TOUGHNESS OF HYDROGELS

John L. Bassani*, Konstantinos Garyfallogiannis and Prashant K. Purohit

University of Pennsylvania, Philadelphia, USA
**Presenting Author email: bassani@seas.upenn.edu*

Abstract
Hydrogels are soft, highly deformable materials with applications ranging from soft actuators to natural and synthetic biomaterials. The rupture of hydrogels generally involves very large deformations that can be strongly coupled to the fluid flow. In this paper, a modified *J*-integral (*J**) is used to calculate the critical energy release rate utilizing either a critical stretch criterion or the measured overall force-extension relation for a SENT specimen of a fibrin gel, which is the primary stress-carying component of blood clots.

Introduction
We consider a poroelastic material modelled as a compressible (porous) solid undergoing large deformations and infiltrated with fluid. The effect of the fluid is described by a Flory-Rehner model in which the stress response depends on an osmotic pressure and chemical potential. The latter drives fluid flow. Motivated by our interest in the rupture of blood clots, i.e. a fibrin gel,, we consider a relatively low volume of porous solid material that, in its reference state, is saturated with fluid. Both 2D and 3D analyses are carried out, where the latter model our experiments.

1. Results
A Mode I crack under both plane strain and 3D deformations is considered for both permeable and impermeable fluid boundary conditions. Through detailed finite element analyses, we investigate the crack-tip mechanical and chemical-potential fields. We show that the stress, strain, and chemical-potential fields at the crack tip display significant differences from solutions for linear poroelasticity.

An experiment on SENT specimens (thickness *d*) of a fibrin gel is shown in Fig. 1. Very large crack tip and overall specimen deformations occur before the gel ruptures associated with crack extension. In our experiments under Mode 1 loading, the crack always propagated directly ahead in its plane.

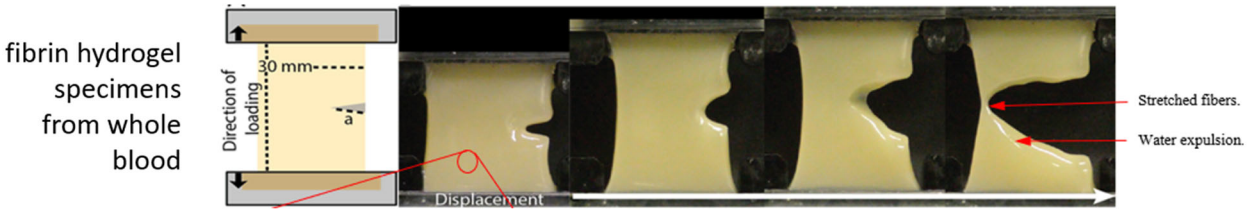

Figure 1. Low density fibrin gel (concentration 2.7 mg/ml) SENT specimen loaded to the onset of crack growth (from V. Tutwiler, J. Singh, R. I. Litvinov, J. L. Bassani, P. K. Purohit and J. W. Weisel, Rupture of blood clots: Mechanics and pathophysiology. *Science Advances*, 6(35), 2020).

The energy release rate with respect to crack advance is computed in terms of a "path" (volume) independent poroelastic integral $J^* = J_{mech} + J_{flow}$. Whereas J_{mech} can be determined directly from experiments, J_{flow}, as far as we know, must be determined from coupled calculations. For large time-dependent deformations, we demonstrate path independence of J^* every instant in transient problems. A key finding is that path independence results from strongly coupled mechanical and liquid-flow (chemical-potential) effects. An important observation is that the strong coupling is not limited to the crack- tip region. The effects of loading rate and solid volume fraction are investigated. With the toughness of the hydrogel defined as the critical energy release rate for crack propagation, we consider a micromechanically-based critical stretch criterion as a function of solid volume fraction.

The energy release rate G is given by the J^* integral defined for any volume V bounded by the surface S than encloses the crack tip:

$$G = J^* = \frac{1}{d}\left[\int_S \left(\Psi N_1 - P_{ij} N_j \frac{\partial x_i}{\partial X_1}\right) dS - \int_V \mu \frac{\partial C}{\partial X_1} dV\right] = J_{\text{mech}} + J_{\text{flow}}$$

where X_i and x_i are material points in the reference and deformed configuration, N_i is the outward unit normal to S, P_{ij} is the 1st Piola-Kirchhoff stress, Ψ is the free energy, C is the volume fraction of liquid, and μ is the chemical potential of liquid in the porous solid. The relative contributions of J_{mech} and J_{flow} depend on a number of factors including the gel permeability, loading rate, and fluid boundary conditions. An example from our finite element analyses for a compressible (porous) neo-Hookian solid containing liquid is plotted in Fig. 2 (K. Garyfallogiannis, P. K. Purohit, and J. L. Bassani, "Energy release rate for cracks in hydrogels undergoing finite deformations," *J. Mech. Phys. Solids,* vol. 167, p. 105009, 2022), where R_{J^*} denotes the radius of a circular cylinder centered at the crack tip for an initial crack length divided by specimen with a/w=0.2, overall displacement rate v=10^{-3} m/s, and permeability K=3.3×10^{-6} m/s. Note that the contribution of J_{flow} depends strongly on the boundary conditions, where the permeable case assumes that the specimen is embedded in liquid.

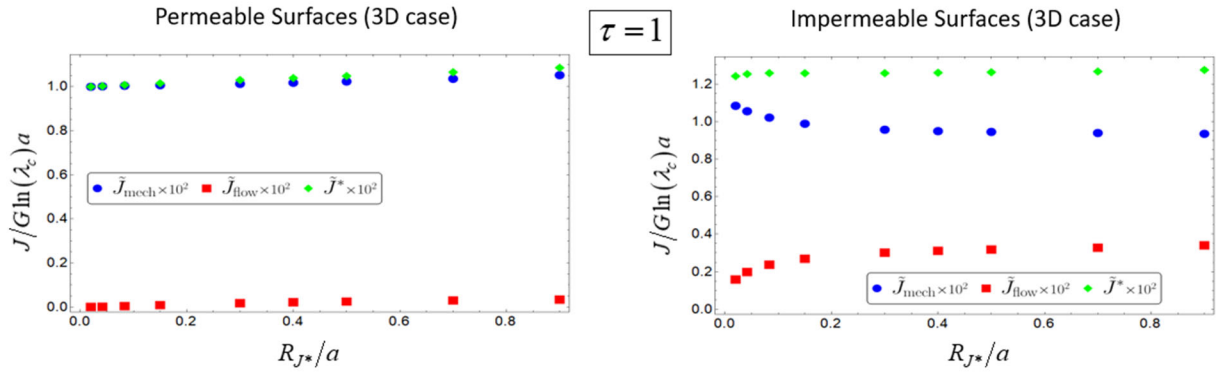

Figure 2. Contributions of J_{mech} and J_{flow} to the energy release rate J^* as a function of the radius of the cylinder enclosing the crack tip, where). τ=1 corresponds to an 50% overall extension of the specimen. The permeable case corresponds to a specimen emmersed in liquid whereas no liquid flows through the external surfaces of the specimen (including crack surfaces) in the impermeable case (from Garyfallogiannis et al., 2022).

Comparisons with the experiments on fibrin gels will also be presented. Since only J_{mech} can be directly determined experimentally, a coupled approach is adopted where a fiber-based material model is fitted to the overall force-extension data from experiments and J_{flow} is calculated from finite element analyses.

2. Conclusions

We have demonstrated the importance of fluid permeation in determining the energy release rate for hydrogel (poroelastic) materials. An open question is how to measure J_{flow} directly from experiments. Our results will be applicable to both synthetic and biological hydrogels and could inform their design in applications and also may be applicable to other poroelastic solids sufferinglarge deformations, such as, geological materials and battery materials.

Acknowledgements

Research reported in this publication was supported by the National Institutes of Health under award number R01 HL148227. The content is solely the responsibility of the authors and does not necessarily represent the official views of the National Institutes of Health.