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Multiscale structure, interfacial cohesion, adsorbed layers, miscibility and properties in dense polymer-particle mixtures

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A major goal in polymer nanocomposite research is to understand and predict how the chemical and physical nature of individual polymers and nanoparticles, and thermodynamic state (temperature, composition, solvent dilution, filler loading), determine bulk assembly, miscibility and properties. Microscopic PRISM theory provides a route to this goal for equilibrium disordered mixtures. A major prediction is that by manipulating the net polymer-particle interfacial attraction, miscibility is realizable via the formation of thin thermodynamically stable adsorbed layers, which, however, are destroyed by entropic depletion and bridging attraction effects if interface cohesion is too weak or strong, respectively. This and related issues are quantitatively explored for miscible mixtures of hydrocarbon polymers, silica nanospheres, and solvent using x-ray scattering, neutron scattering and rheology. Under melt conditions, quantitative agreement between theory and silica scattering experiments is achieved under both steric stabilization and weak depletion conditions. Using contrast matching neutron scattering to characterize the collective structure factors of polymers, particles and their interface, the existence and size of adsorbed polymer layers, and their consequences on microstructure, is determined. Failure of the incompressible RPA, accuracy of PRISM theory, the nm thickness of adsorbed layers, and qualitative sensitivity of the bulk modulus to interfacial cohesion and particle size are demonstrated for concentrated PEO-silica-ethanol nanocomposites. Temperature-dependent complexity is discovered when water is the solvent, and nonequilibrium effects emerge for adsorbing entangled polymers that strongly impact structure. By varying polymer chemistry, the effect of polymer-particle attraction on the intrinsic viscosity is explored with striking non-classical effects observed. This work was performed in collaboration with S.Y.Kim, L.M.Hall, C.Zukoski and B.Anderson.