

Friction/Fracture of Hydrogels

Tristan Baumberger

Christiane Caroli, Olivier Ronsin, Lionel Bureau
 Institut des NanoSciences de Paris,
 Universities of Paris 6 & 7, CNRS, France

- Friction and sliding dynamics at a multicontact interface
- Plasticity of confined material in frictional junctions
- Friction/fracture of transient networks (hydrogels)

What are Hydrogels ?

ex. gelatin, polyacrylamid, ... cartilage

=
 crosslinked polymer network + solvent

Elasticity :

$$G = kT/\xi_{el}^3$$

$$\xi_{el} \approx 10 \text{ nm} ; G = 1-10 \text{ kPa}$$

$$\nu \approx 0.5 \text{ (water)} \Rightarrow E = 3G$$

Diffusion :

$$D_{coll} = kT/(\eta_s \xi_{hydro})$$

$$\xi_{hydro} \sim \xi_{el} ; D_{coll} \approx 10^{-11} \text{ m}^2/\text{s}$$

A poroelastic medium ...
 gels vs. rocks ?

$$\kappa \approx \xi^2 \approx 10^{-17} \text{ m}^2 \text{ (} 10^{-5} \text{ darcy)}$$

$$D_{coll} \ll D_{rock} = E\kappa/\eta_s \approx 10^{-2} \text{ m}^2/\text{s}$$

Gel families...

Chemical gels
(typ. polyacrylamid)
covalent crosslinks
→ permanent



Physical gels
(eg. gelatin)

weaker crosslinks (eg. H-bonds)
→ usually thermoreversible

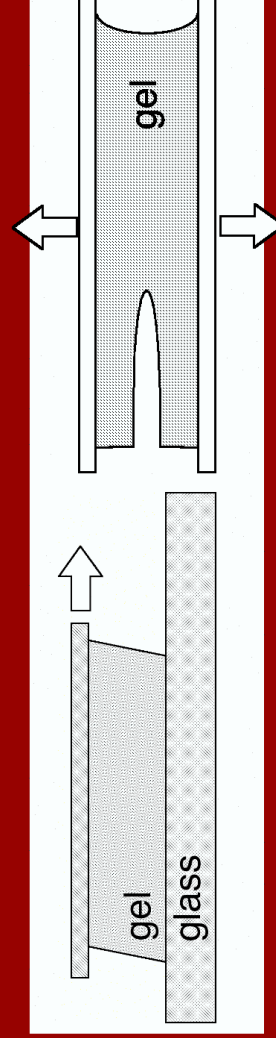
- Transient networks
- Thermal history-dependent
- Neverlasting aging $G(t)$
- Slow creep

Behavior upon sliding ...
adhesive vs. repulsive surface/gel interaction
→ solid friction (threshold) vs. Lubrication
(cf Gong & Osada)

Gelatin gel : a model system

- $c = 5-15$ wt% → $G(c) = 1-10$ kPa ($G''/G' < 10^{-1}$ @ $f < 1$ kHz)
- water/glycerol → tunable (1:4) solvent viscosity η_s

easy to cast, easy to handle ...



Interfacial fracture/sliding

Mode I crack propagation

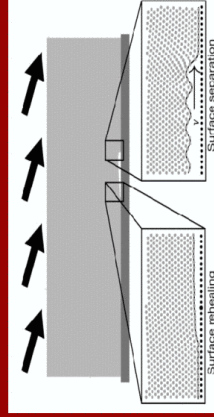
The Friction/Fracture symbiosis...

Sliding at *extended* interfaces

- compliant materials, e.g. gels
- smooth surfaces, e.g. float glass
- large systems, e.g. faults

Friction and fracture are intricate

- Friction law \Rightarrow mode II crack regularisation and healing (Rice & Cochard, Ben-Zion, ...)
- Fracture feature \Rightarrow Amontons' friction law (Gerde & Marder)

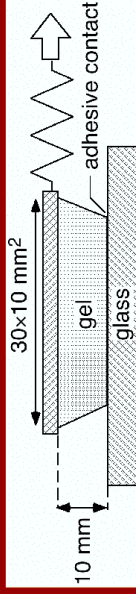


- Schallamach (decohesion) waves
 - strong adhesion
 - large deformations
 - see also Gerde & Marder
- Heaton (slip) pulses
 - from inversion of seismic data
- Adams pulses (theoretical)
 - elastic contrast
 - coulomb $\sigma = \mu p$
 - related to « ill-posedness »
- Still widely open problems :
 - role of the friction law ?
 - role of elastic dissymetry
 - location in parameter space (e.g. $V \dots$) ?

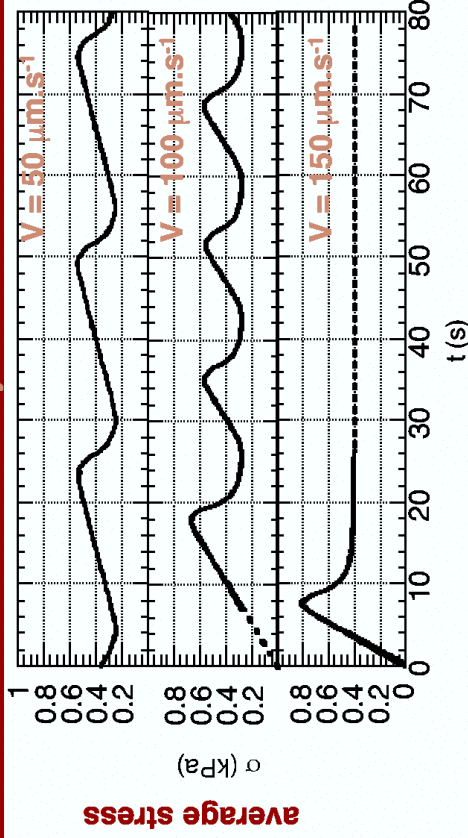
Self-healing slip pulses at a gelatin/glass interface

Baumberger et al. PRL 88 (2002) 75509 ; Eur. Phys. J. E 11(2003) 85

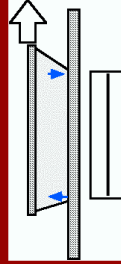
Finite-size gel blocks driven at constant remote velocities $V = 10\text{--}2000 \mu\text{m/s}$



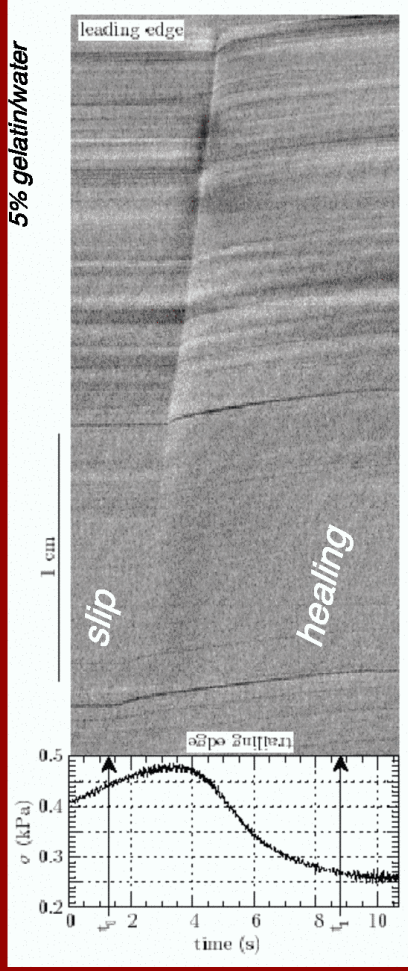
Global dynamics



→ at $V = V_c$: a finite amplitude bifurcation...without hysteresis suggests internal degrees of freedom ...

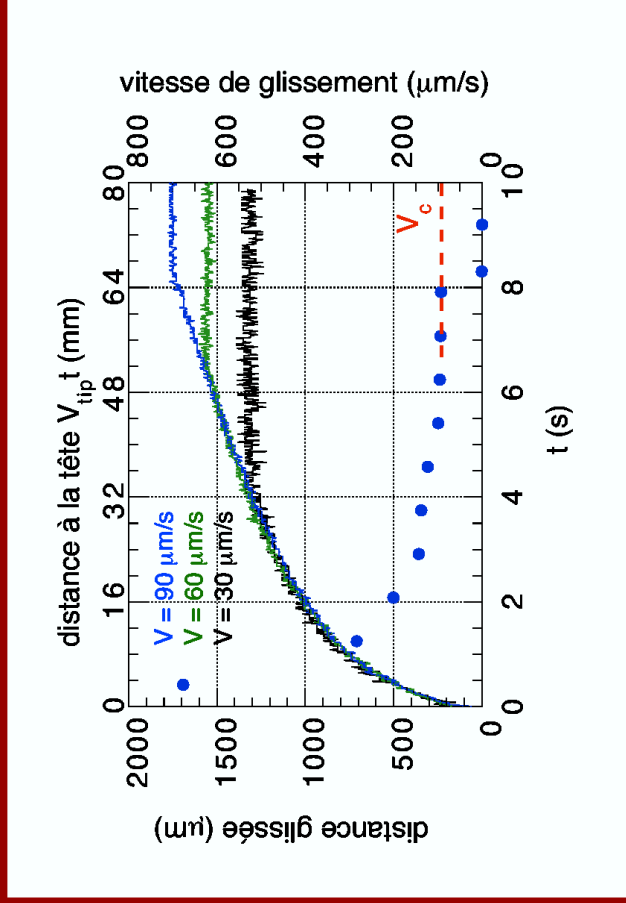


Spatio-temporal representation of the slip field



- $V_{\text{tip}} = 8 \text{ mm/s} \Rightarrow \text{subsonic crack } (c_s = (G/\rho)^{1/2} \sim \text{m/s})$
- no decohesion \Rightarrow slip pulse

Healing occurs at *finite* slip velocity



→ sliding is *not* possible at $v < V_c$

Physical origin of V_{tip} and V_c ?

NB. $V_{tip} \ll V_c \ll C_s$

→ V_{tip} scales with the *poroelastic* velocity $V_{poro} = D_{coll}/\xi$

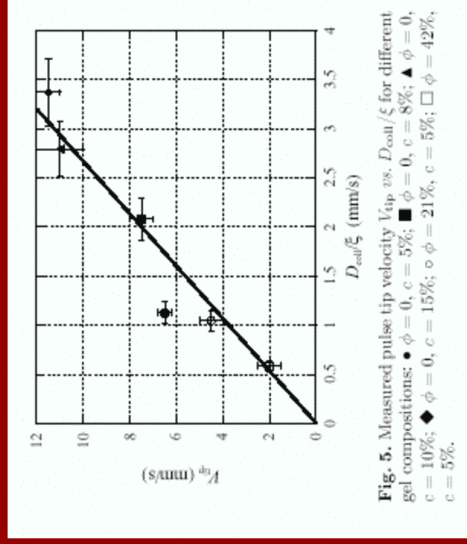


Fig. 5. Measured pulse tip velocity V_{tip} vs. D_{coll}/ξ for different gel compositions: \bullet $\phi = 0$, $c = 5\%$; \blacktriangle $\phi = 0$, $c = 8\%$; \blacklozenge $\phi = 0$, $c = 10\%$; \circ $\phi = 0$, $c = 15\%$; \square $\phi = 21\%$, $c = 5\%$; \diamond $\phi = 42\%$, $c = 5\%$.

Qualitative picture :

- once nucleated the crack accelerates
- bond rupture generates a signal at $\omega \sim V_{tip}/\xi$ with $q \sim \xi$ (here, the process zone size)
- it resonates with the diffusive, slow Biot mode $\omega = D_{coll} q^2$ when $V_{tip} \approx V_{poro}$
- excess energy is then very efficiently dissipated

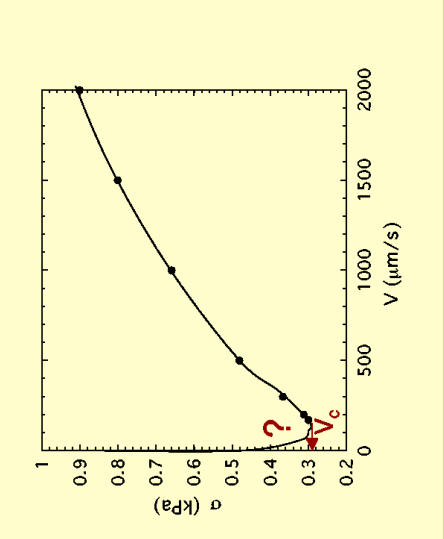
Question :

Is it possible to propagate a crack at $V > V_{poro}$?

→ sudden healing, on the spot, at $v = V_c$
 suggests an *instability* mechanism ...

Rheological instability ?

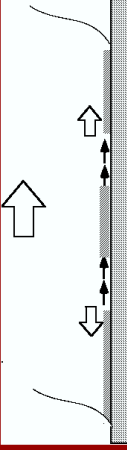
Qualitative picture « à la Schallamach »



- The number of adhesive bonds $n(t)$ is a dynamic variable
 With :
 - ✓ τ = rebonding time
 - ✓ r/V = time of flight over an adhesive site of capture radius r
 - $n(V > r/\tau) = 0$
 - $\sigma \approx \eta(V) V/\xi$ (OK with experiments)
- ⇒ $V_c \approx r/\tau$
- Taking for $r \approx$ a few Å yields $\tau = r/V_c \approx$ a few 10^{-6} s $\sim \tau_{\text{Rouse}}$

Sum-up of the self healing pulses phenomenology

- SHP are observed in a finite size block with strong elastic contrast
 - nucleation at the edge under tension
 - *propagation at a poroelastic velocity*
 - process zone $\sim \xi$
- healing a finite slip velocity V_c fixed by some characteristic pinning time
 - *the existence of a minimum in $\sigma(v)$ is crucial*
 - no SHP for $V > V_c$
- the work provided by the remote drive is essentially dissipated by friction (i.e. viscous dissipation in the fluid interfacial layer at a shear rate $\sim V_c/\xi$)
- SHP are also observed in an edge-free annular geometry
 - cf. Rubio & Galeano [PRE 50 (1994) 1000, Europhys. Lett. 49 (2000) 410]
 - \exists steady states with n pulses running at constant $V_n < V_{\text{poro}}$
 - nucleation from stressed or weak patches
 - *two pulses may counter propagate*

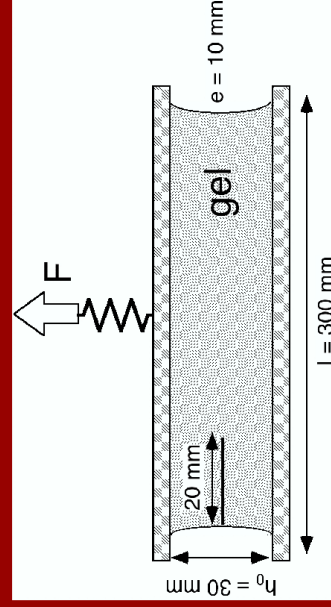
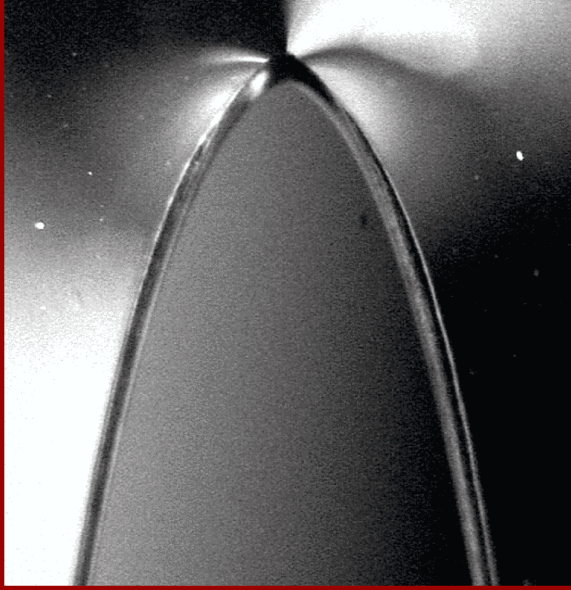


[cf. e.g. Ben-Zion et al, 2005 submit. Geophys. J. Int]

→ Is there anything relevant to fault dynamics ?

Mode I crack dynamics in gelatin gels

[Unpublished material]



Single notch

Fixed grip

« semi- infinite » slab

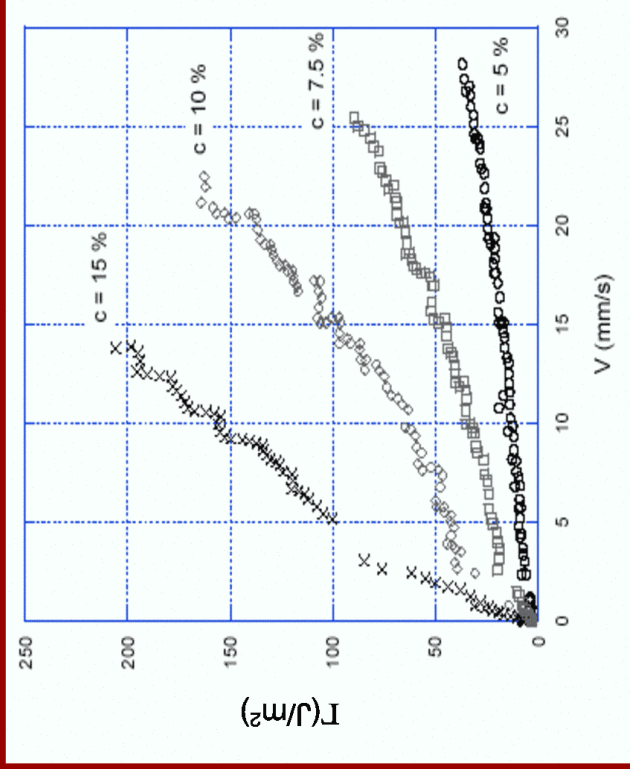
Fracture energy :

$$\Gamma_{\infty} = 1/2 E^*(\Delta h)^2/h_0$$

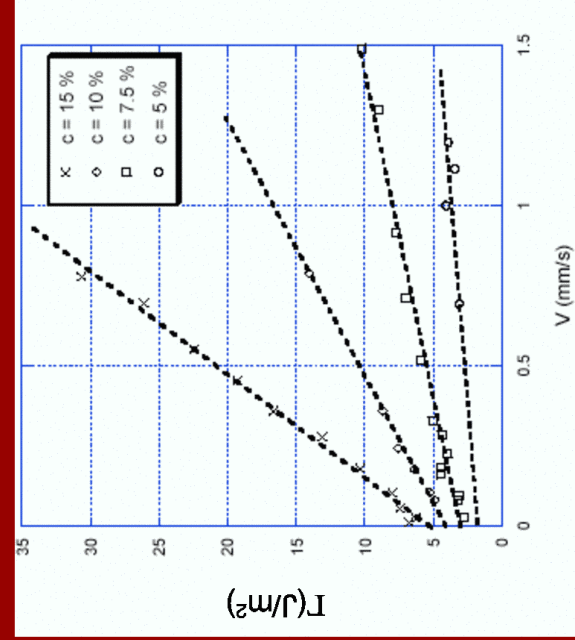
$$\Gamma = - 1/2 \Delta h/e \, dF/dx$$

tip velocity $V = dx/dt$

$$\Gamma(V ; c_{\text{gelatin}}, \eta_{\text{solvent}}, \dots) ?$$

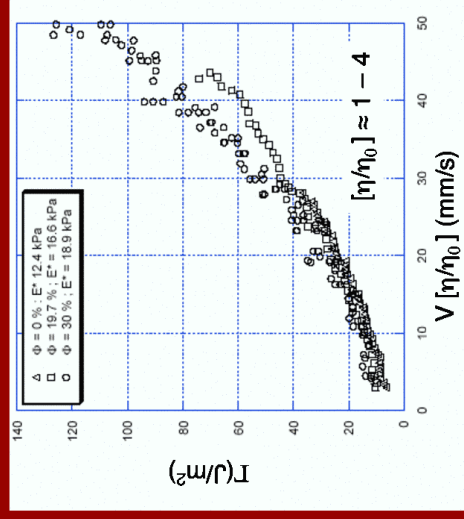
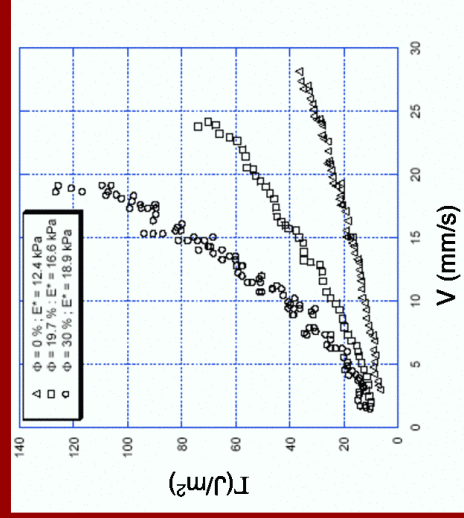


Γ and E increase with $c \Rightarrow$ the stiffer the tougher

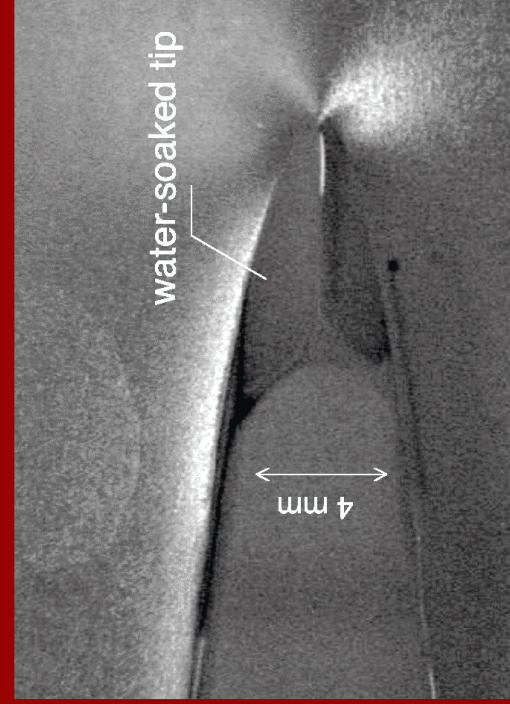


$\Gamma_0 \gg 2\gamma_{\text{water/air}}$
 $\Gamma_0 \xi^2 \sim 10^3 \text{ eV}$

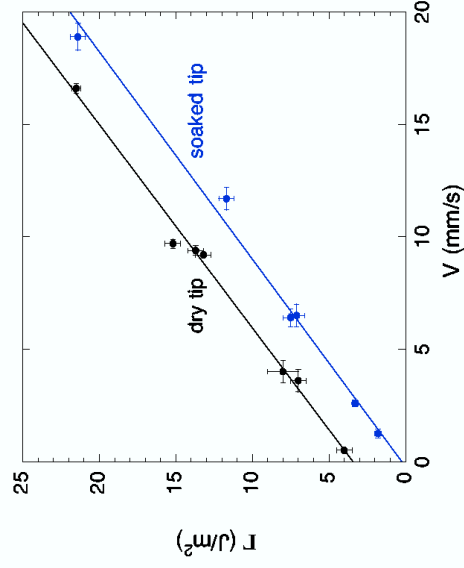
Role of solvent (1)



Role of solvent (2)



NB. $\Delta P_{\text{Laplace}} = \gamma/r \ll \sigma_{\infty}$



NB. for viscoelastic fracture (cf.e.g.Persson & Brenner, PRE 71 (2005) 036123) one would expect $\Gamma(V) = \Gamma_0[1+f(V)]$... seems to be ruled out here.

Possible routes to interpretation of $\Gamma(V)$...

1. Keep in mind...

- $\Gamma_0 \sim$ a few J/m²
- $\Gamma_0 \gg 2\gamma_{\text{water/air}}$
- $\Gamma_0 \xi^2 \sim 10^3$ eV \gg sission energy for a chain ($U \sim$ a few eV)

2. Lake & Thomas argument

- Breaking a chain of n elements requires stocking $nU \gg U$, ultimately released

[Lake & Thomas, Proc. Roy. Soc. A 300 (1967) 108]

$$\rightarrow \Gamma_0 \sim nU\Sigma$$

$$\rightarrow \text{ideal, semi-dilute, gel : } S \sim \xi^{-2} \sim 1/(na^2) \Rightarrow \Gamma_0 \sim U/a^2 \sim \text{a few J/m}^2$$

\rightarrow OK but ... incompatible with the weakness of the crosslinks

3. Poroelasticity

- if the process zone extends over L ,
drained/undrained transition (Ruina) at $V \sim D_{\text{coll}}/L$
- In PAAm gels Y. Tanaka see a change in fracture profile for $V \sim D_{\text{coll}}/\xi$
- $L \sim \xi$ in PAAm (strong chemical gels) ?
- at first sight nothing happens here (cf. point 5 below) : $L \gg \xi$?

4. Viscoelasticity

- Essentially ruled out by soaked-tip experiment
- $\Gamma(V)$ should then be attributed to rate-dependent dissipation within the process zone...

5. Crazing

[see e.g. Rottler & Robbins PRE 68 (2003) 011820]

- Could it occur here ?
- hint : the fracture profile is not mirror-like ($Ra \sim \mu m$), whatever the velocity
- The gel network can be plastically deformed, probably by unwinding (?)
the crosslinks : favorable to drawing fibrils ?
- A highly speculative issue !!