



Constructing continuous networks by branched alumina for enhanced thermal conductivity of polymer composites

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ABSTRACT

Efficient heat dissipation performance of thermal management materials has become one of the most critical challenges in the development of modern microelectronic devices. However, traditional polymer composites display limited enhancement of thermal conductivity even when highly loaded with thermally conductive fillers due to the lack of efficient heat conductive channels. In this study, branched alumina (b- Al_2O_3) is first used as the filler to improve thermal conductivity of phenolic resin (PR) and the preparation of the Al_2O_3 with branched structures is simple and high efficient. It is found that PR composites with b- Al_2O_3 present excellent thermal conductivity (up to $1.481 \text{ W m}^{-1} \text{ K}^{-1}$), which is equivalent to a dramatic enhancement of 7 times compared to neat matrix. The increased thermal conductivity should be attributed to that the branched structures of embedded b- Al_2O_3 particles tend to overlap each other and form continuous networks, which can act as efficient heat transfer pathways in PR matrix. Furthermore, PR composites with b- Al_2O_3 own improved thermal stability and decreased coefficient of thermal expansion (CTE) of $23 \times 10^{-6} \text{ K}^{-1}$ compared to neat PR ($71 \times 10^{-6} \text{ K}^{-1}$). Meanwhile, composites with decreased dielectric loss tangent are achieved because of the incorporation of b- Al_2O_3 , which is extraordinary and hopeful result for thermal management materials. This strategy provides an insight for the development of high-performance composites with potential to be used in electronic packages fields.

1. Introduction

During the past few decades, polymer is widely used as substrate to support and link electronic devices in the field of electronic packaging owing to its excellent mechanical properties, thermal stability, and chemical resistances [1,2]. With the continual miniaturization and increase of power density in the rapidly-developing electronic devices, a great deal of heat generated during operation are setting higher and stricter requirements for heat dissipation performance of advanced packaging materials [3–5]. However, polymer usually exhibits poor thermal conductivity between 0.1 and $0.3 \text{ W m}^{-1} \text{ K}^{-1}$, which restricts the fast heat dissipation for electronic products [5]. It has been reported that the thermal conductivity of composite materials can greatly increase by introducing inorganic fillers with high thermal conductivity, such as aluminum nitride [6–8], boron nitride [3,9–11], silicon carbide [12,13]. This versatile method synergistically combines the advantages of polymer matrix and inorganic fillers, thus the thermal as well as electrical properties of composites could be enhanced by proper

selection of the filler type, shape, and concentration [14].

Besides these as-known various fillers for polymer, Al_2O_3 has drawn great interest because of its variety of advantages: a relatively high thermal conductivity ($36 \text{ W m}^{-1} \text{ K}^{-1}$), low dielectric constant (9.8 at 1 MHz) and low dielectric loss (0.0002 at 1 kHz) [15]. More importantly, Al_2O_3 displays high electrical resistivity ($> 10^{14} \Omega \text{ cm}$), which was used to decorate graphene for thermally conductive and electrically insulating epoxy composites [16]. In addition to its low cost of the resource, features with natural abundance and high-temperature stability make Al_2O_3 to be used in a vast range of applications [17,18]. Researchers had studied effect of Al_2O_3 filler on the thermal conductivity of composite materials. Lee and co-workers introduced Al_2O_3 into epoxy matrix and reported the thermal conductivity of $0.6 \text{ W m}^{-1} \text{ K}^{-1}$ with 60 wt% filler content [19]. Yao et al. prepared polymer composite with 70 wt% Al_2O_3 filler and showed a thermal conductivity of close to $0.8 \text{ W m}^{-1} \text{ K}^{-1}$ [18]. However, compared with the intrinsic thermal conductivity of Al_2O_3 ($36 \text{ W m}^{-1} \text{ K}^{-1}$), the improvement of polymer composite is still limited even with a high

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loading. This is because the filler particles are isolated in the matrix [20], which is difficult to form effective thermal transfer pathways. These pathways are desired for increasing thermal conductivity as phonon heat conduction can occur with much less scatterings among fillers when compared to polymer matrix [21]. Zhang et al. added Al_2O_3 fibers into high density polyethylene to further enhance the thermal conductivity of polymer composite, nevertheless, the achieved value was only $0.40 \text{ W m}^{-1} \text{ K}^{-1}$ at a loading of 50 wt% [22]. The unsatisfactory reinforcement is affected by the orientations of fibers in matrix, as disordered randomly dispersed fibers lead to a relatively greater number of gaps and a small contact area between fillers, thus a higher contact thermal resistance is generated [23,24]. However, achieving an orderly arrangement of fibers is a more thorny and uncontrollable problem in preparation process. Latest studies demonstrated that constructing three dimensional continuous networks using filler was an efficient way to improve the thermal conductivity of composite, for which can serve as the shortcut for phonon transfer [25–28]. Graphene and boron nitride platelet had got the most extensive researches to achieve continuous networks in polymer matrix and thermal conductivity was largely enhanced [26,29,30]. However, the cost of both fillers is high and the preparation of the composites is quiet complexity. To the best of our knowledge, constructing continuous networks by low-cost Al_2O_3 has not been reported as the available conventional granular filler is difficult to form interconnected phase in matrix for heat transfer.

In this work, branched Al_2O_3 (b- Al_2O_3) is first used to reinforce the phenolic resin (PR) matrix and the branched structure is achieved via an easy and feasible method. The continuous networks are formed via overlaps of b- Al_2O_3 particles in matrix and be utilized as the secondary phase to construct efficient thermal transfer channels. At a loading of 75 wt% (48.5 vol%), the b- Al_2O_3 /PR composite possesses a dramatic improved thermal conductivity ($1.481 \text{ W m}^{-1} \text{ K}^{-1}$), corresponding to an enhancement of 7 times compared to that of neat PR. Additionally, b- Al_2O_3 /PR composites present improved thermal stability and greatly reduced coefficient of thermal expansion (CTE) of $23 \times 10^{-6} \text{ K}^{-1}$ compared to neat PR material. It is also worth mentioning that composites display lower dielectric loss tangent with the addition of b- Al_2O_3 , which is rare and required for composite materials used in electronic devices. These results offer new insights into the design and preparation of high-performance composites that are promising for applications in electronic packaging fields, particularly thermal interface materials.

2. Experimental

2.1. Materials

The raw Al_2O_3 powders were prepared by radio frequency induction thermal plasma in our research team. Phenolic resin (PR) was commercially purchased. Hexamethylenetetramine (HMTA) produced by Tianjin Kemiou Chemical Reagent Co. Ltd was used as the curing agent.

2.2. Fabrications of b- Al_2O_3 and Al_2O_3 /PR composites

The b- Al_2O_3 particles were obtained by treating the above raw Al_2O_3 directly at 1250°C in a pressure-less air furnace and the evolution process of branched structure is presented in Fig. 1. After treatment at 1250°C for 2.5 h, comparatively smaller raw particles were sintered

together while larger particles still remained spherical because of their low sintering activity. When the hold time was prolonged to 4.5 h, even larger spheres were connected to adjacent particles through sintering necks. Therefore, it was thanks to the sintering behavior of particles in random directions that Al_2O_3 with branched structures were achieved effectively.

Composites with different Al_2O_3 loading were prepared by the following procedures. Firstly, Al_2O_3 particles were added into liquid PR directly and accompanied by continuous mechanical stirring at room temperature for 30 min. For the composites with high loading of 70 wt% and 75 wt%, grinding the mixtures for 10 min after stirring to achieve a better dispersion. Then, HMTA with a weight ratio of 10: 1 (PR: HMTA) was added into the collosol, and obtained mixtures were further degassed for 20 min under vacuum atmosphere. Subsequently, the degassed mixtures were poured into stainless steel mold for cure reactions (130°C for 2 h). Finally, Al_2O_3 /PR composites could be obtained after demolding process.

2.3. Characterization

The morphologies of Al_2O_3 particles and fracture surfaces of Al_2O_3 /PR composites were investigated using scanning electron microscope (SEM, JSM-7001 F, JEOL, Tokyo, Japan) equipped with an energy dispersive X-ray spectrometer (EDX) and transmission electron microscope (TEM, JEOL JEM-2100). The structure of Al_2O_3 was analyzed by X-ray diffractometer (XRD, Philips X' Pert PRO MPD) operated at 40 kV and 40 mA with Cu K α radiation. Thermal conductivities of composites were measured using the light flash apparatus LFA-427 NanoFlash (NETZSCH, Germany) and the tests were conducted on disk samples with a diameter of 12.7 mm and thickness of 2–3 mm at 25°C . Thermal gravimetric analyses (TGA) of samples were performed with a TG-DTA 6300 instrument (SII, Japan) at a heating rate of $5^\circ\text{C}/\text{min}$ under nitrogen atmosphere, over a temperature range of 25 – 1000°C . The values of CTE of specimens were evaluated using a thermal mechanical analyzer (TMA, Setsys Evo, France) from 25 to 95°C at a heating rate of $5^\circ\text{C}/\text{min}$. Fourier transformed infrared (FTIR) spectrum were recorded between 4000 and 400 cm^{-1} with a spectrometer (T27-Hyperion-Vector22, Bruker, Germany) by casting a thin film on a KBr plate. Dielectric properties were tested using an Agilent 4294A impedance analyzer in the frequency range from 1 kHz to 10 MHz at the temperature of 25°C . Prior to measurement, both sides of the disks were plated with silver to improve electrical contact.

3. Results and discussion

3.1. Al_2O_3 characterization

The morphological and structural information of raw Al_2O_3 and b- Al_2O_3 are shown in Fig. 2. The raw Al_2O_3 particles exhibit uniform spherical shape, smooth surface and without aggregation, as shown in Fig. 2a. After heat treatment at 1250°C for 4.5 h, branched Al_2O_3 particles are obtained, as exhibited in Fig. 2b, which are consistent with the illustration of Fig. 1. Moreover, most particles own three or more branched structures and maintain smooth surface, as the red lines marked in Fig. 2b. The composition of prepared b- Al_2O_3 (sample in Fig. 2b) could be evidenced by EDS spectroscopy (Fig. 2c). It indicates the b- Al_2O_3 mainly consists of Al and O with a ratio of 1.58, close to the theoretic stoichiometric ratio in Al_2O_3 crystal. Fig. 2d shows the XRD

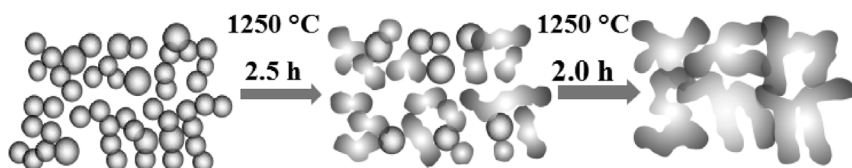


Fig. 1. Schematic of evolution process of branched structures during heat treatment.

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