Contact and friction of thin hydogels films: the role of poroelasticity

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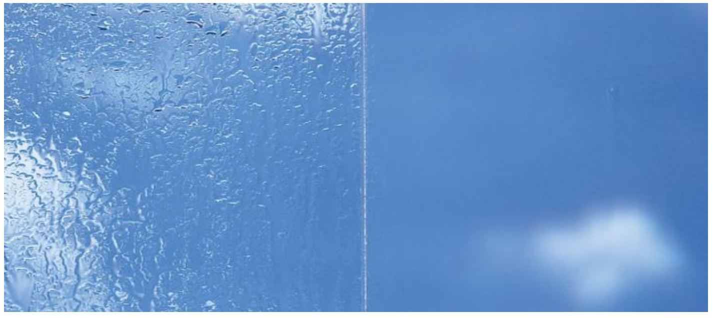
Motivations

Functionalization of glass substrates by hydrophilic coatings

Ex: prevention of mist formation



→ Tribological performance : friction, scratch resistance ?

Thin hydrogels layers mechanically confined within contacts between rigid substrates

⬇

Stress amplification as compared to bulk hydrogel substrates

⬇

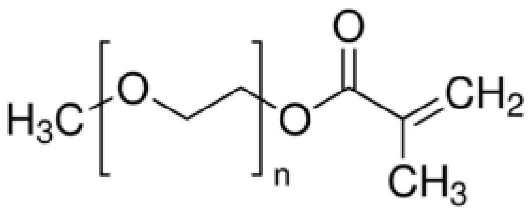
Enhanced drainage of the highly swollen gel network

⬇

Role of poroelasticity on mechanical and frictional properties ?

Model gel networks

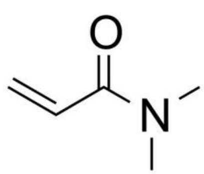
- **Poly(PEGMA)**



poly(ethylene glycol) methyl ether methacrylate
PEGMA (n = 4/5)

Rubbery

- **Poly(DMA)**

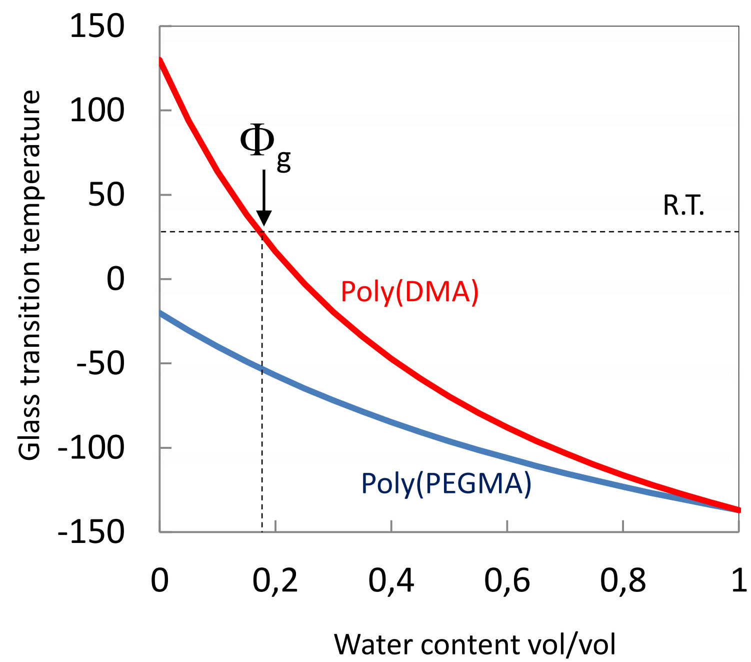


N,N-dimethylacrylamide

Rubbery

$\Phi_g \approx$ 0,18 vol/vol

Glassy

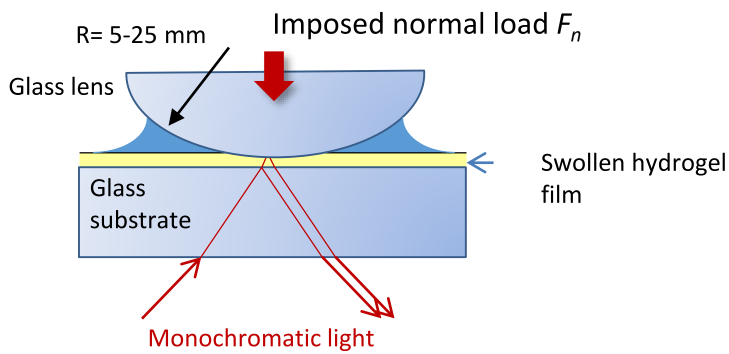


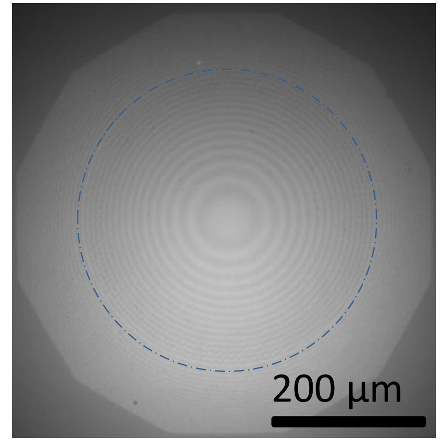
Tiol-ene chemistry route → homogeneous thin films:

Li et al, Langmuir, 2015
Chollet et al, ACS Appl. Mat. Interfaces , 2016

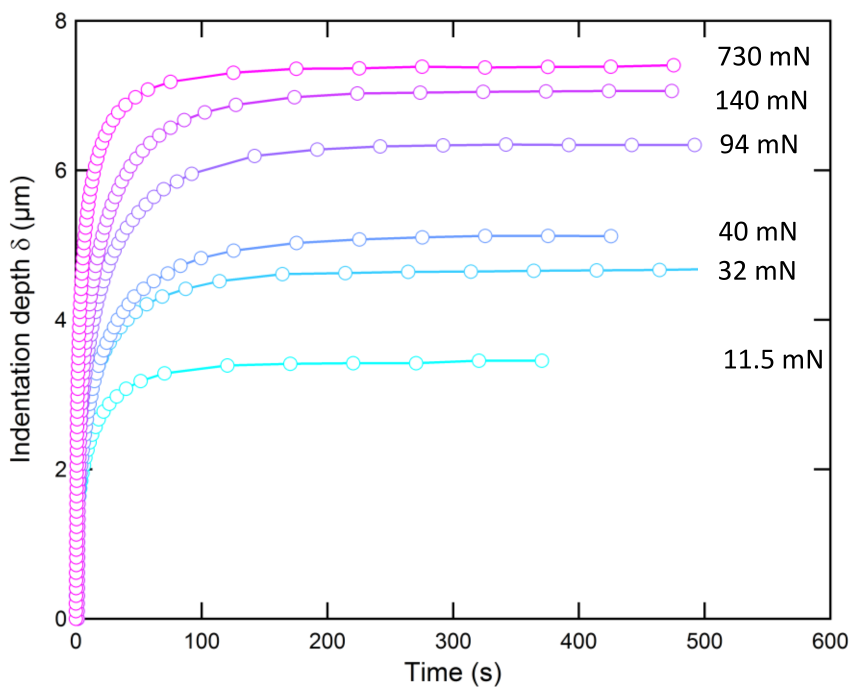
- ✓ grafted to glass or silicon wafer substrates
- ✓ controlled thickness from 250 nm (± 5%) to 2 µm (± 10%)
- ✓ controlled cross-linking (swelling ratio from 2.5 to 4)

Normal indentation response





Newton rings and contact vizualisation

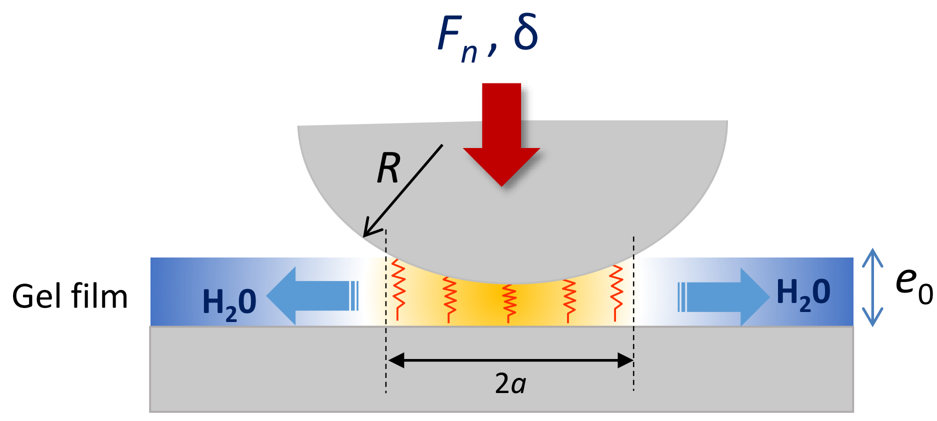


Time-dependent indentation depth

Poly(PEGMA) e_0=9 µm R=5.2 mm

Approximate poroelastic contact model

Within the limits of confined contact geometries $a >> e_0$



- No expansion of film deformation outside the contact
- Compression of the layer within the contact without lateral expansion

  → only vertical displacement components are accounted for

- Rigid substrates

Compressive strain $$\epsilon(r,t) = \frac{e_0 - e(t))}{e_0} = \frac{\delta(t) - r^2/2R}{e_0}$$

Indentation kinetics

Mixture theory developed by Biot (1955)

- Normal contact stress: $\sigma(r,t) = \tilde{E}\epsilon(r,t) + p$

Elasticity of the polymer network — Pore pressure

$$\tilde{E} = \frac{2G(1-\nu)}{1-2\nu}$$ Uniaxial compression modulus

- Water transport driven by Darcy's law: $J_r = -\kappa \frac{dp}{dr}$ $\kappa = \frac{D_p}{\eta}$

- Volume conservation

$$\frac{t}{\tau} = -\frac{\delta}{\delta_\infty} + \frac{1}{2} log\left(\frac{1+\delta/\delta_\infty}{1-\delta/\delta_\infty}\right)$$

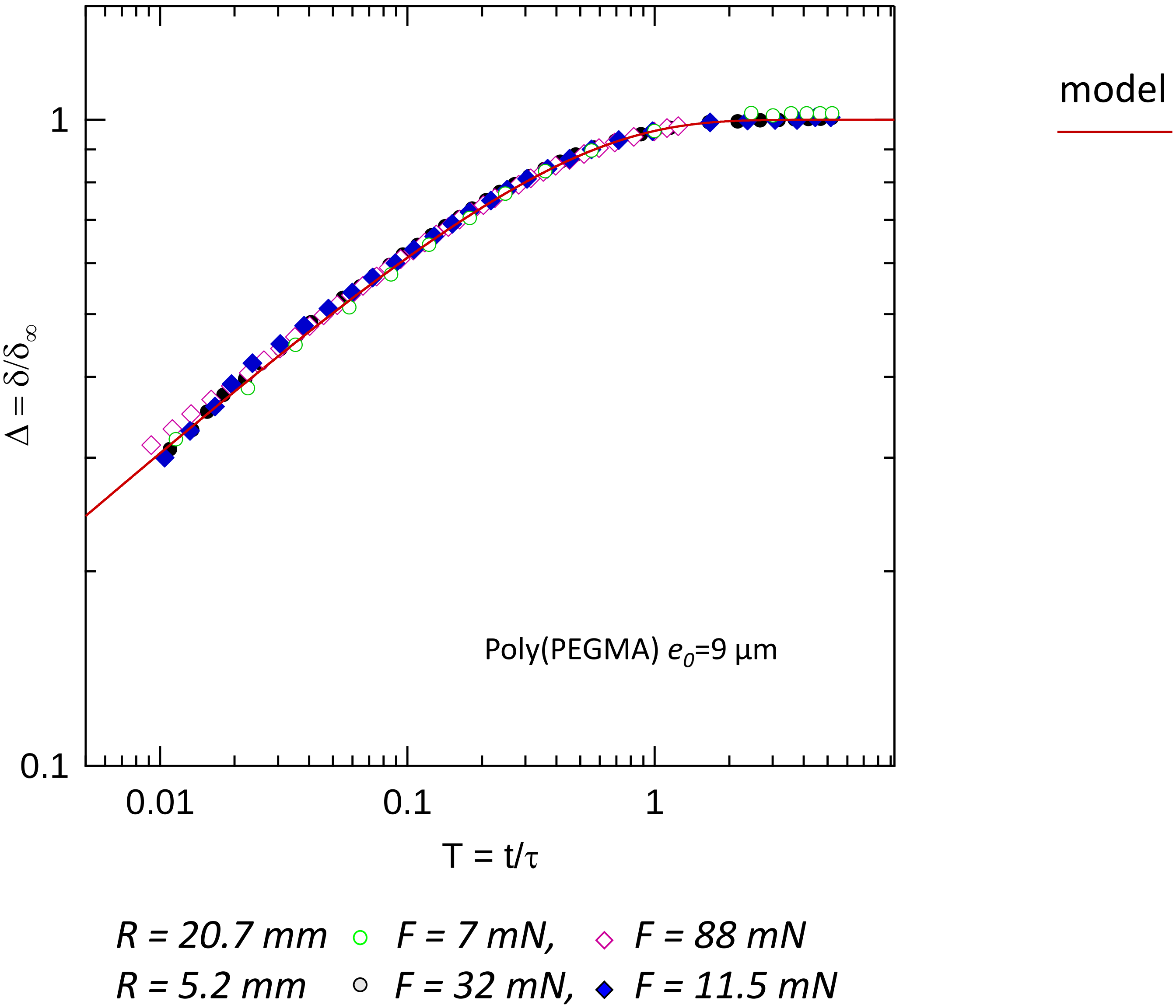
$$\delta_\infty = \left[\frac{F_n e_0}{\pi R \tilde{E}}\right]^{1/2}$$

Equlibrium indentation depth

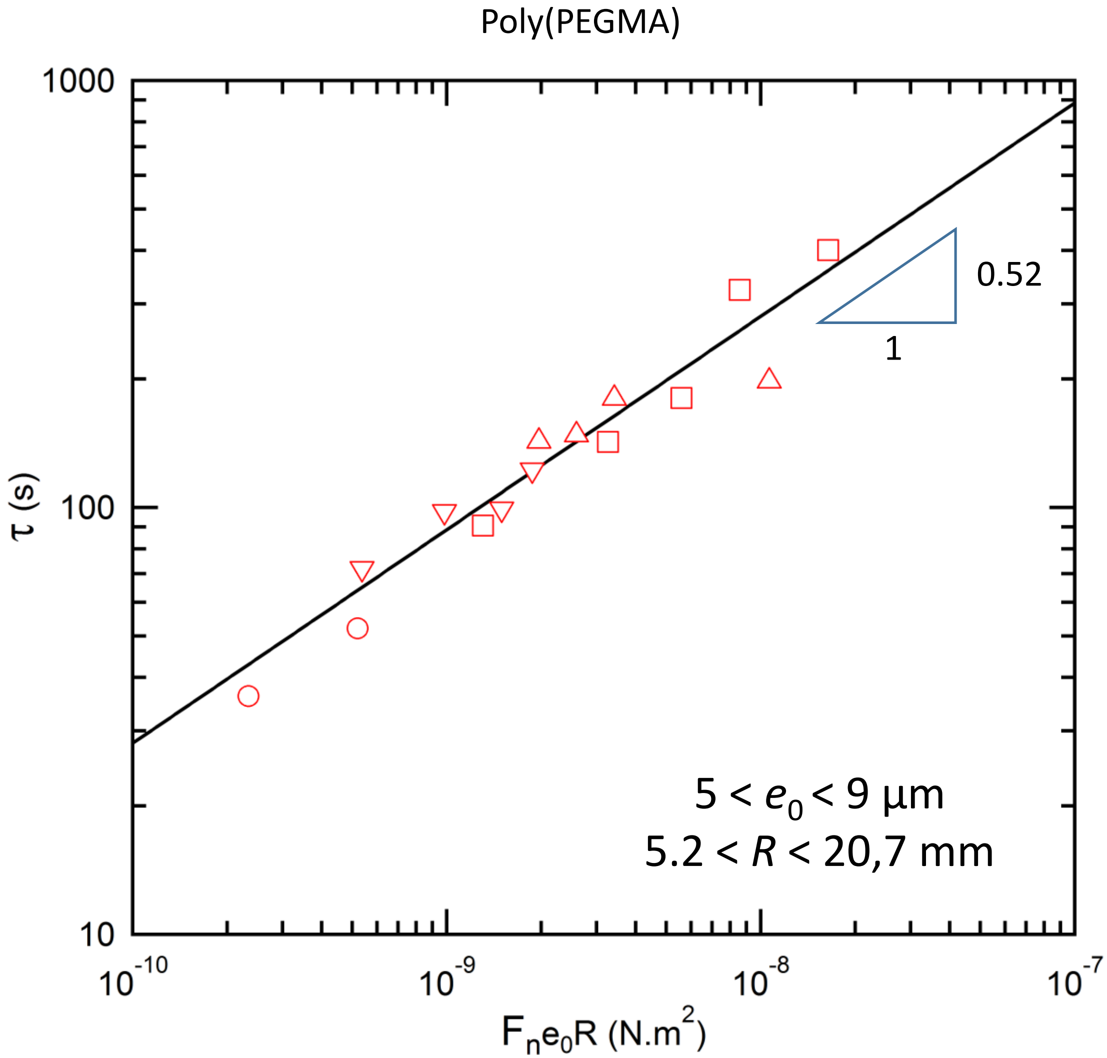
$$\tau = \frac{1}{2\sqrt{\pi}} \frac{\eta}{D_p} \left(\frac{F e_0 R}{\tilde{E}^3}\right)^{1/2}$$

Poroelastic time

Fit of experimental data to the poroelastic contact model



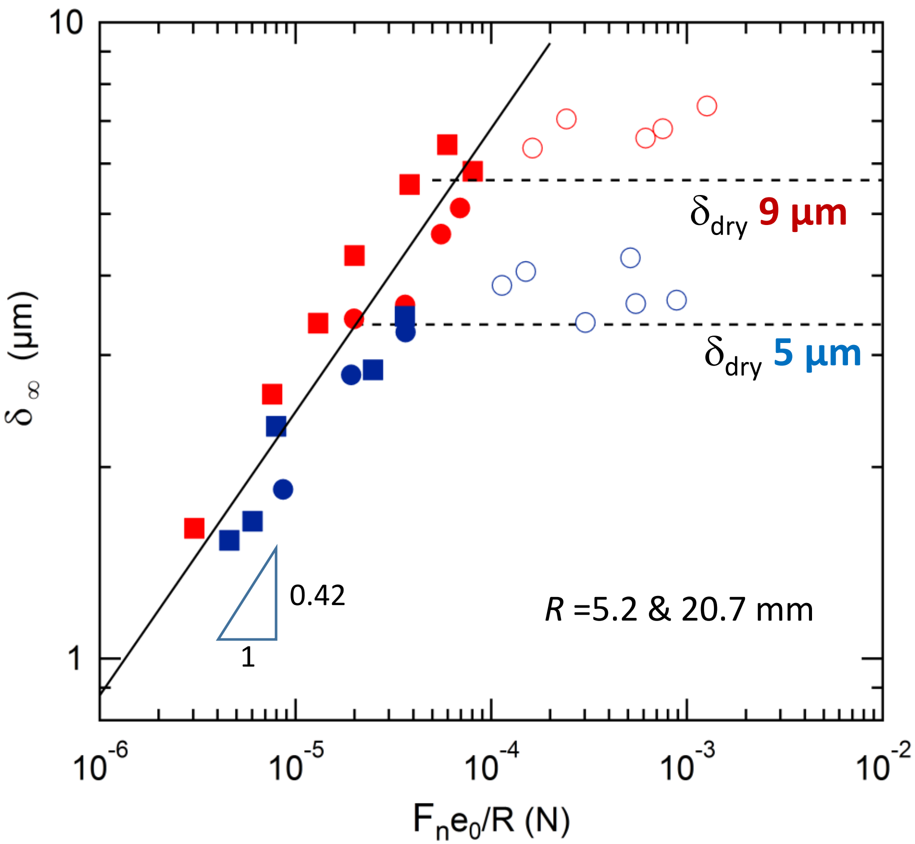
Characteristic poroelastic time τ



✔ $\tau \propto (F_n e_0 R)^{1/2}$

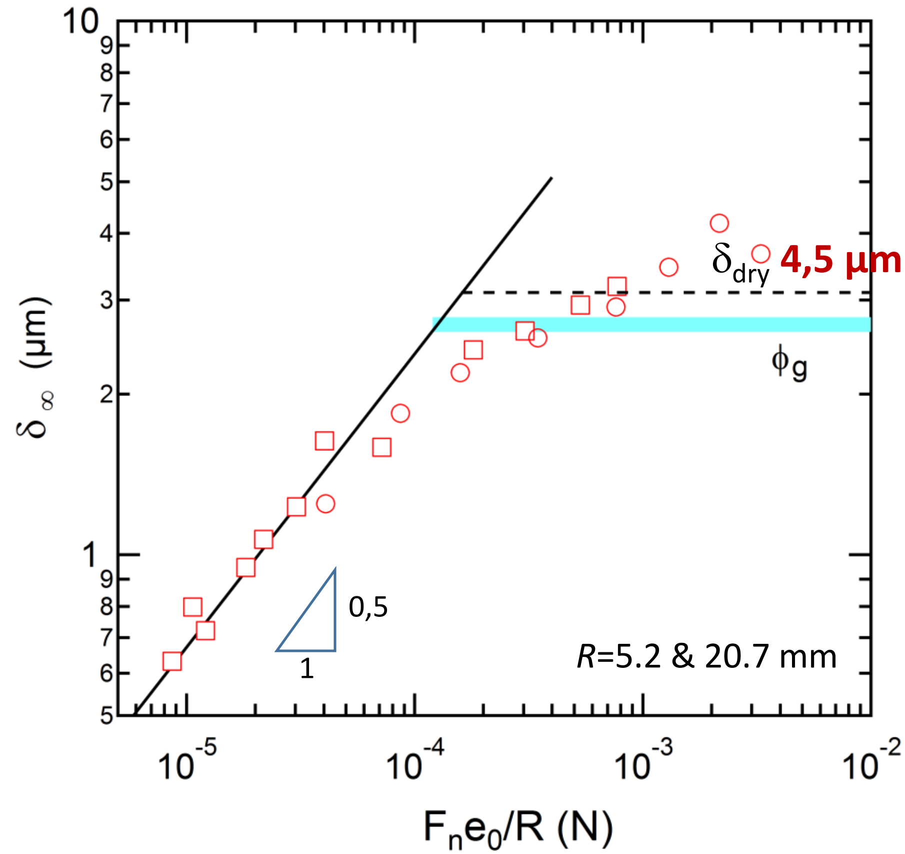
Equilibrium indentation depth

Poly (PEGMA)



✓ $$\delta_\infty \propto \left(\frac{F_n e_0}{R}\right)^{1/2}$$

Poly (DMA)

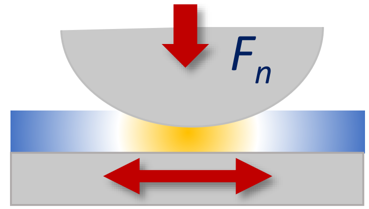


✗ Deviation from the model

Close to ϕ_g

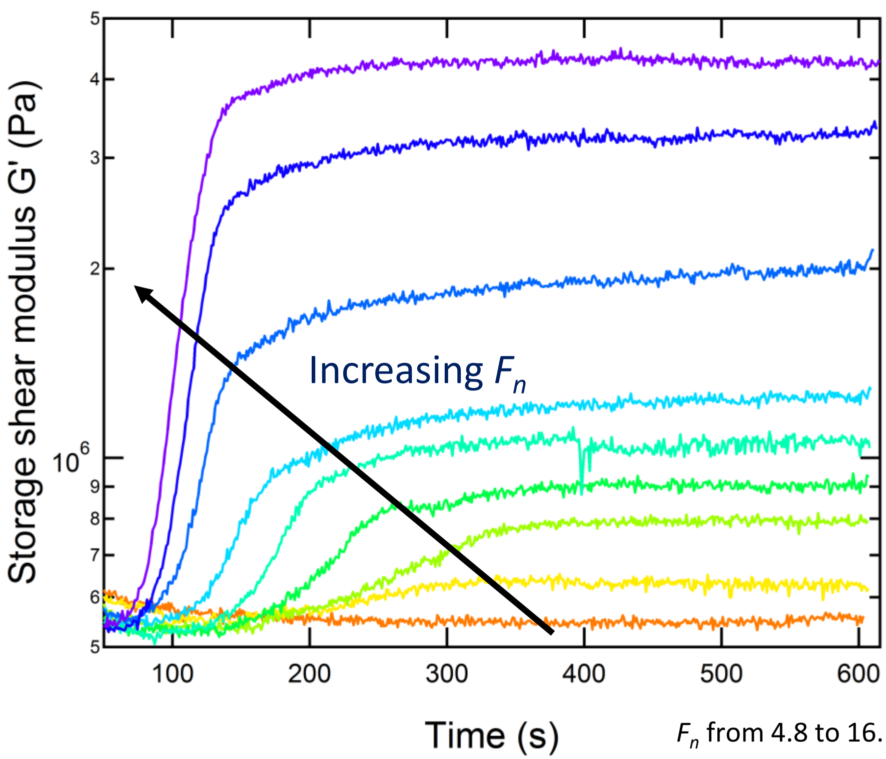
Glass transition induced by poroelastic drainage

- Lateral contact experiments → shear modulus measurements during the course of indentation drainage



$$\Delta = \Delta_0 sin(\omega t) \qquad \Delta_0 \leq \pm 100nm$$

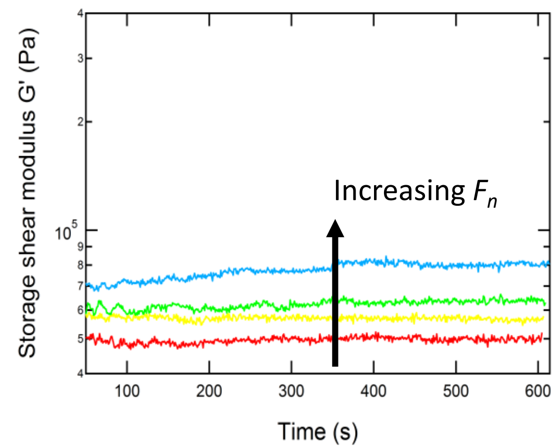
Poly(PDMA)



F_n from 4.8 to 16.1 N

Glass transition

Poly(PEGMA)

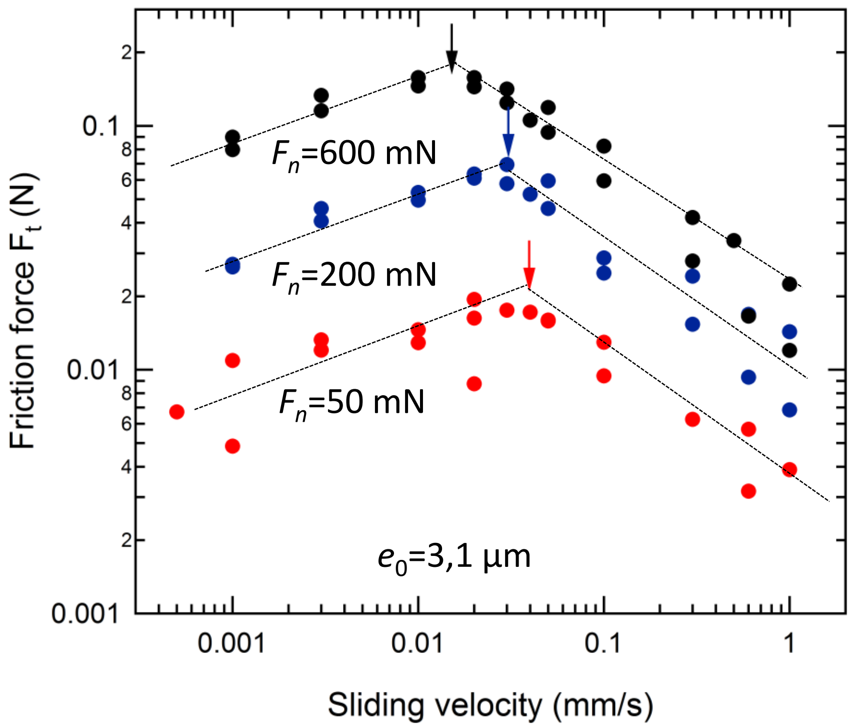


F_n from 4.5 to 13.7 N

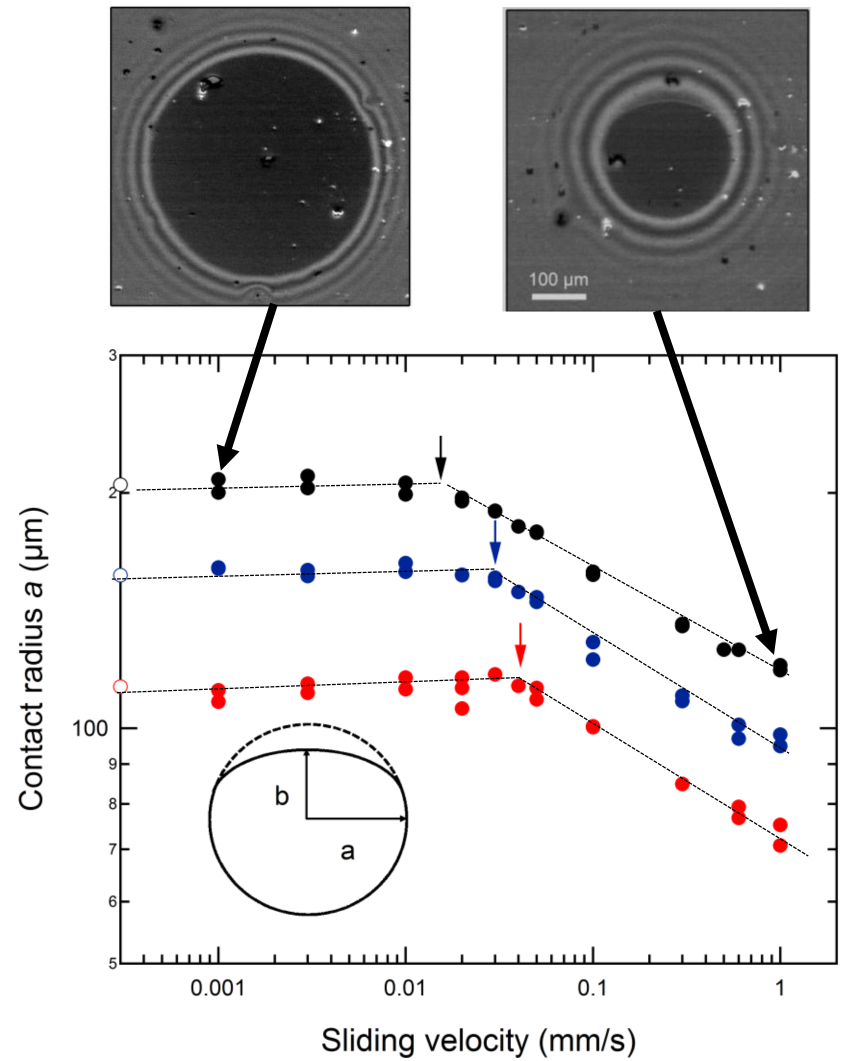
No transition

Steady-state friction : poly(DMA) with $\phi > \phi_g$

Friction force



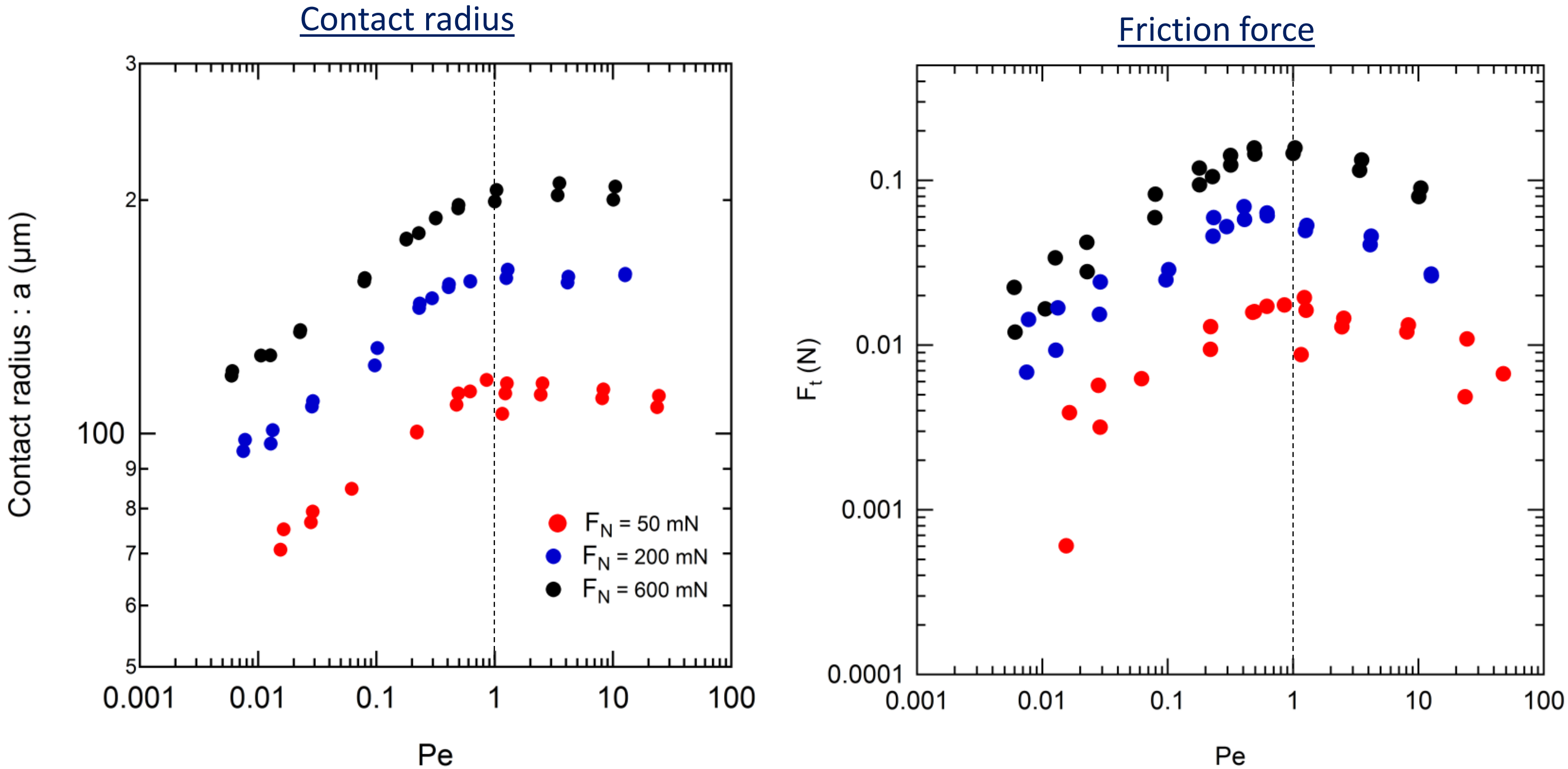
Contact shape



Velocity-dependence: two regimes → poroelastic effect ?

Contribution of poroelasticity : Peclet number

Peclet Number $$Pe = \frac{2a}{v\tau} = \frac{\text{Contact time}}{\text{Poroelastic time}}$$



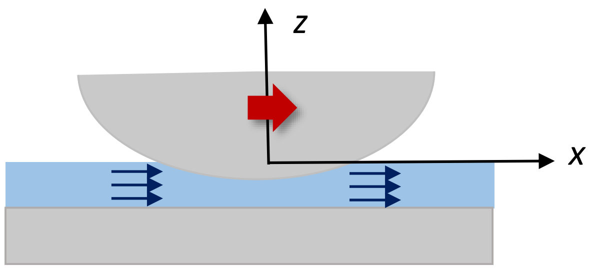
τ determined independently from indentation experiments

Pe > 1 : drainage equilibrium ~ normal indentation

Pe < 1 : out-of equilibrium state → incomplete drainage

Pore pressure distribution

- Extension of the poroelastic contact model to steady-state sliding



Moving coordinate system

$$\left.\frac{\partial}{\partial t}\right|_{X,y} \rightarrow -v\frac{\partial}{\partial x}$$

Pore pressure field induced during lateral motion

$$-v\frac{\partial \epsilon}{dx} = -\kappa \nabla^2 p$$

Darcy's Law

General solution for pore pressure:

$$p = -\frac{v}{8Re_0\kappa} r^3 \cos\theta + \sum_{n=0}^{\infty} a_n r^n \cos n\theta$$

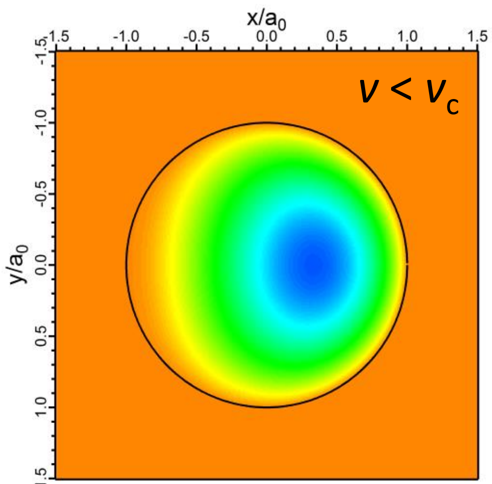
With ***p*=0** on the contact line to be deterrmined

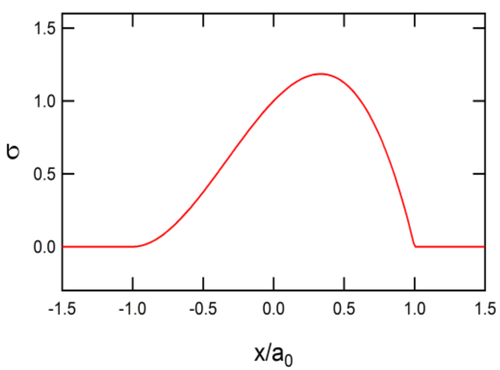
Contact stress: velocity dependence

Hyp: we enforce that contact line is a circle with $p(a_0)=0$

$$\sigma(r,t) = \tilde{E}\epsilon(r,t) + p$$

$$\sigma\left(r,\theta\right) = \frac{\tilde{E}}{2Re_0}\left(a_0^2 - r^2\right)\left[1 + \frac{v}{4\tilde{E}\kappa} r cos\theta\right]$$

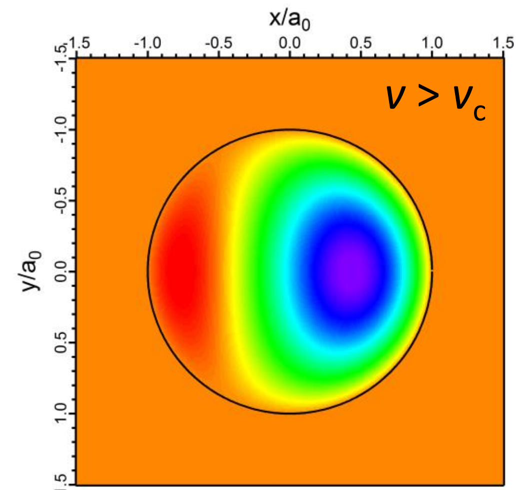


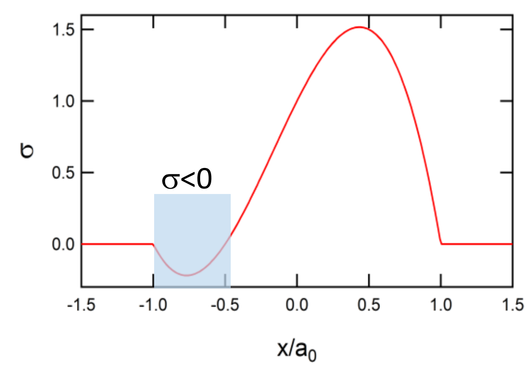


Critical velocity v_c

$$v_c = \frac{4\tilde{E}\kappa}{a_0}$$

$$Pe = 1$$





Above v_c : negative contact stress → contact line shrinks non-uniformly
Reduction in contact size → build-up of pore pressure at increasing velocities

Contact size reduction for Pe < 1

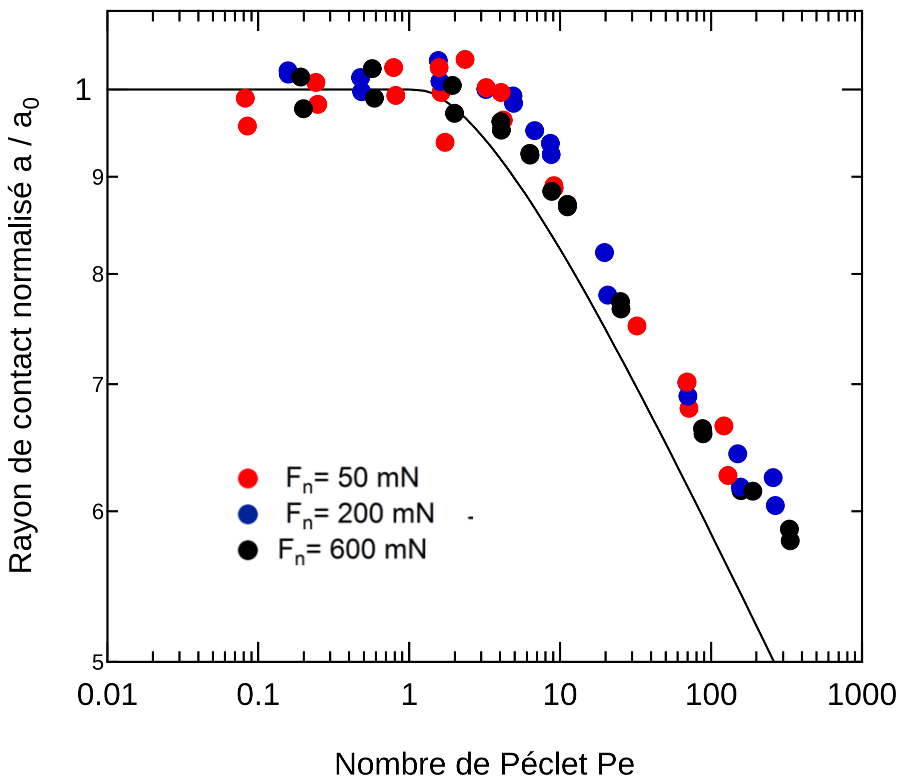
- Numerical solution of the poroelastic contact model

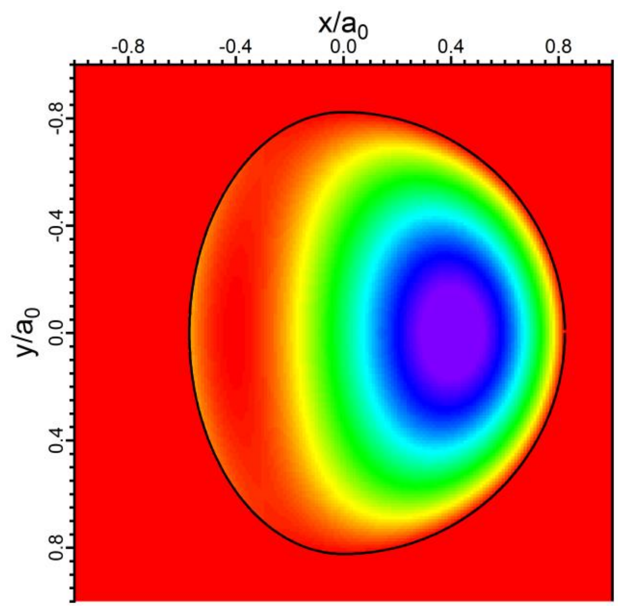
$$\sigma(r,\theta) > 0$$

Contact line a(θ,*v*) ensuring:

$$p(r = a(\theta, v)) = 0$$

$$\int_A \sigma(r,\theta) = F_n$$





Calculated contact shape
and normal contact stress

Pe=0.1

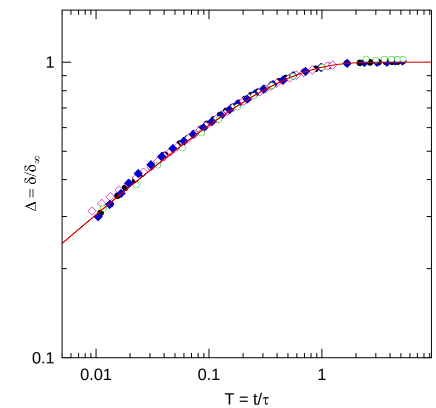
Conclusions & perspectives

- Indentation of thin hydrogels films confined within glass substrates

    ✓ Approximate poroelastic contact model

    → Scaling laws for δ_∞ and τ as a function of - gel mechanical and diffusive properties
    - contact geometry & loading conditions

    ✓ Glass transition induced by drainage

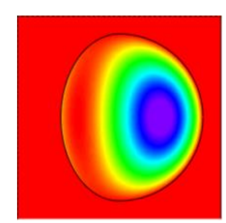


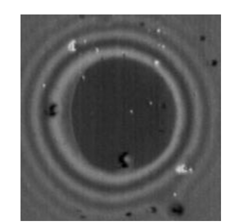
- Frictional properties driven by poroelastic time

    ✓ Two frictional regimes - *Pe* > 1 equilibrium drainage state

    -*Pe* < 1 pressure imbalance resulting in contact asymmetry and size reduction

    → Contact changes accounted for by poroelasticity





Contribution to friction force of viscous dissipation associated to poroelastic flow?

Friction force across glass transition?