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A general model for welding of ash particles in volcanic systems validated using *in situ* X-ray tomography

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ABSTRACT

Welding occurs during transport and deposition of volcanic particles in diverse settings, including pyroclastic density currents, volcanic conduits, and jet engines. Welding rate influences hazard-relevant processes, and is sensitive to water concentration in the melt. We characterize welding of fragments of crystal-free, water-supersaturated rhyolitic glass at high temperature using *in-situ* synchrotron-source X-ray tomography. Continuous measurement of evolving porosity and pore-space geometry reveals that porosity decays to a percolation threshold of 1–3 vol.%, at which bubbles become isolated and welding ceases. We develop a new mathematical model for this process that combines sintering and water diffusion, which fits experimental data without requiring empirically-adjusted parameters. A key advance is that the model is valid for systems in which welding is driven by confining pressure, surface tension, or a combination of the two. We use the model to constrain welding timescales in a wide range of volcanic settings. We find that volcanic systems span the regime divide between capillary welding in which surface tension is important, and pressure welding in which confining pressure is important. Our model predicts that welding timescales in nature span seconds to years and that this is dominantly dependent on the particle viscosity or the evolution of this viscosity during particle degassing. We provide user-friendly tools, written in Python™ and in Excel®, to solve for the evolution of porosity and dissolved water concentration during welding for user-defined initial conditions.

1. Introduction

Magma fragments into particles during explosive volcanic activity. Subsequent welding of these particles can occur at the base of hot pyroclastic density currents (Walker, 1983; Branney et al., 1992), at the walls of volcanic conduits (Gonnermann and Manga, 2003; Rust et al., 2004; Gardner et al., 2017), in tuffisite veins (Tuffen et al., 2003; Kendrick et al., 2016; Gardner et al., 2018), in the hot zone of jet engines (Giehl et al., 2016), and when lightning strikes volcanic ash in the air or on the ground (Cimarelli et al., 2017; Mueller et al., 2018). Despite this wide range of welding scenarios, there has been little work on the physics of welding of volcanic droplets, beyond simple empirical, semi-empirical, or scaling approaches (Friedman et al., 1963; Riehle, 1973; Sparks et al., 1999; Quane and Russell, 2005a; Russell and Quane, 2005; Vasseur et al., 2013; Wadsworth et al., 2014).

Welding involves a reduction of inter-particle pore space (Branney and Kokelaar, 1992; Sparks et al., 1999; Quane and Russell, 2005a; Vasseur et al., 2013). Porosity is therefore a convenient

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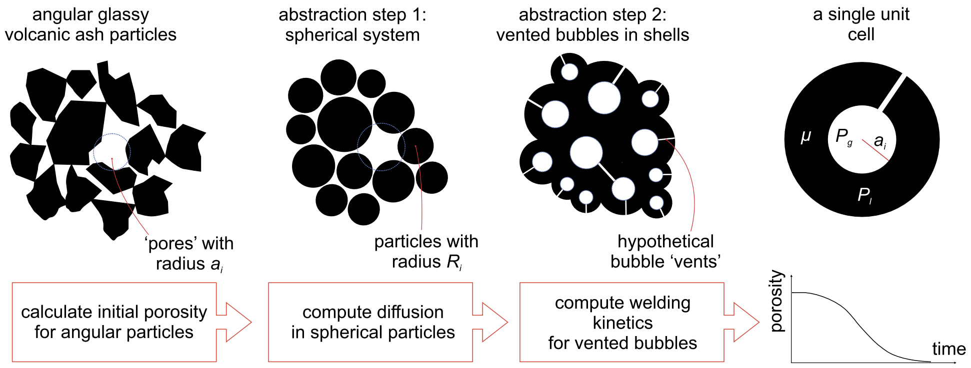

Fig. 1. The development of a diffusion–welding model. The natural system is composed of angular volcanic particles, which are abstracted to a pack of spherical particles with the same initial porosity. The diffusion model is used (Eq. (4)) to compute the average water content as a function of time $\langle C\rangle(R_i,t)$, which is converted to an average viscosity $\langle\mu\rangle$. Then we make a further abstraction to vented bubble geometry and the welding is computed in terms of an evolution of the total porosity with time $\phi(t)$ using Eqs. (1)–(2), accounting for the polydispersivity of the initial particle size distribution (Wadsworth et al., 2017b).

metric for tracking the degree of welding, and has been used to rank the 'grade' of a welded deposit (Quane and Russell, 2005b; Wright and Cashman, 2014). Theoretical models for the evolution of porosity as a function of time in a welding system have been proposed (Frenkel, 1945; Mackenzie and Shuttleworth, 1949) but they do not account for the complexities of welding in magmatic systems, which include non-isothermal behaviour, disequilibrium of dissolved volatile species, and the effect of a confining pressure that pushes the particles together. Non-isothermal behaviour is important because welding in nature may occur as the particles cool (e.g. at conduit margins, within ejected ballistic bombs, or in ignimbrites) or follow more complex heating and cooling pathways (e.g. in a jet engine). Disequilibrium of volatile species – particularly of water – is important because the solubility changes as the pressure and temperature environment of the particles changes, driving diffusion in or out of the particles during welding (Sparks et al., 1999; Gardner et al., 2018). In the case of water, this has a strong impact on the viscosity of the particle (Hess and Dingwell, 1996) affecting welding rate (Grunder et al., 2005; Gardner et al., 2018, 2019). Confining (or lithostatic) pressure resulting, for example, from the weight of aggrading particles at the base of a pyroclastic density current, is important because it provides a stress that pushes the droplets together, accelerating welding. Previous theoretical and quantitative models for welding have focused on cases where welding is driven by surface tension alone (Wadsworth et al., 2016) or, where pressure is considered, have relied on scaling arguments (Sparks et al., 1999) or purely empirical correlations (e.g. Riehle, 1973).

We develop a general and versatile mathematical framework for welding that can be used to predict the textural evolution of a welding pack of particles in a wide range of natural settings. We perform and analyze experiments conducted under non-isothermal, disequilibrium conditions to validate the model.

2. A theoretical model for droplet welding dynamics

2.1. Viscous welding under arbitrary pressure

Previous work on non-volcanic welding (or 'sintering') of spherical particles has shown that in the viscous state (i.e. when particles are droplets), surface-tension-driven welding is well described by a 'vented bubble model', in which the inter-droplet porosity is abstracted as a system of spherical bubbles in liquid shells, which are 'vented' so that the gas can escape as the bubbles shrink (Mackenzie and Shuttleworth, 1949; Wadsworth et al., 2016). The geometric assumptions of the vented bubble model are most valid for highly polydisperse particle distributions (Wadsworth et al., 2017b) of the sort typical in nature, and the approximation becomes increasingly accurate as welding progresses because, as the droplets coalesce, the microstructural geometry continuously diverges from 'droplet-like' towards 'bubble-like' – that is, there is a topological inversion of the pore space (Wadsworth et al., 2017a). We start from the assumption that the vented bubble model also applies to initially-angular particles. The conceptual steps in the geometric abstraction from an ash pack to a system of vented bubbles are shown in Fig. 1.

We extend the vented bubble model to include a confining pressure that acts alongside surface tension stress to drive welding, where we use the term *confining* pressure to refer to an isotropic pressure acting to push the particles together – equivalently, when viewing the particles as viscous droplets, an isotropic pressure in the continuous liquid phase of the coalescing droplets. In their supplementary material, Wadsworth et al. (2016) derive the vented bubble model from the model of Prousevitch et al. (1993) for bubble growth in magma by setting the bubble pressure inside the associated liquid shell to be equal to the gas pressure outside the shell at all times. Here, we relax that assumption, and instead set the pressure difference to a value ΔP. Neglecting inertia, the full equation for the inter-droplet porosity ϕ with time t is then

$$\frac{d\phi}{dt} = -\frac{3\Delta P}{4\mu}\phi - \frac{3\Gamma}{2\mu a_i}\left(\frac{\phi_i}{1-\phi_i}\right)^{1/3}\phi^{2/3}(1-\phi)^{1/3}, \tag{1}$$

where ΔP is the difference between the confining pressure on the liquid droplets P_l and the pressure of the interstitial gas P_g, such that $\Delta P = P_l - P_g$, μ is the particle viscosity, Γ is the interfacial tension between the particles and the gas, a_i is the initial size of the bubble, and ϕ_i is the initial porosity when welding starts. A derivation of Eq. (1) from the Rayleigh-Plesset equation is given in the Supplementary Information.

Eq. (1) can be cast in dimensionless form by normalizing time to a characteristic capillary timescale $\lambda = \mu a_i/\Gamma$, such that $\bar{t} = t/\lambda$, normalizing pressure to a capillary pressure scale $P_c = 2\Gamma/a_i$, such that $\overline{P} = \Delta P/P_c$, and normalizing porosity to its initial value, such that $\bar{\phi} = \phi/\phi_i$, yielding

$$\frac{d\bar{\phi}}{d\bar{t}} = -\frac{3}{2}\left[\overline{P}\,\bar{\phi} + \left(\frac{1-\phi_i\bar{\phi}}{1-\phi_i}\right)^{1/3}\bar{\phi}^{2/3}\right], \tag{2}$$

where a bar above a parameter denotes that it has been rendered dimensionless. The first term within the square brackets represents the contribution of the confining pressure, the second term represents the contribution of the capillary (Laplace) pressure.

The dimensionless time $\bar{t}$ can be generalized to account for non-isothermal temperature–time history, which is especially useful for natural magmatic scenarios. This is achieved by accounting for the change in viscosity μ as temperature varies, via

$$\bar{t} = \frac{t}{\lambda} = \frac{\Gamma}{a_i}\int_{t_i}^{t}\frac{1}{\mu}dt \tag{3}$$

where t_i is the time at which the welding process starts. In casting Eq. (3) this way, we assume that Γ is a constant, independent of time during welding. In reality, Γ is dependent on both water concentration and temperature, however, variations in surface tension are negligible compared with variations in liquid viscosity arising from the effects of both temperature and water concentration (discussed later). Together, Eqs. (2) & (3) represent a universal description of isotropic, viscous particle welding, derived from micromechanical first principles. In the case where $\overline{P} = 0$, this approach has been validated against experimental data across a large range of temperatures (Wadsworth et al., 2016). While other models exist, they are either less easy to use, requiring a switchpoint between two competing processes (Prado et al., 2001), or they rely on bulk properties of the system, which have to be empirically determined and are therefore less general as they are not constructed from the micromechanics involved (Olevsky, 1998; Quane and Russell, 2005a).

The parameter a_i can be difficult to measure, or even define, for what is a complex, interconnected pore network (Fig. 1). We use a relationship between a_i and the distribution of particle sizes $F(R)$ in a pack of particles or droplets (Lu and Torquato, 1992), which is described in detail elsewhere (Wadsworth et al., 2016, 2017b). The relationship relies on knowledge of ϕ_i, and the moments of the distribution of R, denoted $\langle R^n\rangle$, which can be grouped into a polydispersivity factor $S = \langle R\rangle\langle R^2\rangle/\langle R^3\rangle$. The output from this is a pore size distribution $f(a_i)$ that relates to the particle size distribution $F(R)$ and ϕ_i, and is described in the Supplementary Information along with an account of how this is used in conjunction with Eqs. (1)–(2) using convolution techniques (Wadsworth et al., 2017b).

2.2. *Accounting for diffusion of volatiles during welding*

In the model formulation above, the viscosity μ is assumed to be dependent on temperature, T, only. However, viscosity also depends on the concentration of water dissolved in the melt, which, in nature, may vary during welding. Volcanic particles formed at fragmentation may be super-saturated in dissolved water (Giachetti and Gonnermann, 2013), and that super-saturation can grow as the particles ascend rapidly to lower pressures upconduit without time to fully re-equilibrate (Gardner et al., 2017). Similarly, the solubility of water increases as the particles cool, which may cause them to re-hydrate (McIntosh et al., 2014; Ryan et al., 2015). Mass diffusion of water in or out of particles may occur on timescales similar to the timescale of welding (Sparks et al., 1999; Gardner et al., 2017, 2018, 2019); consequently, we anticipate that diffusion of water can affect the rate of welding through its impact on melt viscosity.

In order to account for diffusion, we must define the geometry of the internal welding system. The internal geometry of welding systems is complex and evolves from an initial state of particles in a gas continuum, to bubbles isolated in a liquid continuum. A rigorous solution for mass diffusion through this evolving geometry would require an approach that explicitly resolves both the fluid motion and the diffusion. However, as for the welding model, we simplify the problem by abstracting the geometry. For the purposes of diffusion modelling we assume that the particles remain spherical and simply apply Fick's 2nd law in spherical coordinates,

$$\frac{\partial C}{\partial t} = \frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2 D\frac{\partial C}{\partial r}\right), \tag{4}$$

where C is the concentration of water in the melt particle, D is its diffusivity (which depends on temperature and local water concentration), and r is the radial position from the particle centre. We adopt the assumption that the initial concentration of dissolved water C_i is uniform throughout the particle at the onset of welding, giving the initial condition $C = C_i$ for all r at $t = 0$. At all later times the water concentration at the surface of the particle is given by the equilibrium solubility C_e at the current conditions of gas pressure P_g and temperature T, giving the boundary condition $C = C_e(t)$ at $r = R$ for $t > 0$. We define a zero-flux boundary condition at the centre of the particle: $\partial C/\partial r = 0$ at $r = 0$.

To account for the effect of variable water concentration in the particle we determine a spatial average by integrating C over $0 < r < R$; this integral is $\langle C\rangle = \int_0^1 C\, d\bar{r}$, where $\bar{r} = r/R$. We then use $\langle C\rangle$ to compute an average viscosity $\langle\mu\rangle$ which is used in Eqs. (1)–(3) in place of μ. This approach results in an effective coupling between the diffusion model (Eq. (4)) and the welding model (Eqs. (1)–(3)). In the Supplementary Information, we describe the numerical solution of these equations in detail. We note here that our assumption of spherical particles undergoing diffusion of volatiles may be invalid at large polydispersivity for cases where the smallest particles are in equilibrium while the largest particles are far from equilibrium. This is discussed elsewhere (Gardner et al., 2019).

3. Experimental validation: materials and methods

Our starting material is a metaluminous, tholeiitic natural rhyolitic glass collected from Hrafntinnuhryggur, Krafla (Iceland). We crushed the obsidian to a powder using an agate mortar and pestle and then crushed the resultant chips to a fine powder using a concussion ball mill for short durations to prevent the sample heating significantly. The powder was sieved to <125 μm diameter pieces and the size distribution was measured using a Beckman Coulter LS™ 230 laser refraction particle size analyzer with the measuring range 0.375–1000 μm diameter. The particle size distribution is given in the Supplementary Information, and has a mean radius $\langle R\rangle = 2.2 \times 10^{-5}$ m.

Using a Netzsch Pegasus 404c device for simultaneous thermal analysis, we determined the dissolved volatile concentration that is excess (above solubility) at up to 1325 K to be $C_i = 0.11 \pm 0.02$ wt.%, by the relative loss of mass during heating, consistent with Tuffen and Castro (2009). This determination was performed on single chips ($n = 6$) from within a few millimetres of the subsample of the glass block that was used throughout this study.

To solve the governing equations given in our model, we require a parameterization for D, μ, and C_e relevant to the material in question. We use models relevant to the metaluminous rhyolites (Hess and Dingwell, 1996; Liu et al., 2005; Zhang and Ni, 2010)

$$\begin{aligned}
\log_{10}(\mu) &= -3.545 + 0.833\ln(C) + \frac{9601 - 2368\ln(C)}{T - 195.7 - 32.25\ln(C)} \\
D &= C\exp\left[-18.1 + 0.001888P_l - \left(\frac{9699 + 3.626P_l}{T}\right)\right] \\
C_e &= \frac{354.941P_w^{0.5} + 9.623P_w - 1.5223P_w^{1.5}}{T} + 0.0012439P_w^{1.5}
\end{aligned} \tag{5}$$

where P_w is the partial pressure of water in the interstitial gas phase, and the coefficients given are valid when T is in K, P_l and P_w are in MPa, and C and C_e are in wt.%. In the Supplementary Information we independently verify $\mu(T)$ for our material using a parallel plate compression method (Hess et al., 2007), a calorimetric method using a shift factor of 10.4 (Gottsmann et al., 2002), and a micropenetration method (Hess et al., 1995), which all demonstrate internal consistency, as well as matching the prediction of Eq. (5) for the measured C_i. We take a value $P_w = P_l\alpha$

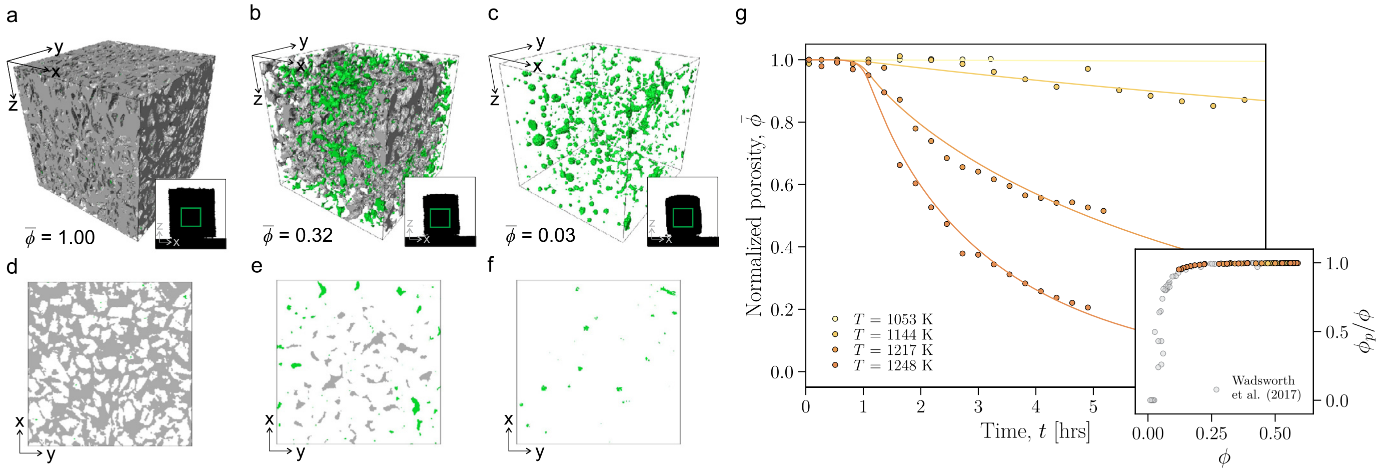

Fig. 2. Constraints of time-dependent welding collected *in situ* using either optical dilatometry or X-ray tomography. (**a**-**c**) 3-dimensional rendered images of the time-dependent welding process segmented on the basis of grayscale gradient filtering from continuous, time-resolved, *in situ* X-ray tomography data. The particle phase is rendered transparent, and the pore phase is divided into a grey and a green component depending on whether it is connected across the sample in any direction (grey) or is isolated from connections (green). Box edge lengths of the sub-volume displayed are 350 μm Displayed is one representative experiment performed at $T = 1350$ K, for which the dimensionless porosity $\bar{\phi}$ is labelled. Inset in each panel is the 2-dimensional side-view of an initially cylindrical sample from an experiment at the same conditions performed in the optical dilatometer (image base length of 5 mm). (**d**-**f**) A 2-dimensional horizontal slice through each of the 3-dimensional rendered images in (**a**-**c**) taken at the midpoint of the z-axis in the sample. (**g**) The porosity as a function of time of the obsidian particles sintered *in situ* using time-resolved X-ray tomography at a range of temperatures (labelled). The curves represent the solutions to Eqs. (1) or (2) with $\overline{P} = 0$ and computing the time-dependent diffusion of volatiles out of the particles (Eq. (4)). *Inset:* the connectivity of the pore phase with porosity showing the collapse from fully connected to isolated as $\phi \to \phi_c$ during welding. Data are compared with welding in synthetic glass systems (Wadsworth et al., 2017a). (For interpretation of the colours in the figure(s), the reader is referred to the web version of this article.)

with $\alpha = 0.2$, representing the typical humidity pressure in a laboratory furnace. Finally, we use $\Gamma = 0.3$ N m^{-1}.

We performed two sets of *in situ* high temperature experiments, which used different methods to image the evolution of a welding pack of obsidian powder, lightly pressed into a free-standing cylinder with 3 mm diameter: (1) synchrotron-source X-ray tomography, providing continuous 3-dimensional microstructural data; and (2) optical dilatometry, providing bulk sample volume changes only. The first set of experiments were performed at the TOMCAT beamline of the Swiss Light Source at the Paul Scherer Institute. Cylinders of obsidian powder were loaded into the imaging window of the X-ray beam path. We used a laser system (Fife et al., 2012) to heat an alumina sleeve (muffle) placed over the samples, thus heating the obsidian pack indirectly. The temperatures measured by a pyrometer were calibrated by comparing the *in situ* welding of a well-studied sample of monodisperse glass beads with *ex situ* characterization of the same process (Wadsworth et al., 2016), resulting in a continuous correction for T, and confirming that temperature gradients on the sample scale were negligible. Full 3-dimensional tomographs were collected at 5.5×10^{-3} Hz with a spatial resolution of 1.6 μm. The second set of experiments were performed using a Hesse Instruments EM-201 optical dilatometer, which continuously records the silhouette of the sample during heating at 1 Hz. Volume is determined from the silhouette as the solid of revolution.

The experiments covered a wide range of conditions, including isothermal experiments at temperatures of 1050–1500 K, and linear heating ramps at rates of 0.04-0.25 K s^{-1}. The data from optical dilatometry are confined to porosity $\phi(t)$, while the tomography provides 3-dimensional data that are used to measure both the total porosity $\phi(t)$, and the porosity that is connected across the sample (or segmented domain) $\phi_p(t)$. After the heating experiments, we repeated the thermal analysis step on a few sub-samples, and no mass loss was observed, demonstrating the samples did equilibrate volatiles during the *in situ* experiments.

Supplementing our datasets collected at relatively low C_i and a small difference between C_i and C_e (small initial supersaturation), we re-analyse the data from Gardner et al. (2018) and Gardner et al. (2019). These data were collected at high P_g such that the equilibrium water concentration is also high. These data also include particles that hydrate and particles that de-hydrate while also welding.

All of the above experiments were conducted without confining pressure, hence $\overline{P} \approx 0$, a situation typical of small-scale laboratory settings. In order to examine the effect of $\overline{P} > 0$, we use data for welding Pyrex™ glass presented previously (Friedman et al., 1963). In those experiments the glass was crushed to a particle size around 100–250 μm (not specified exactly), and welded under a uniaxial liquid pressure of $P_l = 1.52 \times 10^6$ Pa and $P_l = 3.63 \times 10^6$ Pa at temperatures 883–943 K. We re-analyze these data using our model. While the same authors provide data for rhyolite particle welding (Friedman et al., 1963), some doubt exists as to the exact pressures used (Sparks et al., 1999) so we choose not to reanalyze those data.

4. Results, data analysis and model validation

In situ tomography allows us to render the evolving internal pore space of the samples in 3-dimensions through the welding process. Fig. 2 shows a typical experimental result. The pore space is initially fully interconnected, and has a complex geometry between the angular glass fragments. As we heat the sample, the glass particles relax to liquid droplets at high temperature, and the droplet–droplet contacts weld. The porosity decreases with time smoothly and monotonically, and the rate at which it decreases depends strongly on temperature (in the isothermal experiments) or heating rate (in the non-isothermal experiments). The connectivity of the pore space drops during welding from fully connected at the start ($\phi_p/\phi = 1$), to fully closed at volume equilibrium ($\phi_p/\phi = 0$). The porosity at which the connectivity drops to zero is the percolation threshold porosity ϕ_c below which the system is impermeable. We determine this from our experiments as $\phi_c = 0.02 \pm 0.019$, which agrees with theory, simulations (Elam et al., 1984; Vasseur and Wadsworth, 2017), and experiments (Wadsworth et al., 2016) in other welding droplet or overlapping sphere systems. This value is far lower than the percolation threshold for bubbly systems, as has previously been noted for the internal geometry of welding systems (Vasseur and Wadsworth, 2017).

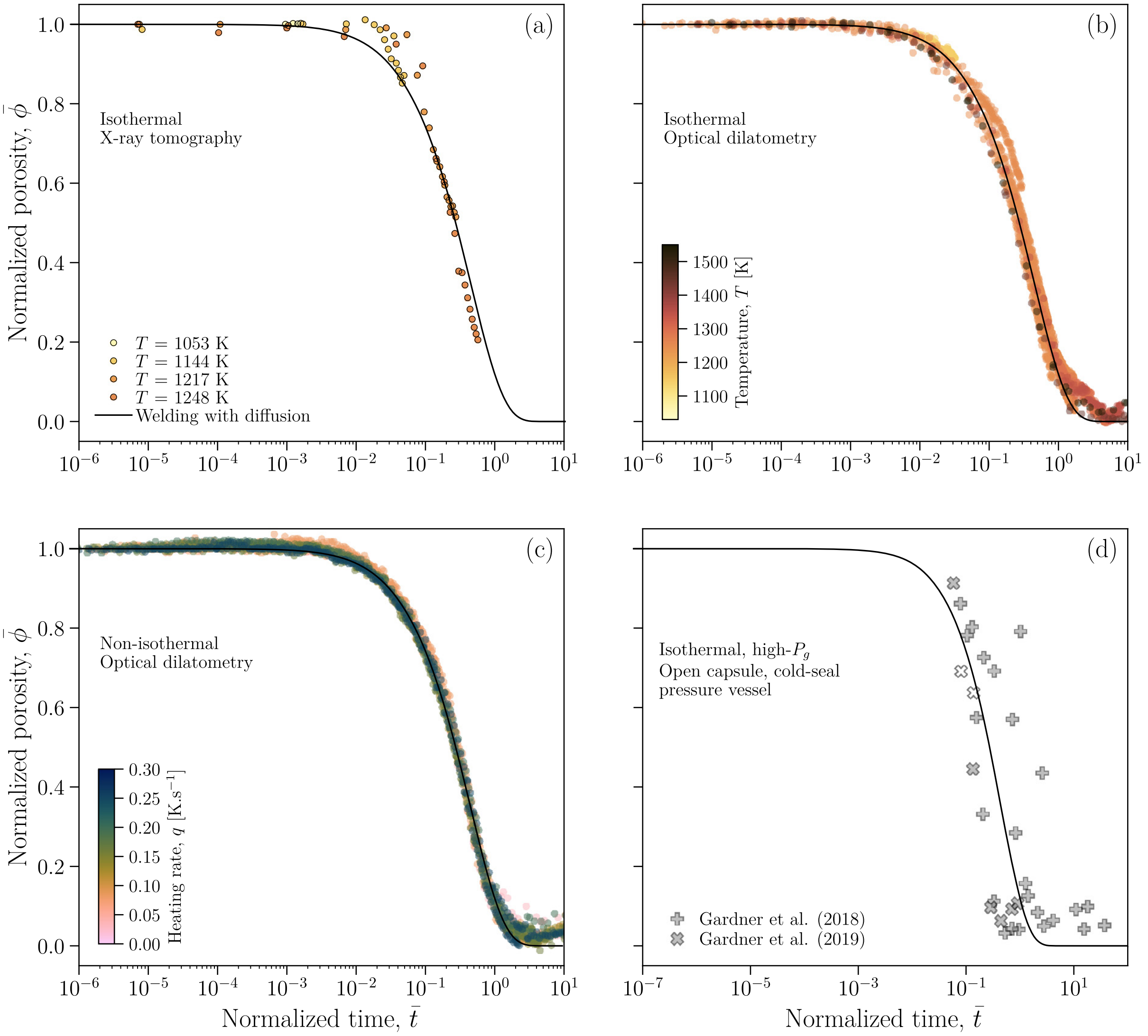

Fig. 3. Modelling the evolution of the pore phase between the welding droplets. (**a**) The porosity $\overline{\phi}$ as a function of time for each experimental T for the X-ray tomography experiments only. These data are compared with the results of Eq. (2) (welding) with Eq. (4) (diffusion), which account for syn-welding degassing of the droplets and which require no fitting parameters. These data are the same as those presented in Fig. 2g. (**b**) The same as (**a**) but for the data collected using optical dilatometry (i.e. without 3-dimensional microstructural information) showing that this technique can be used to capture the bulk decay of porosity with time. (**c**) The same dimensionless plot as in (**b**) but for non-isothermal experiments at different experimental heating rates, showing that regardless of the $T(t)$ path taken by the samples, the efficacy of our model (Eqs. (1)–(4)) is robust. In all panels, the dimensionless time is given by Eq. (3). (**d**) The data from Gardner et al. (2018) and Gardner et al. (2019) collected using a high-P_g cold seal vessel, re-analysed using the diffusion-welding model given here. The filled points represent data for which rhyolite particles are hydrating (from $C_i = 0.15$ wt.% to equilibrium conditions at high P_g and high T) while welding, and the un-filled points represent data for which rhyolite particles are de-hydrating (from $C_i = 2.3$ wt.% to equilibrium conditions at high P_g and high T). See the papers originating the data for more information.

In Figs. 2g and 3, we compare the results for the *in situ* X-ray tomography and optical dilatometry experiments with the model presented in section 2. For these unconfined laboratory-scale tests, the value of $\overline{P}$ is effectively 0. For the isothermal experiments we solve Eq. (2) with $\overline{P} = 0$, accounting for the diffusion of water out of the particles during welding via Eqs. (3) and (4) (Figs. 3a and 3b). For the non-isothermal experiments we additionally account for temperature change via Eq. (3) (Fig. 3c). In both cases, we find good agreement and a reasonable collapse of the data to the model. This result highlights that, in these experiments, welding rates are influenced by temperature and volatile content, both of which control the particle viscosity and can evolve on the same timescale as the welding; hence, they must be solved explicitly (Eqs. (3) and (4)). Welding rate also depends on particle size distribution, interfacial tension, and the initial porosity of the packed particles. Particle angularity may subtly affect both the diffusion rate and the sintering rate when compared with the model, which is based on idealised spherical particles, but, given the good agreement between model and data, this effect does not appear to be of first-order.

The experimental validation of our simple model for $\overline{P} = 0$ appears to be successful. To extend this to conditions where $\overline{P} > 0$, we re-analyse the results from Friedman et al. (1963) in which anhydrous glass was heated under pressure. In the Supplementary Information we give a detailed description of the methods used in Friedman et al. (1963), but note here that Eqs. (1) and (2) are valid in their experiments. We assume that the uniaxial nature of their applied loads can be accounted for using the Trouton ratio, such that our model for isotropic pressurization can be adapted to uniaxial conditions. For their anhydrous experiments, conducted at $\overline{P} = 100$, we apply Eq. (2) directly. We find good agreement across a wide range of temperature (Fig. 4), validating our model up to naturally relevant pressures and across the regime boundary $\overline{P} = 1$. These conditions represent the state where the volcanic

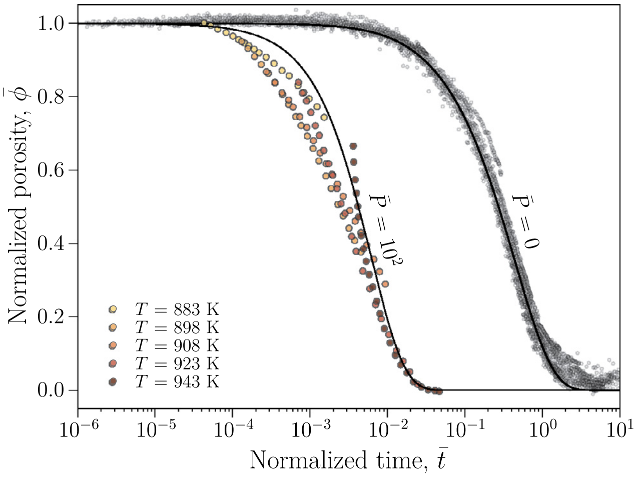

Fig. 4. The effect of pressure on the welding of glassy particles. Shown here are data from Friedman et al. (1963), in which synthetic glass particles (Pyrex™) are welded under a pressure equivalent to $\overline{P} = 100$, and at a range of temperatures (labelled). We use the values of μ, R_i and P_l given in their work, and the curve represents the solution to Eq. (2) without fitting parameters. Shown for reference is the solution for $\overline{P} = 0$ and all data from Fig. 3 given in grey. In all cases, the dimensionless time is given by Eq. (3).

particles are under pressure, but the interconnected gas phase between the particles remain un-pressurized, which is a typical scenario for larger systems in nature. We show that, in this case, the time required for welding to complete is reduced by the elevated confining pressure, as implied by Eq. (2) (note how the data collapse to a model curve to the left of the $\overline{P} = 0$ curve in Fig. 4, and are therefore welding more rapidly).

5. Discussion

5.1. Validity of the welding model

The welding model agrees well with experimental data across a wide range of conditions, validating the model for application to welding systems: 1) with and without applied pressure; 2) under isothermal and non-isothermal conditions; 3) in which dissolved water is in equilibrium or disequilibrium with ambient pressure and temperature conditions. Thus the model is sufficiently general to capture most of the essential features of welding scenarios in magmatic and volcanic systems. There are, however, two limitations. Firstly, the model does not apply directly to welding under high shearing stress. We do use experimental data in which welding particles are under uniaxial compression (Friedman et al., 1963), such that the anisotropy of the pressure applied results in shear stresses internal to the sample (cf. uniaxial experiments in Quane and Russell, 2005a; Heap et al., 2015), which we account for via the Trouton ratio. This gives us some confidence that, under minor local shearing within a system that is loaded anisotropically, our model is valid. Nonetheless, we note that validation for shearing systems requires future systematic study over a larger range of better constrained shear stress. This limitation means that our model does not, for example, explicitly predict the formation of fiammé in welded ignimbrites. Secondly, the model assumes that interstitial gas escapes freely from the welding system, and does not apply when gas escape is significantly hindered by the permeability of the connected pore network. We can determine the conditions under which this second limitation is important.

The characteristic lengthscale beyond which a viscous system is permeability-limited is the compaction length $L_c = (k_r\mu/\mu_g)^{1/2}$ (Michaut et al., 2009; Kennedy et al., 2016), where k_r is a reference permeability and μ_g is the gas viscosity. If we normalize our system length by L_c we have

$$\overline{L} = \frac{L}{L_c} \approx L\sqrt{\frac{\mu_g}{k_r\mu}}. \tag{6}$$

If $\overline{L} \gg 1$, permeability is a rate-limiting parameter, with the consequence that P_g may rise in parts of the system and affect the welding rate (because the welding rate depends is sensitive to $P_l - P_g$); this regime is termed *compaction welding*. If $\overline{L} \ll 1$, gas escape can occur more rapidly than welding occurs and the process is not hindered by sluggish gas escape. The analysis presented via Eqs. (1)–(4) is therefore valid in the regime $\overline{L} \ll 1$.

5.2. Welding regimes in nature

The dimensionless length $\overline{L}$ discriminates between regimes in which welding is or is not limited by permeable outgassing of the interstitial gas phase. For the non-limited regime ($\overline{L} \ll 1$) in which our welding model is valid, we can also discriminate between regimes in which welding is dominated by confining pressure (*pressure welding*), or by capillary pressure arising from the surface tension (*capillary welding*). From Eq. (2), we see that the confining pressure term dominates the capillary pressure term when

$$\begin{aligned} &\overline{P} \gg \left(\frac{1-\phi_i\overline{\phi}}{\overline{\phi}-\overline{\phi}\phi_i}\right)^{1/3} \quad \text{pressure welding,} \\ &\overline{P} \ll \left(\frac{1-\phi_i\overline{\phi}}{\overline{\phi}-\overline{\phi}\phi_i}\right)^{1/3} \quad \text{capillary welding.} \end{aligned} \tag{7}$$

The dependence on the porosity (via $\overline{\phi}$) arises because capillary stress always tends towards infinity as bubble radius tends towards zero, such that a system that starts in the pressure welding regime may end in the capillary regime with no change in the ambient conditions. The value of the terms on the right hand side of Eq. (7) is equal to unity at the start of welding so, in practice we use $\overline{P} \gg 1$ and $\overline{P} \ll 1$ to discriminate between pressure and capillary welding regimes.

Given these constraints ($\overline{P}$ and $\overline{L}$), we can assess the regimes covered by some typical volcanic welding scenarios. For any situation in which welding might occur, we therefore need to know μ, R_i, L, and ΔP (for simplicity, we take a constant $\Gamma = 0.3$ N m^{-1} (Wadsworth et al., 2016), $\mu_g \approx 10^{-5}$ Pa s, and $k_r = k_i \approx 7 \times 10^{-12}$ m^2 for packed particles (Wadsworth et al., 2017a)). For these dimensional considerations we assume that $a_i = R_i$, because there is usually insufficient information provided to compute a_i explicitly, and we justify this by noting that these values are typically of the same order of magnitude (Wadsworth et al., 2016). In Table 1 we give a compilation of estimated values for these parameters gathered from well-studied welded ignimbrites, tuffisites, welded jet engine deposits, and welded obsidian pyroclasts. In each case, these parameters are converted to a quantitative range of $\overline{P}$ and $\overline{L}$ that represents the initial conditions for that particular system, and plotted in Fig. 5. In the case of tuffisites from Volcán Colima, we use the values of particle (droplet) viscosity from Kendrick et al. (2016) which incorporate the effect of crystallinity.

We find that very few systems are in the $\overline{L} \gg 1$ (permeability limited) regime. Exceptions would include welding in particularly large welded ignimbrite systems if the emplacement mode is *en masse* (we give the result for the $L \leq 400$ m Bad Step Tuff, for which $\overline{L}$ can exceed unity). However, under the assumption of the progressive-aggradation model for the sedimentation of ignimbrites (Branney and Kokelaar, 1992) and an estimated, rising $L \approx 2$ m thick welding window (Andrews and Branney, 2011), we find that $\overline{L} \ll 1$ is more typical for ignimbrite emplacement. The *en*

Table 1
Constraints underpinning $\overline{P}$ and $\overline{L}$ from natural deposits or scenarios.

	Particle viscosity[a] μ (Pa s)	System length L (m)	Particle radius[f] R_i (m)	Liquid pressure P_l (Pa)	Gas pressure P_g (Pa)	Initial water content[e] C_i (wt.%)	Emplacement temperature[e] T (K)	References
Welded ignimbrites								
Bad Step Tuff	10^7-10^8	40-400[b]	10^{-5}-10^{-3}	10^6-10^7	10^5-10^6	0.1-0.2	1273	Branney et al. (1992)
TL	10^5-10^6	10.4-28[b]	10^{-5}-10^{-3}	3.1-7.5 × 10^5	10^5-10^6	0.47-0.87	1084-1183	Sumner and Branney (2002)
Grey's Landing	10^7-10^9	2-70[b]	10^{-5}-10^{-3}	10^5-10^6	10^5-10^6	0.1-0.2	1198-1298	Andrews and Branney (2011)
Rattlesnake tuff	10^8-10^{10}	15-70[b]	10^{-5}-10^{-3}	4.1 × 10^5-10^6	10^5-10^6	0.1-0.2	1073-1153	Streck and Grunder (1995)
Tuffisites								
Chaitén (2008)	10^7-10^9	0.005-0.03	1.25×10^{-4} -2.5×10^{-4}	10^6-10^7	4.6×10^6 -8.1×10^6	0.44-1.2	1023-1098	Castro et al. (2012), Saubin et al. (2016)
Cordon Caulle 2011-2013	10^8-10^9	0.005-0.03	10^{-6}-10^{-3}	10^6-10^7	4.6×10^6 -8.1×10^6	0.16-0.25	1168	Castro et al. (2014)
Colima	10^{10}-10^{11}	0.001-0.05	10^{-4}-10^{-3}	10^5-10^6	10^5	0.1-0.2	1213-1253	Kendrick et al. (2016)
Törfajökull	10^9-10^{14}	0.001-0.05	10^{-5}-10^{-3}	10^6-10^7	4.6×10^6 -8.1×10^6			Tuffen and Dingwell (2005)
Obsidian pyroclasts								
Mono craters	10^6-10^{12}	0.01-0.02	2×10^{-5} -1.7×10^{-4}	1.002×10^5	10^5			Gardner et al. (2017)
Jet engine deposits								
Rhyolitic experimental	10^3-10^9	10^{-3}-10^{-2}	3×10^{-5}	4.0002×10^6	4×10^6	0.1	1148-1848	Giehl et al. (2016)

[a] The viscosity is either taken from the references for each case study or otherwise is calculated using Hess and Dingwell (1996) with the C_i and T given for most cases (exceptions are the TL ignimbrite and the basaltic example for the jet engine deposits, both of which are calculated using Giordano et al. (2008) and the composition given in the references; for TL, we use the WTL trachyte zone composition because this is the 'lava like' facies (Sumner and Branney, 2002)).

[b] This system length is assuming *en masse* deposition, but for progressive aggradation we take 2 m for all ignimbrites.

[c] We take this liquid pressure to be the hydrostatic loading pressure assuming a density of 2300 kg m^{-3}. Except for the tuffisite cases, the gas pressure is added to the liquid pressure.

[d] The upper limit of these gas pressures is given by estimates of dynamic pressures during transport in pyroclastic density currents (Clarke and Voight, 2000).

[e] Note that these parameters are only required if the viscosity is not given directly by the originating authors. The value for C_i is approximated as 0.1-0.2 wt.% if other information is not given.

[f] We make the simplifying assumption that $a_i = R_i$ for this scaling analysis.

masse and progressive aggradation models for ignimbrite emplacement represent upper and lower bounds on $\overline{L}$, respectively.

We also find that volcanic welding scenarios span the $\overline{P} = 1$ divide, implying that there are cases for which pressure welding dominates ($\overline{P} > 1$) and cases for which capillary welding dominates ($0 \leq |\overline{P}| < 1$). Pressure welding appears to be typical of tuffisites and ignimbrites, while capillary welding appears to be typical of the formation of obsidian pyroclasts and of undesirable welding in the combustion chamber of jet engines (Fig. 5). Tuffisites, in particular, are known to have variable and complex pressure–temperature histories, implying that they may track through $\overline{P}$ space during their formation and welding (Tuffen and Dingwell, 2005; Castro et al., 2012; Saubin et al., 2016). If the exact evolution of pressure and temperature were known, then our model could be used to determine the degree of welding throughout.

For each of the cases presented in Fig. 5, we can compute a timescale for the porosity to reach the equilibrium value $\phi = \phi_c$. For systems welding at any $\overline{P}$, for $\overline{L} \ll 1$, this timescale includes contributions from the pressure and capillary components, and can be taken as the reciprocal of the sum of the characteristic welding rates associated with the confining pressure and capillary pressure terms

$$\lambda_W \approx \left(\frac{\Delta P}{\langle \mu \rangle} + \frac{\Gamma}{\langle \mu \rangle \langle R_i \rangle} \right)^{-1}. \tag{8}$$

We use Eq. (8) and the inputs in Table 1 to compute λ_W (in seconds) for each case study example given. We find that most systems weld over a timescale of 1 s to 1 day. Exceptions, which require very long timescales to weld, are crystal-rich tuffisites (Kendrick et al., 2016) or obsidian pyroclasts welding under the lowest temperature and gas pressure conditions expected (Gardner et al., 2017). In both of those slow-welding scenarios, it is unlikely that welding will complete before other processes, such as cooling of the particles or deposit, terminate welding. However, remarkably, the welding timescale for most rhyolitic systems investigated appears to span a similar range regardless of the $\overline{P}$ of formation; hence $\langle \mu \rangle$, and therefore the degassing and temperature history, is the most important controlling parameter in welding for rhyolitic magmas.

5.3. *User-friendly computational tools for solving welding problems in volcanic scenarios in Python™ and Excel®*

As part of this contribution, we provide a downloadable executable file for Linux™ and Mac platforms, which solves the full diffusion–welding problem given here. The executable requires the following user inputs: particle size distribution (as a. txt or. csv file), initial porosity ϕ_i, initial dissolved water concentration C_i, initial temperature, gas pressure P_g, pressure differential $\Delta P = P_l - P_g$, surface tension Γ, and the spatial resolution for the diffusion solution (we set a default value of 100 steps, which is sufficient for most cases). We additionally allow the user to input a temperature rate, which should be positive for heating, negative for cooling, or zero for isothermal conditions, and which imposes a linear change in temperature. The outputs of this code are the monodisperse or polydisperse solutions for porosity as a

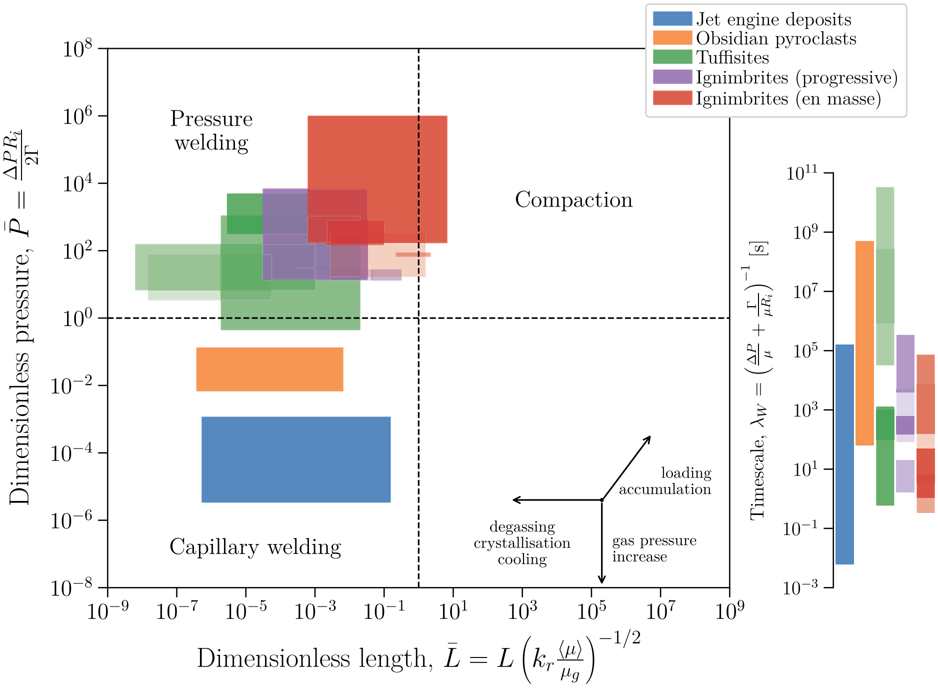

Fig. 5. Scenarios and regimes for volcanic welding. A plot of $\bar{P}$ and $\bar{L}$ regimes for volcanic welding with the range of conditions for natural examples given using constraints compiled in Table 1.

function of time and the value of $\langle C\rangle$. Similarly, we provide an editable Excel® sheet for solving our welding code for isothermal or non-isothermal conditions including for polydisperse particles (or droplets), but without diffusion of volatiles. This code is available via VHub (https://vhub.org/resources/4568).

6. Concluding remarks

We present a universal theoretical model of welding of natural volcanic material at relevant volcanic conditions. The model includes the complex effect of syn-welding dehydration, accounts for the effects of confining pressure and capillary pressure, and is valid for both isothermal and non-isothermal conditions. The welding model is grounded in the microphysical behaviour of a welding system, and requires no fitting parameters.

We use scaling arguments to assess the validity of the model for natural welding scenarios, and conclude that it can be applied to welding in tuffisites, in volcanic conduits, at the base of aggrading pyroclastic density currents, and in jet engines. Our model predicts that volcanic systems span the divide between the regime in which the capillary stress at particle walls drives welding, and the regime in which the driving pressure for welding is the difference between the liquid and the interstitial gas pressures. We find that in most cases examined here, the permeability of the interstitial gas phase does not limit the welding dynamics. Finally, we find that the total time required for complete welding spans seconds to years, and that the viscosity of the particles, or the evolution of viscosity during particle degassing, is the most variable parameter in nature.

The model we present provides a flexible and general tool for investigating welding phenomena across a wide range of volcanically-relevant scenarios. The model solution is given for the specific case of rhyolite welding via a vHub resource.

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Author contributions to this work

The team at the TOMCAT beamline (PSI, Switzerland) included F.B. Wadsworth (team lead), J. Vasseur, E.W. Llewellin, K.J. Dobson, F.W. von Aulock, J. Fife, and F. Marone. J. Schauroth and K.J. Dobson processed the 3D datasets and extracted raw data. F.B. Wadsworth, J. Vasseur, and E.W. Llewellin performed the analysis and model development. F.B. Wadsworth, J. Schauroth, J.E. Gardner, K.-U. Hess, M.J. Heap, H. Tuffen, and D.B. Dingwell provided additional *ex situ* or calibration data for analysis and consulted on the data treatment and the manuscript. F.B. Wadsworth, J. Schauroth, and E.W. Llewellin led the drafting of a manuscript text with help from all authors. T. Havard compiled data from natural deposits.

Appendix A. Supplementary material

Supplementary material related to this article can be found online at https://doi.org/10.1016/j.epsl.2019.115726.

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