



Implication of multi-walled carbon nanotubes on polymer/graphene composites



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ABSTRACT

Graphene sheets stack in polymer matrices while multi-walled carbon nanotubes (MWCNTs) entangle themselves, forming two daunting challenges in the design and fabrication of polymer composites. Both challenges have been simultaneously addressed in this study by hybridizing the two nanomaterials through melt compounding to develop elastomer/graphene platelet/MWCNT (3-phase) composites, where MWCNTs were fixed at 2.8 vol% (5 wt%) for all fractions. We investigated the composites' structure and properties, and compared the 3-phase composites with elastomer/graphene platelet (2-phase) composites. MWCNTs may bridge graphene platelets (GnPs) and promote their dispersion in the matrix, which would provide more interface area between the matrix and the fillers. MWCNTs worked supplementally to GnPs by forming conductive networks, where MWCNTs acted as long nanocables to transport electrons and stress while GnPs served as interconnection sites between the tubes forming local conductive paths. This produced a percolation threshold of electrical conductivity at 2.3 vol% for 3-phase composites, 88% lower than that of 2-phase composites. At 26.7 vol% of total filler content (MWCNTs + GnPs), tensile strength, Young's modulus and tear strength showed respectively 303%, 115%, 155% further improvements over those of 2-phase composites. These improvements are originated from the synergistic effect between GnPs and MWCNTs. The conducting elastomeric composites developed would potentially open the door for applications in automotive and aerospace industries.

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1. Introduction

Of all nanomaterials, carbon allotropes occupy an outstanding position owing to many unique properties. Graphene – a monoatomic layer of sp^2 -hybridized carbon atoms arranged in 2-D honeycomb structure – features a combination of outstanding mechanical and physical properties: tensile strength (100 GPa) and modulus (1 TPa), 25% elongation at break, and excellent thermal (>3000 W/m·K) and electric (6000 S/cm) conductivity [1]. With the discovery of graphene which is considered as the basic building unit for all carbon allotropes, the circle gap in the family of carbon allotropes is now filled: 0-D carbon materials such as fullerenes, 1-D nanotubes, 2-D graphene and 3-D graphite [2,3].

Graphene platelets (GnPs) are few-layer graphene (2–5) with a very low oxidation degree [4–6]. Owing to the high in-plane conductivity, unique graphitized plane structure and more importantly low manufacturing cost, GnPs show promise to the development of polymer composites. The planar structure of GnPs provides a 2-D path for phonon transport and its high surface area offers a large interface area with the polymer matrix as long as the stacking of graphene is prevented, and this results in desired improvements in mechanical and functional properties for polymers as reported [7–9]. Nevertheless, it is a formidable challenge to produce a significant reinforcing effect at a low nanoadditive loading by melt compounding.

Melt compounding [10–13], solution mixing [14] and *in-situ* polymerization [15–19] are the main processing methods to fabricate polymer composites. Since researchers favour the design and fabrication of polymer nanocomposites by the current polymer processing facilities in industry, melt compounding becomes the most attractive and promising selection in spite of its inefficiency

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in preventing GnPs stacking. Polymer nanocomposites would replace increasingly more conventional composites due to their highly improved properties and ease of processability and manufacturing. Actually the improvements at low filler fractions make polymer nanocomposites ideal candidates for applications in high-performance structural composites, such as those in the production of aircraft, automotive, marine, spacecraft composites and sports facility.

High-aspect ratio, rod-like carbon nanotubes (CNTs) are a class of seamless cylindrical fillers showing potential to enhance the electrical and thermal conductivity of hosting polymers [20–23]. There are three major factors limiting the reinforcing effectiveness of CNTs on polymers, including (i) their dispersion in matrices, (ii) the interface strength between CNTs and matrices and (iii) the high fabrication cost of CNTs [24]. At loadings higher than 5 vol%, multi-walled CNTs tend to agglomerate and single-walled CNTs form bundles due to their large aspect ratios (>1000) and the strong van der Waals forces between individual carbon nanotubes [25].

A number of studies investigated the synergetic effects of different fillers on polymers [26–34]. Kim et al. [27] added CNTs and carbon black (CB) into epoxy leading to an electrical percolation threshold at 0.4 wt% (0.2 wt% CB and 0.2 wt% CNTs). In the case of 0.1 wt% multi-walled carbon nanotubes co-used with 0.9 wt% graphite nanosheets, the thermal conductivity of epoxy improved 150% in comparison with 23% when only 1 wt% of graphite nanosheets were used [32]. Three types of fillers, CNTs, CB and expanded graphite, were solution-mixed with epoxy [33] giving a percolation threshold of 0.2 wt%, while the threshold was 0.5 wt% in the case of expanded graphite co-used with CB and it was 1 wt% with expanded graphite only. When CB was added as a compatibiliser between the polar organomodified nanoclay and the nonpolar ethylene–propylene–diene rubber, it improved the thermal and mechanical properties [34].

In a previous work [7] multi-walled carbon nanotubes (MWCNTs) were loosened and disentangled by melt compounding. Inspired by this work, we herein propose to utilize MWCNTs as a third phase for elastomer/GnP nanocomposites fabricated by melt compounding, since MWCNTs may bridge GnPs facilitating stress transfer and electron mobility. From the perspectives of composition and structure, there might be a synergetic effect between CNTs and graphene in terms of structure and cost. We aim to prevent GnPs restacking and produce a significant reinforcing effect at low GnP fraction.

In this study, three-phase elastomer nanocomposites are developed, including: (i) using ethylene–propylene–diene rubber (EPDM) as a nonpolar matrix which features good aging properties and high filler loading capacity [7], (ii) adding graphene platelets (GnPs) as a dominant filler into the elastomer by melt compounding, (iii) incorporating a low fixed-fraction of MWCNTs as the third phase, and (iv) investigating the structure–property relations of the prepared 3-phase composites. Our investigation shows a synergetic effect between GnPs and MWCNTs as proved by the remarkable improvement in mechanical properties and the low electrical conductivity percolation threshold.

2. Experimental details

2.1. Materials

Jilin Petrochemical Limited China supplied ethylene–propylene–diene monomer rubber (EPDM 4045, ethylene content at 53–59%), with a Mooney viscosity of ML (1 + 4) at 100 °C = 38–52. The recipes for fabrication of elastomer composites are listed in Table 1 where curing agents are available in market and used as received. It is known that sulfur and peroxides are widely used to crosslink elastomer. A mixed curing system is often used to

Table 1
Recipes of elastomer compounds.

Material	Weight (g)
EPDM	100
Dicumyl peroxide (DCP)	4
Sulfur	1
N,N'-m-phenylenebismaleimide (HVA-2)	1
Multi-walled carbon nanotubes	5
Graphene platelets	Variable

produce balanced properties, such as aging resistance, compression set, mechanical performance and scorch safety [35]. In this study peroxide dicumyl peroxide (DCP) was added to produce C–C crosslinks while sulfur was responsible for the C–S and S–S crosslinks. N,N'-m-phenylenebismaleimide (HVA-2) is a type I coagent used to increase the curing rate and quality.

Asbury Carbons kindly provided the graphite intercalated compound (Asbury 3494) with a pure carbon content of over 80% and size over 75 µm. Multi-walled carbon nanotubes (MWCNTs) of 95% in purity, 8–25 nm in diameter and ~0.75 µm in length, were provided by Showa Denko, Japan. MWCNTs are preferred as the third phase for EPDM/graphene composites due to their high aspect ratio and cost advantage over single-walled carbon nanotube.

2.2. Preparations

2.2.1. Graphene platelets (GnPs) and multi-walled carbon nanotubes (MWCNTs)

One gram of the graphite intercalated compound was weighed and transferred to a crucible preheated inside a common furnace at 700 °C, and it was treated at the same temperature for 1 min. After cooling down, the expanded product was suspended in acetone at 1 wt% in a metallic container and then sonicated (200 W, 42 kHz) for 1 h in an ice-water bath. The produced GnPs are collected through filtration and dried overnight in a ventilated oven at 60 °C. The detailed process and properties of the produced GnPs can be found elsewhere [7]. MWCNTs were used as received without any further treatment.

2.2.2. Three-phase elastomer nanocomposites

A two-roll mill was used to fabricate EPDM/GnP/MWCNTs composites at room temperature at a speed ratio of 1:1.3 with 1 mm nip clearance. A determined weight of EPDM gum was inserted into the mill to produce a large thin sheet. Firstly a desired quantity of GnPs was gradually added to the matrix. After adding all GnPs, the roll gap was decreased and increased a few times to obtain homogenous filler dispersion. Secondly, MWCNTs were added to each EPDM/GnP compound at 2.8 vol% using a similar procedure. When the blend cooled down to room temperature, the curing agents described in Table 1 were added. Vulcanization was carried out in an electrically heated hydraulic press for 30 min at 150 °C under 3 MPa to form composite sheets (120 × 120 × 2 mm) with different filler fractions. The obtained samples were stored at least 48 h before testing.

2.3. Nanocomposite characterization

X-ray diffraction (XRD): A mini-materials analyzer (MMA) tuned to Cu K α radiation at 35 kV and 12 kW was employed to characterize samples including GnPs, MWCNTs and their composites. The X-ray diffraction patterns were collected at room temperature under a reflection mode with a scanning rate of 1°/min between $2\theta = 2\text{--}45^\circ$.

Transmission electron microscopy (TEM): H-800-1 TEM (Hitachi Co., Japan) was employed to study the internal structure of a

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