1	The permeability of loose magma mush
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12	Models for the evolution of magma mush zones are of fundamental importance for
13	understanding magma storage, differentiation in the crust, and melt extraction
14	processes that prime eruptions. These models are underpinned by calculations of the
15	permeability of the evolving crystal frameworks in the mush, which controls the rate of
16	melt movement relative to crystals. Existing approaches for estimating the crystal
17	framework permeability do not account for crystal shape. Here, we represent magma
18	mush crystal frameworks as packs of hard cuboids with a range of aspect ratios, all at
19	their maximum random packing. We use numerical fluid flow simulation tools to
20	determine the melt fraction, specific surface area, and permeability of our 3D digital
21	samples. We find that crystal shape exerts a first-order control on both the melt fraction
22	at maximum packing, and on the permeability. We use these new data to generalize a
23	Kozeny-Carman model in order to propose a simple constitutive law for the scaling
24	between permeability and melt fraction that accounts for crystal shape in upscaled mush

dynamics simulations. Our results show that magma mush permeability calculated using

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26	a model that accounts for crystal shape is significantly different compared with models
27	that make a spherical crystal approximation, with key implications for crustal melt
28	segregation flux and reactive flow.
29	
30	Keywords: Darcy; volcanic eruption; rhyolite; magma reservoir; silicic magma
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32	INTRODUCTION
33	Substantial volumes of melt are stored in magma mush regions throughout the crust (Hildreth, 1981;
34	Sparks & Cashman, 2017). Models for the evolution of these magma mush zones are of fundamental
35	importance for magma storage timescales, differentiation in the crust, and melt extraction processes that
36	prime eruptions (Bachmann & Bergantz, 2004; Jackson et al. 2018). The initial assembly of crustal
37	magma bodies requires emplacement of, and percolative reactive flow through, crystal mushes (Jackson
38	et al. 2018). The eruption of crystal-poor magmas requires that melts be separated from these mushes
39	(Bachmann & Bergantz, 2004). While the details of these dynamics, the controlling processes, and the
40	overall rates, are all poorly constrained and discussed widely (Petford 2020; Holness 2018), in most
41	models, it is the permeability of the interlocking crystal framework that is a first-order rate-limiter.
42	Leading quantitative models for melt percolation dynamics on crystal scales use variations on a Kozeny-
43	Carman permeability law for which the crystals are assumed to have a single radius (Petford, 1995;
44	Bachmann & Bergantz, 2004; Jackson et al. 2018). Therefore, these constitutive models for mush
45	permeability cannot account for crystal shape or the difference in percolative hydraulic properties
46	between one mush and another if the phenocrysts are of similar size.

47 Petrological and geochronological evidence suggests that melt percolation and extraction prior to the
48 eruption of crystal-poor rhyolites occurs in transient and episodic events rather than continuously over
49 the thermal lifespan of the mush (e.g. Allan et al., 2013). In most known cases, the extraction timescales

50 derived from petrological methods are rapid compared with simple gravitational compaction processes, leading to models that involve additional heat by mafic recharge magmas (e.g. Huang et al. 2015), 51 and/or applied directional stresses and strain (Clemens & Petford, 1999; Bachmann & Bergantz 2008; 52 Holness 2018) resulting in anisotropic dilation of the mush (Liu & Lee, 2021). Differentiating between 53 54 one mechanism and another, or developing predictive frameworks for melt segregation timescales, all 55 depend on rigorous constitutive models for the permeability of real mushes (e.g. Bachmann & Bergantz, 2004), which remains poorly investigated. A key challenge is that the 3D shape of crystals is likely to 56 57 affect both the melt fraction at maximum crystal packing in the mush and the permeability at that melt 58 fraction, such that models should seek to constrain both effects simultaneously.

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# 60 METHODS: PERCOLATION OF MELTS THROUGH MAGMA MUSH

61 Melt extraction rates are given by the volumetric melt flux through a mush, Q, which in turn is governed by Darcy's law  $\nabla P = -\mu_f Q/(kA)$  where  $\nabla P$  is the driving melt pressure gradient,  $\mu_f$  is the melt shear 62 viscosity, k is the permeability of the mush, and A is the area normal to the extraction direction. 63 Throughout we make an isotropic assumption, such that mush permeability can be treated as a pseudo-64 scalar and equal in all directions, in line with previous work (e.g. Bachmann & Bergantz, 2004). 65 Previous models have used simple scaling laws for k that assume all crystals are spherical and can be 66 67 defined by their radius (e.g. Bachmann & Bergantz, 2004; Huber et al. 2010; Hartung et al. 2019; Floess 68 et al. 2019; Pistone et al. 2020). The most widely used model for k is the Kozeny-Carman equation, where k as a function of the solid volume fraction  $\phi$ , the specific surface area of the network s, and a 69 70 dimensionless constant C (c.f. Röding et al. 2020; Vasseur et al. 2021)

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$$k = \frac{(1-\phi)^3}{Cs^2}.$$
 Eq. 1

Using the specific surface area for volumes packed with monodisperse spheres of radius *R* gives  $s = 3(\phi)/R$ , which results in  $k = (1 - \phi)^3 R^2 / [9C(\phi)^2]$  (e.g. Hersum et al., 2015; Lui and Lee, 2021). *C* = 5 has been found to be a typical value for most granular systems (Vasseur et al. 2021; Röding et al. 2020; Torquato, 2013). However, the problem remains that the solid crystals in silicic mushes are often dominated by non-spherical crystals, and indeed may involve highly anisometric crystals such as high aspect ratio plagioclase (see Fig. 1A) or amphibole. Here, our central aim is to find a form of Eq. 1 that accounts for 3D crystal shape, and that can be used widely in mush evolution models.

80 We use numerical periodic domains generated by Liu et al. (2017) of packed and randomly arranged 81 solid cuboids to approximate magma mush, which is a geometry that is closer to natural crystal shapes than spheres (c.f. Fig. 1A). Our cuboids have axis lengths a, b, and c and length aspect ratios  $r_1 = c/a$ 82 and  $r_2 = b/a$ . The cuboids have a square cross-section such that a = b (hence,  $r_2 = 1$ ) and the domains 83 84 are produced at their random maximum packing, given by volume fraction  $\phi = \phi'$  (Figure 1B). Liu et 85 al. (2017) used order parameters to ensure that the packs are isotropic and disordered (i.e. no fabrics or cuboid preferred arrangements are found). We use a marching cubes algorithm to determine the specific 86 surface area of each cuboid pack (Lorensen & Cline 1987), and we use LBflow – a numerical lattice-87 Boltzmann fluid flow simulation tool (Llewellin, 2010a, 2010b) - to characterise steady-state fluid flow 88 89 through the inter-cuboid space and output the permeability of each cuboid domain (details of the 90 numerical analysis are provided in the Data Repository).

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## 92 RESULTS AND ANALYSIS

The results of our permeability determinations show that the permeability is a function of the melt fraction  $1 - \phi$  and the specific surface area as predicted by Eq. 1, which is a function of the crystal aspect ratio  $r_1$ . All raw results are provided in the **Data Repository**. In order to analyze these results in a unified manner across a range of crystal sizes, we introduce the dimensionless permeability  $\bar{k} =$  $k/k_s = ks^2/(2\phi)$ , where  $k_s$  is a generalized Stokes permeability (Vasseur & Wadsworth, 2017; Vasseur et al. 2020) and the specific surface area is measured directly for our packs. Our data for the

normalized permeability  $\bar{k}$  collapse to a single trend as a function of melt fraction, regardless of  $r_1$  and 99 100 crystal size (Fig. 2A), indicating that our non-dimensional approach captures these effects. In this normalized space, Eq. 1 becomes universal for any crystal shape and is  $\bar{k} = (1 - \phi)^3 / (2C\phi)$ ; we find 101 good agreement between the model and the cuboid dataset with the classic C = 5 (Torquato, 2013). To 102 103 calibrate this further, we compare our results with published permeability data for packs of hard spheres 104 normalized in the same way (Fig 2A). The excellent agreement we see between the numerical data and the model is used to validate Eq. 2 as a permeability model applicable to any particle/crystal shape as 105 106 long as s is known. We note that in the dilute limit as  $\phi \rightarrow 0$ , the sphere data deviate from Eq. 1, which is explored and modeled by Vasseur et al. (2021) using a dilute expansion of  $k_s$  (see **Data Repository**). 107

108 The analysis for  $\overline{k}$  relies on our determination of the specific surface area for each sample, which in 109 turn depends on the aspect ratio  $r_1$  and the melt fraction. In order to render this of wide utility in systems 110 for which *s* is not known *a priori*, we test our model using the theoretical specific surface area of a pack 111 cuboids with interstitial melt fraction (Eq.2; see **Data Repository** for derivation).

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$$s = \frac{2\phi}{a} \left( 1 + \frac{1}{r_1} + \frac{1}{r_2} \right)$$
 Eq. 2

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114 which reduces to  $s = 2\phi(2 + 1/r_1)/a$  when  $r_2 = 1$  (square-ended cuboids used here). As with k, we 115 can compare our results for s with the prediction of Eq. 2 across all cuboid packs used here, by making 116 Eq. 2 scale-independent via the normalisation  $\bar{s} = sa/\phi$ , which reduces Eq. 2 to  $\bar{s} = 4 + 2/r_1$ .

We find that our data for *s*, converted to  $\bar{s}$ , collapse to a single trend, which matches the prediction of this  $\bar{s}$  model (Fig. 2B). This shows that the normalized specific surface area  $\bar{s}$  decreases as the cuboids move from oblate (platy-habit such that  $r_1 \ll 1$ ) to prolate (needle-habit such that  $r_1 \gg 1$ ), meaning that rod-like crystals have a lower specific surface area at their maximum packing. Hence, with reference to Eq. 1, the permeability of maximally packed mush consisting of prolate crystals will be higher than that of a mush made from oblate crystals. The result presented in Fig. 2B (i.e. the success

of Eq. 2 in describing the specific surface area of the cuboid packs used here) suggests that the incidence of planar cuboid-cuboid contact surfaces is rare, and therefore justifies our use of Eq. 1 and the generalization of permeability by  $k \propto 1/s^2$ . We propose that Eq. 2 used with Eq. 1, validated herein, represents a universal model for the permeability of packs of cuboids as a proxy for the permeability of magma mush, and given in expanded dimensional form by

$$k = \frac{(1-\phi)^3 a^2}{4C\phi^2} \left(1 + \frac{1}{r_1} + \frac{1}{r_2}\right)^{-2}.$$
 Eq. 3

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# 130 LOOSE MUSH VS MAXIMALLY PACKED MUSH

Using Eq. 3, we can calculate the permeability of percolating mush using measured or estimated crystal 131 aspect ratios and sizes for a given melt fraction thereby accounting for crystal shape. Crystal shape not 132 133 only changes the permeability at a given melt fraction, but also strongly affects the maximum packing fraction itself. Mush maximum packing fraction  $\phi'$  is a function of  $r_1$  (Fig. 3), and for  $r_1 = 1$  (cubes), 134 there is a local minimum in  $\phi'$ , and local maxima at  $r_1 \approx 0.7$  and  $r_1 \approx 1.5$ . Our results are consistent 135 with the general form for previous results for loose random packs of non-spherical particles (e.g. Donev 136 137 et al. 2004; Wouterse et al. 2007; Rudge et al. 2008; Delaney et al. 2010; Meng et al. 2016; Liu et al. 2017). As crystals become highly oblate ( $r_1 \ll 0.7$ ) or highly prolate ( $r_1 \gg 1.5$ ),  $\phi'$  drops, and is 138 symmetric in log( $r_1$ ) around  $r_1 = 1$ . The function  $\phi' = \phi'_0(Ax + 1) \exp(-Bx)$  matches our data, 139 where  $\phi'_0 = 0.641$  is the numerically determined value of  $\phi'$  at  $r_1 = 1$ ,  $x = |\log_{10}(r_1)|$ , and A = 1.26140 and B = 1.04 are best-fit constants. 141

At high melt fraction, crystals do not interact or communicate force (i.e. a 'suspension'). Conversely, at the random maximum packing of crystals, a mush can support load and transmit force through the crystal framework but cannot densify further by compaction or other processes without deformation or re-organisation of the crystal framework. The transition from 'suspension' to a random maximally 146 packed mush can be termed the 'loose mush' region, and we posit that the percolative extraction of melt 147 begins when crystal fractions increase to a critical value  $\phi = \phi_{\tau}$ . We interpret  $\phi_{\tau}$  to be the lowest crystal volume fraction at which crystal-crystal force interactions can occur. Mueller et al., (2010) show that 148  $\phi_{\tau} \approx 0.8 \phi'$  for all  $r_1$ . Using this and our model for how the maximum packing varies with crystal shape, 149 we can quantitatively define the 'loose mush' region. Furthermore, using our general model (Eq. 3), we 150 can predict the permeability at  $\phi_{\tau}$  and  $\phi'$  for all  $r_1$ . We note here that  $\phi_{\tau}$  is an approximate and indicative 151 152 value, and that granular dynamics simulations demonstrate that a single melt fraction may be insufficient to demark the boundary between 'suspension' and 'loose mush' regimes (Deng et al. 2021). 153 Regardless, whatever definition of a lower-bound on  $\phi$  one places to demark 'loose mush', our model 154 155 can predict k for that  $\phi$ .

In Fig. 3 we show the results of our model (Eq. 3) in two modes of application. First, we show the 156 157 general results of our permeability model for any melt fraction and a range of crystal shapes (Fig. 3; 158 a = 1 mm). Second, we show the results of the model specifically for the upper and lower bounds on the 'loose mush' region, defined as when the crystal volume fraction is between the onset of crystal-159 crystal interactions, and the random maximum packing  $\phi_{\tau} < \phi < \phi'$ . This second mode of application 160 of Eq. 3 allows us to deconvolve the two principal effects predicted here: (1) the effect of  $r_1$  on  $\phi'$  or 161  $\phi_{\tau}$ , and (2) the resultant effect of  $r_1$  on the permeability. Fig. 3 shows that crystal shape can play a 162 163 substantial role in controlling the absolute value of the permeability in these 'loose' simulated crystal 164 mushes. A limitation of this model is that anisotropy is not considered, and that in nature, evolution of much from  $\phi_{\tau}$  to  $\phi'$  may well involve crystal rearrangements and fabric development (see Liu & Lee, 165 2021). 166

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### 168 IMPLICATIONS: RATES OF PERCOLATION THROUGH MAGMA MUSH

169 In this study, we have used packs of square-ended cuboids, however, via Eq. 3 our model is extensible 170 to crystals of arbitrary 3D shape. In order to apply our model to mush with real crystals, we use 171 published data for plagioclase phenocryst shapes (Duchene et al., 2008), wherein a, b, and c are 172 measured directly (a:b:c = 1: 6.5: 9.6; Duchêne et al. 2008). In all cases, we normalize all measured crystal shapes so that they are relative to a, which we take to be the shortest of the axes. We note that 173 this approach does not alter  $r_1$  and  $r_2$ . Then we assign a = 1 cm, in order to compute the permeabilities 174 of mushes that comprise those shapes. Using this workflow, we find that for a given melt fraction, the 175 permeabilities of the plagioclase mush fall within an order of magnitude of each other. Importantly, at 176 a melt fraction of 0.5, these datasets occur at predicted permeabilities up to a maximum of 1.5 orders 177 of magnitude greater than the prediction of the Jackson et al. (2018) scaling (Fig. 4). Since such models 178 predict the flux of melt to the shallow crust, we posit that our model has implications for overall melt 179 180 accumulation timescales. Our model (Eq. 3) can be used to predict the melt extraction rates, fluxes, and 181 characteristic timescales, and, importantly, our results suggest that crystal shape plays a first-order role in melt extraction because the timescales  $\lambda$  are proportional to permeability  $\lambda \propto k^{-1} \propto s^2$ . Mush 182 permeability exerts a first order control over the rates of this process, and hence crystal shape effects 183 184 need to be accounted for using our model.

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Figure 1. Mush texture compared with our simulated mush. (A) Ca-concentration map demonstrating the anisometric and cuboidal nature of plagioclase crystals (teal) with interstitial quartz (black) and clinopyroxene (grey; reproduced with permission from Holness et al., 2019, scalebar is 1 mm). (B) 3D visualization of a numerical cuboid pack ( $r_1 = 0.2$ ) with the flow pattern at steady state represented in the melt phase (scalebar is 100 µm).



Figure 2. The permeability and specific surface area of the cuboid packs analyzed. (A) The normalised permeability  $k/k_s$  as a function of melt volume fraction  $1 - \phi$ . The squares are the results from cuboid packs; the circles are for hard spheres (Vasseur et al. 2021) for validation and comparison. The solid curve represents our model using C = 5. The dashed curve is a dilute expansion for the 'suspension' regime at high melt fraction (Vasseur et al. 2021). (B) The scaling for *s* as a function of  $r_1$  for cuboids cast as the normalized  $\bar{s}$ .



Figure 3. (A) The maximum packing crystal volume fraction  $\phi'$  as a function of  $r_1$  (data from Liu et al., 2017) compared with our empirical model for  $\phi'$  (see text). The grey shaded area terminates against the upper bound of  $\phi'$  and a lower bound at  $\phi_{\tau} = 0.8\phi'$ . (B) The model (Eq. 3) solved using a = 1 cm. The black curves represent the result for each aspect ratio at the specific maximum packing value (see A). The cartoons on panel A are a visual representation of the mush at different crystal fractions.



Figure 4. The model (Eq. 3) solved using input 3D crystal shapes for plagioclase (Duchene et al., 2008) phenocrysts (a = 1 cm). Our model is compared with the scaling from Jackson et al. (2018)  $k = a^2\beta\phi^n$  where  $\beta = 1/125$  and  $\alpha = 3$  are the parameters proposed (Jackson et al. 2018). We also give a classical Kozeny-Carman model of the form  $k = (1 - \phi)^3 a^2 / [150(\phi)^2]$  (e.g. Torquato 2013). Both of these comparisons underpredict plagioclase mush permeabilities given here. The vertical line at  $\phi = 0.35$  is for comparison across models (Fig. 3A).

