MAR10-2009-001402

Abstract for an Invited Paper
for the MAR10 Meeting of
the American Physical Society

Crystallization of Unbranched Polyethylene Confined within Block Copolymer Mesophases[1]

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Confinement of polymer crystallites between or within block copolymer microdomains provides an exceptional opportunity for their manipulation and study. Much work over the past three decades has employed hydrogenated low-vinyl polybutadiene (hPBd) as the crystallizable block, whose limited crystal thickness and moderate crystallinity limit the extent of confinement which the microdomains can impose on the crystals. This talk will report the morphology and orientation of the crystals which form within the microdomain structures of diblock copolymers of linear polyethylene and glassy poly(vinylcyclohexane), LPE/PVCH, across the spherical, cylindrical, and lamellar morphologies. Synthesized by ring-opening methathesis polymerization followed by catalytic hydrogenation, LPE is entirely unbranched, and thus capable of producing thick crystals and achieving a high degree of crystallinity. Compared with short-branched polyethylene (such as hPBd), confinement of LPE within spheres, within cylinders, or between PVCH cylinders directly limits the crystal thickness and thereby the crystal melting point. Conversely, crystals formed within LPE lamellae are stacked orthogonally to the LPE/PVCH microdomain layering, so there is no direct limitation imposed on crystal thickness by confinement. As with LPE homopolymer, LPE crystals within lamellae thicken when annealed below the melting point, ultimately forming crystals whose thickness is significantly larger than their lateral extent, set by the bounding PVCH layers. The ribbon-like crystals which form within LPE cylinders or lamellae have a strong orientational coupling to the microdomains; prealignment of the cylindrical or lamellar mesophase by extensional flow yields macroscopic specimens with pronounced *b*-axial and *a*-axial orientations, respectively, after subsequent quiescent crystallization, complementing the usual *c*-axial texture produced by fiber spinning.

[1] with Sasha B. Myers; support from the NSF Polymers Program.