

Drop deformation and breakup in laminar shear flow

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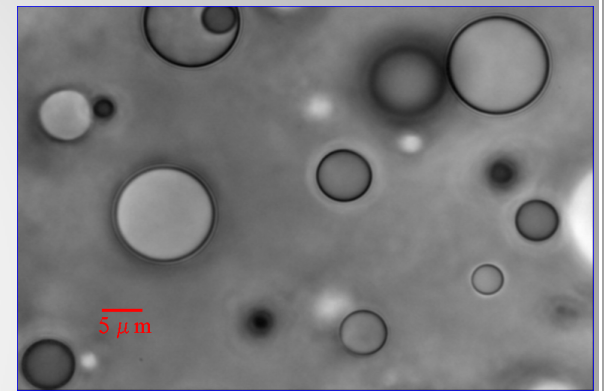
**COST P21 Student Training School “Physics of droplets: Basic and Advanced topics”
Borovets, 13 July 2010**



Background

- **Liquid-liquid biphasic systems**

- Examples: emulsions, polymer blends, water-in-water biopolymer mixtures
- In many cases system microstructure is characterized by **droplets** in a continuous phase



- **Applications:** detergents, personal care, food products, oil recovery

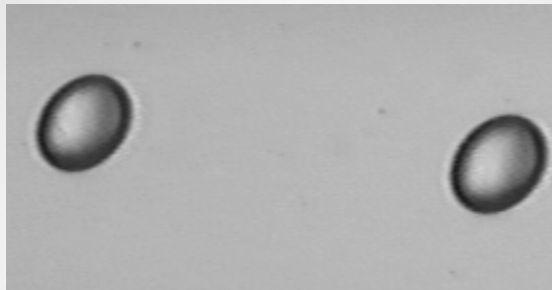
- **Flow-induced microstructure evolution**

- Processing (e.g., mixing)
- Final product properties and usage

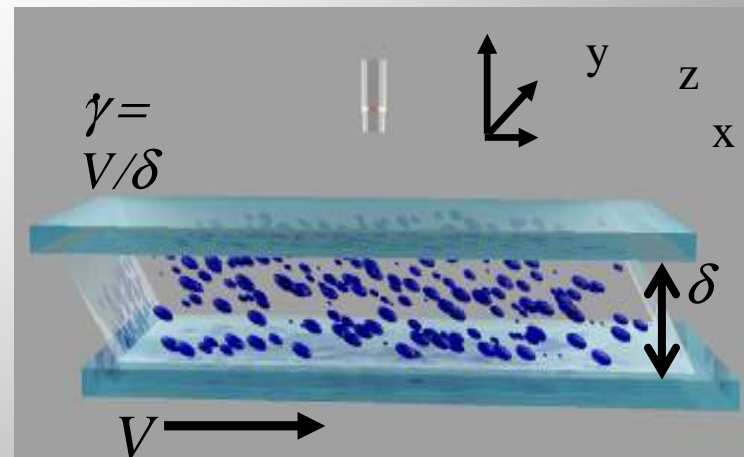
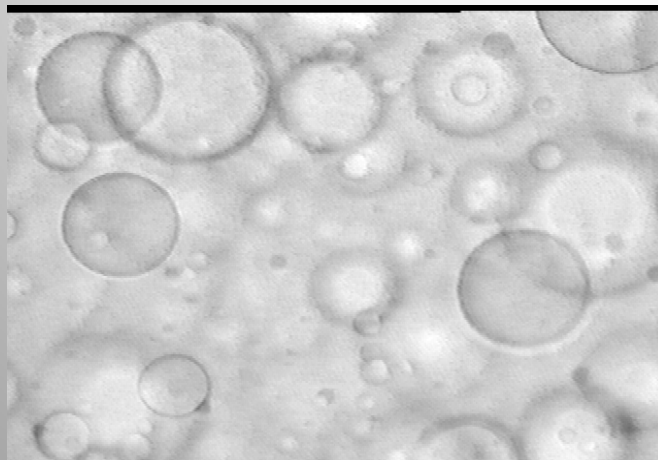


Background

- **Two main mechanisms governing microstructure dynamics under flow**
- Droplet collision and coalescence



- Droplet deformation and breakup (this presentation)





Topics

- **Drop deformation and breakup in simple shear flow**

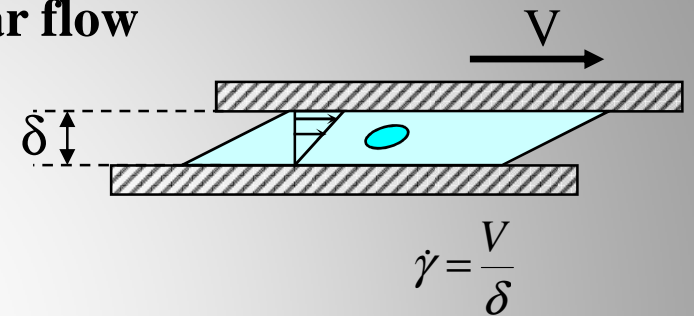
- *Isolated droplets*

- *No interfacial agents*

- *Immiscible, Newtonian fluids*

- *Laminar flow. However, results still apply in turbulent flow when eddies size is larger than droplet diameter (viscous turbulence)*

Vankova N, Tcholakova S, Denkov ND, Ivanov IB, Vulchev VD, Danner Th. J Colloid Interface Sci, 312, 363 (2007)



- **Non-Newtonian effects**

- **Wall effects**

- **Effect of concentration**



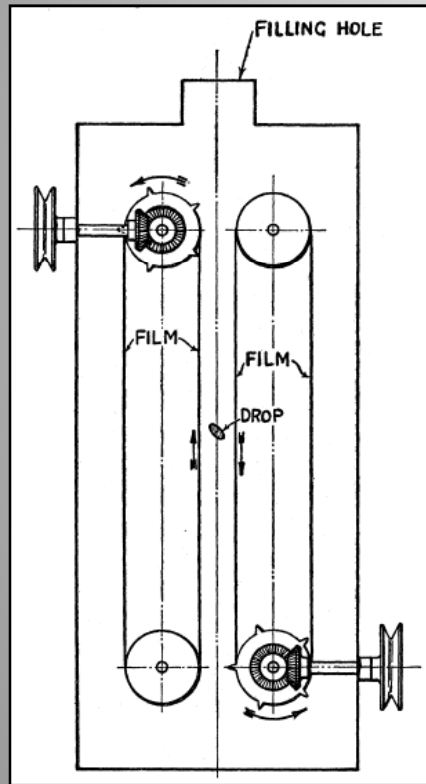
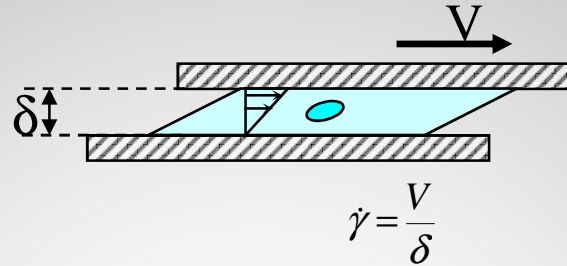
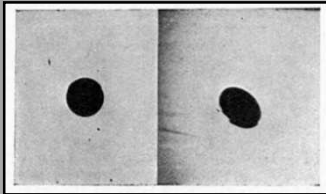
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Reviews

- 1) Rallison J M, “The deformation of small viscous drops and bubbles in shear flows”, *Ann. Rev. Fluid Mech.*, **16**, 45-66 (1984).
- 2) Stone H A, “Dynamics of drop deformation and breakup in viscous fluids”, *Ann. Rev. Fluid Mech.*, **26**, 65-102 (1994) .
- 3) Tucker III C L and Moldenaers P, “ Microstructural evolution in polymer blends”, *Ann. Rev. Fluid Mech.*, **34**, 177-210 (2002).
- 4) S. Guido and F. Greco, “Dynamics of a liquid drop in a flowing immiscible liquid”, in *Rheology Reviews 2004*, Ed. D. M. Binding and K. Walters, The British Society of Rheology, Aberystwyth, pp. 99-142 (2004)
- 5) Fischer P, Erni P, “Emulsion drops in external flow fields -- The role of liquid interfaces”, *Current Opinion in Colloid & Interface Science*, **12**, 196-205 (2007).
- 6) Derkach, S R, “Rheology of emulsions”, *Adv Colloid Interface Sci*, **151**, 1-23 (2009)
- 7) Minale M, “Models for the deformation of a single ellipsoidal drop: a review”, *Rheol Acta*, online article
- 8) Frith W J, “Mixed Biopolymer Aqueous Solutions – Phase behaviour and rheology”, *Adv Colloid Interface Sci*, advanced online article
- 9) Guido S and Preziosi V, *Adv Colloid Interface Sci*, advanced online article

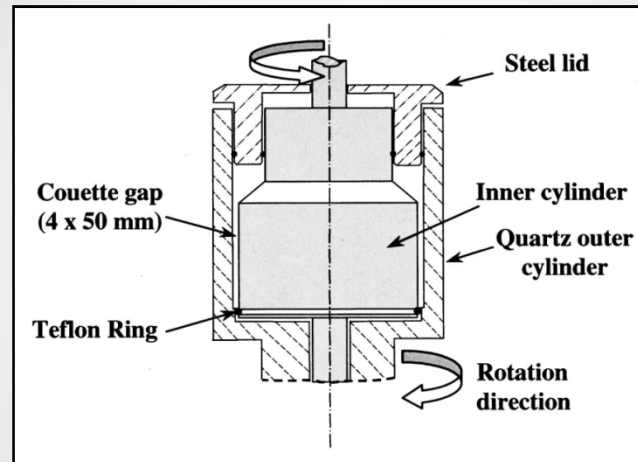


Experimental techniques



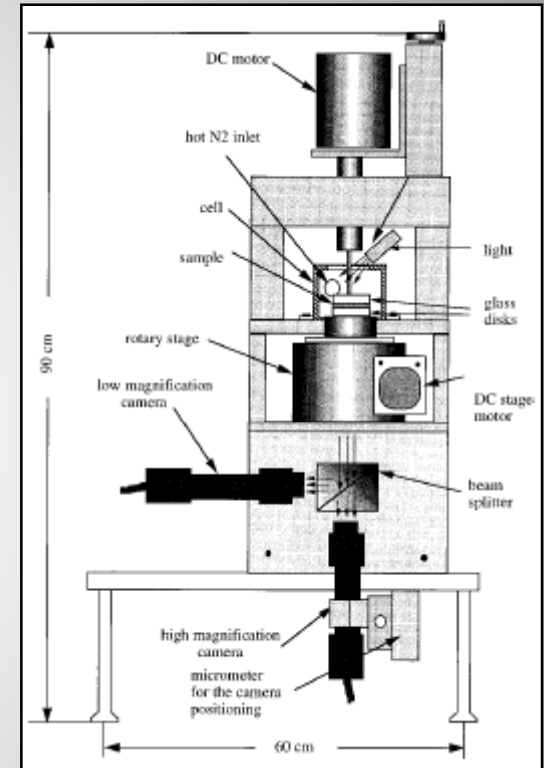
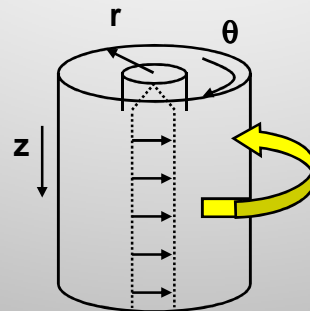
Parallel Band Apparatus

G. I. Taylor, *Proc. R. Soc. Lond. A*,
 146, 501-523 (1934)



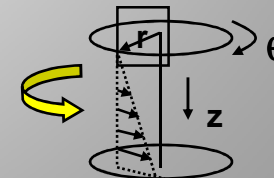
Couette geometry

Mighri F and Huneault M A, *J. Rheol.*, **45**, 783-797 (2001)



Parallel plates (rotational)

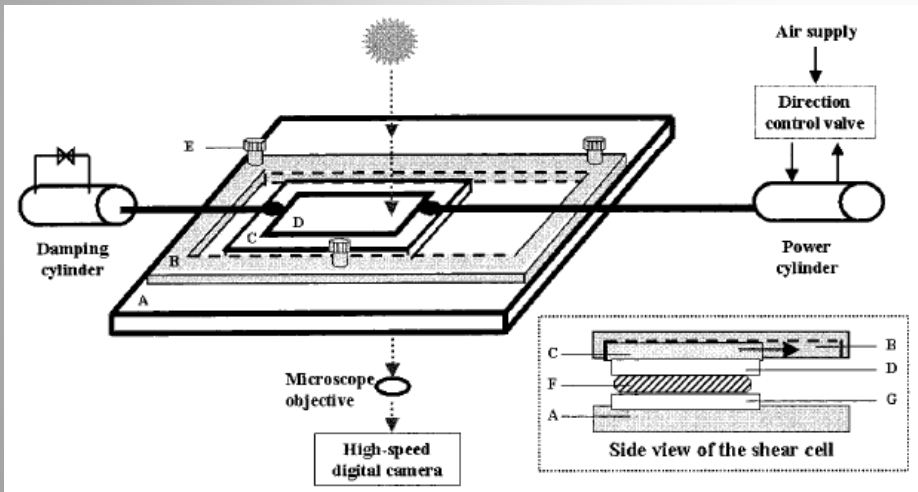
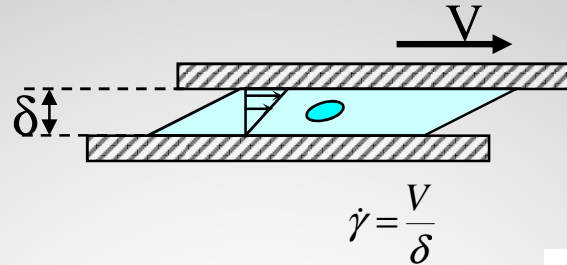
Levitt L, Macosko C W and Pearson S D,
Polymer Eng. Sci., **36**, 1647-1655 (1996)





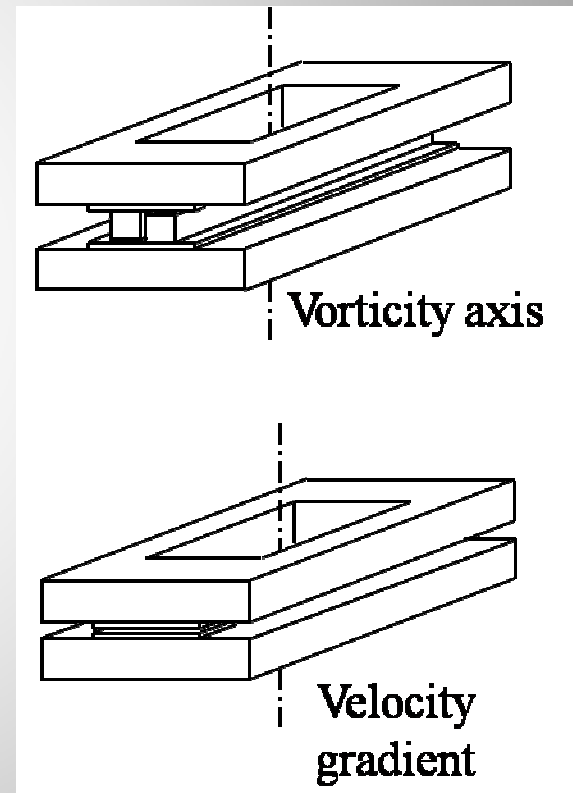
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Experimental techniques



Parallel plates (translating, high-speed)

X. Zhao and J. L. Goveas, *Langmuir*, **17**, 3788-3791 (2001)



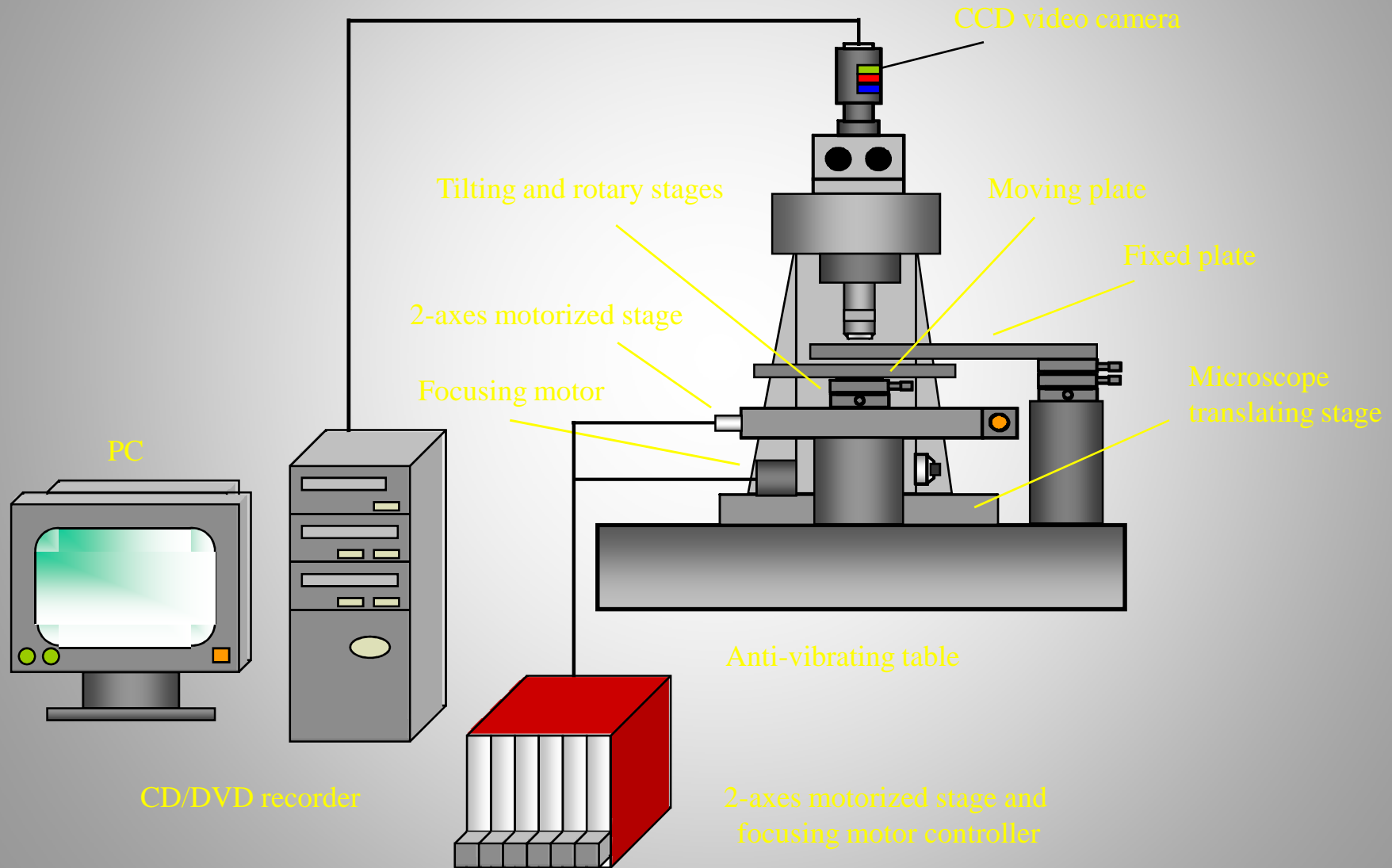
Parallel plates (translating)

S. Guido and M. Villone, *J. Rheology*, **42**, 395-415 (1998)



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Shear flow workstation

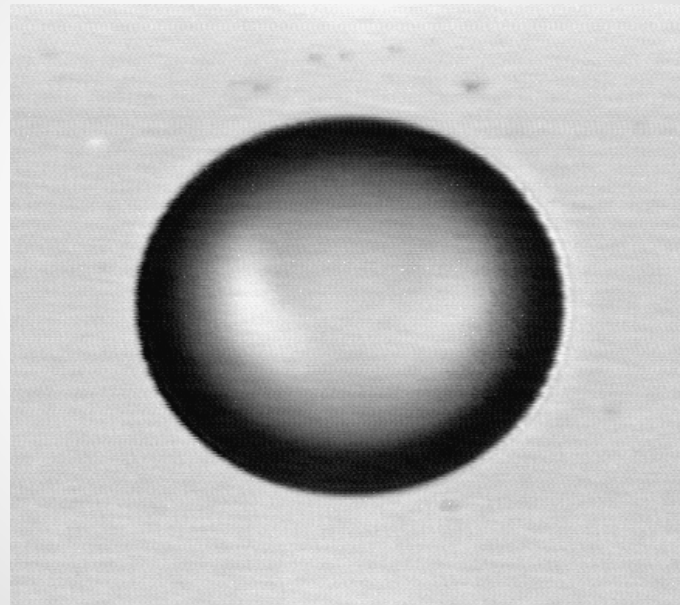
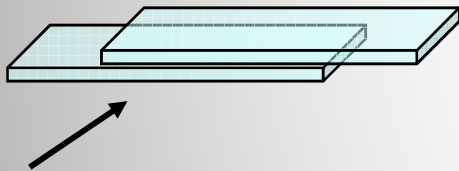




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Drop shape at small deformations

View along vorticity
(video)



Example: drop of polydimethylsiloxane in polyisobutylene

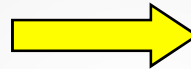


Small deformation theory

Newtonian case - Taylor, Chaffey-Brenner, Greco

Relevant physical quantities

- η_c viscosity of continuous phase
- η_d viscosity of drop phase
- $\dot{\gamma}$ shear rate
- r_0 drop radius at rest
- σ interfacial tension



Nondimensional numbers

$$Ca = \frac{\eta_c r_0 \dot{\gamma}}{\sigma} \quad \text{Capillary number}$$

$$\lambda = \frac{\eta_d}{\eta_c} \quad \text{Viscosity ratio}$$

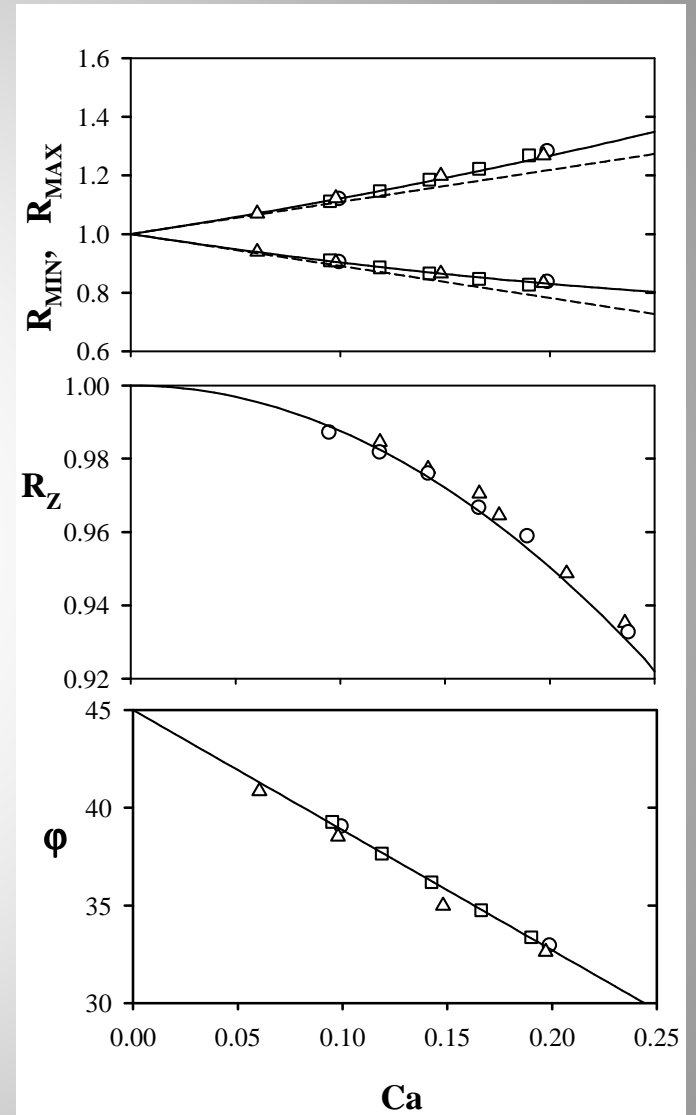
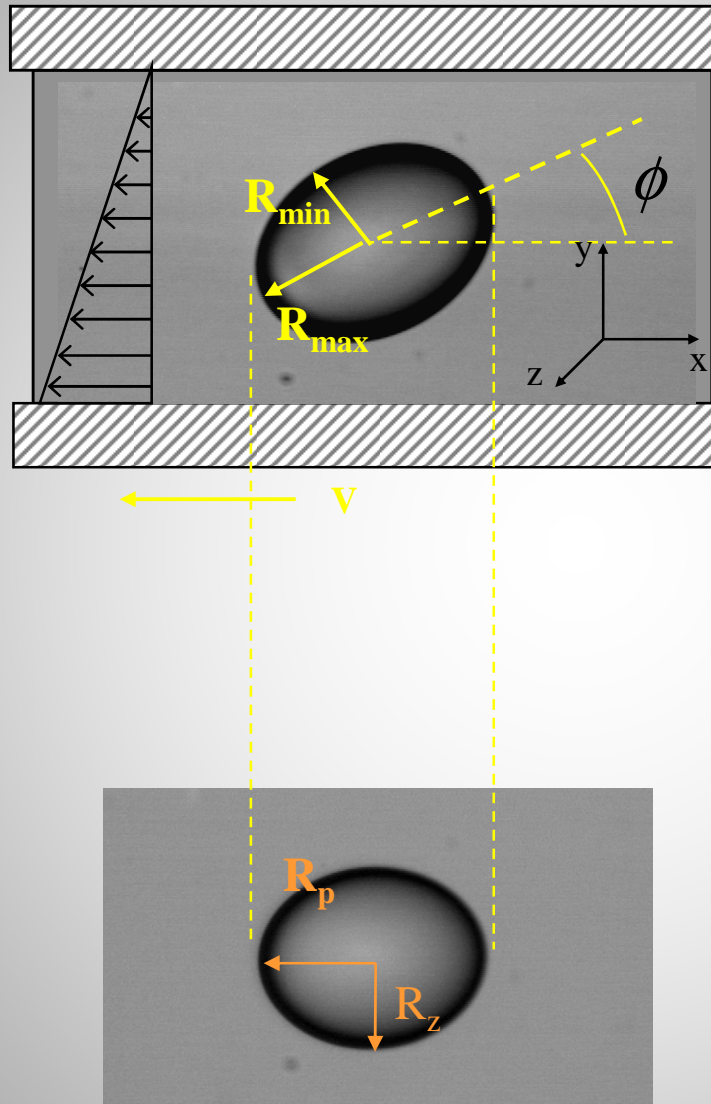
$$Ca = \frac{\tau \sigma}{\tau \dot{\gamma}}$$

Predictions

$$\begin{cases} \frac{r_{MAX}}{r_0} = 1 + f_1(\lambda)Ca + f_2(\lambda)Ca^2 \\ \frac{r_{MIN}}{r_0} = 1 - f_1(\lambda)Ca + f_2(\lambda)Ca^2 \\ \frac{r_z}{r_0} = 1 + f_3(\lambda)Ca^2 \end{cases} \quad \begin{cases} \varphi = \frac{\pi}{4} + \frac{(19\lambda + 16)(2\lambda + 3)}{80(1 + \lambda)}Ca \\ D \equiv \frac{r_{MAX} - r_{MIN}}{r_{MAX} + r_{MIN}} = \frac{19\lambda + 16}{16\lambda + 16}Ca \end{cases}$$



Stationary droplet shape



Drop shape is ellipsoidal up to moderate deformations

S. Guido and M. Villone, *J. Rheology*, **42**, 395-415 (1998)



Ellipsoidal models

Maffettone-Minale model

$$\frac{d\mathbf{S}}{dt} - \text{Ca} (\boldsymbol{\Omega} \cdot \mathbf{S} - \mathbf{S} \cdot \boldsymbol{\Omega}) = -f_1^{MM} [\mathbf{S} - g(\mathbf{S}) \mathbf{I}] + f_2^{MM} \text{Ca} (\mathbf{D} \cdot \mathbf{S} + \mathbf{S} \cdot \mathbf{D})$$

Ellipsoidal droplet described by a second order, positive-definite, symmetric tensor \mathbf{S}

$\boldsymbol{\Omega} = 1/2 \nabla \mathbf{v} - \nabla \mathbf{v}^T$ and $\mathbf{D} = 1/2 \nabla \mathbf{v} + \nabla \mathbf{v}^T$, where $\nabla \mathbf{v}$ is the velocity gradient tensor

$g(\mathbf{S}) = 3\text{III}_S/\text{II}_S$, where III_S and II_S are the third and the second scalar invariant of \mathbf{S} (to preserve droplet volume)

$$f_1^{MM} = \frac{40(1+\lambda)}{(3+2\lambda)(16+19\lambda)};$$

$$f_2^{MM} = \frac{5}{3+2\lambda} + \frac{3\text{Ca}^2}{2+6\text{Ca}^{2+\delta}} \frac{1}{1+\varepsilon\lambda^2},$$

f_i functions chosen to recover Taylor's small deformation limits

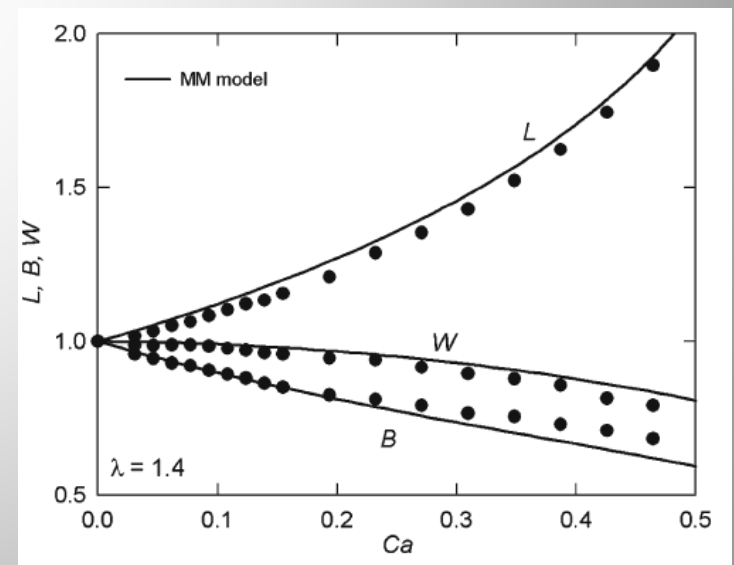
Maffettone PL, Minale M, *J Non-Newton Fluid Mech*, **78**, 227–241 (1998)

Other ellipsoidal models

Wetzel ED, Tucker CL III, *J Fluid Mech*, **426**, 199–228 (2001)

Yu W, Bousmina M, Grmela M, Palierne J, Zhou C, *J Rheol*, **46**, 1381–1399 (2002)

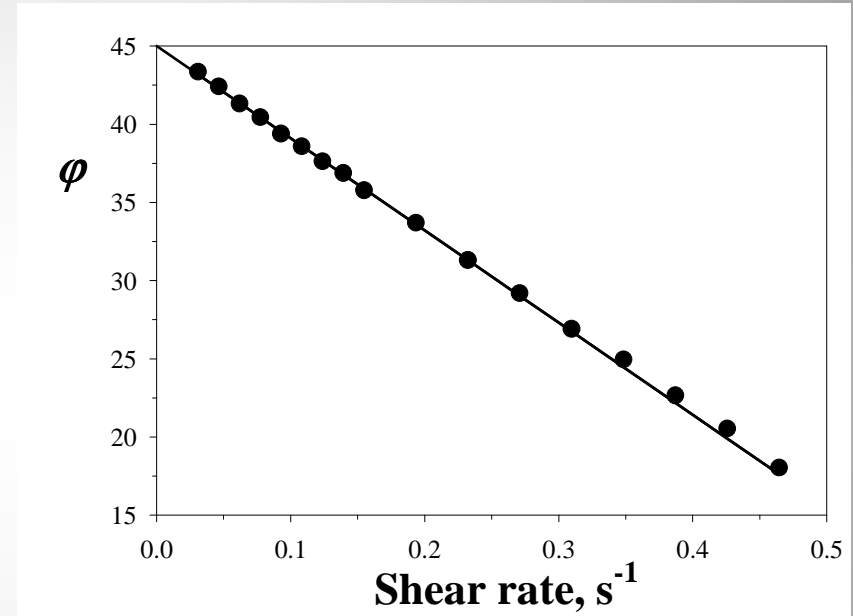
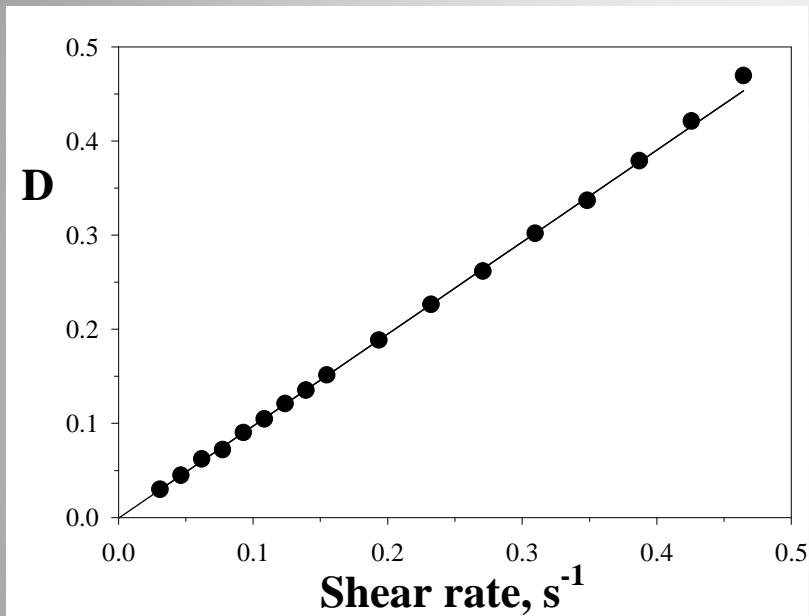
Edwards BJ, Dressler M, *Rheol Acta*, **42**, 326–337 (2003)





Interfacial tension measurement

FROM STEADY STATE SHAPE



$$D \equiv \frac{r_{MAX} - r_{MIN}}{r_{MAX} + r_{MIN}} = \frac{19\lambda + 16}{16\lambda + 16} Ca$$

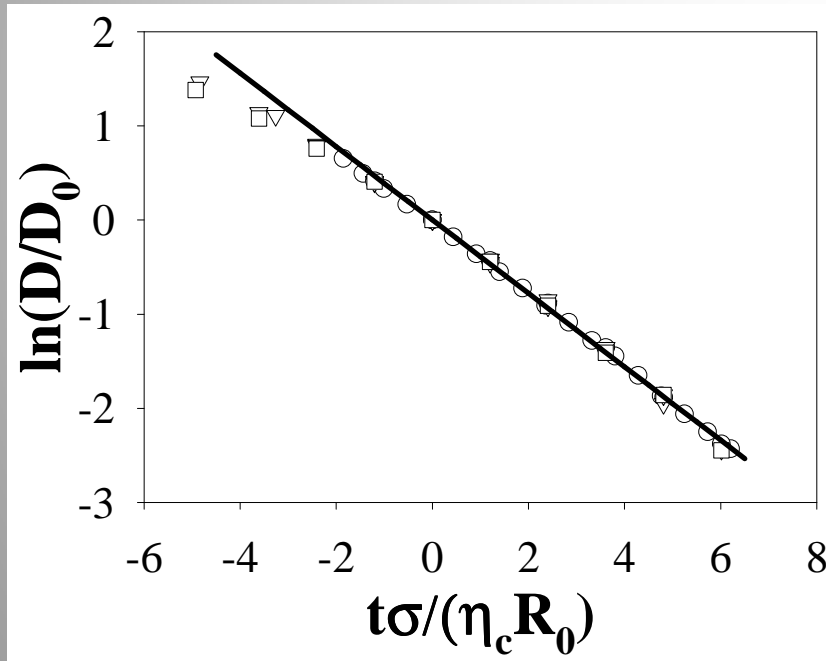
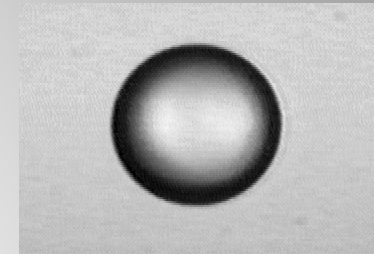
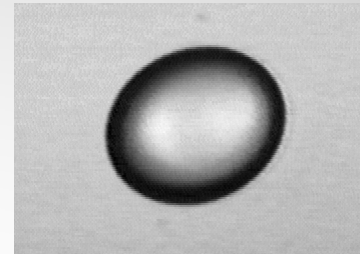
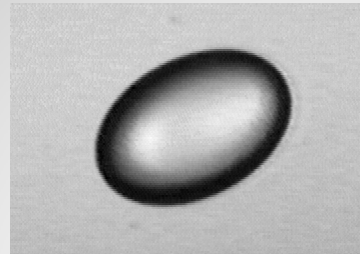
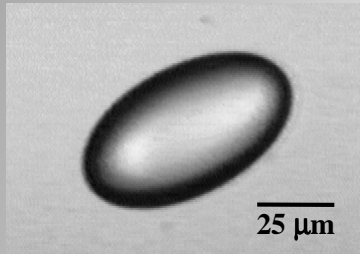
$$\varphi = \frac{\pi}{4} + \frac{(19\lambda + 16)(2\lambda + 3)}{80(1 + \lambda)} Ca$$



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Interfacial tension measurement

FROM DROP RETRACTION



$$D = D_0 \exp\left(-\frac{40(\lambda+1)}{(2\lambda+3)(19\lambda+16)}\tau\right)$$

$$\tau = t\sigma/(\eta_c R_0)$$

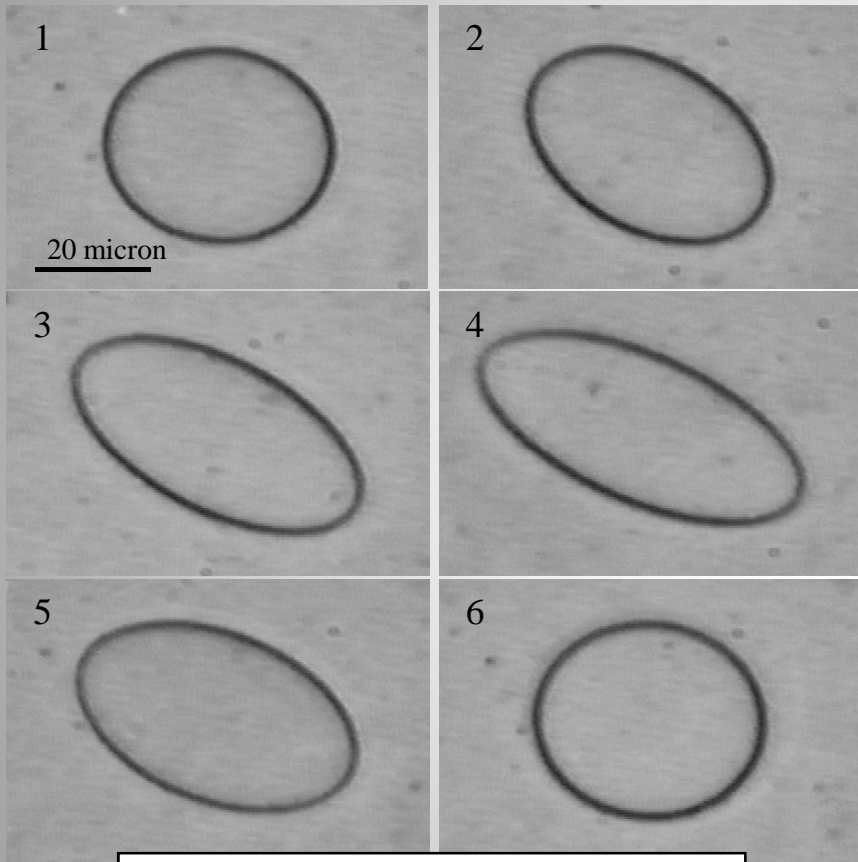
Overall data

Method	σ (mN/m)
Steady state D, Taylor	2.54 ± 0.07
Steady state angle, Chaffey-Brenner	2.62 ± 0.07
Retraction, Rallison	2.58 ± 0.06

Luciani A, Champagne M F and Utracki L A., *J. Polym. Sci. Phys. Ed.*, **35**, 1393-1403 (1997) .
 Guido S and Villone M., *J. Colloid Interface Sci.*, **209**, 247-250 (1999).

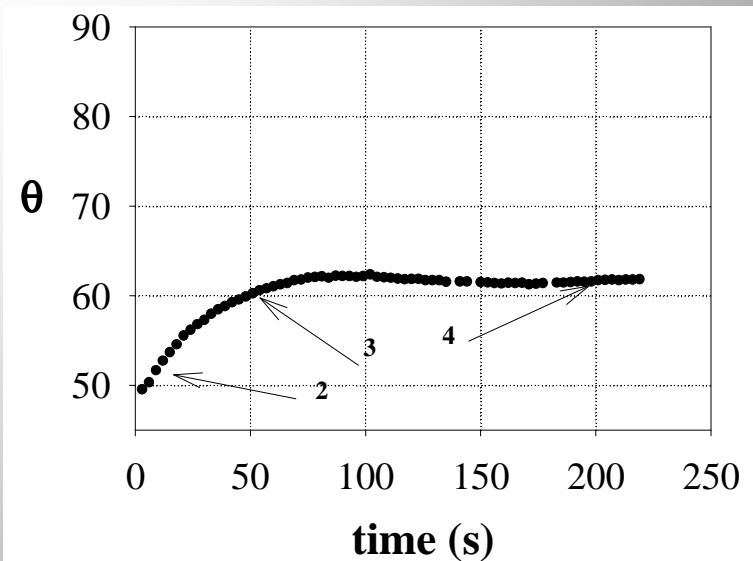
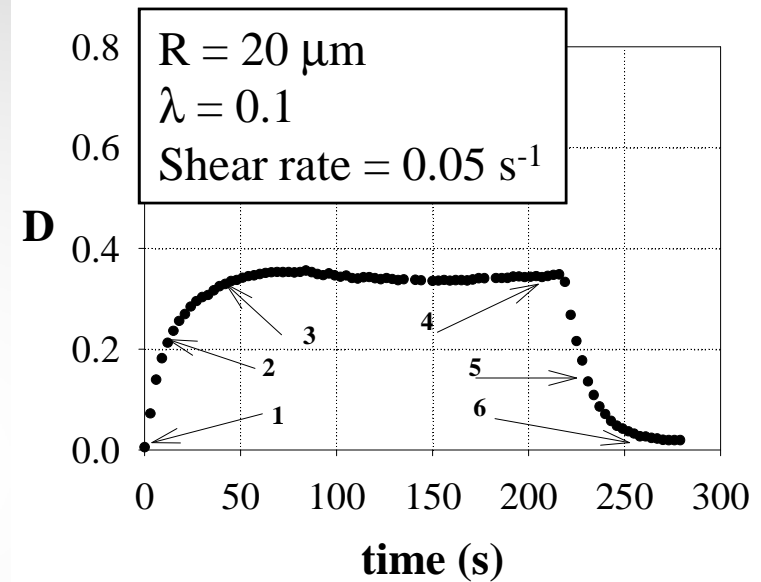


Water-in-water biopolymer mixtures



Drop: Na-caseinate rich phase
Matrix: Na-alginate rich phase

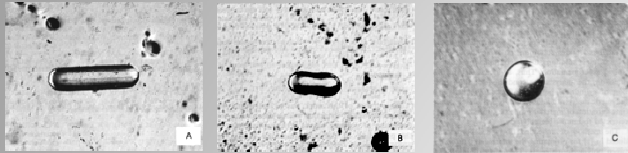
Guido, S Simeone M and Alfani, A, *Carbohydrate Polymers*,
48, 143-152 (2002)





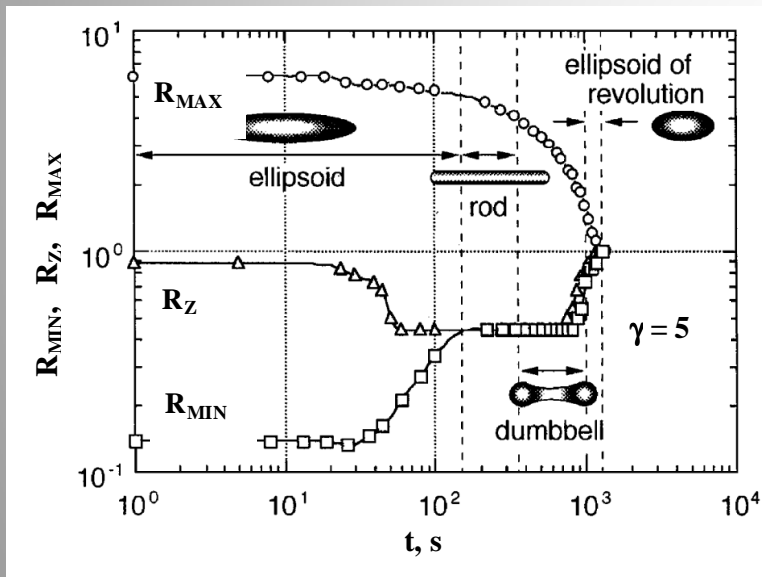
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Droplet retraction after a step strain

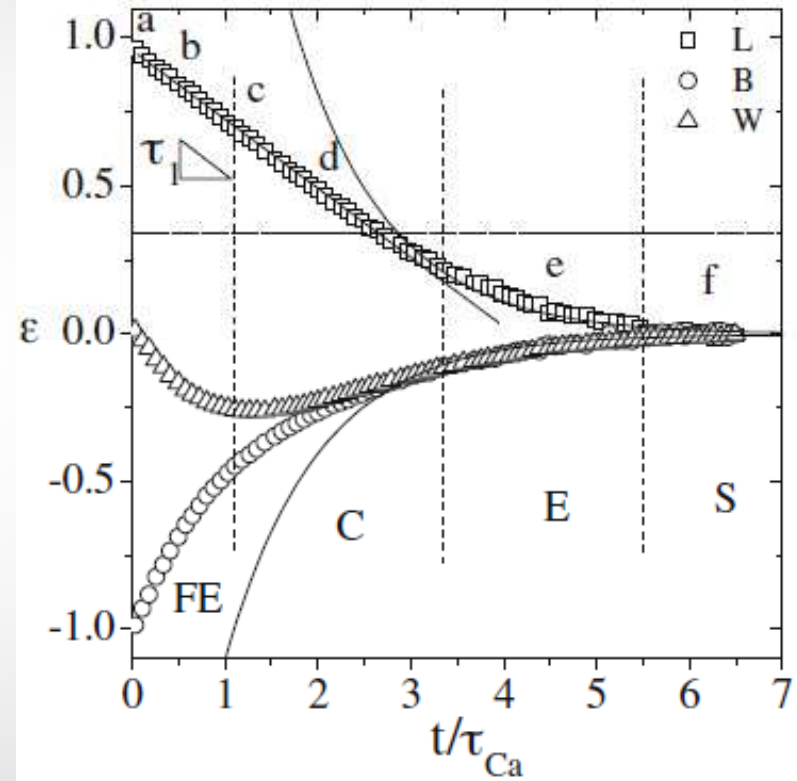


Cohen A and Carriere C J, *Rheol. Acta*, **28**, 223-232 (1989)

$$\varepsilon = \ln(L/2r_0)$$



Yamane H, Takahashi M, Hayashi R, Okamoto K, Kashihara H and Masuda T, *J. Rheol.*, **42**, 567-580 (1998)



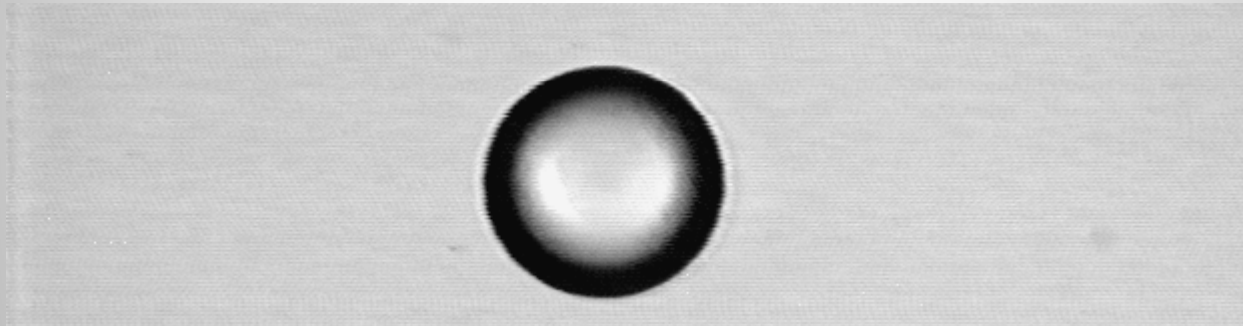
FE flat ellipsoid, C spherocylinder,
 E prolate ellipsoid, S sphere

Assighaou S and Benyahia L, *Rheol Acta*, **49**, 677-686 (2010)



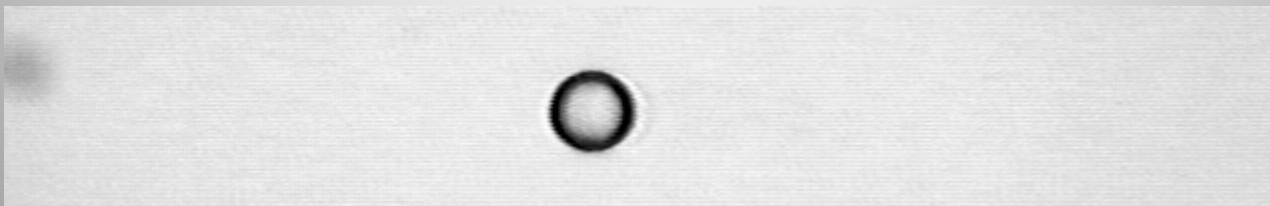
Drop breakup in shear flow

- Upon increasing Ca , a critical condition (Ca_{cr}) is reached where drop shape becomes unstable (video)



$$\lambda = 1$$

$$Ca_1 > Ca_{cr}$$



$$\lambda = 1$$

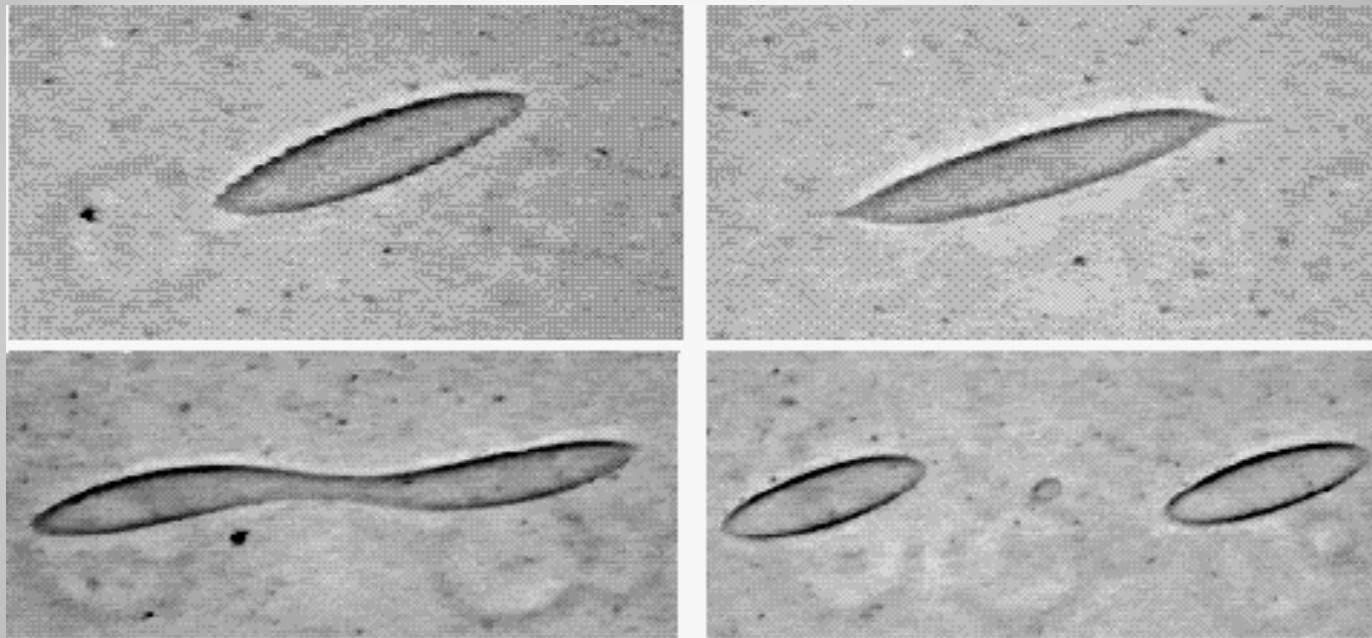
$$Ca_2 > Ca_1$$



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Drop breakup in shear flow

$$\lambda \ll 1$$



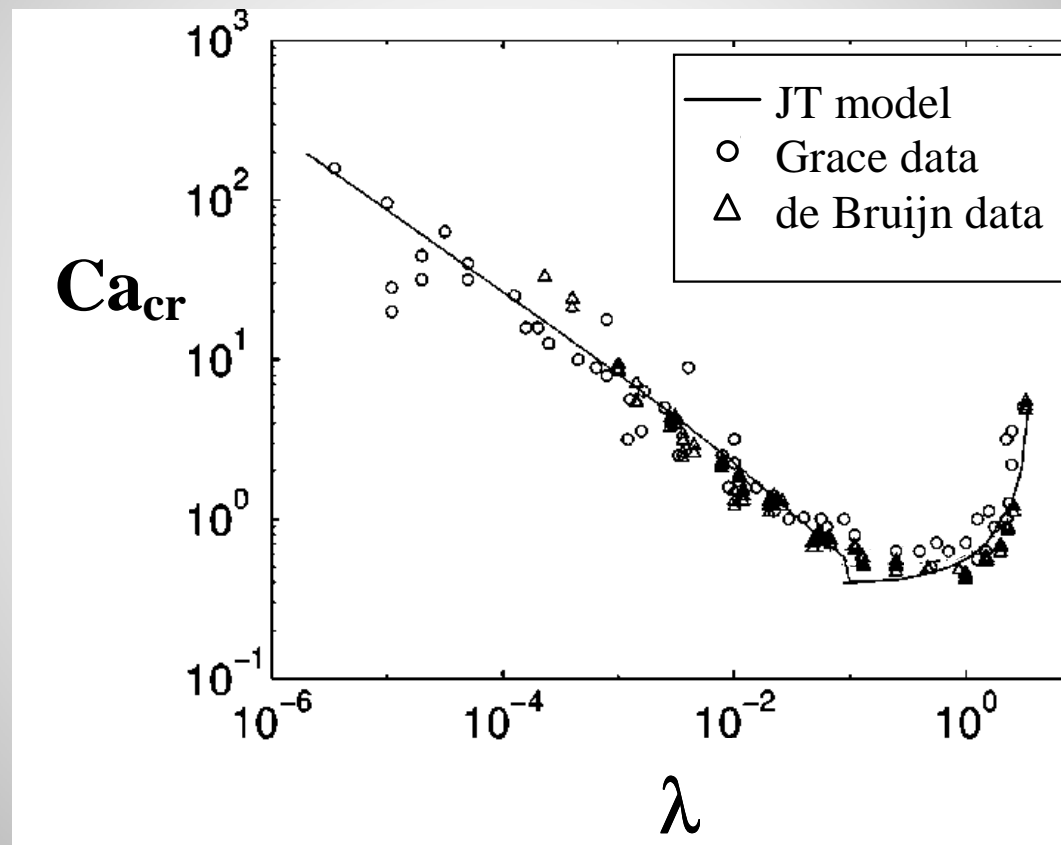
Tip streaming

Drop: Na-caseinate rich phase
Matrix: Na-alginate rich phase



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Drop breakup in shear flow



Grace H P, *Chem. Eng. Commun.*, **14**, 225-277 (1982)

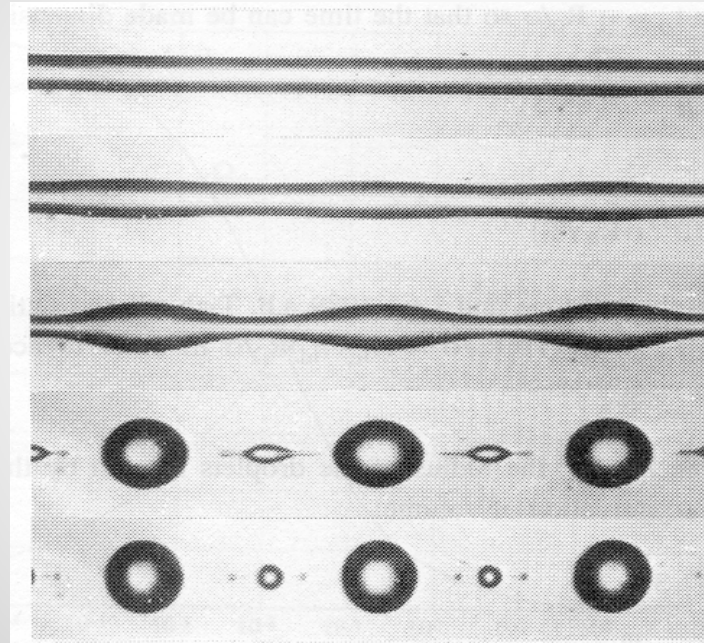
de Bruijn R A, PhD thesis, Technische Universiteit Eindhoven (1989)

Jackson N E and Tucker III C L, *J. Rheol.*, **47**, 659-682 (2003)



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Breakup of a liquid thread



Tomotika S, *Proc. R. Soc. London Ser. A*, **150**, 322-337 (1935)

Elemans P H M, Janssen J M H and Meijer H E H, *J. Rheol.*, **34**, 1311-1325 (1990)



Near critical behavior

Viscous-capillary force balance
 (inertia is negligible)

$$\eta \partial^2 v / \partial r^2 \approx \eta_d \partial^2 v / \partial z^2 \approx \sigma \partial r^{-1} / \partial r$$

Close to breakup, the external viscous shear stresses associated with thread axial motion become comparable to the internal viscous stresses associated with thread extension

$$\eta \frac{\partial v}{\partial r}$$

$$\eta_d \frac{\partial v}{\partial z}$$

$$\left\{ \begin{array}{l} \frac{\zeta}{l} \approx \sqrt{\frac{\eta_d}{\eta}} \\ w \approx \frac{\sigma}{\eta} \end{array} \right.$$

ζ characteristic axial length in the pinch-off region

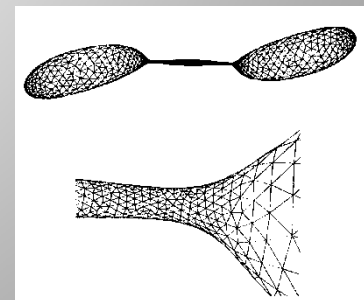
l neck radius

w characteristic velocity

$$\zeta \equiv w(t_{cr} - t) \approx (\sigma/\eta)(t_{cr} - t) \quad \longrightarrow \quad l \approx \frac{\sigma}{\sqrt{\eta\eta_d}} (t_{cr} - t)$$

Lister J R and Stone H A, *Phys Fluids*, **10**, 2758-27 (1998)

Blawdziewicz J, Cristini V and Loewenberg M, *Phys Fluids*, **14**, 2709-2718 (2002)

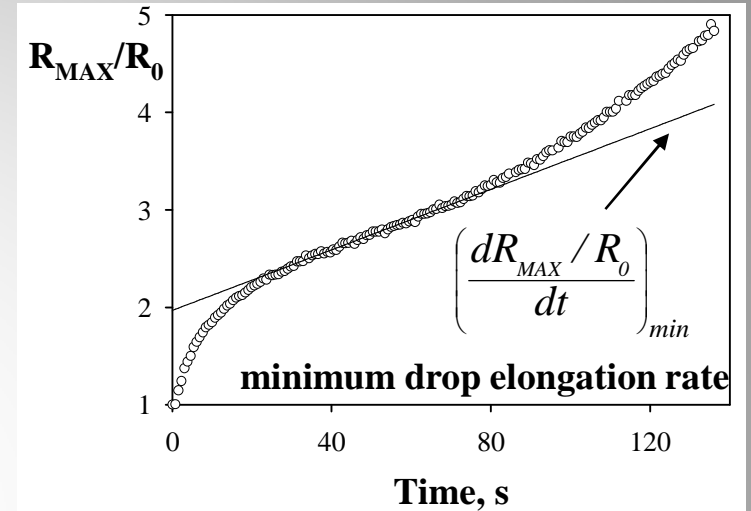
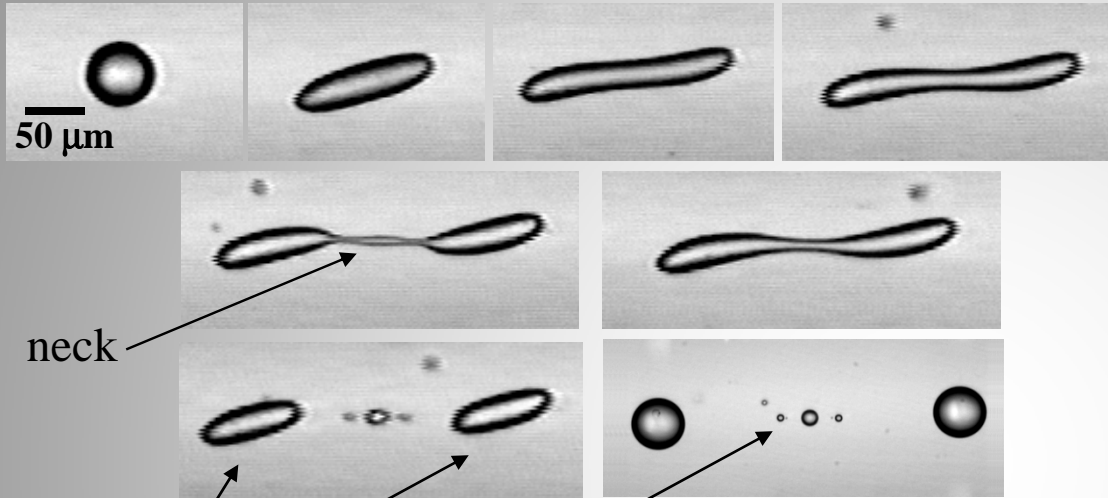




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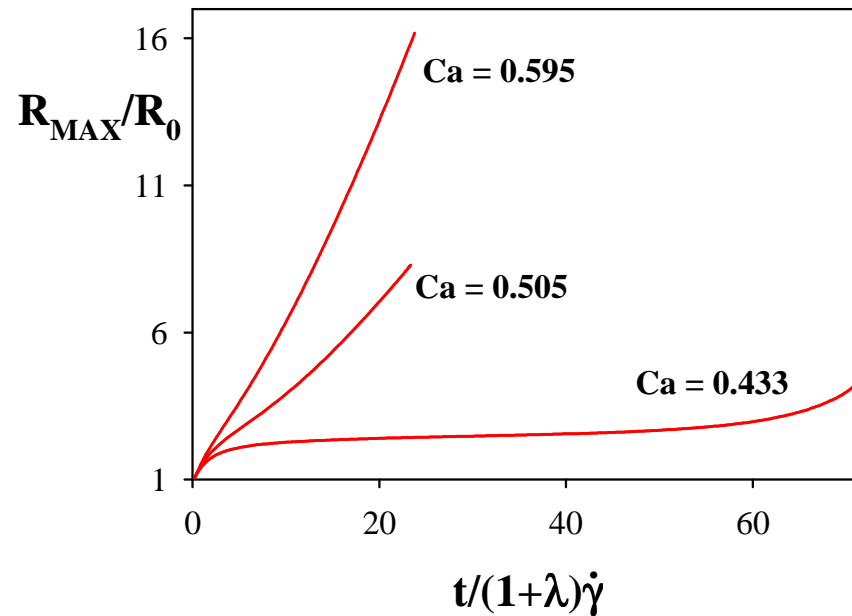
Breakup kinetics

$\lambda = 1$ $Ca = 0.50$



daughters

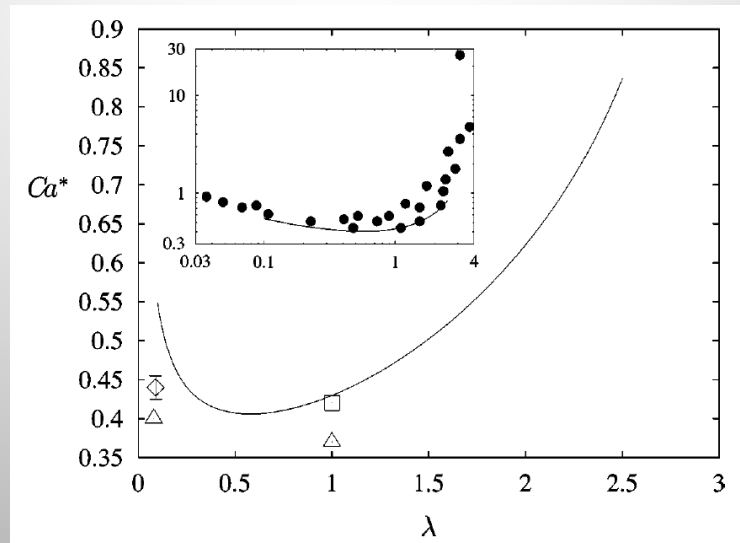
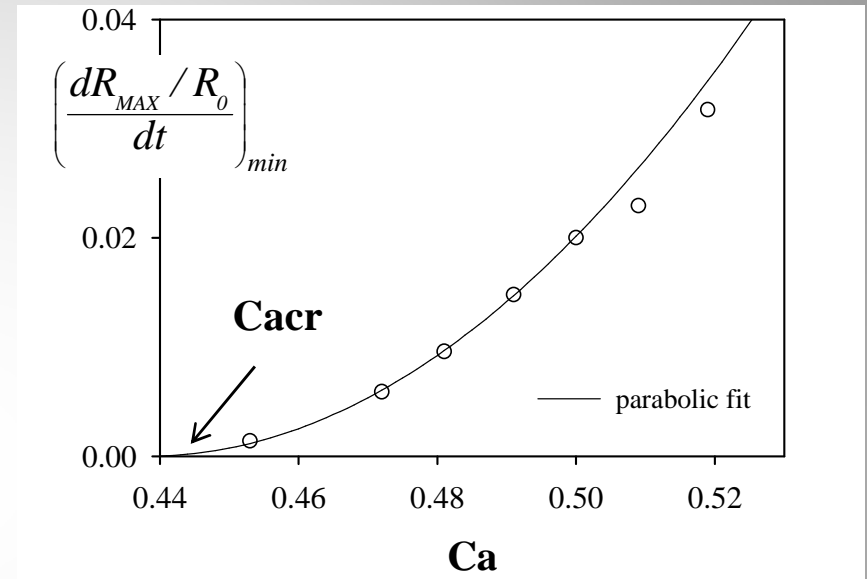
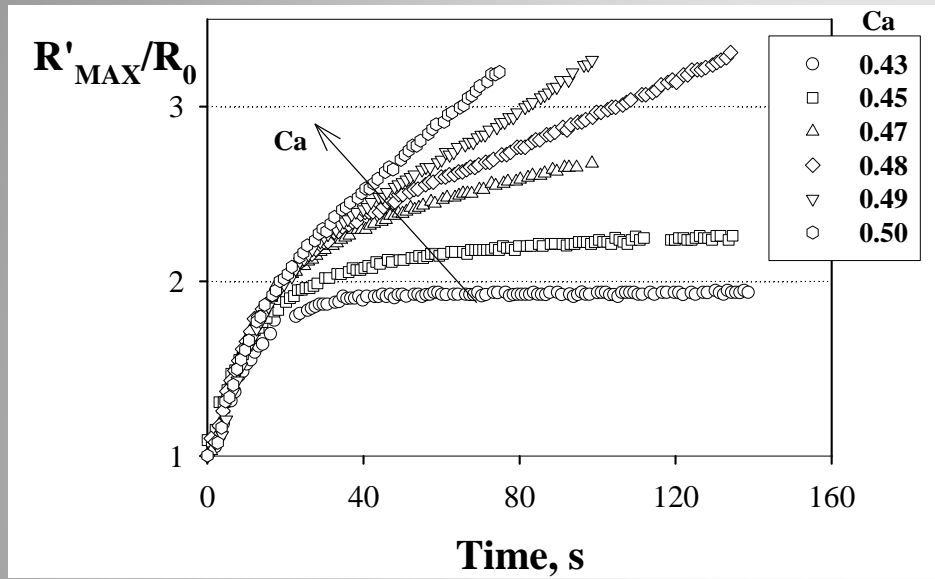
satellites



Cristini V, Guido S, Alfani, Blawdziewicz J and Loewenberg M, *J. Rheol.*, **47**, 1283-1298 (2003)



Determination of Ca_{cr}

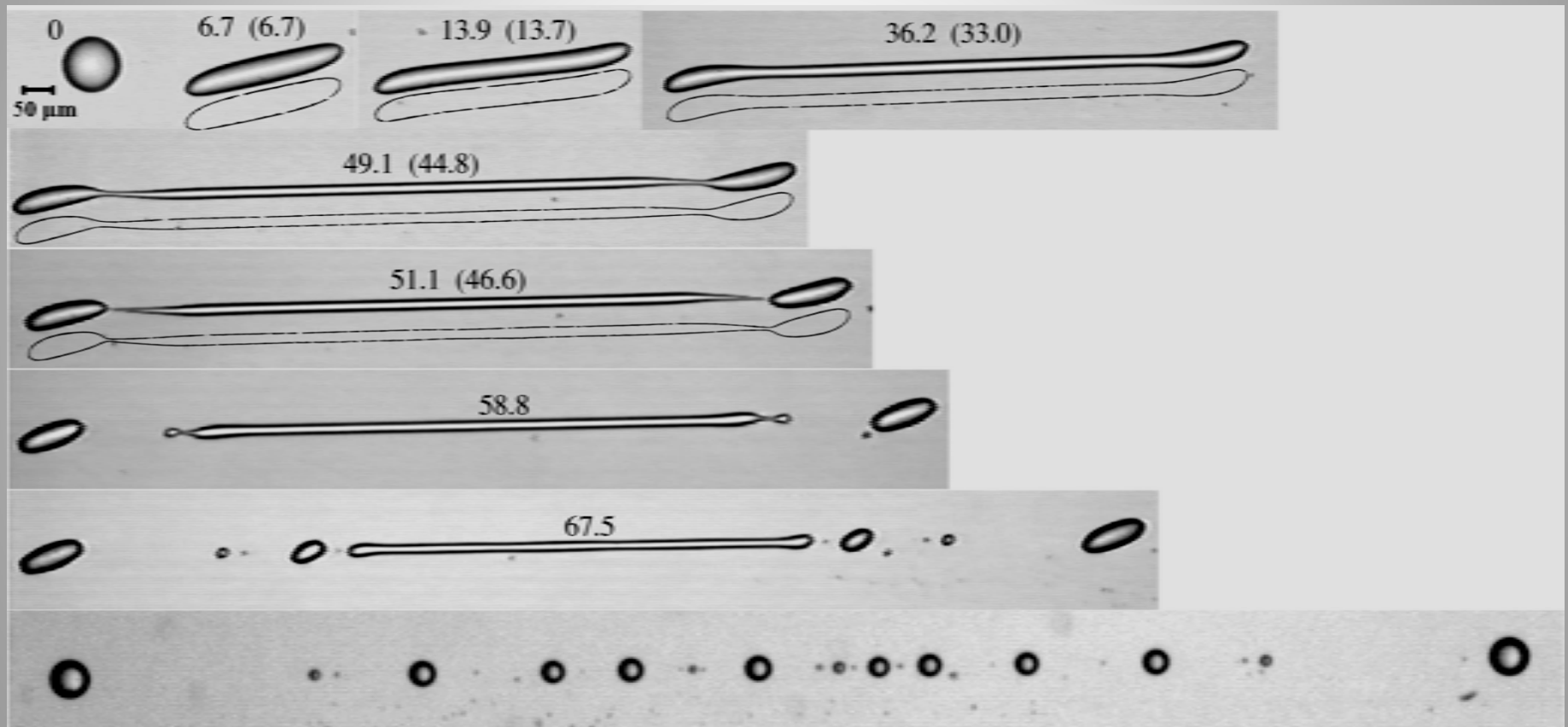


(●) [Grace (1982)]



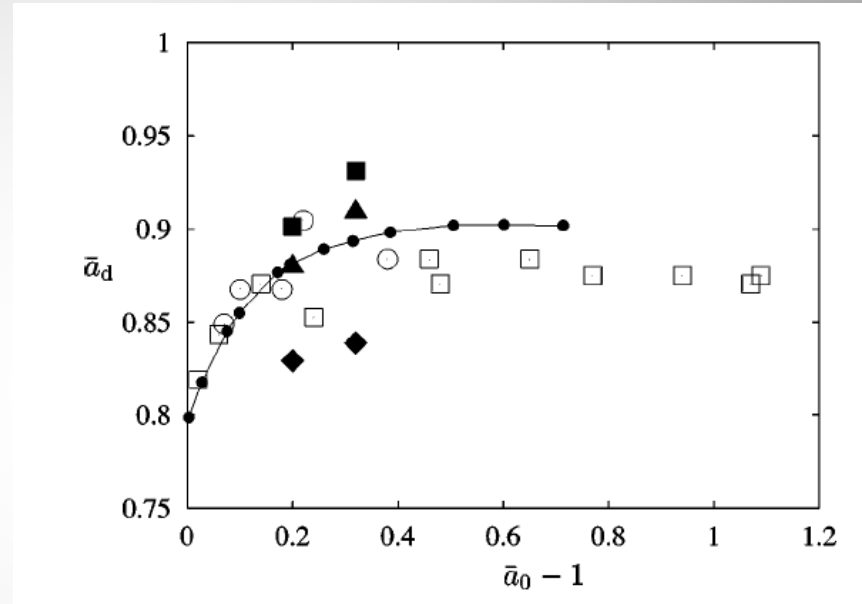
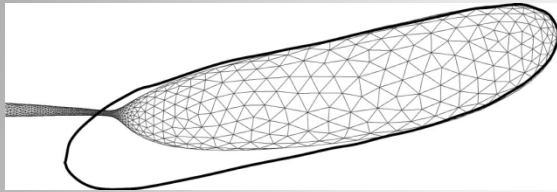
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Shape evolution for $Ca/Cacr = 1.38$





Daughter drop scaling



$\lambda = 0.094$ (\square), $\lambda = 0.5$ (\triangle), $\lambda = 1.02$ (\circ), $\lambda = 2$ (\diamond)

Daughter drop size $\left\{ \begin{array}{l} \text{independent of initial size} \\ \text{scales with critical drop size} \end{array} \right.$



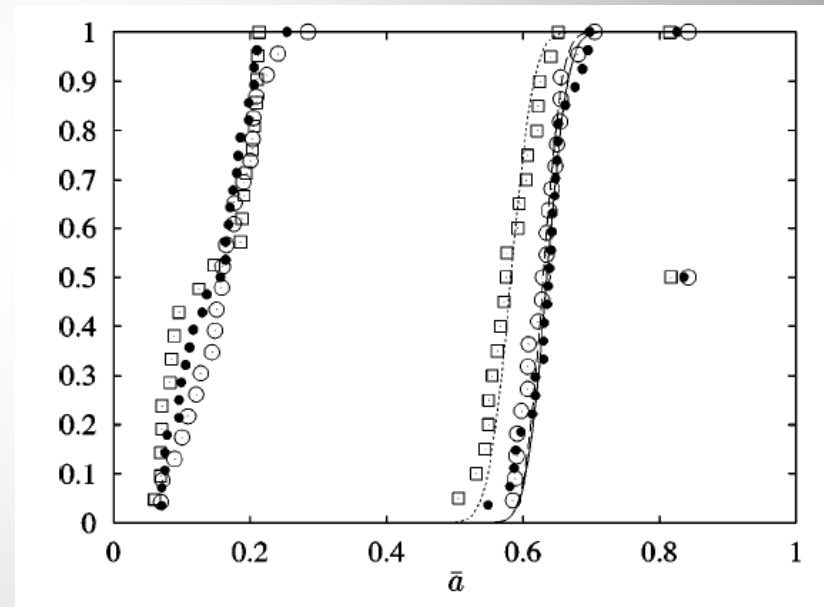
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Drop fragment distribution

Cumulative size distribution



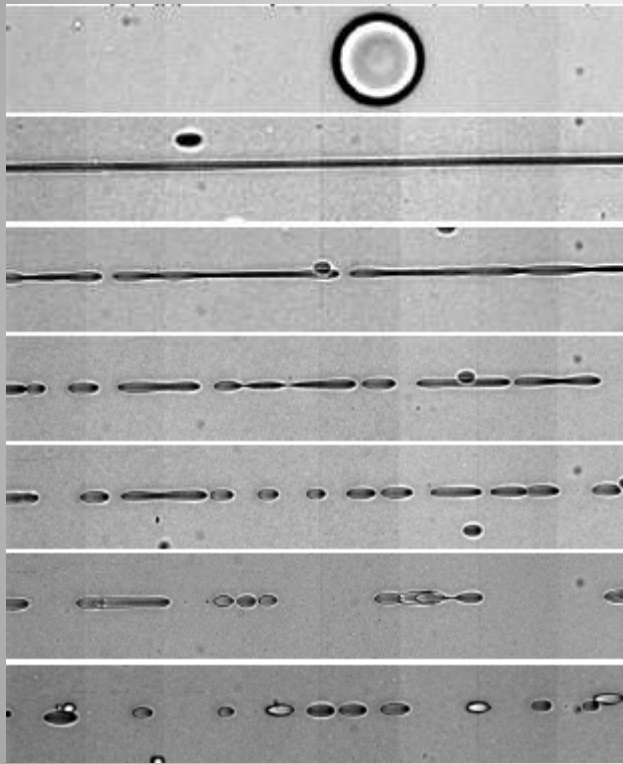
The distributions for each experiment show two distinct daughter drops and three size classes of satellite drops



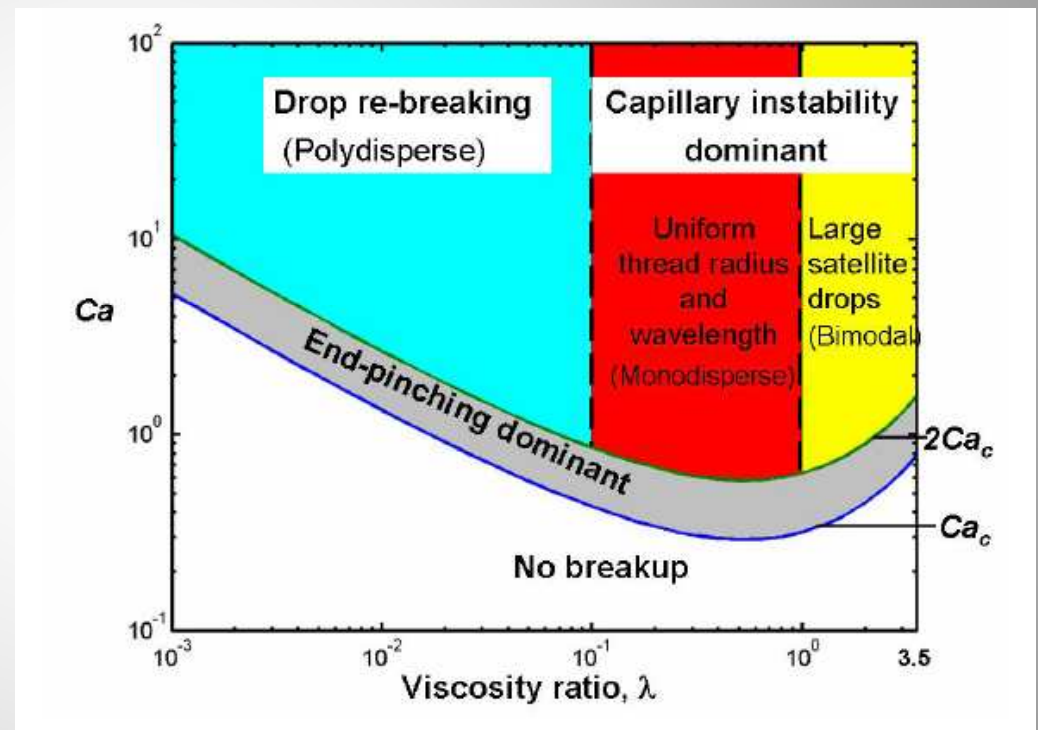
$\lambda = 0.094$; $\bar{a}_0 = 1.22$ (\diamond), $\bar{a}_0 = 1.44$ (\triangle), $\bar{a}_0 = 1.73$ (\square), $\bar{a}_0 = 1.90$ (\circ), $\bar{a}_0 = 2.02$ (\bullet), $\bar{a}_0 = 2.04$ (\otimes).



Drop fragment distribution



$$\lambda = 0.075, Ca = 4.5 Ca_{cr}$$



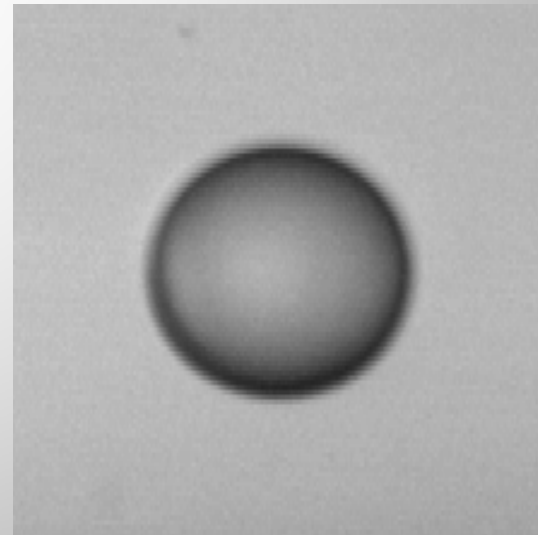


Non-Newtonian effects

- ◆ Liquid-liquid dispersions are *always* viscoelastic systems, due to interfacial tension
- ◆ Our aim is to study the effect of the *intrinsic elasticity* of the fluid components on flow-induced morphology
- ◆ Model fluids: constant viscosity, highly elastic liquids (Boger fluids)

$$\mathbf{Ca} = \mathbf{0.4}$$

**Outer phase: viscoelastic fluid , inner
phase: Newtonian, $\lambda = 1$**





Small deformation theory with elastic fluids

- Constitutive equation of component liquids: second-order fluid

Here **either the outer or the inner phase is non-Newtonian (the other being Newtonian)** \rightarrow 2 additional physical quantities:
 normal stress differences $N_1 = \Psi_1 \dot{\gamma}^2$
 $N_2 = \Psi_2 \dot{\gamma}^2$

- New nondimensional parameters:

$$\mu = \frac{\Psi_2}{\Psi_1} \quad W = \frac{r_0 \Psi_1 \dot{\gamma}^2}{2\sigma}$$

- Small deformation conditions

$$Ca \ll 1$$

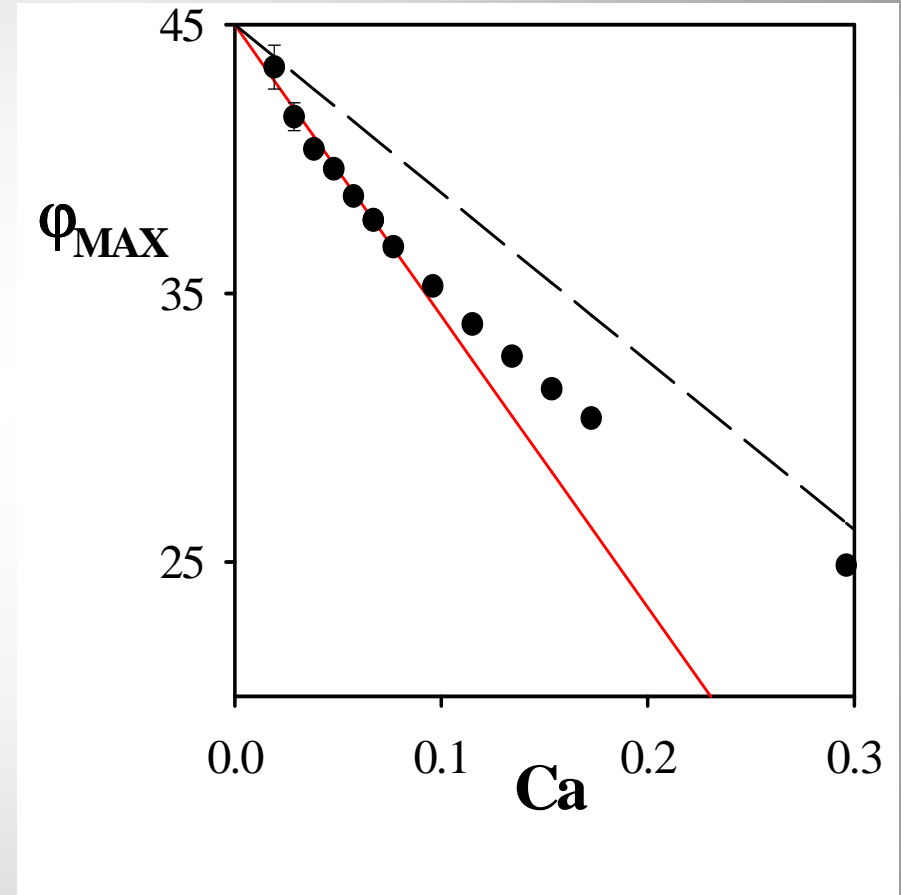
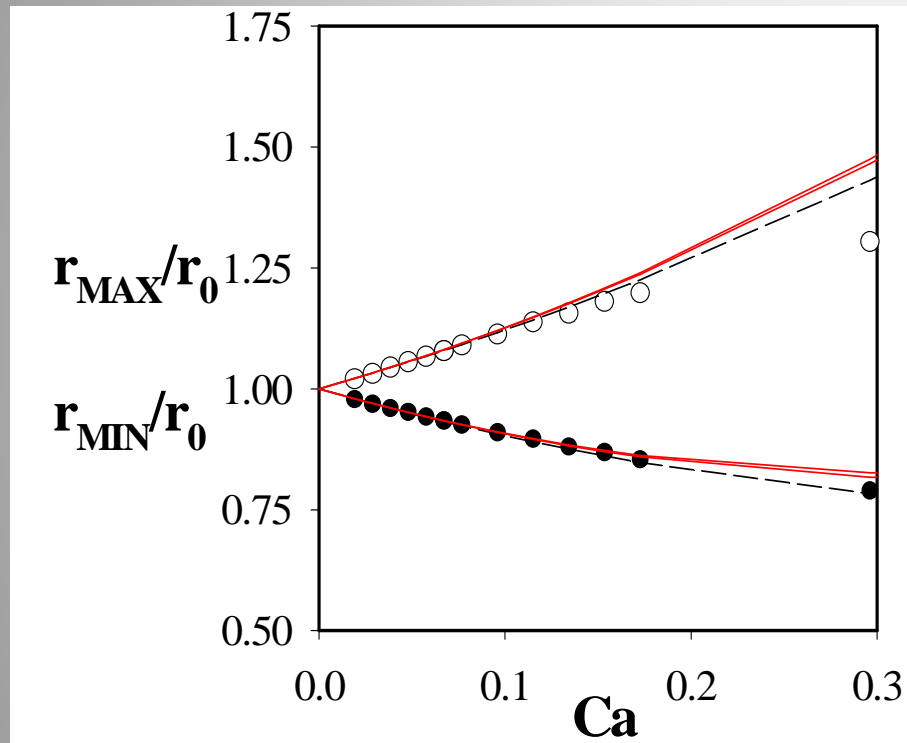
$$W \ll 1$$

Observable non-Newtonian effects \leftrightarrow

$$p \equiv \frac{W}{Ca^2} = \frac{\psi_1 \sigma}{2r_0 \eta^2} \approx 1$$



Comparison with experiments



$$\lambda = 1$$

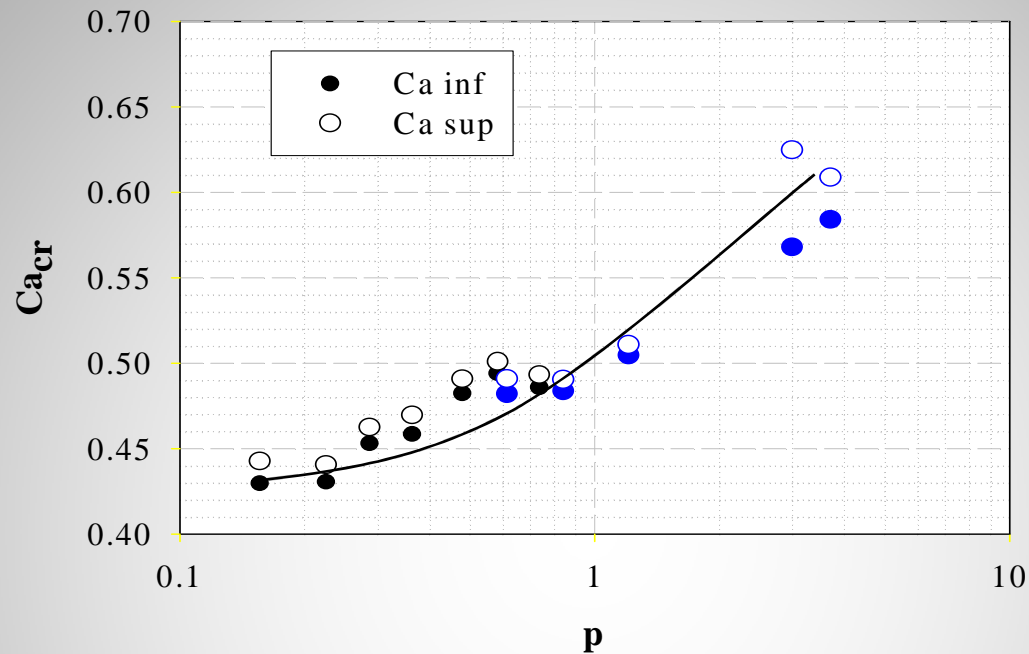
$$p = 1.8$$

Dashed: Newtonian

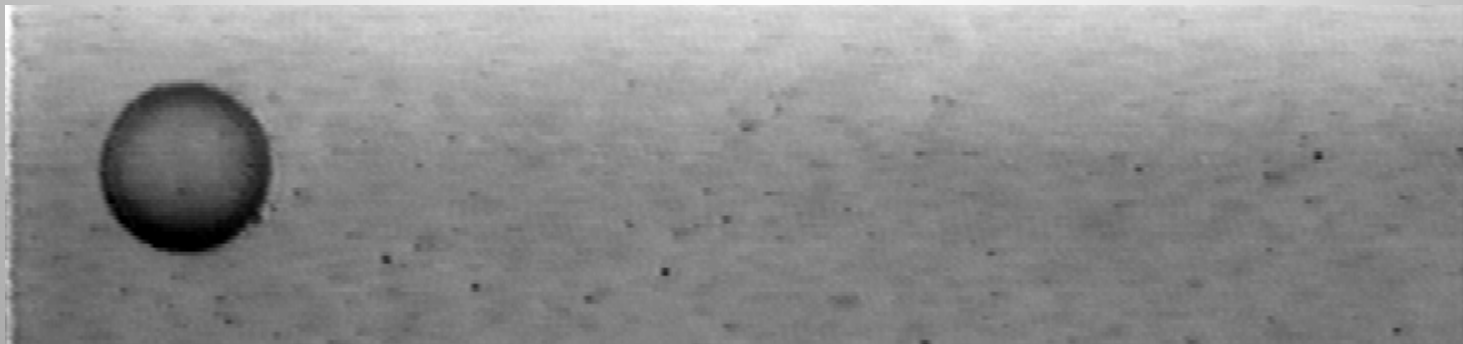
Continuous: non-Newtonian



Breakup



Outer phase: viscoelastic, drop: Newtonian, $\lambda = 0.6$



Outer phase: Newtonian, drop: viscoelastic, $\lambda = 2.6$

Drop breakup is hindered by elasticity of the fluid components



Wall effects

Shape parameters

$$\frac{L}{2R_0}$$

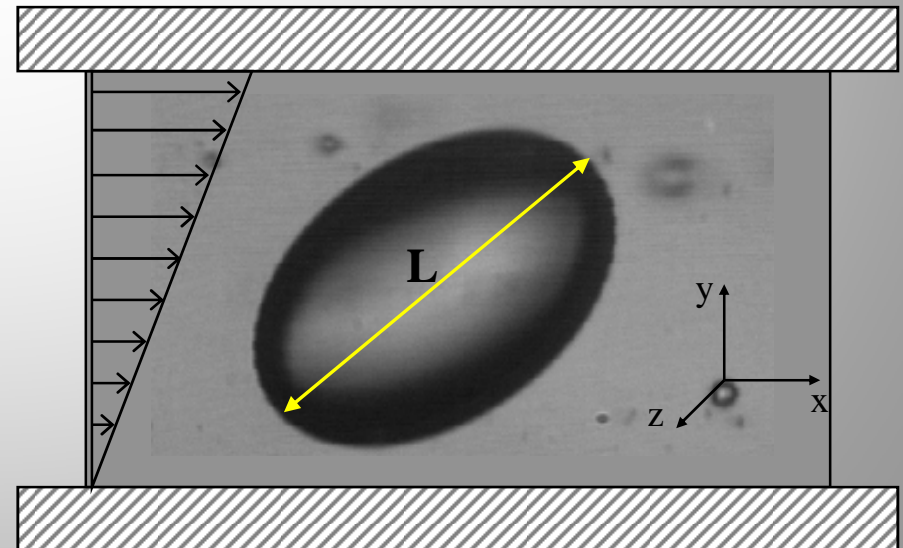
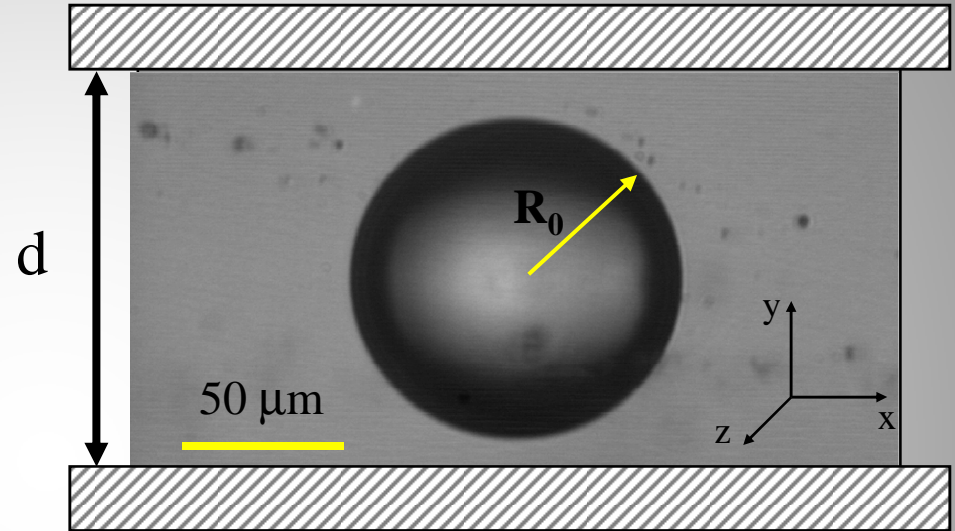
Nondimensional
major axis

$$\frac{d}{2R_0}$$

Nondimensional
gap

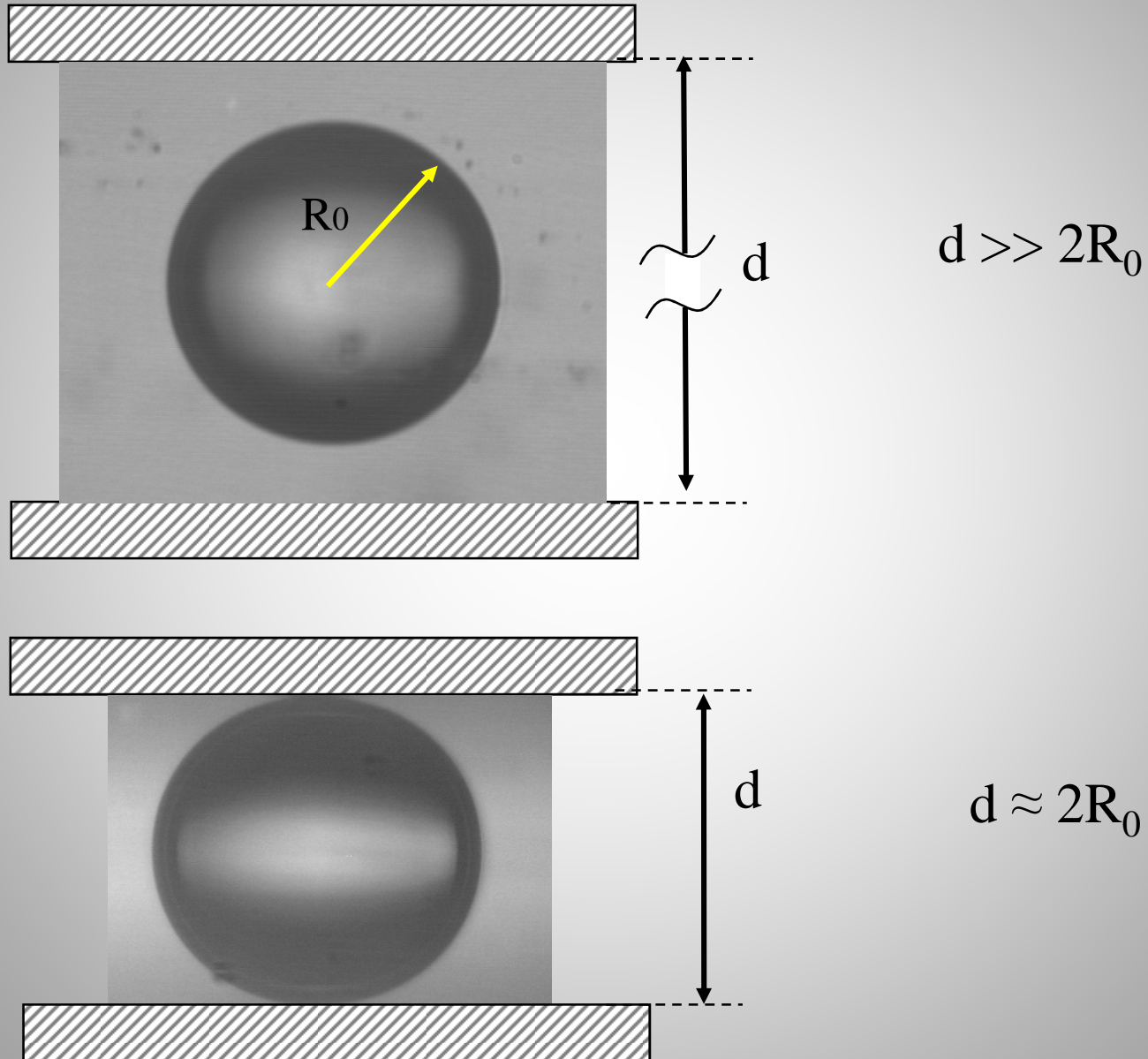
$$D = D_T \left[1 + \left(\frac{2R_0}{d} \right)^3 \frac{1 + 2.5\lambda}{1 + \lambda} \right]$$

Shapira M and Haber S, *Int. J. Multiphase Flow*, **16**, 305 (1990)



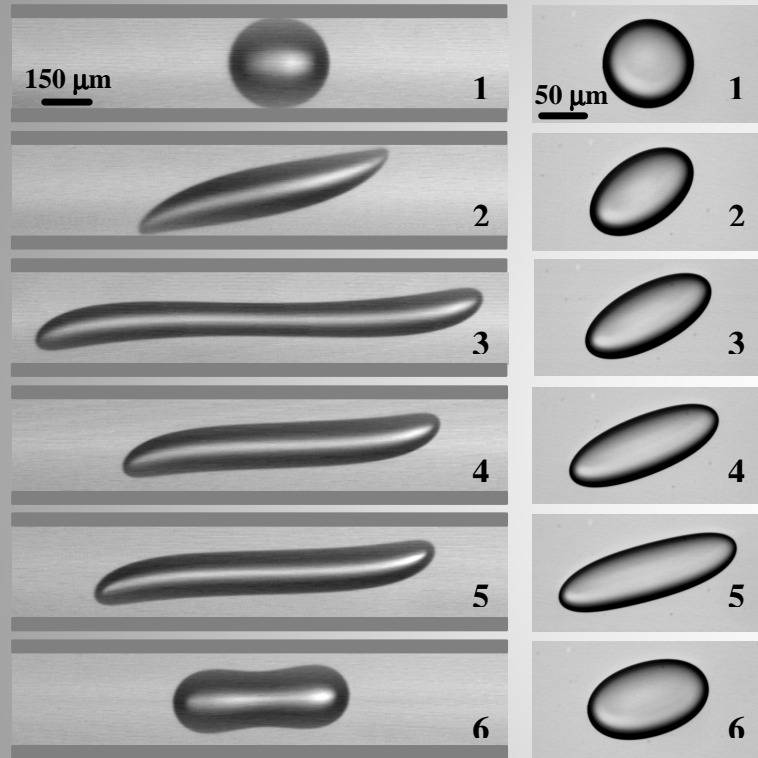


Wall effects



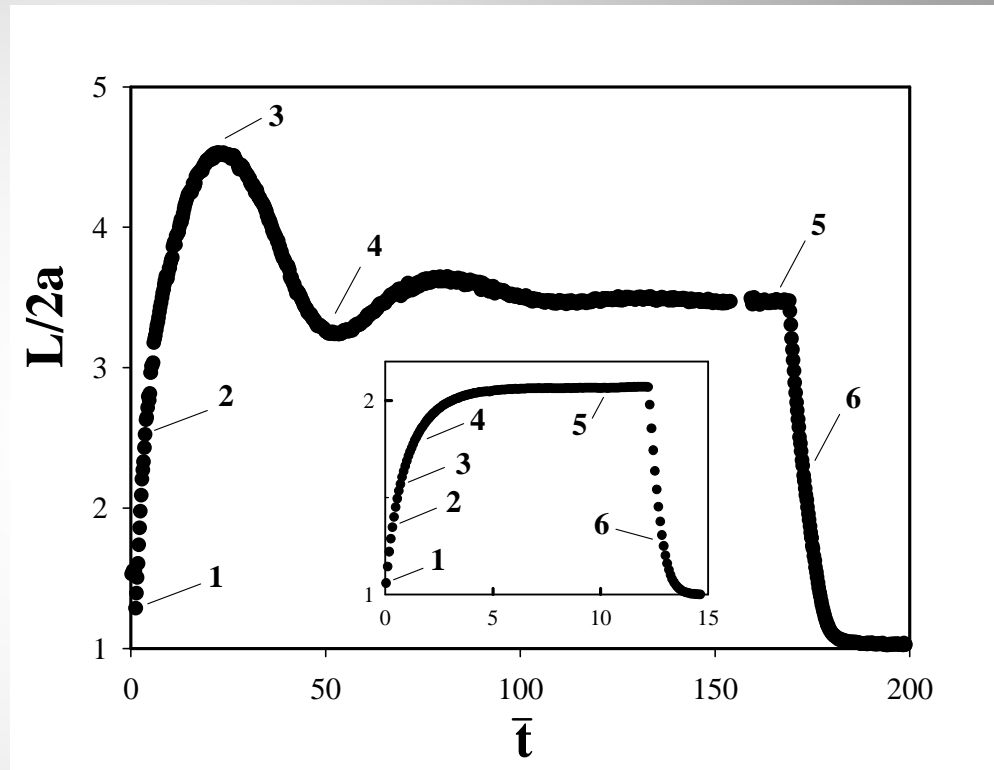


Wall effects



$$d \approx 2R_0$$

$$d \gg 2R_0$$



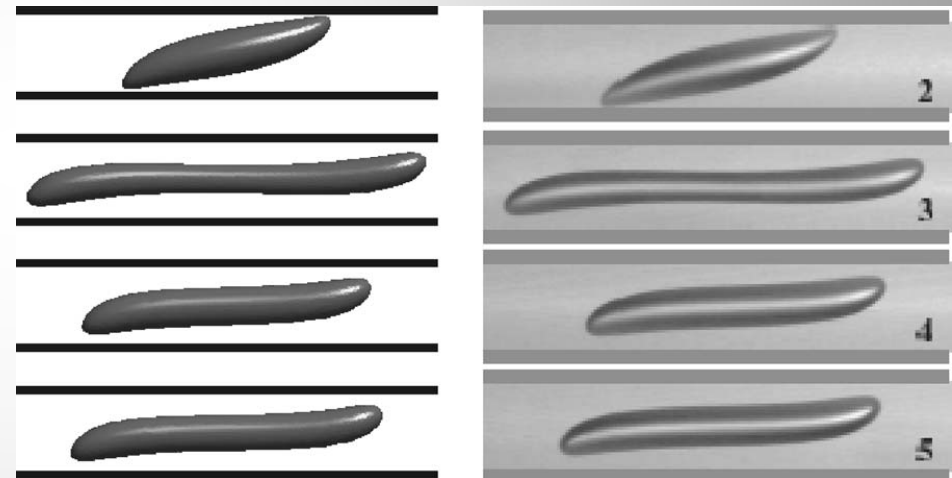
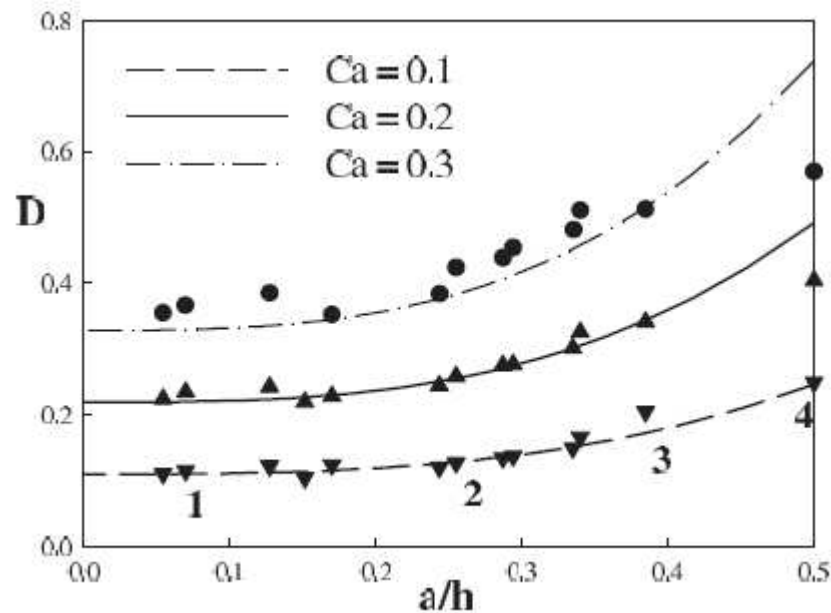
$$Ca = 0.4, \lambda = 1$$



Wall effects

Comparison with theoretical predictions
(left) and numerical simulations (right)

Ca = 0.1



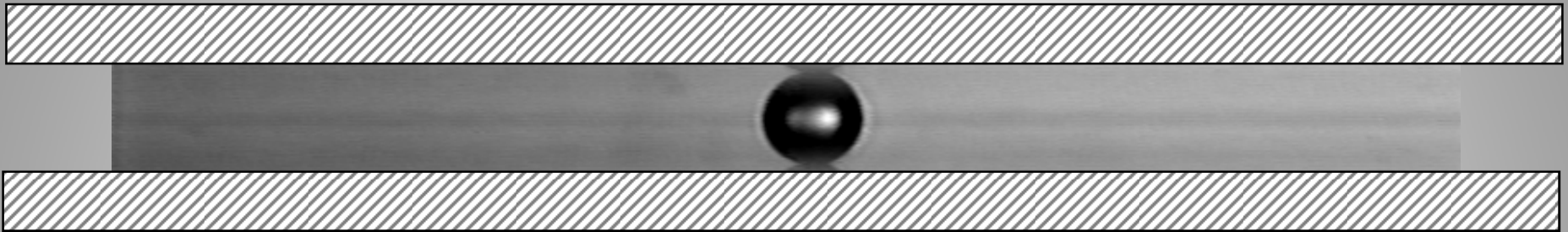
Simulations: Janssen P J A and Anderson P D, *Phys Fluids*, 19, 043602 (2007)

Theory: Shapira M and Haber S, *Int. J. Multiphase Flow*, 16, 305 (1990)

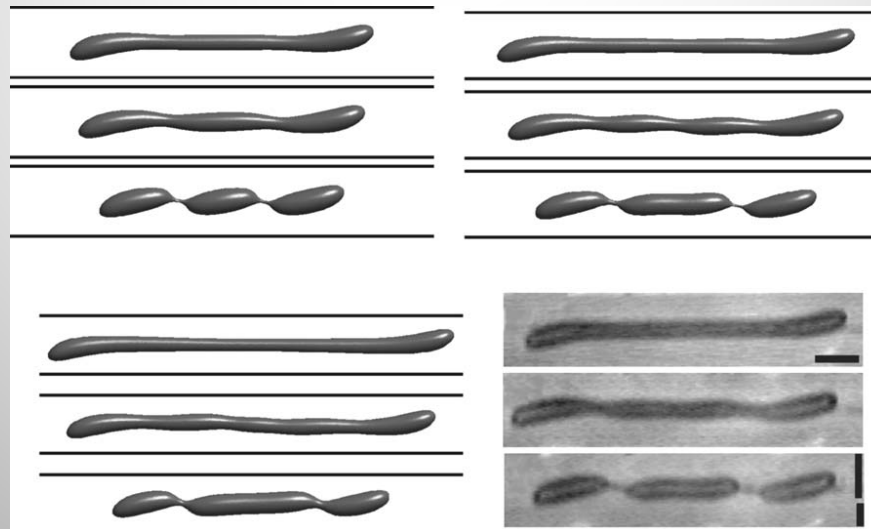


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Wall effects

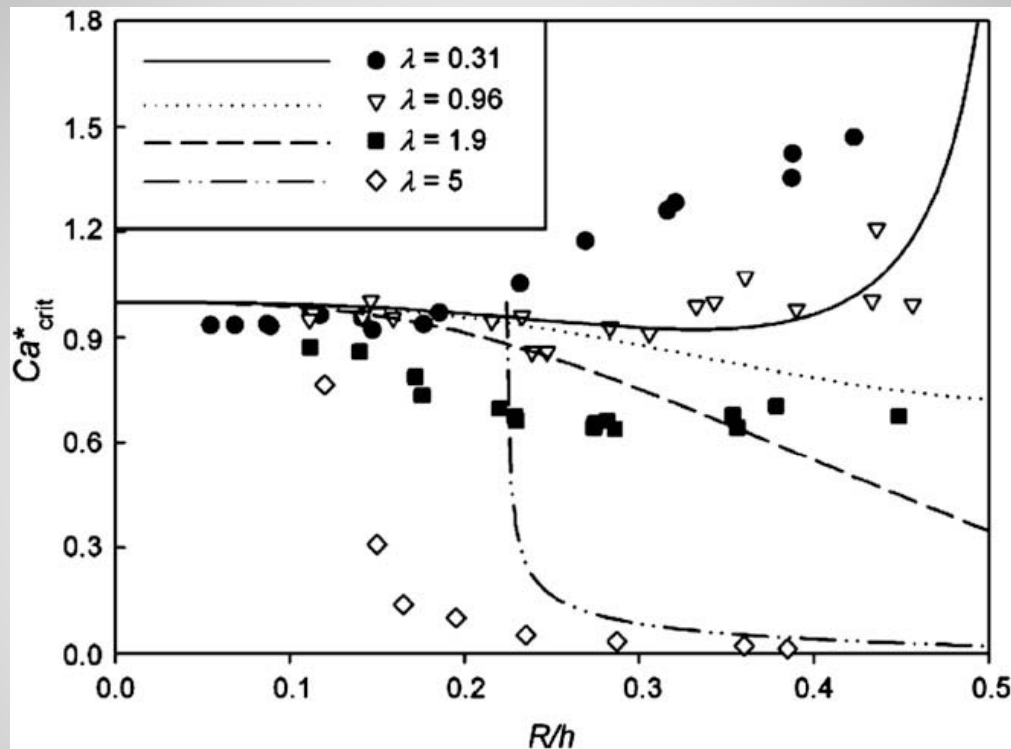


Wall effects act to stabilize drop shape at $\lambda = 1$





Wall effects

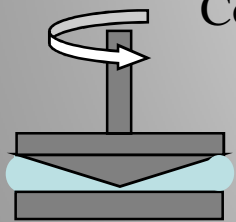
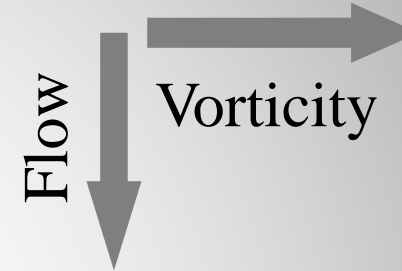
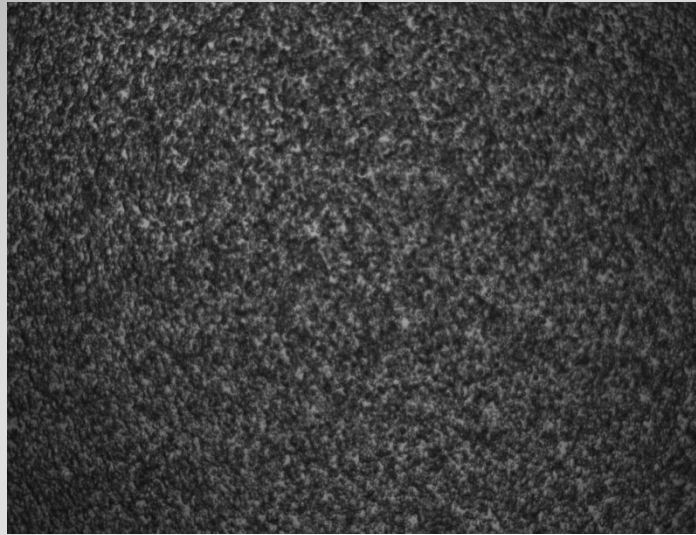
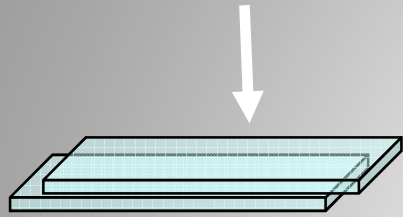


Even at $\lambda > 4$ (no breakup in unbounded shear flow), droplets can still be broken in confined conditions

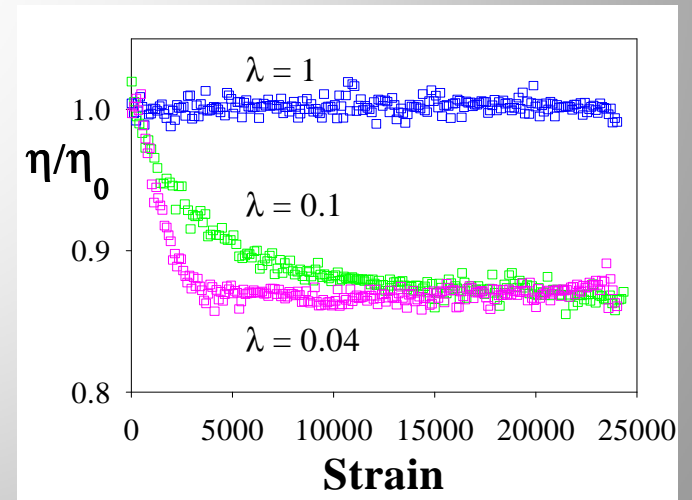
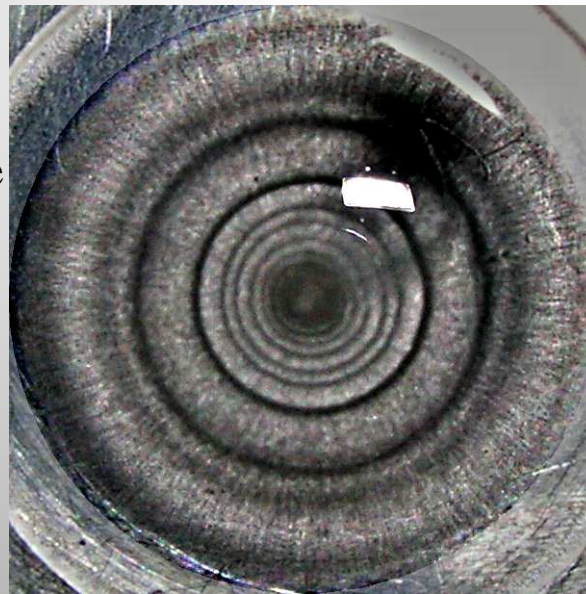


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Wall effects: Shear banding



Cone and plate
rheometer

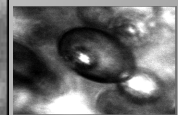
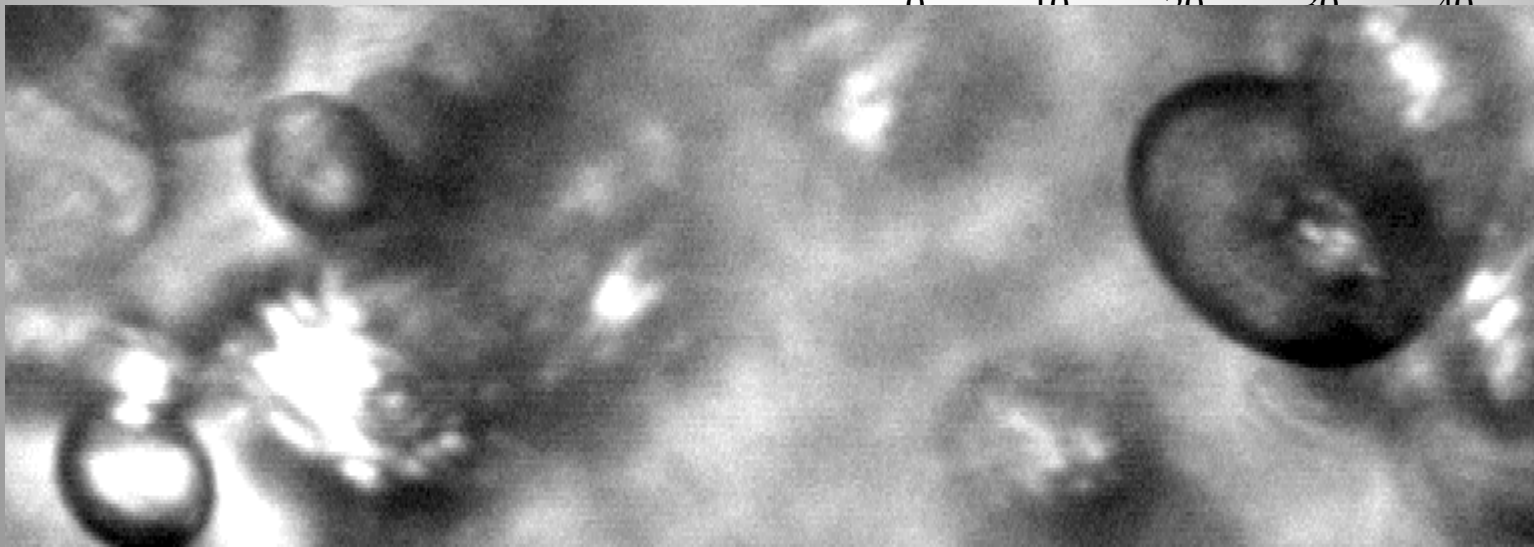
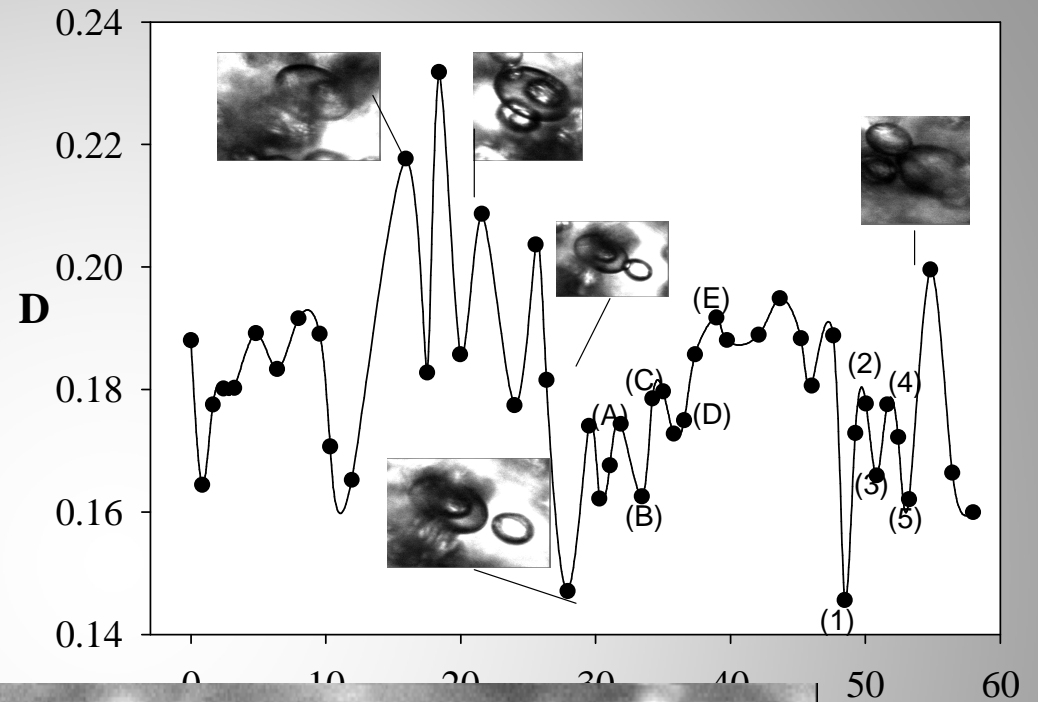


Caserta S, Simeone M, and Guido S, *Phys. Rev. Lett.*, **100**, 137801 (2008)

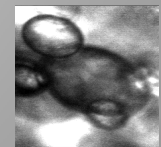


Shape fluctuations due to drop interactions

Concentrated systems



(E)

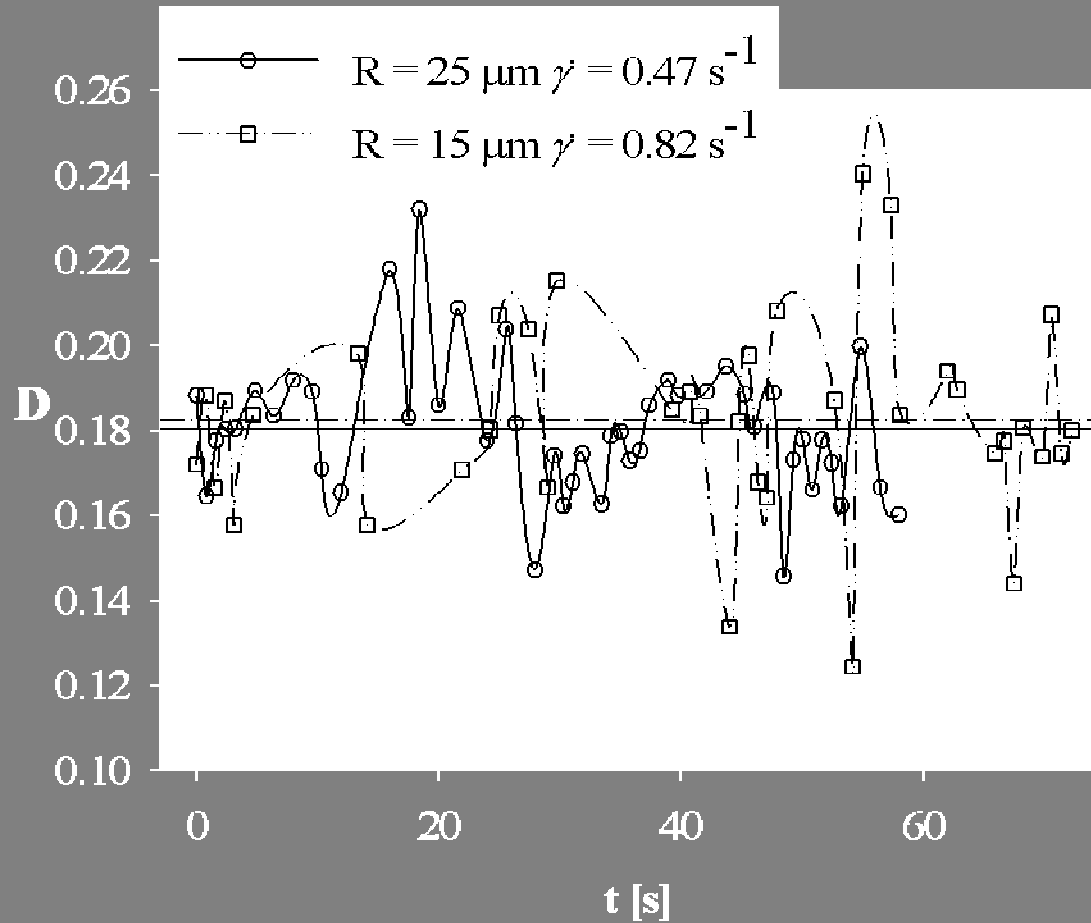


(5)



Concentrated systems

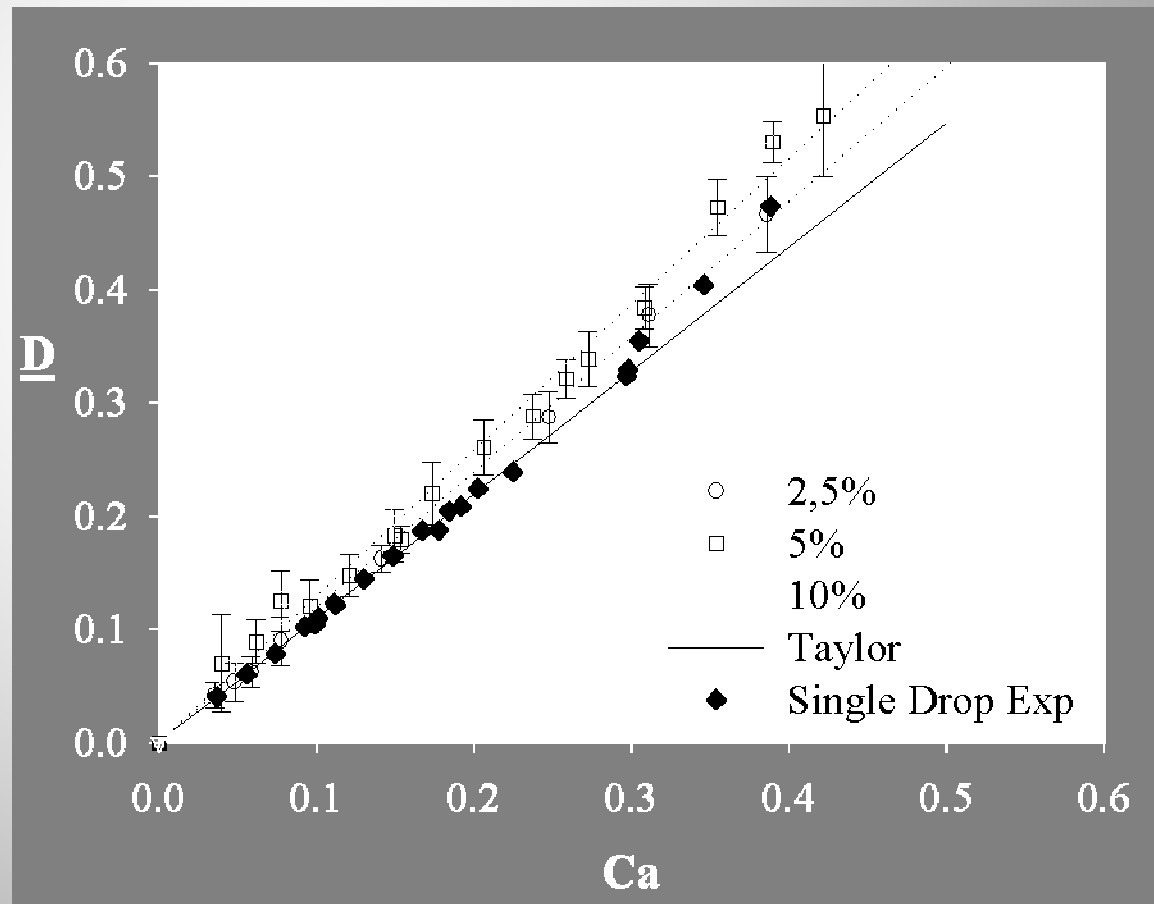
Ca = 0.15



The time-averaged value of D depends on Ca only



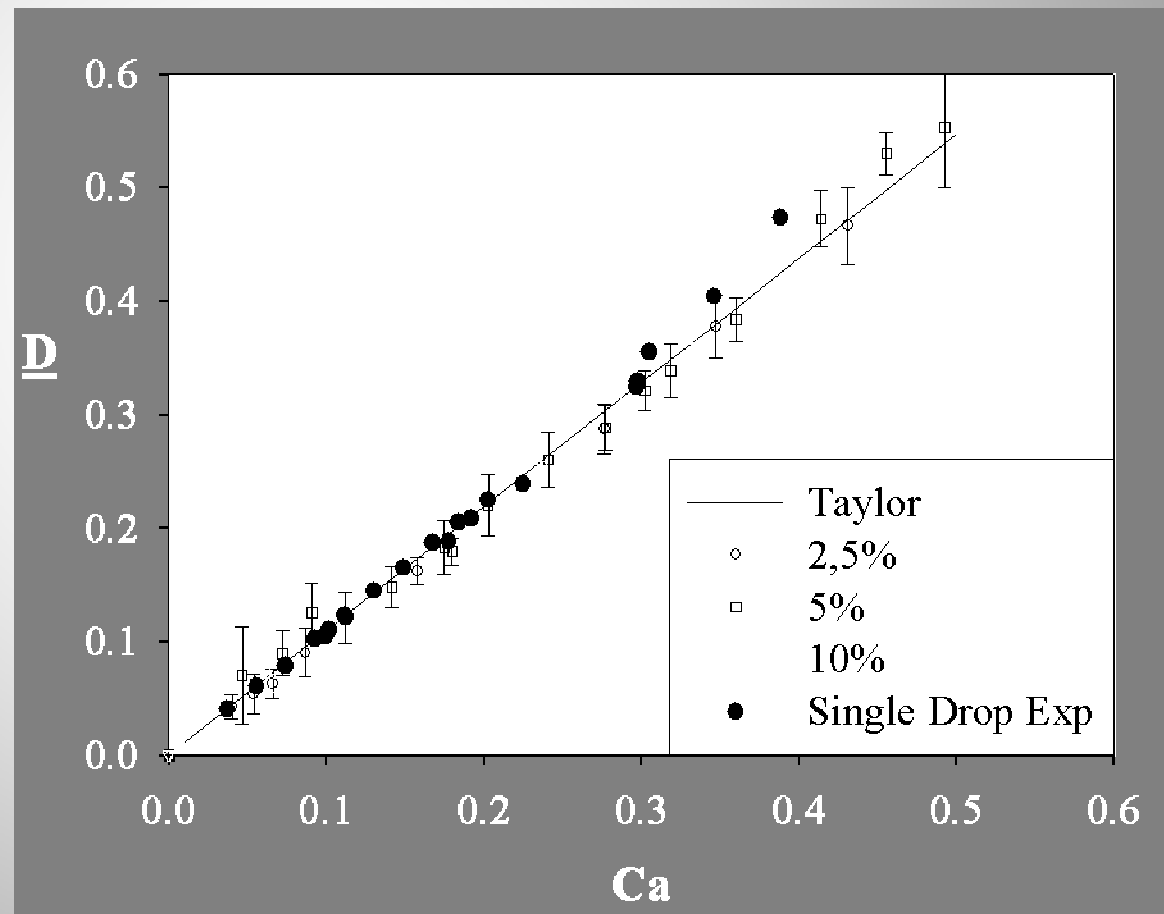
Concentrated systems





Concentrated systems

“Mean field” scaling with blend viscosity



Jansen K M B, Agterof W G M, Mellema J, *J Rheol*, **45**, 227-236 (2001)
Caserta S, Reynaud S, Simeone M and Guido S, *J Rheol*, **51**, 585-774 (2007) (data shown here)



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Conclusions

- Drop shape up to moderate deformations is essentially ellipsoidal and is well represented by small deformation theories and phenomenological models
- Numerical simulations are in good agreement with experiments up to breakup
- Drop fragments distribution can be estimated if the original distribution is known
- Single drop deformation and breakup results can be applied to concentrated systems by using a “mean field” scaling
- Wall effects stabilize drop shape and elicit shear banding phenomena
- Open issue: effects of surfactants