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Aggregation, Steric Stabilization, Bridging and Miscibility of Polymer Nanocomposites

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Microscopic liquid state theory has been employed to study the potential of mean force (PMF), statistical structure, and phase separation of spherical nanoparticles in a dense polymer melt over a wide range of interfacial chemistry, chain length, and filler size and volume fraction conditions. As interfacial cohesion strength increases the nanoparticle organization evolves from contact depletion aggregation, to well dispersed behavior associated with a thermodynamically stable polymer coating, to polymer-mediated bridging of a variable degree of tightness. Near linear scaling of the PMF with the particle/monomer diameter ratio is found, and the spatial range of the interfacial attraction is important in determining nanoparticle organization. Spinodal demixing calculations predict an entropy-driven fluid-fluid phase separation for weak interfacial attractions, and an enthalpically driven network or complex formation type of phase separation in the strong cohesion regime. A miscibility window exists at intermediate interfacial attraction strengths which systematically narrows, and is ultimately destroyed, as particle size and/or direct filler-filler van der Waals attractions increase. The length-scale dependent real space statistical structure is quantified via calculations of the polymer and filler intermolecular pair correlation functions and partial scattering structure factors. At high filler volume fractions interference between the polymer organization near nanoparticle surfaces induces significant changes of filler packing. The presence of bound polymer layers in miscible nanocomposites results in microphase-separation-like features in the small angle collective polymer structure factor. Implications of the theoretical results for the design of thermodynamically and/or kinetically well-dispersed polymer nanocomposites, and the formation of nonequilibrium networks, will be discussed. The theory has also been generalized to treat the consequences of soft intermolecular repulsions, and nonspherical fillers including rod, disk and compact molecule-like shapes.