

Research on Preparation and Performance of Epoxy Resin-Boron Nitride Thermal-Conductive Composite

Xiangnan Wu^{1,a}, Zhi Wang¹, Jiuchuan chen¹, Xiaofeng Wang² and Xiongwei Qu²

¹College of Materials Engineering, North China Institute of Aerospace Engineering, Langfang, Hebei, 065000, China

²School of Materials Science and Engineering, Hebei University of Technology, Tianjin, 300130, China

^a2837084447@qq.com

Abstract. Owing to its excellent mechanical and electrical properties of epoxy resin, epoxy resin is widely used in electronic packaging field. However, thermal conductivity of ordinary epoxy resin is only 0.2W/m·K, heat is easy to be accumulated during operation of components, the stress will increase and a series of questions (e.g. cracking of product) would be caused then. Therefore, enhancing of thermal conductivity of epoxy resin is a focal research point in electronic packaging material field. In this study, hexagonal boron nitride was used as the filler, epoxy resin was used as the matrix, the hexagonal boron nitride thermal-conductive insulating composite of different mass fraction was prepared, and the impact of different boron nitride mass fraction on performance of composite was researched. It was discovered during the research that adding of boron nitride (h-BN) filler enhanced thermal conductivity of composite effectively. When mass fraction of h-BN reached to 15%, thermal conductivity of composite was up to 0.6264W/(m·K), increased by 223.72% than that of pure epoxy resin.

1. Introduction

With rapid development of electric industry, miniaturization and integration has been the development trend of transistors used on microelectronic parts and thermal management has become an important problem for manufacturing of more powerful and reliable components [1-2]. For that purpose, the thermal-conductive and electrically-insulated material was selected as the thermal interface material for thermal dissipation [3-4]. The polymer matrix composite added with inorganic filler is the potential candidate and its machinability and low density has attracted more and more attention [5-7]. Epoxy resin has outstanding adhesion and chemical resistance, is an ideal thermosetting resin that can be used as the matrix composite and has been widely used in electronic packaging material field. However, its poor thermal conductivity (0.2W/m·K) restraints its applications in such as printed circuit board, heat exchanger, thermal interface material and phase-change material fields.

In order to enhance thermal conductivity of epoxy resin, the composite of high conductivity is mainly prepared by filling thermal-conductive filler to resin matrix at present. This method is easy to operate and preparation technology is simple.



This paper mainly researched modification of epoxy resin by means of boron nitride, enhancement of its thermal conductivity, preparation methods and properties of BN/EP composite. The impacts of BN content on thermal conductivity of composite was researched by measurement of thermal conductivity coefficient and SEM testing, etc.

2. Experimental

2.1. Main raw materials to be used during experiments

Epoxy resin (E-51) produced by Nantong Xingchen Synthetic Material Co. Ltd., China; methyl tetrahydrophthalic anhydride (MeTHPA) produced by Puyang Huicheng Electronic Material Co., Ltd. China; 2, 4, 6-tri (dimethylamioethyl) phenol (DMP-30) produced by Puyang Huicheng Electronic Material Co., Ltd., China; boron nitride (BN) produced by Qingzhou Matt Technology Innovation Materials Co., Ltd.

2.2. Preparation of epoxy resin/boron nitride composite

Preparation of composite includes two parts, namely solution mixing and casting molding. Mass fraction of boron nitride is 0%, 2%, 10% and 15%, respectively. The specific preparation procedures are as follows: add a certain quantity of epoxy resin in a flask and preheat it to 80 °C in an oven; weigh and take a certain quantity of boron nitride which has been dried for 24h, add it into a 250ml flask with the epoxy resin which has been preheated to 80 °C together. Then, mix them ultrasonically for 30min in an ultrasonic cleaner under 40 °C and 80Hz, so as that BN was mixed with epoxy resin more evenly; then, stop ultrasonic stirring and place the flask in a vacuum drying oven for vacuum deaeration for 20min at 80 °C; then, the mixture was poured into a mold that has been preheated to 80 °C and cured at 80 °C 1h+120 °C 2h+150 °C 2h; after curing, take out the mold and remove the old after the mixture has been cooled down to room temperature.

2.3. Performance test of epoxy resin/boron nitride composite

Determination of thermal conductivity coefficient: the TC-3000 thermo-conductivity meter produced by Xi'an Xiayi Electronic Technology Co., Ltd. was used during determination. Each specimen was measured 5 times at room temperature and an average was calculated out.

Field emission scanning electron microscopy analysis (FE-SEM): the Nova Nano SEM 450 produced by FEI Corporation was used to observe surface appearance of cracked samples. In order to prevent electrons from focusing on surface of fracture plane, the fracture plane shall be sprayed with metal firstly.

3. Results and discussion

3.1. Thermal conductivity of composite

In this paper, impacts of BN content on thermal conductivity of epoxy resin were researched by changing BN content. Changes of its thermal conductivity caused by changes of BN content were as shown in the figure below. It can be seen from the figure that thermal conductivity coefficient of pure epoxy resin is 0.1935. The reason was that crystalline and ordering of pure epoxy resin is poor, heat conductivity was realized only by thermal motion of macromolecular chain segments, chain links and groups, and the phonons cannot be transmitted rapidly. Therefore, we add BN to change its thermal conductivity.

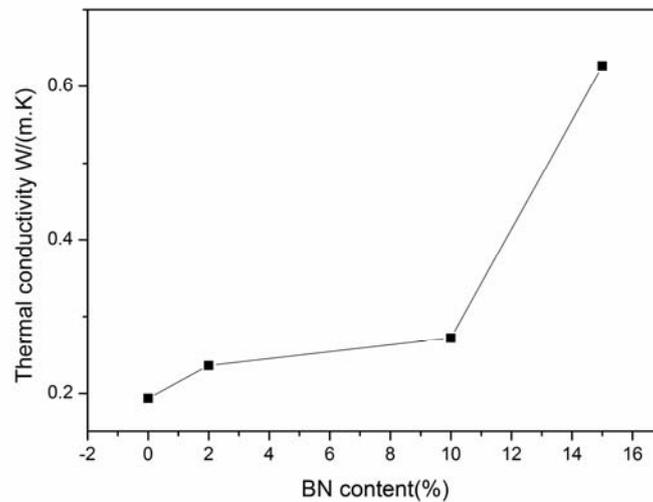


Figure 1. Relation between thermal conductivity and BN content

It can be seen from Fig.1 that the thermal conductivity becomes higher with increasing of BN content. The reason for this phenomenon is that thermal conductivity depends on content of filler. Owing to higher thermal conductivity of boron nitride, the composite will have properties of two materials after boron nitride is added to epoxy resin. Hence, thermal conductivity of composite will be increased accordingly. Moreover, thermal conductivity of the composite will become higher and higher with increasing of boron nitride content. When content of boron nitride is less, the boron nitride particles in resin matrix were mutually dispersed and isolated and an “island” structure was presented. So, when content of boron nitride is less, thermal conductivity of the composite was not enhanced significantly. With increasing of boron nitride, the boron nitride particles will contact mutually in resin matrix and form a thermal-conductive path, the heat spreads in this thermal-conductive path, and the composite shows higher heat transfer efficiency. Therefore, the heat transfer efficiency will change abruptly before and after formation of this thermal-conductive path. We can see from the figure that the thermal conductivity of composite changes abruptly when content of boron nitride is 10%-15%.

Table 1. Thermal conductivity improvement of the epoxy composites

BN content (%)	0%	2%	10%	15%
Thermal conductivity W/(m.K)	0.1935	0.2363	0.2724	0.6264
Improvement percent	---	22.12%	40.75%	223.72%

It can be seen from Table 1 that thermal conductivity of composite increases gradually with increasing of boron nitride content and increasing of thermal conductivity coefficient is more obvious with increasing of boron nitride content. When mass fraction of h-BN reaches to 15%, thermal conductivity of composite is up to 0.6264W/ (m.K), increasing by 223.72% than that of pure epoxy resin.

3.2. Morphology of composite

In order to research morphology of epoxy resin-based composite further, we carry out SEM test on fracture of samples.

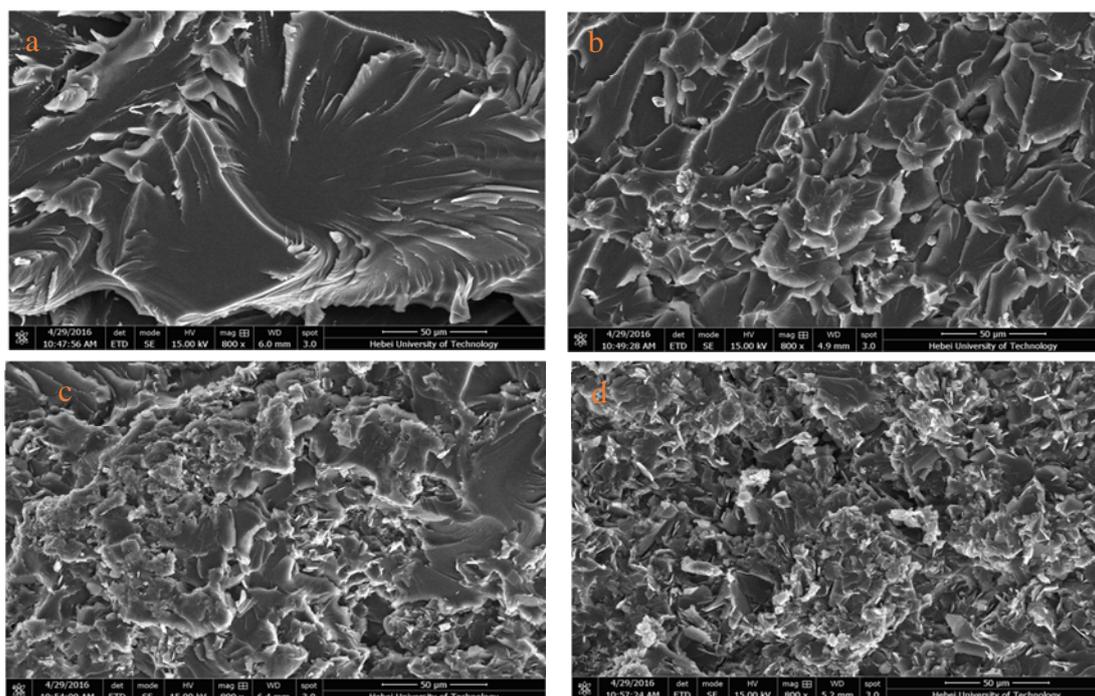


Figure 2. SEM of EP/h-BN composites

a. EP/15wt% BN; b. EP/15wt% BN; c. EP/15wt% BN; d. EP/15wt% BN

Fracture morphology of pure epoxy resin is as shown in Fig.2.a. We can see from the figure that fracture surface of pure epoxy resin is flat, the fracture has river morphology, smooth surface and nearly no plastic deformation. Therefore, the fracture is brittle.

It can be seen from Fig.2.b that some part of the fracture surface is smooth and some is rough after BN is added. When being compared with pure epoxy resin, the fracture surface is obviously rougher, BN particles expose in epoxy resin matrix randomly and form irregular rough pattern. This is because that the boron nitride particles cause deformation or shear yielding of matrix. However, owing to content of added filler is less, BN particles will not contact with each other in matrix, a thermal conductive path cannot be formed and thermal conductivity of composite is enhanced significantly.

It can be seen from Fig. 2.c that the fracture surface becomes rougher, and the defects and number of holes on the surface increases. This is caused by poor wettability and relatively high surface energy of BN plate and the aggregation beams of the micron plates gathered in two-phase interface cause large numbers of defects and holes. This filler will bond with the matrix well, the large phase interface size will, because of boundary scattering and defects scattering, significantly reduces mean free path of phonon transmission, and reduces heater transfer efficiency. Therefore, when 10wt%BN is added, its thermal conductivity will not be enhanced greatly.

It can be seen from Fig.2.d that the fracture surface was very rough and nearly not smooth when content of BN filler reached to 15%, the BN plates overlapped together in the matrix to form a network structure and form a thermal-conductive path in the entire resin matrix, the heat will transfer rapidly along the thermal-conductive path and thermal conductivity of composite is enhanced significantly.

4. Conclusion

In this paper, hexagonal boron nitride was used as the filler, epoxy resin is used as the matrix and the EP/h-BN thermal-conductive insulated composite containing different hexagonal boron nitride mass fraction was prepared. The research showed that thermal conductivity of EP/h-BN thermal-conductive insulated composite increases with increasing of h-BN content. When mass fraction of h-BN reached to 15%, thermal conductivity of composite was up to 0.6264W/(m·K), increasing by 223.72% that that of

pure epoxy resin. It showed that thermal conductivity of composite was effectively enhanced by adding of h-BN filler. It can be seen from fracture appearance when being observed by a scanning electron microscope that fracture appearance of composite was changed and relatively obvious thermal-conductive path was formed by adding of BN.

Acknowledgments

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