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Control of Nanoparticle Organization in Thin Homopolymer Films

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The morphological structure of mixtures of homopolymers with chain-grafted nanoparticles is determined by competing interactions between the nanoparticle cores, the free host chains and the grafted chains. In the bulk, when the nanoparticle grafting density is low, the phase behavior is largely determined by a competition between attractive nanoparticle core-nanoparticle core interactions, mediated by the chains grafted to the surface. At high grafting densities, the entropic brush layer/free host chain interactions are dominant, leading to miscibility or to microscopic/macroscopic phase separation. Thin film mixtures are thermodynamically less stable than their bulk analogs due to the preferential attraction of grafted nanoparticles to the external interfaces. The preferential attraction of the nanoparticles to the interfaces is driven by factors that include: entropic gains of the grafted nanoparticles and linear host chains; van der Waals interactions between the nanoparticles and the interfaces. If the grafted chains and host chains are of dissimilar chemical structure, then the nanoparticles exhibit a tendency to segregate to the free surface, provided its grafts possess a lower surface energy than the host chains. Consequences of these interactions on the overall nanoparticle organization in thin homopolymer films will be discussed.