Liquid migration in shear thickening suspensions flowing through constrictions

Citation for published version:

Digital Object Identifier (DOI):
10.1103/PhysRevLett.123.128002

Link:
Link to publication record in Edinburgh Research Explorer

Document Version:
Peer reviewed version

Published In:
Physical Review Letters

General rights
Copyright for the publications made accessible via the Edinburgh Research Explorer is retained by the author(s) and / or other copyright owners and it is a condition of accessing these publications that users recognise and abide by the legal requirements associated with these rights.

Take down policy
The University of Edinburgh has made every reasonable effort to ensure that Edinburgh Research Explorer content complies with UK legislation. If you believe that the public display of this file breaches copyright please contact openaccess@ed.ac.uk providing details, and we will remove access to the work immediately and investigate your claim.
Liquid migration in shear thickening suspensions flowing through constrictions

Rory E. O’Neill,¹ John R. Royer,¹ and Wilson C. K. Poon¹
¹SUPA and School of Physics and Astronomy, The University of Edinburgh,
King’s Buildings, Peter Guthrie Tait Road, Edinburgh EH9 3FD, United Kingdom
(Dated: August 1, 2019)

Dense suspensions often become more dilute as they move downstream through a constriction. We find that as a shear-thickening suspension is extruded through a narrow die and undergoes such liquid migration, the extrudate maintains a steady concentration \( \phi_{\text{out}} \) independent of time or initial concentration. At low volumetric flow rate \( Q \), \( \phi_{\text{out}} \) is a universal function of \( Q/r_d^3 \), a characteristic shear rate in the die of radius \( r_d \), and coincides with the critical input concentration for the onset of LM, \( \phi_{\text{in}}^{\text{LM}} \). We predict this function by coupling the Wyart-Cates model for shear thickening and the ‘suspension balance model’ for solvent permeation through particles.

Suspensions of granular sized particles (radii \( a \gtrsim 5 \mu m \)) are ubiquitous in industrial applications, e.g. molten chocolate [1], ceramic pastes [2] and cement [3]. Recent experiments, theory and simulations show that the rheology of suspensions of granular hard particles at high concentration is dominated by the formation of interparticle frictional contacts above some critical ‘onset stress’, \( \sigma^* \). Such sliding constraints lead to an increase in viscosity with stress, or shear thickening [4–6].

This new understanding pertains to simple shear, but more complex geometries prevail in applications. Thus, constrictions are frequently encountered, e.g., ceramic paste extrusion through a die or orthopaedic cement injection through a syringe. It is unknown to date how recent advances can be applied to these more complex flows, where the material is subjected to significant stress gradients.

Liquid migration (LM) [7], or self filtration [8], is ubiquitous in flow through a constriction: material becomes more dilute as it moves downstream [9, 10]. The solids buildup above the constriction impedes flow, and may lead to jamming. Downstream dilution seriously impacts material strength and stability in ceramics extrusion, and may be fatal in medical applications [11].

While many have explored LM in extrusion using specific formulations [12–17], few have studied the genericity using model systems with well-understood rheology to probe the underlying physics [8, 18]. We investigate LM during die extrusion of cornstarch suspensions, Fig. 1(a), a model granular shear thickening system [19–24] (mean particle radii \( a \sim 7 \mu m \)) with a steady-state rheology which fits an analytic model [5] for friction-driven thickening [4, 6, 25]. We find that during LM the extruded material (extrudate) maintains a steady solid mass fraction \( \phi_{\text{out}} \), independent of time or initial concentration. Interestingly, \( \phi_{\text{out}} \) is a universal function of \( Q/r_d^3 \) at low to moderate volumetric flow rates \( Q \) and all die radii \( r_d \), which we relate to the cornstarch rheology.

We extruded cornstarch suspensions at various solid mass fractions \( \phi \) (see Supplementary Material for preparation details [26]) using a custom-built extruder or orthopaedic syringe (OrthoD Group Ltd.) driven by a universal testing machine (Lloyd LSS, AmetekTest), Fig. 1(a). The custom-built extruder used interchangeable barrels and dies with radii \( R_b \) and \( r_d \) respectively, while the orthopaedic syringe had fixed \( R_b = 6.75 \) mm and \( r_d = 1.7 \) mm. Barrel and die lengths were generally fixed at 40 mm and 10 mm respectively. We drove the plungers at a fixed speed \( v_p \), giving a volumetric flow rate \( Q = \pi R_b^2 v_p \), and measured the applied force \( F \).

Extrudate was collected in vials and extrusion ceased while material still remained in the barrel, which was recovered by removing the die geometry. The solid mass fraction of the extrudate \( \phi_{\text{out}} \) and material left in the barrel \( \phi_{\text{bar}} \) was measured by comparing wet and dry weights [26].

For fixed \( \{Q, R_b, r_d\} \), LM depends on the initial mass fraction of the suspension \( \phi_{\text{in}} \). With \( R_b = 6.75 \) mm, \( r_d = 1.7 \) mm and \( Q = 0.048 \) mL/s, Fig. 1(b), \( \phi_{\text{in}} \approx \phi_{\text{out}} \), below some critical input mass fraction, \( \phi_{\text{in}} \lesssim \phi_{\text{crit}} \approx 0.49 \). When \( \phi_{\text{in}} \) exceeds \( \phi_{\text{crit}} \), \( \phi_{\text{out}} \) drops below \( \phi_{\text{in}} \), i.e., LM occurs. We collected a time-lapsed sequence of extrudates from a suspension undergoing LM at \( \phi_{\text{in}} = 0.52 \), Fig. 1(c). At fixed \( Q \), \( \phi_{\text{out}} \) remains essentially constant at some \( \phi_{\text{out}}^{\text{LM}} \) even as both \( \phi_{\text{bar}} \), Fig. 1(c), and the driving pressure, Fig.1(d), increase dramatically as LM progresses. Moreover, \( \phi_{\text{out}}^{\text{LM}} \) increases with \( Q \), Fig. 1(c). However, once \( \phi_{\text{in}} \) increases beyond \( \phi_{\text{crit}} \) at fixed \( Q \), \( \phi_{\text{out}} \) remains constant at \( \phi_{\text{out}}^{\text{LM}} \), Fig. 1(a). Non-trivially, to within experimental uncertainties, \( \phi_{\text{out}}^{\text{LM}} = \phi_{\text{crit}} \).

While \( \phi_{\text{out}}^{\text{LM}} \) is independent of compaction within the barrel, it does depend on the flow conditions during extrusion. As \( r_d \) decreases from 4 mm to 0.5 mm at fixed \( \phi_{\text{in}} = 0.52 \) and \( R_b = 7.5 \) mm, \( \phi_{\text{out}}^{\text{LM}} \) steadily decreases, Fig. 2(a). That LM increases in smaller constrictions is known [14]. Note that our smallest \( r_d \approx 70 \mu m \) is well above the range for arching and clogging in micro-channels [27] and granular hoppers [28, 29]. As \( Q \) increases, \( \phi_{\text{out}}^{\text{LM}} \) decreases to a minimum before increasing. Before the minimum, we find data collapse onto a single master curve, Fig. 2(b), using the scaling variable \( Q/r_d^3 \), which sets the shear rate scale in the die. This collapse breaks down at higher flow rates, suggesting a significant change in extrusion dynamics, so that beyond the minimum LM is no longer solely controlled by shear in the die. Below, we
FIG. 1: Onset of LM. (a) Extruder schematic. In (b)-(d), \( R_b = 6.75 \text{ mm} \) and \( r_d = 1.7 \text{ mm} \). (b) Concentration of the extrudate \( \phi_{\text{out}} \) and material remaining in the barrel \( \phi_{\text{bar}} \), varying \( \phi_m \), for \( Q = 0.048 \text{ mL/s} \). (c) \( \phi_{\text{out}} \) at varying intervals of plunger displacements at \( \phi_m = 0.52 \), measured from collected extrudates (blue symbols), and measured post-extrusion \( \phi_{\text{bar}} \) (red symbols). Red lines: calculated \( \phi_{\text{bar}} \), assuming constant \( \phi_{\text{out}} \). For \( Q = 0.024 \text{ mL/s} \) (circles, solid line), 0.048 mL/s (squares, dotted line) and 0.095 mL/s (diamonds, dashed line). (d) Extrusion pressure \( P_{\text{ext}} = F/\pi R_b^2 \) vs plunger displacement for the same experiments in (c).

focus on the low-\( Q \) regime, where \( d\phi_{\text{out}}/dQ \leq 0 \).

Experiments at different \((\phi_m, R_b)\), Fig. 3, show that all liquid-migrated extrudate concentrations fall on a single master curve, \( \phi_{\text{out}}^{\text{LM}}(Q/r_d^3) \). Our data span 6.75 mm \( \leq R_b \leq 12.5 \text{ mm} \), i.e. LM is largely unaffected by dynamics within the barrel. Figure 3 also displays samples that do not exhibit LM (\( \phi\)). Their upper bound defines the critical input volume fraction, \( \phi_{\text{in}}^{\text{crit}} \), reinforcing that \( \phi_{\text{out}}^{\text{LM}} = \phi_{\text{in}}^{\text{crit}} \).

The master curve functions as a ‘phase boundary’. Below it, LM does not occur. Above it, the boundary gives the extrudate concentration. Thus, a sample above this phase boundary (\( \star \), Fig. 3) initially at \( \Phi_0 \) being extruded at \( Q/r_d^3 = \hat{\Gamma}_0 \) gives extrudate at a lower concentration given by the intersection of a downward ‘tie line’ from \( \star \) to the phase boundary; the other end of this ‘tie line’, giving the barrel concentration, moves towards close packing to satisfy mass conservation.

The scaling variable \( Q/r_d^3 \) suggests a link between LM and the suspension shear rheology, which we characterized using a rheometer (see Supplementary Material [26]). Controlling the applied shear stress \( \sigma \), we measured the shear rate \( \gamma \) to obtain \( \eta_r = (\sigma/\gamma)/\eta_b \), the suspension viscosity relative to that of the solvent (\( \eta_b \)).

Below \( \phi = 0.44 \), steady-state flow curves \( \eta_r(\sigma) \) show continuous shear thickening, Fig. 4(a), and can be described by the Wyart-Cates (WC) model for thickening due to stress-dependent frictional constraints [5]. In this model, the viscosity is controlled by two limiting concentrations: \( \phi_0 \) when all contacts are frictionless, where the low-stress viscosity \( \eta_l(\phi) \) diverges, and \( \phi_m \) when all contacts are frictional, where the high-stress viscosity \( \eta_H(\phi) \) diverges. Though edge fracture and interfacial instabilities [6, 23] limit full access to \( \eta_H(\phi) \), our data is consistent with this picture, Fig. 4(b).

Full WC flow curves are given by \( \eta_r = [1 - \phi/\phi_1(\sigma)]^{-2}, \) where the jamming concentration where \( \eta_r \) diverges, \( \phi_j = \phi_0[1 - f(\sigma)] + \phi_m f(\sigma), \) varies between \( \phi_0 \) and \( \phi_m \) as \( \sigma \) increases. We model the increasing fraction of frictional contacts [5, 30] by a stretched exponential \( f(\sigma) = e^{-(\sigma/\sigma^*)^\beta} \) [6, 23], so that such contacts become above an onset stress \( \sigma^* \). We fit our results to this model, extracting \( \phi_0 \) from \( \eta_l(\phi) \) and remaining parameters by simultaneously fitting our full set of flow curves [31], giving \( \phi_0 = 0.538 \pm 0.003, \phi_m = 0.4401 \pm 0.002, \sigma^* = 5.4 \text{ Pa} \pm 0.7 \text{ Pa} \) and \( \beta = 0.62 \pm 0.03 \). To avoid bias towards high viscosities we fit log \( \eta_r \). Though we cannot always reach \( \eta_l \) at high \( \phi \), the inflection in the flow curves at high-\( \phi \) is sufficient to constrain \( \phi_m \).

The WC model predicts backward-bending flow curves for \( \phi \geq \phi_m \), Fig. 4(c), so above a \( \phi \)-dependent maxi-
FIG. 3: LM state diagram. (φ): φ_out = φ_m. Color symbols: (φ_m, Q/r_d^3) for various (φ_m, R_0). For R_0 = 7.5 mm, circles: φ_m = 0.52, squares: φ_m = 0.44, upside-down triangles: φ_m = 0.46 and triangles: φ_m = 0.54. For φ_m = 0.52, + symbols: R_0 = 6.75 mm, open circles: R_0 = 10 mm and * symbols: R_0 = 12.5 mm. Same r_d/a color scheme as Fig. 2. Solid lines: predicted LM phase boundary above φ_m. Black: calculated using ατ_r(φ) and using SBM coupled with the WC model for any r_d/a > 70 (see further Fig. 5(c) and text discussion); color: calculated using SBM + WC below φ_m for r_d/a = 70 (yellow) and r_d/a = 140 (green).

The large pre-factor $\frac{Q}{r_d^3}$ gives steep stress gradients between die and barrel that would then, as is well known [34, 35], drive migration. We assume that $\frac{Q}{r_d^3}$ exceeds the maximum shear rate for stability, $\tau_r(\phi)$, local stresses become arbitrarily large, giving steep stress gradients between die and barrel that would then, as is well known [34, 35], drive migration. We in fact find that $\frac{Q}{r_d^3} = \alpha_t \tau_r(\phi)$, with $\alpha_t \approx 4.8$, fits our phase boundary for $\phi > \phi_m$. Fig. 3(c) (black curve). The large pre-factor $\alpha_t > 1$ indicates that flow in a finite region near the die must exceed $\tau_r(\phi)$ to give measurable LM. This region will extend both radially inward and above the die, and since dense suspensions flow with a blunted velocity profile [36, 37], is likely rather thin.

The stress gradients described above will lead to LM both radially, from center to periphery in the die, and axially, from barrel to die, with the latter dominating the dilution of extrudate. We now construct a quasi 1-D model for particle migration along the axis, $z$, using the suspension balance model (SBM) [38, 39] to quantitatively link migration and local stress gradients. To capture the LM phase boundaries, we neglect time-dependent dynamics and only consider the migration onset starting from an initially uniform suspension. Our ansatz for $\tau_r(z)$ is that it is negligible within the barrel, $\tau_r(\phi) < Qr_d^{-3}$, and transitions to some finite $\tau_r(\phi)$ in the die, Fig. 5(a), in a size of $r_d$ immediately upstream to the die.

During LM, there is a non-zero particle velocity relative to the mean flow, $\Delta u = \frac{k(\phi)}{9n_\eta_0}$, with $k(\phi) = \frac{2}{3}\phi(1 - \phi)^4$ the permeability of the particle packing and $n_\eta = \eta_\eta(\phi, \hat{\gamma})\hat{\gamma}$ the shear-induced particle pressure [38–40]. The normal viscosity $n_\eta$, controlling dissipation due to compressive normal stresses, obeys $\eta_\eta(\phi, \hat{\gamma}) = [\phi_3(\phi) - 1]^2$ [41, 42], and diverges at $\phi_3(\phi)$, which we take from WC theory.

We assume that $\tau_r(\phi)$, and so $\eta_\eta$, are negligible in the barrel outside a small transition zone. In this zone, $\partial_t\Pi(\phi, \hat{\gamma}) \approx \Delta \Pi / r_d \approx \eta_\eta(\phi, \hat{\gamma}_d) \tau_r(\phi, \hat{\gamma}_d)$, where $\hat{\gamma}_d$ is a typical shear rate in the die, Fig. 5(a). From previous discussion, $\hat{\gamma}_d \approx \tau_r(\phi) / Q/\alpha r_d^3$. Normalizing to the mean flow in the die ($u = \frac{Q}{\pi r_d^4}$), we obtain a dimensionless migration speed:

$$\Delta u(\phi, \hat{\gamma}_d) \equiv \frac{\Delta u}{u} = \frac{\pi \kappa(\phi) u^2}{\alpha r_d^3} \eta_\eta(\phi, \hat{\gamma}_d).$$

Like the shear viscosity, $\eta_\eta$ bends backwards at $\tau_r(\phi)$, Fig. 5(b), which manifests as a large, abrupt jump in viscosity in rate-controlled flow. To capture this behavior in fixed-$Qr_d^{-3}$ extrusion, we impose such jumps in
...computed using WC fit parameters. Dotted lines: full backwards bending or ‘S’ shaped flow curves. Solid lines: profiles used to compute ∆\tilde{u} parameters. Dotted lines: full backwards bending or ‘S’ shaped flow curves. (b) Normal viscosity \eta(\gamma) computed using WC fit parameters. Dotted lines: full backwards bending or ‘S’ shaped flow curves. Solid lines: profiles used to compute ∆\tilde{u} with imposed jumps in the unsteady regime. (c) ∆\tilde{u}(\phi,Q/r_d^3), computed using \alpha = 4.8 and r_d/a = 140. Red dashed line: \phi_m. The contour ∆\tilde{u}(\phi,Q/r_d^3) = \epsilon^1 = 0.004 gives the SBM+WC phase boundary in Fig. 3 for r_d/a = 140.

FIG. 5: A 1-D model for LM. (a) Schematic of the ‘dilation zone’ above the die. The resulting stress gradient causes a velocity difference \Delta u = u_p - (u) between the particles and mean flow. (b) Normal viscosity \eta(\gamma) computed using WC fit parameters. Dotted lines: full backwards bending or ‘S’ shaped flow curves. Solid lines: profiles used to compute ∆\tilde{u} with imposed jumps in the unsteady regime. (c) ∆\tilde{u}(\phi,Q/r_d^3), computed using \alpha = 4.8 and r_d/a = 140. Red dashed line: \phi_m. The contour ∆\tilde{u}(\phi,Q/r_d^3) = \epsilon^1 = 0.004 gives the SBM+WC phase boundary in Fig. 3 for r_d/a = 140.

\eta for \gamma_d \geq \gamma_c(\phi) when evaluating ∆\tilde{u}(\gamma, \phi), Fig. 5(b), and similar jumps in the ‘S’-shaped discontinuous shear thickening flow curves [5, 43, 44].

Figure 5(c) shows ∆\tilde{u}(\phi,Q/r_d^3) for \alpha = 4.8. Although there is finite migration, ∆\tilde{u} > 0, for all \phi and Q/r_d^3, the jump from negligible [∆\tilde{u} \sim O(10^{-5})], blue) to strong migration [∆\tilde{u} \sim O(10^{-1})], yellow)] is very sharp for \phi > \phi_m, and is associated with the equally abrupt jump in \eta when \gamma_d \rightarrow \gamma_c(\phi) = Q/\alpha r_d^3, i.e., at precisely the LM boundary. Formally, if we define the transition from negligible to significant migration to occur at some threshold, i.e. when ∆\tilde{u}(\phi, \gamma_d) \geq \epsilon^1, we recover the observed phase boundary above \phi_m for any 10^{-5} \lesssim \epsilon^1 \lesssim 10^{-2} independent of r_d for r_d/a > 70.

Below \phi_m, the WC flow curves approach a limiting high-stress viscosity, so there is no longer a maximum possible shear rate \gamma_c. Now, the transition from low to high ∆\tilde{u} is far less abrupt, Fig. 5(c) (below red dashed line), and where it can be deemed to occur depends on our choice of the threshold, \epsilon^1, and also on r_d/a.

To choose \epsilon^1, we note that the dilation accompanying particle migration is roughly equivalent to ∆V contributing an extra volume ∆V \propto ∆u/\alpha r_d^3 to material in the transition zone above the die, where particles reside for r_d \approx r_d/\alpha. Thus, |∆\phi/\phi| \approx ∆V/Qr_d \sim ∆u/\alpha \equiv ∆\tilde{u}. Experiments on suspensions of larger particles (a \geq 50 \mu m) below \phi_m [18] detected LM for |∆\phi/\phi| \approx 4 \times 10^{-3}, which we take to be our \epsilon^1. We plot the LM boundary using \epsilon^1 = 0.004 for two die radii, r_d/a = 70 and 140, in Fig. 3. The fit to data below \phi_m is credible, but poorer than for \phi > \phi_m. Using smaller \epsilon^1 produces better fits, but is hard to motivate physically. Whatever the choice of \epsilon^1 and r_d/a, our model captures the abrupt change in slope of the LM boundary at \phi_m.

Summarizing, we have characterized LM during extrusion of shear-thickening cornstarch suspensions. The onset concentration at low to moderate flow rates lies on a universal boundary if data for different flow rates and die radii are plotted against Qr_d^{-3}, an estimate of the maximum shear rate in the die. The locus where Qr_d^{-3} \rightarrow \alpha \gamma_c(\phi) fits well the observed LM boundary above \phi_m, with \alpha \approx 4.8, suggesting that flow in a finite region near the die entrance must be unstable for appreciable LM. The instability point \gamma_c(\phi) can be estimated by where \eta dramatically increases in controlled-stress rheology, Fig. 4(c). We therefore have a theory for LM at \phi \geq \phi_m up to a single dimensionless parameter independent of any other theoretical model. It is also possible to obtain \gamma_c(\phi) by fitting bulk rheology data to WC theory. Coupling this to the SBM for particle migration in a simple 1D model gives a semi-quantitative prediction of the boundary below \phi_m. A more sophisticated theory of LM accounting for radial migration [36, 45–47] and extensional flow [48, 49] may obviate the need for \alpha and produce better agreement at \phi < \phi_m.

Previously, LM had been modeled using finite-time-step methods to simulate extrusion, and either a 1-D [50, 51] or 2-D finite-element model [52, 53] to describe the paste. These empirical paste models rely on material parameters not directly extractable from shear rheology. Particle-based simulations of extrusion, which lack an explicit fluid phase, reproduce localized shear and stress gradients near the die entry but not LM, highlighting the importance of such gradient-driven flows [37]. A model for LM in suspensions of larger granular particles based on suspension balance exists [18], but requires measured particle pressures as input. Our model uses bulk rheology data to capture LM.

While we find increasing LM with increasing (low to moderate) flow rate in shear thickening suspensions, LM increases with decreasing flow rates in many other pastes [11, 13, 14, 16], possibly because attractive or adhesive interaction between particles gives rise to yield stress and shear-thinning behavior. A recently-proposed constraints-based extension of the WC model to include such interactions [54] may allow application of our approach here to a broader range of pastes.

Acknowledgments

Funding came from the EPSRC (EP/N025318/1) and the NSF under Grant No. NSF PHY-1748958 through the KITP program on the Physics of Dense Suspensions.