Review of high thermal conductivity polymer dielectrics for electrical insulation

Meng Xiao\textsuperscript{1,2} and Bo Xue Du\textsuperscript{1}

\textsuperscript{1}School of Electrical Engineering and Automation, Tianjin University, Tianjin 300072, People’s Republic of China
\textsuperscript{2}Economic and Technical Research Laboratory, Jinan Power-supply Company, Jinan 250012, Shandong, People’s Republic of China
\textsuperscript{✉} E-mail: tjuxiaomeng@tju.edu.cn

Abstract: Traditional insulation material is thermally insulating and has a low thermal conductivity. The miniaturisation and higher power of electrical devices would generate lots of heat, which have created new challenges to safe and stable operation of the grid. The development of insulating materials with high thermal conductivity provides a new method to solve these problems. The improvement of thermal conductivity would increase the ability to conduct heat and greatly reduce the operating temperature of the electrical equipment, which could reduce the equipment size and extend service life. On the other hand, inorganic thermally conductive particles and the improved thermal conductivity may have great effect on thermal breakdown. In this study, the factors affecting the thermal conductivity of dielectric polymer composites were explored. Intrinsic thermal conductive polymer and particle-filled thermally conductive composites were discussed. Effect of thermal conductivity, shape, size, surface treatment of the particle and prepare process on thermal properties of the composites were illustrated. This study focused on the electrical and thermal properties of thermally conductive epoxy, polyimide and polyethylene composites. Tracking failure caused by thermal accumulation is a typical thermal breakdown phenomenon. The performance of the resistance to tracking failure was studied for these composites. The results showed that thermal conductive particles improved the resistance to tracking failure. Finally, application of thermally conductive epoxy in electrical equipment was discussed.

1 Introduction

Polymer dielectric is thermally insulating and has a thermal conductivity under 0.5 W/(m·K) [1]. The heat generated by overload operation or partial discharge could lead to the temperature rise of insulating materials, which would cause the loss of dielectric performance gradually. With the development of new materials and technology, the voltage level and capacity of the transformer were improved constantly and then the overheat problems became more serious [2, 3]. Moreover, a considerable portion of fault in transformer is believed to be due to thermal accumulation. The development of materials with high thermal conductivity has provided a new approach to reduce operating temperature and then prolong the service life [4, 5].

2 Intrinsic thermal conductive polymer dielectrics

Generally, there are two ways to improve the thermal conductivity of the polymer. Usually, crystal has relatively high thermal conductivity due to its highly ordered structure. So change in the molecule structure of the polymer to form crystal-like structure could improve the thermal conductivity. The crystal-like structure could reduce phonon scattering, so the ability of conducting heat would be improved for the polymer.

Hitachi and Hitachi Chemical have recently developed a novel method for increasing the thermal conductivity of epoxy resin by controlling its higher-order structure [6]. Fig. 1 is a schematic of the higher-order structure of the developed epoxy resin. It shows three main features: (i) microscopic anisotropy with crystal-like structures of oriented mesogens in di-epoxy monomers, (ii) macroscopic isotropy induced by randomly oriented domains of crystal-like structures and (iii) indistinct boundaries between the crystal-like regions, each connected with an amorphous region by covalent bonds. The highly ordered structure would be expected to suppress phonon scattering so that the resin should have high thermal conductivity. Another property resulting from the highly ordered structure is high flexibility, which is important for resins used in manufacturing. The amorphous regions improve mould and process ability but tend to reduce thermal conductivity. Thermal conductivities of epoxy are in the range 0.25–96 W/(m·K), which is ~1.5–5.0 times greater than the conventional ones and have the highest thermal conductivities of all macroscopically isotropic organic insulating substances [0.46–0.51 W/(m·K)]. Fig. 2 shows atomic force microscopy (AFM) and transmission electron microscopy (TEM) images of a conventional resin and of the developed resin. The TEM image shows clearly that the latter has a lattice structure, whereas large domains with sizes of several micrometres are seen in the AFM image.

Studies have shown that the orientation stretching of the polymer can effectively reduce the phonon scattering, and thus significantly improve the thermal conductivity of the polymer [7, 8]. Nysten studied the thermal conductivity of semi-crystalline oriented polymers and proposed the thermal-transfer mechanisms along and across the chains by measuring the thermal conductivity parallel and perpendicular to the polymer chains as a function of temperature. When polyethylene (PE) is stretched to more than 25 times, thermal conductivity of the direction parallel to the molecular chain is 13.4 W/(m·K) at room temperature [9]. The thermal conductivity of unidirectional gel-spun PE fibre-reinforced composites have been measured parallel (K∥) and perpendicular (K⊥) to the fibre axis from 15 to 300 K. The thermal conductivity of gel-spun PE fibre at 300 K is 38 and 0.33 W/(m·K) along and perpendicular to the fibre axis. The axial thermal conductivity is exceptionally high for polymers and this high value arises from the presence of a large fraction of long (>50 nm) extended chain crystals in the fibre [10, 11].

Some researchers studied the thermal conduction anisotropy in polymers by reviewing currently available theories and materials. The improvement of thermal conductivity would increase the ability to conduct heat and greatly reduce the operating temperature of the electrical equipment, which could reduce the equipment size and service life. On the other hand, the high thermal conductivity may have great effect on thermal breakdown. In this study, the factors affecting the thermal conductivity of dielectric polymer composites were explored. Intrinsic thermal conductive polymer and particle-filled thermally conductive composites were discussed. This study focused on the electrical and thermal properties of thermally conductive epoxy, polyimide and polyethylene composites. Tracking failure caused by thermal accumulation is a typical thermal breakdown phenomenon. The performance of the resistance to tracking failure was studied for these composites. The results showed that thermal conductive particles improved the resistance to tracking failure. Finally, application of thermally conductive epoxy in electrical equipment was discussed.
experimental methods for studying oriented polymers. The anisotropic thermal conductivity and diffusivity of oriented polymers originate from the difference between the thermal energy transport mechanisms parallel and perpendicular to their molecules [12]. Kato prepared the liquid crystal acryl compounds which the molecular directions were aligned by the rubbing method. The compounds were polymerised by ultraviolet irradiation and the free-standing aligned films of 200 μm thickness were made. The relation between the thermal conductivity and the aligned molecular direction of the films was investigated. The homogeneous film showed the largest magnitude of the thermal conductivity at the direction along the molecular long axis [0.69 W/(m·K)], which was 3.6 times greater than that of poly(methyl methacrylate). It is indicated that the additional thermal transmission effect, which the increase of the thermal conductivity may be induced, would exist in the twisted films [13, 14].

In addition, an ultra-stretched polymer prepared by a special spinning process also has a high thermal conductivity. Thermally conductive polymer nanowire arrays were prepared by an improved nanoporous template wetting technique. The thermal conductivities of the fabricated high-density PE (HDPE) nanowire arrays with diameters of 100 and 200 nm are about two orders of magnitude higher than their bulk counterparts. The estimated thermal conductivity of a single HDPE nanowire is as high as 26.5 W/(m·K) at room temperature [15]. Cao fabricated high-quality ultra-drawn PE nanofibres with diameters of 50–500 nm and lengths up to tens of millimetres. The thermal conductivity of the nanofibres could be ~104 W/(m·K), which is almost the half of the pure metals. The high thermal conductivity is attributed to the restructuring of the polymer chains by stretching, which improves the fibre quality toward an ‘ideal’ single crystalline fibre [16]. Wang explained the thermal conduction mechanism at the level of individual molecules. When large amounts of heat are transported through a molecule, a crucial process in molecular electronic devices, energy is carried by discrete molecular vibrational excitations. They studied heat transport through self-assembled monolayers of long-chain hydrocarbon molecules anchored to a gold substrate by ultrafast heating of the gold with a femtosecond laser pulse. When the heat reached the methyl groups at the chain ends, a non-linear coherent vibrational spectroscopy technique detected the resulting thermally induced disorder. The leading edge of the heat burst travelled ballistically along the chains at a velocity of 1 km/s. The molecular conductance per chain was 50 pW/K [17].

The intrinsic thermal conductive polymer by means of highly ordered molecular chain arrangement improved the ability to conduct heat without sacrificing their own electrical insulating properties and mechanical properties. Such thermally conductive polymers are potentially useful as heat spreaders and could supplement conventional metallic heat-transfer materials, which are used in applications such as solar hot-water collectors, heat exchangers, electronic packaging and insulation devices. However, there are few applications of intrinsic thermal conductive polymers, especially in the electrical equipment. The manufacturing process is complex and difficult and the cost is very high, which these factors limit the application of these polymers.

3 Particle-filled thermal conductive polymer dielectrics

A widely used approach is to use composites of polymer and fillers. When the fillers were added in polymer, the heat could be conducted through the fillers (usually have high thermal conductivity) instead of the polymer, which can significantly improve the thermal conductivity of the composite [18]. The thermal conductivity of the particles, the loading level, the shape and size of the particle, the particle dispersion and orientation, the interface status and other factors would have a significant effect on thermal conductivity of the composite.

3.1 Polymers

The common polymers used in electrical engineering are epoxy (Epoxy), silicone rubber, polyimide (PI), polypropylene (PP), low density PE (LDPE), HDPE and other new modified polymers. As the matrix of the composites, the polymer should have the following properties: excellent insulating, mechanical and moulding properties; relatively low thermal expansion coefficient and low dielectric constant; high filling fraction of inorganic particles; and cheap and variety of sources.

Thermal conductivity of the composite depends on the number of heat conduction paths and interfacial thermal resistance. If the amount of the added inorganic particles is small, even the particles can be uniformly dispersed in the matrix, the increase of thermal

---

Fig. 1 Schematic of the approach to synthesising macroscopic isotropic resin with high thermal conductivity [6]

Fig. 2 Structure of a developed epoxy resin observed by AFM and TEM [6]
conductivity is not obvious because there is no full contact between the particles thus heat will also be transferred by the matrix. With the further addition of the inorganic particles, when the filling amount exceeds a certain threshold, the heat could be conducted by the particles and then the thermal conductivity of the composite system will increase rapidly [19].

To form the heat conduction path in the matrix is critical for improving the thermal conductivity. The kind, shape, size, surface modification and distribution of the particle would have great effect on forming the paths.

3.2 Thermal conductivity of the particle

As the composites would be used as dielectric polymer, the conductive particles such as metal, carbon and graphite are not suitable for electrical insulation. Usually, inorganic particles such as aluminium oxide (Al₂O₃), magnesium oxide, silicon oxide, zinc oxide, boron nitride (BN), Al nitride (AIN), silicon nitride and silicon carbide are used as fillers in the composite. The difference between the thermal conductivity of different types of inorganic particles is very large. Thermal conductivity of diamond could be 20000 W/(m·K), whereas the thermal conductivity of the alumina is 30 W/(m·K) and the crystalline silicon is only 3 W/(m·K). At the same content, the thermal conductivity of the composite system is usually higher with filling the higher thermal conductivity of particles [20]. However, it does not show a significant improvement when the intrinsic thermal conductivity of the fillers is >100 times the thermal conductivity of the polymer matrix.

In addition to the type and amount of matrix and fillers would affect the thermal properties of the composite, the following aspects would also have great effect on thermal, electrical and mechanical properties.

3.3 Particle shape and size

Bernd [21] had added different fillers to PP and found that different kinds of fillers had little effect on the thermal properties of the composites, while thermal conductivity is mainly determined by the distribution of thermally conductive fillers. When filler content is high, effect of particle size on thermal conductivity would become weak, because the effective thermal conductive paths have been formed in the polymer and the impact of particle size could be ignored.

Mixing different kinds, sizes and shapes of particles in the polymer could decrease the voids between the particles, which the heat conduction path is easier to be formed and then the relatively higher thermal conductivity could be obtained as shown in Fig. 3. Xu et al. [22] used AlN whiskers (and/or particles) and/or silicon carbide whiskers as fillers(s) and polyvinylidene fluoride (PVDF) or epoxy as matrix. The highest thermal conductivity of 11.5 W/(m·K) was attained by using PVDF, AlN whiskers and AlN particles. When AlN particles were used as the sole filler, the thermal conductivity was highest for the largest AlN particle size but the porosity increased with increasing AlN particle size. Al₂O₃ and AIN with different sizes were used alone or in combination to prepare thermally conductive polymer composites. The use of these hybrid fillers was found to be effective for increasing the thermal conductivity of the composite, which was probably due to the enhanced connectivity offered by the structuring filler. At a total filler content of 58.4 vol.%, the maximum values of both thermal conductivities in the two systems were 3.402 and 2.842 W/(m·K) [23]. Sanada [24] investigated thermal conductivity of composites with multi-walled carbon nanotubes (MWN Ts) and alumina nanoparticle and spherical alumina fillers. The results showed that the addition of MWN Ts to the matrix lead to a large increase in thermal conductivity of the composites.

3.4 Surface treatment

Inorganic particles and polymer have completely different properties so the interface compatibility between the two materials is weak. If the particle surface was not treated, it is easy to produce agglomerates in polymer and then affect the dispersion of the inorganic particles, which would reduce the thermal conductivity of the composite. In addition, the filler–matrix thermal contact resistance could cause large phonon scattering here with resulting in the decrease of thermal conductivity.

The thermal conductivity of AlN particle epoxy–matrix composite was increased by up to 97% by silane surface treatment of the particles prior to composite fabrication. The increase in thermal conductivity is due to decrease in the filler–matrix thermal contact resistance through the improvement of the interface between matrix and particles. At 60 vol.% silane-treated AlN particles only, the thermal conductivity of epoxy–matrix composite reached 11.0 W/(m·K) [22]. To improve the thermal conductivity of BNNT filled epoxy composite, admicellar polymerisation was used to coat polystyrene and polymethyl methacrylate on the BN surface to improve the interfacial adhesion in the composite. The results show that the admicellar treatment led to improved wettability of epoxy resin on the treated surface. Thermal conductivity of the composite increased from 1.5 W/(m·K) for untreated BN to 2.69 W/(m·K) when the admicellar-treated BN was used, indicating improvement in the interfacial adhesion between BN and epoxy resin in the composite [32–34]. Huang found that the thermal conductivity enhancement of the composites is not only dependent on the type and physicochemical nature of the modifiers but also dependent on the filler loading. The composites with BN treated by the silane incapable of reacting with the epoxy resin show the most effective enhancement of the thermal conductivity [35].

3.5 Preparation process

When applied electric and magnetic fields in the process of preparing the composites, the distribution of particles may be changed in the polymer. Thus, the composite could obtain a high thermal conductivity at a certain direction. Yan prepared thermal conductive silicone rubbers filled with BN micro-particles assisted

Fig. 3 Schematic of PE/hybrid-BN composites to illustrate microstructures
with electric field assisted curing, and studied the electric field effect [36]. The results indicate that aligned conductive networks have formed between the electrodes under either AC or DC electric fields. Furthermore, the ‘c’ axis of BN particles was found to orient along with the electric field and the orientation degree under AC was higher than that with DC. Under the AC electric field (50 Hz) of 11.0 kV/mm, the thermal conductivity with 20% loading of BN increases by 250% compared with that prepared without electric field. A facile technique was developed to modify BN nanosheets with iron oxides in order to fabricate highly oriented polysiloxane/BN nanosheet composite films under a magnetic field and their thermal properties were evaluated according to the orientation of BN [37]. The results revealed that the modified BN nanosheets were aligned either horizontally or vertically to the film plane, depending on the direction of magnetic flux with high anisotropy. The transmittance and thermal conductivity of the nanocomposite films were improved due to the orientation of the BN nanosheets inside the polymer matrix.

4 Thermal and electrical properties of polymer composite

4.1 Epoxy composite

Epoxy is widely used as insulating materials in electric devices with its excellent insulating properties in spite of its weak ability of thermal conduction. So far, most studies on high thermal conductivity composites had been limited to improve thermal conductivity with lower filler content, while there are few studies on their electrical properties.

The study of electrical properties is mainly resistivity, dielectric constant, electrical breakdown strength and other characteristics. Breakdown strength is significantly affected by the addition of inorganic filler, which depends on the filler kinds, loading, size, shape, surface treatment and preparation process. The electric field distortion and enhancement are caused by the difference in dielectric constant (AC) or electrical conductivity (DC) between the inorganic fillers and polymer matrix. Therefore, to obtain high breakdown strength composites, fillers having similar electrical characteristics as the polymer matrix should be chosen [35]. Andritsch et al. [38] found that combining BN nanofiller (70 nm, Fig. 4 Relation between the sample temperature and the concentration with the time of the applied voltage and the interval of 5 ms. The ambient temperature was 19.3°C [4]

Fig. 5 Typical discharge images and tracking patterns with the different concentration and the pulse frequency [47]

a Typical images of discharge process
b Typical carbonisation pattern in tracking process
10 wt%) with micro-filler (500 nm, 1.5 μm, 5 μm, 10 wt%) gives rise to an increase in the DC breakdown strength of an epoxy composite. Wang fabricated Epoxy/BN composites and a value of 12.3 W/(mK) is the highest thermal conductivity obtained for composite [39]. It was found that nanocomposite has higher breakdown (BD) strength than neat epoxy, whereas micro-filler loaded composites are all lower in BD strength than neat epoxy. Optimum BD performance is obtained if void space is reduced by certain methods such as co-dispersion of different size fillers and addition of nano-filler. Li found that the nano–micro-composite was higher in both BD strength and partial discharge (PD) resistance than the micro-composite. It should be noted that the addition of nano-fillers would provide an excellent approach that can increase the dielectric BD strength and time of micro-filled epoxy composites [40].

As a kind of high thermal conductivity composite, high thermal conductivity is its major advantage. However, few studies have considered the relation between the improved thermal conductivity and the thermal breakdown phenomenon. The insulating materials used in power equipment are usually subjected to severe thermal stresses with sustained heat. A lot of faults in power system are caused by the thermal accumulation [41, 42]. Tracking failure caused by thermal accumulation is a typical thermal breakdown phenomenon [43–46].

The thermal graphs in Fig. 4 represent the thermal phenomena of the tracking. Thermal distribution is detected until tracking failure occurs from back side [4]. The thermal graph analysis is used to analyse the thermal distribution from discharges at discharge area. The ambient temperature was 19.3°C and the results of the thermal graphs with different concentrations under 200 Hz are shown in Fig. 4. It is observed from the back thermal distributions that the maximum temperature decreases with increasing the concentration at different stages. The temperature of undoped sample is the highest. However, the temperature becomes lower with increasing the concentration. BN particles improve the thermal conductivity of the composites. The enhanced characteristics are due to thermal conduction network formed inside of the composites. Here, it is proposed that BN particles allow a larger thermal energy flux to pass through the material, and then the thermal energy dissipates to the air. It is considered that higher concentration of BN particles results in more thermal conduction networks; therefore, the improved thermal properties can prolong the time to tracking failure.

The carbonised points induced by the discharge energy are increasingly accumulated on the sample surface, which finally causes tracking failure between the electrodes, as shown in Figs. 5a and b [47]. The carbonisation patterns with the different concentration are obtained after the experiments. The colour and the area of tracking pattern are clearly different with different samples. The notable changes in the tracking process are observed. Much more dark-colour points can be observed in the tracking pattern of the pure epoxy surface than those with higher BN concentration.

4.2 PI composites

With the advantages of low-temperature tolerance, radiation resistance and excellent dielectric properties, PI that serves as a typical kind of engineering polymer material has been widely used in different fields such as the aerial, nuclear, microelectronic industry, turn-to-turn insulation and turn-to-ground insulation of inverter-fed motors. However, PI film is a thermally insulating material and has a low thermal conductivity.

Many papers have investigated the effect of adding nanoparticles on the resistance to corona discharge of the PI composites [48–51]. Zhang found that the honeycomb structure appeared on the aged surface of original film, and crack and some inorganic like substance exhibit on the aged surface of the nano-inorganic hybrid film. After square pulse corona ageing, the surface of corona region was more scorching than that of after power frequency ageing [52, 53]. A sol–gel process has been developed to prepare PI/Al2O3 hybrid films with different contents of Al2O3. The dimensional stability, thermal stability and mechanical properties of hybrid PI films were improved obviously by an addition of adequate Al2O3 content, whereas dielectric property and the elongation at break decreased with the increase of Al2O3 content. Surprisingly, the corona-resistance property of hybrid film was improved greatly with increasing Al2O3 content within certain range as compared with pure PI film. Especially, the hybrid film with 15 wt% of Al2O3 content exhibited obviously enhanced corona-resistance property, which was explained by the formation of compact Al2O3 network in hybrid film [54].

Research of PI film are mostly focused on nano-filled composite and corona or partial discharge performance. The author investigated the effects of the improved thermal conductivity on tracking process of PI/BN composite. Typical tracking patterns after tracking failure is shown in Fig. 6 [55]. A carbon conducting path is formed on the surface between the electrodes. The colour and the area of tracking pattern are clearly different. The accumulated heat would break the C–H bond of the polymer, which will cause the carbon products decomposed on the surface. When sufficiently intensive discharges last for a considerable time, the carbon products, with some parts of carbonised channel, rapidly form on the dielectric surface. Under the same condition, much more dark-colour points can be observed on the sample with lower mass fraction. It is indicated that the formation of
carbonised products can be restrained by the BN particles. The colour and area of tracking pattern are gradually darkening as the pulse frequency increases. So, the damage condition under higher frequency is much more serious.

The carbonised points decomposed by the discharge energy are increasingly accumulated on sample surface, which is the major reason for tracking failure. The isotherm distribution after applying the discharge for 10 s is shown in Fig. 7. It is observed that the maximum temperature decreases with increasing the mass fraction. The temperature of pure sample is the highest and the maximum temperature at 150 Hz is 420°C, whereas only 180°C when the mass fraction is 80 wt%. As can be seen from Fig. 7, the isotherm distribution of the sample with lower mass fraction is more intensive. Under the same discharge condition, the intensive distribution represents the maximum temperature is much higher and more heat is accumulated in the centre instead of dissipated to the surrounding area. It can be inferred that the BN particles can greatly decrease the maximum temperature. The increase of thermal conductivity improves the ability to conduct heat, so the heat will be conducted out quickly and the effect of thermal accumulation will be reduced, which would make the composite with high mass fraction show a relatively sparse distribution. It can be seen that the high pulse frequency has a great effect on thermal accumulation. When the discharge is 150 Hz, the maximum temperature of the 80 wt% sample is 210°C, whereas only 90°C with 50 Hz. It can also be seen that the isotherm under

![Fig. 7 Relation between the isotherm distribution and the mass fraction after applying different pulse frequencies for 10 s [55]](image)

![Fig. 8 Relation between the maximum temperature on the sample surface and the concentration after applying pulse at 200 Hz for 100 s](image)

![Fig. 9 Relation between the time to dielectric breakdown and the filler concentrations with the application of different pulse frequency](image)
high-frequency discharge shows an intensive distribution, especially in vicinity of the discharge centre, which indicates that the high-frequency discharge is easier to cause thermal accumulation. More heat is accumulated in unit time with the higher frequency. When the heat is not completely dissipated, the next discharge would produce new quantity of heat. So, the shorter interval between adjacent discharges is easier to generate thermal accumulation. The high-frequency discharge would make the molecules chain broken severely and the carbon conductive path formed more quickly, which means the increase of frequency would make tracking failure easier.

The thermal conductivity is improved by the BN particles, and then the heat generated by the arc discharge could be conducted out quickly. The decomposition of composites in tracking process also releases large amounts of heat which is the major reason for the rise of temperature. The interaction between the filler and PI film would increase the physical and chemical cross-linking points, and the thermal-stable BN filler limited the segmental movement of the composites. Therefore, the doped PI film has good high-temperature resistance and excellent thermal stability [20]. In addition, compared with the PI, the inorganic particles have much larger melting point, so BN particles are not involved in the decomposition process. The released heat of the higher mass fraction sample when tracking happened is much smaller than the lower sample. Therefore, the temperature with higher concentration is much smaller. The addition of BN particles inhibits the effect of thermal accumulation at the discharge area.

4.3 PE composites

PE is widely used as the insulting material in those electric devices because of its excellent insulating properties.

In recent years, many researchers have attempted to improve the thermal conductivity of PE composites via adding high thermal conductivity fillers. Tavman investigated the thermal conductivity of HDPE/Al composites in the range of filler content 0–33% by volume. It showed that at higher particle content, the fillers tended to form agglomerates and conductive paths so that thermal conductivity increased rapidly [56]. Krupa et al. pointed out that the thermal conductivity measurements of composites non-linearly increased with increasing the graphite content. In addition, the thermal conductivity of filled HDPE was higher than that of filled LDPE because of high crystallisation ratio [57].

Most research mainly focus on studying insulating PE composites with good thermal properties, especially high thermal conductivity. Nevertheless, few studies have discussed the relation between thermal conductivity and thermal breakdown.

To show the effect of BN fillers on the thermal distribution of samples, the maximum temperature on the sample surface after applying pulse at 200 Hz for 100 s is represented in Fig. 8. The maximum temperature decreases with increasing filler content in the same discharge conditions. When the mass fraction is 10 wt%, the maximum temperature of PE/hybrid-BN composites is 288.4°C, whereas it is 157.6°C at the concentration of 40 wt%. Moreover, the maximum temperature of PE/hybrid-BN composite is lower than that
of the PE sample coated micro-sized BN particles at the same concentration. When filler concentration was 20 wt%, the maximum temperature of PE/micro-BN composite was 247.8°C, whereas only 219.5°C of PE/hybrid-BN sample. It shows that the addition of hybrid-BN particles is more helpful to inhibit the heat accumulation and reduce the maximum temperature on the sample surface.

Fig. 9 shows the relation between the time to dielectric breakdown and filler concentration at different pulse frequencies. It can be seen that with increasing the loading level of fillers, the time to dielectric breakdown shows an increasing tendency at the same pulse frequency. Embedding BN fillers into pure PE matrix will enhance dissipation of heat due to the increase of thermal conductivity. Meanwhile, the incorporation of BN fillers can reinforce the thermal resistance of composites. The improvement of thermal resistance can effectively delay the occurrence of dielectric breakdown.

5 Application of thermally conductive polymer dielectrics

With the wide applications of high thermal conductivity materials in electronic packaging and thermal spreading substrate, very few studies had considered the application of these materials in electrical devices. Yoshitake proposed two new high thermal conductive insulation systems for motors: a glass cross-insulation system and a resin coated epoxy system [58]. Fig. 10 shows the insulation structure of the conventional insulation motor system and the proposed one. Results of experiments on the proposed prototype motors showed that the temperature rise of the motor coils was decreased: their temperature reached 73% of that of the motor coils using normal insulation and normal resin [0.6 W/(m·K)]. The temperature rise between the coil and stator core was only about 4 K in the proposed system. The temperature distributions of the conventional and proposed motors after 2000 s of operation (Fig. 10) were calculated by using thermal conductive and air flow finite element method (FEM). Results showed that the temperature of the conventional motor coil was 20 K degree higher than that of the proposed one.

The effects of epoxy with high thermal conductivity on temperature rise were studied using models that simulated a cast resin transformer. For the cast epoxy, micro-BN particles with grain diameter of 10 μm and nano-BN particles with grain diameter of 50 nm were used as fillers. Thermal conductivity of the epoxy is 0.23(I), 0.35(II), 0.62(III), 0.85(IV) and 1.23(V) W/(m·K) for different winding models.

The calculated thermal distribution of different windings with the thermal conductivities of 0.23(I), 0.62(II) and 1.23 (V) W/(m·K) is shown in Fig. 11. With the increase of the thermal conductivity of epoxy resin, the highest temperatures of different windings are 112.7, 87.7 and 78.8°C. The improvement of thermal conductivity has a significant effect to reduce the maximum temperature of the transformer. In addition, with the increase of the thermal conductivity, the lowest temperatures of different models are 36.4, 48.7 and 56.0°C. As can be seen from the temperature contours, the maximum temperature difference of winding I is 76.3°C, whereas it is 22.8°C in winding V. The temperature distribution of the winding filled with BN particles is more uniform than the conventional transformers.

Temperature distribution measurement is carried out when the transformer is under operation. Thermocouples are located at different positions on transformer winding to measure temperature changes continuously. The inner and outer surface temperature changes of different windings are 112.7, 87.7 and 78.8°C. As can be seen from the temperature contours, the maximum temperature difference of winding I is 76.3°C, whereas it is 22.8°C in winding V. The temperature distribution of the winding filled with BN particles is more uniform than the conventional transformers.

6 Conclusions and future challenges

(i) Minimising the interfacial resistance between the filler particles and the host polymer in a composite remains a major challenge. The ideal situation is to obtain the highest possible thermal conductivity with the lowest amount of the inorganic particles, which the preparation process should be improved to achieve the well dispersion and good dielectric properties.
(ii) Dielectric thermal breakdown characteristics of high thermal conductivity polymer composite should be studied further and the breakdown model and mechanism are not very clear.
(iii) Inorganic particles improved the thermal conductivity and resistance to tracking failure of the composite; however, they also reduce the dielectric breakdown and mechanical properties. How to ensure thermal performance and meet the requirements of electrical equipment is a key issue.
(iv) There are few applications of polymer composites with high thermal conductivity of polymer materials in electrical equipment and the ultimate goal is to achieve the application.

7 Acknowledgments

This work was supported by the Chinese National Natural Science Foundation under the grants 51277131 and 51537008 and the National Basic Research Program of China (Program 973, grants 2014CB239501 and 2014CB239506).

8 References

1 Agari, Y., Ueda, A., Nagai, S., et al.: Thermal conductivity of a polymer composite. J. Appl. Polym. Sci., 1993, 49, (9), pp. 1625–1634
2 Gonzalez, E., Gomez, P., Espino-Cortes, E., F.: ‘Analysis of the electric field distribution on insulating supports of dry-type transformers under high temperature’, IET Electr. Power Appl., 2012, 7 (5), pp. 331–337.

3 Tsekmes, I.A., Kochetov, R., Morshuis, P.H.F., et al.: ‘Modeling the thermal conductivity of polymeric composites based on experimental observations’, IEEE Trans. Dielectr. Electr. Insul., 2014, 21 (3), pp. 412–423.

4 Du, B.X., Meng, X., Zang, J.W.: ‘Effect of thermal conductivity on tracking failure of epoxy/BN composite under pulse stress’, IEEE Trans. Dielectr. Electr. Insul., 2013, 20 (5), pp. 296–302.

5 Wang, Z.B., Iizuka, T., Kozako, M., et al.: ‘Development of epoxy/BN composites with high thermal conductivity and sufficient dielectric breakdown strength part II: breakdown strength’, IEEE Trans. Dielectr. Electr. Insul., 2011, 18 (6), pp. 1973–1983.

6 Ohki, Y.: ‘Development of epoxy resin composites with high thermal conductivity’, IEEE Trans. Dielectr. Electr. Insul., 2005, 12 (1), pp. 48–49.

7 Choy, C.L., Young, K.: ‘Thermal conductivity of semicrystalline polymers-a model’, Polymer, 1977, 18 (8), pp. 769–776.

8 Grishchenko, A.E.: ‘Russian study of the mechanism of polyethylene deformation by the method of anisotropy of thermal conductivity’, Polym. Sci. B (Polym. Phys.), 1991, 36 (5), pp. 872–874.

9 Nysten, B., Gonry, P., Issi, J.P.: ‘Ultrafast breakdown of polyethylene films’, Polymer, 1994, 35 (10), pp. 1419–1425.

10 Wang, Z.H., Jeffrey, A.C., Alexei, L., et al.: ‘High thermal conductivity of polynorbornene nanotubes with very high thermal conductivities’, Nat. Nanotechnol., 2010, 5 (4), pp. 251–255.

11 Cao, B.Y., Yu, W.W., Kong, J., et al.: ‘High thermal conductivity of polyethylene nanowire arrays fabricated by an improved nanoporous template wetting technique’, Polymer, 2011, 52 (8), pp. 1711–1715.

12 Zhang, W.H., Jeffery, A.C., Alexei, L., et al.: ‘Ultrafast flash thermal conductance of polyethylene nanowire arrays’, Appl. Phys. Lett., 2006, 89 (10), pp. 153917–153919.

13 Kato, T., Nagahara, T., Agari, Y., et al.: ‘Relation between thermal conductivity and molecular alignment direction of free-standing film aligned with rubbing method’, Polymer, 2005, 46 (24), pp. 3911–3919.

14 Kato, T., Nagahara, T., Agari, Y., et al.: ‘High thermal conductivity of polynorbornene liquid-crystal acrylic film having a twisted molecular orientation’, Polym. Sci. B (Polym. Phys.), 2006, 44 (10), pp. 1419–1425.

15 Shen, S., Hennedy, D., et al.: ‘Thermal conductivity and mechanical properties of polymer composites filled with ceramic fillers’, J. Mater. Sci., 2010, 45 (4), pp. 689–696.

16 Calebrese, C., Nelson, J.K., Linda, S., et al.: ‘Polyamic-acid-based polymer nanocomposites for high-temperature applications’, J. Appl. Polym. Sci., 2011, 122 (3), pp. 1034–1041.

17 Tang, J.Y., Li, W.W., Wang, Z.H., et al.: ‘Thermal conductivity of highly filled dielectric epoxy/AIN composites’, J. Phys. Chem. C, 2012, 116 (25), pp. 13629–13639.

18 Han, Y.W., Lv, S.M., Hao, C.X., et al.: ‘Thermal conductivity improvement of epoxy resin composites cured under electric field: stacking of shape, thermal conductivity, and particle packing structure anisotropies’, Thermochim. Acta, 2012, 529, pp. 68–72.

19 Cho, H.B., Tokoi, Y., Tanaka, S., et al.: ‘Modification of BN nanosheets and their thermal conducting properties in nanocomposite film with polysulphone according to the orientation of BN’, Compos. Sci. Technol., 2011, 71 (8), pp. 1046–1052.

20 Andritsch, T., Kochetov, R., Gebrekir, Y.T., et al.: ‘Short term DC breakdown strength in epoxy based BN nanofiller composites’. 10th IEEE Int. Conf. on Solid Dielectrics, 2010, pp. 1–4.

21 Wang, Z.B., Iizuka, T., Kozako, M., et al.: ‘Development of epoxy/BN composites with high thermal conductivity and sufficient dielectric breakdown strength part III: sample preparations and thermal conductivity’, IEEE Trans. Dielectr. Electr. Insul., 2011, 18 (6), pp. 1963–1972.

22 Li, Z., Okamoto, K., Ohki, Y., et al.: ‘Effects of nano-filler addition on partial discharge resistance and dielectric breakdown strength of micro-Al2O3 epoxy composite’, IEEE Trans. Dielectr. Electr. Insul., 2010, 17 (3), pp. 653–661.

23 Du, B.X.: ‘Discharge energy and dc tracking resistance of organic insulating materials’, IEEE Trans. Dielectr. Electr. Insul., 2001, 8 (6), pp. 897–901.

24 Reddy, C.C., Ramu, T.S.: ‘Polymer nanocomposites as insulation for HVDC cables: investigations on the thermal breakdown’, IEEE Trans. Dielectr. Electr. Insul., 2008, 15 (1), pp. 221–227.

25 Du, B.X., Zhang, J.W., Gu, L., et al.: ‘Application of nonlinear methods in tracking failure test of epoxy-SiO2 nano-composite’. IEEE Int. Conf. on Solid Dielectrics, 2010, pp. 1–4.

26 Du, B.X.: ‘Effects of atmospheric pressure on de resistance to tracking of polymer insulating materials’, IEEE Trans. Dielectr. Electr. Insul., 2005, 12 (6), pp. 1162–1171.

27 Du, B.X., Zhu, X.H., Gu, L., et al.: ‘Effect of surface smoothness on tracking mechanism in XLPE-Si-ribbon interfaces’, IEEE Trans. Dielectr. Electr. Insul., 2011, 18 (1), pp. 176–181.

28 Li, Z., Okamoto, K., Ohki, Y., et al.: ‘Effects of nano-filler addition on partial discharge resistance and dielectric breakdown strength of micro-Al2O3 epoxy composite’, IEEE Trans. Dielectr. Electr. Insul., 2010, 17 (3), pp. 653–661.

29 Du, B.X.: ‘Discharge energy and dc tracking resistance of organic insulating materials’, IEEE Trans. Dielectr. Electr. Insul., 2001, 8 (6), pp. 897–901.

30 Reddy, C.C., Ramu, T.S.: ‘Polymer nanocomposites as insulation for HVDC cables: investigations on the thermal breakdown’, IEEE Trans. Dielectr. Electr. Insul., 2008, 15 (1), pp. 221–227.

31 Calebrese, C., Nelson, J.K., Linda, S., et al.: ‘Polyamic-acid-based polymer nanocomposites for high-temperature applications’. 10th IEEE Int. Conf. on Solid Dielectrics, Potsdam, Germany, 2010.

32 Maggioni, G., Vornierno, A., Carturan, S., et al.: ‘Structure and optical properties of Oxypolyamide nanocomposites produced by ion implantation’, Appl. Phys. Lett., 2004, 85 (23), pp. 5712–5714.

33 Ha, C.S., Park, H.D., Frank, C.W.: ‘Interactional interface in polyamide/silica hybrid composites by fluorescence spectroscopy’, Chem. Mater., 2000, 12, pp. 833–844.

34 Tong, Y.J., Li, S.Y., Liu, J.P., et al.: ‘Preparation and properties of polyimide films codoped with barium and titanium oxides’, J. Appl. Polym. Sci., 2002, 83, pp. 1810–1816.

35 Zhang, P.H., Fan, Y., Lei, Q.Q.: ‘Conduction current characteristics of nano-inorganic composite polyimide films’. IEEE Eighth IPCADM, 2006, pp. 755–758.

36 Zhang, P.H., Fan, Y., Lei, Q.Q.: ‘Morphology changes of both corona aged original and nano-inorganic hybrid polyimide films characterized by SEM’. IEEE Eighth IPCADM, 2006, pp. 882–884.

37 Ma, P.C., Nie, W., Yang, Z.H., et al.: ‘Preparation and characterization of polyimide/Al2O3 hybrid films by sol-gel process’, J. Polym. Sci., 2008, 46 (2), pp. 705–712.

38 Du, B.X., Xiao, M.: ‘Effects of thermally conducting particles on resistance to tracking failure of epoxy/BN composites’, IEEE Trans. Dielectr. Electr. Insul., 2014, 21 (4), pp. 705–712.

39 Tanman, H.J.: ‘Thermal and mechanical properties of aluminum powder-filled high-density polyethylene composites’, J. Appl. Polym. Sci., 1996, 62 (12), pp. 2161–2167.

40 Krupa, I., Novák, I., Chodák, I.: ‘Electrically and thermally conductive polyethylene/graphite composites and their mechanical properties’, Synth. Met., 2004, 145 (2), pp. 245–252.

41 Yoshitake, Y., Ohata, K., Enomoto, Y., et al.: ‘Experiment and calculation on insulating/thermal characteristics of high thermal conductive materials’. IEEE Int. Symp. on Electrical Insulating Materials, 2011.