

Evolution of Lamellar Structures in Liquid Crystalline and Semicrystalline Polymers Investigated using SAXS Patterns

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Small-angle X-ray Scattering (SAXS) patterns from polymers are useful for studying the organization of the crystalline lamellae and the amorphous regions that influence polymer properties. Despite a long history of observation of SAXS patterns, there is no clear understanding of the underlying structural features, the linkages between the amorphous and the crystalline domains that determines the evolution of the observed patterns, and of the morphological changes during macroscopic deformation. This is because it is not possible to derive a unique structure from a diffraction pattern with just a one or two reflections. In this paper we generate simple structural models using lamellar stacks and 2-D Fourier transform them to simulate the SAXS patterns. To understand how these patterns originate and evolve during deformation, the three distinct SAXS patterns (2-point 'bar', 4-point 'eyebrow', and 4-point 'butterfly'), were generated using microstructural models, and their detailed intensity profiles were compared with the experimental data from poly(ethylene-*co*-octene). It is important to recognize that agreement with the experimental diffraction data proves that the model structure is possible but does not prove it is correct, as such structural solutions are not unique. At lamellar length scales, three modes of deformations have been recognized, chain slip within the lamellae, interlamellar shear, and, whole body rotation of the lamellar stacks. Our simulations show that when stack orientation and chain slip act to rotate lamellae in the same sense, the result is an 'eyebrow' pattern. Opposite senses give a 'butterfly' pattern. Experimental patterns often have reflections where the peak position traces out an ellipse. Adding moderate amounts of rotational disorder to the models produces this effect without any elliptical correlations or deformed lattice in the material. This observation enabled us to explain the deformation and the relaxation behavior of the lamellar structures in terms of the coupling between the amorphous and crystalline regions.