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Gradient Copolymers Yield Uniquely Broad Glass Transition Temperatures in Comparison with Block Copolymers and Polymer Blends JUNGKI KIM, MICHELLE M. MOK, CHRISTOPHER L.H. WONG, ROBERT W. SANDOVAL, JOHN M. TORKEKELSON, Northwestern University, Evanston, Illinois 60208 — Gradient copolymers, which can be made by controlled radical polymerization or ring-opening metathesis polymerization, are distinct from random and block copolymer because of the gradient in comonomer composition along the copolymer backbone. As a result of this gradient along the chain, in the ordered lamellar state gradient copolymers are believed to exhibit a sinusoidal composition profile that is distinct from the “square-wave” composition profile observed in ordered lamellar block copolymers. This difference in the manner in which the local composition varies in the ordered state leads to dramatic differences in the glass transition behavior of gradient copolymers and block copolymers and similarly between gradient copolymers and polymer blends. Five gradient copolymer systems have been examined allowing for study of the effects of the enthalpic incompatibility of the comonomer units, the T_g differences among the homopolymers, and hydrogen bonding effects leading to random copolymers exhibiting higher T_gs than those found in block copolymers. We show via differential scanning calorimetry and dynamic mechanical analysis that single, continuous T_g breadths as large as 70-100 K are possible in gradient copolymers.

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