Host-guest co-amorphous structure of P4MP1 investigated by neutron scattering

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Isotactic poly(4-methyl-1-pentene) (P4MP1) is known to absorb various organic solvents, such as alkanes [1]. Although P4MP1 exhibits nearly identical densities in its crystalline and amorphous regions, it has been revealed that solvent absorption predominantly occurs within the amorphous region [2]. However, because isotropic samples were used in Ref. [2], it was difficult to distinguish the structural changes induced by solvent absorption between the crystalline and amorphous regions. Therefore, in this study, we conducted small-angle neutron scattering (SANS) and X-ray diffraction (XRD) experiments using uniaxially stretched P4MP1 samples.

SANS measurements were performed using TAIKAN at J-PARC, and XRD measurements were carried out at BL40B2 of SPring-8. For both techniques, two-dimensional diffraction patterns of the stretched P4MP1 were acquired before solvent immersion and compared with those obtained after immersion. The SANS results revealed the following insights. For the neat P4MP1 sample, no diffraction peaks were observed due to the lack of contrast in the two-dimensional image, resulting from the nearly identical densities of the crystalline and amorphous regions. Upon dropping deuterated decane, the solvent selectively penetrated the amorphous region, generating a contrast that led to the appearance of a peak derived from the lamellar periodic structure exclusively along the stretching direction. This directional dependence indicates the orientation of the lamellar crystals. From the XRD results, a circular first halo was observed at approximately $q = 0.7\text{Å}^{-1}$ for the neat stretched P4MP1. The absence of orientation in the XRD pattern, contrast to the directional behavior observed in the SANS measurement, indicates that this first halo originates from the microscopic structure within the amorphous region. While it had been suggested that this first halo arises from void spaces between backbones [3], the intensity of the first halo was indeed significantly reduced upon the addition of decane. In the presentation, we will also report on the measurement results obtained from quasi-elastic neutron scattering (QENS).

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