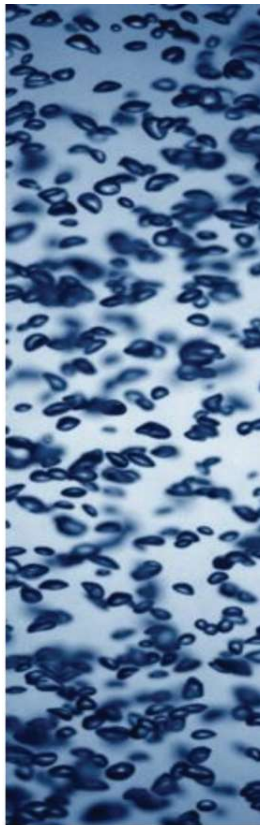
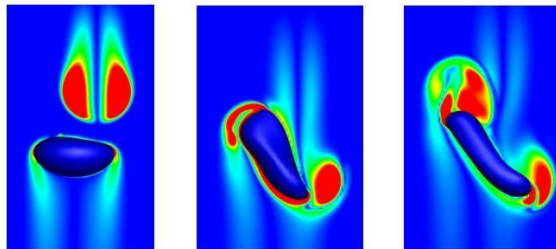
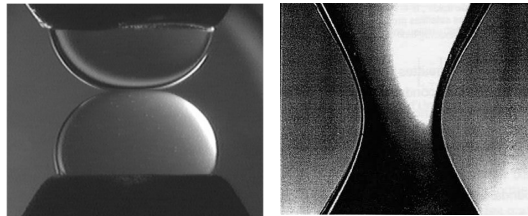


Coalescence & Breakup in emulsions and bubbly flows



Bubble column



Liquid-liquid
pipe flow

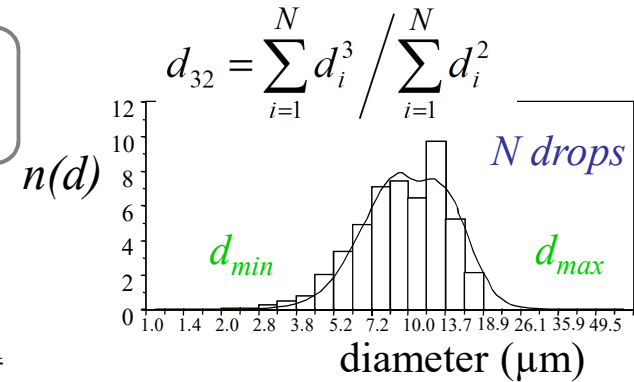


Evolution of interfacial area

Interfacial phenomena make evolve the **size distribution** of droplets, bubbles or particles in a given flow.

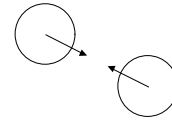
Sauter diameter:
$$d_{32} = \frac{\int_{d_{\min}}^{d_{\max}} n(d) d^3 \delta d}{\int_{d_{\min}}^{d_{\max}} n(d) d^2 \delta d}$$

number density function



$d_{32} \uparrow$

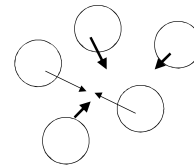
Coalescence



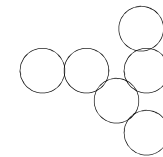
Drops or bubbles



Aggregation

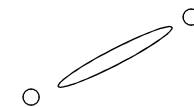
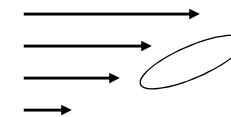
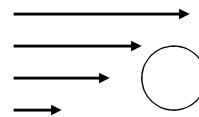


Particles

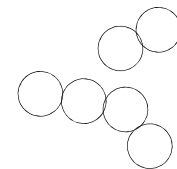
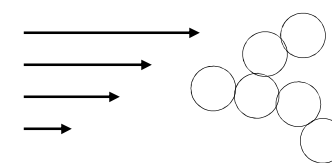
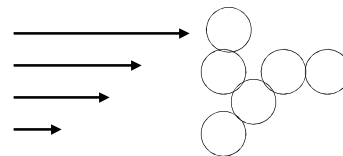


$d_{32} \downarrow$

Breakup (drop or bubble)

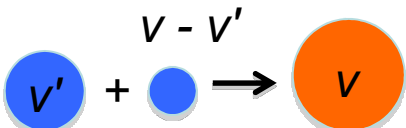


Breakup (aggregate)

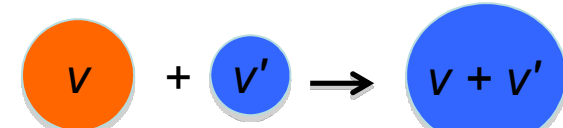


Formulation of the Population Balance Equation (PBE)

Equation of **balance on the number density n of particles v**
 in a perfectly mixed zone (i.e. *with homogeneous hydrodynamics*),
 without mass transfer (i.e. *without growing/dissolution of the particles*):



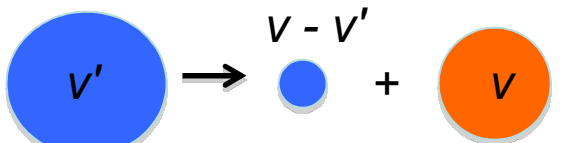
$v' + v - v' \rightarrow v$



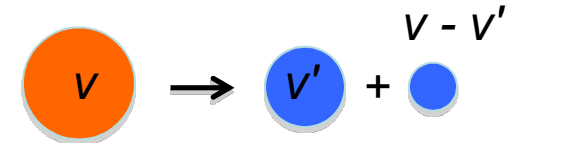
$v + v' \rightarrow v + v'$

$$\frac{\partial n(v, t)}{\partial t} = \underbrace{\frac{1}{2} \int_0^v n(v-v', t) n(v', t) Q(v-v', v') dv'}_{\text{Birth by aggreg./coalesc.}} - \underbrace{n(v, t) \int_0^\infty n(v', t) Q(v, v') dv'}_{\text{Death by aggreg./coalesc.}}$$

$$+ \underbrace{\int_v^\infty \beta(v, v') \Gamma(v') n(v', t) dv'}_{\text{Birth by breakup}} - \underbrace{\Gamma(v) n(v, t)}_{\text{Death by breakup}}$$



$v' \rightarrow v - v' + v$



$v \rightarrow v' + v - v'$

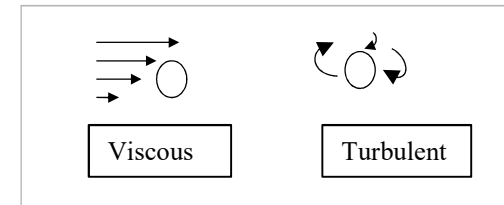
NB: Case of heterogeneous flows: coupling of CFD (Two fluid approach) & PBE
 → Birth/death terms are introduced in a transport equation for $n(v, t)$

Engineering approach

Time evolution of the size distribution: depends on the *local hydrodynamics* and the *physico-chemical properties* of the two phases and the interface.

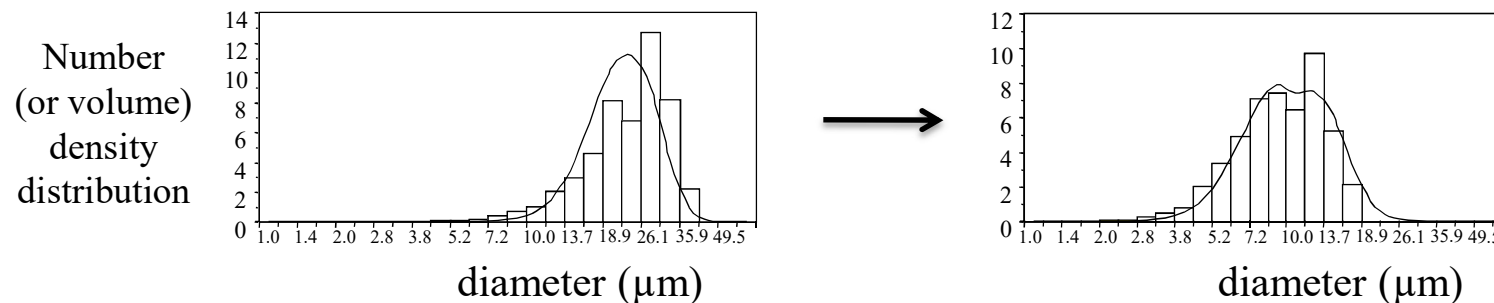
1) Existing phenomena in the device: **breakup** or/and **coalescence**?

2) Determine the cause of deformation/collision.
→ physics at the particle scale



3) Select the accurate **kernel** for the Population Balance Equation (PBE)
→ model for frequency and size distribution

4) Solve the PBE to simulate the time evolution of the population

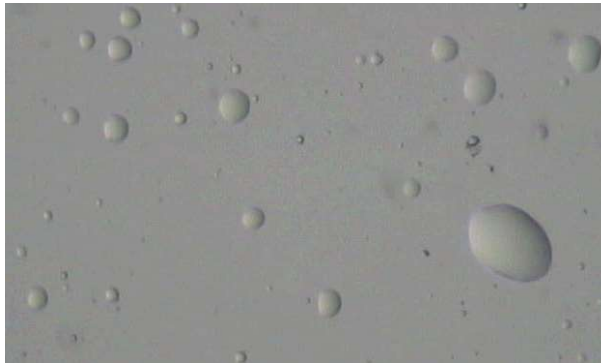


The interfacial tension: a mechanical property of the interface

Interfacial tension → resisting force to surface deformation

Its importance on interfacial phenomena

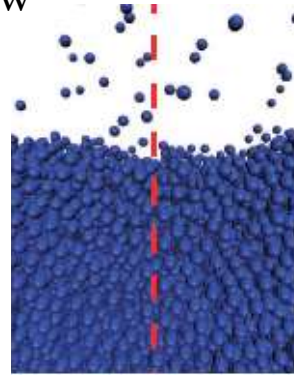
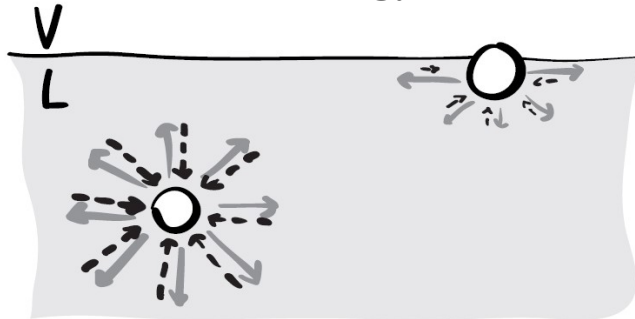
Influence of surfactants



Interfacial tension σ : energy *or* force of the interface

Thermodynamical point of view

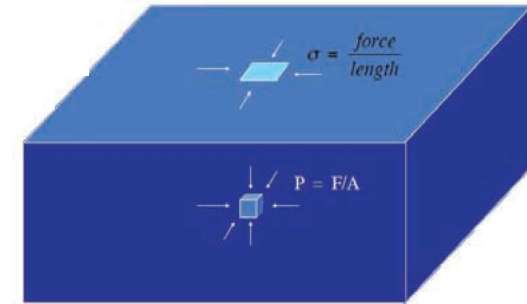
Energy: σ [J/m²]



Sketch showing repulsive (dashed black arrows) and attractive (gray arrows) forces in the bulk and at the surface.

Mechanical point of view

Force: σ [N/m]



A molecule in the bulk is surrounded by neighbors in all directions (equilibrium of energy of attraction and repulsion), while a molecule at the interface has a reduced number of neighbors => it is in an *energetically unfavorable state*: **creation of new surface is energetically costly**, and a **fluid system will act to minimize surface of the interface**.

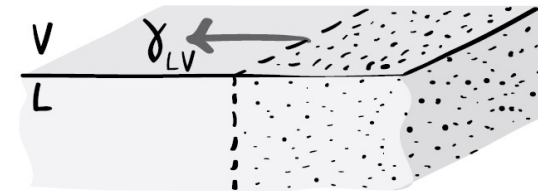
→ **Interfacial tension: work (energy) that must be provided to increase the surface of an interface**, in isothermal, isobaric and reversible conditions, because interactions are different at the interface:

$$dG = \sigma dA$$

G is the Gibbs free energy (or free enthalpy)

→ **Interfacial tension: Force per unit length parallel to the interface** (i.e. in the tangent plane), exerted perpendicular to any line drawn in the surface:

$$dF = \sigma dl$$



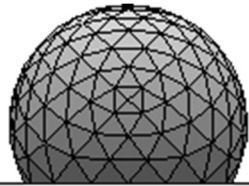
Interfacial tension σ

σ : **Energy** [J/m²] or **force** [N/m] of the interface

→ Explains why free bubbles are spherical (when not deformed by gravity *i.e.* at low Bond number Bo):

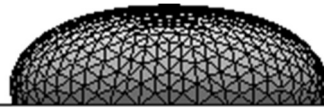
spherical shape = surface that minimizes the surface area for a given volume

→ A droplet on a solid substrate:



Without gravity ($Bo \ll 1$)

http://phyexpdoc.script.univ-paris-diderot.fr/projets/_sites_01_02_2/goutte/Approchetheorique.html



With gravity ($Bo \geq 1$)



Tintin (Hergé) – “Explorers on the Moon” (1976)

→ A whisky drop at $Bo = 0$

→ *Some values of surface tension for clean interfaces:*

Water/air: $\sigma = 72$ mN/m

Water/hexane: $\sigma = 50$ mN/m

Water/butanol: $\sigma = 2.1$ mN/m

$\sigma = 0$: when the phases are totally miscible

Interface G-L:
superficial tension

Interface L-L:
interfacial tension

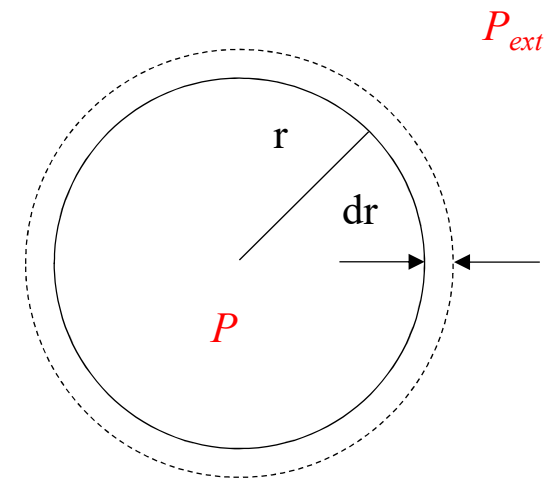
Laplace-Young equation for a bubble or droplet

Pressure jump across a static interface (without flow)

Across a sphere

Volume increase $dG_1 = \sigma d(4\pi r^2) = 8\pi r \sigma dr$
 → Variation of free enthalpy $dG_2 = -(P - P_{ext})4\pi r^2 dr$

At equilibrium: $dG_1 + dG_2 = 0 \Rightarrow$ $P - P_{ext} = 2 \frac{\sigma}{r}$



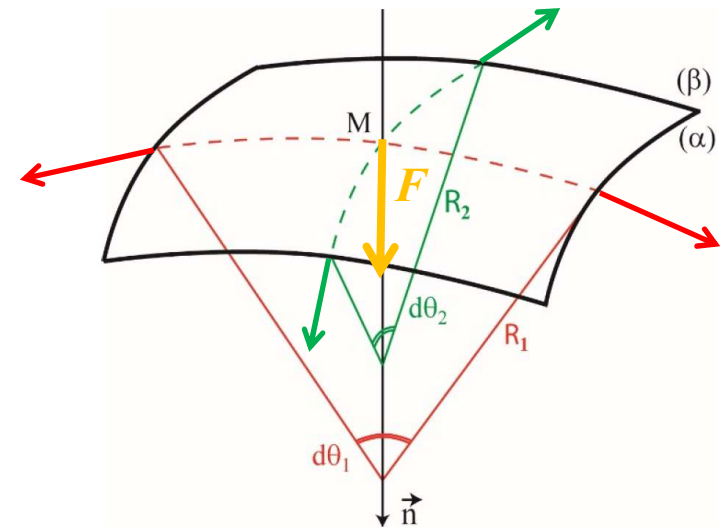
General case – Across a surface of two radii of curvature R_1 and R_2

$\Delta P = P - P_{ext} = \sigma \left(\frac{1}{R_1} + \frac{1}{R_2} \right)$

Laplace law

F : resultant of surface tension forces

- resultant is in the normal direction
- balances ΔP

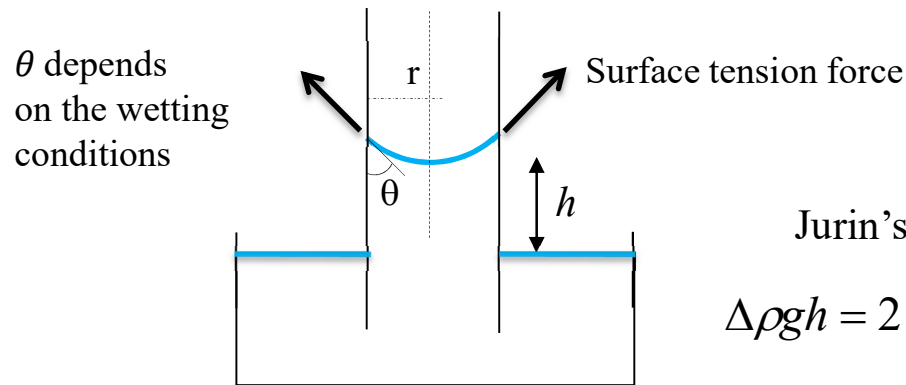


Interfacial tension: examples

Surface tension force: pulling on the surface (tangent to the interface)

Examples:

Capillary rise of a liquid



Jurin's law:

$$\Delta\rho gh = 2 \frac{\sigma \cos \theta}{r}$$

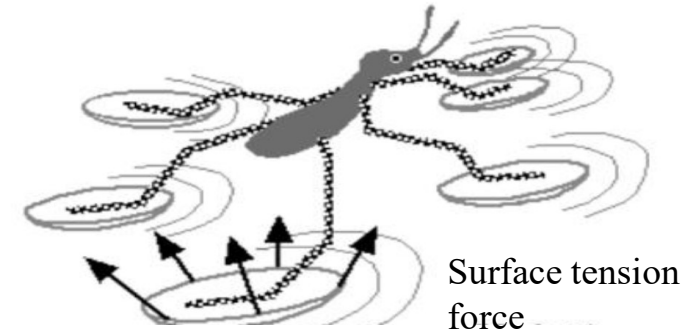
Vertical component of surface tension force:

$$F_s = \sigma \cos(\theta) 2\pi r$$

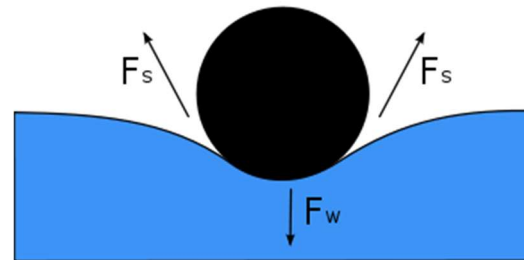
Balanced by the buoyancy force on the raised water:

$$P = \Delta\rho g \pi r^2 h$$

a Gerris walking on the water surface



Flotation of a sphere



https://commons.wikimedia.org/wiki/File:Gerris_ssp_fm1.JPG

http://national.udppc.asso.fr/attachments/Memoires_OdPF/Mmoire_Marcher_sur_l-eau_final.pdf

https://upload.wikimedia.org/wikipedia/commons/thumb/6/6a/Surface_Tension_Diagram.svg/350px-Surface_Tension_Diagram.svg.png

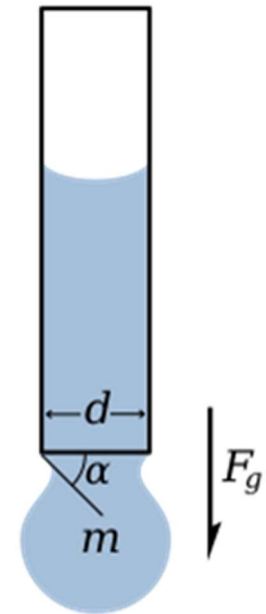
Interfacial tension: examples

A measurement method of surface tension: **pendant drop test**.
Simple method, commonly used.

A drop of liquid is suspended at a capillary tube by surface tension.

α : contact angle with the tube

Write the force balance on the droplet, giving the value of σ .



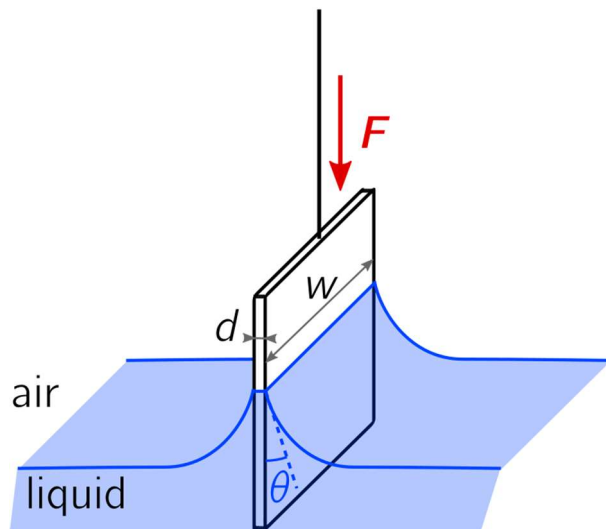
Interfacial tension: examples

Another measurement method of surface tension: **Wilhelmy plate**.

For air-liquid
or liquid-liquid interfaces

Plate perpendicular to the interface

Measure of the **capillary force F**
exerted on the plate



https://en.wikipedia.org/wiki/Wilhelmy_plate

Adapted from Marchand et al.,
(2011)

\vec{F} : Capillary force on the plate

Zoom on the triple contact point

Force balance on the solid plate

Forces at the liquid corner bounded by the contact line

Young's law:

$$\gamma_{LV} \cos \theta = \gamma_{SV} - \gamma_{SL}$$

$F = \gamma_{LV} \cos \theta \cdot l$
wetted perimeter

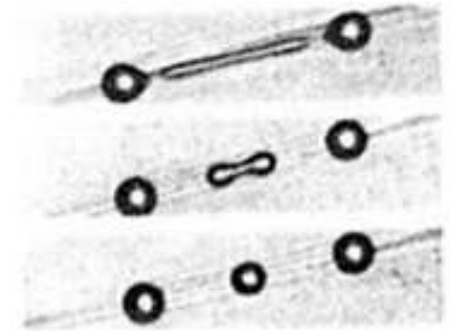
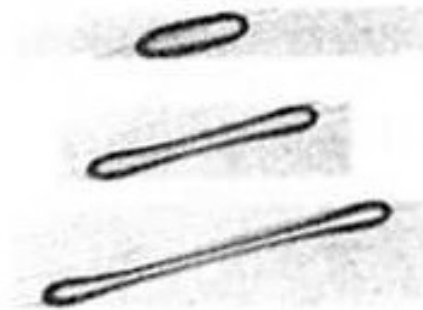
$$l = 2w + 2d$$

(Note: The diagram also shows force vectors γ_{LV} , γ_{SV} , and γ_{SL} at the triple contact point, and a downward arrow for 'Solid attraction'.)

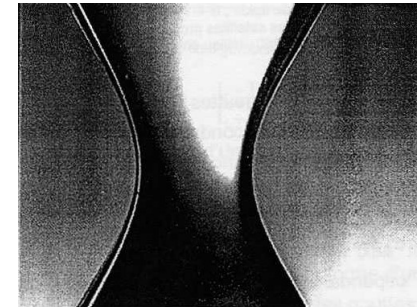
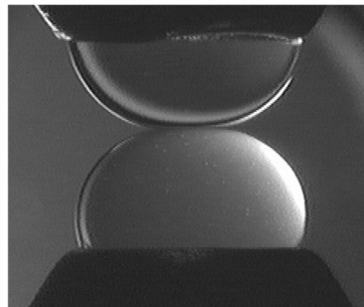
Importance of surface tension in interface phenomena

Surface tension resists to surface deformation, and acts to minimize the area of the interface.

Breakup of a stretched droplet
(Stone, 1994)



Coalescence of two droplets
approaching at constant
velocity
(Klaseboer, 1998)



Wetting of a droplet on
a substrate
(Pétrissans, 1996)



=> contact angle hysteresis effects (advancing and receding angles)

Surfactants: variation of interfacial tension

The Gibbs equation describes the **lowering** of surface tension due to surfactant adsorption
(generally a decrease)

Variation of free enthalpy dG due to a variation of chemical potential (at constant area)

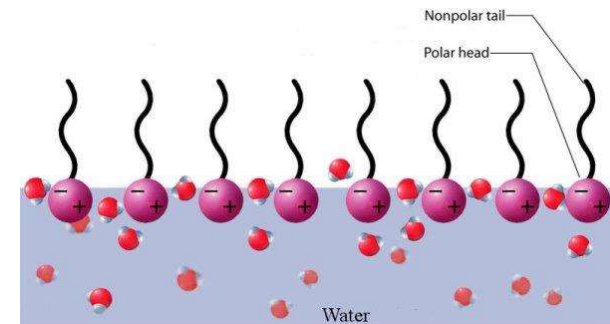
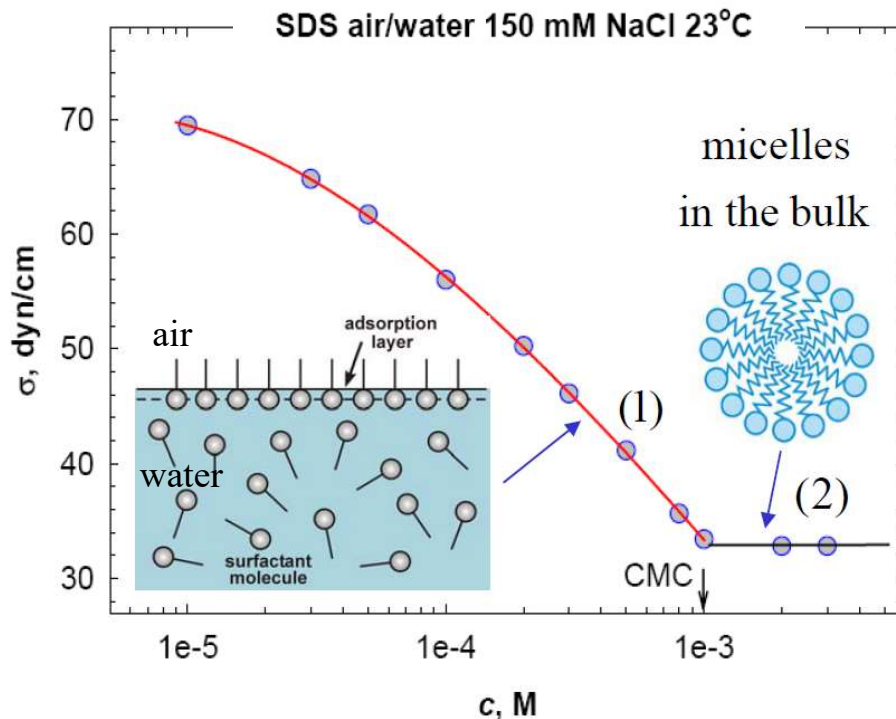
$$dG = Ad\sigma = -\sum_i n_i^S d\mu_i^S$$

Surface concentration of adsorbed species

Bulk concentration

$$\Rightarrow d\sigma = -\sum_i \frac{n_i^S}{A} d\mu_i^L = -\sum_i \Gamma_i d\mu_i^L$$

Adsorption equilibrium: ex, **Langmuir isotherm**
Relationship surface / bulk concentration: $\Gamma = \frac{aC}{1+bC}$

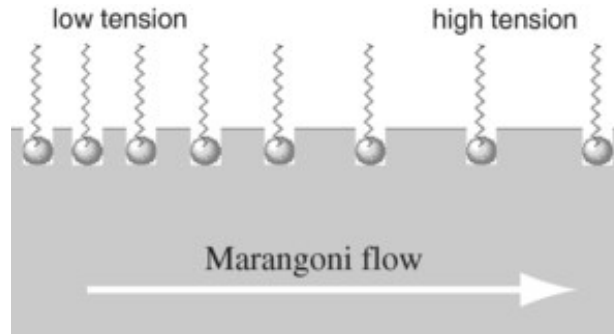


Surfactant: hydrophilic head, hydrophobic tail.
At the interface, it decreases the interactions between molecules of the bulk fluids.

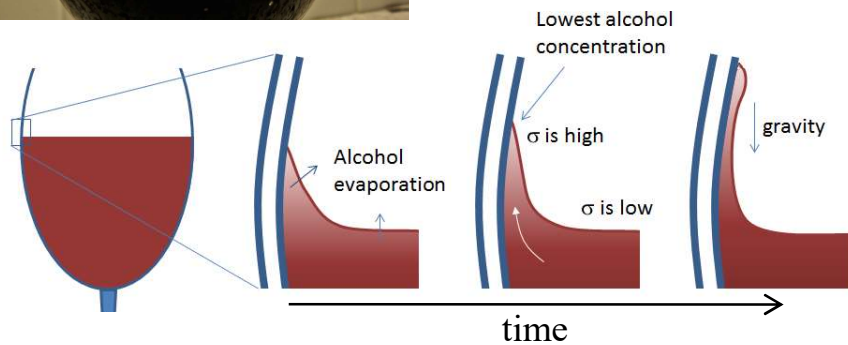
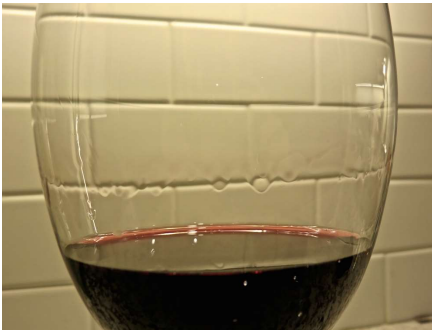
\Rightarrow It becomes *energetically less defavorable* for the system to increase the interface area.

Gradients of interfacial tension: Marangoni effect → change of hydrodynamics at interface

Gradients of σ (due to gradients of surface concentration of surfactants, or temperature) induce surface stresses, which correspond to a **Marangoni flow** along the interface.



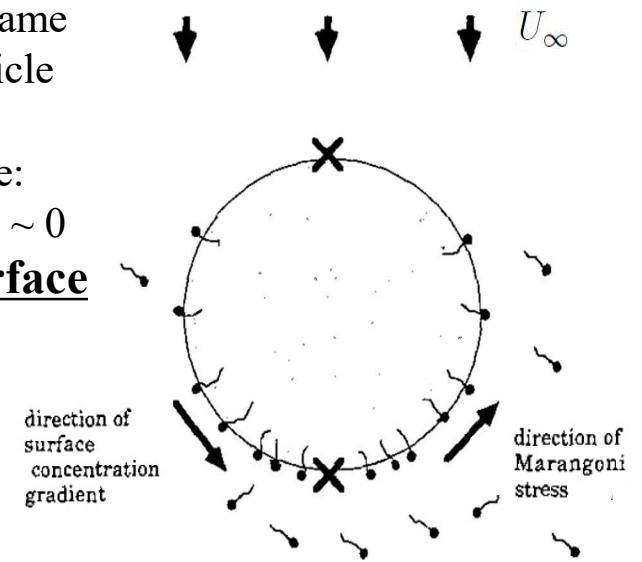
Some consequences:
Tears of wine!



Figures from: <http://iopscience.iop.org/article/10.1088/1751-8113/43/24/242001>
<https://www.comsol.com/multiphysics/marangoni-effect>

Rising drops and bubbles
→ Reduced terminal velocity U_∞

- U_∞ becomes the same as for a solid particle
- At the interface: tangential velocity ~ 0
→ **immobile interface**

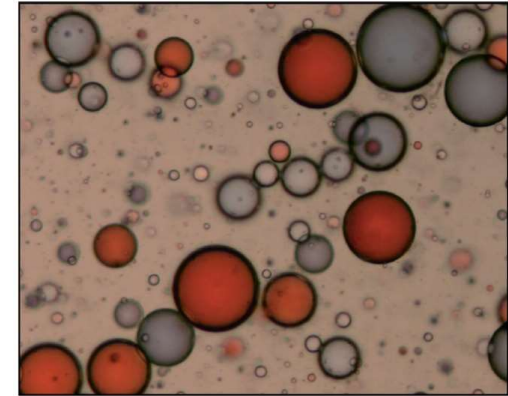


A. Frumkin and V.G. Levich, Zh. Fiz. Khim. **21**, 1183 (1947) 14

Presence of surfactants at an interface, consequences on fragmentation and coalescence

In **emulsions**, surfactants are present (naturally or introduced) in the fluid system.

*They are widely used in the industry (generally at large coverage rate of the interfaces) to **prevent drop coalescence** and **ensure emulsion stability**.*



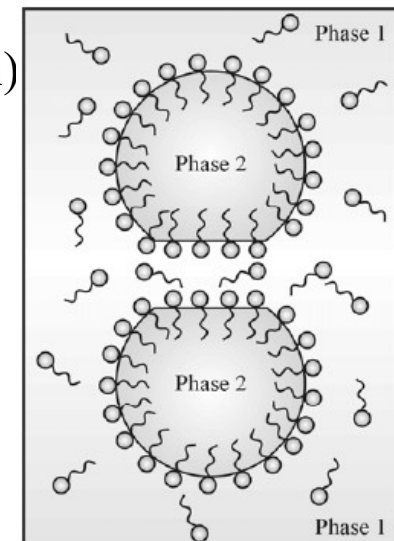
Bremond & Bibette, *Soft Matter* (2012)

Surfactants

- change the interfacial tension
- modify the hydrodynamics at the interface (Marangoni stresses and/or surface viscosity → **interface immobilization**) and give elasticity and viscosity to the interface
→ complex surface rheology not only described by σ
- can modify the molecule surface charges

→ **Strong changes of the coalescence and breakup rates in the presence of surfactants.**

- lower σ ⇒ lower resistance to interface deformation
- drop coalescence considerably reduced.



Danov (2010),
Thin Liquid Films: Drainage and Stability;
Role of Surfactant.

Breakup: physics and modelling

Causes of drop or bubble deformation

Physical parameters describing breakup (rupture)

Breakup kernels: how to model
breakup frequency & daughter drop size distribution



Example of applications

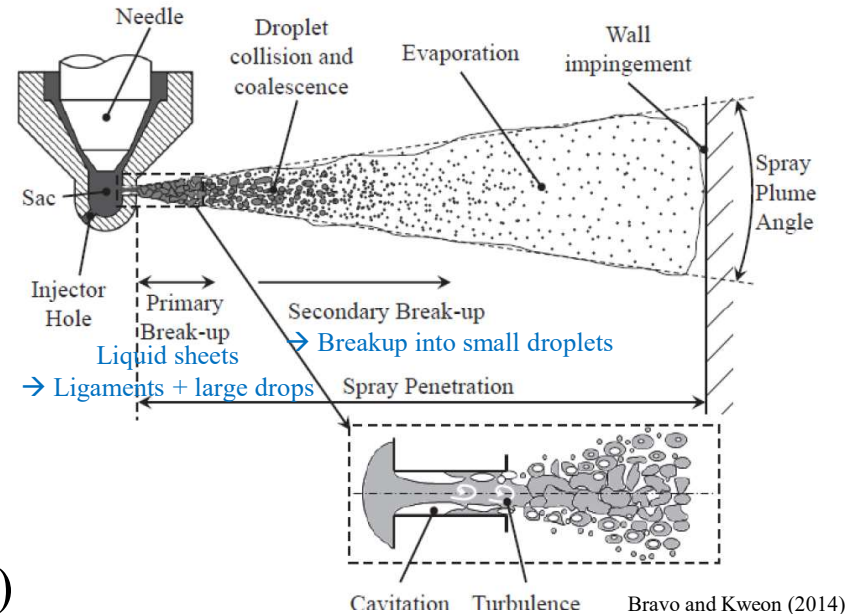
Chemical reactors with bubbly flows, extraction columns:

High rate of breakup desired to increase interfacial area (then mass transfer)

Atomization, sprays

(diesel engines, aerosols...)

High surface area desired for efficient evaporation/combustion



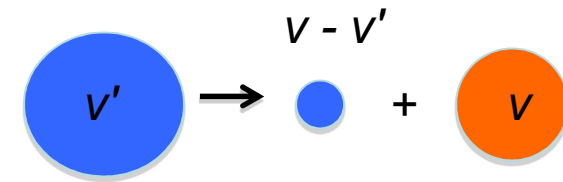
Objective: modelling drop breakup mechanisms – Focus on “secondary breakup”

Problem: Modelling breakup and selecting accurate kernels:

- Breakup frequency: $\Gamma(v)$
- Daughter drop size distribution after breakup: $\beta(v, v')$

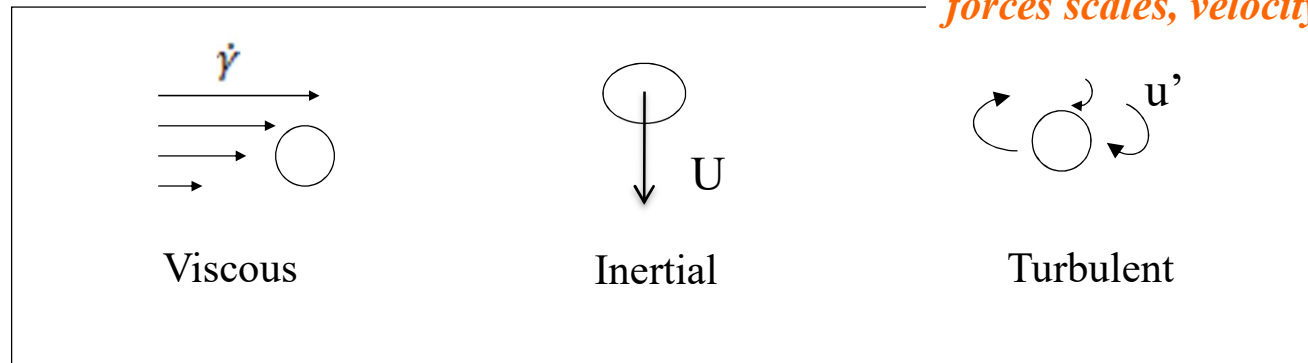
as a function of

- all physico-chemical properties of the fluid system
- local properties of the hydrodynamics field.



Secondary breakup =
Breakup of already formed droplets

Question: important time scales, forces scales, velocity scales?



Methodology:

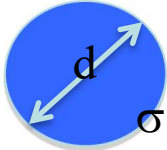
- analysis of breakup phenomena at local scale in controlled hydrodynamics conditions
- derivation of statistical models

Stresses that resist to deformation: surface tension & drop internal viscosity

- Surface tension and viscous stresses are compared in the **Ohnesorge number** of the drop:

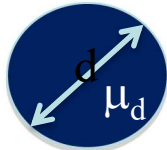
$$Oh = \frac{\mu_d}{\sqrt{\sigma d \rho_d}}$$

$Oh \ll 1$



Surface tension

$Oh \gg 1$



Drop internal viscosity

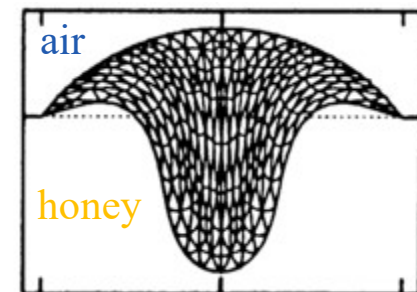
Stresses that **resist to deformation**

- *Provided droplet viscosity is not too high ($Oh \ll 1$), stress that resists to deformation is **due to surface tension** = σ / d [Pa]*

case *mainly* studied
in the following

- *In case of high viscosity of the deforming interface (large Oh): stress that controls deformation is due to viscosity.*

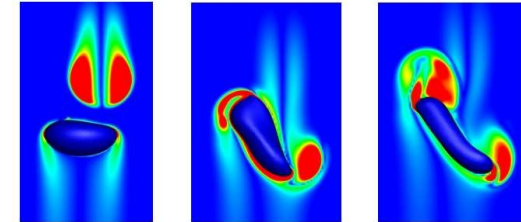
Ex: deformation of an interface **honey** – **air**
 → the relaxation process (after deformation) is controlled
 by viscosity, surface tension effects can be ignored.



Cause responsible for drop or bubble breakup

Breakup: process that converts one drop/bubble into two or more fragments.

- **deformation** with surface increase
- eventually: growing of instabilities
- **fragmentation** in daughter droplets.



Deformation not critical here!

Condition of appearing of a **critical deformation**, given by:

Stress that deforms the droplet $>$ Stress that resists to deformation

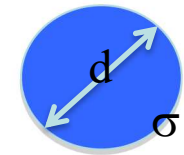
$$\underbrace{T}_{\text{depends on the type of flow}} > \underbrace{\sigma / d}_{\text{when dominated by } \sigma \text{ (= at } Oh \ll 1)}} \quad (\text{Hinze, 1955})$$

depends on the type of flow

when dominated by σ (= at $Oh \ll 1$)

→ Breakup if the ratio $T / \sigma / d$ is larger than a critical value

→ Breakup kernels are related to this critical number



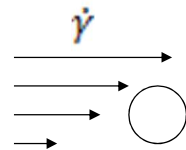
Critical number

Ca_{crit}

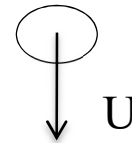
We_{crit}

We_{crit} or Ca_{crit}

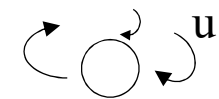
depending
on the stress
responsible
for
deformation



① **Viscous**



② **Inertial**



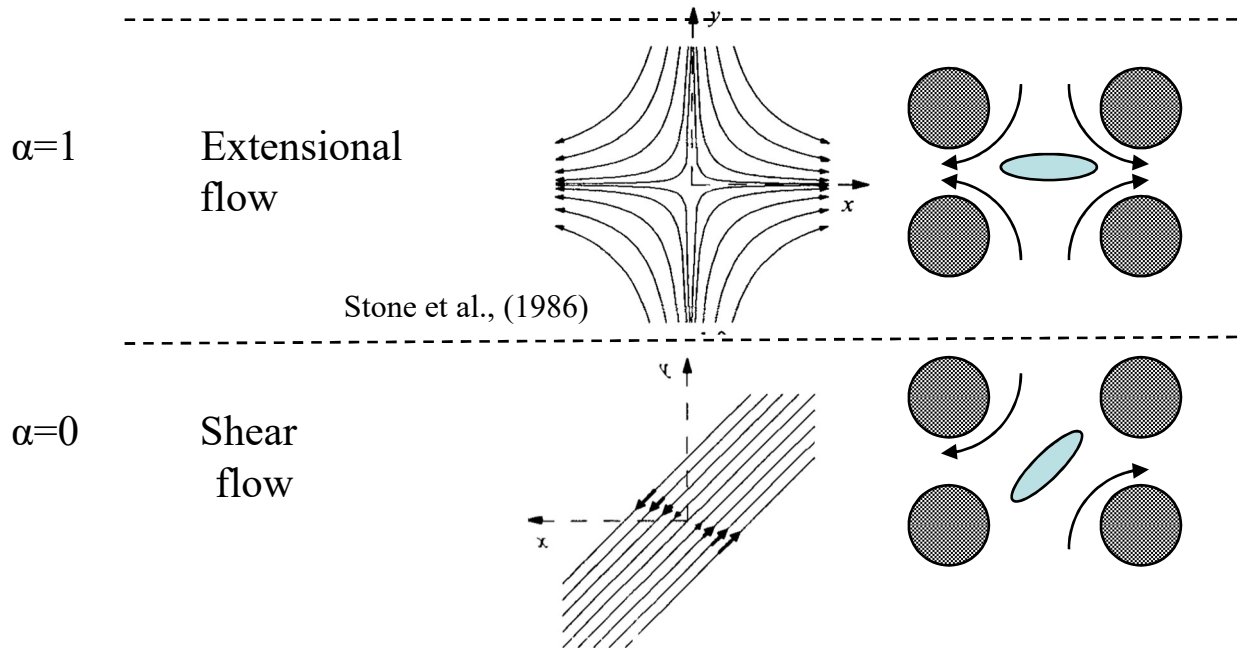
③ **Turbulent**

Hydrodynamics stresses leading to **deformation**

1) Breakup in the viscous regime

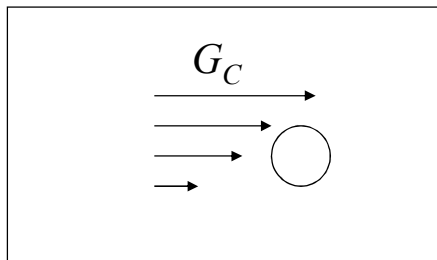
Drop/bubble deformed by a velocity gradient G_c - Without inertia: $Re_p = \frac{\rho_c G_c R^2}{\mu_c} \ll 1$

Experiment of the « **four roll mills** » (Taylor, 1934)



$$\nabla \vec{U} = \frac{1}{2} G_c \begin{pmatrix} 1+\alpha & 1-\alpha \\ -1+\alpha & -1-\alpha \end{pmatrix}$$

Breakup condition: **critical capillary number** Ca_{crit}



$$Ca_c = \frac{\mu_c G_c R}{\sigma}$$

$$\lambda = \frac{\mu_d}{\mu_c}$$

$$\longrightarrow Ca_{crit}(\lambda)$$

Hydrodynamic stress responsible for breakup:

$$T = \mu_c G_c$$

Velocity gradient \nearrow
seen by the droplet (shear rate)

1) Breakup in the viscous regime

Experiment on the « **four roll mills** » device, in steady flow - $Re_p < 1$

→ *With pure shearing*: ‘optimal’ breakup at $\lambda \sim 1$

→ Ca_{crit} ‘high’ at $\lambda \ll 1$:
difficulty in creating internal pressure gradients

→ Ca_{crit} lowest for extensional flow ($\alpha=1$) ;
for $\lambda > 4$, breakup not possible *with pure shearing*
very long filament *in extensional flow*



$\lambda \gg 1$:

Ca_{crit} independent on λ

The filament becomes very thin, and a *capillary instability* develops because surface tension wants to minimize the interfacial area.



Surface tension
becomes *the force that drives breakup at the end!*

Only for **largely extended drops** (length $> 20 R$)

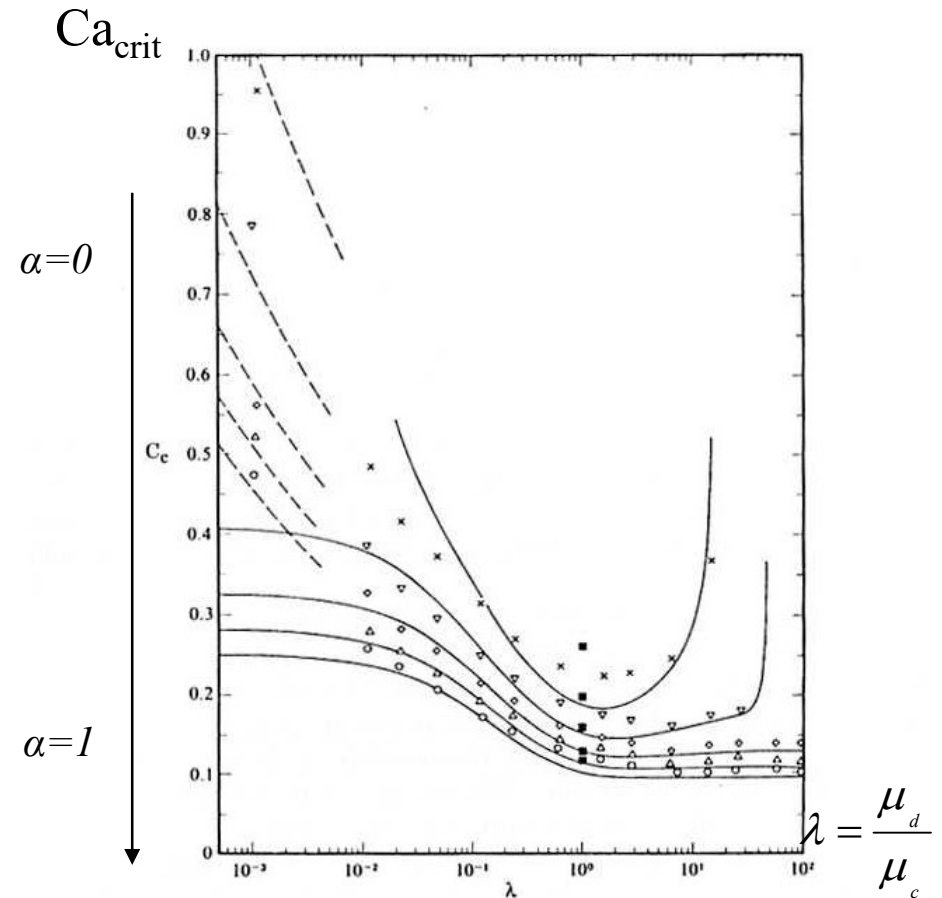
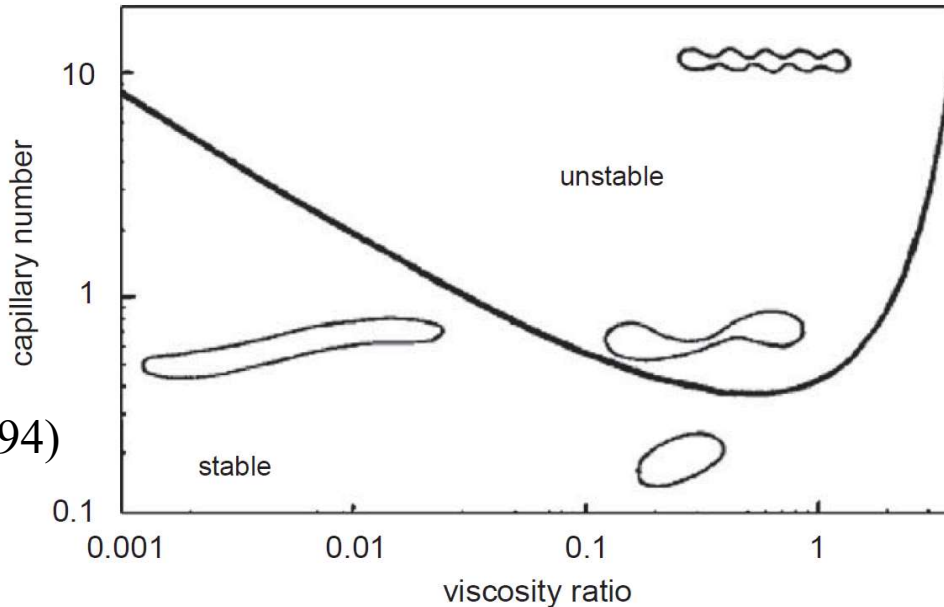


Figure 4 Effects of flow-type on the critical capillary number for drop breakup. Experimental results are presented for flow types $\alpha = 1.0$ (\circ), 0.8 (\triangle), 0.6 (\diamond), 0.4 (∇), 0.2 (\times). The solid curves are the prediction of the $O(C^2)$ small deformation theory and the dashed curves are the prediction of slender body theory (Bentley & Leal 1986b). The \blacksquare symbols are the boundary integral numerical results of Rallison (1981).

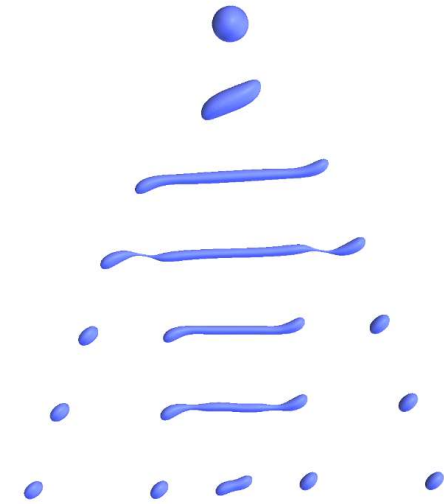
1) Breakup in the viscous regime

Shear flow
 $\alpha=0$

Experiments
 in steady
 conditions
 (Jassen et al., 1994)



A simulation with $\lambda = 2.5$:
 time evolution of drop shape



<https://www.aiche.org/conferences/aiche-annual-meeting/2012/proceeding/paper/361c-lattice-boltzmann-simulations-drop-deformation-and-breakup-shear-flow>

→ Breakup frequency law: $t_b \sim \frac{\mu_c R}{\sigma} f(\lambda)$ breakup time

$$\Gamma(v) \sim \frac{cste}{t_b}$$

Grace (1982),
 Elemans et al. (1993),
 Wieringa et al. (1996)

→ Number and size of fragments after breakup:

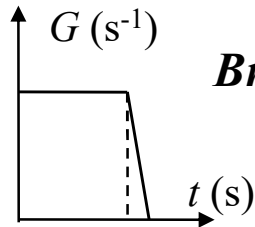
$$Ca_{crit} \leq Ca \leq 2 Ca_{crit}$$

2 fragments + 1 satellite

$Ca_{crit} \ll Ca$
 Series of n fragments
 of same size

1) Breakup in the viscous regime – Unsteady conditions

Steady flows not realistic... → *Influence of abrupt changes in flow conditions*



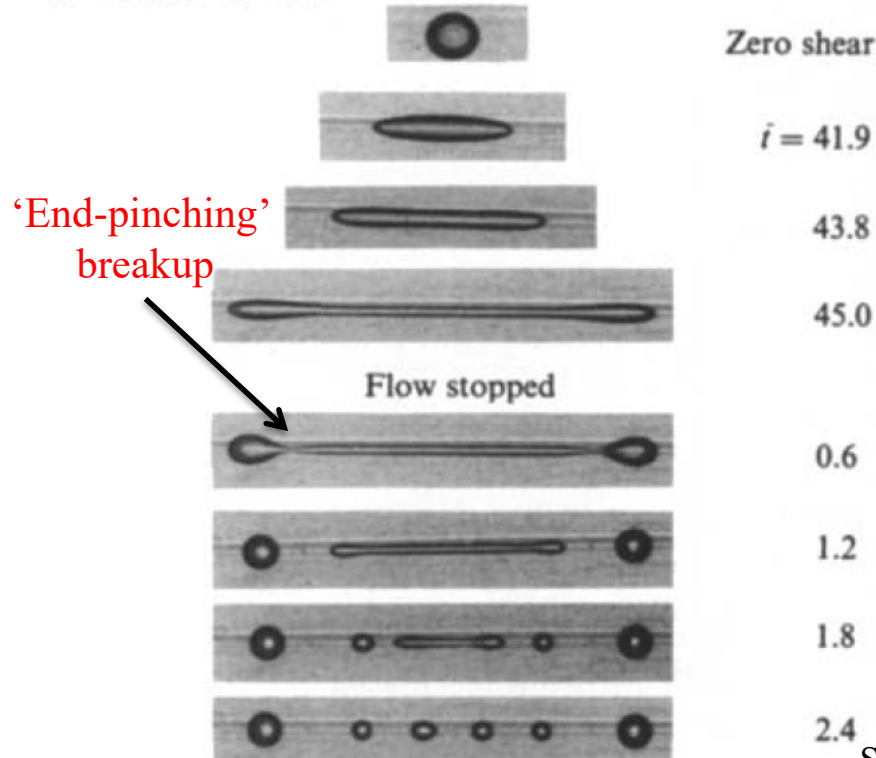
Breakup and drop size distribution depend on the history of the applied flow

Here: applied $G(t)$ (at critical Ca), then flow stoppage → the drop begins to relax.

Low viscous droplet:

Breakup (due to internal pressure gradients)

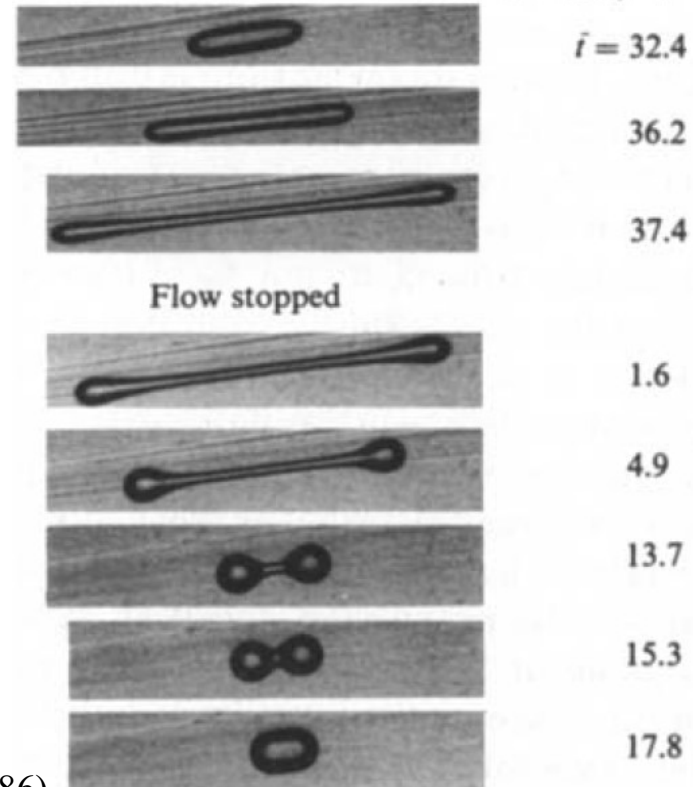
$\lambda = 0.046, \alpha = 1.0$



High viscous drop: internal flows are damped

Breakup inhibited

$\lambda = 11.3, \alpha = 0.6$



1) Breakup in the viscous regime – Case of an emulsion

→ Influence of the volume fraction ϕ of droplets

For suspensions or concentrated emulsions:

At low particulate Reynolds number Re_p ,

assumption of equivalent media:

effective density and viscosity, depending on ϕ .

$$\frac{\mu_e}{\mu_c} = (1 - a\phi)^{-n}$$

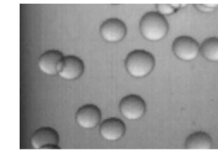
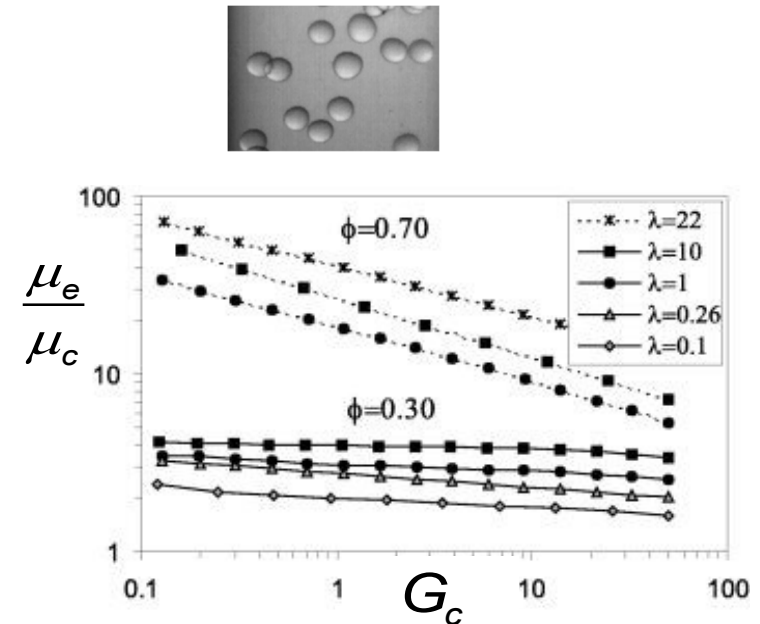
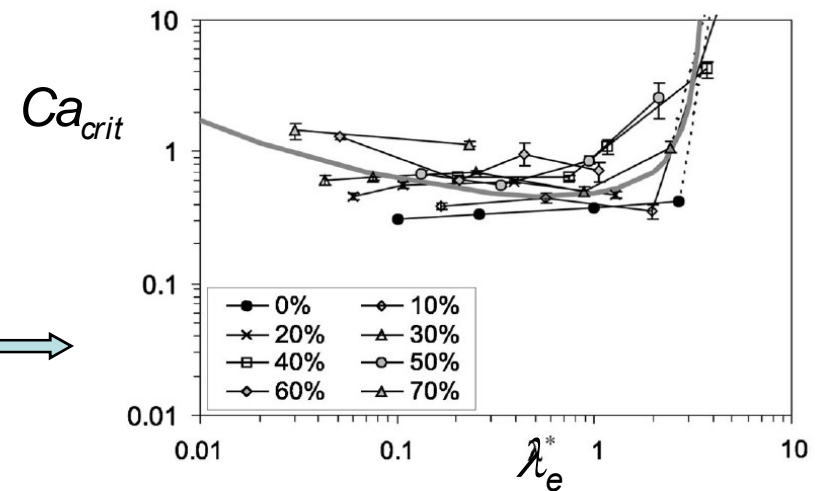
→ Emulsion \equiv non-Newtonian fluid:
shear thinning effects at high ϕ

Experiments of breakup in a *pure shearing flow* show:

same results as for a dilute emulsion,
by using an equivalent capillary number:

$$\mu_c \rightarrow \mu_e$$

$$Ca_{crit} = \frac{\mu_e G_{crit} R}{\sigma} = f\left(\lambda_e = \frac{\mu_d}{\mu_e}\right)$$



Exercise: emulsification in a colloidal mill

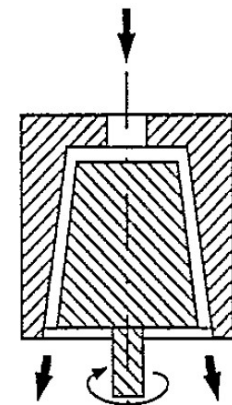
A colloidal mill is a rotor stator device, widely used in food processing or pharmaceutical manufacturing to produce emulsions with desired size.

The shear rate $\dot{\gamma}$ in the thin gap can be adjusted by varying the rotation speed.

- With the target to produce an emulsion with droplets of diameter inferior to $d=100\ \mu\text{m}$, with a continuous phase of density $\rho_c = 1000\ \text{kg/m}^3$ and *effective* viscosity $\mu_c = 0.006\ \text{Pa}\cdot\text{s}$, what minimal value of $\dot{\gamma}$ has to be produced in the gap?
- Verify that the continuous flow regime is laminar in the gap of thickness $h=0.3\ \text{mm}$ (the transition Reynolds number for such a flow in a Couette device with smooth surfaces is superior to 10000).
- Is breakup due to viscous forces or to inertial forces in this system? Comment.

Data available for the fluid system:

- The interfacial tension is found to be $\sigma = 5\ \text{mN/m}$.
- Schuch et al. (2013) report a critical capillary number close to 3 - *based on the biggest droplet diameter of the emulsion* - for an emulsion with a viscosity ratio $\lambda = 16.7$ (inner phase viscosity $\mu_d = 0.1\ \text{Pa}\cdot\text{s}$).



rotor and stator:
smooth or toothed
teeth: various design

2) Breakup in the inertial regime (determinist flows)

Drop/bubble deformed by inertial forces

Resisting stress to deformation is due to *surface tension*.

→ Breakup criterion: based on a **critical Weber number**

We consider determinist flows here:

deformation in extensional, oscillatory, rotating flows,
due to impulsive accelerations,

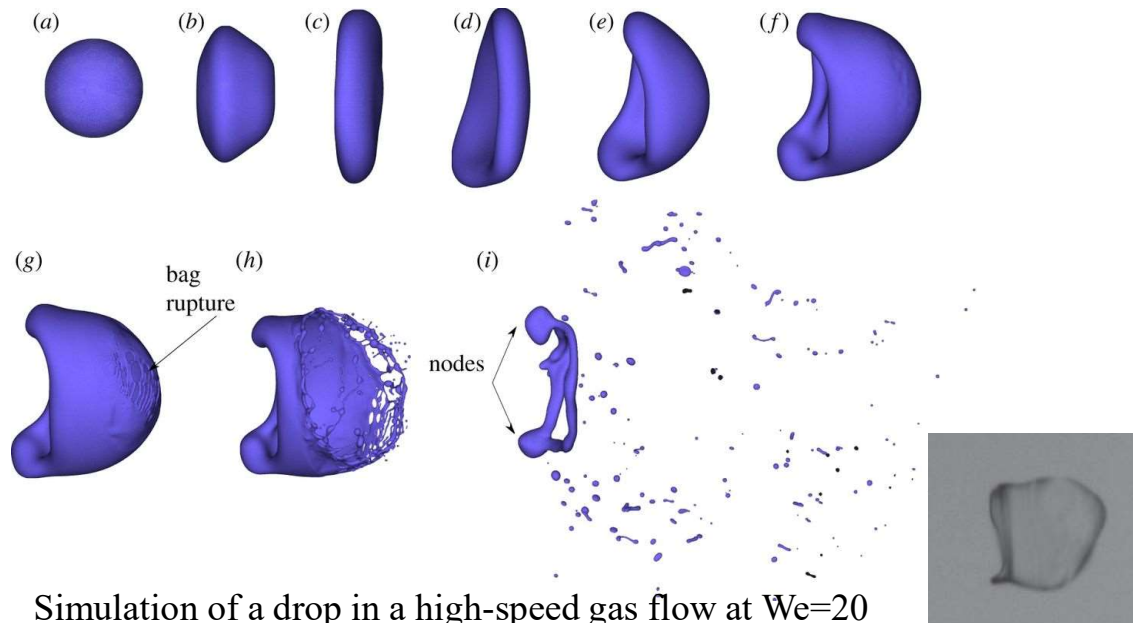
due to the fall or rise of a bubble/drop with strong relative velocity...

$$\text{Inertial stress: } T = \rho_c \mathbb{U}^2$$

$$We = \frac{\rho_c \mathbb{U}^2 d}{\sigma} = Ca \cdot Re_p$$

\mathbb{U} = drop relative velocity
or the velocity based on a
characteristic velocity gradient

Ex: **secondary breakup** in atomization sprays, liquid propellant combustion...

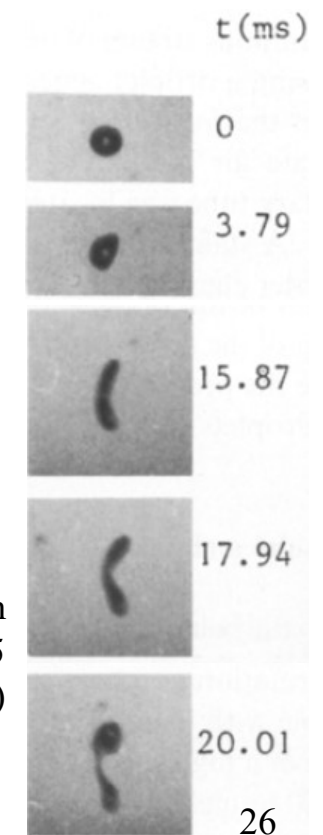


Simulation of a drop in a high-speed gas flow at $We=20$

Experimental picture of bag breakup at $We=20 (> We_{crit})$ - Jain et al. (2015)

'Aerodynamic
fragmentation'

Drop in a gas stream
 $We=12.5$
Wierba (1990)



26

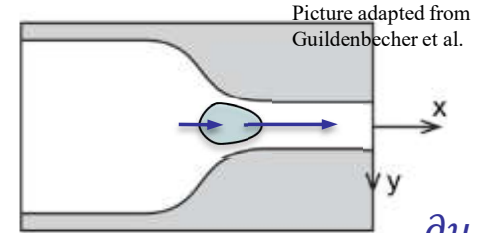
2) Breakup in the inertial regime: examples

Extensional flow: Velocity gradient G_c in the flow direction

→ Inertial stress at the drop scale based on $U = G_c R$

For a bubble:
$$\left(\frac{1}{We_{crit}}\right)^{10/9} = \left(\frac{1}{2.76}\right)^{10/9} + \left(\frac{1}{0.247 Re_p^{3/4}}\right)^{10/9}$$

$$We = \frac{\rho_c (G_c R)^2 d}{\sigma}$$



Ex: convective acceleration $G_c \sim \frac{\partial u}{\partial x}$

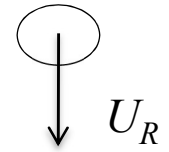
Rising / falling drops or bubbles: $U = U_R$ relative velocity (terminal velocity in a quiescent external phase)

$$Re_p = \frac{\rho_c U_R d}{\mu_c}$$

$$We = \frac{\rho_c U_R^2 d}{\sigma}$$

$$Bo = \frac{\Delta \rho g d^2}{\sigma}$$

$$Mo = \frac{\Delta \rho g \mu_c^4}{\rho_c^2 \sigma^3}$$



Falling drops in quiescent air

$$Bo_{crit} = 12.4$$

$$We_{crit} = 12$$

see Pilch & Erdman (1987), Jain et al. (2015)

Drops at terminal velocity in a quiescent liquid (valid for $Mo < 256$)

$$Bo_{crit} = \frac{16}{1 + 0.5 \left(4.5 \left(\frac{\rho^*}{1 + \rho^*} \right) - 0.35 \left(\frac{2 + 3\rho^*}{1 + \rho^*} \right)^{2.27} (1 + Mo^{1/4})^{0.36} \right)}$$

$$We_{crit} = 0.5 Bo_{crit}$$

$$\rho^* = \frac{\rho_d}{\rho_c} \sqrt[3]{2}$$

Bubbles rising in a quiescent liquid

$$Bo_{crit} = 733 (1 + Mo^{1/4})^{1.66}$$

$$We_{crit} = 0.5 Bo_{crit}$$

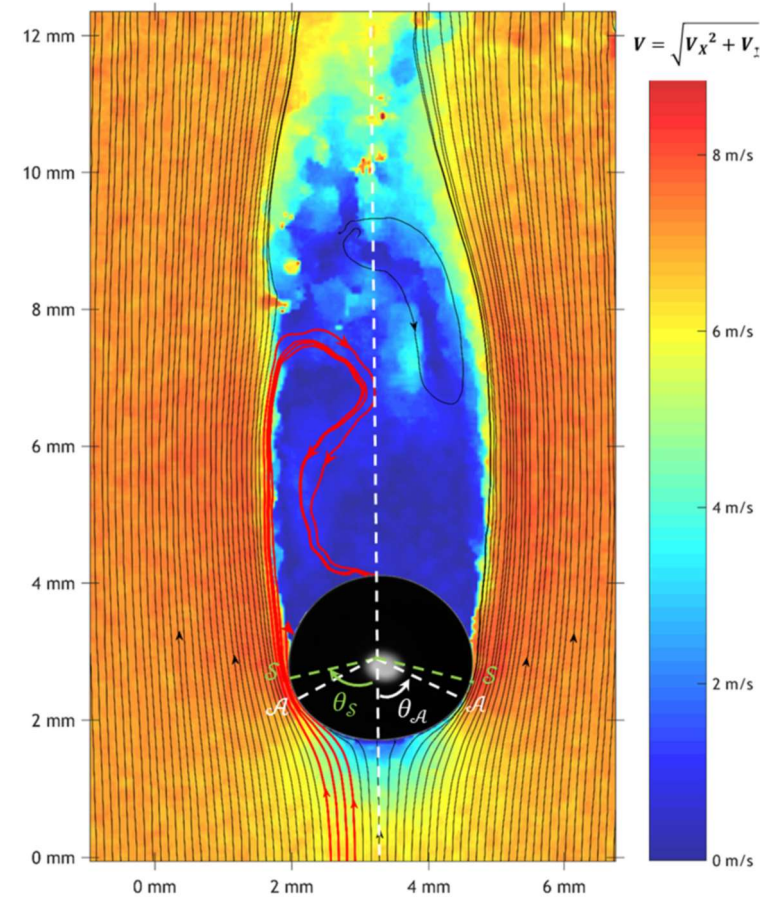
Exercise: breakup due to inertia

Based on the critical breakup condition, estimate (i) the highest possible diameter of a raindrop and (ii) its terminal velocity.

$$Bo_{crit} = 12.4$$

$$We_{crit} = 12$$

(critical value for breakup in the bag regime,
Taylor 1949)

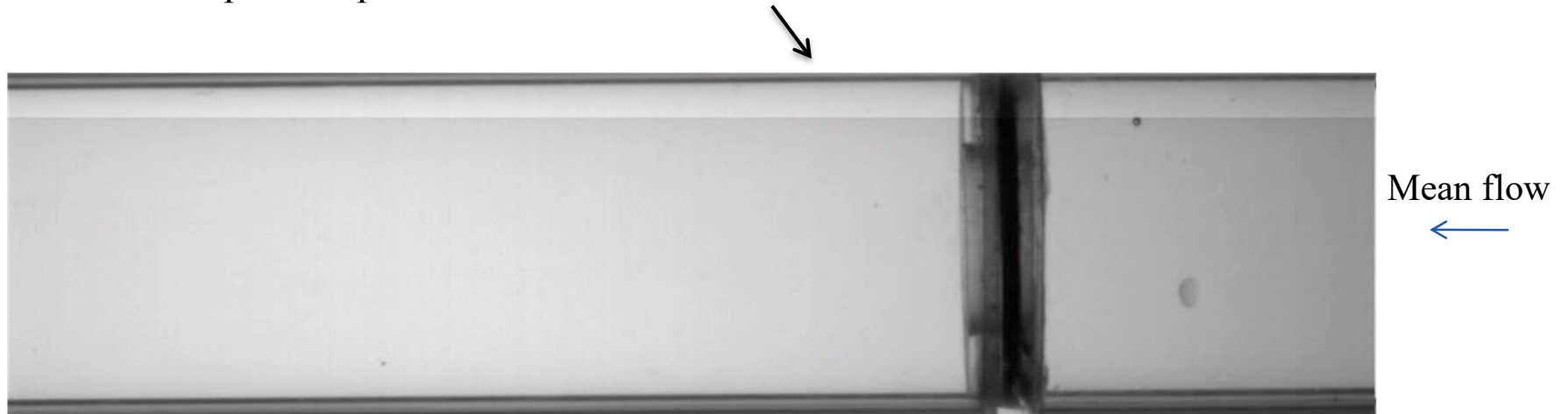


Instantaneous air flow around a raindrop
($Re \sim 2000$, $We < We_{crit}$ – Lemaitre et al., Atmosphere (2020))

3) Breakup in turbulent flows

Drop/bubble deformed by turbulent fluctuations

Breakup of droplets in the turbulent zone downstream of an orifice



In the same device: breakup of an emulsion

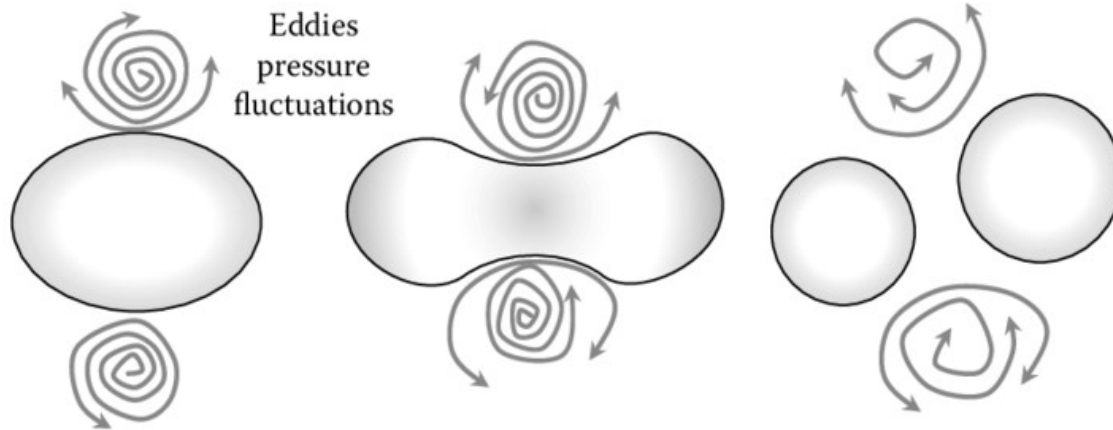


3) Breakup in turbulent flow

Existence of *two regimes*, depending on the drop size compared to the flow length scales:

Turbulent inertial regime

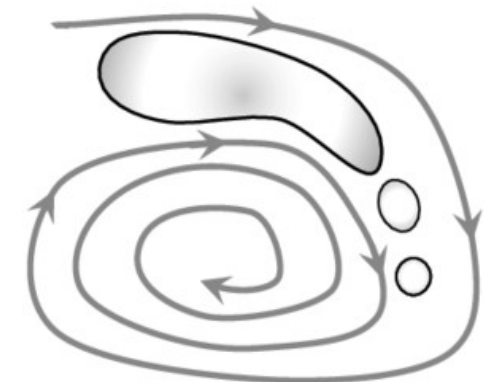
$$\eta < d < L$$



Drops are larger than the smallest eddies and deform by the action of hydrodynamic pressure fluctuations

Turbulent viscous regime

$$d < \eta$$



Drops are smaller than the smallest eddies and deform under the action of viscous stress inside and between the eddies

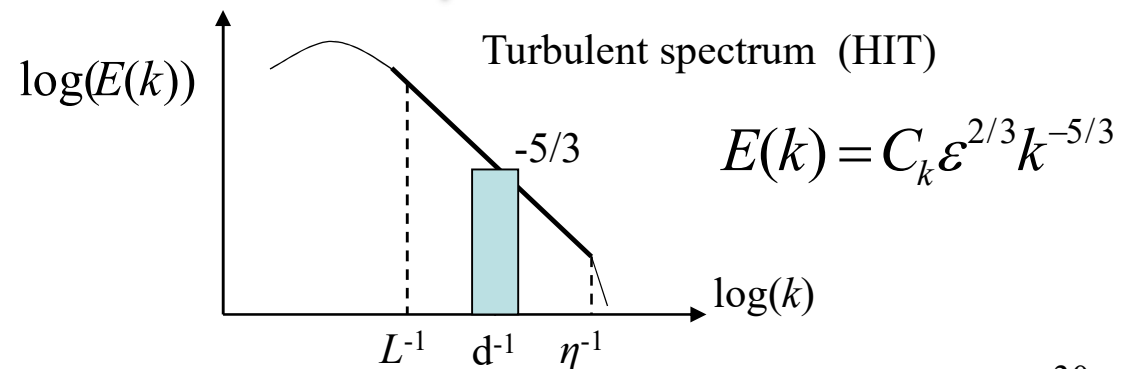
Rayner (2015)

η : Kolmogorov length scale
 L : Integral length scale

Size of the smallest eddies:

$$\eta \sim \varepsilon^{-1/4} \rho_c^{-3/4} \mu_c^{3/4}$$

with ε the dissipation rate



3) Breakup in turbulent flow

Theory proposed by Hinze (1955):

Turbulent inertial regime: $\eta < d < L$

Main assumption: only the turbulent vortices of size comparable to the (initial) droplet diameter d are efficient for breakup.

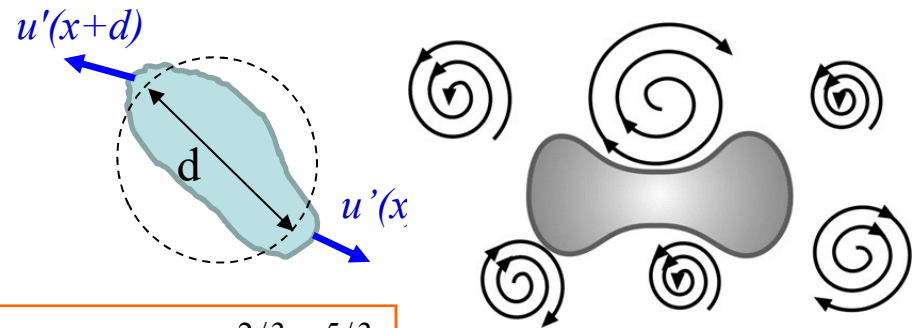
$$\delta u'_c{}^2(d) = \overline{[u'_c(x+d) - u'_c(x)]^2} = 2(\varepsilon d)^{2/3}$$

Inertial stress T (turbulent pressure fluctuation):

$$\rho_c \delta u'_c{}^2 \sim \rho_c (\varepsilon d)^{2/3}$$

a We number to quantify the risk of breakup

$$We = \frac{2\rho_c \varepsilon^{2/3} d^{5/3}}{\sigma}$$



Hinze-Kolmogorov theory

Turbulent viscous regime: $d < \eta$

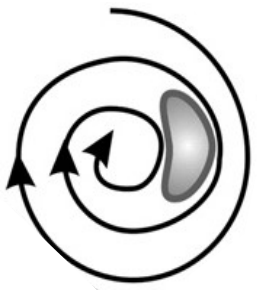
Viscous gradient seen by the droplet: $G_c = \frac{U_\eta}{\eta} \sim \left(\frac{\rho_c \varepsilon}{\mu_c}\right)^{1/2}$ Viscous stress $T = \mu_c G_c$

U_η is given by: inertial = viscous stresses

$$\rho_c (\varepsilon \eta)^{2/3} \sim \mu_c \frac{U_\eta}{\eta}$$

a Ca number to quantify the risk of breakup

$$Ca = \left(\frac{2}{15}\right)^{1/2} \frac{(\rho_c \mu_c \varepsilon)^{1/2} R}{\sigma}$$



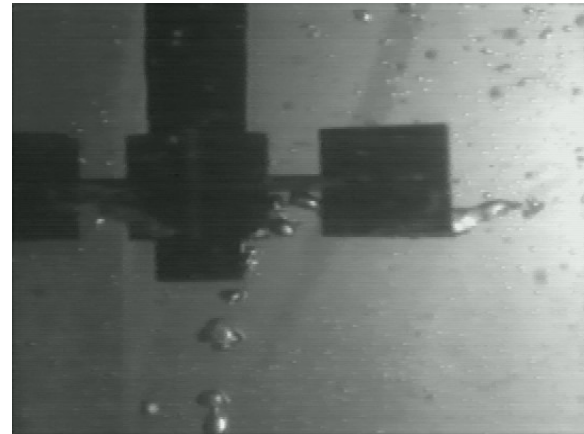
3) Breakup in turbulent flow

Ex : *Breakup of bubbles in a stirred tank*

Assumption: the velocity field created by the agitator in the tank is supposed to be a homogeneous isotropic turbulent flow (HIT).

Dissipation rate by the agitation device:

$$\varepsilon \propto P / \rho_c V_{cuve} \propto N^3 D^2$$



Power consumption:

$$P \propto \rho_c V_{cuve} N^3 D^2$$

Leading to:

$$d_{\max} \propto N^{-1.2}$$

inertial regime: theory predicts $d_{\max} \sim \varepsilon^{-0.4}$

$$d_{\max} \propto N^{-1.5}$$

viscous regime: theory predicts $d_{\max} \sim \varepsilon^{-0.5}$

By varying N , experimental results show that

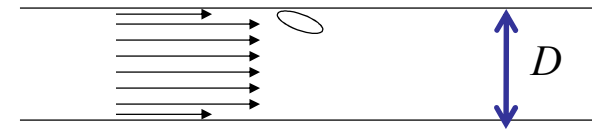
$$\frac{d_{32}}{D} = c_1 We_D^{-0.6}$$

$$We_D = \frac{\rho_c N^2 D^3}{\sigma}$$

→ Turbulent inertial breakup regime
Size of bubbles $> \eta$

Exercise: breakup due to turbulence

Drop breakup in turbulent pipe flow



Breakup probability of a droplet of diameter d is the highest in the near-wall region. We consider a droplet larger than the Kolmogorov scale.

The mean flow, of velocity U , induces an average wall shear rate of about $\dot{\gamma} \approx k \frac{U}{D}$; it is considered here that $k = 4$.

The turbulent field is characterized by a dissipation rate $\varepsilon = 2f \frac{U^3}{D}$, with f the friction factor (or pressure drop coefficient) given by the Blasius law: $f = 0.079 \text{Re}^{-1/4}$.

Give the condition where droplet deformation

- (a) is induced by viscous shearing due to the mean flow;
- (b) is induced by the turbulent fluctuations at the drop scale;

When will the risk of breakup due to turbulent fluctuations be higher than that due to viscous shearing at the wall? Write the condition as a criterion on d/D .

3) Breakup in turbulent flow

Ex : *Emulsification process – Transition between the two breakup regimes.*

Production of an emulsion of micrometer size droplets in a narrow-gap homogenizer

Study of Vankova et al. (2007)

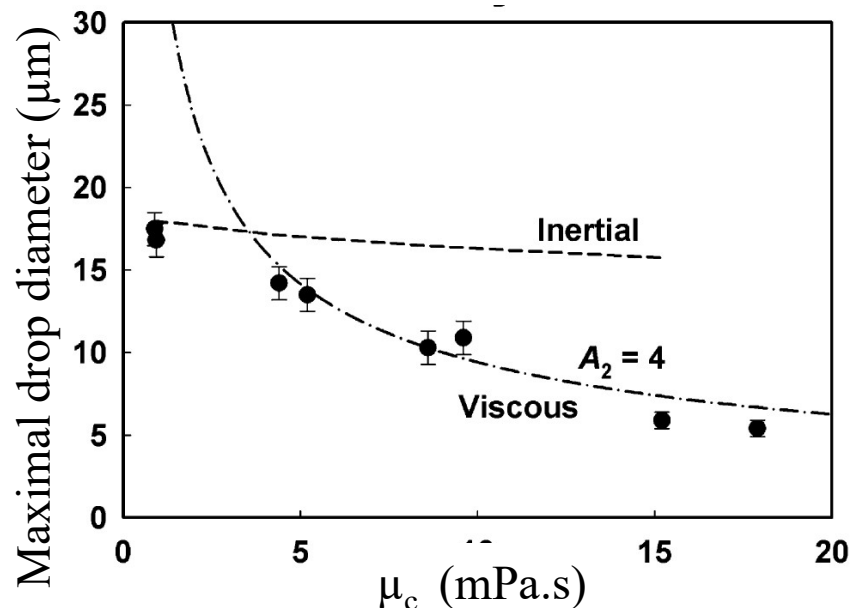
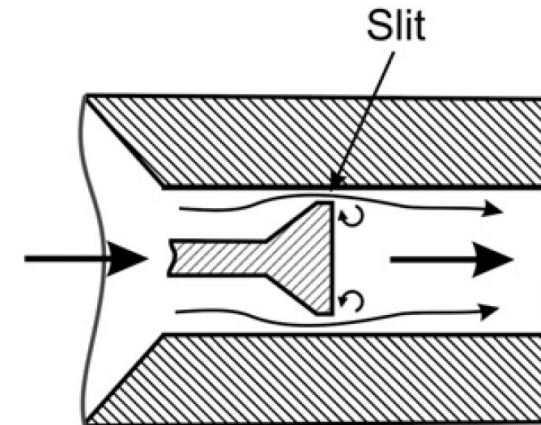
Use of high surfactant concentration

⇒ no coalescence, only breakup *due to turbulence*

An example of experimental observation:

Breakup of a dilute emulsion of silicon oil in water:

average drop size divided by 3 when μ_c is multiplied by 20.



Comparison with theories for the maximal stable diameter in inertial and viscous regimes:

→ the **regime of breakup changes** from **turbulent inertial** (at low μ_c) to **turbulent viscous** (at higher μ_c).

→ Maximal stable diameter in viscous regime:

$$d_{max} \sim cste \cdot \sigma \cdot (\rho_c \mu_c \varepsilon)^{-1/2}$$

Breakup kernels: (A) frequency and (B) daughter drop size distribution

- (A) **Breakup frequency:**

Modelled by a *frequency of collisions* with vortices, times a *probability* that collisions lead to breakup:

$$\Gamma(v) = f_{coll}(v) \cdot P_{rupt}(v)$$

Several kernels available
in the review of Liao and Lucas (2009)

Homogeneous Isotropic Turbulence (HIT):

velocity fluctuation associated to a vortex of size d

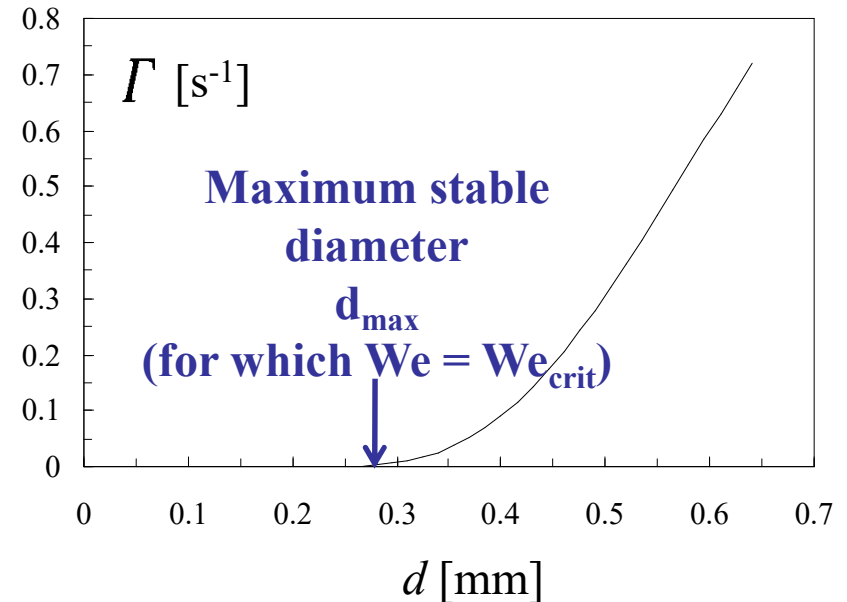
$$u' \cong (\varepsilon d)^{1/3}$$

=> *collision frequency:*

$$f_{coll} = C_1 \varepsilon^{1/3} d^{-2/3}$$

Breakup probability:

$$P_{rupt} = \exp\left(-\frac{C_2}{We - We_{crit}}\right)$$



Constants C_1 et C_2 are not universal and are generally determined from experiments.

Effect of emulsion concentration: taken into account as a modulation of turbulence intensity ε .

Breakup kernels: (A) frequency and (B) daughter drop size distribution

- (B) *Daughter drop size distribution*:

A model that gives the number N_f of fragments after breakup and the daughter size distribution function (or daughter drop volumes)



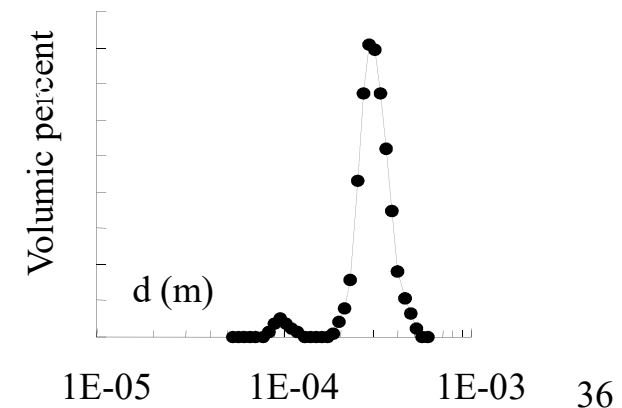
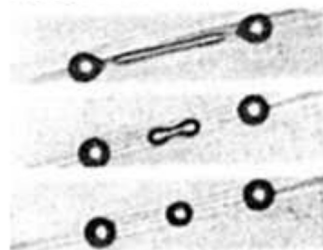
$$\int_0^{v'} \beta(v, v') dv = N_f$$

A distribution is assumed, depending generally on experimental observations: binary breakup, N_f fragments of same size, a Gaussian distribution for the fragments, ...

Ex: Gaussian distribution for N_f fragments; the daughter drops of volume v follow

$$\beta(v, v') = \frac{N_f}{\sigma\sqrt{2\pi}} \exp\left(-\frac{(v - m)^2}{2\sigma^2}\right) \quad \text{Average volume } m = \frac{v'}{N_f} \text{ \& standard deviation } \sigma$$

Ex: Multimodal distribution with 2 characteristic sizes



Open questions on breakup modelling

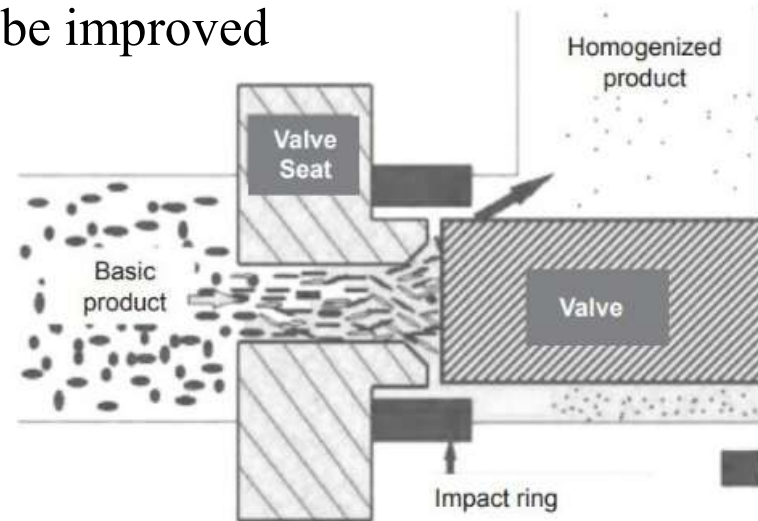
- A lot of existing models for breakup... predicting different breakup frequencies
→ physical reliability of the models to be improved

- Generally: *unsteady* conditions for breakup
Stress is applied during a finite time

→ Importance of *time of residence*

→ Importance of *response time* of drops or bubbles to an instantaneous deformation

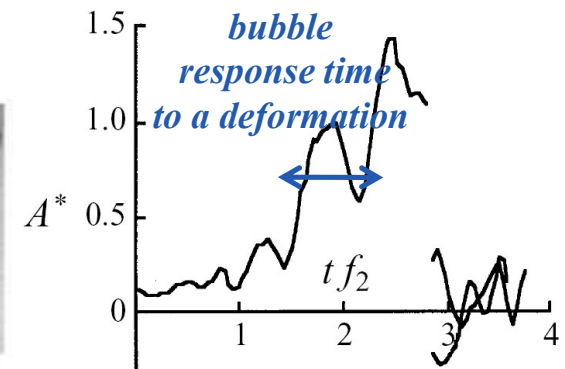
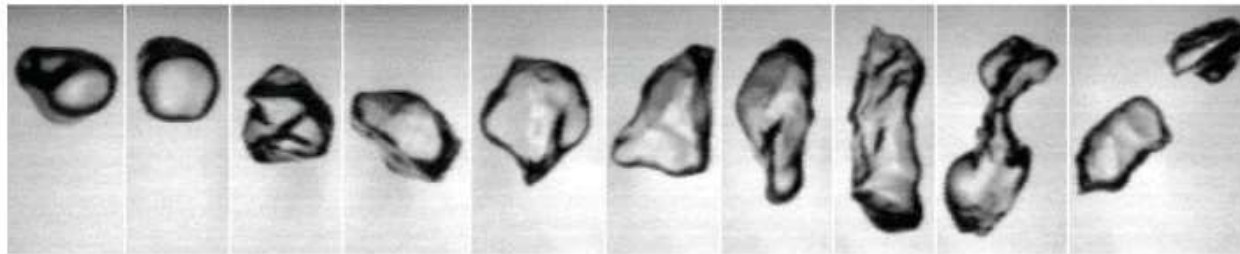
Risso and Fabre (1998) – Lalanne, Masbernat and Risso (2019)



High pressure homogenizer
(widely used in food industry for example)

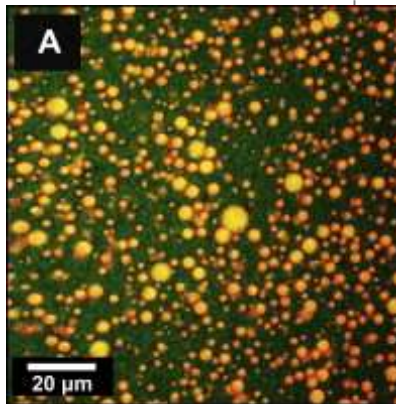
Picture from: <https://korpro.com/product/high-pressure-homogenizer-applications/648/>

A bubble oscillating in an homogeneous turbulent flow



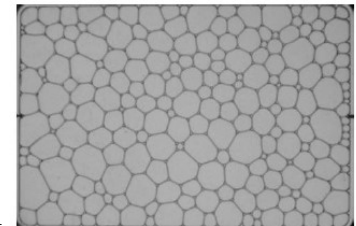
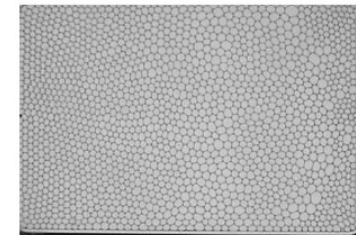
Coalescence: physics and modelling

Physical description of coalescence
Hydrodynamics of film drainage process
Coalescence kernels: how to model
collision frequency, coalescence probability



Milk = natural emulsion of fat globules in an aqueous phase

Garcia et al. (2014)



Time evolution of a foam

Rio (2013)

Example of applications

Stability of food or beverage emulsions (milk, soda...):

Coalescence has to be prevented to avoid ringing, creaming.

Ringling as a result of broken emulsions.



https://sensientfoodcolors.com/en-eu/emulsion_stability_in_rtd_sparkling_beverages/

Separation of emulsions in chemical processes, crude oil recovery:

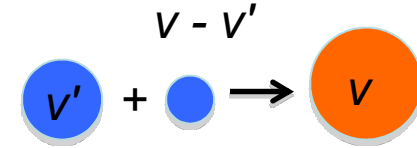
Coalescence has to be fastened to enhance phase separation.

Objective: modelling coalescence process

Problem: Modelling coalescence and selecting accurate kernels for term $Q(v, v')$ of the PBE.

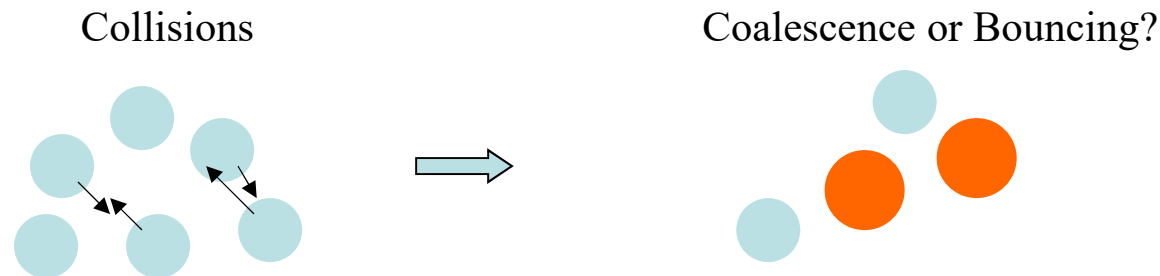
Coalescence rate: $Q(v, v') = C(v, v') P_{coal}(v, v')$ \rightarrow same as for breakup rate

- Collision frequency: C
- Coalescence efficiency or Probability of Coalescence: P_{coal}



which are functions of

- all physico-chemical properties of the fluid system
- local properties of the hydrodynamics field.



Note : Coalescence: only for drops / bubbles

Aggregation, flocculation, coagulation: only for solid particles, not mentioned hereafter.

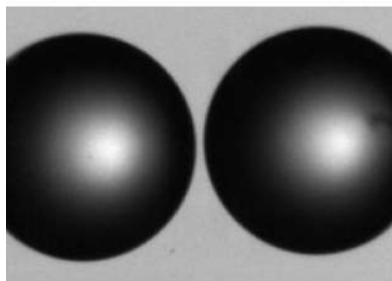
Physical description of the problem

Coalescence: process that merges two droplets or bubbles (→ a way for the system to minimize the surface per unit volume).

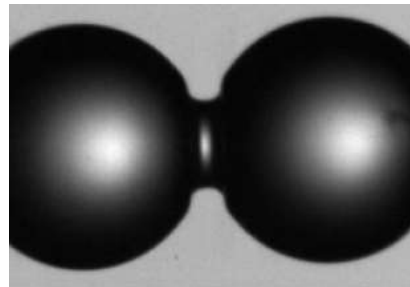
Steps: (each of these steps can limit the coalescence rate.)

1. **Collisions:** approach of two droplets
2. **Drainage** of the film between the droplets
3. **Hole opening:** an hole bridge is established between the drops
4. **Hole growth:** the bridge grows and eventually the droplets merge
5. **Shape relaxation** of the new droplet

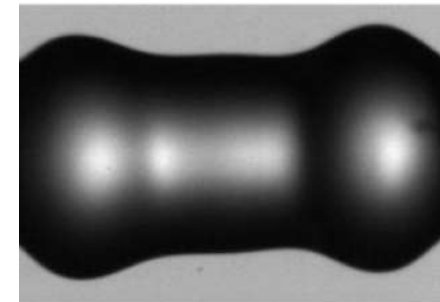
Pictures from Chireux et al. (2017)



Collision and film drainage



Hole opening and growth

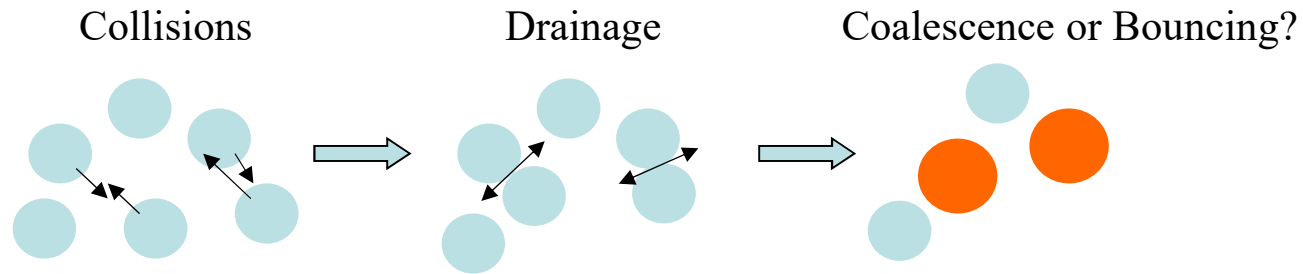


Shape relaxation

This course: case of coalescence limited by **film drainage** (step 2)

→ Generally a relevant case in bubble columns or emulsification processes with moderate surfactant concentrations (so that they do not induce molecular forces that retard hole opening).

During a collision: coalescence or bouncing?

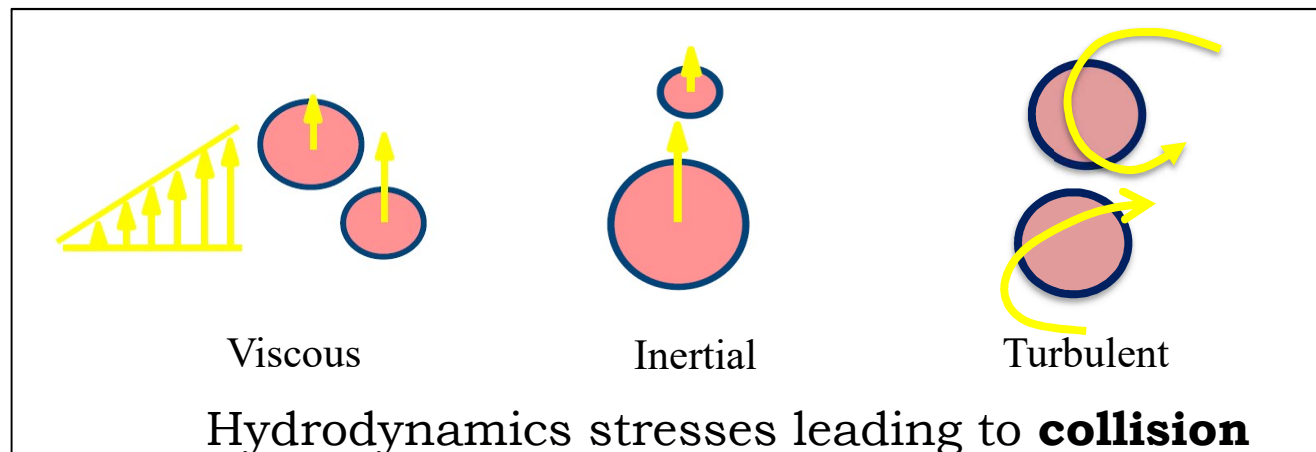


An hydrodynamic stress causes collisions.

- Droplets interact during a characteristic time t_i (**time of interaction** or contact)
- Liquid **film drainage** requires a characteristic time t_d
- Coalescence if $t_d \leq t_i$; otherwise bouncing.

→ **Coalescence probability depends on the ratio of these 2 time scales.**

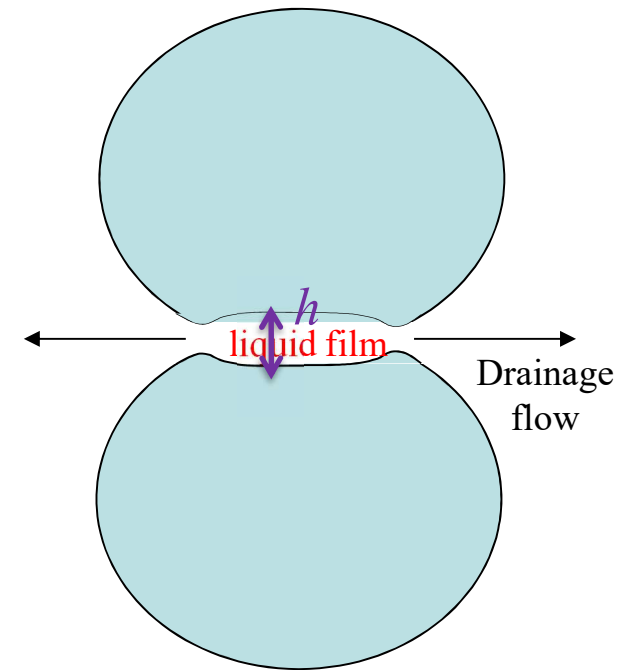
① drainage time ② contact time



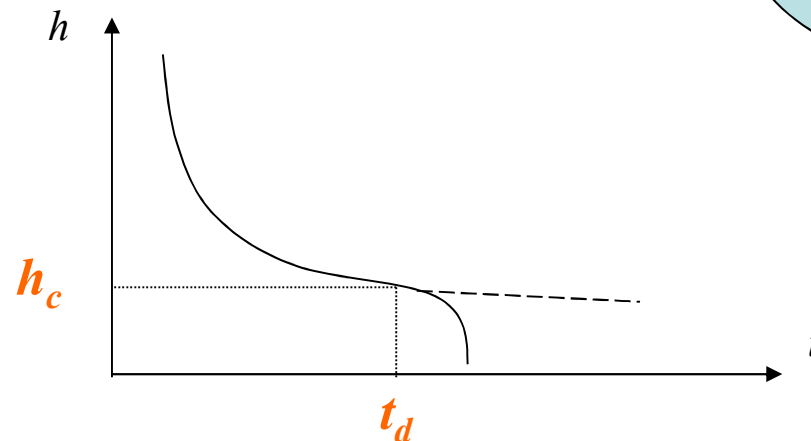
1) Hydrodynamics of film drainage

Kinetics of **film drainage** of the **liquid film** between two bubbles or droplets:

- Decrease of the film thickness h in time
- Until a critical thickness is reached, where the attractive van der Waals forces (short-range forces) become dominant and lead to hole opening *in the absence of repulsive forces to coalescence due to surfactants.*



Order of magnitude:
 $h_c = 10$ nm for a droplet of
 $d = 100$ μ m,
with a decreasing h_c with d .



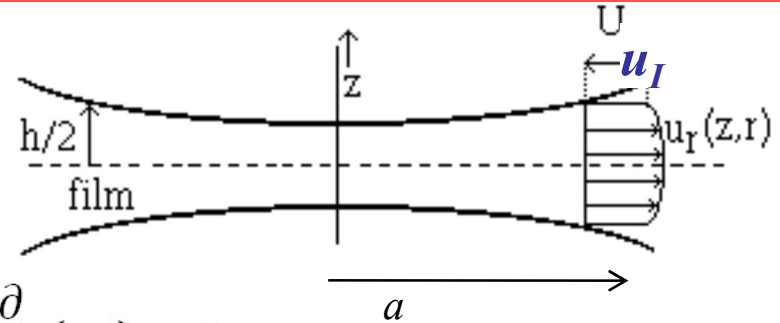
cf Ivanov et al. (1999)

Methodology : Estimation of drainage time t_d required to reach h_c by modelling the hydrodynamics in the liquid film.
→ Theoretical expressions in the following

1) Hydrodynamics of film drainage

Assumptions:

- $h/a \ll 1$ (a is the film radius, h its thickness)
- axial symmetry
- $Re_{film} \ll 1$



Navier-Stokes equations in the film:

$$\frac{1}{r} \frac{\partial}{\partial r} (r u_r) + \frac{\partial}{\partial z} (u_z) = 0$$

$$\frac{\partial P}{\partial z} \cong 0 \quad \longrightarrow \quad P = P(r)$$

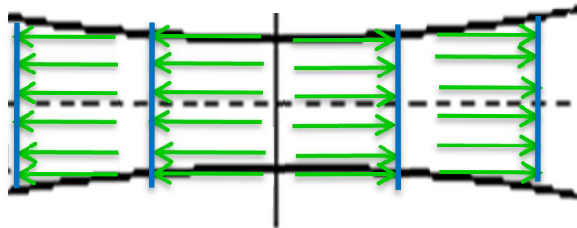
$$\rho_c \left(u_r \frac{\partial u_r}{\partial r} \right) = \mu_c \frac{\partial^2 u_r}{\partial r^2} - \frac{\partial P}{\partial r} + \mu_c \frac{\partial^2 u_r}{\partial z^2}$$

Film between bubbles

Liquid film between droplets

Different boundary conditions for u_I :

Bubbles: $\mu_d \ll \mu_c$
with clean interfaces



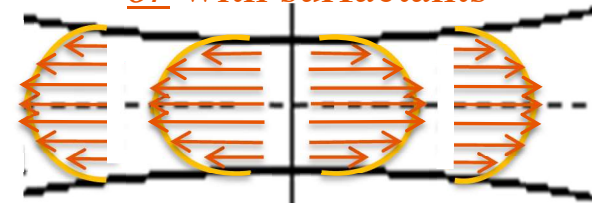
Mobile interface, zero stress

Clean interfaces
(theoretical case)
and $\mu_d \approx \mu_c$

Mobile interface

Droplets

very viscous drops $\mu_d \gg \mu_c$
or with surfactants



Immobile interface: no slip

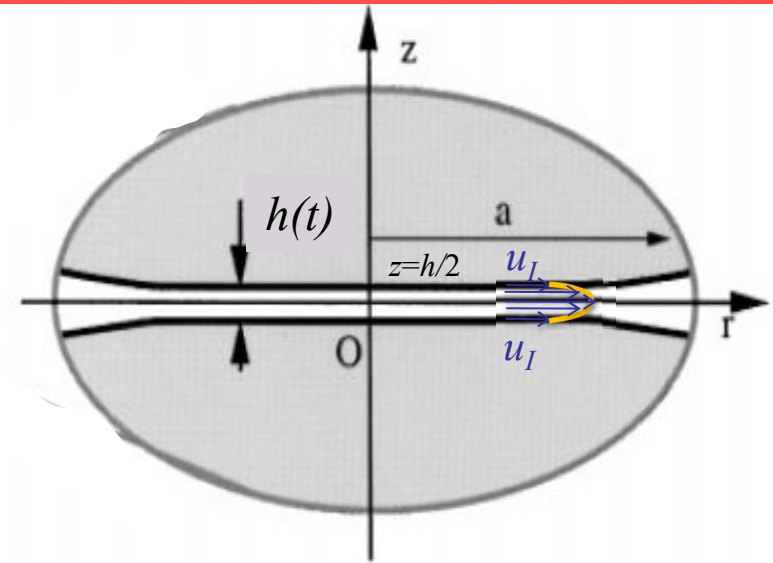
1) Hydrodynamics of film drainage – Reynolds equation

Case of liquid film between droplets

Lubrication flow:

$$Re_{film} \ll 1 \text{ \& } h/a \ll 1$$

Boundary condition: $z=h/2$: $u_R = u_I$
 $= 0$ if immobile interface



Momentum balance in the film:

$$\frac{\partial P}{\partial z} \cong 0$$

Couette-Poiseuille velocity profile in the film if $u_I \neq 0$
 Poiseuille profile if $u_I = 0$

$$\frac{dP}{dr} \cong \mu_c \left(\frac{\partial^2 u_r}{\partial z^2} \right) = cst \quad \longrightarrow \quad u_r = u_I + \frac{1}{2\mu_c} \frac{dP}{dr} \left(z^2 - \left(\frac{h}{2} \right)^2 \right) \quad (1)$$

Mass conservation (continuity equation integrated over z):

$$\frac{\partial}{\partial r} \left(r \int_{-\frac{h}{2}}^{\frac{h}{2}} u_r dz \right) = -r \frac{\partial h}{\partial t} \quad (2)$$

(1) and (2) =>

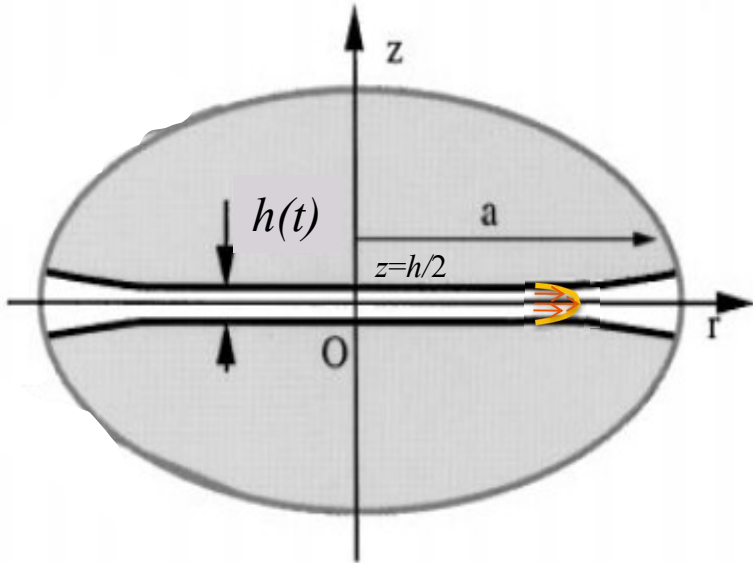
$$\frac{\partial h}{\partial t} = -\frac{1}{r} \frac{\partial}{\partial r} (ru_I h) + \frac{1}{12\mu_c} \frac{1}{r} \frac{\partial}{\partial r} \left(rh^3 \frac{dP}{dr} \right)$$

Drainage velocity

Equation of Reynolds
 = relationship between **drainage velocity** and **pressure gradient in the film**

1) Hydrodynamics of film drainage – Droplets with immobile interface

Case of liquid film between droplets – case of immobile interfaces: $u_I = 0$



Drainage velocity (or droplet approach velocity):

$$V = \frac{dh}{dt} < 0$$

From equation of Reynolds with $u_I = 0$:

$$\frac{dP}{dr} = \frac{6\mu_c V r}{h^3}$$

$$P(r) - P_c \cong \frac{3\mu_c V r^2}{h^3} \quad \text{with } P_c = P(r=0)$$

Expression of the normal force F exerted on the liquid film during drainage:

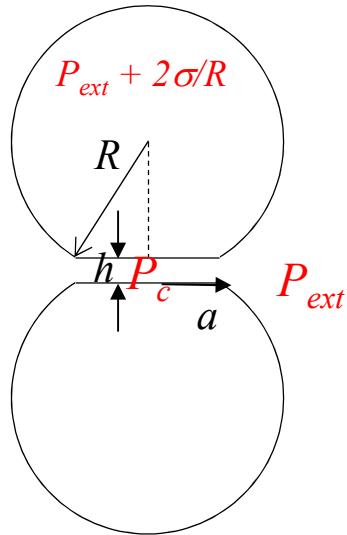
$$F = -2\pi \int_0^r (P(r') - P_c) r' dr' = -\frac{3\pi\mu_c V r^4}{2h^3}$$

→ Relationship between the exerted force and the drainage velocity

1) Hydrodynamics of film drainage – Deformable droplets with immobile interface

Liquid film between droplets - immobile interfaces

Case of deformable interfaces



$$F = -\frac{3\pi\mu_c V a^4}{2h^3}$$

Film with parallel edges $\Rightarrow P_c \cong P_{ext} + 2\sigma/R$
as given by the Laplace pressure:

$$F = \Delta P \pi a^2 \cong \frac{2\sigma}{R} \pi a^2$$

$$\Rightarrow a^2 = -\frac{4\sigma}{3\mu_c R V} h^3$$

$$-\frac{1}{h^3} \frac{dh}{dt} \cong -\frac{8\pi\sigma^2}{3\mu_c R^2 F}$$

$$\frac{1}{h^2} - \frac{1}{h_0^2} \cong \left(\frac{16\pi\sigma^2}{3\mu_c R^2 F} \right) t$$

Drainage characteristic time

$$\Rightarrow t_d \cong \left(\frac{3\mu_c R^2 F}{16\pi\sigma^2} \right) / h^2$$

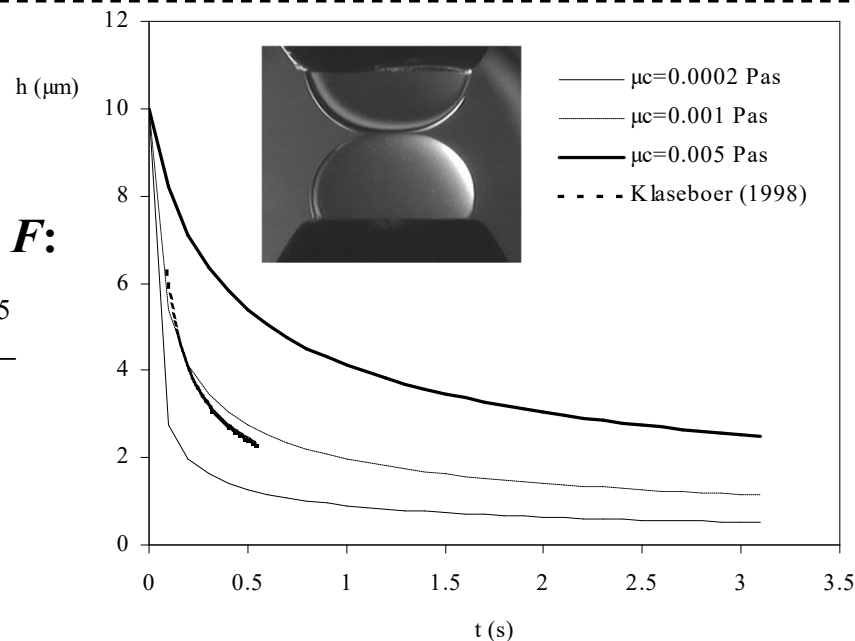
when $h \ll h_0$ (at long time)

Validation against measurements
of film thickness time evolution

By taking the buoyancy force as F :

$$F = \Delta\rho g \frac{4\pi R^3}{3} \Rightarrow t_d \cong \frac{\Delta\rho g \mu_c R^5}{4\sigma^2 h^2}$$

\rightarrow Increase of drainage time with
viscosity of the continuous phase



Comparisons with
experimental data of
Klaseboer et al.
(1998)

Oil droplets
 $R=1$ mm
 $h_0=10$ μm

1) Hydrodynamics of film drainage – Summary of characteristic times of drainage

Immobile interfaces (droplets) - constant force F

From the Reynolds equation:

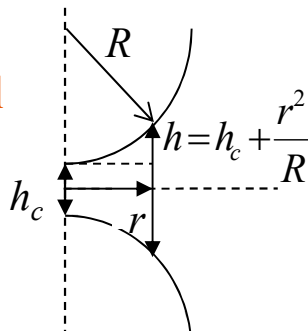
Deformable droplets

(see the previous slide)

$$t_d \cong \left(\frac{3\mu_c R^2 F}{16\pi\sigma^2 h_{cr}^2} \right)$$

→ Reynolds regime

Spherical droplets



$$t_d = \frac{3\pi\mu_c R^2}{2F} \ln(h_0/h_{cr})$$

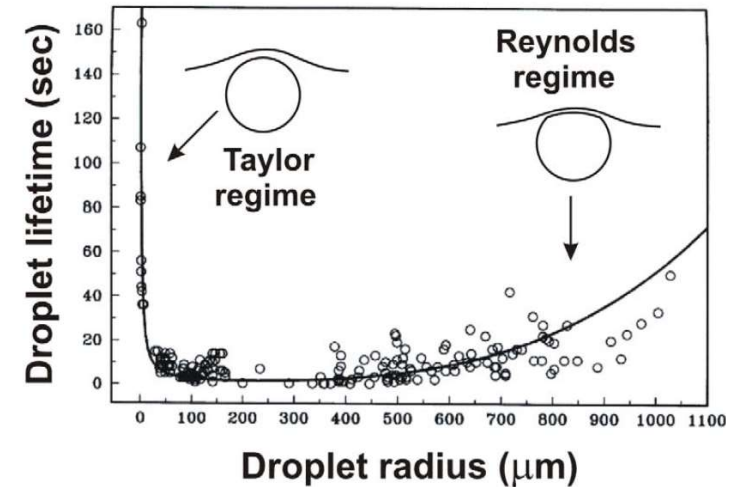
h_0 : initial distance for drainage

→ Taylor regime, valid for small spherical droplets ($Ca \ll 1$)

Note a difference between deformable and spherical droplets:

Deformable => the higher is F , the higher the drainage time because of interface deformation.

Spherical => the higher is F , the lower the drainage time.



Danov et al. (2010)

Fully mobile interfaces (clean bubbles) - constant approach velocity V

Viscous regime
($Re \ll 1$ in the film)

$$t_d = \frac{3\mu_c R}{2\sigma} \ln(h_0/h_{cr})$$

Chesters, 1975

Inertial regime
High Re

$$t_d = \frac{\rho_c V R^2}{8\sigma} \ln(h_0/h_{cr})$$

Summary of the different expressions
in Chesters (1991)

Questions: emulsion destabilization

Case of an emulsion with very small droplets.
Surfactants have been used as emulsion stabilizer.

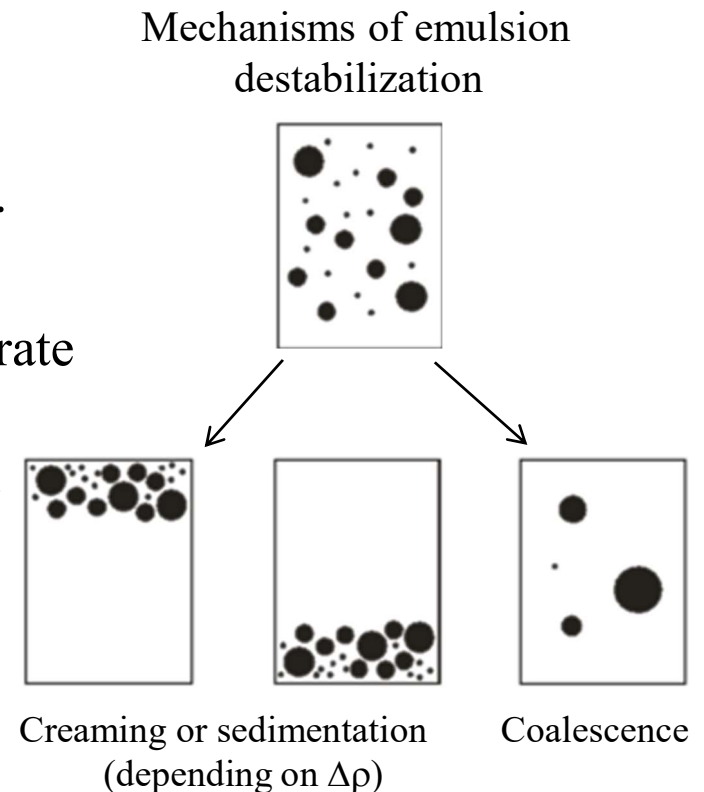
$Bo \ll 1 \Rightarrow$ spherical droplets

$Re_p \ll 1 \Rightarrow$ sedimentation or creaming negligible

Such an emulsion is **stable over a long period of time**.

Centrifugation (“increased gravity”) is used to accelerate the emulsion destabilization (= to observe if creaming/sedimentation/drop coalescence can occur).
 \rightarrow Thus it is an experimental **test to simulate aging**.

- Question:* what is the effect of centrifugation on:
- the drop sedimentation (or creaming) velocity?
 - the drainage time?



1) Hydrodynamics of film drainage

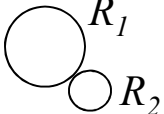
Previous expressions are valid for droplets/bubbles of same radius, in the limit of small deformation $h \ll a \ll R$.

Case of droplets/bubbles of different sizes:

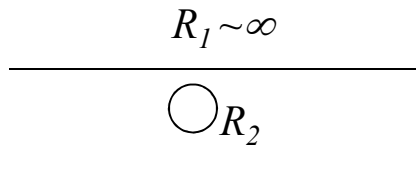
Chesters & Hofman (1982) have shown that these expressions remain valid provided an equivalent radius R_{eq} is used:

$$\frac{1}{R_{eq}} \cong \frac{1}{2} \left(\frac{1}{R_1} + \frac{1}{R_2} \right)$$

Examples:

$$1) \quad R_2 = \frac{R_1}{2} \quad R_{eq} \cong \left[\frac{1}{2} \left(\frac{1}{R_2} + \frac{1}{2R_2} \right) \right]^{-1} = \frac{4}{3} R_2$$


2) Drainage between a droplet and a planar interface:

$$R_2 \ll R_1 \quad R_{eq} \cong \left[\frac{1}{2R_2} \right]^{-1} = 2R_2$$


⇒ *Conclusion:* the radius of the smaller droplet drives the drainage kinetics.

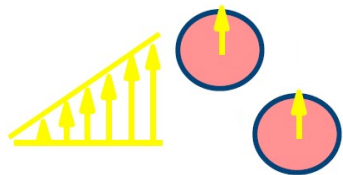
2) Time of interaction (or contact time)

Time of interaction t_i (or contact time):
time spent by the fluid particles in close proximity

Expressions given in Chesters (1991)

Viscous collisions: droplets/bubbles are brought together by the flow

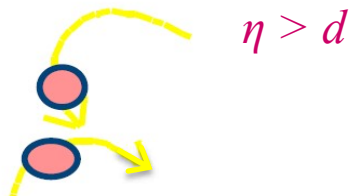
Shear flow



Contact force - Stokes regime	$F \sim 6\pi\mu_c RV \sim 6\pi\mu_c R^2 G_c$	} Solid particles, droplets with surfactants, very viscous droplets.
Contact time	$t_i \sim R/(G_c R) \sim G_c^{-1}$	

For clean bubbles or droplets: $F \sim 6\pi\mu_c R^2 G_c f(\lambda)$ with λ the viscosity ratio

Turbulent viscous



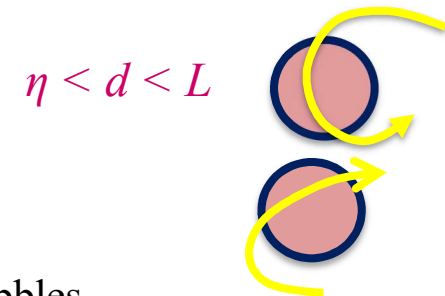
$$G_c = \left(\frac{2}{15} \frac{\rho_c \varepsilon}{\mu_c} \right)^{1/2} \longrightarrow \begin{cases} F \sim 6\pi(2\rho_c \mu_c / 15)^{1/2} R^2 \varepsilon^{1/2} \\ t_i \sim (2\rho_c \varepsilon / \mu_c)^{-1/2} \end{cases}$$

Inertial collisions: moderate and large Weber number

In a turbulent inertial regime (HIT)

Time for 2 droplets/bubbles to pass one another: $d/u'_d \sim d^{2/3} \varepsilon^{-1/3}$ in HIT

But the time of interaction is typically smaller: *because of inertia*, droplets/bubbles need time to deform, and coalescence can only occur during this time... *see next slide.*



2) Time of interaction (or contact time) – Inertial case

In the presence of inertia:

Colliding drops/bubbles have *kinetic energy*

→ Converted into *surface energy*: drops deform and film is created

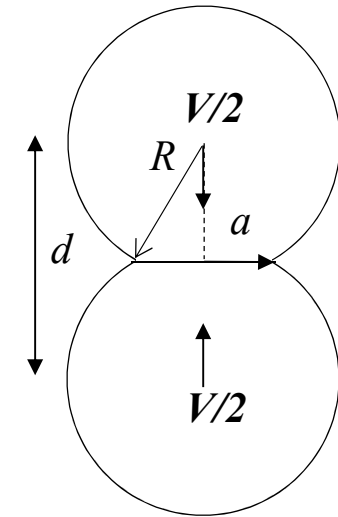
→ If no coalescence, converted again in *kinetic energy* (bouncing).

Actual time of interaction = time required for drop deformation.

Relative increase of surface: $\frac{\Delta s}{s} = \left(\frac{a}{2R}\right)^4$

Balance of energy: $4\pi R^2 \sigma \left(\frac{a}{2R}\right)^4 \approx \frac{1}{2} m \left(\frac{V}{2}\right)^2$

Chesters (1991)
Kamp et al. (2001)



Idealized shape when relative motion is arrested

$m = \frac{4}{3} \pi R^3 \rho_c (C_{am} + \rho_d / \rho_c)$ with C_{am} : added mass coefficient (1/2 for an isolated sphere, rising until 1 for spheres in close proximity)

$\Rightarrow (a/R) \cong (kWe/3)^{1/4}$ with $We = 2\rho_c V^2 R / \sigma$ and $k = C_{am} + \rho_d / \rho_c$

Distance variation $z = 2R - d$ between the drop centers is: $z/R \cong (a/R)^2 \cong (kWe/3)^{1/2}$

→ **Time of interaction = time for drop deformation:**

$$t_i = z/V \cong \left(2k\rho_c R^3 / 3\sigma\right)^{1/2}$$

Coalescence kernels: (A) coalescence probability and (B) collision frequency

- (A) *Probability of coalescence (or coalescence efficiency)*

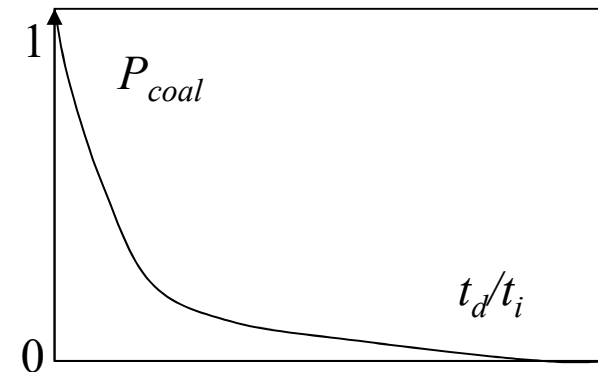
Several kernels available
in the review of Liao and Lucas (2010)

$$P_{coal} = \exp\left(-t_d/t_i\right)$$

t_i : interaction (or contact) time

t_d : drainage time

→ Based on the expressions shown in
the previous slides



General rules:

High contact time between droplets (as for concentrated media with low agitation)

⇒ high P_{coal}

Short contact time as in turbulent flows:

⇒ low P_{coal} - especially in case of high drainage time of film (case with surfactants).

Coalescence kernels: (A) coalescence probability and (B) collision frequency

- (A) *Probability of coalescence*

Several kernels available
in the review of Liao and Lucas (2010)

Drops in viscous simple shear

Partially mobile and deformable interfaces

$$t_d = \frac{\mu_d}{\sqrt{32\pi}} \left(\frac{FR^3}{\sigma^3} \right)^{1/2} \frac{1}{h_c}$$

$$P_{coal} = \exp(-t_d/t_i) \sim \exp(-k_0 \lambda Ca_c^{3/2} (8\pi\sigma R^2/A)^{1/3})$$

$$k_0, k_1 \sim 1$$

$$Ca_c = \frac{\mu_c G_c R}{\sigma}$$

Immobile and deformable interfaces

$$t_d \cong \left(\frac{3\mu_c R^2 F}{16\pi\sigma^2 h_c^2} \right)$$

$$P_{coal} = \exp(-t_d/t_i) \sim \exp(-k_1 Ca_c^2 (8\pi\sigma R^2/A)^{2/3})$$

+ other expressions of t_d on slide 47

Bubbles in turbulent flow

$$t_d \sim \frac{\rho_c V R^2}{8\sigma}$$

$$t_d/t_i = (2k\rho_c R^3/3\sigma)^{1/2}$$

$$P_{coal} = \exp(-t_d/t_i) \sim \exp\left(-\frac{1}{16} (3We/k)^{1/2}\right)$$

Probability of coalescence is higher at low Ca or We

=> coalescence is not expected in zones where intense breakup occurs. 53

Exercise: shearing of an emulsion and drop size evolution

A dilute emulsion of castor oil ($\rho_d = 961 \text{ kg/m}^3$, $\mu_d = 0.742 \text{ Pa}\cdot\text{s}$) dispersed in silicone oil ($\rho_c = 963 \text{ kg/m}^3$, $\mu_c = 0.048 \text{ Pa}\cdot\text{s}$) was produced, having drops of average size $d = 8.1 \text{ }\mu\text{m}$ and an interfacial tension $\sigma = 0.0043 \text{ N/m}$. As surfactants were employed as emulsifiers (at low concentration) and $\mu_d \gg \mu_c$, **interfaces are immobile**.

To study its evolution, **the emulsion is sheared in a cone-and-plate device, at an average shear rate of 11 s^{-1}** .

The drops are **spherical**, not deformed by gravity ($\text{Bo} \ll 1$), sedimentation or the imposed shear flow ($\text{Ca} \ll 1$).

(a) Estimate the critical film rupture thickness h_c by balancing:

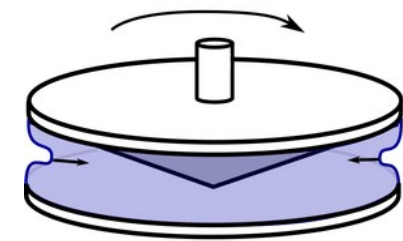
- the contact force exerted by the shear flow (drag in Stokes regime);
- the Van der Waals attraction force between equal spheres:

Hamaker constant $A \sim 10^{-20} \text{ J}$.

$$F = \frac{AR}{12h_c^2}$$

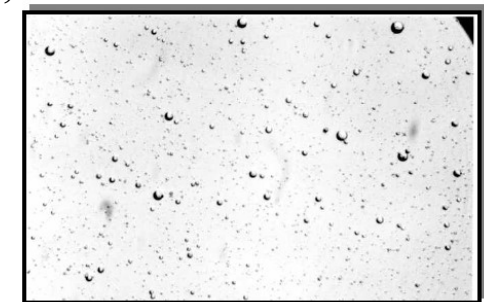
(b) Show that the contact time and the drainage time are of same order by considering that drainage begins at $h_0 \sim \frac{R}{4}$.

(c) How does the coalescence probability changes: (i) if the shear is increased? (ii) when the drop average diameter increases?

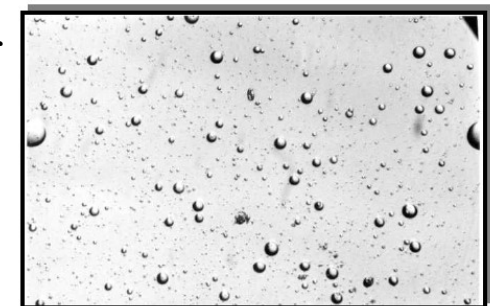


Cone and plate
rheometer

Hemingway et al. (2017)



Fresh emulsion $d = 8.1 \text{ }\mu\text{m}$



After 10mn of shearing: $d = 12.9 \text{ }\mu\text{m}$

Coalescence kernels: (A) coalescence probability and (B) collision frequency

- (B) *Collision frequency between droplets/bubbles in the flow*

It is defined based on the relative velocity u_d of the fluid particles of diameter d and d' , times the section area of collision.

For droplets/bubbles of same size:

$$C(d,d) = -ku_d d^2 n^2$$

Collision frequency
[m³/s]

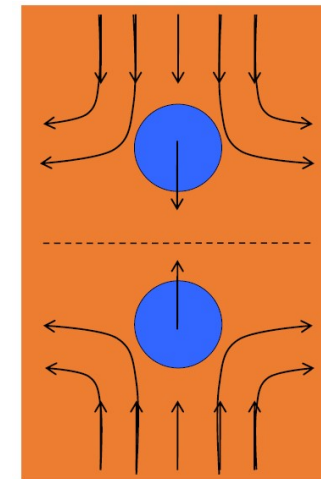
For droplets/bubbles of different sizes:

$$C(d,d') = ku_d (d_{eq}) \cdot d_{eq}^2 n(d)n(d')$$

$$d_{eq}^{-1} = \frac{d + d'}{2dd'}$$

u_d depends on the local properties of the carrier flow.

In a turbulent flow: high $u_d \Rightarrow$ high frequency of collisions.
but low probability of coalescence.



Picture from
Garg and Basaran

Coalescence kernels: (A) coalescence probability and (B) collision frequency

• (B) *Collision frequency between droplets/bubbles*

For dilute media

- Expressions for fluid particles of same size

$$C(d,d) = -ku_d d^2 n^2$$

Viscous shearing

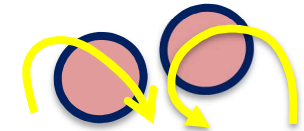
$$u_d \sim G_c d \text{ et } k = 2/3$$



Turbulent inertial collision

$$\eta < d < L$$

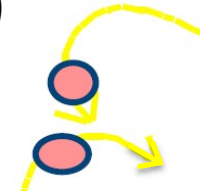
$$u_d \sim (\varepsilon d)^{1/3} \text{ et } k = (8\pi/3)^{1/2}$$



Turbulent viscous collision

$$d < \eta$$

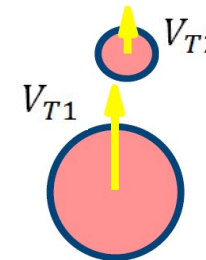
$$u_d \sim (\rho_c \varepsilon / \mu_c)^{1/2} d \text{ et } k = (2\pi/15)^{1/2}$$



- Case of *inertial collision due to a difference in rising velocity*

$$u_d \sim V_{T1} - V_{T2}$$

$$V_{T1} > V_{T2} \\ \text{for } d_1 > d_2$$



For concentrated media

Concept of effective media: use of an effective density and viscosity
for equivalent properties of the continuous phase

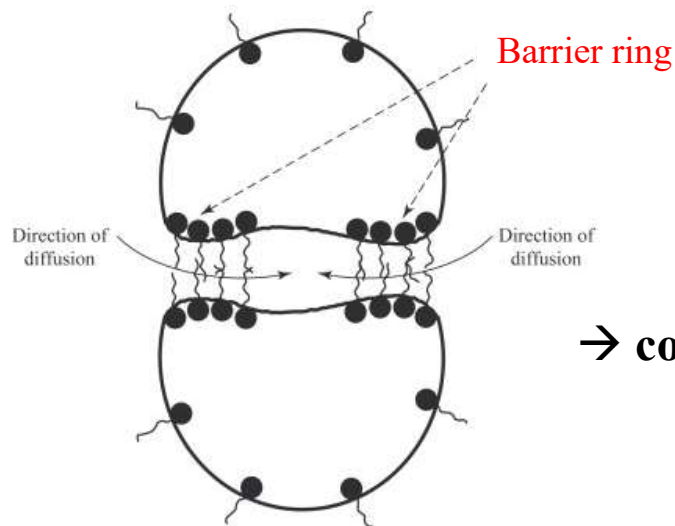
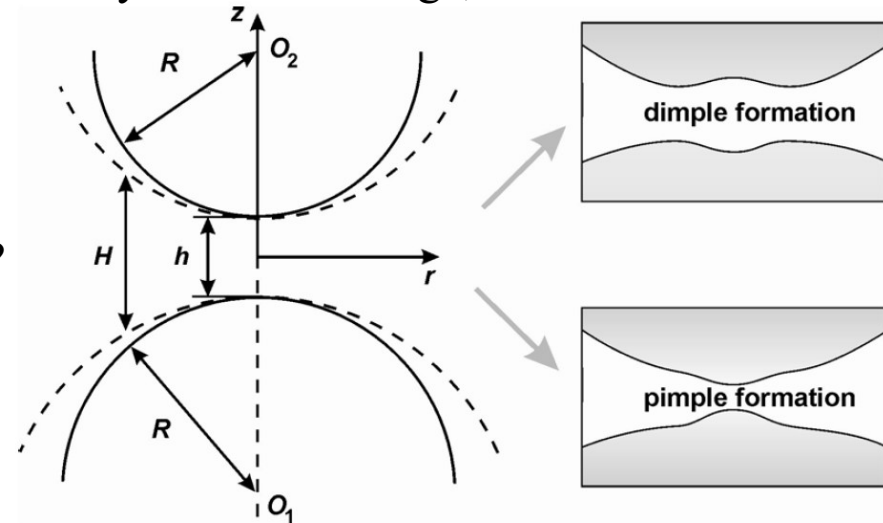
Open questions on coalescence

- Expressions for drainage time (and contact time) depend on theoretical assumptions: interface (im)mobility, drop and film shape, symmetry of film drainage, ...

→ need for experimental validations enabling a better understanding of coalescence

→ effect of mass transfer of a solute on drainage?

An instability in the gap can change the film topology from convex to concave shape
Danov et al. (2010)



Picture from Giribabu, Ghosh (2007)

- Presence of surfactants at high concentrations (generally the case in *emulsions*): coalescence becomes limited by the formation of a hole: ***barrier to coalescence due to molecular repulsive forces.***
- coalescence times become much larger than those of drainage, and experiments show that coalescence becomes ‘stochastic’...
No predictive model of the coalescence time in this case!

Brief summary [*including slides numbers for references*]

Models for Population Balance Equation so as to predict bubble/drop size distribution: [2, 3, 4]

Breakup kernels

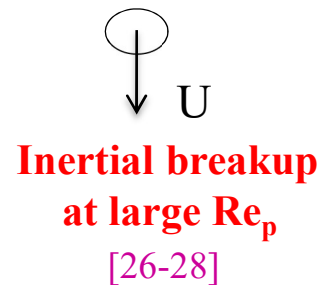
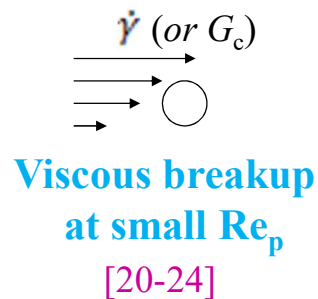
- Breakup frequency Γ [22, 35]
- Daughter drop size distribution β [36]

Coalescence kernels

- Collision frequency C [55, 56]
- Coalescence Probability P_{coal} [53]

Depending on the physics at the bubble/drop scale

Surface tension resists to deformation ($Oh \ll 1$) [12-18]
 A hydrodynamic stress causes **deformation:** [19]

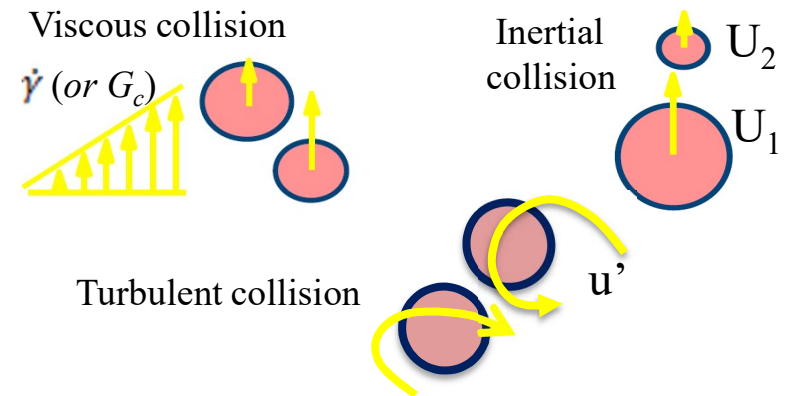


*Critical **Ca** or **We** for breakup*



Turbulent breakup,
 in the **inertial regime** ($d >$ Kolmogorov scale)
 in the **viscous regime** ($d <$ Kolmogorov scale)
 [30-34]

A hydrodynamic stress causes **drop collisions:** [41]



Interaction time (contact time) t_i [50, 51]
 Drainage time for the film t_d [42-47, 49]

→ *When drainage is the limiting step,*
 risk of coalescence when $t_d \leq t_i$ [41, 52]
 → **Surfactants inhibit coalescence** [15,51]

The main dimensionless numbers used to characterize interfacial phenomena

Particulate Reynolds number

$$Re_p = \frac{\rho_c V d}{\mu_c}$$

Inertial force / viscous force

V = relative velocity between drop and bulk fluid

Capillary number

$$Ca_c = \frac{\mu_c V}{\sigma} \quad \text{or} \quad Ca_c = \frac{\mu_c G_c R}{\sigma}$$

Viscous force / interfacial tension

↳ in a velocity gradient G_c experienced by the drop – see slide [20]

see slide [31] for Ca in the case of a turbulent flow

Weber number

$$We = \frac{\rho_c V^2 d}{\sigma} \quad \text{or} \quad We = \frac{\rho (G_c R)^2 d}{\sigma}$$

Inertial force / interfacial tension

$$We = Ca_c \cdot Re_p$$

↳ in a velocity gradient G_c experienced by the drop – see slide [27]

see slide [31] for We in the case of a turbulent flow

Bond (or Eötvös) number

$$Bo \text{ or } E\ddot{o} = \frac{\Delta \rho g d^2}{\sigma}$$

Buoyancy force / interfacial tension

Characterizes the (static) drop deformation due to gravity forces

Ohnesorge number

$$Oh = \frac{\mu_d}{\sqrt{\sigma d \rho_d}}$$

Internal viscous force / (interfacial tension . inertial force)

Characterizes the intensity of internal viscous forces, useful for highly viscous drops

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I- General approach: *Population Balance Equations for dispersed two-phase flows*

Need for closure models of breakup / coalescence

II- The interfacial tension: a mechanical property of the interface

- The interfacial tension: definition, examples and consequences
- Its role on interfacial phenomena
- Influence of surfactants

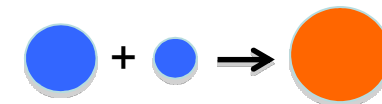
III- Breakup: physics and modelling

- General approach: definition of a critical non-dimensional number
- Breakup due to viscous forces
- Breakup due to inertial forces (laminar flow)
- Breakup due to turbulent fluctuations: turbulent inertial and viscous regimes
- Models of breakup frequency + daughter-drop size distribution



IV- Coalescence: physics and modelling

- Physical description of the coalescence process – Coalescence or bouncing?
- Drainage flow and time scale
- Time of interaction
- Models for coalescence probability + collision frequency



V- Brief summary and main non-dimensional numbers