

Abstract Submitted
for the MAR14 Meeting of
The American Physical Society

Chain-configuration dependent rheological properties in transient networks¹ MICHELLE SING, Massachusetts Institute of Technology, ZHEN-GANG WANG, California Institute of Technology, GARETH MCKINLEY, BRADLEY OLSEN, Massachusetts Institute of Technology — Complex associative networks capable of shear thinning followed by recovery on the order of seconds are of interest as injectable biomaterials. However, there is a limited understanding of the molecular mechanisms that contribute to rheological properties such as the network's yield stress and rate of self-healing. Here we present a transient network theory for associative physical gels arising from the chemical kinetic form of the Smoluchowski Equation capable of modeling the full chain end-to-end distance distribution while tracking the fraction of looped, bridged, and free chain configurations in the gel. By varying the equilibrium association rate relative to the material relaxation time, we are able to track the evolution of loop and bridge chain fraction as the system undergoes stress instabilities. We have evidence that these instabilities result from non-monotonic trends in loop and bridge chain fraction when the end group association rate is high relative to the dissociation rate. This behavior provides insight into the complex kinetic interactions responsible for certain mechanical behaviors while serving as a valuable predictive tool for gel design.

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Date submitted: 15 Nov 2013

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