Novel Design of High-Performance Polymer Materials Using Fibrous Fillers

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The state of fiber dispersion in a matrix polymer is significantly important to design fiber-reinforced plastics (FRPs), which have been studied for a long time in plastic industries. For example, when conductive fibers show network structure, the material has high conductivity. Furthermore, if fibers show nucleating activity for the matrix polymer and are aligned to flow direction, the modulus is greatly enhanced owing to a high level of molecular orientation.

The rotational diffusion, i.e., Brownian motion, of multi-walled carbon nanotube (MWCNT) in molten polymers was firstly investigated in this study by the measurements of rheological properties of a compression-molded plate containing MWCNTs. Before the rheological measurement, MWCNTs were oriented by the applied squeeze flow at compression-molding. Because of the strong squeeze flow, leading to the MWCNT orientation, the oscillatory moduli of the sample prepared at low temperature were lower than those prepared at high temperature. Moreover, the moduli increased during post-processing annealing in the rheometer owing to Brownian motion, which resulted in the interarticular interaction of the MWCNTs, and eventually an MWCNT network. These structure developments of the MWCNTs can be expressed by a simple equation using only one characteristic time, i.e., the time required for MWCNT redistribution by Brownian motion. The obtained result revealed that the MWCNT orientation is barely relaxed at a conventional extrusion process.

Considering the slow relaxation process of the MWCNT orientation, the effect of the addition of MWCNT on the structure and properties for extruded high-density polyethylene (HDPE) was investigated. It was found that the MWCNT addition greatly enhanced the orientation of the HDPE chains with shish-kebab structure, although HDPE without MWCNTs showed no orientation at the same condition. The results demonstrate that the oriented MWCNTs greatly accelerate the flow-induced crystallization of HDPE because they act as shish for HDPE. Moreover, the high level of molecular orientation of HDPE affected the mechanical properties in the solid state greatly.

Poly(vinyl alcohol) (PVA) fiber was focused as another conventional fiber, which has lightweight, good cost-performance, and nucleation activity for some crystalline polymers. The fiber has a great potential to provide extremely high modulus and strength as well. In general, PVA is known to be immiscible with most conventional plastics due to its hydrophobicity and unavailable for melt processing due to the strong hydrogen bonding. Therefore, two novel techniques to produce polypropylene (PP) composites containing PVA fibers were proposed. One was to stretch in a molten state of PP with low-viscous PVA. The other was to introduce PVA aqueous solution into a molten PP directly in a twin-screw extruder. It was found that the PVA obtained from both methods formed fibrous shape. Furthermore, the PVA fibers greatly increased the orientation of PP chains and the modulus of the injection-molded specimen when the fibers aligned parallel to the flow direction.

Also, a new technique to show good electroconductivity was proposed using localization of MWCNTs at the phase boundary in co-continuous immiscible polymer blends of polycarbonate (PC) and ultra-high-molecular-weight polyethylene (UHMWPE). When UHMWPE was added to PC/MWCNT in the molten state in an internal mixer, MWCNTs started to move to the UHMWPE phase. However, MWCNTs require a long time to diffuse into the UHMWPE phase because of a low diffusion constant, and thus they localized at the interface between PC and UHMWPE. As a result, a conductive path was developed by MWCNTs, leading to good electroconductivity when the blends have co-continuous structure, although the MWCNT dispersion at the interphase is not allowed at the equilibrium state.

Keywords: Fiber-reinforced plastics, Molecular orientation, Carbon nanotube, Diffusion, Nucleation activity