27]

$$\tau = k_t \times J_i^{-2/5}. \quad (32)$$

The coefficient $k_t$ depends on melt properties, in particular the diffusion coefficient. Here we assume it is constant because the variability of $k_t$ across the conditions of our experiments is relatively weak [27].

Integration of $J(t)$ through time yields the bubble number density that are nucleated during annealing time as a function of time, $N_{\text{post}}(t)$, given by

$$N_{\text{post}}(t) = \int_0^t J(t)dt' = \Gamma\left(\frac{2}{5}\right) J_i \Gamma\left(\frac{2}{5}, (t/\tau)^{1/2}\right), \quad (33)$$

$N_{\text{post}} = N_0 + N_i$ is related to observed bubble number densities where $N_0$ is the bubble number density at time $t = 0$. In our experiments $N_0$ accounts for bubbles that were nucleated during decompression and is obtained for each suite from $N_m$ in samples with $t_{\text{post}} = 0$. Furthermore, $\Gamma(x)$ is the gamma function

$$\Gamma(x) = \int_0^\infty \zeta^{x-1}e^{-\zeta}d\zeta, \quad (34)$$

and $\Gamma(s, x)$ is the normalized lower incomplete gamma function,

$$\Gamma(s, x) = \frac{1}{\Gamma(s)} \int_0^x \zeta^{s-1}e^{-\zeta}d\zeta, \quad (35)$$

Both $\Gamma(x)$ and $\Gamma(s, x)$ arise out of the integration of $J(t)$ (Equation 31).

At the nucleation time scale, $t \gg \tau$, nucleation rate decreases and bubble number density eventually reaches the equilibrium value of

$$N_{\text{eq}} = N_{\text{post}}(t \rightarrow \infty) = \Gamma\left(\frac{2}{5}\right) J_i \tau. \quad (36)$$

Lastly, the predicted non-dimensional bubble number density is estimated from equations (33) and (36)

$$\frac{N_{\text{post}}(t)}{N_{\text{eq}}} = \Gamma\left(\frac{2}{5}, (t/\tau)^{1/2}\right). \quad (37)$$

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AUTHOR CONTRIBUTION

S.H. carried out the numerical simulations. J.E.G did the experiments. S.H., H.M.G. and J.E.G. involved in interpretation of the results and preparing the manuscript.

DATA AVAILABILITY

The data used are listed in the references. All equations in the numerical simulation are presented in the Methods.

COMPETING INTERESTS

The authors declare no competing interests.

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Supplementary Table 1: Experimental conditions and results. $P_i$ and $P_f$ are initial and final pressures, $T$ is temperature, $t_h$ is hydration duration, $t_d$ is decompression time, $t_p$ is sample hold time before quench, [H$_2$O] is dissolved water concentration and $N_m$ is bubble number density. $^a$Experiments were discussed in Gonnemann and Gardner [31]. $^b$Experiments were discussed in Hajimirza et al. [28]. $^c$Experiments were discussed in Giachetti et al. [26]. $^d$Experiments were discussed in Gardner et al. [51]. $^e$Outliers. 5 out of 35 decompression experiments are considered as outliers because they have a considerably different $N_m$ comparing to $N_m$ in experiments within the same suite.

| Run | Sarting material | $P_i$ (MPa) | $P_f$ (MPa) | $T$ ($^\circ$C) | $t_h$ (hr) | $t_d$ (s) | $t_p$ (s) | [H$_2$O] (wt%) | $N_m$ ($m^{-3}$) |
|-----|------------------|-------------|-------------|-----------------|------------|-----------|-----------|-----------------|-----------------|
| G-1614$^b$ | G-1594A | 161 | 13 | 875 | - | 10 | 0 | - | 1.0 x 10$^9$ |
| G-1616$^b$, $^e$ | G-1594A | 161 | 11 | 875 | - | 15 | 5 | - | 1.1 x 10$^9$ |
| G-1627$^b$ | G-1607 | 161 | 13 | 875 | - | 16 | 18 | - | 1.1 x 10$^{10}$ |
| G-1628$^b$ | G-1607 | 161 | 11 | 875 | - | 11 | 29 | - | 2.7 x 10$^{10}$ |
| G-1796 | G-1780 | 161 | 11 | 875 | - | 11 | 60 | - | 7.1 x 10$^9$ |
| G-608$^e$ | G-595 | 161 | 13 | 875 | - | 11 | 109 | - | 3.2 x 10$^{10}$ |
| G-1746 | G-1736 | 190 | 60 | 850 | - | 54 | 0 | - | 2.4 x 10$^8$ |
| G-1776 | G-1771 | 190 | 58 | 850 | - | 48 | 9 | - | 9.4 x 10$^8$ |
| W-6 | G-1731 | 190 | 60 | 850 | - | 47 | 10 | - | 9.9 x 10$^8$ |
| G-1794 | G-1731 | 190 | 60 | 850 | - | 44 | 22 | - | 1.5 x 10$^9$ |
| W-7$^e$ | G-1570 | 190 | 60 | 850 | - | 55 | 25 | - | 3.4 x 10$^{10}$ |
| G-1798 | G-1779 | 190 | 59 | 850 | - | 40 | 23 | - | 1.3 x 10$^9$ |
| G-1799 | G-1779 | 190 | 59 | 850 | - | 50 | 30 | - | 2.8 x 10$^9$ |
| G-1793 | G-1778 | 190 | 60 | 850 | - | 47 | 44 | - | 2.5 x 10$^9$ |
| W-8$^e$ | G-1579 | 190 | 60 | 850 | - | 55 | 50 | - | 8 x 10$^9$ |
| G-1747 | G-1736 | 190 | 46 | 850 | - | 3 | 0 | - | 4.8 x 10$^9$ |
| W-9$^e$ | G-1579 | 190 | 46 | 850 | - | 3 | 10 | - | 3 x 10$^9$ |
| G-1777 | G-1771 | 190 | 42.5 | 850 | - | 2 | 15 | - | 4.4 x 10$^{11}$ |
| G-1774 | G-1770 | 190 | 42.5 | 850 | - | 2 | 21 | - | 6.2 x 10$^{11}$ |
| G-1775 | G-1770 | 190 | 42 | 850 | - | 1 | 41 | - | 6.9 x 10$^{11}$ |
| W-10 | G-1731 | 190 | 46 | 850 | - | 3 | 50 | - | 1 x 10$^{12}$ |
| G-1613$^b$ | G-1594B | 201 | 75 | 875 | - | 45 | 0 | - | 1.0 x 10$^8$ |
| G-1620$^b$ | G-1594B | 201 | 74 | 875 | - | 28 | 12 | - | 8.0 x 10$^8$ |
| G-1148$^a$ | G-1594B | 201 | 74 | 875 | - | 35 | 25 | - | 5.9 x 10$^9$ |
| G-1623$^b$ | G-1594B | 201 | 75 | 875 | - | 34 | 29 | - | 1.4 x 10$^9$ |
| G-1632$^b$ | G-1608 | 201 | 75 | 875 | - | 34 | 56 | - | 2.3 x 10$^{10}$ |
| G-1481$^c$ | G-1457 | 201 | 54 | 850 | - | 2 | 15 | - | 1.2 x 10$^{12}$ |
| G-1301$^c$ | G-1483 | 201 | 54.0 | 850 | - | 1 | 31 | - | 2.6 x 10$^{12}$ |
| G-1482$^c$ | G-1457 | 201 | 54 | 850 | - | 1 | 60 | - | 2.6 x 10$^{12}$ |
| G-1483$^b$ | G-1456 | 201 | 54 | 850 | - | 2 | 90 | - | 3.4 x 10$^{12}$ |
| G-1791 | G-1741 | 251 | 134 | 850 | - | 2 | 40 | - | 8.0 x 10$^9$ |
| G-1685$^d$ | G-1680 | 251 | 135 | 850 | - | 2 | 58 | - | 2.1 x 10$^9$ |
| G-1797 | G-1742 | 251 | 133 | 850 | - | 3 | 71 | - | 7.1 x 10$^8$ |
| W-1$^e$ | G-1591 | 251 | 135 | 850 | - | 3 | 87 | - | 2.9 x 10$^{10}$ |
| W-2 | G-1732 | 251 | 135 | 850 | - | 2 | 118 | - | 1.1 x 10$^9$ |
Supplementary Table 2: Nucleation rate and time scale predicted for each experimental suite.

| Experimental suite | $P_i$ (MPa) | $P_f$ (MPa) | $J_z$ ($m^{-3}s^{-1}$) | $\tau_n$ (s) | $N_{m,\text{final}}$ |
|--------------------|-------------|-------------|------------------------|--------------|---------------------|
| 1                  | 200         | 54          | $9.5 \times 10^{10}$   | 33           | $2.8 \times 10^{12}$ |
| 2                  | 190         | 42-46       | $2.5 \times 10^{10}$   | 57           | $1.3 \times 10^{12}$ |
| 3                  | 160         | 10-13       | $3.5 \times 10^{9}$    | 319          | $9.9 \times 10^{10}$ |
| 4                  | 200         | 74-75       | $1.2 \times 10^{9}$    | 482          | $5.1 \times 10^{10}$ |
| 5                  | 190         | 58-60       | $6.4 \times 10^{7}$    | 628          | $3.5 \times 10^{10}$ |
| 6                  | 250         | 132-135     | $1.5 \times 10^{7}$    | 1120         | $1.5 \times 10^{10}$ |
Supplementary Figure 1: Supplementary: Schematic representation of bubble nucleation in during experiments. Samples within a given suite have similar initial and final pressures as well as decompression rates, but are quenched at different times. At the final pressure nucleation continues while the sample remains supersaturated. As a consequence bubble number density increases proportionally to annealing time.
Supplementary Figure 2: Observed bubble number density in experiments as a function of supersaturation pressure. For each experimental suite, characterized by similar decompression and decompression rate, bubble number density varies considerably, in some suites over more than 2 orders of magnitude.
Supplementary Figure 3: Predicted bubble number densities compared with the observed values. The predicted values are from equation 30 in the main text. The dashed line represents 1:1 line. All predicted values are within one order of magnitude of observed bubble number densities.