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Localization of Carbon Nanotube in Multi-Phase Polymer Composites

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Immiscible polymer blends have been developed to improve the mechanical properties of polymer materials. Through the addition of nanofillers into the polymer blend, the mechanical properties can be further improved. Among potential nanofillers, carbon nanotubes (CNTs) are of particular interest due to their unique structure and excellent mechanical properties as well as high electrical and thermal conductivity. They can be selectively localized in one of the polymer phases or at the blend interphase, which has a great effect on the performance of a material. This localization is mainly determined by thermodynamics, which is related to the interfacial interaction between CNT and polymers. However, a localization change during melt-mixing can be influenced by kinetic reasons, *i.e.*, mixing protocol, when the CNTs are first filling in a less-preferred polymer phase. Furthermore, the incorporation of CNTs may change the viscosity ratio of polymer components, resulting in blend morphology change. In most of previous studies, the localization of CNTs is controlled by surface modification, although the method presented in this study is different from them.

In this study, localization of multi-walled carbon nanotubes (MWCNTs) in immiscible polymer blends is controlled by adjusting the processing conditions. Firstly, the effect of MWCNTs on the miscibility between polycarbonate (PC) and poly(ethylene terephthalate) (PET) is studied. Although partial miscibility between PC and PET is confirmed, the incorporation of MWCNTs has no/little effect on the miscibility of the blends. Furthermore, it is found that MWCNTs preferentially reside in the PET phase in the composites due to thermodynamic favorable interaction, leading to an apparent increase in crystallization temperature of PET and thus, acceleration of PET crystallization. Consequently, the degree of crystallinity is enhanced, resulting in a great enhancement in the Young's modulus and ultimate strength of the composites.

Besides, the interphase transfer of MWCNTs between polyethylene (PE) and PC is investigated by two methods: Annealing in the laminated sheet and melt-mixing. It demonstrates that MWCNTs prefer to immigrate from PE to PC during annealing treatment, whereas the interphase diffusion from PC to PE does not take place because of a large difference in the interfacial tension between PE and MWCNT. On the contrary, MWCNTs transfer from PC to PE during melt-mixing process, even if MWCNTs are first introduced into PC. The mechanism is attributed to the selective adsorption of PE molecules on the surface of MWCNTs, which is pronounced at high temperature with the aid of oxygen.

Moreover, the localization of MWCNTs in PC/ethylene-propylene copolymer (EPR) and polypropylene (PP)/EPR blends by the processing conditions is further studied. For the MWCNT-filled PC/EPR blend, it is found that MWCNTs are dispersed in the PC matrix at low mixing temperature. In contrast, the transfer of MWCNTs from PC to EPR occurs at high temperature, suggesting that the adsorption of EPR on MWCNT surfaces is accelerated by the high-temperature-mixing. With the same reason, MWCNTs are also found to be distributed in EPR phase in the PP/EPR blend when mixing temperature is high. In order to avoid the adhesion of EPR on surface of MWCNTs, the composite is prepared at low temperature accompanying with the introduction of thermal stabilizer and nitrogen gas. As expected, MWCNTs are exclusively localized in the PP matrix, resulting in the enhancement of Young's modulus and yield strength.

Keywords: Multi-walled carbon nanotube, Immiscible polymer blend, Interphase diffusion, Localization, Mechanical property