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Influence of Deformation on Irreversible and Reversible Crystallization of Poly(ethylene-co-1-octene)

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The effect of uniaxial deformation and subsequent relaxation at ambient temperature on irreversible and reversible crystallization of homogeneous poly(ethylene-co-1-octene) with 38 m-% 1-octene, melt-crystallized at 10 K min^{-1} , has been explored by calorimetry, X-ray scattering, and Fourier-transformed infrared spectroscopy. At 298 K, the enthalpy-based crystallinity of annealed specimens increases irreversibly by stress-induced crystallization from initially 15% to a maximum of, at least, 19% when a permanent set of more than 200% is attained. The crystallinity increases by formation of crystals of pseudo-hexagonal structure at the expense of the amorphous polymer, and as a result of destruction of orthorhombic crystals. The stress-induced increase of crystallinity is accompanied by an increase in the apparent specific heat capacity from $2.44 \text{ J g}^{-1} \text{ K}^{-1}$ to about $2.59 \text{ J g}^{-1} \text{ K}^{-1}$, which corresponds to an increase of the total reversibility of crystallization from, at least, $0.10\% \text{ K}^{-1}$ to $0.17\% \text{ K}^{-1}$. The specific reversibility calculated for 100% crystallinity increases from $0.67\% \text{ K}^{-1}$ to $0.89\% \text{ K}^{-1}$, and points to a changed local equilibrium at the interface between crystal and amorphous phase. The deformation results in typical changes of the phase structure and of the crystal morphology, which involve orientation and destruction of crystals, and the formation of fibrils. The effect of the decrease of the entropy of the strained melt on the reversibility of crystallization and melting is discussed.

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