



Largely improved thermal conductivity of HDPE/expanded graphite/carbon nanotubes ternary composites via filler network-network synergy



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ABSTRACT

Utilizing the synergistic effect of various fillers is an efficient strategy to enhance the thermal conductivity of polymer composites, in which the key is to modulate their dispersion and network formation in polymer matrix. In this work, expanded graphite (EG) was individually added into high density polyethylene (HDPE) to fabricate first the binary composites through melt blending. The electrical conductivity of the prepared composites was measured to determine the percolation threshold for HDPE/EG composites. Then HDPE/EG composites with three compositions, representing below percolation, just percolation and above percolation, respectively, were chosen as matrix and melt mixed with carbon nanotubes (CNTs) to make HDPE/EG/CNTs ternary composites. It was found that adding CNTs results in a linear increase of thermal conductivity for HDPE/EG composites with composition below percolation, along the line by adding the same amount of EG. While a jump of thermal conductivity was observed by adding CNTs for HDPE/EG composites with composition just and above percolation. The electrical conductivity and rheology property were measured and SEM experiment was carried out to explore the filler dispersion and their network formation in HDPE matrix. All the results suggested a possible location CNTs in EG filler network for HDPE/EG composites with composition just and above percolation. Thus the formation of CNTs network within EG network is attributed to the main reason for the largely enhanced thermal property. This work endows a new enlightenment to fabricate the composites with a great thermal conductivity.

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1. Introduction

Nowadays, the thermal conductivity is one of the key properties in many fields. Since polymers are well in corrosion resistance, mechanical properties, good processing ability and so on, especially the polyolefin, they are widely used in electronic, packaging and substrate materials devices. However, polymers are poor in thermal conductivity, most of which are under 0.5 W/mK [1]. Therefore, enhancing the thermal conductivity of polymers is necessary and with a grander prospect [2,3].

There are many strategies to enhance the thermal conductivity of polymers. At present, the most efficient and feasible ways is adding fillers with high thermal conductivity into the polymer matrix [4–6]. Those fillers can be classified into the metallic, the ceramic and the carbonous. Among these fillers, CNTs [7,8] that is one-dimensional structure and EG [9,10] that is two-dimensional structure have drawn a great attention due to their

outstanding thermal conductivity. The conductive value of expanded graphite is more than 300 W/mK, and the CNTs can be almost up to 3000 W/mK in theory [11].

Nevertheless, adding fillers into polymers cannot always get a large increase in thermal conductivity. The reason is that the type of fillers is not the only factors which influence the property. Some others can also play an important role, such as the structure of the matrix, the interfacial thermal resistance, the dispersion of fillers and so on [12–14]. The influential factors of matrix are mainly related to the degree of crystallinity and the orientation of molecular chains. In general, the higher the degree of crystallinity and orientation of molecular chains become, the higher the thermal conductivity is [15,16]. As for interfacial thermal resistance, modifying the surface of fillers can get the interfacial interaction improved so to enhance the thermal conducting property of composites [17–19]. Moreover, optimizing the dispersion of fillers and constructing more conductive network can make a progress in enhancing the conductivity [20–23].

Recently, hybrid fillers have been thought to enhance the thermal conductivity via the synergistic effect of the thermal

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conductive fillers [22–25]. Yu et al. added the single-walled carbon nanotubes and graphitic fillers into epoxy and observed a synergistic effect on the thermal conductivity [26]. This synergistic effect is attributed to the bridging of planar sheets of EG via the CNTs. And Feng et al. fabricated a three-dimensional CNTs/exfoliated graphite block with a remarkable enhancement in thermal conductivity in the cross-plane direction via the synergy of the double fillers [27]. Zhang et al. also found a greatly improved thermal conductivity in the poly(vinylidene fluoride) (PVDF)/CNTs/graphene oxide (GO) composites, compared with the PVDF/CNT composites at the same CNT content [28]. It was demonstrated that the introduction of GO is in favour of the dispersion of CNTs and the formation of denser CNT/GO network structure in the PVDF matrix. And Xiao et al. watched the synergistic effect between BN and MWCNT in the PVDF, where a small amount of CNTs was introduced into PVDF/boron nitride (BN) composites via melt blending processing [29]. The thermal conductivity of the ternary PVDF/BN/CNT composites was greatly improved, compared with the PVDF/BN composites at the same BN content. The mechanism for the largely enhanced thermal conductivity is mainly related to the formation of the denser 3D hybrid BN/CNT network structure.

Concerning the filler dispersion in polymer matrix for polymer composites with two different fillers, there are at least three combinations: (1) both of two filler are separately dispersed in polymer matrix as their filler content is low; (2) one of the filler forms network structure as its content is above the percolation threshold while another filler remains separately dispersed (single filler percolation); (3) both of fillers forms network structure as their content is above the threshold (double filler percolation). One expects totally different synergistic effect for polymer composites with different filler combinations. In our previous research, a sharp increase of thermal conductivity was observed in polypropylene (PP)/EG composites via introducing CNTs, and it is attributed to the formation of double percolated filler network with the dense MWCNT network located within loosened EG network [30]. In that work, only one content of EG (above its percolation threshold) was used to investigate the synergistic effect between EG and CNTs. Thus to investigate different filler synergistic effect, EG content below percolation and just percolation should be also carried out and compared with the ternary composites with EG content above the percolation threshold. More importantly, polymer matrix should be also extended to other ones to check the universality of filler synergy.

In this work, EG was individually added into another widely used polyolefin, high density polyethylene (HDPE) to fabricate the binary composites first through melt blending to determine the percolation threshold. Then HDPE/EG composites with three compositions representing below percolation, just percolation and above percolation, respectively, were chosen as matrix and melt mixed with CNTs to make HDPE/EG/CNTs ternary composites. In this way, the effect of filler combinations, including separated structure, single filler percolation, and double filler percolation on thermal conductivity of HDPE/EG/CNTs ternary composites could be comparatively investigated. We try to find whether the double percolated filler network can be also used to improve the thermal conductivity of HDPE as it works for PP, also for better understanding the synergistic mechanism between different fillers, via comparison study among composites with separated structure, single filler percolation and double filler percolation.

2. Experimental section

2.1. Materials

High density polyethylene (HDPE) was provided by ExxonMobil Chemical, China, which has a melt flow rate (MFR) of 1.9 g/10 min

and a density of 0.923 g/cm³. Multi-walled carbon nanotubes (CNTs, NC7000, 1.5 μm in length, diameter of 9.5 nm) were purchased from Nanocyl S.A, Belgium. Low temperature expandable graphite, the trade mark is ADT KP801, was supplied by Shiji-azhuang ADT Carbonic Material Factory (China).

2.2. Preparation of low temperature expanded graphite (EG)

EG was prepared by using a rapid heat-treatment at 350 °C for 7 min to expand the low temperature expandable graphite to obtain the wormlike graphite. Then, it was smashed to get fine EG sheets at a high speed by the functional grinder (BJ-100, 25,000 rpm) for 30 s and the average lateral size of EG flakes is about 30 μm.

2.3. Preparation of HDPE/EG and HDPE/MWCNT binary composites

HDPE particles with various content of EG or MWCNT were pre-mixed via stirring for several minutes in a container. Melt blending method was applied to prepare the binary composites at 185 °C with 60 rpm/min for 10 min in an internal mixer (HaakeRC-90, Germany). Then, heat compression molding was adopted to prepare samples for measuring electrical conductivity and thermal conductivity, etc. The samples of the composites were molded at the temperature of 185 °C and under the pressure of 10 MPa for 10 min.

2.4. Preparation of HDPE/EG/CNTs ternary composite

HDPE particles, EG and CNTs were pre-mixed by stirring for 5 min in a container. Melt blending method was applied to fabricate the composite with a fixed EG content and various content of CNTs at 185 °C with 60 rpm/min for 10 min in an internal mixer (HaakeRC-90, Germany). Then, hot compression molding was applied to prepare samples for testing electrical conductivity and thermal conductivity, etc. The compositions of the binary and ternary composites have been summarized in Table 1.

2.5. Characterization

The transient place source (TPS) method was adopted to test the thermal conductivity of the composites via a Hot Disk thermal analyzer (Hot Disk 2500-OT, Uppsala, Sweden). Based on this method, the sensor with a diameter of 2 mm was placed between two round samples with the diameter of 25 mm and thickness of 4 mm. P2400 SiC paper was applied to polish the composite's surface on the purpose of accurate thermal conductivity.

The electrical conductivity of the samples was measured with a Keithley 6487 picoammeter under a constant voltage of 1 V to avoid strong electric current within the samples. The samples are rectangular cross-sectional bars cut from the compression-molded plate and have a length of 50 mm, a width of 5 mm and a thickness of 3.5 μm. In order to avoid the contact resistance, silver paint was brushed on the edge of the samples.

The tensile samples are dumbbell-shaped bars and the mechanical properties were measured by a universal tester (SANS, China) at a cross-head speed of 5.0 mm/min according to the ISO 527-3

Table 1
Sample notation and the compositions of the samples.

Sample	HDPE (wt%)	EG (wt%)	CNT (wt%)
HDPE	100	0	0
HDPE/xEG	100 – x	x	0
HDPE/yCNT	100 – y	y	0
HDPE/xEG/yCNT	100 – x – y	x	y

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