Abstract Submitted
for the MAR17 Meeting of
The American Physical Society

Enthalpic and Entropic Competition in Blends of Self-Suspended Hairy Nanoparticles SNEHASHIS CHOUDHURY, AKANKSHA AGRAWAL, LYNDEN ARCHER, Cornell University — Self-suspended hairy nanoparticles, where polymer chains are grafted onto nanoparticles, have attracted significant recent attention. These materials have been reported to manifest several interesting phenomena like thermal jamming, slowing-down of polymer chain dynamics, as well as small-strain stress overshoots during start-up of steady shear. The entropic penalty on tethered polymers produced by the requirement that they fill the space between the nanoparticle cores explain most of these behaviors. Here, we show that the entropic attraction between tethered polymer chains can be manipulated in mixtures of hairy nanoparticles using different polymer chemistry to design materials with unusual characteristics. Specifically, the degree of interpenetration of polymer chains can be controlled by tuning their interaction parameter (χ). For SiO_2-PEG/SiO_2-PMMA blends, oscillatory rheological measurements show that the plateau modulus and yielding energy are significantly increased, while an opposite effect is seen with SiO_2-PEG/SiO_2-PI blends. More subtle effects of this enthalpy-entropy competition are well captured in Dielectric Spectroscopy measurements and SAXS experiments that can be used to quantify the degree of stretch and interdigitation of polymer chains.

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Date submitted: 11 Nov 2016 Electronic form version 1.4