Stretching Colloidal Suspensions: from flow to fracture

M.I. Smith[1,2], R. Besseling[3], M.E. Cates[3], V. Bertola[1]

1. School of Engineering, University of Edinburgh, Kings Buildings, Mayfield Rd, Edinburgh, EH9 3JL, UK
2. School of Physics and Astronomy, Department of Physics, University of Nottingham, University Park, Nottingham, NG7 2RD, UK
3. SUPA, School of Physics and Astronomy, University of Edinburgh, King's Builldings, Mayfield Rd, Edinburgh, EH9 3JZ

e-mail : mike.i.smith@nottingham.ac.uk

Introduction

Concentrated suspensions of particles (CSP) are common in the pharmaceutical, cosmetic and food industries. CSP have been well studied using shear rheology, yet manufacturing processes often involve substantially different geometries.

Using Extensional Rheology, we directly observe dilatancy and granulation, leading to fracture at high extension rates. In addition, CSPs are found to display surprising evidence of viscoelasticity.

Extensional Rheology of Concentrated Colloidal Suspensions

PMMA "Hard Sphere" colloids (r~604nm) at volume fractions ($\varphi \sim 0.603$) in Octadecene are placed between the two plates of an extensional rheometer.

The top plate is retracted at different velocities, giving rise to an approximate extension rate: $d\varepsilon/dt \sim l_{max} / l_0 \Delta t$. The fluid is observed using a high speed camera.

Three types of behaviour are observed:

'Liquid' - At low strain rates the sample thins like a viscous fluid, finally under going capillary induced breakup.

'Jammed' - At high strain rates the sample jams resulting in dramatic granulation and fracture.

'Transition'- At intermediate strain rates the sample initially jams and granulation occurs. Prior to fracture the sample relaxes back into the liquid state.

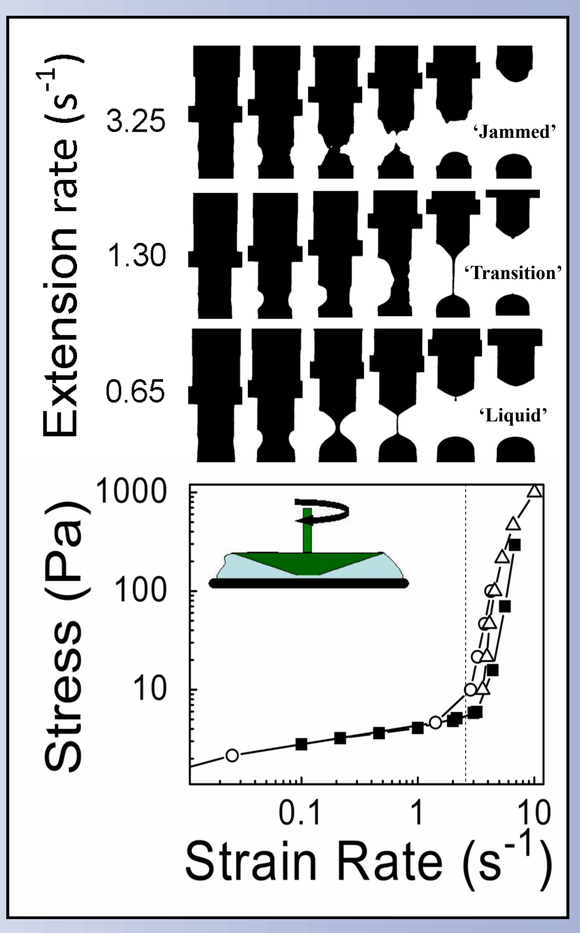


Figure 1: Colloidal Suspension ($\varphi \sim 0.603$) stretched at different rates shows different morphologies above or below a critical strain rate. This critical value compares well with the onset of shear thickening in conventional shear rheology (bottom). Filled Squares, strain rate controlled. Open symbols stress-controlled.

With linear plate separation rate the flow of suspensions has a significant shear component.

Conventional shear rheology shows that the transition between jammed and Liquid states occurs at the onset of shear thickening.

However, fascinating additional effects occur which appear to be related to the large exposed surface area.

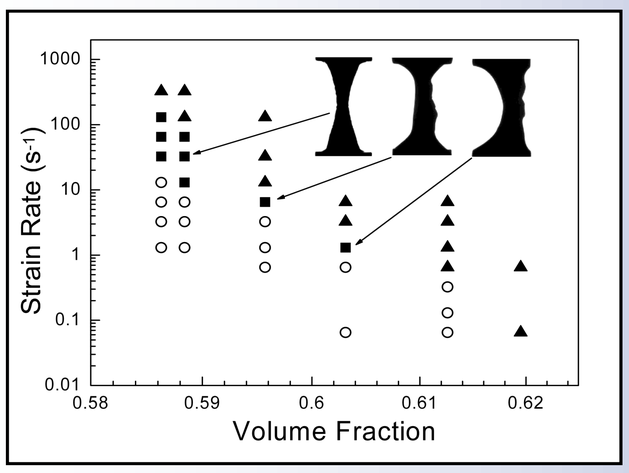


Figure 2: Volume fraction dependence of the transition. ▲ = 'Jammed', ■ = 'Transition', ○ = Liquid'. The inset pictures show the morphology of the transition at the points shown.

Volume fraction dependence

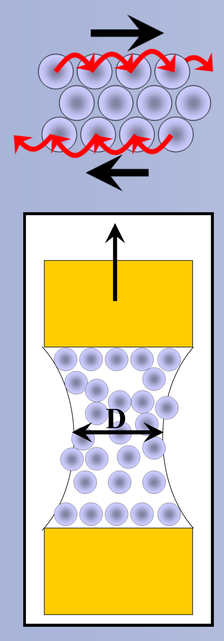
The critical extension rate alters by several orders of magnitude for small changes in volume fraction (φ).

One might expect equivalent behaviour at different φ to be seen at similar locations relative to the transition. However with increasing φ we observe:

- Increasing granulation or 'lumpiness'
- Increasing asymmetry

The role of Dilatancy in granulation & fracture

'Dilatancy' is an effect well known in granular dynamics whereby the volume of a group of particles must increase upon shearing to enable flow.



Dilatancy has been suggested as a possible mechanism for jamming.

Dilation within a fixed volume involves the generation of force transmitting 'clusters'.

The increased occupied volume causes particles to encounter the air-liquid interface and 'poke through'.

This is observed in the change of the surface from gloss to matt (A & B).

The change coincides with granulation and fracture suggesting a strong role for dilatancy in the jamming mechanism.

In order for particles to poke through the surface, the shear driven osmotic particle pressure must exceed the Laplace pressure due to surface tension.

For $\varphi \sim 0.603$ gives $d\varepsilon_c/dt \sim 4s^{-1}$

$$\frac{\sigma}{D} \sim \frac{\eta\dot{\varepsilon}}{\left(1-\varphi/\varphi_0\right)^2}$$

Capillary forces generated at fluid surface result in the dramatic jamming, granulation and fracture by locking the particle clusters in position.

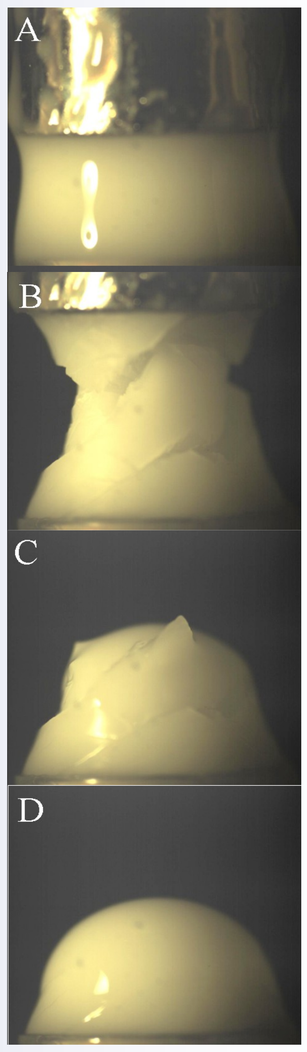


Figure 3 - $\varphi \sim 0.603$, $d\varepsilon/dt \sim 3.25s^{-1}$

Filament Recoil

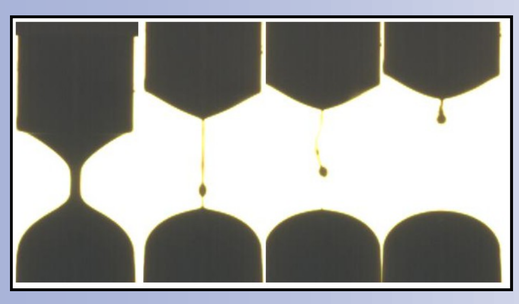


Figure 4 - Filament recoil (φ~0.603, $d\varepsilon/dt \sim 0.65$)

1. Prior to recoil, the filament diameter thins exponentially.

2. As the filament approaches ~ 100 μm the diameter becomes fixed as the filament continues to

3. The filament ruptures recoiling rapidly.

If the colloidal fluid is stretched at a rate just below that which gives rise to the transition the fluid thins to about

The recoil is composed of an initial rapid recoil (~ ms) followed by a longer relaxation which is comparable to the relaxation time of the fluid.

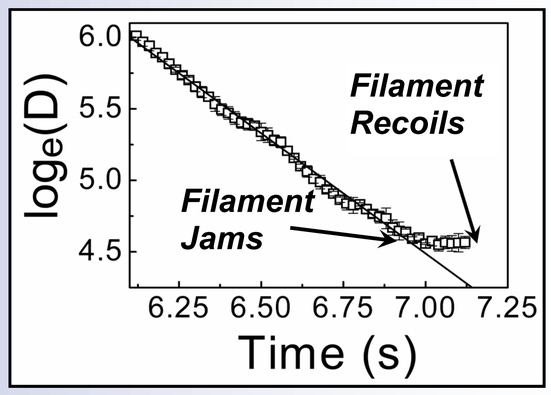
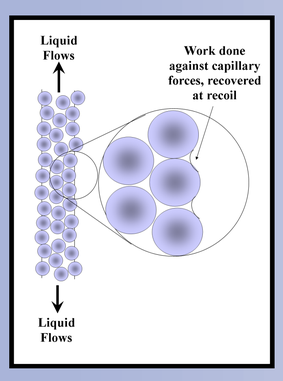


Figure 5 - Filament diameter as a function of time for φ~0.603, $d\varepsilon/dt \sim 0.65$

Self filtration?



If the liquid flows out of the filament faster than the particles ("self-filtration") then the particles in the filament will jam.

As the filament is stretched further, work is done against capillary forces at the surface, storing elastic energy.

Upon rupture the work is recovered in the filament recoil.

Conclusions

- Jamming, granulation and fracture are related to dilatancy
- Colloidal filaments can display visco-elastic recoil

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