

PREPARATION AND PROPERTIES OF THERMALLY CONDUCTIVE EPOXY RESIN/BORON NITRIDE COMPOSITES

X. N. WU^{a,b}, H. F. JI^a, Z. WANG^b, J. C. CHEN^b, J. ZHAO^b, X. F. WANG^a,
X. J. ZHANG^a, X. W. QU^{a,*}

^a*School of Materials Science and Engineering, Hebei University of Technology, Tianjin, 300130, P. R. China*

^b*College of Materials Engineering, North China Institute of Aerospace Engineering, Langfang, Hebei, 065000, P. R. China*

The epoxy resin (EP), due to its excellent mechanical and electrical properties, has been widely used in the electronic packaging. With miniaturization and integration of electronic components, the heat generated will increase significantly and heat dissipation demand of electronic packaging materials increases accordingly. However, thermal conductivity of ordinary epoxy resin is only 0.19W/m·K. In view of this, heat will be accumulated during operation of components, internal stress increases and a series of problems (e.g. cracking of products) may be caused. For this, improvement on thermal conductivity property of epoxy resin is one of the main subjects in electronic packaging materials field. In this paper, the thermally conductive epoxy resin composites with different hexagonal boron nitride (*h*-BN) content were prepared by using *h*-BN as filler and EP as matrix. In addition, the effects of *h*-BN content on thermal conductivity of EP/*h*-BN composites were investigated. The results showed that thermal conductivity of EP/*h*-BN composites was effectively improved through the addition of *h*-BN. Moreover, the composite obtained with 15 wt% *h*-BN particle loading presented a thermal conductivity of 0.6264W/(m·K), 223.72% higher than that of pure EP.

(Received June 8, 2018; Accepted October 16, 2018)

Keywords: Epoxy resin, H-BN, Thermally conductivity, Composites

1. Introduction

With the rapid development of electronic industry, miniaturization and integration of transistors has become a development trend of micro-electronic components and thermal management has become a vital issue for manufacture of more powerful and more reliable components [1-2]. In order to achieve this purpose, thermally conductive and electrically insulated materials are selected as thermal interface materials for heat dissipation [3-4]. The polymer-based composites with inorganic fillers are potential candidates, and their machinability and low density have attracted increasing attention [5-8]. Among them, the epoxy resin is widely used in electronic

*Corresponding author: keyanwxn1@hotmail.com

packaging materials, coatings, adhesives, and composite materials field because it has excellent adhesion and chemical resistance, and is an ideal thermosetting resin, namely the matrix composite material. However, thermal conductivity ($0.19\text{W/m}\cdot\text{K}$) limits its application, such as printed circuit boards, heat exchangers, thermal interface materials, and phase transition materials. In this case, high-conductivity conducting polymers are becoming the most economical and effective means of removing accumulated heat from microelectronics to deal with one of the problems of heat management.

In order to improve the thermal conductivity of epoxy resin, the high crystallization and orientation of the epoxy resin matrix can be made to accelerate the propagation speed of phonon in resin, thus improving its thermal conductivity. The thermally conductive filler can be filled in epoxy resin to prepare the composite materials with good thermal conductivity [9]. And the epoxy resin matrix is not too satisfactory in some respects, for example, it can be melted only at high temperature, its viscosity is very big, the epoxy resin matrix only can be prepared with a lot of effort, high cost and complex technological process, mass production is impossible. Therefore, the composites with high thermal conductivity are mainly prepared by filling thermally conductive filler into resin system at present. This method is convenient for operation, and the preparation technology is simple.

Polymers can provide excellent dielectric properties and excellent processability, but the thermal conductivity is quite low (about 0.19 W/mK). Therefore, the polymer-based ceramic particle composites have the potential to be used in electronic packaging by utilizing the high thermal conductivity of ceramics and the excellent processability of polymers.

Because of its constant broadband gap (approximately 5.5 eV), excellent thermal and oxidation stability, high thermal conductivity and excellent mechanical properties, the hexagonal boron nitride (*h*-BN) nanotubes are the expected nano filler to be used in various applications^[10-11]. In addition, *h*-BN nanotubes are reported to have low permittivity. The polyhedral POSS and *h*-BN nanotubes can induce the local low dielectric constant region in the composite materials. It is expected that the POSS modified *h*-BN nanotube is very suitable for making polymer composites. The polymer produced has the best dielectric properties and high thermal conductivity. In this work, POSS modified *h*-BN nanotubes have proved to be very effective nanometer fillers, which can be used to make the dielectric epoxy resin composites with high thermal conductivity. It was found that this composite material has significantly low dielectric constant and tangential dielectric loss angle [12-13]. In addition, the material has significantly improved thermal conductivity and low thermal expansion coefficient (CTE). This successfully solves the problem that high thermal conductivity and low dielectric loss cannot be realized at the same time. Intrinsic dielectric properties and effective surface modified *h*-BN nanotubes have become a key factor in the manufacturing process of composite materials.

Epoxy resin has good mechanical properties and dielectric properties and can be widely used in electronic packaging, but its thermal performance is poorer, and it is not suitable for the modern highly integrated microelectronics industry. In order to meet the requirements of microelectronics industry, it is necessary to modify it to improve its thermal conductivity. At present, the main modification method is to add thermally conductive filler with high thermal conductivity to improve its thermal conductivity. The 6.0 eV *h*-BN nanotubes with a band gap have a high characteristic surface thermal conductivity between 30 and 300W/m , and the theoretical

thermal conductivity of *h*-BN nanotubes may even be up to about 3000 W/m. With its unique thermal conductivity, *h*-BN has been widely used as the ideal filler of thermally conductive composites.

In this paper, the epoxy resin was modified by using *h*-BN to improve its thermal conductivity, and the preparation methods and properties of EP/*h*-BN composites were studied. The research idea of this paper was as follows:

(1) The suitable curing process and curing agent were selected to prepare the EP/*h*-BN composites and the curing process was determined whether it was suitable for DSC test;

(2) Through the determination of thermal conductivity, TGA, SEM and mechanical performance test, the influence of *h*-BN content on thermal conductivity, insulation performance, thermal stability and mechanical properties of composites was studied.

2. Experimental

2.1 Materials

Epoxy resin (WSR618 (E-51) was purchased from Nantong Xingchen Synthetic Material Co., Ltd. and preheated to 80°C. Methyl tetrahydrophthalic anhydride (MTHPA, content of acid anhydride $\geq 40\%$, neutralization equivalent was 81-85) was provided by Puyang Huicheng Chemical Co., Ltd. 2,4,6- trimethylamine (dimethylamine methyl) phenol (DMP-30, amine content in the yellowish transparent liquid was 600-630mgKOH/g) was provided by Puyang Huicheng Chemical Co., Ltd. And, the hexagonal boron nitride (*h*-BN, 3-5 μ m hexagonal crystal) was purchased from Qingzhou Maitekechuang Materials Co., Ltd. and was dried in a vacuum oven.

2.2 Experimental process

Because of excellent mechanical properties, bonding properties and electrical insulation properties of epoxy resin, the simple forming process, epoxy resin is widely used in the field of electronic packaging. However, the thermal conductivity of ordinary epoxy resin is only 0.2W/m·K, which cannot meet the requirements of use. In this paper, EP/*h*-BN composites with good mechanical properties, dielectric properties and thermal conductivity was prepared by using the *h*-BN with high thermal conductivity as the filler, and a series of performance tests were performed to determine its performance.

2.2.1 Preparation of EP/*h*-BN composites

The preparation of EP/*h*-BN composites consists of two parts: solution mixing and casting. The selected mass fraction of *h*-BN was 0%, 2%, 10% and 15%, respectively. The specific preparation steps were as follows:

1. A certain amount of epoxy resin was taken in a flask and then preheated in vacuum oven to 80 °C;

2. A certain amount of *h*-BN which has been dried for 24h and the EP which has been preheated to 80°C were taken in a 250ml flask, mixed in a ultrasonic cleaner for 30min at 40°C temperature and 80Hz frequency to mix the *h*-BN and EP more evenly;

3. A certain amount of curing agent and accelerant were taken and added into a flask and stirred in an ultrasonic cleaner for 30min at 40°C temperature and 80Hz frequency;

4. Ultrasonic stirring was stopped and the flask was then put into a vacuum drying oven at 80°C for 20min to remove foams;

5. The mixture was then casted into and cured in a die which has been preheated to 80°C for 4h, 120°C for 2h and 150°C for 2h, respectively;

6. The product was taken out from the die after curing and the die has been cooled down to room temperature.

Fig. 1 represents the process flow chart of EP/h-BN composites.

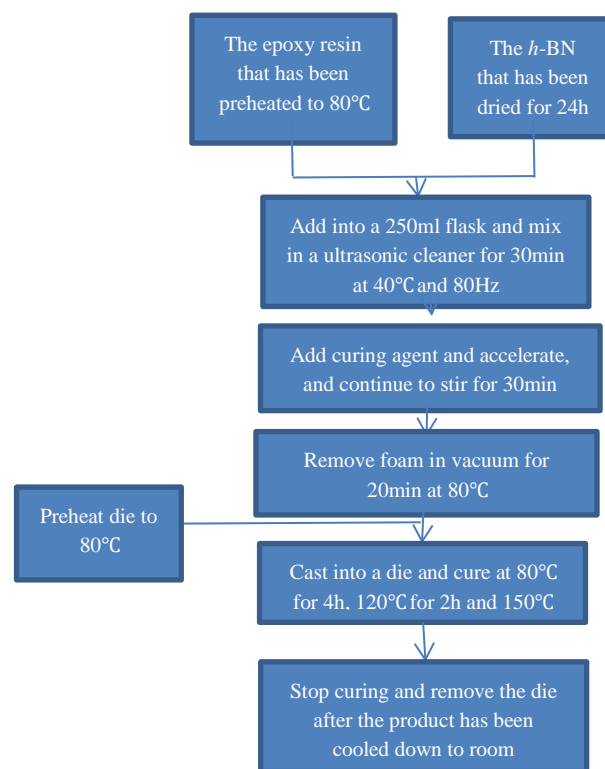


Fig. 1. The process flow chart of EP/h-BN composites.

2.2.2 Performance test of EP/h-BN composites

Differential scanning calorimetry (DSC): The curing behavior of epoxy resin was tested by Diamond DSC of American PE Company. Experimental conditions of cured epoxy resin: the solid epoxy resin was preheated from room temperature to 200°C at an interval of 5°C/min, cooled, and was then heated from 30°C to 200°C again at an interval of 10°C/min. Experimental conditions of uncured epoxy resin: the solid epoxy resin was preheated to 150°C at an interval of 10°C/min, held for 3h, cooled and was heated then to 200°C at an interval of 5°C/min.

Thermal conductivity test: The thermal conductivity is a basic performance parameter measuring the heat passing through the material of unit area and thickness in unit time when the temperature difference is 1 °C. The TC-3000 thermal conductivity meter of Xi'an Xiotech

Electronics Co., Ltd. was used for testing. At room temperature, each sample was measured 5 times and averaged.

Field emission SEM analysis (FE-SEM): The surface morphology of the fracture sample was observed with the Nova Nano SEM 450 of FEI Company. In order to prevent the electron from aggregating on the surface of the fracture surface, it was necessary to spray gold on the fracture surface.

Thermogravimetric analysis (TGA): The thermal stability of EP/h-BN composites was tested with SDT Q600 synchronous thermal analyzer provided by American TA Company. In air atmosphere, the composites were preheated from room temperature to 1,200°C at an interval of 10°C/min.

Bending performance test: The bending performance of samples was tested with the CMF6104 microcomputer control electron universal testing machine provided by Shenzhen Sans Material Test Instrument Co., Ltd., China. The test was performed on a sample of 80mm×10mm×5mm with the three-point bending method at 70% attenuation rate, 18°C and normal atmospheric humidity by referring to the GB/T 9341-2000.

Impact performance test: The impact performance of the sample was tested by ZBC-4 beam shock testing machine provided by Shenzhen Sans Material Test Instrument Co., Ltd., China. The test was performed on a sample of 80mm×10mm×5mm with 1J pendulum bob by referring to the GB/T 1043-2008.

3. Result and discussion

3.1 Analysis of curing behavior of epoxy resin

Epoxy resin curing agent has a lot of kinds, such as amine and acid anhydride, synthetic resin, etc., among them the epoxy resin with acid anhydride curing agent has excellent mechanical properties and dielectric properties, and small volume shrinkage, and is very suitable to be used as the electronic packaging materials. In this paper, bisphenol A type epoxy resin (E-51) and methyl tetrahydrobenzoic anhydride (MTHPA) were used to produce the three-dimensional crosslinking network under the catalysis of the accelerant DMO-30. The curing mechanism reaction was as follows:

1. The accelerant DMP-30 attacked the oxygen atoms on the methyl tetrahydrophthalic anhydride and generated the carboxylate anions;
2. Carboxylate anions attacked epoxy groups of epoxies and generated oxygen anions;
3. The oxygen anions continued to attack other anhydride groups, producing carboxylate anions.

According to the above process, it was gradually added to form a three-dimensional cross-linking network. The equation was as shown in Fig. 2:

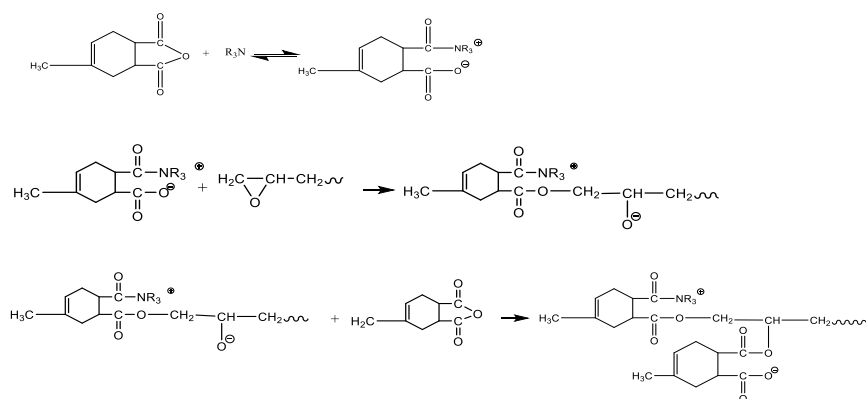


Fig. 2. The cured reaction mechanism of epoxy with anhydride.

The curing behavior of the thermosetting epoxy resin determines the crosslinked structure of the resin and determines the properties of the thermosetting resin further. Therefore, the solidification behavior of matrix resin should be studied before preparing composite materials. The epoxy resin in this paper is solidified according to the curing method. In order to evaluate the ratio of curing matrix and curing agent, curing temperature and curing time, we carried out DSC test on the solidified products. Fig. 3 presents the DSC curve of cured epoxy resin. The figure showed that the glass transition temperature of epoxy resin was 127.6°C and the composite has already been cured completely because no exothermic peak was observed after the glass transition temperature with the increase of temperature.

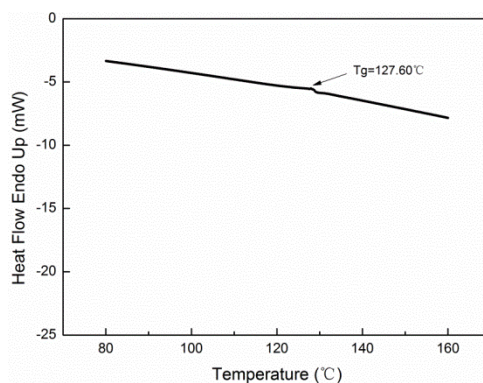


Fig. 3. DSC curve of the cured epoxy resin.

Fig. 4 was the DSC curve of uncured epoxy resin. The figure showed that the system has an exothermic peak at 144.5°C , and the epoxy resin curing temperature was 144.5°C . It showed that the curing process was reasonable.

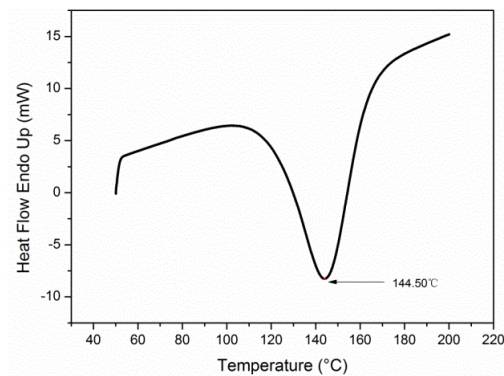


Fig. 4. DSC curve of the uncured epoxy resin.

3.2 Thermal conductivity of the composites

In this paper, the influence of *h*-BN content on thermal conductivity of epoxy resin was studied by changing the content of *h*-BN. The figure below shows the change of thermal conductivity of composite materials with *h*-BN content. It can be seen from the figure that the coefficient of thermal conductivity of pure epoxy resin was 0.1935. This was because that the crystalline and order of pure epoxy resin is poorer, and phonons cannot be transmitted rapidly only by thermal motion of macromolecular chains, links, and the groups. Therefore, it was necessary to add *h*-BN to change its heat transfer efficiency. It can be seen from Fig. 5 that the thermal conductivity of composite materials gets better and better with the increase of *h*-BN content.

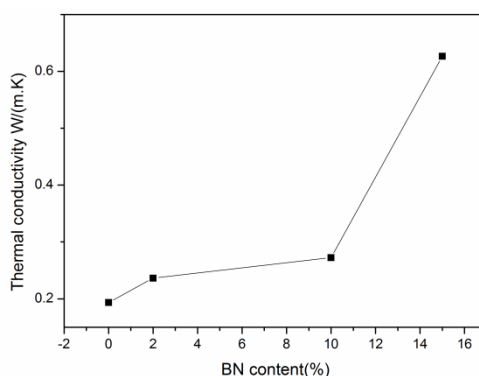


Fig. 5. Change of thermal conductivity with increase of *h*-BN content.

The reason for this phenomenon is the dependence of thermal conductivity on filler content. *h*-BN has higher thermal conductivity. When *h*-BN was added to the epoxy resin, the composite material can have the properties of two kinds of materials. Therefore, the thermal conductivity of composites increased with the increase of *h*-BN, and its thermal conductivity got better and better. When the content of *h*-BN was relatively small, the *h*-BN particles were dispersed and isolated from each other in the resin matrix, presenting an "island" structure. Although the heat transfer was faster in the *h*-BN particles, it was slower to pass through the

interface between *h*-BN and epoxy resin due to the large thermal resistance. It made the heat transfer efficiency in the whole composite material slower. Therefore, when the content of *h*-BN was lower, its thermal conductivity was not much higher. Along with the increase of the content of *h*-BN, *h*-BN particles touched each other in the resin matrix, a heat conduction path was formed, the heat was propagated rapidly through the heat conduction pathway and then the composites showed higher heat transfer efficiency. Therefore, a heat transfer efficiency mutation occurred before and after the thermal conduction pathway. It can be seen from the figure that the mutation of thermal conductivity of composite materials appeared when the content of *h*-BN was between 10% and 15%.

Table 1. Thermal conductivity improvement of the epoxy composites.

	Content of <i>h</i> -BN (wt%)			
	0	2	10	15
Thermal conductivity W/(m·K)	0.1935	0.2363	0.2724	0.6264
Δ (%)	—	22.12	40.75	223.72

Table 1 represents the percentage of the thermal conductivity and the increase in thermal conductivity of the composite with different contents of *h*-BN. Δ represents the percentage of the increase in thermal conductivity. It can be seen from the Table that the thermal conductivity increases gradually with the increase of the content of *h*-BN in the composites, and the higher the content of *h*-BN, the more obvious the thermal conductivity increases. The data in the Table further proves that when the content of *h*-BN is relatively small, the *h*-BN particles present the island structure in the resin matrix, and the thermal conductivity of the composites is not much higher. When the content of *h*-BN increases to a certain amount, the thermal conductive pathway is formed and the thermal conductivity is increased significantly.

3.3 The microscopic appearance of composites

In order to further study the microstructure of epoxy resin matrix composites, we carried out SEM test on the samples. Fig. 6a represents the fracture morphology of pure epoxy resin. It can be seen from the figure that the fracture surface of pure epoxy resin was smooth, the section was fluvial, the surface was smooth, there was almost no plastic deformation, and it was brittle fracture.

Fig. 6b represents the fracture morphology of epoxy resin composite after adding 2wt% *h*-BN. It can be seen from the figure that after adding *h*-BN, some parts of the fracture surface were smooth, some parts were rough, and the fracture surface was obviously rougher than that of the pure epoxy resin. Moreover, the *h*-BN particles were randomly exposed to the epoxy resin matrix, presenting an irregular concave and convex shape, which is due to the deformation or shear yield of the matrix caused by the filled *h*-BN particles. However, due to the small amount of filler, the *h*-BN particles had no contact with each other in the matrix, which cannot form the thermal

conduction pathway, and the thermal conductivity of the material was improved less.

Fig. 6c represents the fracture morphology of EP/10wt% *h*-BN composite material. It can be seen from the figure that the fracture surface was rougher, and its surface defects and holes increased. This was because that the wettability of *h*-BN films was poor and the surface energy was relatively high, and the concentration of microns in the two-phase interface has caused a lot of defects and holes. The combination of the matrix and filler was not good, and the large size of the phase interface can greatly reduce the average free path of phonon propagation due to boundary scattering and defect scattering, thus reducing the heat transfer efficiency. So when 10wt% *h*-BN was added, the thermal conductivity was not improved much higher.

Fig. 6d represents the fracture morphology of EP/15wt% *h*-BN composite material. It can be seen from the figure that when the filling volume of *h*-BN reached to 15%, the fracture surface was very rough and there was almost no smooth part. Moreover, it can be seen that the *h*-BN sheets overlapped together in the matrix, the network structure was formed, a heat conduction pathway was formed throughout the resin matrix, heat can be passed along the thermal conductive pathway quickly, and the thermal conductivity of the composite was greatly increased.

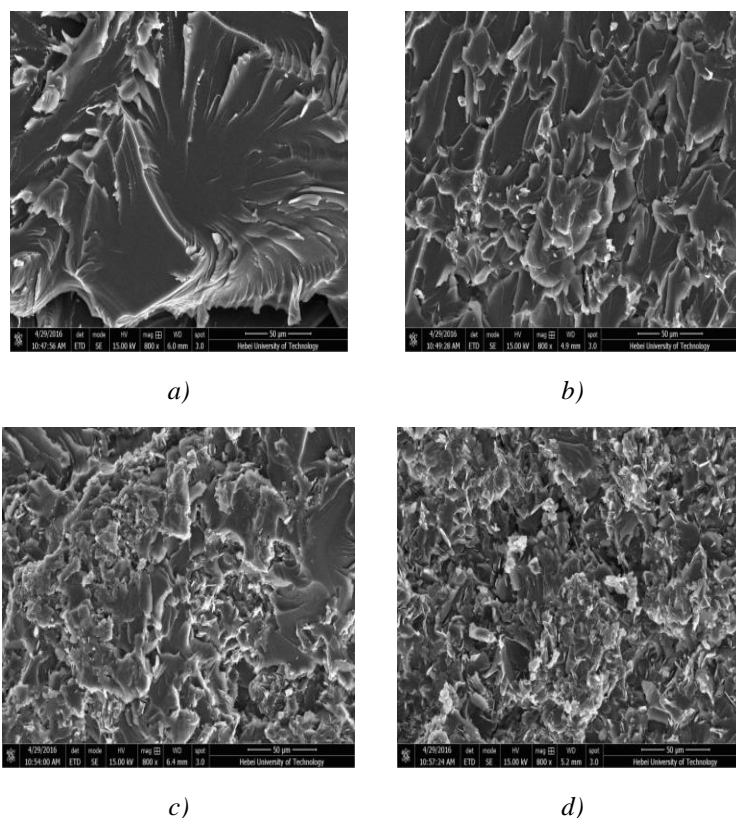


Fig. 6. Fractured surfaces of EP/*h*-BN composites: a. EP/15wt% BN; b. EP/15wt% BN; c. EP/15wt% BN; d. EP/15wt% BN.

3.4 Thermal stability of composites

Because the EP/*h*-BN composites in this paper are going to be used in the field of electronic packaging and the electronic technology develops in the direction of integrated circuit at high speed, the electronic packaging materials have to work under high temperature conditions.

Therefore, the composites should not only have good thermal conductivity, but also have good aging resistance. Only in this way can the composites be better suited for electronic packaging. The heat loss curve of EP/*h*-BN composites in air condition was as shown in Fig. 3.6, and the differential curve of thermal loss was as shown in Fig. 7. The curve 1 in the figure represents the thermo-gravimetric curve of the pure epoxy resin, the curve 2 represents the thermo-gravimetric curve of EP/2wt% *h*-BN composite, the curve 3 represents the thermo-gravimetric curve of EP/10wt% *h*-BN composite, and the curve 4 represents the thermo-gravimetric curve of the EP/15wt% *h*-BN composite. The initial decomposition temperature of 10 wt% ($T_{10\%}$), decomposition temperature of 50 wt% ($T_{50\%}$), decomposition rate of the highest temperature (T_{max}) and the rate of residual carbon under 1,100°C ($Y_{cat\ 1100^\circ C}$) of all samples are summarized in a table. These typical thermal stability parameters can reflect the thermal aging properties of the composites.

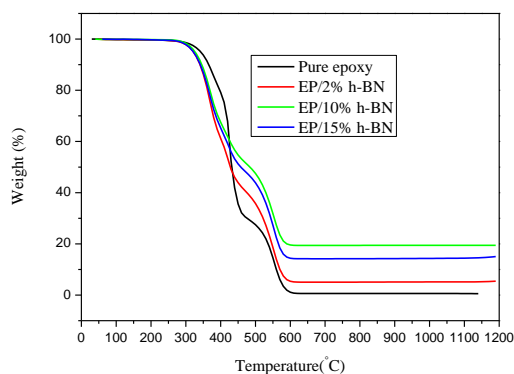


Fig. 7. TGA curves EP/*h*-BN composite.

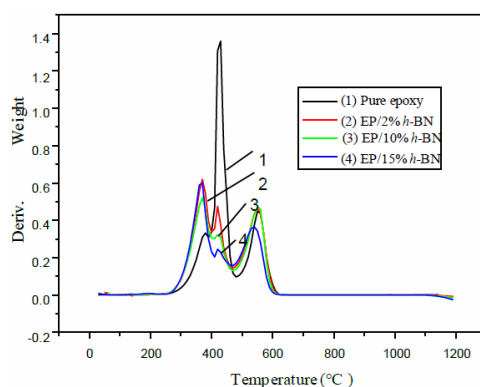


Fig. 8. DTG curves of the EP/*h*-BN composites.

As can be obtained from Fig. 7, the thermal decomposition curve of all EP/*h*-BN composites is similar to that of pure epoxy resin. This shows that the addition of *h*-BN does not affect the thermal decomposition mechanism of materials. It can be seen from Fig. 7, Fig. 8 and Table 2 that: (1) The epoxy resin added anhydride as curing agent has high thermal stability, and

its $T_{10\%}$, $T_{50\%}$, $T_{\max 1}$, $T_{\max 2}$, $T_{\max 3}$ and $Y_{\text{cat } 1100^\circ\text{C}}$ is 365.7, 432.2, 380.1, 430.7, 549.9 and 0.51%, respectively; (2) The addition of *h*-BN delayed the thermal decomposition process of composites. The addition of *h*-BN can improve the thermal stability of the materials, and the more *h*-BN was added, the better the thermal stability will be. The $T_{50\%}$ and $Y_{\text{cat } 1100^\circ\text{C}}$ of EP/15wt% *h*-BN composite were 27.1% and 18.2% higher than that of pure epoxy resin. This is because *h*-BN itself has a high thermal capacity and thermal conductivity, and its large and hard six-square layer acted as a barrier for heat resistance and mass transfer, which can delay the degradation process.

Table 2. Thermal stability of the EP/*h*-BN composites.

Sample	$T_{10\%}$ (°C)	$T_{50\%}$ (°C)	T_{\max} (°C)			$Y_{\text{cat } 1100^\circ\text{C}}$ (%)
			$T_{\max 1}$ (°C)	$T_{\max 2}$ (°C)	$T_{\max 3}$ (°C)	
Pure epoxy	365.7	432.2	380.1	430.7	549.9	0.51
EP/2wt% <i>h</i> -BN	340.7	427.9	369.9	421.1	549.9	5.16
EP/10wt% <i>h</i> -BN	341.4	458.1	369.3	419.9	549.9	14.36
EP/15wt% <i>h</i> -BN	338.2	459.3	369.3	419.9	540.1	18.71

3.5 Mechanical properties of composites

Pure epoxy resin has good mechanical properties and dielectric properties, so it is especially suitable for electronic packaging. However, the addition of *h*-BN filler will change its mechanical properties, so we must study the mechanical properties of the composites and determine whether they can be applied in the field of electronic packaging. In this paper, EP/*h*-BN composites were studied and their texture was hard and brittle. So we mainly studied the bending properties and impact properties of the composites.

3.5.1 Impact properties of composites

Fig. 9 is the schematic diagram of impact process of the composites. It can be seen from the figure that the crack propagation of the sample was divided into three zones: zone A (crack forming zone), zone B (crack slow propagation zone) and zone C (crack rapid propagation zone). In the moment of the experimental shock, the zone A was subjected to the corresponding tensile stress, and the zone C was subjected to the corresponding compressive stress. After the crack was formed, the crack was propagated continuously under the effect of tensile stress in zone A and produced new fine cracks. But before the crack entered the zone C, the front end of the crack was always subjected to compressive stress. Once the crack entered the zone C, the compressive stress transformed into tensile stress and caused rapid propagation of the crack.

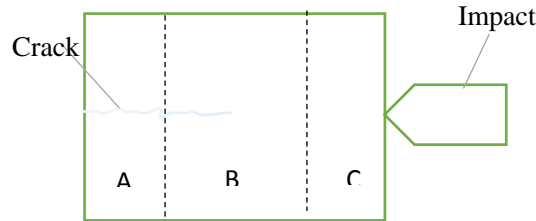


Fig. 9. Schematic diagram of the impact test.

Fig. 10 shows the impact performance schematic diagram of the composites with different *h*-BN contents. As can be seen from Fig. 10, as the content of *h*-BN increased, the impact performance of composites got worse and worse. The impact performance of pure epoxy resin was much lower than that of the epoxy resin with 2wt% *h*-BN. This was because the mechanical properties of the polymer depended to a great extent on the binding force between the two phase interfaces. The compatibility and dispersion of both the *h*-BN and epoxy resin are poor. Therefore, there was a certain gap between the two phase interfaces, and the combination was not strong. When subjected to impact, the two phase interface was easy to crack, and as the crack propagated, the whole specimen cracked. On the macro level, the impact performance was worse.

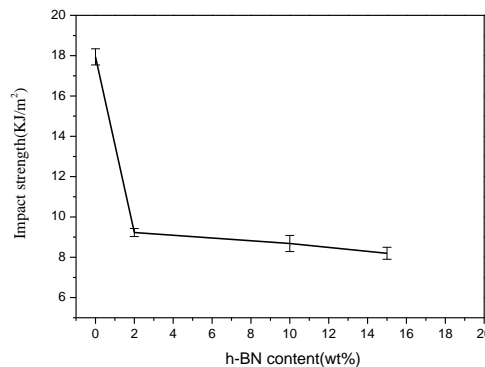


Fig. 10. Impact properties of EP/*h*-BN composite.

In order to be able to accurately characterize the mechanical properties of composites, 4-5 specimens of each composite with different *h*-BN content were tested in the process of testing, an average was taken, and the standard deviation of the test results was analyzed to determine the reliability of the results. The standard deviation of impact strength of each composite is listed in Table 3.

3.5.2 Bending properties of composites

Fig. 11 shows the bending properties of EP/*h*-BN composites. It can be seen from Fig. 11 that its bending strength gets smaller and smaller with the increase of *h*-BN. This is because the mechanical properties of the polymer depend to a great extent on the binding force between the two phase interfaces. The compatibility and dispersion of both the *h*-BN and epoxy resin are poor.

Therefore, there was a certain gap between the two phase interfaces, and the combination was not strong. The three-point bending test method was used in this experiment. The interface of *h*-BN and epoxy resin cracked under the action of shear force. So when *h*-BN was added, the bending strength decreased. With the increase of the amount of *h*-BN, the agglomeration occurred, and the stress concentration was concentrated in the reunite point, causing the bending strength to decrease further.

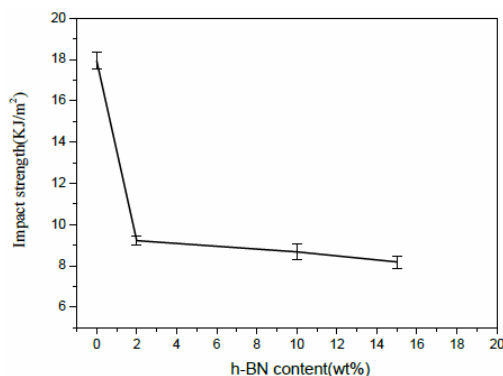


Fig. 11. Bending performance EP/*h*-BN composites.

In practical applications, the composites should not only have good thermal conductivity, but also have good mechanical properties. Therefore, it is necessary to improve the thermal conductivity of the composites on the basis that their mechanical properties meet the application requirements.

4. Conclusions

In this project, *h*-BN was used as the filler and epoxy resin as the matrix to prepare the EP/*h*-BN thermal insulation composites with different mass fraction of *h*-BN. The influence of *h*-BN mass fraction on thermal conductivity, mechanical properties and thermal stability of composite materials was studied. The thermal conductivity of the EP/*h*-BN thermal insulation composites increases with the increase of *h*-BN content. When the mass fraction of *h*-BN reaches to 15%, the thermal conductivity of the composites reaches to 0.6264W/(m·K), which is 223.72% higher than that of pure epoxy resin. It is indicated that adding of *h*-BN filler can effectively improve the thermal conductivity of composites. It is found from the comparison between the thermal conductivity measured during experiment and the thermal conductivity obtained from the Maxwell-Eucken model that when the content of *h*-BN is low, the predicted value of thermal conductivity is similar to that of the experimental value. It can be seen from observation of the scanning electron microscopy (SEM) that the addition of *h*-BN will change the fracture appearance of the composites and form a more obvious thermal conduction pathway. It can be obtained from TGA test that the addition of the filler does not change the thermal decomposition mechanism of composite material, and the addition of filler can improve its thermal stability. The bending

performance and impact performance test show that although the bending properties and impact properties of the composites with *h*-BN were reduced to some extent, it can still meet the requirements of use in a certain range.

Acknowledgements

This work was financially supported by Hebei key research and development project(contact no. ZD2018239), Program for North China Institute of Aerospace Engineering Youth Foundation (contact no. ky201601), and Langfang Science and Technology Supporting Plan Program (contract no. 2016011083).

References

- [1] S. Pal, S. Vivekchand, A. Govindaraj, C. Rao, *Mater. Chem.* **17**, 450 (2007).
- [2] S. Diahm, F. Saysouk, M. Locatelli, et al., *J. Phys. D Appl. Phys.* **48**, 385301 (2015).
- [3] N. Yang, C. Xu, J. Hou, et al., *RSC Adv.* **6**, 18279 (2016).
- [4] F. F. Wang, X. L. Zeng, Y. M. Yao, et al., *Sci. Rep.* **6**, 19394 (2016).
- [5] T. Morishita, H. Okamoto, *ACS Appl. Mater. Interfaces* **8**, 27064 (2016).
- [6] M. Shahrokhi, B. Mortazavi, G. R. Berdiyrov, *Solid State Commun.* **253**, 51 (2017).
- [7] Q. Peng, W. Ji, D. Suvranu, *Comput. Mater. Sci.* **56**, 11 (2012).
- [8] X. Wu, K. Zhang, Y. Zhang, H. Ji, X. Qu, *Digest Journal of Nanomaterials and Biostructures* **13**(2), 399 (2018).
- [9] X. Wu, L. Zhai, N. Wang, X. Qu, *Polymer Materials Science and Engineering* **34**(3), 185 (2018).
- [10] C. Zhi, Y. Bando, C. Tang, D. Golberg, *Materials Science & Engineering R Reports* **70**, 92 (2010).
- [11] J. Yu, Y. Chen, R. Wuhler, Z. Liu, S. Ringer, *In Situ Formation of BN Nanotubes during Nitriding Reactions. Chemistry of Materials.* **17**, 5172 (2005).
- [12] A. Gomathi, S. Hoseini, C. Rao, *Journal of Materials Chemistry* **19**, 988 (2009).
- [13] S. Pal, S. Vivekchand, A. Govindaraj, C. Rao, *Journal of Materials Chemistry* **17**, 450 (2007).