Abstract Submitted
for the MAR14 Meeting of
The American Physical Society

Oxidatively Responsive Chain Extension to Topologically Entangle Engineered Protein Hydrogels BRADLEY OLSEN, SHENGCHANG TANG, MATTHEW GLASSMAN, SHUAILI LI, SIMONA SOCRATE, Massachusetts Institute of Technology — Hydrogels with increased toughness and extensibility have attracted a great deal of interest as mimics for natural tissues in biomedical applications. Artificial protein polymers provide particularly attractive systems for these applications due to their similarity to the chemistry of the natural extracellular matrix. Here, we show that entanglements can be incorporated into physically associating protein gels using simple oxidative chain extension chemistries, producing hydrogels with multiple time and length scales of relaxation. These oxidative chemistries follow the Jacobson-Stockmayer theory and are fully reversible, enabling responsive formation of entanglements within a material. The entangled protein gels demonstrate extensibility up to engineering strains of greater than 3,000%, a toughness of 65,000 J/mˆ3, and significant reductions in creep compliance and increases in elastic recovery. The rheology of the materials is compared to sticky reptation theory as a function of gel concentration, providing insights into the effect of network structure on different modes of molecular relaxation.

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Date submitted: 14 Nov 2013 Electronic form version 1.4