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Structured Nanocomposites: Organization of Particles Templated in Self-Assembled PEO-PPO-PEO Mesophases

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The design of materials with tailored properties and function requires control over their structure in the nanometer scale. It is advantageous to many engineering applications that this is carried out using self-assembly processes that occur at reasonable concentrations ($\sim 10^{20}$ particles/L) and short timescales (ns – μs). In this work, we have developed a novel approach using an ordered template to control the nanoscale structure of materials that would not otherwise order in solution. We use close-packed cubic and cylindrical mesophases of a thermoreversible block copolymer (PEO-PPO-PEO) to impart spatial order on dispersed nanoparticles. The thermoreversible nature of the template allows for the dispersion of particles synthesized outside the template. This feature extends the applicability of this templating method to many particle-polymer systems and also permits a systematic evaluation of the impact of design parameters on the structure and mechanical properties of the nanocomposites. The approach is extremely robust and we have successfully templated solutions of silica and gold inorganic nanoparticles as well as a series of proteins, which act as organic nanoparticles in our system. The influence of relative size (particle to template sites), relative concentration, temperature and shear are experimentally determined using small angle neutron scattering (SANS) and rheology. SANS with contrast variation is used to characterize the structure of the polymer mesophase and the templated particles in a nanocomposite independently. SANS experiments also demonstrate that shear can be used to align the nanocomposites into single-crystal macro-domains; the first demonstration of the formation of single-crystal nanoparticle superlattices. The outcome of this work serves as a basis for designing new soft nanocomposite materials.