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Enhanced thermal conductivity for Ag-deposited alumina sphere/epoxy resin composites through manipulating interfacial thermal resistance

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ABSTRACT

Polymer composites with high thermal conductivity have a great potential application in modern electronics, due to their light-weight, easy process, low cost and stable physical and chemical properties. Nevertheless, most polymer composites commonly possess unsatisfactory thermal conductivity, primarily because of the high interfacial thermal resistance between inorganic fillers. Herein, we report a novel method through silver-deposition on the surface of the fillers to create a silver nanoparticle "bridge", to decrease the interfacial thermal resistance between fillers. The results demonstrate that the out-of-plane thermal conductivity of the epoxy resin/sphere alumina composites is increased to 1.304 $W\,m^{-1}\,K^{-1}$, representing an improvement of 624% compared with pure epoxy resin. This strategy provides an insight for the design of thermally conductive polymer composites with potential to be used in next-generation electronic packaging.

1. Introduction

With the fast development of the electronics towards highly integrated, ultra-light, miniaturization and multi-functionallization, packaging density of the electronic components or the unit cells has dramatically increased. Heat accumulation with running of the electronic products incurs the overheat and malfunction to the electronics, and thus has already become a main drawback to hinder the development of the next generation of electronics. Therefore, heat management has become one of the major fields in the electronic packaging. Polymer-based composites, have been used widely as packaging materials, thanks to their light-weight, easy process, low cost and stable physical and chemical properties. Because pure polymers have low thermal conductivity, different kind of inorganic fillers are introduced into the polymer to enhance the thermal conductivity of polymer-based composite. Carbon-based materials, including graphene [1–6] and carbon nanotubes (CNTs) [7–10] *etc.*, can largely increase the thermal conductivity of the polymer-based composites, due to their intrinsic extremely high thermal conductivity [11]. However, the high electrical conductivity of these carbon-based materials limits their applications in the electronics packaging field, in which electrical insulation is an essential prerequisite. Hexagonal boron nitride, as an analogue to graphite, being a good electrical insulator with the band gap around 5.5 eV, has attracted pretty much interests due to its unique advantage [12–18]. Although this kind of two-dimensional nanomaterials are easily oriented along the plane in the polymer matrices, and significantly enhance the in-plane thermal conductivity of the polymer-based composite, which facilitate the fast heat spreading along the plane of the electronic devices. However, in most cases, increasing out-of-plane thermal conductivity outweighs in-plane thermal conductivity for the thermal management [19]. Therefore, increasing out-of-plane thermal conductivities of polymer-based composites is strongly required for these applications. Ceramic fillers, including alumina (Al_2O_3) [20–23], silicon nitride [24], aluminium nitride [25] and silicon carbide [26] *etc.*, have widely been used as thermally conductive fillers, in the light of their high thermal conductivity, electrical insulation and low cost. However, the obtained polymer composites still have a low thermal conductivity below 2.0 $W\,m^{-1}\,K^{-1}$. High interfacial thermal resistance is believed to be an inevitable barrier to increase the out-of-plane thermal conductivity. In order to minimize the interfacial thermal resistance

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Table 1
The typical composition for Al_2O_3/Epoxy and Al_2O_3-AgNP/Epoxy composites with 70 wt% Al_2O_3 content and 1.96 wt% Ag deposition.

Sample	Component				
	Filler		Matrix		Total
Al_2O_3/Epoxy	Al_2O_3 70	EP828 16.13	MHHPA 13.71	Catalyst 0.16	100
Al_2O_3-AgNP/Epoxy	AgNP + Al_2O_3 1.37 + 68.63	EP828 16.13	MHHPA 13.71	Catalyst 0.16	100

between fillers and polymer matrices, lots of effort has been contributed to the functionalization of the fillers [27–29]. Still, the increase in thermal conductivity is insufficient, because the functionalization of the fillers usually compromises their intrinsic thermal conductivity. Besides, the interfacial thermal resistance between filler and filler turns out to be the main hindrance limiting the increase of the thermal conductivity of the polymer-based composite when filler content reaches high enough to contact each other. From this point of view, it is imperative to come up with an effective and universal method to decrease the interfacial thermal resistance between filler/filler other than surface functionalization.

In our previous group work [30–33], we proposed a novel method through silver-deposition on the surface of the fillers to create a silver "bridge" among the fillers, exploiting electron-phonon coupling mechanism to decrease the interfacial thermal resistance between fillers. It has been proved to be an effective way to highly improve the in-plane thermal conductivity of the composite in terms of the used two-dimensional structure fillers. In this work, commercial Al_2O_3 sphere was chosen as the thermally conductive filler to introduce into the epoxy resin. Through silver deposition after surface modification, we fabricated Ag-deposited Al_2O_3 sphere, and also investigated the influence of the silver deposition on the out-of-plane thermal conductivity of the composite. The result showed that the out-of-plane thermal conductivity of the composite has increased by 35% with silver-deposited Al_2O_3 as the fillers. This increase can be attributed to the silver "bridge" between the fillers facilitating the heat flow across the interfacial boundary, which in turn reduce the interfacial thermal resistance and improve the out-of-plane thermal conductivity of the composite.

2. Experimental

2.1. Materials

Al_2O_3 spheres with an average diameter of 17.4 µm were purchased from Shanghai Baitu Company, China. The coupling agent Glycidoxypropyltrimethoxysilane (GPTS) was purchased from J&K Scientific Co., Ltd (Beijing, China). Liquid epoxy resin, EP-828, was supplied by The Hexion company, USA. 4-methylhexahydrophtahlic anhydride (MHHPA; Sigma-Aldrich Co., Ltd.) and 2-Ethyl-4-methylimidazole (96%, Aladdin Chemistry Co., Ltd) were used as curing agent and catalyst, respectively. Silver nitrate ($AgNO_3$, 99.8%) and absolute ethanol (99.7%) were supplied by Shanghai Ling Feng Chemical Reagent Co., Ltd. Both sodium borohydride ($NaBH_4$, 98%) and sodium citrate dehydrate were supplied by Aladdin Chemistry Co., Ltd. All of the reagents were of analytical grade and used as received.

2.2. Surface modification of Al_2O_3 sphere

Surface modification of Al_2O_3 spheres with GPTS was performed using hydrolysis-condensation method. In detail, 10 g of Al_2O_3 spheres, 40 mL of absolute ethanol, and 5 mL of deionized water were mixed together in a 100 mL breaker, after the mixture was uniformly dispersed under magnetic stirring for 1 h, 0.2 g of GPTS was then added into the mixture. The suspension was stirred for 2 h at room temperature, and was then dried at 110 °C for 2 h in the oven until the solvent evaporated completely. The dried powders were grinded in the mortar with the pestle, and then collected for use.

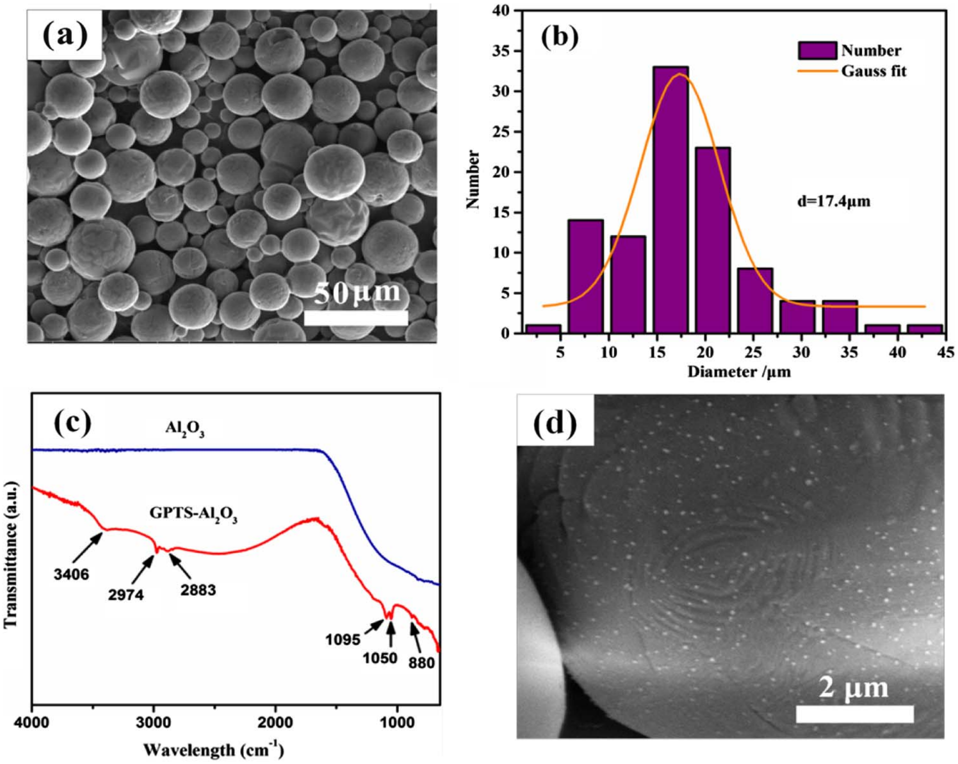

Fig. 1. (a) SEM micrograph of the raw Al_2O_3 Sphere. (b) Size histograms of the Al_2O_3 spheres determined from the SEM image. (c) FTIR spectra for the pristine Al_2O_3 and GPTS-Al_2O_3 spheres. (d) SEM micrograph of the Ag-deposited Al_2O_3 sphere. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)