

High thermal conductivity epoxy composites with bimodal distribution of aluminum nitride and boron nitride fillers

Jung-Pyo Hong^a, Sung-Woon Yoon^a, Taeseon Hwang^a, Joon-Suk Oh^a, Seung-Chul Hong^a,
Youngkwan Lee^b, Jae-Do Nam^{a,c,*}

^a Department of Polymer Science and Engineering, Sungkyunkwan University, 300 Chunchun-dong, Jangnan-gu, Suwon 440-746, South Korea

^b School of Chemical Engineering, Sungkyunkwan University, 300 Chunchun-dong, Jangnan-gu, Suwon 440-746, South Korea

^c Department of Energy Science, Sungkyunkwan University, 300 Chunchun-dong, Jangnan-gu, Suwon 440-746, South Korea

ARTICLE INFO

Article history:

Received 27 October 2011

Received in revised form 6 February 2012

Accepted 6 March 2012

Available online 15 March 2012

Keywords:

Aluminum nitride

Boron nitride

Thermal conductivity

Epoxy composite

ABSTRACT

High thermal-conductivity fillers of aluminum nitride (AlN) and boron nitride (BN) were incorporated in the epoxy matrix in order to identify the effects of the particle size and the relative composition on the thermal conductivity of composites. In the bimodal distribution of polygonal AlN and planar BN particles, the optimal thermal conductive path was strongly affected by the packing efficiency and interfacial resistance of the particles in a sensitive way and, consequently, the maximum thermal conductivity was achieved up to 8.0 W/mK in the 1:1 volume ratio of AlN:BN particles. In the optimal volume ratio of the two fillers at 1:1, the relative filler size, which was represented by the shape factor (or the diameter ratio of the two filler particles, R_D), also influenced the thermal conductivity giving the maximum conductivity at the shape factor $R_D \approx 1$. The optimal morphology and composition of the AlN/BN composite systems were clearly visualized and thoroughly discussed in the filler distribution curves plotting the filler-appearance frequency as a function of particle size. The developed methodology validated that two different particles should be packed well to fill up the interstitial space and, simultaneously, the contact resistance and the contact area of the fillers should be optimized to maximize the thermal conductivity.

© 2012 Published by Elsevier B.V.

1. Introduction

As microelectronic devices become increasingly integrated and used at high powers and high frequencies, a large amount of heat is generated and thus it should be dissipated quickly through the printed circuit boards and/or electronic devices, e.g. in such applications as light emitting diodes (LEDs), highly-integrated memory chips, etc. The generated heat could increase the temperature over the thermal-stability limit of the device to cause fatal damages [1–3]. In addition, the accumulated heat often induces thermal fatigue and chemical reactions, which substantially reduces the service life and operation efficiency. For example, the performance of LEDs is reported to degrade exponentially with increasing temperature above 90 °C due to the thermal degradation of the light-emitting materials [4].

Accordingly, various dielectric polymeric composite systems have been investigated to achieve high thermal conductivity using thermally-conductive but electrically-nonconductive fillers such as silica, aluminum oxide, silicon carbide, aluminum nitride (AlN),

and boron nitride (BN) [1,5–9]. In these filler systems, the particle size and filler content have been reported to be the major factors affecting the thermal conductivity [7,10–12], where the efficient packing increases the loading density of the fillers in the polymer matrices. Compared with a unimodal particle distribution, the bimodal distribution of the fillers has been reported to increase the thermal conductivity by 130% [13]. In the schematic of appearance frequency plotted as a function of particle size (Fig. 1), the bimodal distribution is compared with two separate unimodal distribution curves. In the bimodal distribution, smaller particles can desirably fill the interstitial space of the larger particles so as to increase the packing density of the fillers, which is represented by the continuous valley formed by the overlap of two different unimodal distribution curves. It is believed that the overlapped filler frequency in the bimodal distribution may very well enhance the packing efficiency to give enhanced thermal conductivity of composite materials. In composite preparation, the particle size and composition should be controlled in an appropriate way to make the frequency-distribution curve to be well overlapped and positioned in the desired position of particle size.

Although the intrinsic thermal conductivity of AlN (180–200 W/mK) is higher than BN (60–100 W/mK), the thermal conductivity of BN composites is reported to be higher than

* Corresponding author. Tel.: +82 10 3032 7285; fax: +82 31 299 4069.

E-mail address: jdnam@skku.edu (J.-D. Nam).

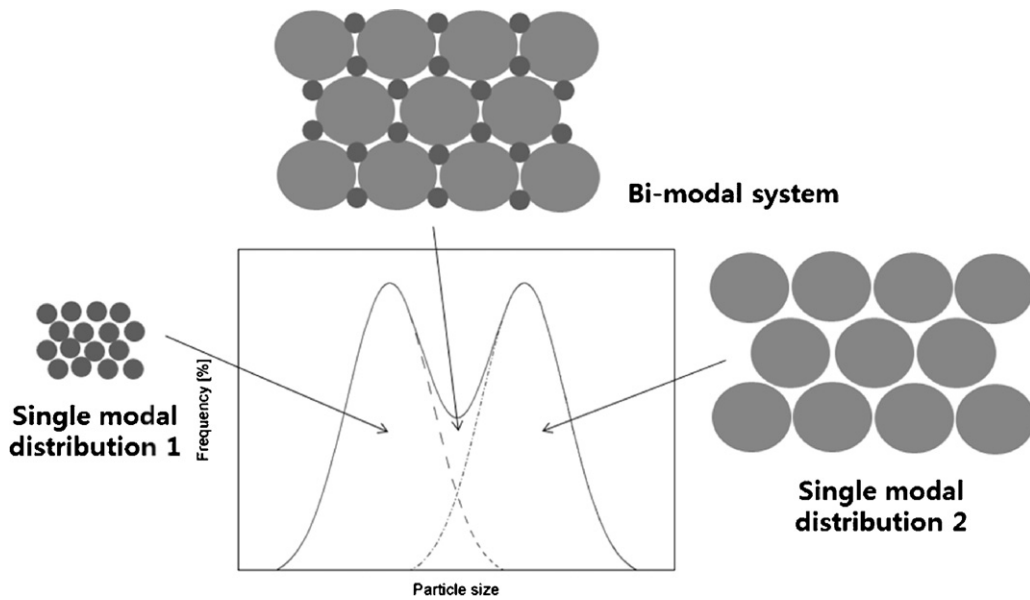


Fig. 1. Schematic of the unimodal and bimodal distributions, and the continuous valley formed by the two overlapped unimodal distribution curves.

that of AlN composites, e.g. 1.2 W/mK and 0.6 W/mK at 30 vol.%, respectively, [14]. It is likely that the BN particles, which have a planar shape, allow a favorable filler packing and network formation, thus providing facile heat dissipation in the in-plane direction of the composites. Since the heat dissipation is greatly influenced by the shape of the fillers, it has been quantified by the aspect ratio of the particles referred to as the “shape factor.” The thermal conductivity of composites has been reported to change with shape factors [14,15]. In addition to the shape, it should be addressed that the particle size may very well influence the thermal conductivity, because it changes the overall contact area of the fillers, interfacial thermal resistance, conducting path, etc.

In this study, hybrid multimodal composite systems composed of AlN and BN were investigated in order to identify the optimal bimodal distribution of two filler particles. The AlN and BN composites were designed to identify the key factors to achieve the optimal heat-conduction paths in the hybrid composite systems. The filler size and relative composition of the two different shaped fillers were thoroughly investigated by measuring thermal conductivities.

2. Experimental

Having different particle sizes, four different types of AlN and three different types of BN particles were used for the hybrid filler systems in this study, as represented by A1, A20, A50 and A150 (SURMET, USA) for the former and B18, B5 and B1 (DENKA, Japan) for the latter. As summarized in Table 1, the mean particle sizes (D_{50}) of AlN and BN were changed in the range of 1.13–25.5 μm

and 1–18 μm , respectively. Their information on particle size was provided from the manufacturer.

The epoxy and hardener used as the matrix system in this study were bisphenol A diglycidyl ether (DGEBA) and methyl tetrahydrophthalic anhydride (MTHPA), respectively, purchased from Kukdo Chemical. The catalyst was 1-methylimidazole (1-MI) and the surface modifier of the fillers was 3-aminopropyl-triethoxy silane (aminosilane), both purchased from Aldrich.

Three different hybrid systems were prepared as summarized in Table 2: in Case 1, AlN is bigger than BN ($D_{\text{AlN}} > D_{\text{BN}}$), in Case 2, AlN is similar to BN ($D_{\text{AlN}} \approx D_{\text{BN}}$), and in Case 3, AlN is smaller than BN ($D_{\text{AlN}} < D_{\text{BN}}$) in terms of their mean particle sizes. In addition, compared with Case 2, Case 2' was designed to evaluate the effect of the particle size of AlN, while maintaining $D_{\text{AlN}} \approx D_{\text{BN}}$. More specifically, the particle sizes of AlN in Case 2 and Case 2' were 14.4 μm and 25 μm , respectively, while keeping the size of BN at 18 μm , so that we could keep the condition of $D_{\text{AlN}} \approx D_{\text{BN}}$.

The surface of the AlN and BN particles was pre-treated using an aminosilane to minimize the thermal resistance at the particle surface [1,16,17]. More details of the pre-treatment can be found elsewhere [1,16]. An epoxy resin system consisting of YD-128, MTHPA, and 1-MI was mixed with the silane-treated fillers at room temperature for 5 min with mechanical stirring. The total filler content was adjusted to 80 vol.%. The mixture was cured in a mold at 3000 psi and 80 $^{\circ}\text{C}$ for 4 h, followed by 2 h of holding at 145 $^{\circ}\text{C}$. The resulting composite samples had a thickness of 1 ± 0.5 mm and a diameter of 12.7 mm.

In order to observe a cross section of composite specimens, the each specimen was molded using EpoxySet (Allied High Tech

Table 1
Summary of physical properties and sizes of fillers used in this study.

	A150	A50	A20	A1	B18	B5	B1
Chemical formula	AlN	AlN	AlN	AlN	BN	BN	BN
Commercial grade	A500–150	A500–50	A500–20	H	SGP	HGP	MBN
Tap density (g/cm^3)	2.09	1.9	1.6	0.43	0.8	0.4	0.3
Particle size (μm)							
D10	4.2	3.3	2.5	–	5.4	1.9	–
D50	25.5	14.4	8.67	1.13	18	6	1
D90	113.2	39.9	18.8	–	41.6	10.6	–
Specific surface area (m^2/g)	–	–	0.052	2.59	2	11	14.4
Oxygen content (wt.%)	–	–	–	–	0.3	1	5.5

Download English Version:

<https://daneshyari.com/en/article/674179>

Download Persian Version:

<https://daneshyari.com/article/674179>

[Daneshyari.com](https://daneshyari.com)