Entropy-Driven Assembly of Polymer-Grafted Nanoparticles within Emulsion-Evaporative Block Copolymer Particles

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The spatial alignment of inorganic nanoparticles (NPs) within the polymer matrix is of great importance due to its potential for building novel hybrid materials. Herein, we demonstrate the spatial distribution tuning of polystyrene-grafted Au NPs (Au@PS) within lamella forming polystyrene-block-poly(4-vinylpyridine) (PS-b-P4VP) particles. Distributions of Au@PS showed the dependence on various entropy-related parameters, including the molecular weight ratio between the polymer matrix to the ligand, the core size of Au NPs, the grafting density of Au@PS. The hybrid particles with weak entropic interactions between Au@PS and polymer matrix showed a "crusted onion" structure with Au@PS forming hexagonal packing on the particle surface. In contrast, strong entropic interactions led to the formation of “alternate-layered onion” in which Au@PS stacked within inner PS domains in hybrid particles. For hybrid particles with intermediate entropic interactions, Au@PS were segregated in central regions of particles, which is designated as “seeded onion”. The phenomena will be explained by considering interdigitations between matrix chains and PS ligands.