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The effect of interfacial state on the thermal conductivity of functionalized Al_2O_3 filled glass fibers reinforced polymer composites

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ABSTRACT

Rapidly increasing packaging density of electronic devices puts forward higher requirements for thermal conductivity of glass fibers reinforced polymer (GFRP) composites, which are commonly used as substrates in printed circuit board. Interface between fillers and polymer matrix has long been playing an important role in affecting thermal conductivity. In this paper, the effect of interfacial state on the thermal conductivity of functionalized Al_2O_3 filled GFRP composites was evaluated. The results indicated that amino groups-Al_2O_3 was demonstrated to be effective filler to fabricate thermally conductive GFPR composite (1.07 W/m K), compared with epoxy group and graphene oxide functionalized Al_2O_3. It was determined that the strong adhesion at the interface and homogeneous dispersion of filler particles were the key factors. Moreover, the effect of interfacial state on dielectric and thermomechanical properties of GFRP composites was also discussed. This research provides an efficient way to develop high-performance GFRP composites with high thermal conductivity for integrated circuit packaging applications.

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1. Introduction

During the past few decades, glass fibers reinforced polymer (GFRP) composites, which are used as substrates to support and link electronic devices are crucial components in the field of electronic packaging. With the continuing development of miniaturization and power density in electronic devices [1–3], efficient heat removal has become more and more important for performance and reliability for GFRP composites. Therefore, GFRP composites must possess good thermal conductivity to dissipate the heat generated and also have electrical insulation to avoid short circuits and delay of signal propagation, which in turn provides better electronic device performance. In order to circumvent the low thermal conductivity of GFRP, addition of thermally conductive fillers is a viable approach for designing new GFRP composites. Up to now, various inorganic fillers with high thermal conductivity have been added into the composites, such as silicon carbide (SiC) [4,5], aluminium nitride (AlN) [6,7], boron nitride (BN) [8–10] and alumina (Al_2O_3) [11–13]. However, simply adding filler particles into the composites did not obtain the ideal materials, mainly due to the high interfacial thermal resistance.

According to earlier study [14], the interfacial thermal resistance partly comes from a barrier to the heat flow induced at the interface, because of the phonon spectra mismatch between the inorganic and polymer phase. Another source of thermal interfacial resistance refers to the imperfect physical contact and a weak interfacial adhesion between inorganic filler and polymer. To solve the problem, surface chemical functionalization of fillers is considered to be an effective way. The surface chemical functionalization of fillers can form covalent bridge bonds, leading to the improved interfacial adhesion, which can minimize the interfacial phonon scattering and decrease the interfacial heat resistance by interpenetrating particle–resin and particle–particle interfaces [15]. Many researches have investigated the role of chemical functionalization of fillers on the reduction of interfacial thermal resistance. For example, Yang et al. [16] demonstrated that carbon nanotubes (CNTs) functionalized with amine groups can effectively reduce the interfacial thermal resistance, leading to improved thermal conductivity of epoxy resin. A recent study demonstrated that [3] polyhedral oligosilsesquioxane (POSS) functionalized boron nitride nanotubes (BNNTs) based epoxy composites show a dramatic enhancement of thermal conductivity (1360% improvement at

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30 wt% loading). The chemical surface functionalization by POSS molecules is mostly responsible for improving the thermal conductivity. Huang et al. reported [15] on a systematic study of the effect of interface on the thermal conductivity of epoxy based composites. They claimed that the thermal conductivity enhancement was mainly dependent on the physicochemical nature of the modifiers and surface functionalization of fillers. Although lots of works have shown that chemical functionalization can improve interfacial adhesion strength and thus higher thermal conductivity, there are few researches focusing on comparison of surface functionalization between chemical grafting and physical adsorption.

In this work, we try to understand this matter by investigating the effect of surface functionalization of Al_2O_3 on the thermal conductivity of GFRP composites. Chemical grafting and physical absorption are achieved by two silane coupling agents and graphene oxide (GO), respectively. Alumina (Al_2O_3) is chosen as filler instead of other thermally conductive fillers for its low cost and high electrical resistivity, which ensures the potential to be used in a vast range of applications. The results indicate that functionalization with amino groups is an effective method for preparing GFRP composites with high thermal conductivity. The strong adhesion at the interface and homogeneous dispersion of fillers are suggested to be the major driving forces. Al_2O_3 particles are successfully encapsulated by GO sheets via electrostatic attraction. However, the improvement in thermal conductivity is restricted by the existence of GO–Al_2O_3 clusters. Moreover, the effect of interfacial state on dielectric and thermomechanical properties of GFRP composites is also discussed.

2. Experimental

2.1. Materials

Alumina (Al_2O_3) particles with average sizes of 200 nm were purchased from Shenzhen Jingcai Chemical Co., Ltd (Shenzhen, China). 4,4′-Bismaleimidodiphenylmethane (BMI) was purchased from Honghu Bismaleimide Resin Factory (Honghu, China), 2,2′-bis (4-cyanatophenyl) propane (BCE) was purchased from Heijang Kinlyuan Pharmaceutical (China), and 2,2′-diallyl bisphenol A (DBA) was purchased from Wuxi Resin Factory (Wuxi, China). The above three raw materials were used to prepare bismaleimide-triazine (BT) resin as the polymer matrix, according to our previous report [17]. Graphene oxide sheets were purchased from Shanxi Institute of Coal Chemistry (Shanxi, China). The major components of BT resin and information of commercial GO are shown in the Supporting Information (see Figs. S1 and S2, Table S1 in Supporting Information). The coupling agents (KH-550 and KH-560) were purchased from Sinopharm Chemical Reagent (Beijing, China). Other materials and reagents were also purchased from Sinopharm Chemical Reagent (Beijing, China).

2.2. Surface functionalization of Al_2O_3

2.2.1. Preparation of epoxy groups-Al_2O_3

Prior to surface functionalization, Al_2O_3 particles were dried in an oven for 4 h at 110 °C. After cooling down to room temperature, 15 g Al_2O_3 particles were suspended in 150 ml of 95% ethanol solution to which 1.5 g of KH-560 was slowly added. The system was stirred at 70 °C for 12 h. The functionalized Al_2O_3 particles were isolated through centrifugation and quickly washed by fresh ethanol. Finally, the product was dried in a vacuum oven for 12 h to yield fried powder.

2.2.2. Preparation of amino groups-Al_2O_3

15 g Al_2O_3 particles and 35 ml dimethylbenzene were mixed into a three-neck flask. Under mechanical stirring condition, the system was then heated up to 115 °C with 1 g KH-550 slowly added. The resulting mixture was stirred at 115 °C for 6 h. The following processes were the same as functionalization of epoxy groups.

2.2.3. Preparation of GO-encapsulated Al_2O_3

50 ml GO aqueous solution and 100 ml ultrapure water were mixed into a three-neck flask, followed by ultrasonic treatment. Amino groups-Al_2O_3 particles were slowly added into the mixture by stirring. After stirring for 3 h, the GO-encapsulated Al_2O_3 hybrid fillers were prepared through filtration. Then the product was dried in a vacuum oven for 12 h to yield fried powder. Fig. 1 shows the scheme of surface functionalization of Al_2O_3 with different modifiers.

2.3. Preparation of functionalized Al_2O_3 filled GFRP composites

A certain amount of functionalized Al_2O_3, BT resin, curing agent and dispersant were suspended in methyl ethyl ketone. After a process of ultrasonic treatment for 30 min, a golden well-distributed solution was prepared. The resulting solution was then roll-coated on glass fibers, followed by drying in oven to evaporate the organic solvent. Then the prepared composite film was put into the laminating machine under condition of high temperature and vacuum for further curing process. Finally, functionalized Al_2O_3 filled GFRP composites were cured by the programmed heating process at high temperature. The detailed composition of the prepared samples is shown in Table S2 in the Supporting Information.

2.4. Characterization

Scanning electron microscopy (SEM) (Nova NanoSEM 450, FEI) was used for observation of fractured morphology of the composite samples. The morphology of GO-encapsulated Al_2O_3 was investigated on FEI Tecnai G2 F20 S-TWIN transmission electron microscope (TEM). Infrared absorption spectra of the functionalized Al_2O_3 were measured using a Fourier transform infrared (FTIR) spectrometer (Bruker Vertex 70). The thermal conductivities of the composites were measured by LW-9389 TIM Thermal Resistance and Conductivity Measurement Apparatus (Long Win Science & Technology, Taiwan) which was based on ASTM D 5470-06 Standard. The thermal conductivity λ was calculated by Eq. (1):

$$\lambda = -\frac{Q \cdot L}{A \cdot \Delta T} \tag{1}$$

where

λ is the thermal conductivity, W/m K;
Q is the heat flux, W or J/s;
L is the thickness of the sample, m;
A is the area of a sample, m^2;
ΔT is the temperature difference between temperature sensors of the hot meter bar, K.

The measurement of dielectric properties including effective dielectric constant (ε_{eff}) and dielectric loss ($\tan\delta$) was taken on an Agilent-4294A impedance analyzer with an applied AC voltage of 500 mv and frequency range of 100 Hz–10 MHz. The used samples were disk shaped, and both sides of the samples were coated with silver paste. Dynamical mechanical analysis (DMA) tests were carried out using a dynamic mechanical analyzer (TA Q800), in multi-frequency strain mode with amplitude of 10 μm. The specimen dimensions were ~20 × 8 × 1 mm^3. Measurements were performed from 30 to 300 °C, at a heating rate of 3 °C min^{-1}.

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