Bubble rising and drainage of thin films of molten glass. Application to the foam stability in glass melting

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Glass melting basics

1st stage: "melting"

Sand (70%) Limestone (13%) Soda ash (12 %)

$SiO_2 + CaCO_3 + Na_2CO_3$ → $Na_2O, CaO, SiO_2 + CO_2$

Glass

Mixing of powders with granulometry 0.5 - 1 mm

- **Strong production of CO_2: 0.2 kg CO_2/1 kg of glass:**
  - **0.1 Nm^3/1 kg of glass ($4 \cdot 10^{-4}$ m^3), 1st source of foam.**
- **Formation of large quantity of bubbles due to the small solubility of CO_2 (10^8 bulles/m^3):**
  - **removing of bubbles.**

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Glass melting basics

2^{nd} stage: "fining"

Requirements in glass quality:

- Flat glass: < 1 bubble/20 m^2 $\Rightarrow$ 10 bubbles/m^3;
- Container (bottle): < 1 bubble/bottle $\Rightarrow$ 10^4 bubbles/m^3.

Rising of bubbles in glass:

- At T=1300°C, $\nu = 10^{-2}$ m^2/s:

Bubble diameter	1 mm	100 μm	10 μm
Rising time on 1 cm	3 mn	5 h	20 j

The aim of fining:

- **To grow the bubbles.**

Use of fining agents:

- **Release of gas (O_2, SO_2) at high temperature:**
  - 2^{nd} source of foam.

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Agenda

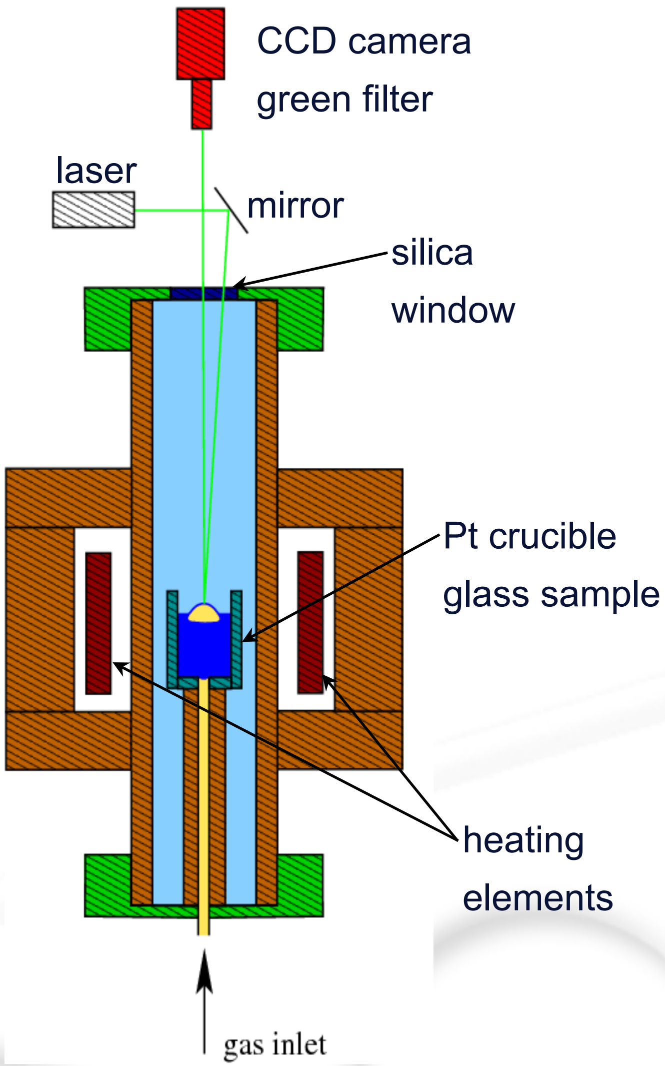
- Experiment on bubble drainage in molten glass
- Numerical simulation of bubble drainage
- Life time of bubbles
- Stability of vertical film

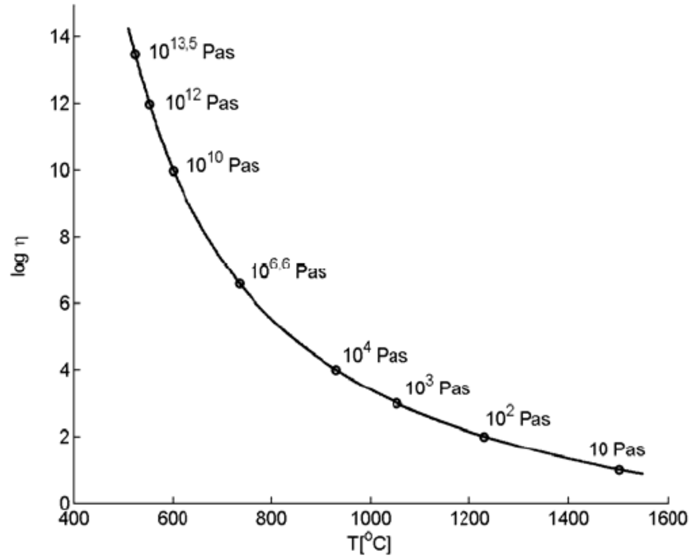
Foam in glass furnaces

- The stability of aqueous foams:
  - Presence of surfactants.
- No surfactant on highly viscous liquids:
  - “bare” films (Debrégeas *et al.*, 1998).
- Why the glass foams exist and are stable?
  - Chemical effect?
  - Thermal effect?

G. Debrégeas, P.-G. de Gennes, and F. Brochart-Wyart *Science* **279**, 1704-1707 (1998)

Experiment





Parame experin

- Bul
- **Temperature**
  - 1100 – 1400°C
  - temperature of the glass sample 1030 - 1330°C
- **Gas in the bubble**
  - N_2
- **Glass composition**

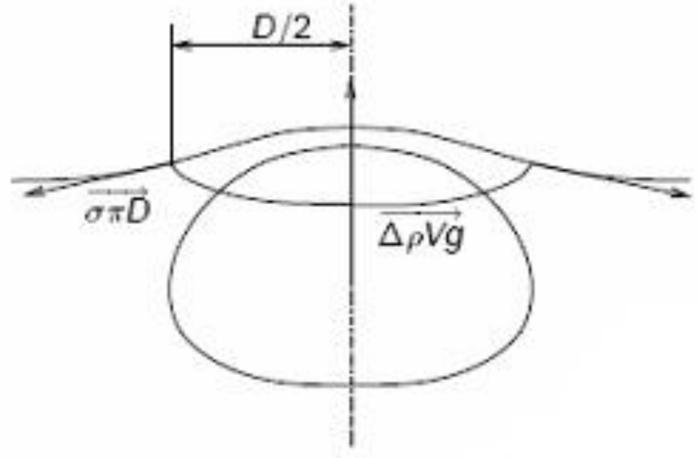
Parameters

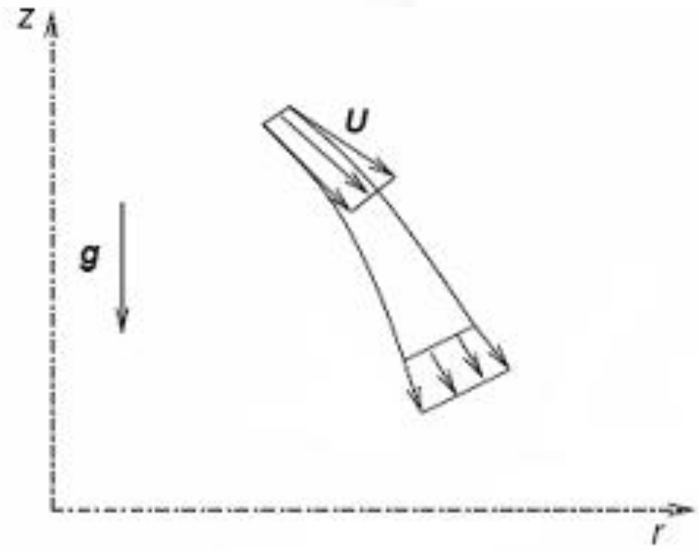
- Hydrodynamics interaction bubble/interface
  - Balance between gravity and surface tension

$$D^3 \Delta\rho g \approx D\sigma \Rightarrow Bo = \frac{\Delta\rho g D^2}{\sigma}$$

  - Balance between gravity and viscosity

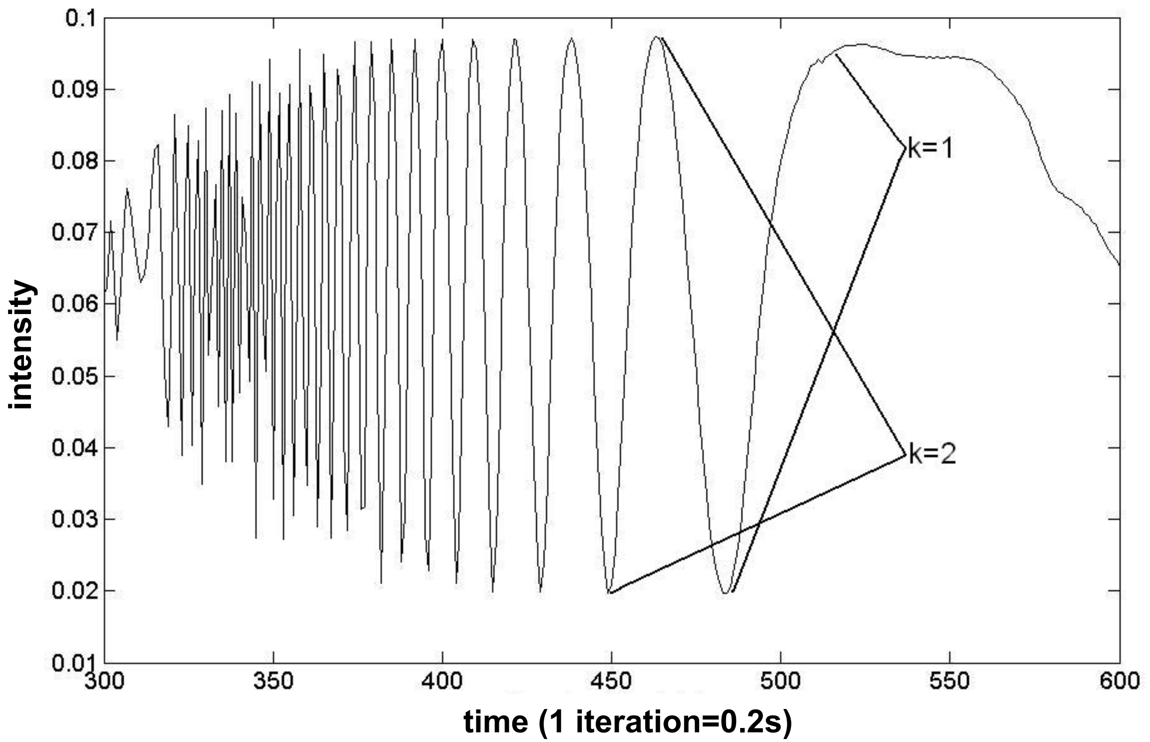
$$\mu \frac{U}{D} \approx \rho g D \Rightarrow U = \frac{\rho g D^2}{\mu} \quad \tau = \frac{\mu}{\rho g D}$$





Experiment – Evolution of thickness

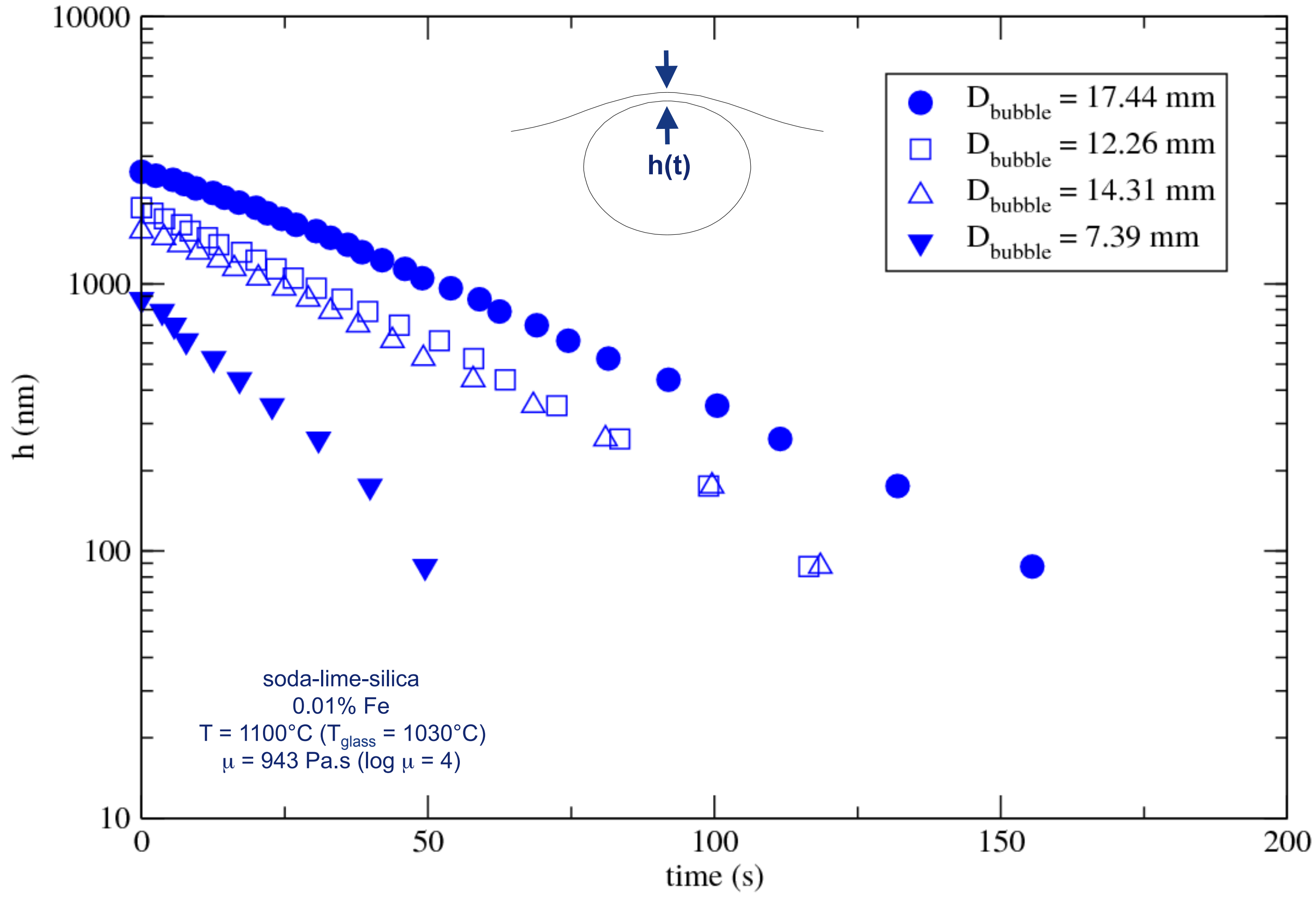
- **Computation of thickness:**



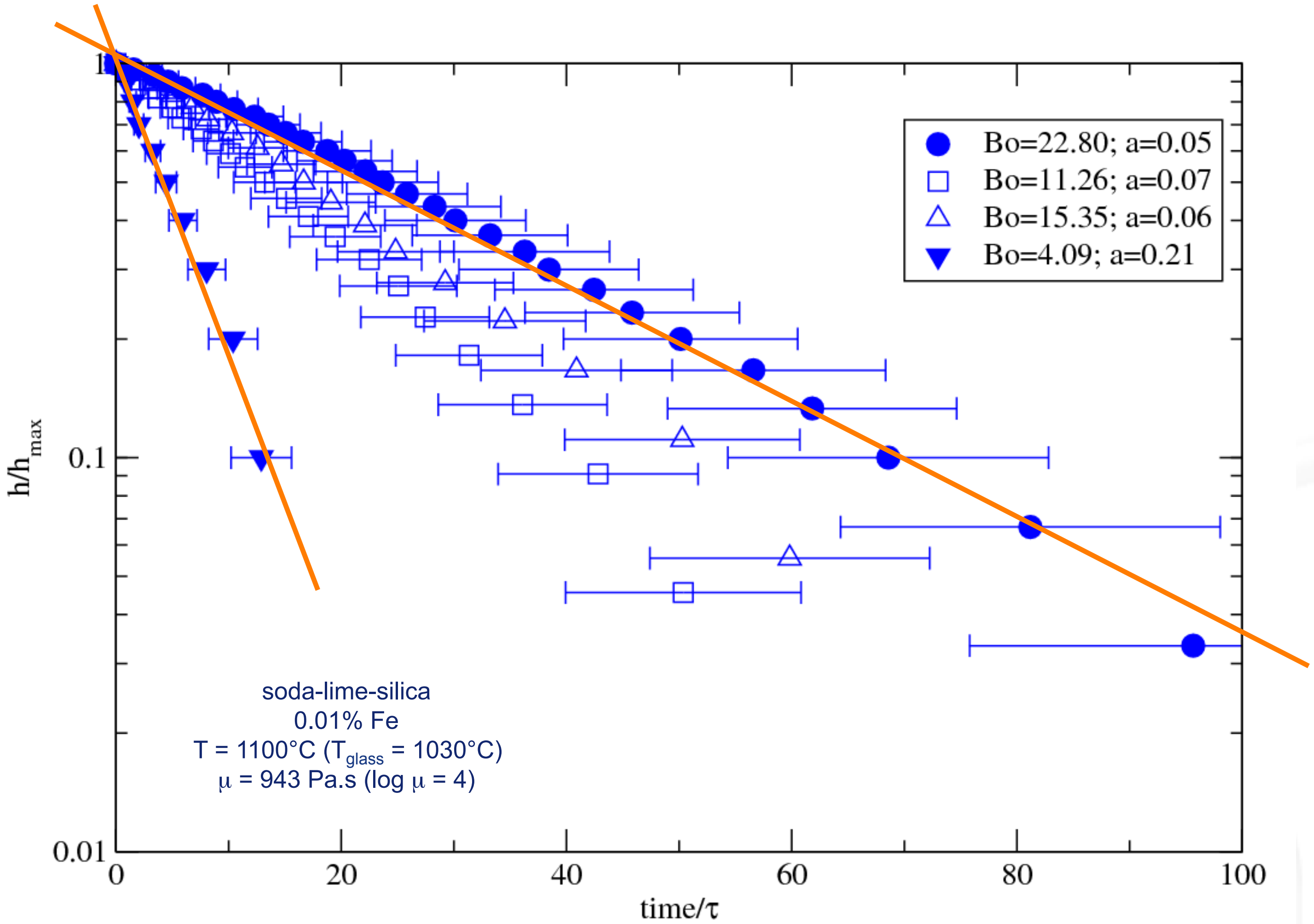
$$h_{I_{max}} = \frac{\lambda}{4n} \cdot (2k-1)$$

$$h_{I_{min}} = \frac{\lambda}{2n} \cdot k$$

Evolution of thickness (Fe cont. 0.01%)



Evolution of thickness (Fe cont. 0.01%)



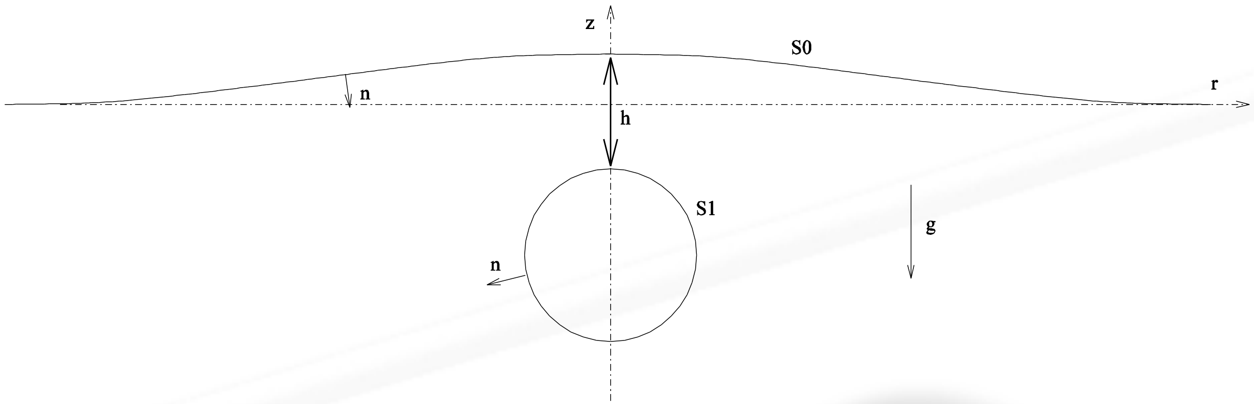
$$Bo = \frac{\rho \cdot g \cdot D^2}{\sigma}$$

$$\tau = \frac{\mu}{\rho \cdot g \cdot D}$$

$$h = h_0 e^{-a\frac{t}{\tau}}$$

Numerical simulation of bubble drainage

- Rising and film drainage of a bubble close to the free surface



- Small Reynolds number → creeping flow

Numerical simulation of bubble drainage

- Stokes equations + boundary conditions

$$div(\vec{u}) = 0,$$
$$\mu\nabla^2\vec{u} - grad(P) = 0,$$
$$\sigma \cdot \vec{n} = \left(\gamma div_s\vec{n} + \rho\vec{g} \cdot \vec{x}\right)\vec{n}$$
$$\vec{u} \cdot \vec{n} = \vec{V} \cdot \vec{n}$$

- Dimensionless form with

$$a, U_T = \frac{\rho g a^2}{3\mu}, a / U_T, U_T a / \mu$$

Numerical simulation of bubble drainage

- Stokes equations + boundary conditions

$$div(\vec{u}) = 0,$$

$$\nabla^2\vec{u} - grad(P) = 0,$$

$$\sigma \cdot \vec{n} = \left(\frac{1}{Bo} div_s\vec{n} + \vec{g} \cdot \vec{x} \right)\vec{n}$$

$$\vec{u} \cdot \vec{n} = \vec{V} \cdot \vec{n}$$

- Bond number

$$Bo = \frac{\rho g D^2}{\gamma}$$

Numerical simulation of bubble drainage

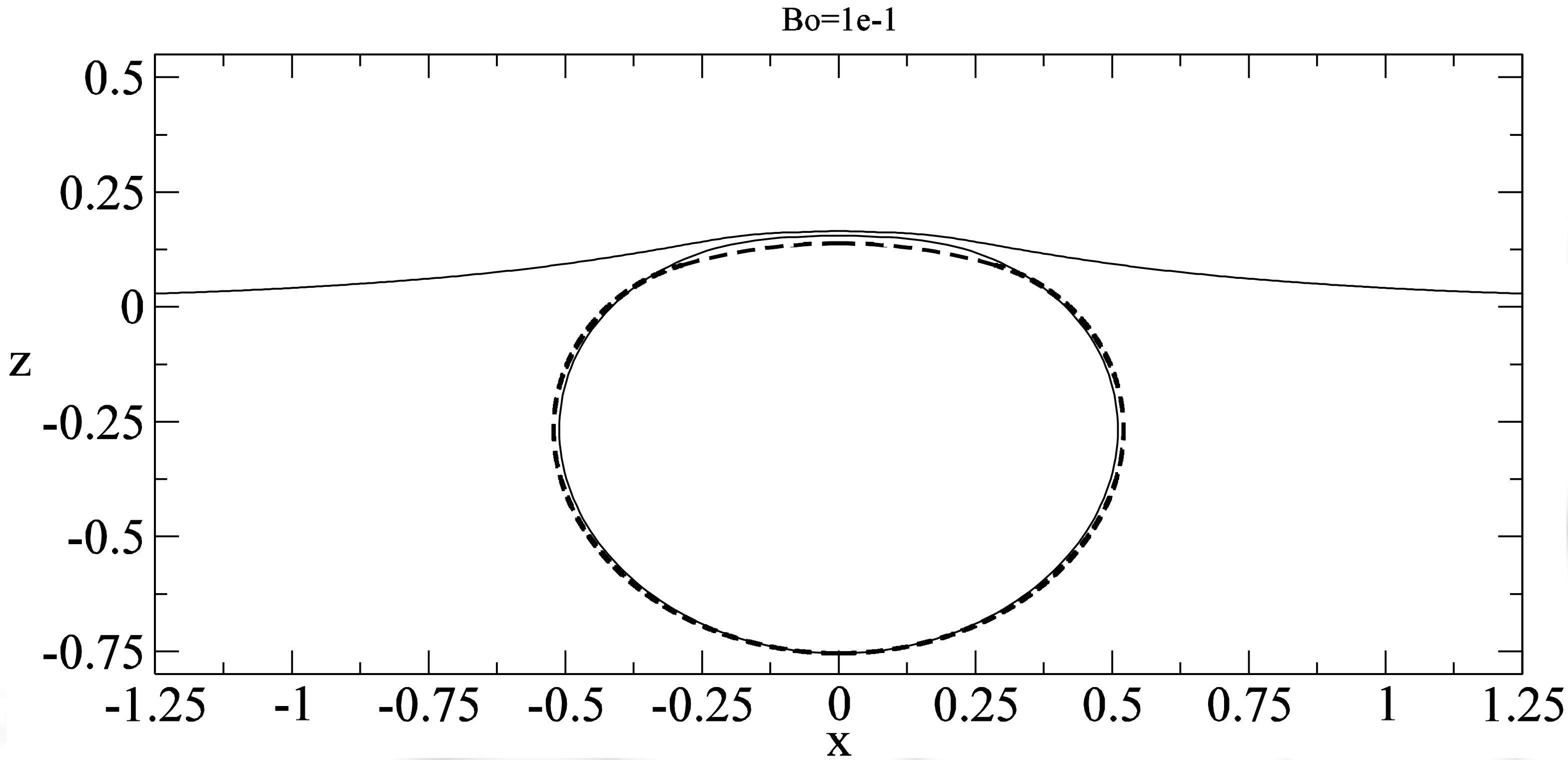
- Integral formulation of Stokes equations

$$\vec{u}(\vec{x}_0) = \frac{1}{4\pi}\int_S \left(\frac{div_s \vec{n}}{Ca} - 12z\right)\vec{n}\cdot G(\vec{x},\vec{x}_0)\,dS(\vec{x}) - \frac{1}{4\pi}\int_S \vec{u}(\vec{x})\cdot T(\vec{x},\vec{x}_0)\cdot\vec{n}(\vec{x})\,dS(\vec{x})$$

- Boundary Integral Method
  - Non conform elements
  - Self adaptive time step
  - Wielandt deflation to remove eigenvalues equal to 1.

Numerical simulation of bubble drainage

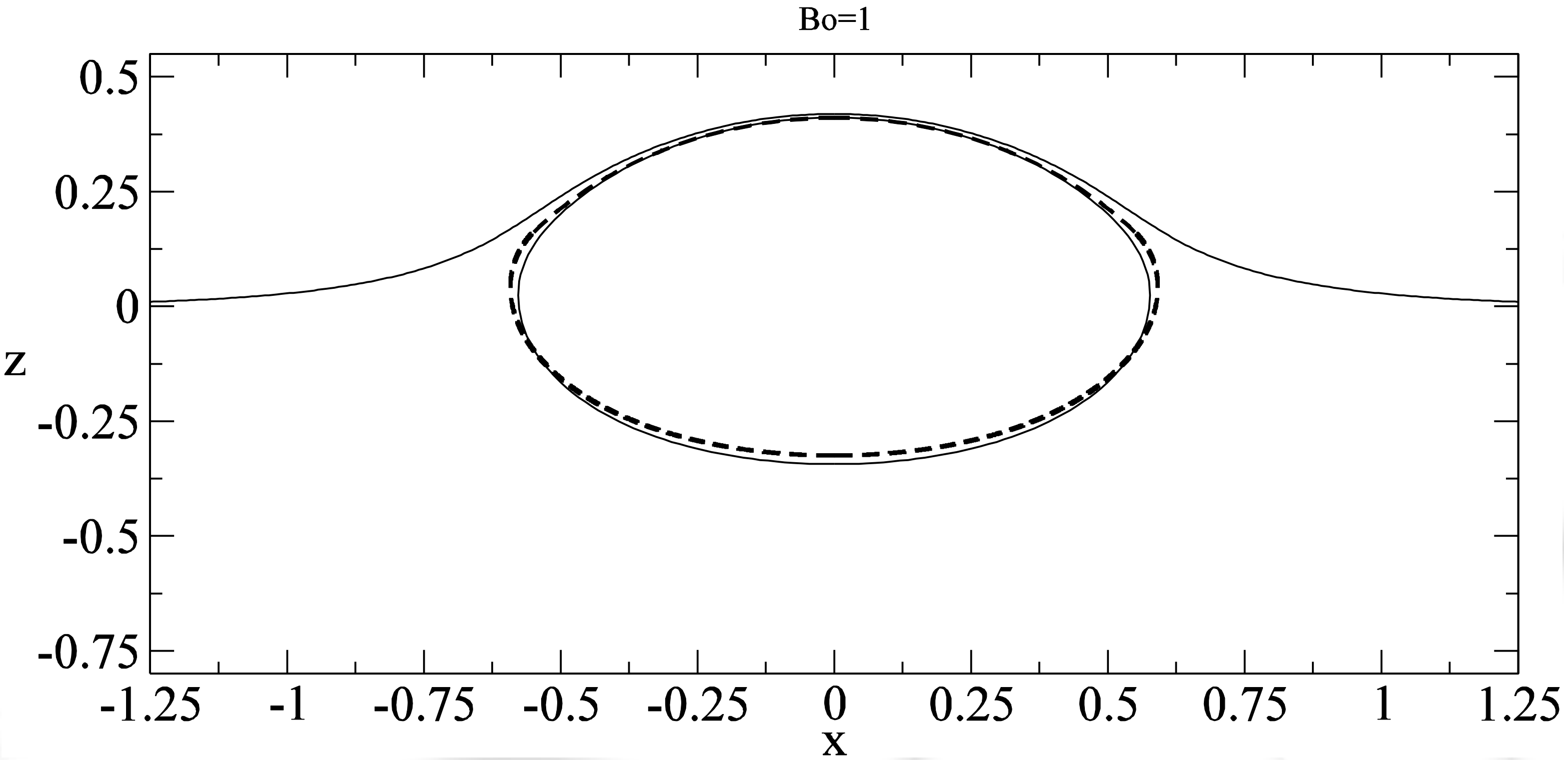
- Bubble shape



H. M. Princen. *J. Colloid Interface Sci.*, **18**:178-195, 1963

Numerical simulation of bubble drainage

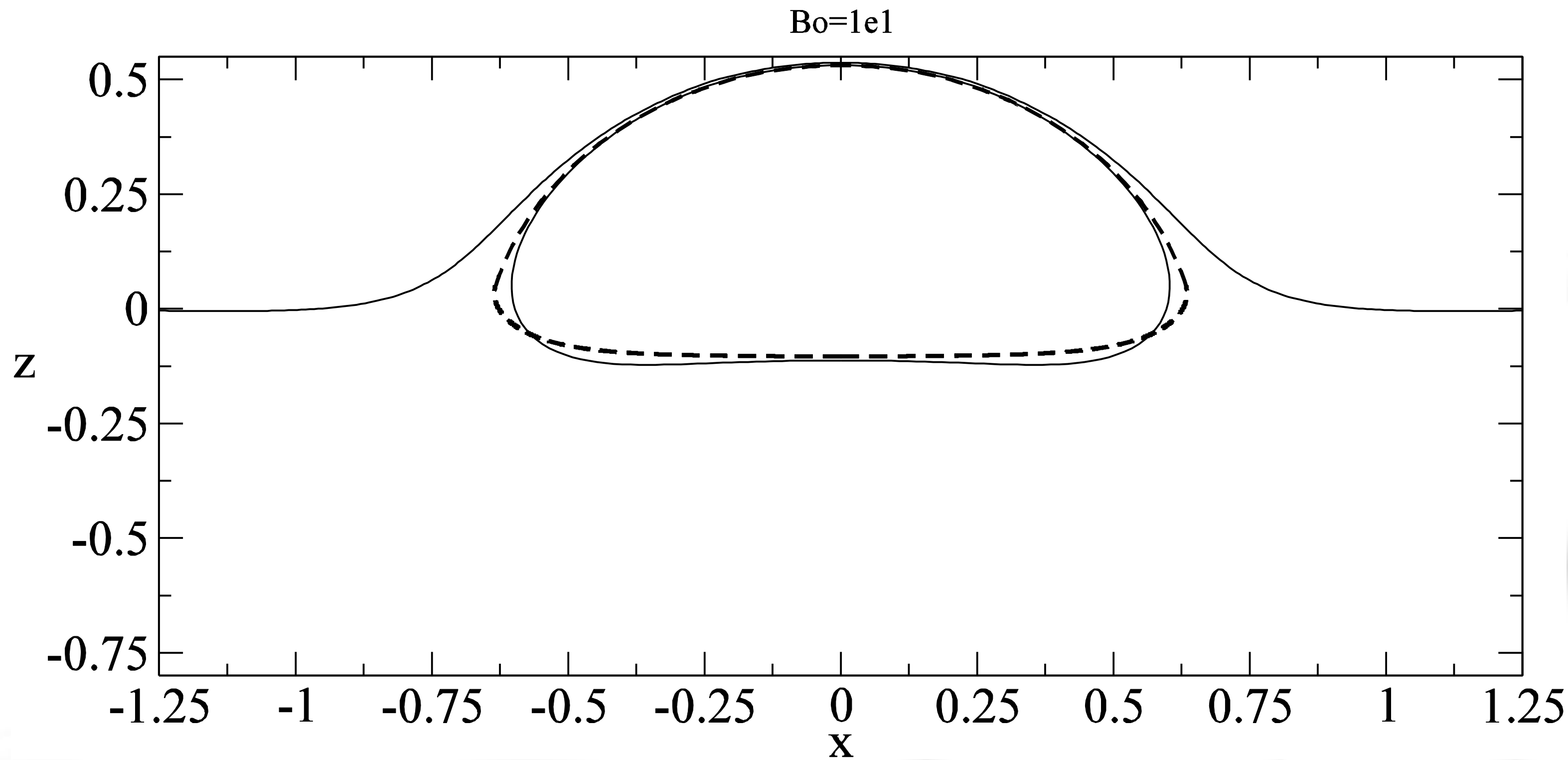
- Bubble shape



H. M. Princen. *J. Colloid Interface Sci.*, **18**:178-195, 1963

Numerical simulation of bubble drainage

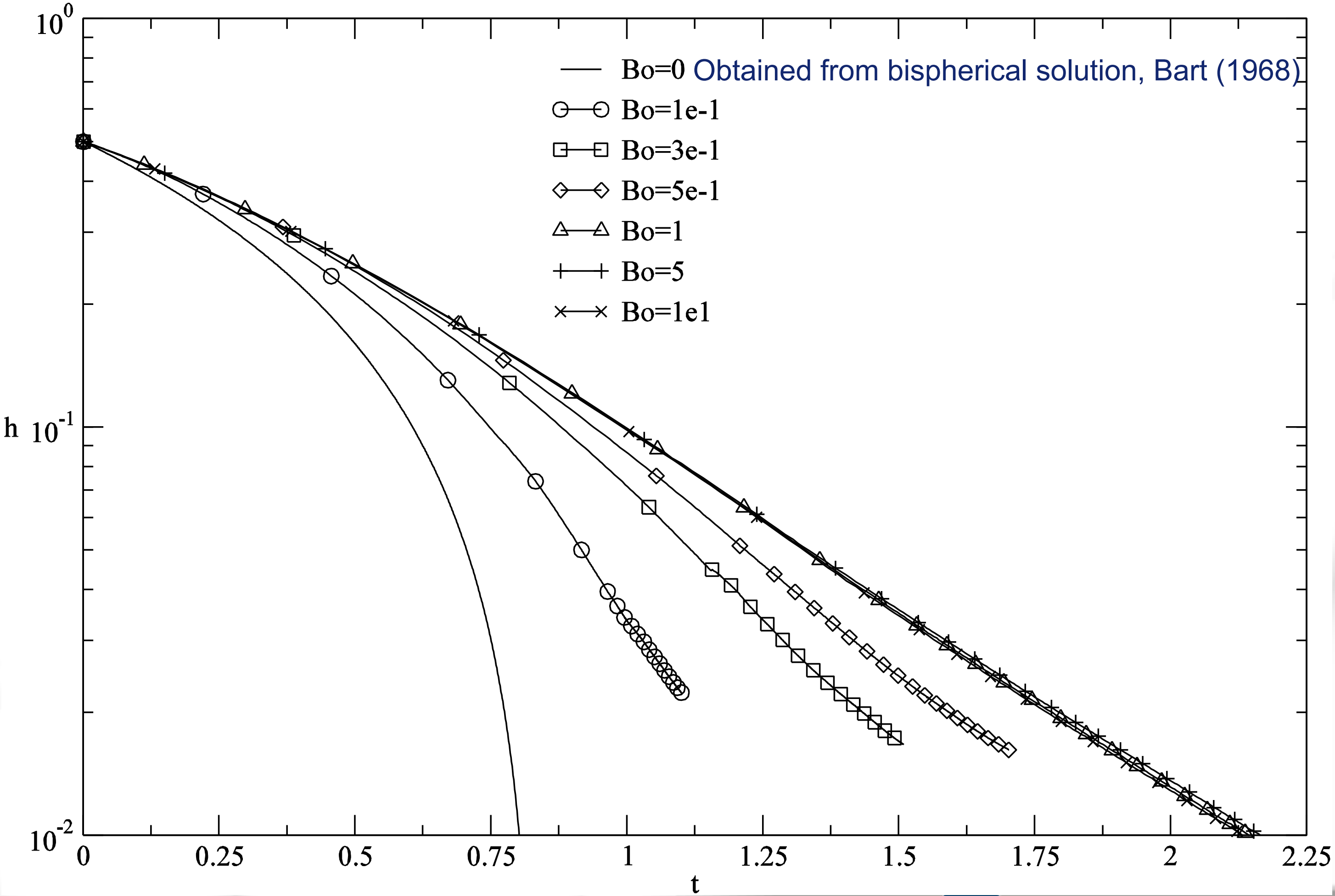
- **Bubble shape**



H. M. Princen. *J. Colloid Interface Sci.*, **18**:178-195, 1963

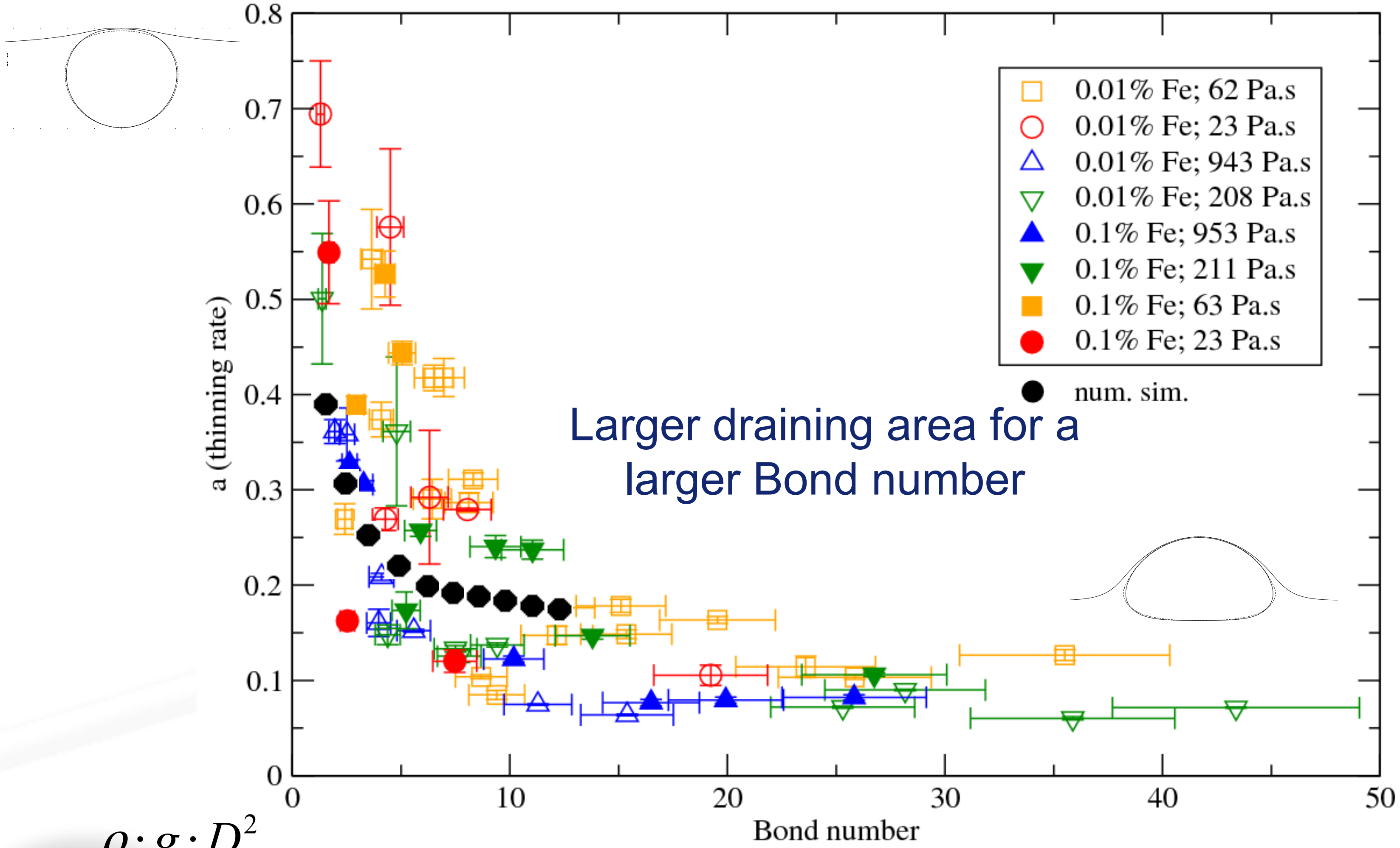
Numerical simulation of bubble drainage

- Film drainage vs time



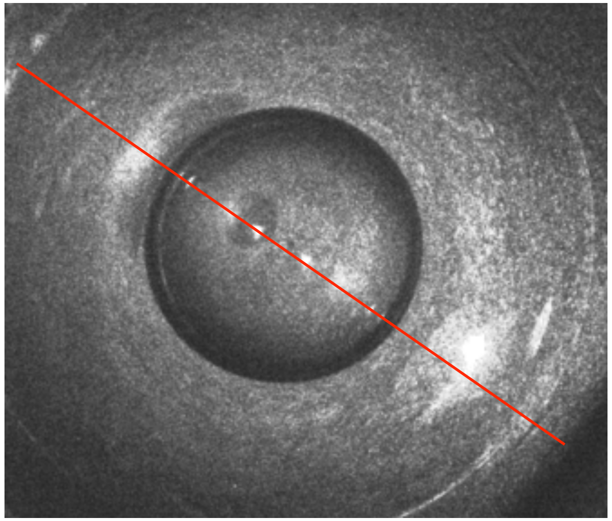
E. Bart. *Chem. Eng. Sci.*, **23**:193-210, 1968

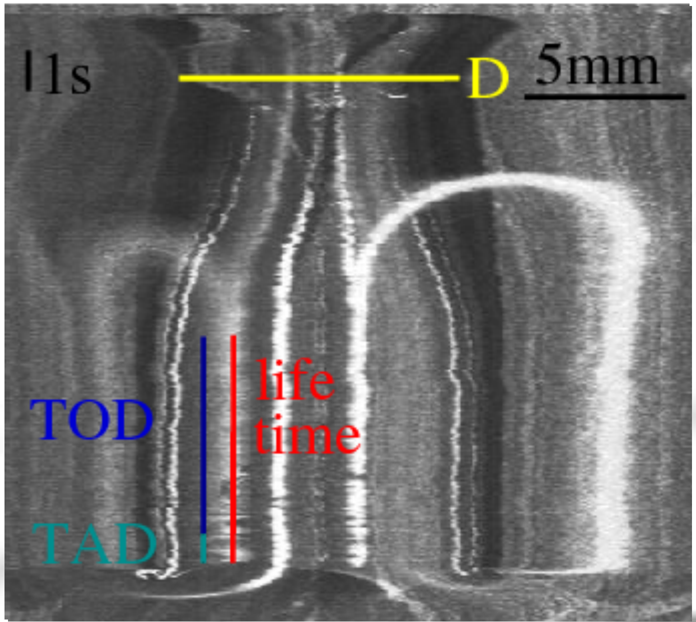
Thinning rate as a function of Bond number (Fe cont. 0.01% and 0.1%) + numerical simulation

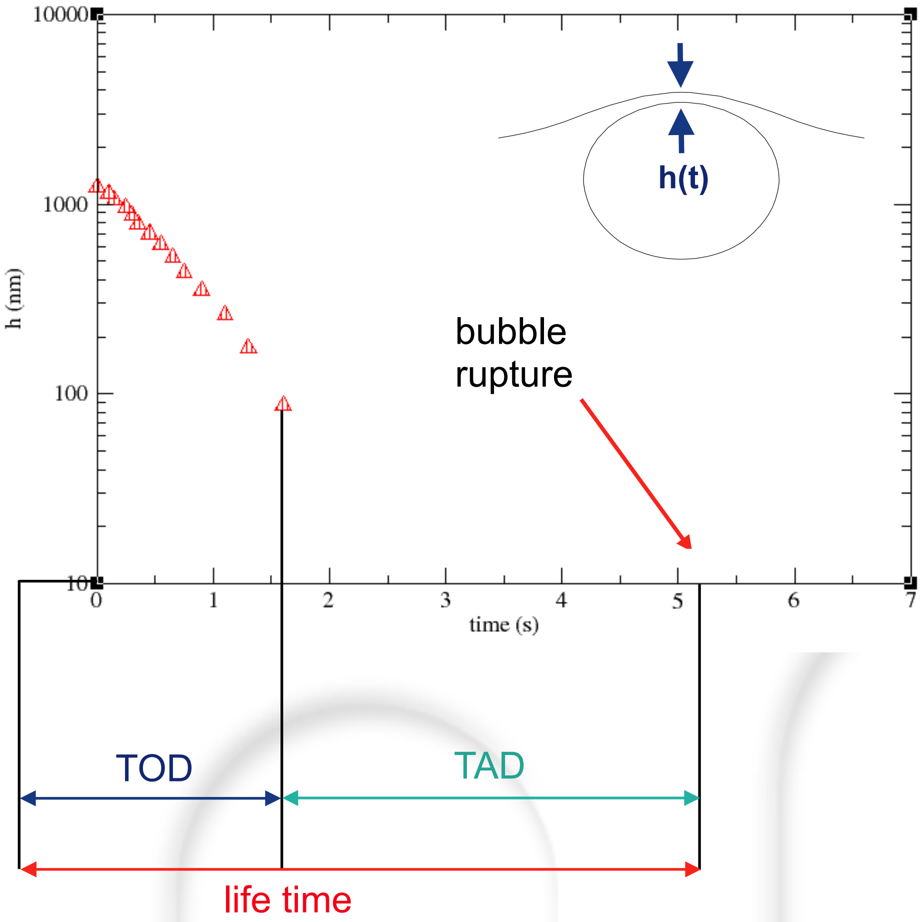


$$Bo = \frac{\rho \cdot g \cdot D^2}{\sigma}$$

Life time




1s
D
5mm
TOD
TAD
life
time

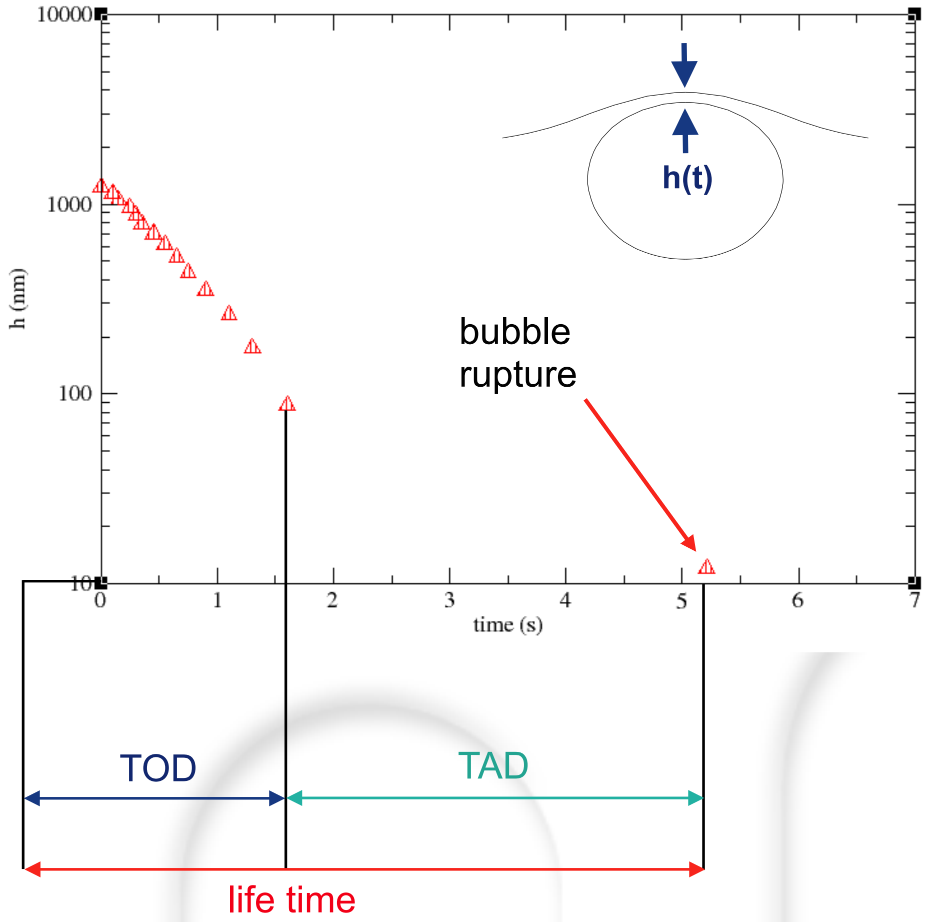

10000
1000
100
10
h (nm)
0
1
2
3
4
5
6
7
time (s)
h(t)
bubble
rupture
TOD
TAD
life time

Life time = time of drainage + time after drainage

- **TOD**
  - function of bubble size and liquid properties
  - predictable

$$TOD = f(\tau; Bo)$$

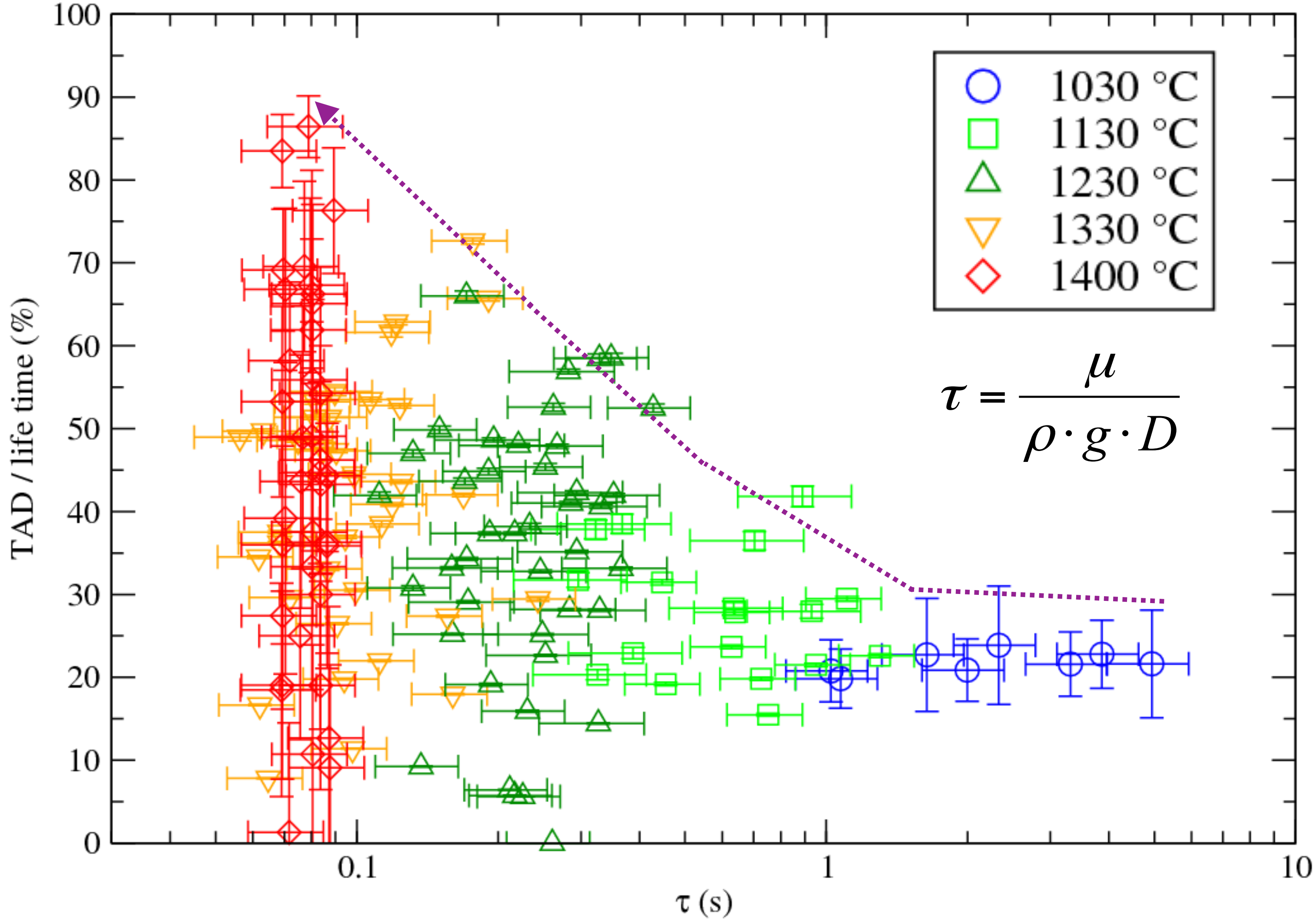
$$\tau = \frac{\mu}{\rho \cdot g \cdot D} \qquad Bo = \frac{\rho \cdot g \cdot D^2}{\sigma}$$



- **TAD**
  - unpredictable
  - changes with temperature

G. Debrégeas, P.-G. de Gennes, F. Brochard-Wyart, Science, vol.279, March 1998

Importance of TAD at a high T



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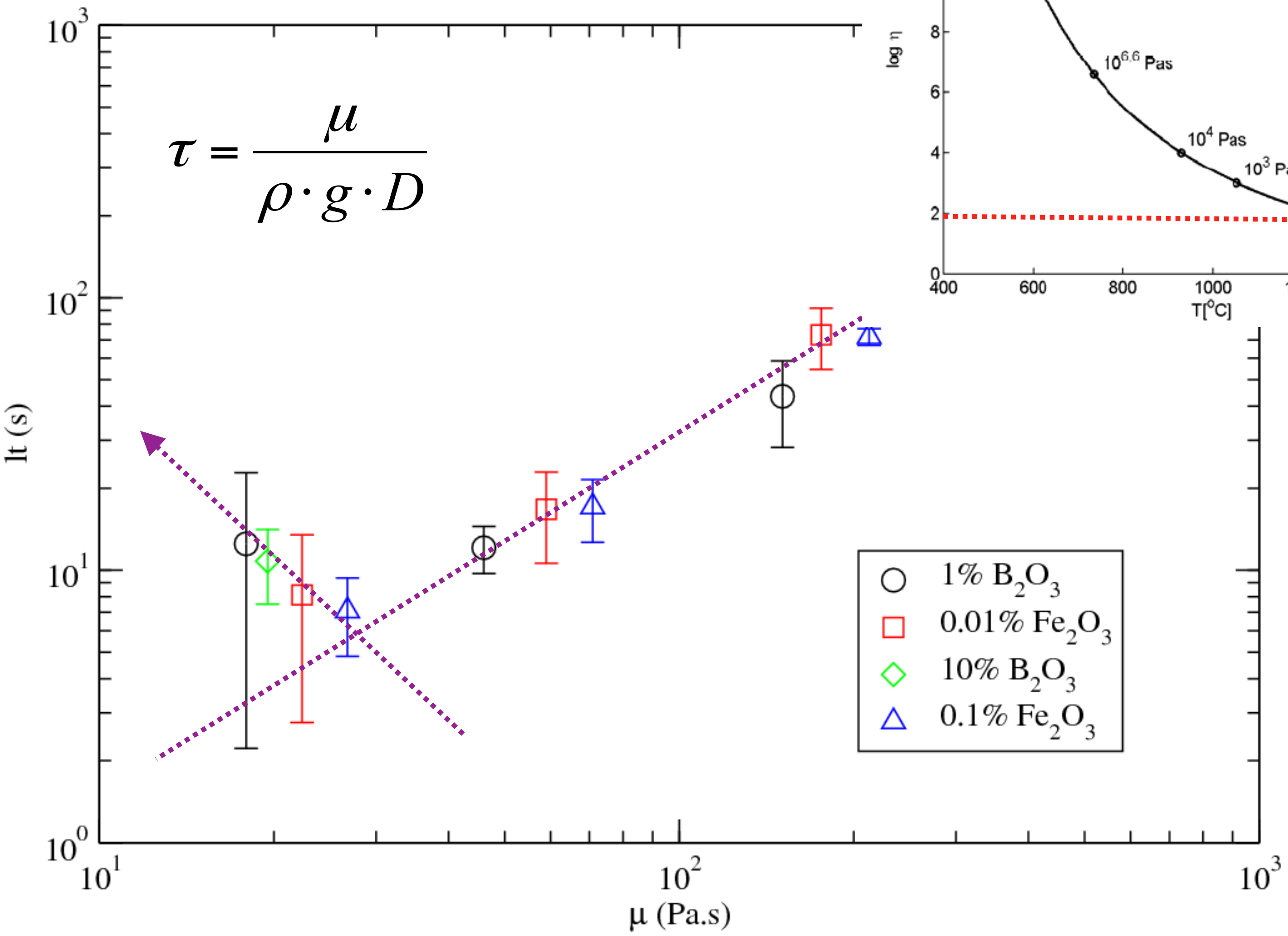
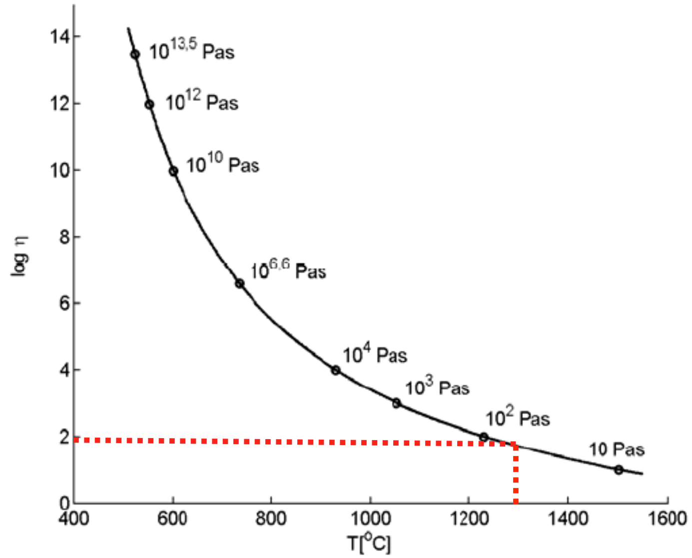
Life time



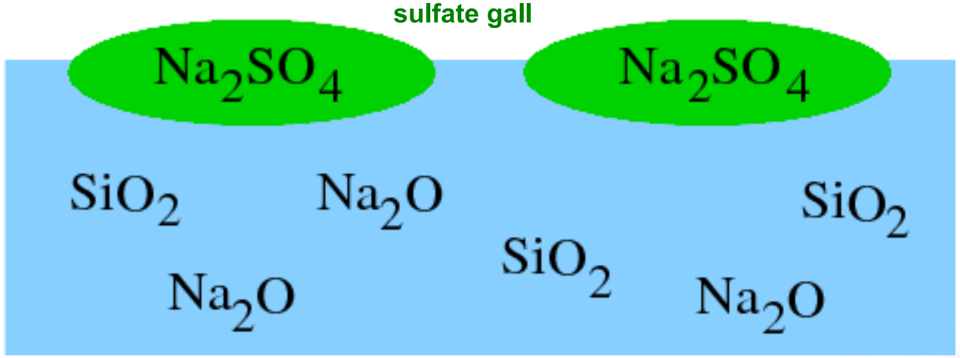
$$\tau = \frac{\mu}{\rho \cdot g \cdot D}$$
10^3
10^2
10^1
10^0
lt (s)
10^1
10^2
10^3
μ (Pa.s)
1% B_2O_3
0.01% Fe_2O_3
10% B_2O_3
0.1% Fe_2O_3



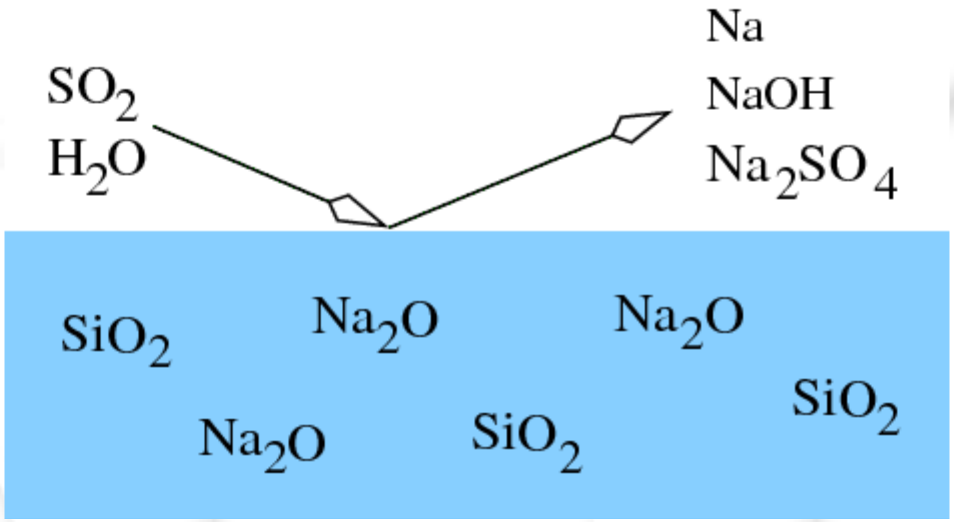
$10^{13,5}$ Pas
10^{12} Pas
10^{10} Pas
$10^{6,6}$ Pas
10^4 Pas
10^3 Pas
10^2 Pas
10 Pas
log η
T[°C]

Chemical behavior of Na_2SO_4

- **Below 1300°C**
  - $\gamma_{glass} = 300$ mN/m
  - $\gamma_{Na2SO4} = 200$ mN/m



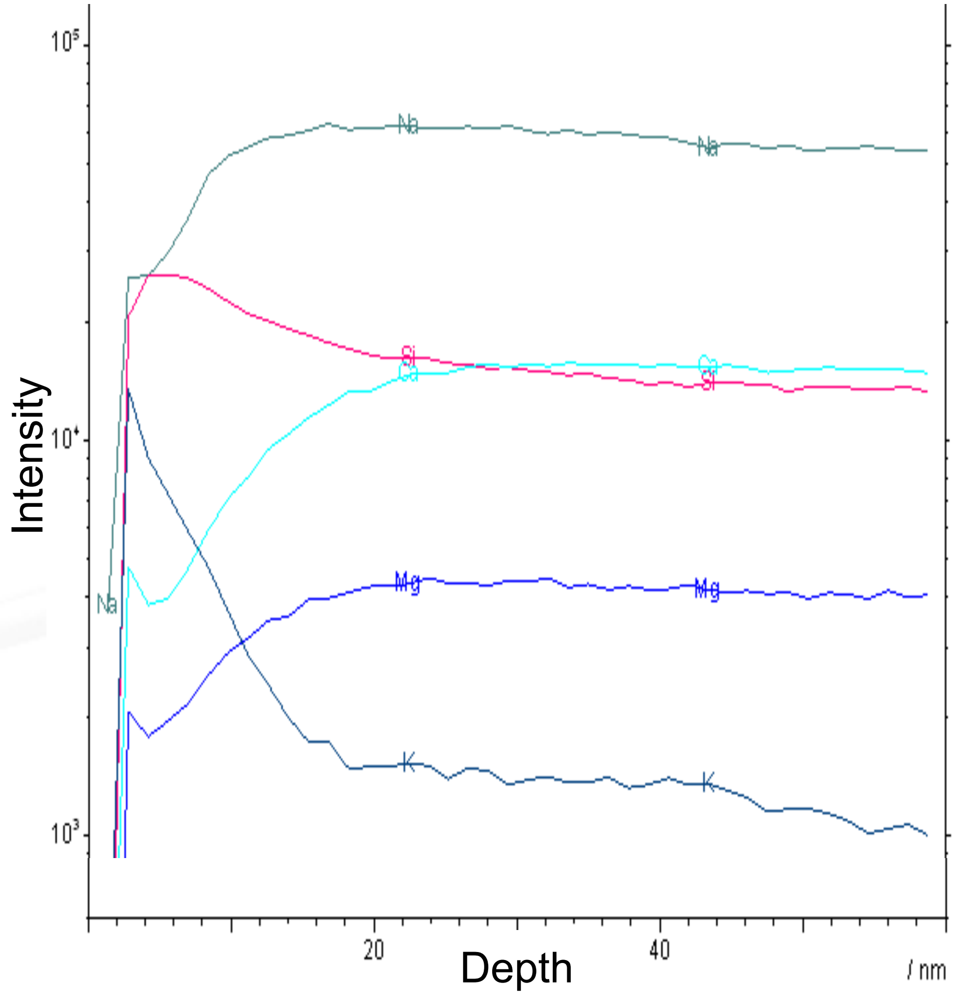
- **Above 1300°C**
  - Decomposition of sulfate
  - Enhanced evaporation of sodium
  - Variation of surface tension



Conroy A.R.; Manring W.H.; Bauer W.C. The Glass Industry; 1966
Bruckner R.; Glastechn. Ber. 1962

Thin film experiment

- Platinum loop 3cm
- Experiment at T = 1200 and 1400 °C
- Chemical composition in 50 nm (SIMS)
- Decrease of Na at the surface layer

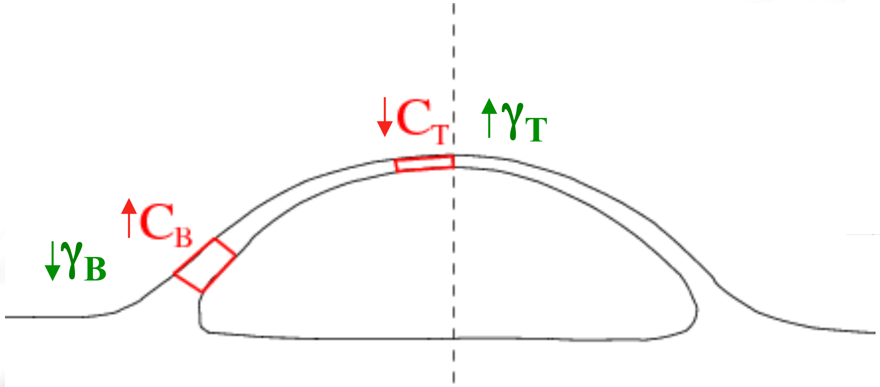


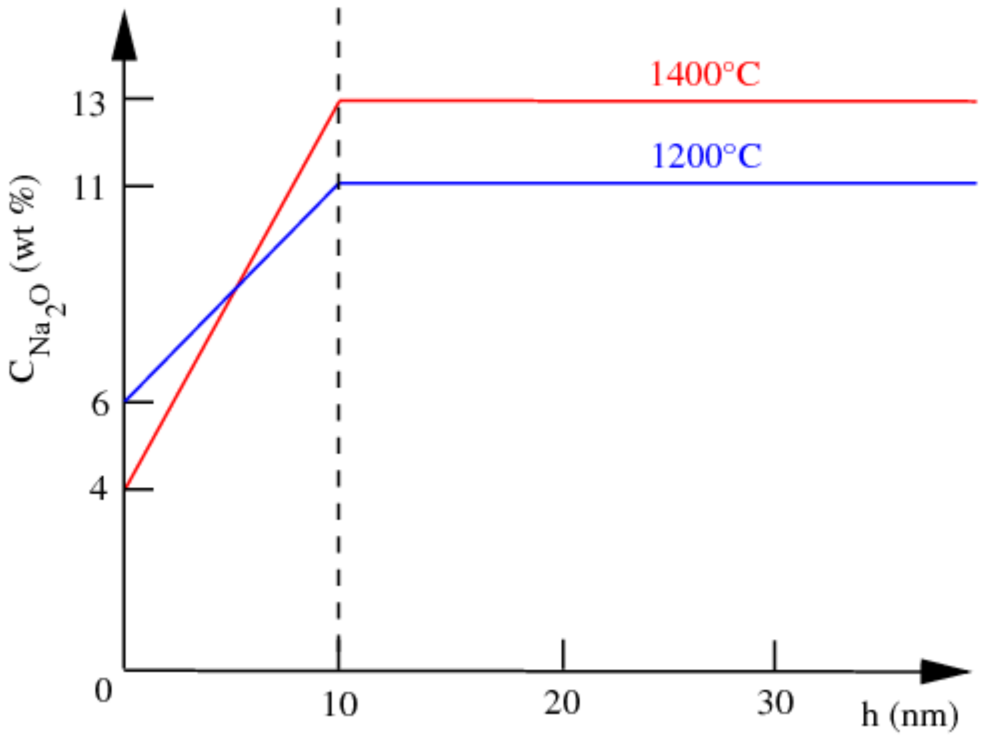
Variation of concentration and surface tension

$$h_T = 100nm$$

$$h_B = 1mm$$

1400°C	1200°C
$\Delta C_{Na_2O} = 1wt\%$	$\Delta C_{Na_2O} = 0.6wt\%$
$\Delta \gamma_{Na_2O} = 2.9\frac{mN}{m}$	$\Delta \gamma_{Na_2O} = 1.2\frac{mN}{m}$





Stability of vertical film

- Surface tension change with the film thickness.
- From a simple model of isotherm adsorption, the surface tension can be written like

$$\gamma = \gamma_0 + \frac{\delta\gamma}{1 + h/(2k)},$$

$$\delta\gamma = \left(\gamma_{\mathrm{SiO_2}} \frac{y_{\mathrm{SiO_2},0}}{y_{\mathrm{SiO_2},0} + y_{\mathrm{CaO},0}} + \gamma_{\mathrm{CaO}} \frac{y_{\mathrm{CaO},0}}{y_{\mathrm{SiO_2},0} + y_{\mathrm{CaO},0}} - \gamma_{\mathrm{Na_2O}}\right) y_{\mathrm{Na_2O},0}.$$

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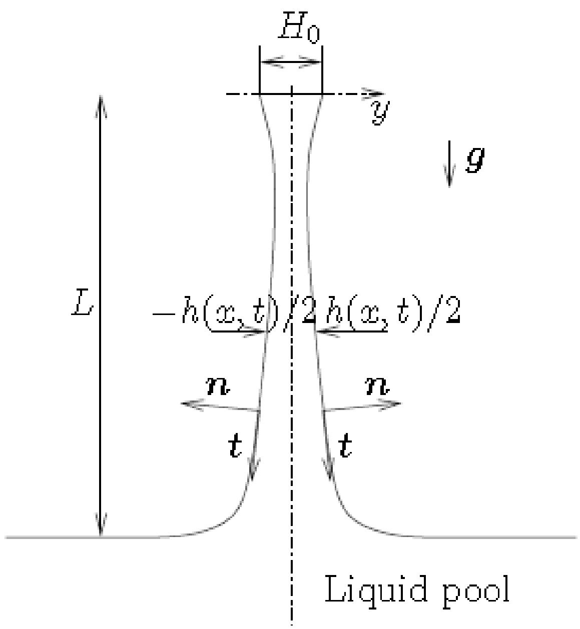
Stability of vertical film

Lubrication model



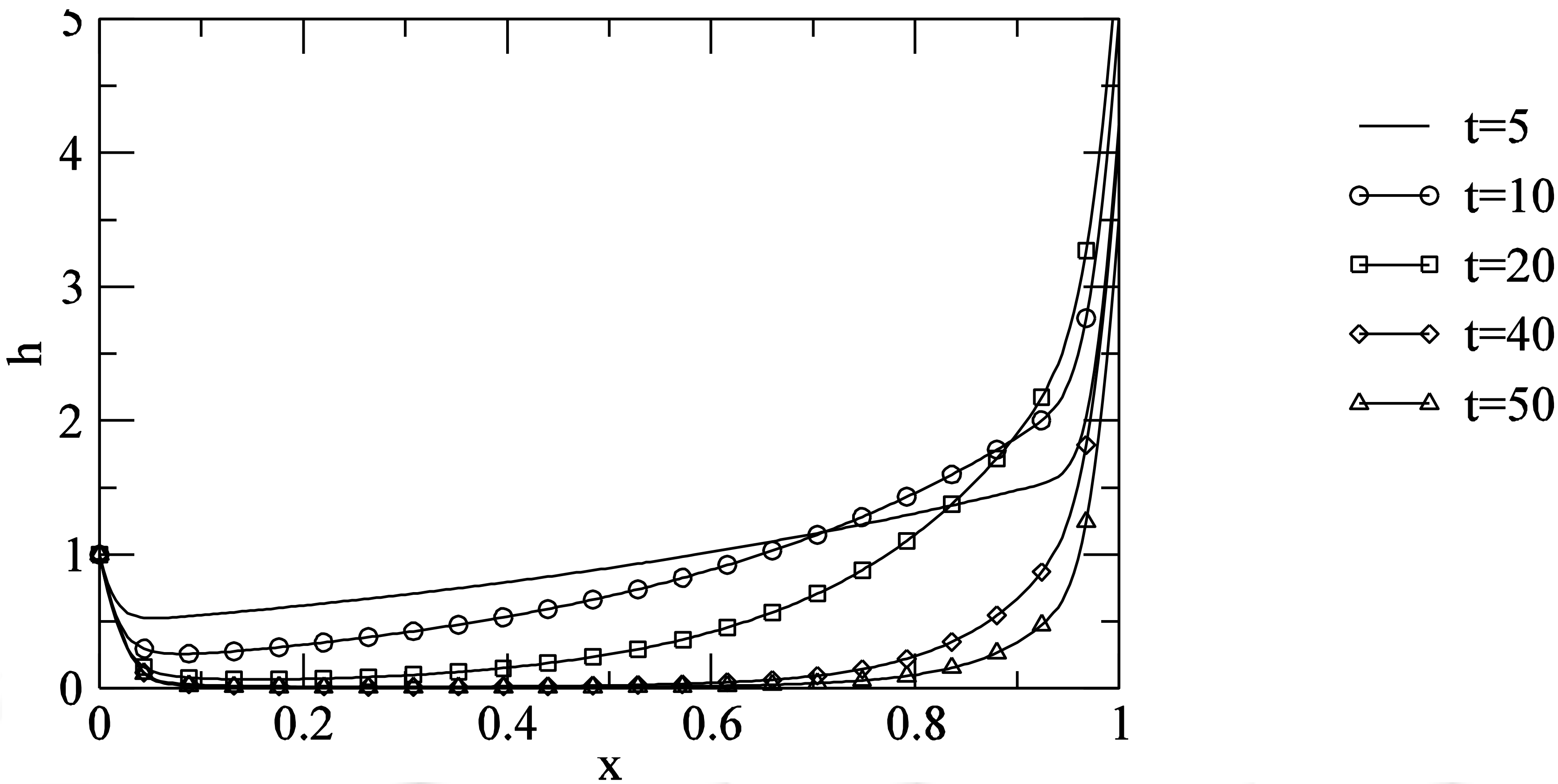
- **Trouton model**

$$
\begin{aligned}
\frac{\partial h}{\partial t} + \frac{\partial (hu)}{\partial x} &= 0, \\
\rho h \left(\frac{\partial u}{\partial t} + u u_{,x} \right) &= 4\mu \frac{\partial (h u_{,x})}{\partial x} + \frac{\gamma h h_{,xxx}}{2} + \delta\gamma \frac{d f_\gamma}{d\chi} h_{,x} + \rho g h,
\end{aligned}
$$

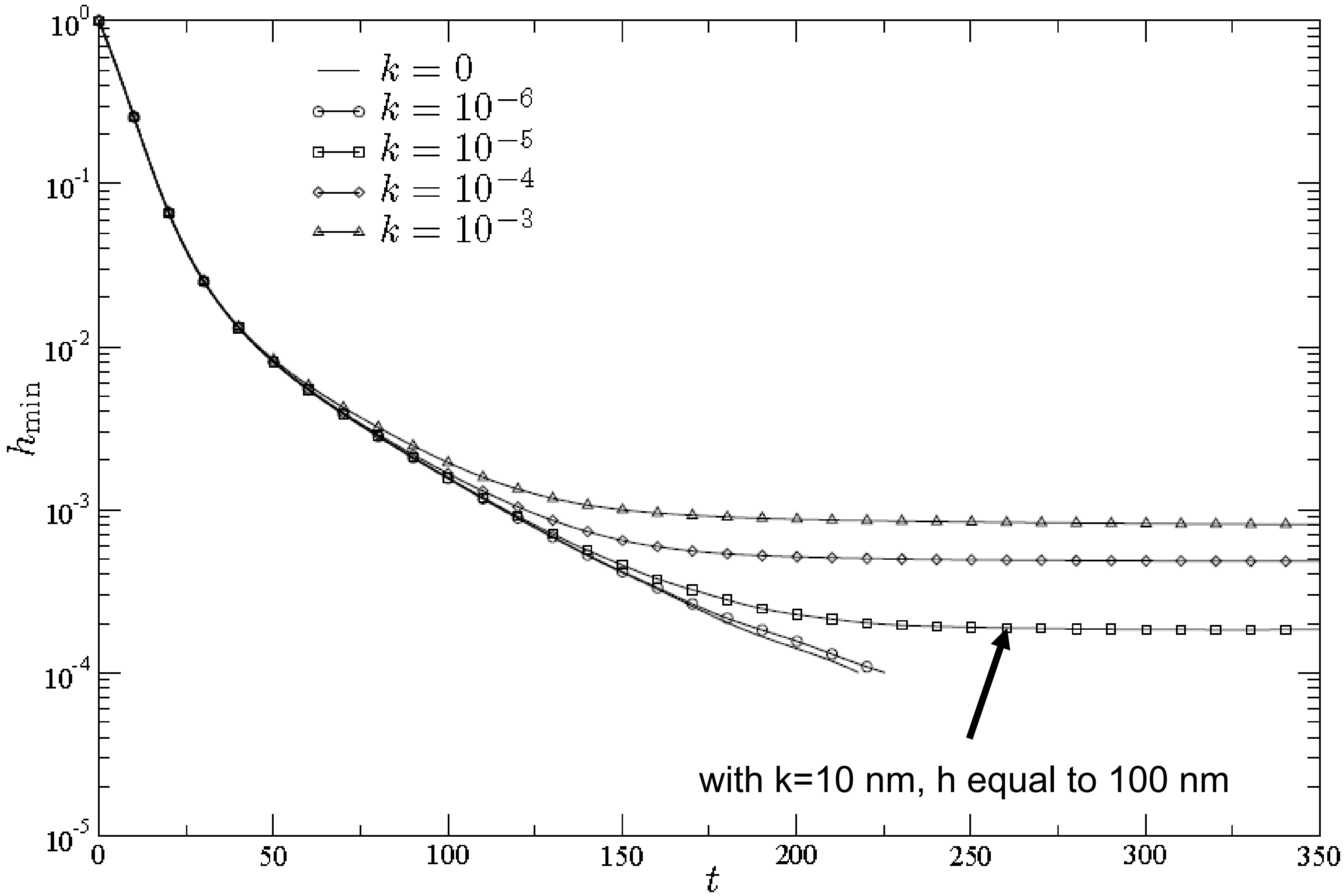
- **Solved numerically with a finite difference method**

Stability of vertical film

Lubrication model



Stability of vertical film
Numerical results



Conclusion

- **Drainage of bubble:**
  - **Exponential decrease of the thin film:**
    - Mobile interfaces.
  - **Bubble size changes:**
    - Thinning rate;
    - Shape.
- **Lifetime of bubble:**
  - **Occurrence of chemical processes;**
  - **Strong effect of**
    - Glass nature;
    - Temperature.
  - **Marangoni stabilization**

Acknowledgments

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