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Title	高結晶性ブロック鎖を有する結晶性ー非晶性2元ブロック共重合体の高次構造に関する研究
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Abstract

In crystalline-amorphous diblock copolymers, microphase separation and crystallization work simultaneously and eventually a complicated morphology is formed in the systems. The morphology is considered to be intimately dependent on the stability of microdomain structure, crystallization behavior, and crystallization method. In this study, the final morphology of melt-quenched, crosslinked, and solution-cast crystalline-amorphous diblock copolymers was investigated by small-angle X-ray scattering (SAXS), wide-angle X-ray diffraction (WAXD), transmission electron microscopy (TEM), and differential scanning calorimetry (DSC). It is expected that the principles of morphological design in polymer alloys are obtained from this study. The results obtained are as follows.

1. The morphological transition (microdomain structure → lamellar morphology) was evidently observed by the crystallization of poly(ε-caprolactone) (PCL) blocks in the melt-quenched poly(ε-caprolactone)-*block*-polybutadiene (PCL-*b*-PB) copolymers with low molecular weights, and the details of the lamellar morphology were significantly dependent on the crystallization temperature. On the other hand, the microdomain structure did not change by the crystallization of the PCL block for PCL-*b*-PB copolymers with high molecular weights, where the PCL crystallinity was extremely small. These facts suggest that the morphology and crystallinity can be controlled by the molecular weight and crystallization temperature.

2. Morphological transition did not occur in PCL-*b*-PB copolymers with low molecular weights when the crosslink was introduced into the PB block, where the PCL blocks crystallized partially within the fixed microdomain structure. The crosslink introduced in the system changes extremely the morphology after crystallization, so that the crosslink reaction is one of the effective methods to control the morphology formed in crystalline-amorphous diblock copolymers.

3. When the molecular weight of PCL-*b*-PB copolymers is low, the lamellar morphology was obtained at any temperatures by the solution-casting method. The repeating distance of the lamellar morphology was little affected by the crystallization temperature, suggesting that the lamellar morphology formed in the solution-cast PCL-*b*-PB copolymers is the equilibrium morphology. When the molecular weight was large, the solution-cast PCL-*b*-PB copolymers had the lamellar morphology or crystallized microdomain structure depending on the crystallization temperature. The PCL crystallinity was extremely different between two morphologies; it was large and comparable to that of PCL homopolymer for the lamellar morphology and small for the crystallized microdomain structure, suggesting the substantially difficulty of crystallization in the curved microdomain. These facts indicate that the morphology and crystallinity can be controlled by the crystallization method.

The results obtained in this study show that the morphology formation in crystalline-amorphous diblock copolymers is complicated and various morphologies are formed depending on the molecular characteristics of the copolymers and also the crystallization condition. This study provides valuable results for morphological design of polymer alloys including crystalline-amorphous diblock copolymers.

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