Thinning or thickening? Complex rheology of dense suspensions

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Nonlinear glassy rheology

- Decreasing $T$ in supercooled liquids: ‘Diverging’ viscosity $\eta(T)$.
- Transition from viscous fluid to solid material (finite yield stress).
- Flow curves at finite shear rate $\dot{\gamma}$ in simple shear flow: $\sigma = \sigma(\dot{\gamma}) = \eta(\dot{\gamma})\dot{\gamma}$, in binary LJ mixture. Viscosity is a function.

[Berthier & Barrat JCP '01]
Dense colloidal suspensions

- Colloidal (Brownian) glass transition upon compression.

- ‘Diverging’ viscosity: $\eta_T(\varphi)$.

- Rheology similar to molecular glasses: $\eta = \eta(\dot{\gamma}, \varphi)$.

[Brambilla et al., PRL ’09]
[Petekidis et al., JPCM ’04]
Granular suspensions

- Jamming transition: compressing macroscopic (non-Brownian) particles.
- Example: granular suspensions. ‘Diverging’ viscosity: $\eta_0(\varphi) \neq \eta_T(\varphi)$.

[Brown & Jaeger, PRL ’09]

- Rigid particles: linear rheology $\eta = \eta_0(\varphi)$.

[Boyer & Pouliquen, PRL ’10]

- Soft particles: Jamming transition from viscous to solid suspension (yield stress) with non-linear rheology: $\eta = \eta(\dot{\gamma}, \varphi)$, at $T = 0$. 
Shear-thickening

- Observed in large variety of systems: soft/hard, large/small... Multiple ‘complex’ explanations: hydrodynamics, micro-structure...

![Graphs showing shear-thickening behavior](image)

- Thickening coexists with other regimes.
- Strong interplay with jamming.
- Role of friction for macroscopic particles: discontinuous thickening.

[Wagner, Brady, Phys. Today ’09]
[Otsubo, Rheol. Acta ’94]

[Brown, Jaeger PRL ’09]
Simple numerical model

• To understand complex systems, one must understand simple ones...

• Langevin dynamics for soft harmonic spheres with shear and temperature in $d = 3$:

$$m \frac{dv_i}{dt} + \xi(v_i - \dot{\gamma}y_i e_x) + \sum_j \frac{dV(|r_i - r_j|)}{dr_i} + \eta_i = 0,$$

with $\langle \eta_i(t)\eta_j(t') \rangle = 2k_B T \xi 1 \delta(t - t')$ and $V(r \leq a) = \epsilon (1 - r/a)^2$.

• Four microscopic timescales:
  (i) dissipation: $\tau_0 = \xi a^2 / \epsilon = 1$, our time unit.
  (ii) thermal time: $\tau_D = \xi a^2 / (k_B T) \to \infty$ when $T \to 0$.
  (iii) velocity damping: $\tau_v = m / \xi$.
  (iv) shear rate: $\dot{\gamma}$, competes with (i)-(iii).

• We can study both finite and zero temperature, both overdamped and inertial regimes within single model. Vary $\dot{\gamma}$ at constant $(T, m)$.

[Ikeda et al., PRL ’12, Soft Matter ’13, Kawasaki et al., arXiv’14]
Multiple rheological regimes

- Complex patterns of Newtonian, shear-thinning, and shear-thickening regimes, with nontrivial density dependence.
- Qualitatively similar to experimental data.
Soft glassy rheology

- Steady state rheology at \((T = 10^{-4}, m = 0)\). Diverging Newtonian viscosity and emerging yield stress as \(\phi\) increases.

- Glassy rheology, as seen in colloidal particles, star polymers, microgels, simple supercooled liquids: \(\gamma \tau_D \ll 1\).

- Theories of driven glasses capture competition between slow glassy dynamics and shear flow (reasonably) well.
Non-Brownian suspension rheology

- Rheology at \((T = 0, m = 0)\). Diverging Newtonian viscosity and emerging yield stress as \(\varphi\) increases.

- Rheological signature of \(T = 0\) jamming transition: \(\dot{\gamma} \tau_D \gg 1\).

- No microscopic theory for rheology. Driven dynamics at \(T = 0\) is difficult to attack from first principles, subtle structural changes under shear.
It is crucial to understand stress & time scales, rather than the shape of the flow curves, in particular $\dot{\gamma} T_D \gg 1$ or $\dot{\gamma} T_D \ll 1$ makes a dramatic difference (from jamming to glass transitions).
Glass – jamming crossover

- Simulations at \((T = 10^{-6}, m = 0)\). Crossover from glassy physics when \(\dot{\gamma}\tau_D \ll 1\) to athermal jamming physics when \(\dot{\gamma}\tau_D \gg 1\).

- Two Newtonian regimes, two distinct viscosities, emergence of yield stress (when \(\dot{\gamma} \to 0\)), with strange density dependence. Complex rheology... even without inertia.
Crossover in experiments

- Microemulsions are ideal to observe the crossover: right size, right softness. Both glass and jamming contributions needed to describe rheology.

- Star polymers and pnipam microgels are too soft, grains/foams are too large. Either glass or jamming, but one must choose the right one...

[from Mason, Bibette, Weitz, 1996]
‘Jamming phase diagram’
‘Jamming phase diagram’
Inertia adds a 3rd timescale

- Rheology for \((T = 0, \varphi = 0.60)\) and increasing \(m\).

![Graph](image)

- Crossover from Newtonian to shear-thickening flow when \(\dot{\gamma} \propto 1/\tau_v = \xi/m\) (prefactor \(\approx 0.01\)).

  - (i) Energy balance: 
    \[
    \sigma \dot{\gamma} \approx \rho L^3 \xi \bar{v}^2 / L^3.
    \]

  (ii) Momentum exchange via collisions: 
  \[
  \sigma \approx \rho L^2 \bar{v} m \dot{\gamma} a / L^2
  \]

  (i) + (ii) \(\rightarrow\) ‘Bagnold’ scaling: 
  \[
  \sigma \approx (\rho m^2 a^2 / \xi) \dot{\gamma}^3, \text{ i.e. } \eta \propto (\dot{\gamma} \tau_v)^2.
  \]

- Softness controls viscosity maximum: 
  \[
  m \bar{v}^2 / 2 \approx \epsilon.
  \]

Dynamic ‘phase diagram’

- Shear-thickening observed when $\tau_v^{-1} \ll \dot{\gamma} \ll \dot{\gamma}_c$, and if no additional stress scale.

- Above jamming at $T = 0$, yield stress $\sigma_Y > 0 \rightarrow$ shear-thinning.

- At finite temperature, thermal yield stress develops: $\rightarrow$ shear-thinning.

- Onset of thickening: $\dot{\gamma} \approx 0.01/\tau_v$. Good agreement for some systems (large emulsions, latex), only qualitative agreement for others (macroscopic hard grains).
Jigsaw falling into place

- Thermal glassy regime: $\dot{\gamma} < 10^{-7}$, athermal overdamped:
  $10^{-7} < \dot{\gamma} < 10^{-5}$, inertial $\dot{\gamma} > 10^{-5}$, softness maximum: $\dot{\gamma} \approx 10^{-3}$, jamming above $\phi = 0.64$. All in a simple model evolving with Langevin dynamics.