Migration and jamming in wide-gap Couette flows of dense suspensions

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Introduction—a bit of history

- Modeling strategy to capture dynamics of colloids (Modified Stokesian Dynamics)
- Flow in a Widegap Couette cell
heterogeneous due to particle migration, and that consequently the macroscopic stress–strain rate relationship cannot be directly related to the local constitutive behavior and thus in particular to the shear thickening.

Here, we compare local and global measurements for what is perhaps the best-known example of a shear-thickening suspension: cornstarch particles suspended in water. We show that the shear thickening can in fact be viewed as a re-entrant solid transition in this system: (i) at rest the material is solid because it has a (small) yield stress; (ii) for low shear rates, shear banding (localization) occurs, and the flowing shear band grows with increasing shear rate, the shear thus liquefies the material; (iii) shear thickening happens at the end of the localization regime, where all the material flows, subsequently it suddenly becomes ''solid'' again. In addition, and (iv) we find a pronounced dependence of the critical shear rate for the onset of shear thickening on the gap of the measurement geometry, which can be explained by the tendency of the sheared system to dilate. This is confirmed by an independent measurement of the dilation of the suspension as a function of the shear rate. It also explains the MRI observations: when flow is localized, the non-flowing region plays the role of a “dilatancy reservoir” which allows the material to be sheared without jamming.

This paper follows up on our earlier work on shear thickening of cornstarch [Fall et al. (2008)] but is much more detailed in that here we present also the MRI measurements of the concentration, more detailed measurements of the velocity profiles, plate–plate measurements, oscillation measurements and more detailed measurements of the variation of the gap of the plate–plate cell under an imposed normal stress. In order for these new data to be comprehensible, we do have to repeat some of the earlier data and discussion. In this way, we obtain a more complete picture of the shear-thickening behavior.

II. MATERIALS AND METHODS

The cornstarch particles (from Sigma Aldrich) are relatively monodisperse particles with, however, irregular shapes (Fig. 1). Suspensions are prepared by mixing the cornstarch with a 55 wt. % solution of CsCl in demineralized water. The CsCl allows one to perfectly match the solvent and particle densities [Merkt et al. (2004)]. We study suspensions of volume fraction ranging between 38% and 46%, and focus here mainly on the variation of the gap of the plate–plate cell under an imposed normal stress.
The maximum sliding-friction forces between spherical surfaces exceed those from aggregate precursors \[32\] [Fig. 2(b)] by pulling on the chain while the other end was produced chainlike structures of 5 to 20 microns in length. However, the stick-slip behavior of rolling Friction forces between spherical microparticles have not been determined experimentally in our knowledge. Rolling-friction forces between spherical particles were highly reproducible and showed a mean square deviation from the ideal sphere. A force \[30\] was used to get information about the surface roughness and heterogeneity. Possibly, indicates the presence of small-scale surface roughness which may decrease the adhesion strength. Results from repeated adhesion measurements with one particle chain oscillation under external forced motion is also available \[33\].

In order to measure rolling-friction forces directly, we performed constant stress tests to remove any loading history by a continuous sheared stress ramp from 0.01 Pa to 1,000 Pa for 3 min for the two lower concentration samples and from 0.1 to 1 Pa. Finally, steady-state stress sweeps were performed for the 0.1 and 1 Pa. Initially, the samples were presheared to ensure a steady-state force was measured. The force \(F\) is given by the equation:

\[
F = \mu \cdot n \cdot S
\]

where \(\mu\) is the coefficient of friction, \(n\) is the number of particles, and \(S\) is the force applied. The frequency of the piezoelectric-crystal motion was kept constant at 0.1 or 1 Hz with peak-to-peak amplitudes of 4 or 8 microns. The piezoelectric stage which is not visible in the images. The forced up-and-down motion through the piezoelectric translator produced an aggregate with the AFM tip until a chain was formed. Figure 2(c) shows an example of 4 or 8 particle chains on which we applied a forced oscillation at 0.1 or 1 Hz with peak-to-peak amplitudes of 4 or 8 microns, respectively. The constant stress tests were performed for the 0.01 to 800 Pa, 0.01 to 1,000 Pa, and 0.1 to 1,500 Pa ranges.

The static case but also a function of shear rate (or stress) and, possibly, the flow history; hence, under shear, USANS yields a one-dimensional projection in the velocity–vorticity (1–3) plane. The Couette along the velocity gradient direction and there was measured in the shear cell due to loading which was measured in the shear cell due to loading...
“On the dilatancy of media composed of rigid particles in contact”

Reynolds 1885

“If in any way the volume be fixed, then all change of shape is prevented.”

from wikipedia
“Experiments on a gravity-free dispersion of large solid spheres in a Newtonian fluid under shear”

Bagnold 1954

spherical droplets of mixture of paraffin wax and lead stearate (~ 1.32 mm)

When shear rate \( \dot{\gamma} \) is low, shear stress \( \propto \dot{\gamma} \)  
viscous effect of solvent

When shear rate \( \dot{\gamma} \) is high, shear stress \( \propto \dot{\gamma}^2 \)  
particle inertia
Hydrodynamic theory

**Einstein 1906**

\[ \eta = \eta_0(1 + 2.5\phi) \text{ for } \phi \ll 1 \]

**Batchelor & Green 1972**

\[ \eta \approx \eta_0(1 + 2.5\phi + 6.9\phi^2) \]

from wikipedia
Hydrodynamic simulation

Stokesian Dynamics
Brady & Bossis 1985

6N-dimensional overdamped Langevin eq.

\[ F_H + F_B = 0 \]

\[ F_H = - R \cdot (U - u) + R' : D \]

\[ u(r) = \nabla u \cdot r = D \cdot r + (\omega/2) \times r \]
- Introduction—a bit of history

 Modeloing strategy to capture dynamics of colloids (Modified Stokesian Dynamics)

- Flow in a Widegap Couette cell
Mechanical properties of fluid materials

Liquid Newtonian

\[ \sigma = -pI + 2\eta D \]

\[ D \equiv \frac{1}{2} \left( \nabla u[r(t), t] + \nabla u^T[r(t), t] \right) \]

Suspensions Non-Newtonian

Stokesian dynamics

\[
\frac{\text{mean free path}^2}{\text{velocity}} \sim 10^{-11} \text{[s]}
\]

\[
\frac{\text{radius}^2}{\text{diffusion constant}} \sim 1 \text{[s]}
\]
microstructure
Rheology

velocity gradient
\[
\begin{pmatrix}
\frac{\partial u_x}{\partial x} & \frac{\partial u_x}{\partial y} & \frac{\partial u_x}{\partial z} \\
\frac{\partial u_y}{\partial x} & \frac{\partial u_y}{\partial y} & \frac{\partial u_y}{\partial z} \\
\frac{\partial u_z}{\partial x} & \frac{\partial u_z}{\partial y} & \frac{\partial u_z}{\partial z}
\end{pmatrix}
\]

stress tensor
\[
\begin{pmatrix}
\sigma_{xx} & \sigma_{xy} & \sigma_{xz} \\
\sigma_{yx} & \sigma_{yy} & \sigma_{yz} \\
\sigma_{zx} & \sigma_{zy} & \sigma_{zz}
\end{pmatrix}
\]

\[\rho \left\{ \frac{\partial u}{\partial t} + (u \cdot \nabla) u \right\} = \nabla \cdot \sigma\]

Constitutive modeling

material functions
scalars
(not coordinate specific)
physical interpretations
\[\eta(\dot{\gamma}), N_1(\dot{\gamma}), N_2(\dot{\gamma})\]
Zero-Reynolds number hydrodynamics

Navier-Stokes equations (non-linear)

$$Re \left\{ \frac{\partial \vec{u}}{\partial t} + (\vec{u} \cdot \nabla) \vec{u} \right\} = -\nabla p + \nabla^2 \vec{u}$$

$$Re \equiv \rho_0 a^2 \dot{\gamma} \rightarrow 0$$

Stokes equations (linear)

$$\vec{0} = -\nabla p + \nabla^2 \vec{u}$$

Hydrodynamic interactions

(6 N dimension)

$$F_H = -\mathbf{R} \cdot (\mathbf{U} - \mathbf{u}) + \mathbf{R'} : \mathbf{D}$$

$$\mathbf{u}(r) = \nabla \mathbf{u} \cdot \mathbf{r} = \mathbf{D} \cdot \mathbf{r} + (\omega/2) \times \mathbf{r}$$

cf. Stokes drag

$$\mathbf{R} = 6\pi \eta_0 a \mathbf{I}$$
Newtonian dynamics vs. Overdamped dynamics

\[ m \frac{dU}{dt} = -6\pi \eta_0 a U \]

\[ U(t) = U_0 \exp(-t/\tau) \]

time step for numerical integration
\[ \Delta t \ll \tau \]

\[ \eta_0 = 0.001 \text{ [Pa s]} \]
\[ a = 1 \text{ [\mu m]} \]
\[ \rho = 1000 \text{ [kg/m}^3] \]

\[ \tau = \frac{m}{6\pi \eta_0 a} \approx 2 \times 10^{-7} \text{ [s]} \]

\[ 0 \rightarrow m \frac{dU}{dt} = -6\pi \eta_0 a U \]
\[ U(0) = U_0 \]
\[ U(\Delta t) = 0 \]
\[ \Delta t \gg \tau \]

cf. time-scale of shear flow
\[ \tau_{\text{shear}} = 1/\dot{\gamma} \]
Purely hydrodynamic suspensions

force balance eq. \[ F_H = 0 \]

hydrodynamic interaction \[ F_H = -R \cdot (U - u) + R' : D \]

Perfect reversibility, if lubrication layers can remain.

\[
F_{\text{Lub}}^{(\text{nor})} \sim -\frac{1}{h} \Delta U^{(\text{nor})}
\]

\[
F_{\text{Lub}}^{(\text{tan})} \sim -\log \left( \frac{1}{h} \right) \Delta U^{(\text{tan})}
\]
Singularity of non-Brownian simulation

Minimum gap vs Shear strain for different discretization steps:

- **Euler, dt=10^{-4}**
- **Euler, dt=10^{-5}**
- **pre. cor., dt=10^{-4}**
- **pre. cor., dt=10^{-5}**

The graph shows the minimum gap decreasing as the shear strain increases, with different curves representing the simulations with various discretization steps. The legends indicate the type of simulation and the corresponding step size.
Regularize the singularity

lubrication force

\[ F_{\text{lub}} = -\frac{3\pi \eta_0 a^2}{2h} (U^i - U^j) \cdot nn \]

\[ \frac{1}{h} \rightarrow \begin{cases} 
\frac{1}{h + \delta} & h > 0 \\
\frac{1}{\delta} & h \leq 0
\end{cases} \quad \delta = 10^{-3}a \]

Particle contacts are no longer forbidden!!
We need a contact force model because particle contacts are no longer forbidden

\[ F_H + F_C = 0 \]

\[ F_H = -R \cdot (U - u) + R' : D \]

\[ |F_C^{\text{tan}}| < \mu |F_C^{\text{nor}}| \]

\[ (F_{ij}^{\text{nor}})^{\text{nor}} = k_n (r_{ij} - 2a)n^{ij} \]

\[ (F_{ij}^{\text{tan}})^{\text{tan}} = -k_t \xi^{ij} \]
Effect of particle softness

\[ F_H + F_C = 0 \]

\[ F_C^n = k_n (r - 2a)n \quad \text{frictionless } \mu = 0 \]

\[ F_H = -R \cdot (U - u) + R' : D \]

\[ \langle \eta/\eta_0 \rangle \]

\[ k_n = 3 \times 10^5 [6\pi \eta_0 a \dot{\gamma}] \]

\[ k_n = 0.5 \times 10^5 [6\pi \eta_0 a \dot{\gamma}] \]
Contact deformation of Silica

\[ F_n = k_n (2a - r)^{3/2} \]
\[ k_n = \frac{4}{3} E \sqrt{a} \]

Young's Modulus of Silica (SiO2)
\[ E = 65 \sim 75 \text{ [GPa]} \]

particle size
\[ a = 10^{-6} \text{ [m]} \]

a large shear stress
\[ \sigma_{xy} = 10^3 \text{ [Pa]} \]

typical force
\[ F_n \sim \sigma_{xy} \times (2a)^2 = 4 \times 10^{-9} \text{ [N]} \]

overlap
\[ \frac{2a - r}{a} \approx 10^{-5} \]

This level of stiffness is very difficult in simulation!!
With and without hydrodynamic lubrication

\[ \langle \eta/\eta_0 \rangle = 1 + \frac{5}{2} \phi \]

[Graph showing the variation of \( \langle \eta/\eta_0 \rangle \) with \( \phi \) for with and without hydrodynamic lubrication.]
Hydrodynamic vs contact force contributions

\[ \mu = 0 \]

\[ \mu = 1 \]

Hydrodynamic vs contact force contributions
Force-chain network in 2D simulation

\[ \bar{F}_{ij} \equiv -\vec{F}^{(ij)} \cdot \vec{n}_{ij} \]

\[ \phi_{\text{area}} = 0.7 \quad \mu = 0.5 \]

\[ \phi_{\text{area}} = 0.8 \quad \mu = 0.5 \]
Angular dependence of normal force

Only hydrodynamics forces are able to be attractive
\[ \phi_{\text{area}} = 0.7 \quad \mu = 0.5 \]

normal force \( \bar{F}_{ij} \equiv - \bar{F}^{(ij)} \cdot \bar{n}_{ij} \)

contribution to shear stress \( \sigma_{xy} \)
\[ \phi_{\text{area}} = 0.8 \quad \mu = 0.5 \]

normal force \( \vec{F}_{ij} \equiv - \vec{F}^{(ij)} \cdot \vec{n}_{ij} \)

contribution to shear stress \( \sigma_{xy} \)
With and without contact friction

\[ \eta/\eta_0 = c(\phi_J^{(\mu)} - \phi)^{-\lambda} \]

Jamming with gear-like contacts
with rolling friction
2D demo for an extreme contact model

no slide and rolling as long as pushing

\[ \mu_{\text{slid}} = \infty \quad \mu_{\text{roll}} = \infty \]
Origin of rate-dependence

\[ F_H + F_C = 0 \quad \text{rate independent} \]
\[ F_H + F_C + F_R = 0 \quad \text{rate-dependent} \]

repulsive force

Laun 1984
Latex particles in water

Viscosity $\eta$ [Pa s]

shear rate $\dot{\gamma}$ [s$^{-1}$]

pH

Salt NaCl [mol L$^{-1}$]

0.287
1.14
0
\[ F_H + F_C + F_R = 0 \quad \text{rate-dependent} \]

repulsive force

Low $\dot{\gamma}$

High $\dot{\gamma}$
How do shear flows bring Brownian hard spheres to non-equilibrium states?

Stokesian Dynamics was introduced to tackle this problem 6N-dimensional overdamped Langevin eq.

\[ F_H + F_B = 0 \]
\[ u(r) = \nabla u \cdot r = D \cdot r + (\omega/2) \times r \]

**Hydrodynamic force** \[ F_H = - R \cdot (U - u) + R' : D \]

**Brownian force** \[ \langle F_B \rangle = 0, \quad \langle F_B(t_1)F_B(t_2) \rangle = 2k_B TR\delta(t_1 - t_2) \]
Modified Stokesian Dynamics

Step 1: Remove lubrication singularity

\[ F_H + F_B + F_C = 0 \]

Step 2: Introduce a contact-force model
Contacts in sheared Brownian motions

\[ \text{Pe} = 1 \quad \text{Pe} = 100 \]

Péclet number: \[ \text{Pe} \equiv \frac{6\pi \eta_0 a^3 \dot{\gamma}}{k_B T} \]
Experimental data

\[ a = 125 \text{ nm} \]
\[ \eta_0 = 0.001 \text{ Pa s} \]

\[ a = 225 \text{ nm} \]
\[ \eta_0 = 0.049 \text{ Pa s} \]

\[ a = 260 \text{ nm} \]
\[ \eta_0 = 0.05 \text{ Pa s} \]

Prof. Wagner’s group
To match experimental data

Stokesian Dynamic + DEM

\[ F_H + F_B + F_C + F_R = 0 \]

Experimental data
black-solid lines

Cwalina and Wagner, JOR (2014)
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Liquid

Newtonian

\[ \sigma = -pI + 2\eta D \]
\[ D \equiv \frac{1}{2} \left( \nabla u[r(t), t] + \nabla u^T[r(t), t] \right) \]

Molecular dynamics

Suspensions

Non-Newtonian

\[ \text{?} \]

Stokesian dynamics

\[ \frac{\text{mean free path}^2}{\text{velocity}} \sim 10^{-11} \text{[s]} \]

\[ \frac{\text{radius}^2}{\text{diffusion constant}} \sim 1 \text{[s]} \]
Macro-scale continuum sim.

\[
\rho \left\{ \frac{\partial u}{\partial t} + (u \cdot \nabla)u \right\} = \nabla \cdot \sigma \quad \text{with} \quad \nabla \cdot u = 0
\]

constitutive eq.

Micro-scale Particle dynamics sim.
Force-balance dynamics with fixed particles

- Velocities of mobile particles to be solved: $U^m = (U^{(1)}, \ldots, U^{(n)})$
- Velocities of fixed particles: $U^f = (U^{(n+1)}, \ldots, F^{(n+m)})$

$F^\text{P} :$ interparticle forces (and torques)

**force balance equations**

$$
\begin{bmatrix}
F_H^m \\
F_f^f
\end{bmatrix} + \begin{bmatrix}
F_H^m \\
F_f^f
\end{bmatrix} + \begin{bmatrix}
0 \\
F_{\text{rct}}^f
\end{bmatrix} = \begin{bmatrix}
0 \\
0
\end{bmatrix}
$$

**known** $(U^f, F_{\text{P}}^m, F_{\text{P}}^f)$

**to find** $(U^m, F_{\text{rct}}^f)$

**step 1** $U^m = (R_{\text{FU}}^{mm})^{-1} (F_{\text{P}}^m - R_{\text{FU}}^{mf} U^f)$  **dynamics**

**step 2** $F_{\text{rct}}^f = R_{\text{FU}}^{fm} U^m + R_{\text{FU}}^{ff} U^f - F_{\text{P}}^f$  **used in rheology**
\( \phi_{\text{area}} = 0.78 \)

\( n = 9000 \)
Migration

\[ j_{\text{radial}} = \frac{2a^2}{9\eta_0} f(\phi) \nabla \cdot \sigma \]
Shear band

Jamming
Monolayer simulation for wide-gap Couette cells

\[ \eta(r) = \eta(\phi(r)) \]

\[ \frac{v_\theta(r)}{R_{in}\Omega} \]

\[ \frac{r}{R_{out}} \]

\[ \frac{\dot{\gamma}_{in}/\gamma_0}{0.01, 0.02, 0.05, 0.1, 0.5} \]

Jammed domain

\[ \phi(r) \]

\[ \frac{r}{R_{out}} \]

\[ \dot{\gamma}_{in}/\gamma_0 = 0.5 \]
Simulation models for colloidal suspensions are very different from MD simulations. (Particles are not points)

We introduced a Modified Stokesian Dynamics simulation.
- A realistic choice to avoid lubrication singularity
- Various possibilities for contact interaction between colloids
- Additional force is essential for rate-dependence

Although we now know rheology of dense suspensions well, we know little about fluid mechanics of dense suspensions