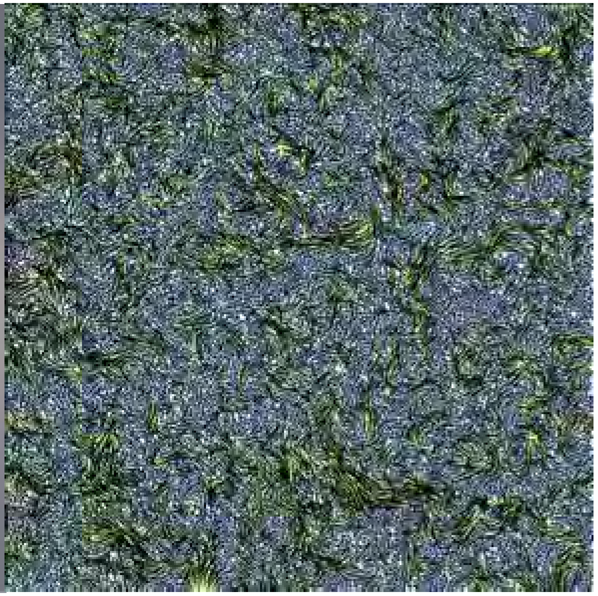
Future Formulation IV, 23 June 2020

Establishing principles for formulation and processing of high-solid-content dispersions of complex compositions in complex flows

Cornstarch dispersed in water



Simulation showing Particle displacement

Future Formulation IV, 23 June 2020

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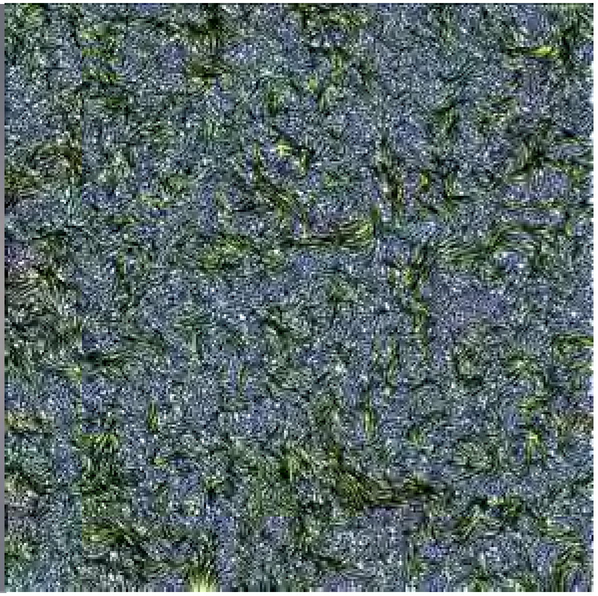

Simulation showing Particle displacement

University of Edinburgh

Jin Sun
School of Engineering
John Royer and Wilson Poon
School of Physics and Astronomy

University of Strathclyde

Mark Haw
Department of Chemical and Process Engineering

EPSRC
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University of Strathclyde Glasgow

Researchers contributed to this presentation

University of Edinburgh
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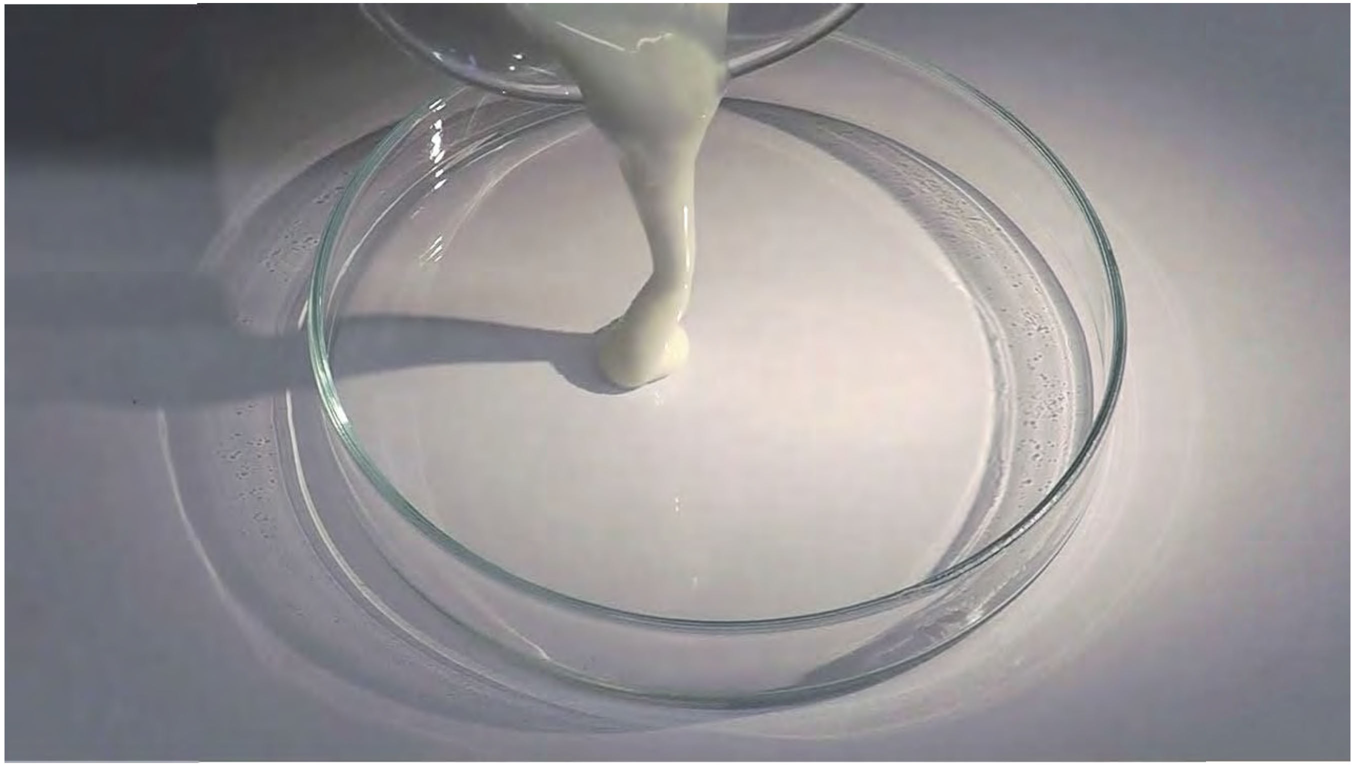
University of Strathclyde
Claire Forsyth, Christopher Boyle, Aditi Mukhopadyay, Leo Lue, Jose Ruiz-Lopez

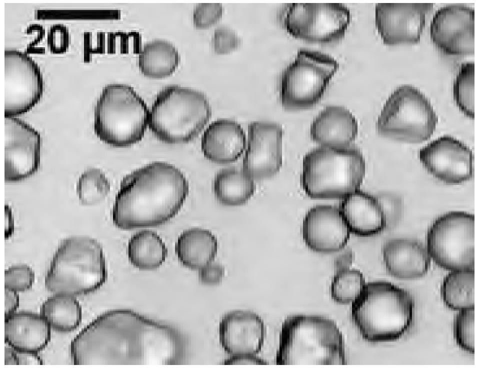
High-solid-content dispersions



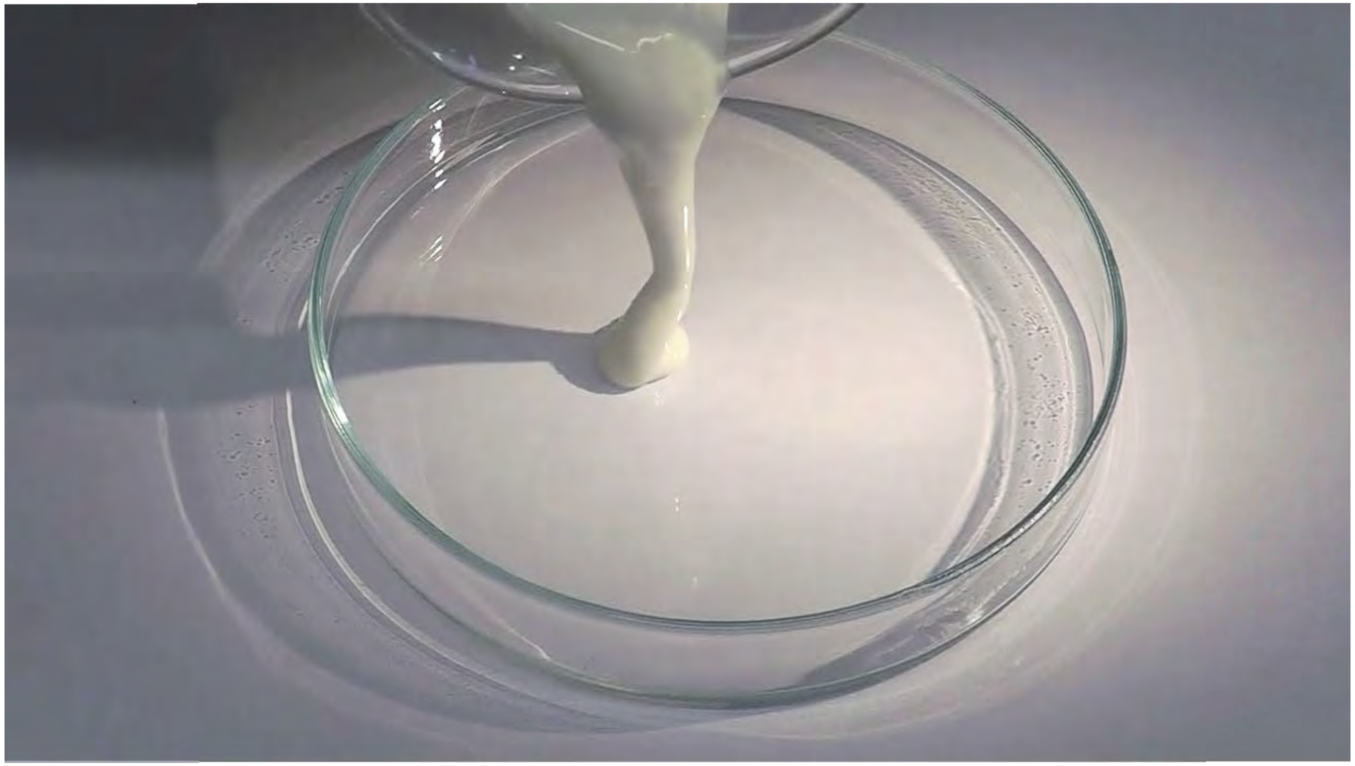
Video from ETH Soft Materials Group

Cornstarch mixed with water at high solid concentration.

Cornstarch particle size ~ 10 μm



High-solid-content dispersions



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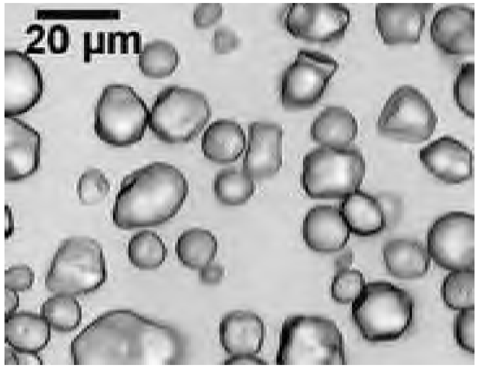

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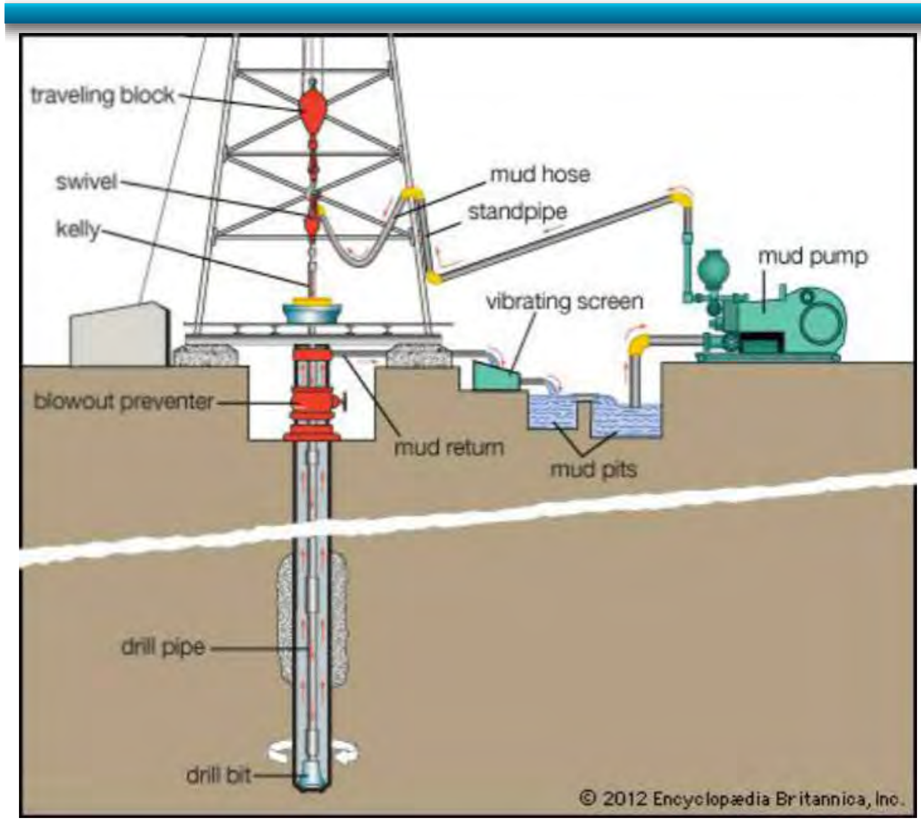
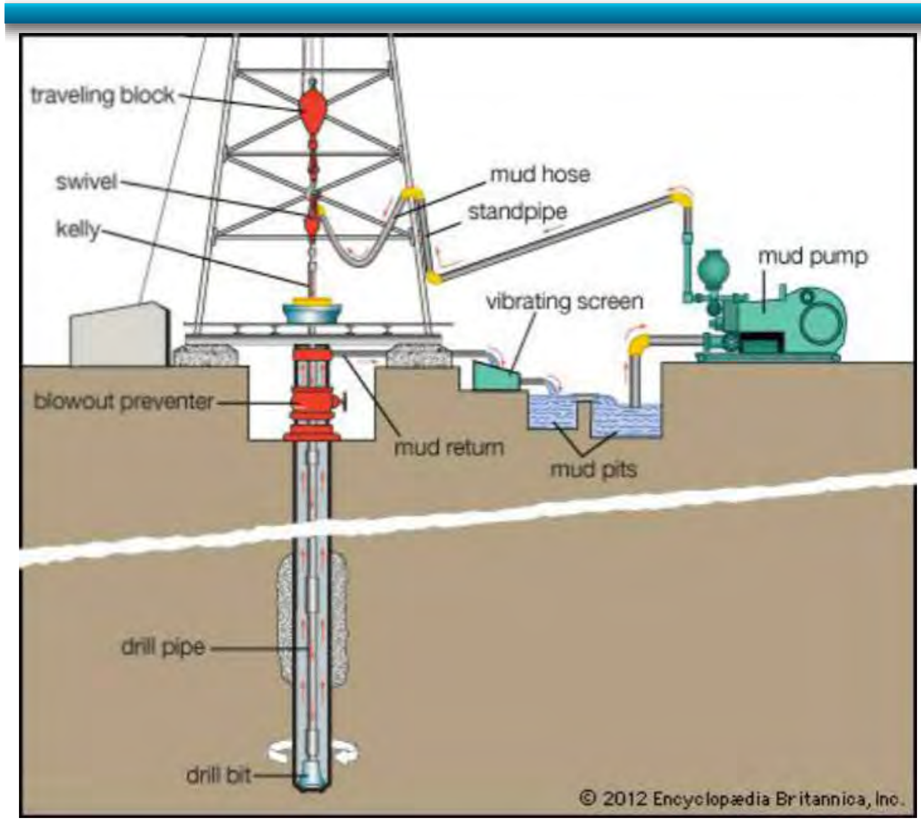
Industrial applications



Drilling fluids

Industrial applications



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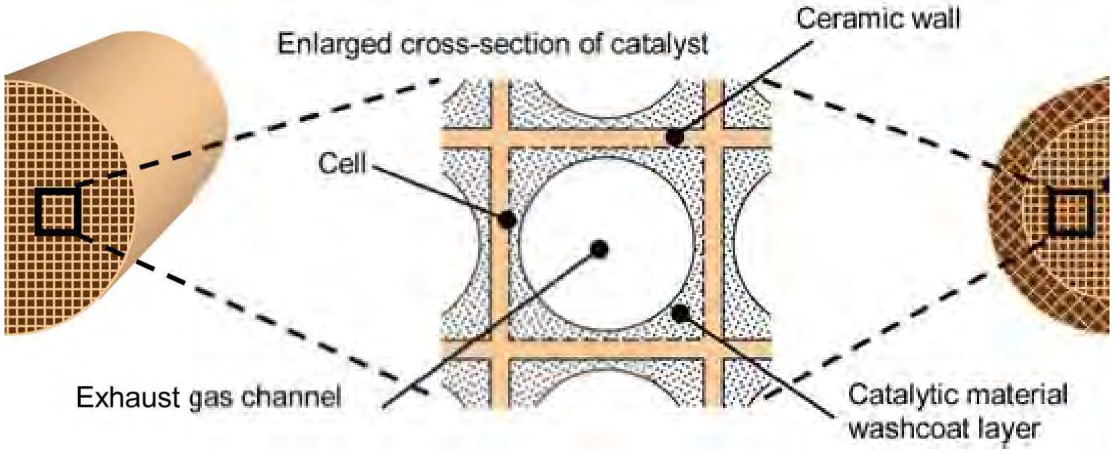
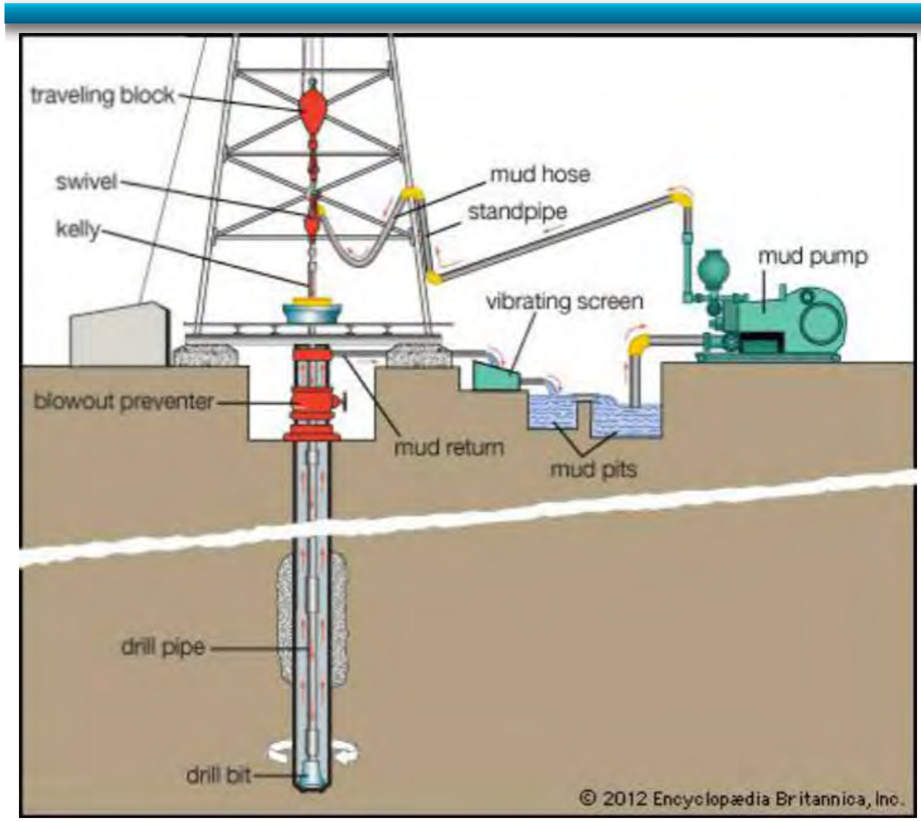


Catalyst production

Industrial applications



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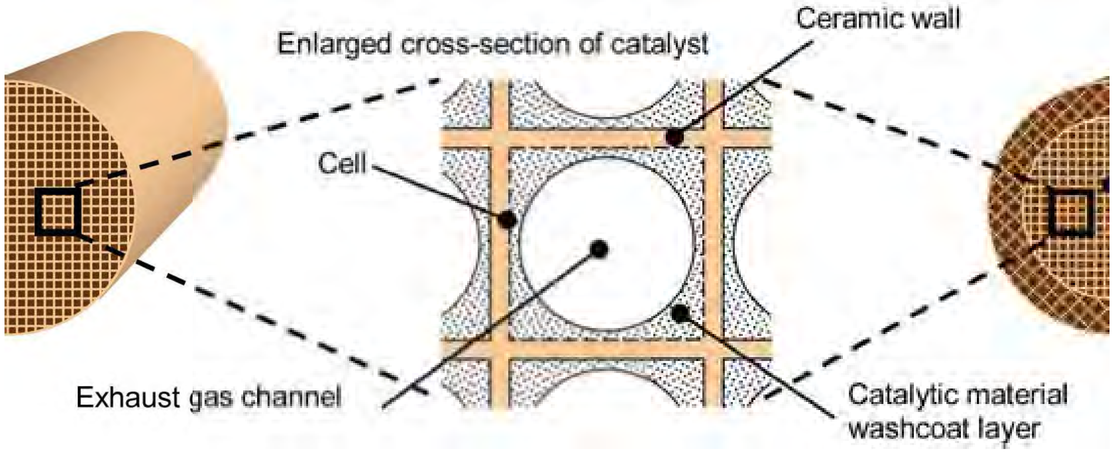
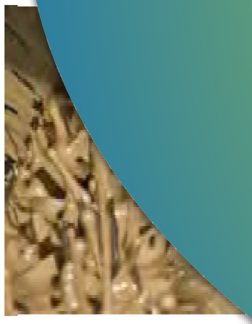
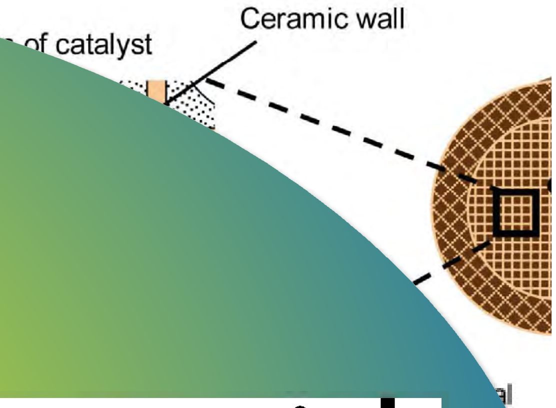


Catalyst production

Paint

Industrial applications



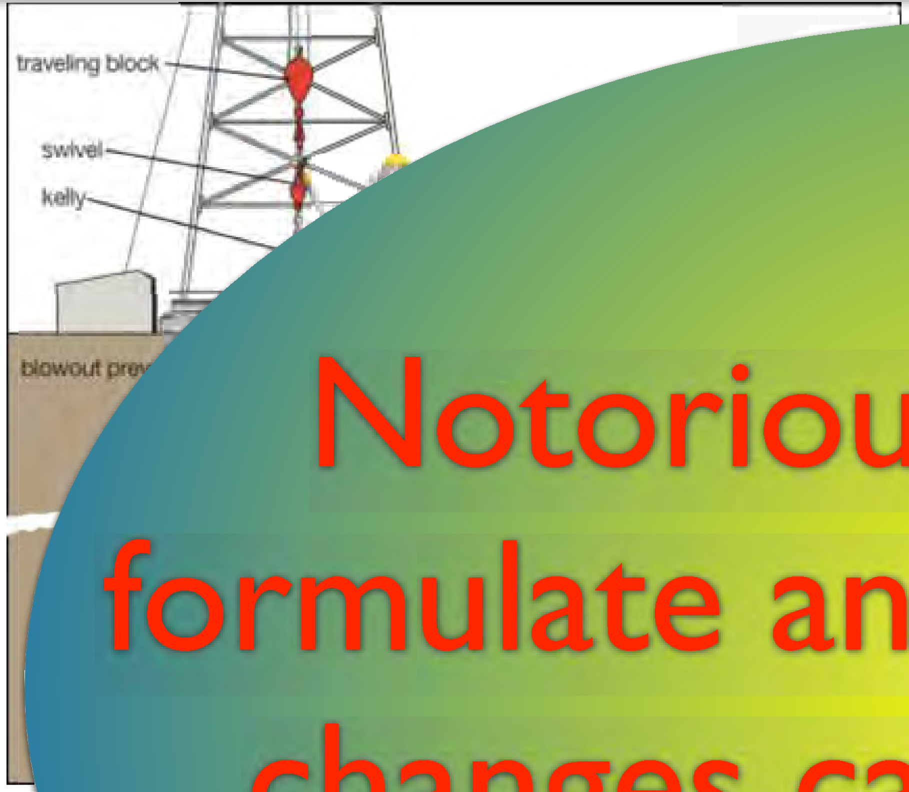


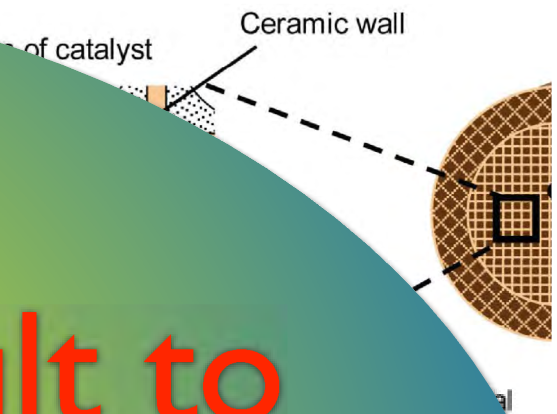
Maximise solid content with desired flow in complex geometries

Drilling

Paint

Industrial applications





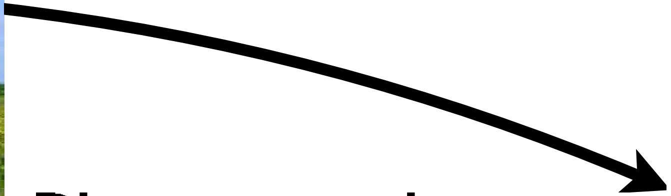
Notoriously difficult to formulate and control — tiny changes can lead to large fluctuations, jams…

Drilli

Paint

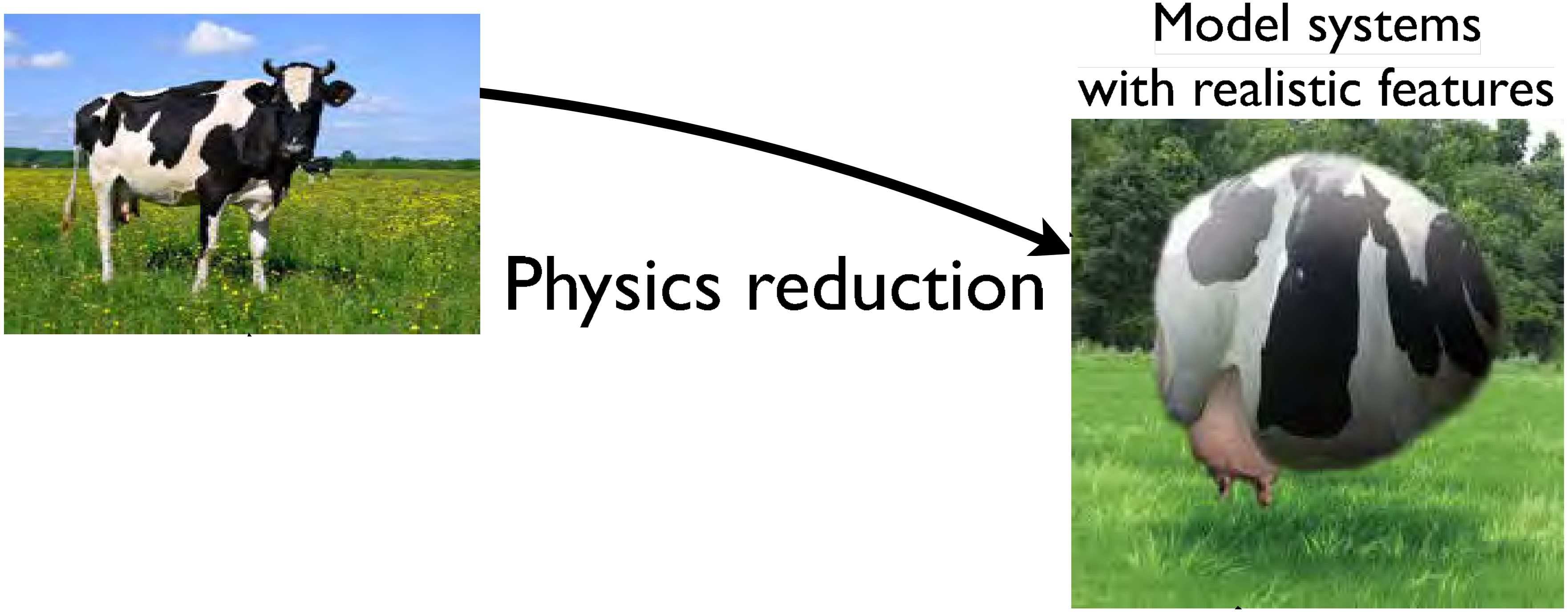
Project logic

Project logic



Physics reduction

Project logic



Physics reduction

Model systems
with realistic features

Project logic

Physics reduction

Model systems
with realistic features

Engineering emergence

Project logic

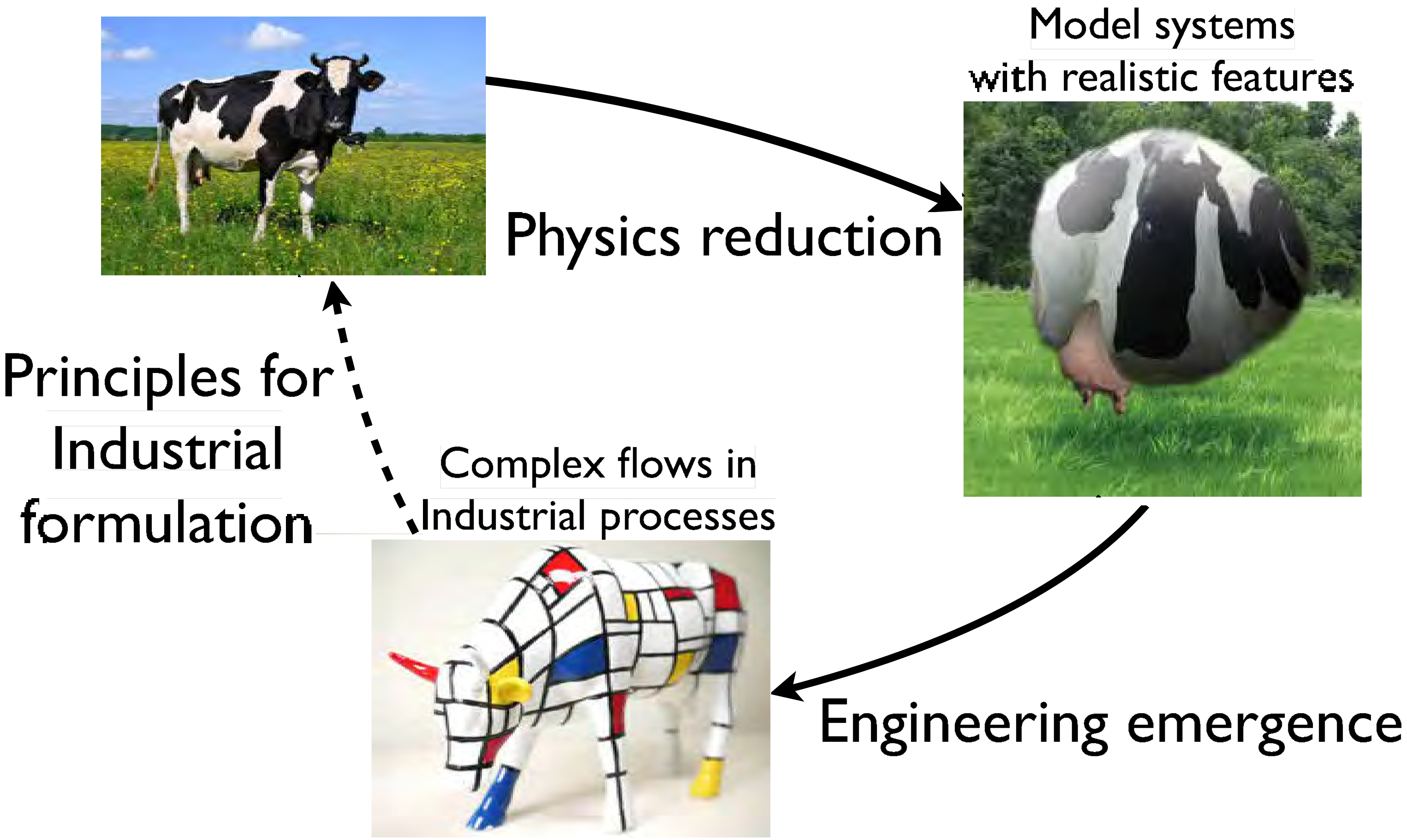
Physics reduction

Model systems
with realistic features

Engineering emergence

Complex flows in
Industrial processes

Project logic



Model systems
with realistic features

Physics reduction

Engineering emergence

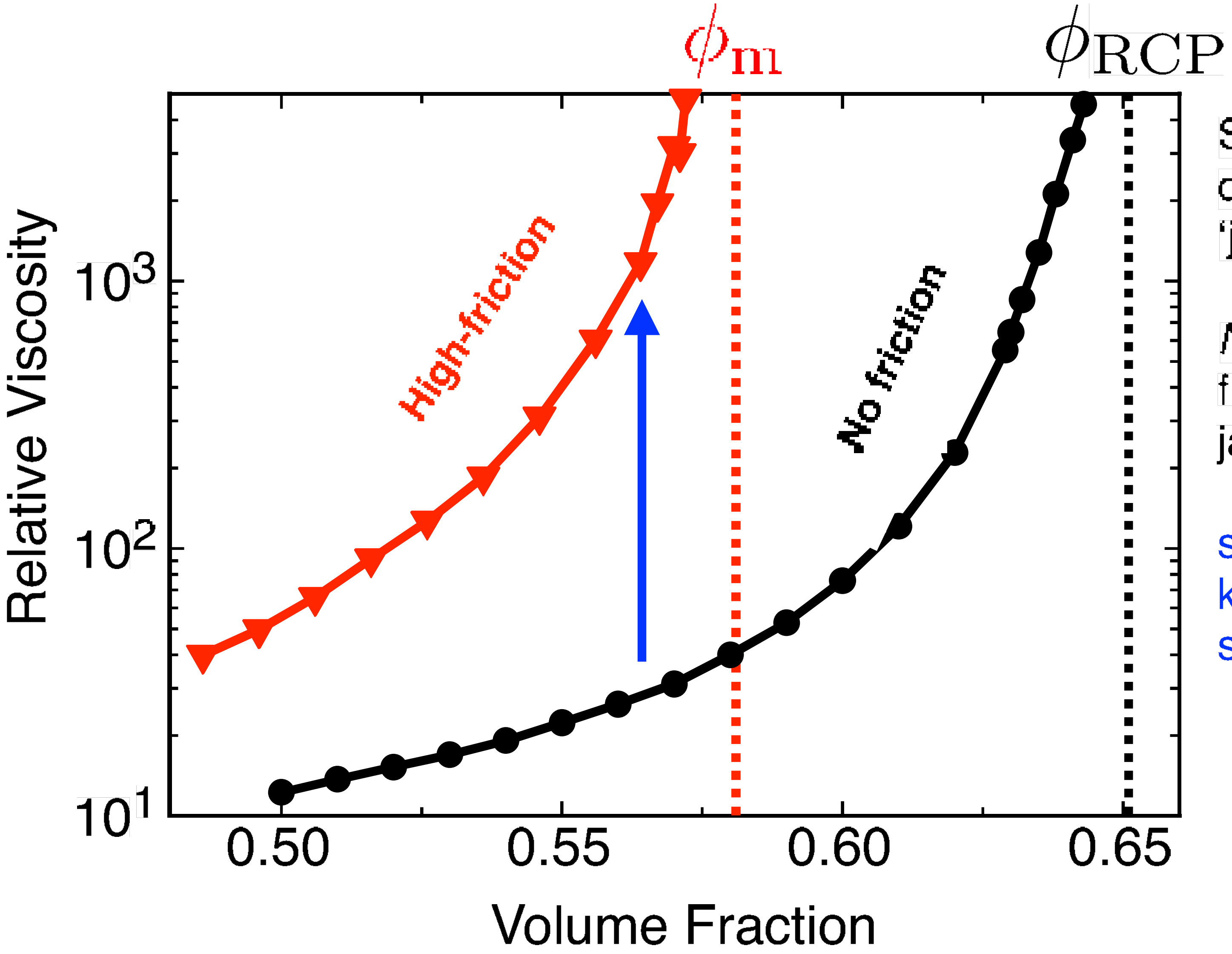
Complex flows in
Industrial processes

Principles for
Industrial
formulation

Research outcomes

- Elucidated roles of particle interactions in determining HSCD rheology, especially shear thickening
- Explored the effects of multiple particles types, including different interactions and sizes
- Connected the rheology to complex flows, including extrusion, oscillations and super-imposed flows

Jamming in dense dispersions



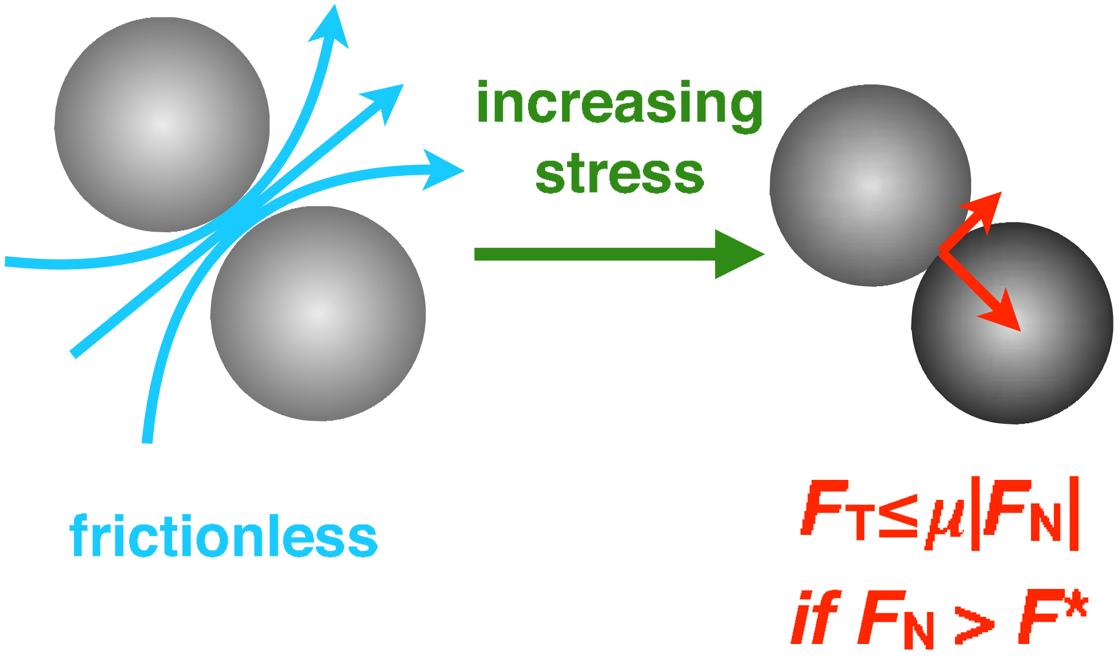
Suspension viscosity diverges at a critical 'jamming' concentration

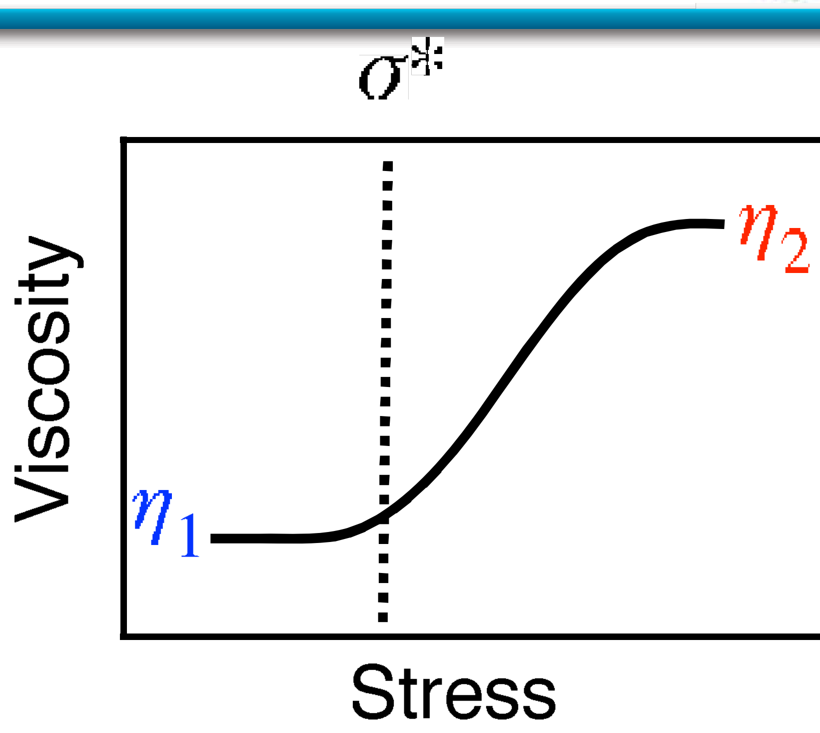
New understanding: frictional contacts shift jamming point

stress-induced friction: key to understanding shear-thickening

Seto, Mari, Morris, PRL 2013, JoR 2014, Wyart and Cates, PRL 2014, Guy, Hermes, Poon, PRL 2015, Royer, Blair, Hudson, PRL 2016 and many others

Shear thickening





Shear thickening is due to stress-driven frictional contacts

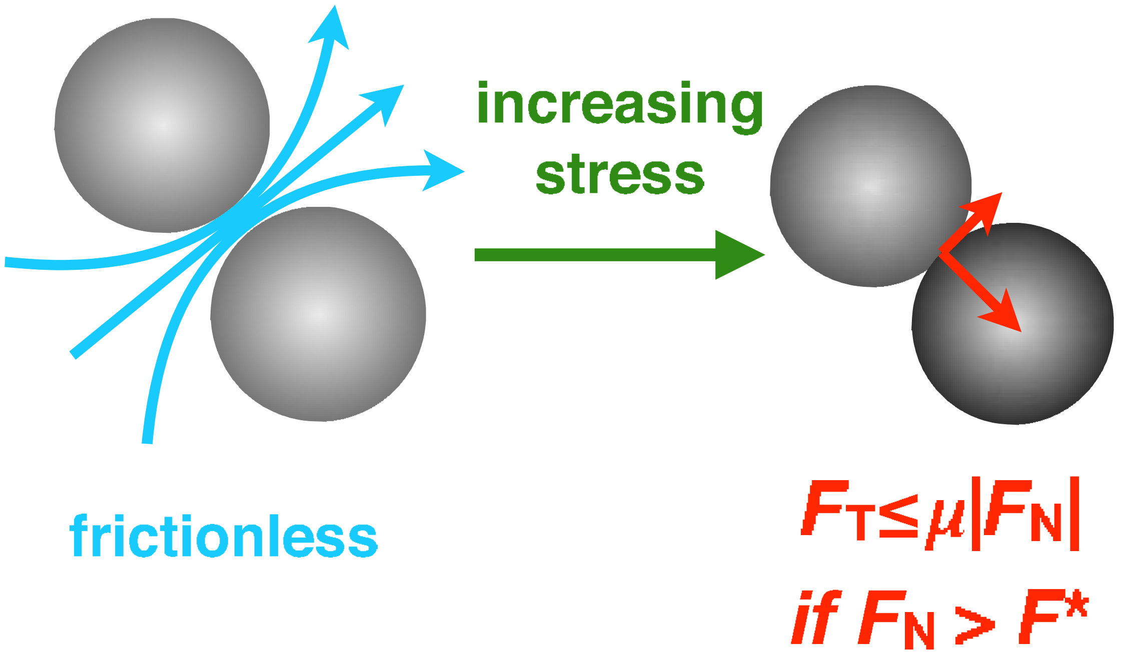
$$\eta_r = \left(1 - \frac{\phi}{\phi_J}\right)^{-2}$$ Wyart-Cates model

$$\phi_J(\sigma) = \phi_{\mathrm{m}} f(\sigma) + (1 - f(\sigma))\,\phi_{\mathrm{RCP}}$$

$f(\sigma) =$ ‘fraction of frictional contacts’

[1] Fernandez et al, PRL 2013, [2] Seto, Mari, Morris, PRL 2013, JoR 2014 [3] Wyart and Cates, PRL 2014, [4] Guy, Hermes, Poon, PRL 2015 [5] Lin et al, PRL 2015, [6] Royer, Blair, Hudson, PRL 2016 [7] Clavaud et al PNAS 2017, [8] Comtet et al, Nat. Comm. 2017

Shear thickening

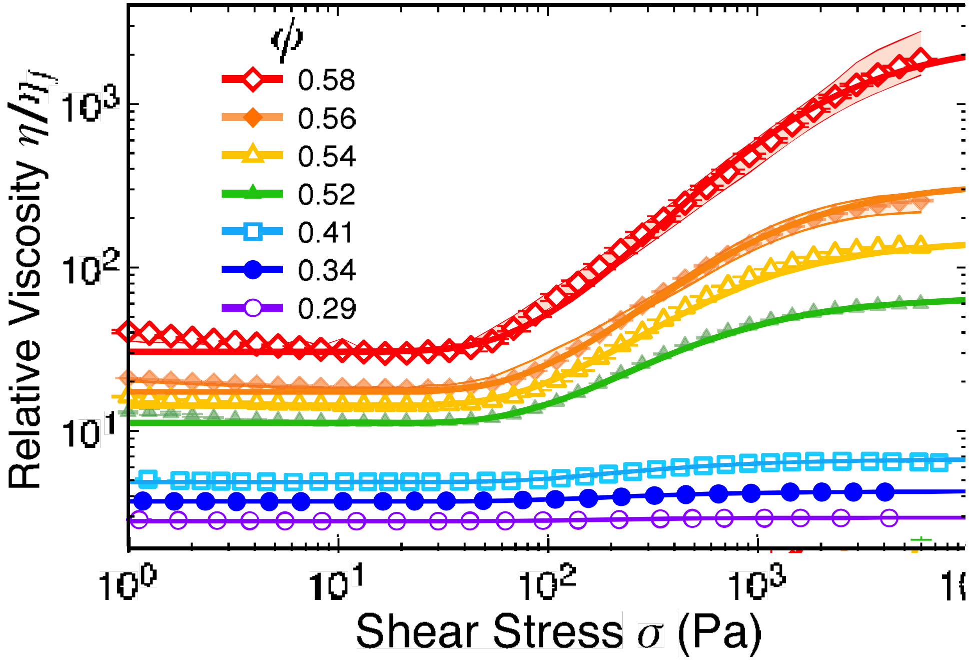


increasing stress

frictionless

$F_T \leq \mu |F_N|$

if $F_N > F^*$



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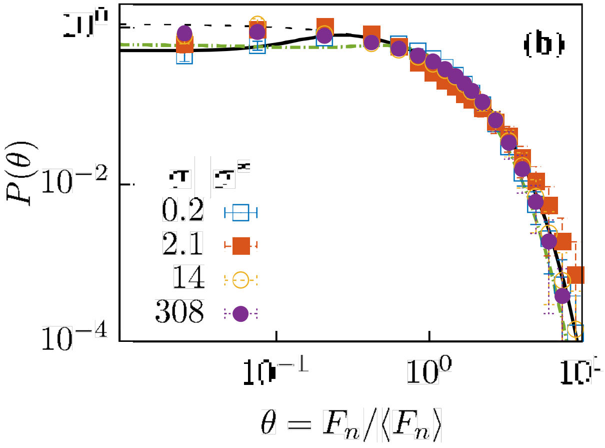
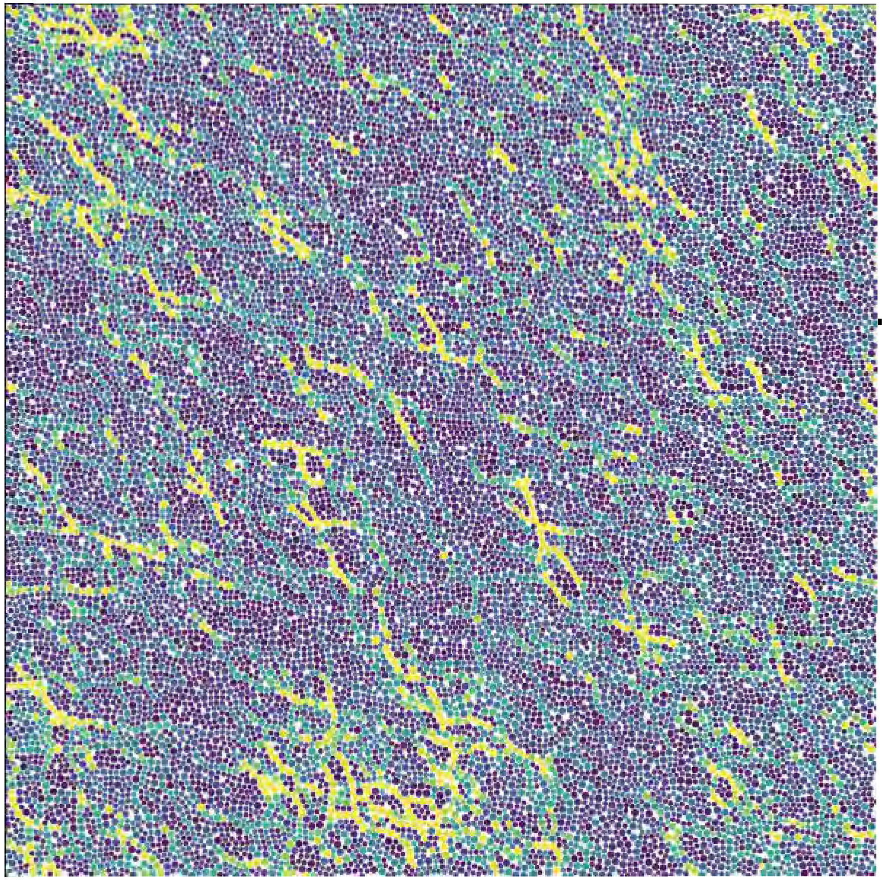
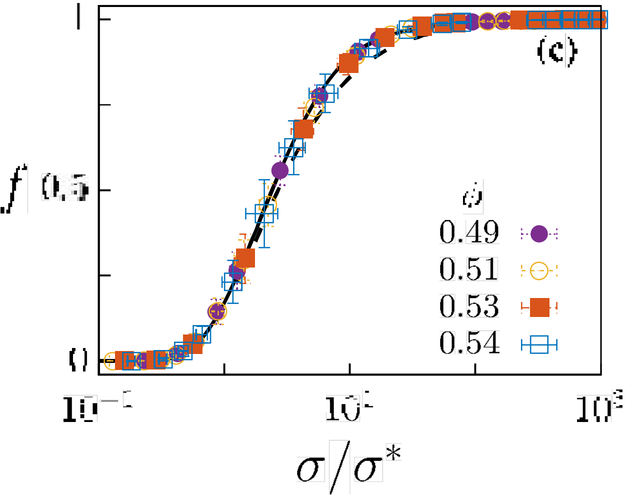
Contacts in dense dispersions



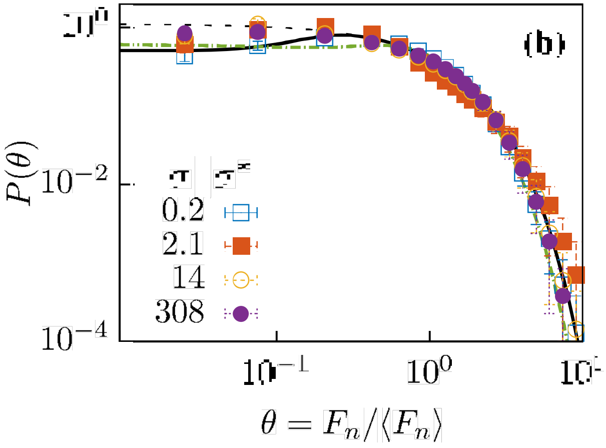


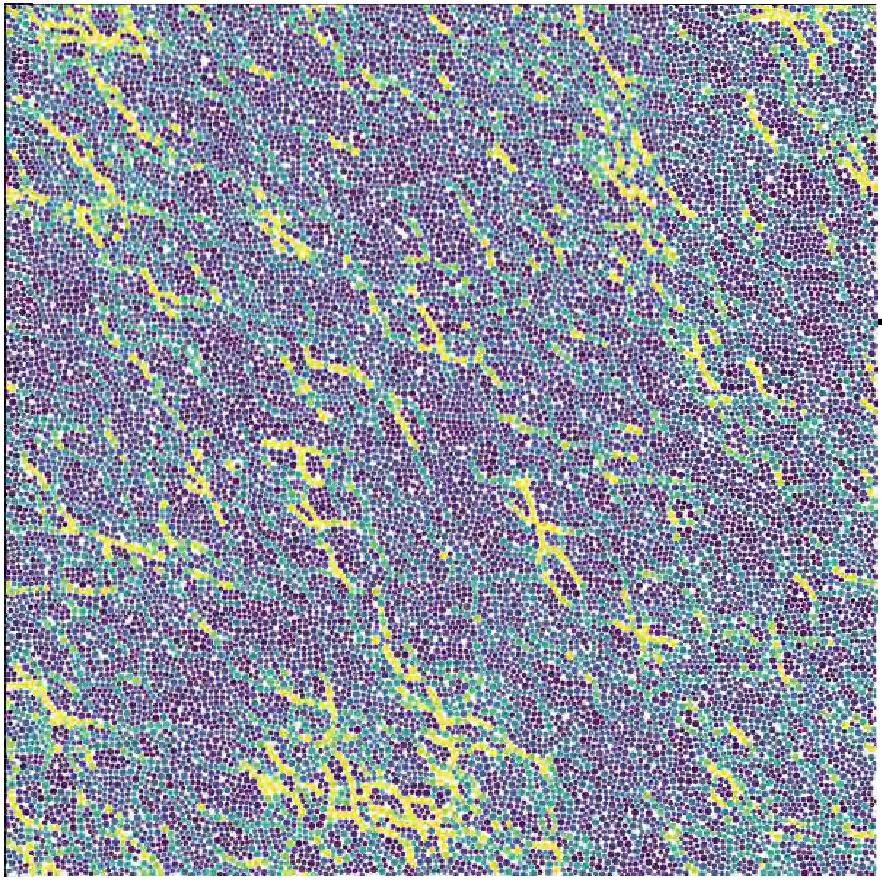


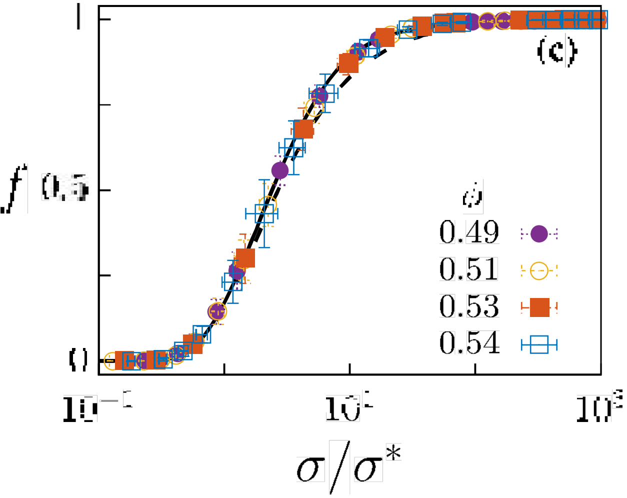
$$f(\sigma) \sim e^{-\sigma^*/\sigma}$$

- Characterise frictional contacts
- Similar force distribution in dry and wet granular dispersions

Contacts in dense dispersions







$$f(\sigma) \sim e^{-\sigma^*/\sigma}$$

- Characterise frictional contacts
- Similar force distribution in dry and wet granular dispersions

Granular Matter (2020) 22:29
https://doi.org/10.1007/s10035-019-0992-5

ORIGINAL PAPER

Force chains and networks: wet suspensions through dry granular eyes

Rangarajan Radhakrishnan[1] · John R. Royer[2] · Wilson C. K. Poon[2] · Jin Sun[1]

Received: 9 April 2019 / Published online: 27 January 2020

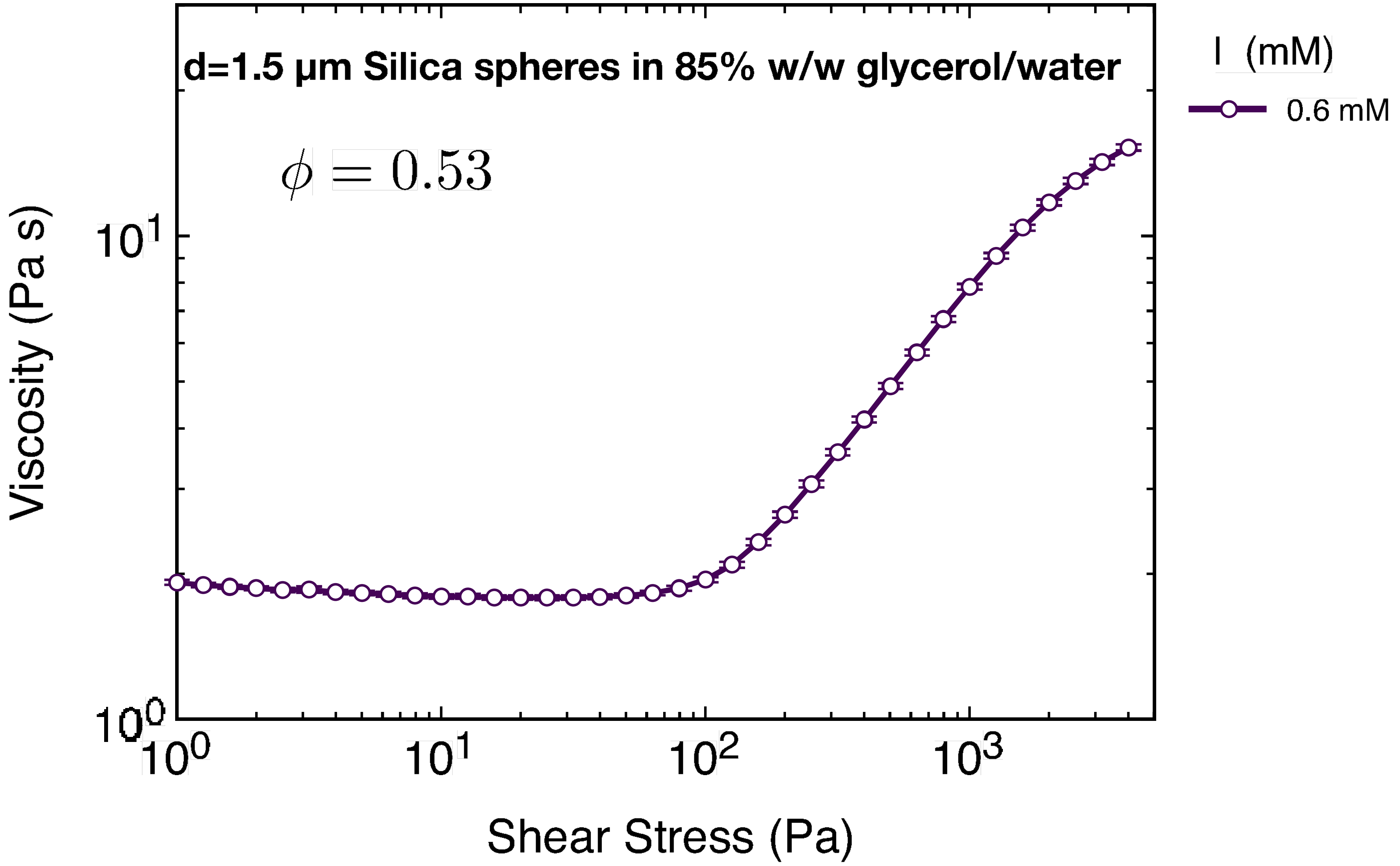
Abstract

Recent advances in shear-thickening suspension rheology suggest a relation between (wet) suspension flow below jamming and (dry) granular physics. To probe this connection, we simulated the contact force networks in suspensions of non-Brownian spheres using the discrete element method, varying the particle friction coefficient and volume fraction. We find that force networks in these suspensions show quantitative similarities to those in jammed dry grains. As suspensions approach the jamming point, the extrapolated volume fraction and coordination number at jamming are similar to critical values obtained for isotropically compressed spheres. Similarly, the shape of the distribution of contact forces in flowing suspensions is remarkably similar to that found in granular packings, suggesting potential refinements for analytical mean field models for the rheology of shear thickening suspensions.

Keywords Suspension rheology · Granular materials · Network properties · DEM simulations

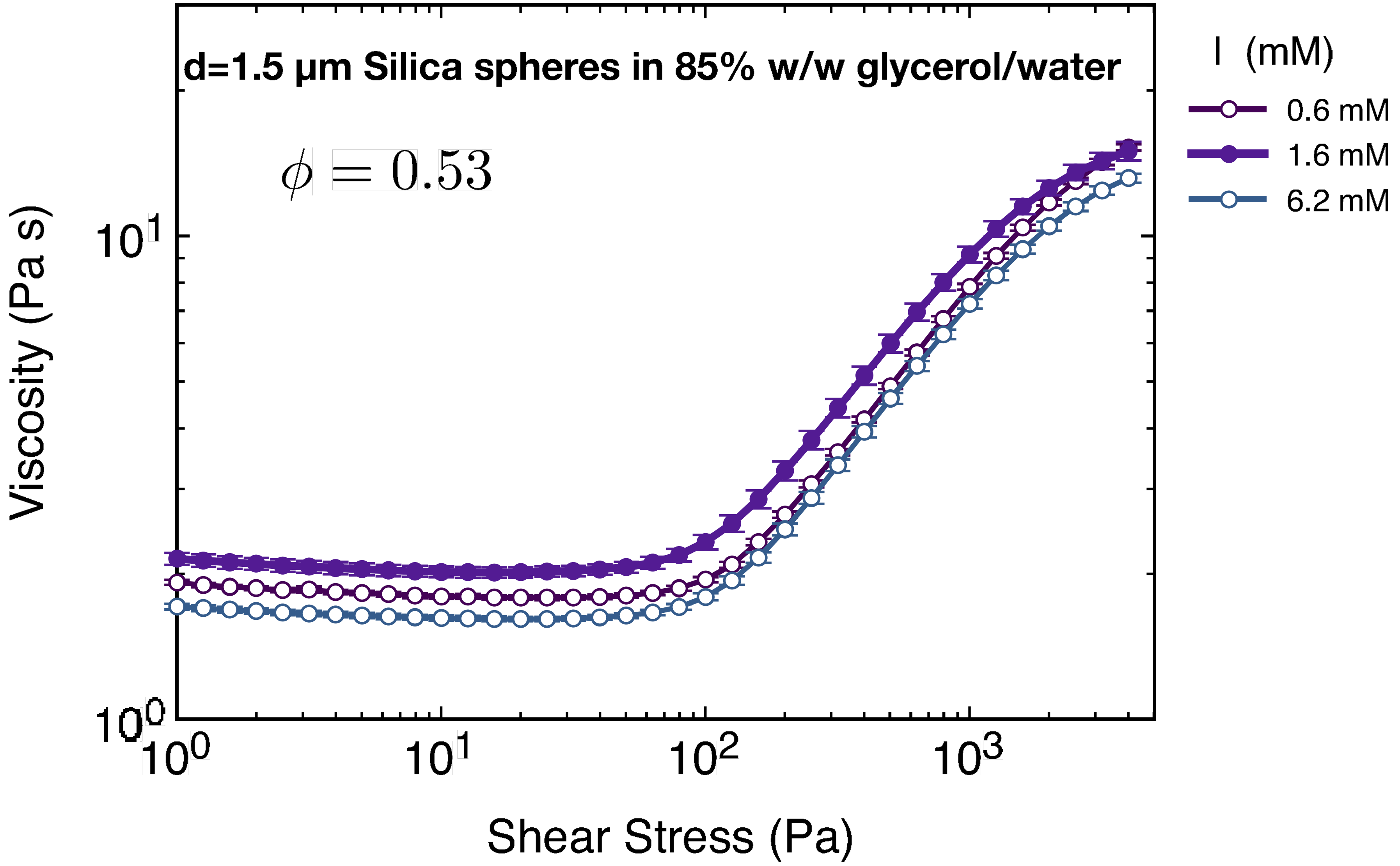
Controlling shear thickening critical stress through tuning particle repulsion

Suspension with tuneable repulsion



- Decreasing repulsion leads to lower critical stress

Suspension with tuneable repulsion



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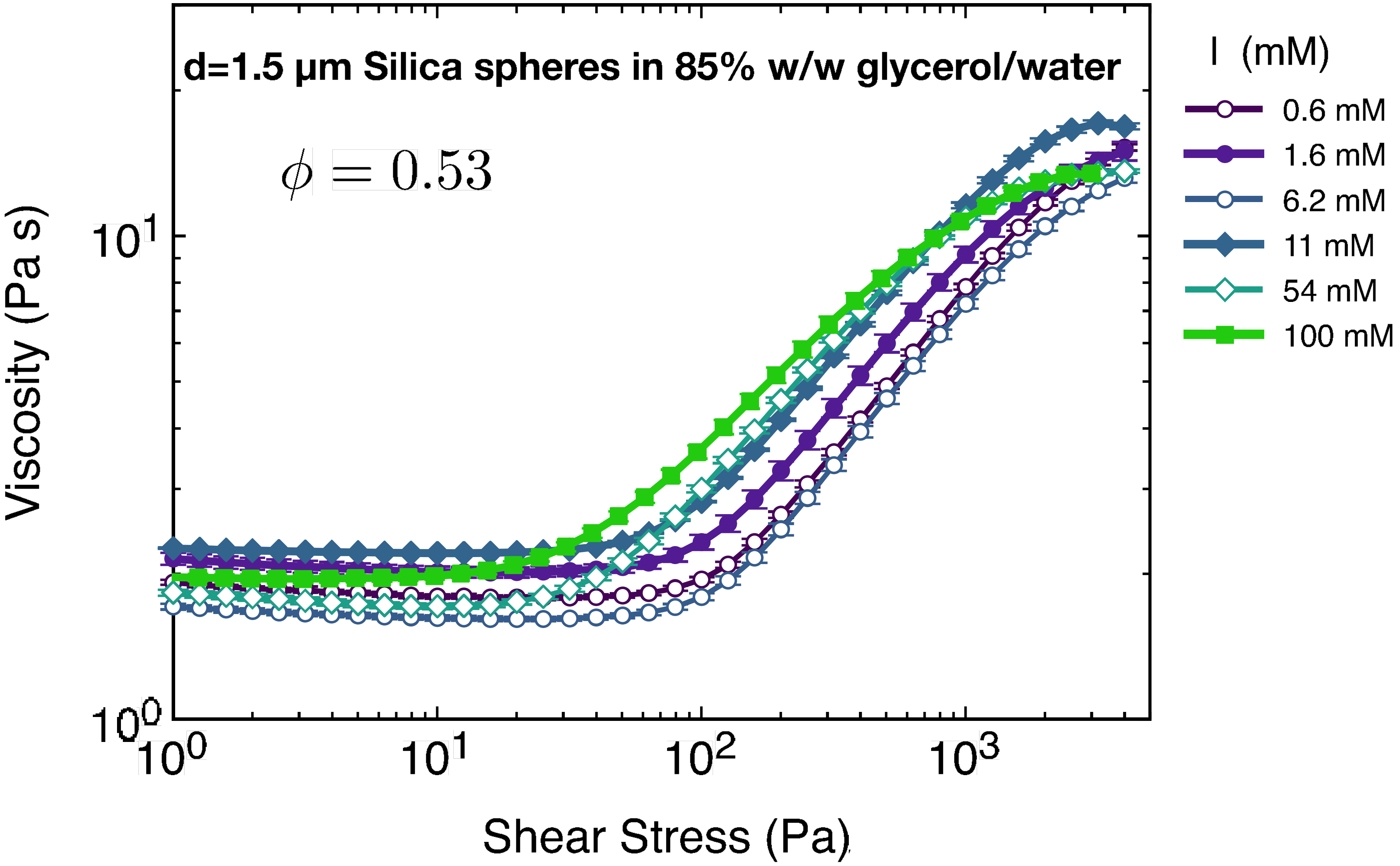

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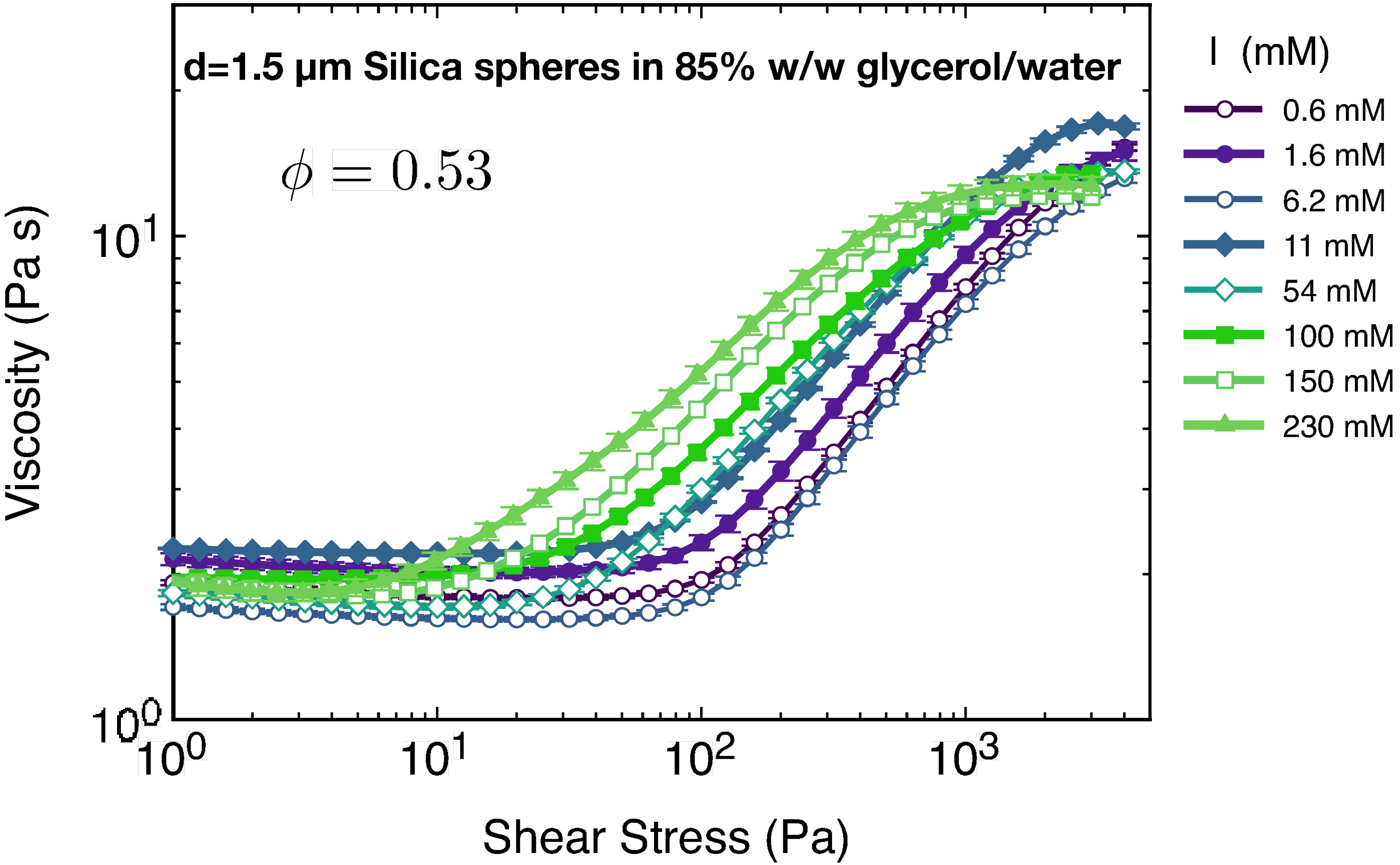
Suspension with tuneable repulsion



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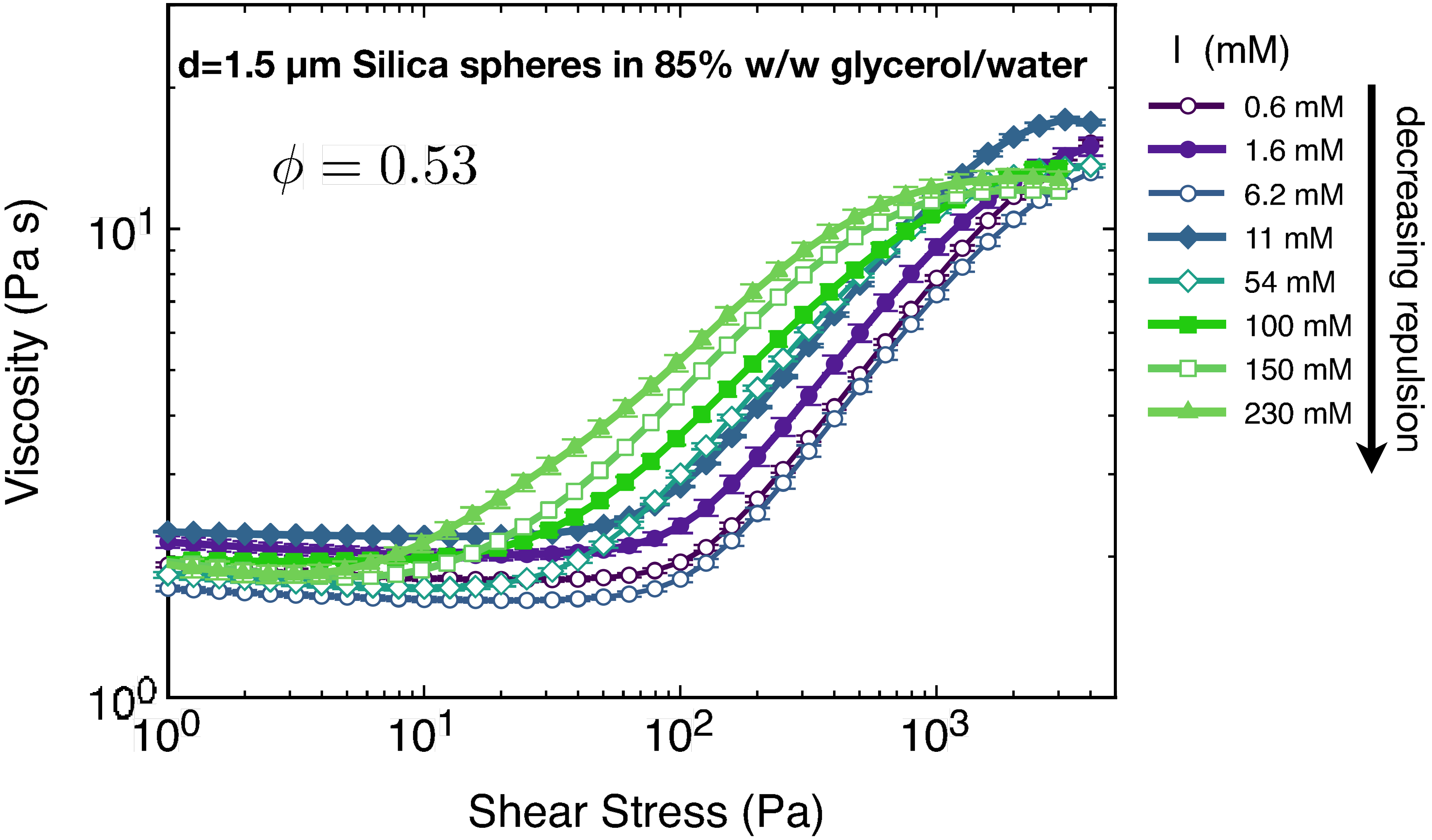
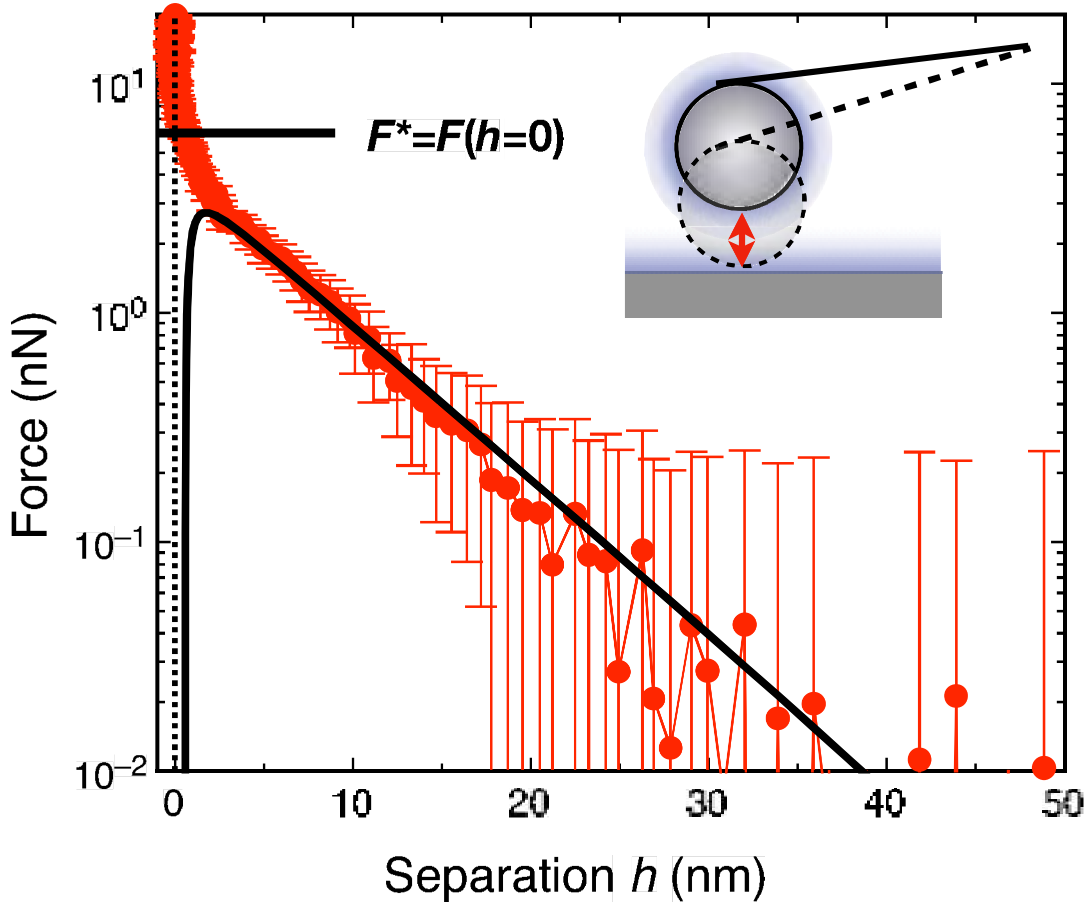
Suspension with tuneable repulsion



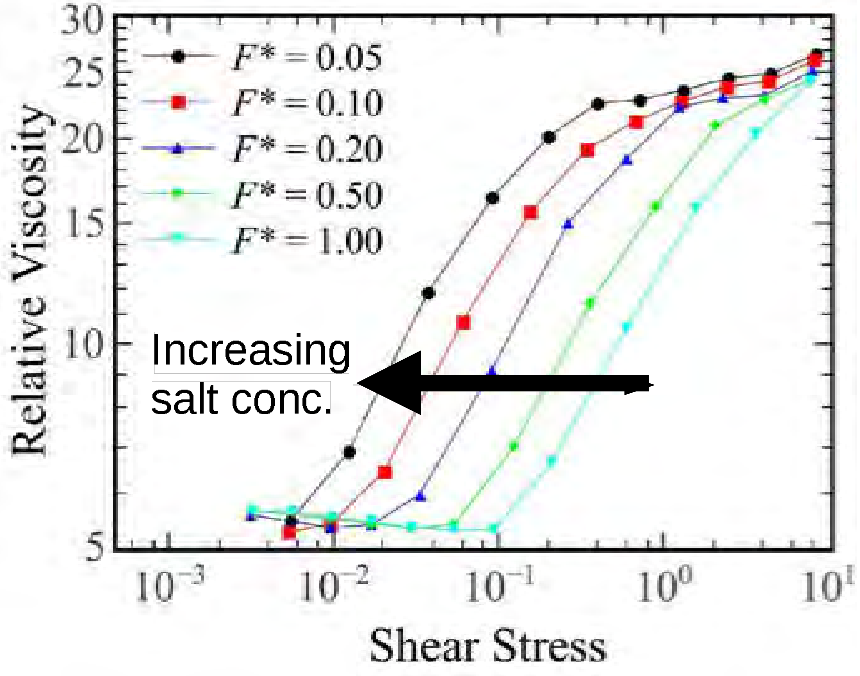
- Decreasing repulsion leads to lower critical stress

Connect rheology to interactions

AFM: force at contact

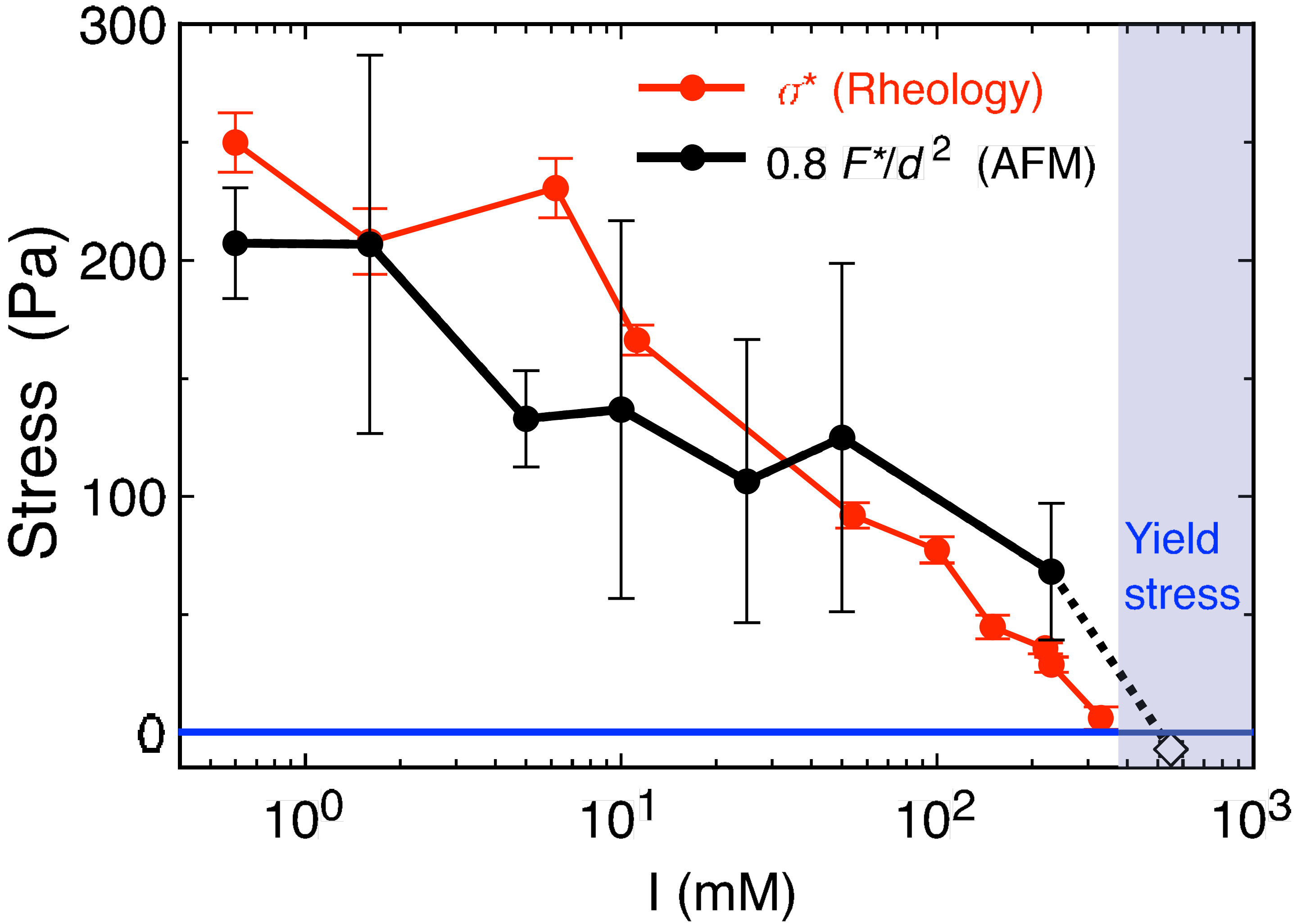


DEM simulations



- Idealized models (eg DLVO) fail at contact - need measure directly

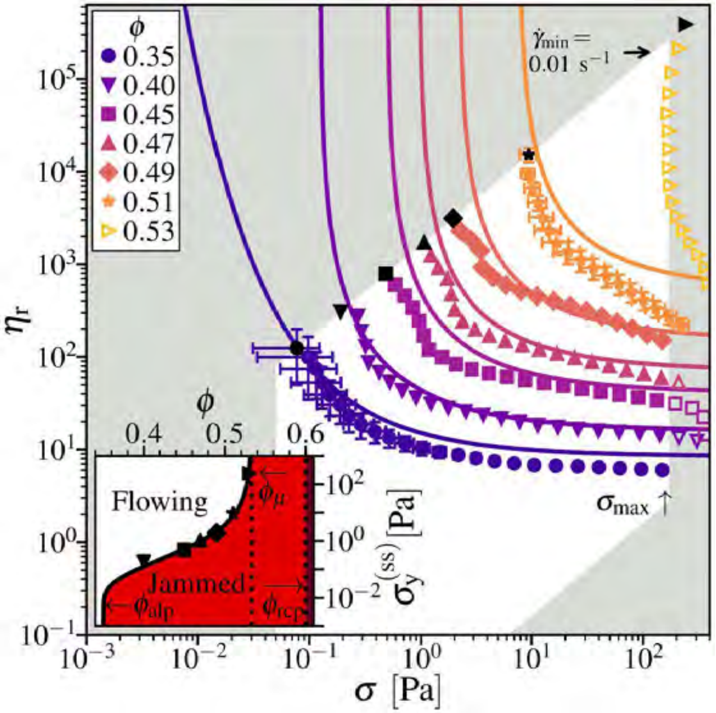
Connect rheology to interactions

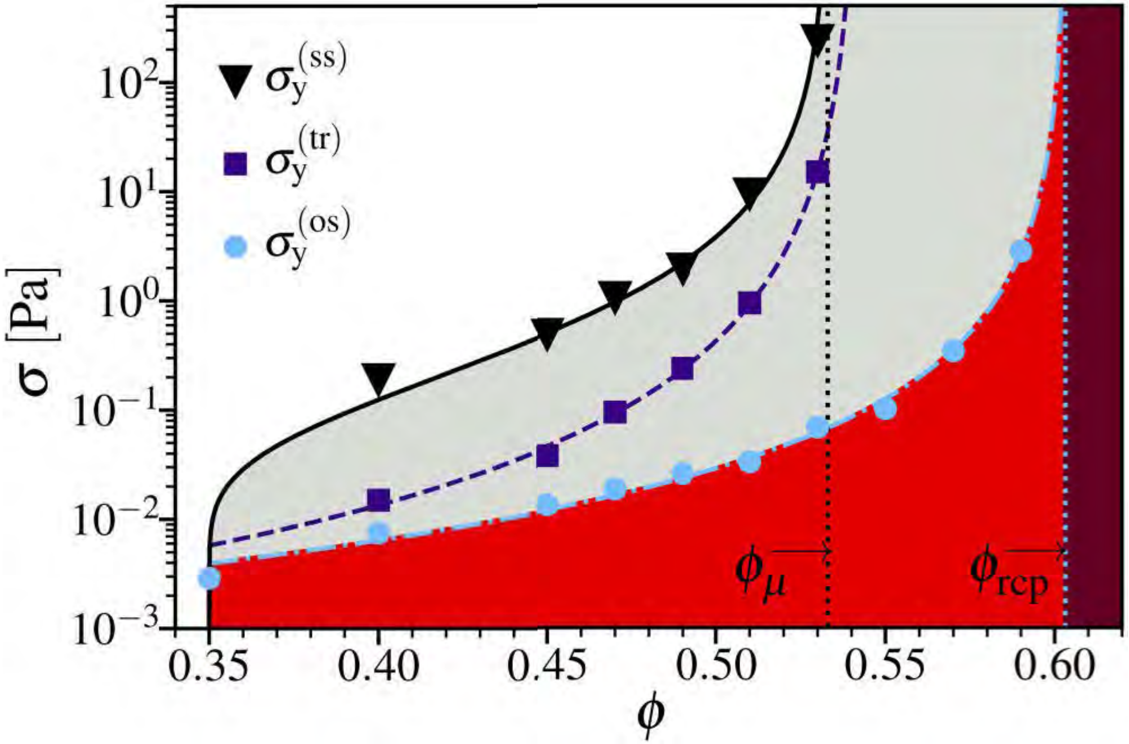


- Contact force sets critical stress for thickening

Adding adhesive particle interactions

Adhesive granular dispersions





- Dispersions with adhesive interparticle bonds due to finite-size contacts, show yielding behaviour
- The volume fraction dependence of the yield stress reveals role of friction

The role of friction in the yielding of adhesive non-Brownian suspensions

J. A. Richards,[1,a)] B. M. Guy,[1] E. Blanco,[1] M. Hermes,[1,2] G. Poy,[1,3] and W. C. K. Poon[1]

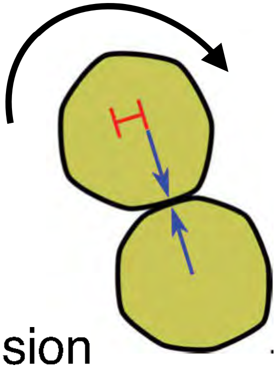
[1]*SUPA and School of Physics and Astronomy, The University of Edinburgh, Peter Guthrie Tait Road, Edinburgh, EH9 3FD, United Kingdom*
[2]*Debye Institute, Utrecht University, Princetonplein 5, 3584 CC Utrecht, The Netherlands*
[3]*Faculty of Mathematics and Physics, University of Ljubljana, Jadranska 19, 1000 Ljubljana, Slovenia*

(Received 17 October 2019; final revision received 7 January 2020; published 5 March 2020)

Abstract

Yielding behavior is well known in attractive colloidal suspensions. Adhesive non-Brownian suspensions, in which the interparticle bonds are due to finite-size contacts, also show yielding behavior. We use a combination of steady-state, oscillatory, and shear reversal rheology to probe the physical origins of yielding in the latter class of materials and find that yielding is not simply a matter of breaking adhesive bonds but involves unjamming from a shear-jammed state in which the microstructure has adapted to the direction of the applied load. Comparison with a recent constraint-based rheology model shows the importance of friction in determining the yield stress, suggesting novel ways to tune the flow of such suspensions. https://doi.org/10.1122/1.5132395

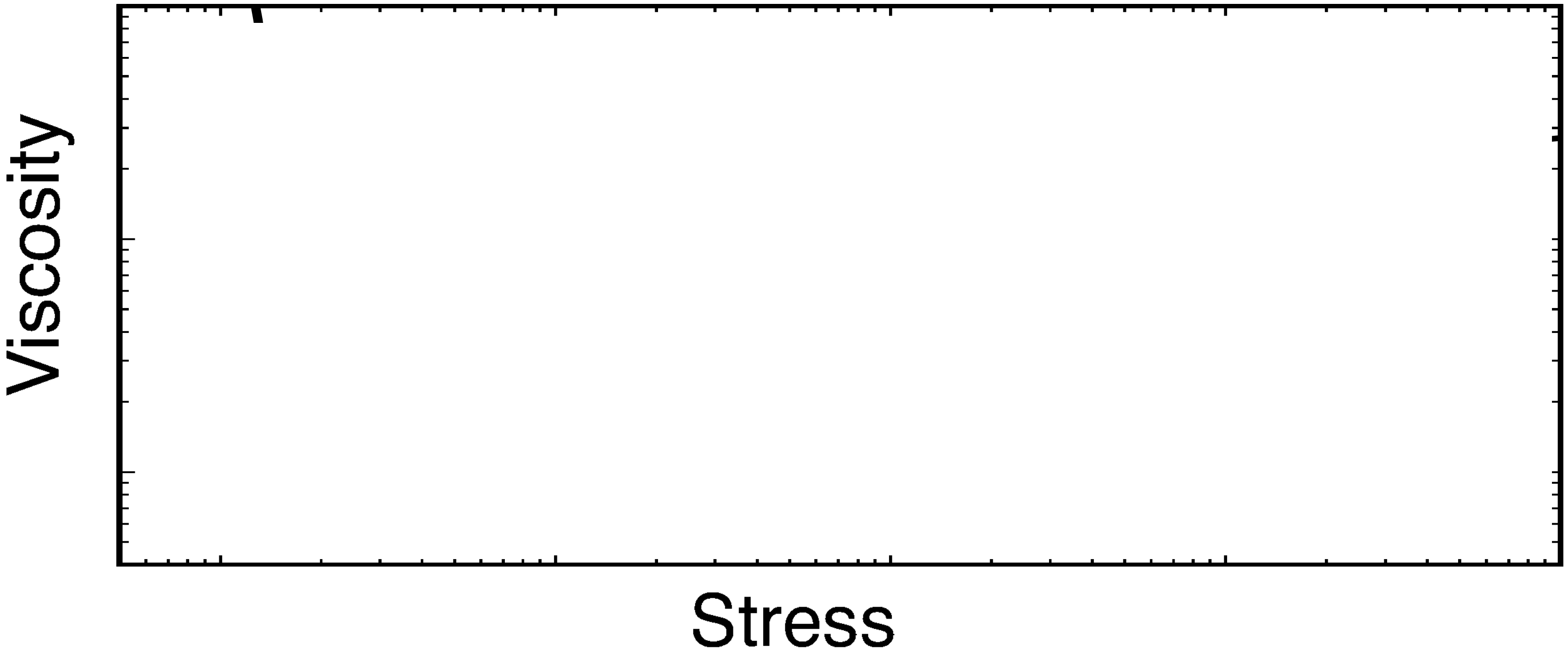
General constraints-based model



Adhesion
= constrained rolling

increasing
stress

$\gtrsim \sigma_a$



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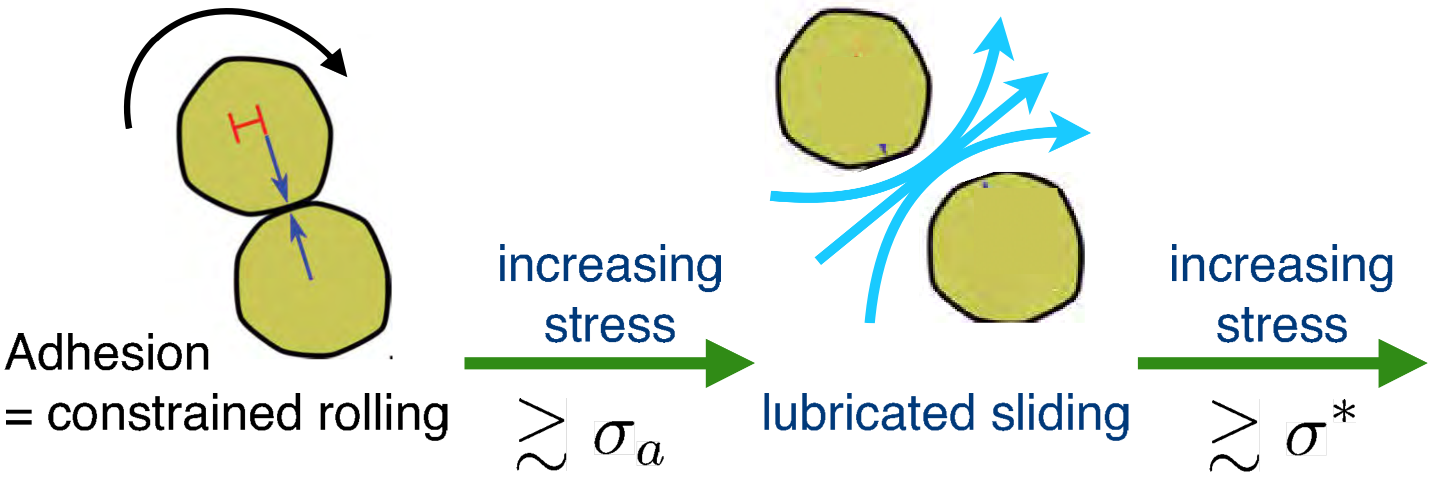


Adhesion
= constrained rolling

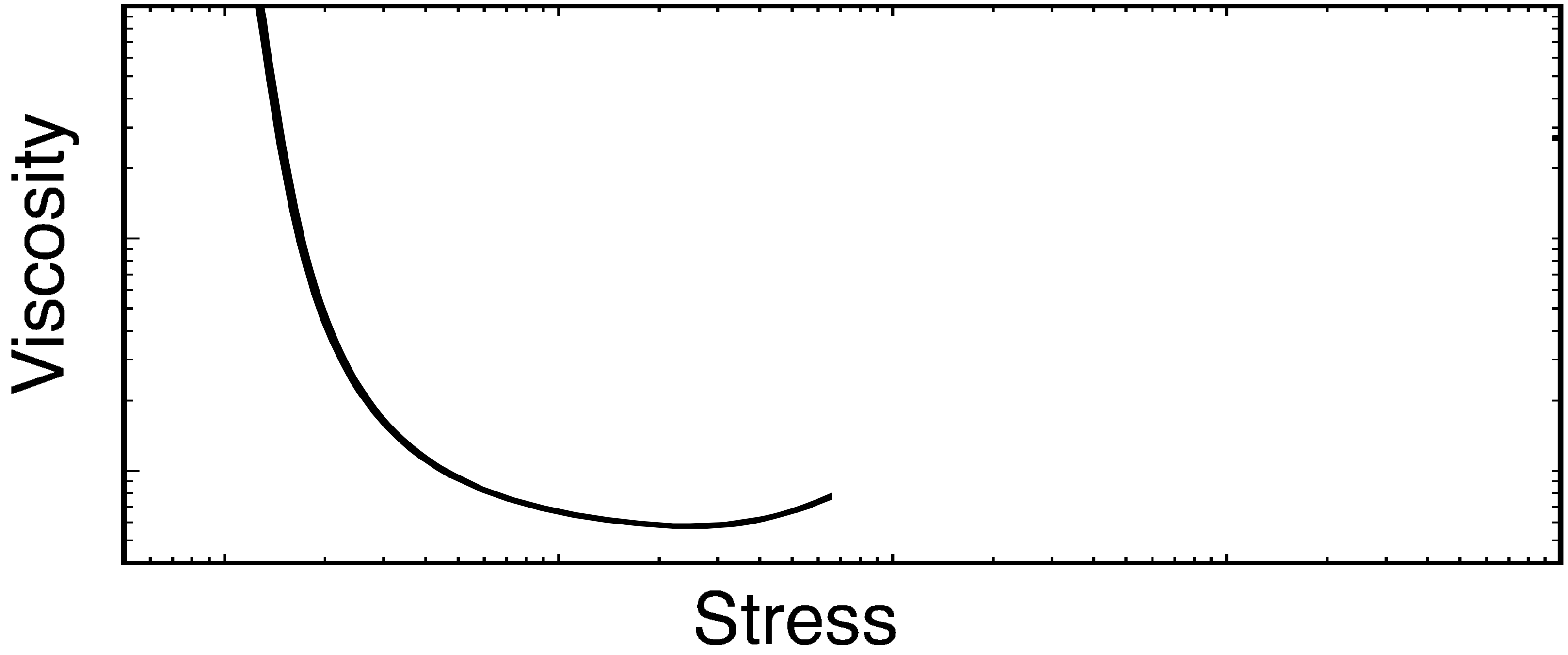
increasing stress

$\gtrsim \sigma_a$

lubricated sliding

increasing stress

$\gtrsim \sigma^*$



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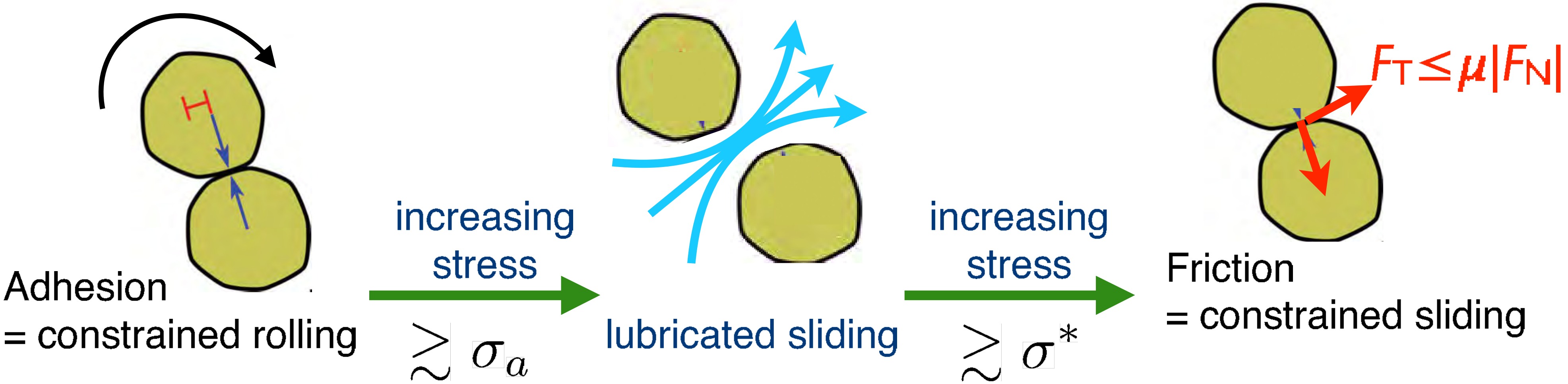


Adhesion
= constrained rolling

increasing stress

$\gtrsim \sigma_a$

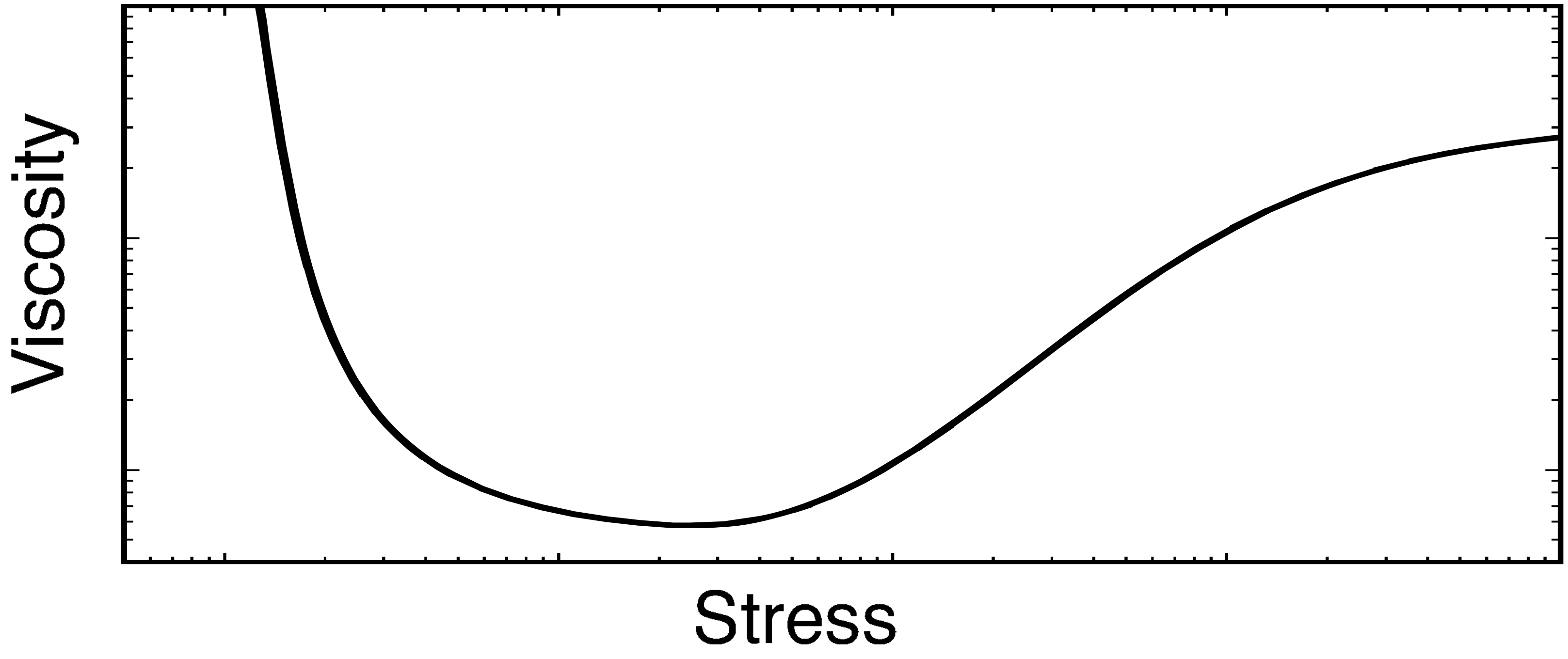
lubricated sliding

increasing stress

$\gtrsim \sigma^*$

Friction
= constrained sliding

$F_T \leq \mu |F_N|$



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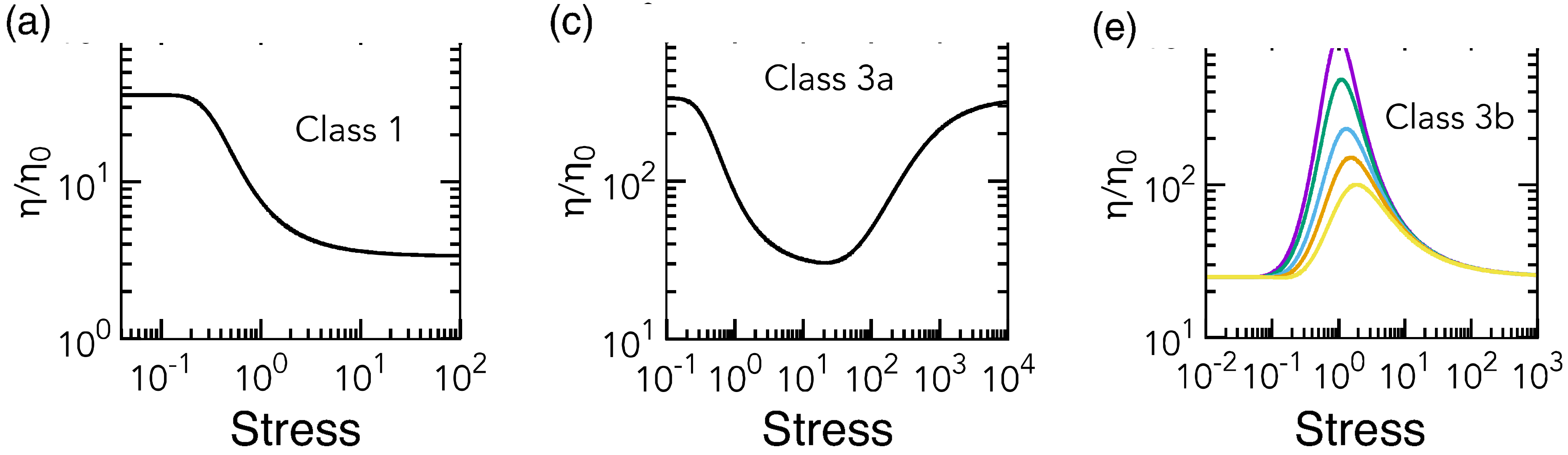


- Generic description of granular dispersion rheology in terms of stress-dependent constraints
- Can capture wide array of flow behavior

PHYSICAL REVIEW LETTERS 121, 128001 (2018)

Constraint-Based Approach to Granular Dispersion Rheology

B. M. Guy,[*] J. A. Richards,[†] D. J. M. Hodgson, E. Blanco, and W. C. K. Poon
SUPA, School of Physics and Astronomy, The University of Edinburgh,
King's Buildings, Peter Guthrie Tait Road, Edinburgh EH9 3FD, United Kingdom

(Received 10 May 2018; published 17 September 2018)

We present a phenomenological model for granular suspension rheology in which particle interactions enter as constraints to relative particle motion. By considering constraints that are *formed* and *released* by stress respectively, we derive a range of experimental flow curves in a single treatment and predict singularities in viscosity and yield stress consistent with literature data. Fundamentally, we offer a generic description of suspension flow that is independent of bespoke microphysics.

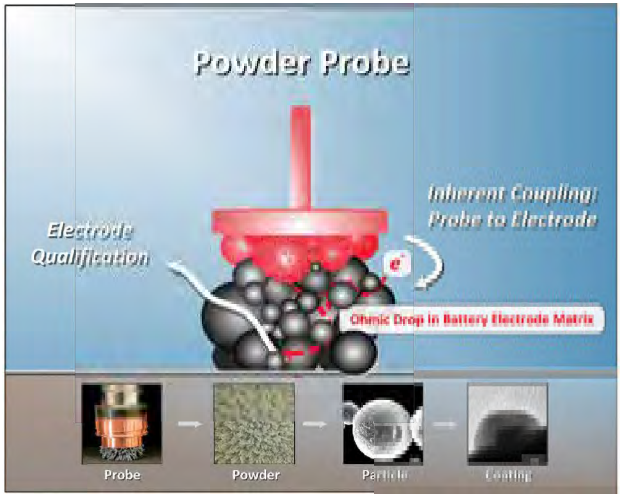
DOI: 10.1103/PhysRevLett.121.128001

Granular particles—colloidal gel composites

Granular—Gel composites

Yield stress - from inter-particle attraction

Suspended large non-sticky particles



Battery electrode

≈ active materials in conducting agents

Concrete

≈ sands in cement paste

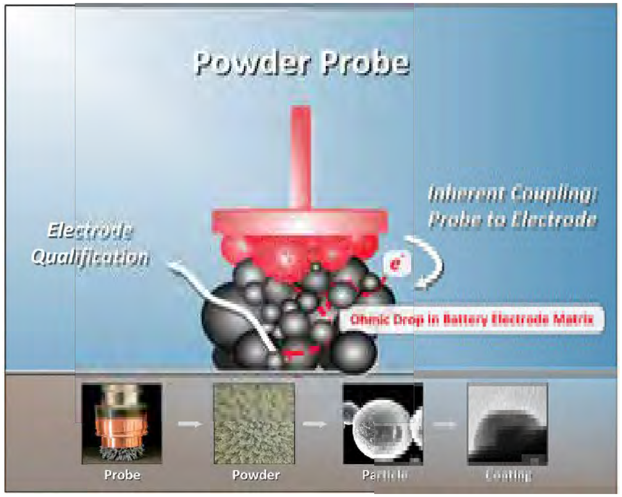
Toothpaste

≈ abrasives in gel phase

Granular—Gel composites

Yield stress - from inter-particle attraction

Suspended large non-sticky particles



Battery electrode

≈ active materials in conducting agents

Concrete

≈ sands in cement paste

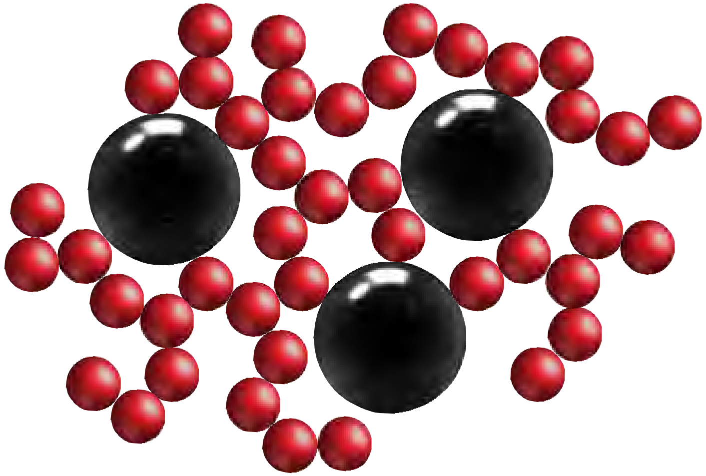
Toothpaste

≈ abrasives in gel phase

Model System

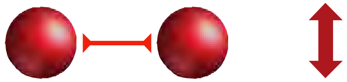
Colloidal gel background

Embedded large particles



Ethanol:water:glycerol = 0.5:1:10

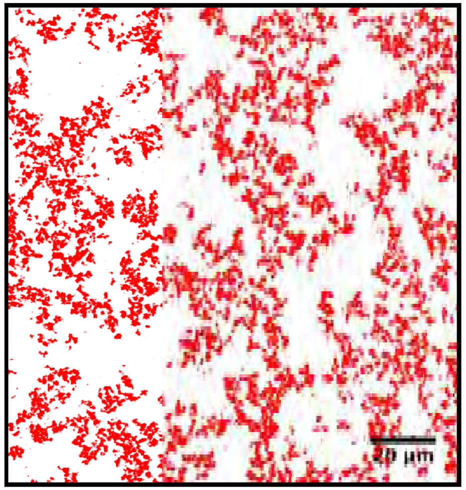
Colloidal gel background



d~500 nm

Hydrophobic silica

Short-range attractive



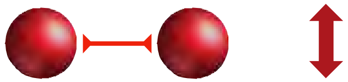
Colloidal gel

$\phi = 10\,\%$

$\sigma_y = 1.8\,Pa$

Ethanol:water:glycerol = 0.5:1:10

Colloidal gel background



d~500 nm

Hydrophobic silica

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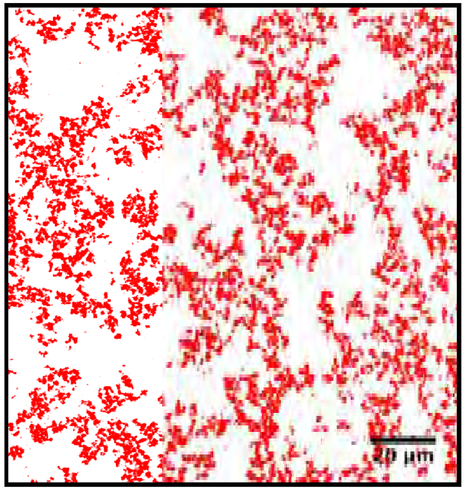
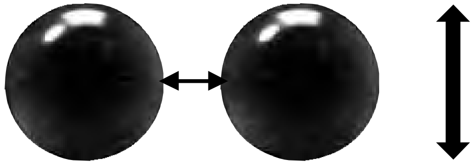


Colloidal gel

$\phi = 10\,\%$

$\sigma_y = 1.8\,Pa$

Embedded large particles



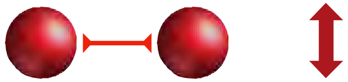
d~4 µm

Silica

Surface charge repulsion

Ethanol:water:glycerol = 0.5:1:10

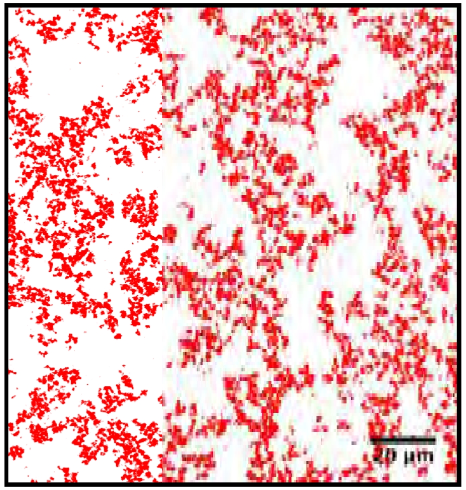
Colloidal gel background



d~500 nm

Hydrophobic silica

Short-range attractive



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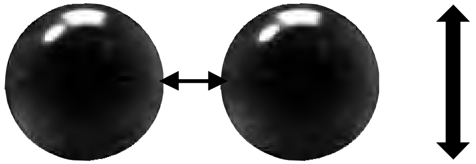

d~4 μm

Silica

Surface charge repulsion

Non-sticky

Ethanol:water:glycerol = 0.5:1:10

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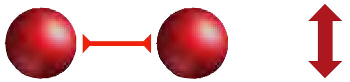
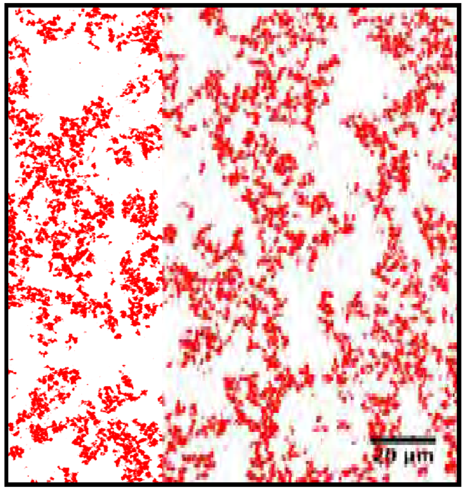


Hydrophobic silica
Short-range attractive

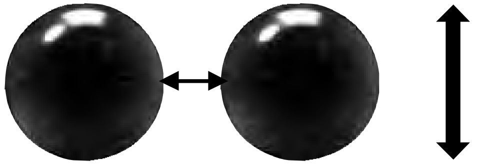


Colloidal gel
$\phi = 10\,\%$
$\sigma_y = 1.8\,Pa$

ϕ_S Smaller attractive particles

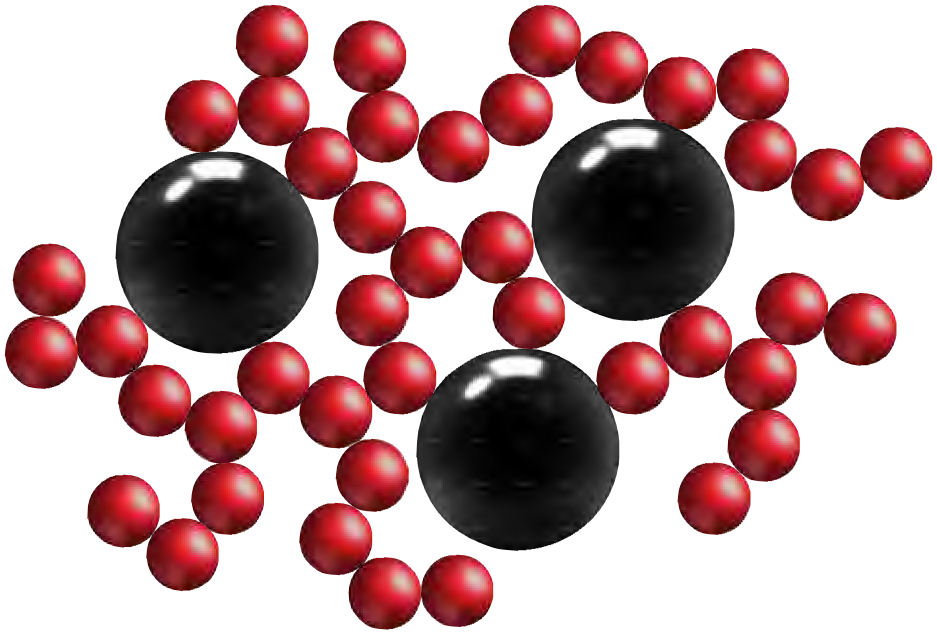
ϕ_L Large repulsive particles

Embedded large particles



Silica
Surface charge repulsion

Non-sticky



History matters!

$\phi_S = 13.5\ \%$, $\phi_L = 35\ \%$

History matters!

$$\phi_S = 13.5\ \%\ ,\ \phi_L = 35\ \%$$

History matters!

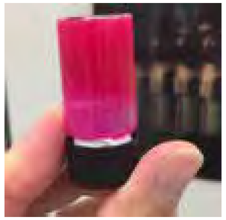
Liquid

After rolling mixing (gentle)

Solid

After vortex mixing (fast)

$\phi_S = 13.5\ \%\ ,\ \phi_L = 35\ \%$



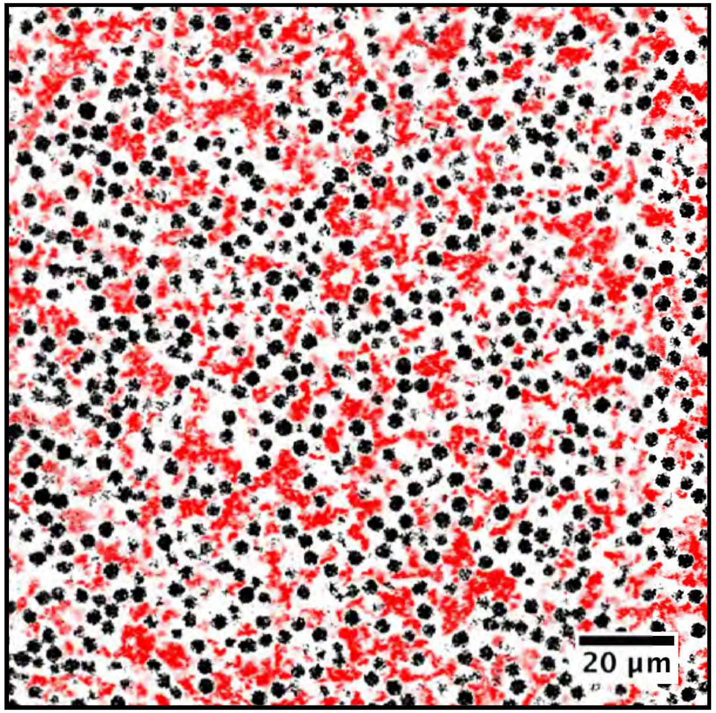
Two States Under Shear

Homogeneous State

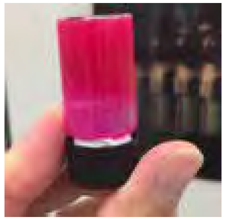
Solid-like

Gel network

Generated by high shear



After high shear ($\dot{\gamma} = 100\ s^{-1}$)



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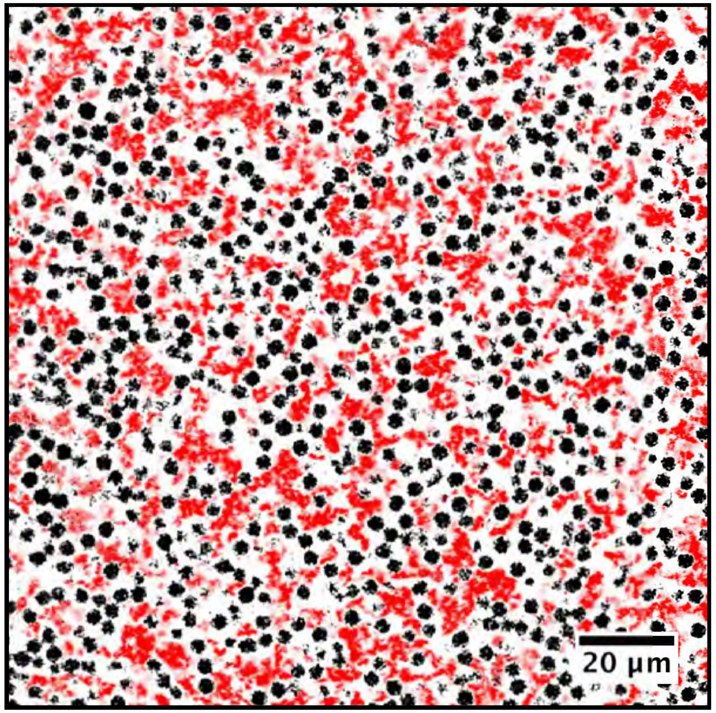


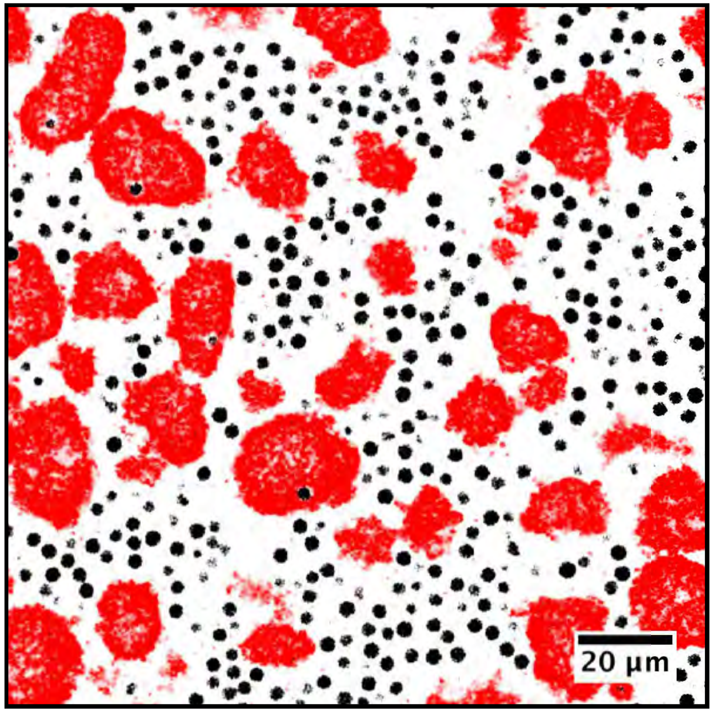
After high shear ($\dot{\gamma} = 100\ s^{-1}$)

Phase-separated State

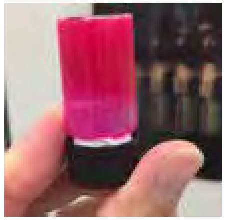
Liquid-like

Disjoint globules

Generated by medium shear



After medium shear ($\dot{\gamma} = 5\ s^{-1}$)



Two States Under Shear

Homogeneous State

Solid-like

Gel network

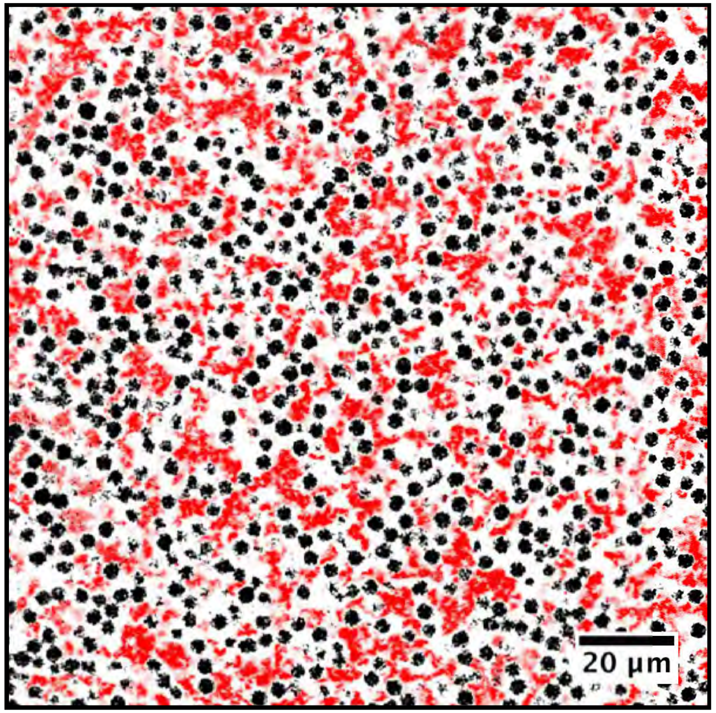
Generated by high shear

Phase-separated State

Liquid-like

Disjoint globules

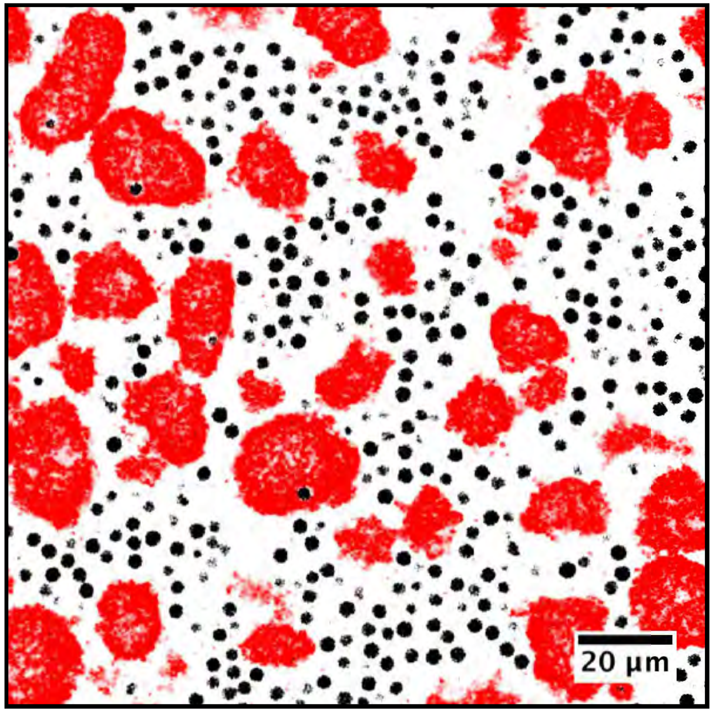
Generated by medium shear



After high shear ($\dot{\gamma} = 100\ s^{-1}$)

Rejuvenation ←

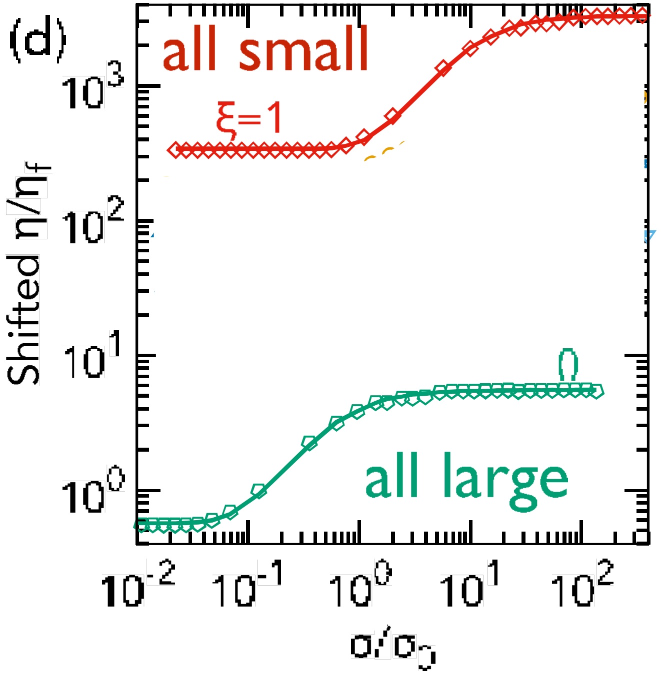
→ Collapse



After medium shear ($\dot{\gamma} = 5\ s^{-1}$)

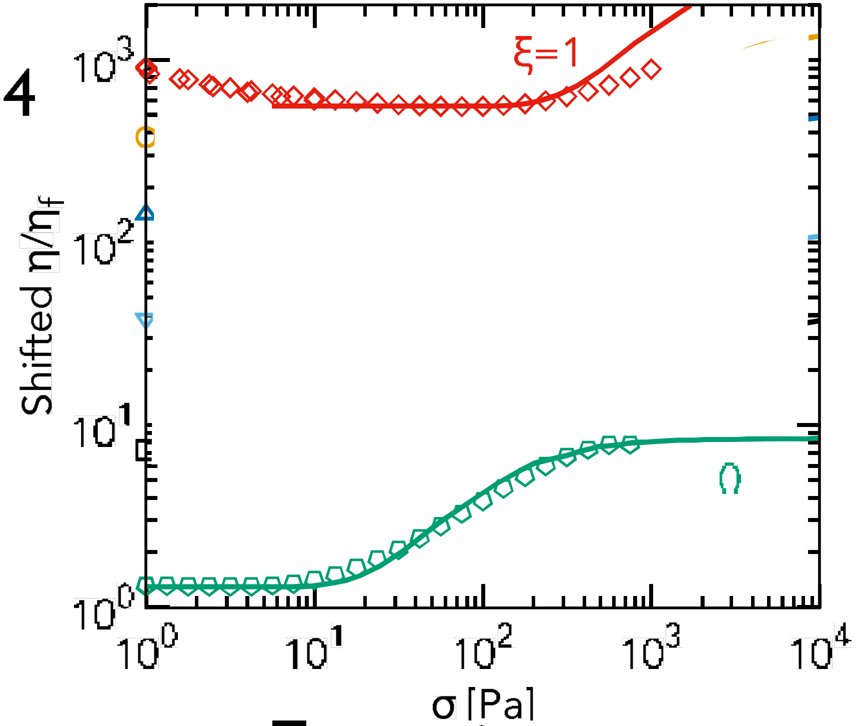
Shear thickening in bidispersed systems: mixture of different sizes and friction

Thickening with different sizes



Simulation

Size ratio 1:4

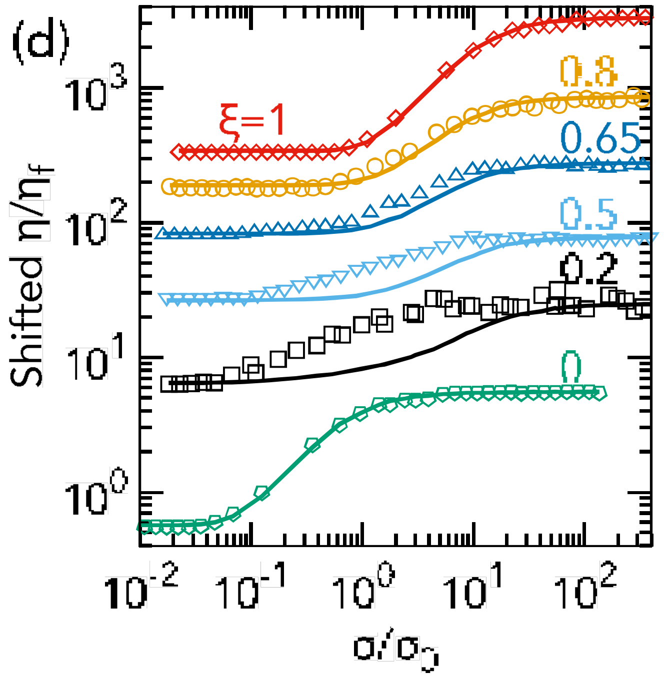


Experiments

Solid lines: Wyart - Cates model

$$f(\sigma) \sim e^{-\sigma^*/\sigma} \qquad \sigma^* \propto d^{-2}$$

Thickening in binary mixtures



Simulation

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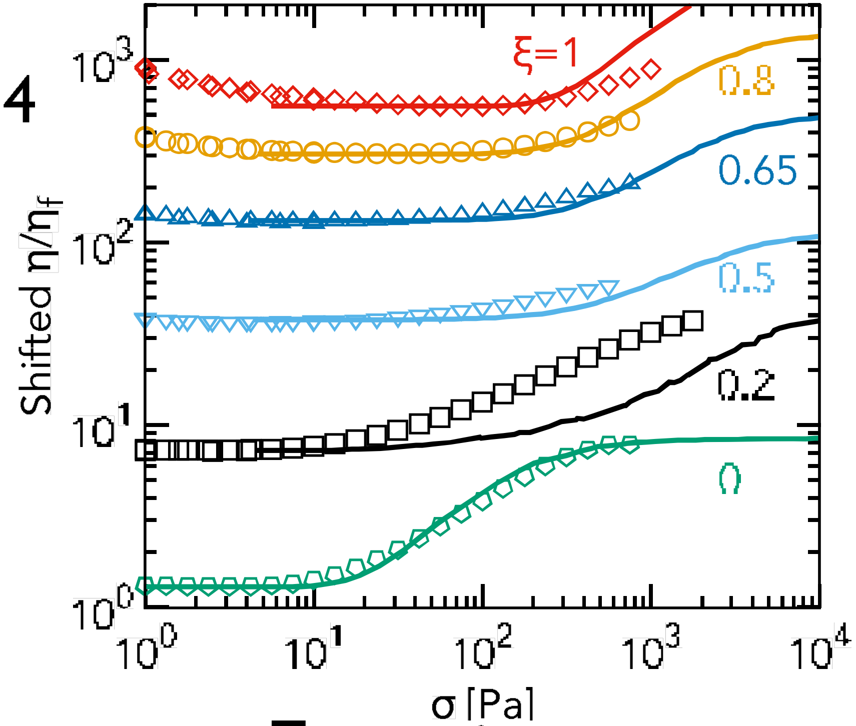
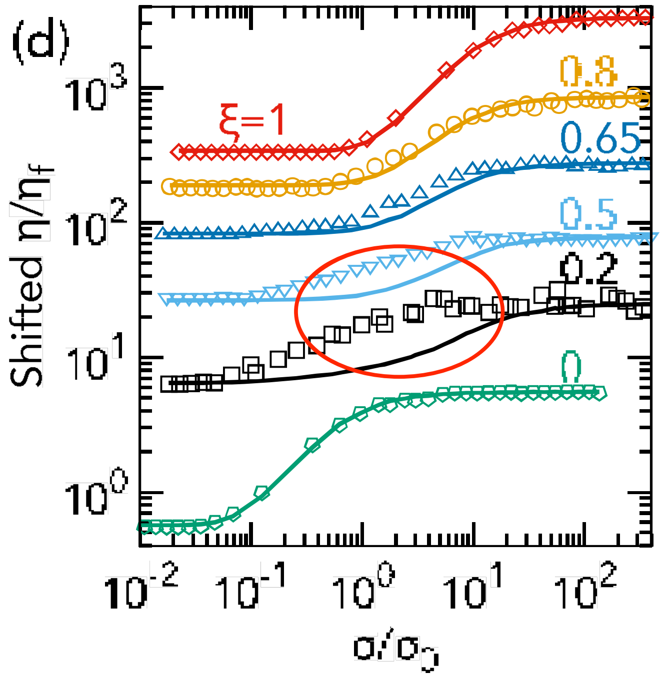


Experiments

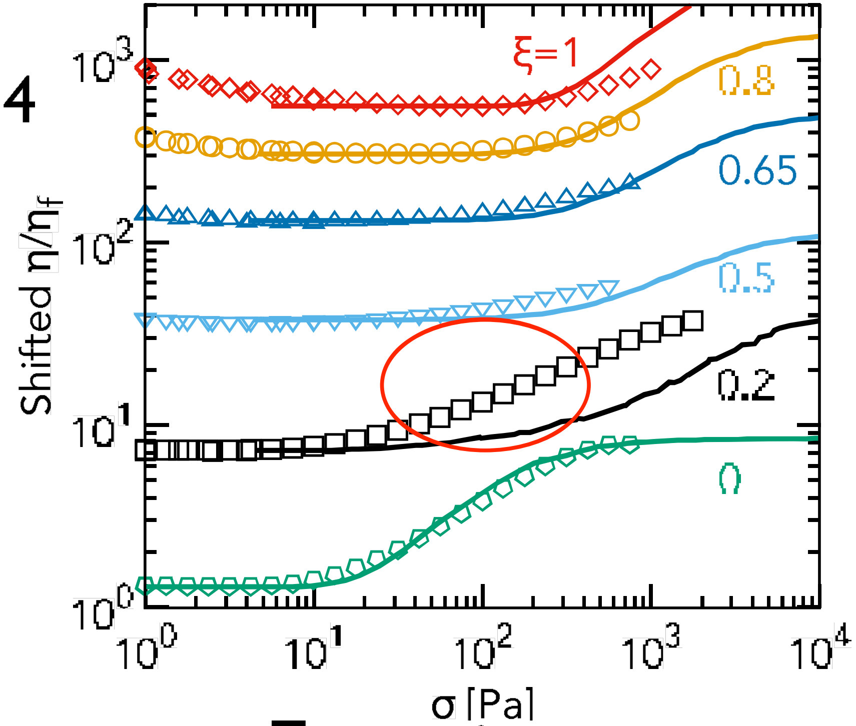
- The WC model cannot capture the transition in mixtures
- Why? Fraction of frictional contacts does not distinguish contact type, but large particles dominate onset.

Thickening in binary mixtures



Simulation

Size ratio 1:4



Experiments

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Soft Matter

ROYAL SOCIETY OF CHEMISTRY

PAPER

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Cite this: *Soft Matter*, 2020, **16**, 229

Received 7th January 2019,
Accepted 15th November 2019

DOI: 10.1039/c9sm00041k

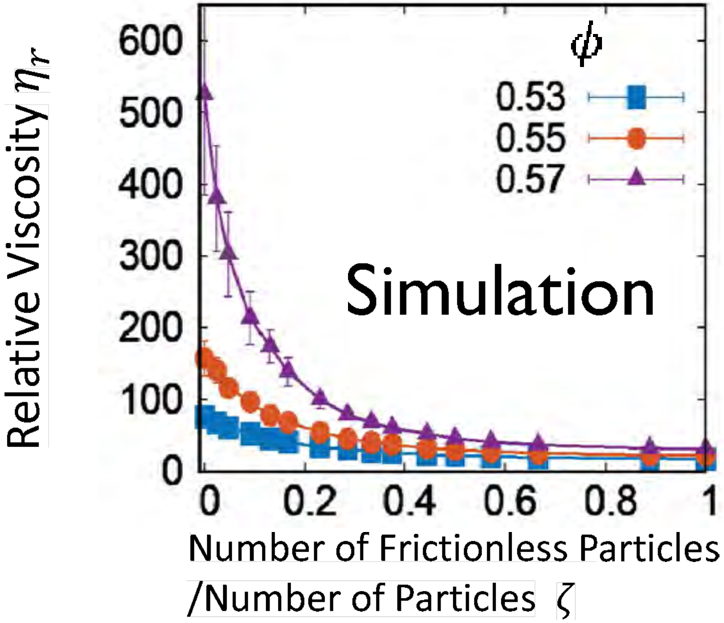
rsc.li/soft-matter-journal

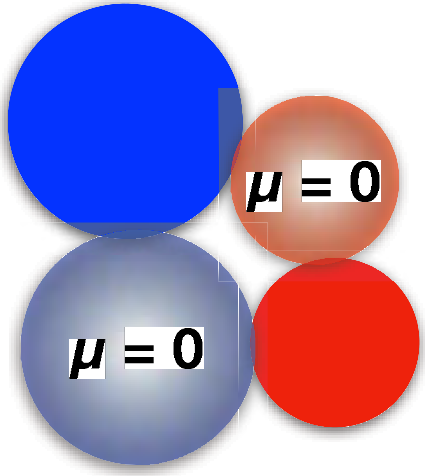
Testing the Wyart–Cates model for non-Brownian shear thickening using bidisperse suspensions†

Ben M. Guy,*[a] Christopher Ness,*[bc] Michiel Hermes,[ad] Laura J. Sawiak,[a] Jin Sun[b] and Wilson C. K. Poon[a]

There is a growing consensus that shear thickening of concentrated dispersions is driven by the formation of stress-induced frictional contacts. The Wyart–Cates (WC) model of this phenomenon, in which the microphysics of the contacts enters solely *via* the fraction f of contacts that are frictional, can successfully fit flow curves for suspensions of weakly polydisperse spheres. However, its validity for "real-life", polydisperse suspensions has yet to be seriously tested. By performing systematic simulations on bidisperse mixtures of spheres, we show that the WC model applies only in the monodisperse limit and fails when substantial bidispersity is introduced. We trace the failure of the model to its inability to distinguish large–large, large–small and small–small frictional contacts. By fitting our data using a polydisperse analogue of f that depends separately on the fraction of each of these contact types, we show that the WC picture of shear thickening is incomplete. Systematic experiments on model shear-thickening suspensions corroborate our findings, but highlight important challenges in rigorously testing the WC model with real systems. Our results prompt new questions about the microphysics of thickening for both monodisperse and polydisperse systems.

Mixtures of frictional and frictionless particles





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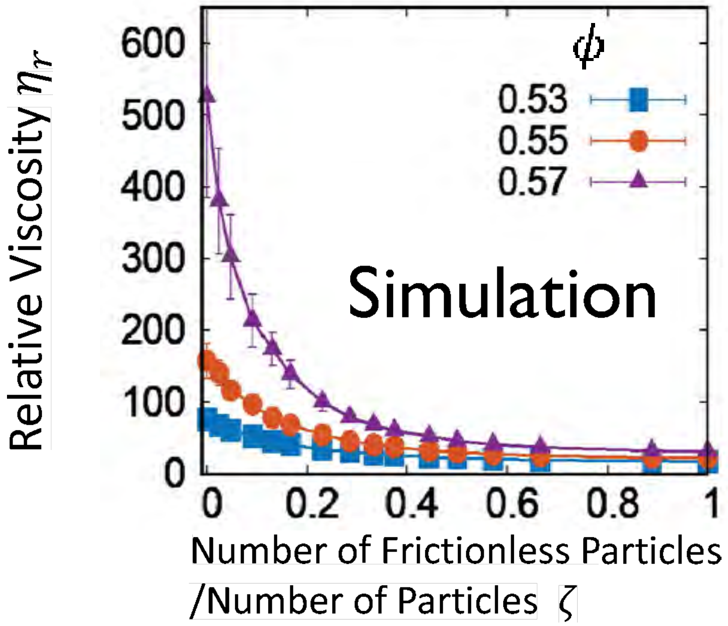

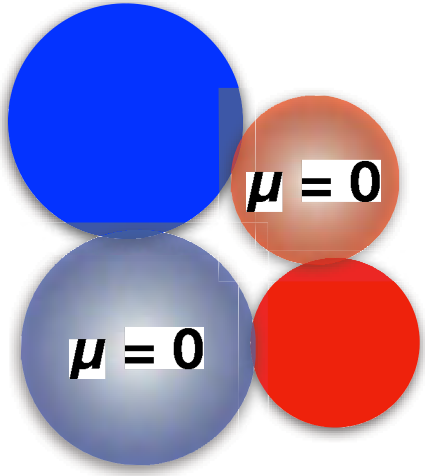

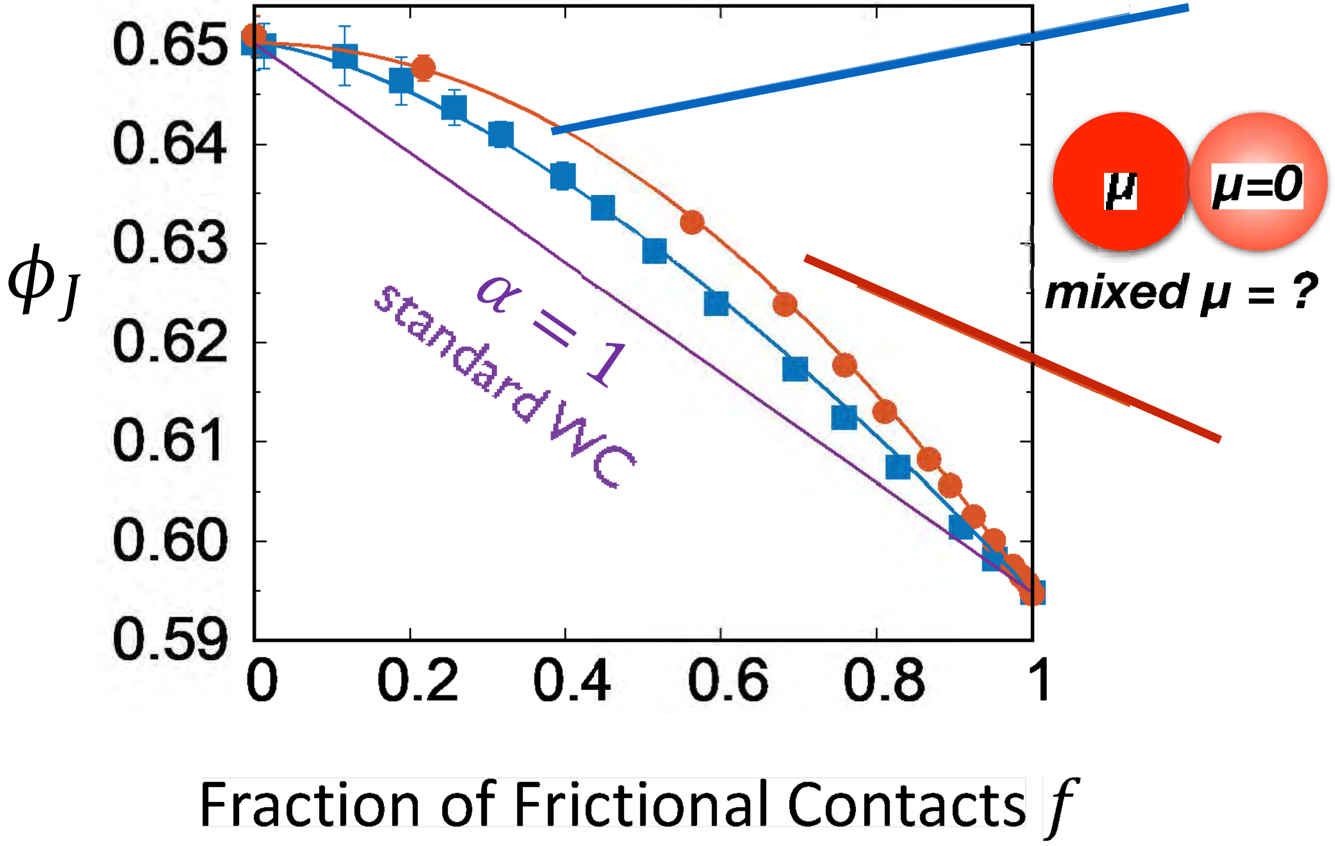

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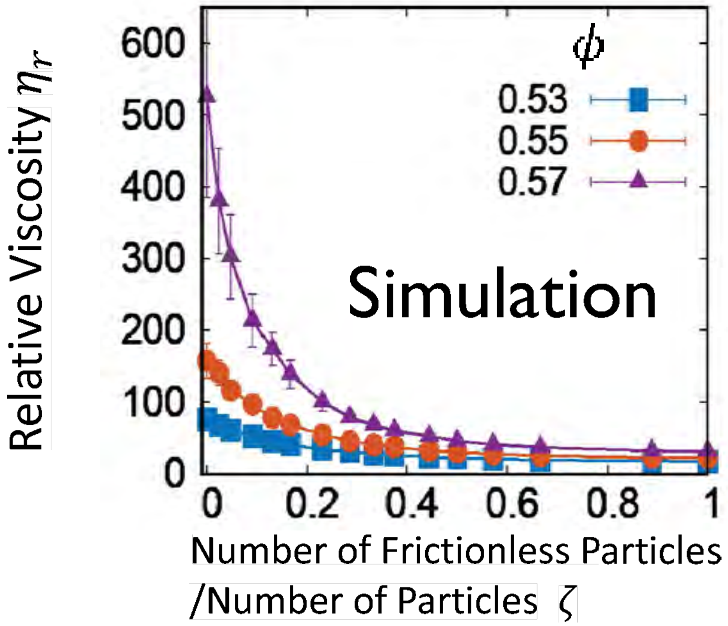

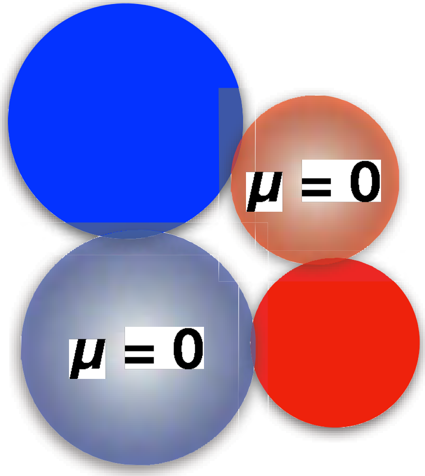

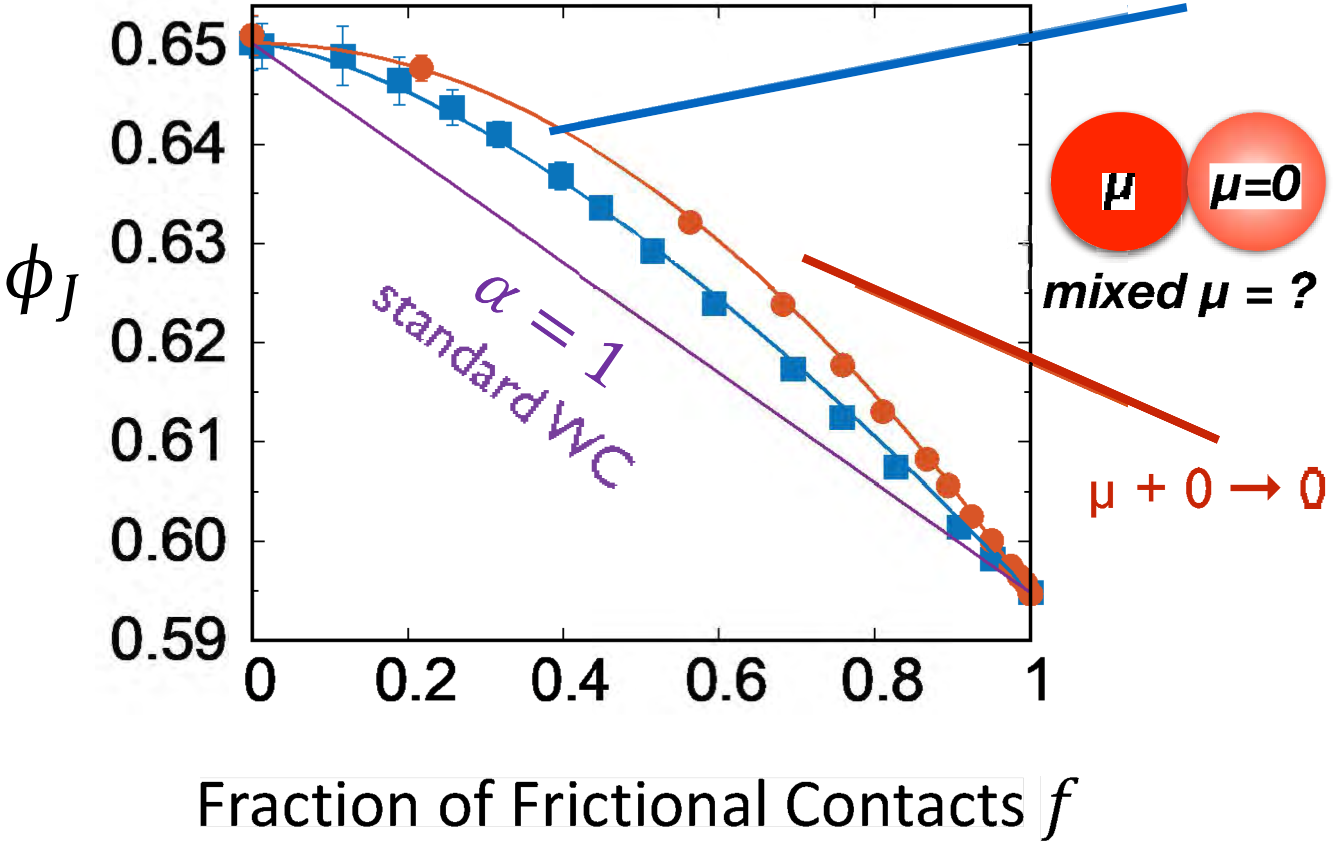

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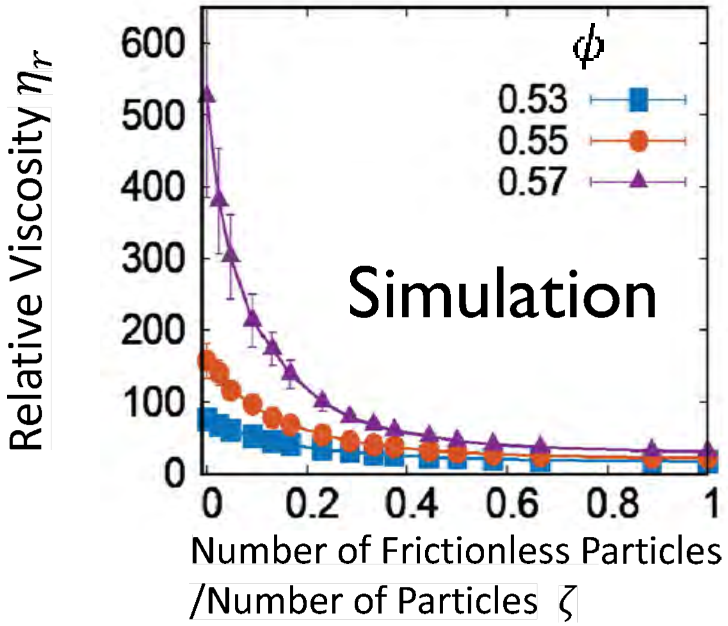

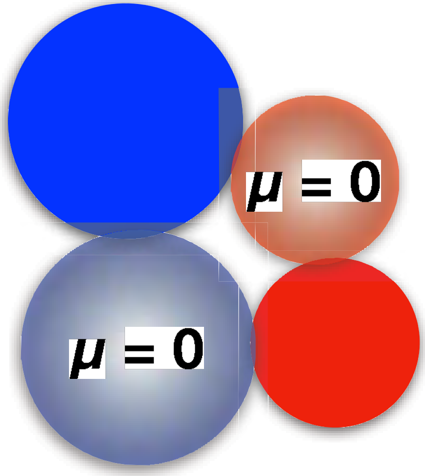

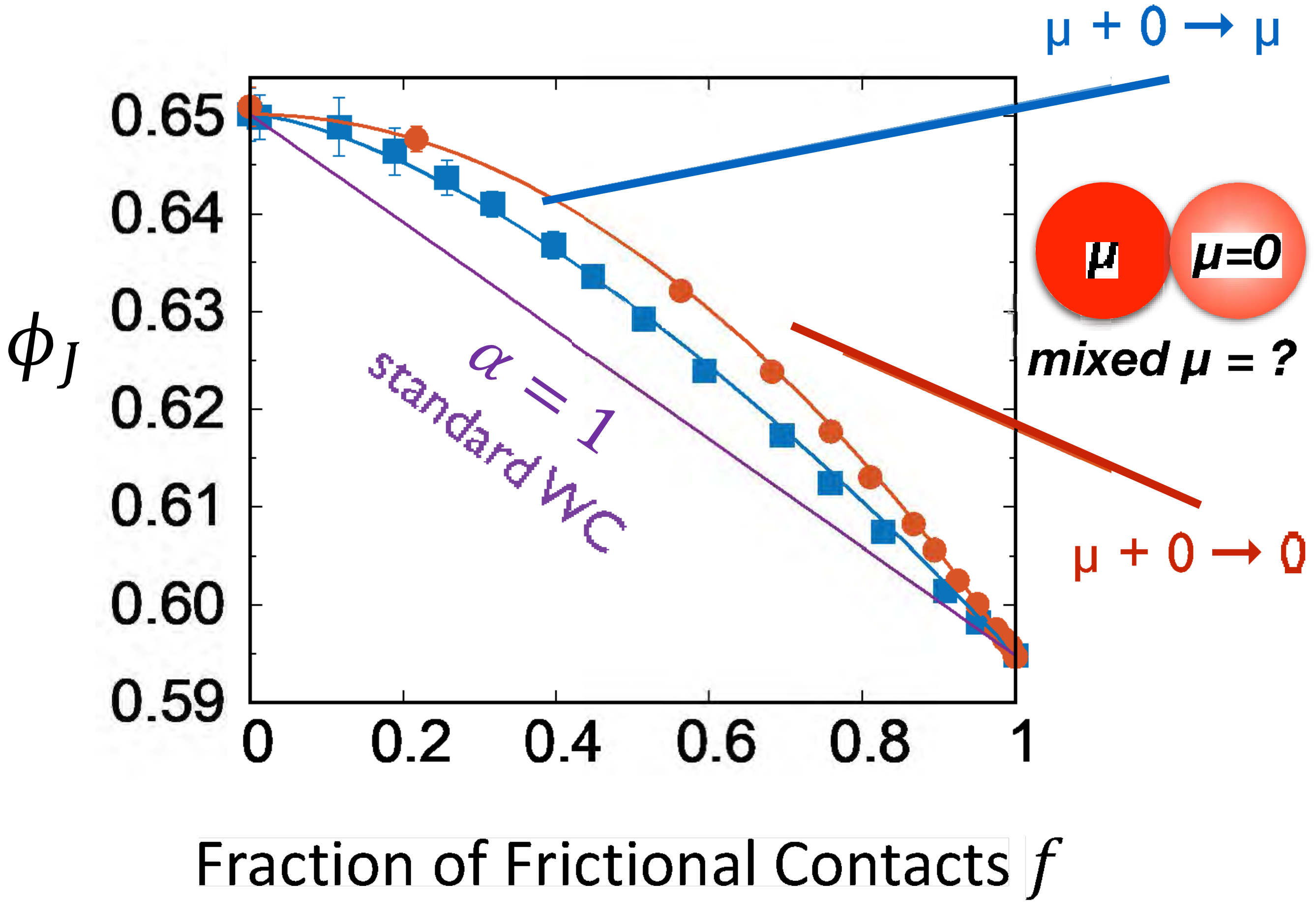

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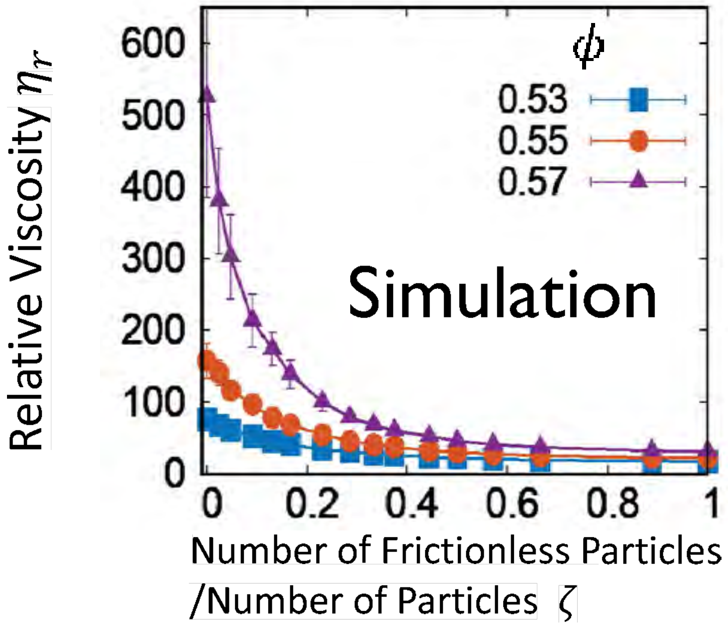
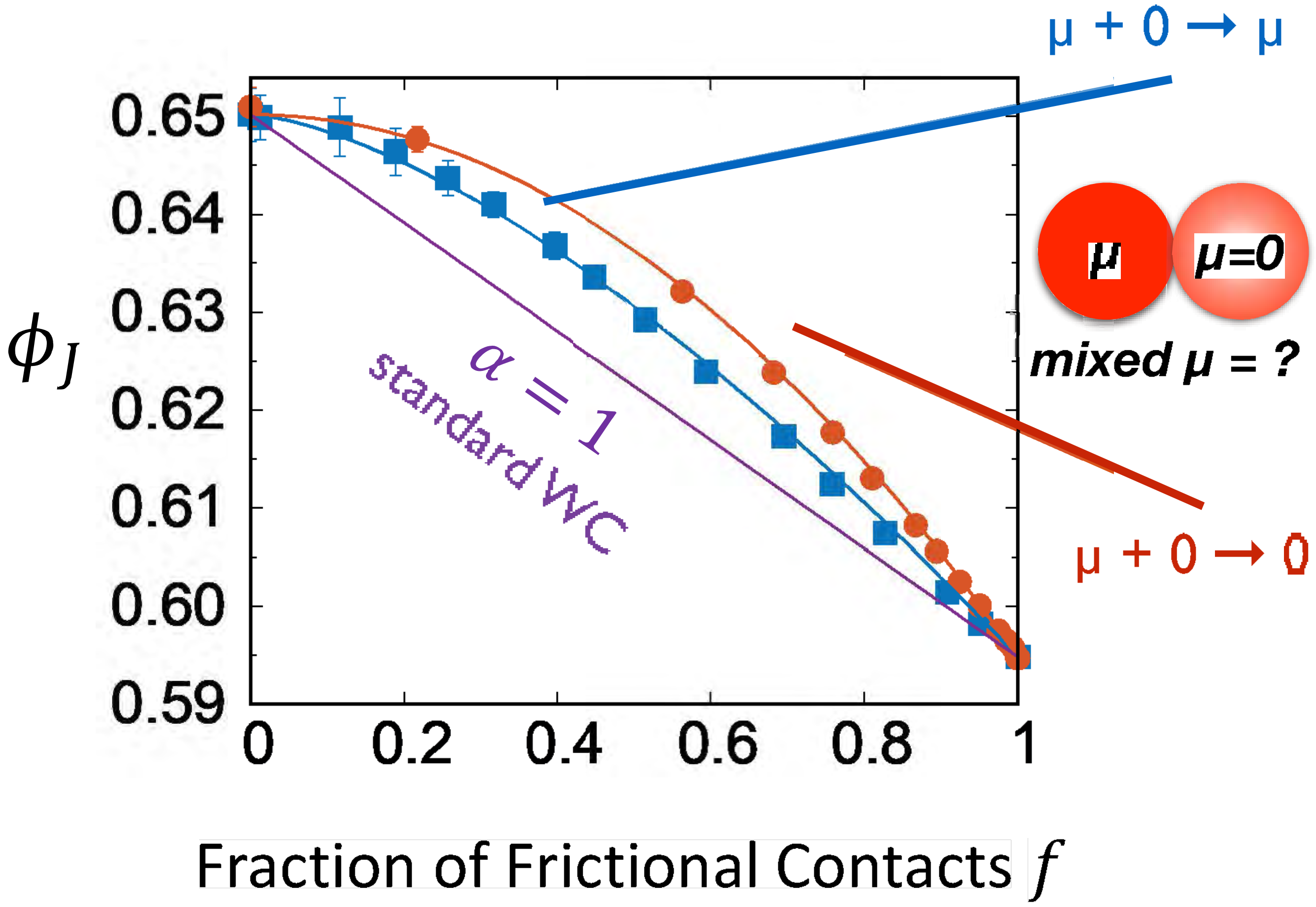
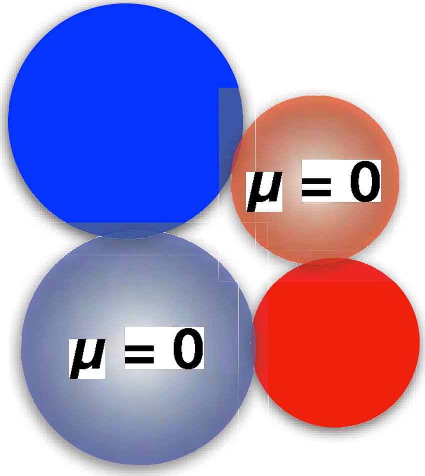


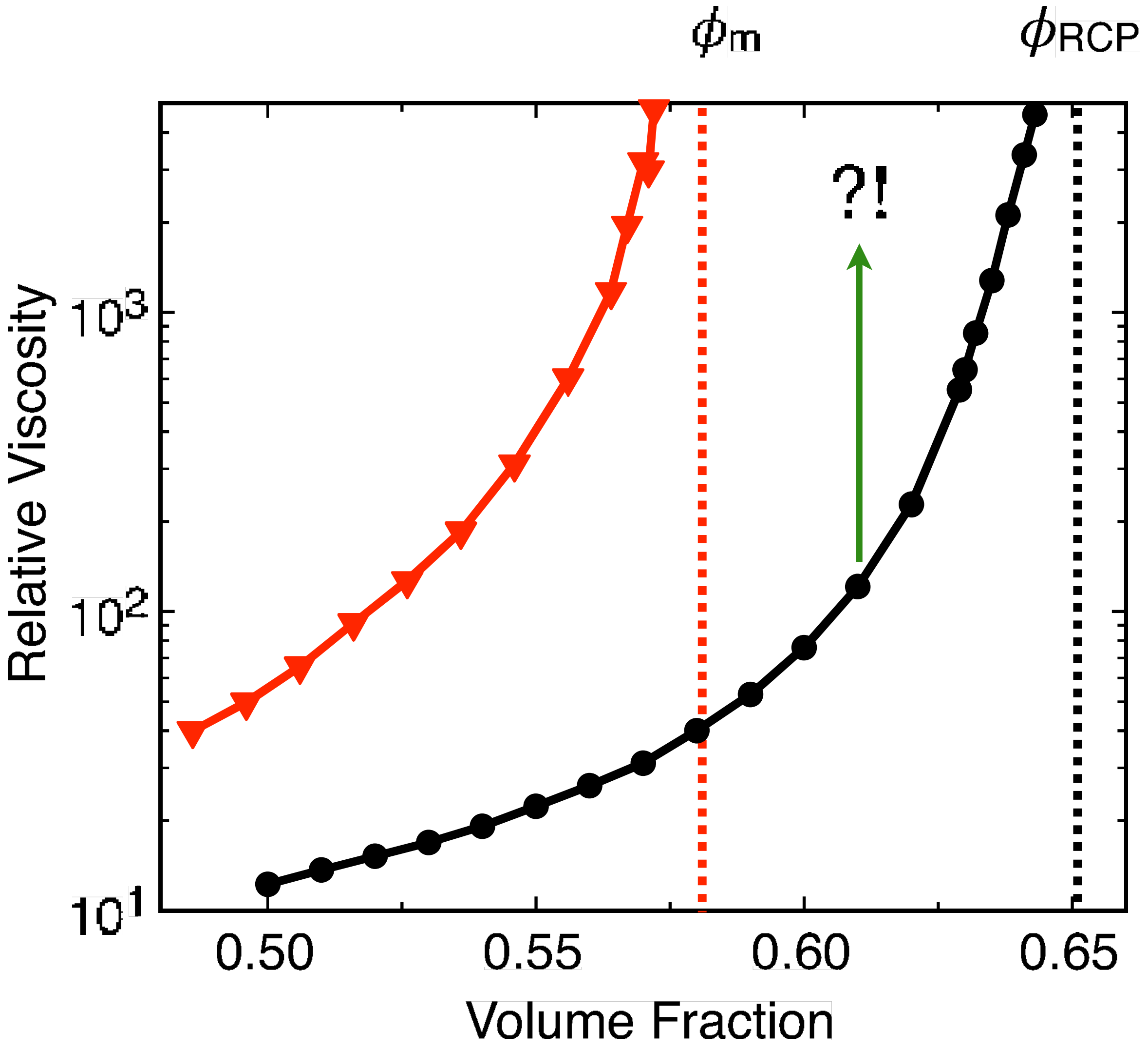




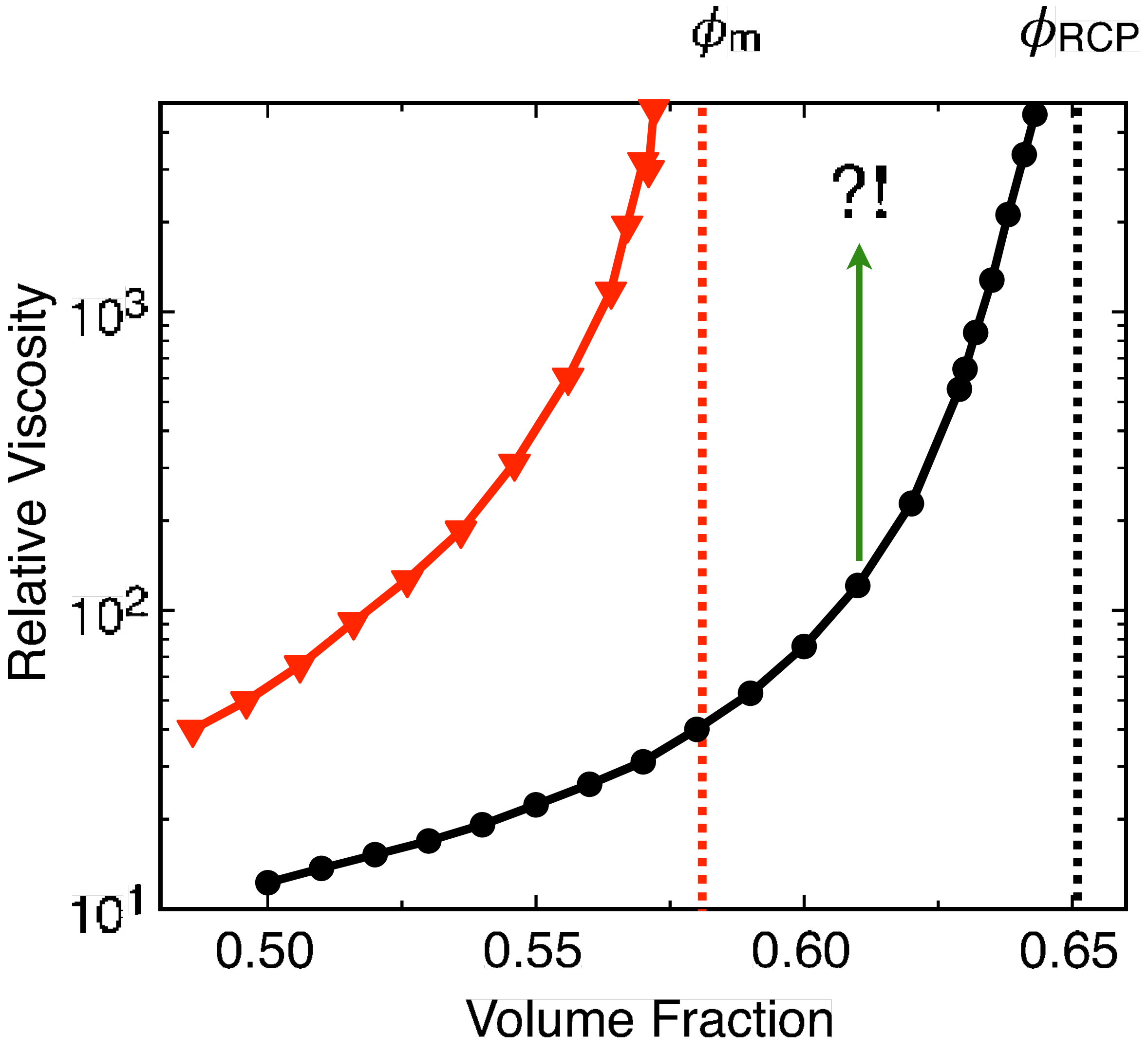
Relation between ϕ_J and f depends on contact details - again standard WC breaks down

Oscillations and fluctuations in rheometric flows

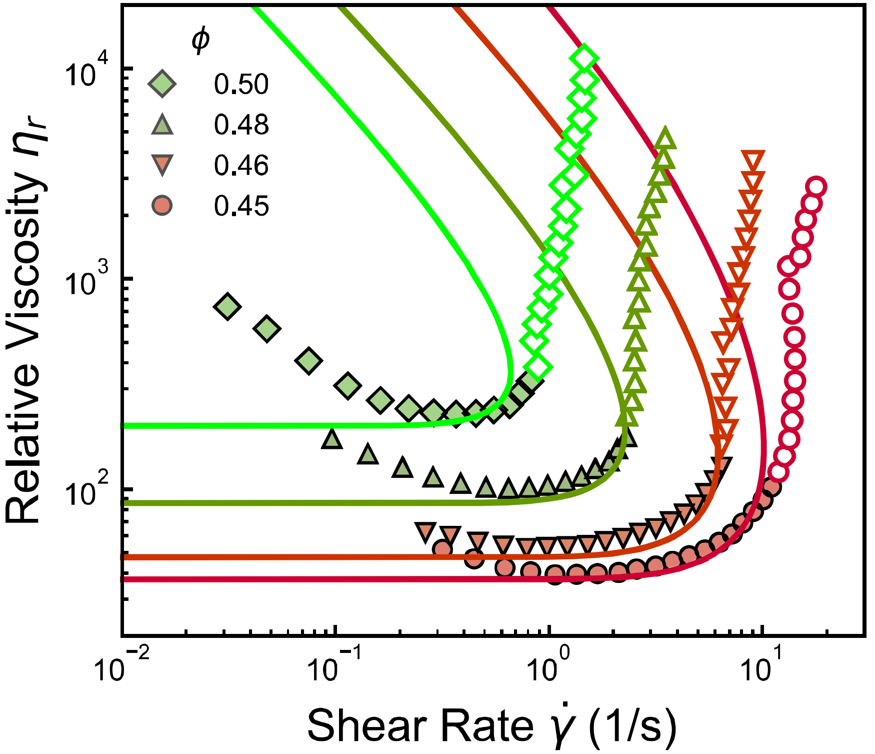
Thickening above ϕ_m - recipe for instability



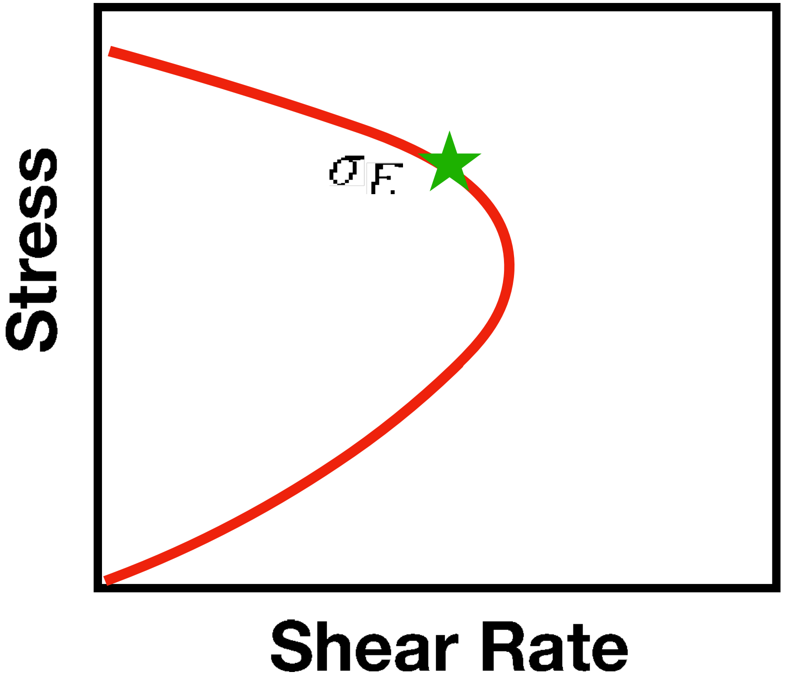
Thickening above ϕ_m - recipe for instability



Wyart and Cates model:
backwards-bending flow curves

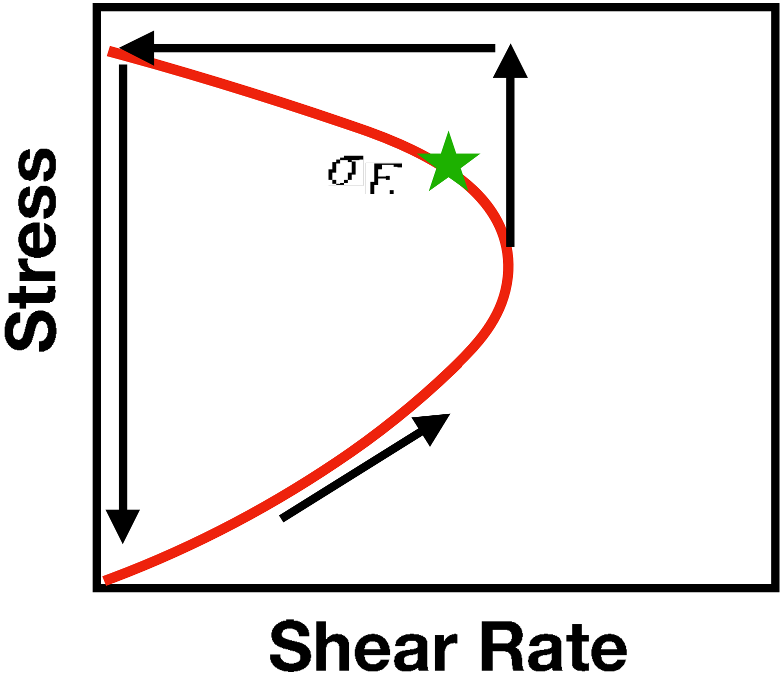


Bulk instability



$$\underbrace{\sigma_E}_{\text{Applied stress}} = \underbrace{\rho_A h \frac{\partial \dot{\gamma}}{\partial t}}_{\text{Plate acceleration}} + \underbrace{\eta(f)\dot{\gamma}}_{\text{Sample stress}}$$

Bulk
instability



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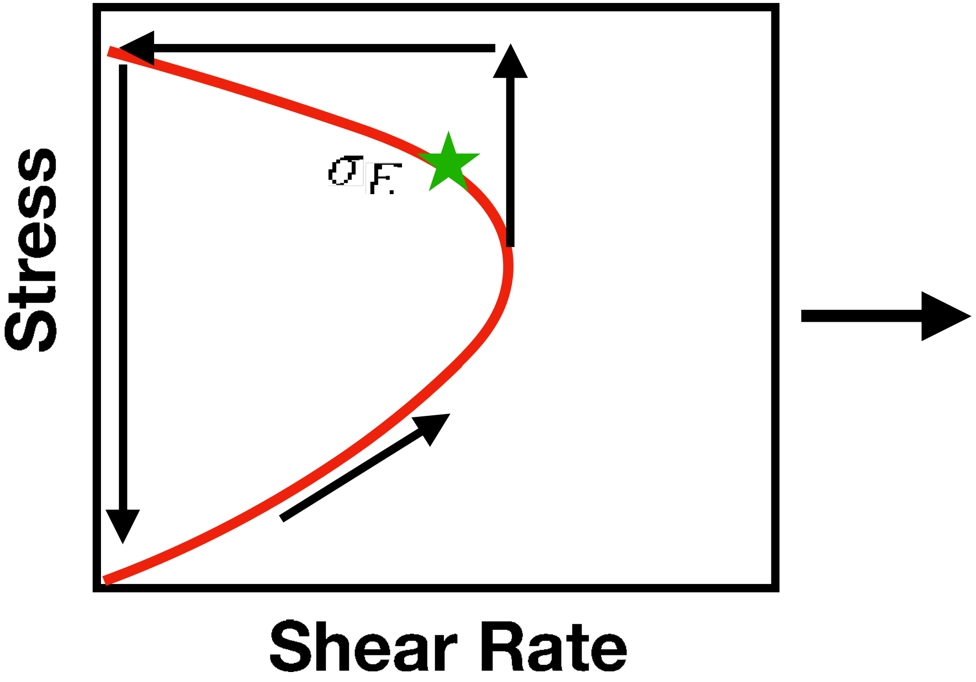

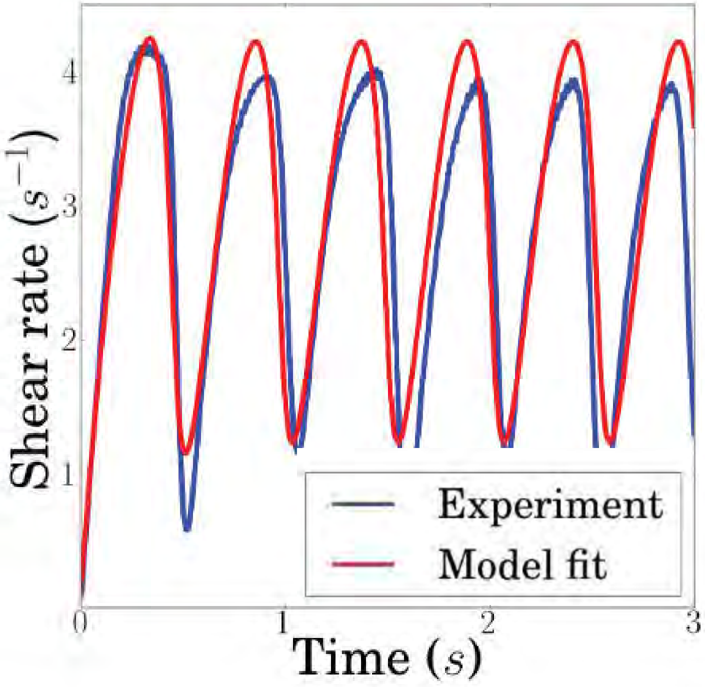

(b) 4.0Pa

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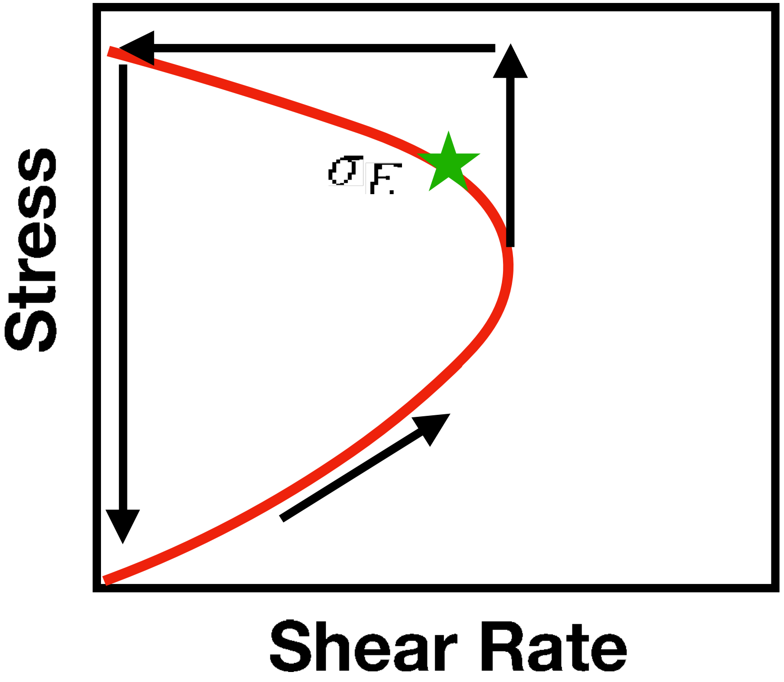

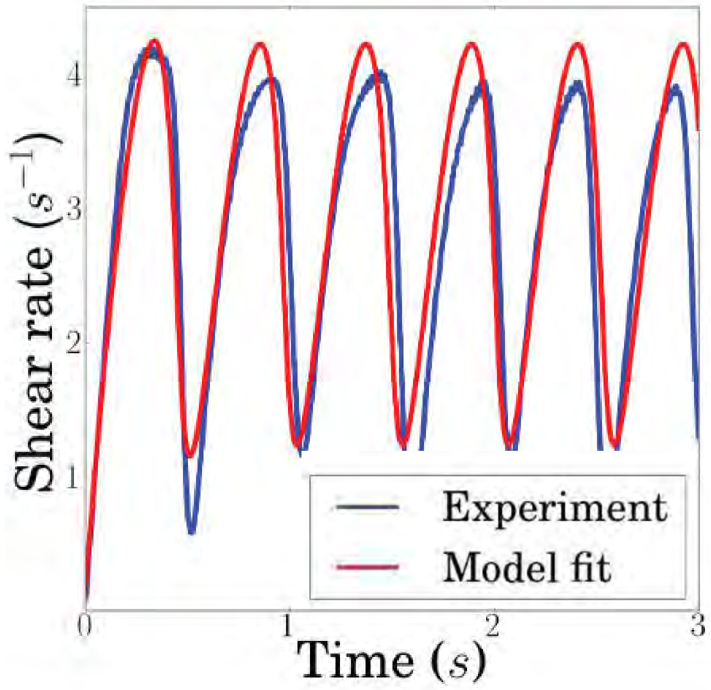

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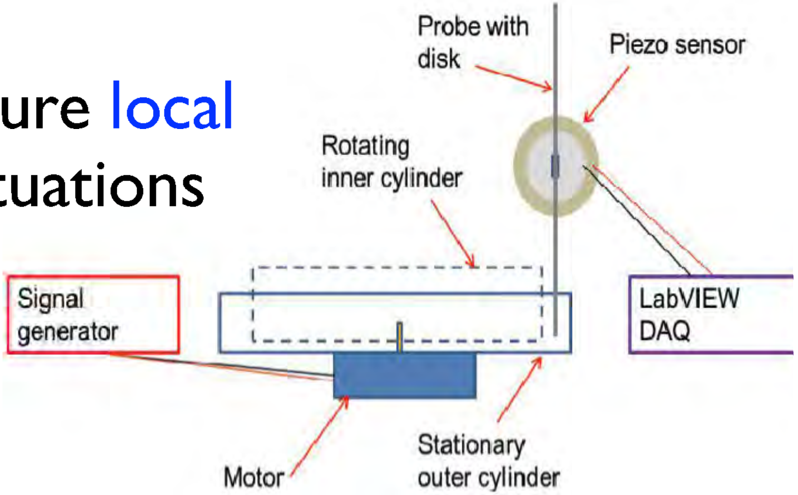
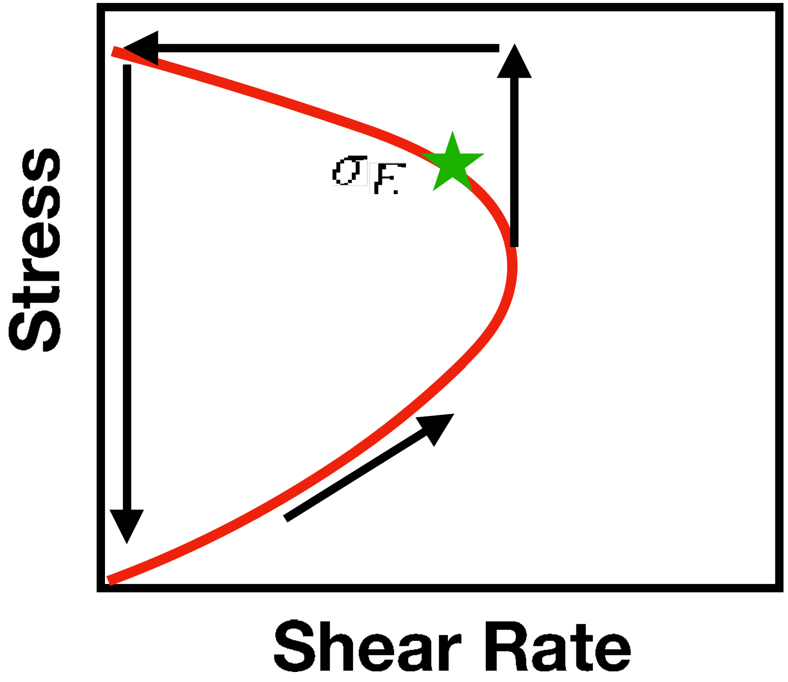
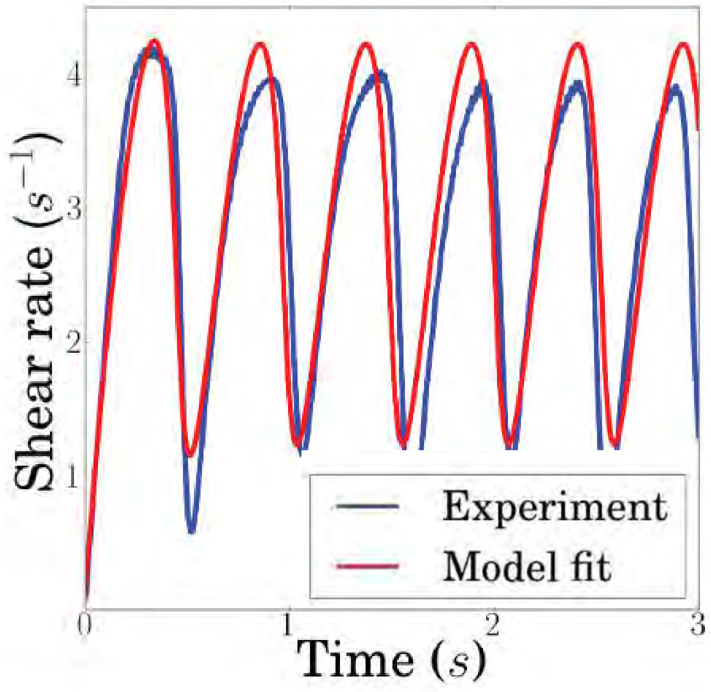
Measure local
fluctuations



Bulk instability





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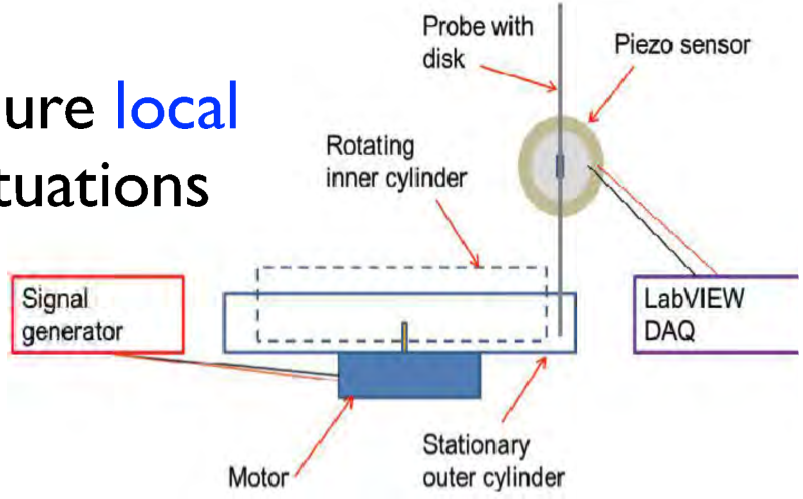
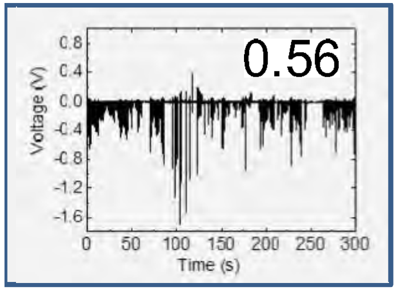
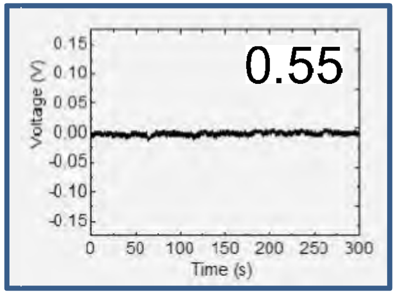
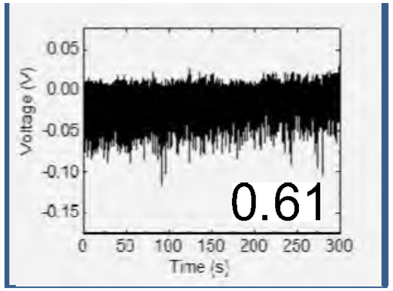








PHYSICAL REVIEW LETTERS **123,** 038004 (2019)

Competing Timescales Lead to Oscillations in Shear-Thickening Suspensions

J. A. Richards,[1,*] J. R. Royer,[1] B. Liebchen,[1,2] B. M. Guy,[1] and W. C. K. Poon[1]
[1]*SUPA, School of Physics and Astronomy, The University of Edinburgh, King's Buildings,*
Peter Guthrie Tait Road, Edinburgh EH9 3FD, United Kingdom
[2]*Institut für Theoretische Physik II: Weiche Materie, Heinrich-Heine-Universität Düsseldorf, D-40225 Düsseldorf, Germany*

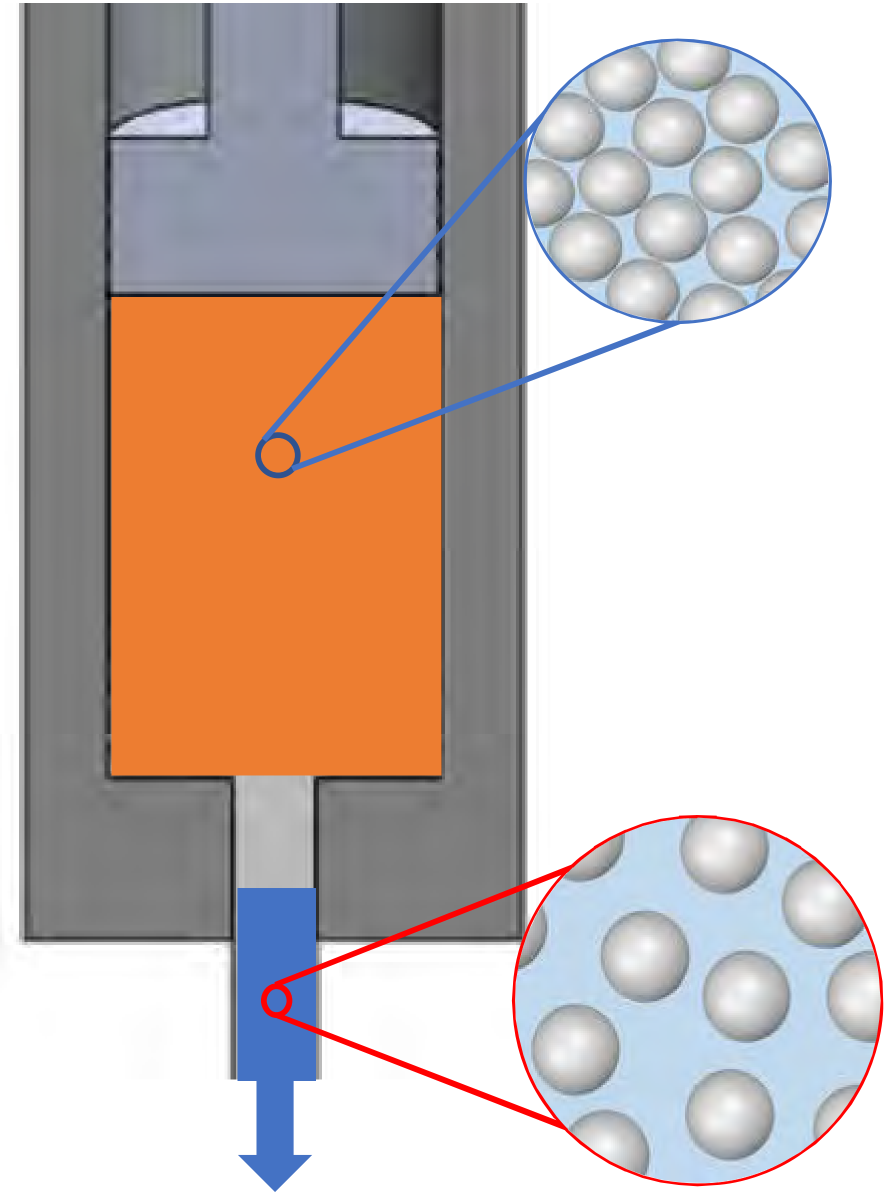
(Received 15 February 2019; published 19 July 2019)

Competing timescales generate novelty. Here, we show that a coupling between the timescales imposed by instrument inertia and the formation of interparticle frictional contacts in shear-thickening suspensions leads to highly asymmetric shear-rate oscillations. Experiments tuning the presence of oscillations by varying the two timescales support our model. The observed oscillations give access to a shear-jamming portion of the flow curve that is forbidden in conventional rheometry. Moreover, the oscillation frequency allows us to quantify an intrinsic relaxation time for particle contacts. The coupling of fast contact network dynamics to a slower system variable should be generic to many other areas of dense suspension flow, with instrument inertia providing a paradigmatic example.

DOI: 10.1103/PhysRevLett.123.038004

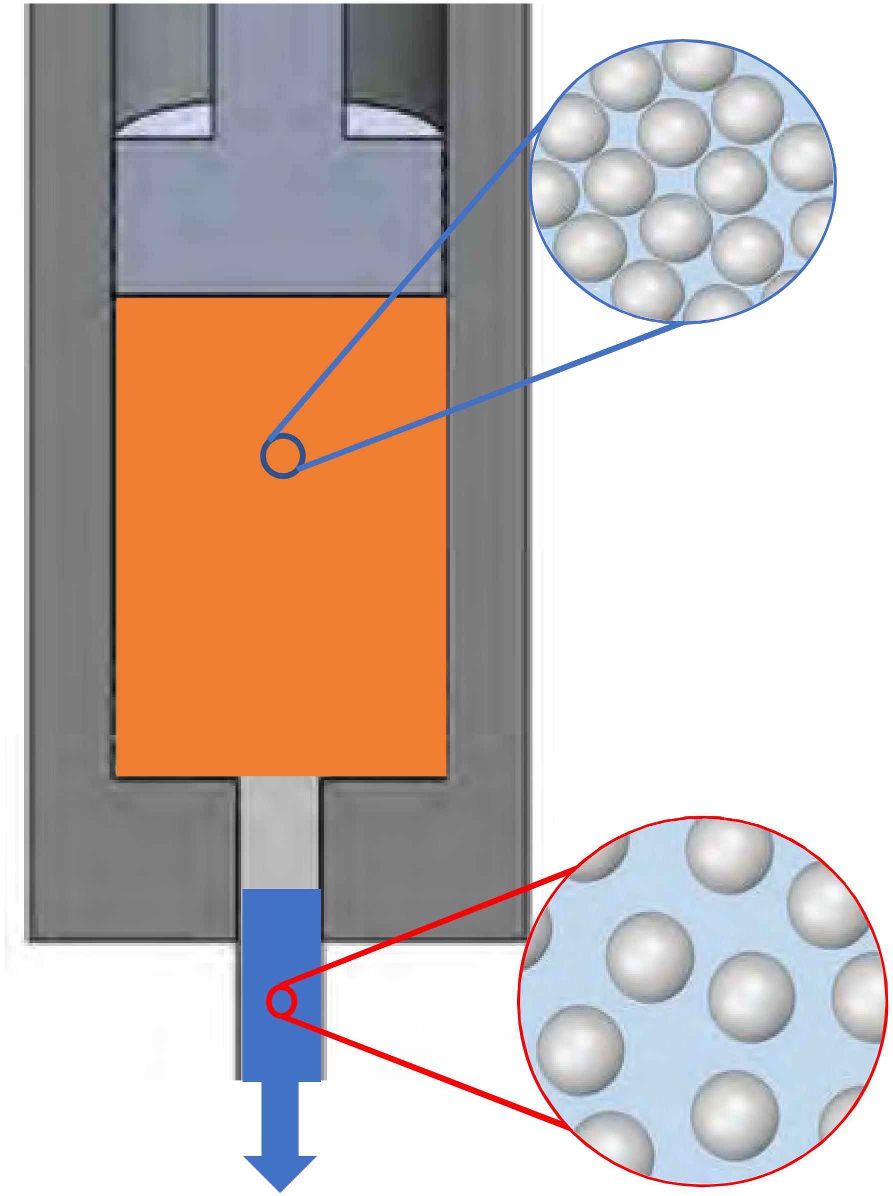
Shear thickening rheology in extrusion flow

Liquid migration

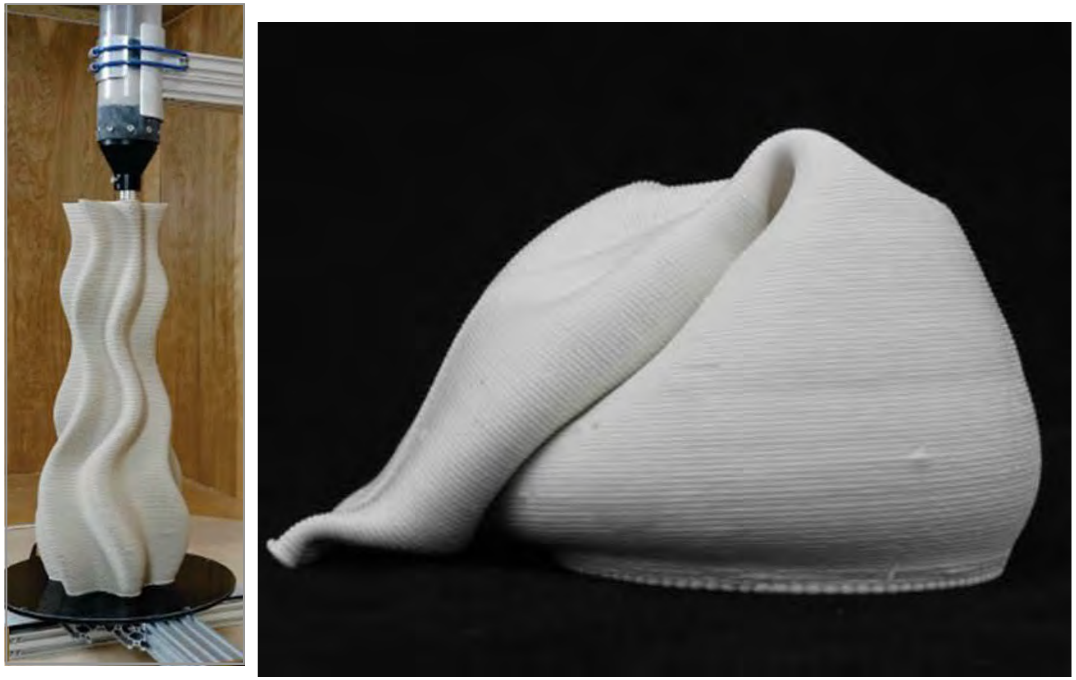


Downstream / extruded material much less concentrated

Liquid migration

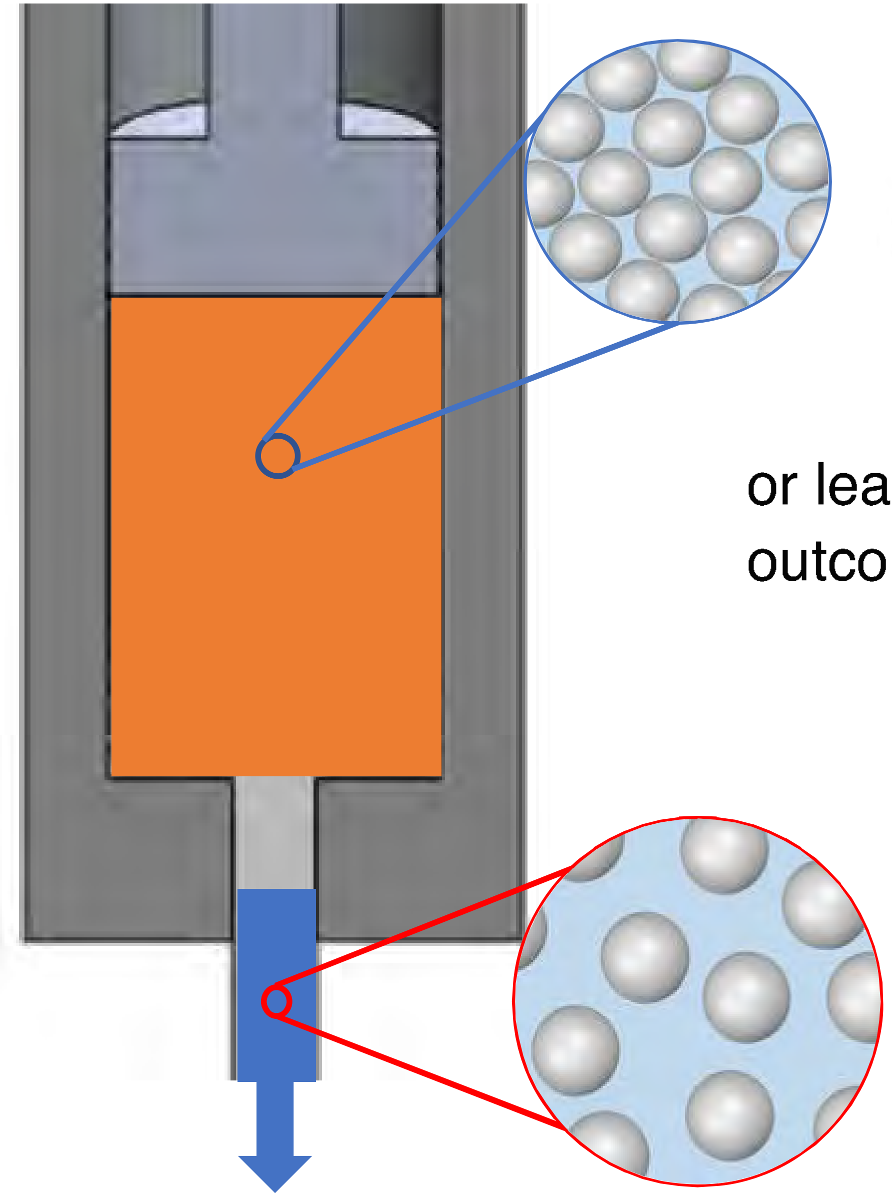


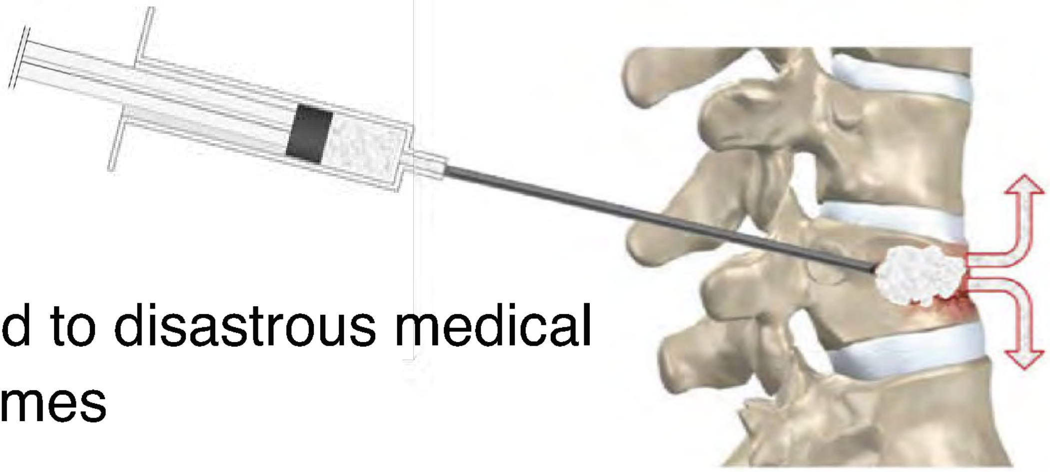
Downstream / extruded material much less concentrated



Loss of solids can impede extrusion-based manufacturing

Liquid migration





or lead to disastrous medical outcomes

Downstream / extruded material much less concentrated

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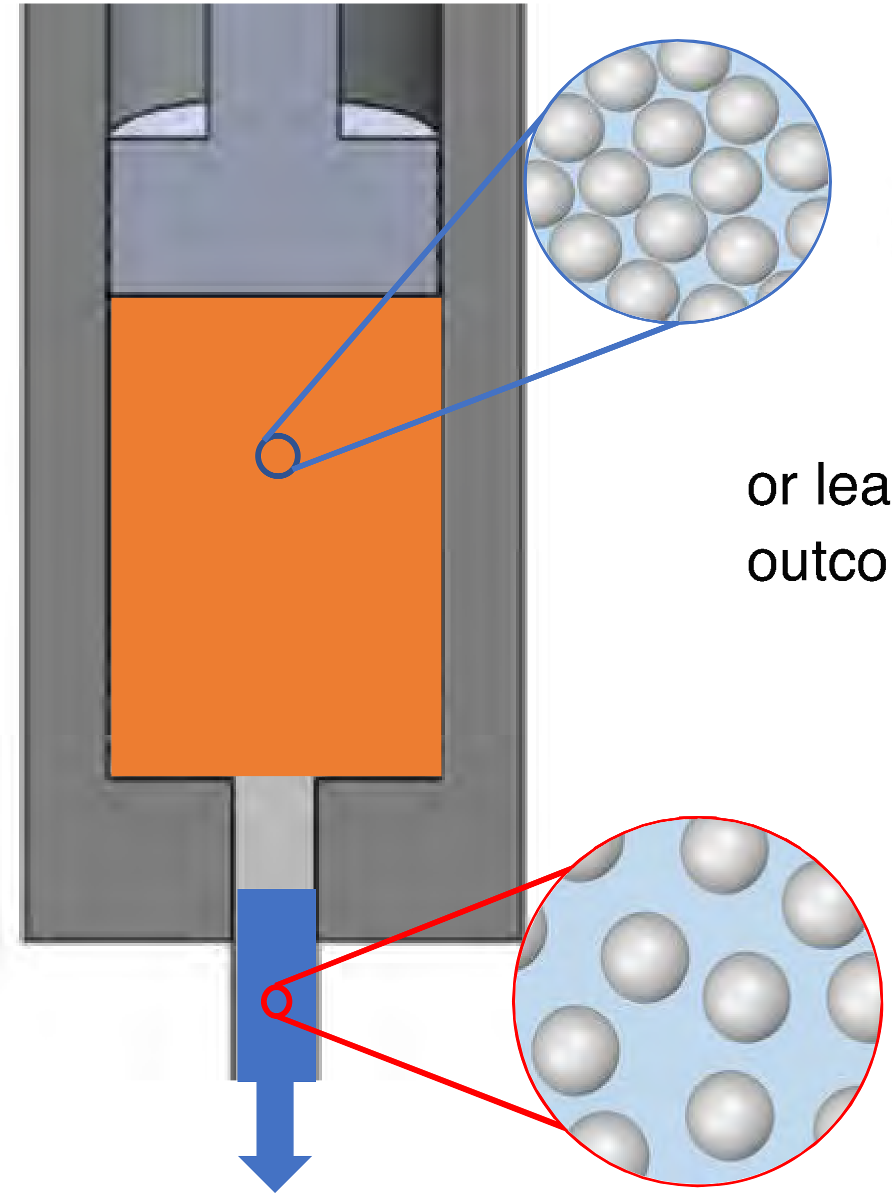
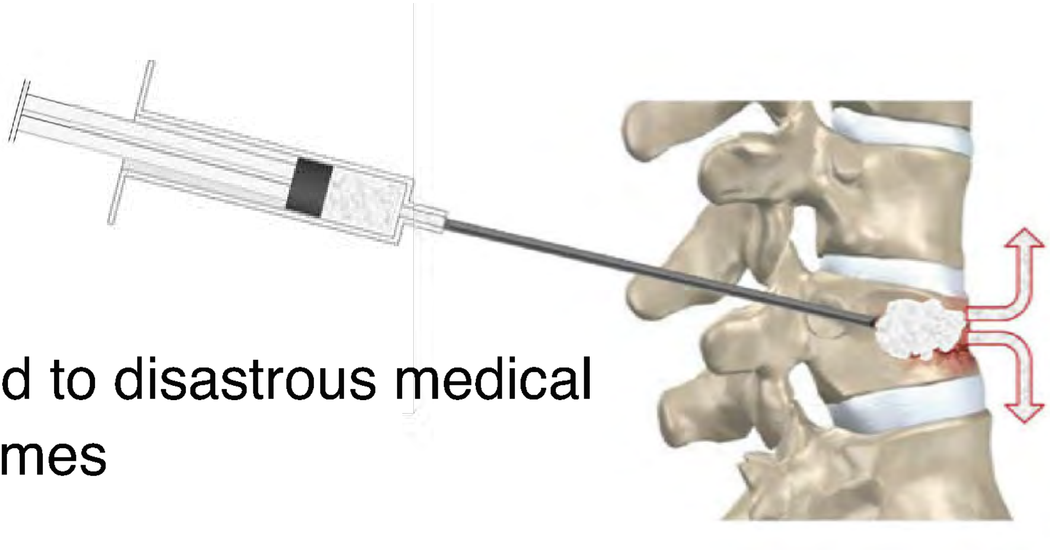


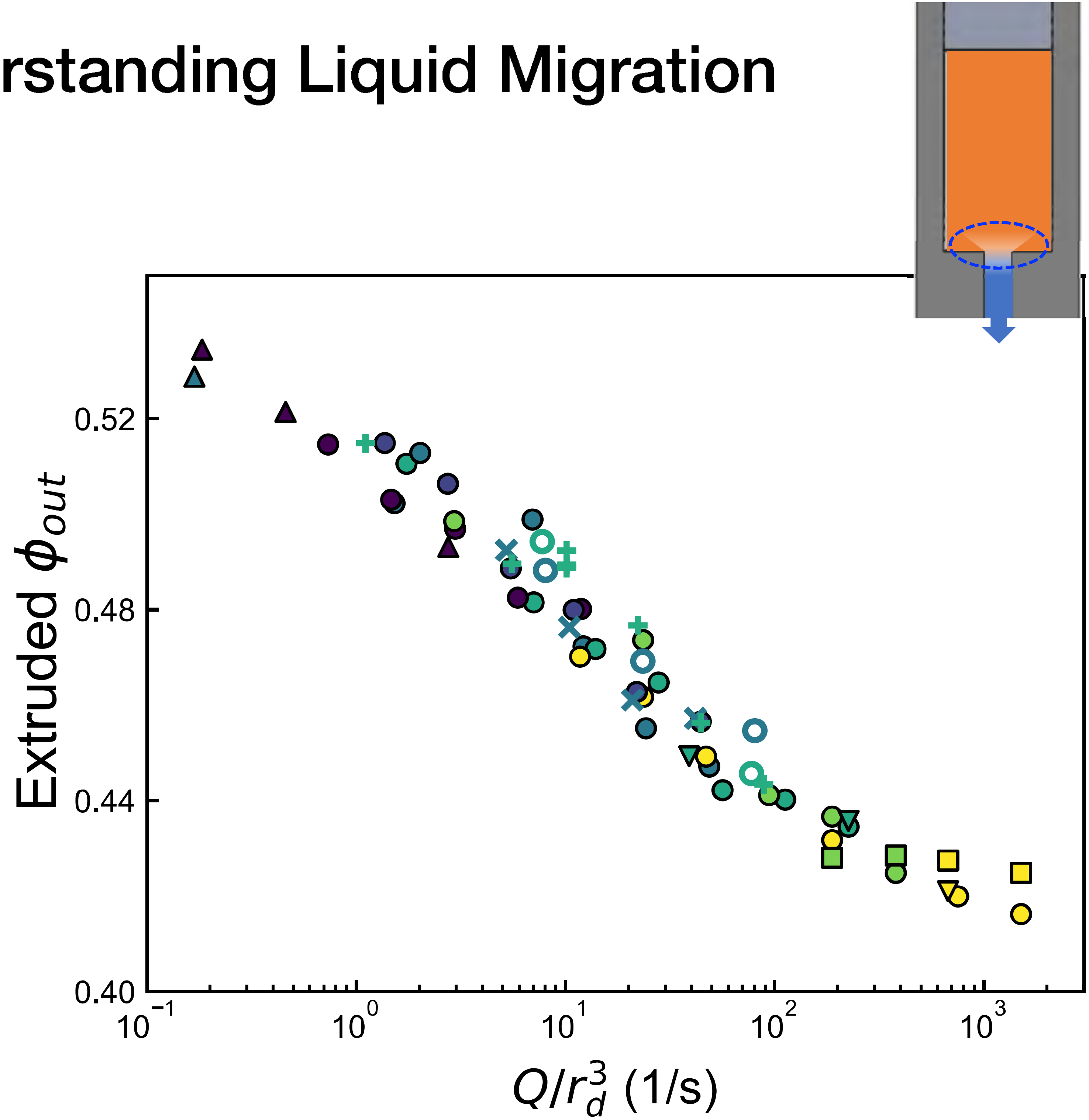


or lead to disastrous medical outcomes

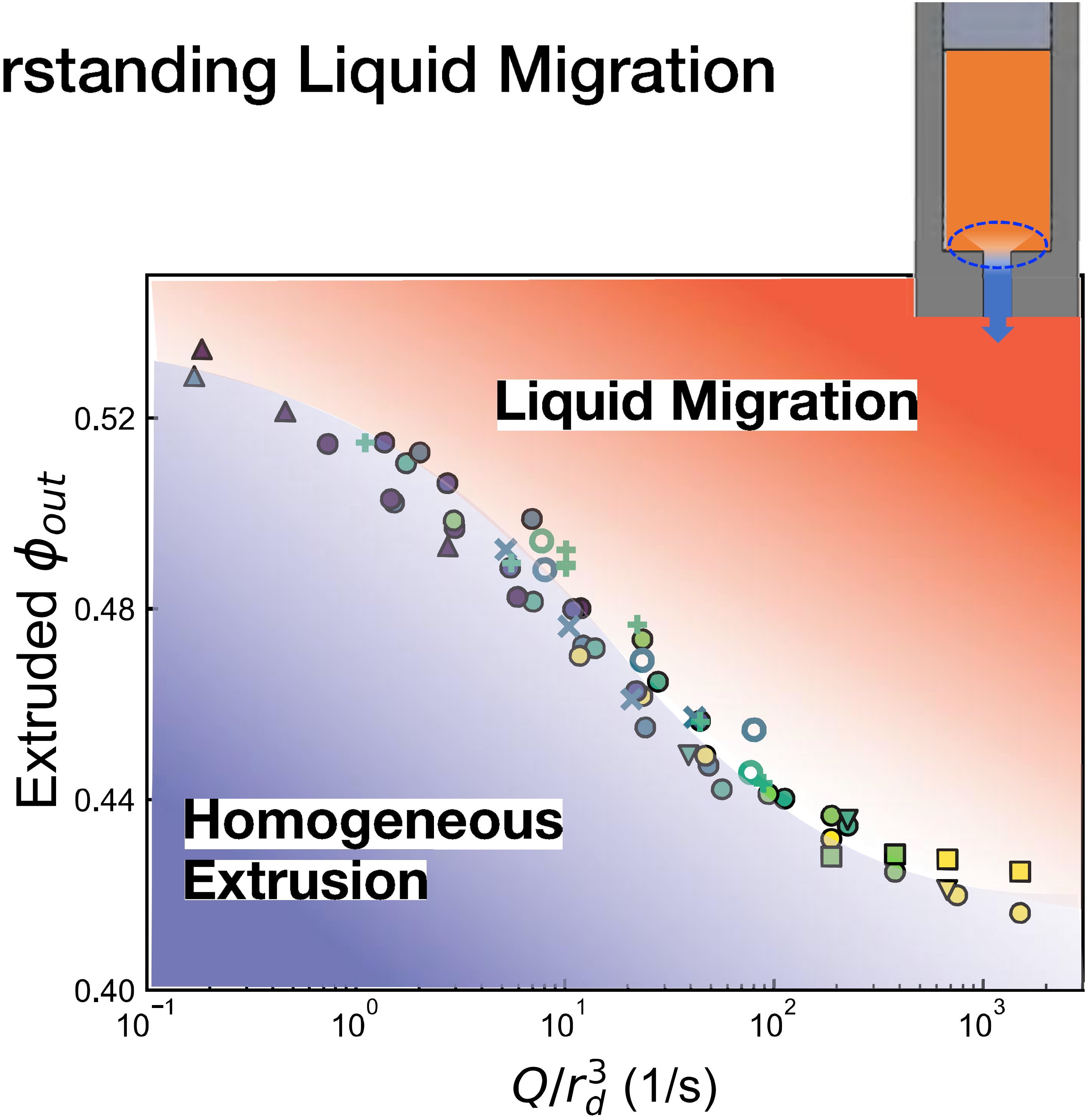
Mechanisms behind Liquid Migration remain poorly understood

Downstream / extruded material much less concentrated

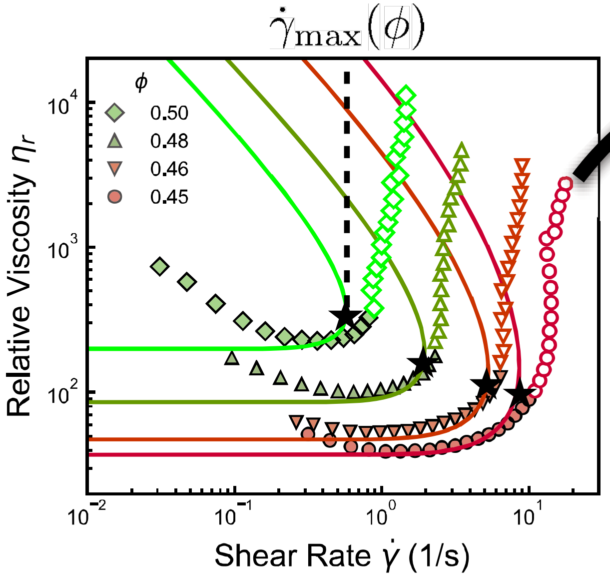
Understanding Liquid Migration

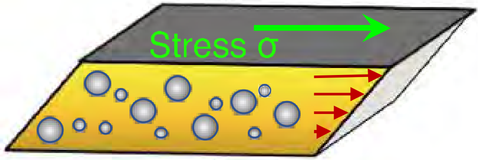


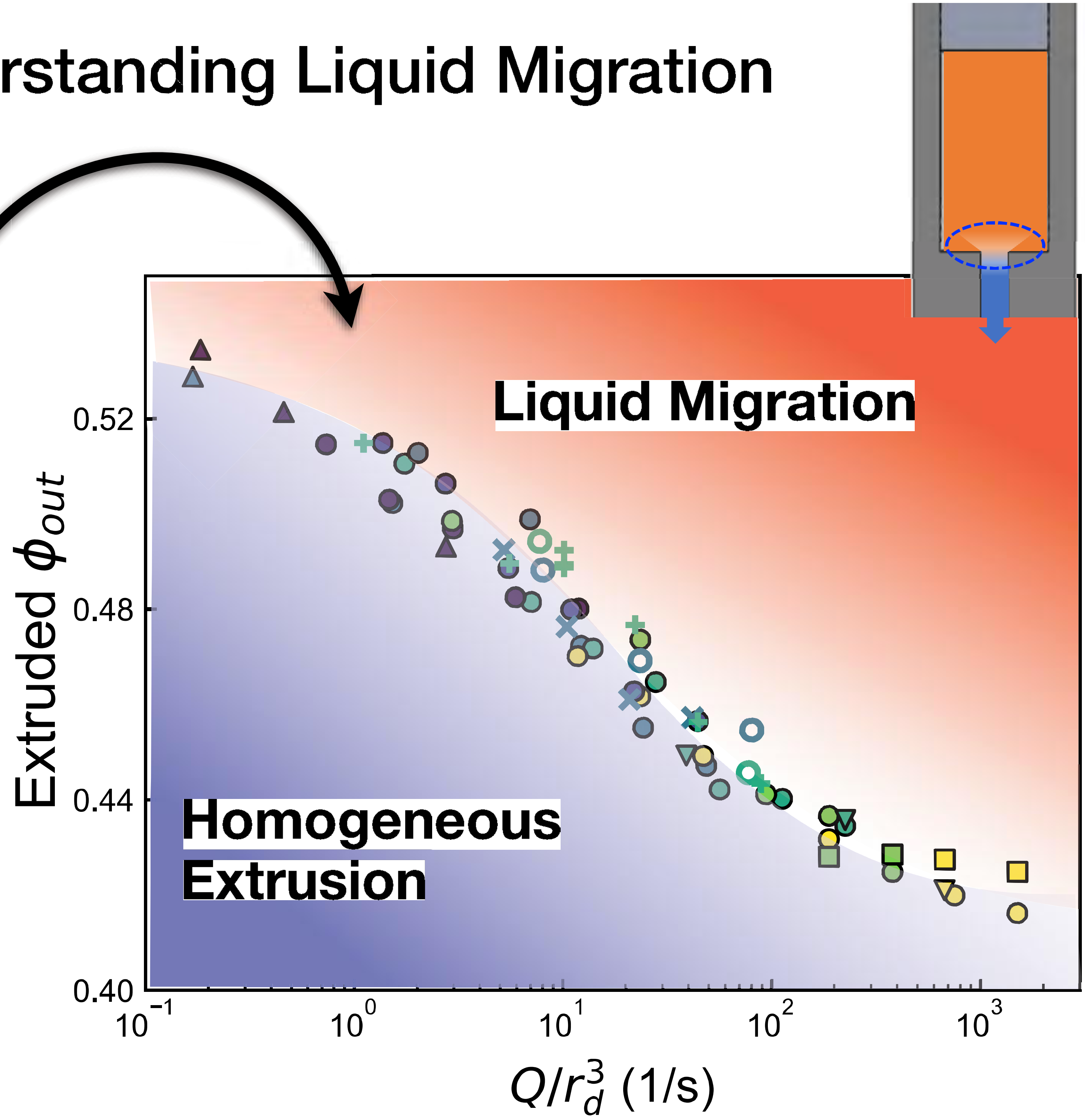
Understanding Liquid Migration



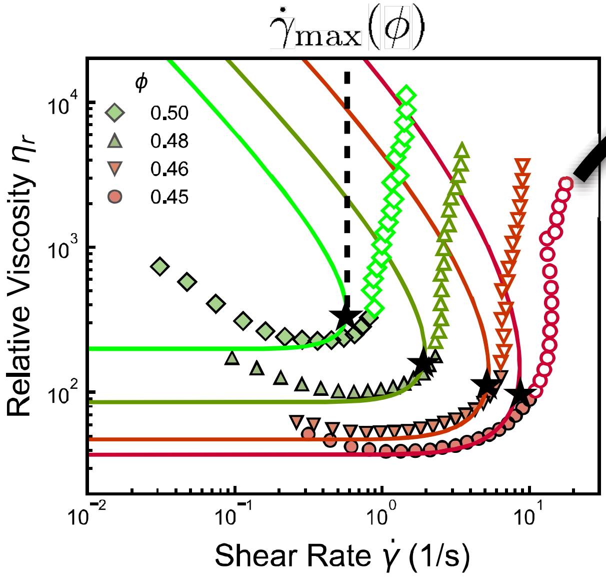
Understanding Liquid Migration

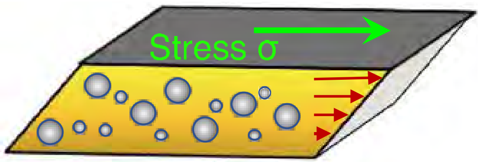


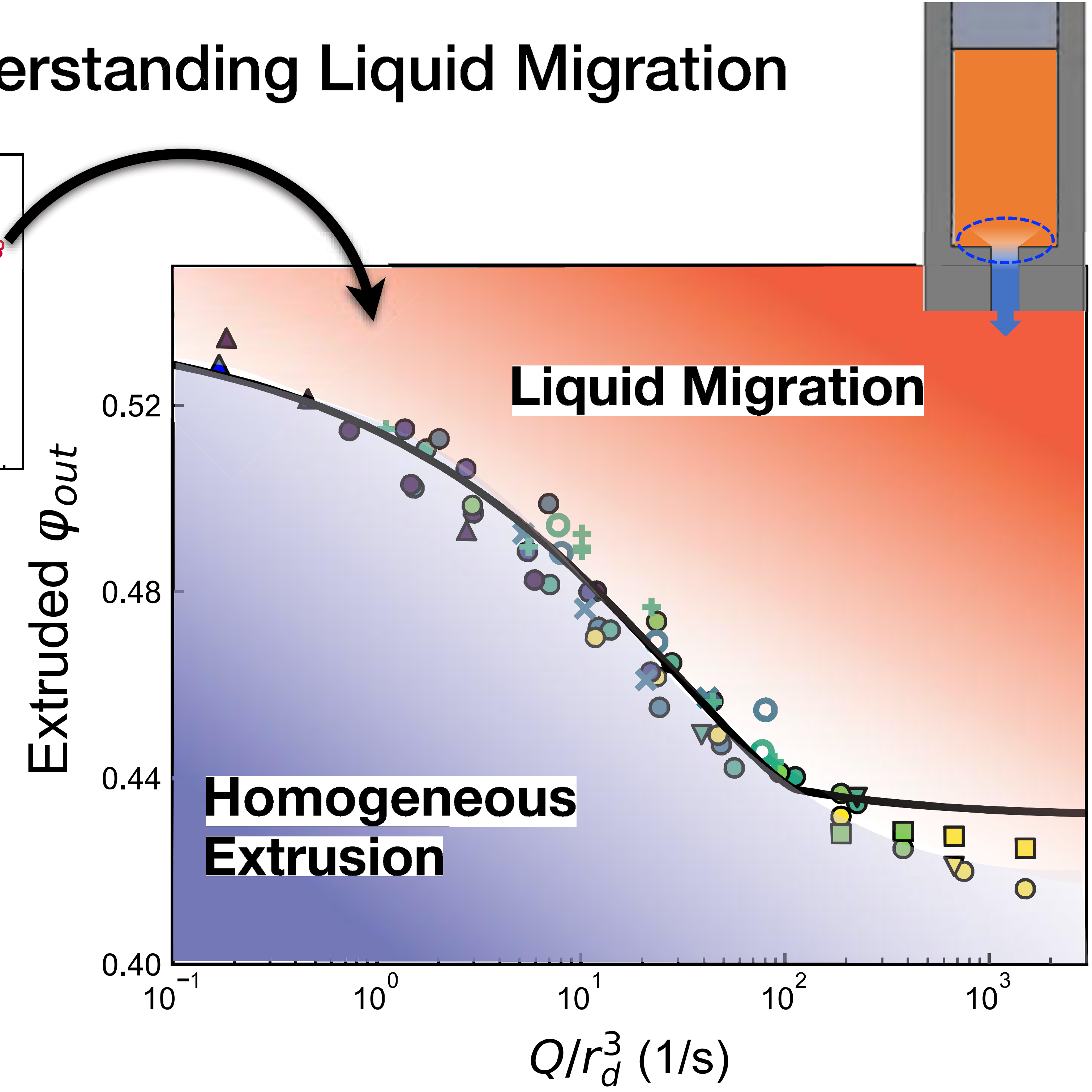




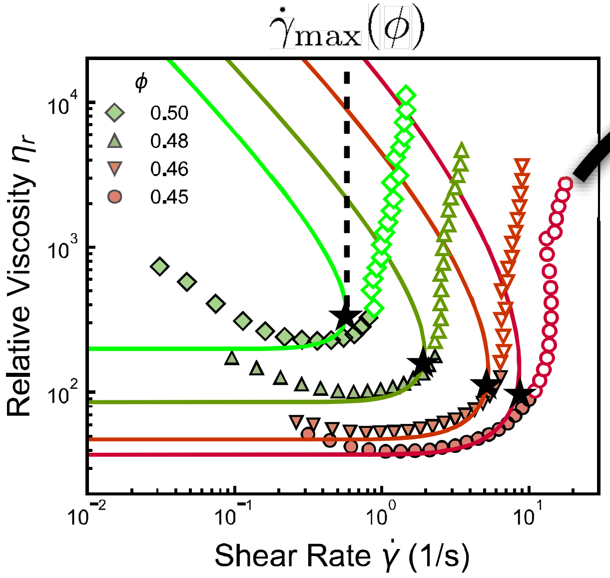
Understanding Liquid Migration

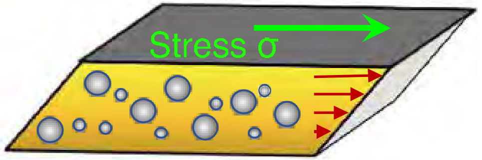




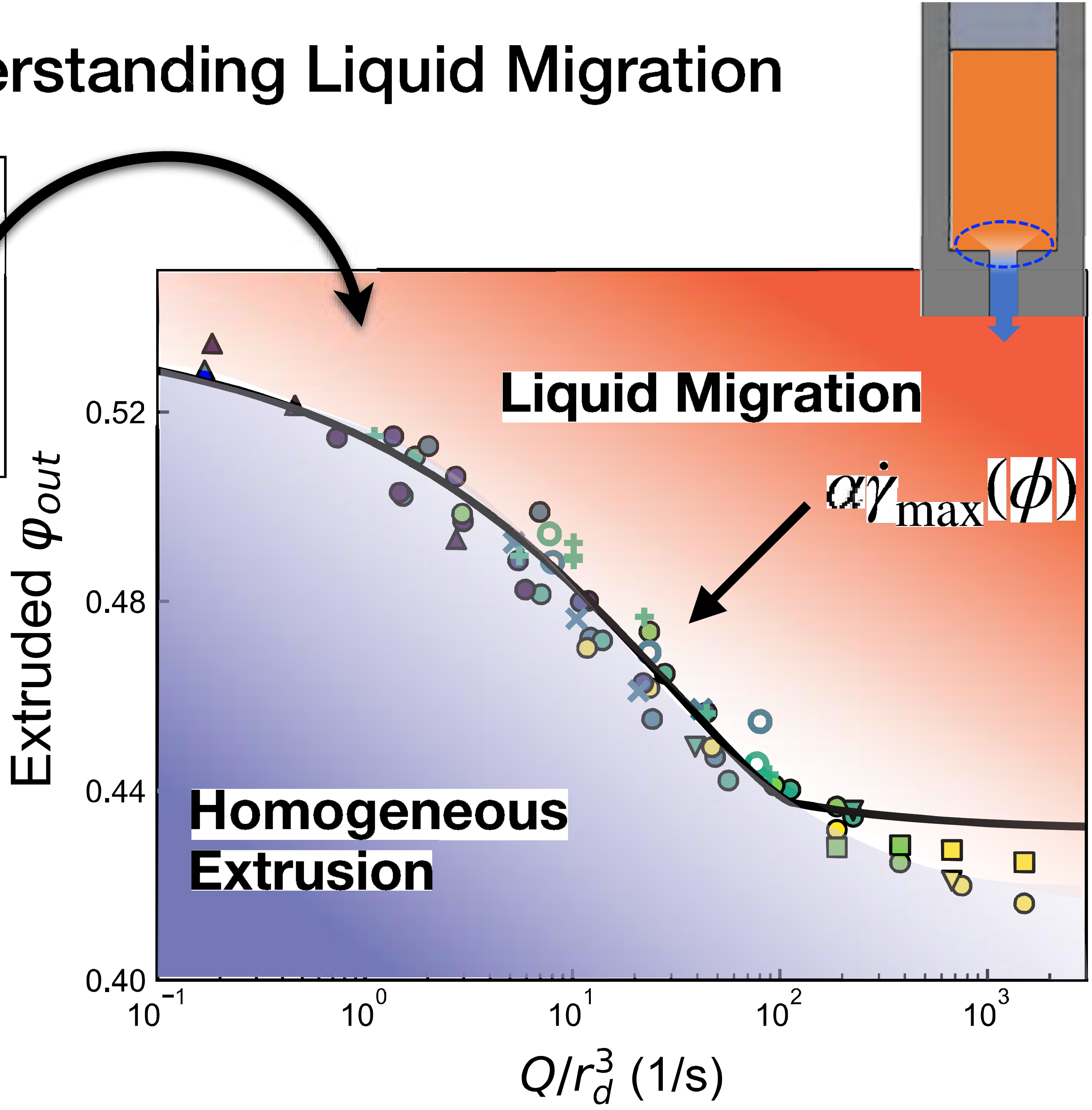


Understanding Liquid Migration





Shear rheology
predicts migration
phase boundary
with single O(1)
free parameter



PHYSICAL REVIEW LETTERS 123, 128002 (2019)

Liquid Migration in Shear Thickening Suspensions Flowing through Constrictions

Rory E. O'Neill, John R. Royer, and Wilson C. K. Poon
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Peter Guthrie Tait Road, Edinburgh EH9 3FD, United Kingdom

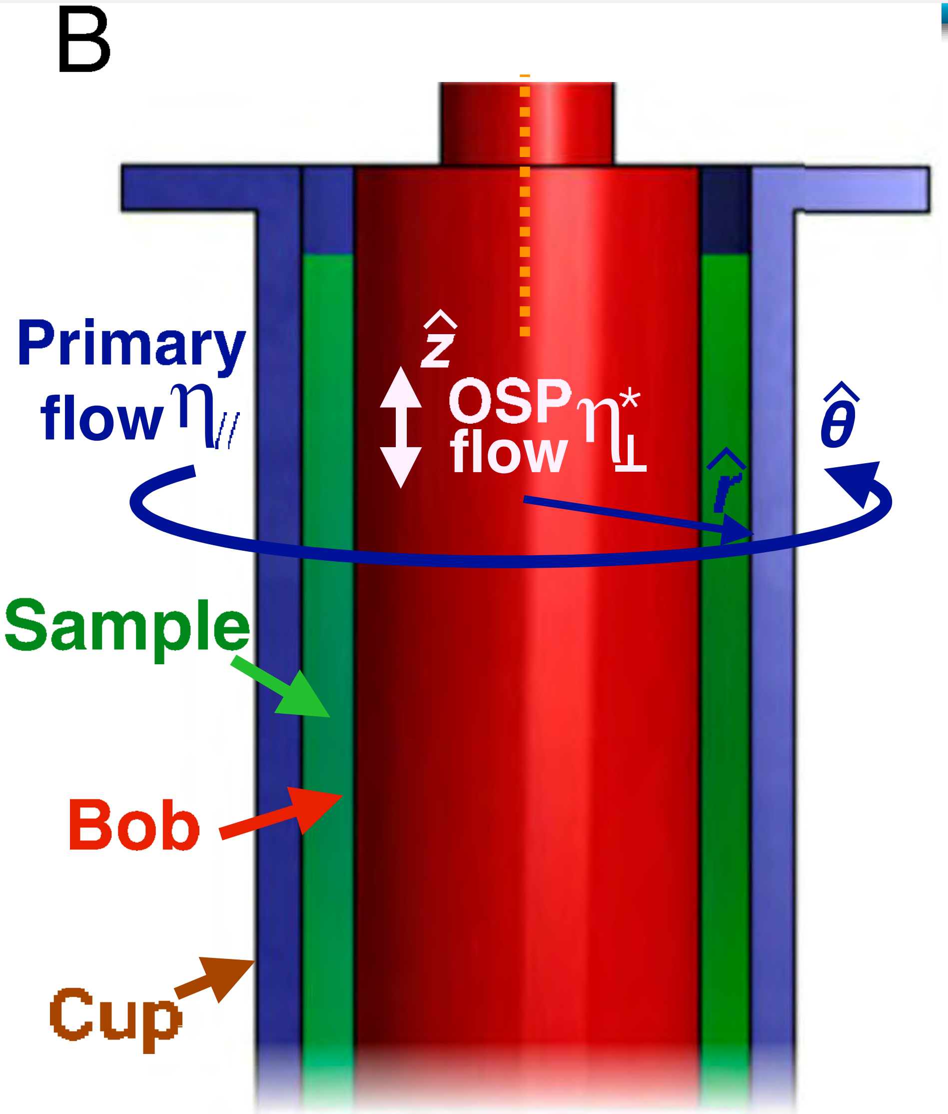
(Received 1 August 2018; revised manuscript received 19 July 2019; published 16 September 2019)

Dense suspensions often become more dilute as they move downstream through a constriction. We find that as a shear-thickening suspension is extruded through a narrow die and undergoes such liquid migration, the extrudate maintains a steady concentration $\phi_{\mathrm{out}}^{\mathrm{LM}}$, independent of time or initial concentration. At low volumetric flow rate Q, $\phi_{\mathrm{out}}^{\mathrm{LM}}$ is a universal function of Q/r_d^3, a characteristic shear rate in the die of radius r_d, and coincides with the critical input concentration for the onset of LM, $\phi_{\mathrm{in}}^{\mathrm{crit}}$. We predict this function by coupling the Wyart-Cates model for shear thickening and the "suspension balance model" for solvent permeation through particles.

DOI: 10.1103/PhysRevLett.123.128002

Tuning viscosity using superimposed flow

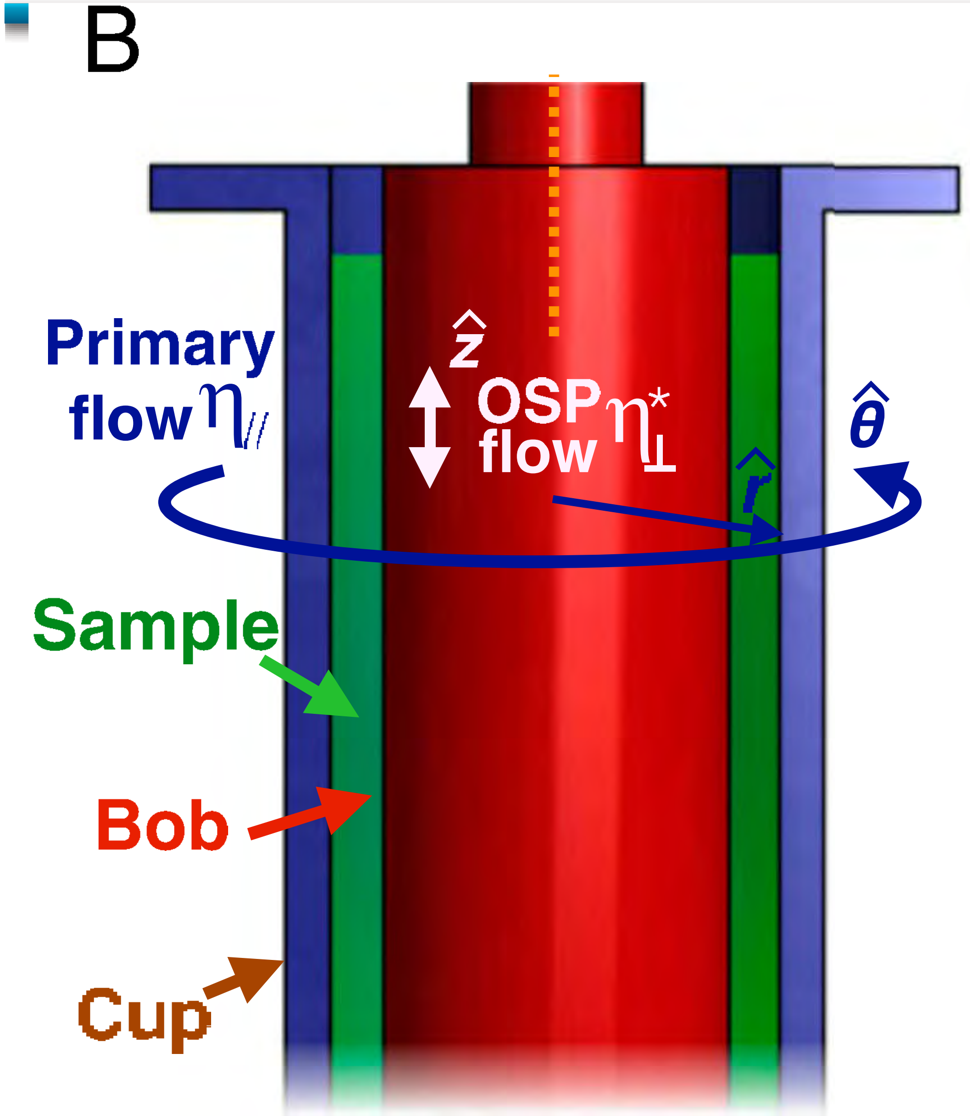
Orthogonal superimposed perturbation

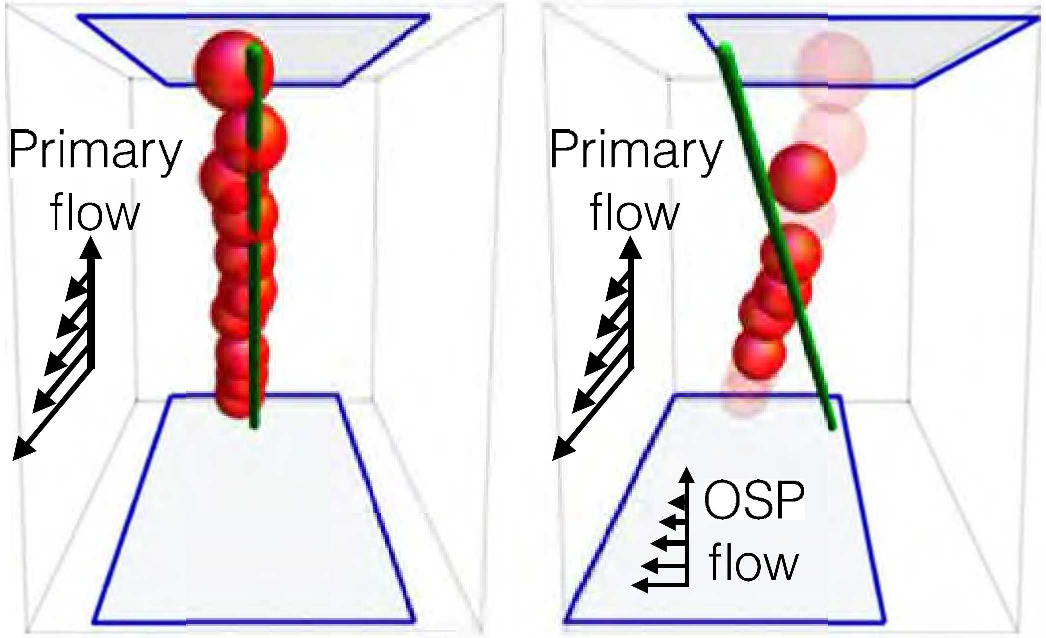


$$\gamma^{\mathrm{OSP}} = \gamma_0^{\mathrm{OSP}} \sin(\omega t) \qquad \dot{\gamma}^{\mathrm{OSP}} = \omega \gamma_0^{\mathrm{OSP}} \cos(\omega t)$$

N.Y.C. Lin, C.J. Ness, M.E.Cates, J. Sun and I. Cohen, PNAS, 2016, 201608348

Orthogonal superimposed perturbation





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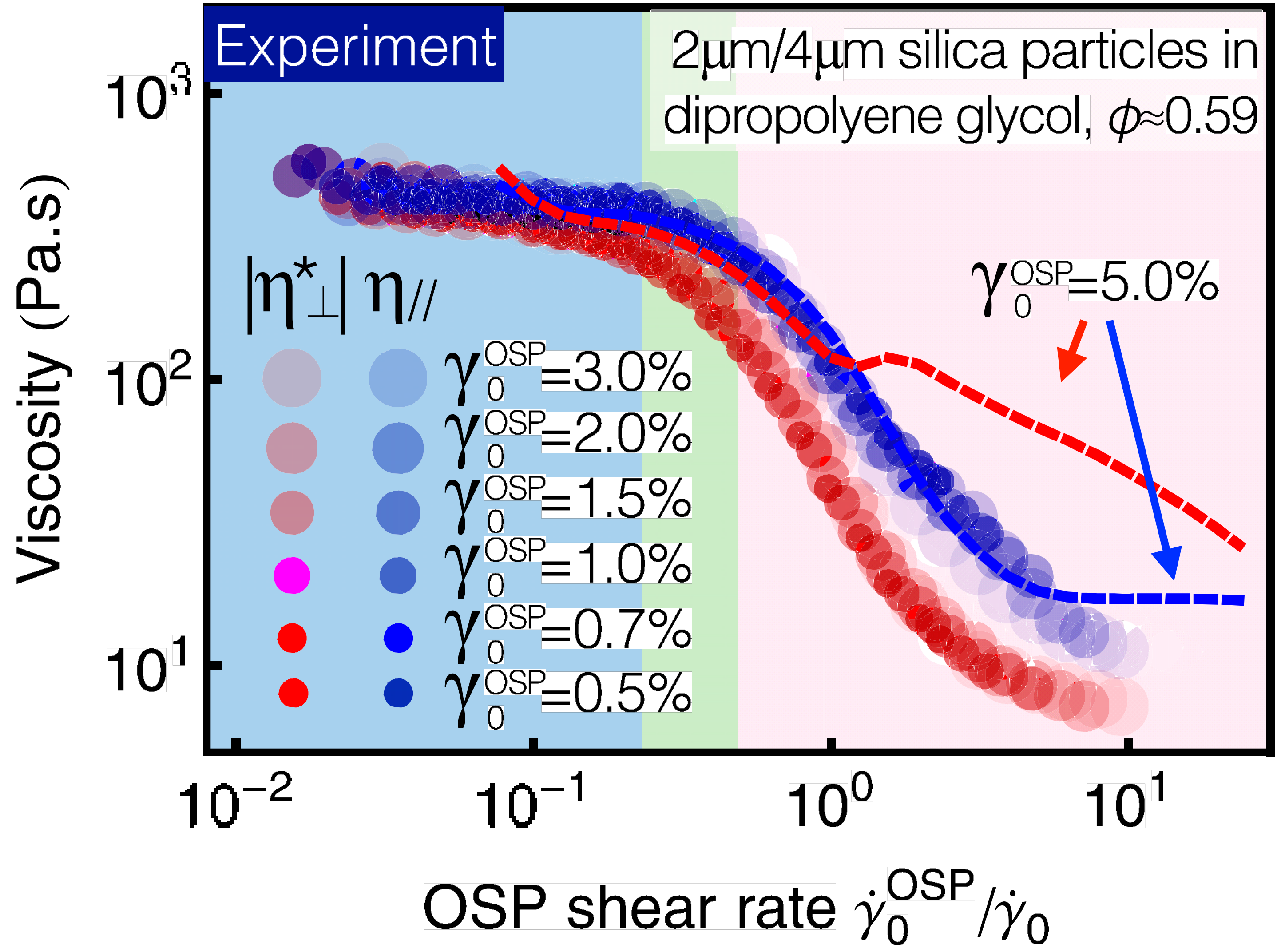
Tuneable viscosity



Blue dots: primary viscosity; red dots: orthogonal complex viscosity

- Shear thickened viscosity can be reduced by orders of magnitude

PNAS

Tunable shear thickening in suspensions

Neil Y.C. Lin[a,1], Christopher Ness[b], Michael E. Cates[c], Jin Sun[b], and Itai Cohen[a]

[a]Department of Physics, Cornell University, Ithaca, NY 14853; [b]School of Engineering, University of Edinburgh, Edinburgh EH9 3JL, United Kingdom; and [c]Department of Applied Mathematics and Theoretical Physics, Centre for Mathematical Sciences, University of Cambridge, Cambridge CB3 0WA, United Kingdom

Edited by David A. Weitz, Harvard University, Cambridge, MA, and approved July 26, 2016 (received for review May 25, 2016)

Shear thickening, an increase of viscosity with shear rate, is a ubiquitous phenomenon in suspended materials that has implications for broad technological applications. Controlling this thickening behavior remains a major challenge and has led to empirical strategies ranging from altering the particle surfaces and shape to modifying the solvent properties. However, none of these methods allows for tuning of flow properties during shear itself. Here, we demonstrate that by strategic imposition of a high-frequency and low-amplitude shear perturbation orthogonal to the primary shearing flow, we can largely eradicate shear thickening. The orthogonal shear effectively becomes a regulator for controlling thickening in the suspension, allowing the viscosity to be reduced by up to 2 decades on demand. In a separate setup, we show that such effects can be induced by simply agitating the sample transversely to the primary shear direction. Overall, the ability of in situ manipulation of shear thickening paves a route toward creating materials whose mechanical properties can be controlled.

rheology | colloidal suspensions | shear thickening | flow control

To that end, we design a biaxial shear protocol that uses an orthogonal flow perturbation to interfere with force chains induced by a primary shearing flow. Our strategy is to maximize the perturbation influence so the force chains usually responsible for thickening cannot establish fully. We conduct biaxial rheometry experimentally and numerically, mapping the response of a hard-sphere suspension as the perturbation rate and amplitude are systematically varied. By integrating our knowledge of the force-chain alignment, mechanical instability, and direct link to the viscosity, we show how this strategy can be optimized. We focus on discontinuous shear-thickening suspensions, as their vast viscosity variations make them most problematic to the engineer (10, 11, 20). Our results show that through suitable regulation, the suspension viscosity at a fixed flow rate may be reduced by up to 2 decades in an active and controlled manner. We finally demonstrate the wide utility of the technique using a simpler flow regulation setup.

The biaxial rheometry experiment is performed using a double-wall Couette geometry that has an outer cup driven continuously by an underneath motor, and an inner bob attached to an oscillating

Conclusions

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- Details matter! Surface interaction details (nano-scale) varying particle friction, adhesion and repulsion can radically change the rheology (metre scale) of high-solid-content dispersions
- The radical effects and ‘noise’ in rheology can be understand and predicted via stress/flow-activated particle interactions
- Flow in complex geometries can in turn be predicted using the rheological models
- Provide principles and predictive tools to guide formulation and processing of high-solid-content dispersions

Predictive formulation of high-solid-content complex dispersions

EPSRC grant EP/N025318/1

Thank you!

Questions?