



Enhancing thermal conductivity and mechanical properties of poly (methyl methacrylate) via adding expanded graphite and injecting water



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ABSTRACT

Poly(methyl methacrylate)/expanded graphite (PMMA/EG) composites are prepared using water-assisted mixing extrusion (WAME) technique. Scanning and transmission electron microscopy micrographs show that the EG exhibits better exfoliation and dispersion in the composite samples prepared with water injection than that in those without water injection. More EG layer networks and stronger PMMA-EG interfacial interaction are formed in the composite samples prepared with water injection, which are confirmed by the results of the FTIR and rheological measurements and the linear fits of experimentally obtained tensile yield stresses. The composite samples with well exfoliated and dispersed EG layers exhibit higher thermal conductivity and mechanical properties, which are ascribed to lower interfacial thermal resistance and stronger interfacial interaction, respectively. Finally, a mechanism for promoted EG exfoliation and dispersion in the PMMA matrix during the WAME is interpreted by analyzing the combined effect of injected high-pressure water with the shear force provided by the extruder screw.

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1. Introduction

Polymers are generally difficult to meet application demands in microelectronic and micromechanical devices due to their low thermal conductivities. Therefore, some efforts are carried out to enhance the thermal conductivities of polymers. Introducing carbon-based thermal conductive fillers, such as expanded graphite (EG) [1–5], carbon nanotube [6–8] and graphene [9–13], into polymer matrices is an effective method. EG, exhibiting two-dimensionally layered structure, is regarded as the most promising candidate due to its relatively low cost and high thermal conductivity. However, EG is not easily exfoliated in polymer matrices due to the van der Waals force between graphite layers. Enhancing interfacial interaction between polymer and graphite [1,5] and/or achieving better exfoliation and dispersion of graphite in polymer matrix [3] can significantly improve the thermal conductivity of polymer composite. Ganguli et al. [5] investigated the effect of 3-aminopropyltriethoxy silane functionalization of graphite on the thermal conductivity of epoxy/graphite composite. The results indicated that grafting silane onto the graphite enhances interfacial interaction between epoxy matrix and graphite by forming covalent bonds, thereby resulting in increased thermal conductivity.

Fu et al. [3] adopted an internal mixer to prepare polyvinylidene fluoride/EG composite. The results showed that the EG is effectively exfoliated to graphite nanosheets in the matrix and the thermal conductivity of the composite is improved due to pre-treatment of EG via ball milling.

Interfacial interaction between polymer matrix and filler affects not only the thermal properties, but also the rheological and mechanical properties for polymer composites. For example, the research by Ronca et al. [14], who prepared high density polyethylene/reduced graphene oxide nanoplatelets composite, demonstrated that the melt rheology and crystallization behavior of matrix are obviously affected by the reduced graphene oxide. This is attributed to strong interfacial interaction between polymer chains and filler. Pukánszky et al. [15] proposed a semi-empirical equation for tensile yield stress of polymeric composites. The equation presents an interaction parameter (B) that is connected with the interfacial properties between polymer and filler. Via fitting experimental data using the Pukánszky equation, parameter B can be obtained to quantitatively characterize the interfacial interaction between polymer and filler [16–18].

In situ polymerization, solution mixing and melt mixing are common methods for preparing polymer/EG composites. The former two methods favor better exfoliation and dispersion of EG in polymer matrices due to solution environment [19]. Poly(methyl methacrylate) (PMMA)/EG composites with better electrical

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conductivity were prepared using in situ polymerization [20] and solution mixing [21] methods. However, the two methods have some drawbacks, such as complex preparation process. To the best knowledge of the authors, there is no report on preparing PMMA/EG composites via direct melt mixing. Water-assisted mixing extrusion (WAME), a method that combines the benefits of both solution and melt mixing methods, favors better exfoliation and dispersion of layered fillers, such as clay [22–26] and graphene oxide (GO) [27,28]. Very recent research performed by the present authors [28] demonstrated that the compatibilization effect of the GO for polystyrene/poly(vinylidene fluoride)-GO blend nanocomposites is promoted by the injected water. Considering the swellability of EG in solution, it is expected to achieve better exfoliation and dispersion of EG in polymer matrix using WAME. So, WAME is used to prepare PMMA/EG composites in this work.

2. Experimental section

2.1. Materials

PMMA resin and EG powder were used in this work. The PMMA (grade Degussa 8H, Germany) has a density of 1.19 g/cm³. The EG (grade XF057, Nanjing XFNANO Materials Tech Co. Ltd, China) has a carbon content of 98%, an expansion coefficient of 350 cm³/g, a density of 2.26 g/cm³ and an average lateral size of about 300 μm.

2.2. Sample preparation

The PMMA pellets were dried in a vacuum oven at 80 °C for 4 h. The PMMA/EG composites were prepared using the WAME setup described in our previous work [28,29]. The mixture of the PMMA pellets and EG powder was added to the hopper of the twin-screw extruder. The PMMA/EG composite samples with three different EG contents (1, 4, and 7 wt%) were prepared by using the WAME. The screw speed was 100 rpm, and the weight ratio of injected water to fed PMMA/EG mixture was 1:2. The extrudate was pelletized at the die exit. After drying in a vacuum oven at 80 °C for 24 h, the pellets were compression molded into sheets at 230 °C under 15 MPa. For comparison, the PMMA/EG composite samples without water injection and the pure PMMA samples with and without water injection were also prepared at the same processing conditions.

The PMMA samples prepared without and with water injection were denoted as P and P-W, respectively; the PMMA/EG composite samples prepared without and with water injection were denoted as P-EG_m and P-EG_m-W (where m represented the weight percentage of the EG), respectively.

2.3. Characterization

Specimens cut from the aforementioned sheets were cryofractured in liquid nitrogen. The fractured specimens were then gold-sputtered and examined using a scanning electron microscopy (SEM; Nova NanoSEM 430, FEI) at an accelerating voltage of 15 kV.

The ultrathin slices (~200 nm) were cut from the sheets using an ultramicrotome (EM UC6+FC6, Leica, Germany). Then transmission electron microscopy (TEM; JEM-2100F, Japan) observation was performed on the slices at an accelerated voltage of 200 kV to characterize the exfoliation and dispersion of the EG in the PMMA matrix.

Wide angle X-ray diffraction (WAXD) measurements were conducted using a D8 Advance X-ray diffractometer (Bruker, Germany; Cu Kα, λ=0.154 nm, 40 kV, 40 mA) for the EG powder and

the composite specimens that were cut from the sheets. The measurements were performed at 2θ angle of 2–40°, a scanning rate of 2°/min, and a scanning step of 0.013°.

Powder (about 1 mg) scraped from the sheets was finely ground with dried KBr at a weight ratio of approximately 1:200 (sample to KBr) in an agate mortar and then pressed into a disk. Fourier transform infrared spectroscopy (FTIR) tests were performed on the disk using a Vertex 70 spectrometer (Bruker, Germany) in transmission mode. The scanning range was from 4000 to 400 cm⁻¹ with a resolution of 4 cm⁻¹.

Thermogravimetric analyses were conducted using a thermogravimetric analyzer (TGA; STA449C, Netzsch). The specimens cut from the prepared pellets were heated from room temperature to 600 °C at a heating rate of 10 °C/min under nitrogen atmosphere.

Thermal conductivity was measured using a Physical Property Measurement System (PPMS-9, USA) at 30 °C. The disks with a diameter of 5 mm and a thickness of 1 mm for measurements were cut from the sheets.

Rheological tests were conducted by a dynamic oscillatory rheometer (Bohlin Gemini 200, Malven Instruments) with 25 mm diameter parallel plate geometry at a temperature of 230 °C. The fixed strain and oscillation angular frequency (ω) range used were 1% and 0.0628–628 rad/s, respectively. The disks with a diameter of 25 mm and a thickness of 1 mm for tests were prepared by compression molding the prepared pellets.

Tensile tests were carried out using a universal testing machine (Instron 5566, USA) at a strain rate of 2 mm/min under room temperature. Five specimens with a dumbbell shape prepared by compression molding the prepared pellets were used in each tensile test and the average value was calculated.

3. Results and discussion

3.1. Microstructure of EG in PMMA matrix

The SEM micrographs of the P-EG7 and P-EG7-W samples are shown in Fig. 1a and b, respectively. The exfoliation and dispersion of the EG in the PMMA matrix are poor and some large aggregates (with diameter up to about 120 μm, as marked by cycles in Fig. 1a) are observed in the P-EG7 sample. In the P-EG7-W sample, although a few aggregates (with a maximum diameter of about 30 μm) exist, better exfoliation and dispersion and even graphite nanoplatelets (as marked by arrows in Fig. 1b) of the EG are achieved. Fig. 2 shows the TEM micrographs for the P-EG7 and P-EG7-W samples, which more clearly show an improved EG exfoliation in the P-EG7-W sample.

As can be seen at the WAXD patterns shown in Fig. 3, the EG characteristic peak in the P-EG7-W sample exhibits lower peak intensity than that in the P-EG7 sample, which further confirms better exfoliation and dispersion of the EG in the former sample. Recent research by Carreau et al. [24] also demonstrated that poly(ethylene terephthalate)/Cloisite 30 B sample prepared by WAME possesses a lower peak intensity of Cloisite 30 B than that prepared without water injection.

3.2. Interfacial interaction between PMMA and EG layers

Fig. 4 shows the FTIR spectra of the P, P-EG7 and P-EG7-W samples. The P sample exhibits an absorption peak at 1734.61 cm⁻¹ corresponding to carbonyl stretching vibration. The absorption peak of carbonyl stretching vibration in the P-EG7 sample is red-shifted to 1729.74 cm⁻¹. This shift to lower energy can be ascribed to the delocalization of carbonyl electrons to the π-clouds of EG, causing a decrease in carbonyl bond order [30]. Interestingly, the

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