Materials Chemistry and Physics 274 (2021) 125151

Contents lists available at ScienceDirect

Materials Chemistry and Physics

journal homepage: www.elsevier.com/locate/matchemphys

Thermal and dielectric properties of epoxy resin filled with double-layer surface-modified boron nitride nanosheets

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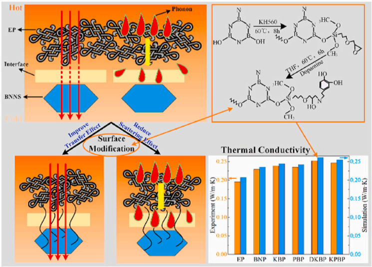
HIGHLIGHTS

- Grafting DA at the end of KH560 is firstly proposed to functionalize BNNSs.
- Thermal conductivity and electrical properties of DKBP are enhanced significantly.
- Improvement mechanism is explored by molecular dynamics simulation.
- This method improves transfer effect and reduces scattering effect at the interface.

GRAPHICAL ABSTRACT



ARTICLE INFO

Keywords:
Epoxy resin
Thermal conductivity
Boron nitride nanosheet
Double-layer surface modification
Interface

ABSTRACT

Owing to the rapid development of advanced power system and modern micro-electronic devices, exploiting composites with excellent thermal conductivity and dielectric properties has become increasingly important. In this work, a double-layer surface modification method of grafting dopamine (DA) at the end of 3-glycidoxypropyl-trimethoxysilane (KH560) is firstly proposed to modify the surface of boron nitride nanosheet (BNNS), for improving the thermal conductivity and dielectric properties of epoxy resin (EP). Besides, the thermal conductivity of EP composites with double-layer surface-modified BNNSs and its improvement mechanism is explored with molecular dynamics (MD) simulation from micro level. The results illustrate that thermal conductivity of EP composites doped with double-layer modified BNNSs by grafting DA at the end of KH560 (DKBP) is 0.252 W/(m·K), 129% higher than EP. Moreover, this double-layer surface modification method of DKBP is beneficial to enhance the heat transfer effect and reduce scattering effect at the interface, which is the essential reasons for improving thermal conductivity of composites. Meanwhile, DKBP composite also presents superior dielectric properties. This work provides a promising idea to develop new insulating materials with high thermal conductivity.

1. Introduction

Due to the excellent adhesion, chemical stability and mechanical properties, epoxy resin (EP) is extensively applied as dielectric materials in manufacturing power equipment and electronic devices [1,2]. With rapidly increasing demand and consumption of electric energy and electron devices, large capacity and miniaturization of power equipment and integration of electronic devices have become the mainstream

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https://doi.org/10.1016/j.matchemphys.2021.125151
Received 14 June 2021; Received in revised form 28 July 2021; Accepted 14 August 2021

direction of power field [3]. However, the heat accumulation problem is more serious in large-capacity facilities and integrated devices, leading to the sharply temperature rise. Insulation failures of solid dielectrics under high temperature can cause permanent damage to the materials and shorten the service life of power equipment or electronic devices [4,5]. Therefore, exploring new insulating materials with high thermal conductivity is extremely essential.

In recent years, there are many researches on improving the thermal conductivity of polymer composites, among which the introduction of thermally conductive nanofillers is an effective method [1,2,6]. Boron nitride nanosheets (BNNS) with good insulation properties, thermal conductivity and chemical stability become nowadays among the most promising and popular inorganic nanomaterials [6]. The thermal conductivity of composites can be enhanced to some degree by improving the preparation technology, but the effect is not expectant [7,8]. Researchers pointed out that the surface modification of nanofillers can significantly improve the thermal conductivity of composites [9–11]. Weng selected 3-aminopropyltriethoxysilane (KH550) and 3-glycidoxypropyl-trimethoxysilane (KH560) to functionalize the BNNSs, and results indicated that the thermal conductivity of BNNS@KH560/EP composite with 20 vol% BNNSs dosage was 0.442 W/(m·K), which was 81% higher than that of pure EP. Meanwhile, BNNS@KH560/EP composites also showed better thermal and mechanical properties than that of BNNS@KH550/EP composites [12]. Qu used poly(dopamine) (PDA) to modify the surface of BNNSs, and a high thermal conductivity of 0.501 W/(m·K) was obtained at 20 wt% PDA@BNNSs content, reaching 2.63 times of the value for polymer (0.176 W/(m·K)) [13]. Recently, double-layer modification of BNNSs was explored to further improve the thermal properties of composites, such as grafting the KH560 at the end of dopamine (DA) and grafting the aminopropylisobutyl polyhedral oligomeric silsesquioxane at the end of KH560 [14,15], which can remarkably improve the thermal conductivity of composites. Although there are many researches on modification methods, the modification mechanism of improving thermal conductivity is not comprehensive.

Interface is the key factor to determine the thermal properties of the thermally conductive polymer composites, and is the main barrier for thermal conduction due to heat loss during heat transfer across interfaces [16,17]. However, it is difficult to understand the interface properties of nanocomposites through experimental methods and hypothetical models. The development of molecular simulation technology provides a powerful tool to directly investigate the role of interface properties in the nanocomposites from the micro perspective [18,19]. In Refs. [20–22], the interfacial molecular models of EP nanocomposites were established by using molecular dynamics method, and the interface density and binding energy were used to characterize the interface characteristics. The results explained that the bonding strength of composites with surface treatment was higher than that of untreated one and the density near interface was also bigger, which had positive effect on the thermal properties of composites. Besides [23,24], supplemented the parameters of cohesive energy density, free volume fraction and molecular chain mobility to further describe the interfacial compatibility and built the correlation between the interface and properties of composites. Therefore, molecular simulation has made great progress in the field of nanocomposite interface, but there are few reports on the correlation between surface modification and thermal conductivity of nanocomposites from micro interfacial characteristics.

In order to enhance thermal conductivity and dielectric properties of EP composites filled with surface-modified BNNSs, the double-layer surface modification method of grafting DA at the end of KH560 is firstly reported to modify the surface of BNNSs. Besides, the corresponding EP composites are prepared as well as the microcosmic morphology, thermal conductivity and dielectric properties are measured. Moreover, the thermal conductivity of composites is also investigated by molecular dynamics (MD) simulation and the improvement mechanism of thermal conductivity for composites with double-layer surface modification is demonstrated from micro interfacial characteristic.

2. Experiment and simulation details

2.1. Materials

The BNNSs (500 nm) are supplied by Shanghai Naiou Nano Technology, Co., Ltd. (China). γ-glycidoxypropyltrimethoxysilane, dopamine hydrochloride and tris-(hydroxymethyl)-aminomethane (Tris) are obtained from Shanghai Macklin Biochemical Technology Co. Ltd. (China). EtOH is obtained from Tianjin Kemiou Chemical Reagent Co., Ltd. (China) while diglycidyl ether of bisphenol A (E−51, Eponex 1513), methyl hexahydrophthalic anhydride (MTHPA) and other reagents are provided by Changzhou Runxiang Chemical Co., Ltd. (China).

2.2. Surface modification of BNNSs

The preparation procedure of DA-KH560-BNNS (DKB) is described in Fig. 1(a). Firstly, 0.2 g KH560 and 2 g BNNSs are added into the EtOH/H_2O solution (EtOH: $H_2O = 1:1$, wt/wt) and stirred by magnetic stirrers with 800 rad/min (at 70 °C for 8 h), then filtrated and washed by EtOH and dried at 70 °C for another 24 h, finally to obtain KH560-BNNS (KB). After that, 2 g KB is poured into the DA/THF solution (0.5 wt% DA) and stirred by magnetic stirrers with 800 rad/min (at 60 °C for 6 h), then filtrated and washed by EtOH and dried in vacuum oven at 70 °C for 24 h, finally to obtain DKB.

The preparation process of KH560-PDA@BNNS (KPB) is described in Fig. 1(b). Firstly, 0.8 g dopamine hydrochloride is added to the 300 mL deionized water at pH = 8.5 (adjusted by Tris). Then, 2 g BNNSs are mixed with above solution and stirred by magnetic stirrers with 800 rad/min (at 60 °C for 12 h), finally to obtain PDA@BNNS (PB). Subsequently, 0.2 g KH560 is added to the above solution and stirred by magnetic stirrers with 800 rad/min (at 60 °C for 8 h). Then the obtained mixture is filtered off, washed with EtOH and dried in vacuum oven at 70 °C for 24 h, finally to obtain KPB.

2.3. Fabrication of composite materials

The preparation of epoxy composites is carried out through the following procedures. Firstly, 60 g EP is heated at 60 °C for 0.5 h and then BNNSs or modified BNNSs (3 wt% EP and curing agent) are added into it. The mixture is then under the ultrasound at 60 °C for 1 h to uniformly disperse the BNNSs. After that, some additives such as methyltetrahydrophthalic anhydride as curing agent (80 wt% EP) and 2,4,6-tris(dimethylaminomethyl)phenol as curing accelerator (1 wt% EP) are added. This mixture is stirred by dispersion emulsifier thoroughly and then poured into the mold by the injector. Then the curing process is successively operated in vacuum at 80 °C for 1 h, 120 °C for 2 h and 150 °C for 2 h. Finally, the molds containing the cured polymers are cooled to 30 °C.

2.4. Characterization

Thermogravimetric analysis (TGA) is carried out on a STA 449-F5 (NETZSCH C Corp.) under N_2 atmosphere. Fourier transform infrared spectro-scopy (FTIR) is carried out using a NICOLET iS10 (Thermo Fisher Scientific). Scanning electron microscope (SEM) is employed to observe the morphology of the epoxy composites by using an Apreo S (Thermo Fisher Scientific). The thermal conductivity of epoxy composites is measured on the DRE-III (Xiangtan xiangyi instrument Co. Ltd) at 30 °C. The temperature dynamic distribution of epoxy composites is tested by using a E6-XT thermal infrared imager (FLIR Systems). The dielectric loss of composites is measured by a high-frequency LCP digital bridge (TH2826; Changzhou Tonghui Electronic Co. Ltd.). Alternating current (AC) breakdown strength of composites is measured using voltage breakdown equipment (HJC-100KV, Huabotech Co., Ltd). (The

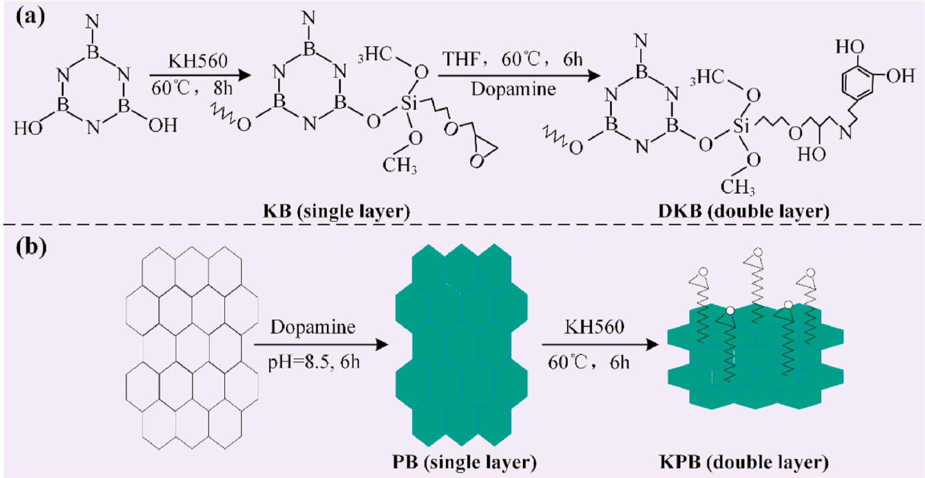

Fig. 1. Schematic diagram for fabrication process of BNNS. (a) DKB, (b) KPB.

experimental details and relevant data sets are shown in section 2 of Supplemental Files).

2.5. Molecular models

The molecular models are built by Materials Studio (MS) software [25,26]. Diglycidyl ether of bisphenol A (DGEBA) and MTHPA are selected as EP molecule and cross-linking agent molecule, shown in Fig. 2 (a). The cross-linked EP models shown in Fig. 2 (b) is built by cross-linking reaction of DGEBA and MTHPA. DA and KH560 are used to modify the surface of BNNSs and the size of BNNS model is 28.4 Å × 29.5 Å, including 175 N atoms and B atoms, as shown in Fig. 2 (c). The pristine BNNSs or surface-modified BNNSs are put into DGEBA and MTHPA models, then the BNNS/EP models are built after cross-linking reaction, as shown in Fig. 2 (d). According to experiments, six models are built, including pure EP, BNNS/EP, KH560-BNNS/EP, PDA@BNNS/EP, DA-KH560-BNNS/EP and KH560-PDA@BNNS/EP, named as EP, BNP, KBP, PBP, DKBP, and KPBP, respectively. Detail information of EP and BNNS/EP nanocomposite models is shown in Table S2 and Fig. S4 of Supplemental Files. In order to avoid the effect of periodic boundary, the vacuum layers (20 Å) are added to the up and bottom of models shown in Fig. 2 (e) when the parameters of interfacial characteristics are calculated.

2.6. Simulation details

All simulations are carried out by large-scale atomic/molecular massively parallel simulator (LAMMPS) and MS software, and the Dreiding force field is employed to calculate [27]. Besides, thermal conductivity of models is calculated by reverse non-equilibrium MD method [28]. Furthermore, the fix/ave/chunk and compute msd/chunk commands are used to calculate radial density distribution (RDD) and radial mean square displacement (RMSD), respectively. Moreover, interface binding energy (IBE) are counted by Equation (1) [26]. Vibrational density of state (VDOS) is widely adopted to analyze the thermal conduction for nanomaterials and the thermal transport behavior at the interface between nanofillers and matrix. It can be calculated from the Fourier transform of velocity autocorrelation function (VACF), as shown in Equation (2) [29]. Then the phonon coupling constant (S) is used to describe the overlap degree of VDOS between EP

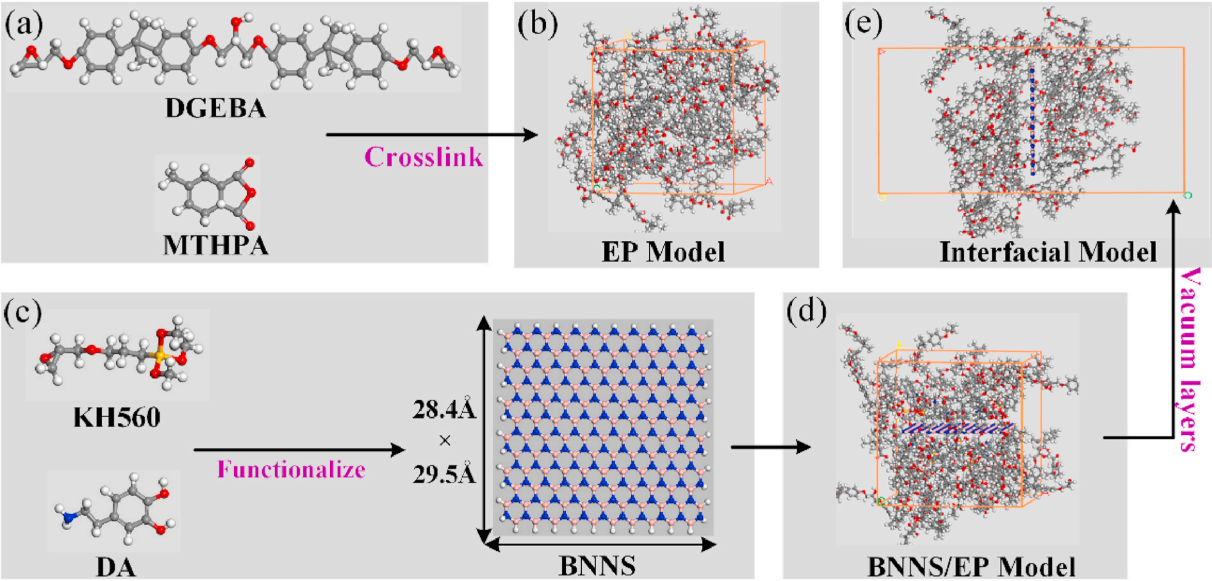

Fig. 2. Molecular models. (a) EP molecule and cross-linking agent molecule, (b) cross-linked EP models, (c) DA, KH560 and pristine BNNS, (d) BNNS/EP models, (e) calculation model of interfacial characteristics.

and BNNSs, as shown in Equation (3).

$$IBE = E_{\text{composite}} - (E_{\text{BN}} + E_{\text{EP}}) \tag{1}$$

where $E_{\text{composite}}$, E_{BN} and E_{EP} are the potential energy of BNNS/EP composites, BNNSs and EP, respectively.

$$P(\omega) = \int_0^t V(t) \cdot \cos(\omega t)\mathrm{d}t) \tag{2}$$

$$S = \frac{\int P_{EP}(\omega) P_{BN}(\omega) \mathrm{d}\omega}{\int P_{EP}(\omega) \mathrm{d}\omega \int P_{BN}(\omega) \mathrm{d}\omega} \tag{3}$$

where ω is frequency and $P(\omega)$ is the VDOS at the frequency of ω. $V(t)$ is the velocity autocorrelation function of all atoms and the computational method is shown in Equation (S2) and (S3) of Supplemental Files.

3. Results and discussion

3.1. Surface modification characterization

The results of FTIR are shown in Fig. 3 (a). Pristine BNNS and surface-modified BNNSs all present strong peaks at 1380 cm^{-1} and 800 cm^{-1}, corresponding to the stretching and bending vibration absorption of B–N, respectively [15]. Besides, both KB and DKB show the anti-symmetric stretching vibration peak of Si–O–Si at 1110 cm^{-1} and KH560 epoxy at 860 cm^{-1} [15]. In addition, the stretching vibration peak of amino group appears between 3571 cm^{-1} and 3625 cm^{-1}. The possible reason is that KH560 is grafting BNNS surface and dopamine is grafting KH560 as well. As for PB and KPB, the characteristic peaks of amino hydroxyl group at 3402 cm^{-1} and the asymmetric stretching vibration and symmetric stretching vibration peaks of KH560 methylene group at 2929 cm^{-1} and 2862 cm^{-1} are important [30]. It proves that KH560 is introduced to the surface of BNNS treated by PDA.

Fig. 3 (b) shows the TGA curves of BNNS and surface-modified BNNSs. When the temperature is lower than 150 °C, the weight loss is attributed to the elimination of water and residual solvent. The mass residues of BN, KB, PB, DKB and KPB at 800 °C are estimated to 98.9%, 94.4%, 92.2%, 93.9%, and 89.3%, respectively and correspondingly grafting weight contents of PDA, KH560, KH560-PDA and PDA-KH560 are 4.5%, 6.7%, 5% and 9.6%, respectively. Above results also imply that the surface of BNNSs was treated successfully.

3.2. Morphologies characterization

In order to accurately characterize the dispersity and agglomerate size of BNNSs, the apparent photography and fracture morphology of EP and BNNS/EP composites are obtained, as shown in Fig. 4 and Figs. S5–S7 of Supplemental files. The fracture of pure EP is fluvial shape and the surface is smooth in Fig. 4(a, g). Besides, Fig. 4(b, h) displays the fracture morphology of BNP composite. Pristine BNNSs occur big size agglomerate and there is obvious gaps between BNNSs and EP in BNP composite. It indicates that the BNNSs without surface modification have bad dispersity in EP matrix comparing with BNNSs with surface modification. Moreover, the agglomerate size and gaps of double-layer surface modified BNNSs is smaller than that of single-layer surface modified BNNSs in EP matrix, which means double-layer surface modification method contributes to improving dispersity and compatibility between BNNS and EP. Due to the small interaction between the non-covalent modified BNNSs and EP matrix, composites doping with BNNSs modified by PDA slide from the interface. As shown in Fig. 4(e, k), the agglomerate size and gaps are significantly smaller than that of other BNNSs in EP matrix, indicating that DKB surface modification method can effectively improve the strong interaction and good compatibility between BNNS and EP matrix. The apparent photography and morphology of EP and BNNS/EP composites are displayed in Figs. S5–S6 of Supplemental files.

3.3. Thermal conductivity

The experiment and simulation results of thermal conductivity are consistent, as shown in Fig. 5(a). At the same content of BNNSs, thermal conductivity of double-layer modified composites is higher than that of the composites treated by single-layer modification. Especially, thermal conductivity of DKBP was 0.25 W/(m·K) and 129% higher than that of pure EP (0.19 W/(m·K)). From Fig. 5(b), the bright area of DKBP is bigger than other composites at the same time, which means that its heat dispersion is best. Additionally, hot-spot temperature distribution in infrared imaging images at different times also reflects heat emission capability of the sample, as shown in Fig. S8 of Supplemental Files. The hot-spot temperature of DKBP is lowest at the same time among all simples.

The calculation formula of interfacial thermal resistance (R_I) and thermal conductivity (λ) is simplified [31], as shown in Equations (4) and (5). From Fig. 5(c), the interfacial temperature gradient of DKBP is lowest, that is, interfacial thermal resistance is smallest, so the thermal conductivity of DKBP is best. The results further demonstrate that double-layer modification method of grafting DA at the end of KH560 is beneficial to reduce the interfacial thermal resistance. In summary, above results manifest that double-layer surface modification method of grafting DA at the end of KH560 is effective to enhance thermal conductivity of EP composites.

$$R_I = \frac{\Delta T}{J} \tag{4}$$

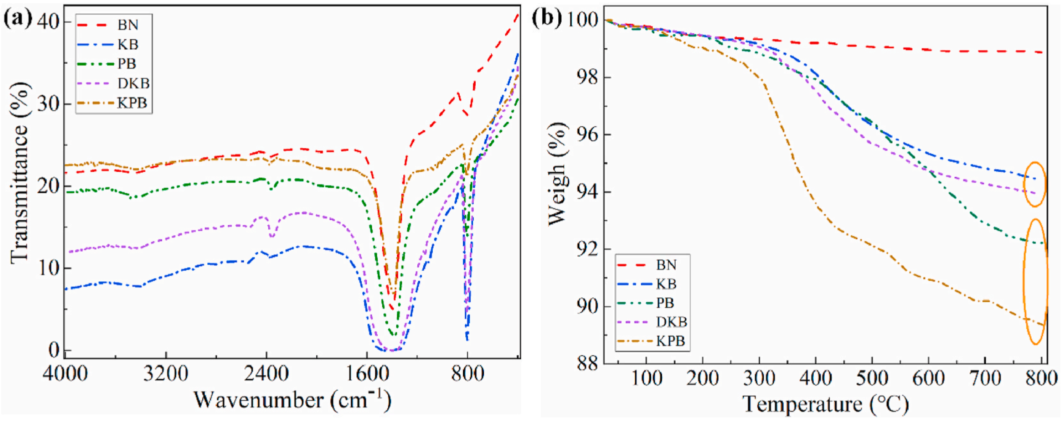

Fig. 3. Characterization of surface-modified BNNSs. (a) Infrared spectra, (b) Thermal weight loss curves.

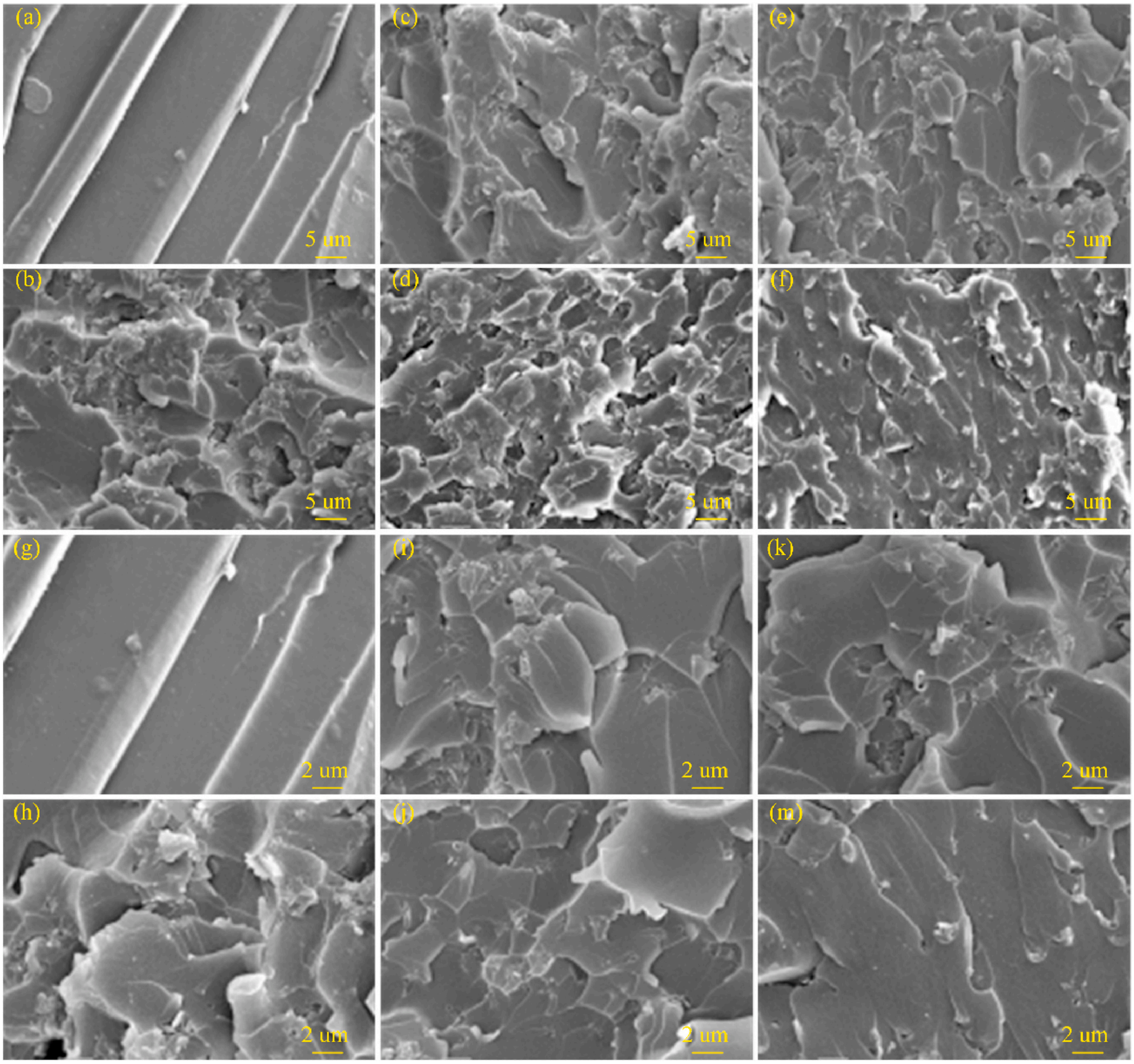

Fig. 4. Fracture morphologies of nanocomposites. (a, g) EP, (b, h) BNP, (c, i) KBP, (d, j) PBP, (e, k) DKBP, (f, m) KPBP.

$$\lambda = \frac{d}{S \times R_I} \tag{5}$$

where ΔT is the interfacial temperature gradient, J is stable heat flow, S is the section area that is perpendicular to the direction of heat flow and d is the model thickness.

3.4. Interfacial effects

The interface is a new phase with the thickness and structure that varies with thermally conductive fillers, significantly different from the polymer matrix. Therefore, interface is the main influential factor for thermal conduction. In order to further understand the effect mechanism of surface modification on thermal conductivity of BNNS/EP composites, the interface parameters are calculated, as shown in Fig. 6.

RDD along the z-axis of composite models is shown in Fig. 6(a). The prominent part of the curve is the mass density of BNNS and nearby this position. RDD of matrix is generally higher than that of pure EP in the vicinity of BNNS. Besides, with the increase of the distance from BNNS, RDD decreased to some extent and finally it is close to that of pure EP. This fluctuation region is the interfacial thickness. It can be seen that the interfacial thickness of DKBP is biggest and the RDD of DKBP smoothly changes, which is beneficial to reducing the phonon scattering caused by structural mutation and enhancing the phonon coupling. From Fig. 6(b), IBE of DKBP nanocomposite model is 912 kcal/mol, which is 137% of the composites without surface modification and higher that other modified composites, indicating that the interfacial adhesion is better. Meanwhile, the larger interfacial bonding strength is conducive to the orientation of molecular chain at the interface and reduce the interspace (proved by Table S3 in Supplemental Files). Moreover, the enhanced IBE of composites contributes to reducing the surface energy of nanofillers, which can improve the mean free path of phonon and phonon coupling.

Molecular chain motion status of composite models is shown in Fig. 6 (c). The RMSD is lower in the near range to BNNS, while the RMSD in the far range to BNNS is basically same as that of pure EP. Besides, RMSD of DKBP composite at the vicinity of BNNS is significantly lower than that of other composites, which indicates that the interfacial interaction is larger and the motion space is smaller. The low RMSD can reduce the phonon scattering at interface caused by molecular chain vibration. Additionally, the phonon coupling constants between EP and BNNSs are shown in Fig. 6(d). The value of S between EP and DKB is 0.0331, higher than other composites, which manifests that the coupling degree of phonon VDOS between EP and DKB is better. The higher coupling degree of phonon VDOS can reduce the phonon scattering at interface

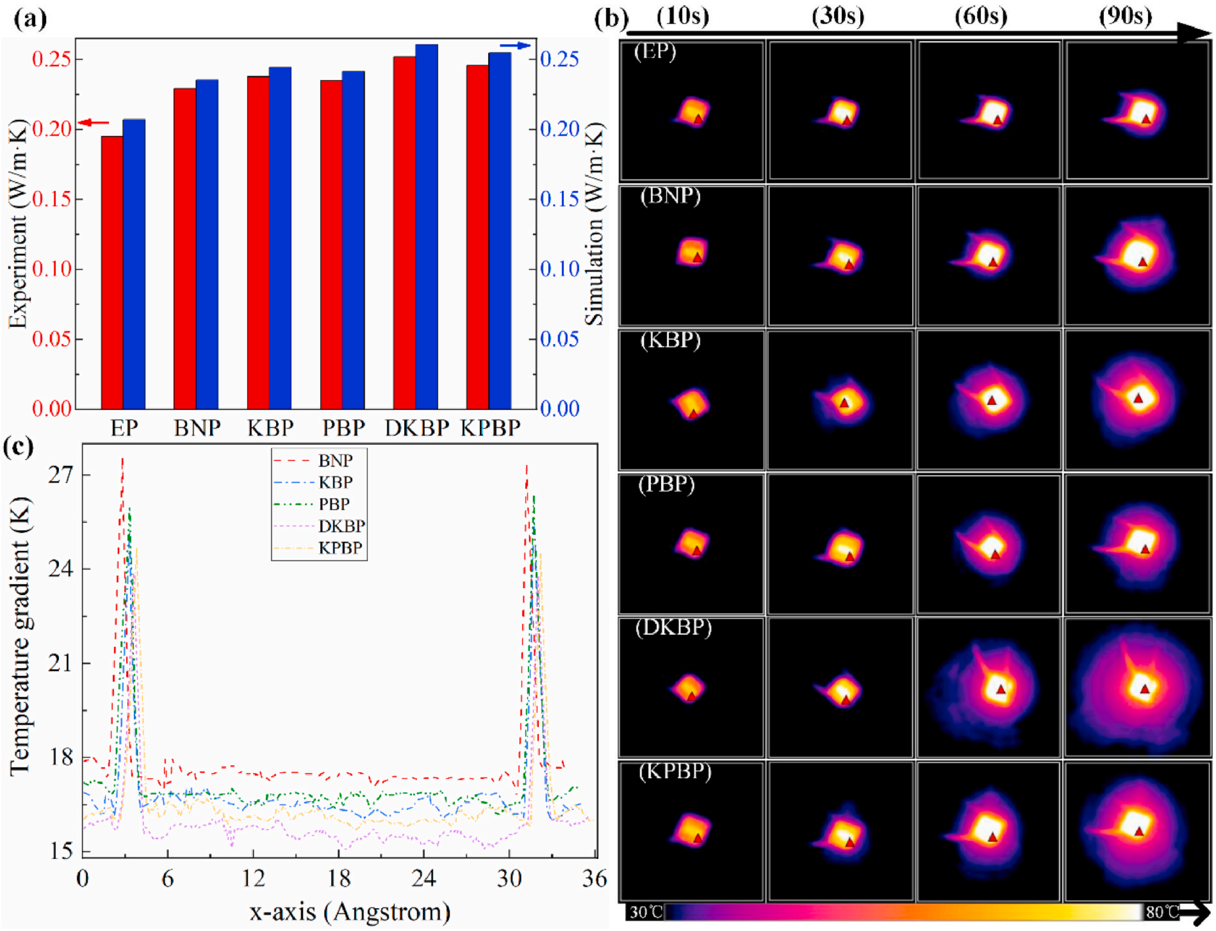

Fig. 5. Thermal conductivity properties of BNNS/EP nanocomposites (a)Thermal conductivity, (b) Infrared thermogram, (c) Temperature gradient.

caused by the difference of phonon vibration frequency.

3.5. Mechanism of surface modification

According to the analysis of interfacial parameters, the roles and effects of the interface are mainly manifested from two aspects: transfer effect and scattering effect, as shown in Fig. 7. Transfer effect means that the interface acts as a thermal bridge between polymer matrix and thermally conductive fillers, maintaining the continuity of the thermal path between matrix and fillers. Scattering effect explains that the interface can scatter the carriers of heat transfer and reduce the mean free path, which slows down heat transfer between the two phases. Therefore, in order to improve the thermal conductivity of composites by introduction of surface-modified nanofillers, enhancing transfer effect, as shown in Path 1, and reducing scattering effect, as shown in Path 2, at interface are the key guiding ideas.

The method of grafting DA at the end of KH560 is proposed to modify the surface of BNNSs in this paper, which is effective to enhance the thermal conductivity of BNNS/EP composites. The reasons for it are as follows. On the one hand, KH560 grafting on BNNSs with covalent bond will enhance the interfacial interaction force. Grafting DA at the end of KH560, leads to producing three hydroxide radicals, which can form hydrogen bonding with EP matrix to enhance the interaction force (proved by Table S4 in Supplemental Files). The strong interaction force and uniform interfacial structure contribute to ensuring the continuity of thermal path, as shown in Path 1. On the other hand, covalent bond modification can improve the coupling degree of phonon vibrational spectral between two phases. KH560 and DA have good affinity with EP matrix, maintaining phonon vibrational spectral and reducing phonon scattering, which is beneficial to ensuring the efficiency of heat transfer, as shown in Path 2. Therefore, this modification method can enhance phonon transfer and reduce phonon scattering, achieving to improve the thermal conductivity of BNNS/EP composites significantly. To sum up, in order to improve the thermal conductivity of composites by introducing nanofillers with surface modification, the variety and length of functional groups and its interaction with polymer matrix should be considered, which can determine the interface properties and thermal conductivity.

3.6. Dielectric properties

The dielectric properties of BNNS/EP composites are shown in Fig. 8. From Fig. 8(a), the dielectric constant of pure EP decreases with the increase of frequency and the change trend of composites is same. Because the dielectric relaxation frequency of dipole polarization and interface polarization is small, some dipole polarization and interface polarization fail to occur when the frequency becomes bigger, which weakens the degree of polarization. Moreover, due to the interaction between BNNS and EP, the directional movement and transport of polar groups in the dielectric materials are restricted under electric field, so dielectric constant of BNNS/EP composites is lower than that of pure EP. But the dielectric constant of DKBP composite is lower than that of other BNNS/EP composites. The main reasons for it are that double-layer covalent modification is more beneficial for enhancing interface bonding strength, resulting in the increase of steric hindrance, so that the polarization of polar groups and molecular chain rearrangement is more difficult to occur. The dielectric loss (tan δ) curves of composites is shown in Fig. 8(b). The interfacial interaction between BNNS and EP

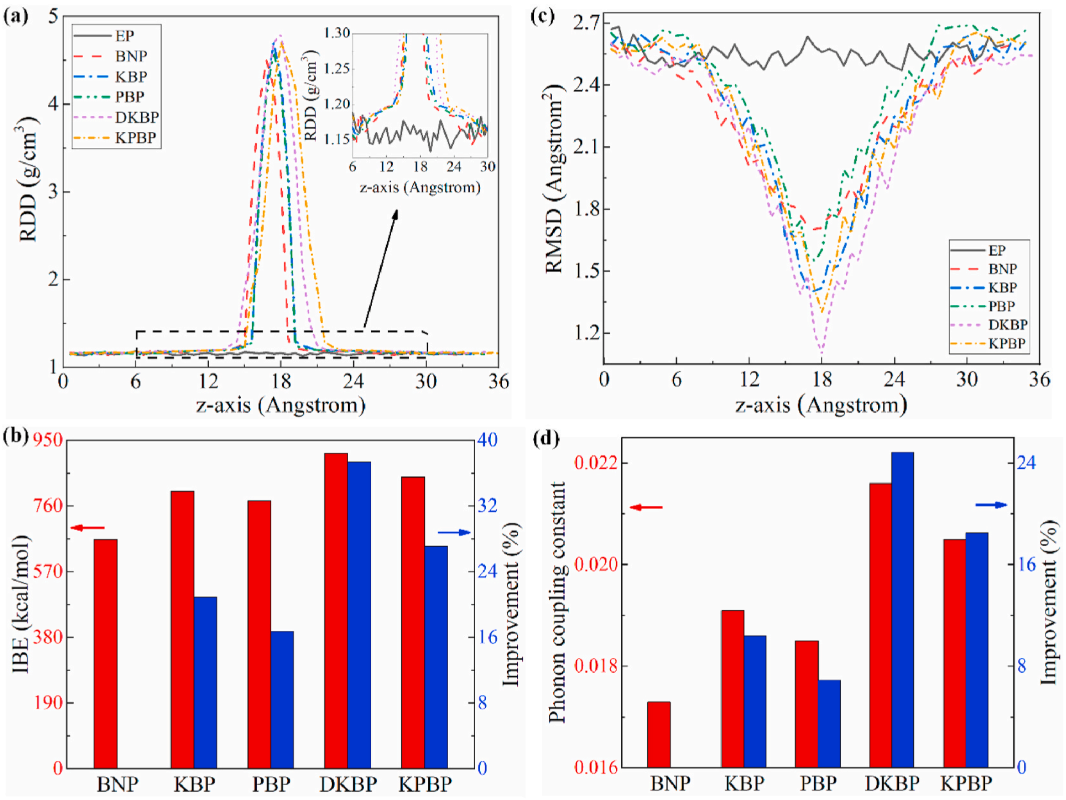

Fig. 6. Interfacial characteristic of BNNS/EP nanocomposites. (a) RDD curves, (b) RMSD curves, (c) IBE, (d) S.

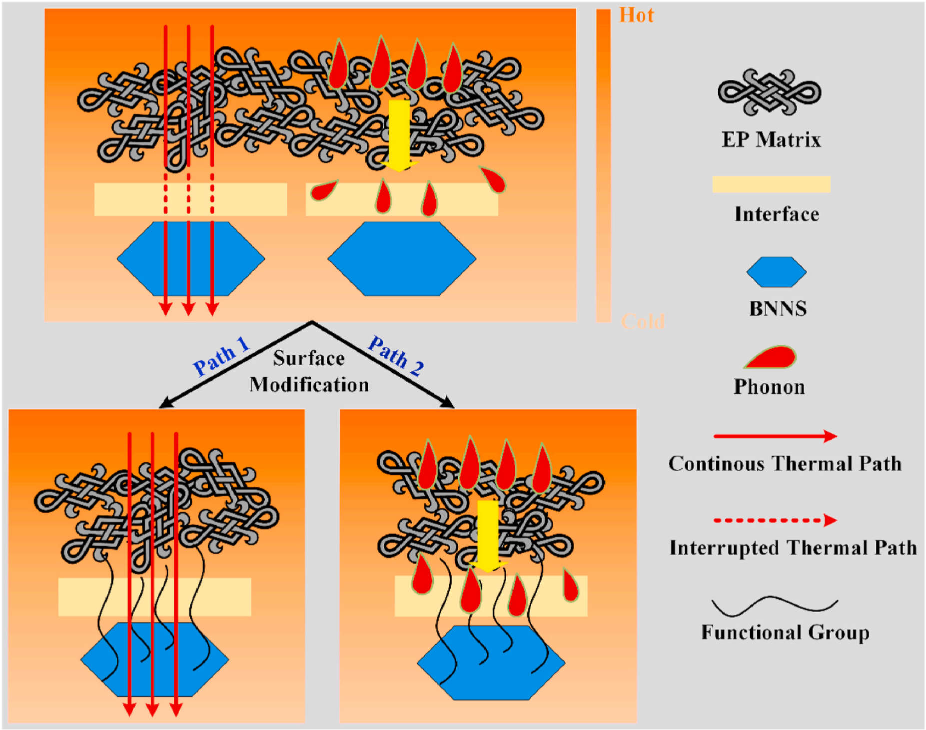

Fig. 7. The illustration of surface modification mechanism of BNNS/EP composites.

limits the movement of dipole and reduces the path of charge, so that $\tan\delta$ of BNNS/EP composites is lower than that of pure EP. Although surface modification may introduce new polar groups, double-layer covalent modification can effectively enhance the interfacial bonding strength, which contributes to the decrease of dielectric constant and loss of DKBP.

Weibull distributions are employed to describe breakdown strength of composites [32], as shown in Fig. 9 and Table 1. The breakdown

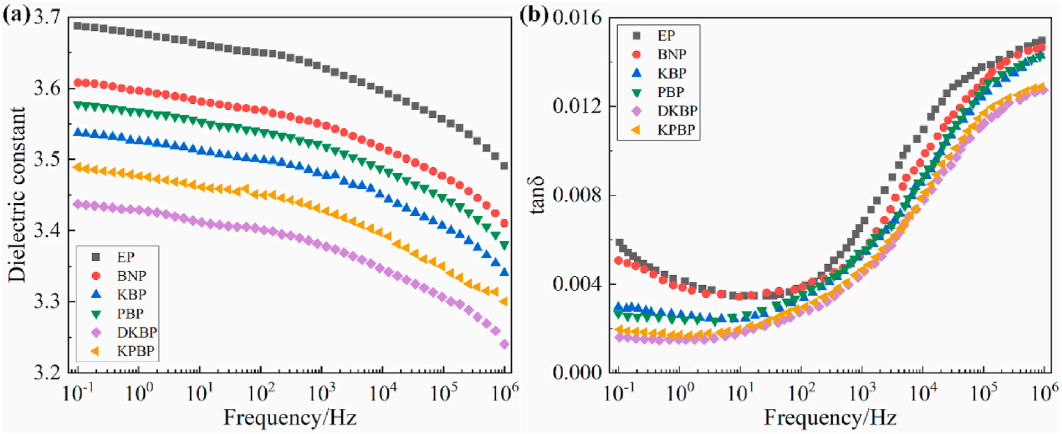

Fig. 8. Dielectric properties of BNNS/EP nanocomposites. (a) Dielectric constant, (b) tan δ.

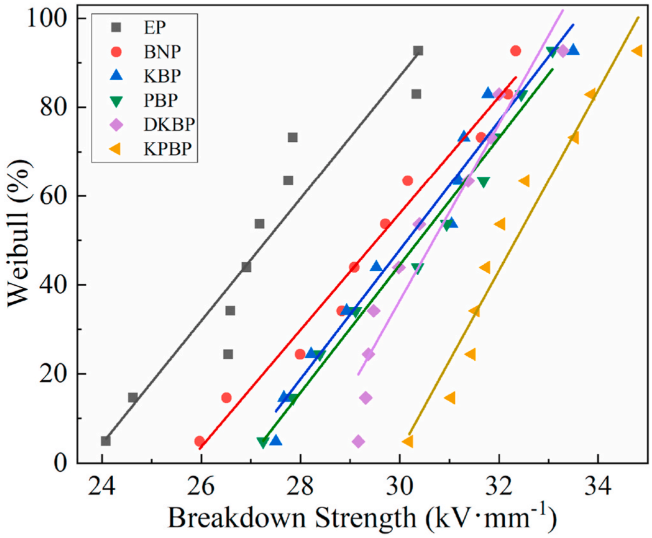

Fig. 9. AC breakdown strength of BNNS/EP nanocomposites.

Table 1
Scale and shape parameters of Weibull distribution for six samples.

Simples	Weibull parameters	
	α(kV/mm)	β
EP	28.15	15.83
BNP	30.42	16.21
KBP	30.96	18.07
PBP	31.20	17.58
DKBP	31.59	23.30
KPBP	32.94	24.28

strength of BNNS/EP composites is higher than that of pure EP (28.15 kV/mm), and the breakdown strength of the DKBP and KPBP composites is greater than 31.5 kV/mm, which is higher than that of other composites. Because the BNNSs modified by double-layer surface treatment can effectively inhibit the injection of space charge and polarization effect of internal dipole. Meanwhile, β of DKBP and KPBP composites is bigger than that of other composites, the reason for which is that the even dispersion of surface-modified nanofillers leads to greater data. The results indicate that EP doped with double-layer surface-modified BNNSs can still maintain excellent dielectric properties.

4. Conclusion

The double-layer surface modification method of grafting DA at the end of KH560 is firstly proposed to enhance thermal conductivity and dielectric properties of EP composites as well as its improvement mechanism of thermal conductivity is investigated by MD simulation in this work. The DKBP composites with 3 wt% DKB presents high thermal conductivity, 129% higher than pure EP. Moreover, the interfacial parameters of micro structure, bonding strength, molecular chain vibration and phonon coupling degree explain that surface modification can be conducive to improving the interface properties and the interface properties have decisive effect on thermal conductivity. Besides, the surface modification method of DKB enhances the transfer effects that can ensure the continuity of heat transfer as well as reduces the scattering effects that can maintain the efficiency of heat transfer at the interface, which contributes to improving the thermal conductivity of composites. Meanwhile, DKBP composites also displays excellent dielectric properties that is superior to other composites. This work can guide the development of thermally conductive materials used in large capacity and miniaturization of power equipment and integration of electronic devices.

CRediT authorship contribution statement

Jiacai Li: Conceptualization, Methodology, Software, Validation, Formal analysis, Investigation, Data curation, Writing – original draft, Writing – review & editing, Visualization. **Shengtao Li:** Conceptualization, Software, Formal analysis, Resources, Supervision, Project administration, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

Computational resources and data analysis for Materials Studio and LAMMPS software packages are provided by High-Performance Computing Centre in State Key Laboratory of Electrical Insulation and Power Equipment (SKLEIPE) of Xi'an Jiaotong University (XJTU).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.matchemphys.2021.125151.

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