



Carbon nanotubes toughened immiscible polymer blends

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ABSTRACT

The incorporation of CNTs into the immiscible blends are increasingly attractive because of high performance, its wide application and great prospects for development in the future. The objective of this review focuses on CNTs toughened immiscible polymer blends based on the selective localization of CNTs in the immiscible blends. Firstly, the thermodynamic and kinetic factors that determines the localization of CNTs are introduced. The former is relative to the wetting coefficient that can be applied to evaluate the favorable polymer component for CNTs; and the latter relates to the migration of CNTs in polymer blends. Combined with the morphology evolution accompanying with the incorporation of CNTs, the selective localization of CNTs can be distributed in dispersed phase, continuous phase or at the interface, exhibiting different unique properties. Among these properties, CNTs toughened the immiscible polymer blends can be found in these two cases: CNTs disperse in one phase (droplets or continuous) or CNTs disperse at the interface. With the presence of CNTs in immiscible blend, the toughening mechanisms involve: 1) the formation of percolative CNTs network; 2) formation of “cross-linked”-like structure; 3) morphology evolution; 4) compatibilization of immiscible blends; 5) nanobridge effect of CNTs at the interface. This review details recent works for the above mentioned selective localization of CNTs in immiscible blend and corresponding the toughening mechanisms of CNTs for immiscible blends, which is proposed to provide a guidance for the polymer toughening.

1. Introduction

Physically blending two or more immiscible polymers has become an important route to obtain high-performance polymer materials via combining the advantages of individual polymer components [1–3]. Due to the combination of physical, mechanical and other properties to the requirements of applications, polymer blends are extensively applied in a wide range of industrial applications such as automobile, aeronautic, and packaging, etc. Mechanical property is often a key property to evaluate the practical application value and the prospects for development. However, for the immiscible polymer blends, the chain entanglement density at the interface is weaker than those in bulk components. Generally speaking, under the load condition, crack initiates and propagates along the interface, resulting in the failure of materials [4].

It is well known that due to the thermodynamic immiscibility of polymer components, the mechanical properties of polymer blends are strongly dependent on the compatibilization of polymer components represented by the morphology and interfacial interaction. The morphologies of immiscible blends may be classified as sea-island and co-continuous structure [5,6]. For the former one, droplet, fiber and

lamellar dispersed phases are included, in which the droplet morphology exhibits stiffness or elasticity, the fiber morphology endows the materials with unidirectional strength, and the lamellar morphology provides excellent barrier properties. For the latter one, the co-continuous morphology can be applied to improve the conductive properties with the aid of conductive fillers. Meanwhile, a great number of articles have exhibited that the change in the size distribution of phase can also result in obviously different mechanical properties or even a brittle-tough transition [7]. Therefore, it is an efficient way to fabricate the blends with excellent properties via tailoring well-defined, stable and producible morphologies. On the other hand, based on the fracture mechanics which aims at determining the response of a cracked material to an applied load, the mechanical properties, especially for the toughness, are usually related to the interface interactions. Most polymer blends are immiscible, leading to weak interfaces with low aptitude in dissipating energy, acting as stress concentrator and failure initiation points. Therefore, fracture of polymer blends is mainly indicated by a weak joint failure, which may greatly affect the toughness of the material [8].

In the pursuit of achieving new polymer blends with high performance, attentions have been drawn to systems with adding

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compatibilizers which can enhance the interfacial adhesion and further tailor the morphology of blends [9,10]. To this end, the following two methods are usually applied: one is the addition of premade binary block or graft copolymer during blending of immiscible blends [11,12], and the other is reactive compatibilization in which the binary block or graft copolymer can be in-situ produced at the interfacial regions [13]. The addition of such compatibilizers can lead to more stable, finer scale morphologies, by reducing the effective interfacial tension, enhancing interfacial adhesion and slowing phase coarsening [14,15]. However, this method is less attractive because copolymers with specific structure are always difficult to synthesize and effective compatibility can only be obtained when the copolymer is adequately diffused to the interface of immiscible polymer blends.

In recent years, CNTs have been considered as an ideal reinforcement agent to improve mechanical, physical, electrical or other functional properties of polymers, due to their outstanding properties, including high elastic modulus and strength, high aspect ratio, high electrical and thermal conductivity and high water and chemical resistance [16,17]. CNTs induced reinforcing mechanisms of polymeric matrix, proposed in the literature, are mainly described as follows: pulling out of CNTs from the matrix [18], the breakage of CNTs [19] and the increased degree of crystallinity of semi-crystalline polymers through heterogeneous nucleation of CNTs [20]. Specifically, for the toughening mechanism, the bridge effect of the CNTs has been emphasized, which has been reported that the CNTs can increase the fracture toughness through inhibition of the crack propagation and crack deviation, as shown in Fig. 1 [21], in which the marks A, B, C and D represent the several scenarios about the break and pull out of CNTs in the material. Illuminated by the toughening mechanisms of CNTs in homogeneous polymer, a question arises: can we introduce CNTs into an immiscible polymer blend to improve the fracture toughness of the blend? It is specially imaged that when CNTs selectively locate at the interface of immiscible blend, the weak interface may be “screwed and mended” by the coil-like CNTs, expecting to exhibit the enhancement in the interfacial adhesion and dissipating energy adsorption. In this case, could the ductility be positively affected?

2. Selective localization of CNTs in the immiscible blends

Introduction of nanoparticles into the immiscible blend is a new strategy that tailors the compatibility of immiscible blends [22–25]. The organic nanoparticles, on one hand, can adsorb polymer chains, giving rise to re-build the thermodynamic balance between polymer components and polymer/nanoparticles, thus the phase morphology and final performance are well optimized. On the other hand, due to the

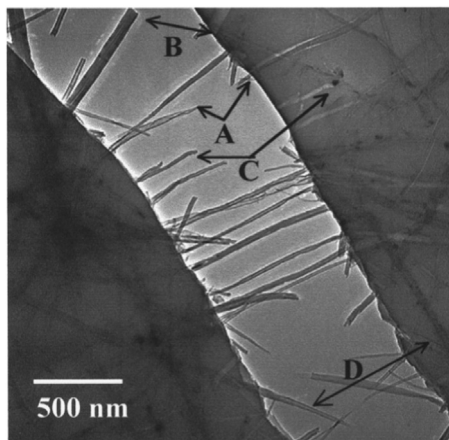


Fig. 1. TEM observation of crack nucleation and propagation in CNTs/PS thin films induced by thermal stresses. The CNTs tend to align and bridge the crack wake then break or pull out of the matrix [21].

preferable adsorption of nanoparticles to individual polymer component, the nanoparticles would like to migrate from the unfavorable polymer component to the favorable one, finally resulting in the selective localization of nanoparticles in polymer blends.

Because of the inherent nature such as large aspect ratio and multi-functions of CNTs, CNTs have attracted great attentions on obtaining polymer blends with superior property via selective localization in blends.

The selective localization of CNTs is the key to endow the blends with desired functions. Commonly speaking, the selective localization of CNTs in immiscible blends is classified as CNTs localization in the dispersed phase, in the continuous phase or at the interface, which may be responsible for unique property correspondingly. For example, when the CNTs are distributed in the continuous phase, the double percolation network, in which the CNTs entangle as network in continuous and the continuous phase penetrate through the whole sample, could be well found and be responsible for the enhancement in the electrically/thermally conductive properties. The excellent toughening effect is also mainly associated with the percolated network structure or interfacial adhesion enhanced by the interfacial localization of CNTs. It is obvious that the selective localization of CNTs in a large extent determines the final performance of polymer blends.

2.1. Thermodynamic predictions of CNTs in immiscible blend

A great amount of publications has exhibited that the selective localization of nanofillers or CNTs in polymer blends can be readily controlled by adjusting thermodynamic and kinetic parameters. As for the former one, preferential localization of CNTs in the one component of immiscible blend is illuminated by the system's tendency to minimize its free energy, and the wetting coefficient w_a which is derived from the Young's equation (Eq. (1)) is generally applied to explain the valid principle [26].

$$w_a = \frac{\gamma_{\text{polymer1,CNT}} - \gamma_{\text{polymer2,CNT}}}{\gamma_{\text{polymer1,2}}} \quad (1)$$

In this equation, the $\gamma_{x,y}$ donates the interfacial tensions between CNTs and polymer components and the interfacial tensions between two polymer components. And the wetting coefficient is applied to present the thermodynamic localization of CNTs in the particular component of immiscible blend, which corresponds to the minimization of the three relevant interfacial tensions. Accurately measurement of the $\gamma_{x,y}$ is based on the contact angle or capillary effects which are significantly influenced by the surface structure, thus the value of contact angles can truly reflect the surface properties of polymer and CNTs. Two main methods are applied to relate the surface energies of polymers and CNTs to the interfacial energies between polymers and CNTs and the polymers. One is the harmonic-mean equation (Eq. (2)), which is valid for the low-energy materials, and the other one is geometric mean equation (Eq. (3)), which is valid to evaluate the interfacial energy between a low-energy material and a high-energy material [27].

$$\gamma_{12} = \gamma_1 + \gamma_2 - 4 \left[\frac{\gamma_1^d \gamma_2^d}{\gamma_1^d + \gamma_2^d} + \frac{\gamma_1^p \gamma_2^p}{\gamma_1^p + \gamma_2^p} \right] \quad (2)$$

$$\gamma_{12} = \gamma_1 + \gamma_2 - 2(\sqrt{\gamma_1^d \gamma_2^d} + \sqrt{\gamma_1^p \gamma_2^p}) \quad (3)$$

In the Eqs. (2) and (3), the γ_i is the surface tension of component i , γ_i^d is the dispersive part of the surface tension of component i , and γ_i^p is the polar part of the surface tension of component i . It should be noted that the surface tension is related to the temperature, thus the values of the surface energies of polymers in the melt state should be extrapolated from the value obtained at room temperature by contacting angle [28,29]. On the other hand, concerning about the wetting characteristics of CNTs, it depends on the intrinsic structures of CNTs. For

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