CRACK TIP BLUNTING AND UNDULATING CRACK PROPAGATION IN GELATIN HYDROGELS

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Abstract

Gelatin hydrogels provide motifs for cell attachment, undergo large deformations, are tunable, and promising scaffold materials for tissue engineering. We characterized the fracture toughness of gelatin hydrogels, crosslinked with glutaraldehyde and methylglyoxal (MGO), using a pure shear notch test and characterize the processes ahead of the crack tip. We present novel processes that link crack growth to the curvature at the crack tip in gelatin hydrogels. We also quantified failures in these gelatin hydrogels using cavitation rheology and observed the growth and dynamics of bubbles. Based on these comparative results, we compare and discuss the advantages and disadvantages of these methods to quantify failures in gelatin hydrogels.

1. Material and methods

Bovine gelatin (Type-B; 0.05 gm/ml) was crosslinked with small amounts of glutaraldehyde (1% v/v) to stabilise the gel structure and cured at 4°C for 6 hours. To increase the crosslink density, these hydrogels were incubated in Methylglyoxal (20 mM) solution at 36°C for 4.5 hours. Pure shear notch tests were performed by stretching a notched hydrogel sheet (15 mm × 70 mm × 2 mm) uniaxially along Y-direction (Fig. 1b) at 0.01 mm/s until failure. A speckle pattern was created using a spray gun, and the strain fields were quantified using digital image correlation. The crack edges and curvatures at the crack tip were calculated using a custom-written MATLAB program. Cavitation experiments were performed by inserting blunt needles of varied gauges into the gel samples. A syringe pump, with a 20ml syringe, introduced an air bubble at 2000 µL/min. The hydrogel cavitation was recorded using a high-speed camera at 2000 fps (Phantom VEO-E 310L).

2. Results

Pure shear notch tests demonstrate that the crack propagates in a “stop-and-go” mechanism in all gelatin samples. MGO samples had higher threshold stress before crack propagation which suggests increased resistance for crack propagation due to the presence of higher crosslinks (Fig. 1a). The strain fields near the crack tip were lower in MGO samples as compared to controls. Fig. 1(b) and 1(c) show strain patterns when the crack first starts propagating in the Control and MGO samples, respectively. Variations in Eulerian strains along the width of a control sample show a clear delineation into four distinct strain domains (Fig. 1d). Strains were approximately zero in region A which shows the load-free configuration. E₉₉, Eₓₓ, and Eᵧᵧ were non-zero in region B which results in a complex strain state near the crack tip. Eₓᵧ and Eₓₓ were near zero in region C, whereas Eᵧᵧ was non-zero. Eᵧᵧ and Eᵧᵧ (not visible in 2D DIC) comprise a state of pure shear in this region. Deviations in region D from the pure shear state occur due to boundary conditions at the free edge.

Figure 1(e) shows the curvature at the crack tip with points marked in red, located towards the lowest values due to blunting, that represent crack propagation points labelled as P,Q,R, and S. These results show that the crack propagation in Control gelatin hydrogels occurs when the curvature is at the lowest value; we note similar patterns in MGO crosslinked gelatin gels. These results show the importance of crack tip curvatures during crack propagation and its correlation with increased crosslink density.

Cavitation experiments showed linear increase in the pressure within the bubbles with inflation time. The maximum pressure before failure varied inversely with needle diameter and was ~154% higher for 75 µm needle (p<0.001; n=12) in MGO gels as compared to Control gels. The fracture energy, computed using the critical pressure, was significantly higher (274%) in MGO samples than Control gels. High-speed
videography shows clear differences in the bubble growth dynamics: the bubble in MGO gels was smaller and penny-shaped as compared to the large and spherical bubble in control samples. The analytical formulation to calculate the fracture energy using cavitation, based on the linear elasticity assumption, merely gives a first-order estimate for a strain-stiffening hydrogel. The expression for fracture energy is unbounded for a large deformation material model, which is a limitation in cavitation rheology method. In contrast, the pure shear notch test uses an energy approach to calculate the fracture energy which circumvents the problem of finite strains and the nonlinearity in the material properties.

![Diagram](image.png)

**Fig.1:** (a) Representative Engg. Stress v/s stretch plot for a control and MGO hydrogel subjected to pure shear notch test. Normal strain fields, quantified using DIC in a Pure-Shear notch Test, are shown for (b) Control, and (c) MGO specimens. (d) The spatial variation of eualrian strains along the sample width in a control gelatin hydrogel. (e) Temporal variation of crack tip curvatures during the crack propagation in a pure shear notch test on a control sample.

### 3. Conclusions

MGO treatment increases the elastic modulus and fracture toughness of gelatin hydrogels. Mode-I failure in gelatin hydrogels using a pure shear notch test shows a “stop-and-go” mechanism of crack propagation. The stops during the crack propagation correlated with the crack tip curvatures and depend on the crosslink density. Higher critical pressures were required for cavitation in MGO hydrogel, and cavity growth dynamics varied significantly between the two gels. Cavitation rheology is useful for samples where geometric dimensions are restricted. On the other hand, the pure shear notch test is useful to visualise the crack tip processes, quantify the spatial strain fields and the fracture energy of the sample using an energy approach. The requirement for specific dimensions for pure shear tests limits use when testing biological samples that have irregular shapes and limited size.

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