

Crystal fractionation by crystal-driven convection

Cansu Culha¹, Jenny Suckale¹, Tobias Keller¹, and Zhipeng Qin²

¹Stanford University

²STANFORD UNIVERSITY

November 24, 2022

Abstract

Fractional crystallization is an essential process proposed to explain worldwide compositional abundances of igneous rocks. It requires crystals to precipitate from the melt and segregate from its residual melt, or crystal fractionation. The compositional abundances of volcanic systems show a bell-curve distribution suggesting that the process has variable efficiencies. We test crystal fractionation efficiency in convective flow in low to intermediate crystallinity regime. We simulate the physical segregation of crystals from their residual melt at the scale of individual crystals, using a direct numerical method. We find that at low particle Reynolds numbers, crystals sink in clusters. The relatively rapid motion of clusters strips away residual melt. Our results show cluster settling can imprint observational signatures at the crystalline scale. The collective crystal behavior results in a crystal convection that governs the efficiency of crystal fractionation, providing a possible explanation for the bell curve distribution in volcanic systems.

Crystal fractionation by crystal-driven convection

Cansu Culha¹, Jenny Suckale¹, Tobias Keller^{1,2}, Zhipeng Qin¹

¹Stanford University, Department of Geophysics, 397 Panama Mall, Stanford CA 94305, USA.

²University of Glasgow, School of Earth and Geographical Sciences, 8NN University Avenue, Glasgow
G12 8QQ, UK.

Key Points:

- Crystals in liquid-rich environments settle efficiently and fractionate as crystal-rich clusters.
- Collective settling increases the efficiency of fractional crystallization compared to individual settling but depends on crystallinity.
- Depending on cluster dynamics, adjacent crystals may have been exposed to different melt environments leading to different zoning patterns.

Corresponding author: Cansu Culha, cculha@stanford.edu

Abstract

Fractional crystallization is an essential process proposed to explain worldwide compositional abundances of igneous rocks. It requires crystals to precipitate from the melt and segregate from its residual melt, or crystal fractionation. The compositional abundances of volcanic systems show a bell-curve distribution suggesting that the process has variable efficiencies. We test crystal fractionation efficiency in convective flow in low to intermediate crystallinity regime. We simulate the physical segregation of crystals from their residual melt at the scale of individual crystals, using a direct numerical method. We find that at low particle Reynolds numbers, crystals sink in clusters. The relatively rapid motion of clusters strips away residual melt. Our results show cluster settling can imprint observational signatures at the crystalline scale. The collective crystal behavior results in a crystal convection that governs the efficiency of crystal fractionation, providing a possible explanation for the bell curve distribution in volcanic systems.

1 Introduction

The worldwide compositional abundances of igneous rocks, including both volcanic and plutonic settings, show bimodal peaks at basaltic and rhyolitic compositions, referred to as the Daly gap (Daly, 1925). One of the key processes contributing to the formation of compositional gaps is fractional crystallization (Clague, 1978; Dufek & Bachmann, 2010; Jackson et al., 2018), a reactive transport process that requires crystals to precipitate from the melt and segregate from their residual melt. While some authors (e.g., Bonnefoi et al., 1995) have hypothesized that fractional crystallization leads to even distributions of composition, studies of compaction show variable efficiency of fractional crystallization (Dufek & Bachmann, 2010; Jackson et al., 2018).

A closer look at the worldwide compositional abundances reveals that their distribution is different in volcanic as compared to plutonic systems. Volcanic systems exhibit a single peak at basaltic compositions, whereas plutonic systems are characterized by a bimodal distribution with peaks at both basaltic and rhyolitic compositions (e.g. Chayes, 1963; Reubi & Blundy, 2009; Lee & Bachmann, 2014; Keller et al., 2015). The fundamental difference in the compositional distributions for the two settings suggests a corresponding difference in the processes governing magma evolution with compaction-driven fractional crystallization being more relevant in the plutonic rather than the volcanic setting.

The goal of this letter is to quantify the variable efficiency of fractional crystallization at low to intermediate crystal fraction. Our study is motivated by understanding the distribution of compositions in volcanic settings, which tend to be characterized by melts with less than 50% crystallinity prior to an eruption (Vigneresse et al., 1996; Jicha et al., 2005; Wieser et al., 2019a). We focus specifically on the segregation component of fractional crystallization, which we refer to as crystal fractionation (e.g., Bowen, 1928). We hypothesize that crystal-crystal interactions increase the efficiency of fractional crystallization by triggering a transition from individual to collective settling.

We test our hypothesis through direct numerical simulations that resolve the crystal-melt interactions at the scale of individual crystals (Suckale et al., 2012a; Qin & Suckale, 2017). As a consequence, we do not require any *a priori* parametrizations of phase-interactions such as effective viscosity, segregation drag, or settling speed. Instead, these quantities emerge self-consistently from simulations. Our simulations hence allow us to quantify the nonlinear ramifications of complex physical processes, such as the dynamic, long-range interaction between crystals, at the scale of individual crystals. An attractive attribute of our model approach is that it is testable against crystalline-scale data and could enable an assessment of the processes leading to fractional crystallization at the field-site

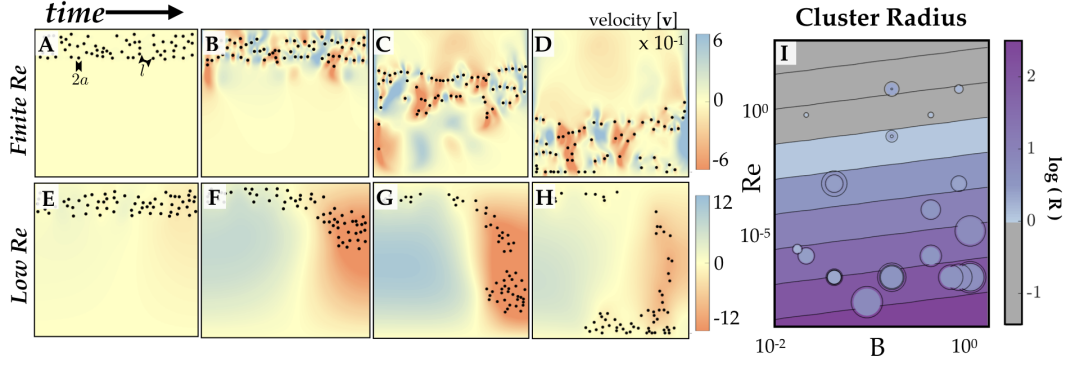


Figure 1. Varying degree of collectivity in crystal settling. Temporal snapshots from two different simulations at finite (A–D) and low (E–H) particle Reynolds number (Re). Both simulations have the same B value. They both show a form of collective motion between the crystals, but the degree of collectivity is more pronounced at low Re . (I) summarizes the degree of collectivity as estimated by the size of the crystal-rich cluster for different Re and B . In (I), the size of the circles indicate the size of the cluster relative to crystal radius. Each circle in the diagram is a simulation. The magnitude size of the clusters, R_m and the background colors, R indicate crystal cluster size relative to crystal radius as described by Supp. Sec. 2.

scale. To demonstrate the potential of the model in this regard, we compare our simulation results to variability observed in compositional profiles of plagioclase crystals in a fractionating basalt from Philpotts et al. (1998).

Our study builds the necessary theoretical foundation for intuition derived from observational studies (Wadsworth, 1973; Mathews et al., 1964; Moore & Evans, 1967) that crystal settling appears to occur either through a single descending column of crystal-rich liquid (Hess, 1960; Irvine, 1980) or “tear-drop-like masses” (Hess, 1960). Similarly, Sparks et al. (1984) suggested that crystal fractionation occurs in a “wide variety of convective phenomena caused by crystallization” such as in crystal-rich downwellings. By isolating the segregation effects of crystal-melt interactions, we are able to focus our contribution on identifying the physical processes leading to crystal-driven convective flow and to quantify the efficiency of melt-crystal segregation in this regime. By zooming into the crystalline scale process, we are also able to record the possible observational signatures of crystal-driven convection. We compare our results to crystalline data from Holyoke basalt flow, which is hypothesized to record fractional crystallization in crystal core to margin profiles (Philpotts et al., 1998).

2 Collective flow dominates in the Stokes limit.

To understand crystal fractionation in crystal-driven convection, we employ an idealized model set up where the upper boundary layer represents a cooling interface with negatively buoyant crystals suspended above a crystal-free melt body. We assume that the melt phase has constant density and viscosity, which means that the ensuing flow is entirely driven by the crystal-melt buoyancy contrast. Therefore, we are able to isolate crystal fractionation due to crystal-driven convection apart from other potentially confounding factors.

A key control on the degree of collective flow during crystal settling is the relative importance of inertial and viscous forces. This relationship is represented in the non-dimensional

Reynolds number, $Re = a\Delta u\rho_\ell/\mu_\ell$, where Δu is the characteristic crystal-melt segregation speed which we take as the Stokes settling speed of a single crystal; a , g , ρ_ℓ , and μ_ℓ are crystal radius, gravity, melt density, and dynamic melt viscosity, respectively. Another important contribution to collective flow is crystal distribution, which we describe by the non-dimensional number $B = \phi a/l$. The number B combines the complementary scales of the crystal radius, the characteristic crystal spacing, l , and the crystal volume fraction or crystallinity, ϕ (Faroughi & Huber, 2015; Shibano et al., 2012). It expresses how varying the crystal spacing, l , at a given crystallinity, ϕ , will distinguish the contrasting scenarios of either a few large or many more small crystals.

For our analysis, we use a dimensional solver (Qin & Suckale, 2017; Qin et al., 2019), but to facilitate a comparison to other contexts, we non-dimensionalize our equations and results using the parameters provided above. We provide details on the methods and set up in Supp. Sec. 1. More details on the numerical method and benchmarks are available in Suckale et al. (2012b); Qin & Suckale (2017) and Qin et al. (2019).

In Figure 1A–H, we compare two simulations with different liquid viscosity but identical parameters in the solid phase. The higher viscosity could represent crustal-scale melt bodies, while the lower viscosity could be applicable to magma oceans.

Figure 1 shows two simulations at different Re with B held constant. We observe a higher degree of collective flow in the viscously-dominated ($Re < 1$) compared to the inertially-dominated ($Re \geq 1$) regime, which is apparent both from the evolving distribution of crystals, as well as from the vertical melt speed. In the inertial regime (panels A–D), crystals begin settling individually, but over time arrange into settling trains forming as a consequence of low-pressure wakes. In the viscous regime (panels E–H), crystals and melt organize into a broad cluster of crystals and melt settling collectively.

To quantify under what conditions collective flow dominates over individual settling, we compute the mean wavelength of the horizontal distribution of vertical speed, the measured cluster radius, R_m , for a range of Re and B values (see Supp. Sec. 2 for measuring R_m). In Fig. 1I, we show how the measured cluster radius varies with Re and B . We observe a continuous transition from individual crystals or crystal clusters consisting of two or three crystals to the formation of mesoscale clusters that include tens of crystals with increasing Re and B (Fig. 1).

To verify the robustness of our results, we run multiple simulations with randomly placed crystals to obtain a range of initial conditions. We find that the stochasticity and the non-linearity of crystal-melt interactions results in some variation in R_m even at identical Re and B . Nevertheless we find an overall consistent trend, with R_m comparable in size to the crystal radius at finite Re and low B , and R_m on the scale of the domain dimensions at low Re and high B .

We derive a characteristic scale for the cluster radius, R , by taking the ratio of the characteristic rates of viscous diffusion and inertial advection of momentum in Supp. Sec. 2. We show this relation as Fig. 1I background color. The observed trend of increasing cluster size, and hence increasing degree of flow coordination, in our simulations is consistent with our dimensional analysis.

3 Crystal rich clusters lead to efficient fractionation

After characterizing collective settling, we test whether crystals fractionate more or less efficiently in collective compared to individual settling. To quantify the degree of crystal fractionation, we run multiple simulations at the same low Re but variable B numbers.

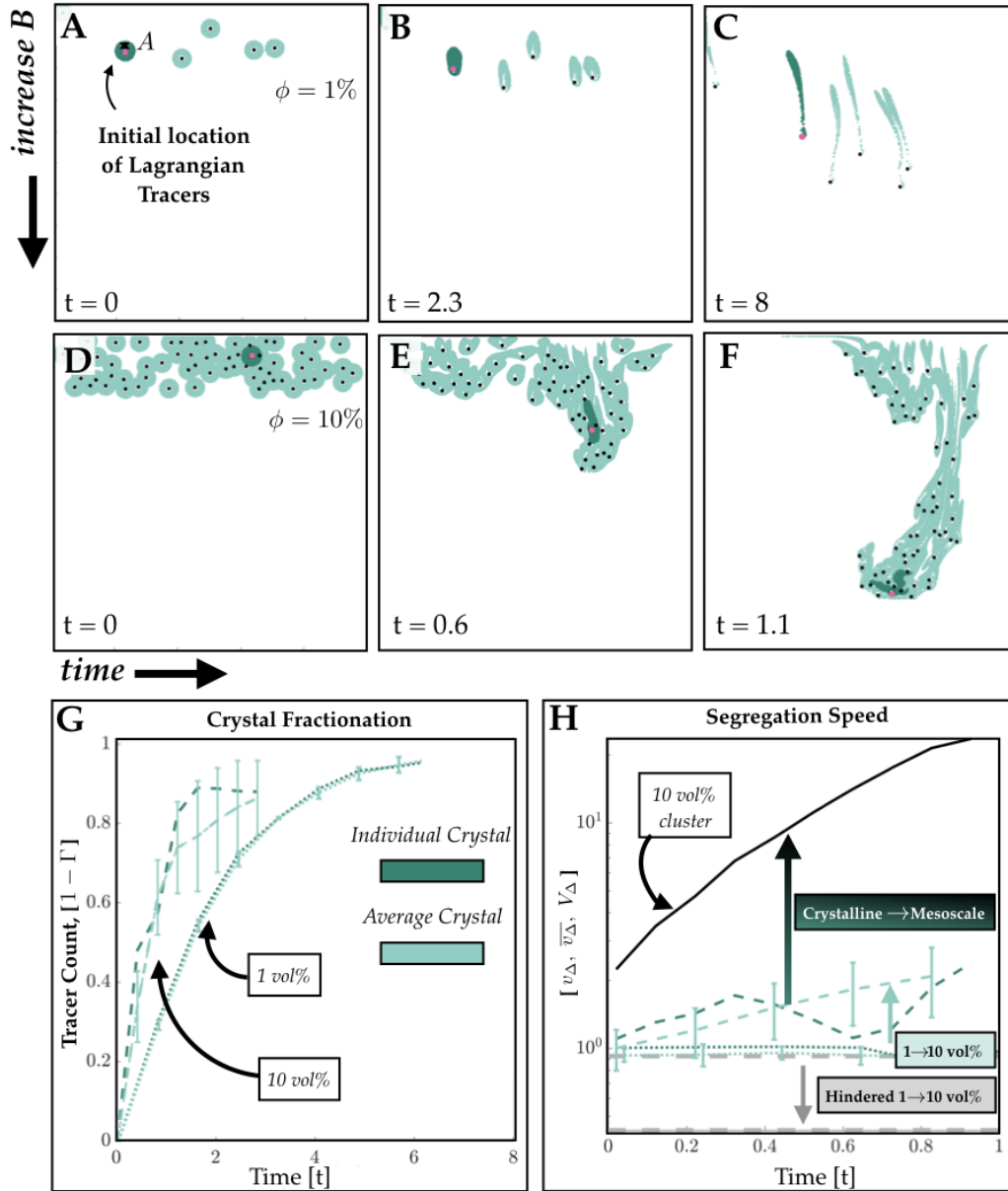


Figure 2. Characterizing crystal fractionation in individual and collective settling.

We show two simulations at the same Re of 4×10^{-6} and different B values of 0.004 (A–C) and 0.4 (D–F). Simulation of crystals segregating from the residual melt (turquoise tracers) at 1 vol% (A–C) and 10 vol% (D–F) crystallinity. We pick a crystal in red and its residual tracers in dark turquoise to track over time. (G) Number of residual tracers in the control volume around the crystals, Γ , to quantify the degree of fractionation, $(1 - \Gamma)$, for the two simulations. The dark turquoise curve highlights the degree of fractionation for the red crystals in (A–F) as a comparison point for the average behavior (light turquoise). (H) Comparison of the segregation speeds. The black line is the cluster speed, identified as the mesoscale segregation speed, V_Δ , from the melt around the cluster. The two gray lines show the hindered-settling speeds at 1 vol% (top line) and 10 vol% (bottom line) crystallinity.

We introduce Lagrangian tracers to track the melt initially surrounding each crystal, which, depending on the element of interest, may be depleted or enriched during crystal growth (turquoise in Fig. 2) as opposed to the background melt unaffected by crystallization (white). We initialize the residual melt tracers in a circular area around the crystal to represent a diffusively depleted rim of radius $A = 3a$ around the crystal (see Supp. Mat. 3 for discussion on A). As crystals settle in the low crystallinity case (1 vol%), individual crystals uniformly strip away the residual melt. Whereas, at 10 vol% crystallinity, crystals fractionate less uniformly.

To quantify the degree of crystal fractionation, we count the number of residual melt tracers that originated around each crystal compared to the total number of tracers within the same control area of radius, A , at each time step. If the ratio of residual tracers to total number of tracers, Γ , in the control area is 1 then no crystal fractionation has occurred. If the ratio is 0, then the crystal is completely stripped of its residual melt, completing crystal fractionation. Therefore, $(1-\Gamma)$ indicates the degree of crystal fractionation for an individual crystal. The average degree of crystal fractionation is the average of this metric over all of the crystals.

Our results show that both individually settling crystals (Fig. 2A–C) and collectively settling crystals (Fig. 2D–F) fractionate from their residual melt. However, clusters generally do not fully reach the same degree of fractionation as individually settling crystals. For the two simulations shown in Fig. 2, the rate of average crystal fractionation, represented by the slopes of curves in Fig. 2 G, is a factor of two faster at 10 vol% as compared to 1 vol% crystallinity. While the precise factor of speedup varies, our full ensemble of simulations with varying A , domain size and initial crystal placement show robustly that the rate of crystal fractionation is comparable to or faster than individual crystal fraction (Supp. Sec. 4).

Next, we quantify the segregation speeds that ultimately control the rate of crystal fractionation in Fig. 2H. We define the individual crystal-melt segregation speed as $v_{\Delta} = |\mathbf{v}_{\Delta}| = |\mathbf{v}_c - \overline{\mathbf{v}}_{\ell}|$, where $\mathbf{v}_c$ and $\mathbf{v}_{\ell}$ are the crystal and melt velocities. We measure the velocity difference between the crystal's center of mass and the average melt velocity within the control area of radius A . In Fig. 2H, the average metric shows a gradual increase in crystal-melt segregation speed with time for 10 vol% crystallinity, whereas the 1 vol% case reaches terminal velocity within a few time steps.

The finding that increasing crystallinity results in faster settling contrasts with the hindered-settling parametrization of average segregation speed. Derived from experiments and theory, hindered-settling implies slower settling speeds at higher crystallinities (e.g., Huppert et al., 1991; Arai & Maruyama, 2017). The crystals within the cluster experience hindered-settling relative to the melt in the cluster, but melt advection outside of the cluster rim increases the segregation speed of crystals along the rim. The average crystal segregation speed is thus greater than suggested by either hindered- or unhindered-settling parametrizations.

4 Crystal scale zoning signatures of convective fractionation

While fractional crystallization is difficult to observe directly in magmatic systems, our models suggest that collective settling imprints subtle, observational clues on crystal clusters (e.g., Wieser et al., 2019b; Schwindinger & Anderson, Jr., 1989) or crystal chains (Philpotts et al., 1998). We observe that each crystal in a collectively settling cluster segregates and thus fractionates at a slightly different rate depending on its location between the cluster center and rim. We first characterize this heterogeneity in individual crystal evolution in dark green lines of Fig. 2 and in more detail in Supp. Sec. 6. We hypothesize that this heterogeneity could be captured in crystalline cross-sectional pro-

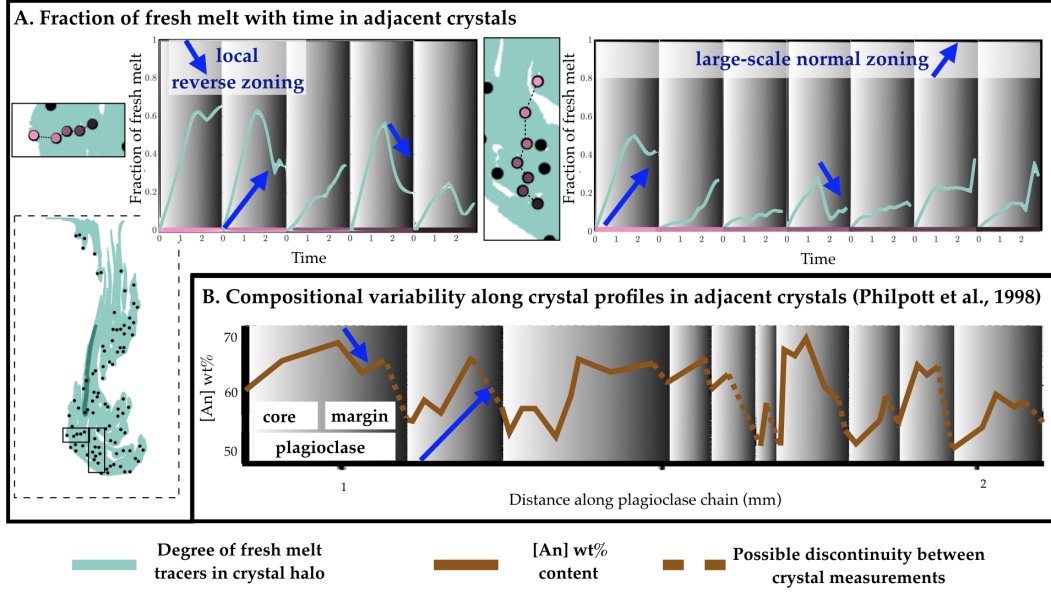


Figure 3. Crystalline scale variability with time and along crystal profile. In (A), we track the number of fresh melt tracers relative to initial number of tracers around each crystal halo through time. Time is from the start of the simulation to the moment of the simulation snapshot shown in the left. We zoom into 2 sets of adjacent crystals, which are identified from light pink to dark pink. We identify a large-scale normal zoning and a small-scale reverse zoning, which are indicated by the blue up and down arrows, respectively. In (B), we zoom into the anorthite content of 10 crystals by Philpotts et al. (1998).

files as crystal zonations, which have been associated with many processes (Wallace & Bergantz, 2005; Longpré et al., 2014; Wolff et al., 2015).

To demonstrate the testability of our model against crystalline-scale data, we compare our results to the 174-m-thick Holyoke basalt flow, which is thought to have experienced fractional crystallization post eruption (Philpotts & Carroll, 1996) with continuous transport of plagioclase crystals from roof to bottom of the flow (Philpotts & Dickson, 2000) at intermediate crystallinity (< 50 vol%; Philpotts et al., 1998). We use the plagioclase crystals in the melt that formed during fractional crystallization (Philpotts et al., 1998) as markers sampling the process.

To quantify the melts that a crystal would sample in our simulations, we track the different types of melt in the system, namely: fresh melt versus evolved melt. We use a complementary description to the analysis in Fig. 2G. Here, all residual melt tracers are identified as evolved melt tracers, $\tilde{\Gamma}$. In Fig. 3A, we plot the fraction of fresh melt tracers, $(1 - \tilde{\Gamma})$, present in the crystal halo for adjacent crystals from the start until the time displayed in the snapshot in Fig. 3A.

We look at the fraction of fresh melt exposure around crystals as it changes with time. A single trajectory of a simulation crystal (first profile in Fig. 3A) shows the crystal losing some of the residual melt and entering into an environment with more fresh melt. Once it reaches about 60 % fresh melt, it gets exposed to more residual melt again before once more entering into an environment with more fresh melt. We observe that (1) on a larger time scale, each crystal gets exposed to greater amounts of fresh melt tracers with time, which would imprint as large-scale normal zoning and (2) on a smaller time scale, each crystal may experience a relative decrease in fresh melt exposure with time,

which would likely imprint as small-scale reverse zoning. Additionally, neighboring crystals may experience different melt histories during collective settling.

Depending on crystal growth rates, crystals may grow or dissolve rims representing the composition of the melt surrounding the crystals (Ruprecht et al., 2008). Crystal growth rates can be as fast as 10^{-6} mm/s (Couch, 2003). At this rate, the small time scale reverse zoning (roughly lasting for 1 dimensionless unit of time) would be preserved in crystalline structure as $10^{-4} - 1$ mm rim for viscosity values of 10^{2-6} Pa·s.

Our results suggest that each crystal may record different sequences of melt environments as it settles through the magma layer with time. We compare the exposure to different melt compositions in our simulation to anorthite content variations in the Holyoke basalt flow plagioclase crystals, where the behavior of large scale normal zoning and small scale reverse zoning in adjacent crystals has been measured (Philpotts et al., 1998). Fig. 3B shows the cross sectional anorthite content of adjacent plagioclase crystals as first shown in Philpotts et al. (1998). The original plot includes 14 crystals with different sizes, obscuring the patterns from crystal core (white) to margin (black). Therefore, we increase the size of 10 crystal profiles to increase visibility of the profiles. Additionally, Philpotts et al. (1998) has the crystal margin connected to the core of the adjacent crystal (brown dashed lines). Since the anorthite content from one crystal margin to another crystal core should be discontinuous, we connect the compositional profiles with brown dashed lines at the last identifiable measurement.

These crystals consistently show a large-scale increase in anorthite content with small zones of decreasing anorthite content. Based on anorthite phase diagrams, batch crystallization could not explain the increase in anorthite content as magma cools. Therefore, increase in anorthite content suggests that the crystals are getting exposed to more mafic magma. The anorthite content is hence analogous to “fresh magma” in our simulations. Although we provide a very simple model where we do not track anorthite content or model the growth of crystals, tracking of residual and fresh melt tracers allows us to capture how crystals can inherit zoning patterns that are comparable to anorthite content patterns captured in Philpotts et al. (1998) by migrating through different domains in a collectively settling clusters and thereby getting exposed to different melt compositions.

5 Efficiency depends on crystallinity and crystal distribution

Reactive processes are essential for describing compositional alterations, but they alone do not explain the change in bulk compositions without crystal fractionation. Therefore, the efficiency of crystal fractionation is significant for quantifying efficiency of fractional crystallization. The efficiency at which crystals collectively strip away from their residual melt is a function of many parameters, but to the first order, it is a function of cluster speed. Additionally, as the cluster sinks, the boundary where crystals initially formed will be replaced by the return flow of the hotter, less evolved magma. As the fresh magma becomes exposed to the cooling boundary layer it will form and grow more crystals, resulting in a self-sustaining convection cell. We use cluster-melt segregation speed as the critical speed to describe the efficiency of collective fractionation.

For the simulations that show collective motion, we measure how fast crystals force magma overturn by measuring the cluster-melt segregation speed, $V_{\Delta} = |\mathbf{V}_{\Delta}| = |\mathbf{v}_{\mathbf{d}} - \overline{\mathbf{v}_{\ell}}|$, where $\mathbf{v}_{\mathbf{d}}$ is the cluster velocity, the mean velocity of the crystals within the cluster area and $\overline{\mathbf{v}_{\ell}}$ is the average melt velocity within $2R_m$ away from cluster center of mass. We approximate the cluster center of mass as the center of mass of the crystals and define the cluster bounds by R_m .

Our analysis suggests that, in the highly simplified case in Fig. 2, a crystal cluster can advect 100 m through melt viscosity of 10^2 Pa·s in 12 days. In contrast, individ-

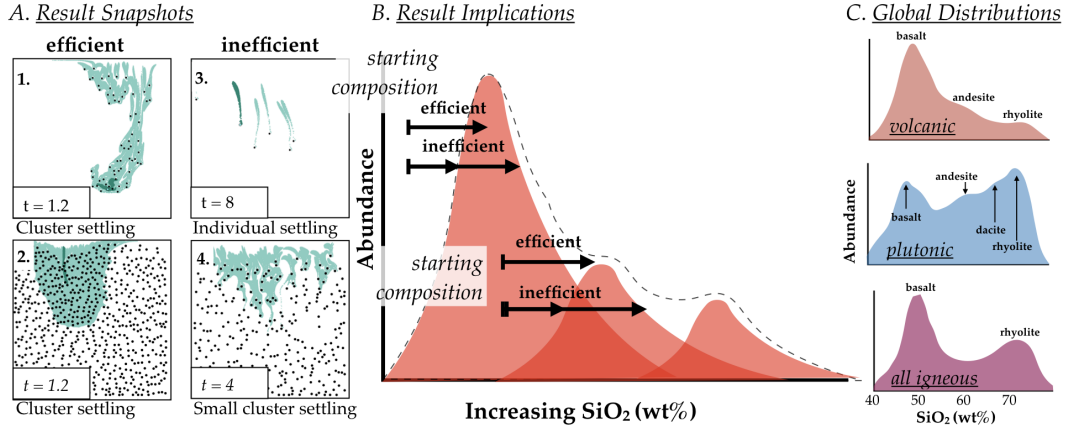


Figure 4. Constant low Re with variable crystallinity and crystal distribution. We show 4 simulation snapshots in (A). We show snapshots with crystallinity difference between top and bottom layers. Simulation (1–3) have 10 over 0, 30 over 20, and 1 over 0 vol% crystallinity, respectively. Sim (4) has 10 vol% crystallinity everywhere. We quantify the efficiency of crystal fractionation in Supp. Sec. 5. In (B), we illustrate the efficiency implications of our results on the compositional abundances as bell curves. The peak composition of the first bell curve would be the new starting composition for further fractional crystallization, forming a new bell curve. Adding these abundance curves could produce the dotted black line. Figure C shows the global distributions of volcanic, plutonic, and all igneous systems as a function of SiO_2 content in arc settings as originally presented in Keller et al. (2015).

ual settling at low crystallinity would cover the same distance in 331 days, resulting in negligible magma overturn, and hence less efficient compositional segregation and fractional crystallization.

We suggest that a hot magma injected into a magma processing zone would initially begin to cool at the boundaries, creating a sharp gradient in crystallinity and efficiently advecting crystal clusters. Our results summarized in Fig. 4A show this efficiency varies depending on crystallinity and crystal distribution. If the magmatic lens reaches equilibrium with its surrounding melt, it will form crystals everywhere (Fig. 44A) or lose most of its crystals to cluster settling (Fig. 43A). It is also possible that the lens has high crystallinity everywhere. Compaction (e.g., Richter & McKenzie, 1984), the crystal fractionation model that best describes high crystallinity regime, would compress melt with viscosity of 10^2 Pa·s 100 m in 600 years, which is orders of magnitude slower than cluster fractionation.

There are many factors that contribute to efficient and inefficient modes of fractional crystallization. In Fig. 4B, we illustrate that the different modes would result in a bell curved bulk composition distribution in large scale data sets. In the figure, we do not provide bulk silica content values on the x-axis because both petrology and crystal fractionation would define the location and distribution of the peak.

We suggest that magmatic lenses from the mantle would fractionate resulting in the first distribution. Volcanic eruptions would sample the initial distribution or stay trapped as new magmatic lenses in disequilibrium. The new lenses would be the starting composition for further fractionation, forming a new bell curve distribution. In the figure, we show each of the bell curves decreasing in abundance with increase in silica content, to tell a simple story that the daughter distributions will be smaller than mother

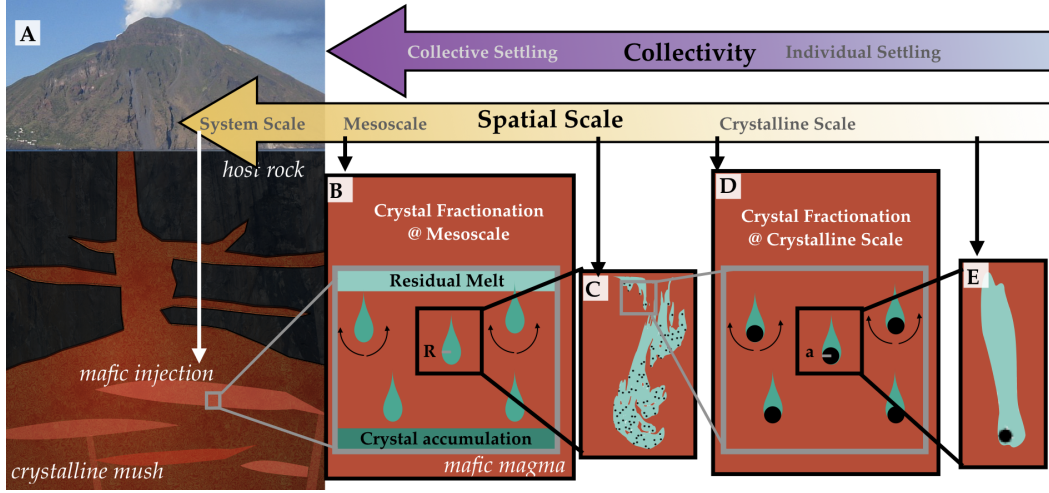


Figure 5. Summary of scales: Zooming into the mafic injection (A), we argue that crystal settling can be conceptualized through mesoscale clusters (B) that lead to efficient but heterogeneous fractionation as demonstrated by our numerical results (C). Further zooming into the cluster would show that melt is stripped from each crystal individually as plotted schematically in (D) and demonstrated in our numerical result in (E) and Fig. 2E-H).

distributions; however, this might not be true if certain areas preferentially have an easier time fractionating than erupting instead of erupting the original distribution. Combining these bell curve histograms would add up to a distribution that resembles the compositional distribution of volcanic systems shown in Fig. 4C. However it does not resemble the plutonic distribution. Plutonic outcrops show vast regions that experienced alteration for millions of years at high crystallinity prior to exposure (Coleman et al., 2004; Deering et al., 2016). The collective settling processes discussed here are hence more pertinent for the volcanic rather than the plutonic context.

6 Conclusions

Large scale convective drivers set the reference frame at which we study crystal cluster settling in trans-crustal mush bodies. Despite differences in dynamics, there is a striking self-similarity between the crystalline-scale and the mesoscale settling. Like a single crystal settling, a cluster strips away the residual melt around itself. In Fig. 5, we provide a conceptual summary of crystal-driven convective fractionation at the mesoscale as understood based on our simulations. The key difference between the two scales is the increased speed at which mesoscale clusters sink through the magma compared to individual crystals. We only study the crystal fractionation component of fractional crystallization. We do not consider reactive processes like melt density differences, which may further enhance the efficiency of fractional crystallization. Outside of crystal resorption and bubble formation, it is reasonable to expect that our estimates for the efficiency of crystal fractionation by collective settling are a lower bound.

Acknowledgments

The authors would like to thank George Bergantz, Shan de Silva, Christy Till, Gail Mahood, Ayla Pamukcu and David Pollard for fruitful discussions throughout the development of the project. CC acknowledges support from the NSF's GRFP and Stanford University's McGee Grant. TK acknowledges support from the Swiss National Science

348 Foundation's Postdoc.Mobility Fellowship 177816. See Supplementary Material and our
349 GitLab repository: `git@zapad.Stanford.EDU:cansu.culha/crystal-fractionation.git` for codes.
350

References

- Arai, T., & Maruyama, S. (2017). Geoscience Frontiers Formation of anorthosite on the Moon through magma ocean fractional crystallization. *Geosci. Front.*, 8(2), 299–308. Retrieved from <http://dx.doi.org/10.1016/j.gsf.2016.11.007> doi: 10.1016/j.gsf.2016.11.007
- Bonnefoi, C. C., Provost, A., & Albarède, F. (1995). The Daly gap' as a magmatic catastrophe. *Nature*, 378(6554), 270–272. doi: 10.1038/378270a0
- Bowen, N. (1928). The evolution of the Igneous Rocks. *Dover Publ.*
- Chayes, F. (1963). Relative abundance of intermediate members of the oceanic basalt-trachyte associatio. *J. Geophys. Res.*, 68(5), 1519–1534.
- Clague, D. A. (1978). The Oceanic Basalt-Trachyte Association : An Explanation of the Daly Gap. *J. Geol.*, 86(6), 739–743.
- Coleman, D. S., Gray, W., & Glazner, A. F. (2004). Rethinking the emplacement and evolution of zoned plutons: Geochronologic evidence for incremental assembly of the Tuolumne Intrusive Suite, California. *Geology*, 32(5), 433–436. doi: 10.1130/G20220.1
- Couch, S. (2003). Experimental investigation of crystallization kinetics in a haplogranite system. *American Mineralogist*, 88(10), 1471–1485.
- Daly, R. A. (1925). The Geology of Ascension Island. *Proc. Am. Acad. Arts Sci.*, 60(1), 3–80.
- Deering, C. D., Keller, B., Schoene, B., Bachmann, O., Beane, R., & Ovtcharova, M. (2016). Zircon record of the plutonic-volcanic connection and protracted rhyolite melt evolution. *Geology*, 44(4), 267–270. doi: 10.1130/G37539.1
- Dufek, J., & Bachmann, O. (2010). Quantum magmatism: Magmatic compositional gaps generated by melt-crystal dynamics. *Geology*, 38(8), 687–690. Retrieved from <http://geology.gsapubs.org/cgi/doi/10.1130/G30831.1> doi: 10.1130/G30831.1
- Faroughi, S. A., & Huber, C. (2015). Unifying the relative hindered velocity in suspensions and emulsions of nondeformable particles. *Geophys. Res. Lett.*, 42(1), 53–59. doi: 10.1002/2014GL062570
- Hess, H. (1960). *Stillwater igneous complex, montana*. The Geologic Society of America Memoir 80.
- Huppert, H. E., Kerr, R. C., Lister, J. R., & Turner, J. S. (1991). Convection and particle entrainment driven by differential sedimentation. *J. Fluid Mech.*, 226, 349–369. doi: 10.1017/S0022112091002410
- Irvine, T. (1980). Magmatic density currents and cumulus processes. *American Journal of Science*, 280(Jackson Volume), 1–58.
- Jackson, M. D., Blundy, J., & Sparks, R. S. (2018). Chemical differentiation, cold storage and remobilization of magma in the Earth's crust. *Nature*, 564(7736), 405–409. Retrieved from <http://dx.doi.org/10.1038/s41586-018-0746-2> doi: 10.1038/s41586-018-0746-2
- Jicha, B. R., Singer, B. S., Beard, B. L., & Johnson, C. M. (2005). Contrasting timescales of crystallization and magma storage beneath the Aleutian Island arc. *Earth Planet. Sci. Lett.*, 236(1-2), 195–210. doi: 10.1016/j.epsl.2005.05.002
- Keller, C. B., Schoene, B., Barboni, M., Samperton, K., Husson, J. M., Hall, G., ... Angeles, L. (2015). Volcanic plutonic parity and the differentiation of the continental crust. *Nature*, 523, 301–307.
- Lee, C. T. A., & Bachmann, O. (2014). How important is the role of crystal fractionation in making intermediate magmas? Insights from Zr and P systematics. *Earth Planet. Sci. Lett.*, 393, 266–274. Retrieved from <http://dx.doi.org/10.1016/j.epsl.2014.02.044> doi: 10.1016/j.epsl.2014.02.044
- Longpré, M.-a., Klügel, A., Diehl, A., & Stix, J. (2014). Mixing in mantle magma reservoirs prior to and during the 2011–2012 eruption at El Hierro, Canary Is-

- lands. *Geology*, 2012–2015. doi: 10.1130/G35165.1
- Mathews, W., Thorarinsson, S., & Church, N. (1964). Gravitative settling of olivine in pillows of an icelandic basalt. *American Journal of Science*, 262(8), 1036–1040.
- Moore, J. G., & Evans, B. W. (1967). The role of olivine in the crystallization of the prehistoric makaopuhi tholeiitic lava lake, hawaii. *Contributions to Mineralogy and Petrology*, 15(3), 202–223.
- Philpotts, A. R., & Carroll, M. (1996). Physical properties of partly melted tholeiitic basalt. *Geology*, 24(11), 1029–1032. doi: 10.1130/0091-7613(1996)024<1029:PPOPMT>2.3.CO;2
- Philpotts, A. R., & Dickson, L. D. (2000). The formation of plagioclase chains during convective transfer in basaltic magma. *Nature*, 406(6791), 59–61. doi: 10.1038/35017542
- Philpotts, A. R., Shi, J., & Brustman, C. (1998). Role of plagioclase crystal chains in the differentiation of partly crystallized basaltic magma. *Nature*, 395(September), 343–346.
- Qin, Z., Alison, K., & Suckale, J. (2019). Rotation matters-direct numerical simulations of rectangular particles in suspensions at low to intermediate solid fraction. *arXiv preprint arXiv:1903.08167*.
- Qin, Z., & Suckale, J. (2017). Direct numerical simulations of gassolidliquid interactions in dilute fluids. *Int. J. Multiph. Flow*, 96(November), 34–47.
- Reubi, O., & Blundy, J. (2009). A dearth of intermediate melts at subduction zone volcanoes and the petrogenesis of arc andesites. *Nature*, 461(7268), 1269–1273. Retrieved from <http://dx.doi.org/10.1038/nature08510> doi: 10.1038/nature08510
- Richter, F. M., & McKenzie, D. (1984). Dynamical models for melt segregation from a deformable matrix. *The Journal of Geology*, 92(6), 729–740. Retrieved from <https://doi.org/10.1086/628908> doi: 10.1086/628908
- Ruprecht, P., Bergantz, G. W., & Dufek, J. (2008). Modeling of gas-driven magmatic overturn: Tracking of phenocryst dispersal and gathering during magma mixing. *Geochemistry, Geophys. Geosystems*, 9(7), 1–25. doi: 10.1029/2008GC002022
- Schwindinger, K. R., & Anderson, Jr., A. T. (1989). Synneusis of Kilauea Iki olivines. *Contrib. Miner. Pet.*, 103, 187–198.
- Shibano, Y., Namiki, A., & Sumita, I. (2012). Experiments on upward migration of a liquid-rich layer in a granular medium: Implications for a crystalline magma chamber. *Geochemistry, Geophys. Geosystems*, 13(3). doi: 10.1029/2011GC003994
- Sparks, R. S. J., Huppert, H. E., Turner, J. S., Sakuyama, M., & O'Hara, M. J. (1984). The Fluid Dynamics of Evolving Magma Chambers [and Discussion]. *Philos. Trans. R. Soc. A Math. Phys. Eng. Sci.*, 310(1514), 511–534. Retrieved from <http://rsta.royalsocietypublishing.org/cgi/doi/10.1098/rsta.1984.0006> doi: 10.1098/rsta.1984.0006
- Suckale, J., Elkins-Tanton, L. T., & Sethian, J. A. (2012b). Crystals stirred up: 2. numerical insights into the formation of the earliest crust on the moon. *Journal of Geophysical Research: Planets*, 117(E8).
- Suckale, J., Sethian, J., Yu, J., & Elkins-Tanton, L. (2012a). Crystals stirred up: 1. direct numerical simulations of crystal settling in nondilute magmatic suspensions. *Journal of Geophysical Research: Atmospheres*, 117(8). doi: 10.1029/2012JE004066
- Vigneresse, J. L., Barbey, P., & Cuney, M. (1996). Rheological transitions during partial melting and crystallization with application to felsic magma segregation and transfer. *J. Petrol.*, 37(6), 1579–1600. doi: 10.1093/petrology/37.6.1579
- Wadsworth, J. (1973). Magmatic sediments. *Mineral, Sci. Engng.*, 5, 25–35.

- 458 Wallace, G. S., & Bergantz, G. W. (2005). Reconciling heterogeneity in crys-
 459 tal zoning data: An application of shared characteristic diagrams at Chaos
 460 Crag, Lassen Volcanic Center, California. *Contrib. to Mineral. Petrol.* doi:
 461 10.1007/s00410-004-0639-2
- 462 Wieser, P. E., Vukmanovic, Z., Kilian, R., Ringe, E., Holness, M. B., MacLennan, J.,
 463 & Edmonds, M. (2019a). To sink, swim, twin, or nucleate: A critical appraisal of
 464 crystal aggregation processes. *Geology*, 47(10), 948–952. doi: 10.1130/g46660.1
- 465 Wieser, P. E., Vukmanovic, Z., Kilian, R., Ringe, E., Holness, M. B., MacLen-
 466 nan, J., & Edmonds, M. (2019b). To sink, swim, twin, or nucleate: A crit-
 467 ical appraisal of crystal aggregation processes. *Geology*, 47(Xx), 1–5. Re-
 468 trieved from [https://pubs.geoscienceworld.org/gsa/geology/article/](https://pubs.geoscienceworld.org/gsa/geology/article/573164/To-sink-swim-twin-or-nucleate-A-critical-appraisal)
 469 573164/To-sink-swim-twin-or-nucleate-A-critical-appraisal doi:
 470 10.1130/G46660.1
- 471 Wolff, J. A., Ellis, B. S., Ramos, F. C., Starkel, W. A., Borroughs, S., Olin, P. H., &
 472 Bachmann, O. (2015). Remelting of cumulates as a process for producing chemical
 473 zoning in silicic tuffs: A comparison of cool, wet and hot, dry rhyolitic magma
 474 systems. *Lithos*, 236–237, 275–286. Retrieved from [http://dx.doi.org/10.1016/](http://dx.doi.org/10.1016/j.lithos.2015.09.002)
 475 [j.lithos.2015.09.002](http://dx.doi.org/10.1016/j.lithos.2015.09.002) doi: 10.1016/j.lithos.2015.09.002

Supporting Information for “Crystal fractionation by crystal-driven convection ”

Cansu Culha^{1*}, Jenny Suckale¹, Tobias Keller², Zhipeng Qin¹

¹Stanford University, Department of Geophysics, 397 Panama Mall, Stanford CA 94305, USA.

²University of Glasgow, School of Earth and Geographical Sciences, 8NN University Avenue, Glasgow
G12 8QQ, UK.

Contents of this file

1. Text Sec. 1 to Sec. 7
2. Supp. Figures 1 to S6
3. Supp. Tables S1 to S2

Additional Supporting Information (Files uploaded separately)

1. Stokes Solver Fortran Code
2. Navier-Stokes Solver Fortran Code

Introduction

In this supplementary material we provide details regarding our nondimensional analysis and show additional simulation data. The codes are provided in our GitLab repository at this link: git@zapad.Stanford.EDU:cansu.culha/crystal-fractionation.git

In the manuscript and through out this supplementary material, we use a set of variables which are summarized in Table. 1 for the readers convenience. We note that all of the values presented in the manuscript are dimensionless, but here we differentiate dimensionless variables from dimensional by including $[\cdot]'$ to the variable, which is absent in the manuscript. We provide the methods section in Supp. Sec. 1, which includes the caveats of non-dimensionalization of our results and of using 2D experiments to describe 3D processes. A summary of all of all of the simulations are shown in Supp Table 2. Additionally, we explain how we calculate the cluster size (Supp. Sec. 2), the impact of the diffusive rim area on quantifying crystal segregation and fractionation (Supp. Sec. 3), the impact of domain size and initial randomness on cluster properties (Supp. Sec. 4), variability in efficiency (Supp. Sec. 5), and variability in crystal populations (Supp. Sec. 6).

1 Methods

To understand crystal fractionation in crystal-driven convection, we employ an idealized model setup where the upper boundary layer represents a cooling interface with negatively buoyant crystals suspended above a crystal-free melt body. We assume that the melt phase has constant density and viscosity, which means that the ensuing flow is entirely driven by the crystal-melt buoyancy contrast. Therefore, we are able to isolate crystal fractionation due to crystal-driven convection apart from other potentially confounding factors.

In the melt phase, we solve the incompressible Navier-Stokes equations,

*Stanford, CA, USA

Corresponding author: Cansu Culha, cculha@stanford.edu

Parameter	Variable
time	t
length	L
velocity	$\mathbf{v}$
pressure	P
gravity	g
force	F
location	X
mass	M
moment of inertia	I
angular momentum	ω
torque	T
viscosity	μ
density	ρ
segregation speed of 1 crystal	Δu
segregation speed	v_{Δ}
average segregation speed	$\overline{v_{\Delta}}$
cluster-melt segregation speed	V_{Δ}
av. spacing between crystals	l
crystallinity	ϕ
crystal radius	a
control area	A
area	Ω
crystal rich layer	Ω_T
volume	Ψ
crystal rich layer	Ψ_T
cluster size	R
Reynolds number	Re
Froude number	Fr
particle distribution	B
speed correction	$n = 1.8$
horizontal direction	$[\cdot]_x$
vertical direction	$[\cdot]_y$
of crystal	$[\cdot]_c$
of melt	$[\cdot]_{\ell}$
of cluster	$[\cdot]_d$
of tracers	$[\cdot]_t$
measured in simulation	$[\cdot]_m$
modeled	$[\cdot]_s$
mixture of crystal and melt	$[\cdot]_{cl}$
dimensionless ⁺	$[\cdot]'$ ⁺

Table 1. ⁺ only applies for supplementary material

$$\nabla \cdot \mathbf{v}'_\ell = 0 \quad (1)$$

$$\text{Re} \frac{D\mathbf{v}'_\ell}{Dt'} = -\nabla P' + \nabla^2 \mathbf{v}'_\ell + \frac{1}{\text{Fr}} \hat{\mathbf{y}}' + \mathbf{F}'_c \quad (2)$$

where $\mathbf{v}'_\ell$ is the non-dimensional velocity of the liquid, P' is the local non-dimensional pressure field, $\hat{\mathbf{y}}'$ is the unit vector parallel to gravity, and t' is non-dimensional time. $\mathbf{F}'_c$ is the non-dimensional force exerted by the crystals on the liquid. $\frac{D}{Dt'} = \frac{\partial}{\partial t'} + \mathbf{v}'_\ell \cdot \nabla$ is the material derivative. We assume that the melt is Newtonian and the non-linear interactions result from the presence of crystals. We describe the crystals as rigid bodies and they obey Newton's laws. For further detail on the approach, benchmarks for low Re number and high Re number, see Qin & Suckale (2017); Qin et al. (2019).

In order to isolate the mechanical aspect of crystal fractionation, all simulations are isothermal and isochemical, and we neglect crystal nucleation, growth, and dissolution. Throughout each simulation, we maintain a constant number of crystals in the domain. The simulations are performed on a 2D domain tens to hundreds of crystal radii wide, and with free-slip boundary conditions on the walls. The parameters for all simulations performed are summarized in Supplementary Table 1 and a summary of the non-dimensionalization relations are summarized in Supp. Sec. 2.

We assume that particles are rigid bodies and each crystal obeys Newton's laws:

$$M_c \frac{d\mathbf{V}_c}{dt} = \tilde{\mathbf{F}}_c + M_c \mathbf{g} \quad (3)$$

$$\frac{d(\mathbf{I}_c \cdot \boldsymbol{\omega}_c)}{dt} = \mathbf{T}_c \quad (4)$$

$$\frac{d\mathbf{X}_c}{dt} = \mathbf{V}_c \quad (5)$$

where c defines an individual crystal, M_c is the mass of an individual crystal, $\mathbf{V}_c$ is the crystal velocity at center of mass, $\mathbf{X}_c$ is the position at center of mass, $\mathbf{I}_c$ is the particle's moment of inertia tensor, and $\boldsymbol{\omega}_c$ is the angular velocity of the crystal. $\tilde{\mathbf{F}}_c$ and $\mathbf{T}_c$ are the hydrodynamic force and torque resulting from the surrounding fluid.

1.1 Nondimensionalization

We introduce dimensionless numbers to characterize the basic physical scales in the problem and present results in that framework for ease of comparison. In this section, we explain how we introduce a correction factor, n , to address the discrepancy in using 3D parameters to non-dimensionalize 2D simulation results. We describe the Stokes speed of a single crystal as the crystal-melt segregation speed and use it as the characteristic speed in our particle Reynolds number, Re. We model the Stokes settling speed as $\frac{2(\rho_c - \rho_\ell)ga^2}{9\mu}$ and hence define the crystal-melt segregation speed as

$$\Delta u_s = \frac{2(\rho_c - \rho_\ell)ga^2}{9\mu}, \quad (6)$$

where ρ_c and ρ_ℓ are crystal and liquid densities, a is crystal radius, and μ the liquid viscosity. This expression for the Stokes speed assumes a 3D flow field, but our simulations are only 2D. Ignoring this inconsistency would lead to a misrepresentation of Re. We hence re-scale the crystal-melt segregation speed, Δu , by our simulation results. We model a single crystal randomly placed in the domain and measure the segregation speed as described in the Manuscript v_Δ . We use the ratio of the stokes speed as shown in eq. 6 and our simulation speed of a single crystal to obtain a ratio, $\frac{\Delta u_s}{\Delta u_m} = n$ where Δu_m is the simulation result. The table below reports 3 simulations in $n = \frac{\Delta u_s}{\Delta u_m}$.

a[m]	μ [Pa·s]	Δu_s eq. 6[m/s]	Δu_m stokes simulation[m/s]	n
0.001	95	1.4488e-5	8.0692e-6	1.8
0.003	95	1.3039e-4	7.6076e-5	1.7
0.001	10	1.3764e-4	6.3561e-5	2.2

For simplicity, we scale all of our simulations by the same segregation speed correction factor and choose $n = 1.8$. Using this speed correction, we update our non-dimensional relationships,

$$L' = \frac{1}{a}L \quad (7a)$$

$$t' = \frac{\Delta u_m}{a}t = \frac{\Delta u_s}{na}t \quad (7b)$$

$$\mathbf{v}' = \frac{1}{\Delta u_m}\mathbf{v} = \frac{n}{\Delta u_s}\mathbf{v} \quad (7c)$$

$$P' = \frac{\Delta u_m}{\mu a}P = \frac{\Delta u_s}{n\mu a}P \quad (7d)$$

$$\text{Re} = \frac{\rho_\ell \Delta u_m a}{\mu} = \frac{\rho_\ell \Delta u_s a}{n\mu} \quad (7e)$$

$$\text{Fr} = \frac{\Delta u_m^2}{ag} = \frac{\Delta u_s^2}{agn^2} \quad (7f)$$

We use the non-dimensional number, B, to characterize the particle distribution. Our definition of B is $B = \frac{\phi a}{l}$, as stated in the paper, where l indicates the average crystal spacing,

$$l = \frac{(\Psi_T - N\frac{4}{3}\pi a^3)^{1/3}}{N} \quad (8)$$

$$= \frac{\Psi_c(\Psi_T(1 - \phi))^{1/3}}{\Psi_T\phi} \quad (9)$$

where Ψ_T , Ψ_c , and N are the total crystal rich fluid volume, individual crystal volume, and the number of crystals, respectively for 3D and

$$l = \frac{(\Omega_T - N\pi a^2)^{1/2}}{N} \quad (10)$$

$$= \frac{\Omega_c(\Omega_T(1 - \phi))^{1/2}}{\Omega_T\phi} \quad (11)$$

where Ω_T and Ω_c are the total crystal rich fluid area and individual crystal area, respectively for 2D .

Our numerical set up for all of the simulations is dimensional and keeps liquid and crystal density constant at 2360kg/m³ and 3000 kg/m³, respectively. Gravity is 9.8 m/s² for all of the simulations. We vary domain size, crystal size, viscosity, and crystal volume fraction to test different nondimensional regimes.

1.2 Implementing Passive Tracers

In order to track the liquid phase and identify the difference between residual melt and ambient melt, we add Lagrangian tracers into the domain. Unlike the crystals, which obey Newton's Laws of Rigid Body motion, tracers do not have mass and volume. They only track the flow field. These tracers are initially placed in a uniform spacing throughout the domain; then are randomly adjusted in space up to a distance that is half way between two tracers. By introducing a component of randomness, we hope

ϕ [%]	a [m]	μ [Pa·s]	l [m]	Re	B	$2R_m$ [m]	$2R'_m$	$\overline{v_{\Delta x}}$ [m/s]	$\overline{v_{\Delta y}}$ [m/s]	$V_{\Delta x}$ [m/s]	$V_{\Delta y}$ [m/s]	$\overline{v_{\Delta}}$ [m/s]	V_{Δ} [m/s]
5.0E-2	1.0E-3	95	1.4E-3	2.5E-7	3.7E-2	7.5E-3	7.5	1.0E-5	6.1E-6	9.8E-5	1.2E-4	8.0E-6	9.8E-5
5.0E-2	1.0E-3	95	1.4E-3	2.5E-7	3.7E-2	8.8E-3	8.8	9.2E-6	4.0E-6	2.6E-5	2.2E-5	6.5E-6	2.0E-5
5.0E-2	1.0E-3	95	1.4E-3	2.5E-7	3.7E-2	7.3E-3	7.3	9.7E-6	4.1E-6	6.8E-5	3.6E-5	6.8E-6	5.1E-5
5.0E-2	1.0E-3	1.0	1.4E-3	2.3E-3	3.7E-2	1.6E-2	16						
5.0E-2	1.0E-3	1.0	1.4E-3	2.3E-3	3.7E-2	9.7E-3	9.7	1.2E-3	4.0E-4	7.2E-3	3.5E-3	8.0E-4	5.3E-3
5.0E-2	2.0E-3	1.0E-1	5.5E-3	1.8	1.8E-2	7.5E-3	3.8	2.1E-2	8.2E-3			1.4E-2	
5.0E-2	2.0E-3	95	5.5E-3	2.0E-6	1.8E-2	1.9E-2	9.3	1.0E-4	3.4E-5	1.8E-4	6.0E-5	6.6E-5	1.1E-4
5.0E-2	2.5E-3	95	8.6E-3	4.0E-6	1.5E-2	1.4E-2	5.6	2.1E-4	4.5E-5	1.1E-4	4.7E-5	1.2E-4	7.1E-5
5.0E-2	2.5E-3	95	8.6E-3	4.0E-6	1.5E-2	1.3E-2	5.4	9.6E-5	2.3E-5	1.6E-4	5.4E-5	5.9E-5	1.0E-4
5.0E-2	2.5E-3	95	8.6E-3	4.0E-6	1.5E-2	1.4E-2	5.4	1.2E-4	2.5E-5	1.3E-4	5.7E-5	7.3E-5	9.5E-5
5.0E-2	2.5E-3	95	8.6E-3	4.0E-6	1.5E-2	1.4E-2	5.6	2.1E-4	4.5E-5	1.1E-4	4.7E-5	1.2E-4	7.1E-5
5.0E-2	4.5E-4	95	2.8E-4	2.3E-8	8.1E-2	1.1E-2	25	3.5E-6	1.6E-6	3.4E-5	3.0E-5	2.5E-6	3.2E-5
1.0E-1	1.0E-3	1.0E-1	6.7E-4	2.3E-1	1.5E-1	3.4E-3	3.4	2.4E-2	1.1E-2	3.9E-2	2.3E-2	1.7E-2	3.1E-2
1.0E-1	1.0E-3	1.0E-1	6.7E-4	2.3E-1	1.5E-1	6.3E-3	6.3	1.4E-2	7.5E-3	9.0E-2	9.2E-2	1.0E-2	8.2E-2
1.0E-1	1.0E-3	1.0E-2	6.7E-4	23	1.5E-1	7.9E-3	7.9	4.8E-3	3.2E-3	8.3E-3	5.1E-3	4.0E-3	6.7E-3
1.0E-1	1.0E-3	1.0E-2	6.7E-4	23	1.5E-1	3.2E-3	3.2	2.2E-2	9.6E-3	3.2E-2	8.5E-3	1.6E-2	2.0E-2
1.0E-1	1.0E-3	95	6.7E-4	2.5E-7	1.5E-1	1.6E-2	16	2.6E-5	1.7E-5	1.2E-4	8.6E-5	2.2E-5	1.0E-4
1.0E-1	1.0E-3	95	6.7E-4	2.5E-7	1.5E-1	2.1E-2	21	1.6E-5	9.0E-6	1.5E-4	1.5E-4	1.2E-5	1.5E-4
1.0E-1	1.0E-3	95	6.7E-4	2.5E-7	1.5E-1	1.4E-2	14	2.2E-5	1.4E-5	1.0E-4	8.2E-5	1.7E-5	9.1E-5
2.0E-1	1.0E-3	95	3.1E-4	2.5E-7	6.4E-1	1.6E-2	16	3.3E-5	2.6E-5	2.6E-4	2.1E-4	3.0E-5	2.3E-4
2.2E-1	1.0E-3	1.0	2.8E-4	2.3E-3	7.8E-1	8.2E-3	8.2	1.7E-3	1.1E-3	6.6E-3	3.7E-3	1.4E-3	5.0E-3
2.2E-1	1.0E-3	95	2.8E-4	2.5E-7	7.8E-1	1.9E-2	19	4.5E-5	2.5E-5	2.8E-4	1.6E-4	3.4E-5	2.0E-4
2.2E-1	2.0E-3	1.0E-1	1.1E-3	1.8	3.9E-1	8.3E-3	4.2	2.2E-2	1.3E-2	5.2E-2	2.6E-2	1.7E-2	3.2E-2
2.2E-1	1.0E-3	1.0E-2	2.8E-4	23	7.8E-1	5.2E-3	5.2	2.3E-2	1.1E-2	3.2E-2	1.5E-2	1.6E-2	2.1E-2
2.2E-1	2.0E-3	10	1.1E-3	1.8E-4	3.9E-1	2.3E-2	11	1.1E-4	6.9E-5	5.0E-4	2.1E-4	8.4E-5	3.3E-4
2.2E-1	2.0E-3	95	1.1E-3	2.0E-6	3.9E-1	2.5E-2	12	1.0E-4	4.8E-5	5.4E-4	2.1E-4	7.2E-5	3.6E-4
2.5E-1	1.0E-3	95	2.4E-4	2.5E-7	1.0	1.9E-2	19	5.1E-5	3.8E-5	2.3E-4	1.7E-4	4.3E-5	1.9E-4
2.5E-1	1.0E-3	95	2.4E-4	2.5E-7	1.0	2.7E-2	27	4.3E-5	3.3E-5	4.8E-4	2.9E-4	3.8E-5	3.7E-4
2.5E-1	1.0E-3	10	2.4E-4	2.3E-5	1.0	2.4E-2	24	2.3E-4	1.6E-4	1.9E-3	1.2E-3	1.9E-4	1.6E-3
1.0E-1	1.0E-3	95	6.7E-4	2.5E-7	1.5E-1	1.2E-2	12	1.6E-5	9.0E-6	5.5E-5	3.4E-5	1.2E-5	4.5E-5
1.0E-1	1.0E-3	95	6.7E-4	2.5E-7	1.5E-1	1.5E-2	15	2.4E-5	1.6E-5	2.8E-4	1.4E-4	1.9E-5	1.8E-4
1.0E-1	1.0E-3	95	6.7E-4	2.5E-7	1.5E-1	2.3E-2	23						

Table 2.

- CS-crystalline scale
- MS-mesoscale

to minimize the effects of tracer clustering and gap formation with time. At each time step, these tracers advect to a new location, $\mathbf{X}_t^{new}$ based on the liquid velocity interpolated to coincide with their original location, $\mathbf{X}_t^{old}$.

$$\mathbf{X}_t^{new} = \mathbf{X}_t^{old} + \mathbf{v}_l * dt \quad (12)$$

1.3 2D versus 3D

In summary, our set of results are conducted as 2D experiments, when the natural process is 3D. The general description of fractional crystallization is consistent between the dimensions. The formation of crystal-rich clusters during settling is consistent with analog experiments in 3D (Michioka & Sumita, 2005). Similarly, the crystal clusters come in different sizes and the crystal clusters sizes are independent of the domain size (Michioka & Sumita, 2005), which is a characteristic often associated with Rayleigh-Taylor Instability. However, quantifying the transition from 2D to 3D of the process is nontrivial. For example, 2D volume fraction of crystals is not the same as 3D volume fraction of crystals. Although we approximate a linear correction factor n , segregation speeds and, hence, the rate of crystal fractionation might not translate linearly. Therefore, these results should be taken to describe the general process. Crystal clusters at low to intermediate crystallinity dominate the terrestrial magmatic systems. They result in crystal fractionation that is more efficient than hindered settling but the efficiency decreases under certain conditions. Crystal clusters can lead to crystals sampling different melt environments. Depending on the dynamic nature of the crystals, neighboring crystals may have different compositional content and profiles.

2 Calculating cluster size

We want to be able to describe when crystals settle collectively versus individually. In order to identify whether crystals are in collective units, we look at the horizontal cross sectional size of the negative vertical velocity and compute a wavelength that is scaled by crystal size. In a crystal cluster, the center of mass of particles does not necessarily define the convective head. We therefore take a horizontal cross section at the crystal center of mass, at 0.1 domain lengths below and 0.1 domain lengths above the crystal center of mass. We collect the dominant widths for the entire time the particles are advecting in the center half of the domain. Then we average over all the widths that are greater than the crystal radius to compute $R'_m = R_m/a$. These values appear as filled circles in Fig. 1I of the Manuscript.

In order to generalize the system and depict the transition of individual to collective settling behavior, we developed a scaling relationship dependent on Re and B . As suggested by our cluster size measurements, the cluster size increases with increase in long range interactions, which is dependent on viscous forces (at low Re) and B . We hypothesize that the cluster formation occurs at the balance of particle speed, which is set by cluster speed, and speed at which long range interactions communicate. We identify the long range interaction speed as $\frac{\eta_{cl} B}{R}$, where $\eta = \frac{\mu}{\rho_{cl}}$ is the kinematic effective viscosity as a function of ϕ and R is the suitably defined size of the cluster. We generalize the Stokes settling speed to clusters to approximate cluster speed.

$$\Delta u_{sd} = \frac{2(\rho_{cl} - \rho_\ell)gR^2}{9\mu} \quad (13)$$

where ρ_{cl} is the mixture of the crystal and melt density, μ is the mixture viscosity which is dependent on crystallinity of the bottom melt and R is a suitably defined size of the cluster. We set the two speeds equal to one another,

$$\frac{\frac{\mu}{\rho_{cl}} B}{R} = \frac{2(\rho_{cl} - \rho_\ell)gR^2}{9n\mu} \quad (14)$$

solving for R , we obtain the dimensional form,

$$R = \left(n \frac{9\mu^2 B}{2\rho_{cl}\Delta\rho_{cl}g} \right)^{\frac{1}{3}}. \quad (15)$$

The plotted relationship in the Manuscript, Fig. 1, is the non-dimensionalized R ,

$$R' = \frac{1}{a} \left(n \frac{9\mu^2 B}{2\rho_{cl}\Delta\rho_{cl}g} \right)^{\frac{1}{3}}. \quad (16)$$

This relationship approximates the pattern of decrease in cluster size with decrease in Re and B . We show the results from this analysis as the background color of Fig. 1I of the Manuscript.

3 Impact of the diffusive rim area on crystal segregation and fractionation

Our approach in measuring segregation speed and crystal fractionation uses a diffusive rim control area of radius A around the crystal. We test the sensitivity of A on crystal fractionation and crystal segregation in Supp. Fig. 1.

In Supp. Fig. 1A–B, we show that the choice of A uniformly alters the degree of fractionation. However, the non-linear effects of crystal-melt interactions result in a slight increase in segregation speed with increase in A for cluster-forming simulations (Supp. Fig. 1C–D). The A we use provides a lower segregation speed than the larger A s we could have picked from such that we can be conservative with our findings.

4 Impact of domain size and initial randomness on cluster properties

Two immiscible fluids that are unstably stratified in density was first proven to have a convective instability by Rayleigh (1883) and Sir Geoffrey (F.R.S.) (1950). The size of the diapiric instability depends on domain size. Since the introduction of Rayleigh-Taylor, many multiphase fluid systems were identified as an instability reminiscent of the original instability. Since our system also includes unstable density stratification, we test the dependence of cluster size on domain size. We find that our results are independent of domain size (Supp. Fig. 2). However, we do find that the randomness of the initial crystal placements results in variable number of clusters and variable cluster sizes as captured by Fig. 1I in the Manuscript. We provide 4 simulations to illustrate this variability in Supp. Fig. 3.

5 Efficiency of crystal fractionation is dependent on crystallinity

In the Manuscript, we notice that there is an increase in crystal-melt and cluster-melt segregation speed with increase in B . Our results show that at a constant Re , increasing B has a positive correlation with increase in segregation speeds. Supp. Fig. 4 summarizes this finding where we plot each of the simulations as a dark spot to indicate cluster-melt segregation and an open circle as crystal-melt segregation. Because the simulations are all at a constant Re , we only vary crystallinity in the top domain in these simulations. There are multiple segregation speeds for each B because each simulation had a different cluster form. The enhanced crystal-melt segregation speeds compared to single crystal Stokes sinking speed may lead to comparable or higher crystal fractionation rates. The formation of the quickly sinking cluster forces the outer rim crystals to also quickly segregate from their surrounding melt compared to individually settling crystals.

However, the efficiency of crystal fractionation decreases with increasing overall crystallinity and removing the crystal gradient. We show the limits of crystal cluster

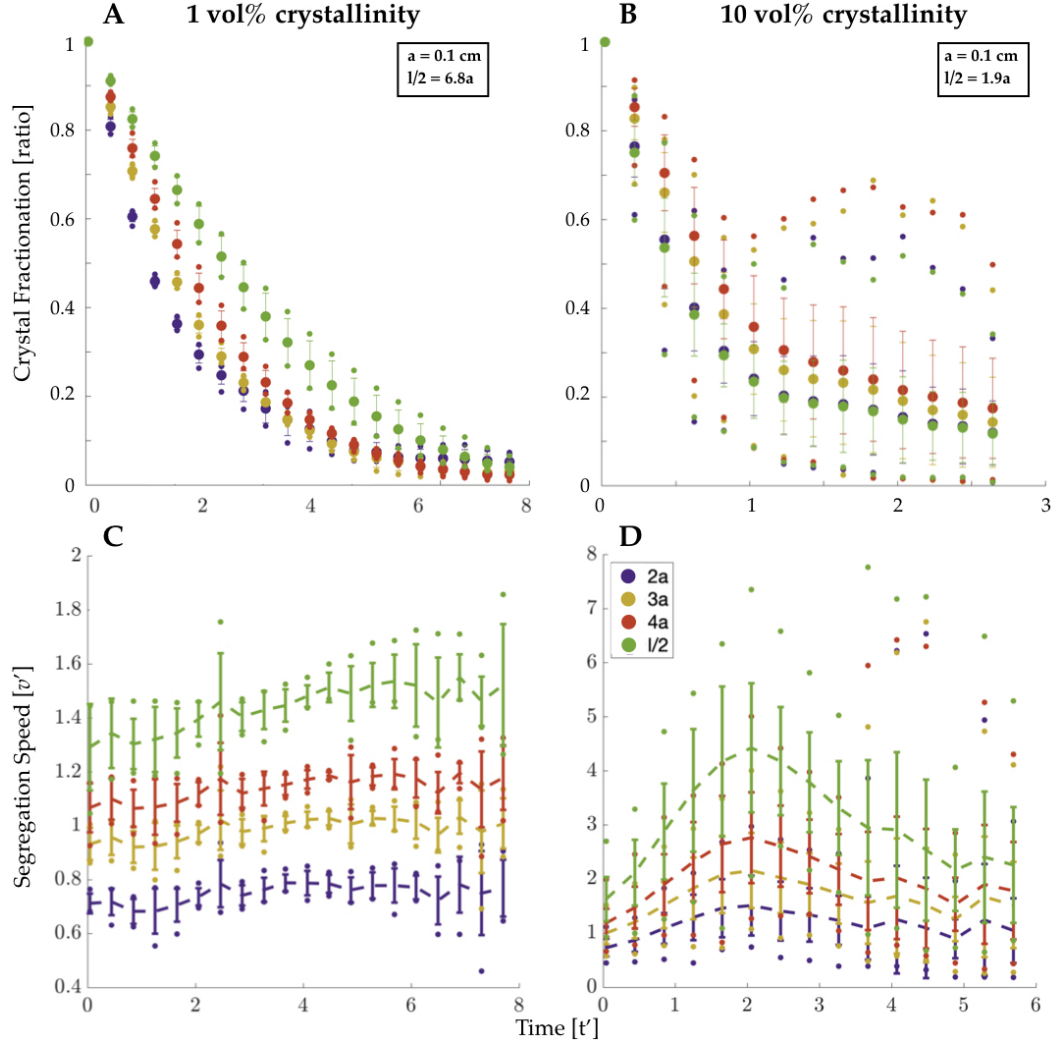


Figure 1. (A–B) The dependence of crystal fractionation on A as defined in the Manuscript. (C–D) The dependence of crystal-liquid segregation speed on A as defined in the Manuscript. (A & C) are for 1 vol% crystallinity whereas (B & D) are for 10 vol% crystallinity.

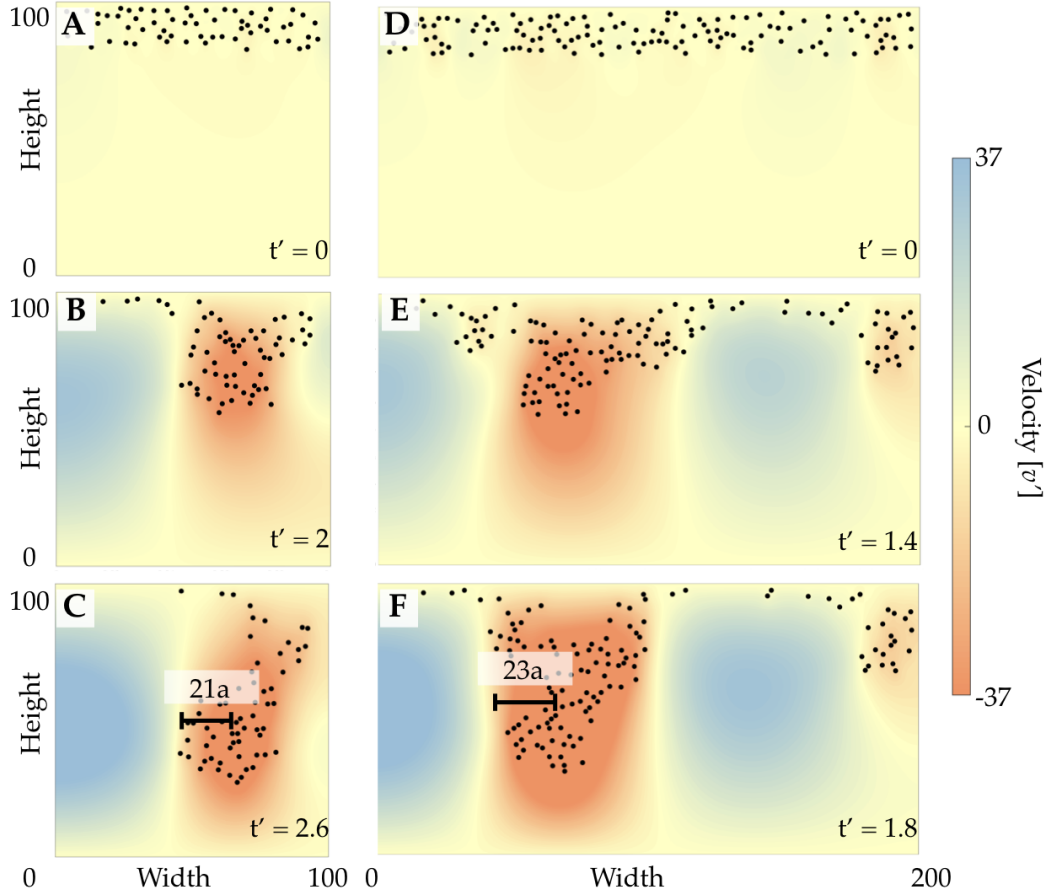


Figure 2. Testing sensitivity of cluster size, R' , to width of the domain. These are both at 10 vol% crystallinity with same viscous liquid properties.

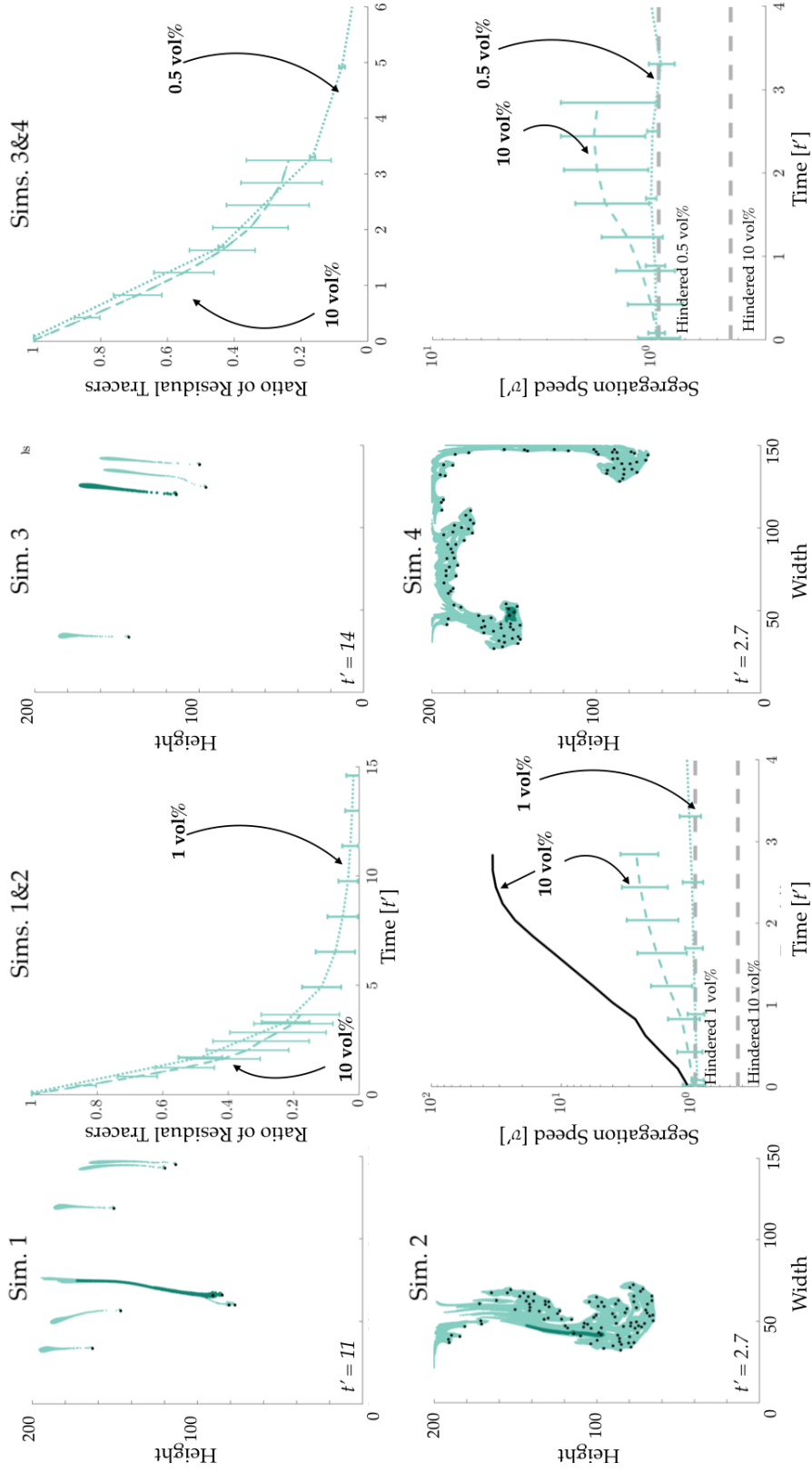


Figure 3. Extra simulation runs. The figure is set up to mimic Fig. 2 in the Manuscript. Simulation 1 and 3 are at 1 and 0.5 vol% crystallinity. Simulation 2 and 4 are at 10 vol% crystallinity. Simulation 2 is the same as Fig. 3 in the Manuscript. We show the segregation speeds and crystal fractionation rates Γ . We only show cluster-melt segregation speed for Simulation 2. Additionally, for figure simplicity, we do not show the individual crystal-melt segregation and crystal fractionation in dark turquoise.

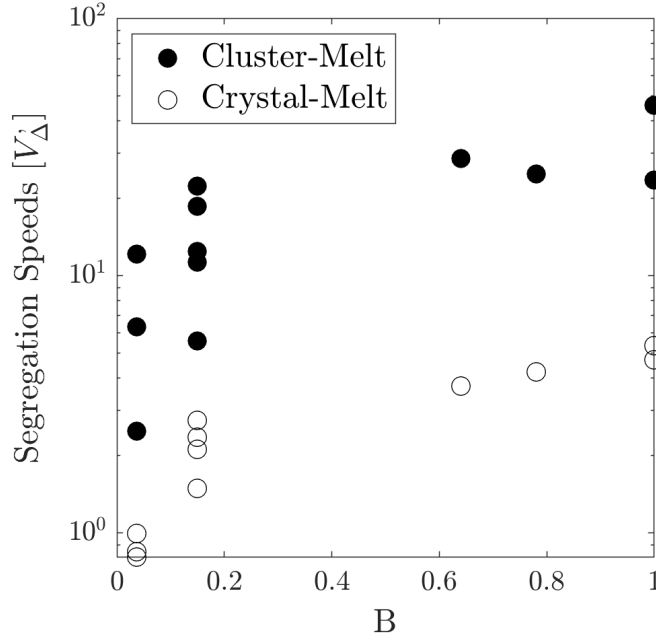


Figure 4. The variation in the average crystal segregation speed, $\overline{v_\Delta}$, and the cluster segregation speed, V_Δ with B .

fractionation in Supp. Fig. 5 for 10 vol% crystallinity gradient and 0 vol% crystallinity gradient. We compare 4 simulations, Sim. (1) with 10 vol% crystallinity on top and 0 vol% crystallinity on the bottom, Sim. (2) with 20 vol% crystallinity on top and 10 vol% crystallinity on the bottom, Sim. (3) with 30 vol% crystallinity on top and 20 vol% crystallinity on the bottom, and Sim. (4) with 10 vol% crystallinity on top and 10 vol% crystallinity on the bottom. We compare different snapshots of the simulations. The crystal fractionation is most efficient for Sim. (1) but the efficiency is comparable to Sims. (2–3) and (4) is the least efficient. Crystal-melt segregation speed is comparable for Sims. (1–3), but crystal-melt segregation speed on average is much lower for Sim. (4). However, segregation speed for Sim. (4) is faster than hindered settling. Cluster-melt segregation speed is comparable for (1–3).

With time, crystals in (2–3) lock up with other crystals, preventing advection of the crystals. Whereas crystals in (1) are able to freely settle through crystal-free melt. This is an explanation as to why the clusters in (2–3) experience less and slower crystal fractionation. Cluster formation still occurs in (4), making it faster than hindered settling would have suggested; however removing the gradient significantly slows down the speed of crystal-melt segregation and hence crystal fractionation.

6 Variability in crystal population

The flow fields that result from cluster settling are unlikely to be preserved in erupted lava because the transport from magma processing zone to volcano conduit will disrupt the collective motion that defines the clusters. Also, because cluster settling is a process unique to liquid-rich regions, the transition from melt-rich to crystal-rich systems could overprint the clusters. Therefore, it is unlikely for clusters to be preserved in plutonic bodies. However, the crystal population in igneous rock samples may preserve indirect signatures of cluster settling. We show one analysis in

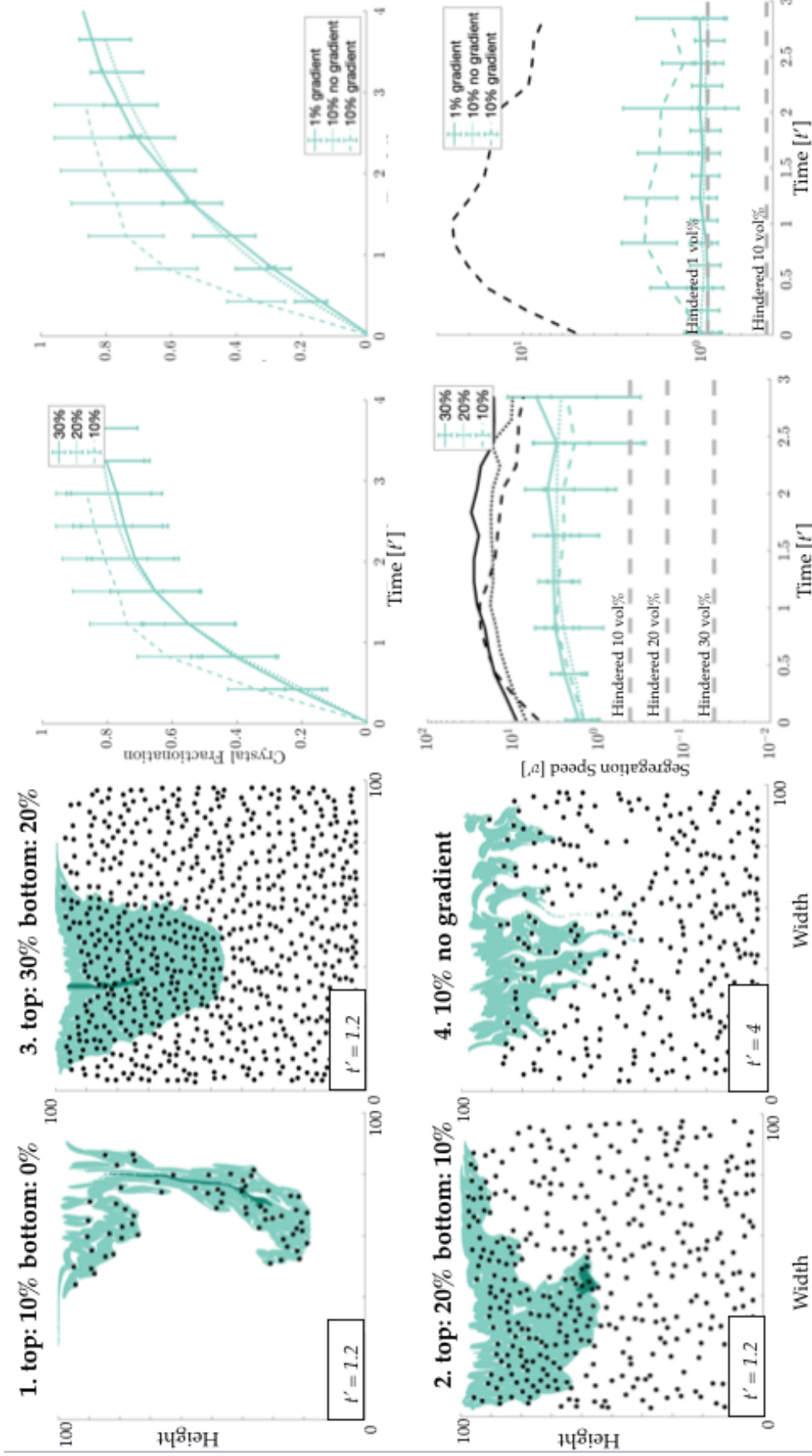


Figure 5. Simulation runs testing efficiency. The figure is set up to mimic Fig. 2 in the Manuscript. Simulations 1–3 have a gradient in crystallinity of 10 vol%. Simulation 1 is the same as the simulation in Fig. 2 of the Manuscript. We show the segregation speeds and crystal fractionation rates Γ . We only show cluster-melt segregation speed for Simulation 1–3, because Simulation 4 does not form a single large cluster. Additionally, for figure simplicity, we do not show the individual crystal-melt segregation and crystal fractionation in dark turquoise.

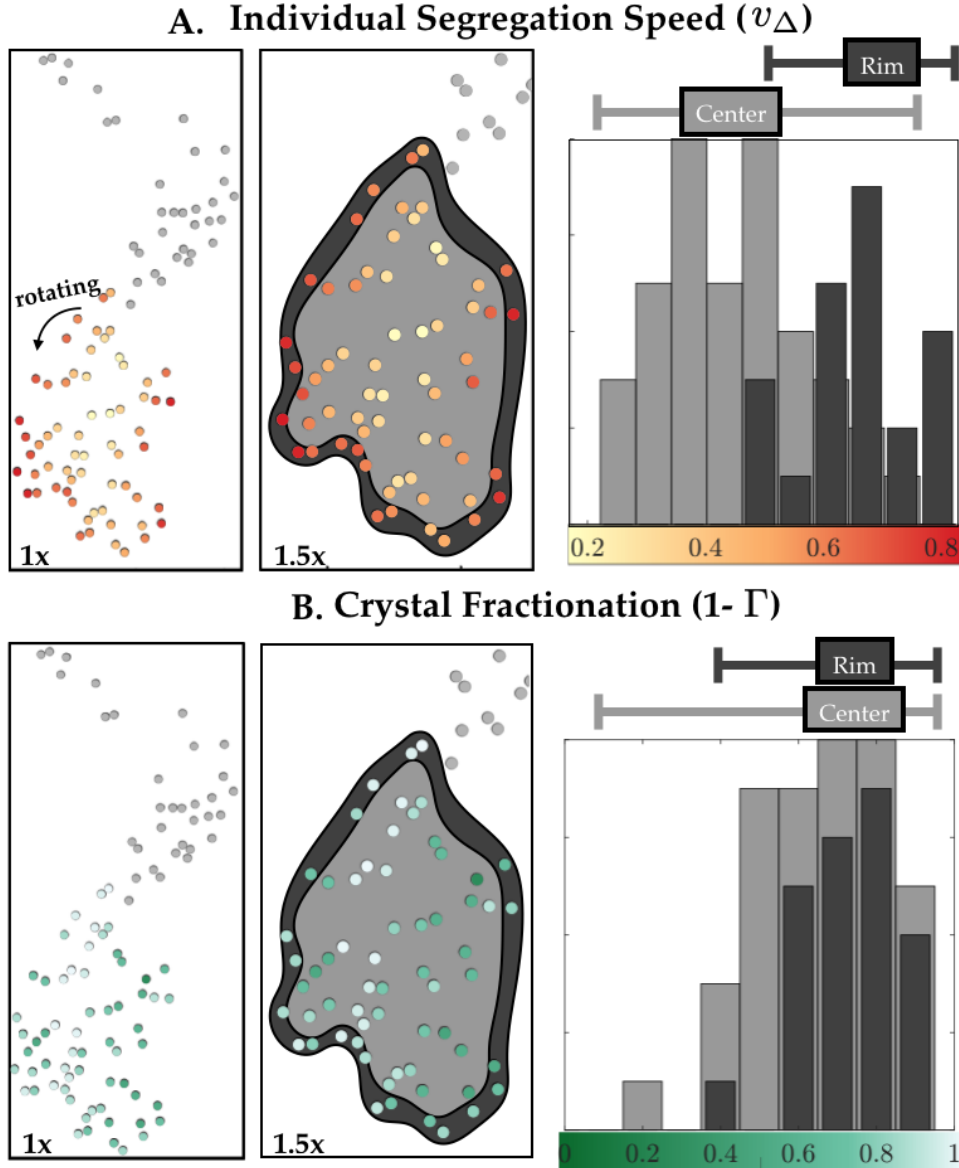


Figure 6. Explaining the observational signatures of convection driven fractionation. Individual crystal segregation speed is shown in (A) and individual crystal fractionation is shown in (B). Figures on the left show a simulation with 10 vol% crystallinity at a single point in time. Individual crystals are colored yellow (slow) to red (fast) to indicate the degree of crystal segregation speeds and dark (no crystal fractionation) to light (full crystal fractionation) green to indicate the degree of crystal fractionation. Dark gray crystals are part of the simulation but are not included in the calculation. The middle figures show which crystals were hand selected to fall in the rim (dark gray) and in the cluster center (light gray). The histograms in the right indicate the distribution of these metrics at either crystal center or crystal rim.

the Manuscript that has direct connection to natural data set. Here we will explore another example that still needs to be connected to natural data set.

We observe that each crystal in a collectively settling cluster segregates and thus fractionates at a different rate depending on its location between the cluster center and rim. We manually identify the crystals that are inside the cluster (light gray) and along the cluster rim (dark gray) in Supp. Fig. 6. On average, the crystals on the rim show higher crystal segregation speeds compared to crystals within the cluster (Supp. Fig. 6A). However, the degree of crystal fractionation is highly variable throughout the cluster (Supp. Fig. 6B).

The degree of crystal fractionation at any given time is a snapshot of the crystal's integrated segregation history. For example, the highlighted red crystal in the 10 vol% crystallinity simulation shown in the Manuscript Fig. 2D-F travels through the cluster neck to the cluster center and finally to the cluster rim. Along this trajectory, its crystal segregation speed is initially higher than the average segregation speed, before slowing down to values similar to the individual settling speed, and finally ending up faster than the average speed again. Each of the crystals continuously shift position relative to one another in the cluster. Clusters are hence dynamically evolving structures. This dynamic evolution allows crystals that formed in opposite ends of the domain to reside next to one another. Examples of olivine crystals with different compositions next to one another could be an example of this subtle variability (e.g., Wieser et al., 2019).

Our results suggest that static clusters would preserve a crystal fractionation population that is similar to the histogram in Fig. 6A, two distinct distributions, whereas dynamic clusters would preserve Fig. 6B. Although growth, dissolution, and nucleation properties of crystals are difficult to model, we hypothesize that histograms of crystalline populations at idealized regions—such as the thick flood-basalts (e.g., Cornwall, 1951; Greenough & Dostal, 1992; Puffer & Horter, 1993) containing horizontal layers of magma that have experienced fractional crystallization post eruption—could determine how dynamic clusters are in magmatic units. This would allow us to better characterize the ideal properties of quickly fractionating magma.

6.1 Quantifying disequilibrium

A relatively low segregation speed between a given crystal and the surrounding melt suggests that the crystal interacts with it for longer than the average crystal interacts with its surrounding melt. This prolonged interaction could translate to an increase in degree of chemical equilibration with the surrounding melt. Since each crystal takes a different path within the cluster, crystals that might come to rest next to each other might show different degrees of chemical interaction with surrounding melt. Generally, the crystals in a cluster will record greater heterogeneity than individual settling crystals. This heterogeneity might be recorded in hand samples as subtle variability in crystal sizes or geochemistry.

At the crystalline scale, the degree of equilibration of crystals with the nearby melt during settling can be characterized by the non-dimensional ratio $D_c/(v_\Delta a)$, a function of the chemical diffusivity, D_c , crystal segregation speed, v_Δ , and crystal size, a . If the segregation speed is large relative to the chemical diffusion rate, the crystal will remain in geochemical disequilibrium. At the mesoscale, since the cluster rims are chemically isolating the cluster center, the cluster rim is most prone to disequilibrium. The crystal-melt segregation speed of crystals in the cluster rim approach V_Δ . Therefore, $D_c/(V_\Delta a)$ would approximate the maximum degree of disequilibrium of crystals in crystal-driven convection.

References

- Cornwall, H. R. (1951). Differentiation in lavas of the Keweenaw series and the origin of the copper deposits of Michigan. *J. Chem. Inf. Model.*, *62*, 159–202. doi: 10.1017/CBO9781107415324.004
- Greenough, J. D., & Dostal, J. (1992). Layered rhyolite bands in a thick North Mountain Basalt flow: the products of silicate liquid immiscibility? *Mineral. Mag.*, *56*(384), 309–318. doi: 10.1180/minmag.1992.056.384.02
- Michioka, H., & Sumita, I. (2005). Rayleigh-Taylor instability of a particle packed viscous fluid: Implications for a solidifying magma. *Geophys. Res. Lett.*, *32*(3), 1–4. doi: 10.1029/2004GL021827
- Puffer, J. H., & Horter, D. L. (1993). Origin of pegmatitic segregation veins within flood basalts. *Geol. Soc. Am. Bull.*, *105*(6), 738–748. doi: 10.1130/0016-7606(1993)105<0738:OOPSVW>2.3.CO;2
- Qin, Z., Alison, K., & Suckale, J. (2019). Rotation matters-direct numerical simulations of rectangular particles in suspensions at low to intermediate solid fraction. *arXiv preprint arXiv:1903.08167*.
- Qin, Z., & Suckale, J. (2017). Direct numerical simulations of gas-solid-liquid interactions in dilute fluids. *Int. J. Multiph. Flow*, *96*(November), 34–47.
- Rayleigh, L. (1883). Investigation of the Character of the Equilibrium of an Incompressible Heavy Fluid of Variable Density. *Proc. London Math. Soc.*, *14*(39), 170–177.
- Sir Geoffrey (F.R.S.), T. (1950). The Instability of Liquid Surfaces when Accelerated in a Direction Perpendicular to their Planes. II. *Proc. R. Soc. A Math. Phys. Eng. Sci.*, *202*(1068), 81–96. Retrieved from <http://rspa.royalsocietypublishing.org/cgi/doi/10.1098/rspa.1950.0086> doi: 10.1098/rspa.1950.0086
- Wieser, P. E., Vukmanovic, Z., Kilian, R., Ringe, E., Holness, M. B., MacLennan, J., & Edmonds, M. (2019). To sink, swim, twin, or nucleate: A critical appraisal of crystal aggregation processes. *Geology*, *47*(10), 948–952. doi: 10.1130/g46660.1