



# Electrically conductive and super-tough polypropylene/carbon nanotube nanocomposites prepared by melt compounding



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## ABSTRACT

Polyethylene–octene copolymer (POE), maleic anhydride-grafted POE (POE-g-MA) and ultrahigh molecular weight polyethylene (UHMWPE) were incorporated as second polymer components with polypropylene/carbon nanotube (PP/CNT) components by melt compounding and their influences on electrical conductivity and toughness of the nanocomposites were studied. POE-containing nanocomposites exhibit not only high electrical conductivity but also super-toughness, which are not considerably dependent on compounding sequences. Though UHMWPE does not largely affect the electrical conductivity of PP/CNT nanocomposites due to its impermeable feature, its toughening efficiency on PP/CNT nanocomposites is rather low. Irrespective of the compounding sequences, the incorporation of POE-g-MA adversely affects the formation of conducting network of CNTs due to its attraction with CNTs, leading to an insulating PP/CNT nanocomposite even with 30 wt% of POE-g-MA. Among the three second polymer components, POE is much preferred in actual applications as it makes PP/CNT nanocomposites super-tough while the conducting feature of the nanocomposites is retained.

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## 1. Introduction

In the past decades, conductive carbonaceous fillers such as carbon black particles, carbon fibers, and graphite flakes were widely used to enhance electrical conductivity of insulating polymers [1–7]. However, high loading of these conductive fillers is usually needed when electrical conductivity is required, which not only increases the final cost of the composites, but also often impairs mechanical properties and processability [8]. Therefore, increasing attention has been paid to nanoscale conducting fillers with high aspect ratios such as carbon nanotubes (CNTs), which endow polymers with high electrical conductivities at low filler loadings [9,10].

Generally, there are mainly three approaches to improve electrical conductivity of a polymer at a given loading of conducting fillers, in other words, to decrease filler loading while keeping a high electrical conductivity. One is to rely on double percolation induced by immiscible polymer blends [11–21]. Sumita et al. [13] showed that the percolation threshold of short carbon fiber-filled high density polyethylene/polypropylene (HDPE/PP, 50/50) blend was lower than those of the individual polymer composites due to the preferential location of the carbon fibers in HDPE phase and the continuity of the HDPE phase in the blend. Gubbels et al. [15,16] endowed immiscible PE/polystyrene (PS) blends with

satisfactory electrical conductivities by using very small amounts of carbon black particles by controlling the localization of the carbon. Compared to the percolation thresholds of 8 wt% for monophase PS and 5 wt% for monophase PE, a co-continuous two-phase PE/PS blend, in which carbon black was spontaneously located in the PE phase, had a reduced percolation threshold of 3 wt%. Interestingly, in the case of the selective localization of carbon black at PE/PS interfaces, the percolation threshold was as low as 0.4 wt%. However, the main drawbacks of using immiscible polymer blends are their unstable morphology and unsatisfactory mechanical properties. Another approach to decrease the loading of conducting fillers is additionally incorporating electrically inert inorganic fillers, such as clay layers and calcium carbonate (CaCO<sub>3</sub>) particles [22–26]. Clay was reported to improve the dispersion of conducting fillers and thus to reduce percolation thresholds [22–24]. In addition, the presence of clay also improved electrical conductivity of co-continuous polycarbonate/PP/CNT nanocomposites, in which CNTs were dispersed homogeneously within polycarbonate phase, by hindering the migration of CNTs from polycarbonate phase to PP phase [25]. Yu et al. [26] incorporated electrically inert CaCO<sub>3</sub> particles into PP/CNT nanocomposites and observed a significant increase in electrical conductivity and a decrease in percolation threshold of the PP nanocomposites, which was due to that the presence of impermeable CaCO<sub>3</sub> particles occupied some space of the matrix and thereby increased the CNT concentration in the PP phase compared to the PP/CNT binary nanocomposite at the same CNT loading. However, it has

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to be noted that the additional incorporation of inert mineral particles may cause the resulting polymer nanocomposites brittle and poor in melt processability. The third approach is to simultaneously use two conducting fillers with different aspect ratios [27–30]. Kim et al. [28] reported an improved conductivity of epoxy nanocomposites containing CNT and carbon black. The addition of carbon black particles effectively filled the gaps between CNTs and benefited the formation of a conductive network.

Apart from the enhanced electrical conductivity, mechanical properties, especially fracture toughness, of the electrically conductive nanocomposites are also a most critical requirement for practical applications. Till now, there are only a few papers focusing on toughening of conductive nanocomposites [10,31–35]. Meinncke et al. [10] investigated the electrical conductivity and mechanical properties of CNT-filled nylon 6/acrylonitrile–butadiene–styrene (ABS) blends. The selective location of CNTs within nylon 6 phase resulted in a remarkable reduction of percolation threshold from 4 to 6 wt% for nylon 6/CNT binary nanocomposites to 2–3 wt% for nylon 6/CNT/ABS ternary nanocomposites. In addition, although the notched impact strength values of the ternary nanocomposites decreased with the increase of CNT loading, the ternary blends still exhibited a sufficiently higher toughness at low CNT loadings. Fu et al. [34] developed electrically conductive and super-tough PP/ethylene-propylene–diene rubber (EPDM) blends filled with carbon black particles using a two-step melt compounding. Their results revealed that carbon black particles tended to be accumulated around EPDM particles to form a conductive network in PP/EPDM/CB (80/20/3) nanocomposite, which dramatically reduced the percolation threshold of the nanocomposites. The notched Izod impact strength of the nanocomposite was also improved. Dasari et al. prepared electrically conductive and super-tough nylon 6/CNT/maleic anhydride-grafted polyethylene–octene copolymer (POE-g-MA) nanocomposites by controlling CNTs dispersing only in nylon 6 matrix [35]. The absence of CNTs inside POE-g-MA particles and their entire presence in matrix improved the electrical conductivity because of the volume-exclusion effect of POE-g-MA; while the toughening efficiency of POE-g-MA particles retained in the ternary nanocomposites, similar to that in nylon 6/POE-g-MA binary blends. It is clear now that, in a rubber-toughened polymer/conducting filler system, the location of the conducting fillers plays a crucial role in improving both conductivity and toughness of the polymer nanocomposites. In the present work, three different polymer components, polyethylene–octene copolymer (POE), POE-g-MA, and ultrahigh molecular weight polyethylene (UHMWPE) were melt-compounded with PP/CNT components. Their influences on electrical conductivity and toughness of PP/CNT nanocomposites were investigated so as to make PP not only electrically conductive but also super-tough.

## 2. Experimental

### 2.1. Materials

PP (S1003) with a melt flow index of 3.0 g/10 min was purchased from Yanshan Petroleum Chemical Co., Ltd., China. The multiwalled CNTs with a purity of >95% were supplied by Chengdu Organic Chemicals Co., Ltd. (China). As declared by the supplier [36], the CNTs were vapor-grown with outer diameters of 20–30 nm and inner diameters of 5–10 nm. POE with a trade name of Engage 8003 was obtained from Dow Chemical (USA); while POE-g-MA with the grafting of 1 wt% maleic anhydride was supplied by Haier Kehua Co., Ltd. (China). UHMWPE with a molecular weight of more than  $2.0 \times 10^6$  g/mol was provided by Shanghai Lianle Chemical Industry Co., Ltd. (China).

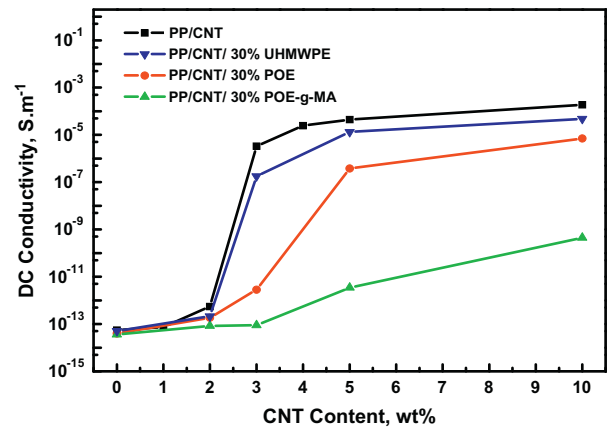


Fig. 1. Electrical conductivity versus CNT content for PP/CNT, PP/CNT/POE, PP/CNT/POE-g-MA, and PP/CNT/UHMWPE nanocomposites. The content of POE, POE-g-MA and UHMWPE is kept at 30 wt%.

### 2.2. Melt compounding

PP nanocomposites and blends were prepared by melt compounding using a two-roll mill at 190 °C for 15 min, followed by compression molding into square plates of  $15 \times 15 \times 3$  mm<sup>3</sup> with a hot press at 200 °C. To investigate the effect of compounding sequence on microstructure and properties of PP/CNT nanocomposites containing POE, POE-g-MA or UHMWPE, two sequences were adopted. C-1: (PP + CNT) + (POE, POE-g-MA or UHMWPE), means PP was firstly compounded with CNTs for 10 min and then the nanocomposite was blended with POE, POE-g-MA or UHMWPE for 5 min. C-2: (PP + POE, POE-g-MA or UHMWPE) + CNT, means PP was blended first with POE, POE-g-MA or UHMWPE and the resulting blend was compounded with CNTs later. All the nanocomposites in the following sections were prepared by C-1 compounding sequence unless otherwise specified.

### 2.3. Characterizations

To investigate their morphological features, the specimens were fractured after immersing in liquid nitrogen for 30 min. Some of them were etched with n-heptane with the help of ultrasonication at room temperature for 48 h to selectively remove POE or

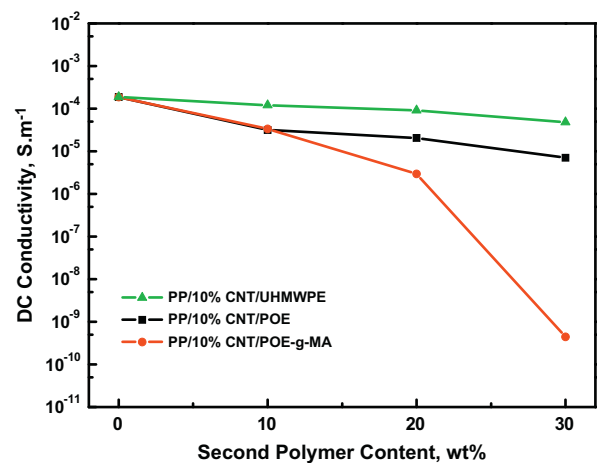


Fig. 2. Effects of second polymer component content on electrical conductivity of PP/CNT/POE, PP/CNT/POE-g-MA, and PP/CNT/UHMWPE nanocomposites at a given CNT loading of 10 wt%.

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