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Responsive Gelation in Physical Double Network Hydrogels from Artificial Protein Polymers B.D. OLSEN, M.J. GLASSMAN, MIT — Artificial protein polymers with responsively associating groups on two different length scales are engineered in order to form physical double networks with potential application as shear-thinning hydrogels that may be toughened after injection. Gel-forming molecules are prepared by conjugating poly(N-isopropylacrylamide) (PNIPAM) at both ends of an artificial protein polymer to form PNIPAM-protein-PNIPAM triblock copolymers. Aggregation of the polymer endblocks forms a longer length scale network, while associating coiled-coil groups within the protein midblock form a shorter length scale network. At low temperatures where the coiled-coil domains are physically crosslinked but the copolymer endblocks are soluble, the materials form soft shear-thinning hydrogels. Elevating the temperature results in self-assembly of the second network, as manifest by stiffening of the gels. The structure of the materials is characterized using light scattering, X-ray scattering, and microscopy. Kinetics of the second network formation are characterized by linear oscillatory shear rheology, and nonlinear rheology is used to characterize the effect of the second network on the yield stress in these gels.

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