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Tethered Nanoparticle−Polymer Composites: Phase behavior and rheology[1] RAHUL MANGAL, LYNDEN A. ARCHER, Cornell University — Polymer nanocomposites with particle radius (a) approaching the radius of gyration (R_g) of entangled host polymer have been reported to exhibit an unusual negative reinforcement effect, which leads to an anomalous reduction in relative an anomalous reduction in relative viscosity at low particle loadings (φ). This so-called Non-Einsteinian flow behavior is understood to be sensitive to the dispersion state of particles in host polymer. We studied suspensions of SiO_2 nanoparticles tethered with polethylene glycol (PEG) in polymethylmethacralate (PMMA) with molecular weights (Mw) from 17 KDa to 280 KDa. Due to strong enthalpic interactions between PEG and PMMA (χ = -0.65), nanoparticles are expected to be well-dispersed, independent of Mw of PMMA. Using small angle x-ray scattering measurements we show that the phase stability of suspensions depends on Mw of the tethered PEG, host PMMA, and φ. Particles functionalized with low molecular weight PEG aggregate at low φ, but disperse at high φ. In contrast, nanoparticles functionalized with higher molecular weight PEG are well dispersed for host chain lengths (P) to tethered chain length (N), (P/N), is as high as 160. The stability boundary of these suspensions extends well beyond expectations for nanocomposites based on tethered PEG chains suspended in PEG. Through in-depth analysis of rheology and x-ray photon correlation spectra we explore the fundamental origins of non-Einsteinian flow behavior.

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