Development of Epoxy-Matrix Composite with Both High-Thermal Conductivity and Low-Dielectric Constant via Hybrid Filler Systems

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ABSTRACT: Materials used for printed circuit boards (PCBs) need to have more multifunctional properties, such as excellent thermal, electrical, and mechanical properties at the same time. Up to now, a sole polymer or single filler-filled polymer composites is hard to satisfy the demand for more multifunctional properties, especially to obtain high-thermal conductivity and low-dielectric constant (D_k) simultaneously. In this study, two hybrid filler systems [i.e., hollow glass microsphere (HGM) and aluminum nitride (AlN), HGM, and boron nitride (BN)] were filled into epoxy matrix in an attempt to reach a composite with high-thermal conductivity and low D_k at the same time. By varying the size, shape, and volume fraction of hybrid fillers, a new kind of epoxy-matrix composite both with high-thermal conductivity and low D_k as well as high-glass transition temperature (T_g) and low coefficient of thermal expansion (CTE) and dielectric loss factor (D_f) were developed as candidate for future PCB markets. In addition, multiphase models describing the thermal conductivity and D_k were developed, and suitable models were recommended for present materials system. © 2009 Wiley Periodicals, Inc. J Appl Polym Sci 116: 518–527, 2010

Key words: polymer-matrix composites; hybrid filler; thermal conductivity; dielectric constant; dielectric loss factor; glass transition temperature; coefficient of thermal expansion

INTRODUCTION

The applications of printed circuit boards (PCBs) have technical demands for the future evolution to high-frequency appliances. As the working frequency of electronic appliances increases, signal intensity losses become more sensitive. Circuit substrates with poor dielectric properties will maintain an unfavorable signal-conveying efficiency. Therefore, demand is high in high-frequency appliance markets for small dielectric constant (D_k) and low-dissipation energy substrates to increase the velocity of signal propagation.[1–4] Meanwhile, thermally conductivity is increasingly important for electronic packaging because the heat dissipation ability limits the reliability, performance, and miniaturization of electronics.[5–7] In addition to high-thermal conductivity and high-electrical resistivity, a low coefficient of thermal expansion (CTE) is needed for resistance to thermal fatigue.[8] Thus, materials used for PCBs need to have more multifunctional properties, such as excellent thermal, electrical, and mechanical properties at the same time. Up to now, a sole polymer material is hard to satisfy the demand for more multifunctional properties because of its high CTE and low-thermal conductivity.[9] Thermal-conductive ceramic particles-filled composites are potentially used as the primary materials for PCBs, because of their high-thermal conductivity. However, they have unacceptable dielectric constant for use in high-frequency appliances.[10–12]

In this study, a combination of high-thermal-conductive filler [aluminum nitride (AlN) and boron nitide (BN)] with low-dielectric filler (hollow glass microsphere, HGM) was filled into epoxy matrix. By varying the size, shape, volume fraction, and composition of fillers, a new kind of polymer-matrix composite both with high-thermal conductivity and good dielectric properties will be developed. A few theoretical issues will also be addressed along with the investigation, namely: a theoretical or empirical three-phase model describing the thermal

TABLE I
Material Properties of Fillers Used

	True density (g/cm^3)	Particle shape	Average particle size (μm)	CTE (ppm/°C)	Dielectric constant	Thermal conductivity (W/mK)
HGM	0.6	Sphericity	30	6–8	2	0.2077
BN	2.28	Platelet	7×0.7^a	<0.5	4	300
		Rough sphericity	0.07			
AlN	3.26	Rough sphericity	2.3	4.5	8.5	270

^{a} The average diameter and thickness of platelet are 7 and 0.7 μm, respectively.

conductivity and D_k. It can be expected that this study will be of importance for new packaging technologies of further increasing of working frequency and miniaturization of electronic devices.

EXPERIMENTAL PROCEDURE

Raw materials

As filler, AlN powders were purchased from Hefei Kiln Nanometer Technology Development Co. (China); BN fillers were supplied by Advanced Ceramics and Zibo ShineSo Chemical New Material Co.; HGMs were provided by 3M Hong Kong. Table I lists the material properties of the fillers. The coupling agent was 3-glycidoxypropyltrimethoxysilane (trade name: Markosi$^@$ Silane KH-560), purchased from Zhenjiang Wholemarket Import and Export Co. The epoxies used as the matrix were Epon 8008 and Epon 1031, purchased from Huntsman Co. The chemical structure of Epon 8008 and Epon 1031 are shown in Figure 1. Epon 8008 is a kind of brominated epoxy resin that has been specially designed to meet the stringent requirements of the PCBs industry. It has an epoxide equivalent of 410–460 g/Eq and a bromine content of 19.0–21.0% w/w. Epon 1031 is a solid multifunctional epichlorohydrin/tetraphenylol ethane epoxy resin with an epoxy group

content of 4350–5130 mmol/kg. Epon 1031 is used to improve the properties of cured epoxy resin systems, particularly at elevated temperatures. In this study, Epon 8008 and Epon 1301 have been dissolved in acetone, and their content in the solution are 80% and 70%w/w, respectively. The curing agent dicyandiamide (DICY, purity >99.5%) and the accelerator 2-methylimidazole (2-MI, purity >99.0%) were obtained from Neuto Products and Tokyo Kasei Kogyo, respectively. The DICY particles had an average diameter of <1 mm. DICY and 2-MI were used as received.

Procedure for composite preparation

The composites were fabricated according to the following steps: (i) The surface of the fillers was pretreated with KH-560 coupling agent, with the amount used being 5 wt % based on the weight of the fillers; (ii) The Epon 8008 solution, Epon 1031 solution, DICY, and 2-MI were weighted at a fixed weight ratio of 100 : 7.349 : 2.552 : 0.056, and different percentages of filler were weighted with respect to the weight of Epon 8008 solution. In the case of HGM and AlN hybrid filler, the HGM percentage increased from 0 to 30 wt % with the interval of 10 wt %, and at every HGM percentage, a set of AlN varied from 0 to 60 wt % with the interval of
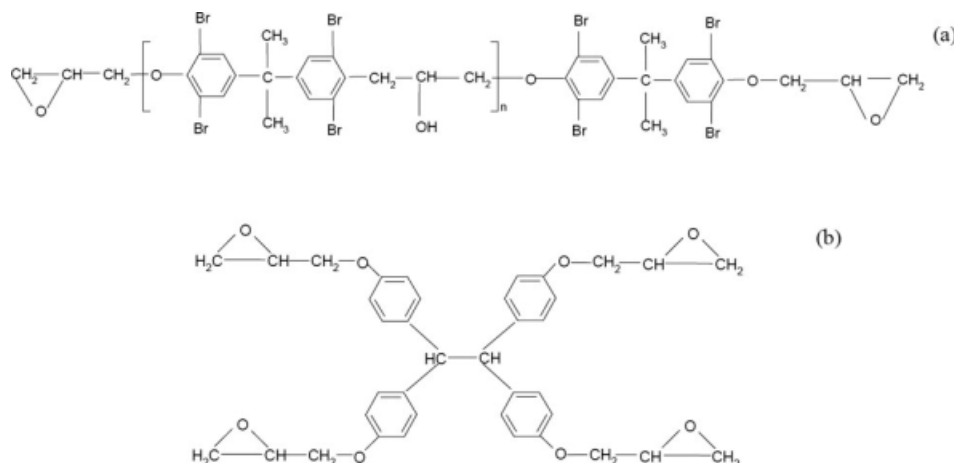

Figure 1 The chemical structure of epoxy resin used are as follows: (a) Epon 8008 and (b) Epon 1031.

20 wt %. In the case of HGM and BN hybrid filler, the HGM percentage also increased from 0 to 30 wt % with the interval of 10 wt %, but at every HGM percentage, a set of BN varied from 0 to 30 wt % with the interval of 10 wt %, in which all the BN used is the mixture of the roughly spherical-shaped BN and platelet-shaped BN with fixed ratio of 1/3; (iii) The mixtures, diluted by a small quantity of acetone, were stirred by a high-speed mixing machine for 1 h; (iv) The uniformly formed mixtures were cast in a mold and were pumped for about 1 h to remove the air bubbles and solvent as the temperature gradually increased from 90 to 150°C; and (v) Finally, they were heated to 175°C for 4 h to complete polymerization.

Characterization

Filler volume fraction

The volume percent of filler in the composites was determined using the following equation:

$$v_{\text{fi}} = \frac{W_{\text{fi}}/\rho_{\text{fi}}}{\sum_{i=1}^{n}(W_{\text{fi}}/\rho_{\text{fi}}) + W_m/\rho_m}, \tag{1}$$

where W_{fi} and W_m are the weight of i filler and matrix in the composite, and they can be determined according to weighted mass; ρ_{fi} and ρ_m are the densities of the i fillers and pure epoxy (1.3846 g/cm^3), respectively.

Thermal mechanical analysis

The CTE and glass transition temperature (T_g) were measured with a Perkin-Elmer thermal mechanical analyser (TMA-7). The temperature range used was from 30 to 220°C, and the heating rate was 10°C/min. All reported TMA data were obtained from a second heating cycle.

Thermal conductivity measurement

The thermal conductivity was given by the product of the thermal diffusivity, specific heat, and density. The thermal diffusivity and specific heat of composites were measured by Flashline$^{\text{TM}}$ 3000 Thermal Properties Analyzer (Anter Corporation). The samples were cylindrical shaped, 12.7 mm in diameter, and 0.8–1.2 mm in thickness.

Dielectric properties measurement

Dielectric constant (D_k) and loss factor (D_f) measurements were taken on an Agilent-4294A impedance analyzer with an applied AC voltage of 500 mV; the
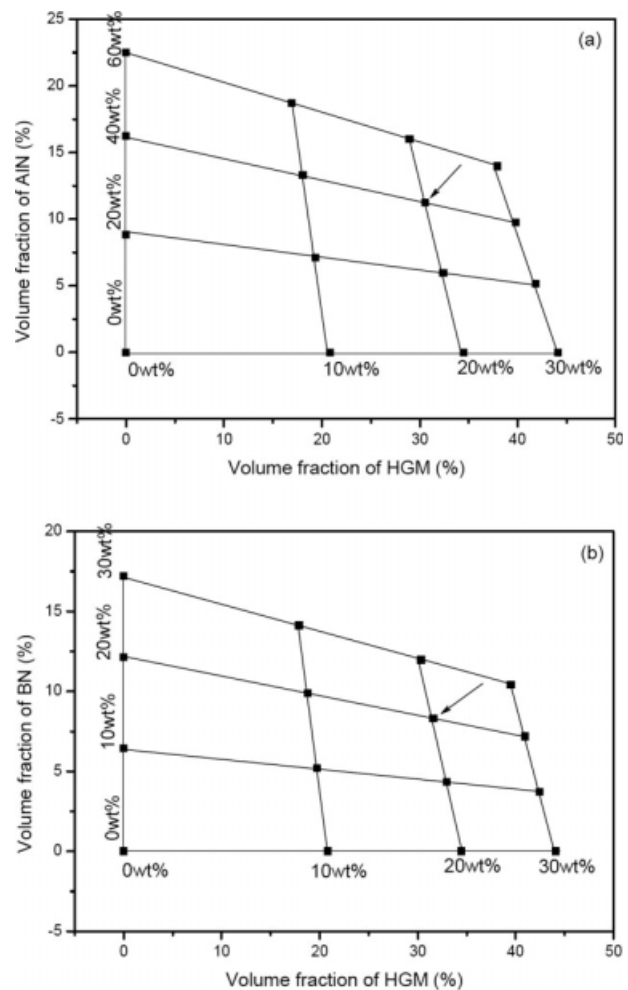
Figure 2 The volume fraction of hybrid fillers in the composites: HGM and AlN (a); HGM and BN (b).

frequency range was 1 KHz–1 MHz. The used samples were disk-shaped, and both sides of the samples were coated with silver paste. The D_k was calculated from capacitance by $D_k = Ct/\varepsilon_0 A$, where t was the thickness of the disks, ε_0 the vacuum dielectric constant, and A the disk area.

EXPERIMENTAL RESULTS AND DISCUSSION

Volume fraction

The volume fraction of the filler in the composites is listed in Figure 2, and weighted filler percentage is also marked for studying the relationship between volume fraction and filler percentage. For single HGM-filled composites (namely, AlN or BN content is zero), volume fractions of HGM are 0, 20.8, 34.5, and 44.1% with increasing HGM percentage from 0 to 30 wt %. For single ALN-filled composites (namely, HGM content is zero), volume fractions of AlN are 0, 8.8, 16.2, and 22.5% with increasing AlN percentage from 0 to 60 wt %. For single BN-filled composites (namely, HGM content is zero), volume

fractions of BN are 0, 6.5, 12.2, and 17.2% with increasing BN percentage from 0 to 30 wt %. At the fixed one filler percentage, the volume fraction of this filler slightly decreases with increasing the other filler percentage, and volume fraction of the other filler relatively increases at the same time. For example, when the HGM percentage is fixed at 20 wt %, its volume fraction gradually decrease from 20.8% to 16.9% with increasing AlN percentage from 0 to 60 wt %, and the volume fraction of AlN increase from 0 to 18.7%. The total volume fraction of the filler increases with increasing other filler percentage at the same percentage of one filler. In the case of HGM and BN hybrid filler, the total volume faction of filler in the composites containing higher percentage HGM is always larger than that in the composites containing lower percentage HGM. For example, the total volume fraction at HGM of 20 wt % (34.5%) is larger than that at HGM of 10 wt % and BN of 30 wt % (32%). As for HGM and AlN hybrid filler, the total volume faction at higher percentage HGM is slightly lower than that at lower percentage HGM and maximal percentage AlN. For example, the total volume fraction at HGM of 20 wt % (34.5%) is slightly lower than that at HGM of 10 wt % and AlN of 60 wt % (35.6%).

T_g and CTE

Figure 3 shows the 3D representation of T_g of the composites as a function of volume fraction of filler. Comparing the T_g between single HGM-, AlN-, and BN-filled composites, it is found that the T_g increase with increasing HGM or AlN content, but it first increases then decreases with increasing BN content. In the case of HGM and AlN hybrid filler, the T_g of the composites increases with increasing the AlN content if only HGM content below 37.9 vol %. When the HGM content above 37.9 vol %, the T_g of the composites first increases then decreases with increasing AlN content. This change trend is also observed when the HGM content is varied and AlN content is fixed at the same time, that is, the T_g of the composites increases with increasing the HGM content if only AlN content below 9.8 vol %, but it first increases then increases with increasing HGM content at AlN content above 9.8 vol %. Thus, lower T_g is observed at HGM above 37.9 vol % and AlN above 9.8 vol %. In the case of HGM and BN hybrid filler, the relation of T_g and filler content shows some differences from that of HGM and AlN hybrid filler. At the fixed HGM content, the T_g of the composites first increases then decreases with increasing BN content, which is similar with that of single BN-filled composites. At the same lower BN content, the T_g of the composites first decreases then increases with increasing HGM content. At the same higher
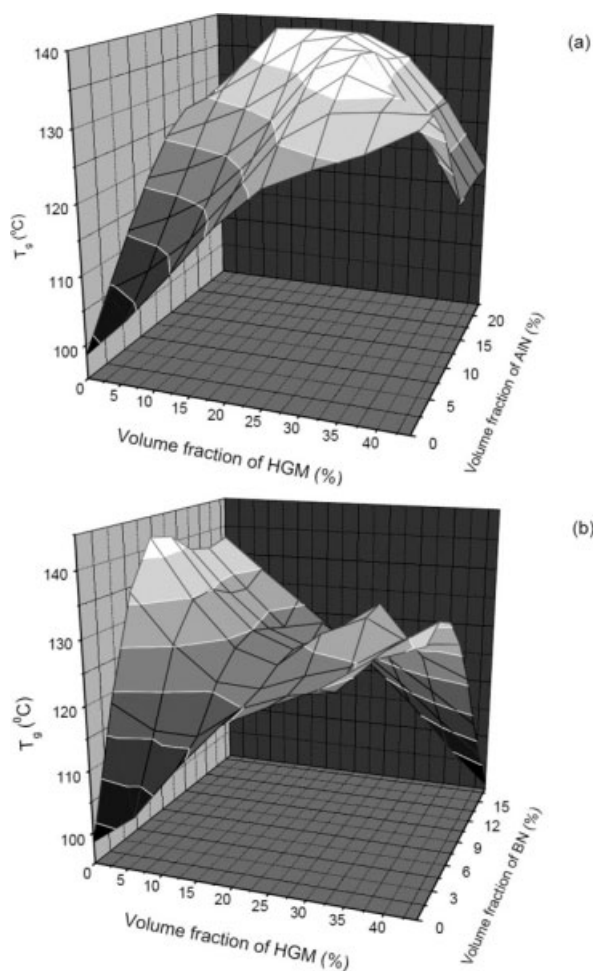

Figure 3 3D representation of T_g of the composites as a function of the volume fraction of HGM and AlN fillers (a) and as a function of volume fraction of HGM and BN (b).

BN content, the T_g show a change trend of decrease-increase-decrease with HGM content. Also, lower T_g of the composites is observed at HGM and BN content above certain values simultaneously.

Theoretically, the removal of epoxy chain can be restricted by filler due to the interaction between filler and epoxy matrix when filler is filled into epoxy matrix, and thus the T_g increases compared with pure epoxy. What's more, the interaction between epoxy matrix and filler enhances with filler content, and thus the T_g increases with increasing filler content. As for single AlN- or HGM-filled composite, it is observed that T_g exhibit increased trend with AlN or HGM content. As for single BN-filled composites, the T_g begins to decrease with high filler content, which indicates T_g is also affected by other factors. During the experiment, it was found that BN can easily cause high viscosity of the mixture compared with HGM and AlN filler. Literature also showed that BN cause remarked increase in viscosity of

mixture than other ceramic filler even if at small volume fraction, and thus the maximum filler content of BN in the composite was relatively small.[13,14] Oppositely, HGM will not remarkably increase the viscosity even if high HGM content. The moldability becomes difficult with increase in viscosity. The removal of solvent and bubble also becomes difficult, and thus result in weak interaction between filler and epoxy matrix. The weak interaction implies free volume of epoxy chain increase and thus T_g decreases. On the other hand, increase in viscosity also results in decrease in crosslinking density, which also cause decrease in T_g.[15] According to above discussion, when single AlN, HGM, and BN fillers are filled into composite in this study, the viscosity of the mixture was not obviously increased by AlN due to its low content, and also it was not increased by HGM due to its intrinsic properties even if at high content, but it was obviously increased by BN due to its intrinsic properties even if at low content. It seems that the effect of viscosity increase can be neglected for single HGM- or AlN-filled composite, and thus the T_g increases with increasing HGM or AlN content. Considering that the effect of the viscosity increase, the T_g show a decreased trend with BN content. When HGM and AlN hybrid filler is filled into composites and their content is above certain value, the effect of viscosity increase will enhance, and thus the T_g begins to decrease.

The effects of filler fraction on the CTE of composites are shown in Figure 4. Because of lower CTE of HGM, AlN, and BN than that of epoxy matrix, the CTE of the composites decrease with increasing HGM, AlN, or BN content. The CTE of pure epoxy is 62 ppm/°C. When single HGM, AlN, and BN filled into composites, CTE can be decrease to 32.92, 39.6, and 38.51 ppm/°C, respectively. Considering that the volume fractions of HGM, AlN, and BN are 44.1, 22.5, and 17.2%, respectively, the efficiency of decreasing CTE of the composites is BN > AlN > HGM, which is consistent with their intrinsic CTE. Obviously, the minimum CTE of the composites is obtained at maximal HGM and AlN content or maximal HGM and BN content.

Thermal conductivity

Figure 5 shows that the thermal conductivity of composites containing hybrid filler systems. In the case of single HGM-, AlN-, and BN-filled composites, the thermal conductivity of HGM-filled composite decrease due to its lower thermal conductivity of HGM than epoxy, but that of AlN- and BN-filled composites increase due to their higher thermal conductivity than epoxy. Compared with the thermal conductivity between AlN- and BN-filled composite,
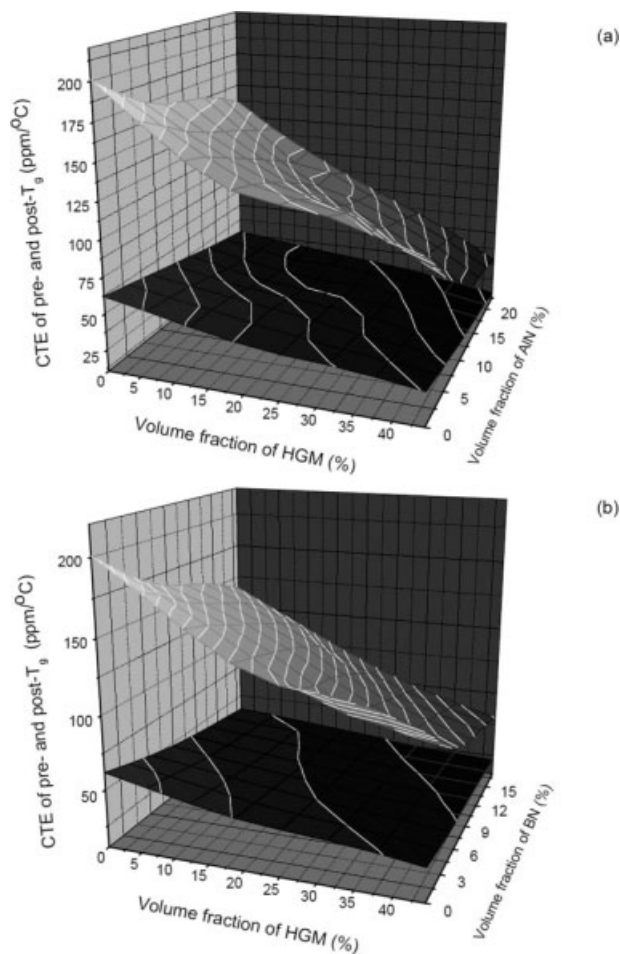

Figure 4 3D representation of CTE of pre- and post-T_g of the composites as a function of the volume fraction of HGM and AlN fillers (a) and as a function of the volume fraction of HGM and BN fillers (b).

it is found that the thermal conductivity of BN-filled composite is higher than that of AlN-filled composites. This is because the intrinsic conductivity of BN is greater than that of AlN, and it is also related to the filler shape and size. BN fillers consist of micro-sized platelet-shaped BN and nanosized spherical-shaped BN, which enhances the formation of the conductive network. In the case of HGM and AlN hybrid filler, the thermal conductivity of the composite increases with increasing AlN content at the fixed HGM content, but it decreases with increasing HGM content at fixed AlN content. In the case of HGM and BN hybrid filler in Figure 5(b), the change trend of the thermal conductivity is similar to that in HGM and AlN system except that the higher thermal conductivity is obtained for HGM and BN hybrid filler. It may be due to higher thermal conductivity of BN than AlN and special shape and size of BN, as mentioned earlier.

To predict the effective thermal conductivity of polymer composite materials, many theoretical and
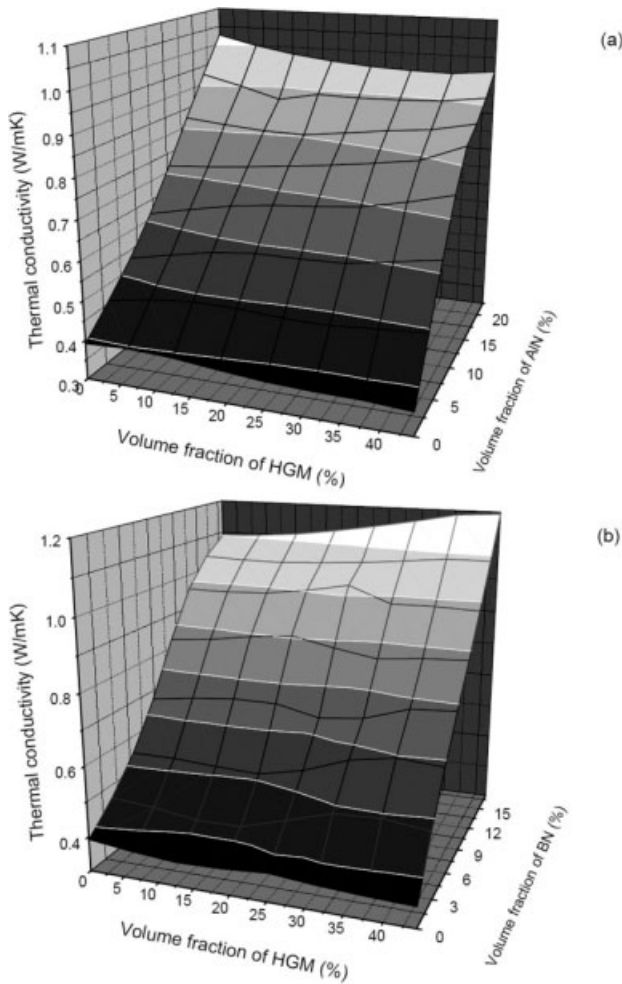
Figure 5 3D representation of thermal conductivity of the composites as a function of the volume fraction of HGM and AlN fillers (a) and as a function of the volume fraction of HGM and BN fillers (b).

semiempirical models have been developed. However, most models were developed based two-phase composite. In this case, we may first establish the relationship between the properties of the composites and one filler, then the composites with one filler can be regarded as a new matrix at fixed filler content, thus the relation between the properties and other filler can be established by using published models, and finally, the three-phase equations will be developed. For some two-phase models, it is easy to develop it to multiphase system. For example, the following eqs. (2)–(4) can be used to predict the thermal conductivity of multiphase system[16]:

$$k^n = \sum_{i=1}^{m} \phi_i k_i^n \tag{2}$$

$$\log k = \sum_{i=1}^{m} \phi_i \log k_i \tag{3}$$

$$\frac{k}{k_1} = \frac{1 + \sum_{i=2}^{m} A_i B_i \phi_i}{1 - \sum_{i=2}^{m} B_i \psi_i \phi_i} \tag{4.1}$$

$$B_i = \frac{k_i/k_1 - 1}{k_i/k_1 + A_i} \tag{4.2}$$

$$\psi_i \cong 1 + \frac{1 - \phi_{mi}}{\phi_{mi}^2} \phi_i \tag{4.3}$$

$$\psi_i \cong 1 + \frac{\phi_1}{\phi_{mi}} [\phi_{mi} \phi_i (1 - \phi_{mi}) \phi_1] \tag{4.4}$$

In all above equations, k is the thermal conductivity of the composite; k_1 is the thermal conductivity of the matrix; k_i is the thermal conductivity of filler i ($i = 2, 3\ldots\ldots m$); ϕ_1 is the volume fraction of matrix material; ϕ_i is the volume fraction of filler i ($i = 2, 3\ldots\ldots m$). In the case of the eq. (2), the exponent n is an empirical constant, varying from $+1$ to -1. In the case of eq. (4), ϕ_{mi} is the maximum packing fraction of filler i ($i = 2, 3\ldots\ldots m$). When $n = 1$ and $n = -1$ in eq. (2), the equation corresponds to the parallel and series models, the simplest alternatives, which suppose that the constituents in composites are arranged in either parallel or series with respect to heat flow. The parallel and series models usually give the upper or lower bounds of effective thermal conductivity of composites, and the measured values lie between them. In most general cases, composite are chaotic or statistical mixtures of several components, thus the n is taken as a certain value between -1 and 1. The eq. (3) is based on geometric mean model, which generally fits the thermal conductivity data better than the parallel and series models. Equation (4) is the Nielsen model in the case of multiphase system. The ψ_i term can be calculated from eqs. (4.3) and (4.4), and eq. (4.4) is referred as the modified term in this article. To use Nielsen's model, two parameters (A and ϕ_m) are chosen for each filler according to their morphology and size (shown in Table II).[16–18]

Figure 6 shows the experimental values and calculated values from the eqs. (2)–(4) for the thermal conductivity of the composites. In the case of HGM and AlN hybrid filler, it is seen that experimental

TABLE II
Parameter A and ϕ_m Values for Different Fillers

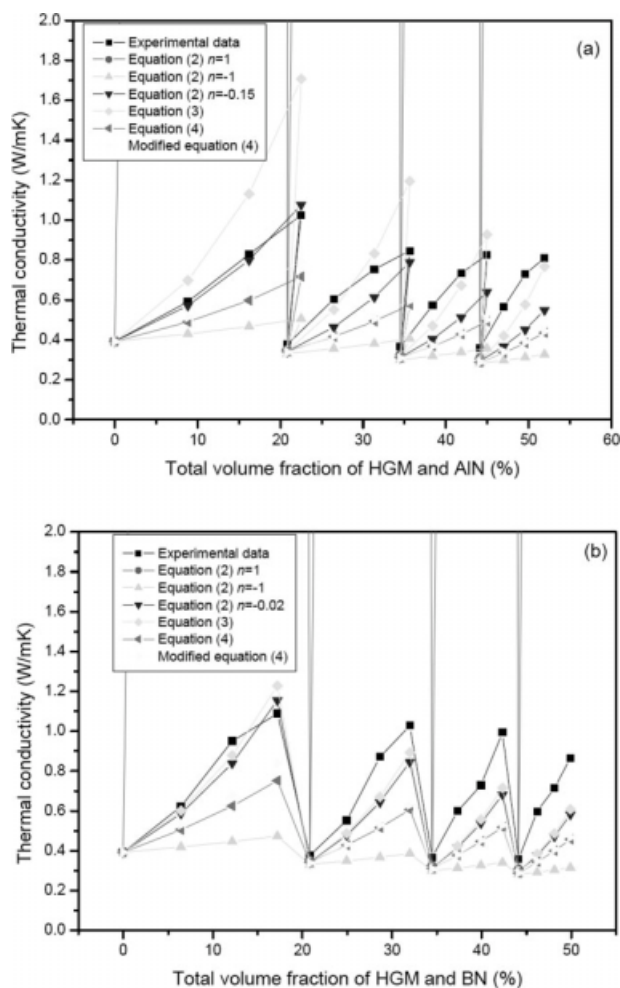|	HGM	AlN	Spherical-shaped BN	Platelet-shaped BN
A	1.5	1.5	1.5	3.99
ϕ_m	0.601	0.637	0.637	0.52

Figure 6 Comparing the experimental values of thermal conductivity with calculated values from different equations for HGM and AlN fillers (a) and for HGM- and BN-(b) filled composites.

values of the thermal conductivity is between that of calculated value from the eq. (2) when $n = 1$ and $n = -1$, as mentioned earlier. When $n = -0.15$ in eq. (2), the calculated values are close to the experimental values at low total volume fraction, namely, single AlN-filled composite. Further increasing total volume fraction, namely, single HGM- or HGM- and AlN-filled composites, the calculated values are lower than experimental values. At low total volume fraction, the calculated value from eq. (3) is higher than experimental values, but it is gradually lower than experimental values with increasing total volume fraction. The calculated values from modified eq. (4) are slightly higher than that from eq. (4), but both of them are lower than experimental values. In the case of HGM and BN, the change trend is similar to that of HGM and AlN hybrid filler. Also, when n is taken a certain value ($n = -0.02$), the calculated values are close to the experimental

values at low total volume fraction, but are obviously lower experimental values at high total volume fraction. In addition, the calculated values from eq. (3) are close to that from eq. (2) when $n = -0.02$. It can be observed that the predictions of parallel, series, and Nielsen model deviated significantly from the experimental data, and these models underestimate the thermal conductivity. The reason for the deviations is probably that the theoretical models do not take into account the state of filler dispersion in the composite. Another factor is probably that each model shows good agreement with experimental data, only in certain composite systems. The experimental data are well fitted into the eq. (2) when n is taken a certain value, as discussed earlier. This is not surprising because the empirical nature of the equation makes it suitable for most filled systems by the correct use of the adjustable constant. At the same time, this indicates that the formation of conductive chains may be taken into account in eq. (2).

Dielectric properties

Figure 7 shows the D_k of the composites as a function of filler content. For single HGM-, AlN-, or BN-filled composites, the D_k decrease with increasing HGM content and it increases with increasing AlN content, but it shows a slight change with increasing BN content. In the case of HGM and AlN or HGM and BN hybrid filler, the D_k increases with increasing AlN or BN content at the fixed HGM content, but it decreases with increasing HGM content at the fixed AlN or BN content. The D_k of HGM- and BN-filled composites is smaller than that of HGM- and AlN-filled composites at the same filler content. All these results can be attributed to the D_k order of the epoxy matrix and fillers being AlN > epoxy ≈ BN > HGM.

To predict the D_k of the composites, many models have been developed. However, most models are based on two-phase system. Also, some models can be developed to use in multiphase system. The following eqs. (5) and (6) are typical theoretical models that can be used to predict D_k of multiphase system[19,20]:

$$\varepsilon^n = \sum_{i=1}^{m} \phi_i \varepsilon_i^n \qquad (5)$$

$$\log \varepsilon = \sum_{i=1}^{m} \phi_i \log \varepsilon_i \qquad (6)$$

where ε is the dielectric constant of the composite; ε_1 is the dielectric constant of the matrix; ε_i is the dielectric constant of filler i ($i = 2, 3…..m$); ϕ_1 is
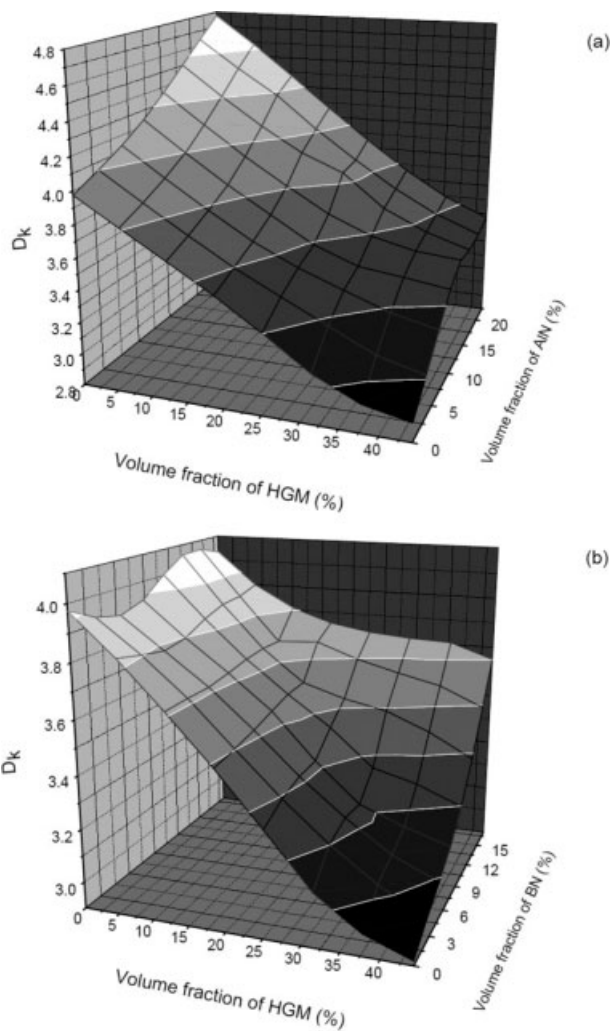
Figure 7 3D representation of D_k of the composites as a function of the volume fraction of HGM and AlN fillers (a) and as a function of the volume fraction of HGM and BN fillers.

the volume fraction of matrix material; ϕ_i is the volume fraction of filler i ($i= 2, 3…..m$). In eq. (5), the exponent n is an empirical constant, varying from $+1$ to -1. As similar as the thermal conductivity of the composites, in the simplest case $n = 1$ and $n = -1$, the equation corresponds to parallel and series models, respectively, which assumes that dielectric system consists of two different homogeneous dielectrics connected in parallel or in series.

Figure 8 shows the experimental values and calculated values from the eqs. (5) and (6) for the D_k of the composites. In the case of HGM and AlN hybrid filler, it is seen that experimental values of the D_k is between that of calculated value from the eq. (2) when $n = 1$ and $n = -1$, as mentioned earlier. When $n = 0.1$ in eq. (5), the calculated values are close to the experimental values at all the total volume fraction. The calculated values from eq. (6)

are also close to the experimental values, as shown in Figure 8(a). These results show that the eqs. (5) and (6) are suitable to predict the D_k of the HGM and AlN hybrid filler-filled composite. For the HGM and BN hybrid filler-filled composites, the experimental value of the D_k is not entirely between that of calculated value from the eq. (2) when $n = 1$ and $n = -1$. When the high BN content is in the composites, the experimental values are higher than the calculated value from the eq. (5), which is related to the high viscosity caused by BN. Because of high viscosity, the solvent may not be removed completely during preparation. The remaining solvents possess of high polarity, thus the D_k of the system obviously increases. Using the eq. (6), the calculated values are relatively close to the experimental data. In fact, the geometric rule of mixtures model (also called geometric mean model) typically fits the experimental data better than the parallel and series models discussed.
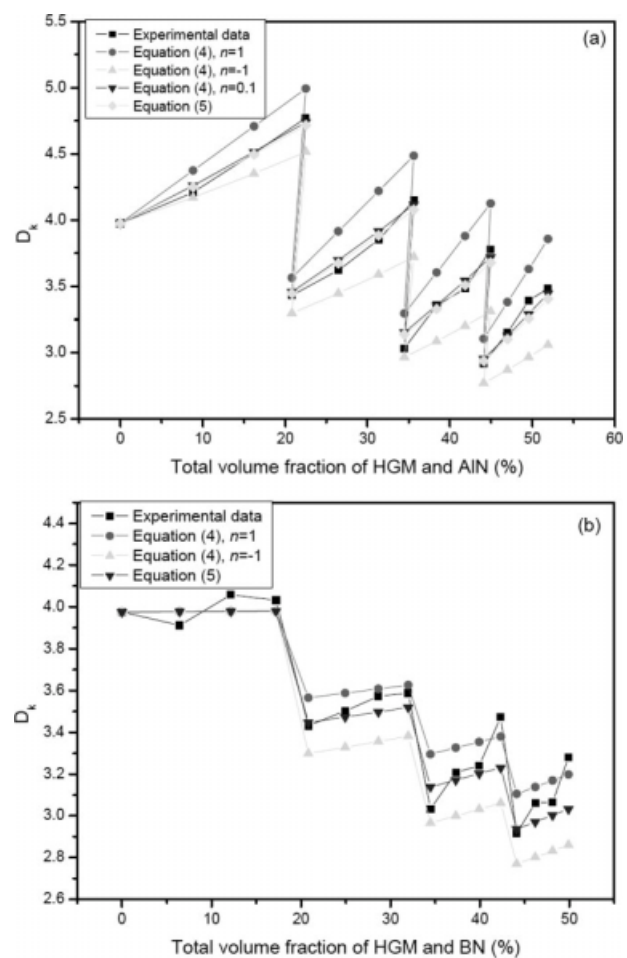

Figure 8 Comparing the experimental values of D_k with calculated values from different equations for HGM and AlN fillers (a) and for HGM- and BN- (b) filled composites.
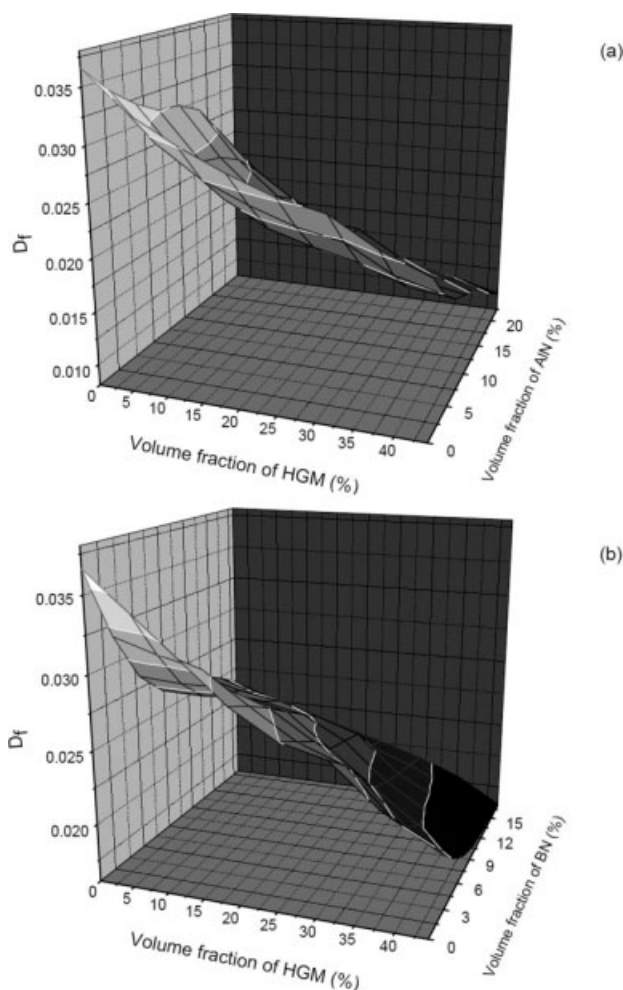
Figure 9 3D representation of D_f of the composites as a function of the volume fraction of HGM and AlN fillers (a) and as a function of the volume fraction of HGM and BN fillers (b).

Figure 9 shows the D_f of the composite as a function of filler content. It is found that the D_f decreases with increasing filler content regardless of filler type. This is because the D_f of HGM, AlN, or BN is smaller than that of epoxy matrix. When single filler filled into composite, the BN show a more efficiency than AlN for decreasing D_f. However, the D_f of HGM- and BN-filled composites is larger than that of HGM- and AlN-filled composites, indicating that the D_f is also affected by the high viscosity caused by BN. High viscosity causes more defects exist in the composite, thus D_f of the composites increases.

Finally, we can determine the optimal filler content in this study according to earlier discussion. As for CTE and D_f, the optimal properties, namely, minimum CTE and D_f are obtained at maximal content of HGM and AlN or BN filler, as mentioned earlier. However, the processability of the composite became difficult at maximal filler content, and the T_g

decrease at the same time. Thus, total filler content should be taken as intermediate value. To obtain relatively high-thermal conductivity and relatively low D_k at the same time, volume ratio of hybrid filler should be also taken as an intermediate value. According to Figure 1, it is easy to find the optimal filler content, namely, 30.6 vol % HGM and 11.3 vol % AlN or 31.6 vol % HGM and 8.3 vol % BN (marked by arrow in Fig. 1).

CONCLUSIONS

In this study, various inorganic fillers including AlN, BN, and HGM with different shape and size were used alone or in combination to prepare epoxy composites for PCBs. The results show that synthesized composite can satisfy the requirement of new PCBs materials when AlN or BN with HGM are simultaneously filled into epoxy matrix, namely, high T_g and thermal conductivity, low CTE, D_k, and D_f. Integrating the processability and properties of the composites, optimal filler content are 30.6 vol % HGM and 11.3 vol % AlN or 31.6 vol % HGM and 8.3 vol % BN.

Compared with pure epoxy, T_g increases about 35°C, and CTE, D_k and D_f decrease about 50%, 12–18%, and 43–53%, respectively, and thermal conductivity increases about 85% for composites with above two filler content. According to the comparison of the properties (thermal conductivity and D_k) by theoretical models reported in the literature with the measured properties, it is believed that selecting suitable models will effectively predict the properties of composites in future studies.

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