

Shear-induced inhomogeneities in concentrated suspensions

Guillaume Ovarlez, LOF, CNRS-Solvay-Univ. Bordeaux

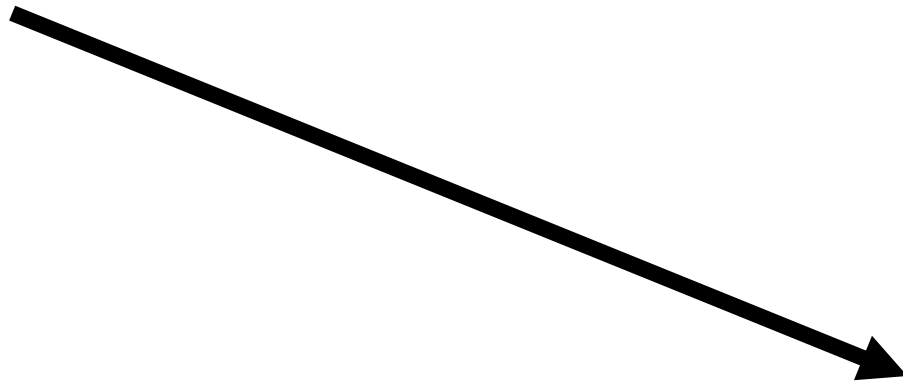
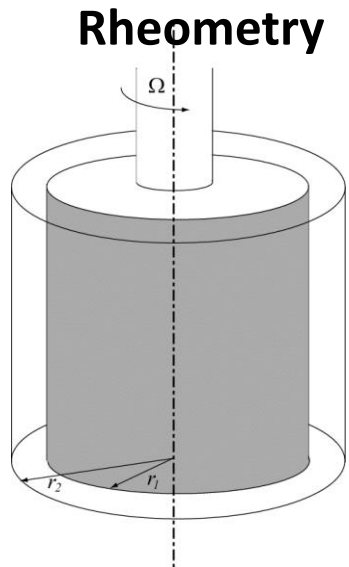
with contributions from

Elisabeth Guazzelli

Abdoulaye Fall, Daniel Bonn & Anaël Lemaître

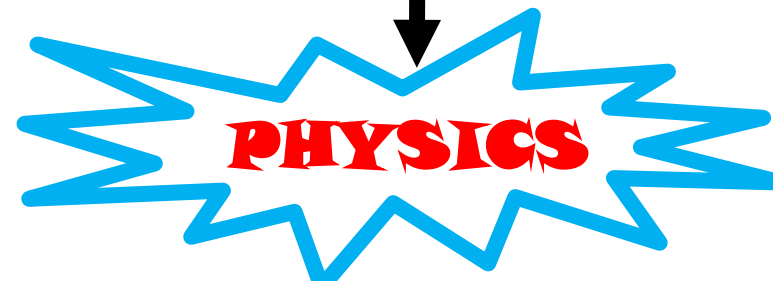
Guillaume Chatté, Nicolas Lenoir & Annie Colin

From rheometry to physics...

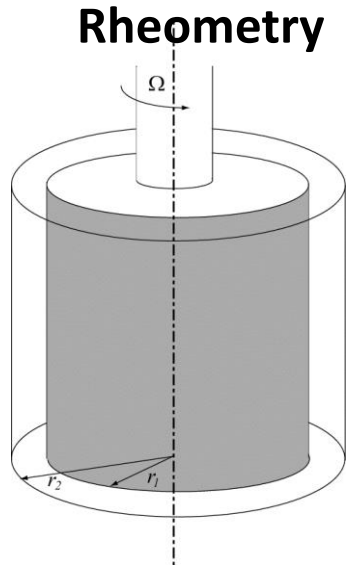


Constitutive law:

$$\tau(\dot{\gamma})$$



From rheometry to physics...

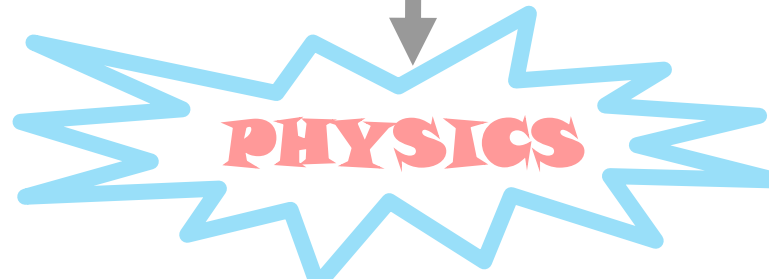


The (boring?) rheologist question

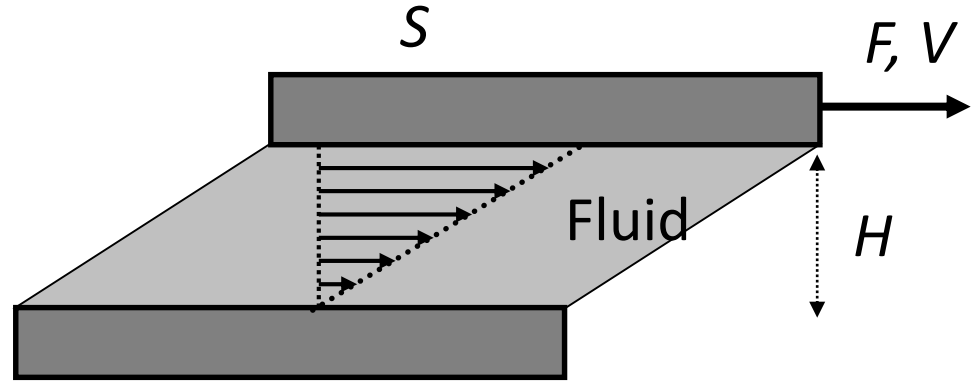
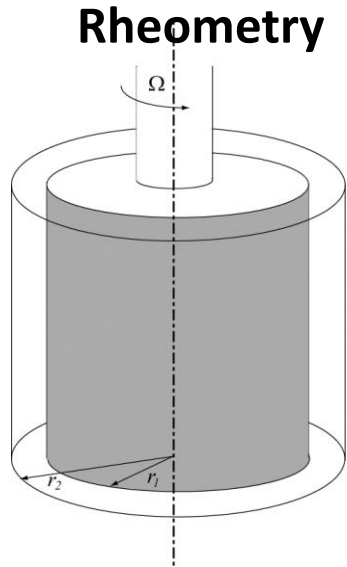
Constitutive law:

$$\tau(\dot{\gamma})$$

nature



From rheometry to physics...



$$\tau = \frac{F}{S} \quad \dot{\gamma} = \frac{V}{H}$$

The rheologist question

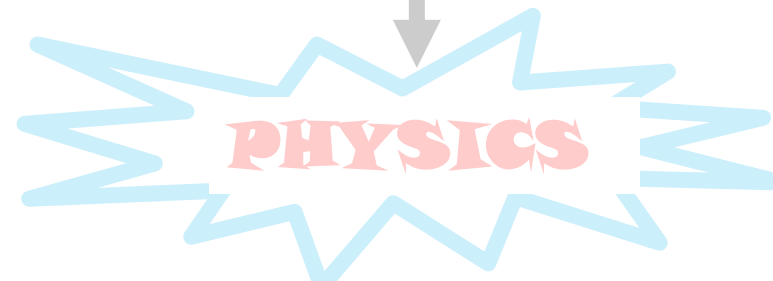
Hypotheses :

- homogeneous shear
- homogeneous material

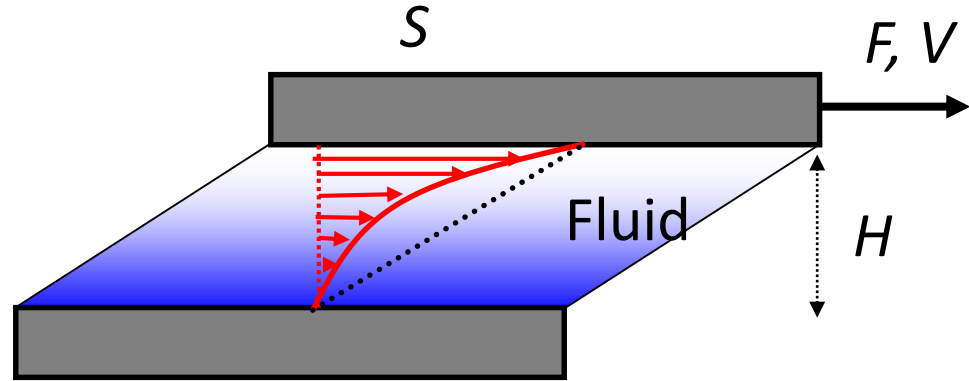
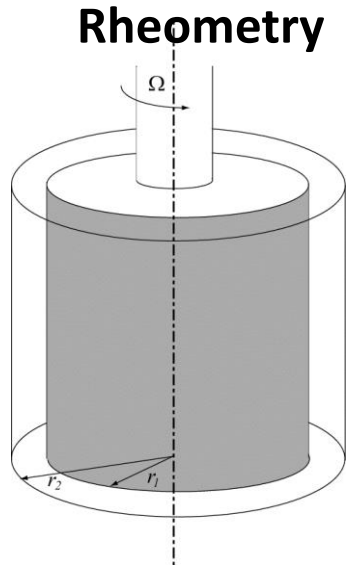
Constitutive law?????

$$\tau(\dot{\gamma})$$

nature



From rheometry to physics...



$$\tau = \frac{F}{S}$$

~~$$\dot{\gamma} = \frac{V}{H}$$~~

The rheologist question

Macroscopic measurement:

F(V)
(or T(Ω))

Hypotheses :

- ?
- homogeneous shear
 - homogeneous material

Constitutive law:

$$\tau(\dot{\gamma}, x)$$

Bad evaluation of shear rate

Bad knowledge of the material

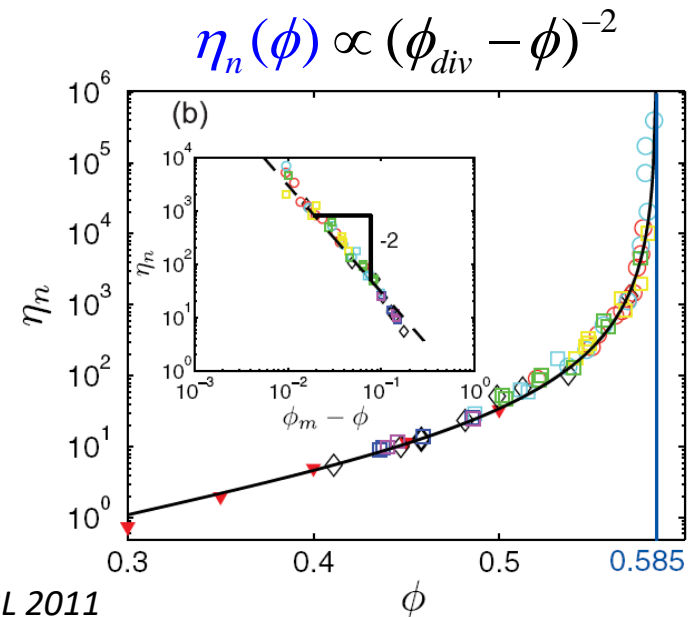
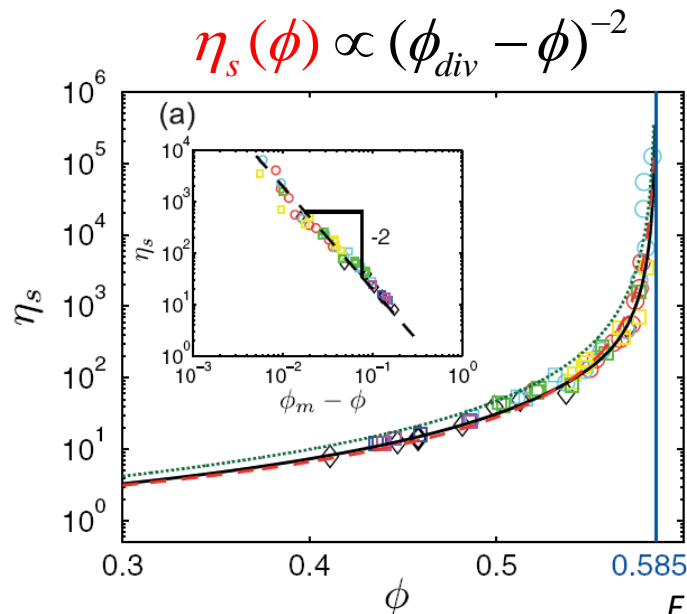
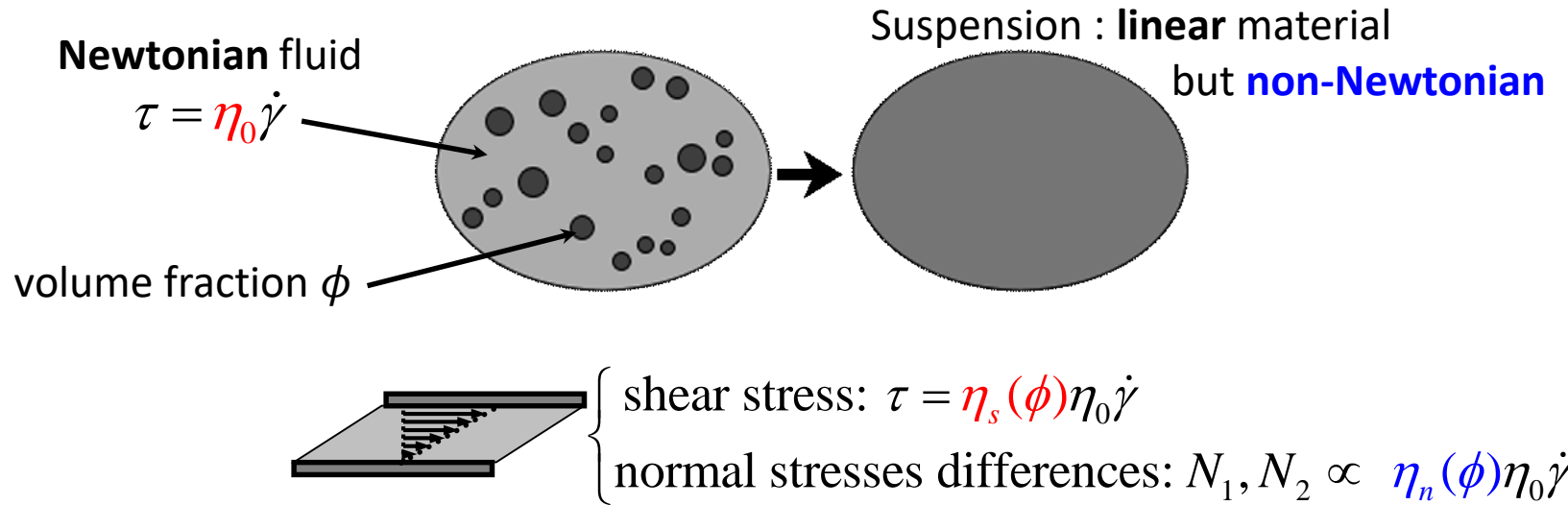


Shear-induced migration in viscous suspensions

**Shear-induced migration in shear-thickening suspensions:
(1) Role of interparticle contacts**

**Shear-induced migration in shear-thickening suspensions
(2) Jamming**

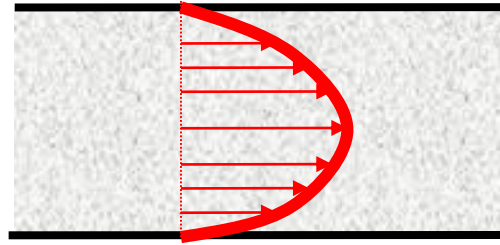
Macroscopic behavior of **viscous nonBrownian** suspensions



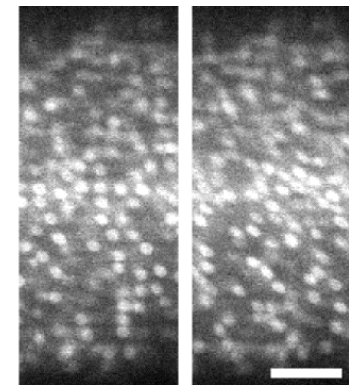
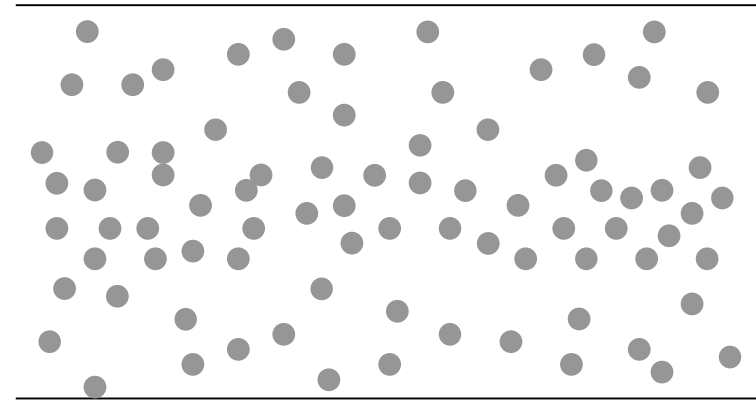
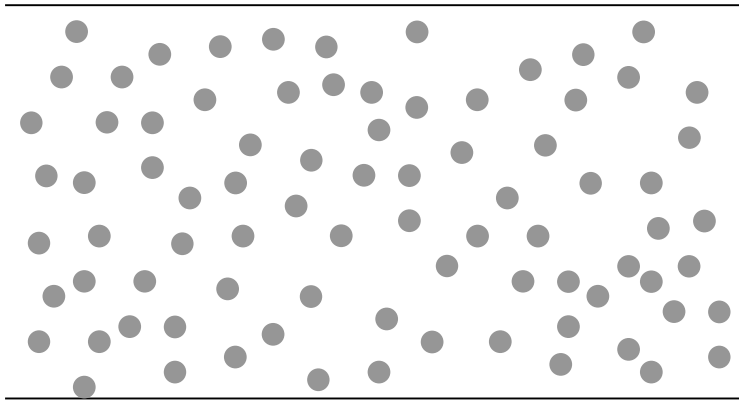
Migration in **viscous** noncolloidal suspensions

Leighton & Acrivos (1987), Phillips et al. (1992), ...

Ex: pipe flow



Migration towards zones of low shear rate



Frank et al. (2003)

⇒ Need for a **diphasic description**

Shear-induced migration and normal stresses

suspension fluid particles

diphasic description: $\Sigma_{ij} = \sigma_{ij}^f + \sigma_{ij}^p$

Lhuillier (2009):
the relevant stress
driving migration is the
contact stress

stress equilibrium on the **particle phase**:

if $\nabla \sigma^p \neq \mathbf{0} \rightarrow$ fluid filtration ; $\nabla \sigma^p$ balanced by drag force

mass conservation \rightarrow kinetic equation for the particle volume fraction

Nott and Brady, JFM (1994)

Mills and Snabre, J. Phys. II (1995)

Morris and Boulay, J. Rheol. (1999)

Lhuillier, Phys. Fluids (2009)

Nott et al., Phys. Fluids (2011)

Experimental setup

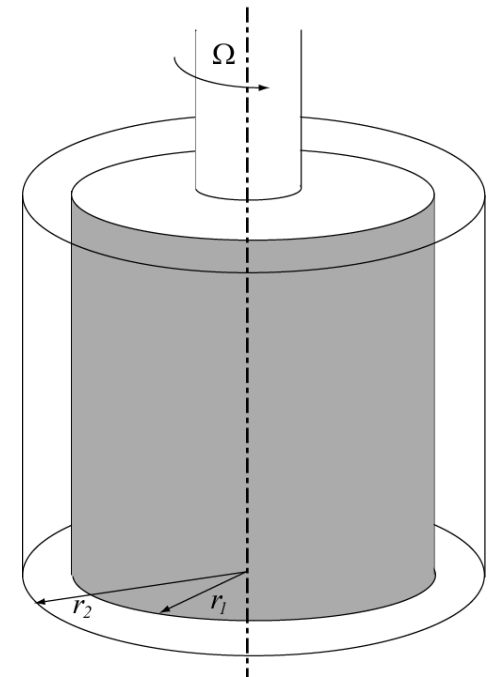
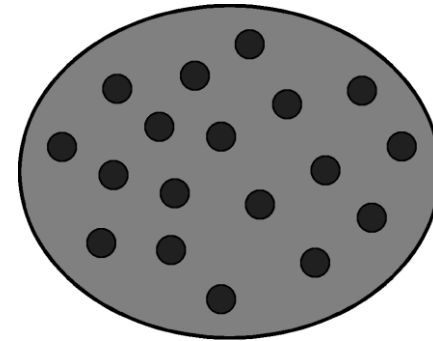
- PS beads : \varnothing 140 μm , $\rho = 1.05 \text{ g.cm}^{-3}$
 - PEG: $\eta = 2.15 \text{ Pa.s}$, $\rho = 1.05 \text{ g.cm}^{-3}$
- Volume fraction : from 5 to 56%

**Same system as Boyer et al. 2011
and Garland et al. 2013**

- macroscopic behavior fully characterized
- no free parameter for the model...

Geometry:

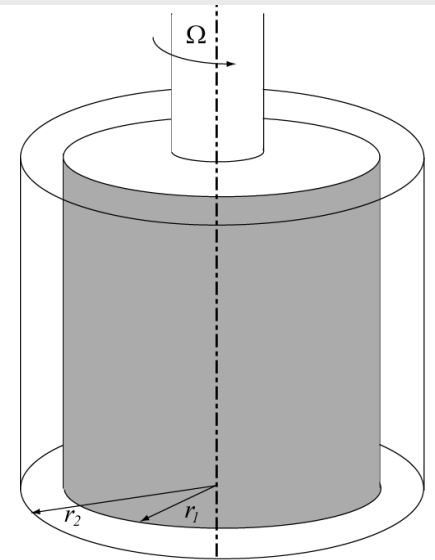
Wide gap Couette ($R_{\text{inner}}=3\text{cm}$, $R_{\text{outer}}=5\text{cm}$)



Experimental setup



MRI
Lab. Navier
Champs sur Marne

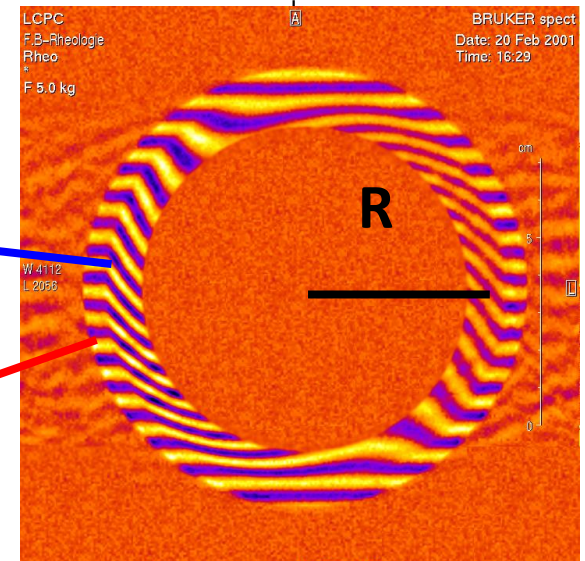


View from above

Torque measurement T

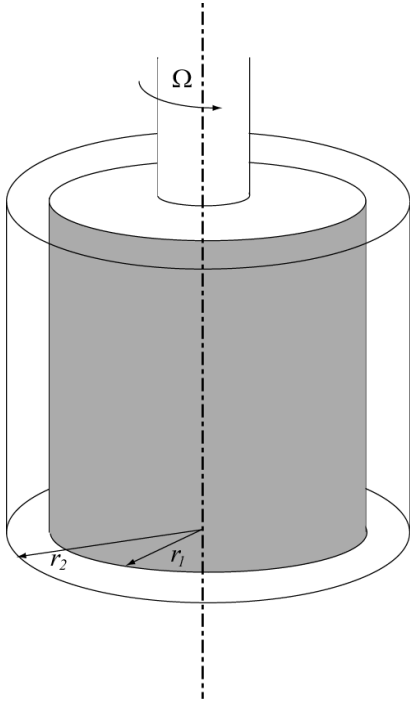
Velocity profile $V(R,t)$ measurement
→ kinetics

Volume fraction profile $\phi(R)$ measurement
→ stationary profile

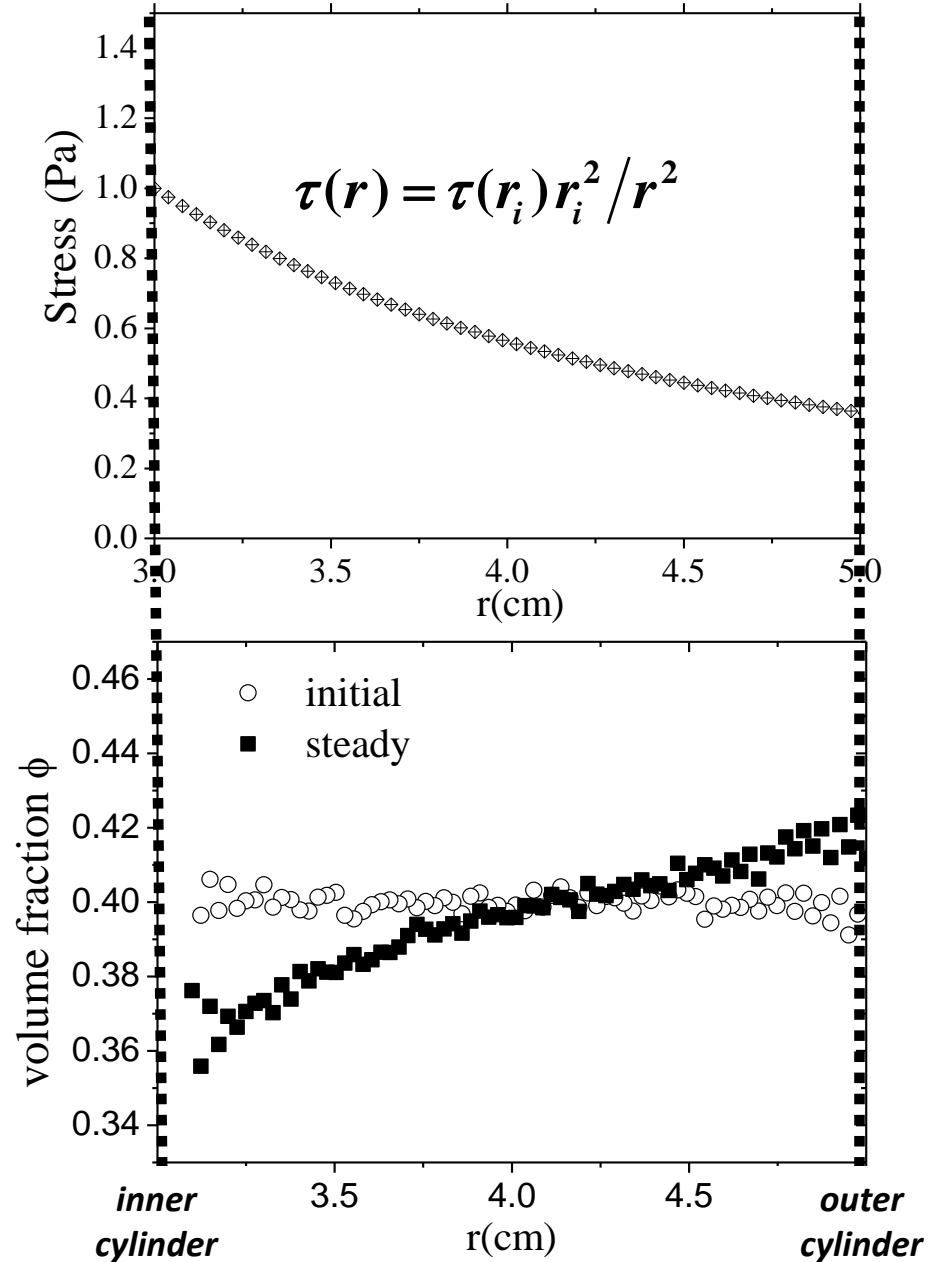


Steady-state: volume fraction

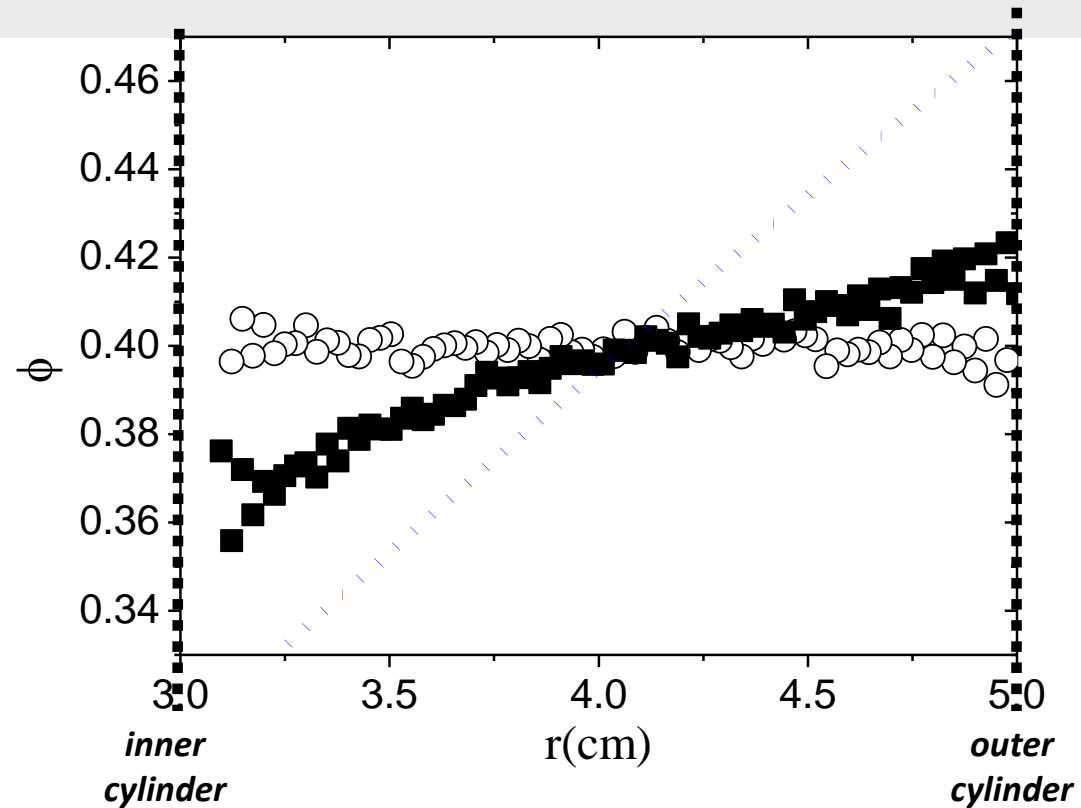
Ex : vol. fraction 40%



**Migration towards
zone of low shear rate**



Migration in dense viscous suspensions: steady-state



normal stresses:

$$\underline{\underline{\sigma}}_n^p = \begin{bmatrix} \lambda_1^p & 0 & 0 \\ 0 & \lambda_2^p & 0 \\ 0 & 0 & \lambda_3^p \end{bmatrix} \eta_n(\phi) \eta_0 \dot{\gamma}$$

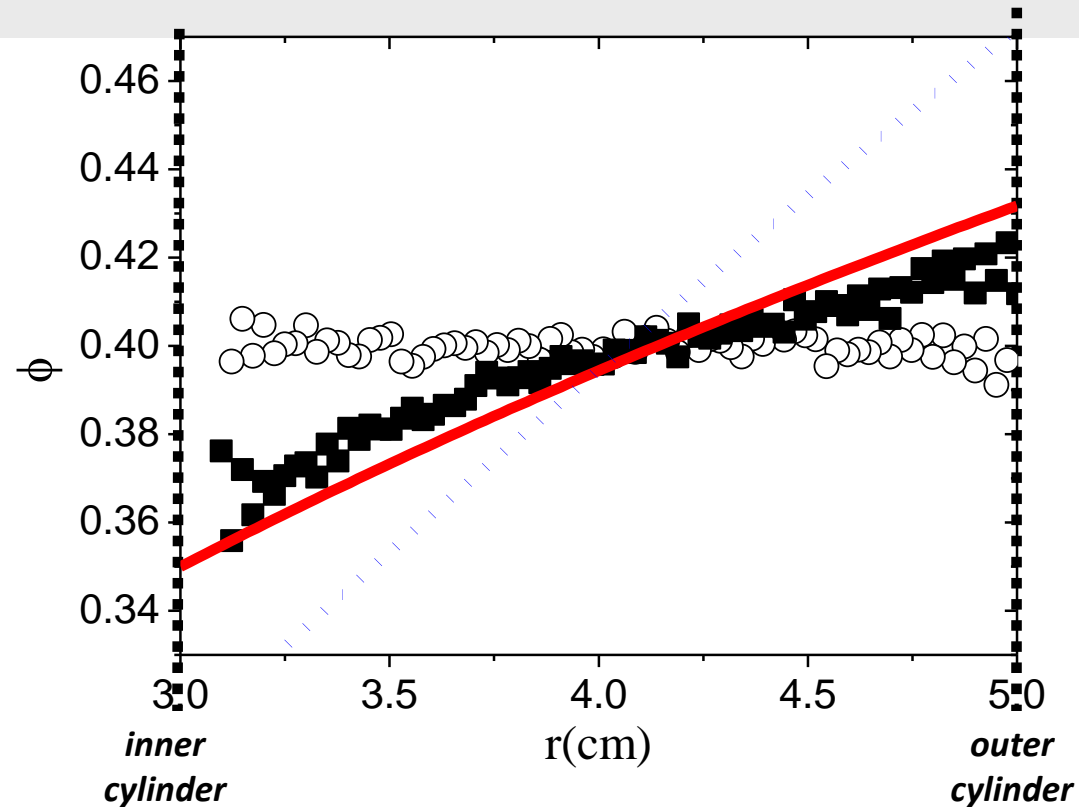
Recent measurements suggest

$$\lambda_1^p \approx \lambda_2^p \approx 1$$

Boyer et al. (2011), Dbouk et al. (2013)

Migration in dense viscous suspensions: steady-state

$$\lambda_2^p \gg \lambda_1^p$$



normal stresses:

$$\underline{\underline{\sigma}}_n^p = \begin{bmatrix} \lambda_1^p & 0 & 0 \\ 0 & \lambda_2^p & 0 \\ 0 & 0 & \lambda_3^p \end{bmatrix} \eta_n(\phi) \eta_0 \dot{\gamma}$$

Doubt on what the measured particle stresses are...

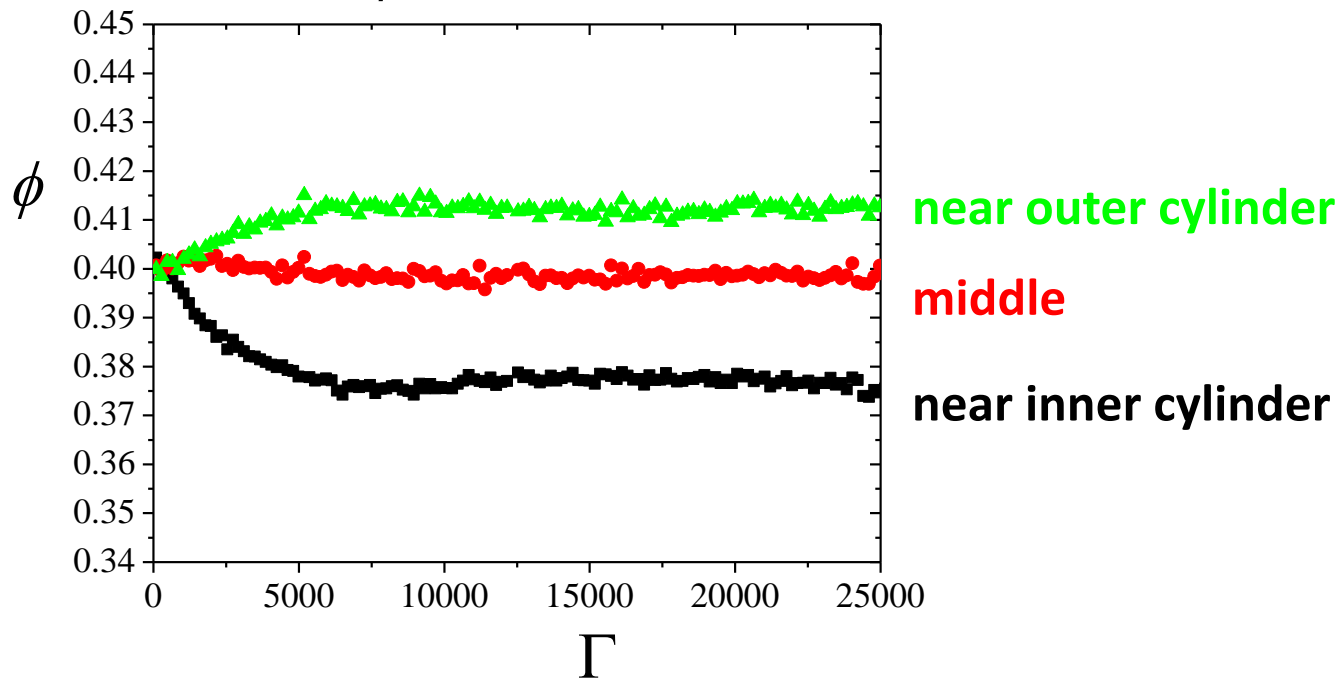
Recent measurements suggest

$$\lambda_1^p \approx \lambda_2^p \approx 1$$

Boyer et al. (2011), Dbouk et al. (2013)

Migration in dense viscous suspensions: kinetics

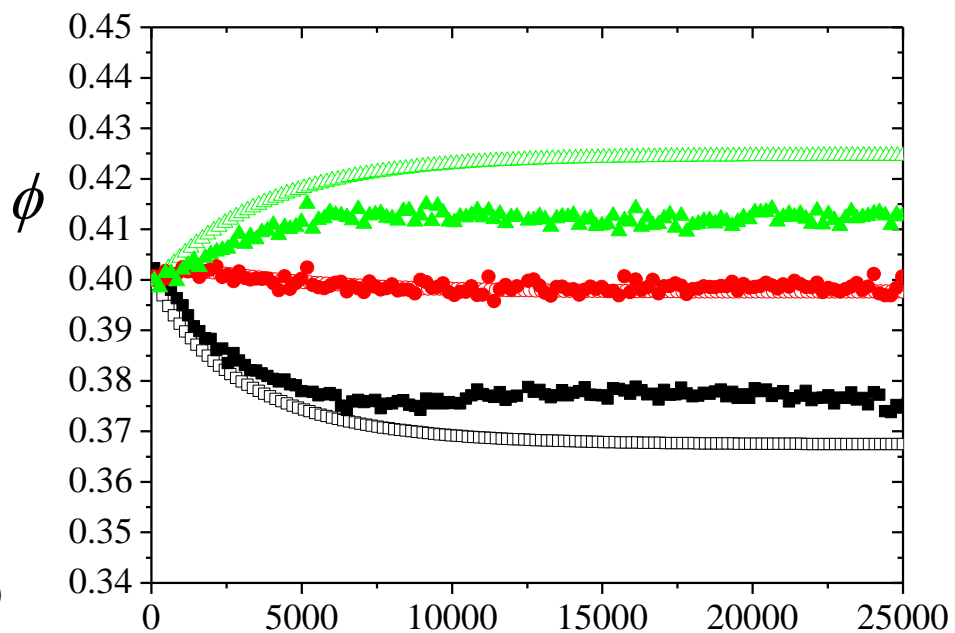
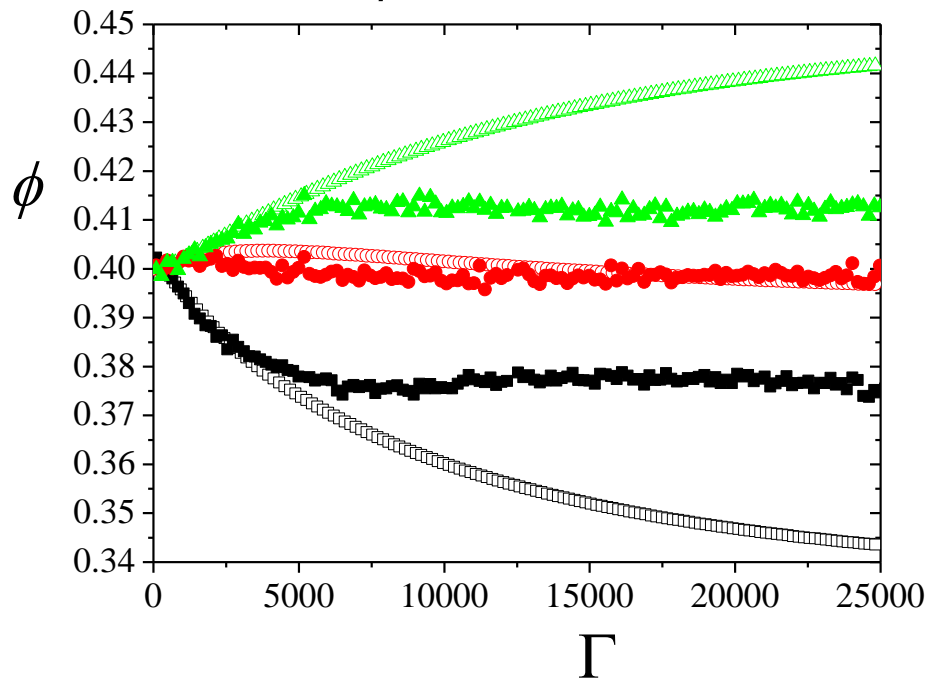
ϕ evolution in 3 different radial positions



Migration in dense viscous suspensions: kinetics

$$\frac{\partial \phi}{\partial t} = -\frac{2}{9} \frac{a^2}{\eta} \frac{1}{r} \frac{\partial}{\partial r} \left[f(\phi) \left(r \frac{\partial \sigma_{rr}^P}{\partial r} - (\sigma_{\theta\theta}^P - \sigma_{rr}^P) \right) \right] \quad \underline{\underline{\sigma}}_n^P = \begin{bmatrix} \lambda_1^P & 0 & 0 \\ 0 & \lambda_2^P & 0 \\ 0 & 0 & \lambda_3^P \end{bmatrix} \eta_n(\phi) \eta_0 \dot{\gamma}$$

ϕ evolution in 3 different radial positions



suggested values (Boyer et al. 2011)

$$\lambda_1^P \approx \lambda_2^P \approx 1$$

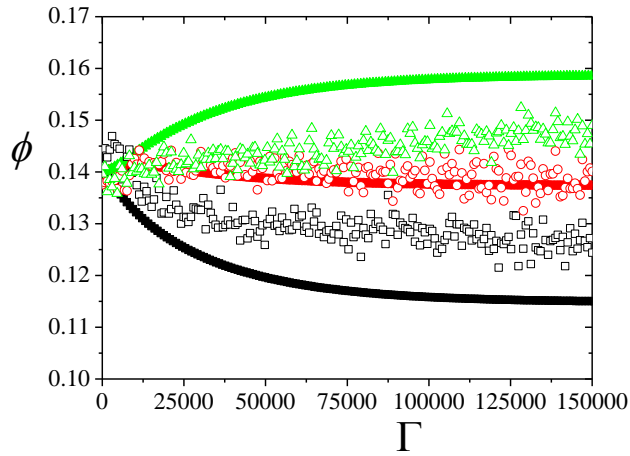
Possible values ?

$$\lambda_2^P \gg \lambda_1^P$$

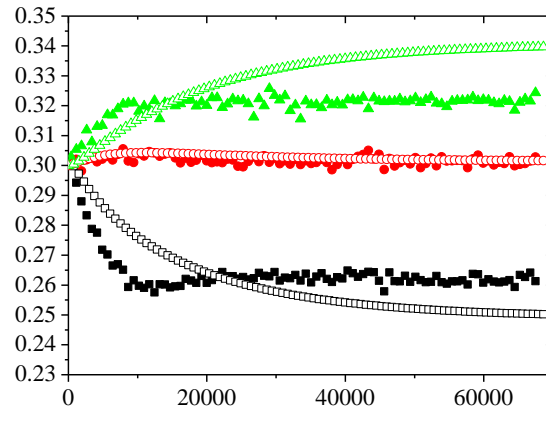
$$\lambda_2^P \sim 3-5$$

Other volume fractions: kinetics

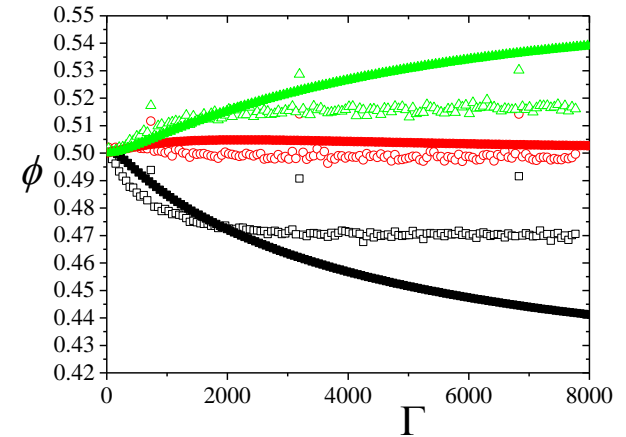
model with $\lambda_1^P \approx \lambda_2^P \approx 1$



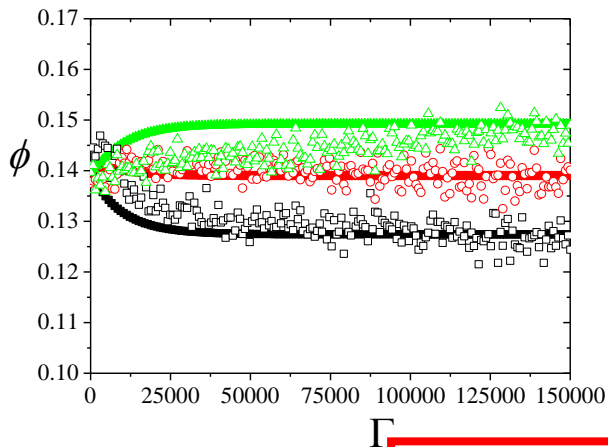
14%



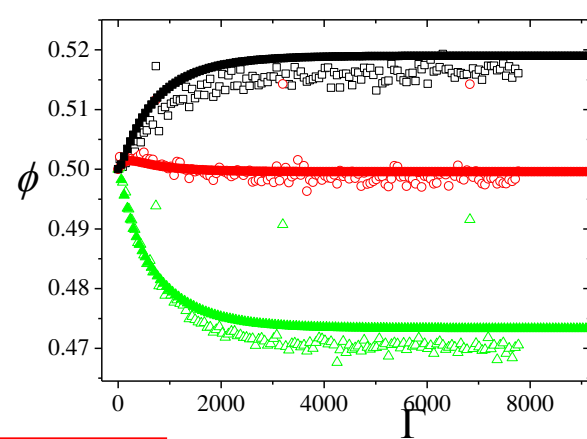
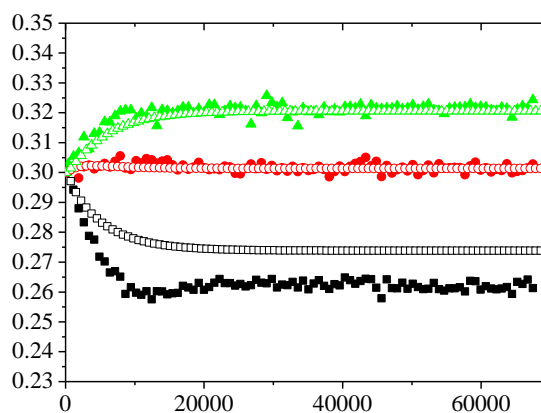
30%



50%



model with $\lambda_2^P \gg \lambda_1^P$, $\lambda_2^P \sim 3-5$



Other possibility: interphase force with gradients

suspension fluid particles

$$\sum_{ij} = \sigma_{ij}^f + \sigma_{ij}^p$$

diphasic description:

Balance equations for the particle phase

$$\frac{\partial \phi}{\partial t} + \nabla \cdot (\phi \langle \mathbf{u} \rangle^p) = 0,$$

$$\nabla \cdot \langle \boldsymbol{\sigma} \rangle^p + \mathbf{F}^{\text{drag}} = 0,$$

Inter-phase drag force

$$\mathbf{F}^{\text{drag}} \approx -\frac{9\eta_f}{2a^2} \frac{\phi}{f(\phi)} (\langle \mathbf{u} \rangle^p - \mathbf{u})$$

Migration equation

$$\begin{aligned} \frac{\partial \phi}{\partial t} + \mathbf{u} \cdot \nabla \phi &= -\nabla \cdot [\phi (\langle \mathbf{u} \rangle^p - \mathbf{u})] \\ &= -\frac{2a^2}{9\eta_f} \nabla \cdot [f(\phi) \nabla \cdot \langle \boldsymbol{\sigma} \rangle^p] \end{aligned}$$

Additional terms of interphase force in inhomogeneous field :

$$-\eta_f \phi \dot{\gamma} D_{ij} \frac{\partial \phi}{\partial x_j} + \eta_f \phi B_{ijkl} \frac{\partial^2 U_l}{\partial x_j \partial x_k}$$

Lhuillier, Phys. Fluids (2009)

Nott and Brady, JFM (1994)

Mills and Snabre, J. Phys. II (1995)

Morris and Boulay, J. Rheol. (1999)

Lhuillier, Phys. Fluids (2009)

Nott et al., Phys. Fluids (2011)

Other possibility: interphase force with gradients

Accounting for vol.
frac. inhomogeneity
in interphase force

$$- \eta_f \phi \dot{\gamma} D_{ij} \frac{\partial \phi}{\partial x_j}$$

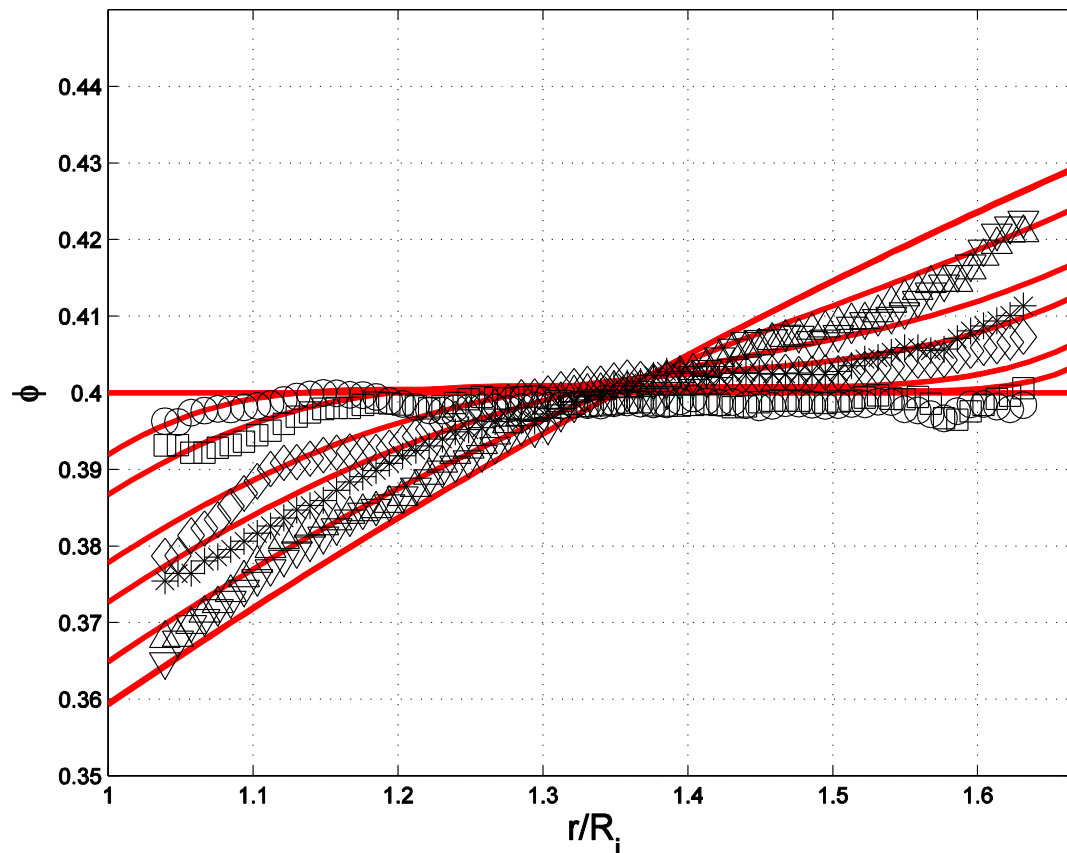
Lhuillier, *Phys. Fluids* (2009)

A **single (free...) parameter**

→ Decrease of migration magnitude

→ Increase of kinetics

Evolution of ϕ in the gap



Similar to adding a stress
to the contact stress

Migration in a Couette cell: a complex 2D problem?

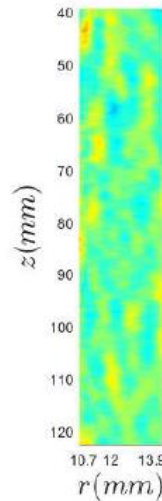
with A. Rashedi & S. Hormozi

Time evolution of the 2D volume fraction maps in a wide gap Couette cell

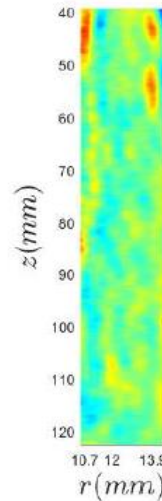
Height=25*gap

Unavoidable z-inhomogeneities due to imperfect boundary conditions ?

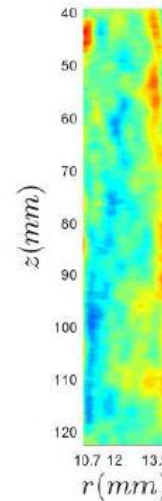
Impact on the evolution at long time only ?



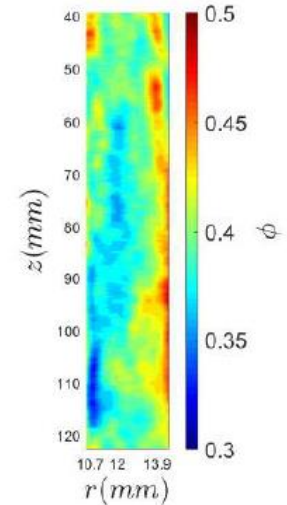
(a) $t=157s$



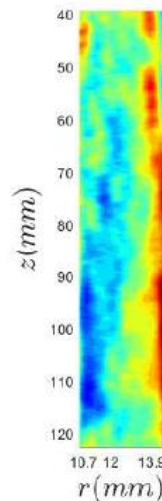
(b) $t=232s$



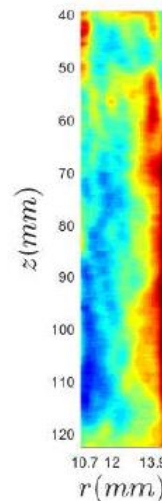
(c) $t=402s$



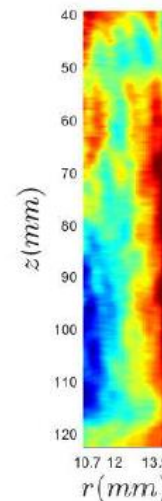
(d) $t=602s$



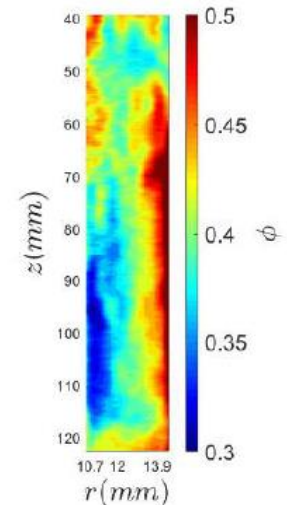
(e) $t=877s$



(f) $t=2877s$



(g) $t=8377s$



(h) $t=10627s$

Shear-induced migration: strainscale

For a viscous suspension,

$$\text{strainscale} = \frac{(\text{gap})^2}{(\text{particle size})^2} f(\phi)$$

ϕ / ϕ_m	strainscale
-----------------	-------------

0.15	100 000
------	---------

0.5	10 000
-----	--------

0.85	2 000
------	-------

0.96	50
------	----

case investigated:

100 particles in the gap \rightarrow strainscale = 10000 $f(\phi)$

**Strain scale seems to decrease down to 0 near jamming
 \rightarrow unavoidable...**

Nonlinear suspension: strainscale = $f(\phi, \dot{\gamma})$

Migration in viscous suspensions : Conclusion

- steady-state : migration **less important** than expected from model
- **kinetics: migration much faster** than expected from model
- Theory: **crucial role of contact stress**
- Experiments: is $\sigma_{22}^p \gg \sigma_{11}^p$???
- Possible **role of density gradients in the hydrodynamic force?**

Strain scale seems to decrease down to 0 near jamming

→ what about small particles? In the shear-thickening regime?

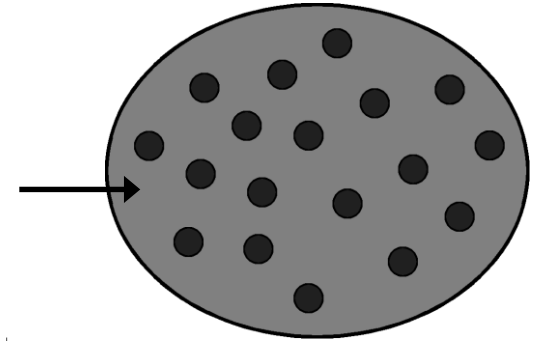
Shear-induced migration in viscous suspensions: test of model

**Shear-induced migration in shear-thickening suspensions:
(1) Role of interparticle contacts**

**Shear-induced migration in shear-thickening suspensions
(2) Jamming**

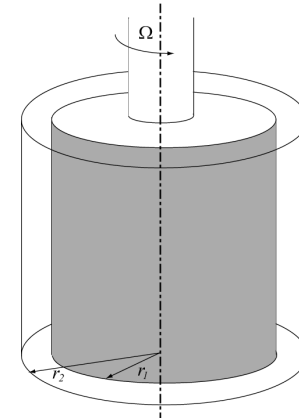
A shear-thickening dense suspension

- *Polystyrene beads* : ϕ **40 μm** , $\rho = 1.05 \text{ g.cm}^{-3}$
- *Water + Cesium chloride*: $\rho = 1.05 \text{ g.cm}^{-3}$, $\eta = 1 \text{ mPa.s}$
- $\phi = 56$ à 60%



Geometry:

Wide gap Couette ($R_{\text{inner}}=4\text{cm}$, $R_{\text{outer}}=6\text{cm}$)



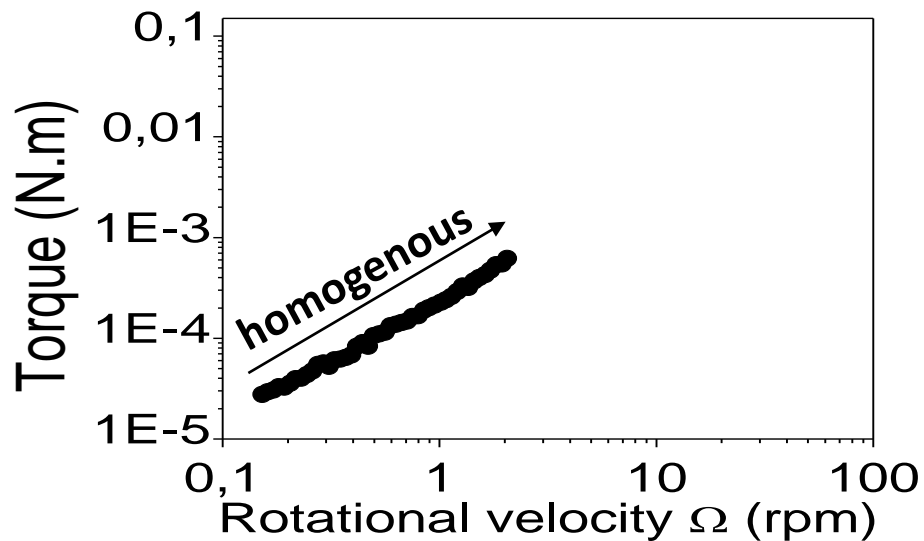
Migration ?

$$\text{strainscale} \propto \frac{(\text{gap})^2}{(\text{particle size})^2} \times f(\phi)$$

→ strainscale expected ~ 1000

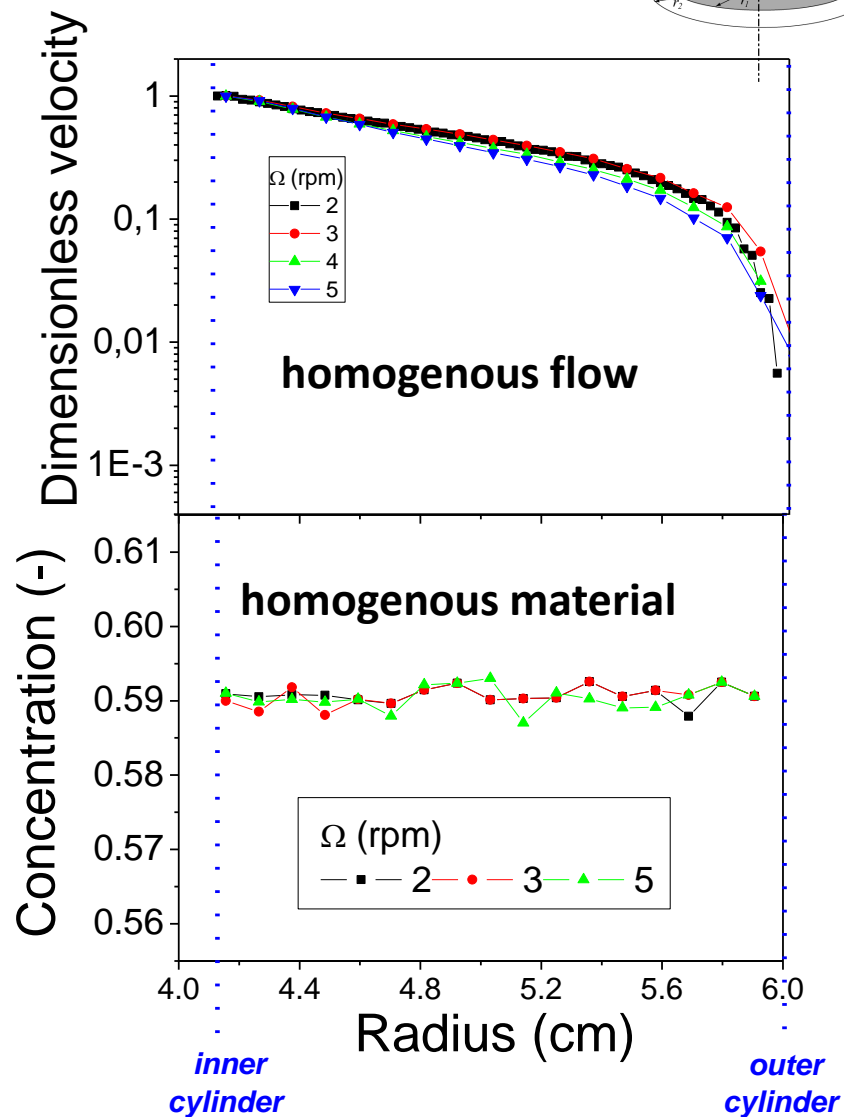
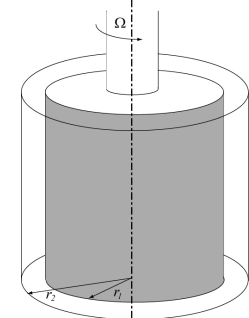
Macroscopic response

when increasing rotational velocity



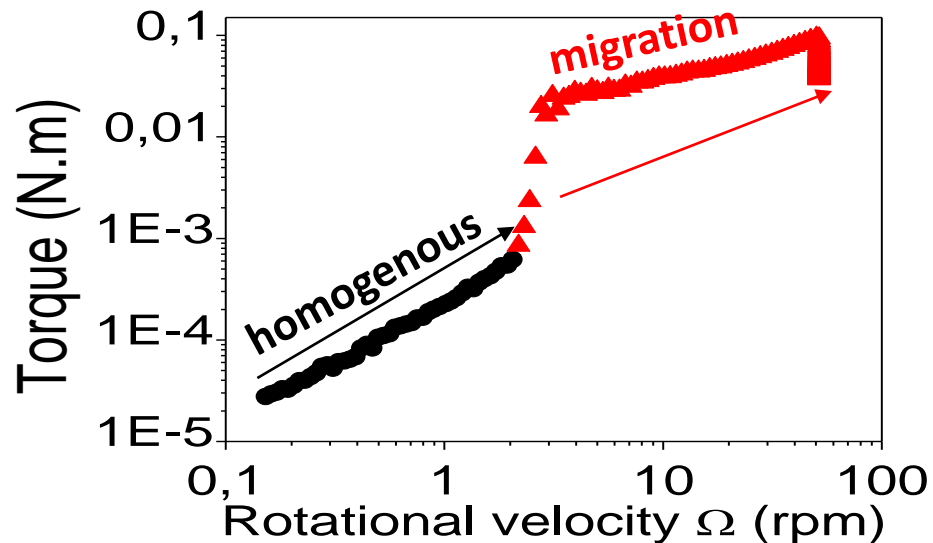
Viscous response of the homogeneous material

Local behavior



Macroscopic response

when increasing rotational velocity

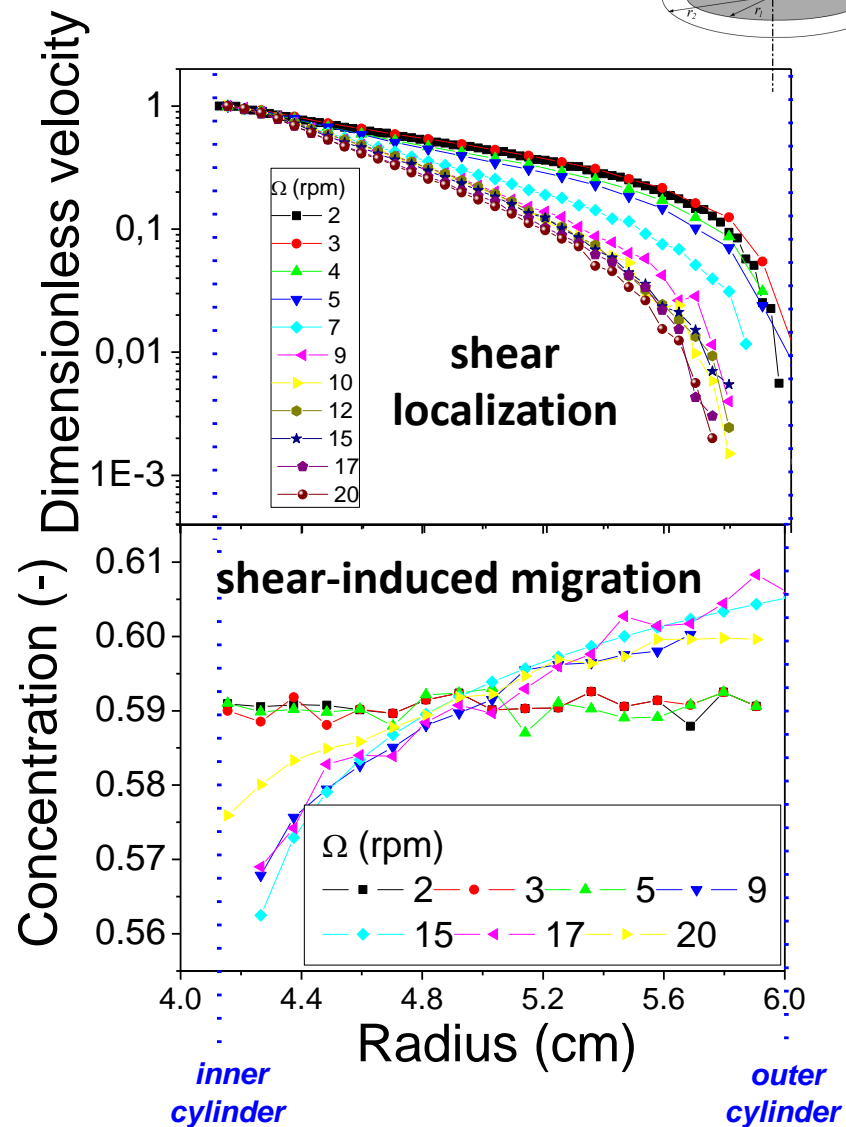
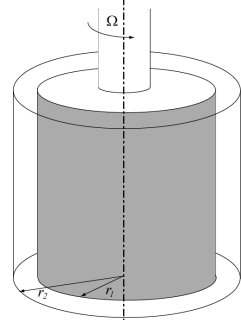


Discontinuous shear-thickening:
transient response linked to migration

Strainscale $\ll 1000$

Consistent with the microscopic picture proposed by Seto, Mari, Wyart... (sudden emergence of contacts) but **what is the true local behavior?**

Local behavior

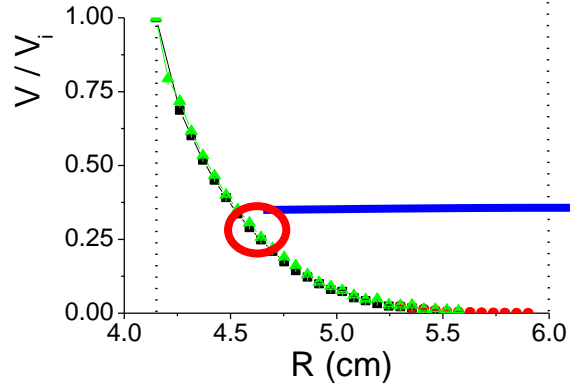


Local measurements of the intrinsic behavior

Ovarlez et al., J. Rheol. 50, 259-292 (2006)

Fall et al. (2010)

Velocity profiles $\Rightarrow \dot{\gamma}(R, \Omega)$



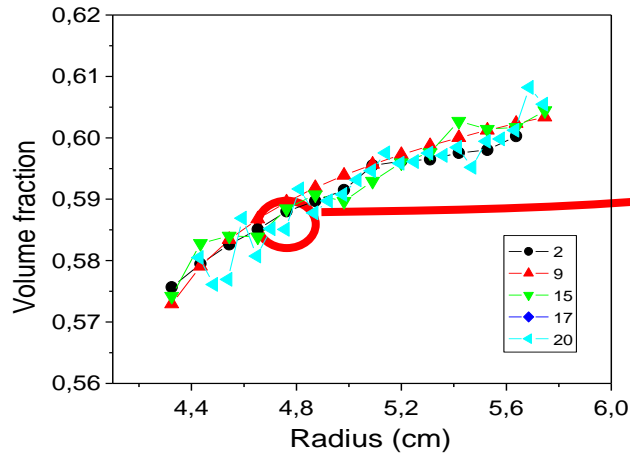
Local behavior at constant volume fraction 59%

Torque $\Rightarrow \tau(R, \Omega) = \frac{T(\Omega)}{2\pi HR^2}$

$\tau(\varphi(R), \dot{\gamma}(R))$

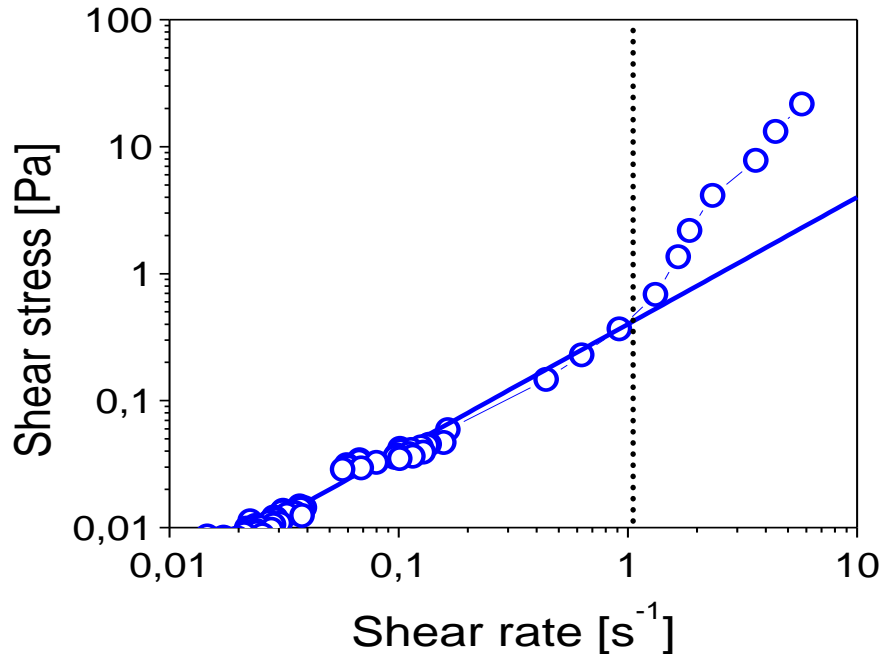
Concentration profiles

$\varphi = f(R, \Omega) \Rightarrow R \leftrightarrow \varphi$



Continuous shear-thickening: viscous \rightarrow granular transition

Local measurement at constant volume fraction (59%)



$$\tau \propto \dot{\gamma} \quad \longrightarrow \quad \tau \propto \dot{\gamma}^2$$

Viscous

« Granular »

Viscous forces + Contact forces ;
negligible inertia

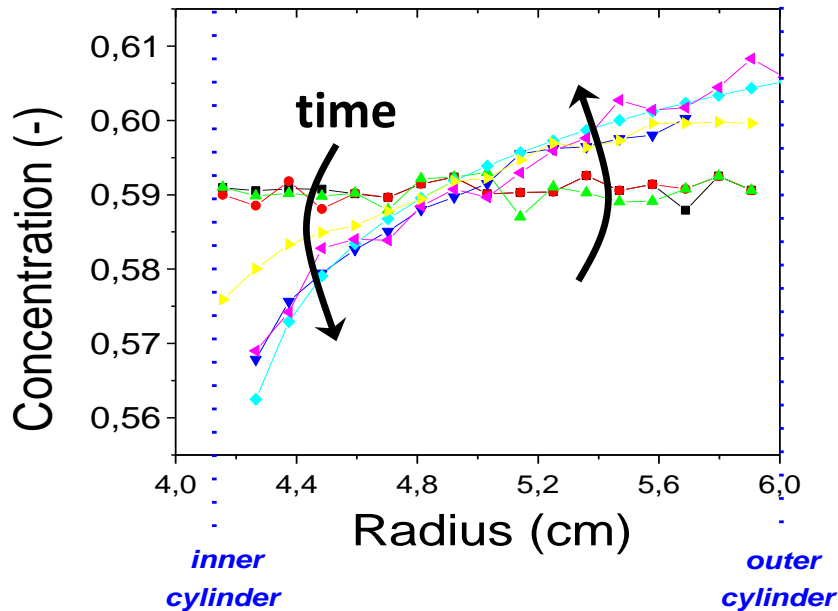
Contact forces + Grains inertia;
negligible viscous forces

$$\mathbf{0} = \sum_j \mathbf{F}_{ij} + \mathbf{F}_i^{visc} \Rightarrow \mathbf{F}_{ij} \propto \dot{\gamma}$$

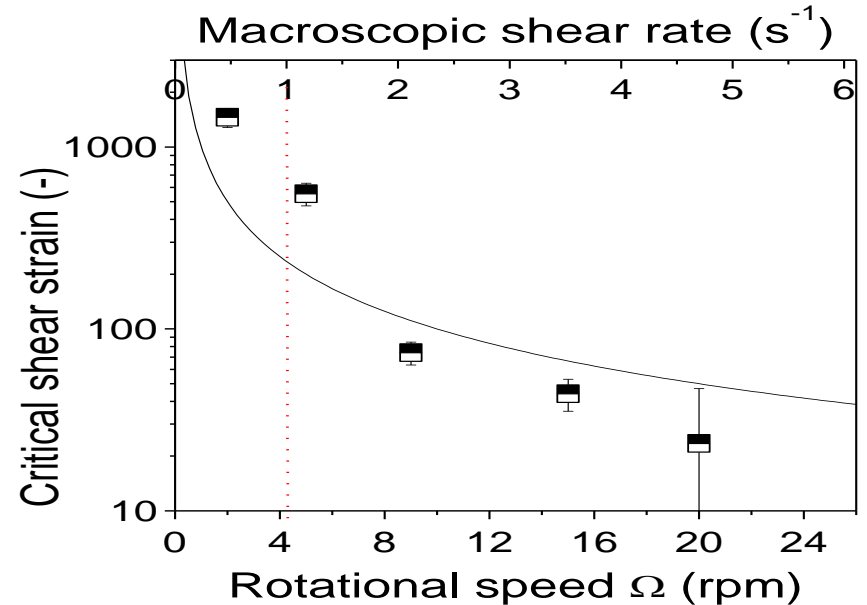
$$m \frac{d^2 \mathbf{r}_i}{dt^2} = \sum_j \mathbf{F}_{ij} \Rightarrow \mathbf{F}_{ij} \propto \dot{\gamma}^2$$

Kinetics of migration

59% suspension of 40 microns beads



Strain at the end of migration

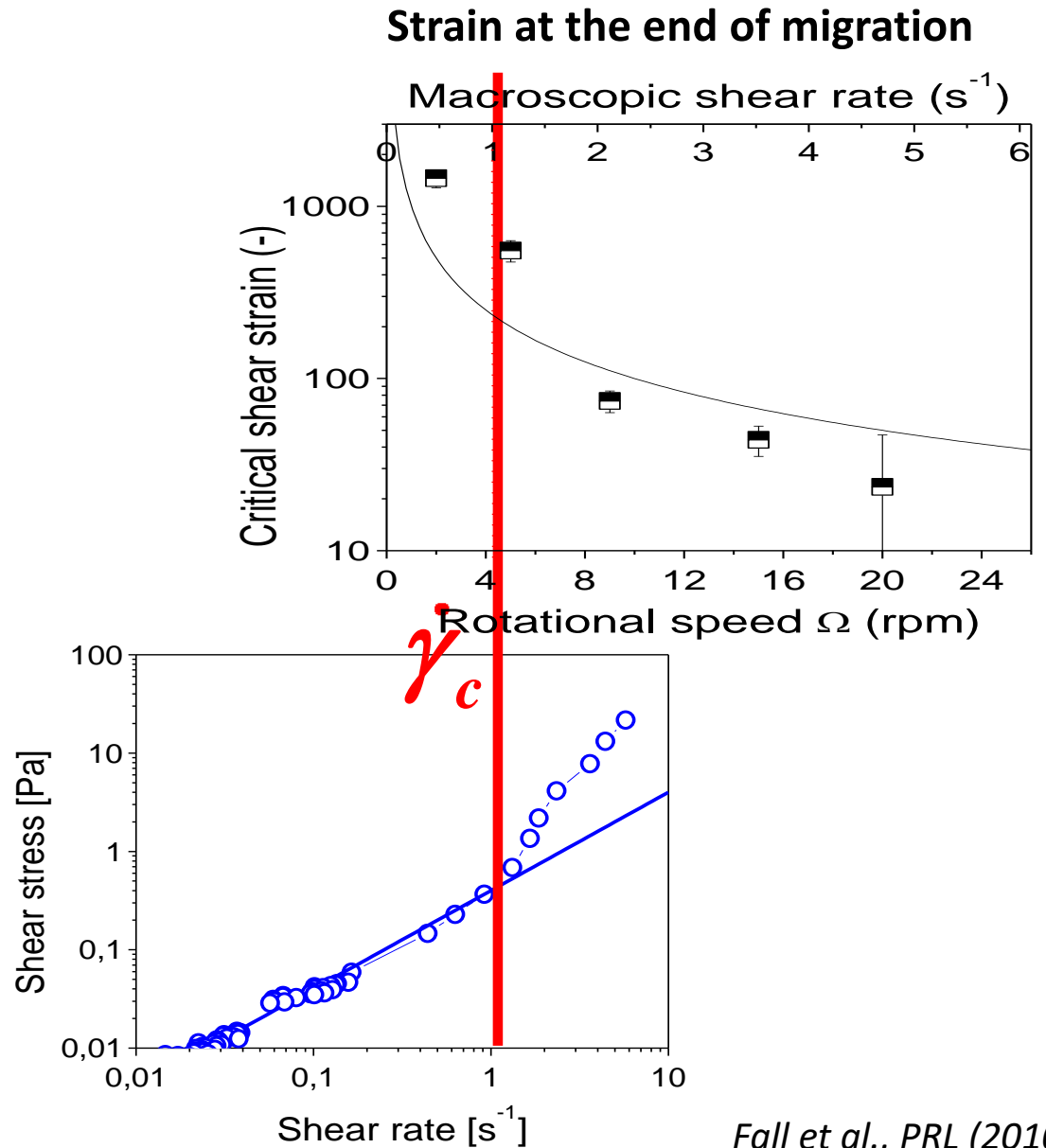


- ✓ **Slow** at low shear rate (strain ~ 1000)
- ✓ **"Instantaneous"** at high shear rate (strain < 50)

~~Theory for viscous suspensions~~

- ~~• timescale = $1/\dot{\gamma}$ \rightarrow **Unique strainscale**~~
- ~~• strainscale expected ~ 1000~~

Interplay between continuous shear-thickening / migration



Interplay between continuous shear-thickening / migration

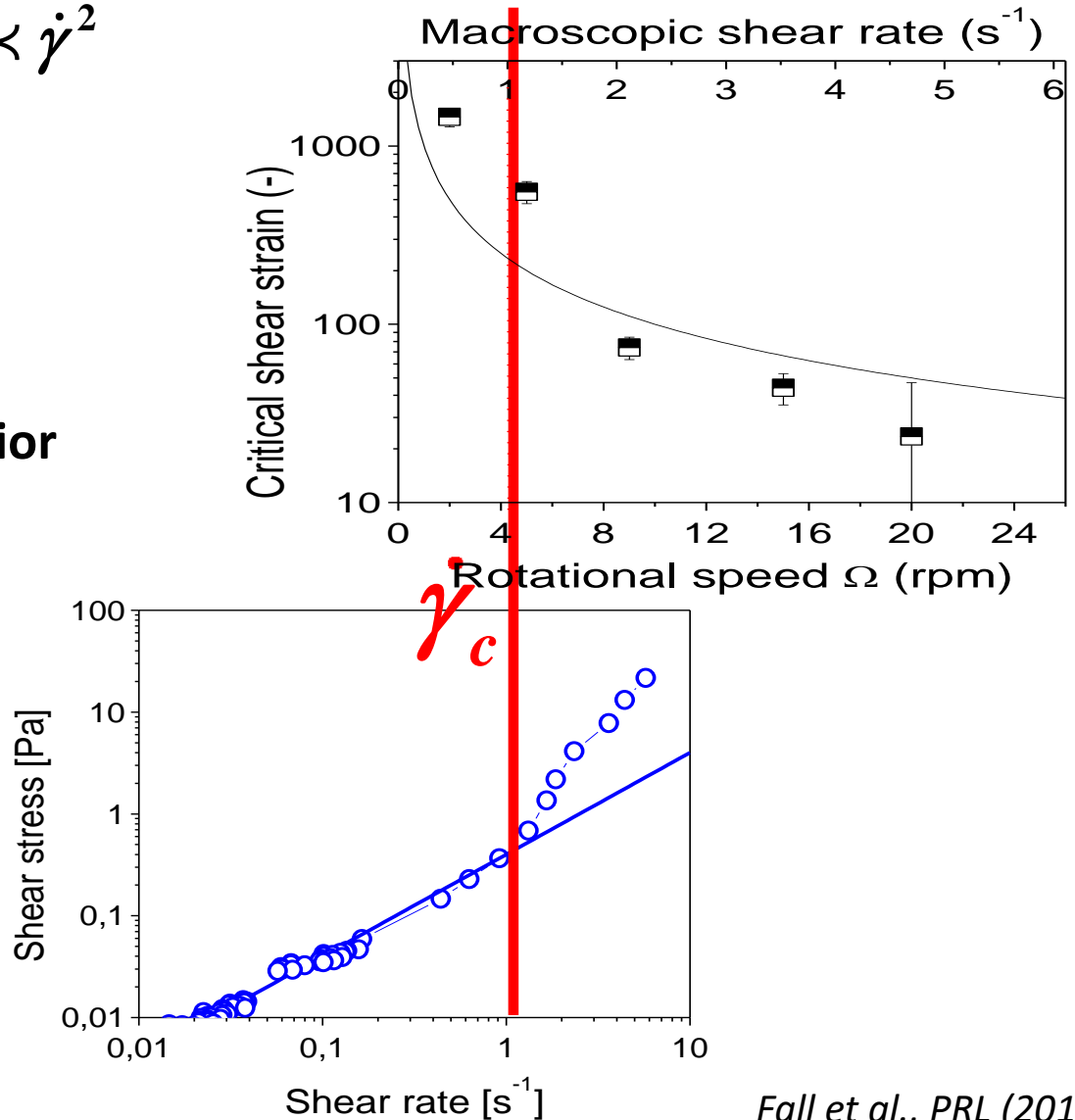
Normal stresses: $\sigma_{11}^p, \sigma_{22}^p \propto \dot{\gamma}^2$

⇒ **Acceleration** of migration

Strainscale $\propto 1/\dot{\gamma}$

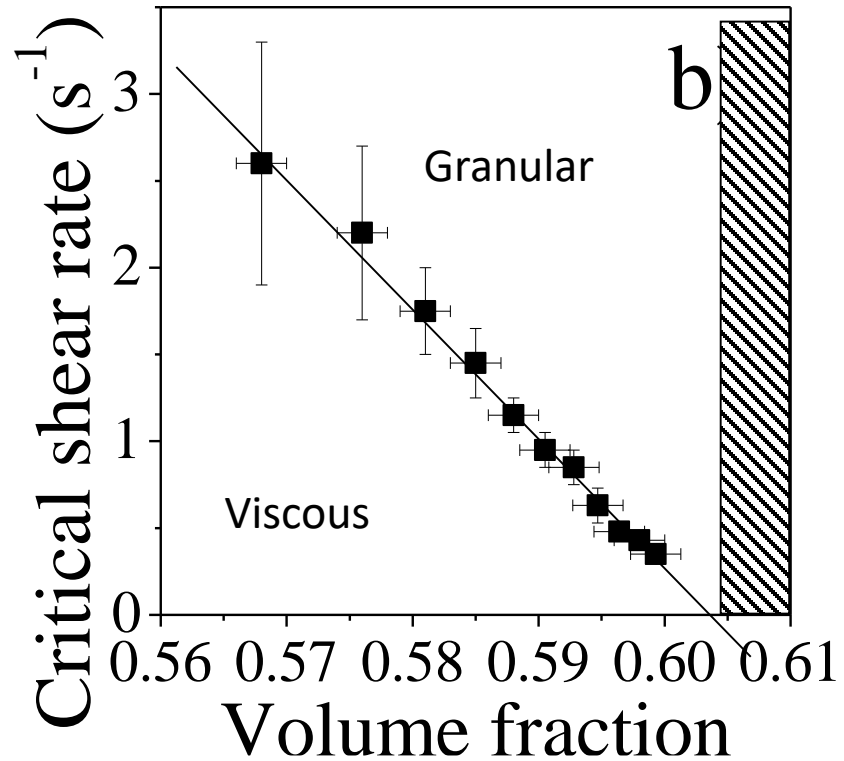
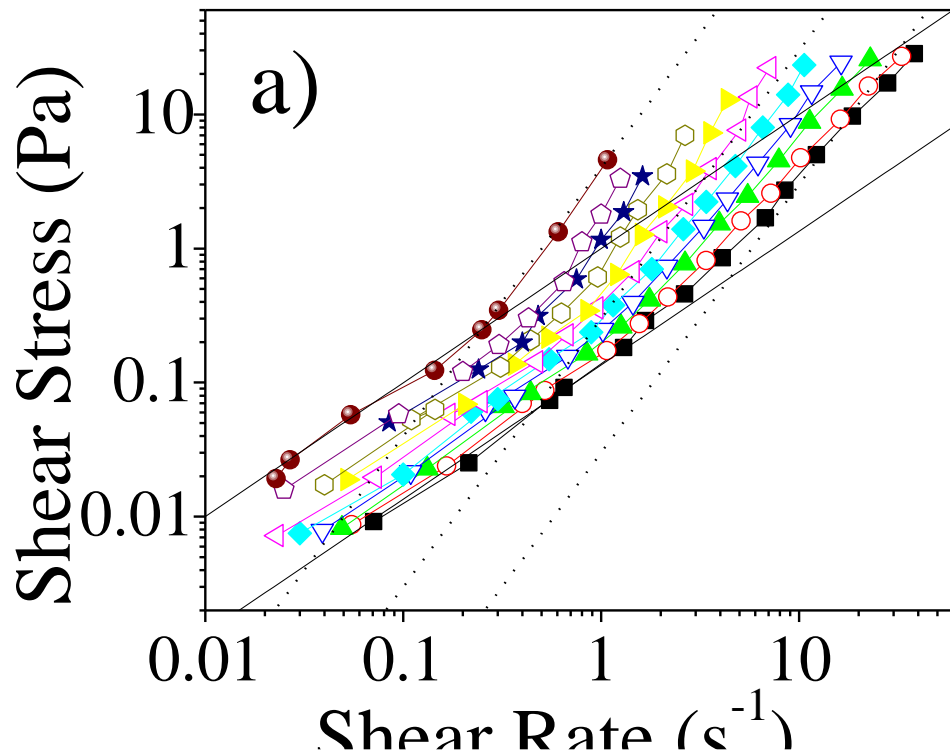
Signature of “granular” behavior

Strain at the end of migration



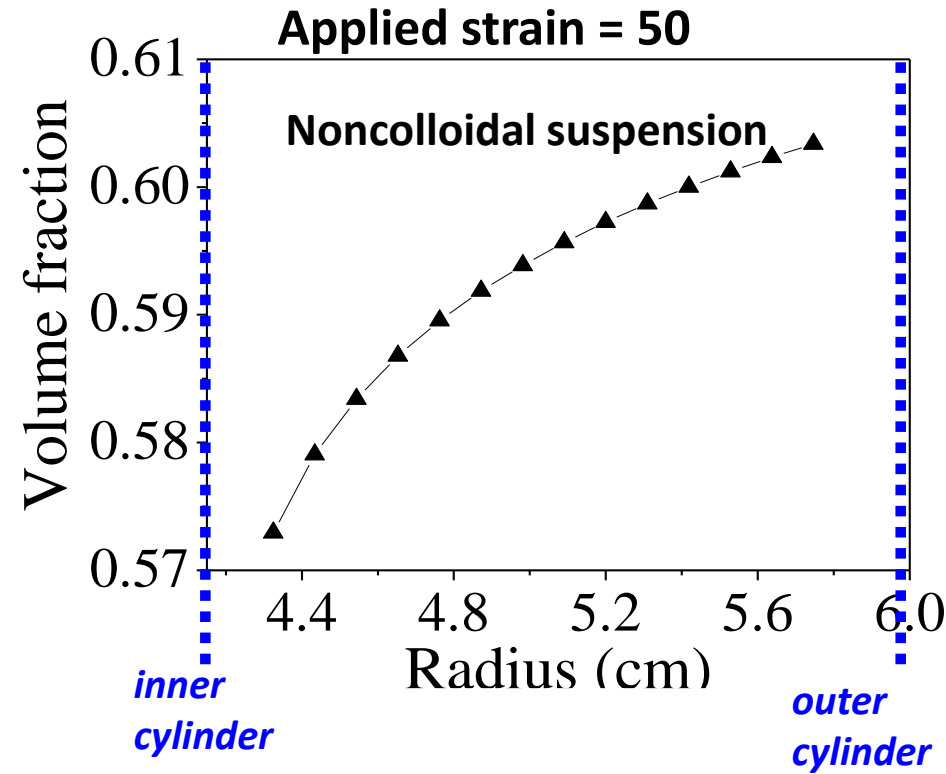
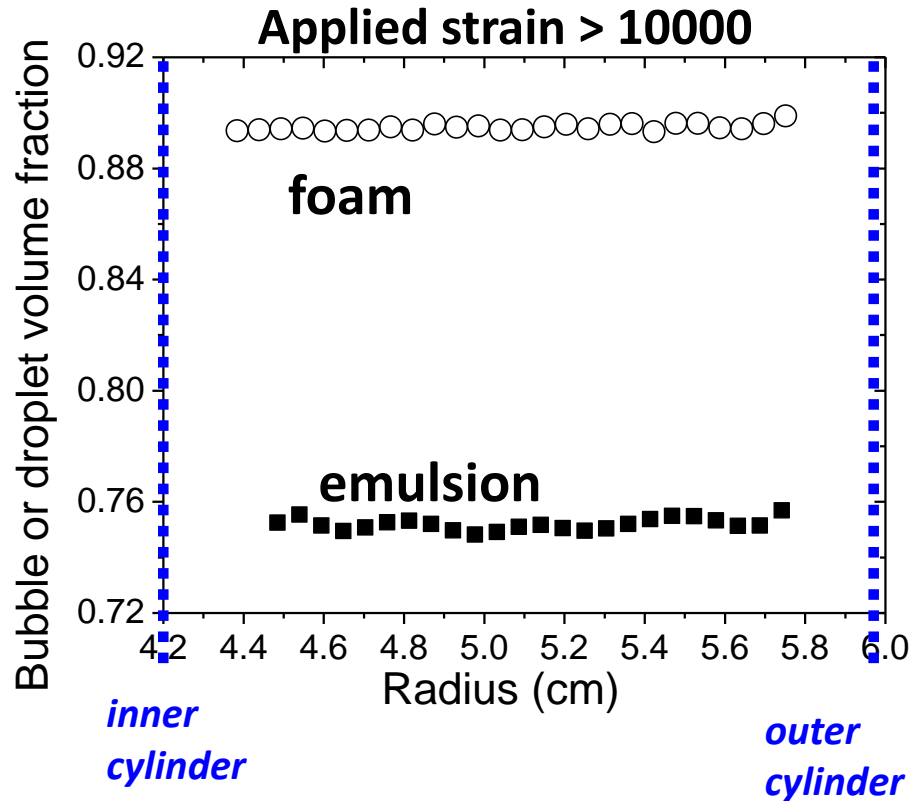
Shear-thickening: viscous \rightarrow granular transition

Vanishing of critical shear rate at jamming; similar to other thickening systems



$$\left. \begin{array}{l} \text{viscous regime: } \Sigma_V(\phi) \propto (\phi_m - \phi)^{-1} \\ \text{granular regime: } \Sigma_I(\phi) \propto (\phi_m - \phi)^{-2} \end{array} \right\} \text{transition: } \dot{\gamma}_c(\phi) \propto (\phi_m - \phi)$$

Dense suspensions of **soft** particles: inhomogeneities?



Ovarlez et al., PRE 2008

Suspensions of soft particles remain homogeneous

→ no flow/concentration coupling mechanism

Consistent with diphasic model of Lhuillier (Phys. Fluids 2009):
migration driven by interparticle forces.

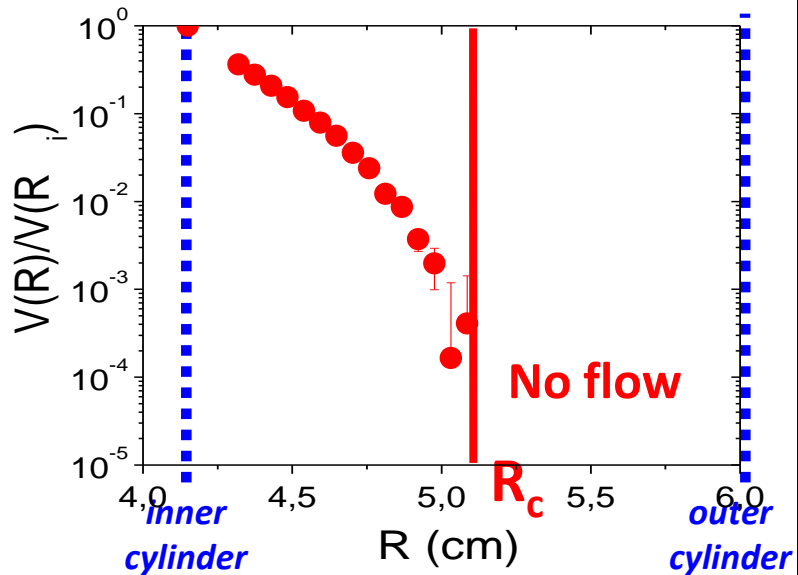
Shear-induced migration in viscous suspensions: test of model

**Shear-induced migration in shear-thickening suspensions:
(1) Role of interparticle contacts**

**Shear-induced migration in shear-thickening suspensions
(2) Jamming**

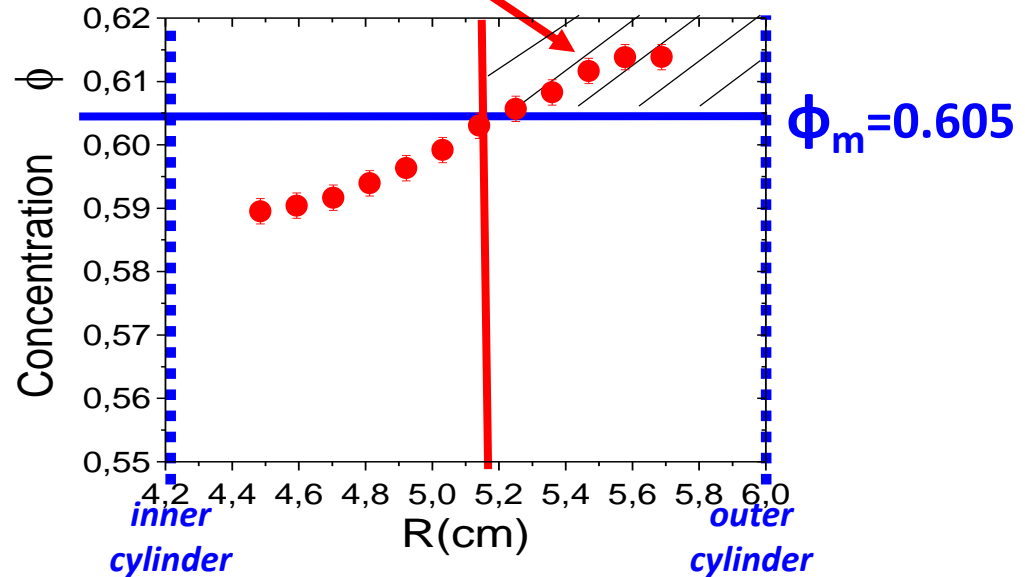
Shear induced migration and Shear induced jamming

Velocity profile in dense suspensions
of mean concentration 60%



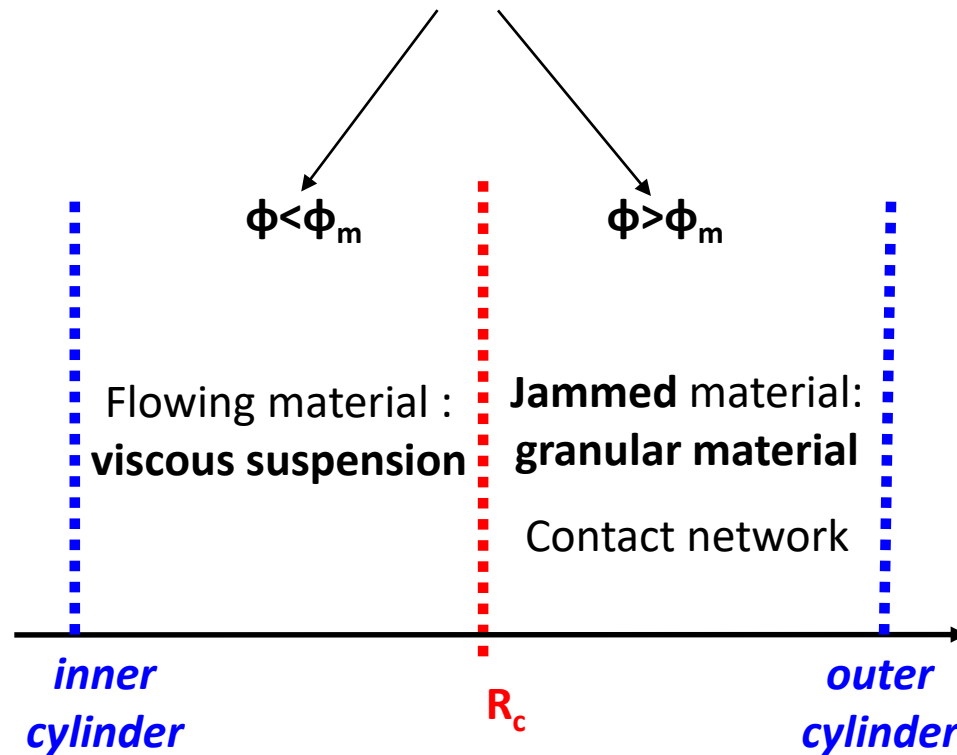
Shear localization at high shear rates:
no flow allowed beyond a critical radius R_c

mean concentration 60%,
local concentration $> \phi_m$
no flow allowed



Shear induced migration and Shear induced jamming

Shear-induced migration generates 2 different zones



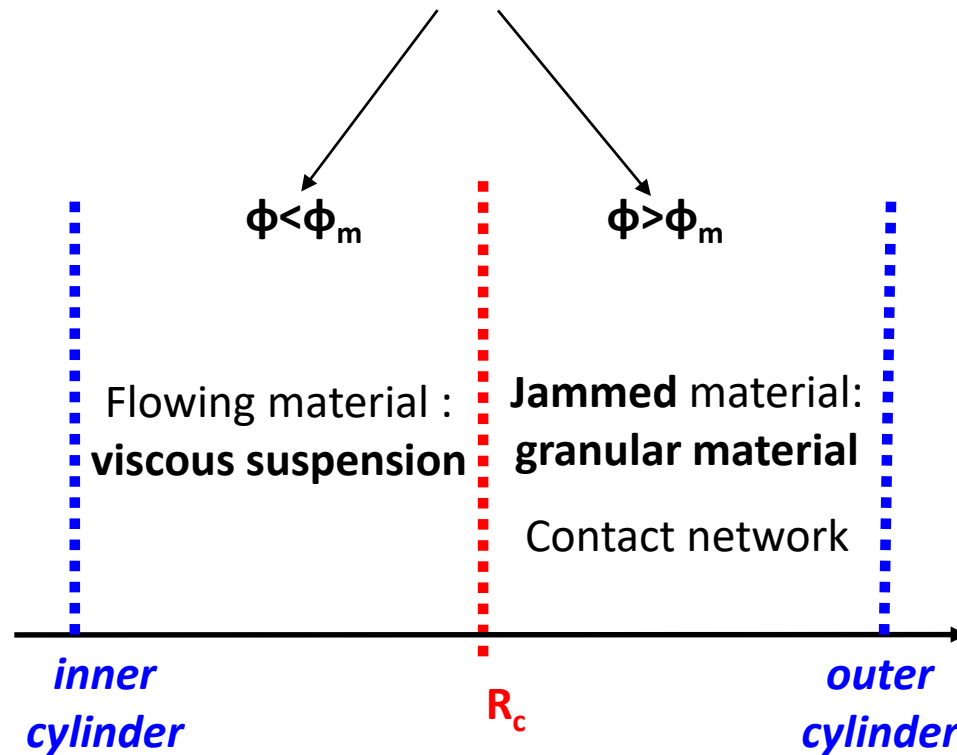
Explains why one can find viscosity measurements above 60% in the literature
(the concentration in the flowing region may be much lower than the mean concentration)

Ovarlez et al., J. Rheol. (2006)

Fall et al., PRL (2010)

Shear induced migration and Shear induced jamming

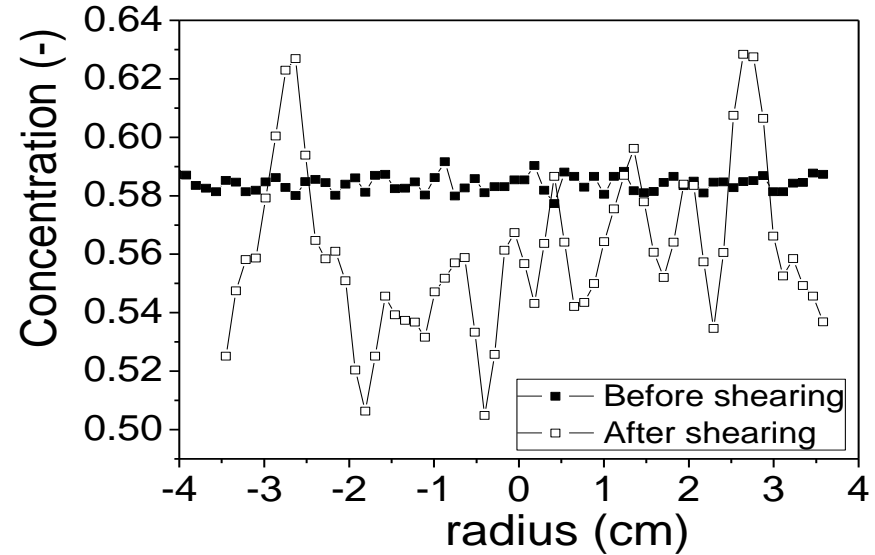
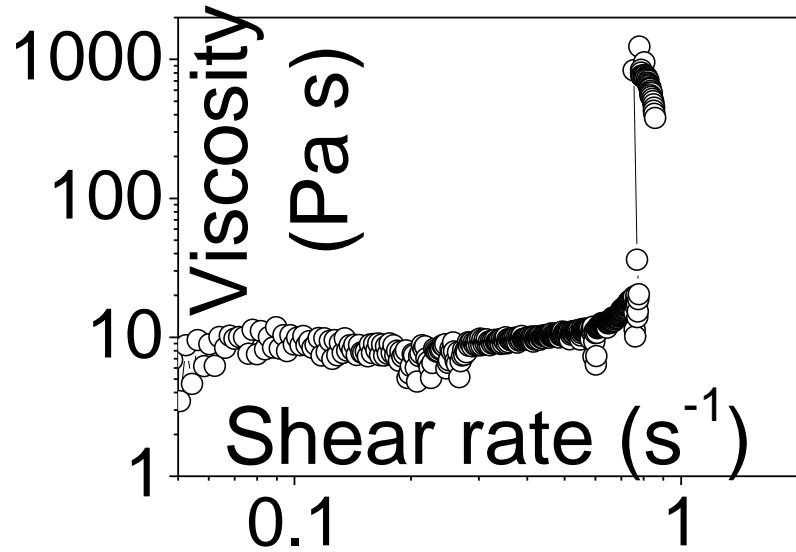
Shear-induced migration generates 2 different zones



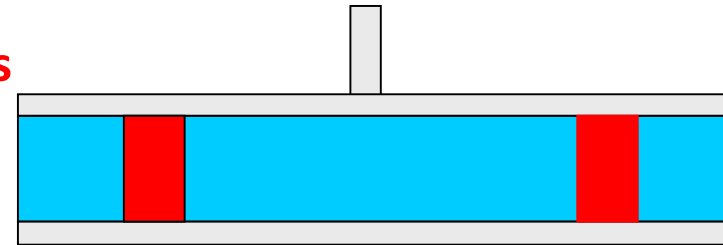
A diphasic model accounting for the granular regime is necessary

→ see Lecampion, Garagash, JFM (2014) !

Shear-induced jamming due to density changes



Friction of **packs of jammed dense granular materials** between the two plate surfaces leads to **apparent viscosity jump**

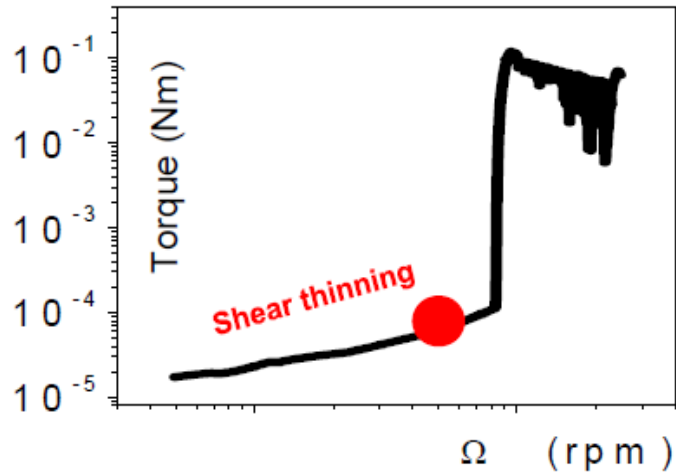


→ Not to be interpreted as an intrinsic behavior !

→ Same thing happens during a transient in the Couette cell?

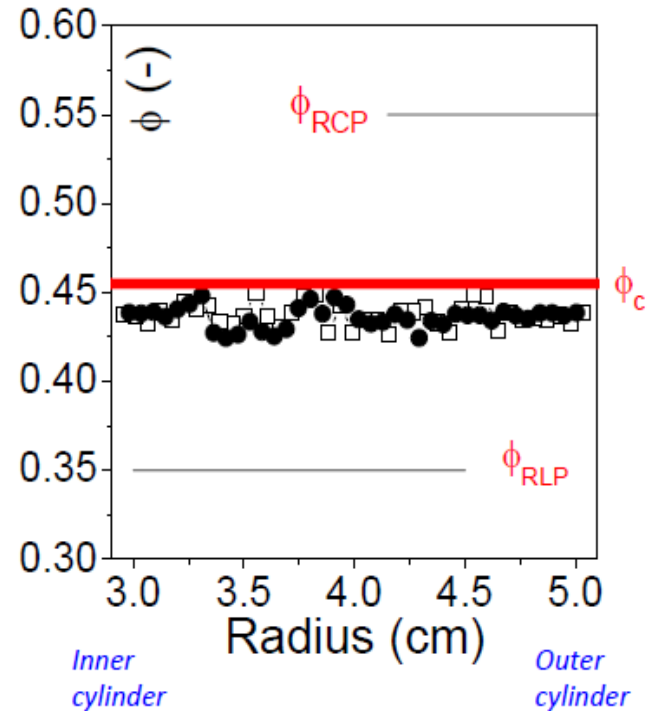
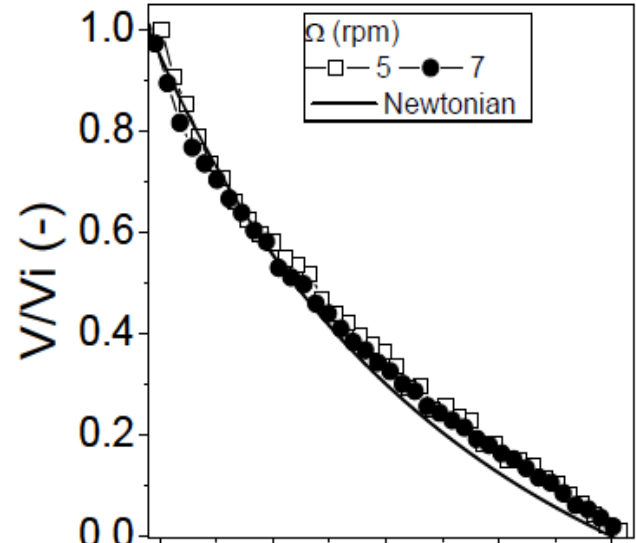
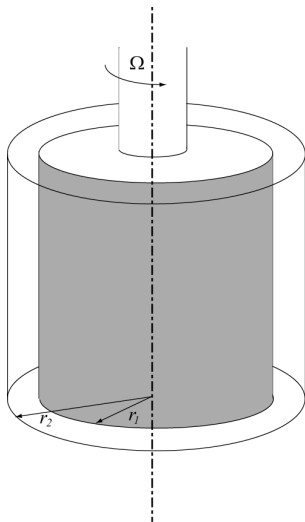
Cornstarch suspension in a wide gap Couette

For a 43.9% mean volume fraction



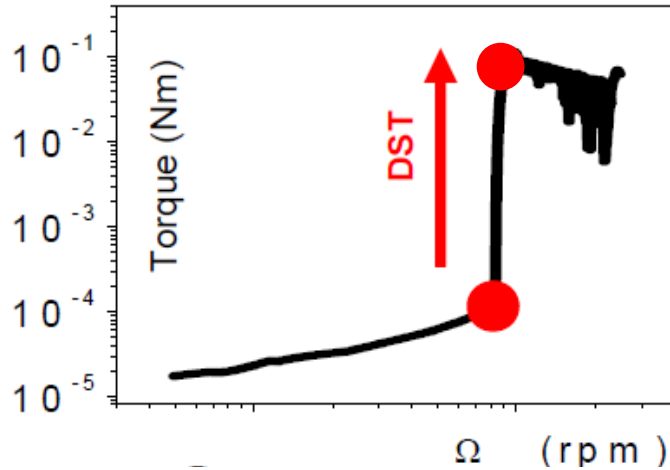
$\Omega < \Omega_c \rightarrow$ Shear thinning

• Homogeneous flow



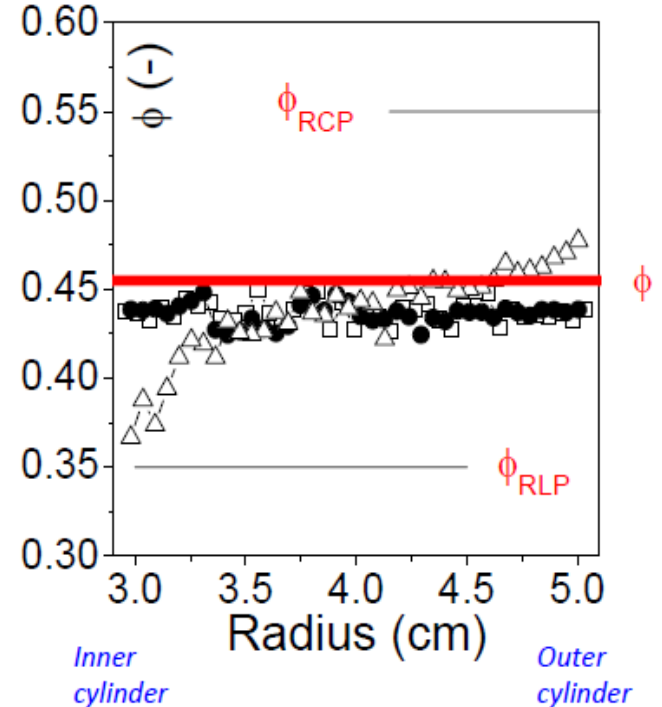
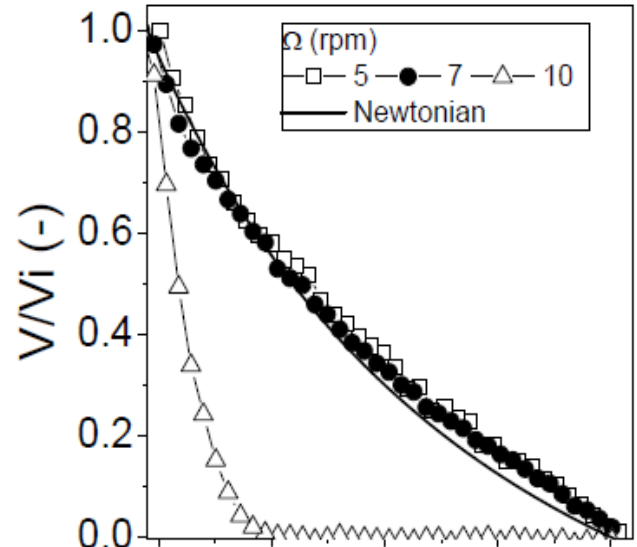
Cornstarch suspension in a wide gap Couette

For a 43.9% mean volume fraction



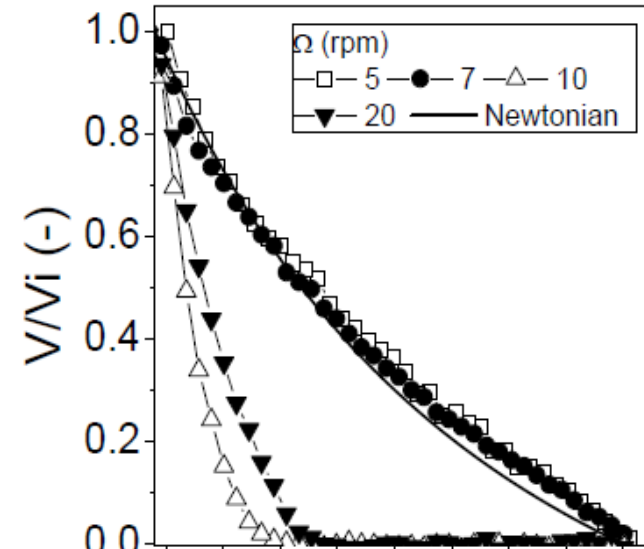
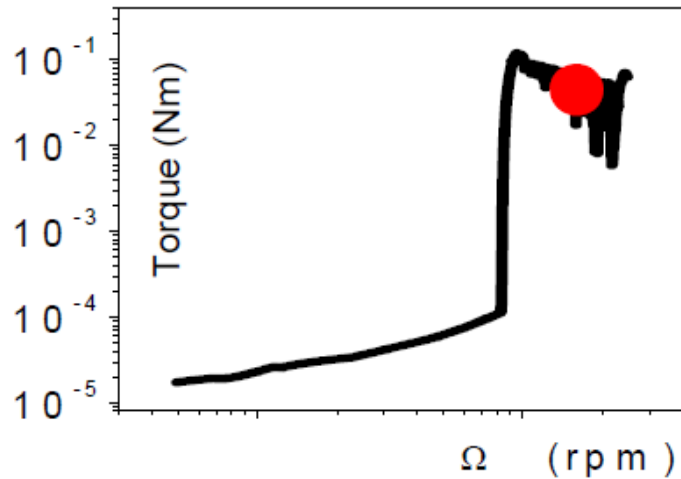
Sudden **shear localization**
(creation of a **dead zone**)

Link with emergence of volume fraction
inhomogeneities

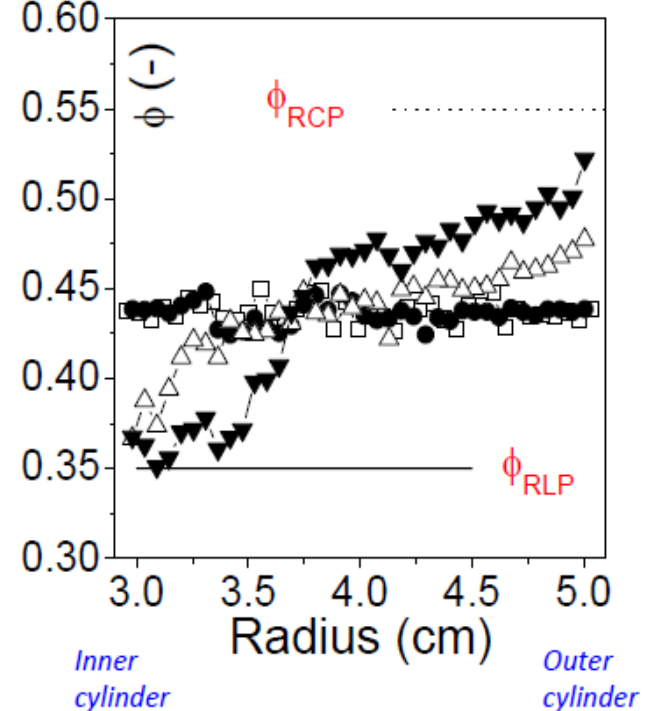


Shear-thickening in cornstarch: local measurements

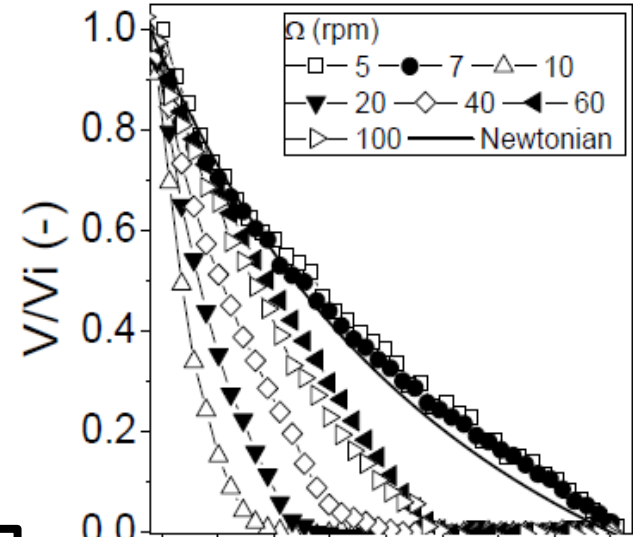
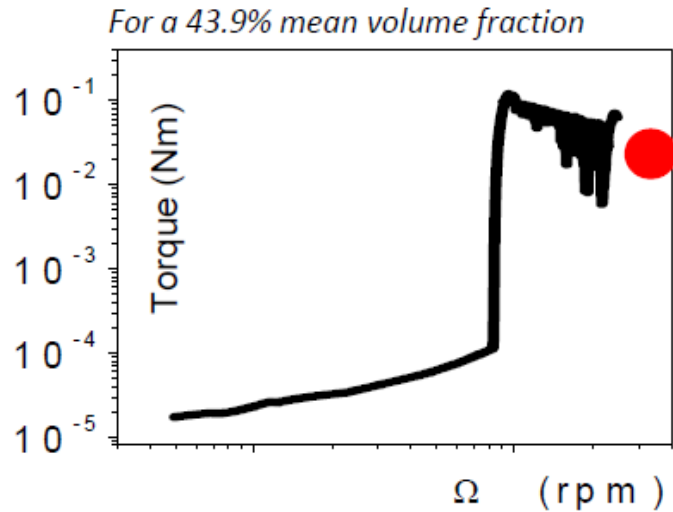
For a 43.9% mean volume fraction



- o The sheared region grows up with Ω
- o Broadening of the low-density region
- o Compaction of the Jammed region towards ϕ_{RCP}

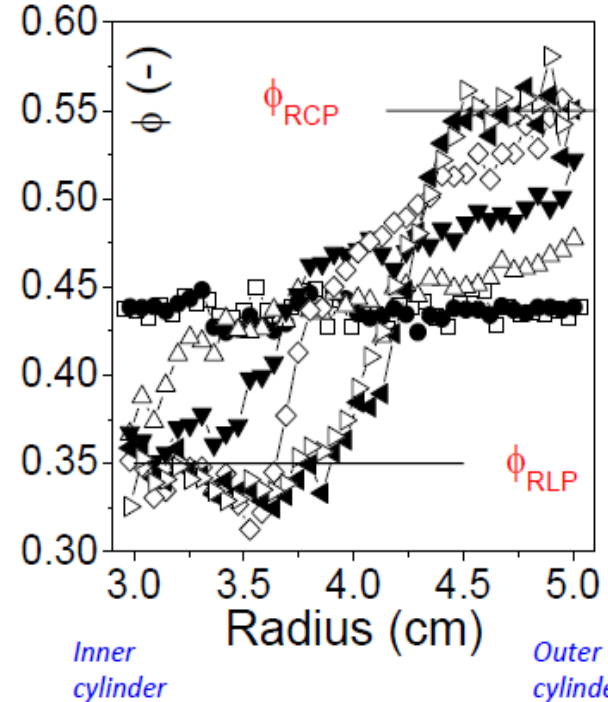


Shear-thickening in cornstarch: local measurements



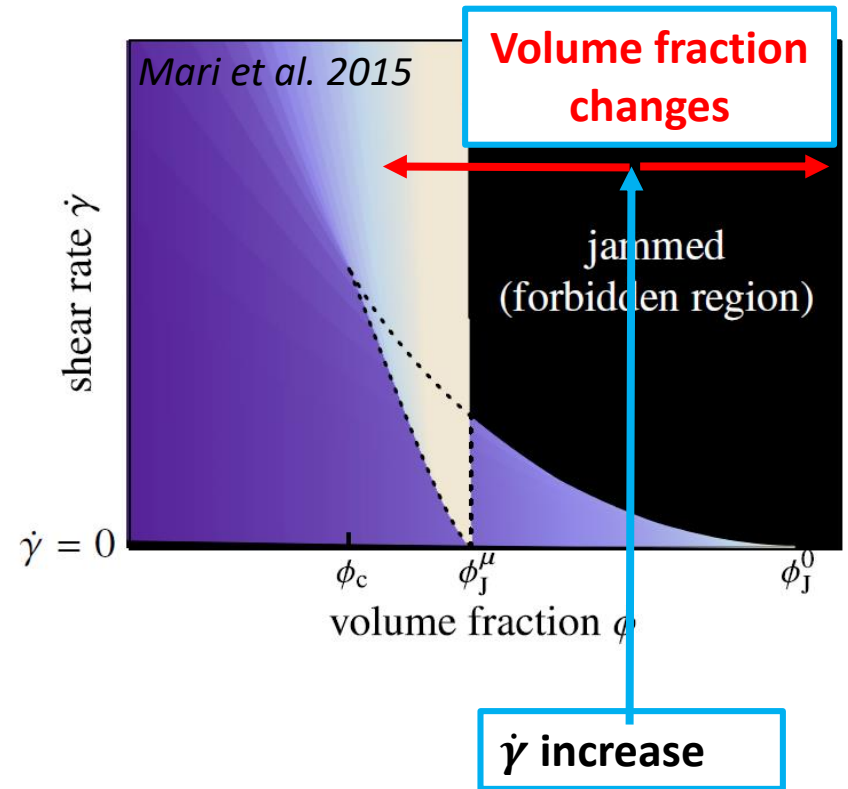
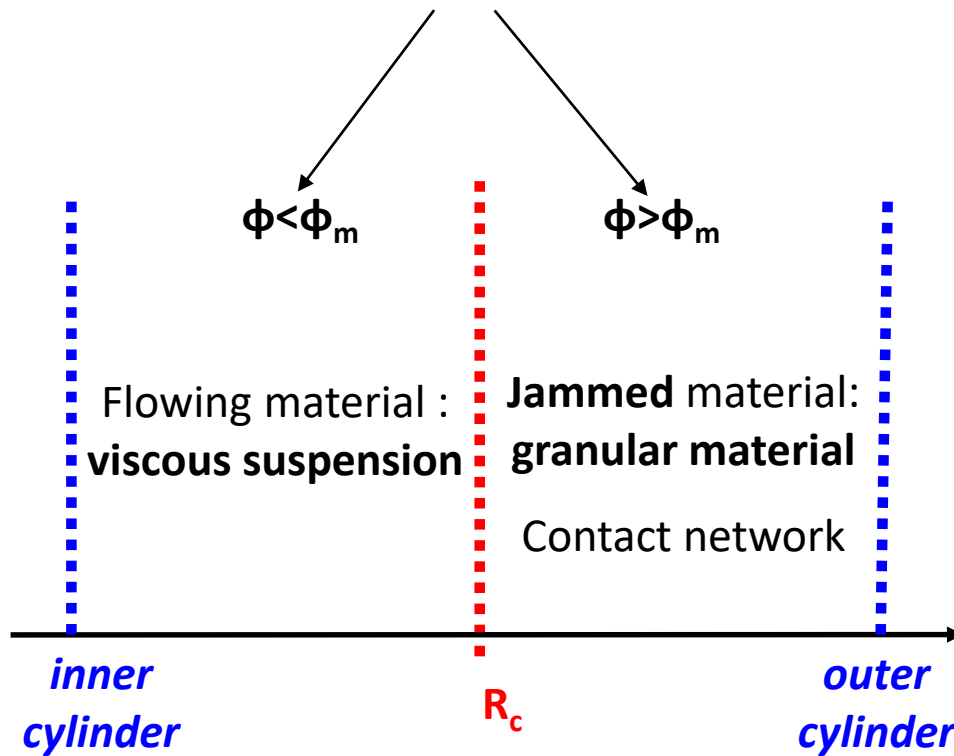
What can we learn on the local behavior from all this mess???

→ Extract all $(\phi, \dot{\gamma}(\phi))$
to build a « state diagram »



Shear induced migration and Shear induced jamming

Shear-induced migration can generate 2 different zones

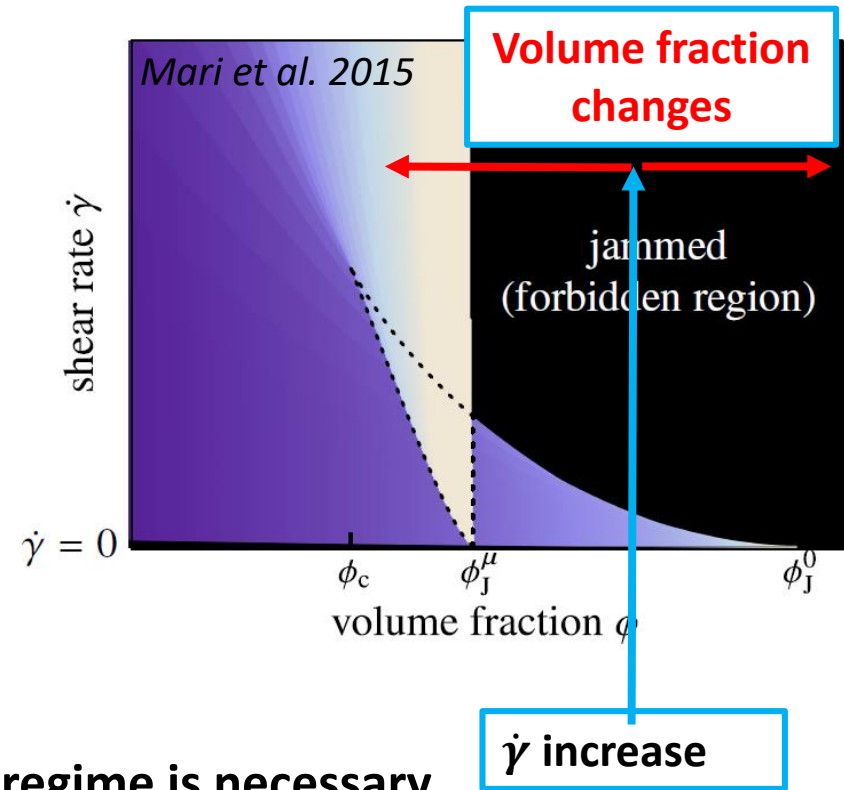
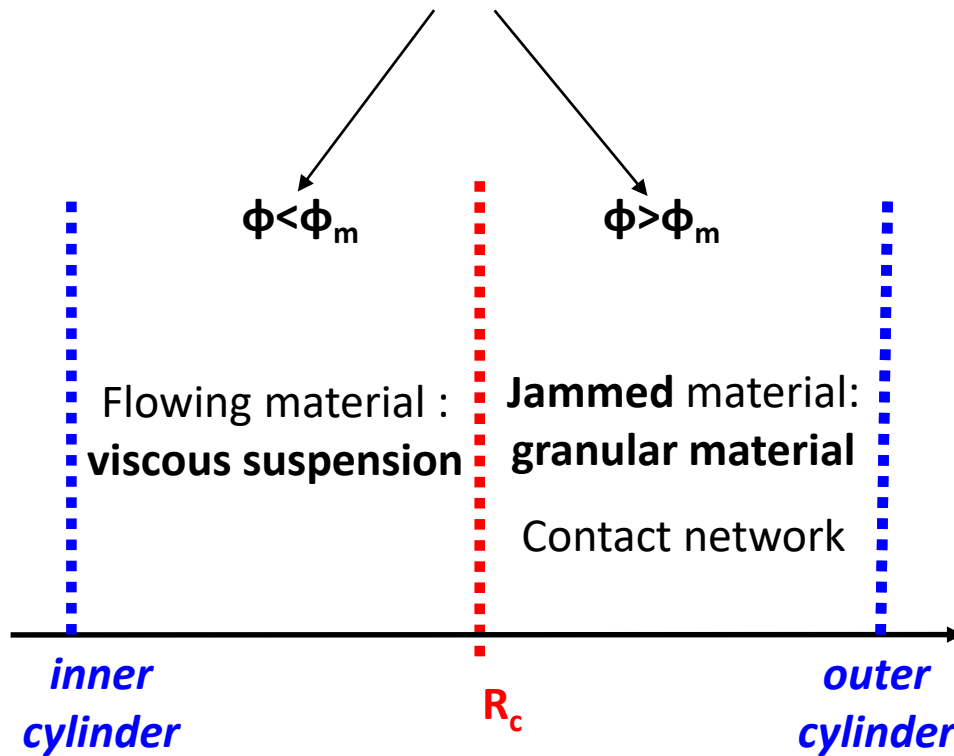


Discontinuous shear thickening = (shear-?) jamming.

ϕ decreases locally to allow for flow near the moving boundary

Shear induced migration and Shear induced jamming

Shear-induced migration can generate 2 different zones

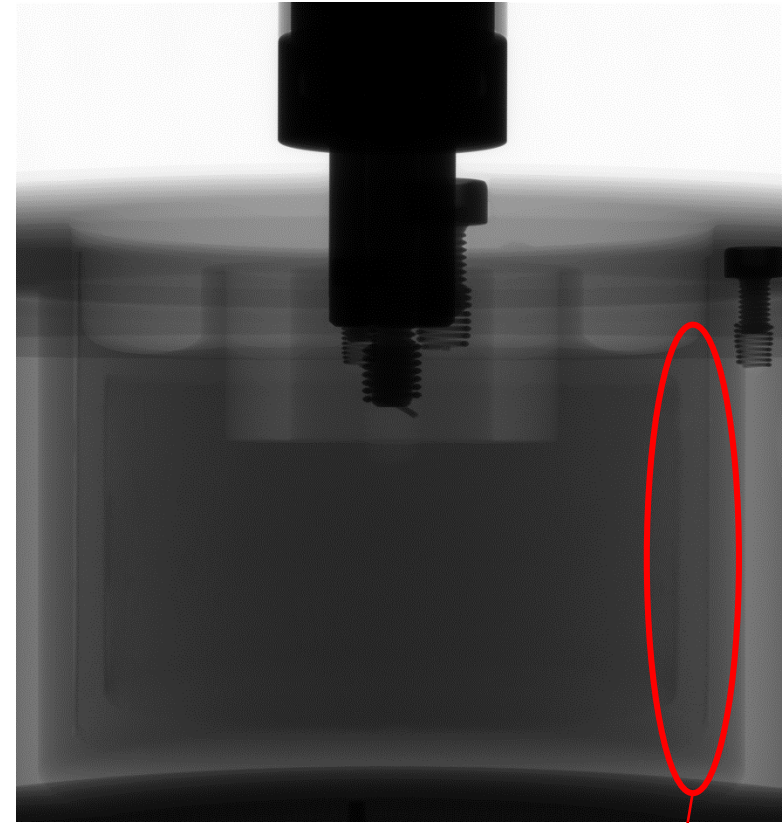
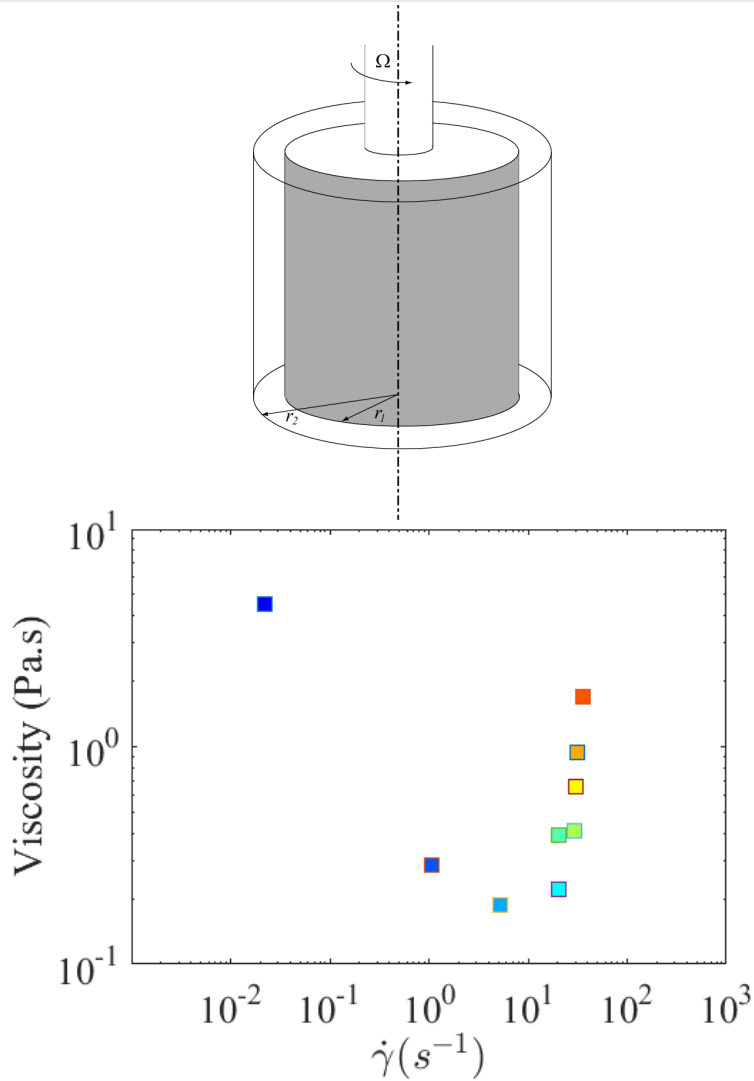


A diphasic model accounting for the granular regime is necessary

→ see Lecampion, Garagash, JFM (2014)

Rheology with X-ray imaging

@Placamat, Bordeaux



Volume fraction field $\phi(r, z, t)$
extracted

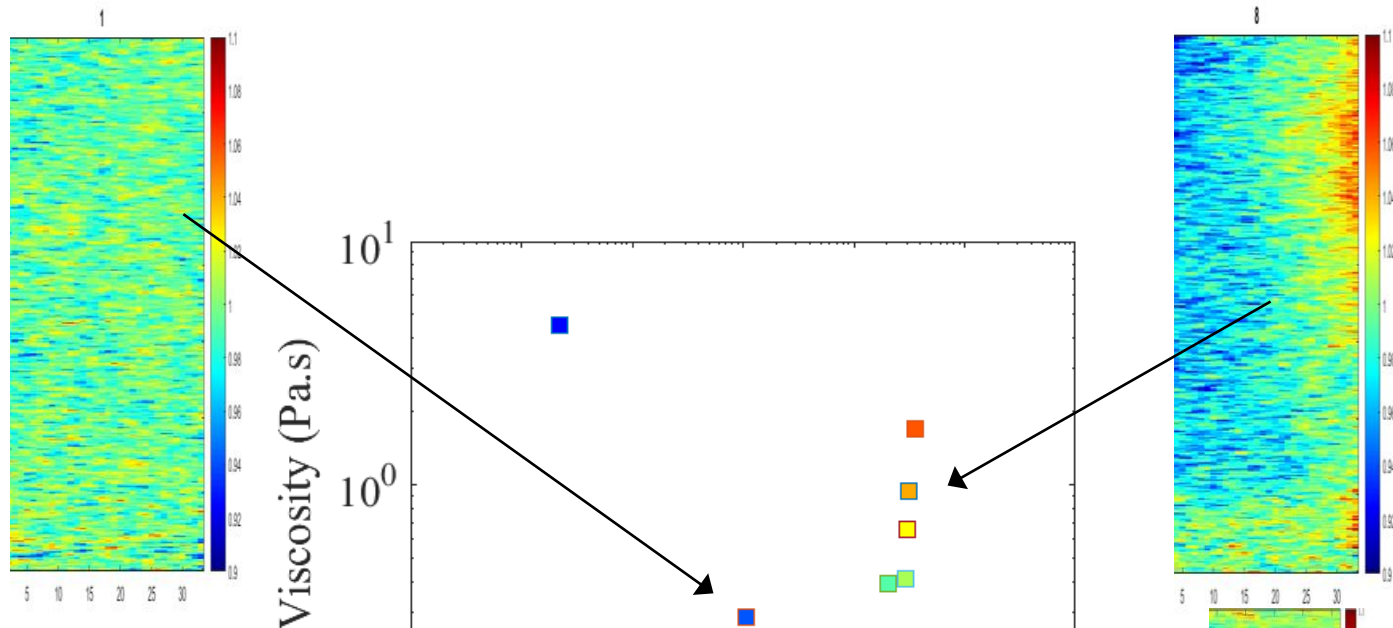
Real-time measurement during rheology experiment

X-ray imaging: application to shear thickening

41% of cornstarch in water/CsCl in 1mm gap Couette (*stress inhomogeneity* : 8%)

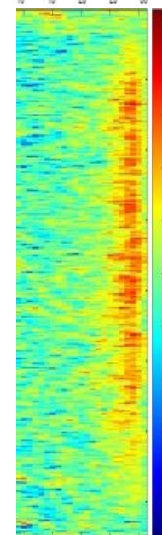
Volume fraction field abruptly changes in the thickening regime

Homo
geneous



$\pm 5\%$
variation
in the gap

Same feature
and magnitude
in a
500 μm gap,
with
smooth/rough
boundaries

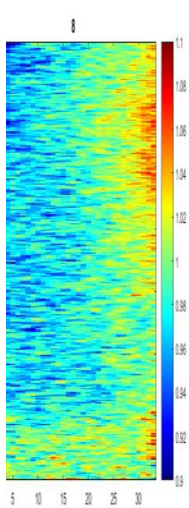
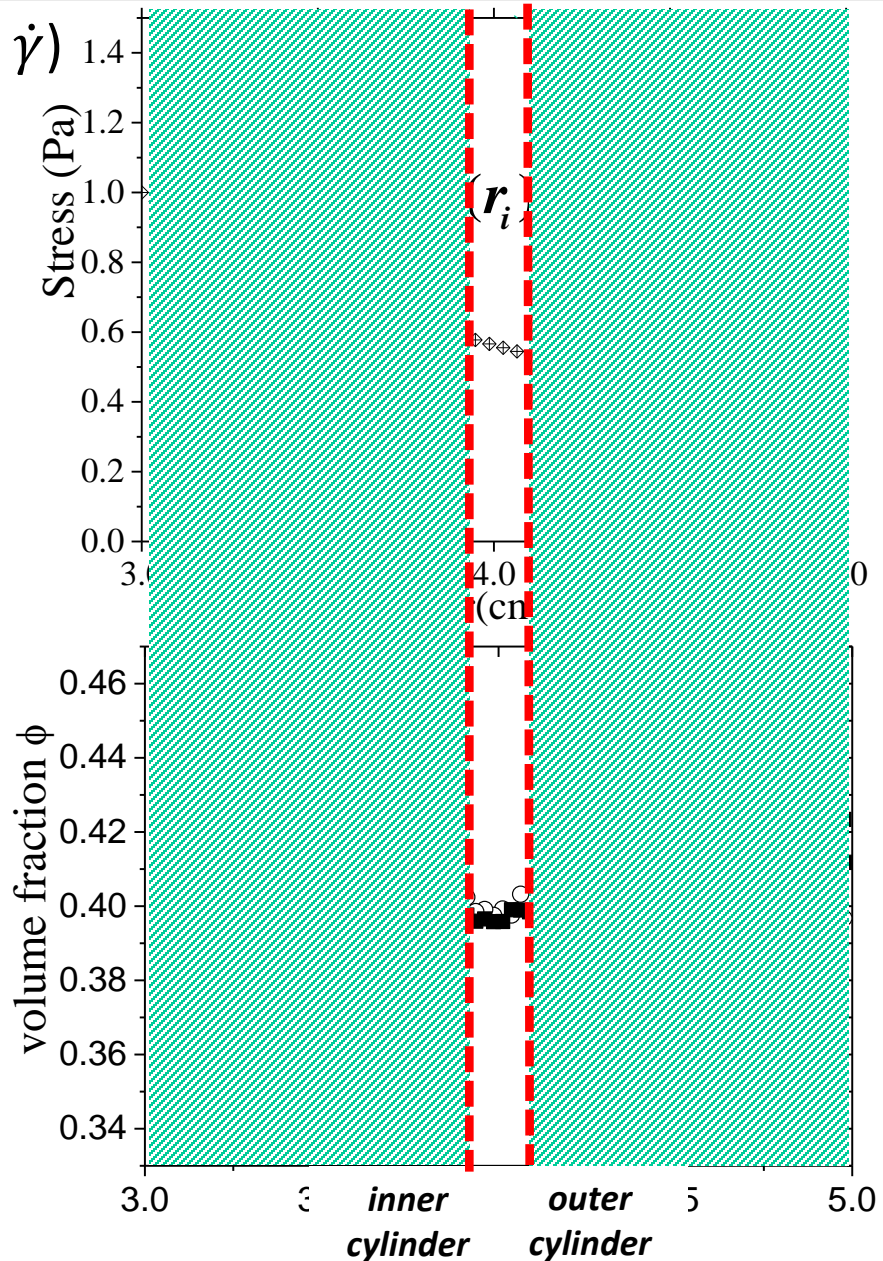


Steady-state: volume fraction in a thin gap Couette

Ex : viscous suspension ($\Sigma \propto \dot{\gamma}$)
40% of PS beads

For a 8% stress inhomogeneity
(case for the thickening suspensions investigated)

$\pm 0.25\%$ relative vol. fraction expected

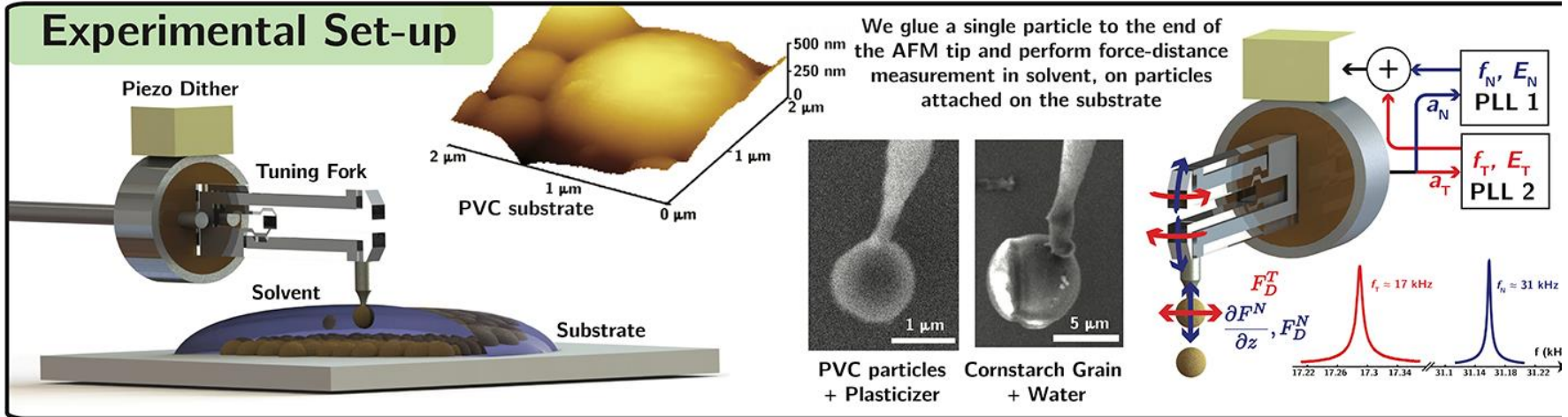


**$\pm 5\%$ variation
observed
in cornstarch !**

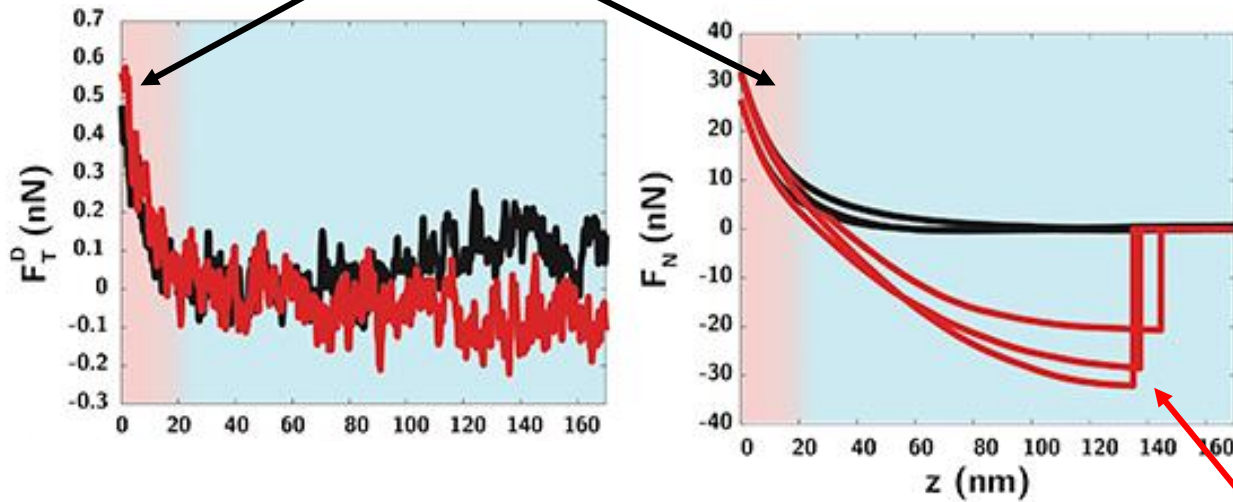
Now, physics can be discussed...

Force measurements between cornstarch particles

Experimental Set-up



Repulsion with friction



**Hysteresis:
adhesion force**

Proposed mechanism

Not a revolution!...

...same idea as

Seto et al., PRL 2013

Wyart and Cates, PRL 2014...

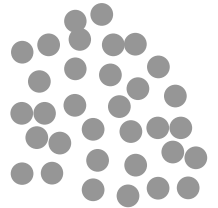
... with adhesion

Transition from

Low stress / shear rate :
repulsion, frictionless particles

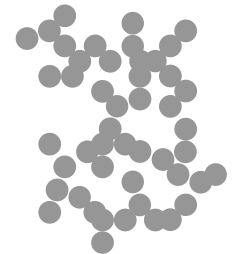
→ suspension of frictionless spheres
with **viscous behaviour**

to



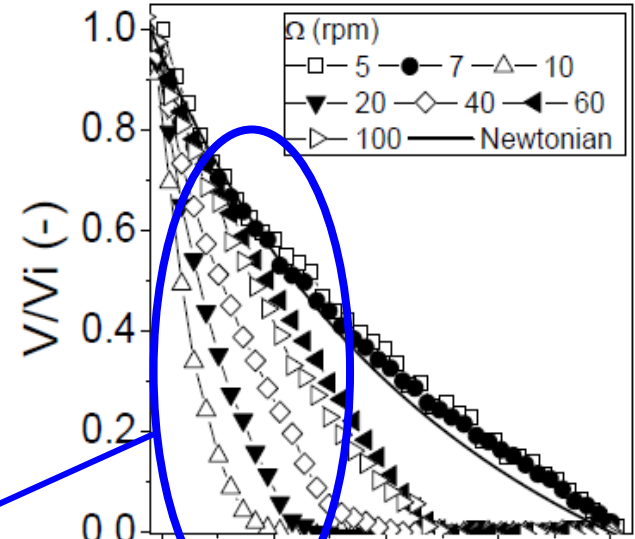
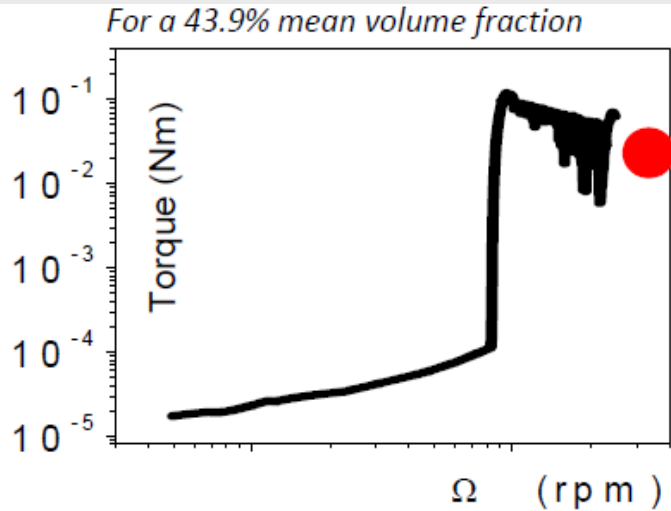
High stress/shear rate:
shear-induced adhesion

→ **cohesive granular material**
(or colloidal gel)
with **yield stress**



Order of magnitude, from adhesion force : $\tau_y \simeq 500 \text{ Pa}$ consistent with post thickening behavior

Linking yield stress behavior/local behavior



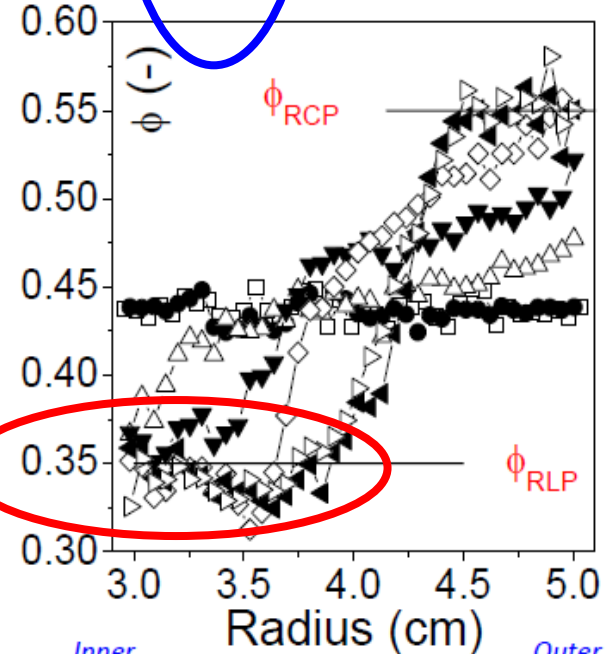
Velocity profiles

of the material

@ local vol. fraction $\approx 35\%$

consistent with
yield stress fluid behavior
(Herschel-Bulkley)

Yield stress \approx hundreds of Pa

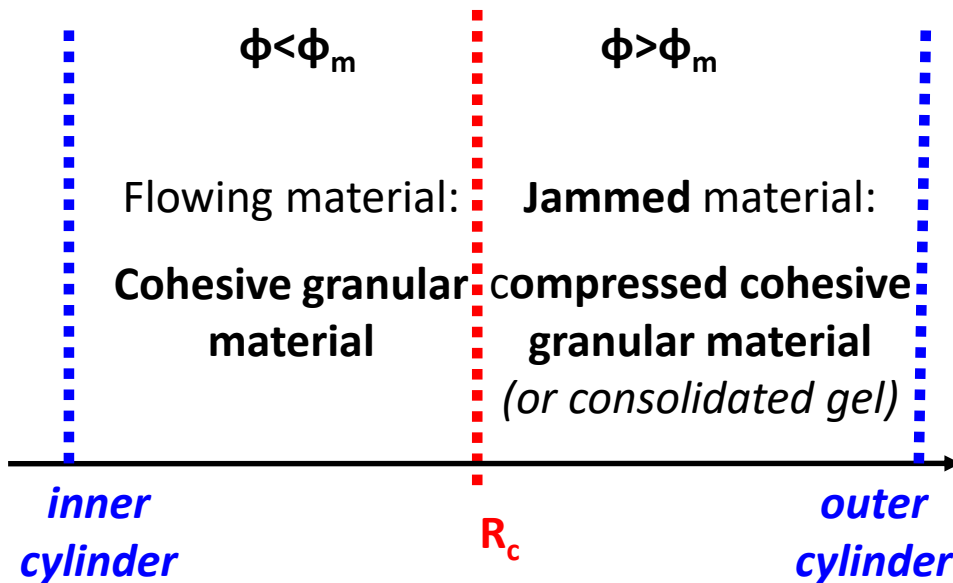
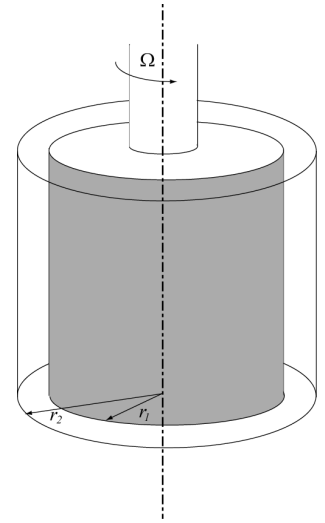


Inner
cylinder

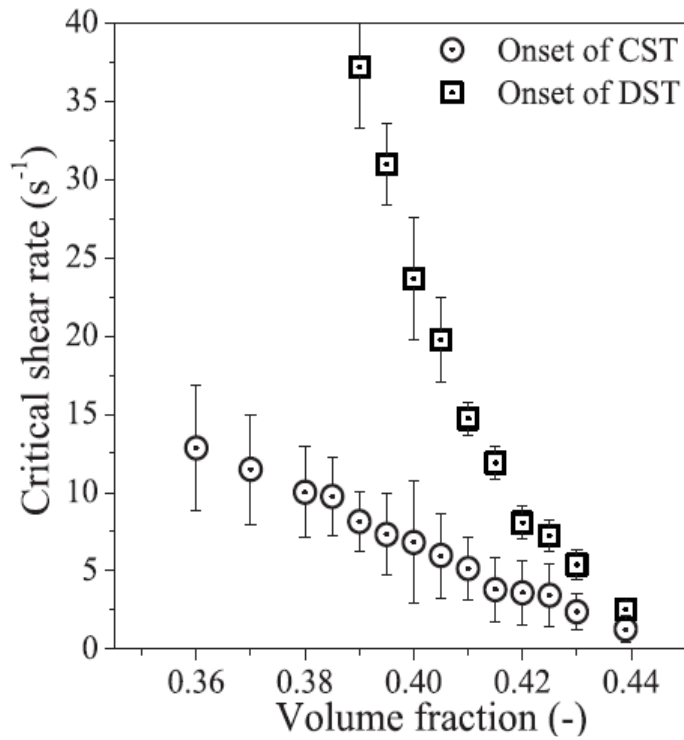
Outer
cylinder

Linking shear-induced inhomogeneities/adhesion

- shear-thickening = shear-induced adhesion
- the cohesive granular material is irreversibly compressible up to ϕ_{rcp}



Accessible volume fractions



Jammed states

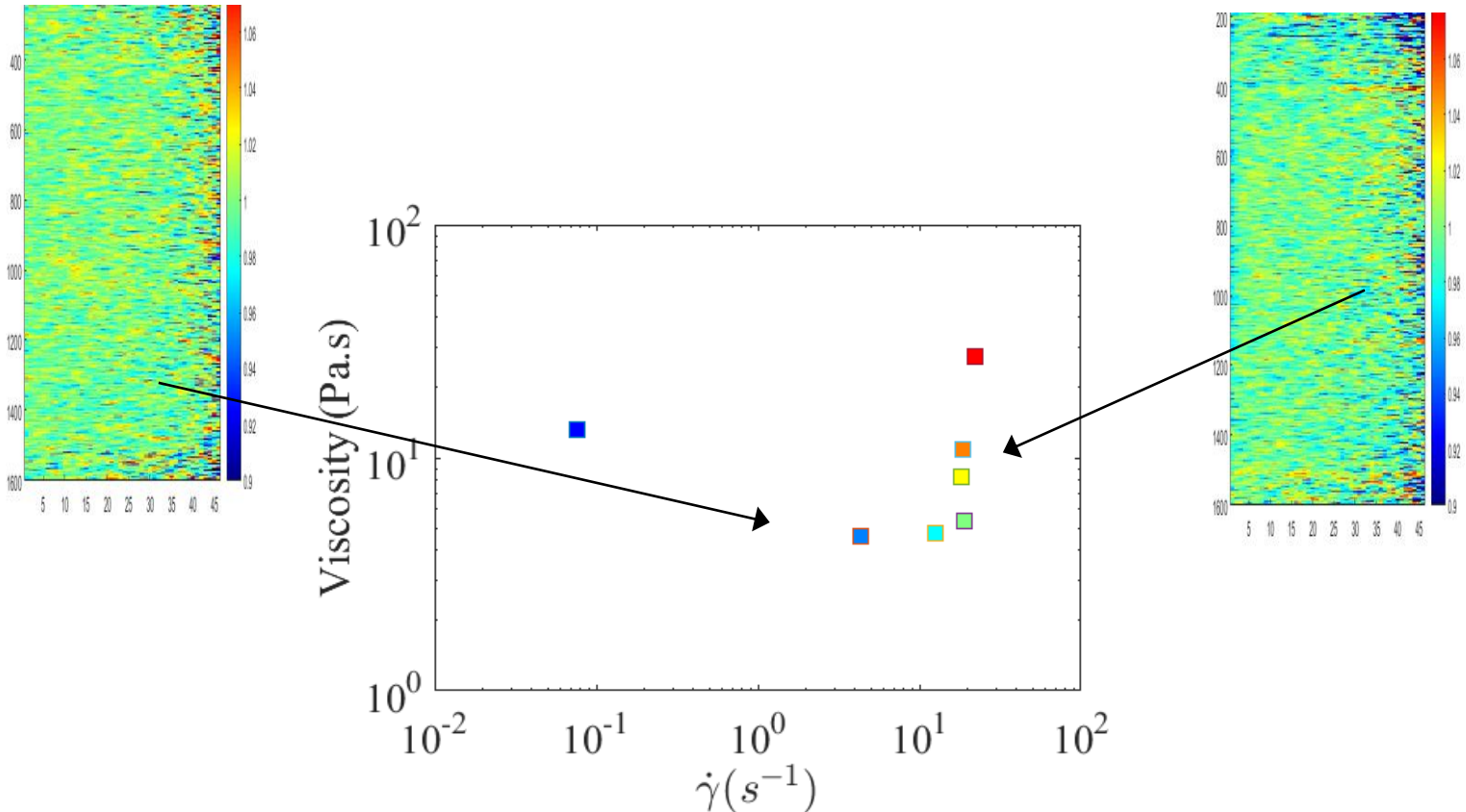
Compressed
cohesive materials

$$\phi_0 = 45\% \neq \phi_{rcp} = 55\%$$

X-ray imaging: application to shear thickening

61% of PVC particles in plasticizer 1mm gap Couette (*stress inhomogeneity : 8%*)

Volume fraction fields in the gap of the Couette cell remain **constant**

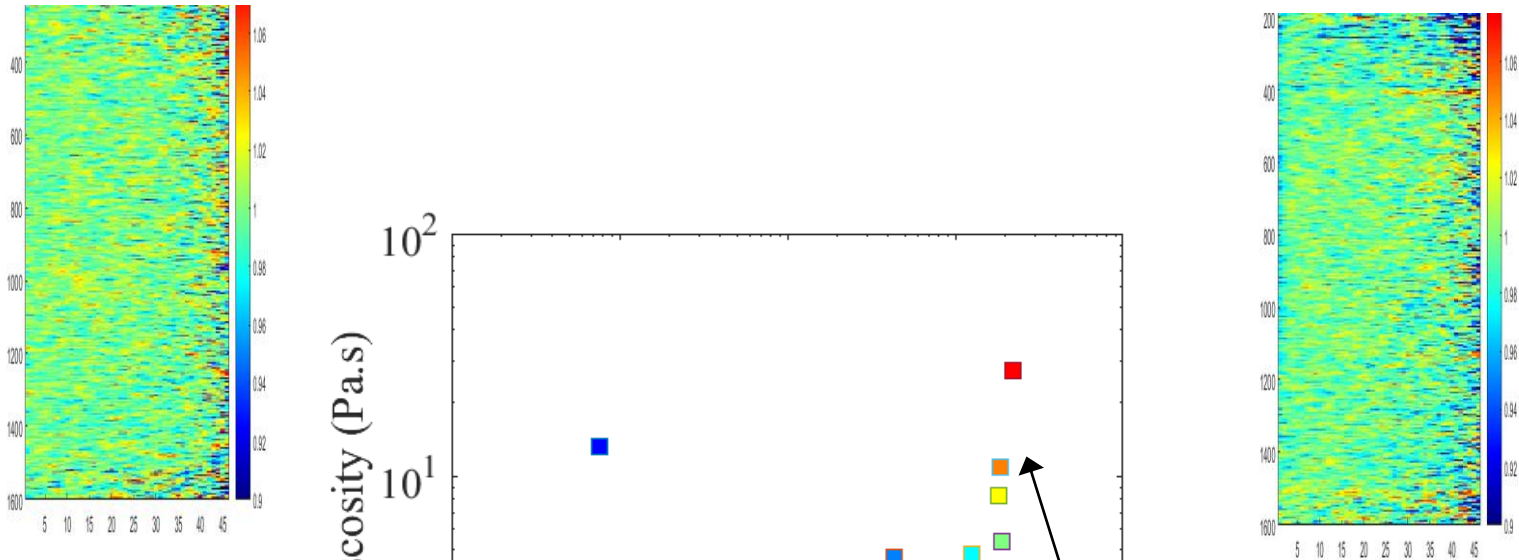


**Discontinuous Shear Thickening is a viscosity jump
in the constitutive behavior of the homogeneous material**

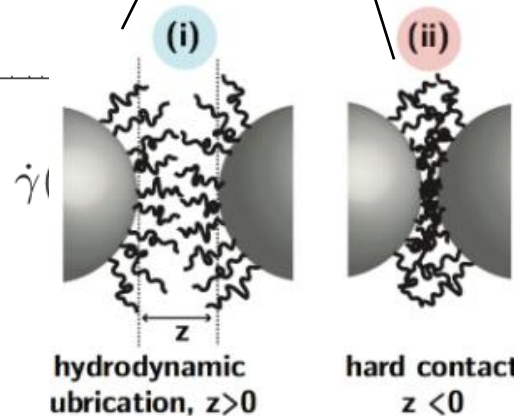
X-ray imaging: application to shear thickening

61% of PVC particles in plasticizer 1mm gap Couette (*stress inhomogeneity : 8%*)

Volume fraction fields in the gap of the Couette cell remain **constant**



Interpreted as transition from lubrication to contacts



Consistent with *Seto et al, PRL 2013*

Comtet et al., Nature communication 2017

Conclusion

Strong interplay between migration / shear thickening / (Shear-) jamming

« Discontinuous shear-thickening »



jump in viscosity **OR** shear-induced yield stress **OR** shear jamming...
depends on interparticle forces

hard to know from only macroscopic measurements

Need for volume fraction measurements

Conclusion

Cornstarch:

- Revisit the origin of the **s-shape**?
In which range of local vol. fraction does s-shape exist?
- Impacts on the surface of a Couette cell (*Peters et al., Nature 2016*)
→ **which volume fractions are truly investigated?**

Other systems investigated:

- **PMMA suspensions** → **inhomogeneous** at DST... }
 - **Silica suspensions** → **inhomogeneous** at DST... }
- To be studied in more depth
- **PVC suspensions** (Comtet et al., Nature. Comm 2016) :
« **THE** » canonical system!