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

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

\textsuperscript{a} Department of Polymer Science and Engineering, Sungkyunkwan University, 300 Chunchun-dong, Jangan-gu, Suwon 440-746, South Korea
\textsuperscript{b} School of Chemical Engineering, Sungkyunkwan University, 300 Chunchun-dong, Jangan-gu, Suwon 440-746, South Korea
\textsuperscript{c} Department of Energy Science, Sungkyunkwan University, 300 Chunchun-dong, Jangan-gu, Suwon 440-747, South Korea

\textbf{A R T I C L E   I N F O}

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

\textbf{A B S T R A C T}

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, \(R_0\)), also influenced the thermal conductivity giving the maximum conductivity at the shape factor \(R_0 \approx 1\). 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.

\begin{itemize}
\item[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