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Clay/carbon nanotube hybrid mixture to reduce the electrical percolation threshold of polymer nanocomposites

Mohammed H. Al-Saleh [a, b, *]

[a] Department of Chemical Engineering, Jordan University of Science and Technology, P.O.Box 3030, Irbid 22110, Jordan
[b] Department of Chemical and Petroleum Engineering, United Arab Emirates University, Al-Ain, United Arab Emirates

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ABSTRACT

Creating electrically conductive polymer composites with extremely low nanofiller concentration by melt compounding is a major research challenge. At low nanofiller concentration, the valuable properties of polymers are preserved and the feasibility of the composite is promoted. In this work, an organically modified clay (OMC) was utilized to alter the structure and consequently the electrical resistivity of carbon nanotubes (CNT)/polypropylene (PP) composite. As a result of OMC incorporation, the electrical percolation threshold concentration (EPTC) was reduced from 1.0 wt% CNT for the CNT/PP composite to 0.5 wt% CNT for the CNT:OMC/PP composite, corresponding to 50% reduction in the amount of CNT. The macro-dispersion analysis did not reveal any significant difference between the dispersion of CNT in the CNT/PP and CNT:OMC/PP composites. However, the processing behavior analysis showed a significant decrease in mixing torque and consequently mixing energy due to the addition of OMC. The decrease in mixing torque and/or mixing energy decreases the destruction of CNT aspect ratio. In addition, the DSC analysis showed a decrease in composite crystallinity due to OMC addition. This finding reveals a thinner insulating crystalline layer at the surface of CNT particles and consequently higher electrical conductivity. Based on these experimental findings, it can be speculated that the addition of OMC promoted the conductivity of the composite by decreasing the mixing shear stress and/or polymer crystallinity.

1. Introduction

Conductive composites are attractive materials for many applications in the electronics, packaging, health and energy sectors [1–4]. Conventional polymers, such as polypropylene (PP) and polystyrene (PS), are insulators with electrical resistivity around 10^{16} Ohm cm [5]. However, proper incorporation of a critical amount of conductive nanofiller within these polymers creates 3D conductive nano-networks that lead to sudden and remarkable decreases in the system electrical resistivity [6,7]. This critical amount of the conductive nanofiller at which the insulator-conductor transition occurs is known as the electrical percolation threshold concentration (EPTC). Any conductive nanofiller can be used to create the 3D nano-network. However, in the case of nanofillers with low aspect ratio, such as the low-structure carbon black and spherical metal nanoparticles, very high nanofiller loading (~40 vol%) is required to reach the EPTC. At high EPTC, the composite becomes brittle with poor mechanical properties. Thus, composites reinforced with nanofillers of high aspect ratio, such as the 2D graphene nano-sheets and the 1D carbon nanotubes (CNT), have been widely investigated [8–12]. For example, experimental findings showed that CNT/polymer composites prepared by melt-compounding are conductive at nanofiller loading less than 1 wt% [5]. However, we are still in need for further reduction in the EPTC to reduce the cost of the final composite material.

In addition to their high aspect ratio, CNTs are very conductive [13,14]. The high conductivity is required in applications such as the transparent conductive electrodes and electromagnetic interference (EMI) shielding [15–17]. However, since the CNT are very expensive compared to the hosting polymer matrix, we are still in need to reduce the EPTC and enhance the electrical conductivity of the CNT-based composites [18]. It is well known that by solution processing, a CNT-based nanocomposite with extremely low EPTC can be obtained [18–20]. Nevertheless, solution processing is not suitable for large-scale production because it needs huge volumes of solvents. In addition, solution processing is appropriate for a

* . Department of Chemical Engineering, Jordan University of Science and Technology, P.O.Box 3030, Irbid 22110, Jordan.
E-mail address: mhsaleh@just.edu.jo.

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limited number of polymers because of the difficulty in finding suitable co-solvent for the polymer/CNT pairs [21]. Thus, herein the focus is on reducing the EPTC by the melt compounding, which is the favorable production method in the industry because it can handle almost all thermoplastics.

Recently, several researchers have investigated the concept of conductive nanofiller/non-conductive nanofiller hybrid mixture aiming at reducing the EPTC and enhancing the electrical conductivity of composite materials [22–25]. Non-conductive nanofillers, such as clay [22,26–29], have been utilized aiming at affecting the CNT dispersion-state (Fig. 1b) or distribution-state (Fig. 1c), as shown schematically in Fig. 1. In the case of affecting the dispersion-state as shown in Fig. 1b, the non-conductive filler is required to enhance the dispersion of the conductive nanofiller, i.e. reduce the CNT aggregates. While in the case of affecting the distribution-state Fig. 1c, the non-conductive nanofiller is required to create a CNT-free zone; thus enhancing the concentration of CNT particles is a smaller domain. Ultimately, this will lead to higher effective concentration and lower EPTC [22,26,30,31].

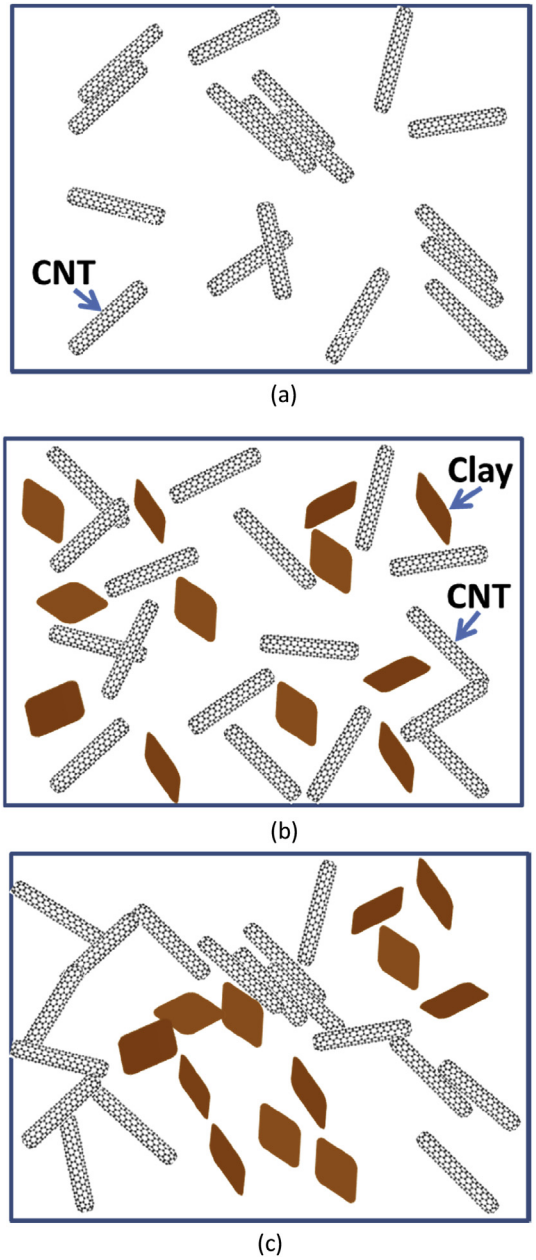

Fig. 1. Dispersion of CNT particles (a) before addition of clay (b) Clay improving the dispersion of CNT (c) Clay creating CNT-free zones.

For Clay:CNT/epoxy composites, a significant reduction in the EPTC was reported upon the addition of 2 wt% clay due to the enhancement in the dispersion of CNT [26]. Recently, Wu et al. [31] reported that tourmaline:CNT/polyurethane (PU) composite has lower EPTC and electrical resistivity than the CNT/PU composite. For example, a 3 wt% CNT/PU composite exhibited two orders of magnitude reduction in electrical resistivity due to the incorporation of 5 wt% tourmaline particles. This remarkable decrease in the electrical resistivity was ascribed to a reduction in the potential barrier between the adjacent conductive nanoparticles due to the presence of polar tourmaline particles. In contrast, Bao et al. [28] and Plaza et al. [27] reported an increase in the EPTC of CNT/PP composite due to the addition of clay since it prevents the formation of CNT-network. Similar observation was reported by Besco and coworkers [32] for CNT:OMC filled polymer blend of polycarbonate/acrylonitrile-butadiene-styrene (PC/ABS). The CNT and OMC particles were observed in PC phase, and the OMC particles prevented the formation of the conductive CNT-network within the PC phase [32]. In this work and due to the controversial results reported in the literature regarding the influence of non-conductive nanofillers on the electrical resistivity and percolation behavior of CNT/polymer composites, the influences of OMC on the electrical resistivity of melt-compounded CNT/PP nanocomposite were investigated.

2. Experimental details

2.1. Materials and fabrications

One polymer and two different fillers were used in this study. The polymer was PP (504P, homopolymer, SABIC, Saudi Arabia); and the fillers were NC-7000 multiwalled CNT (Nanocyl S.A., Belgium) and 1.44P OMC nanoplatelets (Nanocor Inc., USA). According to the manufacturer, the CNT particles are 9.5 nm in diameter and 1.5 μm in length. The OMC is functionalized with quaternary ammonium and is recommended for use with polyolefins. The nanocomposites were compounded in 50-EHT batch mixer connected to Plastograph EC torque-rheometer (Brabender, Germany). Before melt compounding, the PP and CNT were dried in a vacuum oven. The OMC powder was not dried in order to avoid the degradation of the functional groups at the clay surface. In a typical experiment, the mixing chamber was preheated to 180 °C then 28.3 g of PP pellets were charged into the chamber and mixed for 3.0 min at 100 rpm. Then the nanofiller/s were added to the molten PP and mixed for additional 10 min. The nanofiller was either CNT or CNT:OMC mixture. If not otherwise stated, the CNT:OMC weight ratio in the formulation is 1:1. Parts for electrical conductivity characterization were prepared by compression molding at 200 °C and 27.5 MPa for 10 min. The parts were ($40 \times 20 \times 1\ mm^3$) rectangular plates. After the 10 min of compression molding, the mold platens were cold under the applied pressure using water stream.

2.2. Characterization tools

The dispersion state of CNT within the PP matrix was characterized by an optical microscope (MC50, micros, Austria). For the optical microscopy analysis, thick sections (~1.0 μm) were cut from the mold specimens using Reichert-Jung Ultracut E ultramicrotome. The electrical resistivity was measured using Keithley 2010 digital multimeter connected to a 4-wire probe test fixture and