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Multi-scale Multi-mechanism Toughening of Hydrogels¹

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Hydrogels are widely used as scaffolds for tissue engineering, vehicles for drug delivery, actuators for optics and fluidics, and model extracellular matrices for biological studies. The scope of hydrogel applications, however, is often severely limited by their mechanical properties. Inspired by the mechanics and hierarchical structures of tough biological tissues, we propose that a general principle for the design of tough hydrogels is to implement two mechanisms for dissipating mechanical energy and maintaining high elasticity in hydrogels. A particularly promising strategy for the design is to integrate multiple pairs of mechanisms across multiple length scales into a hydrogel. We develop a multiscale theoretical framework to quantitatively guide the design of tough hydrogels. On the network level, we have developed micro-physical models to characterize the evolution of polymer networks under deformation. On the continuum level, we have implemented constitutive laws formulated from the network-level models into a coupled cohesive-zone and Mullins-effect model to quantitatively predict crack propagation and fracture toughness of hydrogels. Guided by the design principle and quantitative model, we will demonstrate a set of new hydrogels, based on diverse types of polymers, yet can achieve extremely high toughness superior to their natural counterparts such as cartilages.

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