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Enhanced Thermal Conductivity for Mesophase Pitch-Based Carbon Fiber /Modified Boron Nitride/Epoxy Composites

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Abstract

Carbon fiber reinforced polymer composites are important in advanced components which require high thermal conductivity (TC). However, the epoxy-rich interlayer makes even highly thermal conductive mesophase pitch-based carbon fibers (MPCF) less effective in the out-of-plane direction. In this study, boron nitride (BN) particles with high TC are incorporated in MPCF/epoxy composites and after an efficient surface treatment by coupling agent KH550, TC of composite has achieved 7.9 W/(m*K) which enhances 61% compared with the control sample without BN (4.9 W/(m*K)). It is possibly attributed to the reinforced adhesion among modified BN (mBN), epoxy and MPCF achieved by the silanol groups of KH550 which enables to reduce the thermal resistance as well as increase efficient packing between fillers and polymer matrix. Besides, TC of composites with mBN also is more stable with temperature's changing compared with the one incorporated by pristine BN.

Key words: surface treatment, boron nitride, thermal conductivity, carbon fibers

Introduction

Carbon fibers (CF) are the most promising materials in the field of aerospace and aeronautics due to the low density and high performances including the good mechanical property, flexible processability, and corrosion resistance, etc, compared with conventional materials like metal alloys or ceramics.^[1,2] Moreover, as an excellent thermal conductor, CF can also meet the requirements of high thermal conductivity (TC) for aerospace and aeronautics devices to guarantee efficient heat diffusion conduction and avoid locally substantial overheating and degradation of aircrafts.^[3] CF's thermal properties largely depend on the process condition and the organic precursors, namely the polyacrylonitrile (PAN)-based and the mesophase pitch (MP)-based.^[4-6] The mesophase pitch-based carbon fibers have a much higher in-plane TC (up to 1000 w/(m*K) as reported) than that of PAN-based ones owing to the highly crystalline and oriented microstructure.^[7, 8] Thus, the mesophase pitch-based carbon fibers (MPCF) are attractive for reinforcement materials in a variety of structural and functional components where the thermal conduction is highly required, such as artificial satellite antenna, heat sink and other thermal controls in electronics devices, etc. Unfortunately, the high TC of MPCF is largely limited once fabricated into composites due to the intrinsic low TC (around 0.2 w/(m*K)) and insolate nature of epoxy matrix. Therefore, it is highly desired to find an efficient way to improve the out-of-plane TC for the MPCF reinforced epoxy composites.

Enormous efforts and attempts have been endowed on how to address the problem of heat transfer in epoxy-rich zones of the composites. For instance, the liquid crystalline epoxy with high TC 1.16 w/(m*K) is developed to improve TC of epoxy from the view of polymer's structure directly,^[9, 10] but it is worthwhile to point out that the liquid crystalline epoxy usually has a high melting point which restricts the further application as a matrix for the composites. Another common and popular way is to prepare composites by incorporating fillers with high TC into the epoxy matrix including carbon based materials such as carbon nanotubes (CNT),^[11, 12] graphite nanoplatelets (GNP),^[13, 14] ceramic fillers such as boron nitride (BN),^[15, 16]

aluminum nitride (AlN)^[17, 18] and metal particles^[19, 20] such as silver, copper, etc. In order to achieve high TC for the composites, a large volume fraction of the fillers (over 50%)^[1, 15-18, 24, 25] is usually needed but the enhancement on TC sometimes is slight since the interface adhesion between fillers and matrix is hard to be controlled well. The poor adhesion introduces more defects, voids and much area of interfaces which unavoidably lead to low TC for composites because the possibility of phonons scattering increases and then the heat transfer in the inner of composite is weakened. For the epoxy matrix, the heat transfer is mainly conducted by the energy quanta of atomic lattice vibration or phonon which is different from the metal by electrons as the charge carries.^[1, 2] Hence, from the perspective of heat transfer, for pursuing a higher TC of MPCF/epoxy matrix composite, a better adhesion for fillers and epoxy matrix is the doubtlessly key factor for the composite to achieve high thermal property.

Surface treatment of fillers is a widely-used method to optimize the adhesion of fillers-matrix and modify fillers' dispersal.^[21-23] There are various surface modifiers for ameliorating filler's surface property and reducing thermal interfacial resistance with matrix including surfactants, coupling agents, functional polymers and inorganic coating.^[1] Experimental results have shown that an appropriate surface treatment for fillers including type's choosing and content's controlling enables to strengthen the interface regions' bonding. Furthermore, the surface modifiers can reduce the viscosity of the epoxy during the processing which can modify filler's dispersal in the epoxy. Even humble surface treatments for fillers may enhance the global TC for the composites as reported by researchers.^[1]

In this study, in order to achieve high out-of-plane TC for the composite, MPCF, BN and epoxy are chosen as the raw materials to prepare composites (MPCF/BN/epoxy) with double layers structure. We firstly investigate different BN's volume fractions from 5% to 20% and moreover, an organo-silanes coupling agent (KH550) is applied for a surface treatment on BN particles (remarked as mBN) so as to compare with TC of the composite incorporated by pristine BN. The TC results illustrate that due to forming better interfacial adhesion after the surface treatment, the

out-of-plane TC of composite with merely 20% volume fraction of mBN (MPCF/mBN-20%/epoxy) has reached 7.9 W/(m*K) which is much higher than that of control sample MPCF/epoxy (4.9 W/(m*K)). Besides, the TC of MPCF/mBN-20%/epoxy also shows good stability with the temperature changing.

Experimental

Materials: The polymer matrix used in this study is epoxy: the resin is 1080S (Resoltech Ltd., France), bisphenol F-(epichlorhydrin) (number average molecular weight ≤ 700). The curing agent is 1084 (Resoltech Ltd., France), 4-4'-methylenebis (cyclohexylamine) (MCHA). The mass ratio of resin and curing agent is 3:1. Boron nitride (BN) particles with the size of 1 μm and the coupling agent, (3-Aminopropyl) triethoxysilane (KH550), are purchased from Sigma Aldrich directly. CW-300 is as the MPCF tissues used in the composite's preparation. MPCF tissue used in the study is ThermalGraph EWC-300X bought from Cytec and its TC are about 250-300 W/(mK).

Preparation of composites: The surface treatment for BN by KH550 can be found in elsewhere [\[24, 25\]](#) and in this study the addition of KH550 was 3% calculating to BN's mass and the solvent used was ethanol. The preparation of the composite were divided into three steps: first, the mixture of fillers (BN or mBN) and resin with calculated weight fractions were mixed by three rolls mill for 20 min at the room temperature and after that the curing agent was dropped into the mixture. The obtained slurry of fillers, resin and curing agent was applied on two MPCF tissues carefully to form the composite with double layers. Second, the composite was put into two aluminum molds under vacuum for 30 min to remove the bubbles. Afterward, it was put into a vacuum bag and moved into a hot-press machine under the pressure of 0.8 Pa at 60 °C for 2 h with pump. Finally, the further curing of epoxy was conducted in an oven at 60 °C for 10 h. The whole preparing process and epoxy's chemical reaction are presented in Fig.1.

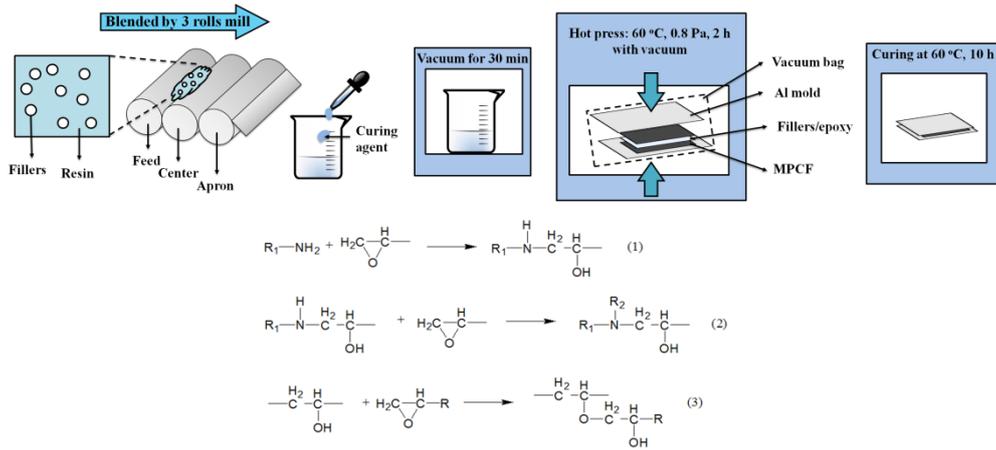


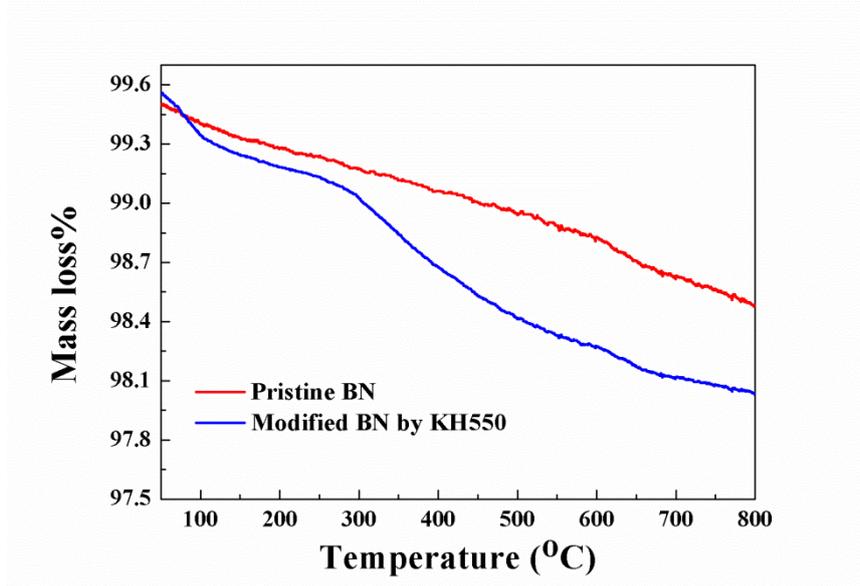
Figure 1. Schematic figure of the composite preparation process and the chemical reaction for epoxy matrix.

Characterization: The morphology for the surface fracture of the composite was conducted by scanning electron microscope (SEM) (LEO Gemini 530). Thermo gravimetric analysis (TGA) characterization was conducted by NETZSCH STA 449F3 in a wide temperature range from 30 to 800 °C with 10 °C/min heating rate. The out-of-plane TC was calculated by flash method (Nanoflash LAF 440) at room temperature (20 °C) and the evaluation was conducted by Cowan fitting with finite pulse correction. Dynamic mechanical analysis (DMA) for samples of the composite was conducted in three-point bending mode by NETZSCH DMA 242C. The measurement was from 30 °C to 150 °C and with 2 °C/min heating rate at 1 Hz. The size of the sample for the measurement was 40 mm × 10 mm × 1 mm. The loss tangent for the modulus is defined as $\tan \theta = \frac{E''}{E'}$, where E' and E'' correspond to the real and imaginary parts of the storage modulus E, respectively.

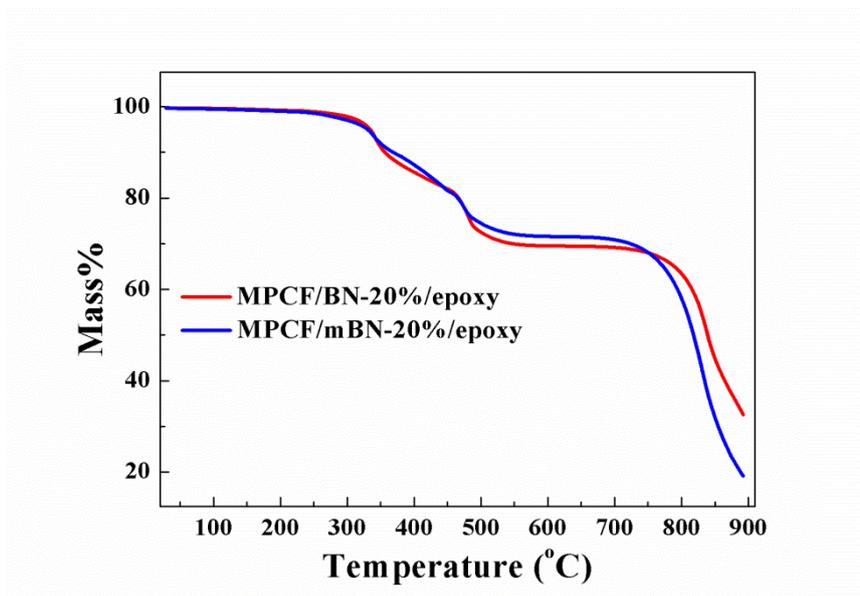
Results and discussions

The TGA for pristine BN and mBN are presented in Fig.2 (a). mBN in the blue line has a clear mass loss in the temperature range from 200 to 350 °C due to the addition of KH550 whose boiling point is about 217 °C while the pristine BN does not this in such a temperature range. This difference of the mass loss shows that KH550 is existed in mBN particles after simple surface treatment. We also do TGA for

MPCF/BN-20%/epoxy and MPCF/mBN-20%/epoxy composites and it can be found the weight ratio between epoxy composite and MPCF is about 1:3 since the weight loss until 500 °C for both two composite is around 25%.



(a)



(b)

Figure 2. (a) TGA for pristine BN and mBN from 30 to 800 °C. (b) TGA for two composites, MPCF/BN-20%/epoxy and MPCF/mBN-20%/epoxy from 30 to 900 °C.

The SEM images of surface fractures for two kinds of composites with 20% volume fraction of filler's incorporation are presented in Fig.3. It can be found that

MPCF can be adhered by epoxy with the fillers but the qualities of adhesion are different between MPCF/BN-20%/epoxy and MPCF/mBN-20%/epoxy. Compared with the Fig.3 (a) and Fig.3 (c), the adhesion in the boundary region shown in Fig.3 (c) of MPCF/mBN-20%/epoxy are better than that of MPCF/BN-20%/epoxy since a denser structure can be figured out in Fig.3 (c). Meanwhile, observing Fig.3 (b) and Fig.3 (d), there are many little points remained in the surface of mBN in Fig.3 (d). Instead, the surface of BN is flat and clean in MPCF/BN-20%/epoxy as shown in Fig.3 (b). These little points remained after epoxy's breaking indicate that mBN has formed modified bonding with epoxy matrix after treated by the KH550 coupling agent. But the clean and flat surface of BN infers a weak bonding with epoxy. Therefore, it can be viewed that MPCF/mBN-20%/epoxy has better interfacial adhesion among MPCF, mBN and epoxy than that of MPCF/BN-20%/epoxy.

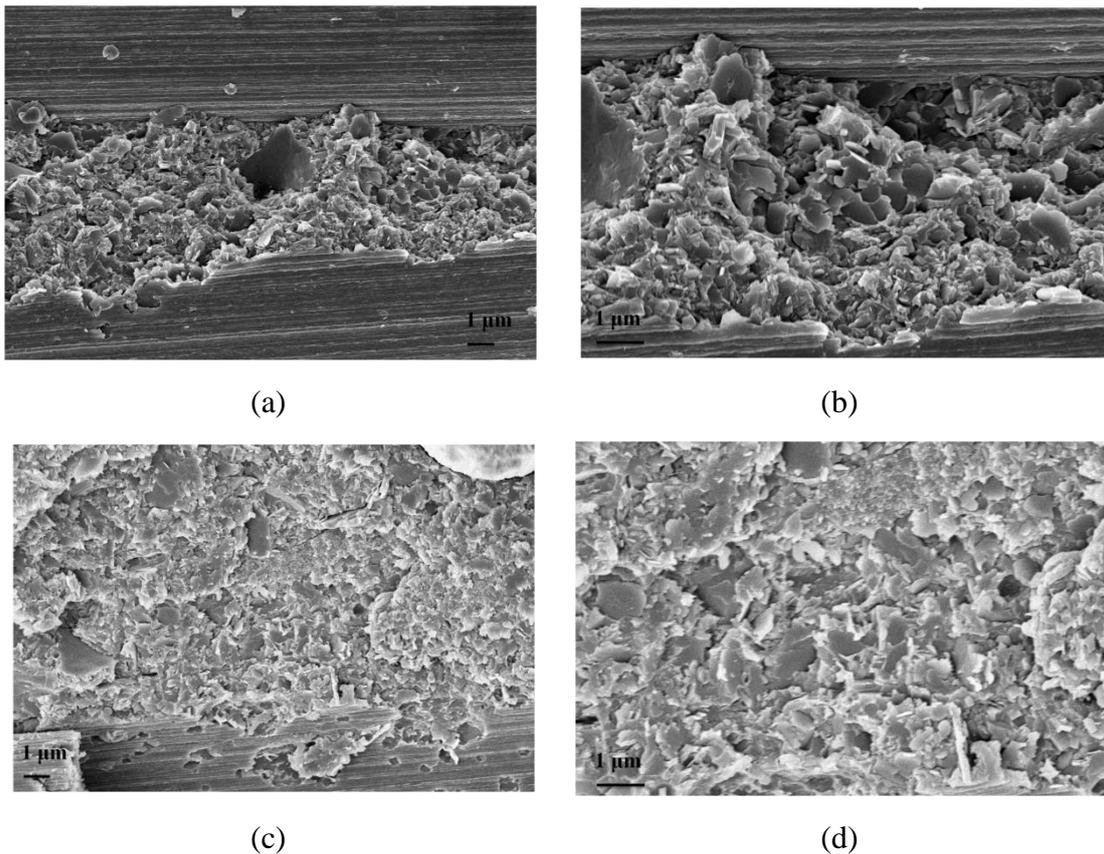


Figure 3 (a) and (b) are SEM images for MPCF/BN-20%/epoxy and (b) is an enlarged image. (c) and (d) are SEM images for MPCF/mBN-20%/epoxy and (d) is an enlarged

one.

The thermal properties of MPCF/BN/epoxy and MPCF/mBN/epoxy composites with different volume fractions are measured by a Nanoflash method and the results including α and TC are presented in the Figure 4 (a) and (b). For the reference, the TC of MPCF/epoxy without BN fillers is 4.9 W/(mK). The calculated enhancement of TC (TCE%) for MPCF/mBN/epoxy composites compared with the MPCF/BN/epoxy composites is follows the relations (1) and the results are listed in the Table 1.

$$TCE\% = \frac{(TC_{MPCF/mBN/epoxy} - TC_{MPCF/BN/epoxy})}{TC_{MPCF/BN/epoxy}} * 100\% \quad (1)$$

Table 1 TCE% for MPCF/mBN/epoxy composites compared with MPCF/BN/epoxy composites with different volume fraction of fillers.

Volume fractions of mBN	5% (vol)	10% (vol)	15% (vol)	20% (vol)
TCE%	2.7%	22.9%	56.6%	78.9%

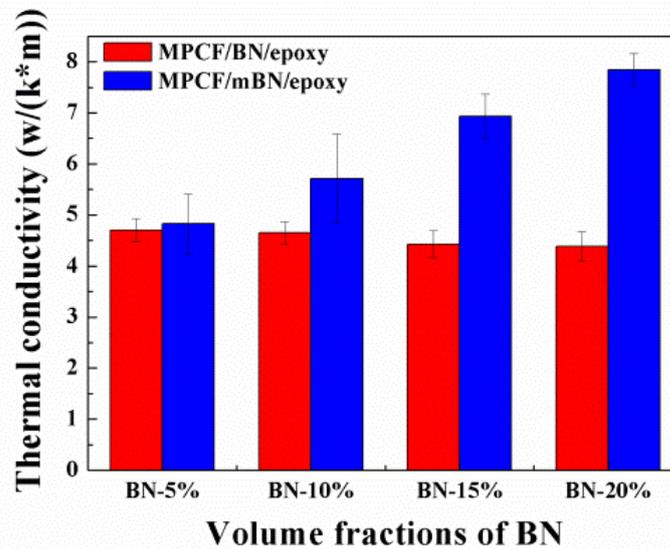


Figure 4. TC of MPCF/BN/epoxy and MPCF/mBN/epoxy composites with different volume fractions of fillers.

Results in Fig.4 illustrate that simply mixing BN with epoxy is not an effective

way for MPCF epoxy matrix composites since the thermal properties of MPCF/BN/epoxy do not show obvious improvement as f_{BN} 's increasing. Instead, only after the surface treatment by KH550 for BN particles, the thermal properties of MPCF/mBN/epoxy increase obviously as the $f_{\text{m-BN}}$ increasing, especially the TC for MPCF/mBN-20%/epoxy (7.9 w/(m*K)) whose TCE% is 79% compared with MPCF/BN-20%/epoxy (4.4 w/(m*K)) as presented in Table 1. In order to explain the function of the surface treatment by KH550, it needs to retrospect the phonon's heat transfer mechanism in epoxy matrix. As inorganic fillers, the dispersion of BN particles in epoxy matrix creates huge amounts of interfaces as shown in the SEM images. The adhesion of BN particles and epoxy in the interfaces greatly determines interfacial thermal resistances which control phonon scattering and affect TC of the composite. According to the thermal property theory, the interfacial thermal resistance is generally classified in two ways: (1) by thermal contact resistance and (2) by thermal boundary resistance. Firstly, the thermal contact resistance usually results from several mismatches in the surface conditions of BN, epoxy and MPCF layers. Meanwhile, it also depends on how well these three materials are bonded together and the contact areas among three. Then the other part, the thermal boundary resistance, is caused by the mismatch in the vibration harmonics of phonons and/or the flow of phonons among BN, epoxy and MPCF. In the MPCF/BN/epoxy composites, with the volume fraction of BN increasing, these mismatches become gradually hard to adjust and simultaneously voids caused by the mechanical incompatibilities between BN and epoxy increase.^[26] As the results of these factors, the TC's enhancement for MPCF/BN/epoxy is limited despite increasing BN's incorporation. In contrast, the results show that mBN treated by KH550 can reduce these mismatches and consequently modify the interface contacting. This modification can be explained by the formation of silanol groups (Si-OH) attached with the surface hydroxyls of BN. With the formation of chemical bonding with epoxy matrix, the adhesion among epoxy, mBN and MPCF can be ameliorated and effectively decrease the possibility of interfacial scattering of phonons. Besides, with the small molecules of KH550 on the surfaces of BN particles, some voids perhaps can be filled and improve the

compatibility between matrix and fillers which makes the packing more efficient especially for the case of 20% volume fractions. Therefore, the surface treatment by KH550 is an effective method for enhancing the TC of MPCF/mBN/epoxy composites.

In order to further study the function of KH550 from the perspective of epoxy molecular motions, the temperature dependence of TC for MPCF/BN-20%/epoxy and MPCF/mBN-20%/epoxy composites are investigated from 30 to 120 °C. Before discussing the behavior of TC's temperature dependence, the study of the glass transition of epoxy matrix for these two composites is conducted by DMA to detect the interaction between fillers and matrix which helps to understand the behavior of TC's temperature dependence more straightforward.

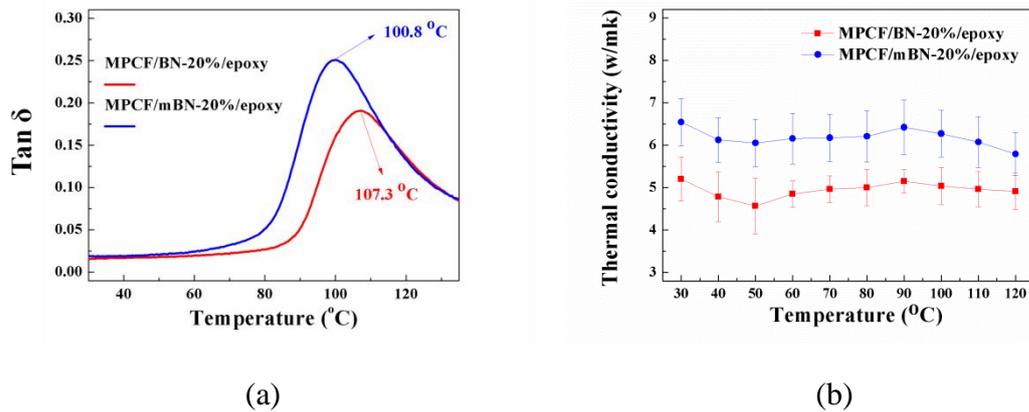


Figure 5. (a) Dynamic mechanical analysis for MPCF/BN-20%/epoxy and MPCF/mBN-20%/epoxy composites. (b) Temperature dependence of TC for the two composites.

The temperature of $\tan \delta$ peak for the composite is generally viewed as the glass transition for its polymer matrix in the DMA measurement. As illustrated in Fig.5 (a), T_g for MPCF/BN-20%/epoxy are 107.3 °C which is about 7 °C higher than that of MPCF/mBN-20%/epoxy (100.8 °C) due to the addition of KH550. The shift of T_g in a composite is usually governed by two primary factors: the dispersion state of the fillers and the modified mobility of polymer in the interfacial region which is also known as the interaction between matrix and fillers. After KH550's treating, the

organic-silance functions as the plastic which increases more free volume around the epoxy and consequently reduces the energy of the mobility for polymer segments. Besides, due to the base chemical property, KH550 may also influence the cross-linking degree of the epoxy which may also reduce Tg. Thus, the glass transition for MPCF/mBN-20%/epoxy happens in advance at lower temperature than that of MPCF/BN-20%/epoxy.

The temperature dependence of TC for two composites is presented in Fig.5 (b) and the effect of temperatures on TC can be divided into three stages: firstly TC for the two composites decreases when the temperature increases from 30 to 50 °C but later increases a bit as the temperature reaches 90 °C. Differently, the change of TC for MPCF/mBN-20%/epoxy is less obvious than that of MPCF/BN-20%/epoxy as the temperature increasing. As shown by the equation (1), the obtained TC is dominated by three factors: thermal diffusivity, heat capacity and density. Thermal diffusivity and heat capacity are related with the phonons scattering and epoxy's segment motions. When the temperature increases from 30 to 50 °C, phonon's scattering increases because of the thermally activated process which causes the thermal diffusivity decreases clearly, but in this procedure, the heat capacity does not increase clearly since epoxy's segment's motion is slight in such a temperature range which can also be found from DMA in the Fig. 5 (a). Thus, the reducing of thermal diffusivity directly leads to TC's decrease. But as the temperature increasing, in the second stage from 50 to 100 °C, the heat capacity of composites increases which can not only offset the decrease of thermal diffusivity but also increase the global TC. TC's temperature dependence for two composites can reflect the cooperative molecular motions involving both intra- and intermolecular interactions and we use the activation energy as a tool to understand this procedure.^[27] The variation of activation energies for two composites can be obtained by fitting TC at different temperatures (50 to 100 °C) with the well-known Arrhenius relation expressed in equation (3) and the fitting results are presented in the Fig.6.

$$TC \propto \frac{1}{T} \exp \left[-\frac{E_a}{k_b T} \right] \quad (3)$$

Where E_a is activation energy, T is the absolute temperature and K_b is the Boltzmann constant. These parameters are significant to differentiate the nature of various conduction mechanisms. When the values of TC are applied from 50 to 100 °C, E_a for MPCF/BN/epoxy and MPCF/mBN/epoxy are obtained as 0.0527 and 0.0458 eV as shown in the inset column graph of Fig.6. A smaller E_a of MPCF/mBN/epoxy indicates that silanol groups (Si-OH) of KH550 in the interface region of mBN and epoxy enter into the epoxy cross-linking network which not only helps to form better adhesion but also fill some voids in the composite. This better adhesion and denser packing enable to decrease phonon scattering and stabilize the TC of the composite so that weaken the temperature effect compared with pristine BN's reinforcement.

As the temperature reaches over 100 °C, the values of TC for MPCF/BN/epoxy and MPCF/mBN/epoxy decrease as the temperature increasing since composites start the glass transition. In this stage, the TC of MPCF/mBN/epoxy reduces more than that of MPCF/BN/epoxy for the increase of free volume around epoxy's segments after modified by KH550 as we have mentioned in DMA part. More free volumes make the epoxy segments easier to move which may increase thermal motion and consequently decreases TC for the composites. Thus in the third stage from 100 to 120 °C, TC for two composites both decrease a bit and the value for MPCF/mBN/epoxy decreases more. But generally, two composites' TC do not reduce a lot and both of them show a good stability on TC during the whole temperature changing procedure due to the excellent thermal property of MPCF.

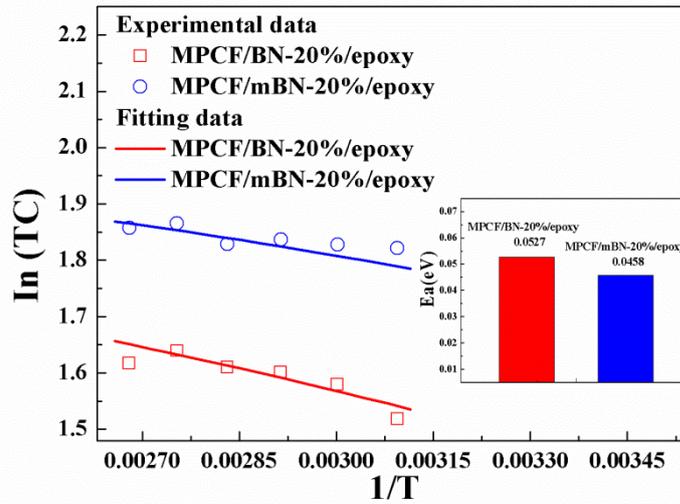


Figure 6 ln (TC) verse temperatures for MPCF/BN/epoxy and MPCF/mBN/epoxy composites with 20% volume fraction fillers' incorporation. The inset graph shows the fitted active energies for two composites.

Conclusion

The MPCF/BN/epoxy composite achieves a high TC with a low volume fraction of BN. Moreover, after an effective surface treatment by KH550 coupling agent, MPCF/mBN-20%/epoxy's TC has improved further and reaches 7.85 w/(m*K) which has 79% increase compared with MPCF/BN/epoxy without surface treatment. Such a high enhancement on TC is attributed to the reinforced adhesion among mBN, epoxy and MPCF achieved due to the silanol groups in KH550 which enables to reduce the thermal resistance as well as increase efficient packing between fillers and polymer matrix. Additionally, MPCF/BN-20%/epoxy and MPCF/mBN-20%/epoxy display a good stability on TC as the measurement temperature increasing because of the excellent thermal property of MPCF, before glass transition (< 100 °C), TC of MPCF/mBN-20%/epoxy is more stable to temperature change due to the KH550's surface treatment.

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