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Improvement of thermal conductivity and mechanical properties for polybenzoxazine composites via incorporation of epoxy resin and segregated structure

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Abstract

Developing tendency of increasing integration in electronic components filed led to the demand for thermally conductive polymer materials. Forming a segregated structure with thermally conductive fillers in the matrix has been proven to be a promising technique. However, the mechanical properties of the material prepared via this technique are commonly sacrificed because the interaction between fillers and matrix are weak. We present a simple method to improve the mechanical properties while maintaining the segregated structure via the collaboration of epoxy and polybenzoxazine (PBz). The fillers, epoxy resin, and benzoxazine (BOZ) were mixed and then hot pressed under 12 MPa of pressure with a certain heating strategy. The thermal conductivity, density, mechanical properties, morphology, and thermal stability of the composites were investigated. With 25 wt% filler, the value of thermal conductivity coefficient of EP/PBz composites reached 0.811 W mK⁻¹, while that of PBz composites was 0.762 W mK⁻¹. The tensile strength of the matrix improved from 58.3 MPa to 99.7 MPa.

1. Introduction

Considering the rapid development in 5G industries, billions of microchips will be needed shortly. However, the increasing density of electronic components and rising operating power leads to the accumulation of a huge amount of heat, which will undermine the stability and performance of the devices [1]. Therefore, it is critical to developing polymeric packaging materials with splendid heat dissipation ability to remove excessive heat. However, most polymer materials are reckoned as thermal insulators due to their ultralow thermally conductive coefficient value. Thus, highly thermally conductive polymer composites have been widely concerned in recent decades [2]. Conventionally, inorganic particles with high thermal conductivity (TC) are introduced into a polymer matrix, including boron nitride (BN), alumina (Al₂O₃), carbon nanotubes (CNTs) [3–6]. However, to achieve higher thermal conductivity coefficient (λ), excessive addition of fillers (>30 vol%) is needed, which will provide challenges to mechanical, processing properties of polymer composites. Therefore, to achieve effective usage of thermally conductive fillers, Turner initially developed the concept of forming a segregated conductive network for nickel/high-density polyethylene composites in 1971 [7, 8]. In a segregated structure, the thermally conductive fillers are primarily located at the interfaces between the polymeric matrix particles instead of being randomly dispersed throughout the entire composite system [9]. A facile way to construct the segregated structure is dry-mixing of powder materials followed by hot-compression molding, reported by Agari and co-workers [10]. For example, Bo Li fabricated aluminum oxide/polypolyethylene composite with two different processing techniques: mechanical grinding (MG) and melt mixing (MM). The segregated structure can be observed clearly with scanning electron microscopy in the composite system prepared through MG followed by
hot pressing. The TC of samples prepared via MG was greater than those prepared via MM once the segregate structure was formed. However, the strength of MG samples decreased rapidly at the same time because the network structure impairs interaction between the polymer particles. Gu fabricated highly thermally conductive polyphenylene sulfide (PPS) via mechanical ball-milling followed by hot-compression method with micrometer boron nitride/nanometer boron nitride (mBN/nBN). The λ value increased from 0.286 W mK\(^{-1}\) for pristine PPS matrix to 2.638 W mK\(^{-1}\) for PPS composites with 60 wt% filler content. At the same filler loading, the strength of the composite was less than 40 MPa while the pristine PPS matrix was over 88 MPa. Therefore, it can be concluded that the construction of segregated structure would be favorable to λ values of the polymer composites while impairing the mechanical properties.

In this research, we proposed a facile way to enhance the adhesion between network structure and polymer matrix while the segregated structure can still be maintained. Polybenzoxazine (PBz) and epoxy were employed as matrix, and mechanical mixing followed by hot compression was used to prepare the composites. Polybenzoxazine is a novel class of phenolic resin that possesses excellent mechanical properties, tiny volumetric shrinkage upon polymerization, low water absorption, and high char yield. Therefore, PBz resins are promising materials for electronic packaging and circuit boards. Epoxy resins have many advantages, including good mechanical properties, high impact resistance, and good adhesion properties, which are one of the most used materials in the electronic packaging field. The copolymerization between epoxy and PBz can greatly decrease the crosslink density of the matrix and magnificently enhance its mechanical properties. Furthermore, the ring-opening reactions of benzoxazines (BOZ) when being heated, produce phenolic hydroxyl groups, which can act as a curing agent for epoxy resins and provide cross-linking points into the matrix. As the benzoxazine monomers are solid at room temperature, the epoxy and fillers will distribute on the surface of the benzoxazine particles so that the segregated structure will be formed. After being heated, the BOZ particle will start curing itself as well as the epoxy resin. With the formation of strong chemical bonds, the mechanical properties of the segregated system can be improved.

2. Experimental

2.1. Materials

Benzoxazine resin (BOZ), 1.20 g cm\(^{-3}\), was purchased from ACO Pharm Co., Ltd, (AIBZ682). Epoxy resin (EP), 1.19 g cm\(^{-3}\), was purchased from WuXiShi Changgan chemical Industry Co. LTD, (E-51). Hexagonal Boron Nitride (99.9%) (h-BN), 2.25 g cm\(^{-3}\), with a diameter of 50 \(\mu\)m was supplied by Zibo Jonye Ceramic Technologies Co., Ltd.

2.2. Specimen preparation

The BOZ and E51 were firstly mechanically mixed at the mass ratio of 6:4 (which was label as ZE64). Then BN was added into the mixture and mechanically stirred for 30 min After that, the compound was moved into a certain mold and hot pressed under 12 MPa. The heating strategy is as follows: 160 °C/1 h + 180 °C/1 h + 200 °C/1 h + 220 °C/1 h. The whole process is shown in figure 1. For comparison, PBz composites were prepared at the same filler loading with the same processing. The corresponding formula was shown in table 1.

2.3. Characterizations

Fourier transform infrared (FT-IR) spectra were performed on IR spectrometer (IRAffinity-1, Japan) from 400 cm\(^{-1}\) to 4000 cm\(^{-1}\).
Density test was conducted on electronic densimeter (FK-300DT, Xiamen Fubusi, China) with block samples. Each sample was tested for three times and the results were averaged.

The tensile tests were carried out by means of a mechanical tester (MTS E43). Test specimens were cast into stainless steel molds with dog-bone geometry according to GB/T2567-2008. Each sample was tested for three times and the results were averaged.

The morphology of the fabricated composites were characterized by scanning electron microscope (SEM, S-3400, Hitachi Ltd, Japan). Specimens were fractured in liquid nitrogen and coated with a thin layer of gold before observation.

Thermogravimetric analysis (TGA) of the samples were carried out by WRT-2P (Shanghai Shangping Instrument Co., Ltd, China) at 10 °C min⁻¹ (nitrogen atmosphere), over the range of temperature (50 °C–750 °C).

Thermally conductive coefficient (λ) of the samples were tested via hot wire method with thermal conductivity instrument TC3000E (Xi’an Xiaxi Electronic Technology Co., Ltd, China) at 25 °C. Hot wire method is mainly applicable to measure the λ value of solid polymers samples as a function of temperature [21]. The scale of the specimen was 60 mm × 40 mm × 2 mm. Each sample was polished and then measured for five times. A filmy thermos-resistor wire is set between two samples, on which a 500 g weight is placed to offer constant pressure. During measurement, the stable input of electric power generates heat at a constant rate. Then the heat transfers into the samples through the wire between them mainly by conduction. The basic measurement principle of hot wire method are as follows: the samples can be reckoned as a semi-infinite system and the λ value of the samples can be obtained through the following relationship [22]:

\[
\lambda = \frac{Q}{4\pi} \frac{dT}{d \ln t}
\]

in which Q is the heat generation per unit length of the hot wire, T is the temperature of the samples and t is the corresponding time.

3. Results and discussion

3.1. Curing behavior of epoxy and polybenzoxazine

Figure 2 presents the FTIR spectra of E51, BOZ, and cured ZE64. BOZ resin reacts with epoxy resin through a nucleophilic addition reaction of the phenolic hydroxyl group with the epoxy group, because the phenolic groups of polybenzoxazine acts as an initiator and catalyst for epoxy polymerization [23]. From the FTIR spectra, it can be seen that the peak at 1035 cm⁻¹ for E51 disappeared after cured, which indicated the ringing opening of the oxirane ring. In the meanwhile, the peak at 1225 cm⁻¹ for BOZ (Ph-O-C) shifted to 1239 cm⁻¹ after cured indicated the ring-opening of benzoxazine structure. It was worth noting that the strong peak appeared at 3128 cm⁻¹ and 1400 cm⁻¹ for ZE64 indicated the existence of hydrogen bonding, which can only be formed in polybenzoxazine resin. Therefore, it can be inferred that in the curing process, epoxy and benzoxazine polymerization took place concurrently [24, 25]. The external part of BOZ granules recated with the the epoxy resin while the self-polymerization occurred at the core of BOZ granules because they cannot be exposed to the epoxy resin. The polymerization of BOZ proceed through the ring opening reaction of oxazine ring at high temperature. Then the formed cation led to Friedel-Crafts reaction with aromatics which completely turned BOZ into polybenzoxazine [26]. The curing reactions were shown in figure 3.

| Sample | ZE64 wt% | PBz wt% | BN wt% |
|--------|----------|---------|--------|
| ZE64   | 100      | 0       | 0      |
| ZEB6405| 95       | 0       | 5      |
| ZEB6410| 90       | 0       | 10     |
| ZEB6415| 85       | 0       | 15     |
| ZEB6420| 80       | 0       | 20     |
| ZEB6425| 75       | 0       | 25     |
| PBz    | 0        | 100     | 0      |
| ZB05   | 0        | 95      | 5      |
| ZB10   | 0        | 90      | 10     |
| ZB15   | 0        | 85      | 15     |
| ZB20   | 0        | 80      | 20     |
| ZB25   | 0        | 75      | 25     |
Figure 2. FT-IR spectra of (a) E51; (b) BOZ; (c) cured ZE64.

Figure 3. Curing reaction of (a) ZE64; (b) PBz.
3.2. Thermal conductivity of BN/PBz and BN/ZE64 composites

The $\lambda$ values of the BN/PBz and BN/ZE64 composites and the corresponding enhancement are shown in figure 4. It can be observed that the thermal conductivities of both PBz and ZE64 composites increase gradually with the rising filler loading, which agreed with theoretical expectations. With 25 wt% filler content, the thermal conductivity of PBz composites reached 0.762 W mK$^{-1}$, while that of ZE64 composites reached 0.811 W mK$^{-1}$.

It was worth noting that though the ZE64 composites exhibited better thermal conductivity than the PBz composites at all filler levels, the gap between their thermal conductivities varied at different filler loading. It can be seen that the thermal conductivity of ZE64 composites rised evenly with the increase of filler loading while that of PBz composites exhibited slow growth and then a sharp increase when filler loading was over 20 wt%.

These different tendencies indicated that the segregated structures were not only maintained but also perfected with the introduction of epoxy resin. In the BN/PBz system, BN flakes were dispersed on the surface of BOZ granules and formed direct contact between each other. However, the interaction between thermally conductive fillers and the BOZ particles were so weak that voids and air gaps could not be avoided, which caused thermal boundary resistance at the interface between them [27]. When epoxy resin was introduced into the system, the strong shearing force would eliminate the voids to a certain degree and prevent fillers from agglomerating to achieve even distribution of fillers on the surface of the BOZ granules. Figure 5 illustrates the formation of segregated structure in BN/ZE64 composites. At first, the BN flakes were fixed on the surface of BOZ granules. After being heated, epoxy resin cured with BOZ granules, and the cores of BOZ granules cured itself because they cannot connect with epoxy as well as the BOZ was overdoes. After curing, the segregated structure was maintained, and each part of the core–shell structure was connected through chemical bonds.

![Figure 4](image1.png)

**Figure 4.** (a) Thermal conductivity of the BN/PBz composites and BN/ZE64 composites; (b) enhancement in the thermal conductivity concerning pristine matrix.

![Figure 5](image2.png)

**Figure 5.** Illustration of mechanism for ZE64 in enhancement in thermal conductivity.
3.3. Density of BN/PBz and BN/ZE64 composites

To demonstrate the changes in voids between fillers and matrix, a density test was conducted, and the results were shown in figure 6. It can be seen that the densities of the composites increased with the rising filler loading though none of them reached theoretical value, which indicates the existence of air gaps and voids. It can be observed that in BN/PBz system, the gap between theoretical and experimental values maintained at a certain level, which indicated the porosity showed little dependence on content of filler. In the BN/ZE64 system, the gap between theoretical and experimental values increased with the increase in content of filler, resulting in increased porosity. Besides, ZE64 composites possess higher densities than PBz composites at all filler levels, which indicated that the introduction of epoxy enhanced the interaction between fillers and matrix and eliminated air gaps and voids. This was because the contact between fillers and matrix were insufficient while powder mixing as a result of the irregular surface of BOZ particles. Then, during hot pressing, without further shearing force, the elimination of voids can only depend on the flow of the matrix. Therefore, some of the air gaps and voids were sealed in the composites, resulting in serious phonon scattering. However, with the introduction of epoxy, the blending transferred from powder into an emulsion, in which shearing force would be generated while blending. Therefore, the air gaps and voids can be eliminated greatly at first. With higher filler loading, the incompatibility between filler and matrix would eventually cause higher porosity.

3.4. Mechanism research on thermal conductivity

To fully demonstrate the effect of epoxy on forming the segregated structure, Foygel’s model was adopted to describe the formation of thermally conductive network by the fillers. The model is based on the random filler entanglement and heat dissipation among fillers, which can be used to estimate the $R_c$ at high filler loadings. The $\lambda$ value of composites can be described by the following function [28]:

$$\lambda - \lambda_m = K \left( \frac{V_f - V_c}{1 - V_c} \right)^\beta$$

(2)

$$R_c = \frac{1}{K d V_c^\beta}$$

(3)

in which $K$ stands for a pre-exponential factor ratio which means the expected contribution of individual filler networks, $\beta$ represents a thermal conductivity exponent that is dependent on the aspect ratio of the fillers, $V_c$ represents the critical volume fraction of fillers, and $d$ represents the diameter of filler which is 50 $\mu$m according to the supplier. The fitted curves and corresponding results were shown in figure 7 and table 2. The obtained $R_c$ value for BN/ZE64 composites was $7.488 \times 10^6$ K · W$^{-1}$, which was nearly one-tenth to that for BN/PBz composites, $8.090 \times 10^7$ K · W$^{-1}$. The magnificent decrease in $R_c$ value also indicated that the interfacial surface conditions of BN platelets were tuned by the introduction of epoxy resin. The main reason was that with the strong shearing force in processing resulted from the collaboration of epoxy resin, most gaps and air voids between BN platelets and matrix can be eliminated. Therefore, mismatch of lattice between fillers caused by the vacancy defects between BN in the formed network was moderated with the increased affinity.
3.5. Mechanical properties of BN/PBz and BN/ZE64 composites

Figure 8 shows the mechanical properties of the BN/PBz composites and BN/ZE64 composites. It can be observed that with the introduction of epoxy, the tensile strength of the matrix increased while the modulus decreased, which attributed to the fact that the reaction between the oxirane ring and hydroxyl functional groups formed tough ether linkages and extended molecules between crosslinks, which also reduced the steric inhibition between BOZ monomers [19]. The modulus of BN/PBz composites and BN/ZE64 composites both increased with the increase of BN fractions and the modulus of the latter one grew faster than that of the former one, which can be explained by the combined action between the formation of the segregated structure and interfacial defects. The formation of a segregated structure results in a core–shell structure that can enhance stiffness, which could be offset by the existence of defects caused by air voids [29]. With the introduction of epoxy, the air voids and gaps can be eliminated greatly, which explained the faster growth in modulus. The tensile strength of them showed the opposite tendency. The main reason was that BN separated the matrix into dozens of regions, which impaired interaction between the polymer particles, degrading the toughness of the composites [11]. Due to the fact that BN lack interfacial compatibility with the matrix, the interface bonding

|                | $V_c$ | $\beta$ | $K$   | $R_s$ ($K \cdot W^{-1}$) |
|----------------|-------|---------|-------|-------------------------|
| BN/PBz         | 0.100 | 1.510   | 0.008 | $8.090 \times 10^7$     |
| BN/ZE64        | 0.080 | 0.9160  | 0.027 | $7.488 \times 10^6$     |
strengthen between the core–shell structure was weak. In the meanwhile, with the increasing filler loading, the incomplete dispersion of BN and existence of air voids would occur, which led to more defects. Therefore, the tensile strength of PBz and ZE64 composites further decreased at higher filler loading [30].

3.6. Morphology of BN/PBz and BN/ZE64 composites

Figure 9 shows the SEM images of cross-sections of PBz composites and ZE64 composites filled with different amounts of BN. Though segregated structure can be observed in both systems, the segregated pathways are different for different matrix. It can be observed that when filler loading was at 5 wt% and 15 wt%, the smooth parts, which referred to the matrix, in PBz composites were larger than that in ZE64 composites. In the meantime, the segregated pathways in PBz composites were denser than those in ZE64 composites, and big nodes can be observed in PBz composites. This was mainly because, in powder blending, the BN flakes cannot be fixed on the surface of BOZ granules so that BN flakes tend to fill in the gaps between BOZ granules instead of completely covering the matrix. It can be inferred that the big nodes in PBz composites were the gap between several BOZ granules filler with BN. In ZE64 composites, an evener distributed segregated structure can be observed because the epoxy can help to fix BN on the surface of BOZ granules, and the gaps between them will partially be filled with epoxy. In the magnified images of figures 9(a), (b), (d) and (e), it can be observed that the edges of the BN are blurry in ZE64 composites while they are clear in PBz composites. However, in magnified image of figure 9(c), the edges of BN became clear as well. This was because without epoxy resin, the contact between BN platelets and BOZ granules were ‘solid-solid phase contact’, which was unfavourable for the formation of affinity interface between BN and BOZ. With the introduction of epoxy, the BN platelets can be immersed due to the shearing force in the mixing process so that the compatibility between fillers and matrix can be improved. When the content of filler was over 25 wt%, there was not enough epoxy to immerse the BN flakes so that the interface between filler and matrix became clear.

3.7. Thermal stability of BN/PBz and BN/ZE64 composites

Thermogravimetric analysis was carried out to evaluate the thermal stability of PBz composites and ZE64 composites. The corresponding TGA curves and data are shown in figure 10 and table 3. It can be obviously observed that PBz composites exhibited better stability than ZE64 composites did at all filler loading. And the residual mass of PBz composites is higher than that of ZE64 composites. This was because epoxy possesses lower heat resistance comparing with PBz. Also, with the incorporation of epoxy, the flexible \(-C=O\) linkages in the backbones of E51 extended the main chain of PBz resin, which in turn decrease the effective crosslink density [31], which made it easier to decompose. With the increase of filler loading, the residual mass of both PBz and ZE64 composites grew gradually because BN was stable under 750 °C and the weight loss was totally caused by the matrix. However, the tendency of $T_{\text{Heat-resistance index}}$ with increasing filler loading in both composites were different. In BN/PBz composites, the $T_{\text{Heat-resistance index}}$ firstly decreased then increased with increasing filler loading, while that of BN/ZE64 composites grew slightly with increasing filler loading. This was because in BN/PBz composites when the filler loading was not sufficient to form complete segregated structure, the existence of fillers would impair the interaction between matrix and hinder the crosslink density. In BN/ZE64 composites, the adjacent BOZ granules would be connected through curing reaction.
with epoxy so that the crosslink density can be maintained. Therefore, the thermal stability of ZE64 composites was maintained while that of PBz composites was hindered when BN was first added. However, with the increase of filler loading, a core–shell structure can be formed. The shielding effect of continuous BN particles would block the spread of free radicals and delay the degradation process. Also, the excellent thermal stability and high specific capacity of BN contribute to the absorbing heat, resulted that the matrix degraded at higher temperatures.

4. Conclusions

Collaboration of epoxy and PBz resin has been proven to be a facile way constructing segregated structures with improved interfacial compatibility, which results in the enhancement in thermal conductivity and mechanical properties. SEM and density analysis confirmed the segregated structure and improved interfacial compatibility. With 25 wt% content of filler, the thermally conductive coefficient of ZE64 composites reached 0.811 W mK⁻¹, while that of PBz composites was 0.762 W mK⁻¹. FT-IR results indicated that epoxy resin copolymerized with BOZ monomers, leading to a lower crosslinking density. Therefore, the tensile strength of the matrix was improved while modulus was decreased. In addition, the THeat-resistance index of the matrix decreased from 212 °C to 191 °C.

Table 3. Corresponding characteristic thermal data of TGA curves.

| Sample name | T₅ (°C) | T₃₀ (°C) | THeat-resistance index (°C) | ω% |
|-------------|--------|---------|-----------------------------|----|
| PBZ         | 393    | 460     | 212                         | 47.9 |
| ZB05        | 368    | 456     | 207                         | 51.3 |
| ZB10        | 363    | 466     | 208                         | 54.1 |
| ZB15        | 376    | 473     | 212                         | 58.1 |
| ZB20        | 376    | 480     | 214                         | 61.0 |
| ZB25        | 375    | 494     | 219                         | 63.9 |
| ZEB64       | 349    | 417     | 191                         | 45.0 |
| ZEB6405     | 350    | 418     | 191                         | 46.4 |
| ZEB6410     | 350    | 420     | 192                         | 47.8 |
| ZEB6415     | 351    | 420     | 192                         | 49.0 |
| ZEB6420     | 352    | 423     | 193                         | 50.7 |
| ZEB6425     | 354    | 429     | 195                         | 52.9 |

1 \( T_{\text{Heat-resistance index}} = 0.49 \times [T₅ + 0.6 \times (T₃₀−T₅)] \).
2 \( T₅ \) and \( T₃₀ \) is corresponding decomposition temperature of 5 wt% and 30 wt% weight loss, respectively.
3 ω% is residual mass of the sample at 700 °C.
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References

[1] Jiang Y, Liu Y, Min P and Sui G 2017 Compos. Sci. Technol. 144 63—9
[2] Bigg D M 1986 Polym. Compos. 7 125—40
[3] Yu A, Ramesh P, Sun X, Bektasova E, Ikiz M E and Haddon R C 2008 Adv. Mater. 20 4740—4
[4] Bian W, Yao T, Chen M, Zhang C, Shao T and Yang Y 2018 Compos. Sci. Technol. 168 420—8
[5] Zhang K, Tao P, Zhang Y, Liao X and Nie S 2019 Carbohydr. Polym. 213 228—35
[6] Ng H Y, Lu X and Lau S K 2005 Polym. Compos. 26 778—90
[7] Kusy R P and Turner D T 1973 J. Appl. Polym. Sci. 17 1631—3
[8] Malliaris A and Turner D T 1971 J. Appl. Phys. 42 614—8
[9] Pang H, Xu L, Yan D-X and Li Z-M 2008 Adv. Mater. 20 4740—4
[10] Li W, Chu J, Heng L, Wei T, Gu J, Xi K and Jia X 2013 Polymer 54 4909—22
[11] Dumas L, Bonnau L, Olivier M, Poorteman M and Dubois P 2014 Eur. Polym. J. 58 218—25
[12] Takeichi T, Saito Y, Agag T, Muto H and Kawauchi T 2008 Polymer 49 1173—9
[13] Ng H Y, Lu X and Lau S K 2005 Polym. Compos. 26 778—90
[14] Ishida H and Allen D J 1996 Polymer 37 4487—95
[15] Shuai Z, Qichao R, Qiang F and Yi G 2019 React. Funct. Polym. 139 75—84
[16] Zhang X, Zhang J, Xia L, Wang J, Li G, Xu F, Zhang X, Wu H and Guo S 2018 Chem. Eng. J. 334 247—56
[17] Fonteneau R, Anez D, French S and Sobolev V L 2005 Phys. Rev. B 71 104201
[18] Fang H, Zhang X, Zhao Y and Bai S-L 2017 Compos. Sci. Technol. 152 243—53
[19] Gu J, Yang X, Lu Z, Li N, Liang C and Zhang Q 2016 Int. J. Heat Mass Transf. 92 15—22
[20] Karikal Chozhan C, Chandramohan A and Alagar M 2019 J. Macromol. Sci., Part A 56 1—16