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1 **The influence of olivine settling on the formation of basaltic cumulates revealed by micro-**  
2 **tomography and numerical simulations**

3

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5

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14

15

16 **Abstract**

17 The speed at which crystals settle in magmatic reservoirs affects the solidification rate of magmas and

18 their differentiation. Despite extensive prior work on the subject, most of our quantitative

19 understanding of the process is still restricted to treating crystals as spherical particles and does not

20 address the geometric complexities of natural crystals. Here, we use three-dimensional (3D) X-ray

21 microcomputed tomography (X- $\mu$ CT) observations on olivine crystals from Kīlauea Volcano (Hawai'i)

22 to document their highly intricate and variable geometries, and the textural growth relationships

23 between the olivine crystals and their inclusions (melt, spinel, and fluid/vapor bubbles). Olivine crystals

24 generally have clustered polyhedral or skeletal shapes, which reflect variable magmatic conditions (or

25 growth rates) during their formation. The cumulative presence of spinel, melt, fluid, and vapor

26 inclusions affects the density of the host crystals by up to 6% relative, and thus plays a limited role on

27 modifying crystal settling rate. In contrast, the overall crystal shape plays a major role. We performed

28 numerical simulations employing a finite element method to investigate the effect of crystal

29 morphology on settling rate and the evolution of the particle volume fraction in a magmatic convective

30 layer. We show that for all olivine geometries investigated, the settling velocity is highest when the

31 long axis of the crystal is aligned with the flow direction of the melt. Increasing the aspect ratio of the  
32 olivine tends to decrease its settling velocity and results in an increase in the influence of its  
33 orientation on the terminal velocity ( $U_T$ ). Extrapolation of the simulation results to variable particle  
34 volume fractions ( $\Phi = 0-0.5$ ) indicates high crystal settling rates ( $U_T = \sim 9.7 \times 10^{-6} - 1.4 \times 10^{-5}$  m/s) that  
35 are used to estimate the timescales for the formation of olivine cumulates in natural melt-dominated  
36 basaltic systems. The formation of olivine cumulates is therefore rapid, potentially leading to the  
37 accumulation of a crystal layer at the bottom, where the frictional contacts between the crystals exert a  
38 rheological lock-up acting against further convection. Crystal accumulation in the locked layer  
39 (parametrized with the solid/liquid volume ratio in the reservoir) is a function of the reservoir size and  
40 crystal fraction, and takes a few years in small reservoirs (less than 1 km thick) and a few decades in  
41 larger reservoirs (several kms thick). We propose a calibration of olivine suspension timescales for  
42 mafic magma reservoirs (based on the knowledge of the particle volume fraction, reservoir height, and  
43 olivine morphology). This calibration is used to estimate the rate of cumulate build-up, and can help  
44 interpret crystal size distributions in the framework of crystal suspension times.

45

#### 46 **Keywords**

47 Olivine; crystal settling; crystal morphology; X-ray microcomputed tomography; numerical simulations

48

#### 49 **Highlights**

- 50 • Solidification of a basaltic reservoir strongly depends on the settling rate of olivine
- 51 • Olivine terminal velocity is highest when the crystal long axis is aligned with the melt flow.
- 52 • Inclusions in olivine play a limited role on modifying crystal settling rate.
- 53 • Crystal accumulation in mafic systems takes a few years to a few decades.

54

#### 55 **1. Introduction**

56 Crystal settling has been considered a ubiquitous process in magma reservoirs for a long time (Murata  
57 and Richter, 1966). Crystal settling is thought to control the mode of solidification of magmas on Earth  
58 (Martin and Nokes, 1988; Koyaguchi et al., 1990; Pankhurst et al., 2018a) and in planetary magma  
59 oceans (Elkins-Tanton, 2012). Quantifying the rate at which magma reservoirs solidify is essential for  
60 the understanding of their chemical differentiation through time. Differentiation of a cooling magma is a

61 mediated process by crystal nucleation and growth or dissolution, settling and reentrainment (by  
62 magma replenishment or assimilation). Crystal growth rates in basaltic magmas are relatively well  
63 quantified (e.g., Jambon et al., 1992; Arzilli et al., 2015; Mourey and Shea, 2019) and the  
64 reentrainment of negatively buoyant particles (entrainment of grains in convective layers) has been  
65 addressed experimentally (Solomatov et al., 1993a; Lavorel and Le Bars, 2009) and theoretically  
66 (Solomatov and Stevenson, 1993b). In most cases, the accumulation of crystal layers at the bottom of  
67 mafic reservoirs involves growth or reprecipitation of olivine crystals while in suspension in the melt,  
68 followed by mechanical settling, compaction through pressure, dissolution at grain contacts, and  
69 expulsion of the interstitial melt (e.g., Schmidt et al., 2012; Holness et al., 2017). Experimental studies  
70 have previously reproduced textures of olivine-rich layers (Schmidt et al., 2012), but the influence of  
71 crystal shape on settling remains poorly quantified.

72

73 The shape of natural olivine crystals is highly variable (e.g., equant, elongated, tabular, skeletal,  
74 hopper, dendrite) and is related to undercooling, cooling rate and the melt composition (e.g.,  
75 Donaldson, 1976; Faure et al., 2003; Mourey and Shea, 2019). The well-faceted, polyhedral, tabular  
76 and hopper morphologies have been associated with relatively low undercooling ( $-\Delta T < 25^{\circ}\text{C}$ ; Faure et  
77 al., 2003; Mourey and Shea 2019, where  $-\Delta T$  is the temperature difference between the olivine  
78 liquidus and a temperature of interest below the liquidus), whereas skeletal and dendritic crystals are  
79 formed at moderate to high undercooling ( $-\Delta T > 25^{\circ}\text{C}$ ; Faure et al., 2003; Mourey and Shea 2019).  
80 Polyhedral crystals were initially assumed to crystallize at near-equilibrium conditions (Pearce, 1984)  
81 and in a concentric “tree-ring” fashion (e.g., Clark et al., 1986). However, morphological remains of an  
82 early rapid growth (e.g., stepped hopper cavities) are commonly observed in polyhedral olivine  
83 phenocrysts (e.g., Welsch et al., 2013), challenging the assumption of near-equilibrium, low  
84 undercooling conditions. Instead, polyhedral olivine phenocrysts are considered to have an initial rapid  
85 growth history, forming a skeletal olivine framework that is progressively infilled as undercooling and  
86 growth rates decrease (e.g., Welsch et al., 2013; Mourey and Shea, 2019; Wallace et al., 2021).

87

88 Olivine is the most abundant mineral in Hawaiian eruptions, and the only mineral (along with Cr-spinel)  
89 within active summit reservoirs (e.g. Thornber et al., 2015). Moreover, both olivine and spinel are the

90 first minerals to crystallize in basaltic melts with >7 wt.% MgO (Roeder et al., 2001). Unsurprisingly,  
91 spinel inclusions are common in olivine crystals.

92 In this study, we demonstrate how modern X-ray microcomputed tomography (X- $\mu$ CT) scans of olivine  
93 crystals can provide a wealth of new information on the three-dimensional (3D) distribution of melt  
94 inclusions, vapor bubbles, and spinels, in addition to the true 3D shape of crystals. The complexities of  
95 the inclusions network trapped in the crystals can help document crystal growth histories and thermal  
96 (undercooling) conditions.

97 We collected high-resolution X- $\mu$ CT scans of three populations of olivine crystals from the 1820  
98 Golden Pumice eruption (unit K1 from the Keanakākoʻi Tephra) at Kīlauea Volcano (Hawaiʻi). We  
99 document the internal complexities (melt inclusion, vapor bubble and spinel fractions) of well-faceted  
100 polyhedral crystals and more skeletal crystals, and interpret those complexities in terms of crystal  
101 growth conditions. Using numerical simulations, we test how different olivine morphologies affect  
102 settling velocities, with implications for the timescales of olivine-rich mush and dunite cumulate  
103 formation. We quantify how the volume fraction of inclusions impacts a crystal's density and terminal  
104 velocity. To investigate the influence of crystal morphology on settling, realistic crystal morphologies  
105 identified with X- $\mu$ CT scans are used to define initial crystal meshes with equal volumes and mass.  
106 The meshes are then integrated into numerical simulations of melt flow around the crystal employing  
107 the finite element method. The simulations explore the influence of the orientation of olivine crystals  
108 with the flow direction of the melt and the influence of their shape on the settling velocities. These  
109 velocities are then used to estimate settling timescales as a function of the crystal fraction and the  
110 reservoir size. Those timescales bring important new constraints on the rates at which cumulates and  
111 mush piles may form in mafic reservoirs, and can help petrologists better interpret crystal population  
112 characteristics (e.g. size distributions) in the light of suspension and accumulation rates.

113

## 114 **2. Methods**

### 115 **2.1. Sample description**

116 We used a sample from the Unit K1 (formerly Golden Pumice) eruption that occurred around 1820  
117 C.E. during the explosive Keanakākoʻi Tephra eruptive period at Kīlauea (Fig.1; Swanson et al., 2012).  
118 The eruption consisted in high lava fountains that produced fall deposits comprising chiefly rapidly  
119 cooled golden pumice with sparse lithic materials. The sample is glassy with 6.8 – 9.1 wt.% MgO in

120 the glass (Lynn et al., 2017) and contains different olivine populations ( $Fo_{80-89}$  ( $Fo = Mg/[Mg+Fe]$  in  
121 mol.%; Lynn et al. 2017; Mourey et al., 2022). No other phenocryst phases were found.

122

## 123 **2.2 X-ray micro-tomography**

124 In volcanology, X- $\mu$ CT has been used to measure the 3D shape and distribution of vesicles in volcanic  
125 rocks (e.g., Polacci et al., 2006), quantify bubbles nucleation and growth in multiphase systems  
126 (Oppenheimer et al., 2021), and characterize the morphology and chemical zoning of olivine crystals  
127 (Pankhurst et al., 2014; 2018b; Mourey and Shea, 2019). Traditionally, the interpretation of mineral  
128 textures is based on 2D images obtained with optical and electron microscopy. 2D images may lead to  
129 inaccurate or erroneous interpretations when addressing the complexities of 3D morphologies (e.g.,  
130 Mourey and Shea, 2019). The extrapolation to 3D from 2D images can be made by time-consuming  
131 and destructive techniques involving serial sectioning and grinding method (e.g., Byron et al., 1995;  
132 Daniel and Spear, 1999; Mock and Jerram, 2005). This method however results in low resolution in  
133 the direction perpendicular to the observed plane, and most of the material is unfortunately destroyed  
134 during polishing. Therefore, only partial records of the complexities of a 3D crystal are observed in 2D  
135 sections. Recent advances in X-ray computed microtomography (X- $\mu$ CT) during the last three  
136 decades better allow accessing real 3D information. This technique affords proper interpretation of the  
137 spatial relationships between textural and, sometimes, geochemical characteristics. X- $\mu$ CT is non-  
138 destructive and high spatial resolutions (sub-micron) can be attained.

139

140 The 1820 C.E. golden pumices were first gently crushed with a mortar and pestle, and a  
141 representative variety of olivine morphologies were hand-picked for X- $\mu$ CT scans. The crystals were  
142 coated with glass and, to make crystal faces more apparent, they were immersed in a 1 normal HF  
143 solution and cleaned (e.g., Lynn et al., 2017). Crystals were then mounted on carbon tape, carbon  
144 coated, and imaged with a JEOL-8500 field emission gun electron microprobe at the University of  
145 Hawai'i. A selection of crystals ( $n=13$ ) were scanned at the X- $\mu$ CT facility at the University of Austin  
146 Texas using an X-ray tube-based scanner (cone-beam geometry, Xradia MicroXCT-400). Each crystal  
147 was individually mounted on the rotating sample stage for X- $\mu$ CT scans. For each tomographic scan,  
148 1261 X-ray projections were acquired with a constant angular step over a  $360^\circ$  rotation. The effective  
149 pixel size of the images varied between 2.19 and 2.83  $\mu\text{m}$  for a sample-to-detector distance between 8

150 and 54 mm. A maximum beam energy of 70 keV was used for most of the olivine scans but was set at  
151 40 or 50keV for some rare crystals showing more beam artifacts (all conditions are reported in  
152 supplementary material). 3D reconstructions were done iteratively (e.g., adjustment of beam  
153 hardening, filter smooth). The images were imported in the ImageJ software to improve the contrast  
154 and brightness. For each scan, a brightness threshold is attributed for each phase (olivine, spinel, melt  
155 and vapour) in the Avizo™ software. Each image is then checked individually for additional  
156 segmentation corrections. After segmentation, melt and vapour renderings were smoothed during the  
157 surface reconstruction process using the filter packages in Avizo™ software. Textural measurements  
158 (size and volume) of the different phases (olivine, melt inclusions, vapor bubbles, spinels) were done  
159 using the Avizo™ software. The accuracy of the X-μCT technique to retrieve the phase volume (and  
160 number) is higher than classical techniques using point counting in transmitted light on 2D  
161 petrographic thin sections. For instance, most olivine crystals in a typical thin section are off-center  
162 intersected (Shea et al., 2015), and so measurements from thin sections generally underestimate the  
163 actual maximum dimensions of the crystals and their numbers.

164

### 165 **2.3. Numerical modeling**

166 Next, we aimed to use morphological constraints gleaned from 3D scans of natural olivine to  
167 investigate the influence of olivine morphology on rates of crystal settling in a basaltic melt. We  
168 simulated the flow of melt around crystals numerically, using morphologies representative of the range  
169 observed in 3D scans.

170 The equation of motion of a crystal suspended in melt (Carrara et al. 2019) may be written as:

$$171 \frac{dv}{dt} = \frac{\Delta\rho}{\rho_c} \mathbf{g} - \frac{\mathbf{F}_D}{m} \quad (1)$$

172 where  $\rho_c$  is the density of the crystal,  $\Delta\rho$  is the density contrast between the crystal and the surrounding  
173 melt ( $\Delta\rho = \rho_c - \rho_l$ ,  $\rho_l$  being the density of the melt),  $\mathbf{g}$  is the gravitational acceleration vector,  $m$  is the mass  
174 of the crystal, and the drag force imposed by the melt on the crystal  $\mathbf{F}_D$ . On the right-hand side of Eq.  
175 (1), the first term comprises both the gravitational and Archimedes' forces (reduced weight), while the  
176 second term corresponds to the drag force imposed by the melt on the crystal,  $\mathbf{F}_D$  divided by the crystal  
177 mass. The drag force can be further decomposed as the product of that of an isolated crystal and a  
178 voidage function (Di Felice, 1994):

179  $\mathbf{F}_D = \mathbf{F}_{D,0} f(\phi)$  (2)

180 with  $\mathbf{F}_{D,0}$  the drag force of an isolated crystal and  $\phi$  the voidage function defined as:

181  $f(\phi) = (1 - \phi)^{-\beta}$  (3)

182 where  $\phi$  is the solid volume fraction around an isolated crystal, and  $\beta$  is the coefficient of momentum  
 183 exchange between the crystal and the melt ( $\beta$  increases with crystal size and particle volume fraction;  
 184 see supplementary material). The coefficient of momentum exchange can be further decomposed as  
 185 (Di Felice, 1994):

186  $\beta = 3.7 - 0.65 \exp\left(-\frac{[1.5 - \log_{10}(\text{Re})]^2}{2}\right)$  (4)

187 which gives the following expression for the drag force:

188  $\mathbf{F}_D = \mathbf{F}_{D,0} (1 - \phi)^{-\left(3.7 - 0.65 e^{-\frac{(1.5 - \log_{10} \text{Re})^2}{2}}\right)}$  (5)

189 where  $Re$  is the particle Reynolds number.  $\beta_0$  depends on the viscosity and density of the melt, and the  
 190 shape, size, and orientation of the crystal. The second part of the right-hand side of Eq. (5) is a correction  
 191 that depends on the solid volume fraction (Di Felice, 1994). The particle Reynolds number (He and Tafti,  
 192 2018) is:

193  $Re = \frac{d_{eq} \|\mathbf{v} - \mathbf{u}\| \rho_l}{\eta}$  (6)

194 where  $d_{eq}$  is the diameter of a sphere having the same volume as the crystal, and  $\eta$  is the dynamic  
 195 viscosity of the melt.

196 For the conditions relevant to crystals settling in melt,  $Re < 0.1$ , such that Eq. (5) can be simplified as:

197  $\mathbf{F}_D = \mathbf{F}_{D,0} (1 - \phi)^{-3.7}$  (7)

198 The influence of the shape and orientation of the crystal on  $\mathbf{F}_{D,0}$  may be isolated from the influence of  
 199 its size and properties on the liquid phase by expressing it as a function of the drag coefficient,  $C_D$   
 200 (Zastawny et al. 2012; Sanjeevi et al. 2018):

201  $\mathbf{F}_{D,0} = \frac{1}{2} C_D A \rho_l |\mathbf{v} - \mathbf{u}| (\mathbf{v} - \mathbf{u})$  (8)

202 with  $A$  representing the cross-sectional area of the crystal,  $\mathbf{u}$  is the velocity vector of the melt,  $\mathbf{v}$  is the  
 203 velocity vector of the crystal. The drag force of an isolated crystal for which the cross-sectional area is  
 204 assumed to be that of an equivalent sphere of diameter  $d_{eq}$  such that  $A = \frac{\pi}{4} d_{eq}^2$  is defined as:

205  $\mathbf{F}_{D,0} = \frac{\pi}{8} C_D \rho_l d_{eq}^2 |\mathbf{v} - \mathbf{u}| (\mathbf{v} - \mathbf{u})$  (9)



206  $C_D$  depends on the density and viscosity of the liquid, the size of the particle, the shape of the particle,  
 207 and on the relative velocity between the flow and the particle. In the viscous regime, when  $Re < 1$  (almost  
 208 always the case for magmas),  $C_D$  is proportional to  $Re^{-1}$  (Sanjeevi et al. 2018) and is expressed as:

$$209 \quad C_D = \frac{\xi}{Re} \quad (10)$$

210 where  $\xi$  is a coefficient that depends on the shape and orientation of the particle. When the crystal  
 211 reaches its terminal settling velocity,  $U_T$ , the drag force equals the reduced weight of the particles such  
 212 as:

$$213 \quad \frac{\Delta\rho}{\rho_c} \mathbf{g} = \frac{\mathbf{F}_D}{m} \quad (11)$$

214 Inserting Eqs. 7 & 9 into Eq. (11) gives:

$$215 \quad \frac{\Delta\rho}{\rho_c} \mathbf{g} = \frac{\pi C_D \rho_l d_{eq}^2 (1-\phi)^{-3.7} |\mathbf{v}-\mathbf{u}|(\mathbf{v}-\mathbf{u})}{8m} \quad (12)$$

216 Inserting Eqs. (6 & 10) into Eq. (12):

$$217 \quad \frac{\Delta\rho}{\rho_c} \mathbf{g} = \frac{\pi \xi \eta d_{eq} (1-\phi)^{-3.7}}{8m} U_T \quad (13)$$

218 which gives the following expression for  $U_T$  after balancing the forces:

$$219 \quad U_T = \frac{8\Delta\rho g m}{\pi \xi \rho_c \eta d_{eq} (1-\phi)^{-3.7}} \quad (14)$$

220 where  $g$  is the magnitude of  $\mathbf{g}$ . Increasing the crystal volume fraction decreases the permeability of the  
 221 suspension and increases the velocity and tortuosity of the flow of liquid in the intergranular spaces,  
 222 which in turn increases the drag force and decreases the settling velocity (Carrara et al., 2019).

223 To determine the shape and orientation factor,  $\xi$ , we conducted numerical simulations of the flow of the  
 224 melt around the crystal and computed the total force applied by the melt on the surface of the crystal.  
 225 We simulated the flow of melt around single crystals using the software MIGFLOW (Constant et al.,  
 226 2018), which solves the Navier-Stokes equations for an incompressible liquid employing a finite element  
 227 method. To generate a tetrahedral mesh around the crystal, we employed the software GMSH  
 228 (Geuzaine and Remacle, 2009). The computational domain is rectangular with the crystal located at its  
 229 center (Fig. 2a). The melt is injected at the base of the domain at constant velocity,  $U_{inj}$ , and can flow  
 230 outside the domain thanks to an outflow at constant pressure located atop. The sides of the domain are  
 231 free-slip walls and the surface of the crystal is a non-slip wall. Simulations are performed with no gravity  
 232 so that the force generated by the hydrostatic pressure gradient (i.e., the Archimedes' force) is null and  
 233 does not influence the estimation of  $\xi$ .

234 We computed the total drag force applied to the crystal by the melt,  $\mathbf{F}_D$ , by summing the contributions of  
235 the liquid force on all the element faces located at the surface of the crystal (Fig. 2b). The integration of  
236 the total drag force may be written as (Zastawny et al. 2012):

$$237 \mathbf{F}_D = \int_S (-p\mathbf{n}_s + \boldsymbol{\tau}) dS \quad (15)$$

238 where  $p$  is the melt pressure at the surface of the crystal (initially equal to 0),  $\boldsymbol{\tau}$  is the viscous stress  
239 tensor,  $\mathbf{n}_s$  is the outward-pointing normal vector of the surface of the crystal, and  $dS$  is the incremental  
240 area.  $p$  is positive on the surface facing the flow and is negative on the faces on the other side of the  
241 crystal. The viscous stress is calculated as:

$$242 \boldsymbol{\tau} = \eta \frac{\partial \mathbf{u}^t}{\partial \mathbf{n}_s} \quad (16)$$

243 where  $\mathbf{u}^t$  is the tangential velocity of the melt to the surface of the crystal, and  $\partial \mathbf{u}^t / \partial \mathbf{n}_s$  is the gradient of  
244 the tangential velocity along  $\mathbf{n}_s$ . The shape and orientation factor of the isolated crystal is then calculated  
245 using a combination of Eq. (9) and Eq. (10):

$$246 \xi = \frac{8|\mathbf{F}_D|}{\pi\eta d_{eq} U_{inj}} \quad (17)$$

247 Three different characteristic olivine morphologies were tested, with variable aspect ratios along the  
248 three crystallographic axes: *a*- and *c*-elongated crystals, as well as an equant crystal. The choice of  
249 these morphologies is partly based on previous experimental studies and natural olivine crystals  
250 (Donaldson, 1976; Welsch et al., 2013; Mourey and Shea, 2019; Gordeychik et al., 2020), including  
251 those imaged in this study. All the crystal geometries were generated using MATLAB and have the same  
252 volume ( $\sim 4.19 \text{ mm}^3$ , which is equivalent to a 1-mm radius sphere). For each crystal morphology, we  
253 performed three simulations with one of the crystallographic axes aligned with the flow direction of the  
254 melt. For all the simulations and computations of settling velocity, we used the same physical properties  
255 for the melt and crystal (see Table 1). The ability of the numerical model to constrain settling velocities  
256 was validated by comparing the terminal velocity of a sphere using our numerical model and using the  
257 Stokes velocity (relative error of 0.89%; see supplementary material).

258

### 259 **3. Results**

#### 260 **3.1. 3D textures of Keanakāko'i Tephra olivine**

261 The Keanakāko'i Tephra (unit K1) contains a large variety of olivine morphologies (Table 2 and Fig.3),  
262 which were classified in three morphological populations:

263

264 (1) Clustered polyhedral crystals are elongated along the *a*- or *c*-axis (4 clusters analyzed) and have  
265 the largest olivine grains (>2 mm; Fig. 3a). The complex morphology of these crystals is evaluated  
266 using the surface-area to volume (SA:V) ratio, measured by Avizo™. Clustered polyhedral crystals  
267 have low SA:V values between 1 mm<sup>-1</sup> and 2 mm<sup>-1</sup>. The clusters analyzed consist in 4 to 7 principal  
268 olivine phenocrysts (>500 μm), with up to 20 additional microphenocrysts- to microlite-sized crystals.  
269 Individual phenocrysts within polyhedral clusters have a more equant shape (relative to the other  
270 crystal morphologies discussed below) and large SA:V ratios (up to 20 mm<sup>-1</sup>).

271 (2) Clustered skeletal crystals (2 clusters analyzed; Fig. 3b) are elongated along the *a*-axis and  
272 connected by melt inclusions that typically track the direction of primary and secondary growth  
273 branches (Fig. 3b). These crystals have high SA:V ratios between 7 mm<sup>-1</sup> and 14 mm<sup>-1</sup>. Spinel  
274 inclusions are concentrated near the olivine crystal edges. The crystal morphology and the abundance  
275 of melt inclusions suggests that clustered skeletal olivine formed at moderate-to-high undercooling  
276 conditions (i.e., -ΔT = 40-60°C; Mourey and Shea, 2019).

277 (3) Single skeletal crystals (7 crystals analyzed) are generally elongated along the *a*-axis (Fig. 3c).  
278 They are characterized by small, elongated melt inclusions (<50 μm) that are fully enclosed and not  
279 connected to vapor bubbles. Larger melt inclusions (up to 400 μm) usually form a tubular melt network  
280 that wraps vapor bubbles from both sides of the olivine crystals and may be open to the olivine surface  
281 (Fig. 3c). Spinel inclusions are only observed near the crystal edges. These crystals have medium  
282 SA:V values between 2 mm<sup>-1</sup> and 6 mm<sup>-1</sup>. Similar to the clustered skeletal crystals, single skeletal  
283 crystals likely formed at moderate-to-high undercooling conditions (i.e., -ΔT = 40-60°C).

284

### 285 **3.1.1. Volume of melt, vapor bubble and spinel inclusions**

286 The volume fraction of melt inclusions (reported in the supplementary material) varies from 3.6 to 22.1  
287 vol.% in the single and clustered skeletal crystals (Fig. 4), but it is much smaller in the polyhedral  
288 clusters (between 0.45 and 1.4 vol.%; Fig. 4). Spinel makes up 0.3 - 1.4 vol.% of the total crystal  
289 volume. It is mostly located on the edges of skeletal crystals, and in both the cores and rims of  
290 polyhedral clusters (Fig. 3a). The volume occupied by vapor bubbles is usually low in polyhedral  
291 crystals (0 to 0.15 vol.%), medium in skeletal clusters (1.1 to 1.8 vol.%) and highly variable in single  
292 skeletal olivine crystals (0.24 to 4.8 vol.%). The vapor bubbles are generally attached to melt

293 inclusions (but some small bubbles are isolated as fluid inclusions as in k1820-ol8A; see  
294 supplementary material). Spinel, melt and vapor bubble inclusions modify the density of individual  
295 crystals in polyhedral clusters by < 0.5 % (Table 2; see supplementary material for detailed  
296 calculations). These inclusions affect the density of skeletal crystals (single or clusters) to a larger  
297 extent (up to 6% less dense). In the next sections, we compare the dimensions of the inclusions  
298 (spinel, vapor bubble, melt) of the different olivine crystal morphologies.

299

### 300 **3.1.2. Size distributions of melt, vapor bubble and spinel inclusions**

301 For ease of reporting and discussion, we express the size of individual objects (melt inclusions,  
302 spinels, vapor bubbles) as the diameter of a volume-equivalent sphere ( $d_{eq}$ ).

303 The morphology of olivine-hosted melt inclusions is different in the three olivine types, suggesting  
304 variable magmatic conditions (or growth rates) for their formation and/or maturation. The size of melt  
305 inclusions (most of which are fully enclosed) in single skeletal olivine is highly heterogeneous (Fig. 5a).  
306 The largest melt inclusions in single skeletal olivine vary from  $d_{eq} < 5 \mu\text{m}$  to  $d_{eq} = 415 \mu\text{m}$ . As discussed  
307 in previous studies (Faure and Schiano, 2005; Mourey and Shea, 2019; Wallace et al., 2021), small  
308 melt inclusions may be inherited from larger inclusions that were split during crystal maturation and  
309 closing in of the skeletal framework. Melt inclusions in clustered olivine crystals are for the most part  
310 highly segregated (and highly oblate), with  $d_{eq} < 100 \mu\text{m}$  (on average,  $d_{eq} = 29 \mu\text{m}$ ). Melt inclusions in the  
311 clustered olivine are equant, slightly larger than those in the skeletal crystals ( $d_{eq} < 200 \mu\text{m}$ ,  $d_{eq} = 46$   
312  $\mu\text{m}$  in average).

313 Vapor bubbles are usually connected to melt inclusions in clustered and single skeletal olivine crystals.  
314 Bubbles in clustered, *c*-elongated crystals are not common. They can be associated with melt  
315 inclusions or isolated as fluid inclusions (Fig. 3). The largest vapor bubbles (on average  $d_{eq} = 56 \mu\text{m}$ ;  
316 maximum  $d_{eq} = 275 \mu\text{m}$ ) are observed in single skeletal olivine crystals elongated along the *a*-axis  
317 (Fig. 5b). Smaller bubbles are present in clustered skeletal ( $d_{eq} = 44 \mu\text{m}$  average size) and *c*-  
318 elongated olivine clusters ( $d_{eq} = 27 \mu\text{m}$  average size).

319 Most spinel crystals are completely enclosed within their host olivine, or in some cases, partially  
320 exposed at the olivine surface. Their average and maximum sizes vary between the three olivine  
321 morphologies (Fig. 5c). In clustered and single skeletal crystals, they are generally smaller ( $d_{eq} = 9 \pm 2$   
322  $\mu\text{m}$  and maximum at  $d_{eq} = 40\text{-}50 \mu\text{m}$ ). In the polyhedral clusters dominated by *c*-elongated olivine

323 crystals, spinel crystals are on average larger ( $d_{eq}=15\ \mu\text{m}$ ) with some well faceted microphenocrysts  
324 (up to  $d_{eq}=170\ \mu\text{m}$ ; e.g., k1820-13A or k1820-5B). It is not possible to detect spinels smaller than  $\sim 2$   
325  $\mu\text{m}$  (voxel resolution) using X- $\mu\text{CT}$  scans or objects smaller than  $8\ \mu\text{m}^3$ . The number density of spinel  
326 is low in polyhedral crystal clusters (128 to 2,093 spinels / $\text{mm}^3$ , or 405 to 3,184 spinels/cluster; Fig.  
327 5d), medium-high in skeletal crystal clusters (3,461 and 4,289 spinel / $\text{mm}^3$ , or 448 to 627  
328 spinels/cluster), and highly variable for the single skeletal crystals (1,953 to 17,736 spinel / $\text{mm}^3$ , or 106  
329 to 2,093 spinels/crystal). Spinel size and volume in single and clustered skeletal crystals overlap with  
330 those of polyhedral crystal clusters (*c*-elongated, more mature crystals; Mourey and Shea, 2019). The  
331 volume of Cr-spinel observed in the olivine is likely related to the kinetics of olivine growth but through  
332 the mechanism is not yet understood. Welsch et al. (2013) proposed that Cr enrichments formed  
333 around rapidly growing olivine as Cr behaved incompatibly, leading to the crystallization of Cr-spinel  
334 that are later progressively buried in the inner part of the olivine crystal during slower growth.  
335 However, Cr generally shows partitionless or compatible behavior in the olivine-basalt system  
336 (Schreiber 1979; Lang et al. 2022) and therefore cannot easily pile up during rapid growth. It is  
337 possible that Cr-spinel simply forms prior to olivine and gets incorporated during rapid or slow growth  
338 (e.g., Roeder et al. 2006). The variation in the number density of spinel in the crystal may be related to  
339 changed in growth kinetics but further experiments are required to elucidate the exact mechanism  
340 through which Cr-spinel is included in olivine, often in high numbers.  
341 Overall, the crystals scanned by X- $\mu\text{CT}$  highlight the negligible effect of inclusions (melt/fluid, vapor  
342 bubble, Cr-spinel) on crystal settling (inclusions have little contribution to the particle density). Next, we  
343 leverage the morphologies identified in the 3D scans to define initial olivine crystal meshes that are  
344 integrated into numerical simulations (see section 2.3.). These simulations explore the influence of  
345 crystal shape and its orientation respective to the flow direction of the melt on the settling velocities.  
346

### 347 **3.2. Settling velocities**

348 The results of the simulations are reported in terms of the coefficients of momentum exchange and  
349 settling velocities (Table 3) in the absence of other crystal (i.e., clusters). Figure 6 displays the evolution  
350 of the terminal velocity as a function of the solid volume fraction computed with Eq. (10) for all the  
351 orientations we simulated. In the context of a shear flow at low volume fraction, crystals tend to align

352 along the streamlines. At intermediate crystal volume fractions, the contacts between the crystals disturb  
353 this alignment and generate bimodal distributions of the shape-preferred direction (Arbaret et al., 2007).  
354 The preferential orientation of the crystals along their elongation direction compared to the melt flow  
355 minimizes friction with the melt and leads to an increase in the crystal's settling velocity. Overall,  
356 increasing the aspect ratio of a crystal tends to exacerbate the influence of its orientation on the settling  
357 velocity (Schwindinger and Anderson, 1989).

358 For the two non-equant crystals (elongated parallel to *a*- and *c*-axis; Fig. 2), the settling velocity is  
359 the highest when the long axis is parallel to the flow of the melt, and the lowest when the short axis is  
360 oriented in the flow direction. For the *a*-elongated crystal, the gap between the two lowest settling  
361 velocities (runs A2 and A3 in Fig. 6) is significant because the difference between the lengths of the  
362 short and intermediate axes (*b*-axis < *c*-axis) affects the projected areas of the crystal in the direction of  
363 the flow. The projected area is largest when the flow is along the *b*-axis (see *a*-elongated olivine in Fig.  
364 2) and the settling velocity at a minimum (Fig. 6). This is illustrated by the larger contribution of the  
365 pressure component of the drag force in run A3 (~50% of the total drag force; Table 3) compared to the  
366 other simulations. On the contrary, the olivine elongated along the *c*-axis has its short and intermediate  
367 axes with similar lengths, such that the projected areas and settling velocities are more similar when the  
368 flow is aligned along the *a*-axis or *b*-axis (runs B1 and B2). The projected area of the crystal in the  
369 direction of the flow is, however, a bit larger when the *a*-axis is aligned with the flow, as illustrated by the  
370 larger contribution of the pressure component to the total drag force in run B1 (Table 3). The effect of  
371 the larger projected area on the settling velocity is balanced by the influence of the crystal morphology,  
372 which is more protruding along the *a*-axis than the *b*-axis (Fig. 2c). This makes the crystal more  
373 hydrodynamic along the *a*-axis than the *b*-axis, balancing the influence of the projected area and  
374 resulting in comparable settling velocities.

375 For the equant crystal, the difference between the settling velocities is controlled by the morphology  
376 of the surface of the crystal. The projected area of the crystal is the largest when the flow is along the  
377 *b*-axis, resulting in a lower settling velocity (Fig. 6) and a larger contribution of the pressure component  
378 to the total drag force (Table 3). The crystal has almost the same projected area when the flow is aligned  
379 along the *a*-axis or *c*-axis. The difference between the two settling velocities in these orientations results  
380 from the morphology of the crystal, which is more protruding along the *a*-axis.

381 To summarize, the simulations showed that the settling velocity is highest when the long axis of the  
382 crystal is aligned with the flow direction of the melt. Settling velocity decreases with the increasing  
383 aspect ratio of the crystal and leads to an increase in the influence of the crystal orientation (with the  
384 flow direction) on the terminal velocity. Based on these simulations, we find relatively high crystal  
385 settling rates ( $U_T$ ) of  $\sim 9.7 \times 10^{-6}$  to  $1.4 \times 10^{-5}$  m/s (or 3.5 to 5.0 cm/hour) for particle volume fractions  
386 ( $\Phi$ ) of 0 to 0.5. The complex olivine geometries decrease by factors of 3 to 5 the settling rates to that  
387 of ideal spheres with the same diameter ( $\hat{U}_T$ ; Fig. 6). Overall, our estimated settling rates are more  
388 refined than previous Stokes velocities for Kilauea olivine ( $6.5 \times 10^{-8}$  m/s to  $4 \times 10^{-5}$  m/s;  
389 Schwindinger 1999). In the following sections, we exploit our new settling rate estimates to constrain  
390 the timescales for the formation of olivine cumulates in mafic magma reservoirs. Then, we consider  
391 how crystal suspension times in magmatic reservoirs may affect real crystal size distribution (CSD)  
392 measured in Kilauea samples.

393

## 394 **4. Discussion**

### 395 **4.1. Suspension time of olivine crystals in basaltic melts**

396 In this section, we evaluate the timescales over which olivine crystals can stay in suspension in a basaltic  
397 melt. We term this timescale the suspension time. We consider olivine crystals with realistic  
398 morphologies that undergo vigorous melt convection following an updated version of the model of Martin  
399 and Nokes (1988). This model considers a suspension of particles convecting within a viscous liquid of  
400 a given volume. A rheologically-locked layer, where the frictional contacts between the crystals oppose  
401 motion (Huber et al., 2010) is located at the base of the convecting volume (Fig.7a). Martin & Nokes  
402 (1988) model is valid at the base of the fluid where the convective velocity is zero, presumably meaning  
403 that that fluid thickness must be rather small. However, their model holds to a very good approximation  
404 even when the settling velocity is comparable to the convective velocity over the whole depth of the  
405 fluid. Therefore, we consider a convective layer of relatively small thickness (i.e. 200 m; Fig.7a)  
406 representative of the inferred size of most of Hawaiian sills (Ryan et al., 1983). The evolution of the  
407 number of particles located within the convective volume,  $N$ , is calculated using Eq. (6) of Martin and  
408 Nokes (1988):

$$409 \frac{dN}{dt} = \frac{-U_T}{h} N \quad (18)$$

410 where  $U_T$  is the terminal settling velocity, and  $h$  is the height of the convective layer. Considering all the  
411 crystals as having the same volume, the number of particles within the convective layer,  $V_c$ , can be  
412 expressed as a function of  $\phi$ :

$$413 \quad N = \frac{\phi V}{V_c} \quad (19)$$

414 where  $V$  is the total volume of the convective layer (considered constant). Inserting Eq. (19) into Eq.(18)  
415 and neglecting the growth or dissolution of crystals ( $V_c$  constant) gives:

$$416 \quad \frac{d\phi}{dt} = \frac{-U_T}{h} \phi \quad (20)$$

417 Inserting Eq. (9) into Eq. (20) gives the following differential equation describing the evolution of  $\phi$  in the  
418 convective layer:

$$419 \quad \frac{d\phi}{dt} = \frac{-\Delta\rho g m \phi}{h \rho_c F_{D,0} (1-\phi)^{-3.7}} \quad (21)$$

420 To compare our model to that of Martin and Nokes (1988), we consider a convective layer with a  
421 height of 200 m with an initial particle volume fraction  $\phi_0 = 0.5$ . We considered a dilute regime (where  
422 particle volume fractions in suspension represent <1 vol.%) and an intermediate regime (with particle  
423 volume fractions between 1-50 vol.%) but did not considered concentrated regimes where particle  
424 volume fractions is close to the maximum packing and where crystal-crystal interactions become  
425 important for the overall effective viscosity, the mobility of magmas and decrease magma convection  
426 (Bachmann and Bergantz, 2004; Faroughi and Huber, 2023). The coefficient of momentum exchange  
427 and terminal velocity used as input for the models are taken from the result of the 9 numerical  
428 simulations (Table 3). Unlike the model of Martin and Nokes (1988), our calculations account for the  
429 dependence of crystal settling velocity on  $\phi$ . Therefore, the suspension time of crystals in the  
430 convective layer is a function of the initial particle volume fraction ( $\phi_0$ ; Fig. 7).

431 We find that if the settling velocity is constant (with a settling velocity fixed at  $\phi = 0$ , as predicted by Eq.  
432 13), the suspension time tends to be underestimated (predicted by Eq. 20; Fig. 7) and the settling rate  
433 of the particles is overestimated in comparison with the Martin and Nokes (1988) model. We neglect  
434 the decrease of magma convection caused by the high fraction of particles ( $\phi$  up to 0.5) that can  
435 decrease our suspension times and therefore we propose maximum estimates for the suspension  
436 times. Moreover, the model does not consider the reentrainment of some crystals near the interface on  
437 top of the convective layer after their settling. The volume fraction of particles in suspension  $\phi$   
438 decreases with time for a given reservoir height (Fig. 7).  $\phi$  in the convecting layer reaches less than



439 0.1 after 3 to 4 years for a 200 m magma body. Crystal settling of most of the particles ( $\phi$  close to 0) in  
440 a small reservoir (i.e., sill with a height of 200 m) is reached in around 5 years in all the simulations  
441 performed.

442 Magma mixing and the addition of gas bubbles in the magma leads to complications for the  
443 understanding of crystal settling in natural volcanic systems. In natural magmas, crystals and silicate  
444 melt are interacting with exsolved volatiles at shallow depths. Adding a supercritical fluid such as CO<sub>2</sub>  
445 bubbles increases the settling rate of crystals (Schwindinger, 1999). Magmas with initially low volatile  
446 contents at depths (i.e., 0.3 wt.% CO<sub>2</sub> at >1 kbars for Kīlauea) can exsolve large amounts of volatile at  
447 shallower depths (in our previous example, up to 35 vol.% at 50 bars (Holloway, 1977; Flowers, 1979;  
448 Silver et al., 1990; Dixon et al., 1995; Anderson, 1995; Schwindinger, 1999). The upward streaming of  
449 the gas, associated with an intruding MgO-rich melt, can initiate a lateral segregation of the crystals and  
450 enhance the sedimentation rate of the olivine crystals by a factor of 5-10 (Schwindinger, 1999).  
451 Therefore, our settling rate estimates should be viewed as minimum values. Considering the likely  
452 presence of exsolving volatiles in natural basaltic reservoirs, the suspension time of 1-mm crystals,  
453 similar to those modeled in this paper, are on the order of a few years even for large mafic reservoirs  
454 (>1 km thick).

455

#### 456 **4.2. Application to mafic systems and the formation of olivine mushy zones**

457 Our crystal settling simulations can be used to constrain the timescales required for the formation of  
458 olivine cumulates in basaltic systems. One limitation in our numerical simulations is that we did not  
459 consider the lateral motion of crystals expected to occur in natural sills (DiBenedetto et al., 2020).  
460 Crystal settling timescales in such sills are assumed to be short, and therefore cumulates can form  
461 rapidly in layers where convection is not effective. Notwithstanding, our results have implications for  
462 interpreting the formation of mush systems in active magma reservoirs. In mafic systems like Kīlauea,  
463 the solidification rate of a magma reservoir depends on olivine nucleation, growth and settling rates.  
464 Recrystallization can also affect the differentiation of a solidifying reservoir. Settling rates of olivine  
465 crystals, and thereby the timescales to form olivine cumulates, can only be constrained with  
466 information about their 3D geometry and the respective proportions of each crystal morphology (e.g.,  
467 same number of crystals elongated along *a*-, *b*-, and *c*-axis). Future work is needed to document the

468 3D morphology of olivine crystals in exposed olivine cumulates to access information about their  
469 formation.

470 The relatively short suspension times of olivine crystals in a convective layer (a few years depending  
471 on the crystal volume fraction and reservoir size) could explain why most Pele's hairs, tears, scoria  
472 and tephra collected from lava lakes are typically aphyric (e.g., Thornber et al., 2015). Lava lakes like  
473 those at Kīlauea produce occasionally glassy juvenile ejecta falling as Pele's hair and tears, and small  
474 clots of spatter on the rim of Halema'uma'u (Wilson et al., 2008), but never sample the large  
475 proportions (~14 vol.%; Clague and Denlinger, 1994) of olivine cumulates that are present beneath  
476 Kīlauea summit, aerial and submarine Rift Zones (Delaney et al., 1990; Johnson, 1995; Denlinger,  
477 1997). One reason may be that olivine settling is fast and efficient, which could be enhanced by  
478 exsolved volatiles (Schwindinger 1999). We propose that a rapid increase in the proportion of locked  
479 crystals that are not convecting and are not reinjected into the convecting melt is responsible for the  
480 formation of olivine aggregates observed in Hawaiian and other basaltic volcanoes. An increase in  
481 magma supply rate (as recently observed during the 21<sup>st</sup> century at Kīlauea, ~0.2 km<sup>3</sup>/yr; Poland et al.,  
482 2014) increases the amount of heat provided to the reservoir and in turn the vigor of the convection,  
483 and it can trigger the reentrainment of some crystals near the interface between the locked layer and  
484 the convective layer. However, Kīlauea olivine crystals in the inner part of the locked layer may be well  
485 compacted and possibly deformed (Clague and Denlinger, 1994) and therefore not easily unlocked.

486 The preponderance of magma mixing at Kīlauea (Lynn et al., 2017; Mourey et al., 2023) also favors  
487 undercooling and heating of the existing magma (and its crystal cargo) that counterbalance the  
488 solidification of the resident magma.

489

490 Using previous estimates of the vertical extent of olivine cumulate mush piles at Kīlauea (~180–720 m  
491 with ~40% porosity; Wieser et al., 2019), the constraints on Kīlauea reservoir radius from  
492 interferometric synthetic aperture radar (~2-4 km by InSAR; Baker and Amelung, 2012), and for olivine  
493 crystals equivalent to a 1 mm-radius sphere (same volume as in our simulations), we can estimate that  
494 a locked layer of mush may contain about  $2.2 \times 10^{17}$  to  $3.5 \times 10^{18}$  crystals. The convective melt may in  
495 turn contain  $1.6 \times 10^{16}$  to  $2.6 \times 10^{17}$  crystals, assuming an olivine volume fraction of 0.05 (Thornber et  
496 al., 2015). The locked layer (or mush piles) therefore contains ~13times more crystals than the  
497 convecting layer. Using our olivine crystal settling rates ( $U_T = 7.5 \times 10^{-6}$  to  $1.1 \times 10^{-5}$  m/s at  $\phi = 0.05$ ),

498 we can estimate that 5 vol.% of olivine crystals in the convecting layer can reach the bottom of the  
499 180–720 m reservoir in ~6 months to 3 years (ignoring the influence of volatiles and possible lateral  
500 segregation). Assuming continuous nucleation of olivine crystals in the magma column, we estimate  
501 that olivine mush piles at Kīlauea can be formed in < 5 years. Suspension timescales for olivine  
502 crystals are much shorter than crystal storage timescales retrieved from melt inclusion studies (e.g.,  
503 scavenging of century-old primitive olivine crystals with new olivine cargo during the Maunaulu 1969-  
504 1974 eruption; Wieser et al., 2019) and a negligible fraction of the tholeiitic shield stage period at  
505 Kīlauea Volcano (starting around 240 ka years ago; Garcia et al., 2017). The low crystal fraction in  
506 Kīlauea summit lavas (up to 5 vol.%) therefore implies that most of olivine cargo that nucleated at the  
507 summit is stored either at the bottom of the summit reservoirs or in the Rift Zones. The poor constraint  
508 on the intensity of magma convection at Kīlauea potentially strongly increases our crystal suspension  
509 times (longer suspension) in the case of a highly convective magma reservoir. Our model also  
510 assumes homogeneous nucleation in the magma column and do not consider any preferential crystal  
511 nucleation or the formation of crystal cluster by growth (branching or surface nucleation) or synneusis  
512 (reorientation and attachment of the grains to minimize the surface energy) that could also increase  
513 our crystal settling estimates (Culha et al., 2020).

514

### 515 **4.3. Application to CSD studies**

516 Our olivine settling rates can also be used to evaluate the time frames over which crystal size  
517 distributions (CSD) produced by crystal nucleation, growth, and ripening can evolve by gravitational  
518 settling in mafic reservoirs. For a fixed reservoir height ( $h$ ) and terminal velocity ( $U_T$ ), the suspension  
519 time of olivine crystals is a function of the particle volume fraction  $\phi$  (Fig. 8). For olivine spanning  
520 diameters equivalent to phenocryst sizes observed at Kīlauea (250  $\mu\text{m}$  to 8 mm; Vinet and Higgins,  
521 2010, 2011; Mourey and Shea, 2019), the suspension times vary from <1 year to a few years for mm-  
522 sized crystals, up to a few centuries for smaller phenocrysts (250  $\mu\text{m}$ ). For instance, the phenocryst  
523 fraction (>250  $\mu\text{m}$ ) from CSDs extracted from samples of the Maunaulu 1969-1974 eruption (Vinet and  
524 Higgins, 2010) correspond to suspension times ranging from ~6 months (for larger phenocrysts) to  
525 ~120 years (for crystals close to 250  $\mu\text{m}$  in size). Our calibration could therefore be used to re-interpret  
526 CSDs from melt-dominated basaltic systems in terms of crystal suspension times.

527

## 528 **Conclusion**

529 We conducted X- $\mu$ CT scans of olivine crystals from Kīlauea Volcano to elucidate the complexities of  
530 olivine internal textures. The crystal scans reveal complex melt, spinel and vapor bubble shapes, and  
531 a large diversity of inclusion volumes. We show that mineral, melt and gas inclusions only modify the  
532 density of crystals by up to 6% relative, which has a limited effect on crystal settling rates in  
533 comparison with the crystal shape. We thus explore the effect of crystal morphology on settling rate  
534 using numerical simulations. We find that the settling velocity is higher when the elongated  
535 crystallographic axis is aligned with the flow of the melt. We evaluate the time of the olivine crystals  
536 suspended in a reservoir undergoing vigorous convection. Suspension times vary with the initial  
537 particle volume fraction, but in general, olivine crystals have short (<5 years) suspension times in a  
538 convective layer. The short suspension times may in turn affects the rates of solidification at the base  
539 of a basaltic reservoir in synergy with magma cooling and crystallization. We also propose a  
540 calibration of olivine suspension times (based on known particle volume fraction, reservoir height,  
541 olivine morphology) that can be used to interpret the CSDs in terms of crystal suspension times in a  
542 mafic reservoir. Crystal settling is a potentially key melt differentiation mechanism, as has been  
543 highlighted at some ocean island volcanoes (e.g., Pankhurst et al., 2018a) but may also be of  
544 importance at other melt-dominated mafic volcanoes.

545

546

## 547 **CRedit authorship contribution statement**

548 **Adrien J. Mourey**: Conceptualization, Data curation, Formal analysis, Investigation, Methodology,  
549 Software, Visualization, Writing – original draft. **Alexandre Carrara**: Conceptualization, Funding  
550 acquisition, Investigation, Methodology, Software, Visualization, Writing – original draft, Writing –  
551 review & editing. **Thomas Shea**: Funding acquisition, Investigation, Methodology, Software, Writing –  
552 review & editing. **Fidel Costa**: Conceptualization, Funding acquisition, Writing – review & editing.  
553 **Marc-Antoine Longpré**: Conceptualization, Funding acquisition, Writing – review & editing.

554

## 555 **Declaration of competing interest**

556 The authors declare that they have no known competing financial interests or personal relationships  
557 that could have appeared to influence the work reported in this paper.

558

559 **Data availability**

560 All data obtained in this study are included in the supplementary data tables.

561

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809 **Fig. 1. Location of Kīlauea Volcano (left panel) and map of Kīlauea's summit (right panel).**  
810 Background digital elevation model from August 2018, with the 2018 collapse area shaded in pink.  
811 Black cross represents coordinate system at 19.4073° N, 155.2784 °W. The location of the Golden  
812 Pumice eruption unit (Unit K1 from the explosive Keanakāko'i Tephra), the source of olivine  
813 phenocrysts used for this study, is indicated by the red star.

814 **Fig. 2. Numerical simulations of olivine settling.** (a) Numerical simulation of the flow of melt around  
815 an olivine crystal. The black box corresponds to the limit of the computational domain. The two gray  
816 surfaces represent the mass inflow and pressure outflow boundary conditions. The crystal is located at  
817 the center of the domain. The curves located within the domain are the melt flow streamlines color-  
818 coded for magnitude of flow velocity. (b) Zoom on the crystal. The red line indicates the edges of the  
819 particles. The mesh corresponds to the surface elements on which the force applied by the liquid is  
820 computed. (c) Geometry of the olivine crystals we have tested. Crystals used in the numerical  
821 simulations are either elongated along the *a*-axis, elongated along the *c*-axis or equant. The three  
822 crystal morphologies have the same volume ( $\sim 4.19 \text{ mm}^3$ ) equivalent to a 2-mm sphere diameter.  
823

824 **Fig. 3. Representative secondary electron images (in grey) and 3D renderings of olivine**  
825 **crystals from the 1820 Keanakāko'i (unit K1) eruption.** (a) Clustered polyhedral olivine elongated  
826 along the c-axis; (b) Clustered skeletal olivine elongated along the a-axis; (c) Skeletal olivine  
827 elongated along the a-axis. Numbers in the pie charts are relative volume % of the different phases in  
828 the crystal (melt inclusions, Cr-spinel, vapor bubbles) retrieved from the Avizo™ software. Note that  
829 only inclusion fractions are represented in the pie charts (not the olivine fraction).  
830



831 **Fig. 4. Melt inclusion and vapor bubble relative volume % extracted from 3D renderings. Cr-**  
832 **spinel in olivine makes up <1.5 volume % in all cases.**

833

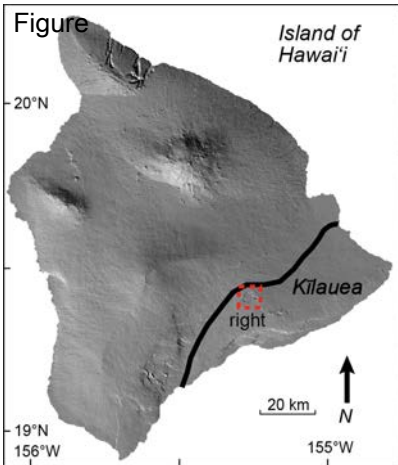
834 **Fig. 5. Size of inclusions in olivine.** Distribution (illustrated as violin plots) of equivalent sphere  
835 diameters of (a) melt inclusions, (b) vapor bubbles, (c) Cr-spinel crystals and (d) density of Cr-spinel /  
836 mm<sup>3</sup>) in skeletal crystal clusters, polyhedral crystal clusters and single skeletal olivine crystals  
837 elongated along the *a*-axis. Cr-spinels are larger in polyhedral clusters (both the mean and maximum  
838 diameter) than in skeletal clusters and skeletal single crystals. Most melt inclusions have an equivalent  
839 sphere diameter <100 μm (segmented melt inclusions) but larger melt inclusions (up to 415 μm) are  
840 also observed in skeletal olivine crystals elongated along the *a*-axis. The filled white circles indicate  
841 the median value of the distribution, the filled grey areas are the interquartile ranges, and the thin  
842 black lines represent the rest of the distribution.  
843  
844

845 **Fig. 6. Variation of the settling velocity ( $U_T$ ) of olivine crystals with particle volume fraction ( $\phi$ )**  
846 **for different crystal geometries and orientation with respect to the melt flow direction.** Inset:  
847 ratio of olivine settling rates with different geometries to that of ideal spheres with the same diameter  
848 ( $\hat{U}_T$ ) as a function of  $\phi$  ( $\phi = 0$  for the Stokes' velocity). The highest speeds are those when the  
849 elongated crystal axis is aligned with the flow. Complex crystal geometries decrease settling rates by  
850 factors of 3 to 5 compared to spherical shapes.  
851

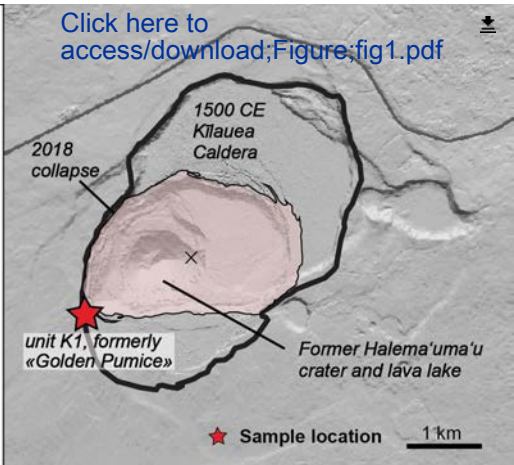
852 **Fig. 7. Time evolution of the particle volume fraction ( $\phi$ ) in suspension in the convective melt**  
853 **layer as a function of the crystal morphology.** (a) We consider a convective layer (in blue), in which  
854 convection occurs and with particle volume fraction  $\phi=0-0.5$ , and a locked layer (in red), in which no  
855 convection occurs (with  $\phi >0.5$ ). The black circles represent the particles. The red arrows indicate the  
856 convection. The green dashed line indicates the limit between the convective and locked domain. A  
857 relatively small convective layer of 200 m is considered for the calculations since the fluid thickness  
858 must be rather small for the Martin & Nokes (1988) model. (b) Suspension times (in years) of olivine  
859 crystals in the convective layer as a function of the particle volume fraction ( $\phi$ ), the crystal morphology  
860 (see the parameters for the simulations in Tables 1 & 3). The blue, red and black curves correspond  
861 to the numerical solution of Eq. (20) for the different simulations.  
862

863 **Fig. 8. Effect of crystal size ( $d_{eq}$ ) on suspension time and as a function of the particle volume**  
864 **fraction ( $\phi$ ).** Only phenocryst sizes between 250  $\mu\text{m}$  and 8 mm (in diameter) are displayed. The size  
865 of the reservoir ( $h$ ) is fixed at 200 m for the calculations. The terminal velocity ( $U_T$ ) is fixed at  $1.36 \times 10^{-5}$   
866 m/s (olivine elongated along a-axis with the a-axis aligned with the flow, run A1 in Table 3). This  
867 calibration for the suspension timescales can be adapted for a given magmatic system with a known  
868 magma reservoir height and olivine morphology.

Figure

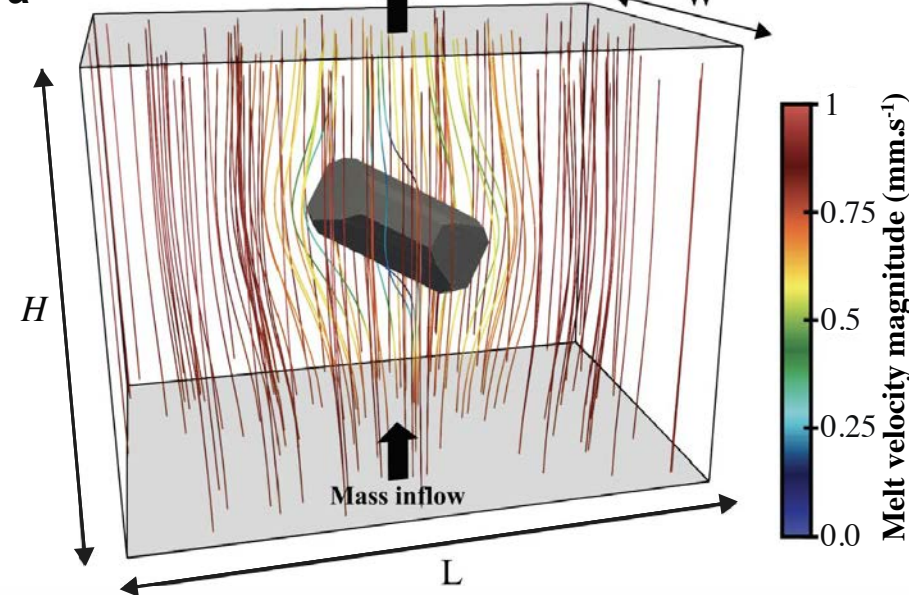


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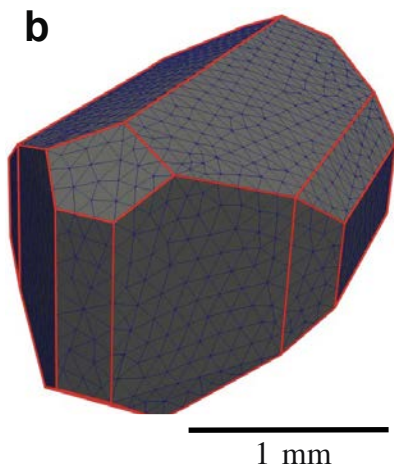


Figure

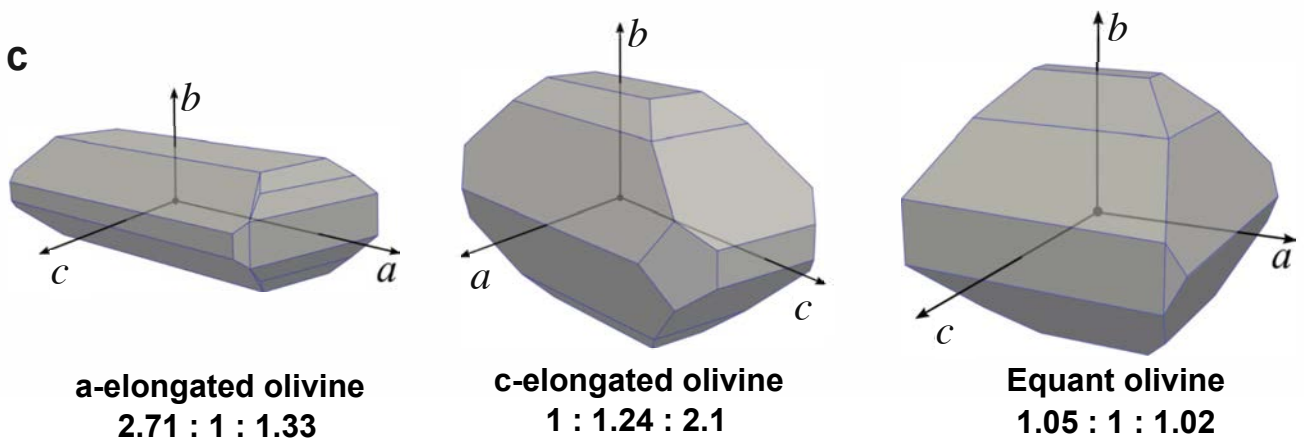
**a**



**b**



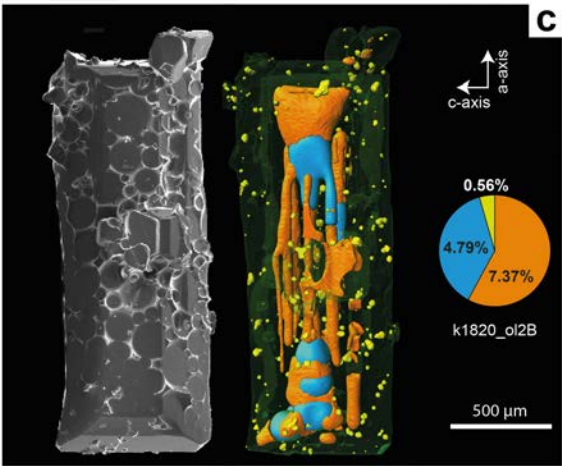
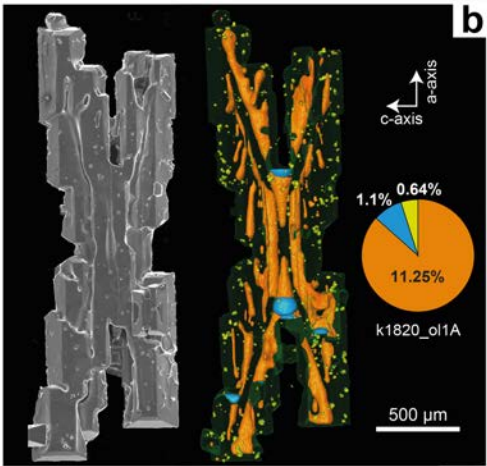
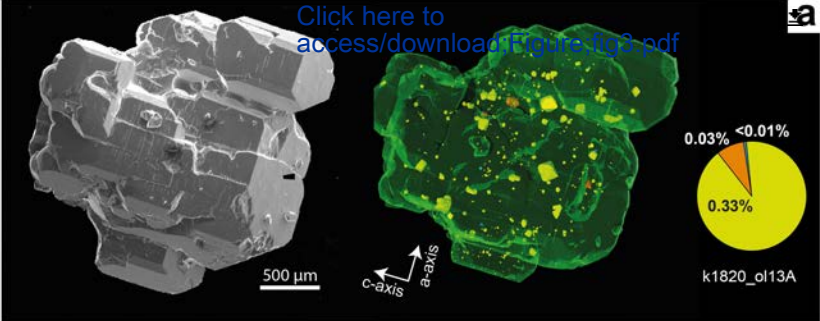
**c**



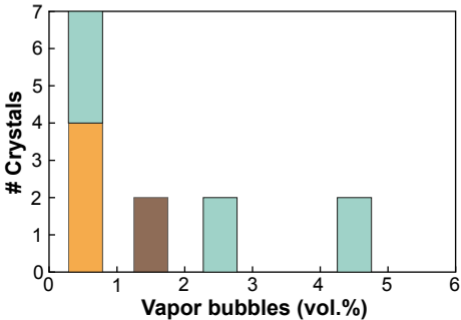
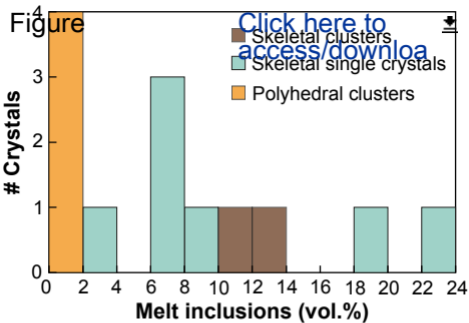
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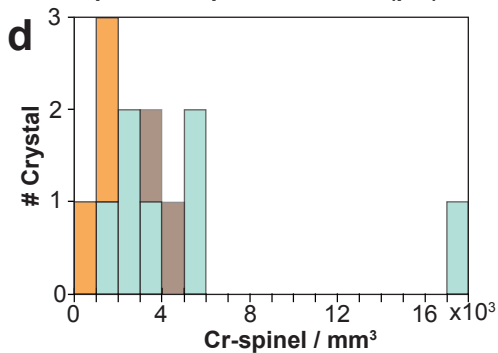
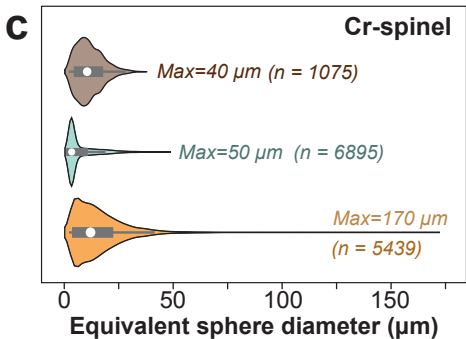
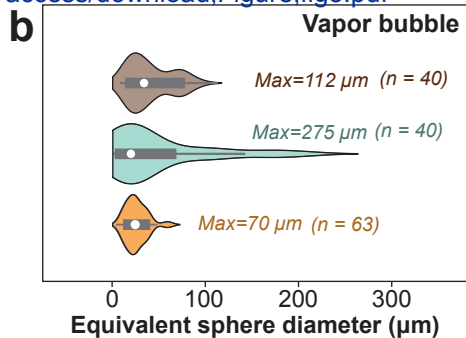
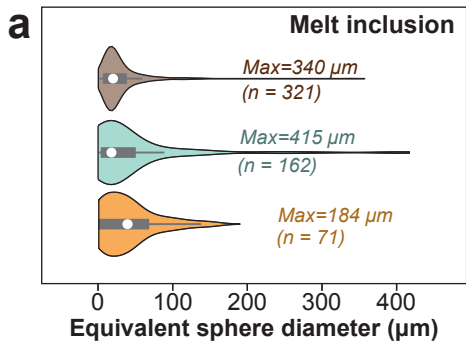
Figure  
**Legend for**  
**X- $\mu$ CT scans**

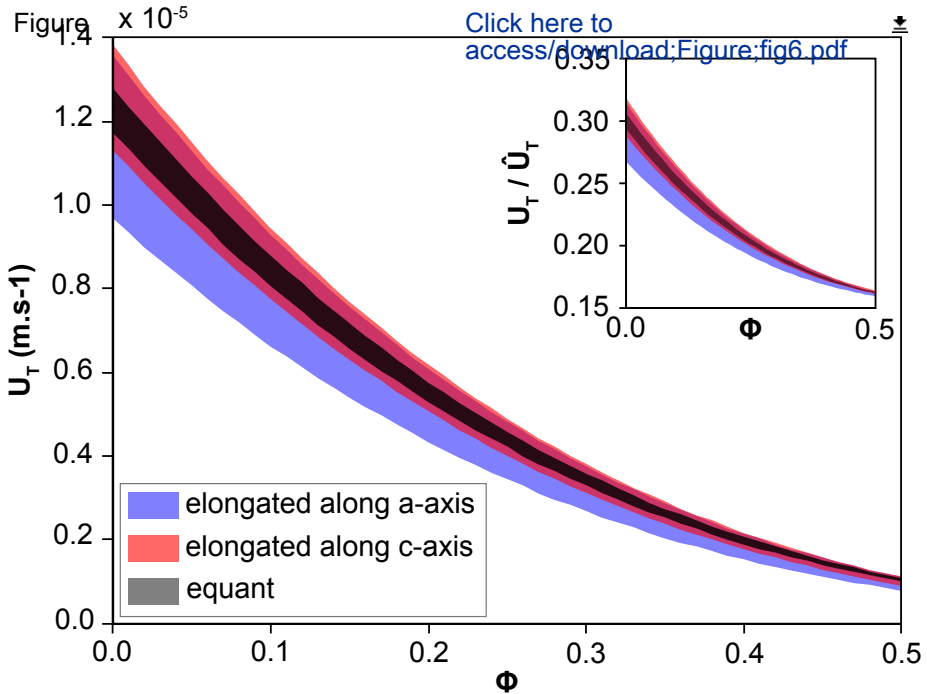
- Olivine
- Melt inclusions
- Cr-spinel
- Vapor bubble











Figure

$\phi = 0.5$

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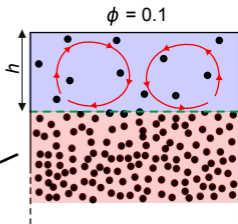
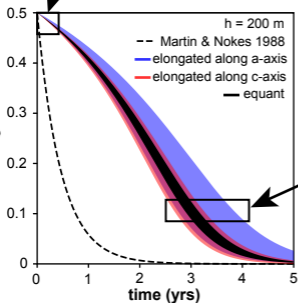
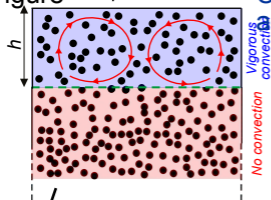
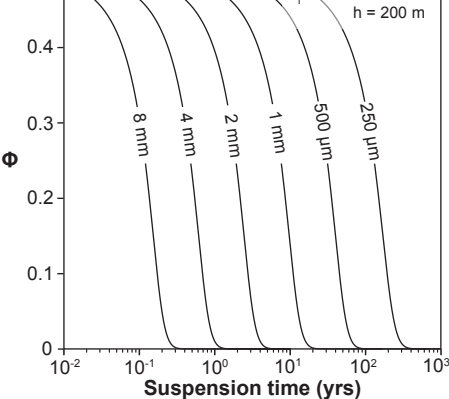


Figure 9.5 Olivine equivalent spherical diameter  $\rightarrow$   
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**Table 1:** Physical properties of the crystal and melt. Symbols used are  $\rho_l$  (melt density),  $\eta$  (melt viscosity),  $\rho_c$  (crystal density),  $U_{inj}$  (injection velocity).

Physical properties	Value	Reference
$\rho_l$	2750 kg m <sup>-3</sup>	Mourey and Shea (2019)
$\eta$	30 Pa s	Giordano et al. (2008)
$\rho_c$	3400 ± 50 kg m <sup>-3</sup>	Lynn et al. (2017); Mourey et al. (2022)
$U_{inj}$	10 <sup>-3</sup> m s <sup>-1</sup>	–

**Table 2:** Olivine crystal dimensions along a = [100], b = [010], c = [001] axis, equivalent sphere diameter, density of olivine crystal, effect of inclusions (melt, Cr-spinel, vapor bubbles) on crystal density. Details on density calculations are in the supplementary material.

Olivine name	Morphology	[100] length (μm)	[010] length (μm)	[001] length (μm)	Density crystal with inclusions (kg m <sup>-3</sup> )	Olivine/crystal density difference due to inclusions (%)
k1820_ol1A	Skeletal cluster	2035	159	701	3253-3340	-3.20 to -2.90
k1820_ol4A	Skeletal cluster	1487	289	849	3212-3296	-4.45 to -4.11
k1820_ol2B	Skeletal crystal elongated along [100]	1916	353	729	3152-3239	-6.12 to -5.92
k1820_ol3A	Skeletal crystal elongated along [100]	1735	452	844	3312-3405	-1.32 to -1.15
k1820_ol14A	Skeletal crystal elongated along [100]	1517	832	330	3155-3230	-6.38 to -5.83
k1820_ol9A	Skeletal crystal elongated along [100]	872	351	479	3246-3328	-3.55 to -3.09
k1820_ol15A	Skeletal crystal elongated along [100]	705	297	347	3236-3326	-3.60 to -3.42
k1820_ol11A	Skeletal crystal elongated along [100]	1040	362	612	3143-3229	-6.41 to -6.17
k1820_ol10B	Skeletal crystal elongated along [100]	953	304	581	3339-3434	-0.47 to -0.33
k1820_ol8A_1	Polyhedral crystal elongated along [100]	923	391	600	3381-3480	+0.87 to +0.94
k1820_ol12A_1	Polyhedral crystal elongated along [100]	1565	580	1207	3381-3480	+0.11 to +0.16
k1820_ol12A_2	Polyhedral crystal elongated along [100]	1220	556	817	3355-3454	+0.11 to +0.16
k1820_ol12A_3	Polyhedral crystal elongated along [001]	1030	826	1238	3355-3454	+0.11 to +0.16
k1820_ol13A_1	Polyhedral crystal elongated along [100]	2311	961	1404	3353-3453	+0.08 to +0.09
k1820_ol13A_2	Polyhedral crystal elongated along [100]	631	464	619	3353-3453	+0.08 to +0.09
k1820_ol13A_3	Polyhedral crystal elongated along [100]	927	596	516	3353-3453	+0.08 to +0.09
k1820_ol13A_4	Polyhedral crystal elongated along [100]	1082	634	650	3353-3453	+0.08 to +0.09
k1820_ol5B_1	Polyhedral crystal elongated along [001]	1172	813	1261	3358-3457	+0.20 to +0.25
k1820_ol5B_2	Polyhedral crystal elongated along [100]	971	408	612	3358-3457	+0.20 to +0.25
k1820_ol5B_3	Polyhedral crystal elongated along [001]	532	350	684	3358-3457	+0.20 to +0.25
k1820_ol5B_4	Polyhedral crystal elongated along [001]	725	613	1209	3358-3457	+0.20 to +0.25
k1820_ol5B_5	Polyhedral crystal elongated along [001]	491	467	582	3358-3457	+0.20 to +0.25
k1820_ol5B_6	Polyhedral crystal elongated along [001]	307	250	371	3358-3457	+0.20 to +0.25
k1820_ol5B_7	Polyhedral crystal elongated along [001]	87	77	97	3358-3457	+0.20 to +0.25
k1820_ol5B_8	Polyhedral crystal elongated along [001]	81	233	250	3358-3457	+0.20 to +0.25

1 **Table 3:** List of the simulations and results.  $\xi$  is a coefficient that depends on the shape and  
 2 orientation of the particle (see Eq. 17), %press. is the percentage of the contribution of pressure drag  
 3 to the total drag force ( $\int_S -p \mathbf{n}_s dS$  in Eq. 15), %frict. is the percentage of the contribution of viscous  
 4 friction to the total drag force ( $\int_S \tau dS$  in Eq. 15),  $U_T$  is the terminal settling velocity,  $\phi$  is the solid volume  
 5 fraction at the location of the  
 6 crystal.  
 7

Run	Crystal	Orientation	Axis lengths a, b, c ( $\mu\text{m}$ )	$\xi$	% press.	% frict.	$U_T$ at $\phi = 0$ ( $\text{m s}^{-1}$ )
A1	Elongated along a-axis	<i>a</i> -axis aligned with the flow	3414; 983; 1648	83.6	21.3	78.7	$1.36 \times 10^{-5}$
A2	Elongated along a-axis	<i>b</i> -axis aligned with the flow	3414; 983; 1648	102.8	31.9	68.1	$1.10 \times 10^{-5}$
A3	Elongated along a-axis	<i>c</i> -axis aligned with the flow	3414; 983; 1648	117.2	50.3	49.7	$9.67 \times 10^{-6}$
B1	Elongated along c-axis	<i>a</i> -axis aligned with the flow	1426; 1742; 3022	100.2	44.3	55.7	$1.13 \times 10^{-5}$
B2	Elongated along c-axis	<i>b</i> -axis aligned with the flow	1426; 1742; 3022	99.7	38.0	62.0	$1.14 \times 10^{-5}$
B3	Elongated along c-axis	<i>c</i> -axis aligned with the flow	1426; 1742; 3022	82.1	22.5	77.5	$1.38 \times 10^{-5}$
C1	Equant	<i>a</i> -axis aligned with the flow	2040; 1942; 1978	88.4	35.6	64.4	$1.28 \times 10^{-5}$
C2	Equant	<i>b</i> -axis aligned with the flow	2040; 1942; 1978	96.4	35.6	64.4	$1.18 \times 10^{-5}$
C3	Equant	<i>c</i> -axis aligned with the flow	2040; 1942; 1978	90.8	38.3	61.7	$1.25 \times 10^{-5}$

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