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Molecular origins of reinforcement in responsively nanostructured, shear thinning double network hydrogels MATTHEW GLASSMAN, Massachusetts Institute of Technology, JACQUELINE CHAN, California Institute of Technology, BRADLEY OLSEN, Massachusetts Institute of Technology — Triblock copolymers containing associative protein midblocks and thermoresponsive endblocks have recently been shown to form reinforceable, nanostructured hydrogels. Triggered self-assembly of orthogonal physical crosslinks causes a reversible transition from a shear thinning material at low temperatures to a toughened state at high temperatures with resistance to creep, erosion, and failure in uniaxial compression. In this study, properties of the individual networks were varied to investigate the relationships among association density in the protein network, nanostructure formation, and ultimate mechanical reinforcement that could be realized in this double network architecture. Through a broad survey of materials, large changes in static and dynamic mechanical properties were identified, some leading to a 14-fold increase in plateau modulus and a decrease in creep compliance by more than two orders of magnitude over the range from 5-50^{o}C. Detailed investigation of the structure and relaxation behavior of the underlying network of micelles with associative coronae reveals important parameter constraints for achieving high performance in these double network gels.

Matthew Glassman
MIT

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