Research Article

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Thermal conductivity of silicone elastomer with a porous alumina continuum

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Abstract: In this paper, porous alumina continuum (PAC) was prepared with alumina powders (APs) by the gel-casting method and was applied to obtain silicone elastomer (SR) composites (PAC/SR) by the impregnating process. The structure was investigated by X-ray diffraction (XRD) and scanning electron microscopy (SEM). The influences of PAC on thermal conductivity and dielectric permittivity of PAC/SR composites were studied in comparison with AP/SR composites. When the alumina content was 14 vol%, the thermal conductivity of the PAC/SR composites reached $0.84 \text{ W} \cdot (\text{m} \cdot \text{K})^{-1}$, which was 3.1 times higher than that of the AP/SR composites. The thermal conductivity of PAC/SR and AP/SR was simulated by theoretical models, and the interfacial thermal resistance was calculated by effective medium theory, which indicated the advantages of PAC in enhancing the thermal conductivity of SR-based composites and the reduced interfacial thermal resistance between PAC and SR.

Keywords: porous alumina continuum, thermal conductivity, three-dimensional network, silicone elastomer, thermal interface materials

1 Introduction

The increasing demand for thermally conductive materials has inspired to meet the rapidly developing requirements of thermal management in electronic devices with integrated, miniaturized, and functionalized characteristics (1). Due to high heat accumulation inside the devices, heat dissipation becomes crucial to maintain the stability and reliability of the devices (2). Thermal interface materials (TIMs) are thus applied to improve the thermal conductivity between electronic components and heat sinks. As a kind of typical TIM, polymer-based composites have been widely investigated in facing the challenges of the poor thermal conductivity of polymers and their potential applications as substitutes of metals in some industrial scenarios of lightweight, low cost, high specific modulus, and easy to manufacture (3). Fillers with high thermal conductivity, such as carbon fillers (4), ceramic (5), and metal particles (6), are usually introduced to obtain thermally conductive polymer-based composites. Much effort has been devoted to build an efficient pathway for heat dissipation in thermally conductive composites by closest packing of fillers (7), hybrid fillers (8), orientations of fillers (9), and thermally conductive framework (10).

Recently, the construction of a thermally conductive network in polymers has attracted much attention. The addition of hybrid fillers into composites was investigated as an efficient way to form a thermally conductive network and achieve a synergistic effect on the enhancement of thermal conductivity of polymer-based composites (11). A three-dimensional network was constructed with modified multiwall carbon nanotubes and graphene in the PVDF matrix and the composites presented a thermal conductivity of $1.46 \text{ W} \cdot (\text{m} \cdot \text{K})^{-1}$ at 10 wt% filler loading (12). 3D phonon skeleton was built with stacked boron nitride (BN) platelets and reduced graphene oxide (rGO) by the combination of ice-templated and infiltrating methods, which resulted in a through-plane thermal conductivity of $5.05 \text{ W} \cdot (\text{m} \cdot \text{K})^{-1}$ for epoxy-based composites at a filler loading of 13.16 vol% (13). The oriented carbon fiber/alumina/silicon rubber composites were fabricated

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by 3D printing, which showed a thermal conductivity of $7.36 \text{ W} \cdot (\text{m} \cdot \text{K})^{-1}$ at 12 vol% carbon fiber and 30 vol% alumina loadings (14). 3D printing was also applied to construct the filler networks with oriented BN platelets and spherical alumina and resulted in $3.64 \text{ W} \cdot (\text{m} \cdot \text{K})^{-1}$ in-plane thermal conductivity of the polydimethylsiloxane (PDMS) composites with hybrid fillers of 35 wt% BN and 30 wt% alumina (15).

Alumina (Al_2O_3) is one of the most popular thermally conductive fillers owing to its good thermal conductivity, electric insulation, and low cost (16). The morphology, crystalline structure, and loading fraction of alumina exert strong influences on the thermal conductivity of polymer-based composites (17–19). High thermal conductivity is usually attained by increasing the loading fraction of alumina fillers in the composites. A thermal conductivity of $6.7 \text{ W} \cdot (\text{m} \cdot \text{K})^{-1}$ was achieved at a high alumina fraction of up to 79 vol% in epoxy-based composites by using spherical particles with different size scales (20). A thermal conductivity of $13.46 \text{ W} \cdot (\text{m} \cdot \text{K})^{-1}$ was reported at a high alumina fraction of up to 70 vol% in epoxy-based composites by using sintered porous alumina ceramic skeletons (21). Thermal conductivity of $2.92 \text{ W} \cdot (\text{m} \cdot \text{K})^{-1}$ of silicone-based composites with 80 vol% hybrid alumina powder was obtained through *in situ* polymerization (22). In addition, attempts were also made to fabricate a 3D alumina scaffold in order to decrease the loading fraction of alumina while reaching a high thermal conductivity. A continuous alumina framework in epoxy was realized through a protein foaming technique followed with infiltration, and a thermal conductivity of $2.58 \text{ W} \cdot (\text{m} \cdot \text{K})^{-1}$ was obtained at a filler fraction of 23.32 vol% (23). A thermally conductive 3D framework in silicone rubber (SR) was constructed by foaming of alumina/SR composites with NH_4HCO_3 and a thermal conductivity of the composites with 32.6 wt% alumina reached $0.747 \text{ W} \cdot (\text{m} \cdot \text{K})^{-1}$ (24).

Silicone resin, owing to its low toxicity, excellent electrical insulation, good mechanical properties, high chemical stability, and thermal stability, has been widely applied as one of the typical polymeric materials to develop TIM (25,26). In view of the poor thermal conductivity of silicone resin and the insulation requirements of TIM applications, alumina is considered to be an ideal candidate filler to prepare silicone-based composites with good thermally conductive and electrically insulating properties (27). To obtain thermally conductive silicone-based composites with relatively low dielectric permittivity at a low fraction of alumina, this paper aims at constructing a porous alumina continuum (PAC) with alumina powders (AP) by the gel-casting method and forming a three-dimensional thermally conductive network in silicone

elastomer by the impregnating process. Using these methods, a new solution would be proposed for the development of silicone-based TIM.

2 Materials and methods

2.1 Materials

Alumina powder (AP) with a purity of 99.9% and an average size of $3 \mu\text{m}$ was purchased from Dongguan Dongchao Material Technology Co., Ltd. Magnesium oxide (MgO) with a purity of 99.9% was purchased from Shanghai Macklin Biochemical Technology Co., Ltd. Agarose was obtained from Shanghai Lanji Technology Development Co., Ltd. Sodium dodecyl sulfate (SDS) was purchased from Tianjin Guangfu Fine Chemical Research Institute. Silicone elastomer (Sylgard 184) was purchased from Dow Corning Corp.

2.2 Preparation methods

Alumina powder (AP) was used as a raw material to prepare the porous alumina continuum (PAC) by the gel-casting method. The ceramic slurry with an alumina content of 40 vol% was prepared by ball milling for 4 h with the addition of 1.0 wt% agarose as a monomer, 0.6 wt% SDS as a dispersant, 1.0 wt% MgO as a sintering aid, and deionized water as a solvent. The well-dispersed slurry was foamed with 3.0 wt% SDS under stirring with a blender for 30 min. The foamed slurry was then cast into a rubber mold and matured at room temperature for 48 h to form a green body. The green body was transferred into the tube furnace and sintered under an air atmosphere. The furnace was heated to $1,250^\circ\text{C}$ at a rate of $5^\circ\text{C} \cdot \text{min}^{-1}$ and maintained at $1,250^\circ\text{C}$ for 1 h. After the furnace was naturally cooled, PAC was obtained and sanded to the required size. The content of AP in the slurry was adjusted to obtain the PAC for the applications in thermal conductive composites.

The PAC was immersed in the silicone elastomer (SR) solution to prepare PAC/SR composites by the impregnating process. SR solution was obtained by blending two components together according to the ratio recommended by the supplier. The mixture of PAC and SR solution was treated under a vacuum environment for 1 h and then cured at 100°C for 1 h. The composites of PAC/SR were thus obtained. The processes of preparing PAC and

PAC/SR composites are schematically illustrated in Figure 1. The samples with PAC contents of 5.0, 7.5, 10, and 14 vol% were prepared and named PAC5.0/SR, PAC7.5/SR, PAC10/SR, and PAC14/SR, respectively.

The composites of AP and SR (AP/SR) were also prepared to make a comparison with PAC/SR composites. AP was mixed together with the components of SR and treated under a vacuum environment for 1 h and cured at 100°C for 1 h. The samples with AP contents of 5.0, 7.5, 10, 14, and 41 vol% were prepared and named as AP5.0/SR, AP7.5/SR, AP10/SR, AP14/SR, and AP41/SR, respectively.

2.3 Characterization

A field emission scanning electron microscope (SEM, Sirion200, FEI) was used to observe the morphological structure of PAC and the composites with the cryo-fractured section metalized with gold. X-ray diffractometer (XRD, X'Pert PRO, PANalytical) was used to characterize the crystalline structure of the AP and PAC with Cu K α radiation ($\lambda = 1.5406 \text{ \AA}$) at a voltage of 40 kV and a current of 40 mA with a step of 0.013° and a scanning speed of $8^\circ \cdot \text{min}^{-1}$ in the range of $5\text{--}80^\circ$. The thermal conductivity (λ) of the composites was tested by using a thermal conductivity meter (LFA447, Nanoflash, Netzsch) with a sample diameter of 12.7 mm and a thickness of 2.0 mm. A thermoviewer (E8, FLIR) was used to capture the infrared thermal images and record the thermographic observations.

The samples with a thickness of 2.0 mm were placed horizontally on the plate preheated to a constant temperature of 80°C. The images on the top surface of samples were recorded at 2, 10, 20, 40, and 60 s after the samples were exposed to the heated plate. A broadband dielectric spectrometer (Alpha-A, Novocontrol, Germany) was used to analyze the dielectric constant and loss factor at room temperature at a voltage of 3 V in the frequency range of $1\text{--}10^6 \text{ Hz}$ on the sample coated with circular aluminum electrodes with a diameter of 25 mm using a high-vacuum resistance evaporation coating machine (Model ZHD-400, Beijing Techno Technology Co., Ltd.).

3 Results and discussion

The SEM micrographs of the PAC are shown in Figure 2. From the full view of the PAC, as shown in Figure 2a, the continuous network was observed with a uniform pore structure. The cells in Figure 2b were connected by the thin alumina wall and the windows were observed between two neighboring cells. The joint of the cell walls is clearly noticed in Figure 2c, which was used to build the three-dimensional structure of PAC. The alumina particles in the cross-section of the joint were found to be well connected in Figure 2d and present in good continuity. The closely bonded particles of alumina were attributed to the high-temperature sintering with the aid of MgO, which can be also used as a good thermally

Figure 1: Schematic process to prepare PAC and PAC/SR composites.

Figure 2: SEM micrographs of PAC at different magnifications: (a) a network structure of PAC at a magnification of 160 $\times$, (b) three-dimensional cell structure of PAC at a magnification of 500 $\times$, (c) a continuous wall structure of PAC at a magnification of 1,000 $\times$, and (d) the micro-morphological structure of alumina particles in the joint of PAC at a magnification of 10,000 $\times$.

conductive filler. Accordingly, PAC with a well-structured scaffold was obtained and its good thermal conductivity could be expected by decreasing the interfacial thermal resistance between fillers due to the bonded alumina particles (28).

The thermal conductivity of alumina shows a strong dependence on the crystalline structure (17). The XRD patterns of AP and PAC are shown in Figure 3. The same diffraction peaks were observed for AP and PAC, which were assigned to the reflections of α - Al_2O_3 according to the standard pattern of Corundum (ICDD PDF#83-2080). The results revealed the same crystalline structure of PAC as AP after the gel-casting process and indicated their same intrinsic thermal conductivities as thermally conductive fillers.

3.1 Morphology of the composites

Figure 4 illustrates the photographs and SEM micrographs of AP/SR and PAC/SR composites. Comparing the photographs of SR, PAC, and PAC14/SR in Figure 4a, the integrated structure of PAC was demonstrated in the composites of PAC14/SR, which showed the transparent parts of SR and the macropores of PAC. Meanwhile, the composites of AP14/SR presented the homogeneous appearance in white, thanks to the well-dispersed alumina

particles in SR as shown in Figure 4b. The SEM micrograph of cryo-fractured sections of AP14/SR in Figure 4c evidenced the continuous structure of PAC in the SR matrix. The joints and the cell walls of PAC are clearly observed in Figure 4d. Furthermore, there were no noticeable voids in the composites. The good compatibility between the cell wall of PAC and the matrix of SR is revealed in the inserted SEM micrograph in Figure 4d. Consequently, PAC can provide numerous interpenetrated channels for the access of SR, thanks to its ample open pores and high porosity,

Figure 3: XRD patterns of PAC and AP.

Figure 4: Morphological images of the composites: (a) photographs of SR, PAC, AP14/SR, and PAC14/SR, (b) SEM micrograph of the cryo-fractured section of AP14/SR, (c) SEM micrograph of the cryo-fractured section of the PAC14/SR composite at low magnification and (d) that at high magnification with inserted micrograph of the cell wall in the SR matrix.

which is quite a contrast to the randomly distributed alumina particles in AP/SR composites by adding thermal conductive fillers.

3.2 Thermal conductivity

Figure 5 presents the thermal conductivity of PAC/SR and AP/SR as a function of the filler content. A continuously increasing thermal conductivity of the composites was observed as the filler content increased. The PAC/SR composites exhibited significantly higher thermal conductivity than the AP/SR composites. The thermal conductivity of the PAC/SR composites with the alumina content of 14 vol% reached $0.84 \text{ W} \cdot (\text{m} \cdot \text{K})^{-1}$, which increased by 3.1 times in comparison to the AP/SR composites with the same alumina content. The remarkable advantage of PAC was evidenced in improving the thermal conductivity of the composites. PAC as an integrated filler prompted the formation of interconnected three-dimensional networks of alumina in PAC/SR composites and exerted the effective function on the thermal conductivity. In contrast, the randomly dispersed alumina particles in AP/SR composites arranged disorderly and failed to construct a good thermally conductive pathway.

The enhancement of thermal conductivity (η) was adopted to make a comparison between PAC/SR and

AP/SR according to Eq. 1 (15). As shown in Figure 6, the thermal conductivity enhancement of PAC/SR in contrast to AP/SR increased with the filler content. The enhancement increased from 30% to 236% as the filler content increased from 5 to 14 vol%. Thanks to the effective thermal conducting network of PAC, the high thermal conductivity of PAC/SR composites was realized at a much lower content of alumina. While considering the contribution per unit volume fraction of PAC and AP to

Figure 5: Thermal conductivity of AP/SR and PAC/SR at different filler contents.

Figure 6: The thermal conductivity enhancement of PAC/SR in contrast to AP/SR.

the thermal conductivity of the composites with an alumina content of 14 vol%, a value of $0.054 \text{ W} \cdot (\text{m} \cdot \text{K})^{-1}$ per unit volume was obtained for PAC in contrast to that of $0.013 \text{ W} \cdot (\text{m} \cdot \text{K})^{-1}$ per unit volume for AP. The results indicated the importance of the three-dimensional continuum of alumina on the enhancement of thermal conductivity of composites:

$$\eta = \frac{\lambda_{\text{PAC/SR}} - \lambda_{\text{AP/SR}}}{\lambda_{\text{AP/SR}}} \times 100 \quad (1)$$

where $\lambda_{\text{PAC/SR}}$ and $\lambda_{\text{AP/SR}}$ are the thermal conductivities of the PAC/SR and AP/SR composites, respectively.

The thermal conductivity of the composites was simulated according to four theoretical models, viz., the series model, parallel model, Maxwell model, and EMT model as expressed in Eqs. 2–5 (29,30). As can be seen in Figure 5, the values of thermal conductivities of PAC/SR and AP/SR were situated between the simulated lines of the series model and parallel model. The simulated line of the series model represented the lower bound of the composites while that of the parallel model gave the upper bound. Good fitting was found in simulating the thermal conductivity of AP/SR by the Maxwell model, which was proved to be useful for the homogeneous composites with randomly dispersed particles. Taking the thermal conductivity of PAC/SR into account, none of the models could fit the experimental results of PAC/SR. The three-dimensional continuum of alumina was challenged in the description of its size and shape during simulations. On the other hand, the experimental thermal conductivity of PAC/SR was much close to the simulated results by the parallel model. In view of the co-continuous structure of PAC

and SR in the composites, we used a combined model of the series and parallel models (S-P model) to simulate the thermal conductivity of PAC/SR according to Eq. 6 (31, 32). After simulations with the experimental results of PAC/SR, a good fitting curve was obtained when $n = 0.38$. As a result, a model was obtained, which was of good predictability in simulating the thermal conductivity of PAC/SR.

$$\frac{1}{\lambda_c} = \frac{V_f}{\lambda_f} + \frac{(1 - V_f)}{\lambda_p} \quad (2)$$

$$\lambda_c = V_f \lambda_f + (1 - V_f) \lambda_p \quad (3)$$

$$\lambda_c = \lambda_p \left[\frac{\lambda_f + 2\lambda_p + 2V_f(\lambda_f - \lambda_p)}{\lambda_f + 2\lambda_p - V_f(\lambda_f - \lambda_p)} \right] \quad (4)$$

$$V_f \frac{\lambda_f - \lambda_c}{\lambda_f + 2\lambda_c} + (1 - V_f) \frac{\lambda_p - \lambda_c}{\lambda_p + 2\lambda_c} = 0 \quad (5)$$

where V_f is the volume fraction of the filler; λ_p , λ_f , and λ_c are the thermal conductivities of the polymer matrix, the filler, and the composites, respectively. The thermal conductivities of $0.16 \text{ W} \cdot (\text{m} \cdot \text{K})^{-1}$ and $28 \text{ W} \cdot (\text{m} \cdot \text{K})^{-1}$ were adopted for SR and alumina during simulations, respectively.

$$\lambda_c^n = V_f \lambda_f^n + (1 - V_f) \lambda_p^n \quad (6)$$

where n is the coefficient in the range of -1 and 1 . When $n = -1$, the series model is obtained; when $n = 1$, the parallel model is obtained.

3.3 Interfacial thermal resistance

As well known, the parallel model overestimates the contribution of the fillers because it assumes the independent contributions from each phase to the total conductivity and the perfect contact between particles. Even though a good continuous structure was designed in PAC, the defect can hardly be avoided and weakened the thermal conductivity of PAC. Furthermore, the influences of interfacial thermal resistance (R_B) between the filler and the matrix should be taken into consideration while taking the advantage of the three-dimensional network of PAC and maximizing its contribution at a much lower content. Here, the Maxwell-Garnett effective medium theory (MG-EMT) was introduced to calculate the value of R_B according to Eq. 7 (33):

$$\lambda_c = \lambda_f \left[\frac{3\lambda_p + 2V_f(\lambda_f - \lambda_p)}{(3 - V_f)\lambda_f + V_f\lambda_p + \frac{R_B V_f \lambda_p \lambda_f}{H}} \right] \quad (7)$$

where R_B is the interfacial thermal resistance at the filler/matrix interface and H is the thickness of the filler.

Considering the particle size of alumina, $3\ \mu\text{m}$ was adopted as H during calculations.

Figure 7 presents interfacial thermal resistance of AP/SR and PAC/SR as a function of the filler content. The composites of PAC/SR showed much lower interfacial thermal resistance than that of AP/SR. The positive influence of PAC on the thermal conductivity could be attributed to the decreased interfacial thermal resistance. Meanwhile, the interfacial thermal resistance could also dramatically alter the thermal conductivity of the composites, which gave rise to the much lower thermal conductivity of the composites than that of alumina (34). The high interfacial thermal resistance was induced by the phonon scattering in the filler network and the interface between the fillers and polymer matrix. The dispersed alumina in AP/SR failed to form an effective path in the SR matrix and contributed little to heat conduction due to the high interfacial thermal resistance of the interface between the particles and the matrix (21). PAC, owing to its good integrity and interconnected structure, was used to build a three-dimensional network in the SR matrix and played a vital role in heat conduction.

3.4 Heat dissipation

In order to further illustrate the heat transfer performances of the composites, infrared thermal imaging was applied to characterize the temperature profiles of SR, AP14/SR, and PAC14/SR, as shown in Figure 8. The temperature distribution on the top surface of PAC14/SR was obviously different from that of SR and AP14/SR. At 10 s, PAC14/SR

Figure 7: Interfacial thermal resistance of AP/SR and PAC/SR by the MG-EMT model.

Figure 8: The temperature profiles vs time of SR, AP14/SR, and PAC14/SR; inset: photos of infrared thermal imaging.

showed higher temperatures than SR and AP14/SR. It took 40 s for PAC14/SR to reach 70°C , while it took 55 s for AP14/SR and 66 s for SR. According to the temperature profiles vs time, the temperature on the surface tended to level off after 60 s. At 90 s, the temperature of PAC14/SR settled at 79°C , while those of AP14/SR and SR settled at 75°C and 72°C , respectively. As can be seen, the temperature of PAC14/SR increased much faster in comparison with AP14/SR and SR and the temperature at 90 s was much closer to the plate temperature (80°C). The results revealed better heat transfer performances of PAC14/SR than AP14/SR and SR, which can be beneficial to accelerate heat dissipation.

3.5 Dielectric constant and loss factor

As TIM, dielectric properties are very important parameters to be considered. Thanks to the good electrically insulative characteristics of alumina, the composites usually present good insulation properties in terms of resistivity. Recently, attention has been paid to composites with certain dielectric permittivity and dielectric loss due to the rapid development in the fields of high frequency. Considering the opportunities in these fields, the dielectric permittivity and loss factor were investigated by a dielectric spectrometer on two samples with similar thermal conductivities, AP63/SR and PAC21/SR. Figure 9 shows their curves as a function of frequency. The permittivity of PAC21/SR was 4.30 at 100 Hz while that of AP63/SR was 6.06. A remarkable increase in permittivity was observed due to a higher content of alumina in AP63/SR. The loss factor curve of PAC21/SR showed a peak around 45 Hz, which gave rise to a higher loss factor than AP63/SR. As a result, the loss factor of PAC21/SR was 0.009 at 100 Hz while that of AP63/SR was 0.003. Taking the continuous

Figure 9: The dielectric permittivity (ϵ') and the loss factor ($\tan \delta$) of AP63/SR and PAC21/SR composites as a function of frequency.

network structure of PAC into account, the dielectric loss peak could be attributed to the interface polarization between alumina particles. The loss factor of AP63/SR and PAC21/SR tended to be similar after 10^4 Hz.

4 Conclusion

PAC with a structured scaffold was obtained by the gel-casting method and applied in SR to construct a thermally conductive network for heat dissipation. The good compatibility between PAC and SR was revealed, and no voids were found in the composites. The integrated structure of PAC in the SR matrix was demonstrated by the continuous joints and cell walls, which were used to build a three-dimensional alumina network in the composites. A remarkable enhancement of thermal conductivity was revealed for PAC/SR composites in comparison with AP/SR composites and neat SR. The thermal conductivity of the PAC/SR composites with an alumina content of 14 vol% reached $0.84 \text{ W} \cdot (\text{m} \cdot \text{K})^{-1}$, which was 3.1 times that of the AP/SR composites with the same alumina content and 5.3 times than the neat SR. The Maxwell model and S-P model were found to well fit the experimental results of AP/SR and PAC/SR, respectively. The effects of PAC on the thermal conductivity of the composites can be explained by the decreased interfacial thermal resistance between the filler and the matrix, as well as that between the fillers due to the densely packed alumina particles. Such an effective thermal conductive network was beneficial to accelerate the heat dissipation and resulted in the better heat transfer performances of PAC14/SR. As a result, much lower loading of PAC would be required to obtain a certain thermal conductivity of the silicone-based composites in contrast to AP, which gave rise

to a lower dielectric permittivity of the composites and would meet the developing demands for TIM.

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