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Short Communication

Improved thermal conductivities in polystyrene nanocomposites by incorporating thermal reduced graphene oxide *via* electrospinning-hot press technique

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ABSTRACT

Thermal reduced graphene oxide (TRG) was performed to obtain the TRG/polystyrene (TRG/PS) composite fibers by solution blending followed by electrospinning technique. And then the thermally conductive TRG/PS nanocomposites were fabricated *via* hot press method. The thermally conductive coefficient (λ), thermal diffusion coefficient (α), glass transition temperature (T_g) and heat-resistance index (T_{HRI}) of the obtained TRG/PS nanocomposites were all improved with the increasing addition of TRG. The addition of 15 wt% TRG could increase the λ value of pure PS from 0.226 W/mK to 0.689 W/mK, a value from 0.2157 mm^2/s to 0.6545 mm^2/s, T_g value from 90.3 °C to 95.0 °C and T_{HRI} value from 184.2 °C to 194.3 °C.

1. Introduction

Polystyrene (PS) has been widely applied in the fields of electronic industry and food packing, *etc.*, owing to its stable physical and chemical properties, outstanding chemical corrosion resistance, as well as excellent thermal stability [1–4]. However, the intrinsic low thermally conductive coefficient (λ) value of pure PS matrix has restricted its broader application in the thermal conduction fields [5–8]. Under this circumstance, the λ value of the PS matrix needs to be improved, effectively to prolong its working life and expand its application fields [9–11].

At present, the most common method to prepare PS-based thermally conductive composites is to introduce single or hybrid thermally conductive fillers [12,13]. Wu et al. [14] fabricated the aluminum nitride/PS (AlN/PS) composites *via* the method of powder blending. The obtained λ value of the AlN/PS composites with 25 wt% AlN was 0.418 W/mK, two times than that of pure PS matrix. Han et al. [15] also introduced boron nitride (BN) fillers into PS matrix *via* solution blending. When the mass fraction of BN fillers was 30 wt%, the obtained λ value of the BN/PS composites was 0.692 W/mK, about three times than that of pure PS matrix.

To our knowledge, by incorporating hybrid thermally conductive fillers with different shape is in favor of forming effective contacts, finally to improve the λ value of the composites with the same addition of thermally conductive fillers. Cui et al. [16] incorporated hybrid fillers of BN and graphene nanosheets (GNS) into PS matrix to prepare thermally conductive BN/GNS/PS composites. The obtained λ value of the BN/GNS/PS composites with 21.5 wt% hybrid fillers (20 wt% BN + 1.5 wt% GNS) was improved to 0.660 W/mK, higher than that of PS composites filled with the same loading single BN or GNS. Wu et al. [17] also adopted multi-wall carbon nanotubes (MWCNTs) and graphite nanosheets (GNPs) as hybrid fillers to prepare the thermally conductive (PS/MWCNTs)@GNPs composites. When the volume fraction of hybrid fillers was 9.0 vol% (5.5 vol% MWCNTs and 3.5 vol% GNPs), the obtained λ value of the PS composite was 1.05 W/mK, higher than that of PS composites filled with the same loading single MWCNTs or GNPs. In our previous work [18], functionalized silicon carbide whisker (SiCw) and silicon carbide particle (SiCp) were employed to prepare PS/SiCw/SiCp thermal conductivity composites. The obtained λ value was improved from 0.180 W/mK for pure PS matrix to 1.290 W/mK for the PS composites with 40 vol% SiCw/SiCp (volume fraction, 3:1) hybrid fillers, higher than that of PS composites with the

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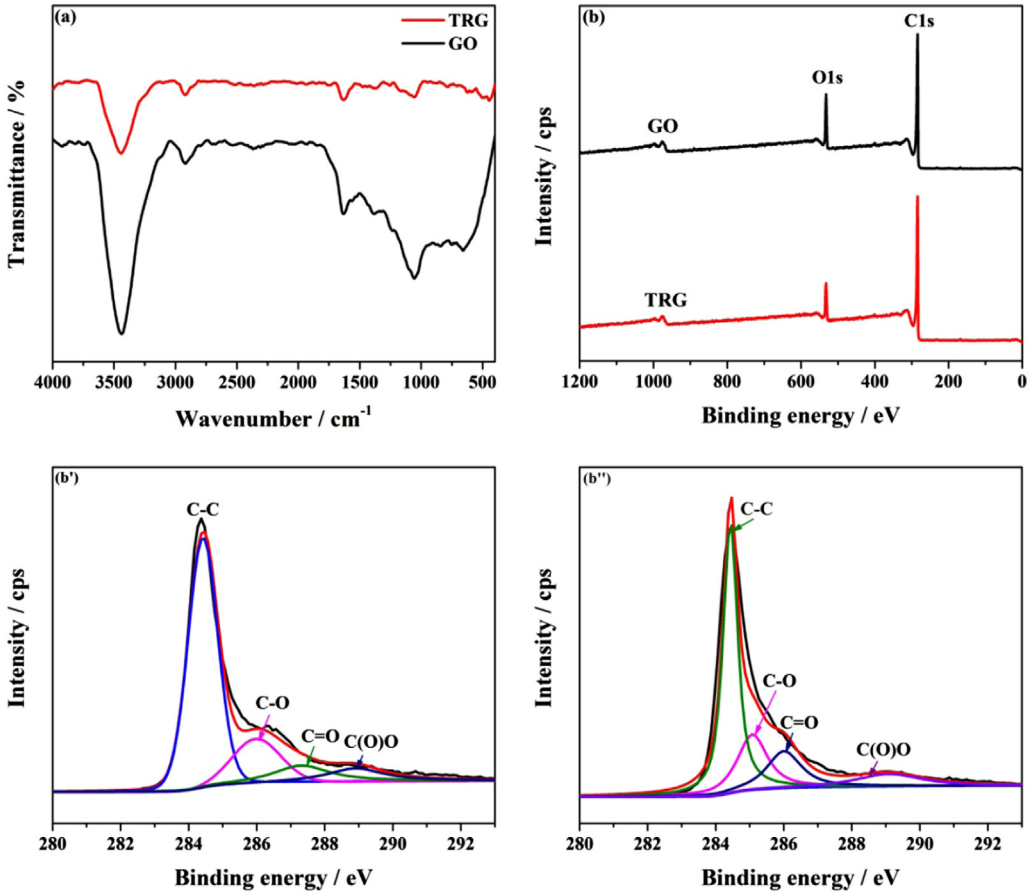

Fig. 1. FTIR (a) and XPS (b) curves of the GO and TRG.

same loading single SiCw or SiCp. In addition, compounding thermally conductive fillers with different particle size can also effectively reduce the gaps between fillers, to form thermally conductive networks or pathways more easily, finally to enhance the λ value of the composites [19,20]. However, the reported λ value of the filled PS-based composites is still well below expectations, mainly attributed to relatively poor interfacial compatibility and inevitable interfacial thermal barriers between thermally conductive fillers and polymeric matrix [12,13].

Reports revealed that the intrinsic λ value of the graphene was extremely high (theoretical λ value for monolayer graphene, about 5300 W/mK) [21–24], higher than that of carbon nanotubes [25–27], but also much higher than that of silver [28], copper [29] and alumina [30]. And the methods for preparing graphene mainly include mechanical exfoliation method [31,32], redox method [33,34], silicon carbide epitaxial method [35] and chemical vapor deposition method [36], *etc.*, Herein, Hummers method is reported as a simple and high-yield method [37–39].

Furthermore, compared with common methods (powder blending, solution blending and melt blending, *etc.*,), electrospinning technique is extremely simple and universal, presenting to be another effective method for improving the uniform dispersion and alignment of fillers [40–42]. In our previous work, silicon carbide particle/polystyrene (SiCp/PS) composites [43], boron nitride/polyimide (BN/PI) composites [44] and chemically modified graphene/polyimide (CMG/PI) nanocomposites [45] were fabricated based on electrospinning technology. Results revealed that the thermally conductive fillers could disperse uniformly inner polymeric matrix and align along the same direction as that of adopted polymeric fibers.

In our present work, thermal reduced graphene oxide (TRG) was firstly prepared from natural graphite flake *via* modified Hummers method, which was then performed to obtain the TRG/polystyrene (TRG/PS) composite fibers by solution blending followed by electrospinning technique. Finally, the corresponding thermally conductive TRG/PS nanocomposites were fabricated *via* hot press method. Fourier transform infrared (FTIR) and X-ray photoelectron spectroscopy (XPS) were adopted to analyze and characterize the structure and components of the graphene oxide (GO) and TRG. Scanning electron microscope (SEM) was also performed to observe the morphologies of the PS fibers and the fractures of the PS nanocomposites. Furthermore, the mass fraction of TRG influencing on the thermal conductivities and thermal properties of the thermally conductive TRG/PS nanocomposites were also discussed and investigated.

2. Experimental section

2.1. Materials

Natural graphite flake, 325 mesh, was supplied from Alfa Aesar Co., Ltd. (Shanghai, China); Polystyrene (PS), molecular weight 120,000, was purchased from Formosa PS Co., Ltd. (Ningbo, China); N, N-Dimethylformamide (DMF) and tetrahydrofuran (THF), analytical reagent, were both received from Guangzhou Guanghua Sci-Tech Co., Ltd. (Guangzhou, China).

2.2. Fabrication of thermally conductive TRG/PS nanocomposites

GO was firstly prepared by acid oxidation of natural graphite flake according to modified Hummers method [46]. TRG was then obtained by placing GO into a 200 °C tube furnace under a mixture of hydrogen-argon for 2 h. PS matrix was dissolved in the mixed solvent of DMF/THF (1/1, wt/wt), and TRG fillers were then added, to obtain the corresponding electrospinning solution after magnetically stirred for