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Effect of nano-fillers on the thermal conductivity of epoxy composites with micro-Al₂O₃ particles

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ABSTRACT

Nano-AlN particles, AlN/graphene nano-hybrids (AlN/GE) and AlN/carbon nanotubes nano-hybrids (AlN/ CNTs) were prepared. The structures, morphologies of synthesized nano-materials were examined by X-ray diffraction (XRD), scanning electron microscope (SEM) and transmission electron microscope (TEM). The results showed that the morphologies of the synthesized nano-materials were obviously different. In addition, the thermal conductivity of epoxy composites could be effectively improved by adding the produced nano-fillers. Especially, the epoxy composite with AlN/GE nano-hybrids had the highest enhancement in thermal conductivity comparison to the pure epoxy. Moreover, the density of epoxy composites with the synthesized nano-fillers was decreased and the corresponding thermal stability was enhanced.

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1. Introduction

As the global demands for high-density electronic devices rise, high thermal conductive polymer materials have became more and more important in heat dissipation applications due to their lightweight, good chemical resistance, excellent insulation performance and economic fabrication [1,2]. Unfortunately, the thermal conductivity of conventional polymer materials was low, which restricted their applications. Thus, developing polymer-based composites with high thermal conductivity and low fabrication cost was of primary importance [3]. For the aim of producing the polymers with higher thermal conductivity, various fillers such as Al₂O₃, SiO₂, AlN, BN, and SiC had been added into polymers [4–6]. Theoretically, the high thermal conductivity was obtained by adding high fillers loading. However, the high content of fillers would result in a high density and poor mechanical properties of polymers.

Recently, carbon-based nano-fillers such as multi-walled carbon nanotubes, single-walled carbon nanotubes, graphene, and graphite nano-powder had been gradually applied as thermal conductive fillers due to their low specific gravity and super thermal conductivity [7–13]. Pak et al. had investigated the thermal conductivity of polyphenylene sulfide (PPS) composites with a mixture of boron nitride (BN) power and multi-wall carbon nanotube (MWCNT) fillers [8] and revealed that the thermal conductivity of PPS composites with 50 wt% BN was $1.00 \text{ W} (\text{mK})^{-1}$ while that of PPS composites with 50 wt% BN and 1 wt% MWCNT was increased to $1.45 \text{ W} (\text{mK})^{-1}$. Teng also [14] suggested that the thermal conductivity of epoxy composites containing 25 vol.% modified AlN and 1 vol.% functionalized MWCNTs could reach to $1.21 \text{ W} (\text{mK})^{-1}$, comparable to that of epoxy composites containing 50 vol.% untreated AlN ($1.25 \text{ W} (\text{mK})^{-1}$), which could reduce the half quantity of AlN filler used. Besides, they also reported that the thermal conductivity of the epoxy composites with 1 phr graphene ($0.86 \text{ W} (\text{mK})^{-1}$) was superior to that of MWCNT/epoxy composites ($0.32 \text{ W} (\text{mK})^{-1}$), which was due to that the graphene possessed higher surface area to contact with polymer in comparison with MWCNT [3].

The carbon nanotubes possessed the large aspect ratios and the graphenes had distinctive 2D structures [15]. These special structures could easily form the bridges or networks in polymers, known as thermal conductive chains and networks. The formation of random bridges or networks would facilitate phonon transfer and improve the thermal conductivity of polymer composites. Although these materials had such advantages, there were two main problems limiting the applications of them as the thermal conductive fillers: (1) the poor dispersion in polymer matrix, (2) the high cost. In order to overcome these drawbacks, the inorganic nano-particles were coated on CNTs, which was a convenient way to improve the quality of the interface between CNTs and polymer matrixes. Zhao et al. had synthesized silica coated on MWCNTs with different coating thicknesses and revealed that the silica layer could improve the interfacial interaction between MWCNTs and







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polymers and then enhance the dispersion of MWCNTs in the matrix [16]. Besides, hybrid filler system was another way to manufacture high performance polymer composites with low carbonbased nano-fillers content. Yang et al. denoted that the hybrid filler system with MWCNTs and SiC not only produced epoxy composites with high thermal conductivity, but also restricted the existence of the big filler agglomeration [17]. These studies indicated that the functionalization of CNT surfaces could be employed to promote well-dispersed conditions, which was beneficial to the establishment of thermal conductive paths and increased the thermal conductivity of composites. Moreover, the hybrid filler system could be utilized to obtain high thermal conductivity with relatively low cost.

It was well known that AlN had been considered as an attractive candidate among the conventional inorganic filler materials because of its excellent thermal conductivity (320 W/(mK)) [7,18], high electrical resistivity [19], and high mechanical strength [20,21]. In this present work, a strategy was to synthesize nano-fillers (AlN nano-particle, AlN/graphene (AlN/GE) nano-hybrids and AlN/carbon nanotubes (AlN/CNTs) nano-hybrids) for applying as fillers in thermally conducting composites. Then, the synthesized nano-fillers combining untreated micro-Al₂O₃ particles were added into epoxy composites. The effect of synthesized nano-fillers on the thermal conductivity of epoxy composites was investigated. The density and the thermal stability of composites were also studied.

2. Materials and methods

2.1. Materials

The aluminum chloride (AlCl₃·6H₂O, 97%, Sailboats Tianjin Chemical Reagent Co., Ltd.), urea (CON₂H₄, 99.0%, Sailboats Tianjin Chemical Reagent Co., Ltd.), anhydrous ethanol (C₂H₅OH, 99.5%, Tianjin Chemical Reagent Factory), nitrogen (N₂, 99.999%), graphene solution (Nanjing Nano Materials Technology Co., Ltd.) and carbon nanotubes (Beijing DK Nano Technology CO., Ltd) were used as raw materials to prepare AIN nano-particle, AIN/GE and AlN/CNTs nano-hybrids. All starting materials were purchased from commercial sources and were used without further purification. The concentration of the commercial graphene dispersion was 0.2 mg/ml and the dispersant was hexadecyl trimethyl ammonium bromide (CTAB). The commercial carbon nanotubes had the diameter of 10–30 nm and the length of 10–15 μm. Epoxy resin utilized in this work was a bisphenol-A epoxy resin and anhydride-curing agent (Tianjin Synthetic Material Research Institute). The micro-Al₂O₃ was provided by Zibo Linkai Chemical Co. Ltd. (China), which had the average diameter of 5 μ m.

2.2. Preparation of the nano-fillers

The fabrication process of AlN nano-particles, AlN/GE and AlN/ CNTs nano-hybrids was introduced in our previous studies [22,23]. The basic procedure employed for the synthesis of nano-fillers was to heat compounds precursor at an appropriate temperature. Firstly, aluminum chloride and urea were dissolved in anhydrous ethanol to obtain a concentrated solution, respectively. The urea/metal (urea/AlCl₃·6H₂O) molar ratio was set to be 6 for every sample. The graphene (carbon nanotubes) solution was sonicated for 1 h before used, and then, was introduced into the urea solution. Three kinds of urea solution were obtained: pure urea solution, urea solution with 1.0 mg graphene, and urea solution with 0.04 g carbon nanotubes. Then, the aluminum chloride solution was added slowly into the three urea solution at 70 °C, respectively. The obtained compounds were separated by filtration

Table 1

The name of therma	l conductivity epoxy	composites specimen.
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Sample	EP	Al ₂ O ₃	AlN	AlN/GE	AIN/CNTs
name	(wt%)	(wt%)	(wt%)	(wt%)	(wt%)
EP AO AO/AN AO/AN/GE AO/AN/CNT	100 50 50 50 50 50	50 45 45 45	5	5	5

and dried at 80 °C for 5 h. Finally, the compounds were put into a tube furnace and heated under flowing N₂ at 1000 °C for 5 h. The resultant nano-fillers fabricated from different urea solution samples were named as nano-AlN, AlN/GE nano-hybrid and AlN/CNTs nano-hybrid, respectively.

2.3. Preparation of epoxy composites

Before blending, the fillers (Al₂O₃, nano-AlN, AlN/GE nanohybrid and AlN/CNTs nano-hybrid) were dried at 85 °C in the vacuum oven for 5 h to remove the moisture. After that, the composites were prepared by solution blending and casting method, which involved in (a) stirring epoxy resin and curing agent (weight fraction ratio of epoxy resin to curing agent was 4:3) for 20 min and then pumping in vacuum oven at 40 °C, (b) adding the pretreated filler systems to the resin and continuing stir for 30 min, (c) casting the mixture in the mould and repeatedly degassing the mixture in vacuum drying oven at 40 °C until no air bubble appears on the surface of mixture, (d) curing the mixture at 90 °C for 5 h and (e) cooling to room temperature and then demoulding. The different filler systems and the corresponding prepared epoxy composites were summarized in Table 1.

2.4. Characterization

The nano-fillers were characterized by X-ray diffraction (XRD) for phase identification using Cu K₂-radiation. XRD measurements were performed on a Bruker D8 Advanced X-ray diffractometer from 20° to 80° with a scan speed of 4°/min. The morphologies and microstructures of nano-fillers, commercial graphene nanosheets and commercial carbon nanotubes were characterized by scanning electron microscope (SEM; FEI Nanosem 430, FEI, Hillsboro, OR) and transmission electron microscope (TEM; FEI Tecnai G2 F20, FEI). The thermal conductivity of the epoxy composites was examined by TC 3010 apparatus (Xi'an Xiatech Electronic Technology Co. Ltd., China), which was a Hot Disk thermal analyzer. The equipment was based upon the transient plane source method. The measurements were carried out according to the ISO 22007-2:2008 test method for the determination of the thermal conductivity and thermal diffusivity [24,25]. Fig. 1 showed the apparatus for the thermal conductivity measurement. The sensor was placed between two identical samples which had dimensions of $40 \text{ mm} \times 30 \text{ mm} \times 2 \text{ mm}$. During testing, the senor supplied a heat pulse to the samples and then the associated change in temperature as a function of time was recorded. Finally, the thermal conductivity of the samples was obtained by fitting the recorded data. Moreover, the hot disk technique had been proven to be a highly effective and accurate method to measure the thermal conductivity of various materials, which had been applied in many Refs. [9,13,26-29]. In this paper, for the aim of reducing the testing error, the thermal conductivity was conducted using five samples and each sample was tested three times. The given thermal conductivity was the average of the obtained results. Therefore, the results of the measured thermal conductivity were credible. The densities of composites specimens were measured Download English Version:

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