Melt and Solid-State Structures of Polydisperse Polyolefin Block Copolymers

RICHARD REGISTER, SHENG LI, Princeton University — Recent developments in coordinative chain transfer polymerization have enabled the synthesis of ethylene-co-octene block copolymers, where the blocks are either crystallizable (an ethylene-co-octene random copolymer block with low octene content) or amorphous (analogous block with high octene content). With a suitable choice of catalyst type(s) and reactor train configuration, accessible chain architectures include diblock, where each block ideally has the most-probable distribution of chain lengths, and multiblock, where both the individual blocks and the number of blocks per chain follow the most-probable distribution. With a sufficiently large interblock octene differential, block copolymers of both architectures, containing roughly equal masses of the two types of block, self-assemble in the melt into well-ordered lamellar structures, despite the large polydispersity. Interblock mixing, induced by the modest Flory interaction parameter and the broad distribution of block lengths, yields an enormous domain spacing (> 100 nm) despite the relatively low average block molecular weights (< 50 kg/mol). Extensive interblock mixing also allows the polyethylene crystals to grow freely and nearly isotropically across the domain interfaces, while preserving the domain structure present in the melt; in the solid state, the optical and x-ray contrasts between dissimilar domains are greatly enhanced due to their different levels of crystallinity. (Work conducted in collaboration with Jeffrey Weinhold, Philip Hustad, and Brian Landes of Dow Chemical Core R&D.)

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Richard Register
Princeton University

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