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**Block Copolymers with Composite Crystalline-Glassy Hard Domains Formed from Single-Phase Melts** RICHARD REGISTER, JOHN BISHOP, Princeton University — Thermoplastic elastomers (TPEs) are typically ABA triblock copolymers made up of a rubbery midblock (B) and glassy endblocks (A). If instead the hard segments were crystalline, microphase separation could be driven by crystallization from a lower-viscosity single-phase melt. We use living ring-opening metathesis polymerization and subsequent hydrogenation to synthesize TPEs consisting of crystalline hydrogenated polynorbornene (hPN), rubbery hydrogenated poly(5-hexylnorbornene) (hPHN), and glassy hydrogenated polymethyltetracyclododecene (hPMTD). By suitable control of the block lengths, we achieve a single-phase melt in a symmetric pentablock TPE with the architecture hPN-hPMTD-hPHN-hPMTD-hPN, where microphase separation is driven by crystallization. The single-phase melt is confirmed by both a featureless small-angle x-ray scattering pattern, and a relatively low and Newtonian viscosity at modest shear rates. When the hPN block crystallizes, the attached hPMTD block forms a glassy layer surrounding the crystals, limiting their lateral growth. The resulting pentablock TPE shows excellent solvent resistance at room temperature due to the hPN crystallinity, while the composite crystalline/glassy hard domains impart strain-hardening behavior to the material, with only modest unrecoverable deformation.

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