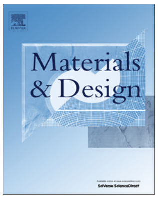
Materials and Design 51 (2013) 257–261

Contents lists available at SciVerse ScienceDirect

Materials and Design

journal homepage: www.elsevier.com/locate/matdes



Preparation and properties of polyamide 6 thermal conductive composites reinforced with fibers

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ARTICLE INFO

Article history:
Received 31 October 2012
Accepted 23 March 2013
Available online 10 April 2013

Keywords:
Polyamide 6
Hybrid fillers
Fiber reinforced
Thermal conductivity
Mechanical
Heat deflection temperature

ABSTRACT

The majority of inorganic particles-filled thermal conductive composites highlight thermal conductivity in detriment of mechanical properties. In this work, magnesium hydroxide ($Mg(OH)_2$), alumina (Al_2O_3) and flake graphite-filed polyamide 6 (PA6) composites prepared by twin-screw extruder, were reinforced with carbon and glass fibers separately. Effects of fiber type and content on thermal conductivity, mechanical properties and heat deflection temperature (HDT) of the PA6-based composites were investigated. The results showed that the thermal conductivity of the composites improved with increasing carbon fiber content, while decreased slightly with glass fiber loading. Furthermore, strength, modulus and HDT of the PA6-based composites increased with the increase of fiber content. The reinforcing effects of the two fibers on the thermal and mechanical properties of the composites were compared and interpreted in this paper. By incorporating simultaneously high thermal conductive fillers and high-strength fibers, the combined composites hold a good potential in heat dissipation applications.

1. Introduction

Thermal conductive composite materials become more and more important in heat dissipation applications because of their lightweight, good chemical resistance, excellent insulation performance and economic fabrication [1–3]. A great attention was given to the filled thermal conductive polymer composites because they combined features of polymers and thermal conductive fillers, such as easy processing and the high thermal conductivity. In order to improve thermal conductivity of the composites, many kinds of thermal conductive fillers were introduced into the composites, such as aluminum nitride (AlN) [4–7], silicon nitride (Si_3N_4) [8,9], alumina (Al_2O_3) [10], boron nitride (BN) [11–13], graphite [14], and carbon nanotube [15,16].

Thermal conductivity of the polymer composites mainly depended on the thermal conductive chains or networks produced by the fillers, the high thermal conductive performance could be obtained by adding high fraction of fillers. However, high fraction of fillers deteriorated the mechanical properties of the filled polymer composites [17]. Efforts should be carried out to retain high thermal conductivity and good mechanical properties of the filled composites. Gu et al. [18] pointed out that the silicon carbide whisker (SiCw) and silicon carbide particle (SiCp) hybrid fillers were more favorable to improve the thermal conductivity of the composites, and high thermal conductivity could be obtained at a low SiCw/SiCp hybrid filler content. A low content of fillers could reduce the detrimental influence on mechanical properties of the composites. On the other hand, some reinforcements can be added to the composites to improve mechanical properties. For instance, Zhou et al. [19] reported that the composites with Al_2O_3 short fiber and Si_3N_4 hybrid fillers performed better mechanical properties than the original Si_3N_4-filled polymer.

In this work, magnesium hydroxide ($Mg(OH)_2$), Al_2O_3 and flake graphite hybrid fillers were used as thermal conductive fillers to prepare polyamide 6 (PA6)-based thermal conductive composites by twin-screw extruder. Carbon fiber and glass fiber were introduced separately to the composites in an attempt to obtain high mechanical properties while keeping high thermal conductivity. Effects of fiber type and content on thermal conductivity, mechanical properties and heat deflection temperature (HDT) of the composites were investigated to provide some practical guidance for the preparation of composites with high thermal conductivity and good mechanical performance.

2. Experimental details

2.1. Materials

Polyamide 6 (PA6) with the trade mark of IMNC 101, supplied by Shanghai Hersbit Chemical Co. Ltd. (China), was used as matrix resin in this work. Alumina (Al_2O_3) and magnesium hydroxide

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http://dx.doi.org/10.1016/j.matdes.2013.03.076

($Mg(OH)_2$) were used as thermally conductive fillers and particle reinforcements, which were provided by Zibo Linkai Chemical Co. Ltd. (China) and Dalian Yatai Science and Technology New Material Co. Ltd. (Chna), respectively. Flake graphite used as the highly thermally conductive fillers in the present study, was supplied by Nanjing Grf Carbon Material Co. Ltd. (China). Chopped glass fiber and carbon fiber were used as reinforcements, which were supplied by Jushi Group Co. Ltd. (China) and Shanghai Langyu Industry Co. Ltd. (China), respectively. The properties of the PA6 resin, fillers and chopped fibers are shown in Table 1.

2.2. Surface treatment of the fillers

In order to improve the binding quality of the filler/matrix interface, the Al_2O_3 and $Mg(OH)_2$ particles were subjected to a surface treatment using the silane coupling agent KH-550 ($NH_2-(CH_2)_3Si-(OC_2H_5)_3$). The silane coupling agent (2 wt.% of filler) was mixed with alcohol evenly, and then the solution was added to the fillers and stirred for 30 min with double cone mixer (Wuxi Xinbiao Powder Machinery Co. Ltd., China). At last the particles were dried in a vacuum oven at 85 °C for 10 h.

2.3. Composites preparation

Before blending, PA6, fillers and short fibers were dried at 85 °C in the vacuum oven for 4 h to remove the moisture. Fiber reinforced thermally conductive PA6 composites were manufactured by a SJ-36 twin-screw extrude machine (Nanjing Giant Machinery Co. Ltd.) at a screw speed of 30 Hz, melt temperature at 270 °C. The screw diameter in the extrude machine was 36 mm, and the length–diameter ratio was 50. First, Al_2O_3, $Mg(OH)_2$ and graphite were mixed by the weight ratio of 2:2:1. Then, PA6, the hybrid fillers and fibers were added to the twin-screw extrude machine through extruder feed system 1, 2 and 4, respectively (as shown in Fig. 1). The fixed weight fraction of hybrid filler was 45% and the weight fraction of fiber was controlled at 3% and 5% by changing the extruder feed rate. Finally, the granulated fiber reinforced thermally conductive PA6 composites were obtained. After that they were dried at 85 °C for 4 h, the granular composites were injection molded to obtain specimens with standard shapes.

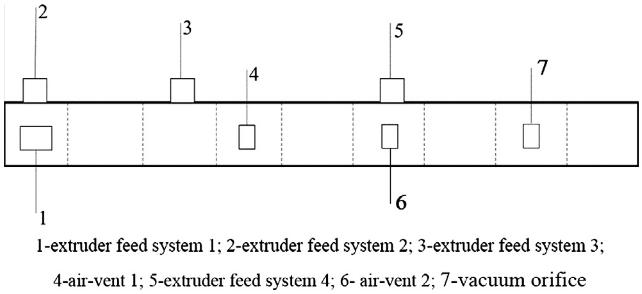

Fig. 1. Simple diagram of the extruder barrel part.

2.4. Characterization

Thermal conductivity of the composites was examined by TC 3010 apparatus (Xi'an Xiatech Electronic Technology Co. Ltd., China), and the thermal conductivity test was carried out according to ASTM C1113/C1113 M-09∗. The testing samples had dimensions of $40 \times 30 \times 2\ mm^3$.

BX41M-LED metallurgical microscope (Olympus Corporation, Japan) was used to observe the distribution of fibers in the composites after the surfaces of composites were polished.

Tensile tests were carried out at room temperature according to ISO 527-2:2012 [20] by using a computer controlled CMT-4304 universal testing machine (Shenzhen Suns Co. Ltd. Stock Technology, China) with a cross-head speed of 5 mm/min. Flexural tests were also conducted on the universal testing machine at room temperature according to ISO 178:2010 [21]. The notched impact strengths of the composites were tested with a XCJD-50 charpy impact tester (Chengde Puhui Testing Instrument Co. Ltd., Hebei, China) at room temperature according to ISO 179-1:2010 [22]. At least five samples were tested for each reported value.

Hatchi S-4800 field scanning electron microscope (SEM) was used to analyze the tensile fracture surfaces of the composites. All fracture surfaces were sputter coated with gold to reduce the incidence of surface charging in the scanning electron microscope.

HDT testing was conducted according to ISO 75-2:2004 [23] by means of HDT/V-110 hot deformation, vicat softening point temperature detector series (Chengde Jinjian Testing Instrument Co. Ltd., Hebei, China). In the experiments, the flexural stress was 1.80 MPa.

3. Results and discussion

3.1. Thermal conductivity of the composites

Fig. 2 shows thermal conductivity of the composites reinforced with different types and contents of fibers. For a given filler weight fraction, thermal conductivity of the composites increases with increasing content of carbon fiber, but decreases slightly with increasing content of glass fiber.

The effective thermal conductivity of polymer composites mainly depends on the interactions and connections among fillers. Thermal conductive fillers will produce thermal conductive chains or networks in the polymer matrix as the concentration of the fillers is up to a high level [17,24]. The thermal conductivity of the composites without fibers is 1.95 $W\ m^{-1}\ K^{-1}$ in the study, which is about seven times higher than the thermal conductivity of pure PA6. It illustrates that thermal conductive chains and networks are produced by fillers in the composites. When carbon fibers are added to the composites, the particles and carbon fiber hybrids can be used to enhance the thermal conductive chains and networks, where carbon fiber plays an important role as the thermal conductive bridge to connect the particles with each other, as carbon fiber has excellent thermal conductive performance. As the

Table 1
Properties of the PA6 resin, fillers and chopped fibers.

Properties	PA6	Al_2O_3	$Mg(OH)_2$	Graphite	Glass fiber	Carbon fiber
Thermal conductivity ($W\ m^{-1}\ K^{-1}$)	0.27	30	80	>190	1.1	26
Density ($g\ cm^{-3}$)	1.13	3.97	3.23	2.29	2.60	1.78
Purity (%)		⩾99	⩾99	⩾99		
Diameter (μm)		5–10	1–2	150	8	6
Length (mm)					6–8	4–6
Tensile strength (MPa)	68				1500	2000
Tensile modulus (GPa)	1.5				75	250