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Responsive Gel-Gel Phase Transitions in Artificially Engineered Protein Hydrogels

B.D. OLSEN, Massachusetts Institute of Technology

Artificially engineered protein hydrogels provide an attractive platform for biomedical materials due to their similarity to components of the native extracellular matrix. Engineering responsive transitions between shear-thinning and tough gel phases in these materials could potentially enable gels that are both shear-thinning and tough to be produced as novel injectable biomaterials. To engineer a gel with such transitions, a triblock copolymer with thermoresponsive polymer endblocks and an artificially engineered protein gel midblock is designed. Temperature is used to trigger a transition from a single network protein hydrogel phase to a double network phase with both protein and block copolymer networks present at different length scales. The thermodynamics of network formation and resulting structural changes are established using small-angle scattering, birefringence, and dynamic scanning calorimetry. The formation of the second network is shown to produce a large, nonlinear increase in the elastic modulus as well as enhancements in creep compliance and toughness. Although the gels show yielding behavior in both the single and double network regimes, a qualitative change in the deformation mechanism is observed due to the structural changes.