

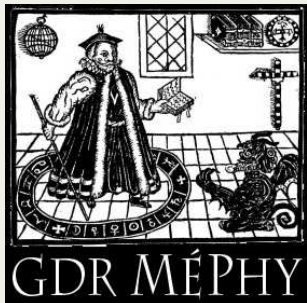
FRICITION OF POLYMER GELS

O. Ronsin, T. Baumberger, C. Caroli

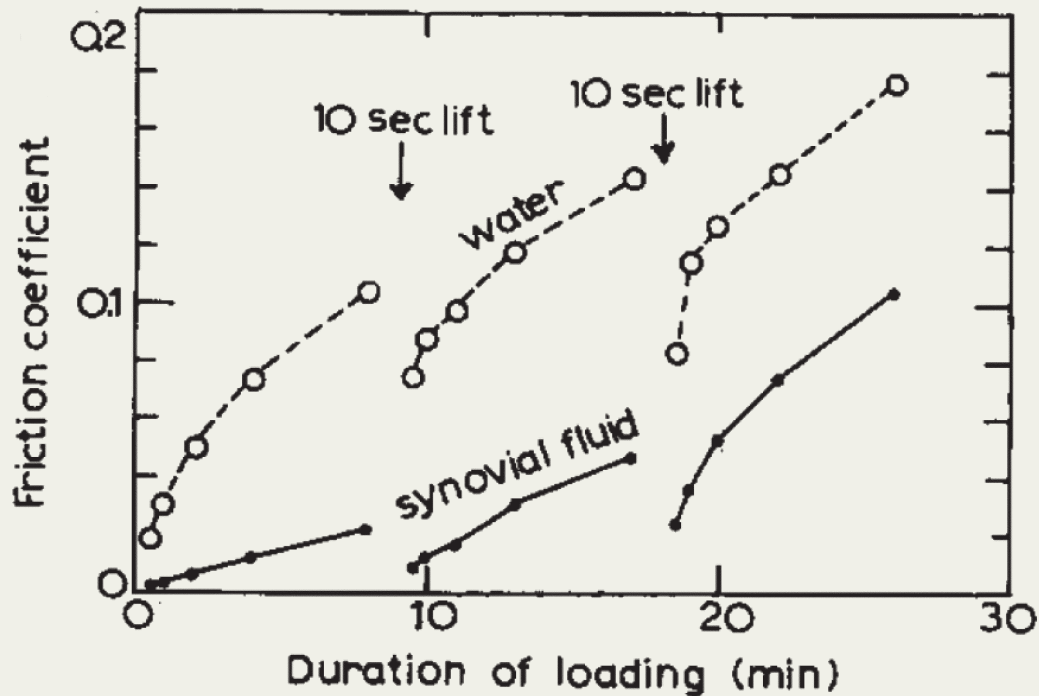
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Gels = soft elastic solid with high content of fluid = poroelastic solid
⇒ specific frictionnal properties

- e.g. : gels in biological systems : very low friction (e.g. articular cartilage)
- Network / solvent contributions to friction ?



POROELASTIC EFFECTS : ARTICULAR CARTILAGE (COLLAGEN GEL)



Poroelastic transfer of fluid across cartilage

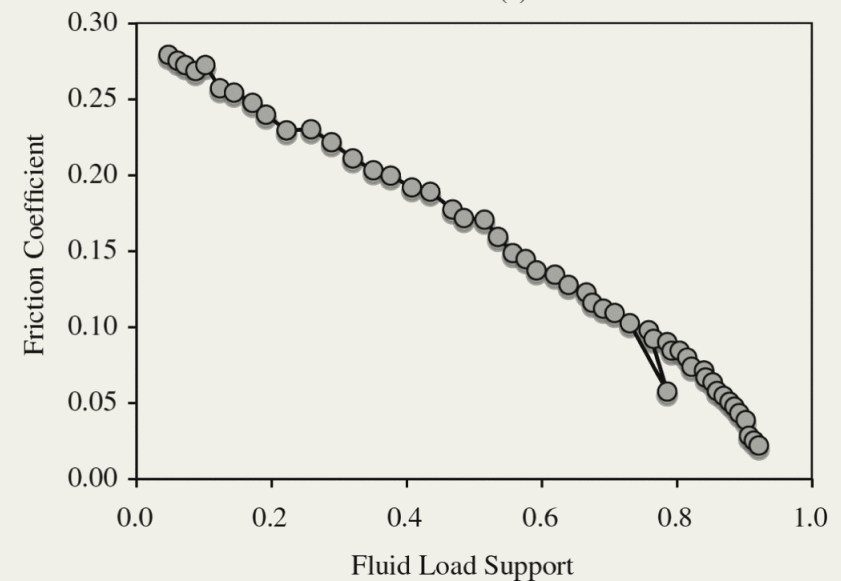
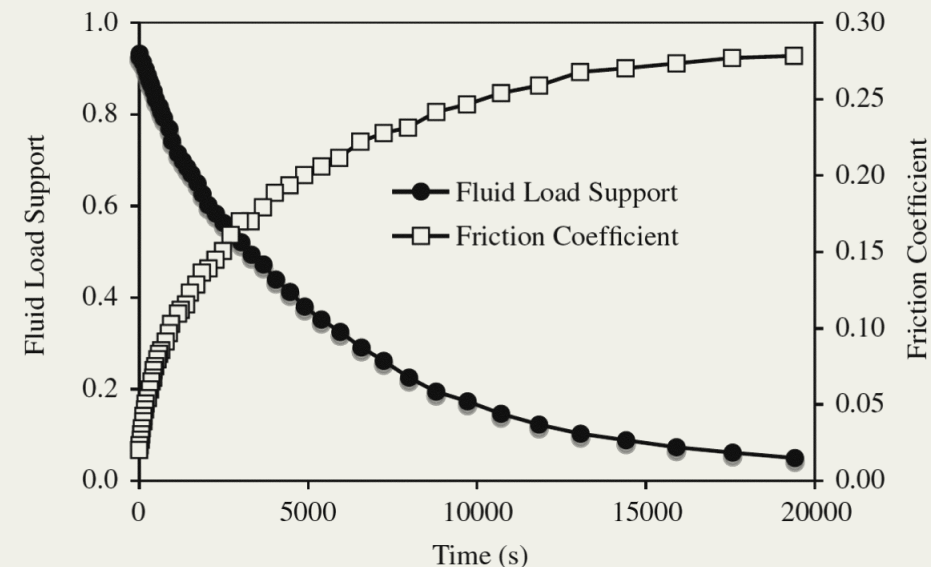
→ load supported by fluid at short times

low friction : **fluid lubrication**

→ increasing contribution of matrix as $t \nearrow$

higher friction : **solid friction**

$$\mu = \mu_{\infty} \left(1 - (1 - \phi) \frac{W_{\text{fluid}}}{W_{\text{total}}} \right)$$



C. W. McCutchen, *Wear* **5**, 1 (1959).

G. A. Ateshian, *Journal of Biomechanics* **42**, 1163 (2009).

POLYMER GELS

- network = chains + cross-links ★
(physical or chemical), distance ξ
- solvent (90–99%), viscosity η

⇒ "poro-elastic" material

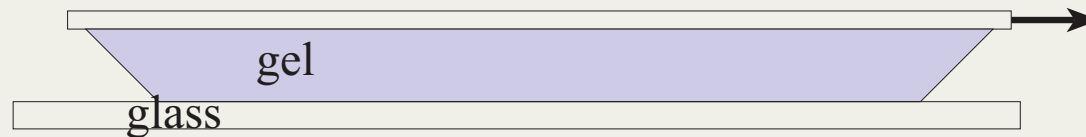
- elasticity of entropic origin (rubber)
 $G \simeq \frac{k_B T}{\xi^3} \simeq 10^2 - 10^5 \text{ Pa}$, $\xi \simeq 3 - 30 \text{ nm}$.
- solvent/network relative motion ⇒
diffusive mode
 $D_{\text{coll.}} \simeq 10^{-9} - 10^{-11} \text{ m}^2 \cdot \text{s}^{-1}$
for aqueous gels ($10^{-7} \text{ m}^2 \cdot \text{s}^{-1}$ for
cartilage)
⇒ $10^5 - 10^7 \text{ s}$ for 1 cm thick sample
⇒ get rid of time dependence of fluid
loaded part.



HYDRODYNAMIC FRICTION OF GELATIN GELS

Model system : gelatin gel on clean flat glass in plane/plane contact.

→ control almost independently solvent viscosity $\eta_S \simeq 1 - 4$ Pa.s (water + glycerol) and network mesh size $\xi \simeq 6 - 12$ nm (gelatin concentration c)



impose

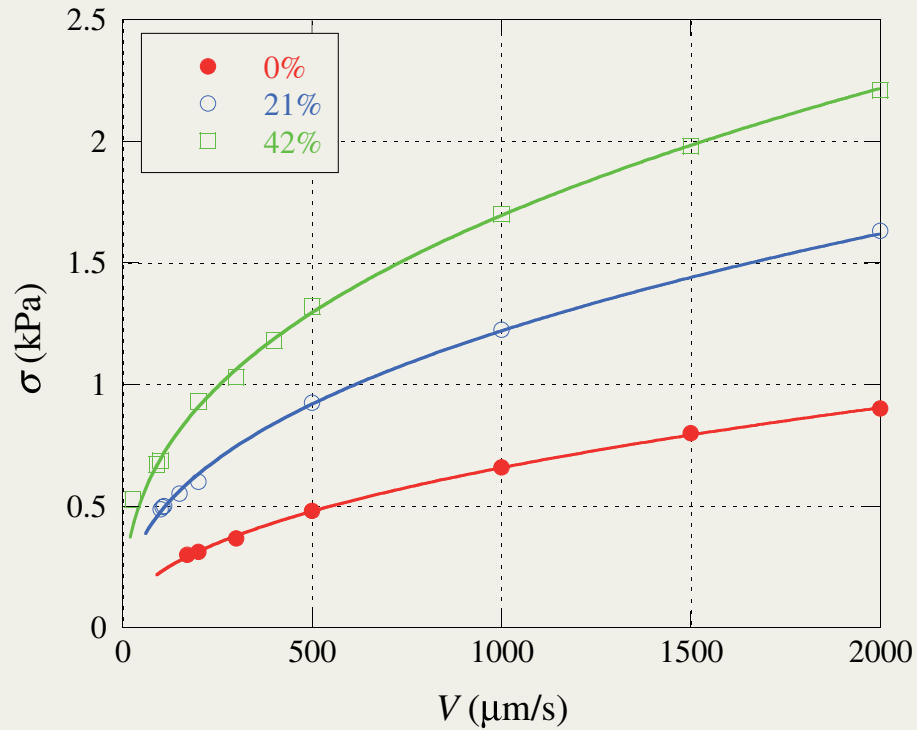
- sliding velocity V ($1 - 2000 \mu\text{m/s}$)
- normal pressure p ($0 - 2$ kPa)

measure

- stationary frictional shear stress $\sigma(V)$.
- interfacial slip.

HYDRODYNAMIC FRICTION OF GELATIN GELS

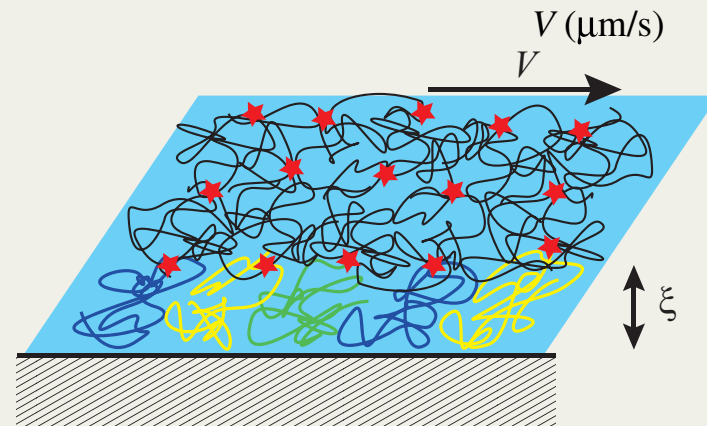
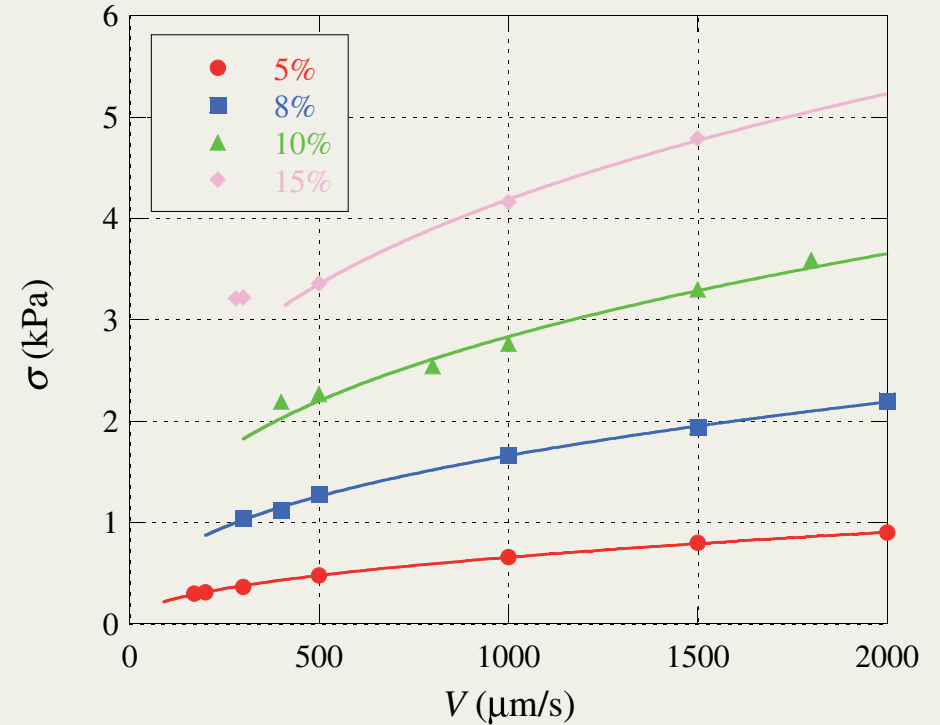
Changing solvent viscosity η_s



\Rightarrow Response of a confined layer of thickness $\simeq \xi$?

Changing gelatin concentration

$$c \nearrow \Rightarrow G \nearrow \Rightarrow \xi \searrow$$

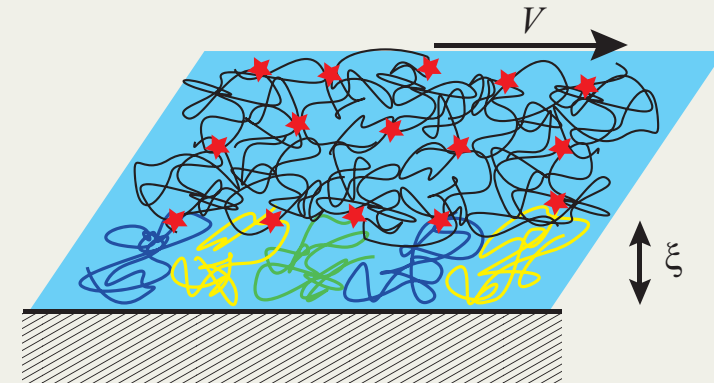


HYDRODYNAMIC FRICTION OF GELATIN GELS

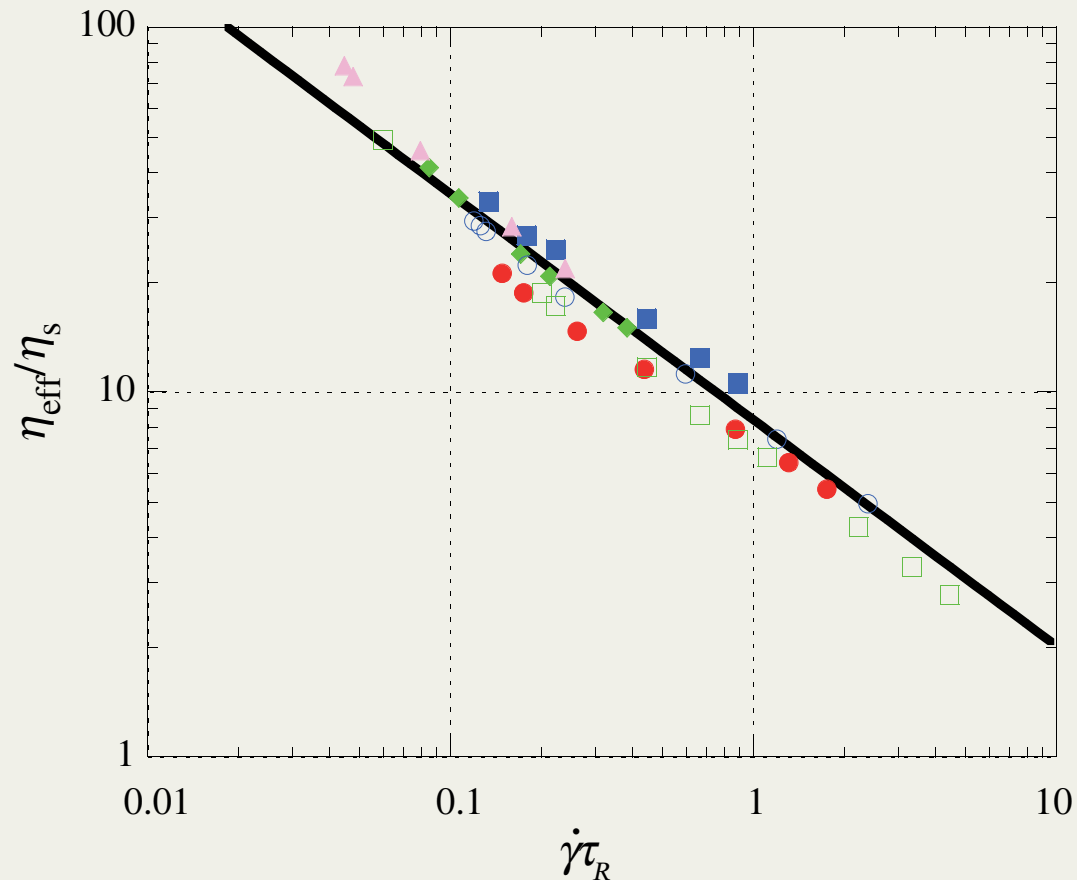
layer sheared at a rate $\dot{\gamma} = V/\xi$

measured effective viscosity $\eta_{\text{eff}} = \frac{\sigma \xi}{V}$

relaxation time τ_R of a chain of size ξ (Rouse)



$\frac{\eta_{\text{eff}}}{\eta_s} \sim (\dot{\gamma} \tau_R)^{-\alpha}$, $\alpha = 0.6$ compatible
with macroscopic rheology of solutions.

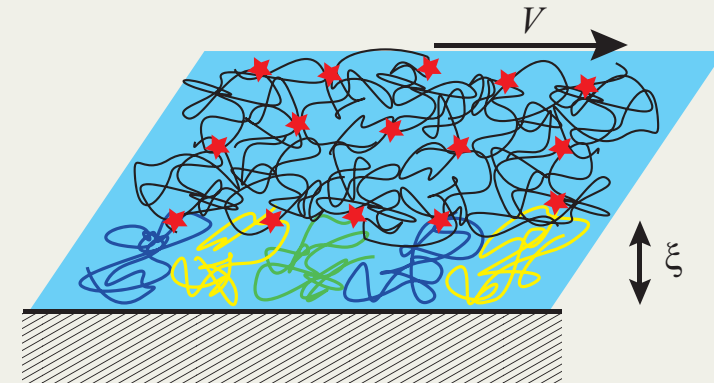


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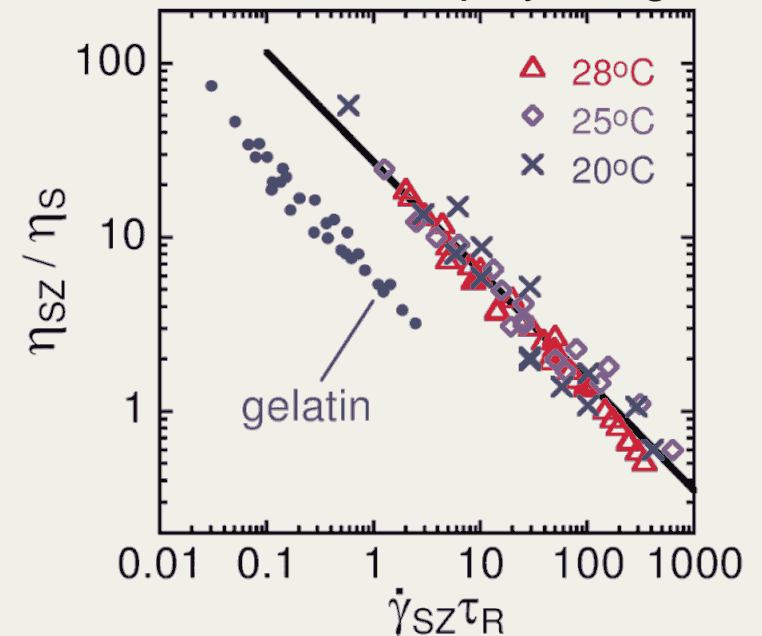
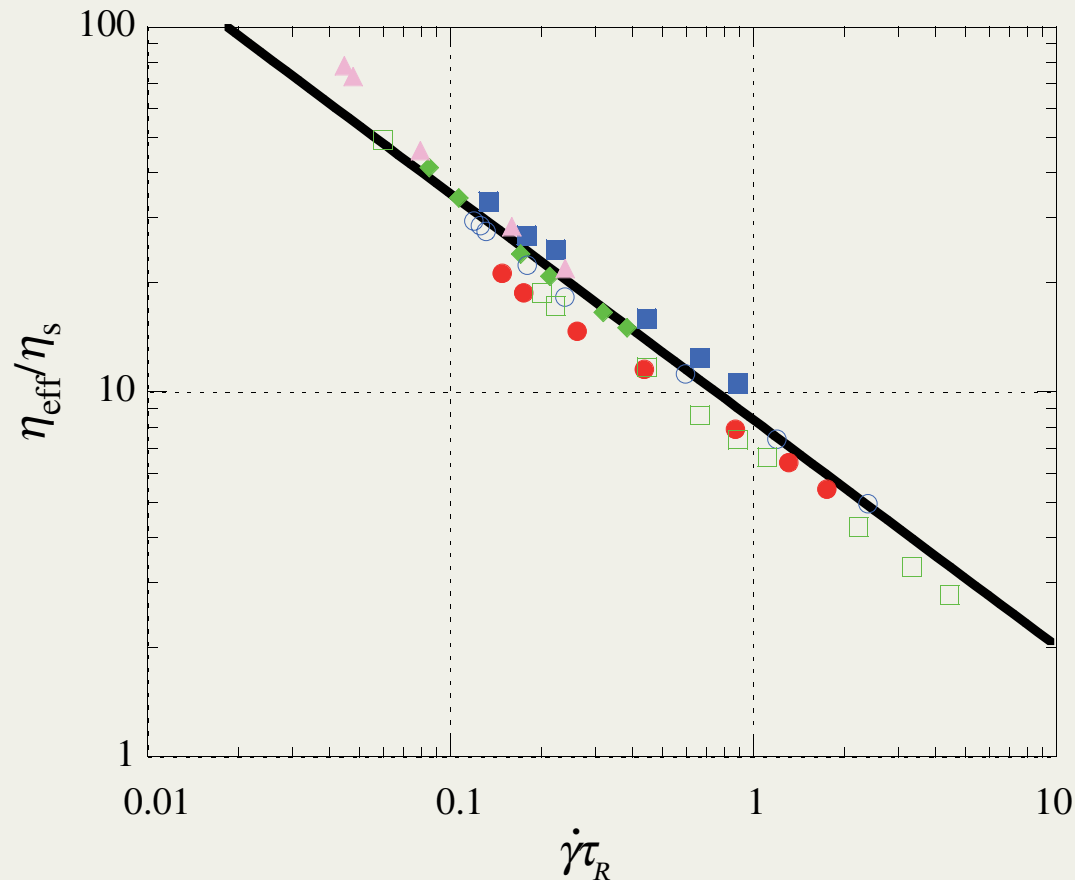
relaxation time τ_R of a chain of size ξ (Rouse)



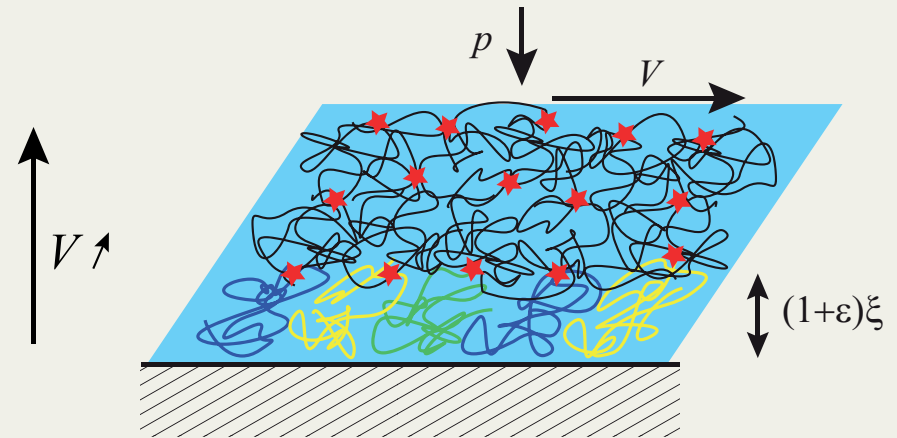
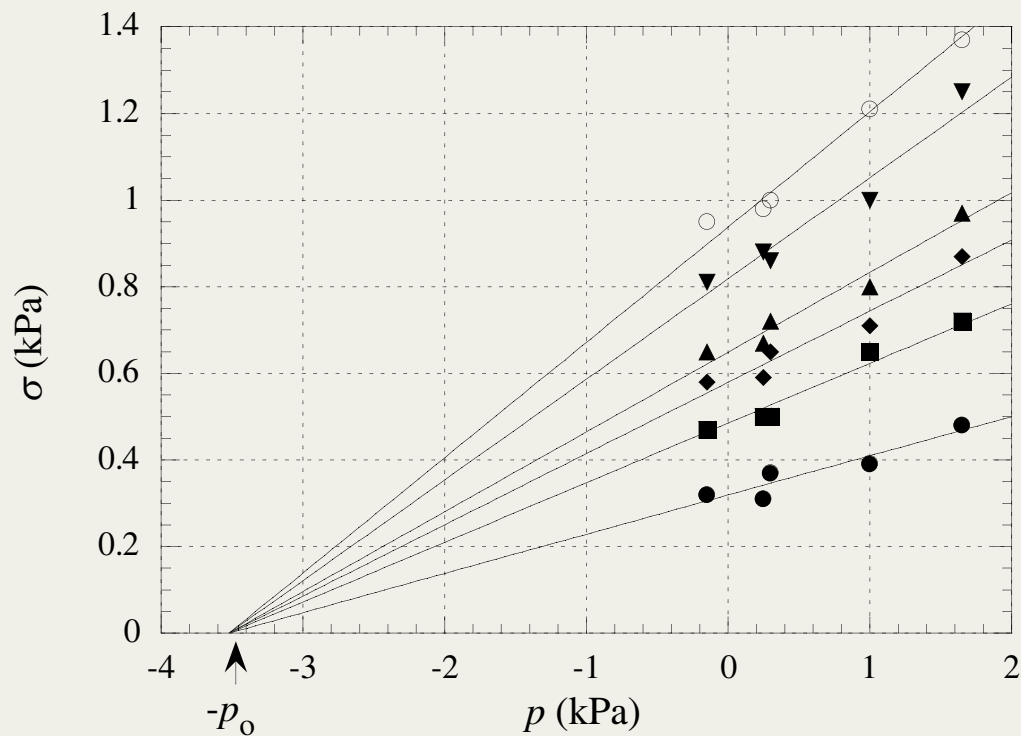
$$\frac{\eta_{\text{eff}}}{\eta_s} \sim (\dot{\gamma} \tau_R)^{-\alpha}, \quad \alpha = 0.6 \quad \text{compatible}$$

with macroscopic rheology of solutions.

Also observed with block copolymer gels



EFFECT OF NORMAL STRESS



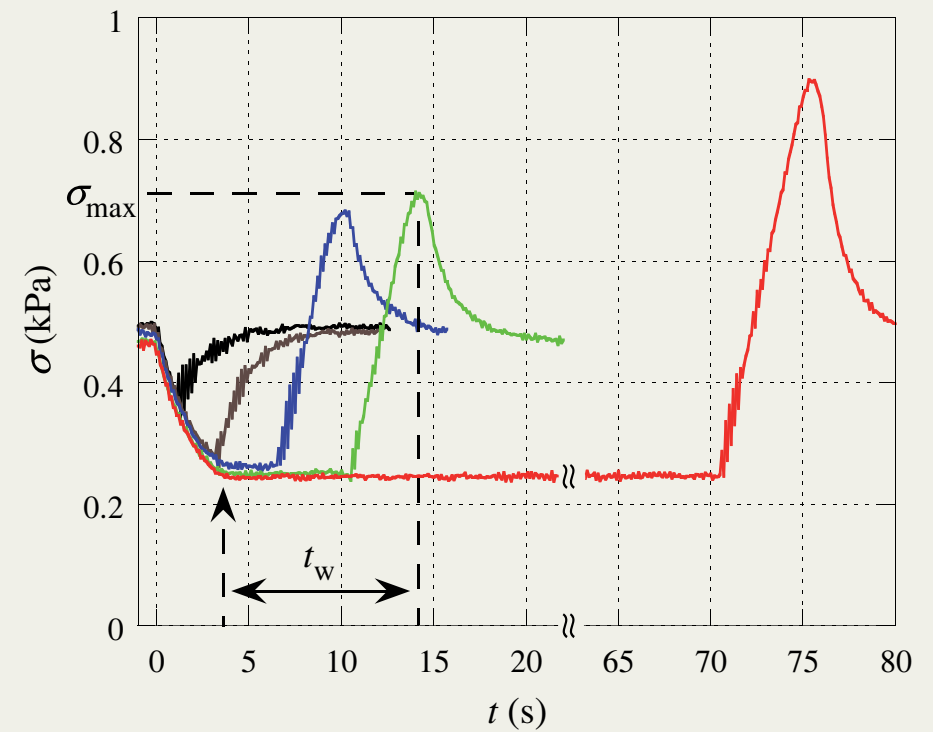
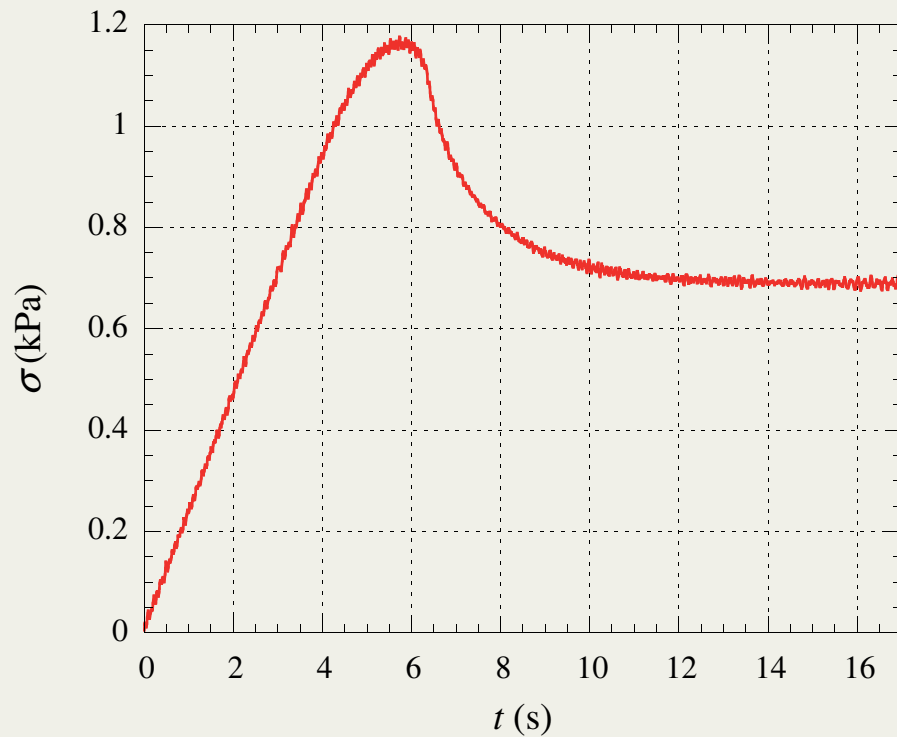
deformation under compression $\epsilon = -\frac{p}{E} = -\frac{p}{3G}$

\Rightarrow layer thickness $(1 + \epsilon)\xi$

$\Rightarrow \sigma(V, p) = \sigma(V, p = 0) \left[1 + (1 + 3\alpha) \frac{p}{3G} \right]$

$p_0 = \frac{3G}{1 + 3\alpha} \simeq G$

STATIC FRICTION

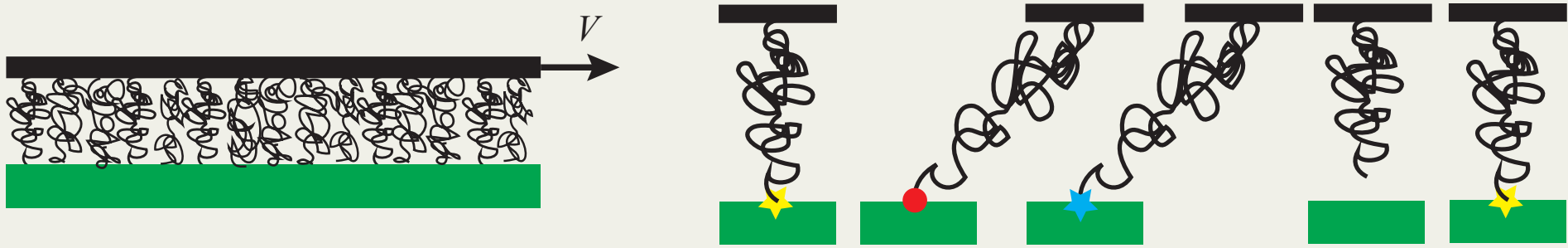


Increase of the number of pinned chains with time.

\Rightarrow Decrease of the number of pinned chains with velocity.

ELASTIC CONTRIBUTION TO GEL FRICTION

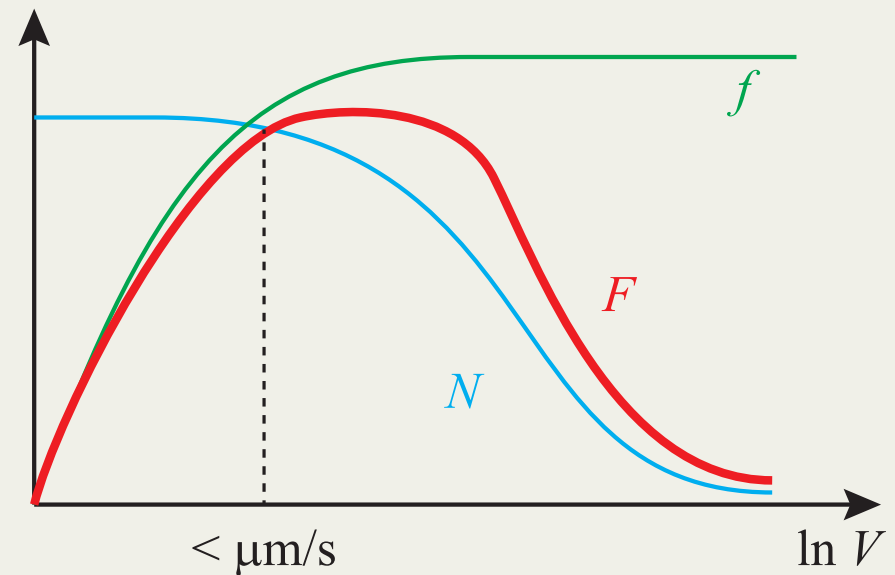
Schallamach model of rubber friction : dynamics of chain pinning/depinning



thermally activated depinning
+ advection at velocity V

\Rightarrow number of pinned chains \searrow with V

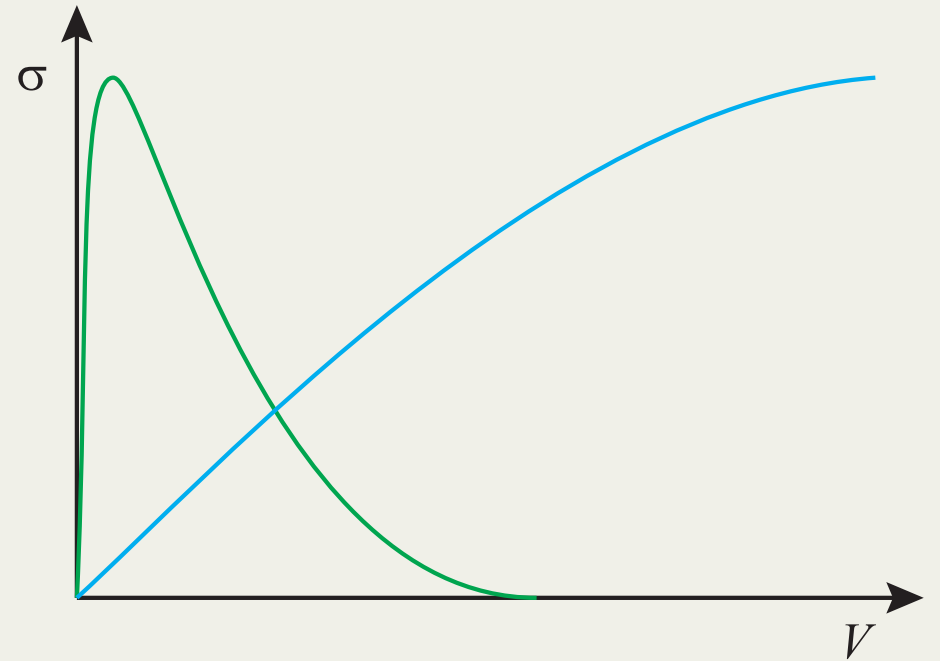
\Rightarrow force per chain \nearrow with V



POLYMER GELS FRICTION

two contributions:

- chain pinning
- viscosity of interfacial layer

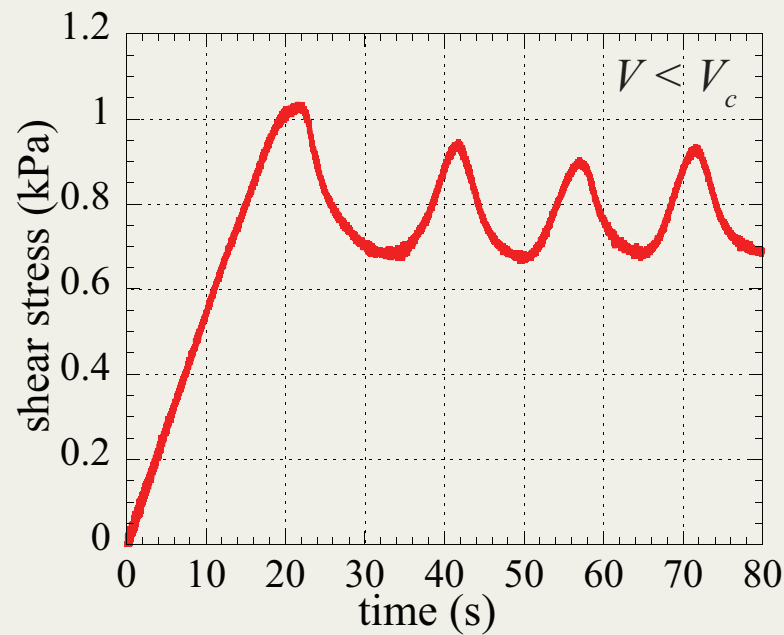
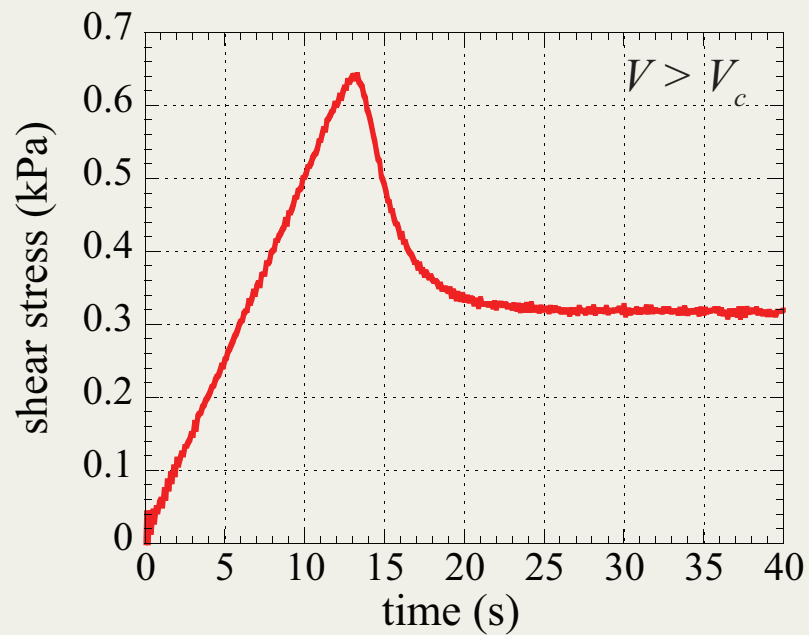
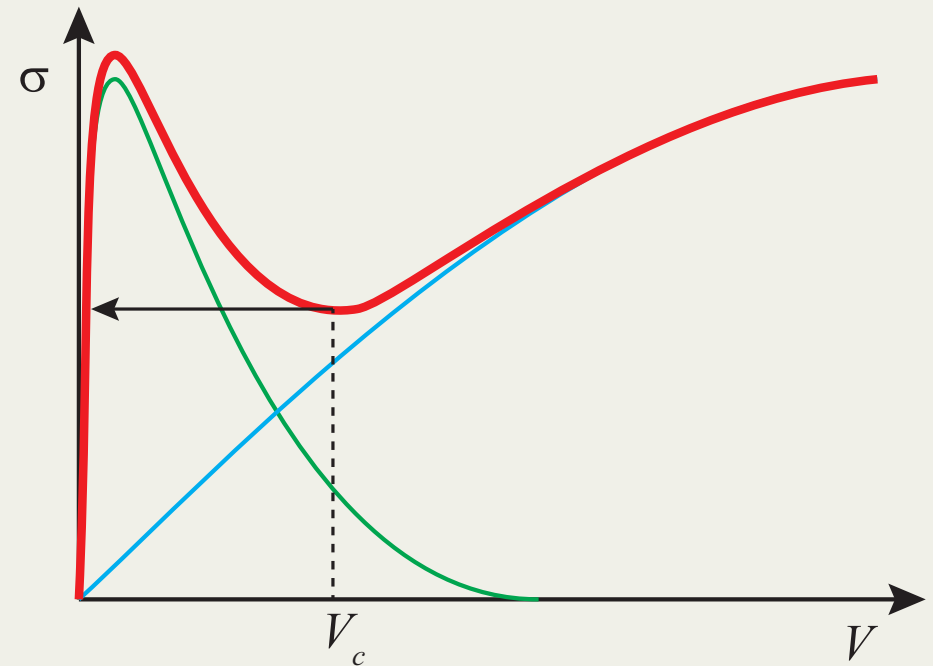


POLYMER GELS FRICTION

two contributions:

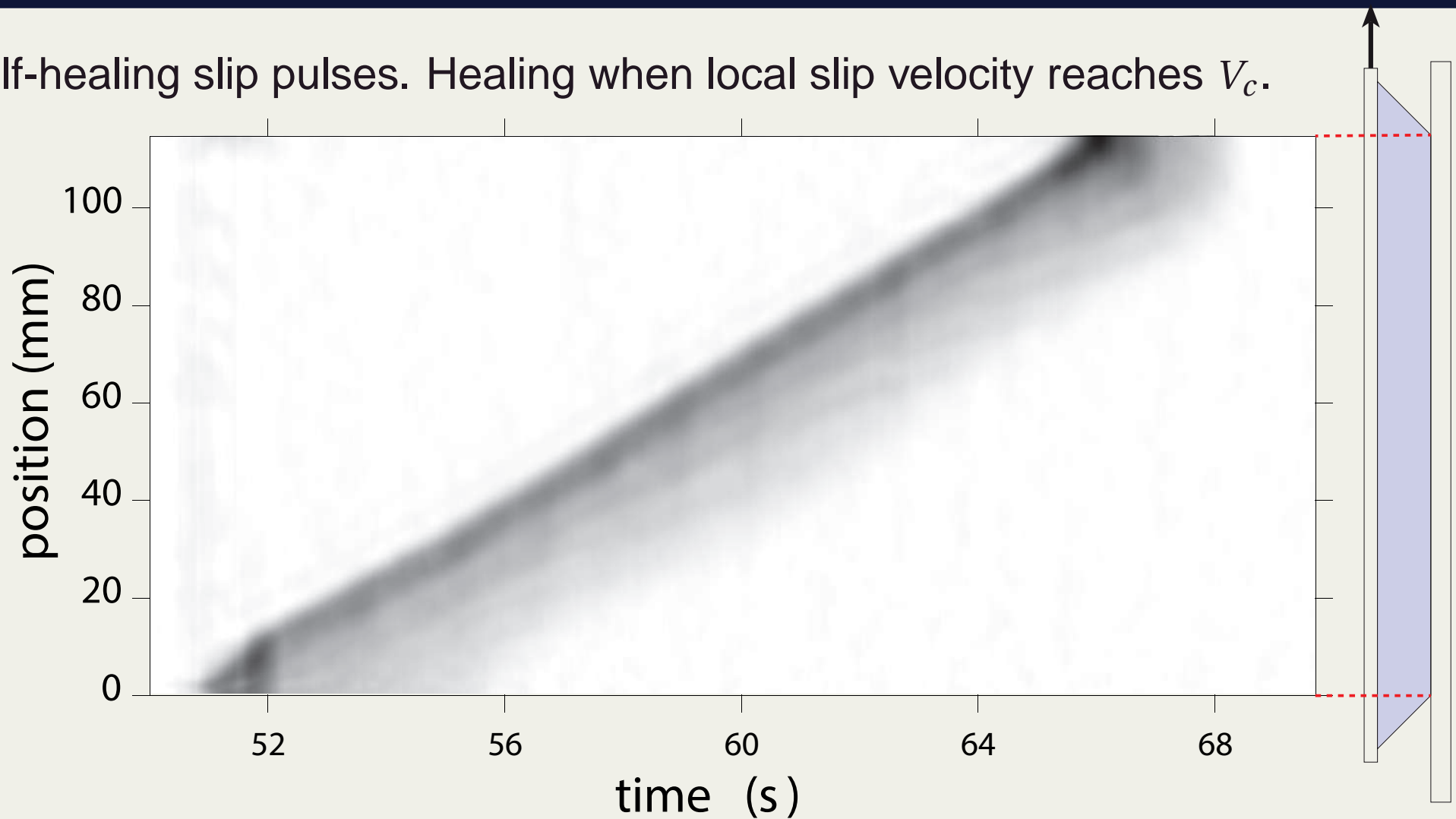
- chain pinning
- viscosity of interfacial layer

⇒ instability at V_c (compliant system
 \propto imposed stress)



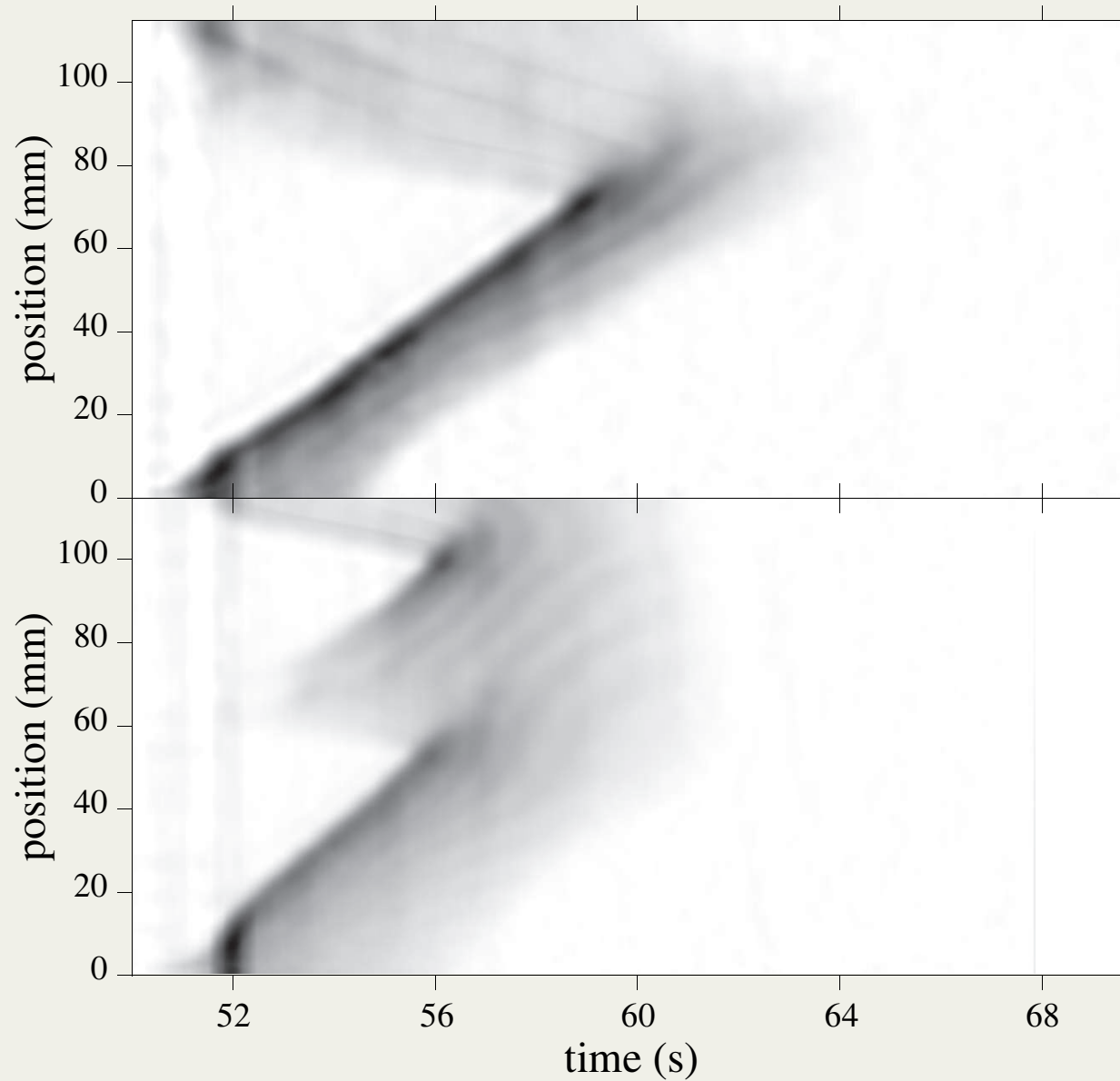
SLEF-HEALING SLIP PULSES IN GELATIN

self-healing slip pulses. Healing when local slip velocity reaches V_c .



SLEF-HEALING SLIP PULSES IN GELATIN

nucleation favored at the edge(s) of the contact, but also within the contact.



CONCLUSION

biphasic nature of gels \Rightarrow 2 contributions to friction

- elastic : (rate-dependent) stretching of adsorbed polymer chains
- hydrodynamic : viscous flow of fluid interfacial layer

relative contributions influenced by many parameters (velocity, pressure, charges, surface chemistry, roughness...)

- Tuning these parameters to achieve very low friction \rightarrow artificial cartilage : need for tougher gels \Rightarrow double network gels (J.P. Gong *et al.*).
- their competition can lead to instabilities \Rightarrow model system for earth crust (F. Corbi *et al.*, *J.G.R. Solid Earth*, vol. 118, p. 1483 (2013)).

