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Soft Semicrystalline Thermoplastic Elastomers by Arrested Crystallization ADAM BURNS, RICHARD REGISTER, Princeton University — Thermoplastic elastomers (TPEs) marry the solid-state behavior of vulcanized rubbers with the melt processability of thermoplastics. Archetypal soft TPEs consist of triblock copolymers comprising a rubbery mid-block flanked by two identical glassy end-blocks. Incorporating crystalline blocks into TPEs can confer solvent resistance as well as reduce the processing costs by giving access to single-phase melts. However, simply substituting crystalline for glassy end-blocks dramatically degrades the solid-state mechanical properties, particularly at large strains. We seek to integrate the benefits of crystallinity into TPEs, while maintaining the desired mechanical properties, using the block architecture: crystalline-glassy-rubbery-glassycrystalline. Methods have been developed to synthesize highly symmetric, narrowdistribution block copolymers with this architecture using anionic polymerization of butadiene, styrene, and isoprene followed by hydrogenation. Judicious choices of block molecular weights indeed yield homogeneous melts above the melting point of the crystalline component. Upon cooling, crystallization—rather than interblock repulsion—establishes the solid-state microstructure which physically crosslinks the rubbery mid-block, ultimately conferring elasticity. Subsequent vitrification of the adjacent glassy blocks arrests the growth of the crystallites, and protects them from yielding under applied load. As a result, our materials show low initial moduli, strain hardening, and high extensibility, typical of commercial TPEs.

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