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Shear thickening of dense suspensions: The role of friction

Shear thickening of particle suspensions is caused by a transition between lubricated and frictional contacts between the particles. Using three-dimensional (3D) numerical simulations, we study how the interparticle friction coefficient ($\mu$) and hence the microstructure and rheology of dense shear thickening suspensions. We propose expressions for $\mu$ in terms of distance to jamming for varying shear stresses and $\mu_m$ values. We find $\mu$ to be rather insensitive to interparticle friction, which is perhaps surprising but agrees with recent theory and experiments. Unifying behaviors were observed between the average coordination numbers of particles across a wide range of viscous numbers and $\mu_m$ values.

I. INTRODUCTION

Understanding the rheological properties of shear thickening suspensions is scientifically challenging and highly relevant from the viewpoint of several applications.\textsuperscript{1-12} The phenomenon of shear thickening,\textsuperscript{11} in which the viscosity increases with increasing shear rate and shear stress is attributed to the formation of frictional contacts between the particles as suggested by computational results\textsuperscript{2-14} and confirmed by experiments.\textsuperscript{15-19} Shear thickening suspensions can be characterized by their macroscopic friction coefficient $\mu$, given by $\mu = \sigma_{\text{shear}}/P$, with $\sigma_{\text{shear}}$ the shear stress and $P$ the confining pressure. Using suspensions under constant confining pressure, Boyer et al.\textsuperscript{17} demonstrated that $\mu$ is a unique function of a viscous parameter $I_v$ defined as $I_v = \eta_f \gamma/P$, where $\eta_f$ and $\gamma$ are the fluid viscosity and the shear rate, respectively. They observe similar $\mu(I_v)$ behavior for different materials (polystyrene, PMMA) and particle sizes. Gallier et al.\textsuperscript{20} studied $\mu(I_v)$ rheology in simulations for $\phi < 0.45$ ($\phi$ being the particle volume fraction) and their simulations agree quantitatively with the experimental results. Recent studies by Chévremont et al.\textsuperscript{21} and Trulsson et al.\textsuperscript{22} have shed light upon the influence of the microscopic interparticle friction coefficient $\mu_m$ on $\mu$, viscosity, and the jamming volume fraction. However, a more detailed analysis of $\mu$ and associated changes in the microstructure of the suspension is needed to further understand the behavior of the macroscopic friction coefficient $\mu$ and notably its relation with the microscopic friction coefficient $\mu_m$. Here, we perform 3D numerical simulations of dense shear thickening suspensions with varying interparticle friction coefficients to study the associated changes on $\mu$. Based on recent results on constitutive relationships for shear thickening systems,\textsuperscript{12,23} we propose analytic expressions for $\mu$ in terms of the distance to jamming ($\phi_m - \phi$, where $\phi_m$ is the jamming volume fraction) for constant volume systems with varying pressure, shear stress, and $\mu_m$ values. Using the average coordination number as a parameter, the microstructure of the particles in the system is analyzed to assess its influence on $\mu$. Finally, simulations of nonspherical particles are performed to study the effect of nonsphericity on the behavior of the macroscopic friction coefficient.

II. METHODS

The numerical simulations were performed using the simulation framework SuSi.\textsuperscript{24} We use the Lattice Boltzmann Method (LBM) based fluid to simulate the fluid field and Lagrangian particles as the solid phase. The fluid-particle interactions are modeled with the Noble Torczynski Method.\textsuperscript{25} Lubrication forces are calculated explicitly at particle gaps smaller than the LBM lattice spacing. Adaptive refinement of time steps is performed in order to ensure...
numerical stability and accuracy, as the interparticle forces diverge at small particle gaps. The contact normal force \( F_{ep} \) between particles is calculated from the overlap of a contact repulsion layer of specified thickness \( d_c \approx 0.001R \), where \( R \) is the mean radius of particles,

\[
F_{ep} = \begin{cases} 
-c_0 \frac{(d - d_c)^2}{dd_c^2} c_n, & d \leq d_c, \\
0, & \text{otherwise}, 
\end{cases}
\]

where \( c_0 \) is the repulsion coefficient, \( d \) is the gap between the particles, \( d_c \) is the repulsion layer thickness, and \( c_n \) is the connecting unit vector between the particles. The static and kinetic friction between particles is modeled as proposed by Luding.\(^{27}\) Upon initiation of frictional contact between particle pairs, a linear spring of length \( \xi \) is initialized between the closest surface points to model static friction and is updated using the relative tangential velocity between the two contacting surface points. The maximum static friction is \( F_s \leq \mu_0 |F_{norm,frc}| \), as given by Coulomb’s Law. The spring force \( F_{ep} \) is applied if the amplitude of \( F_{ep} = -k d \) is smaller than the maximum possible static friction force \( F_s \). Kinetic friction \( F_k = \mu_k |F_{norm,frc}| \) is applied as a tangential force at the surface points if \( F_{ep} \) exceeds \( F_s \).

For kinetic friction, the static friction spring length is rescaled so that \( F_{ep} = F_k \). In our simulations, we keep \( \mu_k = \mu_m \), where \( \mu_m \) is referred to as the microscopic friction coefficient.

The interacting particles are deemed frictional based on a critical load model,\(^{27} \) where two particles are considered to be in friction if the normal force \( |F_{ep}| \) between the contacting particles exceeds a threshold value \( (F_{CL}) \). The static and kinetic friction is based on the normal force for friction \( (F_{norm,frc}) \), calculated as\(^{12} \)

\[
F_{norm, frc} = \begin{cases} 
|F_{ep}| - F_{CL}, & \text{if } |F_{ep}| \geq F_{CL}, \\
0, & \text{otherwise}. 
\end{cases}
\]

For the simulations discussed in Sec. III, a 96 \( \mu \text{m} \times 64 \mu \text{m} \times 96 \mu \text{m} \) system is used, which contains \( \approx 650 \) particles for \( \phi = 0.56 \). The particles have a mean diameter of 8 \( \mu \text{m} \) with a standard deviation of 0.4 \( \mu \text{m} \) to avoid crystallization. The particles are neutrally buoyant in the suspending fluid, which mimics water (fluid viscosity \( \eta_f = 1.002 \times 10^3 \text{ Pa s} \), density \( \rho_f = 1000 \text{ kg/m}^3 \)). The simulated systems have a characteristic stress for frictional contacts, given by \( \sigma_0 = F_{CL}/(6\pi R^2) \), where \( R \) is the average particle radius. \( F_{CL} \) is 0.2 nN in all simulations, and the tangential spring constant \( k = 0.0361 \text{ N/m} \). For the performed analysis, we choose instances of the system with the average shear stress greater than \( \sigma_0 \) so that frictional interactions are significant.

III. RESULTS AND DISCUSSION

A. Viscosity and normal stress differences

The viscosity of suspensions increases with the particle volume fraction and diverges when \( \phi \to \phi_m(\mu_m) \) where \( \phi_m \) is the jamming volume fraction associated with \( \mu_m \). Scalings such as the Maron-Pierce expression \(^{18} \) \( \eta_\text{f} = (1 - \phi/\phi_m)^{-2} \) and the Krieger—Dougherty expression \(^{19} \) \( \eta_\text{f} = (1 - \phi/\phi_m)^{-2.5\phi_m} \) have been used to describe the viscosity divergence with increasing \( \phi \). We compare our simulation results across various \( \mu_m \) values against previous experiments.\(^{12,20} \)

B. Viscous number rheology

The macroscopic friction coefficient \( (\dot{\gamma}) \) of suspensions is characterized by the viscous number \( (I_v) \) of the suspension flow. \( I_v \) is defined as

\[
I_v = \eta_\text{f} \dot{\gamma}/P,
\]

\( \text{FIG. 1.} \) Relative viscosity \( \eta_f \) vs particle volume fraction \( \phi \), normalized by the jamming volume fraction \( \phi_m \) for our simulations (colored symbols), compared against: Maron-Pierce \((\eta_f = (1 - \phi/\phi_m)^{-2})\) scaling, Krieger and Dougherty \((\eta_f = (1 - \phi/\phi_m)^{-2.5\phi_m})\) scaling \( \sigma_2 \) and \( \sigma_3 \) are the normal stresses in the shear gradient and vorticity directions) has been a topic of debate since there are considerable differences in the available experimental data.\(^{23,24,30} \)\(^{11} \)\(^{25} \)\(^{26} \)\(^{31} \)\(^{32} \)\(^{33} \) N_2 is generally smaller in magnitude compared to \( N_2 \). The behavior of \( N_2 \) is less well understood since even the algebraic sign of \( N_2 \) appears to depend on experimental conditions.\(^{26} \) We compare \( N_1 \) observed in our simulations against the existing literature in Fig. 2(b). \( N_1 \) obtained in our simulations closely resembles the works by Royer et al.\(^{17} \) who observed a negative to positive transition for \( N_1 \) with increasing \( \phi \). Recent studies\(^{17} \) have made advances to address some of the outstanding questions about \( N_1 \).
where \( \eta \) is the fluid viscosity, \( \dot{y} \) is the shear rate, and \( P \) is the pressure in the system. The viscous number can be seen as the ratio of the internal time scale of microscopic particle rearrangements in a viscous system (\( \eta/\dot{y} \)), to the macroscopic flow time scale (\( 1/\dot{y} \)). Boyer et al.\(^{11} \) used pressure imposed flows to study variation in \( \mu \) with \( I_v \), where systems of hard spheres were sheared at constant pressure (\( P \)) and shear rate (\( \dot{y} \)) while the system was allowed to dilate (changing \( \phi \)) in order to keep \( P \) constant. They demonstrated that \( \mu \) of suspensions is the sum of contact (\( \mu_c \)) and hydrodynamic (\( \mu_h \)) stress contributions, as shown in the following:

\[
\mu(I_v) = \mu_c + \frac{\mu_h - \mu_c}{1 + I_0/I_v + \frac{5}{2} \phi_m I_v^2 / \mu_c}
\]

where, \( \mu_c \) is the limit of the particle contact contribution to the macroscopic friction (\( \mu_c \)) at vanishing viscous numbers, and \( \mu_h \) is the maximum \( \mu_h \) at \( I_v \to \infty \) as observed in granular flows. \(^{10,11} I_0 \) represents the scale over which \( \mu_c(I_v) \) changes and is observed to be constant for a given particle shape. \( \phi_m \) is the jamming volume fraction. \( \mu_c(I_v) \) is designed to reproduce the Einstein viscosity at low \( \phi \) and be nonsaturating at high \( I_v \). Here, simulations of constant \( \phi \) and \( \dot{y} \) with varying \( P \) are performed to study \( \mu(I_v) \). In this study, we define \( P \) as the average of the diagonal elements of the stress tensor in the system, i.e., \( P = \sum_{i=1}^{3} \sigma_{ii}/3 \). We systematically vary the microscopic friction coefficient \( \mu_c \) and compare to the predictions of \( \mu(I_v) \) rheology [Eq. (4)] to see if the constant \( \phi \) and \( \dot{y} \) simulations conform to the predictions of \( \mu(I_v) \) rheology.

At vanishing \( I_v \), we find high corresponding \( \phi \) values similar to that in experiments.\(^{11} \) Under constant \( \phi \) settings, the range of \( I_v \) values accessible for each \( \phi \) value is limited [as seen in Fig. 3(b)].\(^{31,40} \) Vertical and horizontal error bars correspond to variation in \( \mu \) and \( I_v \), in the data, in each interval.\(^{31} \) Variation in \( \phi \) vs. viscous number \( I_v \). Dots represent simulation results, and the line represents results from Boyer et al.\(^{11} \) Error bars represent the range of \( I_v \) values observed for a given \( \phi \).

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**FIG. 2.** Ratio of the normal stress difference to the shear stress (a) \(-\sigma_2/\sigma_{22}\) and (b) \(N_1/\sigma_{22}\) as a function of \( \phi \) for various \( \eta_{in} \) values. Simulations (colored symbols) are compared against experimental results of Dai et al.,\(^{24} \) Doucai et al.,\(^{36} \) Couturier et al.,\(^{25} \) Zarraga et al.,\(^{11,20} \) Singh and Nott,\(^{20} \) and Royer et al.\(^{32} \) simulation results of Gallier et al.\(^{11} \) and the fits suggested by Dai et al.\(^{24} \) Relevant information about the particle diameter \( d \) and \( \mu_c \) are shown in the legends. Results from simulations are taken over a range of \( \sigma_{22}/\sigma_{12} < 0.01, 100 \). The values of \( N_1 \) have significant fluctuations, and the averaged values are presented.

**FIG. 3.** (a) Macroscopic friction coefficient \( \mu \) vs viscous number \( I_v \) comparison between simulation and the model. Dots represent \( \mu \) prediction from simulations of \( \phi \) corresponding to its color. The dashed line shows the \( \mu(I_v) \) prediction from Eq. (4) with \( \mu_c = 0.34 \) (minimum \( \mu \) observed), \( \mu_2 = 0.7 \), and \( I_0 = 0.009 \) providing a good fit to the simulation results. The microscopic friction coefficient \( \mu_c = 0.5 \). Triangles represent the experimental results from Boyer et al.\(^{11} \) Vertical and horizontal error bars correspond to variation in \( \mu \) and \( I_v \) in the data, in each interval. (b) Variation in \( \phi \) vs viscous number \( I_v \). Dots represent simulation results, and the line represents results from Boyer et al.\(^{11} \) Error bars represent the range of \( I_v \) values observed for a given \( \phi \).
experiments. The variation in $\phi$ with $I_v$ is shown in Fig. 3(b), along with the experimental observation from Boyer et al.\cite{Boyer2019} The simulations show good agreement with the previous experimental results and the theoretical scalings.

C. Viscosity variation and effect on macroscopic friction

Viscosity and macroscopic friction coefficient are inherently related. Simple algebraic manipulation of Eq. (3) shows that $\mu$ and $I_v$ are related to the relative viscosity $\eta_r$ as [see Eq. (5), from Boyer et al.\cite{Boyer2019}]

$$\eta_r = \frac{\mu}{I_v}. \tag{5}$$

Figure 4(a) shows the relative viscosity $\eta_r$ ($\eta_r = \eta/\eta_f$, $\eta$ being the suspension viscosity) as a function of the shear rate in our simulations (same system as in Fig. 3) for various particle volume fractions. With increasing $\phi$ and $\gamma$, an increased relative viscosity $\eta_r$ is observed in the system. At lower $\phi$ values ($\phi < 0.56$), we see CST and subsequently DST at higher $\phi$ values. The macroscopic friction coefficient $\mu$ also shows variation with $\phi$ and $\gamma$, as shown in Fig. 4(b). With increasing $\phi$, pronounced reduction in $\mu$ is observed, and when the system undergoes shear thickening, we find $\mu$ to reduce again. Similar observations were made in two dimensional systems by Thomas et al.\cite{Thomas2014}. This happens as a result of the difference in scaling of the pressure compared to $\sigma_{\text{shear}}$ during shear thickening. Viewed from the perspective of viscous number rheology, this is a result of the reduction in $I_v$ during shear thickening as a result of the pressure increasing at a faster rate compared to the increase in $\gamma$ [see Eq. (3)]. This can also be interpreted in terms of distance to jamming, as shown in Sec. III F. As shown in Sec. III F, the closer a system is to jamming (in terms of say, $\phi_m - \phi$), the lower the value of $\mu$. With increasing $\phi$, the value of $\phi_m - \phi$ reduces and the system moves closer to jamming. Consequently, a reduction in $\mu$ is observed. When the system undergoes shear thickening, $\phi_m$ decreases as more particles become frictional [see Eq. (9)] and as a result, $\phi_m - \phi$ reduces further; the system again moves closer to jamming and $\mu$ is reduced again. Next, we compare the variation in $\eta_r$ with $\mu$ in Fig. 4(c). As viscosity increases, we see reduced $\mu$ values. As $\phi$ approaches $\phi_m = 0.59$, $\mu$ approaches $\mu_1$ and $\eta_r$ diverges. The collapse of the data onto the same curve might be surprising but can be rationalized. As shown in Eq. (5), $\eta_r$ is related to $\mu$ and $I_v$. Since $\mu$ is a function of $I_v$ described by Eq. (4), $\eta_r$ is simply a function of $I_v$. By substituting Eq. (4) in Eq. (5), we can describe this relationship quite well, as shown in Fig. 4(c). In Fig. 4(d), we analyze the frictional ($\sigma_{\text{shear}} > \sigma_0$) and nonfrictional ($\sigma_{\text{shear}} < \sigma_0$) states of the same system separately. We see that $\eta_r(\mu)$ follows slightly different curves for frictional and frictionless states in comparison, as frictionless states effectively have a microscopic friction coefficient $\mu_m = 0$. Thus, $\eta_r(\mu)$ would diverge at a lower $\mu_1$ (see the Sec. III E), but since we do not simulate $\phi$ values close to the $\phi_m$ value associated with $\mu_m = 0$ ($\phi_{m(\mu)} = 0.64$), the divergence for the nonfrictional states is not approached.

D. Effect of varying the critical load for friction

Varying the value of the critical load $F_{CL}$ changes the onset force for friction between contacting particles. Consequently, the characteristic stress $\sigma_0$ for friction and shear thickening depends on $F_{CL}$, as $\sigma_0 \propto F_{CL}/R^2$. We present the results of varying $F_{CL}$ over three orders of magnitude in Fig. 5. Changing $F_{CL}$ does not change the results of $\mu(\gamma)$ in viscous number rheology in our constant $\phi$ systems. The viscous number is given by $I_v = \eta_r F/\mu$ and the macroscopic friction coefficient $\mu = \sigma_{\text{shear}}/P$. These quantities change depending on the scaling of $\sigma_{\text{shear}}$, $P$ and $\gamma$ with respect to each other. Prior to or post shear thickening, the scaling of $P$ and $\sigma_{\text{shear}}$ with $\gamma$ does not change, as evidenced by the constant viscosity in these states. Thus, $\mu(\gamma)$, which depends on the ratio of these terms does not change prior or post shear thickening. During shear thickening, the scaling of $P$, $\sigma_{\text{shear}}$ and $\gamma$ changes with respect to each other due to the additional force scaling (compared to the nonfrictional state) due to the progressive increase in the fraction of frictional contacts.\footnote{As a result, variation in $\mu$ and $I_v$ are observed only during shear thickening. This can be readily observed in Fig. 4(b) where the $\mu$ varies during shear thickening and remains constant prior to and post shear thickening.}

![FIG. 4. Relative viscosity $\eta_r$ and macroscopic friction coefficient $\mu$ variation during shear thickening, for $\mu_{m(\mu)} = 0.5$. (a) Relative viscosity $\eta_r$ vs shear rate $\dot{\gamma}$. Shear rate $\dot{\gamma}$ is normalized by $\dot{\gamma}_0 = \sigma_0/\eta_f$. (b) $\mu$ vs normalized $\gamma$. (c) $\mu$ variation with $\mu$, $\mu_1 = 0.34$ is indicated by the dashed vertical line. Black line represents predictions from Eqs. (4) and (5), with parameters the same as that in Fig. 3. Colors represent the same $\phi$ values in (a, b, c). States of the system where $\sigma_{\text{shear}} > \sigma_0$ are chosen in order to avoid nonfrictional states. (d) $\eta_r$, $\mu$ for frictional ($\sigma_{\text{shear}} > \sigma_0$) and frictionless ($\sigma_{\text{shear}} > \sigma_0$) states analyzed separately (note the difference in the divergences).}
E. Effect of varying microscopic friction coefficient

The properties of the particle contacts have significant influence on the rheology of suspensions. Purposeful roughening of particles has shown to increase the viscosity of suspensions in the works of Moon et al.44 and Lootens et al.44 Hsu et al.44 and Lootens et al.44 also found the jamming transition to happen at lower volume fractions upon using rougher particles, as rougher particles have higher microscopic friction coefficients.

The microscopic friction coefficient \( \mu_m \) has a significant impact on shear thickening process. Experimental results from Lootens et al., simulations by Mari et al.,45 and theoretical models by Singh et al.46 show that with decreasing \( \mu_m \), the strength of shear thickening reduces in dense suspensions and at \( \mu_m = 0 \), shear thickening disappears altogether. In order to study the effect of changing \( \mu_m \) on \( \mu \), we perform simulations of 0.01 \( \leq \mu_m \leq 10 \), while keeping all other system parameters the same. This amounts to over 500 individual simulations. In agreement with the literature, we find the microscopic friction coefficient to affect the viscosity significantly, as shown in Fig. 6(a). With lower \( \mu_m \) values, shear thickening observed becomes progressively less pronounced.

Earlier simulation studies of the role of the microscopic friction coefficient \( \mu_m \) in viscous number rheology were performed at large viscous numbers \( (I_v > 0.1) \) with limited overlap between \( I_v \) ranges studied in experiments.47 Here, a larger range of \( I_v \) values is accessed, allowing comparisons with experimental results at lower \( I_v \) values, as shown in Fig. 3. In the previous works of Gallier et al.,48 it was postulated that the master curve for \( \phi(I_v) \) observed by Boyer et al. [Fig. 2(d) in Ref. 11] was possibly not unique across various friction coefficients, as the microscopic friction coefficients in their experiments may not have been varied significantly. Since we are able to access the low \( I_v \) regime, we find evidence that this is indeed true, as shown in Fig. 6(b). With varying \( \mu_m \), we find different curves of \( \phi(I_v) \), each saturating at low \( I_v \) at different \( \phi_m \) values corresponding to the \( \mu_m \) used. Upon normalizing \( \phi \) values with \( \phi_m \), we see in Fig. 6(c) that the results obey Eq. (11) as suggested by Boyer et al.,47 and that this relationship is insensitive to \( \mu_m \). This was also reported by Trulsson et al.45 in their 2D simulations and recently observed by Chéremont et al.44 in their 3D simulations. It should be noted that the works of Chéremont et al. and Trulsson et al. have constant pressure with varying \( \phi \), while our system has constant \( \phi \) with varying pressure.

We now look at the influence \( \mu_m \) has on the \( \mu(I_v) \) relationships. In Fig. 7(a), the simulation results of \( \mu(I_v) \) for various \( \mu_m \) values are shown. At large \( I_v \) values \( (I_v > 0.1) \), \( \mu(I_v) \) is similar for all \( \mu_m \) values. At vanishing \( I_v \) values \( (I_v < 10^{-3}) \), the minimum \( \mu(I_v) \) (i.e., \( \mu_l \)) reduces with decreasing \( \mu_m \), as shown in Fig. 7(c). This observation is in agreement to that made in past simulations of 2D granular and suspension flows.49,50 Interestingly, the relationship between \( \mu - \mu_l \) and \( I_v \) collapses to the same curve for all \( \mu_m \) values in this system [see Fig. 7(b)]. Such a collapse was not observed when spherical particle suspensions studied in this section are compared against nonspherical particle suspensions (see Sec. III H), suggesting that particle shape is a factor here. The change in \( \mu_l \) with \( \mu_m \) follows a sigmoidal relationship, as observed in Fig. 7(c). The collapse of \( \mu - \mu_l \) for \( I_v < 10^{-3} \) with the viscous number is obviously due to \( \mu \) being constant and equal to \( \mu_l \) in this range. Within the intermediate viscous number range \( (10^{-3} \leq I_v \leq 10^{-1}) \) where the particle contact contribution \( [\mu_c, \text{Eq. (4)}] \) to \( \mu \) remains dominant, the variation in \( \mu \) with the microscopic friction coefficient \( \mu_m \) is dictated by the variation in \( \mu_c - \mu_l \) with \( \mu_m \). Seeing that \( \mu_c \) is rather insensitive to microscopic interparticle friction coefficients \( \mu_c \), varies between 0.7 and 0.8 for completely frictionless and frictional particles,
respectively, we estimate that the largest difference in $\mu - \mu_1$ between systems of $\mu_m = 0.01$ and $\mu_m = 10.0$ should be $\approx 0.2$, which agrees with the observed variations in $\mu - \mu_1$ with $\mu_m$ at $I_v \approx 10^{-1}$. For large viscous number range ($I_v > 10^{-5}$), the variations in $\mu$ are dominated by the hydrodynamic component [$\mu_h$ in Eq. (4)], and does not depend on the friction.

The observation that $(\mu - \mu_1)$ as a function of $I_v$ collapses onto the same curve for $0.01 \leq \mu_m \leq 10$, and $(\mu - \mu_1) \to 0$ the closer the system is to jamming, suggests that $\mu - \mu_1$ could be considered as a measure of the distance to jamming for these systems. In other words, the value of $\mu - \mu_1$ is dictated by the “closeness” of a system to jamming. At the same microscopic to macroscopic particle rearrangement time scale ratios (i.e., $I_v$), all systems have the same distance to jamming, regardless of their microscopic friction coefficient. This also entails that if $\mu - \mu_1$ is a measure of the distance of a system from jamming, it should have a mapping to some other measure of distance to jamming, such as $\phi_m - \phi$. We shall explore this in Sec. III F.

**F. Macroscopic friction coefficient and distance to jamming**

In the simulations, a range of shear stresses ($\sigma_{\text{shear}}$), volume fractions ($\phi$) and microscopic friction coefficients ($\mu_m$) are studied. From previous experiments and simulations, we understand the effect of changing each of these parameters on the rheology, especially on the jamming volume fraction ($\phi_m$). Shear thickening is due to the formation of system spanning frictional networks, and the best way to describe this is to look at the fraction of frictional particles in the system. Beyond a characteristic shear stress $\sigma_0$, the fraction of particles in the system that have frictional contacts ($f$) increases until all particles become frictional.

This increase in $f$ with shear stress $\sigma_{\text{shear}}$ can be described as

$$\sigma_0 = F_{\text{CL}} / 6\pi R^2,$$

$$\sigma = \sigma_{\text{shear}} / \sigma_0,$$

$$f = e^{-\ln(1/\phi)},$$

where $R$ is the average radius of the particles, $F_{\text{CL}}$ is the characteristic stress for the onset of friction. Increasing the fraction of frictional particles leads to a lower jamming volume fraction $\phi_m$, as $\phi_m$ for frictional particles is lower than nonfrictional particles. This is a result of the frictional particles requiring a smaller number of interparticle contacts to be arrested in comparison with frictionless particles. The average coordination number for jamming ($Z_J$) in suspensions varies continuously between $Z_J(\mu_m = \infty) = 4$ and $Z_J(\mu_m = 0) = 6$ in suspensions. Increasing the fraction of frictional particles in the system reduces the jamming volume fraction $\phi_m$ from that of a lubricated, nonfrictional suspension ($\phi^\infty$) to that of a frictional suspension ($\phi_f$). $\phi_m(\mu_m)$ is the jamming volume fraction in a suspension with all particles in frictional contact and is a decreasing function of the microscopic friction coefficient $\mu_m$. Hence, the volume fraction associated with jamming varies with $\mu_m$ and the fraction of frictional particles $f$ in the system, and can be described by

$$\phi_m(\sigma, \mu_m) = \phi_f(\mu_m) f(\sigma) + \phi_f^\infty (1 - f(\sigma)).$$

Here, $\phi_f(\mu_m)$ represents the jamming volume fraction when $f = 1$ for a given microscopic friction coefficient $\mu_m$. $\phi_f^\infty$ is the jamming volume fraction when $f = 0$, which is equivalent to a $\mu_m = 0$ (frictionless) state. Changing the microscopic friction coefficient $\mu_m$ influences $\phi_m$, as lowering $\mu_m$ increases $\phi_f$, according to Eq. (10),

$$\phi_f(\mu_m) = \phi_f^\infty (1 - (\phi_f - \phi_f^\infty) e^{-\mu_m/\mu_0}).$$

Here, $\phi_f^\infty$ is the jamming volume fraction at large $\mu_m$ values, and $\mu_0$ is a constant. Boyer et al. proposed a model for $I_v$ in terms of $\phi_m$ and $\phi$ as

$$\phi(I_v) = \phi_m \frac{1 + I_v}{1 + I_v^2},$$

and when substituted in Eq. (4), this gives $\mu$ as a function of $\phi_m$ and $\phi$,

$$\mu(\phi, \phi_m) = \mu_1 + \frac{\mu_2 - \mu_1}{1 + I_v \phi_f(\phi_m - \phi)^2} \mu_h + \frac{\mu_h}{\phi_f} + \left(\frac{\phi_m - \phi}{\phi}\right)^2 \mu_2 \phi_m - \phi^2.$$

Under constant volume settings, the fraction of the frictional contacts varies with shear stress (or shear rate) in the system, which
in turn varies $\phi_m$. We can account for this variation in $\phi_m$ by employing Eqs. (6)–(10). This helps to predict $\phi_m$ in our constant volume system in terms of $\sigma_{\text{shear}}$, and $\mu_m$ which in turn enables an analysis of $\mu$ as a function of $\phi_m - \phi$ (i.e., a distance to jamming metric) and compare against the predictions from Eq. (12).

Figure 8(a) shows the $\mu - \mu_1$ as a function of $\phi_m - \phi$ compiled over a range of $\sigma_{\text{shear}}$, $\phi$, and $\mu$ values. The simulation results show agreement with the predictions from theory outlined in Eqs. (6)–(12). The changes in $\psi$ with $\mu_m$ are taken into account by using their relationship outlined in Eq. (10), as shown in Fig. 8(b). The simulation results agree with the theoretical assumption that, by accounting for changes in $\psi$ with $\sigma_{\text{shear}}$ and $\mu_m$, the values of $\mu$ across different $\sigma_{\text{shear}}$ and $\mu_m$ values collapse to the regime outlined in Fig. 8(a). The change in the frictional jamming volume fraction $\psi$ with $\mu_m$ is shown in Fig. 8(b), along with the model presented in Eq. (10). The results also show that $\mu - \mu_1$ is indeed a measure for the distance to jamming, as suggested in Sec. III E.

G. Microstructure changes

The microscopic friction coefficient plays an important role in the nature of contact networks formed at jamming. The mean coordination number at which the suspension jams ($Z_J$) is inversely dependent on $\mu_m$, as $Z_J(\mu_m = 0) = 6$ and $Z_J(\mu_m = \infty) = 4$. The evolution of $\mu$ with the average coordination number ($Z$) under varying $\mu_m$ values, thus, is of interest. It is also compelling to view $\mu(Z)$ rheology in terms of the evolution of $Z$.

Figure 9(a) shows average coordination number $Z(I_v)$ under various $\mu_m$ values. $Z$ is calculated per particle by counting the number contacts it makes, i.e., cases where $r_{ij} - R_i - R_j \leq d$, where $r_{ij}$ are the distance between the particles and $R_i$, $R_j$ are their radii. Even though the data is compiled from various $\phi$ and $\sigma_{\text{shear}}$ values, $Z(I_v, \mu_m)$ collapses to unique curves depending on $\mu_m$. The maximum coordination number is $Z = 4$ at $\mu_m = 10.0$ and saturates at higher maximum values ($Z_\infty$) with reducing $\mu_m$ as expected from $Z(I_v)$ relationship described before. The low $Z$ values at large $I_v$ sheds light on the insensitivity of $\mu(I_v)$ rheology to changes in $\mu_m$ in these $I_v$ ranges. $\mu(I_v)$ rheology hence is essentially the process of varying coordination numbers between zero and $Z(I_v)$. Upon normalizing $Z$ by $Z(I_v)$, the different $Z(\mu_m)$ curves collapse to a single curve, which can be modeled as

$$Z/Z_f = 1 - (1 + I_v^{\alpha_1})^{-\beta_1},$$

where $\alpha_1 = 0.77$ and $\beta_1 = 0.3$. The variation in $Z_f$ between 6 and 4 depending on $\mu_m$ can also be modeled using the expression

$$Z_f = 6 - 2(1 + I_v^{\alpha_2})^{-\beta_2},$$

where $\alpha_2 = -1.72$ and $\beta_2 = 0.27$. Figure 9(b) shows $Z/Z_f$ as a function of $I_v$, and it can be observed that the data collapses to a single curve, modeled by Eq. (13). The variation in $Z_f$ with $\mu_m$, modeled by Eq. (14) is shown in Fig. 9(c). It is relevant to note that the variation...
in \( Z_I \) with \( \mu_m \) is found to be quite similar to the change in the coordination numbers associated with minimum random loose packing (RLP) limit observed in dry granular systems. The minimum RLP coordination number corresponds to the minimum coordination number required to obtain a disordered, mechanically stable jammed system. As the limits of jamming are prescribed entirely by the properties of the particles, it is conceivable that the characteristics related to jamming in granular systems devoid of fluid is to be expected in suspensions as well.

The effect of changing \( Z \) on \( \mu \), under various \( \mu_m \) values is shown in Fig. 10(a). \( \mu(Z) \) values reasonably collapses into a single curve for all values of \( \mu_m \) studied. This demonstrates that the minimum \( \mu \) achieved at low \( I_v \) values (i.e., \( \mu_1 \)) is determined by \( Z_I \). As \( Z_I \) is inversely related to \( \mu_m \), the relationship between \( \mu_1 \) and \( \mu_m \) depicted in Fig. 7(b) can be rationalized. Assuming a range of \( I_v \) values, one can calculate and compare \( \mu \) against \( Z \) for a given \( \mu_m \) value using the relationships outlined in Eqs. (4), (10), (13), and (14). As shown in Fig. 10(a), the theoretical predictions of \( \mu(Z, \mu_m = 0.5) \) is in agreement with the simulation results. Consequently, the variation in \( \mu \) with \( \phi \) also collapses reasonably onto a simple curve across the various \( \mu_m \) values studied, as seen in Fig. 10(b). This behavior is observed in 2D simulations of sheared suspensions and dense granular systems\(^\text{12,17} \) and experimentally by Boyer et al.\(^\text{11} \). With increasing volume fraction, under a given shear rate, the shear stress and normal stresses become larger, but their ratio (\( \mu \)) reduces till \( \mu = \mu_1 \) at jamming [see Figs. 10(d)-10(f)].

This implies that the jamming volume fraction determines \( \mu_1 \), the minimum macroscopic friction coefficient. The lower the jamming volume fraction, the higher the observed \( \mu_1 \); see Fig. 10(c). Our simulations of nonspherical particle suspensions (see Sec. III H) that jam at a lower volume fraction compared to spherical particles also agree with this observation, as shown in Fig. 10(c).

### H. Nonspherical particles

Particle shapes have significant effects on the shear thickening behavior of the suspensions. Cornstarch particles are observed to shear thicken at much lower \( \phi_m \) values (\( \phi_m \approx 0.44 \)) in comparison to suspensions of spherical particles which shear thicken around \( \phi_m = 0.56 \). Simulation results\(^\text{25} \) show that frictional jamming volume fraction \( \phi_J \) is lowered when particles shapes become “cornstarch-like.” In the interest of comparing the macroscopic friction coefficient \( \mu \) variation in spherical particles to that of nonspherical particles, simulations of “cornstarch-like” nonspherical particle suspensions were performed. The “cornstarch-like” particles were created using overlapping spheres of varying sizes, as outlined in Ref. 25. The nonspherical particles are bimodal with diameters of 8 \( \mu m \) (50% by volume) and 11.2 \( \mu m \) (50% by volume) and a standard deviation about 0.01 \( \mu m \). As \( \phi \) increases, the properties of the particles, it is conceivable that the characteristics related to jamming in granular systems devoid of fluid is to be expected in suspensions as well.

The minimum volume fraction \( \phi_m \) for various \( \mu_m \) values (i.e., \( \mu_1 \)) is determined by \( \phi_m \). As \( \phi_m \) increases, the coordination numbers associated with minimum random loose packing (RLP) limit observed in dry granular systems. The minimum RLP coordination number corresponds to the minimum coordination number required to obtain a disordered, mechanically stable jammed system. As the limits of jamming are prescribed entirely by the properties of the particles, it is conceivable that the characteristics related to jamming in granular systems devoid of fluid is to be expected in suspensions as well.

**Fig. 10.** (a) Variation in the macroscopic friction coefficient \( \mu \) with average coordination number \( Z \) compiled across different \( \mu_m \) and \( \phi \) values. The solid line and shaded area represent the mean and range of values \( \mu(Z) \) under various \( \mu_m \) values predicted by Eqs. (4), (10), (13), and (14). (b) Variation of \( \mu \) with \( \phi \) for various \( \mu_m \) values. The solid line and shaded area represent the predicted range of \( \mu(\phi) \) for various \( \mu_m \) values by Eqs. (9)–(12). Green triangles represent the experimental results from Boyer et al.\(^\text{11} \) (c) Minimum macroscopic friction coefficient \( \mu_1 \) achieved at jamming as a function of the jamming volume fraction \( \phi_J(\mu_m) \). Black triangle represents the \( \mu_1 \) observed at jamming for suspensions of nonspherical particles as discussed in Sec. III H. (d) and (e) Pressure \( P \) and shear stress \( \sigma_{\text{shear}} \) normalized by \( \sigma_g \) scaling with volume fraction for various shear rates for \( \mu_m = 0.5 \). (f) Macroscopic friction coefficient \( \mu \) measured for the pressure and shear stresses shown in (d) and (e). Vertical black lines show the jamming volume fraction.
suspensions, for any constant $\mu_m$ value. Naturally, these deviations become apparent at $I_v$ values where particle interactions become relevant, i.e., $I_v < 10^{-1}$. Results suggests that the macroscopic friction coefficient of nonspherical particle suspensions plateaus to $\mu_1$ at higher viscous numbers in comparison to the spherical particle suspensions. Also, at vanishing viscous numbers, the macroscopic friction coefficient of the nonspherical particle suspensions saturates to a higher $\mu_1$ in comparison with spherical particle suspensions, for a given $\mu_m$ value. This agrees with measurements of the macroscopic friction coefficient for cornstarch suspensions close to jamming,

$$I_v \approx 0.62$$ in the experimental systems and $\mu_1 \approx 0.6$ in the simulations. In Sec. III G, it was concluded that the jamming volume fraction determines the minimum value of the macroscopic friction coefficient. Considering that the nonspherical suspension simulated here jams around $\phi_{\text{spherical}}^{\text{non-spherical}} = 0.53$, which is lower than the jamming volume fraction for spherical particles ($\phi_{\text{spherical}} = 0.576$) at the same $\mu_m$ value ($\mu_m = 1$), the larger $\mu_1$ observed here can be rationalized.

It is intriguing to see whether one can generalize these variations in $\mu$ with particle shapes and microscopic friction coefficients to arrive at a common curve for all available data. By (a) normalizing $I_v$ with $I_v^2$ [where $I_v^2 = I_v(\mu = 2\mu_1)$] to account for the shift in $I_v$ values at which $\mu$ plateaus to $\mu_1$ and (b) setting upper and lower bounds to the variation in $\mu$ by using $(\mu - \mu_1)/\mu$ as the measure of the variation of $\mu$ with $I_v$, the results collapse nicely to a single curve, for both spherical and nonspherical particle suspensions, across varying $\mu_m$ values [see Fig. 11(b)]. The results of Boyer et al.\textsuperscript{11} are shown for comparison and also agrees with the curve. This common relationship can be fitted using the empirical expression

$$\frac{\mu - \mu_1}{\mu} = \frac{\sqrt{I_v}}{\sqrt{I_v + \sqrt{I_v^{2m}}}},$$

which in turn gives:

$$\mu = \mu_1 \left(1 + \sqrt{\frac{I_v}{I_v^{2m}}} \right).$$

The terms $\phi_m$, $\mu_1$, $I_0$ from Eq. (4) are incorporated in $I_v^{2m}$ in Eq. (16). Since $I_0$ and $\mu_2$ do not change significantly with $\mu_m$, $I_v^{2m}$ becomes a function of the free parameter $\phi_m$. Even though the simulation results reasonably conform to the expression given by Eq. (15), it should be mentioned that the validity of the expression at high viscous numbers $(I_v > 0.5)$ is suspect, as we have no experimental data in this regime. Experimental data for nonspherical particles at viscous numbers high enough to obtain $I_v^{2m}$ are also absent, which prevents us from further validation.

IV. CONCLUSION

We analyze the behavior of the macroscopic friction coefficient ($\mu$) under different microscopic friction coefficients ($\mu_m$) using 3D numerical simulations. The predictions of $\mu$ from simulations agree with earlier predictions of viscous number granular suspension rheology. We find that when $\mu_m > 0.3$, the viscous number rheology is largely insensitive to the value of $\mu_m$. By changing the jamming volume fraction $\phi_m$ with the changes in shear stresses and $\mu_m$, we analyze $\mu$ in terms of distance to jamming ($\phi_m - \phi$) and provide phenomenological but analytic formulas that match the observations. Our results also suggest that the behavior of $\mu$ across various $\mu_m$ and viscous numbers ($I_v$) can be reduced to effects of the distance to jamming. The study of changes in the average coordination number ($\langle Z \rangle$) with the viscous number ($I_v$) shows that $\langle Z \rangle$ smoothly decreases from $\langle Z \rangle$ (at jamming) to zero with increasing viscous number, where $\langle Z \rangle$ is again determined by $\mu_m$. Our results suggest that the minimum $\mu$ achieved is inversely related to the jamming volume fraction and $\langle Z \rangle$. Finally, we show that with appropriate scaling, a common curve for the variation of $\mu$ with $I_v$ emerges for both spherical and nonspherical particles under varying $\mu_m$ values.

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