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► To cite this version:

Imen Naassaoui, Olivier Ronsin, Tristan Baumberger. A poroelastic signature of the dry/wet state of a crack tip propagating steadily in a physical hydrogel. *Extreme Mechanics Letters*, 2018. hal-01755515

HAL Id: hal-01755515

<https://hal.science/hal-01755515>

Submitted on 30 Mar 2018

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A poroelastic signature of the dry/wet state of a crack tip propagating steadily in a physical hydrogel

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Abstract

We report detailed measurements of the rate-dependent fracture energy Γ of poroelastic gelatin gels for two distinct boundary conditions on the crack faces. When the crack tip is in contact with a reservoir of solvent, Γ increases linearly with the crack velocity V . When the tip is exposed to air, $\Gamma(V)$ exhibits a marked departure from linearity at low enough velocities. We show that viscous dissipation associated with poroelastic flow in the tip vicinity plays a minor role in this phenomenon. We interpret the dry crack peculiar behavior as resulting from the rate-dependent hydration state of the gelatin chains in the cohesive zone, where they are pulled-out. At large velocities, the cohesive zone is fully dry and Γ is larger than in the wet tip case. At velocities low enough for solvent to flow from the bulk of the gel, partial rehydration occurs and reduces the difference between the dry and wet fracture energies.

Keywords: hydrogels, poroelastic fracture

1. Introduction

Hydrogels, made of self-assembled biopolymer networks in aqueous solvents, are increasingly involved in load-bearing structures, such as implantable scaffolds for tissue engineering or patches for active wound dressings and drug delivery. In such applications, their interface with physiological fluids is clearly an issue since, with such highly stretchable materials, even subcritical surface flaws may widely open, their blunted tips being exposed to aggressive solutes. The extreme environmental sensitivity of crack growth in physical

(non-covalently crosslinked) hydrogels has been revealed through several experimental studies [4, 6, 7]. It has been shown that water-soluble species can diffuse from the crack-tip opening into the process zone where they modify e.g. the solvent viscosity or the strength of the solvent/polymer interaction. Consequently, they can strongly affect (usually weaken) stationary or slowly propagating cracks.

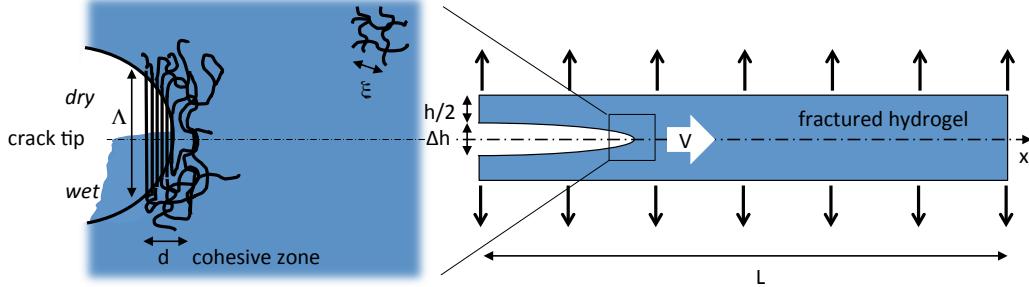
The complex role of solvent flow on the mode I fracture toughness of a polymer gel containing a semi-infinite growing or stationary crack has recently motivated several theoretical and numerical studies [19, 11, 8, 14, 15] accounting for the poroelastic nature of the soft solids. It has been predicted that the fracture energy could be modified by (i) the time-dependent load transfer between the pore pressure and the network tension, and (ii) the rate-dependent viscous dissipation associated with solvent flow. Interestingly, the poroelastic effect is found to depend on whether the sample, hence the crack tip, is in contact either with air or with a solvent bath. It has also been pointed out [15] that the existence of a finite size cohesive zone driving solvent flow in the near-tip region has profound consequences on the calculated toughness. The emerging picture is that of a rich and complex issue with lacking connections between experiments and theory.

In this paper, we revisit and extend previous experimental results on gelatin gels [4, 3]. These physical networks are known to fracture without chain scission. Chains are entirely pulled-out in the cohesive zone and exposed either to air (*dry* crack) or to a drop of the gel solvent itself (*wet* crack). Extracting chains into air, rather than into their aqueous solvent, is known to induce an extra energy cost. We find that at low enough velocities, the corresponding excess of fracture energy for nominally dry cracks decreases and tends to vanish. We provide experimental evidence in favor of a simple picture where the cohesive zone of dry cracks is partially rehydrated by the poroelastic flow from the bulk of the sample, while for wet cracks, imbibition of the chains from the tip reservoir is complete, showing no measurable delay with respect to the crack advance.

2. A reminder on the fracture energy of single network, physical hydrogels.

The low velocity (strongly subsonic) fracture dynamics in hydrogels for which physical, non-covalent crosslinks act as mechanical fuses preventing chain scission has been extensively described [4, 3, 2]. To a first approx-

Figure 1: Right: Fracture setup. The gel sample of length L , initial height h and thickness e (not shown) is loaded (displacement Δh) in pure shear configuration. The crack tip propagates at a velocity V along the x direction. Left : Schematic blow-up representation of the gel tip. In the cohesive zone of extension d along the fracture plane, the triple helix crosslinks (meshsize ξ) are unzipped and the chains are extracted taut (contour length Λ). In the “dry tip” configuration (upper half) the chains are exposed to air while in the wet tip” one (lower half) they remain hydrated by a solvent drop wetting the crack opening.



imation, the fracture energy $\Gamma(V)$ of a mode I crack steadily propagating at a velocity V in a gelatin gel is well described by a linear law: $\Gamma(V) = \Gamma_{\text{wet}}^{(0)} + \Delta\Gamma^{(0)} + \tilde{\Gamma}\eta V$, where the shift factor $\Delta\Gamma^{(0)}$ is a constant, positive when the crack is “dry” i.e. in contact with air and null when the crack is “wet” i.e. in contact with a reservoir of the gel solvent. A cohesive zone model has been proposed to account for this expression. In brief, $\Gamma_{\text{wet}}^{(0)}$ corresponds to the energy paid for unzipping the extended crosslinks (triple helices in the gelatin case). The velocity-dependent term, featuring the solvent viscosity η , stems from the viscous dissipation cost of chain pull-out. $\tilde{\Gamma}$ is a dimensionless geometrical factor. The shift $\Delta\Gamma^{(0)}$ was attributed [4] to the extra energy cost for exposing the extracted chains to air — a poor solvent for hydrophilic polymers. In this picture, owing to the scissionless rupture mechanism, the maximum crack-tip opening is the contour length $\Lambda \lesssim 1 \mu\text{m}$ of the polymer chains which are stretched taut upon extraction. The depth d of the cohesive zone (Fig. 1) has been estimated experimentally [4] to be typically $d \simeq 100 \text{ nm}$. It is important to emphasize that $d \ll \Lambda$, i.e. that the cohesive zone extends perpendicular to the crack plane. This behavior is characteristic of elastically blunted cracks [10] in soft solids for which the rupture stress is much larger than the elastic modulus, hence where *non-linear* elasticity is at work far beyond the cohesive zone [12, 13, 2].

3. Materials and methods

Gel samples are prepared as described previously[4, 3] by dissolving gelatin powder (type A, from porcine skin, Sigma) in either a mixture containing $\phi = 60$ wt.% glycerol in deionized water or pure deionized water ($\phi = 0$), under continuous stirring at 60° C for 30 min. The hot pre-gel solution is then poured into a mold made of a rectangular metallic frame sandwiched between Mylar films covered by hard flat plates. The inner faces of the long sides of the frame are covered with Velcro tape which ensures good gripping of the gelled sample. The mold is then kept at 5° C during 15 h for gelation to proceed, before being clamped to the mechanical testing setup and left for 2 h at room temperature ($20 \pm 0.5^\circ \text{ C}$). The Mylar films are removed just before running the experiments to avoid solvent evaporation.

The resulting gel sample (length $L = 300$ mm, height $h = 30$ mm, thickness $e = 10$ mm) is first stretched by moving one of the gripping bars along the sample height direction up to $\Delta h_{\max} = 8$ mm at a rate of 12 mm.s^{-1} , then immediately unloaded. The corresponding loading force $F(\Delta h)$ is measured by a stiff dynanometer made of a double spring cantilever, coupled with a capacitive displacement gauge. This enables us to measure the small strain shear modulus G as well as the total elastic energy $\mathcal{E}(\Delta h)$ stored in the sample [3].

The sample is then notched in the middle of one of its edges with a razor blade and loaded by a displacement $\Delta h \leq \Delta h_{\max}$ such that a crack grows from the notch in the middle of the sample (see figure 1). Wet crack experiments are performed by adding a drop ($\simeq 50 \mu\text{L}$) of the gel solvent into the crack tip with a micropipette.

The sample aspect ratio ($L \gg h$) is such that the loading configuration is the “pure shear test” one [16]. Accordingly, the energy release rate of the crack is uniform in a large region, away from the sample edges, where it can be approximated by $\mathcal{G}(\Delta h) = \mathcal{E}/Le$. Note that, although physical hydrogels are known to relax stress, this expression assumes that no dissipation occurs in the bulk of the gel during crack growth. However, it has been shown[17] that, in gelatin gels, stress relaxation slows down as aging proceeds, with a characteristic time which scales with the aging time itself. As our samples are aged for a total of 17 h, keeping them loaded for durations shorter than a few minutes ensures that no significant stress relaxation occurs.

The loading Δh is decreased or increased stepwise in order to scan different values of \mathcal{G} . The crack is followed by recording its propagation with a

video camera. From the tracking of the crack tip position $x(t)$ at constant Δh from the video record, we check that the crack grows at a constant velocity $V = dx/dt$ and we thus have access to the gel fracture energy $\Gamma(V) = \mathcal{G}(\Delta h)$ for dry and wet cracks on the same sample.

The collective diffusion coefficients D_{coll} of the gels were measured by dynamic light scattering[5] on samples having experienced the very same thermal history as the corresponding fractured ones. The physical characteristics of the different samples used in this study are shown in Table 1.

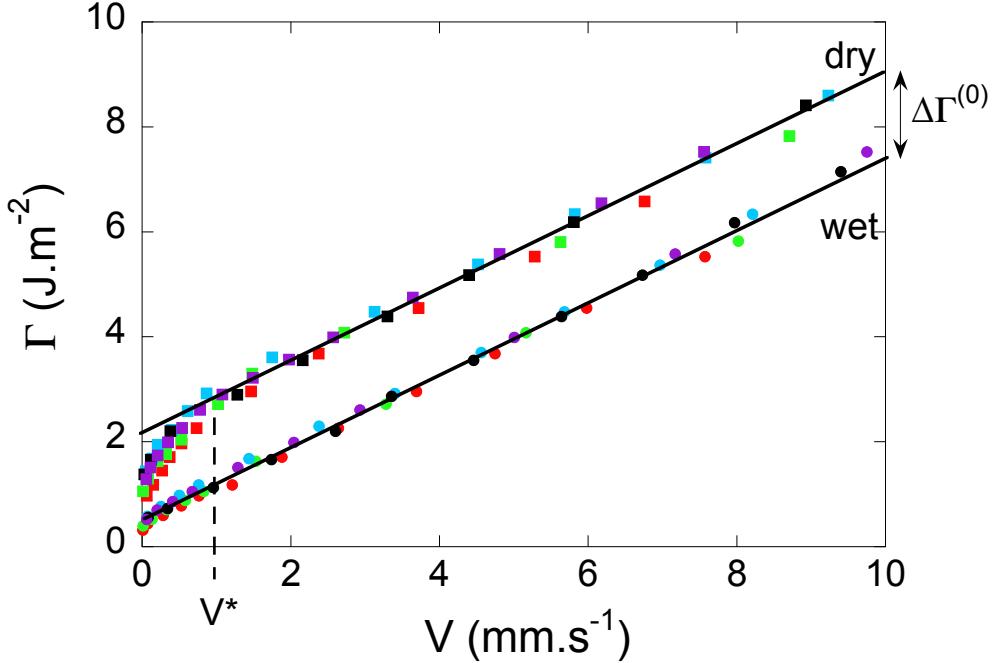
Table 1: Characteristics of the samples used in this study. Gels contain a weight fraction c of gelatin. A solvent with a volume fraction ϕ of glycerol in water has a viscosity η . The shear modulus of the gel at $T = 20^\circ\text{C}$ is G from which the mesh size ξ is estimated according to $G = k_B T \xi^{-3}$ with k_B the Boltzmann constant. The characteristic poroelastic velocity is $V_{poro} = D_{coll}/\xi$ with D_{coll} the collective diffusion coefficient.

c [wt.%]	ϕ [%]	η [mPa.s]	G [kPa]	ξ [nm]	D_{coll} $[10^{-11} \text{ m}^2.\text{s}^{-1}]$	V_{poro} [mm.s $^{-1}$]
5	0	1	2.5	12	2.5	2.1
5	60	11	4.5	9.7	0.4	0.4
10	0	1	9.4	7.6	2.7	3.6
15	0	1	15	6.4	2.9	4.4

4. Experimental results

Figure 2 displays the $\Gamma(V)$ characteristics of mode I cracks for $c = 5$ wt.% gelatin gels in pure water ($\phi = 0$). For each sample, the crack dynamics was assessed under dry and wet crack tip conditions. The high reproducibility of the data over $n = 5$ samples firmly establishes the main result of this study, namely the striking qualitative difference between the $\Gamma(V)$ curve for dry and wet cracks. First of all, at high enough crack velocities one recovers the previously reported behavior, namely both $\Gamma(V)$ characteristics are linear with identical slopes, the dry crack curve being merely shifted upwards from the wet crack one by a constant amount $\Delta\Gamma^{(0)} \simeq 1.6 \text{ J.m}^{-2}$. However, whereas the data for wet cracks remain remarkably aligned over the whole experimental velocity range $0.01 < V < 10 \text{ mm.s}^{-1}$, a systematic deviation from linearity is observed for dry cracks at low velocities $V < V^* \simeq 1 \text{ mm.s}^{-1}$.

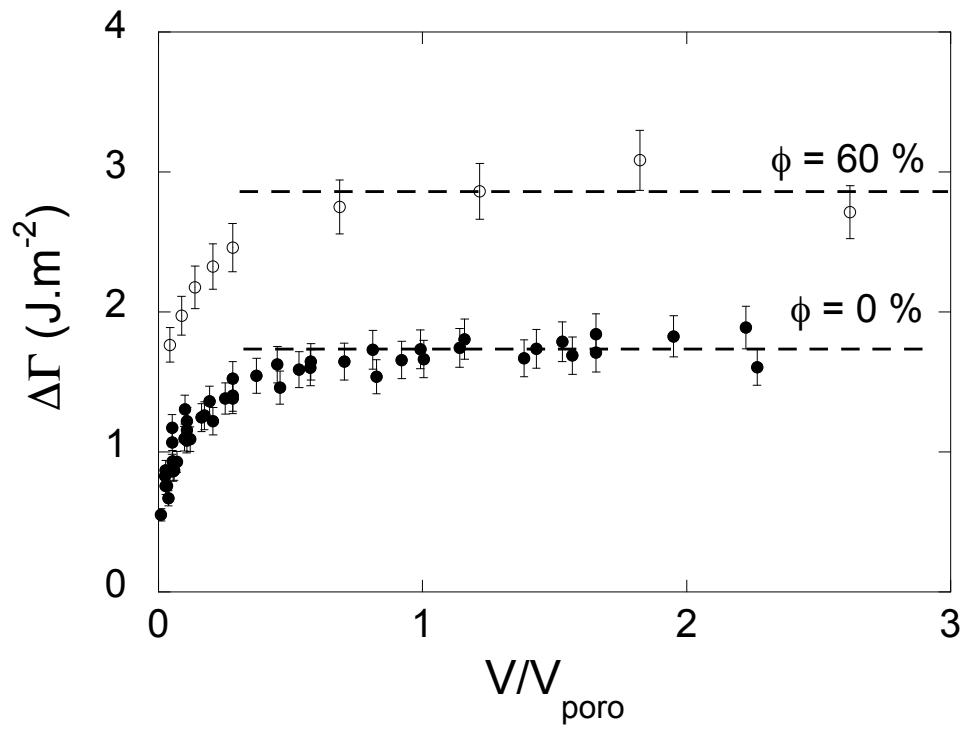
Figure 2: Fracture energy Γ with respect to the steady crack tip velocity V for five, nominally identical samples (different colors on line) in both the dry (squares) and wet (circles) crack tip configurations in a $c = 5$ wt.% $\phi = 0$ gel. The velocity V^* corresponds to the departure from linearity of the $\Gamma_{dry}(V)$ curve. For $V \gg V^*$ data are aligned on parallel straight lines shifted by $\Delta\Gamma^{(0)}$.



This can be described conveniently by a velocity-dependent hydration shift factor $\Delta\Gamma(V) = \Gamma_{dry}(V) - \Gamma_{wet}(V)$ with $0 < \Delta\Gamma < \Delta\Gamma^{(0)}$. Dry cracks are therefore weaker and weaker as their velocity decreases, their toughness approaching that of wet crack ones as $V \rightarrow 0$. This suggests that nominally dry cohesive zones get partially rehydrated at low enough velocities. Since the only available source of hydration for a dry crack is the solvent contained inside the sample itself, this naturally points to a poroelastic draining phenomenon.

Let us make a few scaling remarks. Poroelastic transport is controlled by the collective diffusion coefficient D_{coll} , which scales as $k_B T / (\eta \xi)$, where ξ is the network mesh size. In the frame of the crack tip moving at velocity V , solvent diffusion affects a skin of depth $d_{poro} \sim D_{coll}/V$. For

Figure 3: Shift function $\Delta\Gamma = \Gamma_{dry} - \Gamma_{wet}$ plotted with respect to the reduced velocity V/V_{poro} for two gels with $c = 5$ wt.% differing by their glycerol content ϕ . Dashed lines correspond to $\Delta\Gamma^{(0)}$ (see Fig.2).



$V > V_{\text{poro}} = D_{\text{coll}}/\xi$, the skin depth is smaller than the mesh size ξ . Certainly, for $V \gg V_{\text{poro}}$, the poroelastic flow becomes irrelevant and the crack tip remains undrained. It is therefore legitimate to expect that the velocity V^* above which $\Delta\Gamma(V)$ saturates is on the order of V_{poro} .

In order to test this guess we have taken advantage of the ability of gelatin to form gels in water-glycerol mixtures. Increasing the fraction ϕ of glycerol results in a decrease of V_{poro} due to the increase of η , mitigated by a decrease of ξ which depends on the solvent composition [18] (see Table 1). Using a water-glycerol ($\phi = 60\%$) solvent, V_{poro} is decreased by a factor of 5 with respect to the pure water gel with the same gelatin content ($c = 5$ wt.%). Fig.3 displays the corresponding shifts $\Delta\Gamma$ as a function of V/V_{poro} . It is clear that both factors reach their large velocity values $\Delta\Gamma^{(0)}$ for $V/V_{\text{poro}} = \mathcal{O}(1)$ which lends strong support to our claim that the qualitative difference between the dry and wet cracks is of poroelastic origin. The remaining vertical shift between both curves in Fig.3 is reasonably attributable to the difference between the areal densities ($\sim \xi^{-2}$) of chain crossing the fracture plane, and thus exposed to air, for the networks in their respective preparation states.

5. Discussion

5.1. The wet crack case

First of all, let us stress that $\Gamma_{\text{wet}}(V)$ showing no measurable departure from linearity for $V < V_{\text{poro}}$ deserves attention in itself. Many authors [19, 8, 15] have shown that for free draining boundary conditions, solvent is pumped from the crack faces so as to swell a zone centered around a point located about d_{poro} ahead of the crack tip. The viscous dissipation associated with this flow contributes an amount $\Delta\Gamma_{\text{poro}}^{\text{wet}}$ to the effective fracture energy $\Gamma_{\text{wet}}(V)$ (defined so as to match the energy release rate \mathcal{G} during quasistatic crack propagation). For $V \gg V_{\text{poro}}$ draining is irrelevant, $\Delta\Gamma_{\text{poro}}^{\text{wet}}$ should tend to zero in this limit, therefore preserving the linearity of $\Gamma_{\text{wet}}(V)$. Since this remains so even for $V < V_{\text{poro}}$, we are led to conclude that within experimental accuracy $\Delta\Gamma_{\text{poro}}^{\text{wet}}(V)$ is negligible with respect to the contribution from chain/solvent friction in the cohesive zone. Noselli et al. [15] have computed $\Delta\Gamma_{\text{poro}}^{\text{wet}}/\Gamma_{\text{wet}}$ and found that it depends essentially on the drained poisson ratio ν . The ratio naturally vanishes for $\nu = 1/2$ but can becomes of order unity for osmotically compliant networks ($\nu < 1/2$). Nevertheless, this analysis was performed within the *linear* poroelasticity framework and we will argue that in our experiments *non-linear* elasticity is at work, even outside

the cohesive zone. Thus, we cannot make use of the quantitative result of [15] and rather consider our experimental result as a strong indication that the fracture energy of a wet crack in gelatin gels is dominated by the pull-out of chains in a thin cohesive zone, extending along the crack front, and remaining imbibated by solvent flowing readily from the reservoir-drop.

5.2. The dry crack case

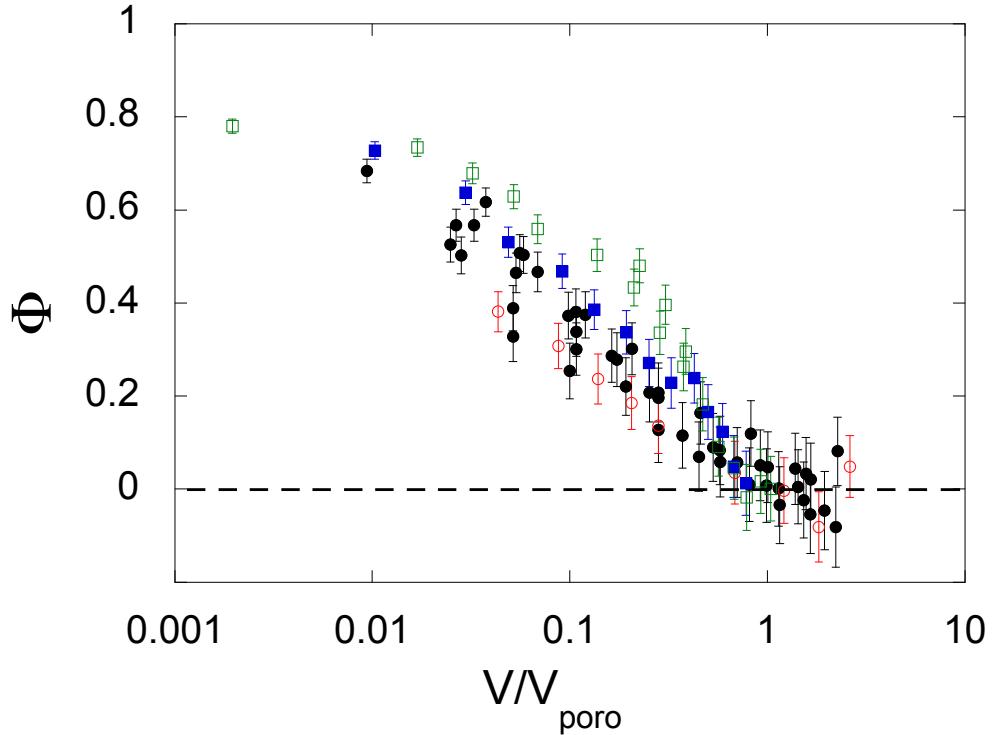
Let us now focus on the dry crack case for which solvent is merely redistributed inside the bulk of the sample, in the crack tip region. The effective fracture energy now reads $\Gamma_{wet}(V) = \Gamma_{wet}^{(0)} + \tilde{\Gamma}\eta V + \Delta\Gamma^{(0)} + \Delta\Gamma_{poro}^{dry}$ where $\Delta\Gamma^{(0)}$ is the hydration term and $\Delta\Gamma_{poro}^{dry}$ stems from the dissipation associated with the internal solvent flow. In order to gain some feeling about the importance of this term, we have extended the analysis of Noselli et al. [15] to the dry case. We show (Appendix) that, within the linear poroelasticity limit, $\Delta\Gamma_{poro}^{dry} \simeq \Delta\Gamma_{poro}^{wet}$. We take it as a hint that, in the dry case as well, viscous dissipation out of the cohesive zone has negligible effect on $\Gamma_{dry}(V)$. We propose that the decrease of the fracture energy of the dry crack below V_{poro} originates from partial rehydration of the extracted chains by solvent flowing into the cohesive zone from the bulk of the gel.

In order to test this hypothesis, we have computed $\Phi = 1 - \Delta\Gamma(V)/\Delta\Gamma^{(0)}$ as an estimate of the volume fraction of the cohesive zone which has been rehydrated by the poroelastic flow. Fig.4 shows a plot of Φ with respect to V/V_{poro} for the two gels $c = 5$ wt.% with $\phi = 0$ and 60 % (data of Fig.3) and two gels $c = 10$ and 15 wt.% with $\phi = 0$. All data reasonably collapse onto a single curve meaning that, in full agreement with our scenario: (i) it is the hydration contribution $\Delta\Gamma^0$, rather than viscous dissipation which controls the dry/wet fracture energy shift; (ii) the partial rehydration dynamics is of poroelastic origin.

5.3. Extension of the draining basin and relevance of non-linear elasticity

Figure 4 shows clearly that for $V/V_{poro} < 1$, and over at least two decades in reduced velocity, Φ decreases quasi-logarithmically. Extrapolating this singular behavior to $\Phi = 1$ would predict full rehydration to occur for $V/V_{poro} \simeq 2 \times 10^{-3}$ corresponding to a draining basin of extension $\simeq 500\xi$. This lengthscale, of order 5 μm , is larger than the extension $d \times \Lambda$ of the cohesive zone. Also important is the extension of the zone ahead of the crack where the network behaves as a strongly *non-linear* elastic medium. A conservative estimate of this length is $\mathcal{L}_{NL} = \Gamma/E$ with E the Young

Figure 4: Reduced shift function $\Phi = 1 - \Delta\Gamma/\Delta\Gamma^{(0)}$ with respect to the reduced velocity V/V_{poro} . Closed circles: $c = 5\%, \phi = 0\%$; open circles: $c = 5\%, \phi = 60\%$; closed squares: $c = 10\%, \phi = 0\%$; open squares: $c = 15\%, \phi = 0\%$. The dashed line $\Phi = 0$ corresponds to the high velocity, undrained tip limit.



modulus, corresponding to the distance from the crack tip where LEFM would predict stresses to become of order the Young modulus [10]. For this study, $\mathcal{L}_{NL} \gtrsim 100 \mu\text{m}$. Consequently, the whole draining basin lies within the strongly non-linear elastic zone, so that, as already mentioned, a linear poroelastic analysis cannot be expected to be quantitatively relevant.

6. Concluding remarks

The experimental results described in this study turn out to be simple and robust enough to serve as a touchstone for theoretical and numerical models of cracks in poroelastic soft solids. As mentioned in the discussion, none of the existing studies [8, 15, 19, 11] accounts for all the basic requirements of our system, e.g. steady crack growth and non-linear elasticity. As noted by several authors [10, 12, 6, 13], the need for resorting to non-linear elastic fracture mechanics is a general requirement for hydrogels as highly stretchable materials. Expressions for the elastic energy density accounting for the strong strain hardening encountered in polymer gels have been given and studied in [12]. How to take into account the effect of large strain on the poroelastic characteristics is less clear and we are not aware of any model or measurements which describe the anisotropic mobility tensor relating the fluxes to the gradients of chemical potential [8, 9].

More specific to gelatin gels is the fact that they are underswollen in their preparation state. Since physical gelation is basically an arrested phase separation, many hydrogels tend to expel solvent out of the network during the polymer self-assembly process. This so-called syneresis results in gel surfaces covered with a layer of solvent which may play the role of a reservoir for the crack cohesive zone. This is presumably the case for calcium alginate gels which we found to show [7] no hydration effect ($\Delta\Gamma^{(0)} = 0$). In the case of gelatin, the cohesive zone of a dry crack tends to remain dry; i.e. the chains are exposed to air. The menisci at the solvent/air interface in the dry pores generates a negative Laplace pressure of order $-\Delta\Gamma^{(0)}/\Lambda$ [2]. It is the balance between the average negative pressure $\bar{p} \simeq -\Delta\Gamma/\Lambda = -(1 - \Phi)\Delta\Gamma^{(0)}/\Lambda$ over the cohesive zone and the pore pressure in the bulk of the gel which controls partial rehydration. It is therefore important to control the chemical potential of solvent in a reference state which is the preparation one (as usual in experimental studies) rather than the swelling equilibrium one (as usual in numerical studies).

A final remark concerns an unexpected implication of a such qualitative signature as a diagnosis of the dry/wet state of the crack. When the tip opening is wet by a solution of particles small enough to be brownian but too large to diffuse into the gel network, it is likely that the poroelastic flow which drains solvent into the gel will leave a layer of nanoparticles clogging the crack tip. If the hydraulic resistance of such a porous layer is large enough, it will prevent the cohesive zone of the nominally wet crack to be fully hydrated. The fracture energy of such a clogged crack should therefore exhibit the specific signature studied in this paper. Such a phenomenon has actually been observed and will be published elsewhere.

Acknowledgements

We thank Christiane Caroli for careful reading of the manuscript and insightful discussions.

Appendix

In this appendix, we evaluate the contribution to dissipation of the poroelastic solvent flow for a mode I crack, steadily propagating at velocity V in a linear poroelastic material. We extend the calculation of Noselli et al. [15] performed for a wet crack, to the case of a dry crack. For simplicity, we chose to characterize the poroelastic material properties in terms of the shear modulus G , the (drained) Poisson coefficient ν and the collective diffusion coefficient D_{coll} .

The derivation is based on the assumption that solvent flow is confined to a region surrounding the crack tip, on the order of d_{poro} that remains small as compared to the size fo the specimen. So, outside this region $r > d_{poro}$, the gel behaves as an elastic (undrained) material and there exists an enclosing domain where the linear elastic fracture mechanics asymptotic stress field is dominant. For $r < d_{poro}$, the stress field and chemical potential are given by the linear poroelastic asymptotic expressions given by Atkinson and Craster[1].

With these assumptions, the dissipation resulting from solvent flow through the gel network reads (eq. (35) of [15], rewritten with our notations)

$$\Delta\Gamma_{poro} = \int_{\mathcal{R}_\infty} \frac{\mu}{\Omega} \frac{\partial\epsilon}{\partial x} dA \quad (1)$$

with μ the chemical potential, Ω the solvent molar volume (μ/Ω is the pore pressure), ϵ the dilation and \mathcal{R}_∞ a region where dissipation essentially occurs, which is taken as a disk of radius γd_{poro} with γ a number of order unity.

In order to evaluate (1), [15] derived the asymptotic solutions of ϵ and μ from the equations of poroelasticity with the boundary conditions imposed by the absence of normal and shear stress on the crack faces. To leading order, this gives in polar coordinates (eq. (17) of [15])

$$\epsilon = \frac{1-2\nu}{G} \frac{K_{\text{tip}}}{\sqrt{2\pi r}} \cos \frac{\theta}{2}$$

where K_{tip} is the stress intensity factor at the crack tip, and (eq. (18) of [15], with a corrected typo)

$$-V \frac{\partial \epsilon}{\partial x} = \frac{1-2\nu}{G} \left(\frac{\partial K_{\text{tip}}}{\partial t} \cos \frac{\theta}{2} + \frac{v K_{\text{tip}}}{2r} \cos \frac{3\theta}{2} \right) \frac{1}{\sqrt{2\pi r}} \quad (2)$$

The chemical potential μ is then found by solving, with the appropriate boundary conditions, equation (13.2) of [15], which in steady state reduces to

$$\frac{1}{\Omega} \Delta \mu = -\frac{2G(1-\nu)}{D_{\text{coll}}(1-2\nu)} V \frac{\partial \epsilon}{\partial x}$$

where $\Delta \mu$ is the laplacian of μ .

Wet case

and (ii) , which, for the wet case reduces to

In the wet case, the thermodynamic equilibrium of solvent across the crack faces imposes $\mu = \mu_0$ for $\theta = \pm\pi$, with $\mu_0 = 0$ the chemical potential of the reservoir of solvent (the drop of solvent in our experiments) and the solution is to leading order [15, 1]

$$\frac{\mu}{\Omega} = -\frac{V K_{\text{tip}}(1-\nu)}{2D_{\text{coll}}} \frac{r^{1/2}}{\sqrt{2\pi}} \cos \frac{3\theta}{2} + K_2 r^{1/2} \cos \frac{\theta}{2} \quad (3)$$

Inserting (2) and (3) into (1) gives

$$\Delta \Gamma_{\text{poro}}^{\text{wet}} = K_{\text{tip}}^2 \gamma \frac{1-2\nu}{16G} \quad (4)$$

Dry case

In the dry case, the boundary conditions require that there is no solvent flux across the crack faces, that is $\partial\mu/\partial y = 0$ for $\theta = \pm\pi$. The solution now reads [1]

$$\frac{\mu}{\Omega} = -\frac{VK_{\text{tip}}(1-\nu)}{2D_{\text{coll}}}\frac{r^{1/2}}{\sqrt{2\pi}}\left(\cos\frac{3\theta}{2} + 3\cos\frac{\theta}{2}\right) \quad (5)$$

which gives

$$\Delta\Gamma_{\text{poro}}^{\text{dry}} = K_{\text{tip}}^2\gamma\frac{1-2\nu}{16G} \quad (6)$$

Note that, in (4) and (6), K_{tip} and γ have no reason to be the same for both cases. They however are on the same order, so that, eventually

$$\Delta\Gamma_{\text{poro}}^{\text{dry}} \simeq \Delta\Gamma_{\text{poro}}^{\text{wet}}$$

References

- [1] C. Atkinson and R. V. Craster. Plane strain fracture in poroelastic media. *Proceedings: Mathematical and Physical Sciences*, 434(1892):605–633, 1991.
- [2] T. Baumberger and O. Ronsin. From thermally activated to viscosity controlled fracture of biopolymer hydrogels. *The Journal of Chemical Physics*, 130(6):061102, 2009.
- [3] Tristan Baumberger, Christiane Caroli, and David Martina. Fracture of a biopolymer gel as a viscoplastic disentanglement process. *The European Physical Journal E: Soft Matter and Biological Physics*, 21(1):81–89, 2006.
- [4] Tristan Baumberger, Christiane Caroli, and David Martina. Solvent control of crack dynamics in a reversible hydrogel. *Nature materials*, 5(7):552–555, 2006.
- [5] Tristan Baumberger, Christiane Caroli, and Olivier Ronsin. Self-healing slip pulses and the friction of gelatin gels. *The European Physical Journal E*, 11(1):85–93, 2003.
- [6] Tristan Baumberger and O Ronsin. A convective instability mechanism for quasistatic crack branching in a hydrogel. *The European Physical Journal E: Soft Matter and Biological Physics*, 31(1):51–58, 2010.

- [7] Tristan Baumberger and O Ronsin. Cooperative effect of stress and ion displacement on the dynamics of cross-link unzipping and rupture of alginate gels. *Biomacromolecules*, 11(6):1571–1578, 2010.
- [8] Nikolaos Bouklas, Chad M Landis, and Rui Huang. Effect of solvent diffusion on crack-tip fields and driving force for fracture of hydrogels. *Journal of Applied Mechanics*, 82(8):081007, 2015.
- [9] Wei Hong, Xuanhe Zhao, Jinxiong Zhou, and Zhigang Suo. A theory of coupled diffusion and large deformation in polymeric gels. *Journal of the Mechanics and Physics of Solids*, 56(5):1779–1793, 2008.
- [10] C-Y Hui, A Jagota, SJ Bennison, and JD Londono. Crack blunting and the strength of soft elastic solids. In *Proceedings of the Royal Society of London A: Mathematical, Physical and Engineering Sciences*, volume 459, pages 1489–1516. The Royal Society, 2003.
- [11] Chung-Yuen Hui, Rong Long, and Jing Ning. Stress relaxation near the tip of a stationary mode i crack in a poroelastic solid. *Journal of Applied Mechanics*, 80(2):021014, 2013.
- [12] Venkat R Krishnan, Chung Yuen Hui, and Rong Long. Finite strain crack tip fields in soft incompressible elastic solids. *Langmuir*, 24(24):14245–14253, 2008.
- [13] Ariel Livne, Eran Bouchbinder, Ilya Svetlizky, and Jay Fineberg. The near-tip fields of fast cracks. *Science*, 327(5971):1359–1363, 2010.
- [14] Alessandro Lucantonio, Giovanni Noselli, Xavier Trepat, Antonio DeSimone, and Marino Arroyo. Hydraulic fracture and toughening of a brittle layer bonded to a hydrogel. *Physical review letters*, 115(18):188105, 2015.
- [15] Giovanni Noselli, Alessandro Lucantonio, Robert M McMeeking, and Antonio DeSimone. Poroelastic toughening in polymer gels: A theoretical and numerical study. *Journal of the Mechanics and Physics of Solids*, 94:33–46, 2016.
- [16] R. S. Rivlin and A. G. Thomas. Rupture of rubber. i. characteristic energy for tearing. *Journal of Polymer Science*, 10(3):291–318, 1953.

- [17] O. Ronsin, C. Caroli, and T. Baumberger. Interplay between shear loading and structural aging in a physical gelatin gel. *Phys. Rev. Lett.*, 103:138302, Sep 2009.
- [18] Olivier Ronsin, C Caroli, and Tristan Baumberger. Preferential hydration fully controls the renaturation dynamics of collagen in water-glycerol solvents. *The European Physical Journal E*, 40(5):55, 2017.
- [19] Xiao Wang and Wei Hong. Delayed fracture in gels. *Soft Matter*, 8(31):8171–8178, 2012.