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Thermally conductive phenol formaldehyde composites filled with carbon fillers

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ABSTRACT

Thermally conductive phenol formaldehyde resin (PF)/graphite composites were prepared and the effects of a small amount of carbon fibers and multi-walled carbon nanotubes on the through-plane and in-plane conductivities of the composites were investigated. The use of graphite flakes increases the thermal conductivity of PF resin. The increase in in-plane thermal conductivity is much higher than that in through-plane thermal conductivity due to the layered structure of graphite. By replacing 10 wt% of graphite with carbon fibers, the through- and in-plane thermal conductivities of the PF composite are as high as 8.6 W/(mK) and 31.5 W/(mK), respectively. The partial replacement also improves the flexural properties of the composites.

1. Introduction

With the rapid development of high density electronic devices, the growing demand for dissipation of generated heat has prompted great interest in thermally conductive materials [1–4]. The poor conductivity of polymers hinders their further applications in electronic fields despite the unique lightweight and good processibility. Therefore, thermally conductive fillers, such as aluminum nitride, boron nitride, graphite, carbon fibers (CFs) and carbon nanotubes (CNTs), were used to enhance thermal conductivities of polymers [1–3,5,6]. Among these fillers, cost-effective natural graphite has advantages in endowing polymers with high thermal conductance due to its high thermal conductivity and large aspect ratio [1,5]. CFs also exhibit significant reinforcement to polymers [6,7].

Thermally conductive fillers with different shapes are easy to form an efficient heat transfer network and thus two or more types of inorganic fillers were combined to further improve the thermal conductance of polymers [1–3,8]. Synergistic improvement in thermal conductivity of polyphenylene sulfide composites was achieved by using the mixture of boron nitride platelets and multi-walled carbon nanotubes (MWNTs) [8]. The well-dispersed CNTs effectively connect the adjacent CFs and significantly improve thermal conductance of the composites [6]. The present work aims to improve the thermal conductivity of PF/graphite composites by introducing small amounts of CFs or MWNTs. The influences of the graphite size and the surface modification of MWNTs on thermal conductivities of PF composites were investigated. The possible synergistic effect of graphite with CFs or MWNTs was also explored.

2. Experimental details

Graphite powders with mean sizes of 13 μm and 48 μm were provided by Huadong Graphite Factory (China) and designated as G13 and G48, respectively. The synthesis of PF resin was described elsewhere [9] and PF composites were prepared by blending carbon fillers with PF resin. PF resin was dissolved in ethanol in a beaker and then mixed with 10 wt% hexamethylene tetramine (curing agent) and carbon fillers for 10 h. After the solvent was removed, the obtained composite was dried in a vacuum oven. The crushed powders were finally compression-molded into specimens for property measurements. To investigate the synergistic effect of different fillers on thermal conductivity, the total filler content of PF composites is fixed at 70 wt% and CFs (diameter:10 μm, length: 200 μm), pristine MWNTs (p-MWNTs) (diameter: 20–30 nm, length: 10–30 μm) or carboxyl-functionalized MWNTs (f-MWNTs) were used to replace part of graphite G48.

The morphologies of the fillers and their PF composites were observed with a Hitachi S4700 scanning electron microscope (SEM). The through-plane and in-plane thermal conductivities of disk-shaped specimens with a diameter of 60 mm and a thickness

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of 5 mm were tested with an EKO HC-074 thermal meter and Hot Disk TPS1500 thermal analyzer, respectively. Flexural strength and modulus of the composites were measured on an Instron1185 universal testing machine using a three-point bending mode according to ASTM D790. At least five rectangular specimens with dimensions of $75 \times 10 \times 4$ mm^3 were measured.

3. Results and discussion

Fig. 1a shows the thermal conductivities of the PF composites filled with graphite flakes with average diameters of 13 μm (G13) and 48 μm (G48). It is clear that both through-plane and in-plane thermal conductivities increase gradually with the increase of graphite content, which would decrease the thickness of thermally insulating PF resin between adjacent graphite flakes and facilitate the formation of thermal transfer pathways. At low contents, the difference between through-plane and in-plane thermal conductivities is not considerable. With increasing the graphite content, however, the increase in in-plane thermal conductivity is faster than that of through-plane thermal conductivity due to the anisotropic feature of graphite [10]. With 80 wt% of G48, the through- and in-plane conductivities of its PF composite are 5.2 W/(mK) and 27.9 W/(mK), respectively. In addition, PF/G13

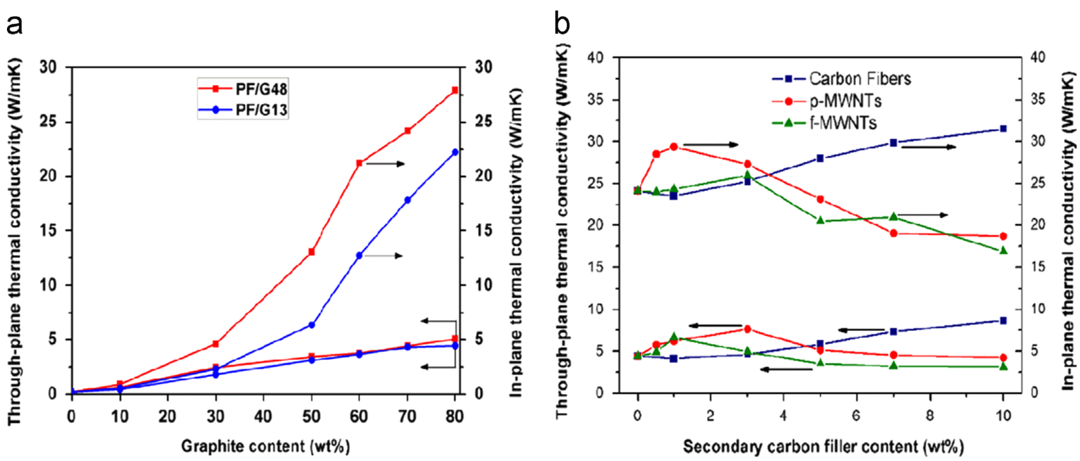

Fig. 1. (a) Plots of thermal conductivity versus content of graphite for PF/graphite composites; (b) plots of thermal conductivity versus content of CFs, p-MWNTs and f-MWNTs for PF/graphite (G48) composites. The total content of carbon fillers is kept at 70 wt%.

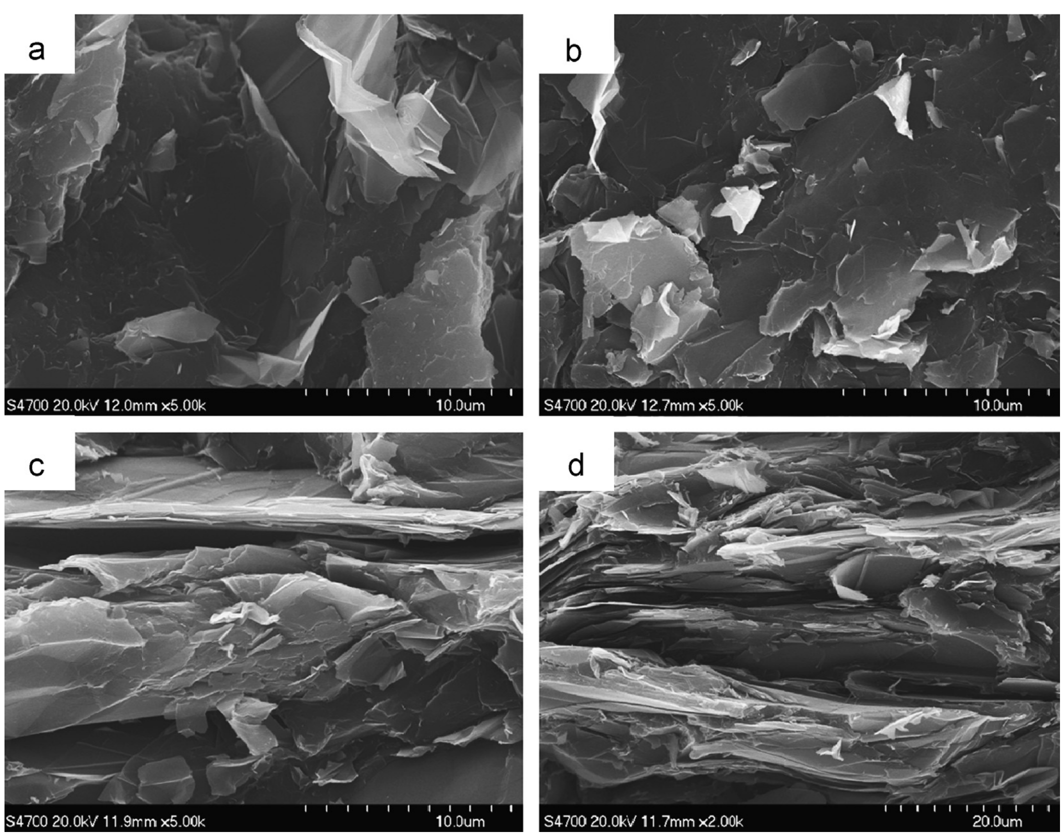

Fig. 2. SEM microphotographs of the PF composites with (a) 10 wt%, (b) 30 wt%, (c) 60 wt%, and (d) 80 wt% of graphite (G48).