ME – PhD Thesis Defense

Role of Crosslinks and Fiber Reinforcement in Hydrogel Mechanics

Mr. Anshul Shrivastava, Department of Mechanical Engineering, IISc Bengaluru

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ABSTRACT

Hydrogels are soft materials that exhibit nonlinear mechanical behavior under large deformations but can fail catastrophically, limiting their applications. Developing tough, biocompatible hydrogels with high strength and fracture resistance is a challenge in tissue engineering. In this thesis, we explored the role of covalent crosslinks in the fracture and mechanical properties of soft materials, including hydrogels and biological tissues. Our first study investigated the fracture toughness and material properties of differentially crosslinked gelatin hydrogels, exploring their potential as biomimetic scaffolds for tissue engineering. Bovine gelatin hydrogels were crosslinked using glutaraldehyde (Control) and methylglyoxal (MGO), a metabolic by-product during non-enzymatic collagen crosslinking. Monotonic compression tests showed that highly crosslinked MGO samples had a 96% higher modulus compared to Control hydrogels, with a modulus of 4.77 ± 0.73 kPa. The first-order Ogden model fits the data from both Control and MGO hydrogels well, while the Mooney-Rivlin and neo-Hookean hyperelastic models fit only the Control samples. Cavitation rheology was employed to quantify the maximum pressure for bubble failure in the hydrogels using blunt needles with varying inner radii. Critical pressures were used to quantify the fracture energies of the hydrogels. We next used pure shear notch tests and 2-D digital image correlation to characterize fractures and strain fields near the crack tip. A numerical method based on Taylor's series expansion measured the crack tip curvatures. Results showed that cracks in gelatin hydrogels underwent frequent arrests during propagation, with MGO hydrogels exhibiting 85% enhanced fracture toughness and a significantly higher number of stalls compared to the Control group. Crack initiations following stalls correlated with low tip curvatures in both groups. Mechanical stretching blunted the crack tip before propagation, with the degree of blunting independent of the cross-link density and elastic modulus. We also studied the influence of viscoelastic properties on bubble growth dynamics during cavitation in hydrogels. Polyacrylamide hydrogels with different viscous components but similar elastic moduli were prepared. Cavitation experiments showed a change in symmetry of the cavity shape from spherical in hydrogels with higher viscous components to ellipsoidal in hydrogels with lesser viscous components, suggesting the importance of viscous dissipation, which delays the onset of fracture. The critical pressure to cause cavity failure was significantly higher in more viscous hydrogels compared to the hydrogels having less viscous dissipation. Additionally, an ex-vivo model of goat cornea was used to investigate the effects of collagenase on collagen content, crosslinks, and microstructure. Results demonstrated that the material properties of the cornea decreased with collagen degradation, and collagen fiber waviness changed due to collagenase treatment, altering tissue properties and potentially influencing the mechanobiology and disease phenotype. These studies provide insights into the development of tougher hydrogels and discuss the comparative approaches for fracture characterization of hydrogels. These results also offer potential strategies for controlling cavitation dynamics by understanding the role of viscous contributions in hydrogel failure. Further, these results suggest that the altered load transfer from collagen fibers to cells in biological tissues such as the cornea can influence the mechanobiology and, consequently, the altered disease phenotype.

ABOUT THE SPEAKER

Anshul Shrivastava is a Prime minister's research fellow in the Mechanical Engineering department at the Indian Institute of Science, Bangalore. His research aims to investigate and gain insights into the mechanics of soft materials, such as hydrogels and biological tissues. He uses methods in experimental techniques and mathematical modeling to examine the mechanical and fracture behavior of these materials. His research further includes the biomechanical assessment of biological tissues, such as the cornea, under diverse pathological conditions.