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Thermal Behavior and Dielectric Property Analysis of Boron Nitride-Filled Bismaleimide-triazine Resin Composites

Xiaoliang Zeng, Shuhui Yu, Rong Sun
Shenzhen Institutes of Advanced Technology, Chinese Academy of Sciences , Shenzhen 518055, China
Correspondence to: S. Yu (E-mail: yuushu@gmail.com)

ABSTRACT: Bismaleimide-triazine (BT) resin/hexagonal boron nitride (h-BN) composites are prepared, and the effects of h-BN content on the thermal and dielectric properties are studied in the view of structure–property relationship. It is found that the introduction of the BN in the BT resin dramatically improve the thermal conductivity of BT resin. The thermal conductivity of the composites is up to 1.11 W/m.K, with an h-BN concentration of 50 wt %, which is increased by six times compared with the pure BT resin. The BT resin/h-BN composites also exhibit excellent thermal properties, with the glass transition temperatures above 200°C, and thermal decomposition temperatures over 390°C. Moreover, the composites possess good dielectric properties. Their dielectric constant and loss tangent (tan δ) are less than 4.5 and 0.015, respectively. The results indicate that the BT resin/h-BN composites are promising as efficient heat-releasing materials in the high-density electronic packaging technology. © 2012 Wiley Periodicals, Inc. J. Appl. Polym. Sci. 000: 000–000, 2012

KEYWORDS: composites; dielectric properties; thermal properties; thermosets

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INTRODUCTION

Bismaleimide-triazine resins (BT resins) are widely used in the circuit boards, electric motor, and semiconductors encapsulates due to its excellent thermal stability, low water absorption, and good retention of mechanical properties at elevated temperatures, especially in hot/wet environment. Owing to the miniaturization trend of electronic circuits in the electronics industry and the drive toward higher element density, the dissipation of heat generated in electronic components has attracted much attention. However, the BT resin materials in the electronic components are inherently poor thermal conductors.[1] They must be modified to assist in heat removal from electronic parts. The addition of thermally conductive fillers in an insulation polymer matrix can effectively increase the thermal conductivity of the polymer. A number of inorganic fillers, such as carbon nanotubes,[2] silicon carbide (SiC),[3] silicon nitride (Si_3N_4),[4–6] aluminum nitride (AlN),[7–9] boron nitride (BN),[10–13] and alumina (Al_2O_3)[14,15] have been used as thermal conductive materials embedded in a polymer matrix.

Epoxy resins are the most used polymers, while unfortunately their thermal resistance and thermal conductivity cannot completely satisfy the needs of high-performance products any more. There are only few articles reporting on the high heat resistance of the compounds with high thermal conductivity. Li and Hsu[16] enhanced thermal conductivity of polyimide (PI) films via a hybrid of microsized and nanosized BN. The thermal conductivity of the PI/BN composite was up to 1.2 W/m.K. The glass transition temperature (T_g) and thermal decomposition temperatures were above 360 and 536°C, respectively, showing excellent thermal properties. Ling et al.[17,18] prepared new composite based on cyanate (CE) resin and AlN, with high thermal conductivity and thermal stability. The thermal conductivity of CE/AlN composite was up to 2.0 W/m.K, and the T_g values were above 200°C. Ishida and Rimdusit[19] studied BN-filled polybenzoxazine composites, showing that a very high thermal conductivity value of 32.5 W/m.K was achieved with BN loading of 78.5% by volume. The composites compound also exhibit high with a high T_g value of 220°C. However, so high inorganic fillers (78.5 vol %) reduced the mechanical property consequentially, limiting its application.

Although much effort has been directed toward preparing high-thermal conductive materials using different polymers, no one has reported the BT resin/h-BN composites to enhance thermal conductivity of the BT resin, while maintaining good thermal stability of the composites. In this work, we report the preparation and properties of BT resin composite with BN as the thermal conductive fillers. The effects of the filler content on thermal conductivity, thermal stability, and dielectric properties were discussed in the view of structure–property relationship.

Furthermore, the main factors influencing the thermal behavior of the composites, including the BN defects, the interfacial thermal resistance, and the phonon were discussed. To better understand the reasons limiting enhancement of the thermal conductivity, Lewis-Nielsen and Agari models were used to simulate the thermal conductivity of the composites and comparison was made with the experimental data.

EXPERIMENTAL

Materials

4,4'-bismaleimidodiphenylmethane (BMI, Honghu Bismaleimide Resin Factory, China, purity 95%), 2,2'-bis (4-cyanatophenyl) propane (BCE, Heijang Kinlyuan Pharmaceutical, China, purity 90%), and 2,2'diallyl bisphenol A (DBA, Wuxi Resin Factory, China,purity 90%) were used as starting materials to prepare the modified BT resin. Hexagonal BN (h-BN, Sanhe Yanjiao Xinyu Hi.-Tech Ceramic Material, China) was used as ceramic particles, which have a density of 2.27 g/cm^3, a thermal conductivity of 250 W/m.K, and the average size of 1.0 μm. γ-Glycidoxypropy1 trimethoxysilane (KH-560, Sinopharm Chemical Reagent, China.) was used to modify BN particles. Other materials and reagents were also purchased from Sinopharm Chemical Reagent, China.

Surface Modification of BN by KH-560

First, 5 g of BN particles were suspended in 50 mL of 95% ethanol solution to which 0.5 g of KH-560 were slowly added. The mixture was stirred for 12 h at 70°C. The modified BN was isolated through centrifugation and washed three times with the ethanol. Finally, the modified BN was dried at 50°C for 24 h to yield dried powder.

Modification of BT Resin by DBA

The modified BT was prepared by a melt method, as reported previously.[20] Typically, 5 mol BMI, 3 mol BCE, and 2 mol DBA were mixed by stirring at 120–150°C for 30 min to form transparent, amber-colored liquid and cooled to room temperature.

Preparation of BT/h-BN Composites

In the BT/h-BN composites, the h-BN fillers are used in the range of 10–50 wt %. First, the modified BT resins were dissolved in an organic solvent (methyl ethyl ketone). Then, the surface-modified h-BN particles were dispersed into the BT solution under stirring condition and ultrasonicated for 1 h. The obtained BT/h-BN suspension was then poured into molds and dried at 80°C for 30 min to evaporate the solvent. Finally, the dried BT/h-BN composites were cured by the programmed heating process: 180°C/2 h + 200°C/2 h + 220°C/2 h.

Characterization

The chemical status of the h-BN surface after modification was investigated with Fourier transform infrared spectroscopy (FTIR) (Thermo Nicolet308 System). The fractured morphology of the composite samples was examined using scanning electron microscope (SEM) (4800 S, Hitachi).

The thermal conductivity was measured with LW-9389 TIM Resistance and Conductivity Measurement (Long Win Science & Technology, Taiwan), which was fabricated according to the ASTM D-5470-06 standard. Then, the thermal conductivity was calculated from thermal diffusivity according to eq. (1):

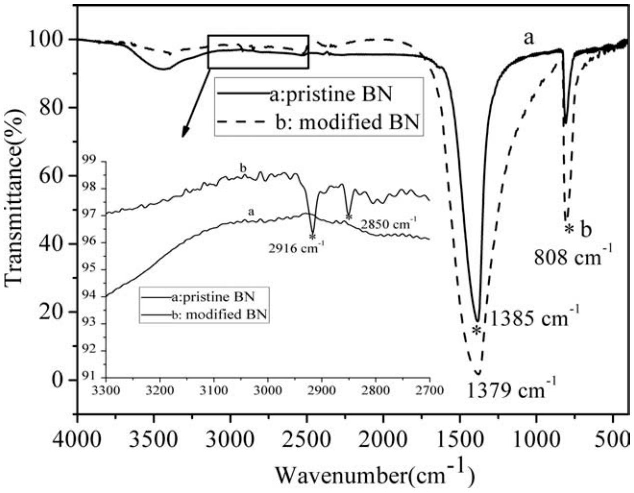

Figure 1. FTIR spectra of pristine h-BN and modified h-BN.

$$\lambda = -\frac{Q.L}{A.\Delta T} \tag{1}$$

where λ is the apparent thermal conductivity, W/m·K, Q is heat flux, W or J/s, L is the thickness of specimen, m, A is the area of a specimen, m^2, and ΔT is the temperature difference between temperature sensors of the hot meter bar, K.

The glass transition temperature (T_g) of the BT/h-BN composites and pure BT resin were measured with the differential scanning calorimetry (DSC) at a heating rate of 10°C/min in nitrogen (TA Q20). The T_g is read at the initial change in the slope of the heat capacity of the DSC thermogram. The thermogravimetric analysis (TGA) was performed using TA Q600 Thermogravimetric analyzer at a heating rate of 10°C/min in nitrogen.

The dielectric properties including effective dielectric constant (ε_{eff}) and dielectric loss (tanδ) were measured using an impedance analyzer (Agilent 4294 A) in the frequency range of 1.0 kHz to 10 MHz, in which much noises from environment or testing circuits could be avoided. At least three points were measured to ensure repeatability.

RESULTS AND DISCUSSION

Microstructure of BT/h-BN Composites

The dispersion of the h-BN particles into the polymer matrix is an important issue for the composite preparation. Blending h-BN particles without modification with BT resin will result in the aggregation of h-BN particles due to the different polarities between the organic and inorganic phases. One of the best ways to uniformly disperse the inorganic phase into a polymer matrix is to functionalize the surface of the inorganic phase with organic surfactants. In this study, γ-Glycidoxypropy1 trimethoxysilane (KH-560) was used as modifier to functionalize the h-BN surface. Figure 1 presents the FTIR spectra of unmodified and modified h-BN with KH-560. The absorption band of the unmodified h-BN at 3600–3200 cm^{-1} is attributed to the valence vibration of the —OH group on the BN, which could be the reactive site interacting with the coupling agent. The broad

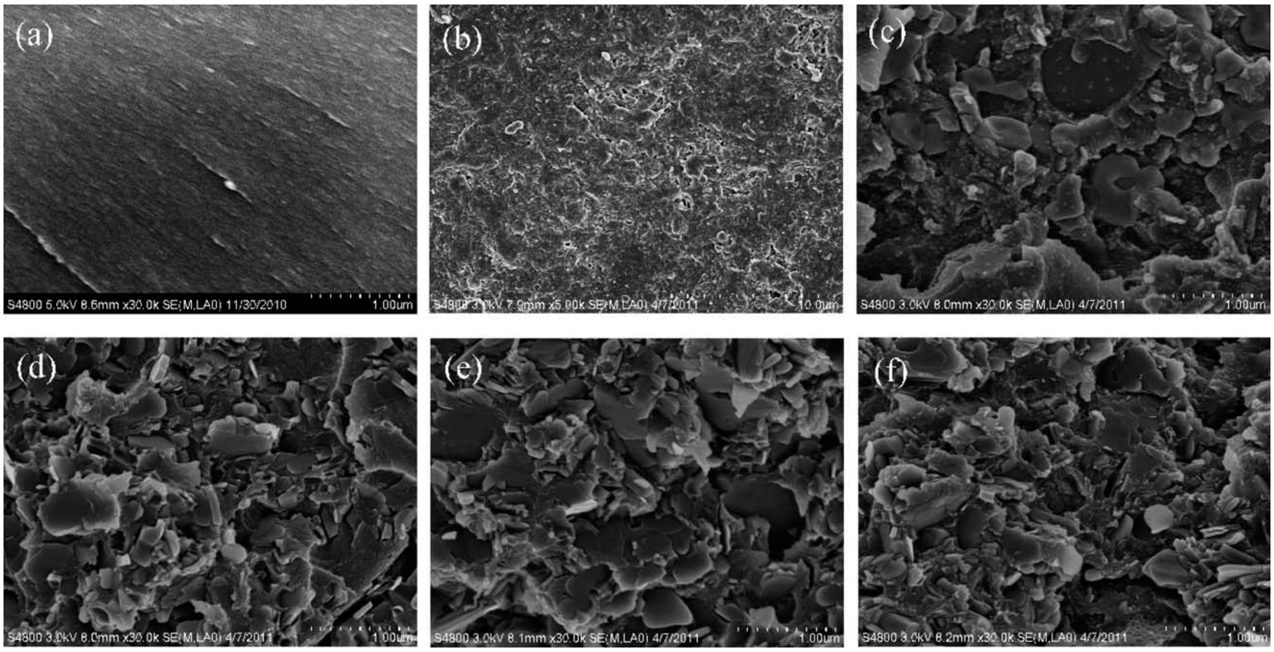

Figure 2. SEM images of the fractured morphology of BT/h-BN composites: (a)0 wt % h-BN; (b) 10 wt % h-BN; (c) 20 wt % h-BN; (d) 30 wt % h-BN; (e) 40 wt % h-BN; (f) 50 wt % h-BN.

peak around 1385 cm^{-1} could indicate in-plane stretching vibration of hexagonal BN, and the peak at 808 cm^{-1} represents the out-of-plane bending vibration. However, compared with the spectrum of the unmodified h-BN, the absorption band of the modified h-BN at 3600–3200 cm^{-1} decreases, due to the reaction between the —OH group and the coupling agent. Meanwhile, the spectrum of the modified h-BN displays a new absorption band at the wave number of 2916 and 2850 cm^{-1}, corresponding to the characteristic peaks of the $—CH_2$- unit from KH-560. A frequency shift of the B—N bond vibration from 1385 cm^{-1} (pristine BN)[16] to 1379 cm^{-1} (modified BN) and a weakening of the shoulder of B—N bond vibration at 1385 and 808 cm^{-1} are important information to support the occurrence of modification by KH-560. Based on the above results, we conclude that KH-560 successfully reacted on the BN particles surface.

The microstructure of the BT/h-BN composites is shown in Figure 2. The platelet-shaped h-BN filler with 1 μm in size is embedded in the BT resin and no pores can be seen in the composites, confirming interface adhesion with the h-BN in BT resin matrix. At low h-BN content, the h-BN particles are separated from each other and dispersed uniformly in the matrix. With increasing the particles content, the h-BN particles contact with each other and a continuous network is formed, indicating formation of a thermally conductive network.

Thermal Conductivity of the Composites

The dependence of the thermal conductivity of BT/h-BN composites on the filler content is shown in Figure 3. It can be seen that the thermal conductivity of BT/h-BN composites increases with increasing the h-BN content. The BT/h-BN composites exhibit the highest thermal conductivity of 1.11 W/m.K with 50 wt % of h-BN, which is about six times of that of the pure BT resin. It is worth noting that, there are many researches on using h-BN particles as the thermal fillers to improve the thermal conductivity of the polymer, however only low thermal conductivity (<2.0 W/m.K) can be obtained. Li and Hsu.[16] developed a new thermally conductive PI composite film with both microsized and nanosized BN as fillers. The thermal conductivity of the PI/BN composite was up to 1.2 W/m.k, for a mixture containing 30 wt % of microsized and nanosized BN fillers in the PI matrix. In the case of epoxy resin embedded with functionalized BN, Teng et al.[21] observed the improvement from 0.18 to 1.12 W/m.K, with BN powder fractions increased from 0 to 30 vol %, which agrees with our results. For high-density polyethylene, Zhou et al.[22] observed the improvement

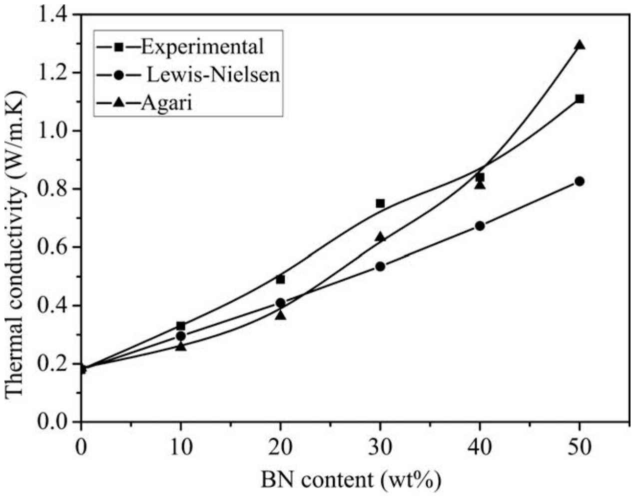

Figure 3. The experimental and theoretical thermal conductivity of BT/h-BN composites according to the h-BN content.

from 0.26 to 1.12 W/m.K, when 0 and 35 vol % BN were added, which also agrees with our results. From the above results, we can see that although the theoretical thermal conductivity of h-BN is very high (250 W/m.K), the experimental thermal conductivity of composites using h-BN as thermally conductive fillers is low (<2.0 W/m.K). Several factors probably are responsible for affecting the ability of h-BN fillers to enhance the thermal conductivity of BT resins, leading to the relatively low thermal conductivities of polymer/BN composites compared with expectations from the inherent thermal conductivity of h-BN.

First, the quality of h-BN intensely affects its inherent thermal conductivity and then impairs the thermal conductivity of composites. The very high thermal conductivity of pristine h-BN will lead to good properties of the composites embedded with h-BN. However, the possible defects within individual h-BN particle, including vacancy defects, topological defects, lattice defects, impurities, or exposed edges, could lead to reduced quality of h-BN, and weaken the effective thermal conductivity of h-BN.

Second, the interfacial thermal resistance between BT resin and h-BN filler impacts the effective thermal conductivity of the BT/h-BN composites. It is well-known that the interfacial thermal resistance, known as the Kapitza resistantce after Kapitza's discovery of temperature discontinuity at the metal-liquid interface,[23] hinders the transfer of phonon caused by the phonon mismatch at the interface of the h-BN and the BT resin, thus leading to dramatically reduced effective thermal conductivity. Although the h-BN surface was functionalized by KH-560 to improve the compatibility between BT resin and h-BN, the thermal resistance is still existent.

Third, the phonon, which is the main mode to transfer heat, can be damped and scattered by the BT resin and thus reducing the inherent thermal conductivity of h-BN. Gojny et al.[24] obtained a higher thermal conductivity in the multiwalled carbon nanotubes (MWCNTs) filled epoxy than that filled with single-walled carbon nanotubes. This phenomenon was attributed to the fact that the MWCNTs have inner shells which conduct phonons efficiently despite the outer shell interacting with BT resin. In the case of our study, the BT resin can also impair the phonons of h-BN surfaces. Therefore, because of the above reasons, the thermal conductivity (1.11 W/m.K) of the BT/h-BN composites is still lower than 2 W/m.K even with high h-BN content. However, it means that there is still room for thermal conductivity enhancement if we reduce the interfacial thermal resistance.

The modeling of thermal conductivity of the composite is important for the analysis of filler effect on the thermal conducting behavior. For illustration and comparison, two approximation models are used in this study to further analyze the thermal conductivity of the composite. The two models are as followed:

(a) Lewis-Nielsen model[25]

$$\lambda = \lambda_p \left[\frac{1 + A.B.V_f}{1 - B\psi V_f} \right] \qquad (2)$$

where

$$B = \frac{\frac{\lambda_f}{\lambda_p} - 1}{\frac{\lambda_f}{\lambda_p} + A} \quad \psi = 1 + \left(\frac{1 - \varphi_m}{\varphi_m^2} \right).V_f$$

where A is a function of the geometry of the filler particles, and Φ_m is the maximum packing fraction, which is 0.52 for three-dimensional random packing.

(b) Agari model[26]

Agari developed a model based on the generalization of models of series and parallel conduction in composites.

$$\log \lambda = V_f C_2 \log(\lambda_f) + (1 - V_f) \log(C_1 \lambda_p) \qquad (3)$$

where λ, λ_f, λ_p, and V_f are defined as same as in eq. (2), C_1 is a factor relating to the effect of the filler on the secondary structure of the polymer, and C_2 is a factor relating to the ease in forming conductive chains of the filler. In eq. (3), logarithms of the thermal conductivity of the composite increases linearly with the volume fraction of the filler, constants C_2 and C_1 are experimentally determined.

Figure 3 shows the experimental and theoretical thermal conductivity of BT/h-BN composites. As shown in the figure, the experimental data do not agree with Lewis-Nielsen model. Although Lewis-Nielsen model considers the particle size, geometry, and dispersion, it underestimates the thermal conductivity of the BT/h-BN composites. The reason is that the Lewis-Nielsen model ignores the effect of filler orientation on thermal conductivity. Hill and Supancic[27] have proposed that the deformation of particles takes place during the formation of h-BN-filled composites, which leads to the unexpected improvement in thermal conductivity through the formation of chains of particles, a factor not accounted for by the Lewis-Nielsen model. In the case of Agari model, as shown in Figure 3, the experimental data also cannot be exactly accordant with Agari model, with a discrepancy in the percentage of about 20%. The values of C_1 and C_2 are 0.97 and 0.65, respectively, calculated by eq. 3. The C_1 and C_2 values here are in good accordance with those proposed by Agari which are speculated to be around 1.0 and 0.75, respectively. Although there is a disparity between prediction by Agari and experimental data, the Agari model still gives a relatively better prediction than the Lewis-Nielsen model, in the whole experimental weight fraction range. In our future research, a new theoretical model for the prediction of the thermal conductivity of BT/h-BN composites will be proposed, accounting for the variations in the size, defects of h-BN, the interfacial thermal resistance, and filler orientation.

Thermal Properties of the Composites

Figure 4 shows the DSC thermograms of BT/h-BN composites with various h-BN contents. The T_g value of the BT/h-BN composites are presented in Table I. It is apparently shown that the T_g shifts to lower temperature with the addition of h-BN. The T_g value for the composite containing 50 wt % h-BN is 206.58°C, which is decreased by 36.43°C compared with the BT resin. On the one hand, as a large amount of h-BN is

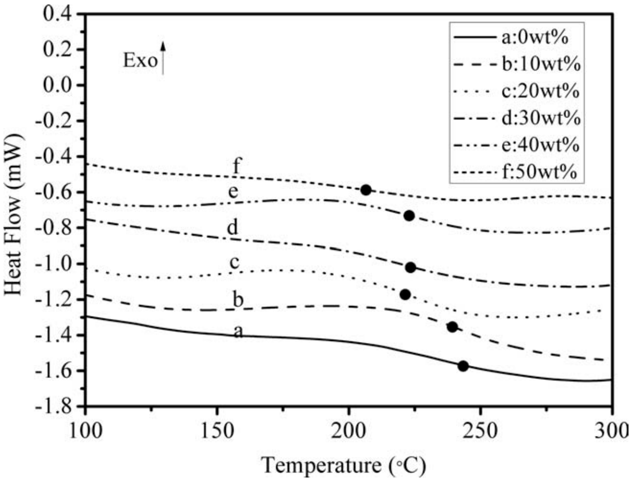

Figure 4. DSC thermograms of BT resin and BT/h-BN composites.

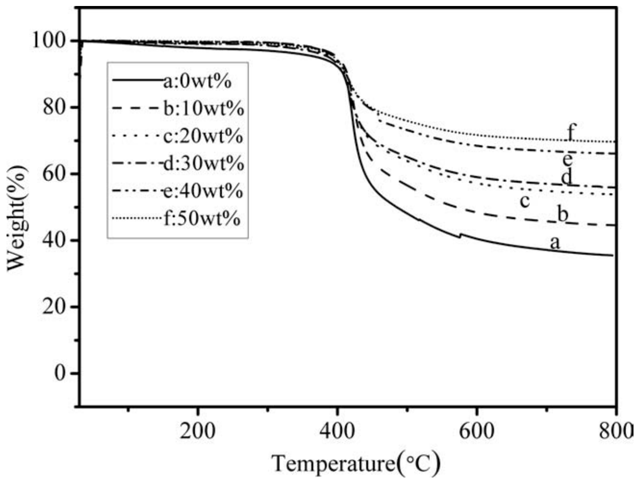

Figure 5. TGA thermograms of BT resin and BT/h-BN composites.

incorporated into the BT resin, the crosslinkage density of BT resin would decrease, leading to decrease of T_g. On the other hand, addition of inorganic fillers will occupy a certain part of volume for the movement of polymer chains and the modified fillers may also interact with the polymer chains leading to a possible increase of T_g. However, in the case of this study, because the negative effects of the decreased crosslinkage density counteract the positive effects of the restriction of segmental motions, the $T_g$values of the composite decrease with increasing the loading of h-BN fillers. The results are in agreement with the previous report on CE ester/AlN composites.[18] Although T_g of the composite decreases with the h-BN addition amount, the T_g values (above 200°C) are still higher than that of the conventional epoxy resins and can satisfy the utilizing requirements of prolonged service at high temperatures.

Figure 5 shows the TGA thermograms of BT/h-BN composite with various amounts of h-BN. The TGA data, temperatures at which the total mass loss of 5 and 10% occur and the char yield at 800°C for BT/h-BN composites are shown in Table I. As shown in the table, with the increase of the filler content, the $T_{d5\%}$ and $T_{d10\%}$ of the composite shift to higher temperatures, and the residues at 800°C increase with increasing h-BN content in spite of the decreased crosslinkage density of BT resin. The $T_{d5\%}$ and $T_{d10\%}$ of the composite with 50 wt % h-BN are 395.61 and 412.06°C, higher by 27.54 and 10.25°C than that of the pure BT resin, respectively. The phenomenon should be caused by the decrease in permeability due to "tortuous path" effect of the filler. The existence of the fillers leads to a dense layer when the composites are degraded. This dense layer delays

Table I. The Thermal Properties of BT/h-BN Composites

Sample	T_g(°C)	$T_{5\%}$(°C)	$T_{10\%}$(°C)	Residues at 800°C (wt %)
BT resin	243.01	372.54	408.53	35.5
10 wt %	241.30	399.88	414.30	44.57
20 wt %	222.89	392.59	411.03	53.83
30 wt %	223.05	387.74	408.48	55.93
40 wt %	221.03	397.81	413.03	66.12
50 wt %	206.58	395.61	412.06	69.65

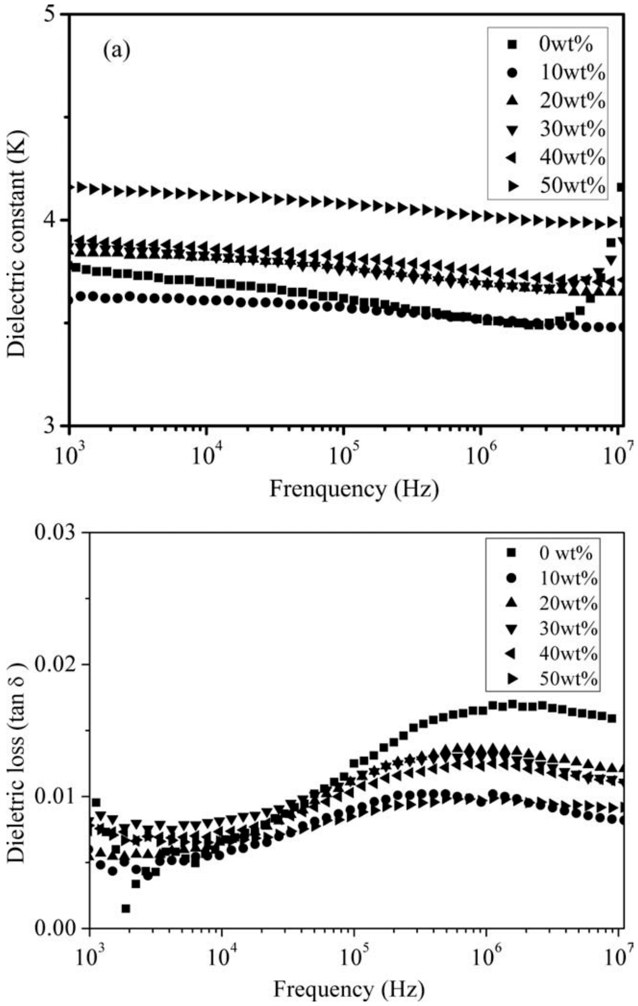

Figure 6. Effective dielectric constant (a) and loss tan δ (b) of BT/h-BN dielectric composites at various frequencies and h-BN content.

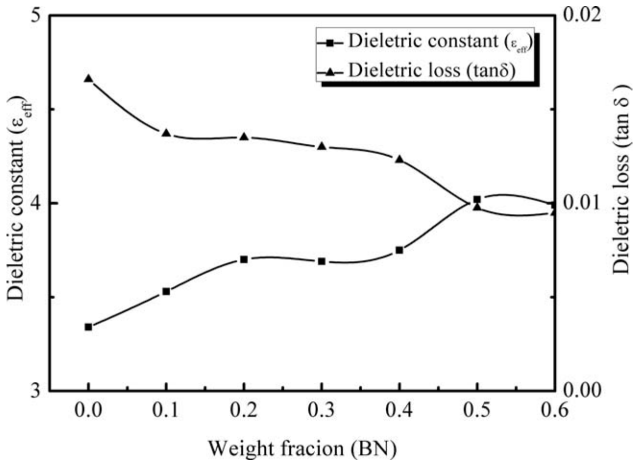

Figure 7. Effective dielectric constant ε_{eff} and dielectric loss tan δ of BT/h-BN composites according to h-BN weight fraction measured at 1.0 MHz.

the escape of volatile degradation products. As a consequence, retarded effect of h-BN on the degradation can compensate for the lower stability of BT resin in the composite caused by the decreased crosslinkage density, leading to the enhanced thermal stability of BT/h-BN composites with h-BN content. The same phenomena have also been observed in other composite,[28] where the T_g decrease was observed in all the clay-modified epoxy systems, but the $T_{d5\%}$, $T_{d10\%}$, and the residues at 800°C for the composites increased.

Dielectric Properties of the Composites

Figure 6 presents the frequency-dependence of effective dielectric constant (ε_{eff}) and loss tan δ of BT/h-BN composites with different weight fractions of h-BN at 25°C. As shown in Figure 6(a), the composites demonstrate characteristically low dielectric constant due to a low polarity of the central triazine ring of BT resin and low dielectric constant of h-BN. Moreover, the h-BN addition results in a minimal increase in dielectric constant, with ε_{eff} increased from 3.34 for the pure BT resin to 3.99 for the 50 wt % of BN fillers, as shown in Figure 7. The increase of ε_{eff} could be interpreted in several aspects. First, the dielectric constant (4.2) of h-BN is higher than pure BT resin. Second, the addition of fillers decrease crosslinkage density of BT resin, leading to decreased number of triazine rings. In this regard, the whole polarity of BT composites was increased due to less number of triazine rings. Third, as the fillers used in this article are modified, the polar groups on the fillers may further increase the whole polarity of the resulted BT composites. The higher dielectric constant of BN and the increased polarity of BT/h-BN composites result in increased dielectric constant of BT/h-BN composites. However, the dielectric constants of the composites are still less than 4.0.

The tan δ of the BT/h-BN composites increase with frequency in the range of 1.0 kHz to 10 MHz, as revealed in Figure 6(b). Figure 7 shows the effect of loading of the BN fillers on the tan δ of the BT/h-BN composites. As shown in the figure, the tan δ decrease from 0.0166 for the pure BT resin to 0.00949 for the 50 wt % of BN fillers. These results are in agreement with the previous study on the h-BN/polymer composites.[29] This is because, the positive effects of low dielectric loss of BN can counteract the negative effects of increased polarity of BT/h-BN composites, leading to decreased dielectric loss with increasing the loading of BN fillers. Such a composite with low dielectric constant and loss is desired for the application in semiconductor devices package.

CONCLUSIONS

BT resin/h-BN composites with h-BN content ranging from 0 to 50 wt % were prepared and characterized. According to the SEM analysis, with increasing the particles content, the h-BN contacts with each other and a continuous network is formed, indicating the formation of thermal conductive network. As expected, the thermal conductivity increases with the increase of h-BN content. With an h-BN concentration of 50 wt %, the composite exhibits a thermal conductivity of 1.1 W/m.K, which increases by six times of the pure BT resin. However, there exists the possibility to enhance the thermal conductivity if we improve the quality of h-BN and to reduce thermal interfacial resistance. The DSC thermal analysis has determined that the T_g decreases with the addition of h-BN into BT resin matrix. It decreases to 206.58°C when the composite contains 50 wt % h-BN, which is decreased by 36.43°C compared with the BT resin. However, the thermal decomposition temperatures of the composites shift toward the higher temperature as the h-BN content increases, which is attributed to the decrease of permeability due to “tortuous path” effect of the filler. As the content of h-BN increases from 0 to 50 wt %, the dielectric constant of BT resin/h-BN composites slightly increases, while the loss tangent (tan δ) decreases. Two theoretical models for the prediction of BT resin/h-BN composites thermal conductivity are available. The Agari model gives a relatively better prediction than the Lewis-Nielsen model, with a discrepancy in the percentage of about 20%. Based on the results obtained, the BT resin/h-BN composites are potential for use in high-temperature microfabrication of heat dissipative components in microelectronic industry.

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