Stretching dense colloidal suspensions: from flow to fracture

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Talk Overview

- A brief detour
- Introduction
- Liquid
- Jamming / Granulation
- Frictional contacts
- Stability – Ductility & Brittle Fracture
- Conclusions
- Acknowldegements

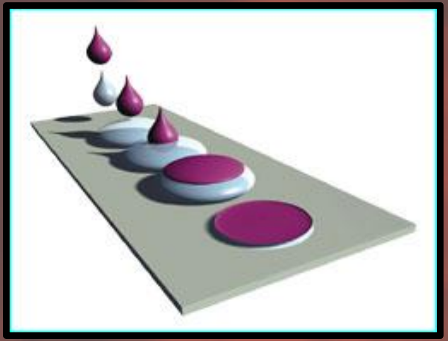
Introduction

Concentrated colloidal suspensions

Concentrated suspensions of particles (CS) are common every day fluids

CS can exhibit pronounced increases in viscosity with strain rate (shear thickening)

Under certain conditions fluids may even jam solid



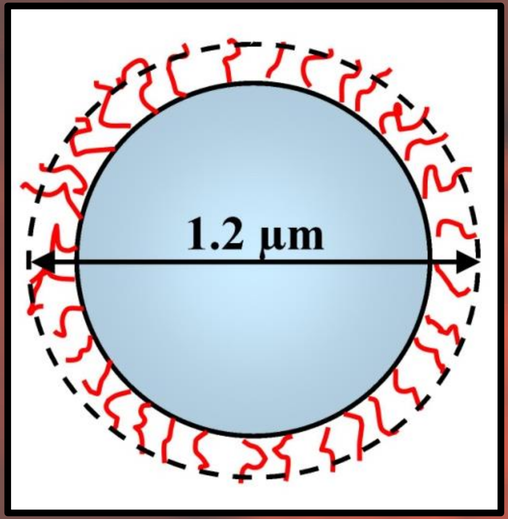
Model Hard Sphere Colloids

"Hard spheres" are commonly used as a model system.

This simplifies the interactions present:

- *Attractive VdW forces are very small*
- *Hydrodynamic interactions*

Insights gained from hard spheres, provide insight into more complicated systems



<u>Our Colloids</u>

Poly-methyl methacrylate particles (D ~ 1.2 μm, 2μm)

Sterically stabilised with 10nm
Poly-12- hydroxystearic acid chains

Practical Flows

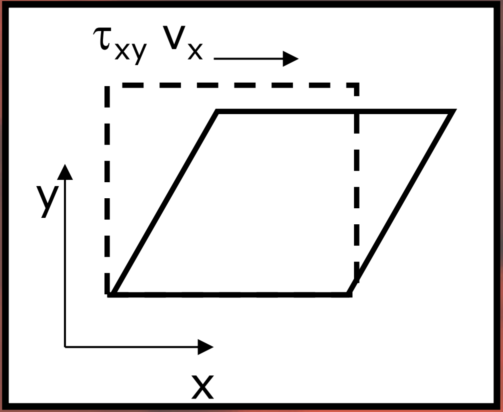
Concentrated suspensions are generally studied using a shear rheometer:

- Shear flow
- Steady State
- Hard boundaries

However, many practical flows e.g nozzle flows are:

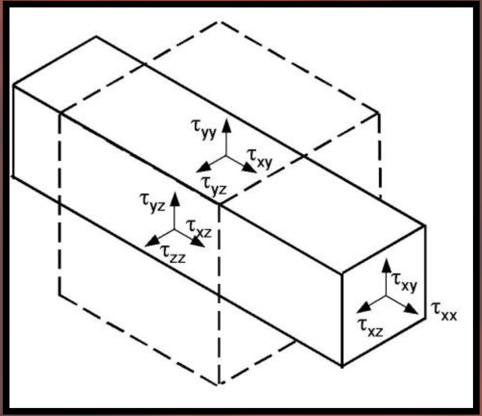
- extensional or mixed.
- transient
- Liquid-gas interface

Shear flow:



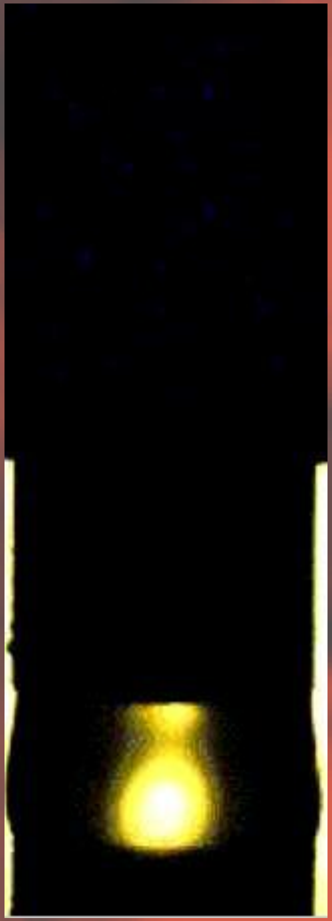
$$\eta = \frac{\tau_{xy}}{\dot{\gamma}_{xy}}$$

Extensional flow:



$$\eta_e = \frac{\tau_{xx}-\tau_{yy}}{\dot{\varepsilon}_{xx}}$$

Extensional Rheometer



Liquid sample is placed between two cylindrical plates (Diameter 6mm)

The upper plate is moved upwards at a constant velocity 0.1 - 500mms^{-1}

As the plates move apart the fluid thins and breaks up

The flow is imaged with a high speed camera so it can be analysed

Liquid break up

Liquid
($\varepsilon = 0.65\ s^{-1}$)

Suspension of 1.2μm diameter particles suspended in ultra-low volatility solvent (Octadecene)

Volume fraction ~0.603

At low strain rates the colloidal suspension thins forming a long filament

Viscoelastic Filament

We observe visco-elastic recoil of filaments

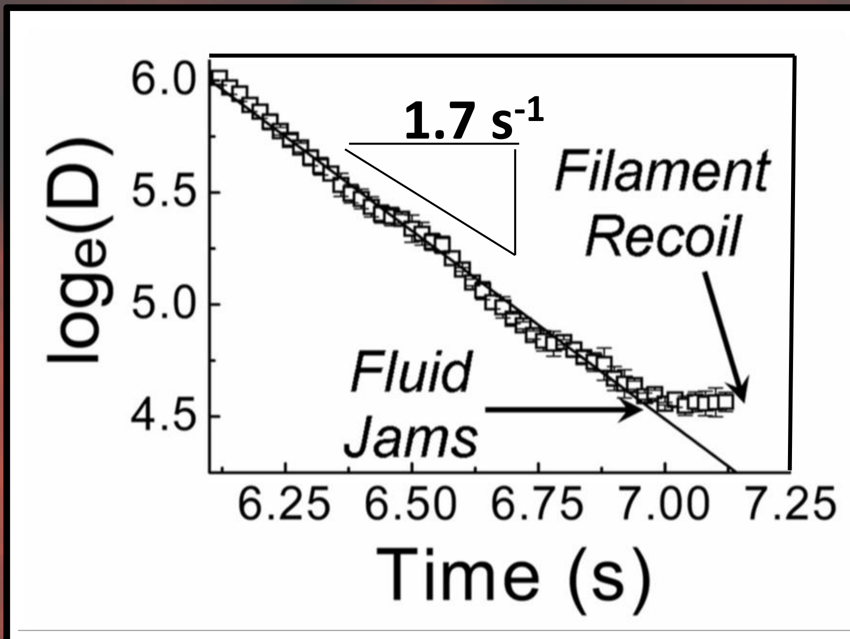
Two stages:

1) Rapid elastic recoil after filament ruptures

2) Slow viscous movement on timescale of fluid relaxation.

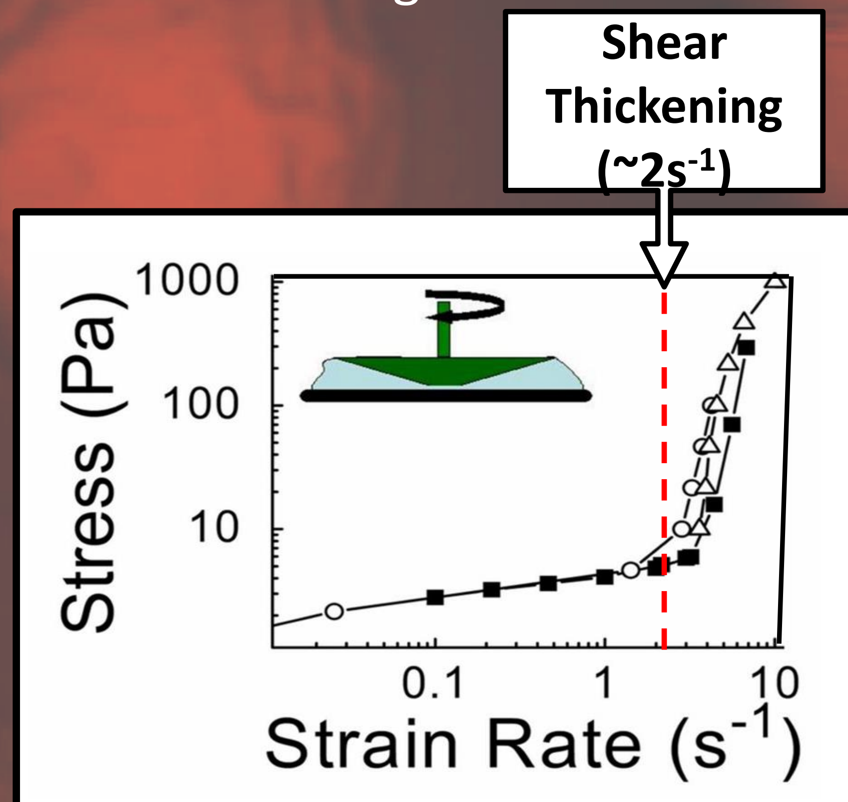
Recoil occurs for $\phi > 0.595$

Viscoelastic filament



Diameter of filament narrows exponentially with time

Timescale correlates with onset of shear thickening



Driven by Laplace pressure σ/D

Prior to recoil, filament diameter stops thinning → Jamming

Self-filtration

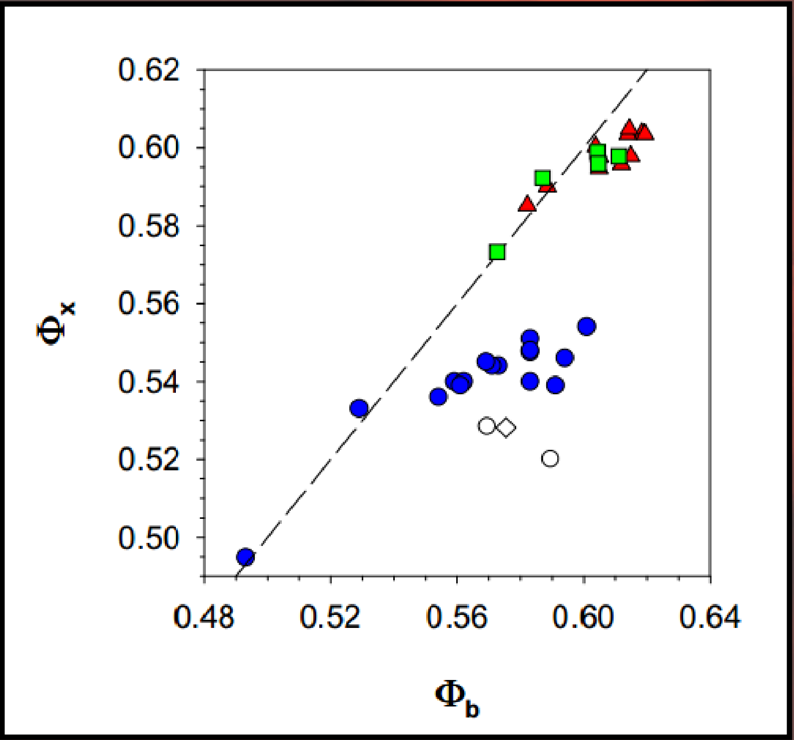
Jamming may be due to self-filtration

- Haw et al observe self filtration at these volume fractions

- Recoil only occurs at $\phi > 0.595$

$-u \sim (\sigma/D^2)ka^2/\eta$ → % change in volume fraction

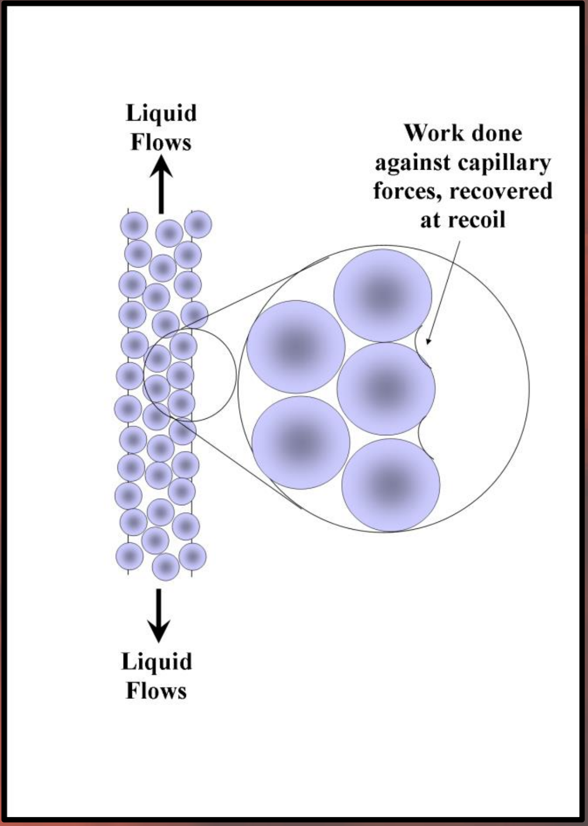
- Roche et al (PRL 2011) observed heterogeneous ϕ close to break up in a similar setup



[M. Haw PRL 92, 185506 (2004)]

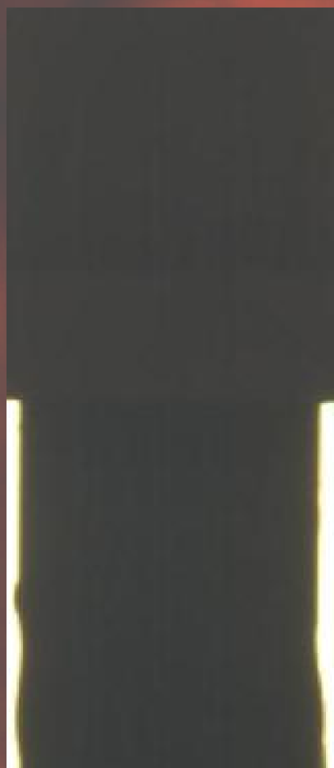
Viscoelastic filament

- Upon jamming the rheometer continues to stretch the filament
- This performs work against capillary forces which is recoverable upon rupture (ie elastic)
- If the rheometer is stopped prior to breakup, the recoil is much smaller but still exists so this is not a complete explanation.

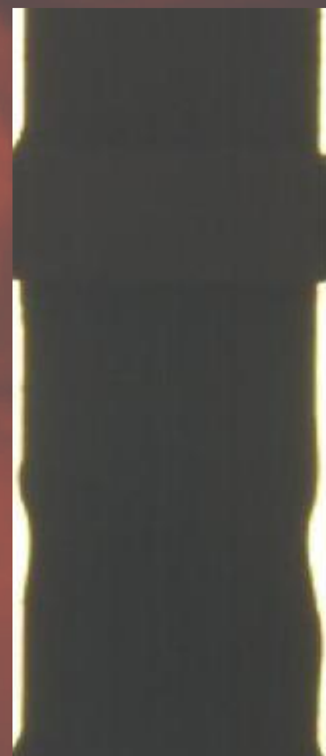


Jamming / granulation

Strain-rate dependent behaviour of Concentrated Suspensions of Colloids (φ~0.603)



Transition
($\dot{\varepsilon}$ = 1.3 s^{-1})



Jammed
($\dot{\varepsilon}$ = 3.25 s^{-1})

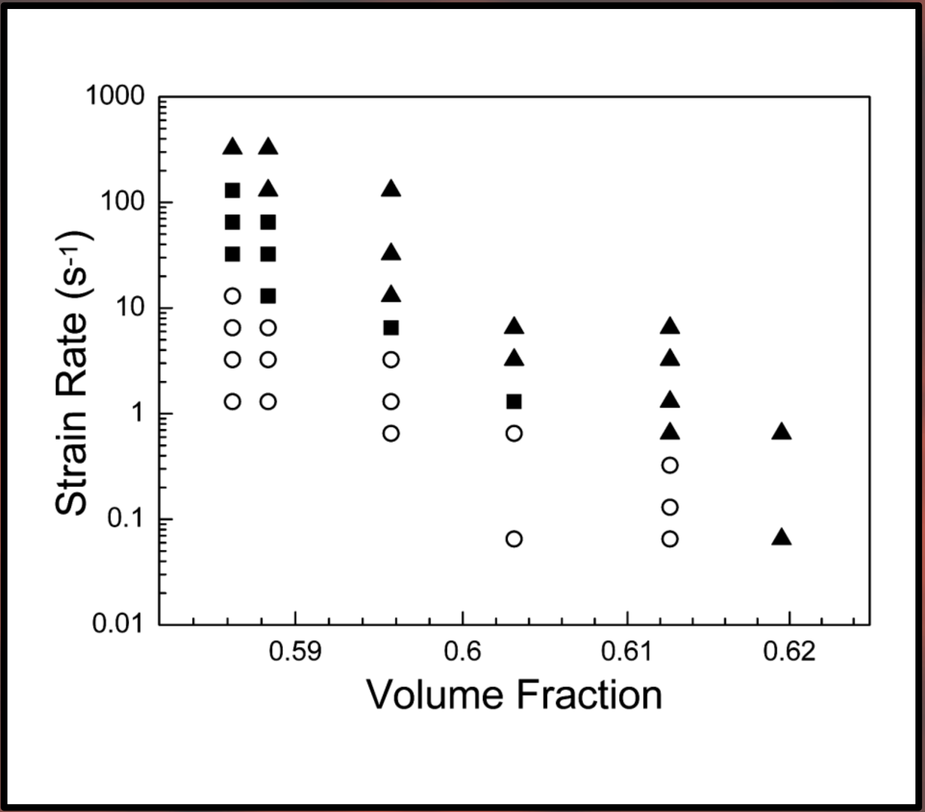
At low strain rates CS flow smoothly

Above a critical strain rate $\dot{\varepsilon}_c$ the sample jams resulting in granulation

$\dot{\varepsilon}_c$ depends critically on the volume fraction

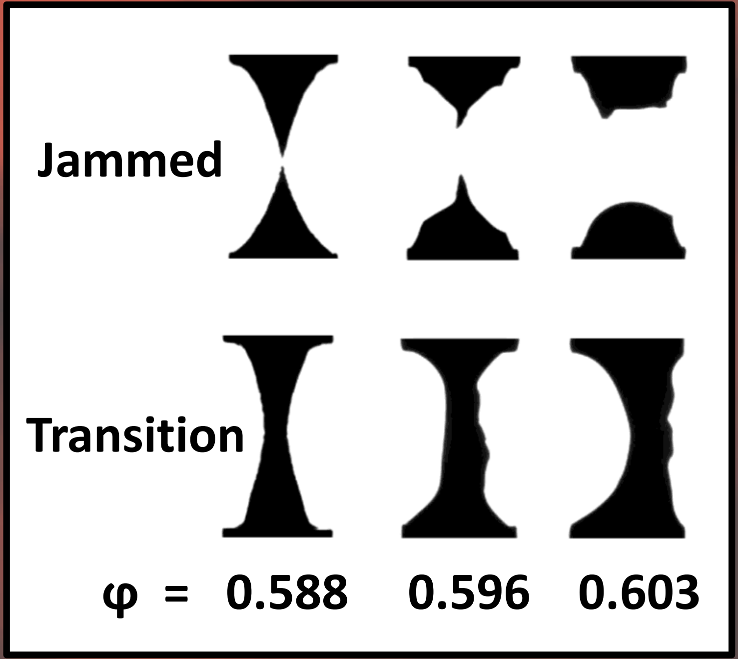
$\dot{\varepsilon}_c \sim 1.3 s^{-1}$

Volume Fraction Dependence



A 2% change in volume fraction alters critical strain rate by ~2 orders of magnitude

With increasing volume fraction we see increasing asymmetry and larger granules



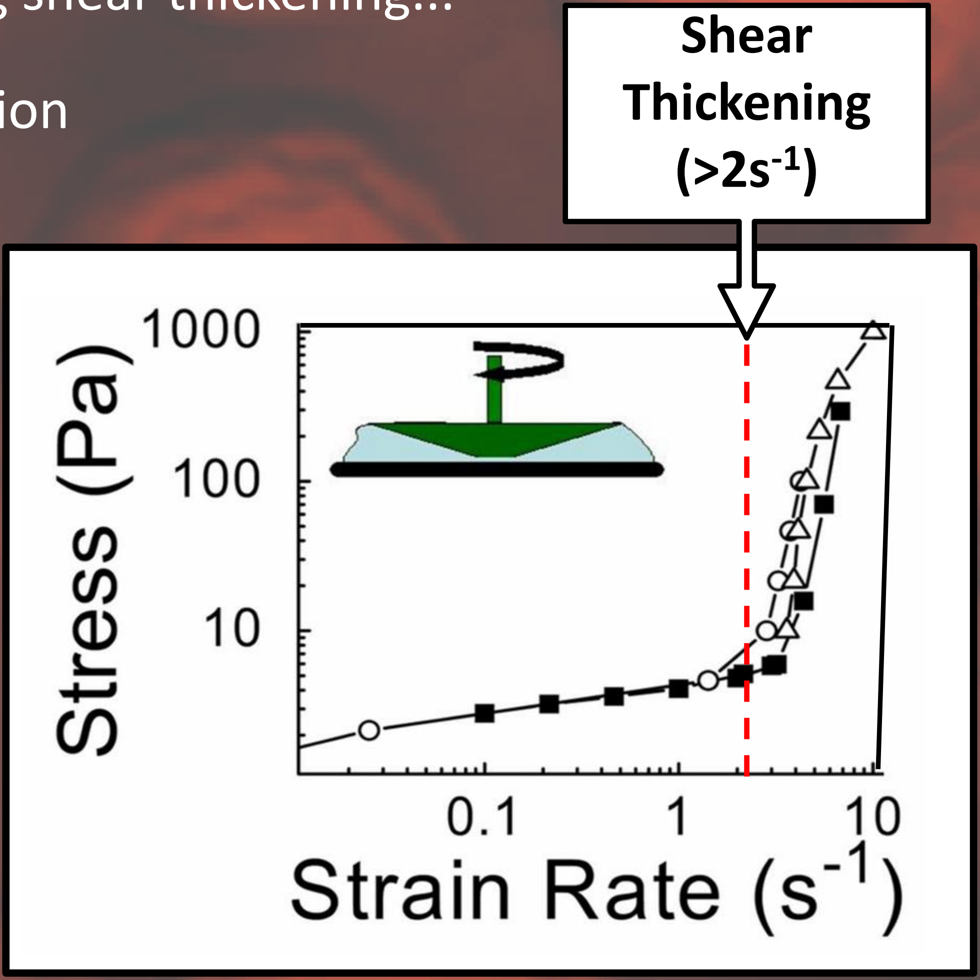
Relationship to Shear Rheology

For φ ~ 0.603 we observe strong shear thickening...

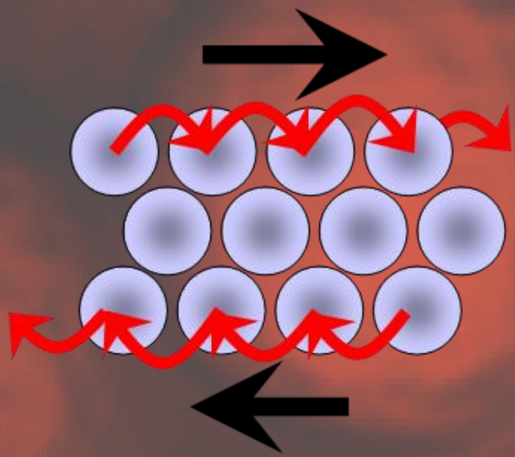
...but **not** jamming and granulation

Shear rate for the onset of shear thickening closely matches the transition between liquid and jammed samples (~ 1-2 s^{-1})

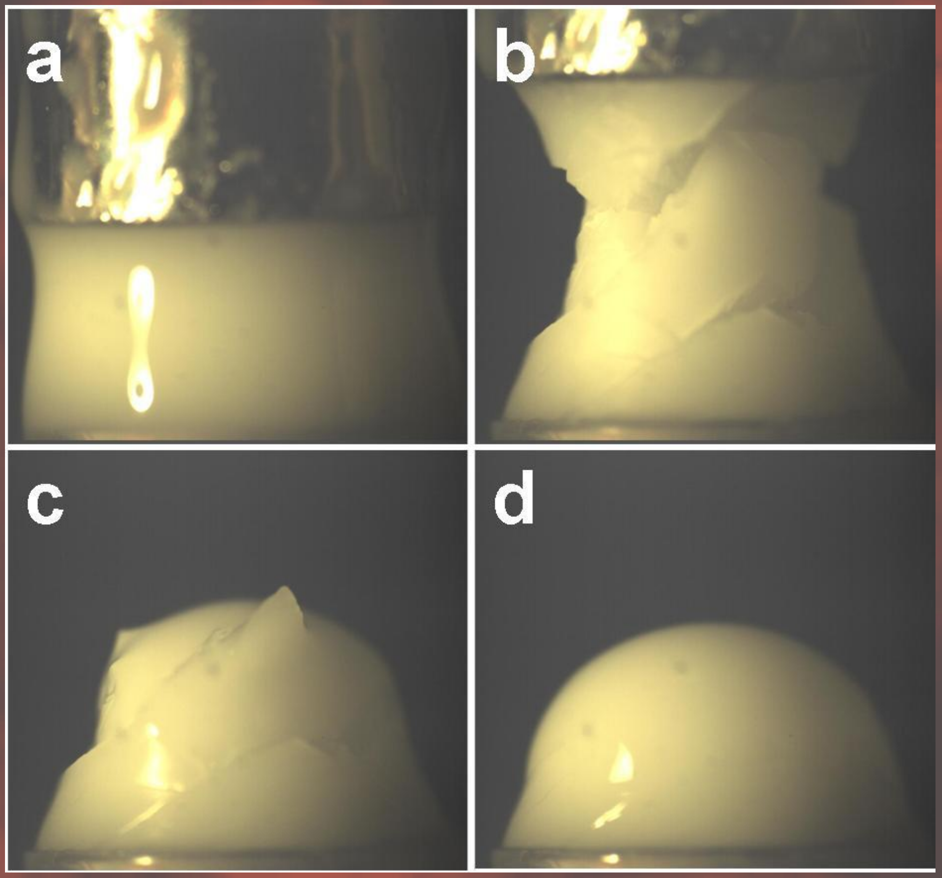
So what causes the dramatic jamming and granulation in the extensional geometry?



Dilatancy



In order for a packed bed of particles to flow the volume must increase.

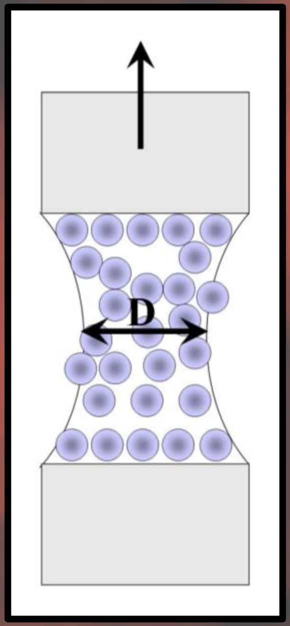


Particles poke through the liquid surface resulting in scattering

→ Surface appears dry and matt

Air-Liquid interface plays a key role in granulation

Jamming



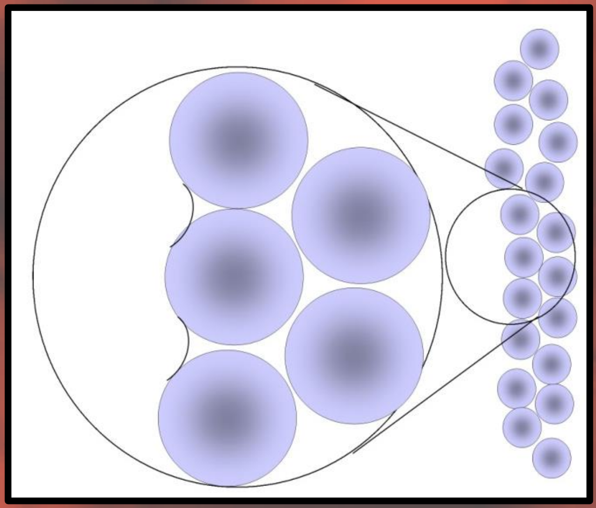
Dilatancy forces particles into contact with the air-liquid interface.

As volume filled by the particles expands, particles will poke through the air-liquid interface .

Capillary forces generated between exposed particles confine and arrest flow of suspension.

→ jamming and granulation

Jamming



Simple estimate of the onset strain rate:

$P_{osmotic} = \eta\dot{\varepsilon}/(1-(\varphi/\varphi_0))^2 \sim P_{LaPlace} = \gamma/D$

$\rightarrow \dot{\varepsilon}_{Critical} \sim 8\ s^{-1}$ for $\varphi \sim 0.603$

Similar to experimental value of ~ $1.3 s^{-1}$

$P_{LaPlace} = \gamma/D \sim 6\ Pa$

This is a long way below a capillary stress ~ 100kPa!

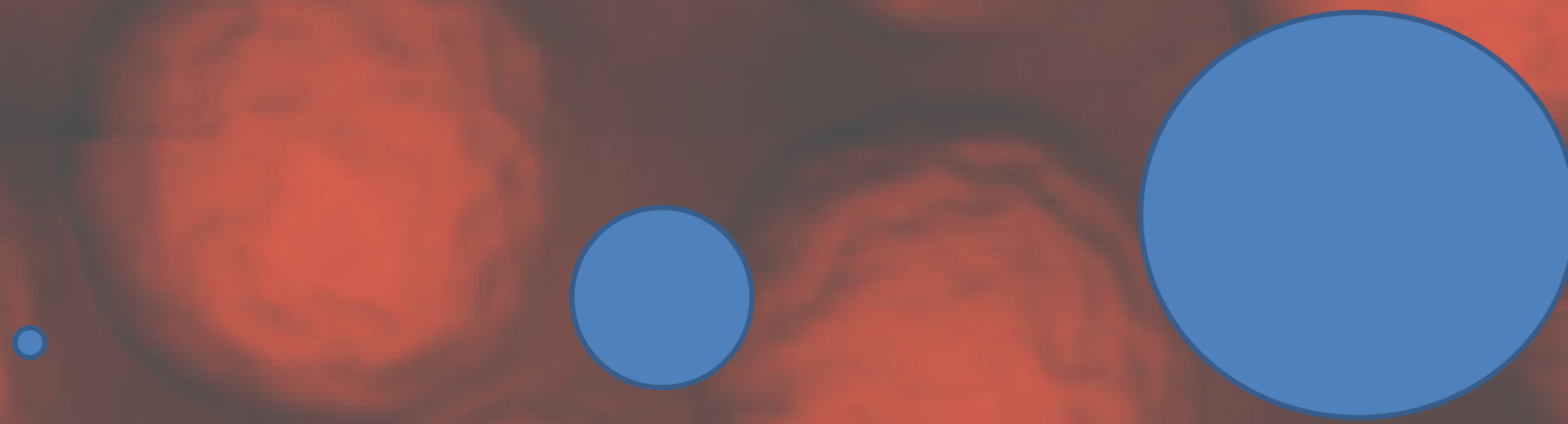
Are such large forces really being generated and how?

Frictional contacts?

Different types of shear thickening

- Continuous Shear Thickening
  - Smooth monotonic increase in the viscosity
  - Caused by hydrodynamic induced cluster formation
  - Occurs over wide range of volume fractions
- Discontinuous Shear Thickening
  - Large and rapid increase in viscosity with strain rate
  - non-monotonic flow curve
  - Characterised by an onset stress
  - Only occurs for concentrated suspensions
  - Depends on boundaries

Common Physics?



<u>Nanoparticles</u>
Size ~ 1-200nm
Thermal motion dominates
Hydrodynamic
Lubrication forces

<u>Concentrated colloidal suspensions</u>
Size ~ 1 μm

<u>Granular fluids</u>
Size ~ mm
"Zero temperature"
Frictional contacts

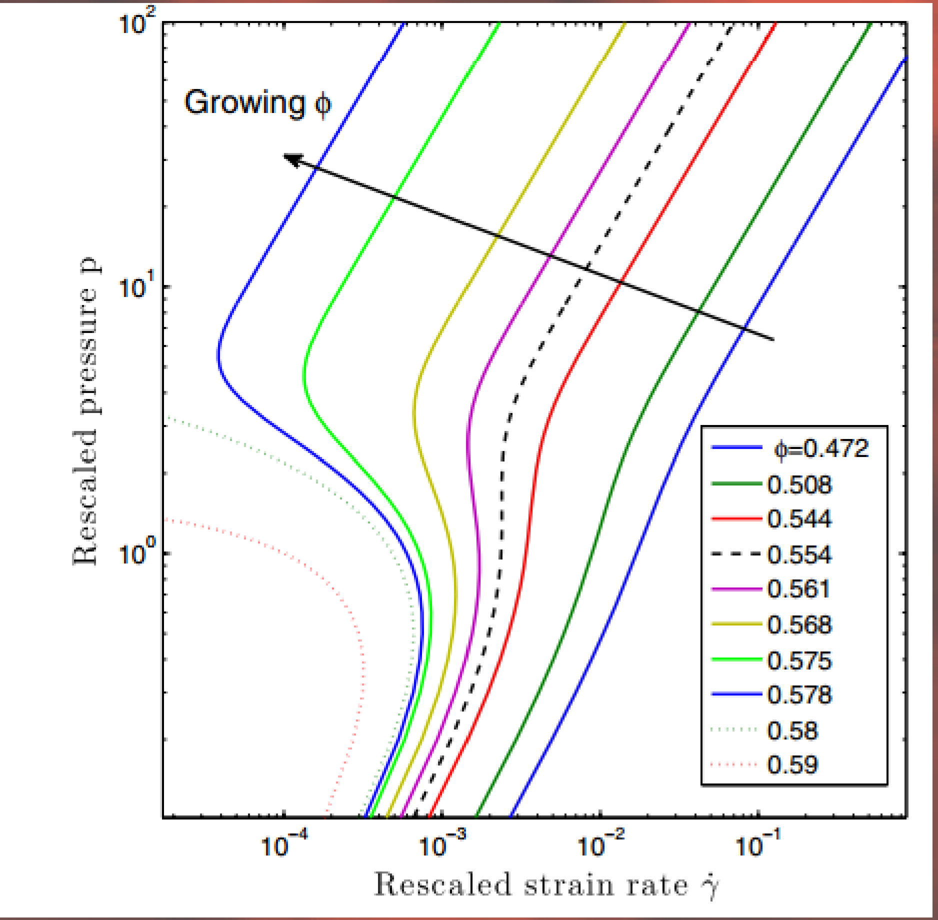
Frictional contacts in DST / jamming

"Above some stress scale lubrication films which normally keep particles separated convert to frictional contacts"

[Wyart et al PRL 2014]

Emulsions & Foams do not display DST

The onset of DST can be influenced by tuning inter-particle interactions

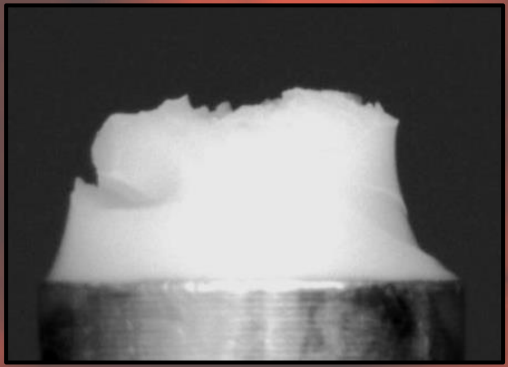


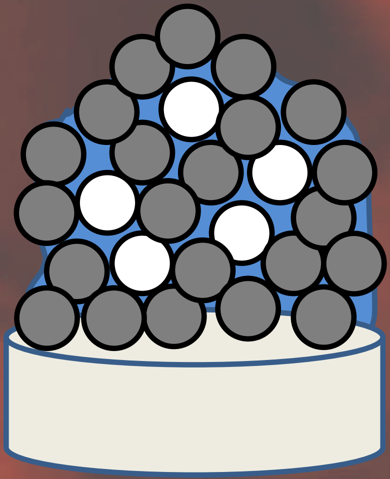
Granulation

Using particles of diameter 2μm at high enough volume fractions the granules produced <u>remain</u> in a jammed state

Capillary forces due to the menisci between exposed particles place the sample under compression.

Dynamics are very slow enabling quantitative measurements of the forces to be made

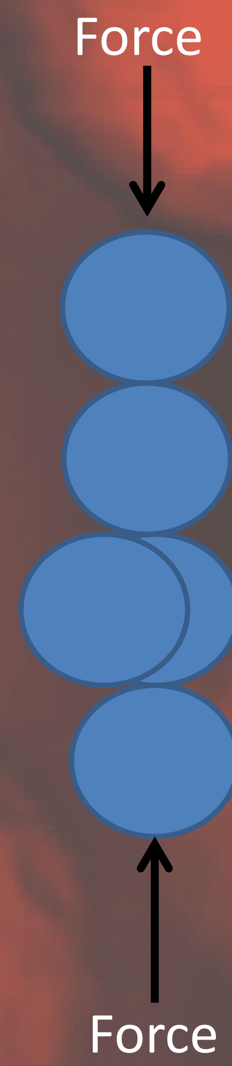




Granules & Frictional Contacts

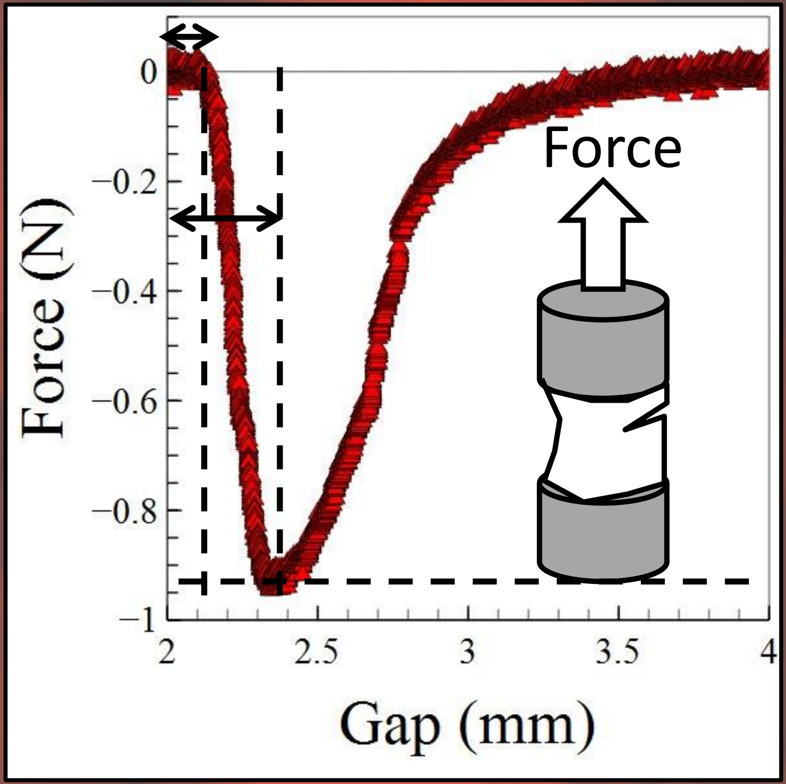
Chains of load bearing particles ("Force Chains") in the granule interior balance the forces leading to an arrested or jammed state

A static structure without bonding suggests frictional contacts



Quantifying Granulation

Measure the force directly using the normal force sensor of a Shear rheometer

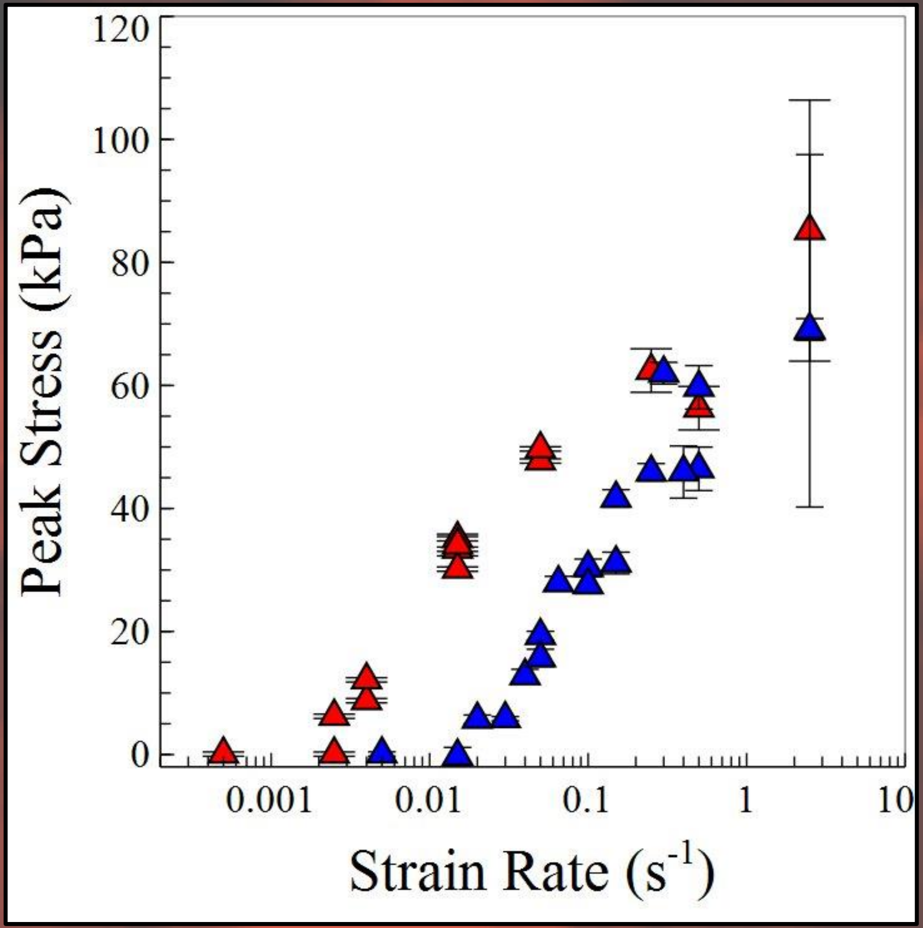


Always a small strain ~ 5% before significant forces are measured

Maximum measured force occurs at macroscopic fracture

For different strain rates we measure the peak force and the gap at this peak force.

Quantifying Granulation



Stress scale is large
Cf shear rheology ~ 100Pa

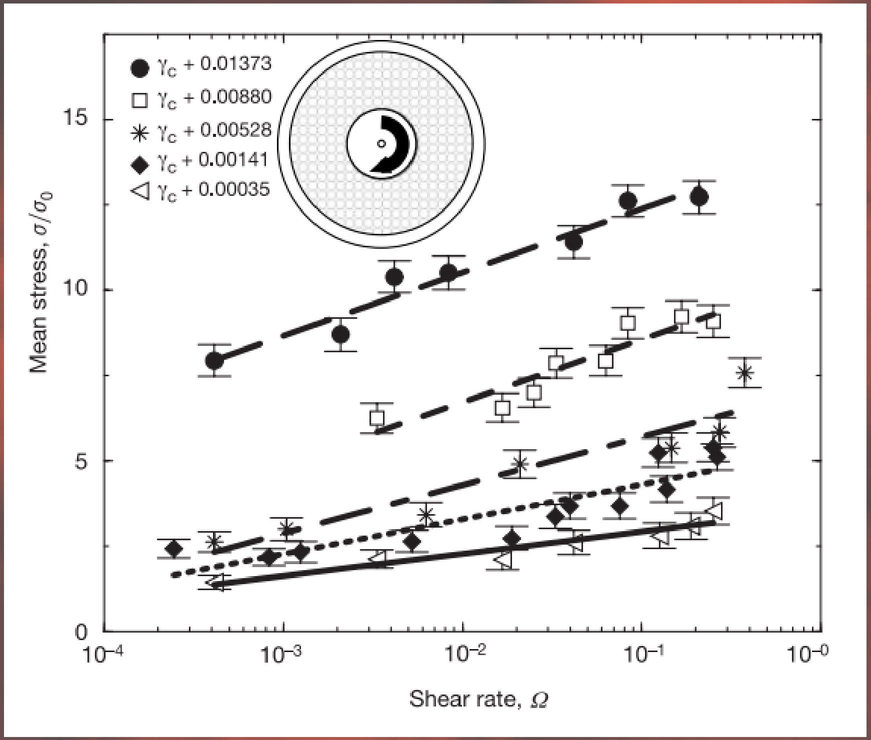
Comparable with maximum capillary stress ~ $5.3\gamma/a$ = 162kPa

Stress has a weak dependence on strain rate

Comparison with 2D granular experiments

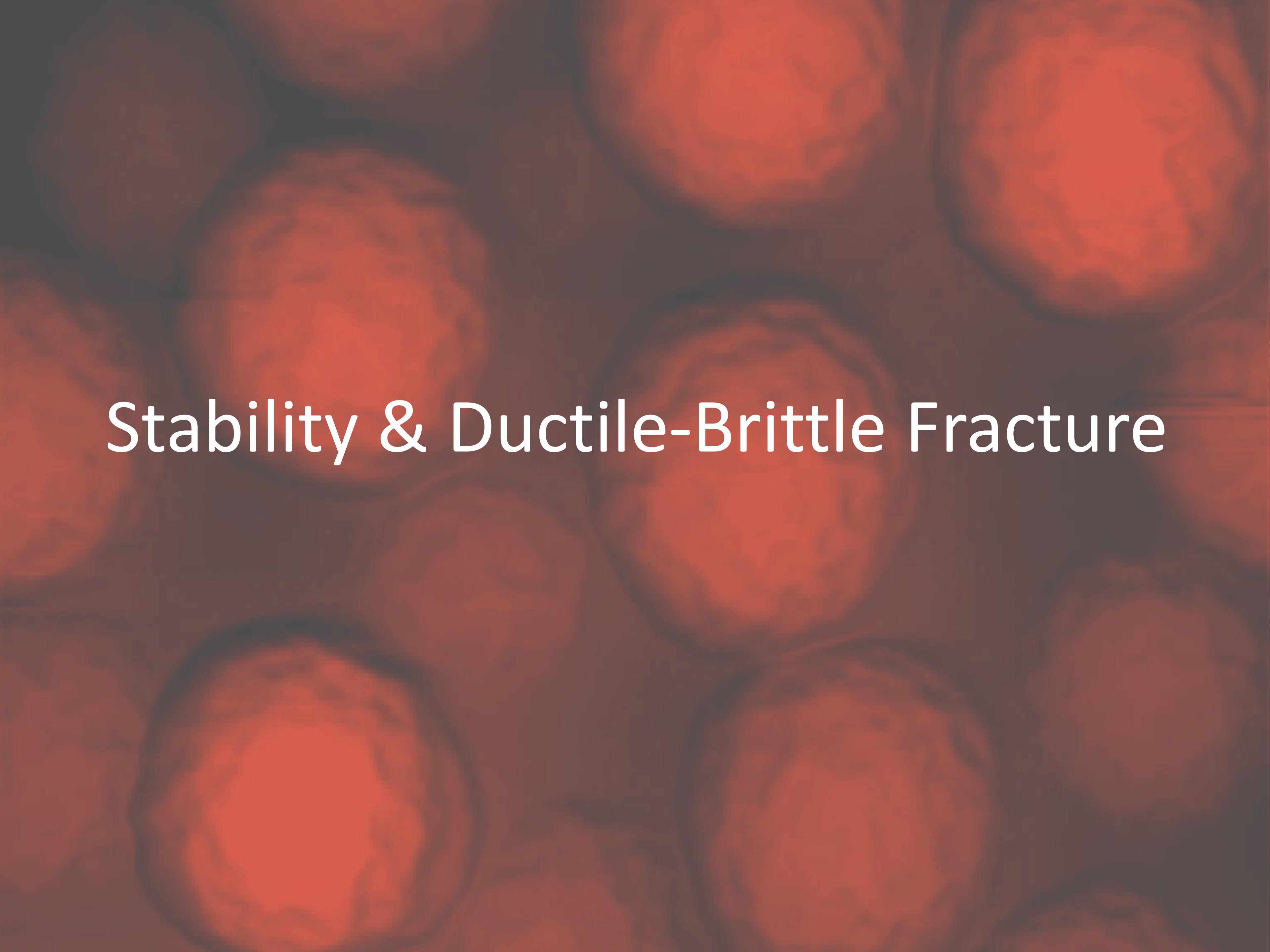
Logarithmic dependence of stress consistent with frictional contacts model

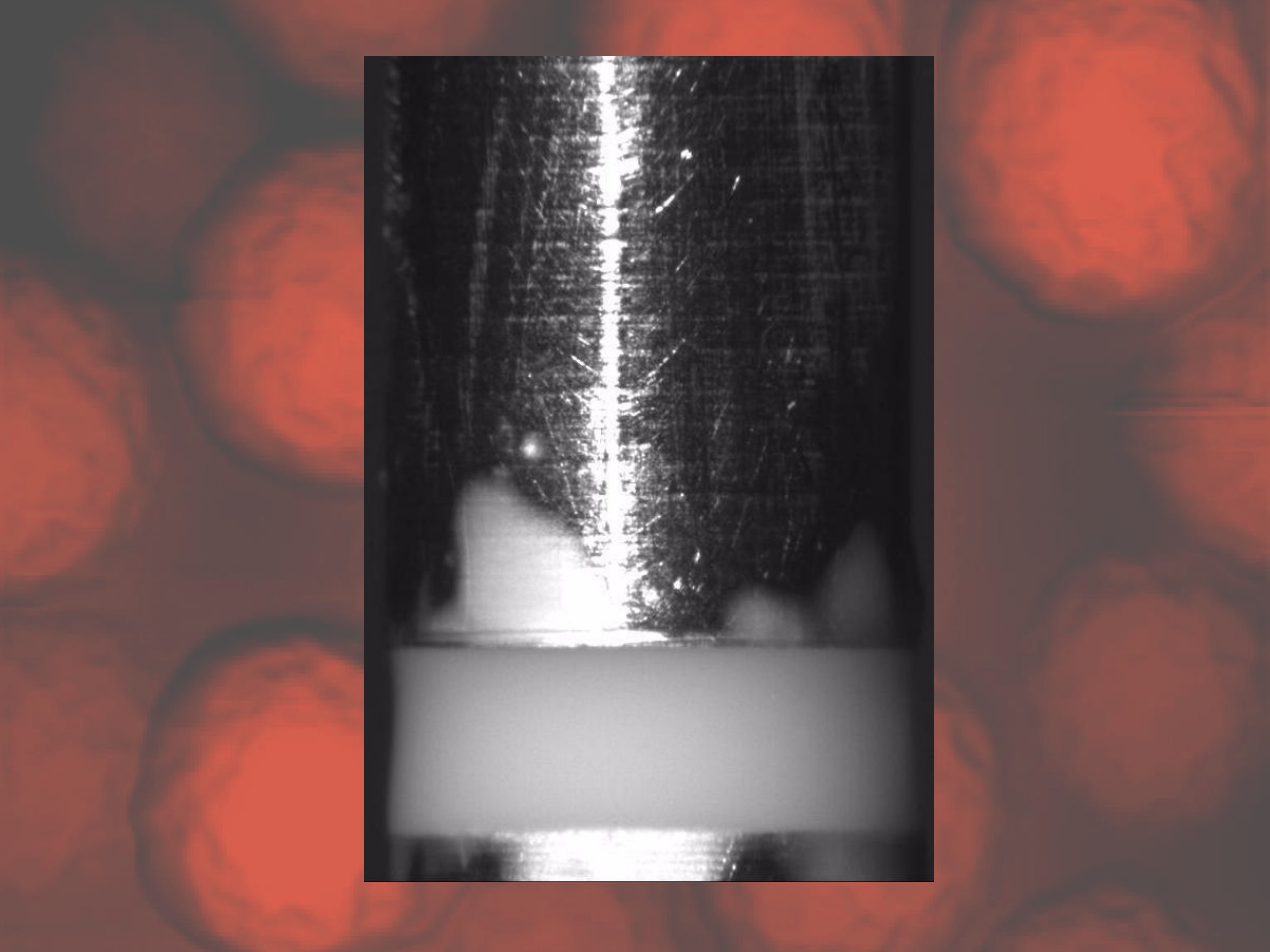
"Irreversible rearrangements of force chains leading to a strengthening of the network"?



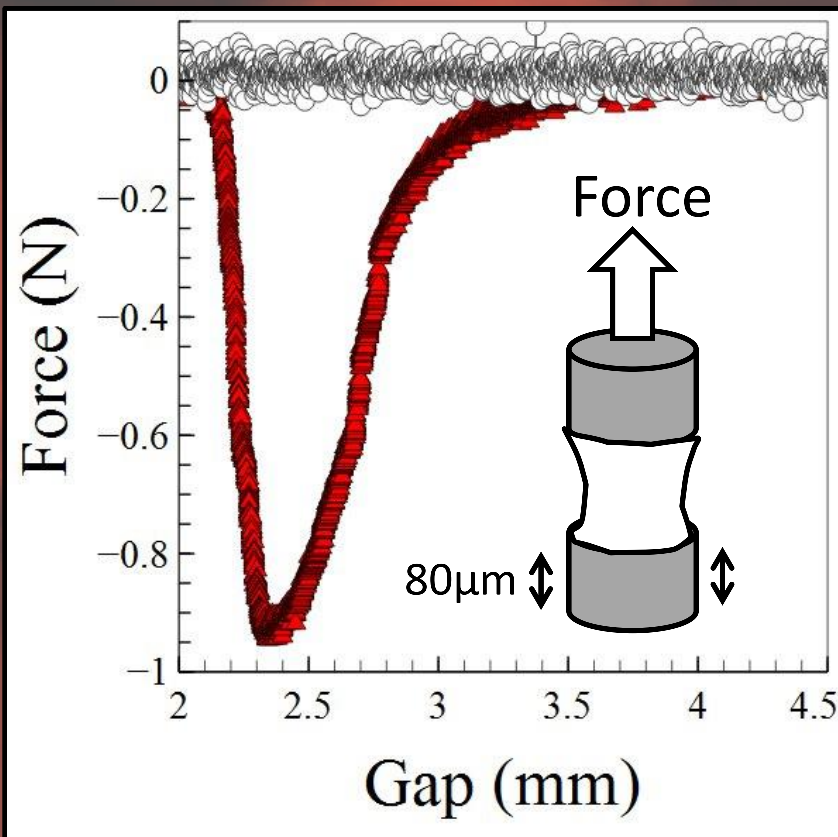
Hartley et al 2003

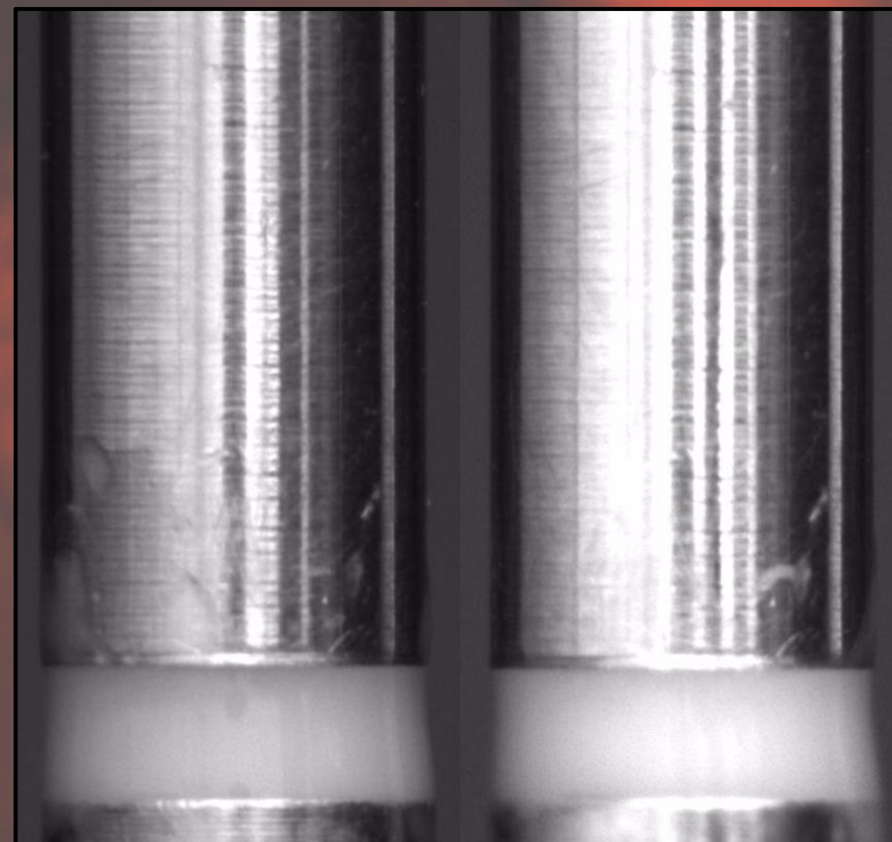
Stability & Ductile-Brittle Fracture



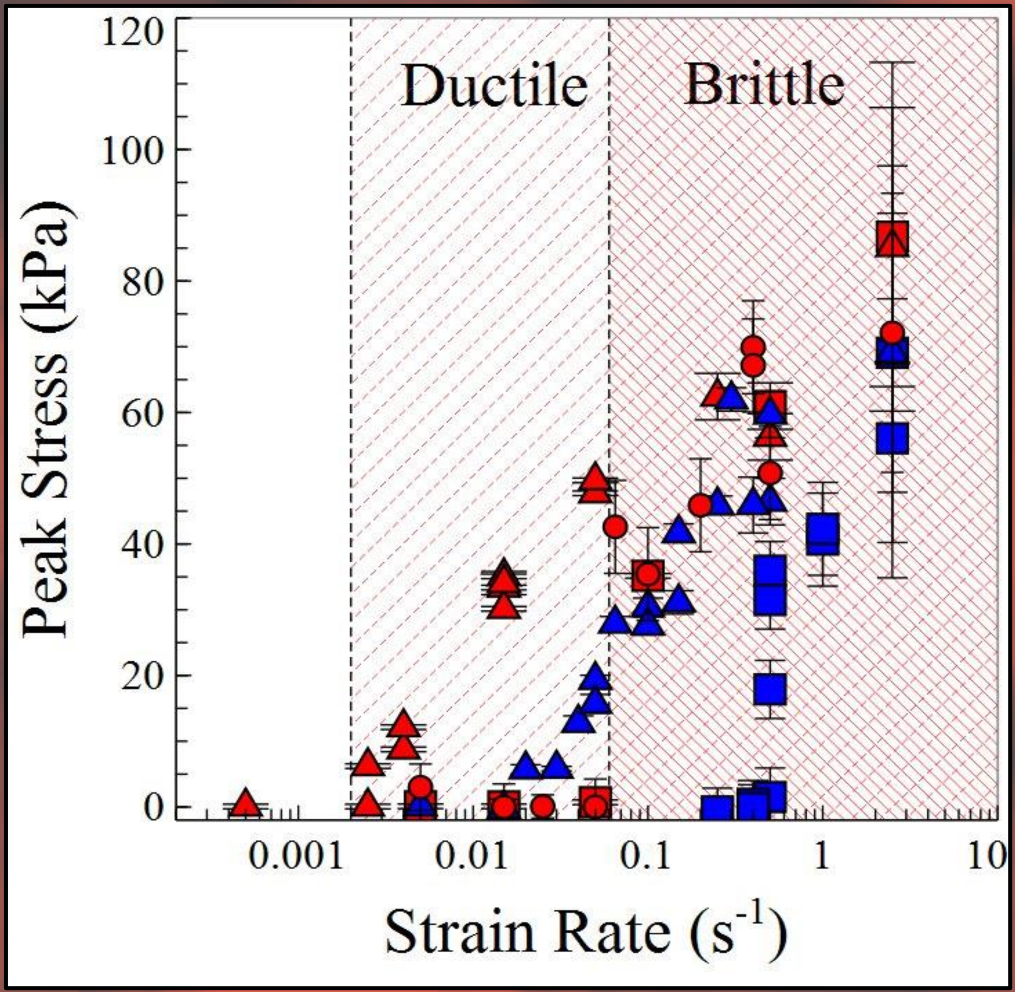


Using vibrations to modify the jamming transition





Two types of Jammed granule?

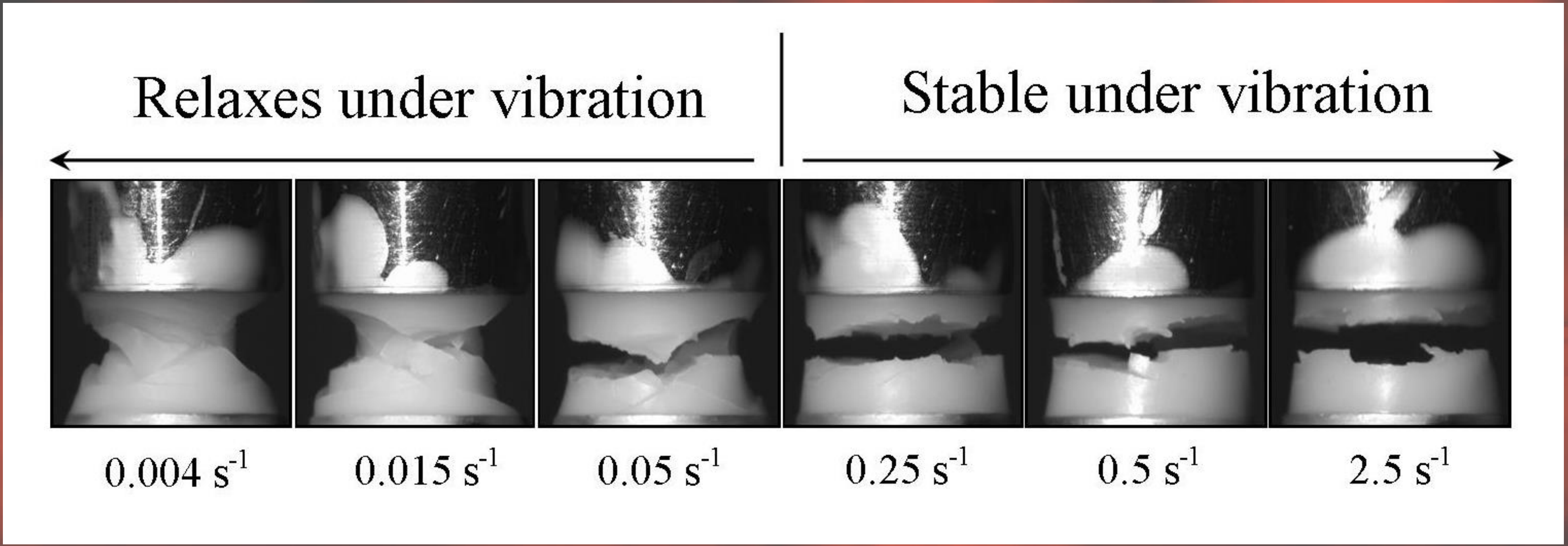


At low strain rates the jamming transition is suppressed

At higher strain rates the vibrations have little or no effect

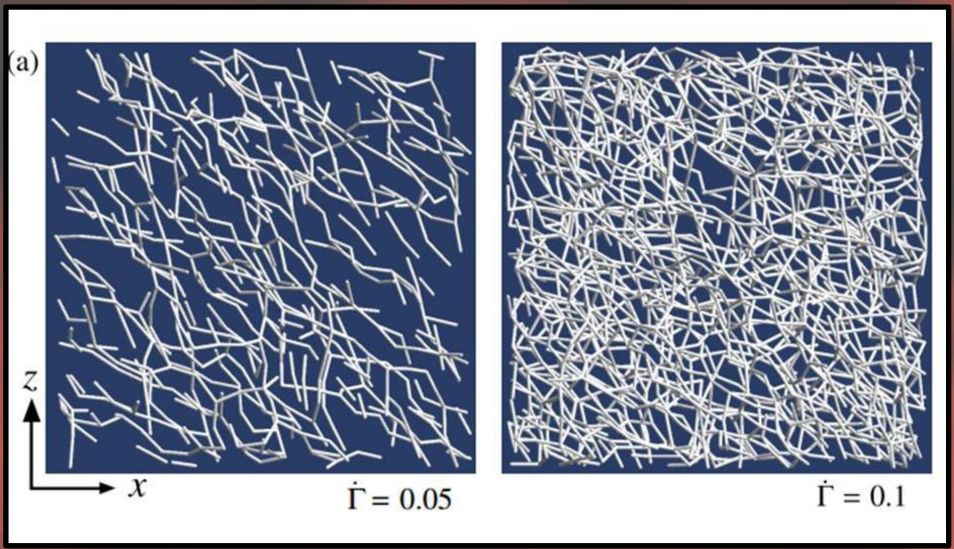
Two types of jammed state?

Fracture modes in the absence of vibration

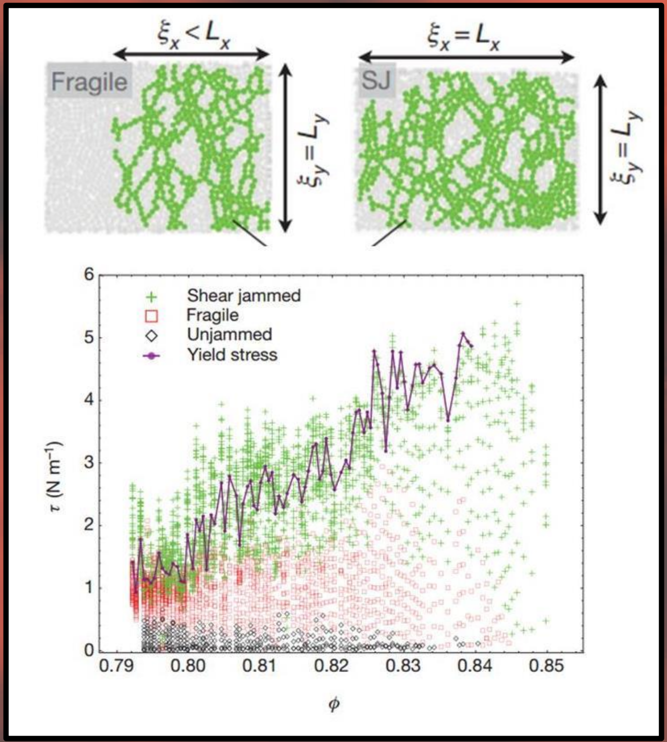


- $\dot{\varepsilon} < 0.1 s^{-1}$ fractures occur diagonally (maximum resolved shear stress)
- $\dot{\varepsilon} > 0.1 s^{-1}$ fractures form perpendicular to applied force
- Since force chains assemble to resist the applied flow this implies differences in the transient network in each case

Force Chain Isotropy



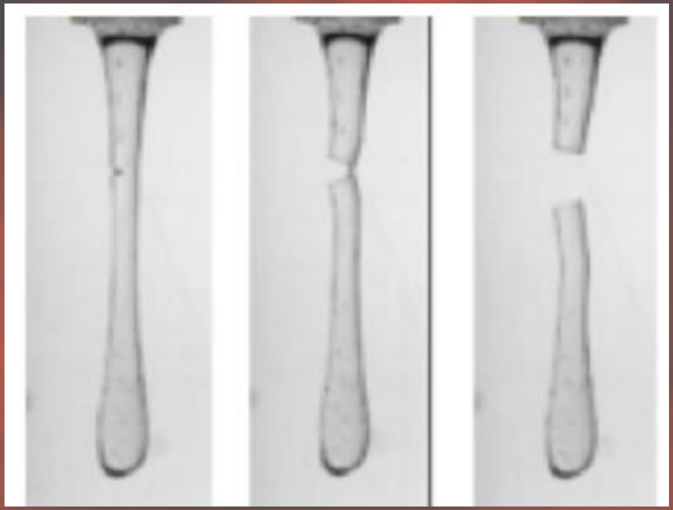
[Seto et al PRL 2013]



[Bi et al Nature 2011]

Above some critical strain rate / stress there is a change from anisotropic to isotropic particle contact networks

Fracture Modes in Complex Fluids



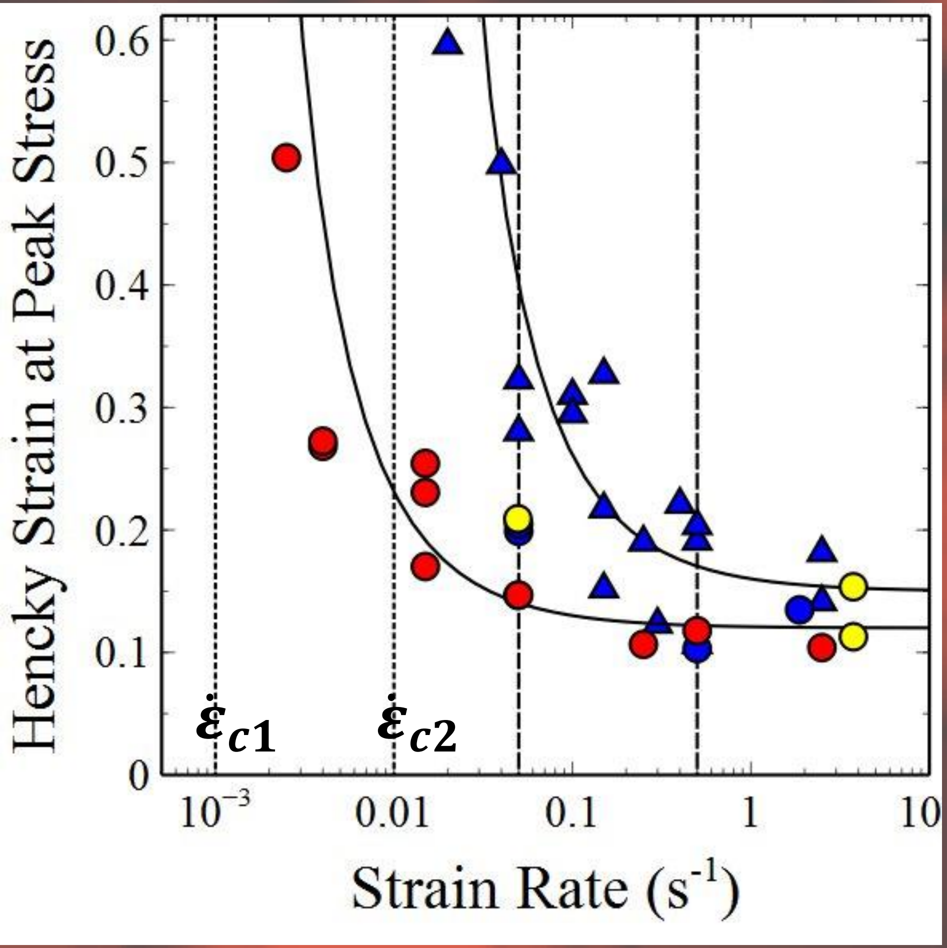
"Brittle fracture"

A crosslinking microemulsion transient network

Two timescales : Network formation & Bond rupture (related to applied stress or strain rate)

In a force chain network we expect network formation to be related to strain rate

Gap at fracture



$$\varepsilon(\dot{\varepsilon}) = \frac{1}{\left(\dot{\varepsilon}/\dot{\varepsilon}_c - 1\right)} + \varepsilon_0$$

Brittle fracture occurs when the relaxation rate of the network is insignficant.

May suggest that the ductile-brittle transition is a limiting case of the same physics underlying the jamming transition

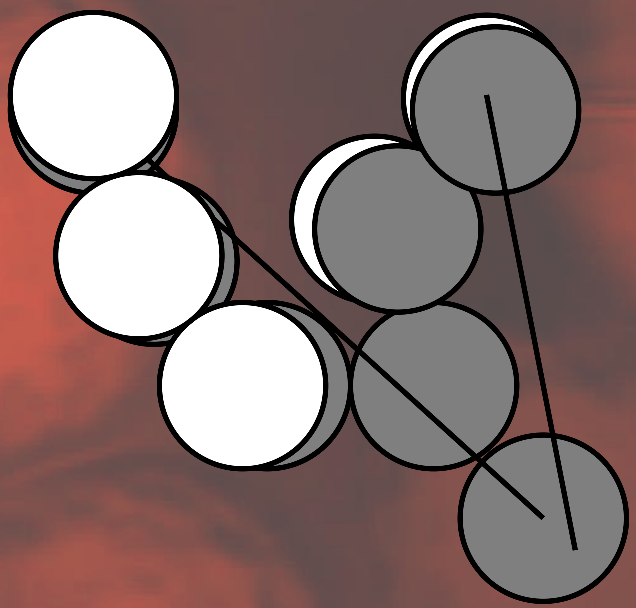
Why do vibrations suppress jamming in the ductile region?

Ductile system:

- small irreversible changes in particle position can lead to large changes in the network of force chains

Brittle network:

- Reversibility leads to more consistent contact network
- Force chains are likely to be more isotropic; even if the structure changes the network can resist the confining capillary stresses.



Conclusions

- Above a critical strain rate hard sphere colloidal suspensions undergo dilatancy induced jamming and granulation.

- Above the jamming transition we observe a transition from plastic to elastic responses in granules.

M.I. Smith, R. Besseling, M.E. Cates, V. Bertola
Nature Communications 1, 114 (2010)

Acknowledgements

School of Engineering, University of Liverpool, UK

Dr Volfango Bertola

School of Physics, University of Edinburgh, UK

Professor Mike Cates

Dr Rut Besseling

Dr Andrew Schofield

School of Physics, University of Nottingham, UK

Dr James Sharp

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