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Creation of a multilayer aluminum coating structure nanoparticle polyimide filler for electronic applications

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ABSTRACT

Polymer-based composites were prepared by the embedding of core-shell Al@Al₂O₃@SiO₂ (noted as Al*@SiO₂) filler particles into a polyimide (PI) matrix. The effects of Al*@SiO₂ filler content (0.1–0.4 volume fraction (vf)) on the thermal conductivity, relative permittivity, and dielectric loss were then investigated. The obtained Al*@SiO₂/PI composites showed remarkably improved high thermal conductivity and low relative permittivity. The maximum thermal conductivity of the composites with a 0.4 vf Al*@SiO₂ filler was 3.1 W/mK, while the relative permittivity and dielectric loss were 20.7 and 0.015 at 1 MHz, respectively. Compared with pure metal Al particles composites, the use of core-shell Al*@SiO₂ nanocomposites increased composite thermal conductivity and reduced composite permittivity. The relative mechanism was studied and discussed. This work helps pave the way for the identification of proper modifier fillers for polymer matrix nanocomposite materials.

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1. Introduction

Microelectronic packaging continues to play an increasingly important role in the rapid progress of electronic and electrical technologies. The heat generated by these devices during use must be efficiently directed to a heat sink and then dissipated into the ambient environment. As such, in addition to the traditional physical and mechanical properties, packaging materials must possess good thermal conductivity, low relative permittivity, and low dielectric losses to reduce the signal propagation delay to ensure better device performance.

Common polymers such as polyethylene, polypropylene, polyamide, acrylonitrile–butadiene–styrene, and epoxy resins generally have low thermal conductivities. As a result, they cannot effectively dissipate heat when used in various devices and their high thermal expansion coefficients result in thermal failure. Therefore, increasing the thermal conductivity of packaging materials would open up large new markets. Polymer-based composite materials with high thermal conductivity and low dielectric

permittivity have attracted considerable attention [1–4]. The addition of thermal conductive fillers to polymer remarkably improves the thermal behavior and dielectric properties of packaging materials. Studies concerning thermal conductive polymer composites are particularly focused on the use of different kinds of ceramics, such as boron nitride (BN) [5,6], aluminum nitride (AlN) [7–9], silica (SiO₂) [10], silicon nitride (Si₃N₄) [11,12], alumina (Al₂O₃) [13–15], and silicon carbide (SiC) [16,17]. However, in most of these studies, their thermal and other properties seem insufficient, leaving much room for improvement in the search for balance in electrical and thermal management.

In this work, metal Al nanoparticles were chosen as fillers due to their high thermal conductivity ($\lambda=237$ W/mK). To ensure the insulating property of the packaging materials, SiO₂-coated Al nanoparticles (Al*@SiO₂) with core-shell structures were made and used as fillers. Polyimide (PI), a well-known material with low permittivity and dielectric loss, high strength, and thermal stability, is widely used as a packaging material. In this article, Al*@SiO₂/PI composites with 0.1–0.4 vf were prepared and the thermal and dielectric properties of the resulting composites were studied.

2. Experimental

The metal Al particles with an average diameter of 800 nm were supplied by Beijing DK Nano technology Co. Ltd. Polyvinylpyrrolidone (PVP; Aladdin Company) was chosen as a coupling agent.

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Tetraethoxysilane (TEOS) and PI powder were obtained from 3F Co. The three-step synthesis procedure for creating the modifier fillers is illustrated in Fig. 1 (top left inset). First, uniform metal Al particles were coated with alumina through the classical metal passivation method of air oxidation (obtaining Al* or Al@Al₂O₃). Second, the resulting Al@Al₂O₃ particles were further treated with a PVP surface active agent. Finally, the obtained Al@Al₂O₃@PVP particles were further coated with silica through a facile classical sol–gel approach using TEOS as a precursor, then centrifuged and dried, leading to the formation of the multilayer Al@Al₂O₃@SiO₂ coating structure consisting of Al cores surrounded by an alumina shell and a silica shell.

The Al*@SiO₂ particles were separately dispersed into the PI matrix with filler (0.1–0.4 vf) to form Al*@SiO₂/PI composites. The four different Al*@SiO₂/PI composites were treated by drying in an

oven at 300 °C for 25 min and then molded by hot pressing at about 300 °C under uniaxial pressure of 50 MPa for 25 min. The final samples were made as disks that were 12 mm in diameter and 1 mm thick for dielectric measurement and φ is equal to 20 × 6 mm² for thermal conductivity. Al/PI composites made of pure Al particles (0.1 vf) were prepared for comparison.

Thermal conductivity (λ) was measured using a thermal constant analyzer (TPS-2500s; Hot Disk, Sweden). The dielectric properties were measured using an impedance analyzer (4980A; Agilent, USA) with a 16034B dielectric test fixture in the frequency range of 1 kHz–1 MHz. Field emission scanning electron microscopy (JEOL 7401F, Japan) and transmission electron microscopy (JEM-1230; JEOL, Japan) were used to observe the nanoparticle dispersion and particle sizes within the composite samples.

3. Results and discussion

The thermal conductivity and density (bottom right inset) of the composites with the volume fraction of Al*@SiO₂ measured at room temperature are shown in Fig. 1. The thermal conductivity increased obviously with the volume fraction of Al*@SiO₂, and the thermal conductivity of pure PI and Al was 0.23 and 27.4 W/mK, respectively. The maximum value of the thermal conductivity of the composites with the 0.4 vf Al*@SiO₂ filler was 3.1 W/mK. The density curve showed the same trend. The thermal conductivity can be described by the following equation:

$$\lambda = \alpha \rho C_p \quad (1)$$

where λ is the thermal conductivity of the material, α is the thermal diffusivity of the material, ρ is the density, and C_p is the specific heat capacity under constant pressure. For the same components in the composites, ρ and λ are related [18].

Fig. 2 shows scanning electron microscopy (Fig. 2a and b) and transmission electron microscopy (Fig. 2c and d) photographs of 800-nm uncoated Al metal particles and 0.4 vf Al*@SiO₂/PI composites, respectively. Fig. 2b shows that the Al*@SiO₂ particle fillers are highly dispersed throughout the PI polymer, while

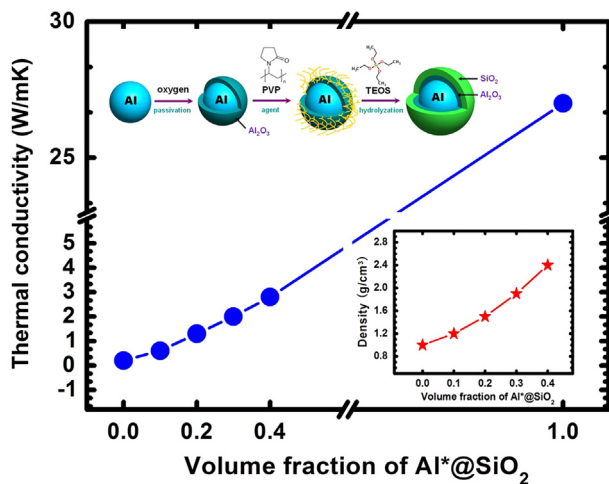


Fig. 1. Schematic illustration depicting the formation process of the Al*@SiO₂ double-shelled core-shell microspheres (top left inset), thermal conductivity and density (bottom right inset) of Al*@SiO₂/PI composites as a function of the volume fraction of Al*@SiO₂.

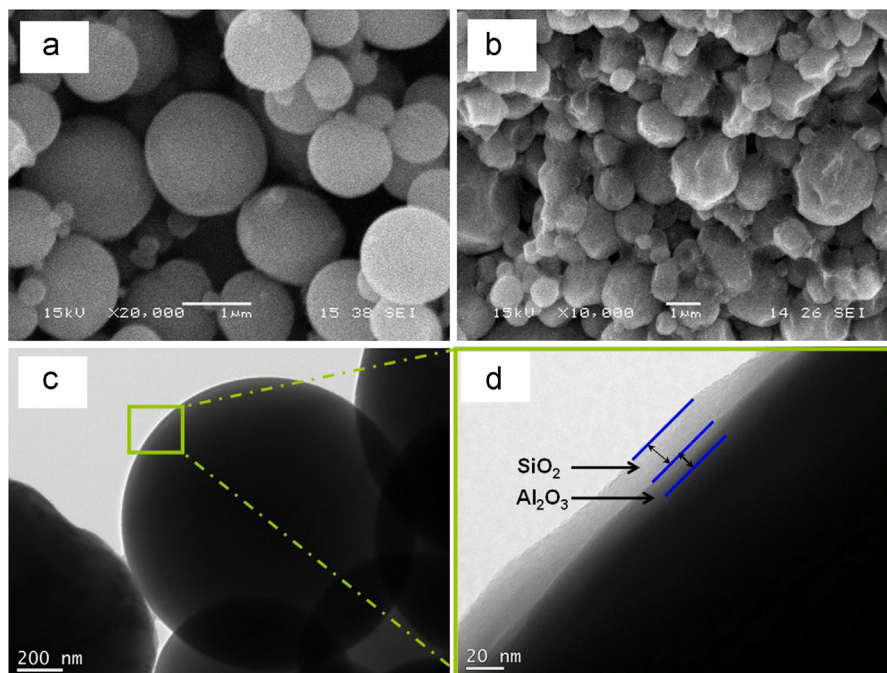


Fig. 2. SEM images of (a) average size 800 nm pure metal aluminum particles, (b) 0.4-vf Al*@SiO₂/PI composites and TEM images of (c) multilayer coating aluminum microsphere, and (d) partial enlarged drawing multilayer coating aluminum microsphere.

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