

A general constitutive model for dense, fine-particle suspensions validated in many geometries

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Fine-particle suspensions (such as cornstarch mixed with water) exhibit dramatic changes in viscosity when sheared, producing fascinating behaviors that captivate children and rheologists alike. Examination of these mixtures in simple flow geometries suggests intergranular repulsion and its influence on the frictional nature of granular contacts is central to this effect—for mixtures at rest or shearing slowly, repulsion prevents frictional contacts from forming between particles, whereas when sheared more forcefully, granular stresses overcome the repulsion allowing particles to interact frictionally and form microscopic structures that resist flow. Previous constitutive studies of these mixtures have focused on particular cases, typically limited to 2D, steady, simple shearing flows. In this work, we introduce a predictive and general, 3D continuum model for this material, using mixture theory to couple the fluid and particle phases. Playing a central role in the model, we introduce a microstructural state variable, whose evolution is deduced from small-scale physical arguments and checked with existing data. Our space- and time-dependent model is implemented numerically in a variety of unsteady, nonuniform flow configurations where it is shown to accurately capture a variety of key behaviors: 1) the continuous shear-thickening (CST) and discontinuous shear-thickening (DST) behavior observed in steady flows, 2) the time-dependent propagation of “shear jamming fronts,” 3) the time-dependent propagation of “impact-activated jamming fronts,” and 4) the non-Newtonian, “running on oobleck” effect, wherein fast locomotors stay afloat while slow ones sink.

discontinuous shear thickening | constitutive model | two-phase flow

The behavior of granular materials suspended in fluid media has been a major topic of study for over a century. These types of mixtures are present in many industrial, geotechnical, and biological engineering problems (e.g., transport of waste material, erosion of shorelines and riverbeds). Of particular interest in this work is the behavior of chemically stable, hard, frictional particles suspended in viscous fluids as found in industrial processes and studied in soil mechanics.

Although thoroughly studied and classified, a unifying constitutive model relating mixture stresses, strains, and strain rates across all material types and flow regimes remains elusive (1). One challenge for such a model is capturing the non-Newtonian, shear-thickening behavior that is often observed in mixtures like water–cornstarch and water–poly(methyl methacrylate) when the mean particle diameter, d , is less than $\sim 10\ \mu\text{m}$. At low volume fractions, the apparent viscosity of such mixtures grows steadily with increasing shearing rate (CST); however, at high volume fractions, the apparent viscosity of these mixtures can jump several orders of magnitude with very little change in measured shearing rate (DST).

Theoretical work in refs. 2 and 3, experimental observations in refs. 4–8, and simulations reported in refs. 9–12 have shown that the shear-thickening behavior observed in these mixtures is a direct result of intergranular repulsion inhibiting the formation of frictional granular contacts and the resulting effect on the dilation behavior of the mixture. In the presence of rel-

atively small applied stresses, the particles in the mixture will interact through lubrication forces in the suspending medium and behave like a granular material with low interparticle friction. In the presence of relatively large applied stresses, the particles in the mixture will be forced into frictional contact, drastically increasing the bulk resistance to flow. In dense suspensions, this frictional transition can cause the granular skeleton to dilate. Further, recent observations reported in refs. 13–16 and simulations shown in ref. 17 have indicated that the growth and decay of these dilating structures in the granular skeleton plays an important role in the time-dependent behavior of these mixtures.

Prior work modeling general fluid–sediment mixtures in ref. 18 has focused on hard, frictional, non-Brownian, nonrepulsive particle suspensions ($d \gtrsim 100\ \mu\text{m}$ for common engineering mixtures). The modeling framework proposed in that work combines the empirical relations presented in refs. 1, 19, and 20 with the 2-phase mixture theories developed in refs. 21 and 22 into a single constitutive theory. In this work, we build upon this framework to produce a more general model for granular suspensions that accounts for the effect of intergranular repulsion on mixtures with mean grain diameters $d \lesssim 100\ \mu\text{m}$. Using a custom numerical scheme, we validate this model in transient and inhomogeneous flows, in both 2D and 3D geometries. The model is shown to replicate the unusual landmark features of these shear-thickening suspensions, such as the ability to run across them but not walk.

Significance

Shear-thickening suspensions display peculiar phenomena, exhibiting solid-like and fluid-like behaviors at the same volume fraction. These materials have puzzled researchers since the 1930s and have seen renewed interest due to advances in computational and experimental methods as well as new infrastructural and military applications that might exploit these behaviors. We introduce a continuum model for shear-thickening suspensions, which correctly predicts realistic 3D flows including transient and inhomogeneous cases. Our approach builds the microscopic physics of particle repulsion into a framework for general fluid–grain mixtures, bridging to the fields of geomechanics and granular rheology. Using a material point method routine, we provide a robust model implementation and demonstrate the method’s broader utility for a variety of fluid-particle flow problems.

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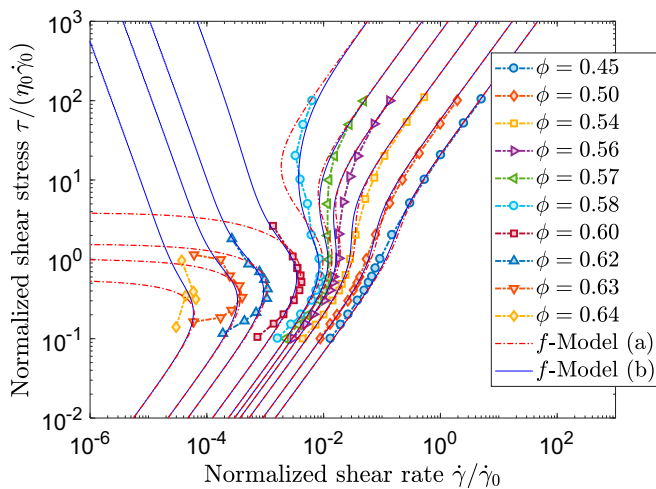


Fig. 1. Comparison of steady-shearing response of model described in *Model Development* to data reported in ref. 12 for $\mu_c = 1$. Results are normalized by $\eta_0 \dot{\gamma}_0$ and associated shearing rate $\dot{\gamma}_0$ as reported in ref. 12. The simulated data are represented by the square, triangular, and circular markers (see legend). Our model is shown with red dashed lines for the parameters in Table 1 (a) and with blue solid lines for those in Table 1 (b) at the packing fractions [0.45, 0.50, 0.54, 0.56, 0.57, 0.58, 0.60, 0.62, 0.63, 0.64].

Model Development

General Theory. In continuum modeling of fluid-particle mixtures, we consider the materials that constitute the mixture independently. The individual solid particles (or grains) are homogenized into a single continuum body called the “granular phase,” which has a density $\bar{\rho}_s = \phi \rho_s$ and velocity components v_{si} . Here, ϕ defines the volume fraction of the granular phase, and ρ_s defines the density of each individual grain. Similarly, the fluid that fills the space between the grains is homogenized into a single continuum body called the “fluid phase” with density $\bar{\rho}_f = (1 - \phi)\rho_f$ and velocity components v_{fi} . Here, ρ_f is the true density of the fluid. A thorough theoretical description of this process of homogenization can be found in refs. 18 and 22.

From the basic laws of mass conservation, momentum and energy balance, and entropy imbalance, it is possible to define a complete set of governing equations for the evolution of density, velocity, and stress within each phase. These equations account for 1) momentum exchange between the phases through a buoyant force and a Darcy-like interphase drag, 2) dilation and contraction of the granular phase through a specialized elastic-plastic constitutive model, and 3) separation of mixture stresses into components from the granular skeleton, $\bar{\sigma}_{ij}$, and components from the pore fluid, τ_{fij} and p_f . These equations are combined to form the model proposed in ref. 18. This model is shown to be accurate in a wide range of flow regimes and geometries; however, it does not account for the effects of intergranular repulsion on the frictional nature of granular contacts. In this work, we show that reformulating 2 previously constant parameters governing the dilation and critical state behavior of the mixture, a and ϕ_m (Eqs. 1 and 2 below), is sufficient to capture the physics of particle repulsion and accurately model steady CST and DST as well as several transient and dynamic behaviors observed in the dense, fine grain suspensions we seek to model.

The full set of equations that define our proposed 3D, time-dependent, 2-phase model can be found in *SI Appendix, Table S1*; however, several key features of this model are easily illustrated by considering its behavior in steady, constant volume, homogeneous, 2D (or quasi-2D) shearing flows. This behavior

is described by the relationship between the steady mixture shearing rate $\dot{\gamma}$, the mixture shear stress τ , and the granular-phase pressure $\bar{p}$. For flows dominated by viscous effects (e.g., water–cornstarch; $(\rho_s d^2 \dot{\gamma})/\eta_0 \ll 1$), that steady-state relationship is identical to the model proposed in ref. 23 and presented in ref. 19 and is expressed as follows,

$$\frac{\tau}{\eta_0 \dot{\gamma}} = \eta_r = 1 + \frac{5}{2} \phi \left(\frac{\phi_m}{\phi_m - \phi} \right) + 2\mu_c \left(\frac{a\phi}{\phi_m - \phi} \right)^2, \quad [1]$$

$$\frac{\bar{p}}{\eta_0 \dot{\gamma}} = \eta_n = 2 \left(\frac{a\phi}{\phi_m - \phi} \right)^2, \quad [2]$$

with η_0 , the fluid-phase viscosity; and μ_c , a dimensionless measure of flow resistance due to frictional granular contacts. The dilation parameters ϕ_m and a have a significant effect on the mixture stress and granular-phase pressure.

The parameter ϕ_m limits the range of volume fractions where steady flow is possible (i.e., for $\phi \geq \phi_m$, shearing the material results in continuous growth of the shear stress, sometimes referred to as a “jammed” state), and the parameter a modifies the critical state volume fraction (see ϕ_{eq} in ref. 18 and *SI Appendix, Table S1*) and associated Reynolds’ dilation. ϕ_m and a are often treated as material constants; however, as proposed in ref. 3 and fully defined in ref. 12, ϕ_m and a are better described as functions with ϕ_m bounded between ϕ_j and ϕ_c (with $\phi_j > \phi_c$) as follows,

$$\phi_m = \hat{\phi}_m(f) = \phi_j + (\phi_c - \phi_j)f, \quad [3]$$

and a bounded between a_0 and a_∞ as follows,

$$a = \hat{a}(f) = a_0 + (a_\infty - a_0)f, \quad [4]$$

where $f \in [0, 1]$ is the fraction of frictional contacts within the granular skeleton. Conceptually, f represents a measure of granular structure: as f increases, ϕ_m will decrease toward ϕ_c , producing higher resistance to flow (see η_r in Eq. 1) and causing “jamming” through a wider range of volume fractions. The material parameters ϕ_j and ϕ_c then correspond to the range of volume fractions where steady-shearing flow can occur. For $\phi < \phi_c$, steady flow can occur at all shearing rates. For $\phi \geq \phi_j$, no steady flow is possible; the mixture is jammed. For $\phi_c \leq \phi < \phi_j$, steady flow is only possible at some shearing rates. a_0 and a_∞ are fitting parameters.

Unlike in previous models where f was a function of granular stress directly, we model f as a spatial field that evolves over time according to the following rule,

$$\frac{\dot{f}}{K_0 \dot{\gamma}} = H(f_m - f) - Sf, \quad [5]$$

with K_0 , a dimensionless parameter; H , a dimensionless hardening rate; S , a dimensionless softening rate; and f_m , an upper bound on the steady value of f given by $f_m = \hat{f}_m(\phi)$. Such an upper bound on f was first proposed for low volume fractions in ref. 24 and is an important feature in this model, especially as it relates to “rheo-chaos” (25, 26).

The first term in Eq. 5, $H(f_m - f)$, describes the evolution of f toward f_m and, by Eqs. 1 and 3, the associated increase in

Table 1. Material parameters for curves in Fig. 1

Fit	ϕ_c	ϕ_j	a_0	a_∞	$\tau^*/(\eta_0 \dot{\gamma}_0)$	η_0/η_B
(a)	0.585	0.65	0.455	0.841	1.74	0
(b)	0.585	0.65	0.455	0.841	1.44	$0.254(\phi_j - \phi)$

$\mu_c = 1$ for all fits. μ_h was determined from a and ϕ_m (18).

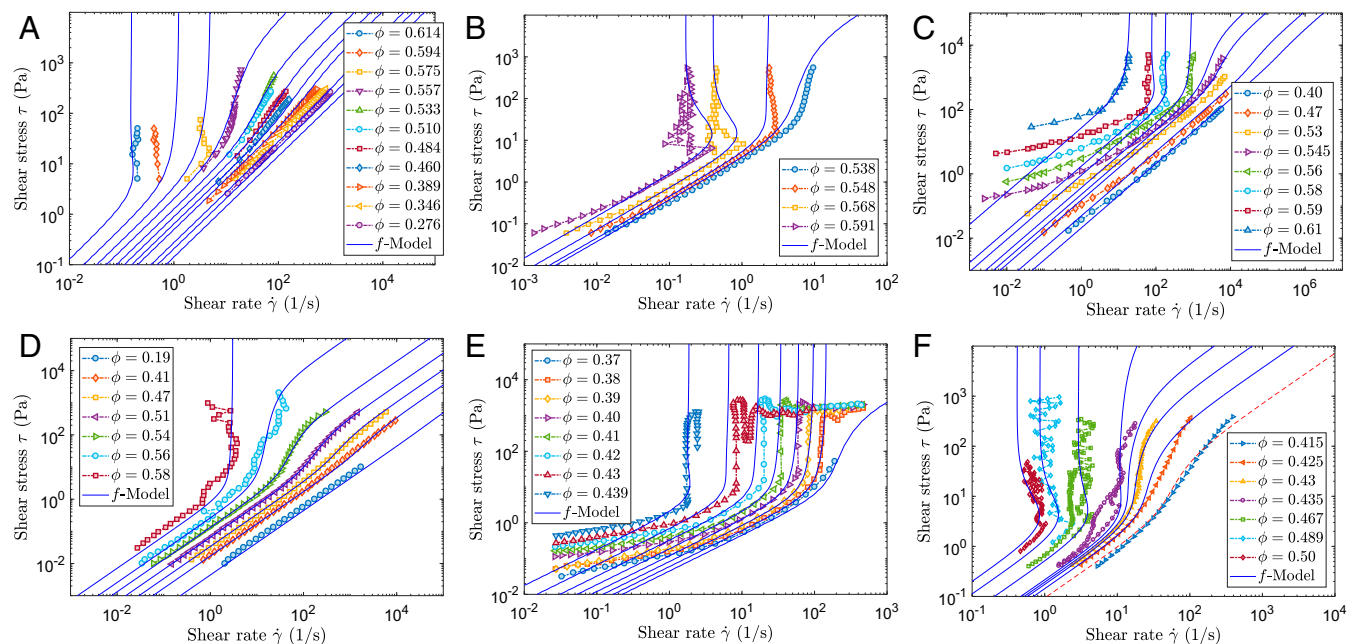


Fig. 2. Comparison of steady-shearing response of model described in *Model Development* to experimental data reported in ref. 32 (A), ref. 30 (B), ref. 5 (C and D), ref. 31 (E), and ref. 25 (F). Experimental data are represented by colored markers at volume fractions noted in the associated legends. The response of our model is shown with the solid blue curves at the relevant volume fractions using the parameters in Tables 2 and 3. The dashed red curve in F corresponds to the model response at a volume fraction of 0.39. The high-stress plateau present in the experimental data shown in E is a consequence of the granular normal stress reaching the limit of what can be supported at the boundaries, causing a deviation from the idealized flow conditions that this plot assumes; see refs. 8 and 31 for further details.

effective viscosity, η_r . Based on the work of ref. 10 (*SI Appendix, Fig. S1*), we propose the following form for H ,

$$H = \left(\frac{\bar{\tau}}{\tau^*} \right)^{3/2}, \quad [6]$$

with τ^* , a repulsive stress scale with units of Pa (see τ_c in ref. 4, $\tau_{\min}$ in ref. 27, and σ^* in ref. 12); $\bar{\tau} = (\mu_c + \mu_h)\bar{p}$, the effective granular shear stress; and μ_h , a dimensionless measure of flow resistance due to fluid-mediated granular interactions (19). This form of H captures the strengthening of the granular phase as the granular stress forcing grains together overcomes intergranular repulsion. The second term in Eq. 5, $-Sf$, describes the evolution of f toward 0 and the associated decrease in effective viscosity, η_r . We propose the following form for S ,

$$S = 1 + \frac{\bar{\tau}}{\eta_B \dot{\gamma}} + \frac{\dot{\xi}_e}{\dot{\gamma}}, \quad [7]$$

with η_B , a measure of the resistance of the granular skeleton to buckling induced degradation with units of Pa·s; and $\dot{\xi}_e$, a rate scale for Brownian or electrostatic repulsion with units s^{-1} . The 3 terms in S reflect 3 proposed mechanisms that will break down granular structure and thereby decrease the fraction of grains in frictional contact. First, macroscopic shearing of the mixture will cause grains to slip past each other, altering their structure (see ref. 28 for discussion of “plastic rearrangement” in jammed materials). Second, we postulate that force chains can undergo load-initiated reorganization ($\bar{\tau}/\eta_B \dot{\gamma}$), presumably through chain buckling, inducing structural degradation without macroscopic deformation. In systems of grains with large but finite elastic stiffness G_p , the small relative contact area A_c/d^2 of the particles means the buckling load $\tau_c \sim G_p A_c^2/d^4$ need not be large. The proposed form might then be seen as the product of a statistical measure of critical chains ($\sim \bar{\tau}/\tau_c$) and a time scale of buckling degradation given by the properties of the mixture ($t_c \tau_c/\eta_0 = \dot{\varphi}(\rho G_p d^2/\eta_0^2, A_c/d^2, \phi)$, i.e., $\eta_B \propto \tau_c t_c$). Lastly,

Brownian diffusion or electrostatically driven particle reorganization may move grains out of frictional contact ($\dot{\xi}_e/\dot{\gamma}$); in flowing materials at room temperature, this contribution to $\dot{f}$ should be negligible.

Steady Behavior of Rheologically Stable Flows. We begin analyzing the proposed model by considering the simulated 3D shearing flows in ref. 12. These flows were composed of between 500 and 2,000 stiff, spherical particles undergoing simple shear in a periodic domain. The discrete particle interactions were modeled using standard contact laws in conjunction with additional forces to account for lubrication and intergranular repulsion. The steady-state relationship between $\dot{\gamma}/\dot{\gamma}_0$ and $\tau/(\eta_0 \dot{\gamma}_0)$ (with $\eta_0 \dot{\gamma}_0 \propto \tau^*$) reported in that work (Fig. 1) contains several regimes where $\partial \dot{\gamma}/\partial \tau \leq 0$. These regimes can lead to unstable behavior under imposed shearing rates (29) and imposed shear stresses (26). The latter instability manifests in rheologically chaotic behavior (i.e., transient inhomogeneities in the flow occur; rheo-chaos). The lack of reported rheo-chaos in ref. 12 suggests that the behavior of larger systems of particles may differ at packing fractions near and above ϕ_c .

Table 2. Material parameters for curves in Fig. 2

Fit	ϕ_c	ϕ_j	a_0	a_∞	τ^* [Pa]	η_0 [mPa·s]
Ref. 32	0.57	0.65	1.08	1.08	5.21	56
Ref. 30	0.548	0.642	1.08	1.08	17.93	130
Ref. 5(a)	0.56	0.617	1.577	1.577	423.0	2.4
Ref. 5(b)	0.563	0.610	1.09	1.538	3.5	2.4
Ref. 31	0.38	0.454	1.00	1.00	8.55	0.89
Ref. 25	0.436	0.535	0.8	0.8	5.35	6

$\mu_c = 0.3$ for refs. 5, 30, and 32. $\mu_c = 1.19$ for refs. 25 and 31. $\Delta = 0$ for all experiments. Ref. 5(a) and (b) correspond to data for $d = 404$ nm and $d = 3,770$ nm particles, respectively.

Table 3. Parameters of $\hat{\eta}_B(\phi)$ for curves in Fig. 2

Fit	φ_1 [(Pa·s) ⁻¹]	α_1	φ_2 [(Pa·s) ⁻¹]	α_2
Ref. 32	8.83×10^6	6	0	0
Ref. 30	1.79×10^{11}	12	4.5×10^{-3}	0.1
Ref. 5(a)	3×10^6	5	9×10^{-2}	0.1
Ref. 5(b)	1.0×10^6	6	0	0
Ref. 31	4.0×10^4	3	0	0
Ref. 25	1.86×10^4	4	9.3×10^{-3}	0.1

Ref. 5(a) and (b) correspond to data for $d = 404$ nm and $d = 3,770$ nm particles, respectively.

We can, however, still examine the steady-state response of the model in Eq. 5 for such rheologically stable shearing flows. To do this, we first define a functional form for $\hat{f}_m(\phi)$. The results in ref. 10 are generated using the method shown in ref. 12 and suggest that f_m is insensitive to changes in ϕ . We therefore let $f_m = 1$ for such mixtures. With f_m determined, we calculate and fit the steady-shear response of our model to the data in ref. 12 by solving for $\dot{\gamma} = 0$ at different volume fractions ϕ and shear rates $\dot{\gamma}/\dot{\gamma}_0$ (with $\tau/\eta_0\dot{\gamma}_0$ determined from Eq. 1), as shown in Fig. 1. The relevant material parameters for these fits are provided in Table 1. (See *SI Appendix* for specific details about our model fitting procedure.)

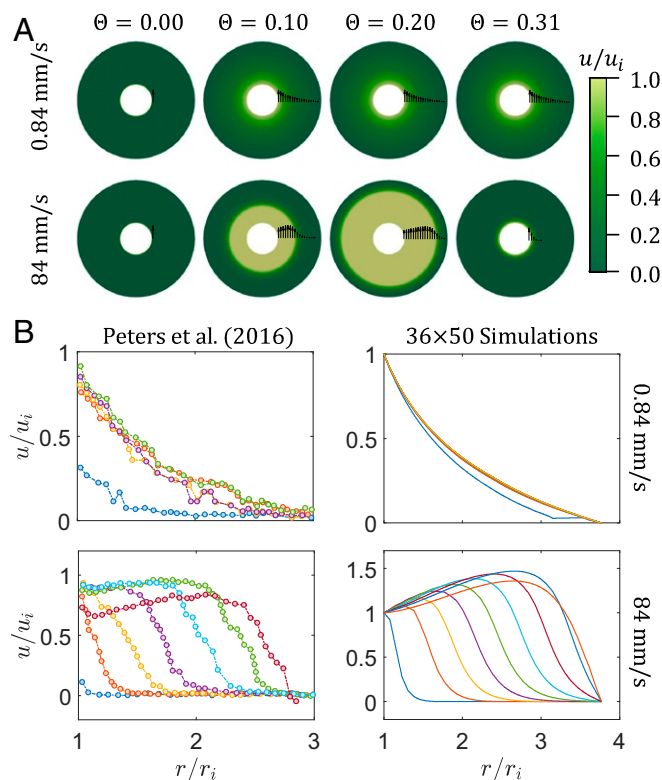


Fig. 3. (A) Comparison of surface-velocity field for annular-shear simulations (Table 4) after 0, 0.1, 0.2, and 0.31 radians of inner-wall rotation for $u_i = 0.84$ and $u_i = 84$ mm/s. The $u_i = 0.84$ mm/s simulation rapidly approaches the diffusive-flow profile of Newtonian fluids in annular shear. However, the $u_i = 84$ mm/s simulation shows rapid growth of a rigidly rotating block of mixture; when the edge of this block reaches the outer edge of the annular-shear cell, all flow ceases. **(B)** Comparison of experimental surface-velocity profiles taken at even intervals of rotation from ref. 13 to simulated velocity profiles. Note, simulations do not account for wall slip or the effects of boundary breakdown.

Pseudo-Steady Behavior of Rheologically Chaotic Flows. We continue analyzing the proposed model by considering experimental analysis of repulsive-grain mixtures. Although many authors have explored the phenomenology of these mixtures, complete characterization of their behavior is complicated by several factors that require additional modeling considerations beyond the ideal case discussed in the prior section. The scaling of τ^* with respect to mean grain diameter d is often reported as $\tau^* \propto d^{-2}$ or d^{-3} (5, 27, 30) and is observed to have vanishing effect on the response of common granular suspensions with $d \gtrsim 100$ μ m; this puts practical limitations on the minimum system size (quantified by number of grains) that can be analyzed. Boundary breakdown (where granular normal stresses exceed operational boundary limits; refs. 8, 13, and 31 and Fig. 2E) is observed when applied shear stresses exceed $\sim 10^4$ Pa, limiting the range of mixture responses that can be probed. Additionally, starches and other porous particles swell significantly in suspension, making the true volume fraction of the mixture unclear (25).

Nevertheless, experimentation is the most direct way to examine the behavior of these particle suspensions and has yielded many important observations. Among these is the rheo-chaos reported in refs. 25, 32, and 33; these rheologically chaotic flows are characterized by large, rapid changes in the measured mixture shearing rate at constant applied stresses and measured relationships between $\dot{\gamma}$ and $\bar{\tau}$ that generally have $\partial\dot{\gamma}/\partial\bar{\tau} \geq 0$. More thorough study is necessary to understand the mechanics and transient effects of rheo-chaos, but if we consider the long-time average behavior of the shearing flows reported in the literature to be representative of the average local behavior, we can examine the steady-state response of the model in Eq. 5 for such rheologically chaotic flows.

We begin this analysis by defining an expression for $\hat{f}_m(\phi)$ based on the experimental observations reported in refs. 25 and 31 as follows,

$$\hat{f}_m(\phi) = \begin{cases} 1, & \text{if } \phi \leq \phi_c \\ \frac{\phi - \phi_j}{\phi_c - \phi_j}, & \text{if } \phi_c < \phi \leq \phi^* \\ \frac{\phi^* - \phi_j}{\phi_c - \phi_j}, & \text{if } \phi > \phi^*, \end{cases} \quad [8]$$

with $\phi^* = \phi_j + (\phi_c - \phi_j)\Delta$ and Δ , an implicit function of the compliance of the granular skeleton. By observation, we expect $\Delta \rightarrow 0$ for large systems of irregular grains (where rheo-chaos is observed) and $\Delta \rightarrow 1$ for small systems of stiff, spherical grains (where rheo-chaos is not observed). In the latter case, $f_m \rightarrow 1$ as in the idealized mixtures of the previous section.

Given this form of $\hat{f}_m(\phi)$, it is possible to determine a critical shearing rate associated with DST, $\dot{\gamma}_{\text{DST}}$, which is the limiting value of $\dot{\gamma}$ as $\bar{\tau}$ increases for the volume fractions in the range $\phi_c \leq \phi \leq \phi^*$ (*SI Appendix*). Based on the distribution of apparent $\dot{\gamma}_{\text{DST}}$ values in the literature and the trends of $\dot{\gamma}_c$ reported in ref. 34, we propose the following form of the structural resistance to buckling induced degradation η_B ,

$$\eta_B = \hat{\eta}_B(\phi) = \left(\sum_{i=1}^{i_{\text{max}}} \varphi_i (\phi_j - \phi)^{\alpha_i} \right)^{-1}, \quad [9]$$

with φ_i representing material parameters with units of (Pa·s)⁻¹ and α_i representing dimensionless scaling factors.

With these expressions determined, we calculate and fit the steady-shear response of our model to experimental results

Table 4. Material parameters for simulations shown in Fig. 3

ϕ_0	η_0 [mPa·s]	ρ [kg/m ³]	G [Pa]	K [Pa]	K_0
0.505	10	1,620	3.8×10^5	8.3×10^5	0.06

Table 5. Material parameters for simulations shown in Figs. 4 and 5

η_0 [mPa·s]	ρ_s [kg/m ³]	ρ_f [kg/m ³]	G [Pa]	K [Pa]	K_0
7 or 1	1,620	1,000	3.8×10^7	8.3×10^7	1.0

reported in refs. 5, 25, and 30–32 by solving for $\dot{\gamma} = 0$ while satisfying the relationship in Eq. 1 at the relevant volume fractions and shearing rates as shown in Fig. 2. All data from the various experiments are seen to be well represented by the generic form proposed. The parameters for these steady-shearing experiments can be found in Tables 2 and 3. The wide range of τ^* values reported in Table 2 is likely linked to the scaling of $\tau^* \propto d^{-2}$ or d^{-3} and changes in surface chemistry between different granular materials (see the effects of hydrogen bonding on “shear jamming” and DST in ref. 35). Refs. 5, 30, and 32 performed experiments using poly(methyl methacrylate) beads with diameters d between roughly 0.3 and 4 μm while refs. 25 and 31 performed experiments using cornstarch with reported grain diameters often between 5 and 20 μm . (See *SI Appendix* for specific details about fitting $\dot{\gamma}_{\text{DST}}$ and determining ϕ from ref. 25.)

Complete, Time-Dependent Expression of Model. The steady-state measurement of DST and CST is only one part of the puzzle of repulsive-grain suspensions. As noted in ref. 36, a fully coupled constitutive model is necessary to capture the observed dynamic behavior of these mixtures as epitomized by the “running on oobleck” effect. This effect describes the ability of a person to run over the surface of a mixture of standard cornstarch and water without sinking (within a certain range of volume fractions); however, if a person walks over the same mixture, they rapidly sink to the bottom.

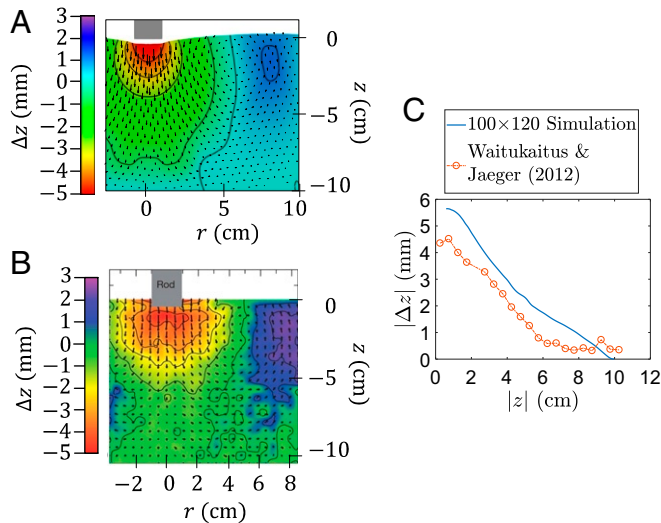


Fig. 4. (A) Plot of z -displacement field and contours for simulated cornstarch–water/glycerol mixture ($\eta_0 = 7$ cP) after $\Delta t = 60$ ms for aluminum rod impacting 10 cm deep, radially symmetric mixture ($r_{\text{mixture}} = 10$ cm; $r_{\text{rod}} = 0.95$ cm) at 1.0 m/s. Simulation is performed on 100×120 element grid with $1 \text{ mm} \times 1 \text{ mm}$ resolution. The red and yellow region near the impactor represents a region of jammed material comoving with the rod. (B) Plot of experimental z -displacement field taken from ref. 36. Reprinted with permission from ref. 36. (C) Comparison of displacement $|\Delta z|$ vs. depth z directly below the impacting rod after $\Delta t = 60$ ms for 100×120 element simulation and results reported in ref. 36. The roughly linear profile observed in the simulated results and in the experiment is indicative of the steadily growing, solid plug of material below the rod.

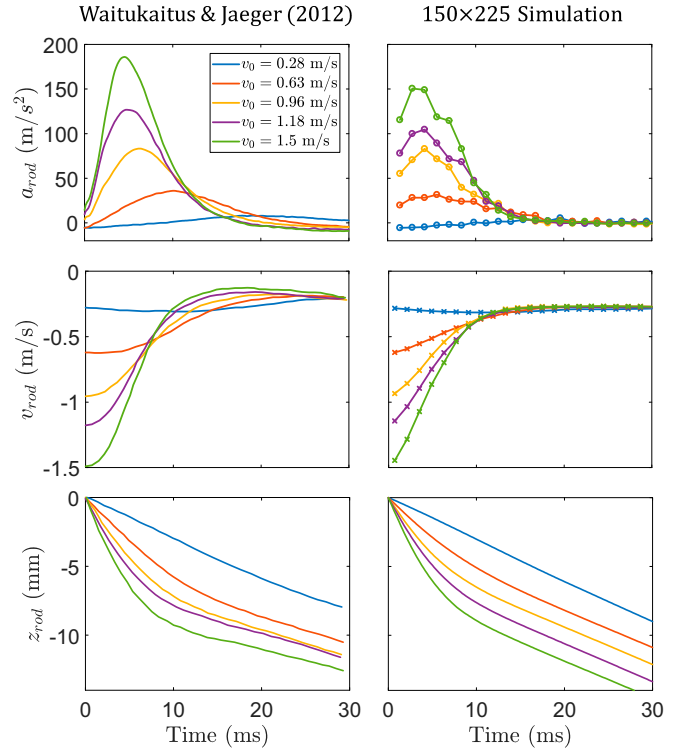


Fig. 5. Comparison of rod motion (acceleration a_{rod} , velocity v_{rod} , and position z_{rod}) from large-container experiments ($\eta_0 = 1$ cP) in ref. 36 to simulated rod dynamics from axisymmetric, 150×225 element Cartesian-grid simulation. Several initial rod velocities v_0 are shown (see legend). Simulated rod-velocity and -acceleration profiles are obtained from the simulated position profiles using a first-order finite-difference scheme. The length of the finite-difference stencil is chosen to remove the effects of elastic waves rebounding off of the simulation boundaries.

In the next section, we will show that the model proposed in this work is capable of capturing this behavior; however, we must first express the time-dependent, 3D form of our model. The complete set of governing equations (many of which are unchanged from the model developed in ref. 18) can be found in *SI Appendix*, Table S1. Here, we focus only on the granular-phase stress components $\tilde{\sigma}_{ij}$, which obey a Maxwell-like model. We assume that the deformation rate tensor, $D_{ij} = \frac{1}{2}(\partial v_{si}/\partial x_j + \partial v_{sj}/\partial x_i)$, can be decomposed into an elastic part $\tilde{D}_{ij}^e$ and plastic part $\tilde{D}_{ij}^p$ with v_{si} the components of the granular-phase velocity. This allows us to express the rate of change of the granular-phase stress $\tilde{\sigma}_{ij}$ as a function of the elastic deformation rate, the granular shear modulus G and bulk modulus K . The components of the plastic part of the deformation rate tensor can be expressed using the form given in ref. 18 as follows,

$$\tilde{D}_{ij}^p = \frac{\dot{\gamma}^p}{2\bar{\tau}}(\tilde{\sigma}_{ij} + \tilde{p}\delta_{ij}) + \frac{1}{3}(\beta\dot{\gamma}^p + \dot{\xi}_1 + \dot{\xi}_2)\delta_{ij}, \quad [10]$$

with,

$$\bar{\tau} = \sqrt{\frac{1}{2}(\tilde{\sigma}_{kl} + \tilde{p}\delta_{kl})(\tilde{\sigma}_{kl} + \tilde{p}\delta_{kl})}, \quad \tilde{p} = -\frac{1}{3}\tilde{\sigma}_{kk}, \quad [11]$$

δ_{ij} , the Kronecker delta function; and $\dot{\gamma}^p$, the scalar (equivalent) plastic shear rate that generalizes $\dot{\gamma}$ to unsteady, 3D flows. The expression in Eq. 10 captures the 2 mechanisms of granular flow: shearing and dilation. The second term in Eq. 10 (representing dilation of the granular phase) captures the effects of Reynolds dilation ($\beta\dot{\gamma}^p$), pure expansion ($\dot{\xi}_1$), and pure compaction ($-\dot{\xi}_2$).

Table 6. Material parameters for simulations shown in Figs. 6 and 7

η_0 [mPa·s]	ρ_s [kg/m ³]	ρ_f [kg/m ³]	G [Pa]	K [Pa]	K_0
4.6	1,620	1,620	3.8×10^7	8.3×10^7	0.2

β , $\dot{\gamma}^p$, and $\dot{\xi}_2$ are functions of the granular stress $\tilde{\sigma}_{ij}$, a , and ϕ_m ; $\dot{\xi}_1$ is a function of $\tilde{\sigma}_{ij}$ and intergranular cohesion. Implementation of the changes made in our proposed model is achieved by replacing a and ϕ_m in the functions for β , $\dot{\gamma}^p$, and $\dot{\xi}_2$ with Eqs. 3 and 4 and substituting $\dot{\gamma}^p$ in place of $\dot{\gamma}$ in Eq. 5.

Results

In this section, we show that the fully 3D, time-dependent, 2-phase form of our model accurately reproduces the observed transient behavior of cornstarch–water mixtures in annular shear and impact experiments as well as the running on oobleck effect. To perform this analysis, we implement our model in the numerical framework shown in ref. 18. This numerical framework is an adaptation of the material point method (MPM) for simulating mixtures as 2 overlapping continua. This version of MPM is very similar to the method shown in ref. 37 and differs from the original method described in ref. 38 in that each continuum phase of the mixture is represented by independent sets of material point tracers. These material point tracers act as quadrature points for solving the weak form of the momentum balance equations on a static background simulation grid. In addition, these tracers move with the continua that they represent. To understand the results shown in this work, it suffices to understand that the velocities and strain rates of the continua are represented on the nodes of the simulation grid, while displacements and stresses are represented on the material point tracers.

As in ref. 18, we assume that the granular phase of the mixture is “elastically stiff” (i.e., elastic moduli G and $K \gg \tau$), so that any significant deformation of the material comes from plastic flow ($\dot{\gamma}^p$). In all of the simulations reported in this work, we choose values of the fluid bulk modulus and the moduli G and K to be large enough such that any effects of elasticity do not alter the flow solution (to do this, we require that all elastic deformations are smaller than $\sim 1\%$) but not so large that they significantly impact the quality of the numerical calculations. In this way, we ensure that our results are representative of elastically stiff mixtures while also avoiding well-known numerical artifacts in MPM (see kinematic locking in ref. 39 and the ringing instability in ref. 40) that affect the smoothness of the spatial stress field. Further discussion of this point can be found in *SI Appendix*, along with a brief note about the validity of the elastically stiff assumption.

Dynamic Shear Jamming. Here, we show that our model can capture the reported propagation of “shear jamming fronts” in annular-shear experiments; in particular, we are interested in the starkly different mixture responses reported at different applied shearing rates. One example of this phenomenon can be found in ref. 13, which reports several experiments involving density-matched mixtures of cornstarch, water, glycerol, and CsCl. In the annular-shear cell described in that work, an applied inner wall velocity of $u_i = 0.84$ mm/s produces shearing flow that rapidly approaches the diffusive-flow profile expected from a Newtonian fluid; however, when the applied inner wall velocity is multiplied by a factor of 100, the mixture begins jamming at the inner wall. This jammed region is characterized by nearly rigid rotation (Fig. 3) and a well-defined boundary that propagates outward with a near-constant rate.

We recreate the reported experiment with our continuum model using an axisymmetric form of MPM on a 36×50

element Cartesian grid with $1 \text{ mm} \times 1 \text{ mm}$ resolution. The upper surface of the simulated mixture is modeled as “free” (zero traction), the inner and outer surfaces are modeled using no-slip, rigid boundaries with assigned velocity (according to the relevant experimental parameters), and the bottom surface is modeled as a rigid wall with slip (zero friction). Our model is calibrated to the experiments using the steady-flow parameters for the cornstarch–water/glycerol experiments of ref. 25 (Tables 2 and 3) and material parameters as in Table 4. The only parameters that were tuned to fit experimental observations were K_0 and ϕ_0 . The values chosen for η_0 , d , and ρ are informed by the materials used in the mixtures (d in all simulations is chosen to be $5.85 \text{ } \mu\text{m}$; ref. 41). K_3 and K_4 are chosen according to the findings of ref. 18 (4.715 and 0, respectively). The simulation results in Fig. 3 show excellent agreement with experimental observations neglecting wall slip or the effects of boundary breakdown that were not implemented in our model.

Impact-Activated Solidification. In this section, we show that our model accurately captures the solidification of cornstarch–water/glycerol mixtures under impact. Similar to the shear jamming observed in the previous section, this “impact-activated solidification” results in 2 strikingly different material responses. At high impact speeds, the mixture responds like a solid, sometimes causing the impactor to bounce (36); at low impact speeds,

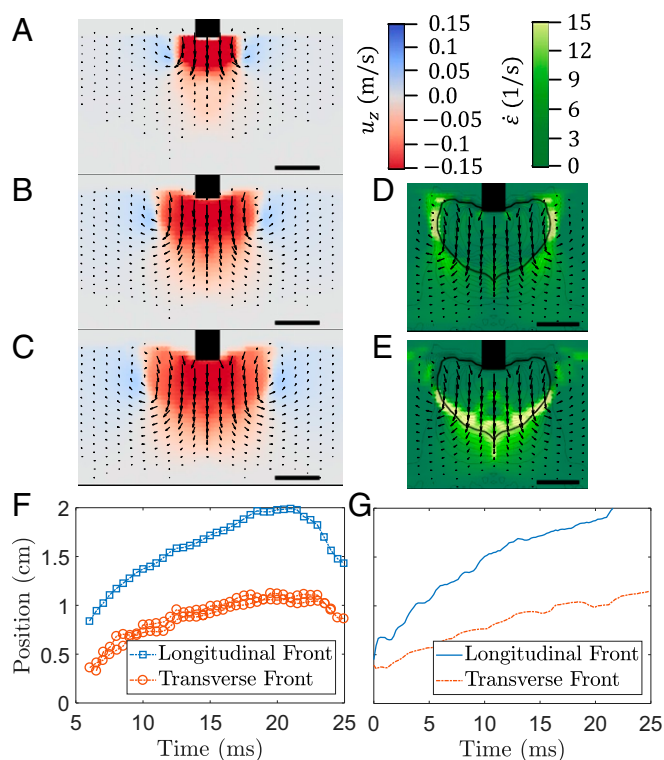


Fig. 6. Visualization of flow field and front motion from axisymmetric MPM simulation of intruder. Black bars provide 1-cm length scale. The driving velocity v_0 of the intruder is 0.175 m/s and mixture volume fraction ϕ is 0.47 for results shown in A–E. The driving velocity v_0 is 0.2 m/s and mixture volume fraction ϕ is 0.46 for results shown in F and G. (A) Flow field colored by vertical component of velocity u_z at $t = 6.2$ ms. (B) Flow field colored by u_z at $t = 13.2$ ms. (C) Flow field colored by u_z at $t = 20$ ms. (D) Visualization of strain rate $\dot{\epsilon} = |D_{rz}|$ at $t = 20$ ms with contours of velocity u_z ($0.5v_0$ in bold). (E) Visualization of strain rate $\dot{\epsilon} = |D_{zz}|$ at $t = 20$ ms. (F) Experimental measurement of front position (defined by $u_z = 0.5v_0$) in the longitudinal and transverse directions. (G) Simulated front position in the longitudinal and transverse directions.

the mixture responds like a fluid, flowing around the intruder with ease. In addition, as predicted in ref. 36 and directly observed in refs. 14 and 42, at high impact speeds, a “solid plug” forms at the impact site and propagates outward (another type of jamming front).

We begin analyzing the response of our (fully dynamic) model under impact by recreating several of the experiments reported in ref. 36 where an aluminum rod ($r_{\text{rod}} = 0.95$ cm, $m_{\text{rod}} = 0.368$ kg) is launched at a mixture of cornstarch, water, and glycerol. Measurement of the rod motion was taken visually and with an accelerometer, and mixture displacement fields were visualized using X-ray imaging. We calibrate our model to the experiments using the steady-flow parameters for ref. 25 in Tables 2 and 3 and material parameters in Table 5 (d , K_3 , and K_4 are chosen as in the previous section). The simulations are performed using axisymmetric MPM on Cartesian grids with $1 \text{ mm} \times 1 \text{ mm}$ resolution. The upper surface of the simulated mixture is modeled as free (except for frictional contact with the simulated rod; ref. 43), while the lower and outer surfaces are bounded by no-slip, rigid walls. The surfaces of the rod are modeled as free (except for the region of contact with the simulated mixture). We only tune K_0 to accurately reproduce the displacement fields (Fig. 4) and rod dynamics (Fig. 5) reported in ref. 36. By using the full continuum momentum balance, our model naturally captures observed added mass effects from material inertia.

The behavior of our model during impact shows remarkable similarity to experiments. The stiff response of the mixture at high impact speeds (as shown by the acceleration curves in Fig. 5) is contrasted by the steady sinking that is observed once the rods have slowed (as shown by the velocity curves in Fig. 5). The X-ray imaging performed in ref. 36 allowed observation of the final displacement field within the mixture (Fig. 4) and the results are indicative of a “solidification front” propagating during impact.

We continue the analysis of our model during impact by recreating the controlled-intrusion experiments described in ref. 14 with a focus on measuring the rate of propagation of the solidification front as the intruder enters the mixture. In these

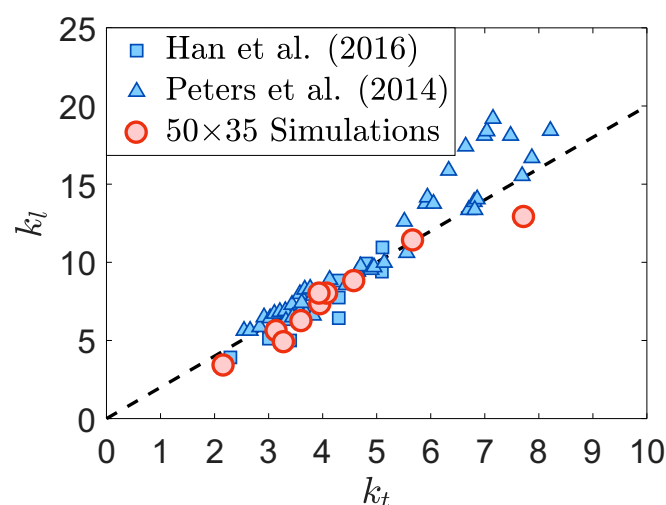


Fig. 7. Scatter plot of front propagation factors k_l and k_t from ref. 14 (blue squares), ref. 42 (blue triangles), and from axisymmetric MPM simulations on a 50×35 element Cartesian grid (red circles). The black dashed line is the proposed front propagation ratio reported in ref. 14, $k_l/k_t \approx 2$. The simulation results shown were run at volume fractions ϕ of [0.46, 0.47, 0.48, 0.49, 0.50] and intruder velocities v_0 of [200 mm/s, 500 mm/s]. Measurement of simulated front propagation speed is performed using the method described in ref. 14.

Table 7. Material parameters for simulations shown in Figs. 8 and 9

η_0 [mPa·s]	ρ_s [kg/m ³]	ρ_f [kg/m ³]	G [Pa]	K [Pa]	K_0
1.0	1,620	1,620	3.8×10^5	8.3×10^5	1.0

experiments, a rod was driven with a constant velocity v_0 into a density-matched cornstarch–water/glycerol/CsCl mixture, and the internal flow of the mixture was visualized using high-speed ultrasound imaging. We calibrate our model to the experiments using the parameters for ref. 25 found in Tables 2 and 3 and material parameters in Table 6 (d , K_3 , and K_4 are chosen as before). The simulations are run using axisymmetric MPM on a 50×35 element Cartesian grid with $1 \text{ mm} \times 1 \text{ mm}$ resolution using the same boundary conditions as in the previous impact simulations.

Tuning K_0 , we are able to closely match the reported flow fields from ref. 14 (Fig. 6). As the intruder enters the mixture, a solid plug of material forms below the intruder and grows outward in all directions. This solid plug is characterized by nearly rigid vertical motion with a well-defined boundary that grows with near-constant rate. The rate of propagation of this boundary in the transverse v_{ft} and longitudinal v_{fl} directions is often quite different (as shown in Fig. 6 *F* and *G*); however, these rates appear to obey a particular scaling rule that is independent of volume fraction: $(v_{fl} - v_0)/v_{ft} \approx 2$. Remarkably, our simple 3D model naturally captures this reported scaling (Fig. 7; results are shown in terms of the transverse and longitudinal front propagation factors $k_t = v_{ft}/v_0$ and $k_l = v_{fl}/v_0 - 1$, respectively). This close agreement of model predictions to experimental observations is unmatched by any other models in the literature.

Running on Oobleck. In this section, we demonstrate that our model can capture the running on oobleck effect by simulating a spoked elastic wheel driving over the surface of a density matched cornstarch–fluid mixture similar to those described above. The simulation is run using 2D MPM on a 150×75 element ($2 \text{ m} \times 1 \text{ m}$) periodic Cartesian grid using the steady-flow parameters for ref. 25 in Tables 2 and 3 with material parameters in Table 7 (d , K_3 , and K_4 are chosen as in the previous sections). The upper surface of the simulated mixture is modeled as a free boundary (except for frictional contact with the wheel; ref. 43), while the lower surface is modeled as a no-slip, rigid wall. The 4-spoked elastic wheel used in this simulation has a radius of 15 cm, spoke width of 5 cm, and density of $3,000 \text{ kg/m}^3$ (weighing $\sim 85 \text{ kg/m}$) and is driven with constant rotation rate ω starting on the mixture’s surface (Figs. 8 and 9).

As shown in Figs. 8 and 9, respectively, the wheel driving at $\omega = 1.0 \text{ s}^{-1}$ will rapidly sink, while the wheel driving at $\omega = 10.0 \text{ s}^{-1}$ will run along the surface. This difference in behavior is caused by the same mechanism explored in the previous section. In the slow case ($\omega = 1.0 \text{ s}^{-1}$), the spokes of the wheel are traveling too slowly to produce a solid-like response; the entire wheel slips into the mixture as it would in a fluid. However, in the fast case ($\omega = 10.0 \text{ s}^{-1}$), when the spokes of the wheel impact the mixture surface, they cause rapid solidification of the surrounding mixture—see the spatial f field in Figs. 8 and 9 (plotted using a postprocessing routine from ref. 44). This allows that spoke to support the weight of the wheel until the next spoke strikes the surface further on; the wheel runs along the top of the mixture. Analysis of the stresses within the spoke of the fast wheel show peak vertical stresses of around 20 kPa after the spoke strikes the mixture, much larger than the approximate Archimedes pressure of 3 kPa due to the displacement of the mixture. (Video of these simulations can be found in [Movies S1–S4](#).)

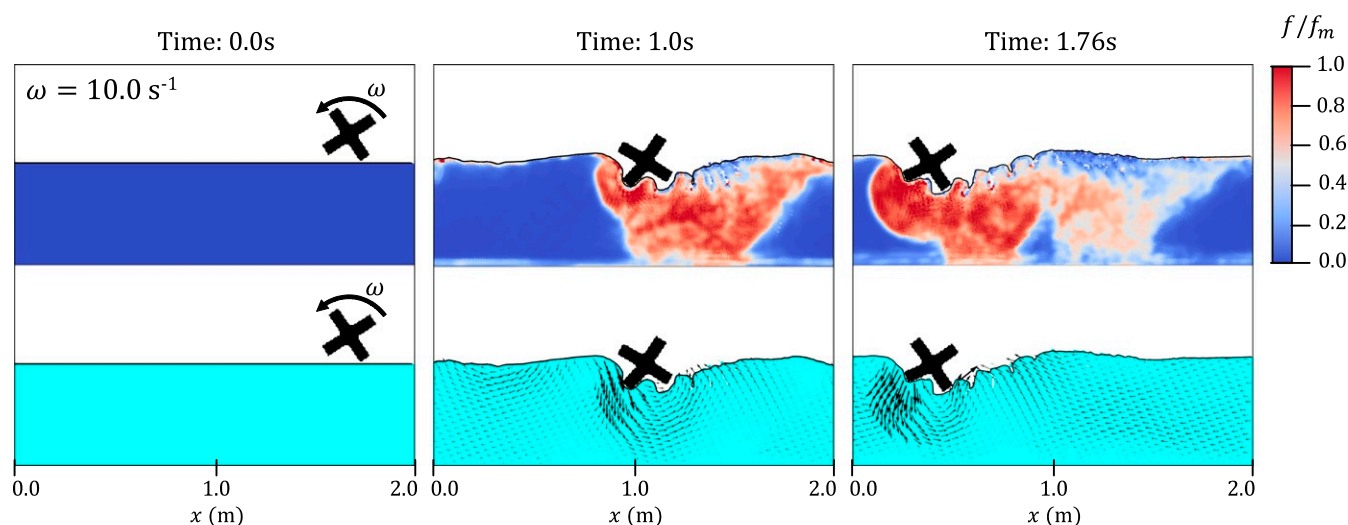


Fig. 8. Visualization of structural variable f and velocity vector field in simulated cornstarch–fluid mixture for wheel driven at $\omega = 10.0 \text{ s}^{-1}$. The value of f is used to color the material point tracers representing the cornstarch phase of the mixture. The black vectors representing the velocity field are drawn with component lengths $l_i = 1.215v_i \sqrt{R/g}$. A black contour line is added to aid in visualizing the free surface of the mixture and is associated with the volume fraction $\phi = 0.1$ as represented on the background grid. This contour does not exactly represent the free surface, as accumulated numerical errors cause disagreement between material point volume fractions and effective background grid volume fractions; these numerical errors are caused by topological changes along the surface of the material but do not significantly affect the qualitative behavior reported.

Conclusion

The model proposed in this work shows remarkable accuracy in predicting the behavior of repulsive-particle suspensions in both steady and unsteady flows. By combining a model for the evolution of granular microstructure with the governing equations for fluid–sediment mixtures presented in ref. 18, we have created a single constitutive model for chemically stable, hard, frictional particles suspended in viscous fluids. In addition, implementation of this model in the numerical framework proposed in ref. 18 allows simulation of the coupled behavior of particles and fluid through a wide range of particle sizes, volume fractions, and flow regimes (including independent motion of each phase and interaction with solid bodies). We have demonstrated that this model captures the landmark features of these mixtures, includ-

ing 1) DST and CST, 2) propagation of shear jamming fronts, 3) the propagation of impact-activated jamming fronts, and 4) the running on oobleck effect.

There are a number of areas remaining for further research and improvement of this model. In this work, we have focused on the time-average behavior of rheologically chaotic flows; however, the basic model may be used to predict the time-accurate behavior of these flows through a fabric-tensor contribution to the evolution of f (as noted in refs. 1 and 32). In addition, future work will need to address the dependence of τ^* , η_B , and K_0 on changes in particle and fluid material properties such as η_0 , d , ρ , and θ (with θ the mixture temperature; ref. 45) and the additional dependence of K_0 on stress scale $\bar{\tau}$ and shearing rate $\dot{\gamma}$. In *SI Appendix*, we describe a simple relaxation experiment

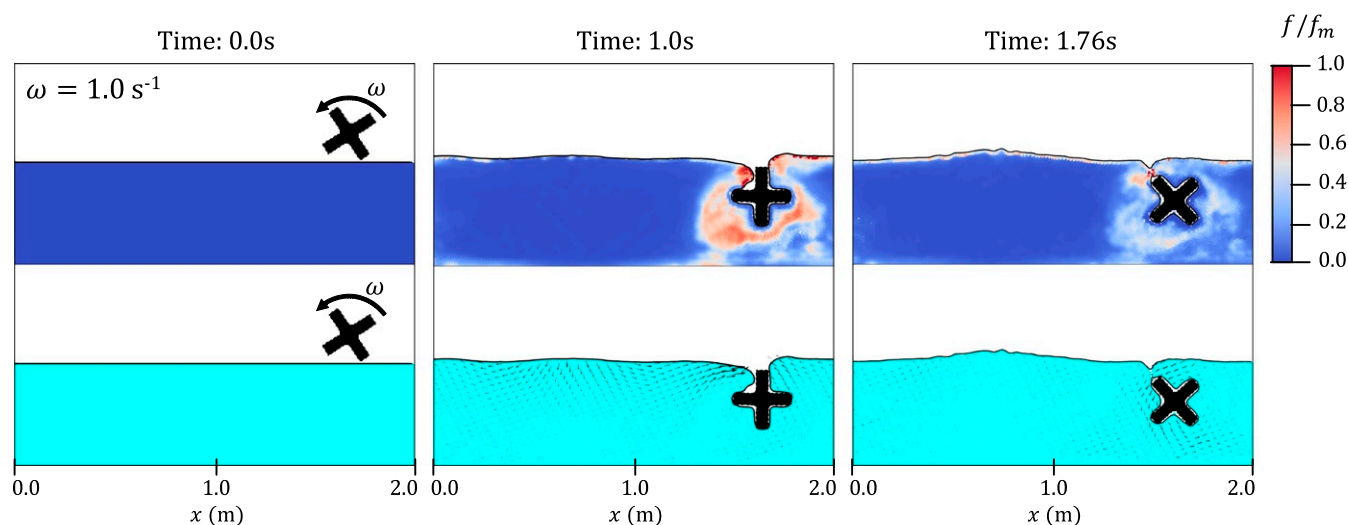


Fig. 9. Visualization of structural variable f and velocity vector field in simulated cornstarch–fluid mixture for wheel driven at $\omega = 1.0 \text{ s}^{-1}$. The value of f is used to color the material point tracers representing the cornstarch phase of the mixture. The black vectors representing the velocity field are drawn with component lengths $l_i = 1.215v_i \sqrt{R/g}$. A black contour line is added to aid in visualizing the free surface of the mixture and is associated with the volume fraction $\phi = 0.1$ as represented on the background grid.

that could help determine these dependencies. Numerically, we have shown that our model can be implemented in MPM (18) and accurately reproduce the time-dependent behavior of these highly nonlinear mixtures in nontrivial geometries. Although our initial tests are promising, further work should also be done to curb known issues in the numerical framework such as kinematic

locking (39) and the ringing instability (40), which can cause spurious artifacts in certain parameter ranges if not dealt with carefully.

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