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# A novel approach for $Al_2O_3$/epoxy composites with high strength and thermal conductivity

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ABSTRACT

Insulating materials for electronic packaging should ideally have high strength and thermal conductivity. Ceramic/polymer composite is one of the candidates for such materials. In this work, $Al_2O_3$/epoxy composites with very high flexural strength and thermal conductivity were prepared using a new processing technique consisting of the gelcasting, sintering and vacuum infiltration methods. $Al_2O_3$ green bodies were first formed by gelcasting. After being degreased at 600 °C, the green bodies were sintered at different temperatures from 1200 °C to 1500 °C, and porous $Al_2O_3$ ceramic skeletons with different porosity were resulted. The porosity was readily controlled by the sintering temperature. Higher sintering temperature resulted in lower porosity and larger average grain size in the $Al_2O_3$ ceramic skeletons. Epoxy was vacuum infiltrated into the sintered porous $Al_2O_3$ ceramic skeletons to form $Al_2O_3$/epoxy composites by curing. High $Al_2O_3$ loadings up to 70 vol.% in the composites were achieved. The flexural strength and thermal conductivity of the $Al_2O_3$/epoxy composites were found to reach 305 MPa and 13.46 W $m^{-1}$ $K^{-1}$, respectively. These results are remarkably higher than those previously reported in the literature. This work provides an effective approach for fabricating high performance ceramic/polymer composites for electronic packaging.



## 1. Introduction

Insulating materials for electronic packaging with outstanding properties, such as high thermal conductivity, high strength, good reliability and more importantly, low cost, are gaining intensive attention in the last decade as high-performance electronic devices are being developed and commercialized at an accelerating pace [1]. In recent years, ceramic/polymer composite materials have been extensively studied because they can be fabricated at relatively low cost and their thermal and mechanical properties can be readily optimized at the design stage [2,3]. In general, as shown in Fig. 1a, the ceramic/polymer composites comprise ceramic particles as fillers which reinforce the polymer matrix [4]. It is well-known that polymers usually have quite poor thermal conduction [5]. Hence, the other major task of the reinforcing ceramic particles in the ceramic/polymer composites is to enhance the thermal conduction of the composites.

Many kinds of ceramic particles such as silica [6,7], alumina [8,9], aluminum nitride [10,11], boron nitride [12–14] have been applied to polymer matrices to form ceramic/polymer composites, and their thermal conductivity was found to be higher than the polymer matrices. Lee et al. [7] employed silica, alumina, boron nitride, and diamond particles as the inorganic fillers and epoxy as the matrices to prepare polymer-based composites with 60 vol.% filler content, and reported that the highest thermal conductivity was 0.994 W $m^{-1}K^{-1}$ at 25 °C. Although the thermal conductivity is low, it is much higher than the thermal conductivity of epoxy (0.23 W $m^{-1}$ $K^{-1}$). Lee et al. [15] prepared AlN/epoxy composite with 57 vol.% filler content and reported a thermal conductivity of 3.39 W $m^{-1}$ $K^{-1}$. Compared with the thermal conductivity of the bulk ceramics such as $Al_2O_3$ (30 W $m^{-1}$ $K^{-1}$) and AlN (320 W $m^{-1}$ $K^{-1}$) [16], the improvement in the thermal conductivity of these above ceramic/polymer composites is still rather small. This is because the ceramic particles are isolated in the composite as shown in Fig. 1a. In order to further enhance their thermal conductivity, researchers have tried to use ceramic whiskers as filler for ceramic/polymer composites. Xu et al. [11] used AlN whiskers as fillers (60 vol.%) and polyvinylidene fluoride (PVDF) or epoxy as matrix to prepare ceramic/polymer composites, and reported a thermal conductivity of 11.5 W $m^{-1}$ $K^{-1}$. Although the

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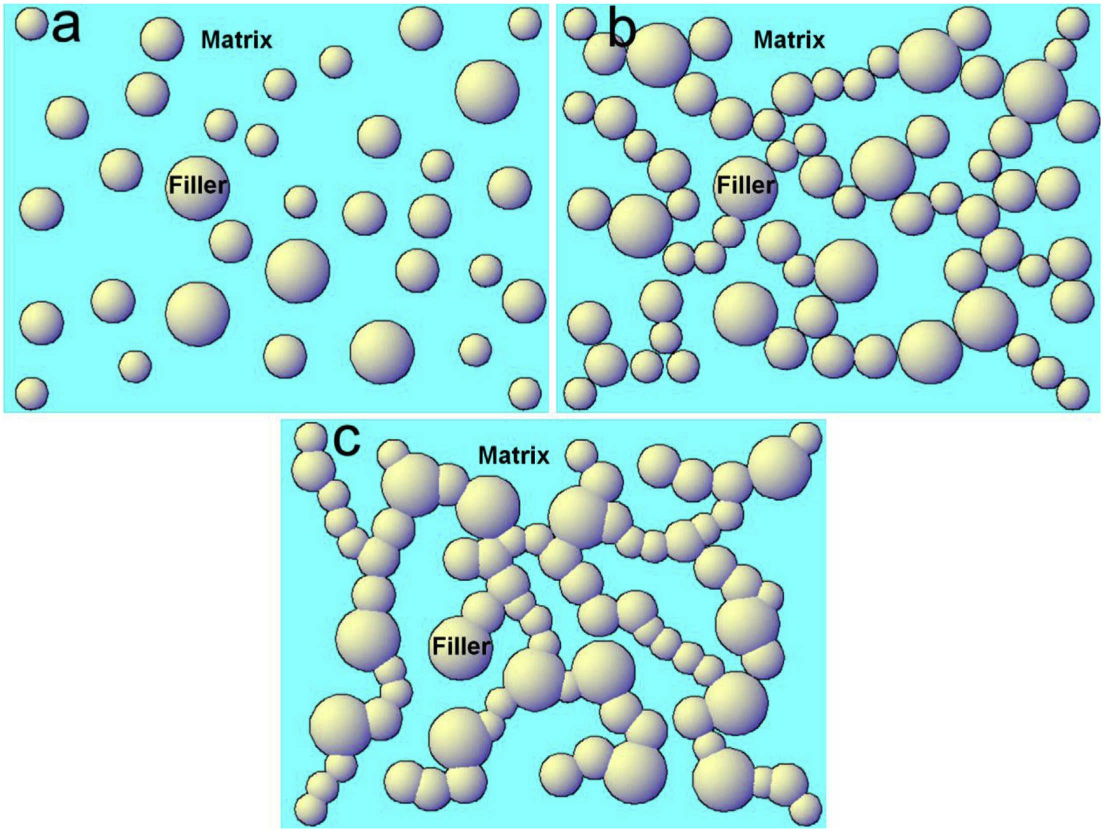


**Fig. 1.** Three filling configurations of ceramics particles in ceramic/polymer composites: dispersed (a), mechanically contacted (b), and chemically bonded (c).

thermal conductivity was largely enhanced, the cost of ceramic whiskers, however, is very high.

As shown in Fig. 1b, the thermal conductivity of ceramic/polymer composites could also be enhanced to a certain extent by dispersing high loadings (up to 70 vol.%) [15,17,18] of ceramic particulate fillers using the polymer molding technique [19,20]. In Fig. 1b, the ceramic particles mechanically contact one another. However, the interface between neighboring filler particles (Fig. 1b) has much lower thermal conduction than the bulk particles, although the filler content is over the percolation threshold. This is a result of the phonon scattering at the filler–filler contacts within the percolated network. Thus, the enhancement in the thermal conductivity of such high loading ceramic/polymer composites is expected to be rather modest as reported by Zhang et al. [17] and Bae et al. [18]. With regards to their mechanical properties, such high loadings, however, will result in inferior mechanical properties (e.g., susceptible to thermal cracking and challenging to process) to the composites [21].

In this work, we intend to prepare ceramic/polymer composites by building both the ceramic skeletons and polymer networks which are interpenetrating as shown in Fig. 1c. The porous ceramic is sintered so that the neighboring ceramic particles form chemically bonded boundaries (Fig. 1c), which are expected to have higher thermal conductivity than the mechanically dispersed particles (Fig. 1a and b). In addition, such ceramic skeletons also possess good mechanical properties because of their strongly bonded grain boundaries. Considering $Al_2O_3$ has relatively good thermal conductivity and is of much lower cost than AlN and BN, we selected $Al_2O_3$ to prepare $Al_2O_3$/epoxy composites in this work. High loadings of $Al_2O_3$ in the $Al_2O_3$/epoxy composites were achieved using the new process consisting of the gelcasting, sintering and vacuum infiltration techniques. The advantage of the gelcasting technique is that ceramics with complex shapes can be conveniently fabricated, such as ceramic gas turbines and rotors [22]. A significant enhancement in the thermal conductivity of the $Al_2O_3$/epoxy composites was resulted, and in the meantime high mechanical properties were achieved. This work demonstrates an effective processing method for fabricating ceramic/polymer composites with very high thermal conductivity, mechanical properties and good reliability.

## 2. Experimental procedure

### 2.1. Material preparation

Commercial $\alpha$-$Al_2O_3$ powder (purity 99.7%, average particle size D50 = 1.36 μm) was used in this study. Commercial nanometer-sized $SiO_2$ powder was used as sinter aid. Ammonium polyacrylate ($NH_4$-PAA), acrylamide (AM, Sinopharm Chemical Reagent Co., Ltd., Shanghai, China), N,N'-methylenebisacrylamide (MBAM, Aladdin Chemistry Co., Ltd., Shanghai, China), N,N, N′,N′-tetramethylethylenediamine (TEMED, Sinopharm Chemical Reagent Co., Ltd., Shanghai, China), and ammonium persulfate (APS, Xilong Chemical Co., Ltd., China) were used in the gelcasting process as dispersant, monomer, cross-linker catalyst and initiator, respectively. After $NH_4$- PAA was added (0.8%, by weigh of $Al_2O_3$ powder), 50 vol.% slurries were prepared by ball milling $Al_2O_3$ and $SiO_2$ powders (99 wt% for $Al_2O_3$, 1 wt% for $SiO_2$) in an aqueous premix solution to which AM (10 wt%) and MBAM (0.5 wt%) were added. Then the catalyst and initiator were added into the suspensions followed by degassing for 10 min in vacuum. The $Al_2O_3$ suspensions were casted into a metal mold and then solidified by heating at