MAR17-2016-020690

Abstract for an Invited Paper
for the MAR17 Meeting of
the American Physical Society

Tunable Multiscale Nanoparticle Ordering by Polymer Crystallization

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Achieving the controlled, multiscale assembly of nanoparticles (NPs), which is known to underpin the unusual mechanical properties of some biomaterials, such as Nacre, remains a major challenge in nanoscience. Here we achieve such Nacre-inspired hierarchical NP ordering in a one-pot approach by leveraging the kinetics of polymer crystallization, which yields a multiscale structure spanning lamellae [(10 nm)], fibrils (m) and spherulites (mm). NPs are engulfed by the growing crystals, ordered into layers in the interlamellar zone [spacing of (10-100 nm)], or assembled into fractal objects at the interfibrillar scale, (1-10 m), with the relative NP populations in this hierarchy readily manipulated by crystallization speed. Adding NPs always increases the Young's modulus, but the effects of multiscale ordering are nearly an order of magnitude larger than those for a randomly mixed state. Since fracture toughness remains practically unaffected, this assembly strategy allows us to create high strength materials that retain the attractive high toughness and low density of polymers.