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Higher order structure of polymer spherulites

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結晶性高分子の一般的な形態である球晶は、それを構成する微結晶（ラメラ晶）が分岐・衝突を繰り返してできた複雑な分岐網となっている。偏光顕微鏡下で特徴的な模様が見られることは古くから知られているが、この形態の定量化はされていない。今回我々は、偏光顕微鏡像の画像解析からこれを定量化しその性質を議論する。

Introduction

Spherulite is a typical morphology of polymers crystallized from the melt. The structure consists of thin lamellar crystallites and amorphous stacks filled between the lamellae. The lamellar crystals undergo branching repeatedly and stop growing on the occasion of collision with other crystallites. The branching and collision evolve a complex network structure of the higher order structure in the spherulites. We examined the complexity in a quantitative manner.

Experimental

Figure 1 shows the typical images of spherulites of poly(butene-1). The images are obtained by the superposition of two images taken by polarizing optical microscopy (POM) with the rotation angle of 45 degrees of the pair of polarizer and analyzer in the position of crossed nicols. The intensities of the images represent the optical directions of crystallites with respect to the radial axis. The white areas are elongated in the radial directions of the spherulite and represent the domains keeping the optical directions. The areas become longer as crystallization temperature is higher. In order to examine the structure in a quantitative way, we calculate the auto-correlation function of the intensity profiles along the radial and tangential directions.

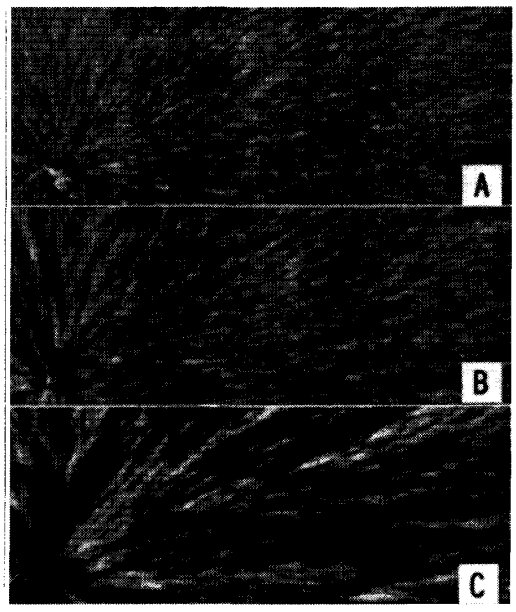

Figure1: POM images of the spherulites of poly(butene-1). Width of the images is 200 μ m. Crystallization temperatures are A : 85℃, B : 90℃, and C : 96℃.

Results and Discussions

When a lamellar crystal undergoes branching, the child lamellae may deviate from the parent one in a random fashion. The sequence of branching resembles a random walk of the direction of the lamellar crystals with respect to the radial axis. If it is the case, the auto-correlation function of the intensity profiles will be represented by a single exponential function. We have assumed that the auto-correlation function for the tangential direction is also represented by a single exponential function. Then, the characteristic lengths of the domains in the radial and tangential directions have been determined from the fittings.

Figure 2 shows the characteristic lengths in the (a) radial and (b) tangential directions plotted against crystallization temperatures. As seen in Fig. 1, the characteristic lengths in both directions become longer as the crystallization temperature becomes higher. The dependence of the radial length is stronger than that of the tangential one, and hence the domains keeping the optical direction become thinner at higher temperatures.

For the thinner domains at higher temperatures, we will be able to think of various possibilities. One possibility is smaller branching angle with the increase in the crystallization temperature, so that the network of the lamellae becomes thinner. Twisting angle may also become smaller, so that the radial characteristic length becomes longer. In order to determine the determining factors, we need to examine the thickness and width of those lamellar crystals, which compose the spherulite.

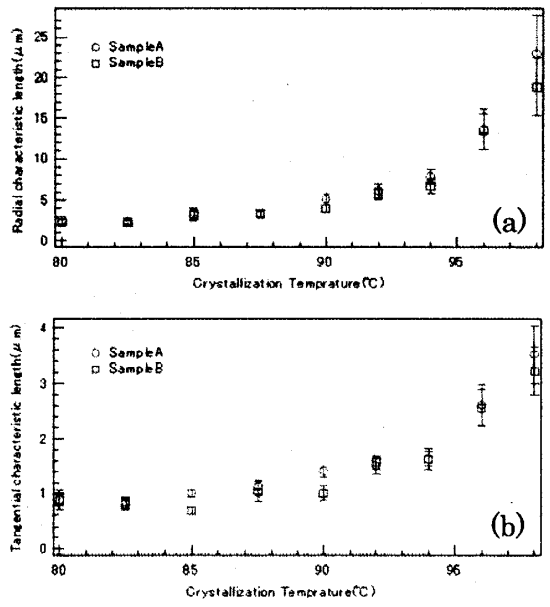

Figure 2: Characteristic lengths in the (a) radial and (b) tangential directions plotted against crystallization temperatures.