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John H. Dillon Medal Lecture: Molecular Heterogeneity in Block Copolymer Self-Assembly

MAHESH MAHANTHAPPA, Department of Chemistry, University of Wisconsin-Madison

Narrow molecular weight dispersity in block copolymers has long been considered necessary for well-defined, periodic structure formation, by analogy to various crystallization processes. Consequently, much attention has focused on narrow dispersity copolymers derived from controlled and “living” polymerization techniques. However, these methods restrict the palette of functional monomers amenable to block copolymerization, thus constraining the physical and chemical properties of the resulting materials. New polymer syntheses enable access to a “Pandora’s Box” of block copolymers with unusual chemical functionalities and useful physical properties, at the expense of introducing significant segmental dispersities into the resulting copolymers. The development and use of these functional materials requires basic understanding of the physical implications of continuous segmental dispersity on block copolymer phase behavior. Our work aims to understand the physical principles underlying polydisperse ABA-type triblock copolymer self-assembly, in order to transform segmental dispersity into a predictable and useful tool for manipulating block copolymer morphology. We have systematically demonstrated that mid-segment dispersity in ABA triblock copolymers does not preclude the formation of classical, structurally periodic, microphase separated morphologies. Mid-segment dispersity instead shifts the locations of the composition-dependent phase windows, dilates the microdomains, and unexpectedly stabilizes the microphase separated ABA triblock copolymer melts. Studies of three different polydisperse copolymer systems have provided general insights into the consequences of chain length heterogeneity on block copolymer self-assembly.