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Tough Block Copolymer Organogels and Elastomers as Short Fiber Composites[1]

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The origins of the exceptional toughness and elastomeric properties of gels and elastomers from block copolymers with semicrystalline syndiotactic polypropylene blocks will be discussed. Using synchrotron X-radiation small angle (SAXS) and wide angle X-ray scattering (WAXS) experiments were simultaneously performed during step cycle tensile deformation of these elastomers and gels. From these results the toughness can be attributed to the formation, orientation and elongation of the crystalline fibrils along the tensile direction. The true stress and true strain ε_H during each cycle were recorded, including the true strain at zero load $\varepsilon_{H,p}$ after each cycle that resulted from the plastic deformation of the sPP crystals in the gel or elastomer. The initial Young's modulus E_{init} and maximum tangent modulus E_{max} in each cycle undergo dramatic changes as a function of $\varepsilon_{H,p}$, with E_{init} decreasing for $\varepsilon_{H,p} \leq 0.1$ and then increasing slowly as $\varepsilon_{H,p}$ increases to 1 while E_{max}increases rapidly over the entire range of $\varepsilon_{H,p}$ resulting in a ratio of $E_{max}/E_{init} > 100$ to 1000 at the highest maximum (nominal) strain. Based on SAXS patterns from the deformed and relaxed gels, as well as on previous results on deformation of semicrystalline random copolymers by Strobl and coworkers, we propose that the initial decrease in E_{init} and increase in E_{max} with $\varepsilon_{H,p}$ are due to a breakup of the network of the original sPP crystal lamellae and the conversion of the sPP lamellae into fibrils whose aspect ratio increases with further plastic deformation, respectively. The gel elastic properties can be understood quantitatively as those of a short fiber composite with a highly deformable matrix. At zero stress the random copolymer midblock chains that connect the fibrils cause these to make all angles to the tensile axis (low E_{init}), while at the maximum strain the stiff, crystalline sPP fibrils align with the tensile axis producing a strong, relatively stiff gel.The evolution of the crystalline structure during deformation is confirmed by WAXS and FTIR measurements.

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