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Nano SiO2/epoxy coating effect on lighting impulse breakdown characteristic in N2

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Abstract

Since SF6 has a very high global warming potential, it is essential to replace SF6 with environmentally friendly insulation gas. However, most readily available insulation gases have low electrical breakdown strength, which needs to adopt additional insulating means to enhance the poor breakdown strength. In this paper, with the aim of improving breakdown strength, filler/epoxy (EP) composites with nano-SiO2 particles in the weight percentage range of 0–5 wt% are used as dielectric coating materials and N2 is applied as insulating gas. As compared to that of neat EP, there is a decline in permittivity of SiO2/EP nanocomposites in the frequency range from 10^{-2} to 10^{6} Hz. Moreover, for SiO2/EP nanocomposites, the permittivity increases at first and then decreases as the filler content increases. With respect to the gas-coating insulation system, 50% breakdown voltage (BDV) U_{50} of the electrode with pure EP dielectric coating material is larger than that of bare electrode. Furthermore, a better BDV performance is obtained when applying SiO2/EP nanocomposites as dielectric coatings, indicating the introduction of SiO2 nanoparticles into EP matrix can reinforce the property of epoxy. The maximum BDV of 93 kV occurs in the electrode with 3 wt% SiO2/EP coating material, which is 29.17% higher than that of bare electrode. Additionally, it is found that the permittivity of coating material has a significant influence on the breakdown performance since it can alter the electric field distribution, and a lower permittivity always gives rise to a higher BDV.

1. Introduction

SF6 gas has been broadly applied to gas insulated switchgear (GIS), gas insulated transmission line (GIL), and gas blast circuit breaker (GCB) due to its extremely good insulating and arc-extinguishing performances [1–4]. However, SF6 has a very high global warming potential of roughly 23 000 times larger than that of CO2, moreover, the production costs of SF6 are relatively high and the handling of SF6 after use is also costly as the environmental constraints. Therefore, it has far-reaching meaning to replace SF6 with a more environmentally friendly insulation gas [5–9]. Unfortunately, most readily available and environmentally friendly insulation gases have a relatively low electrical breakdown strength, which would require the operating pressure or the dimensions of GIS to be significantly increased [10]. However, it would be unfeasible to increase the operating pressure or the dimensions of GIS, hence, the main challenge in the field of GIS is to improve the electrical breakdown strength of GIS without increasing the size or raising the operating pressure above the current design limits.

Recently, the investigation into the improvement of the breakdown strength of GIS has taken two main paths. One is that developing a new replacement gas which possesses a breakdown strength comparable to that of SF6 [11–15], and the other is that introduction of a dielectric coating layer on the electrode inside GIS [16–19]. Generally, when applying eco-friendly gas as an insulating gas, additional insulation technique like dielectric coated electrode is necessary to compensate the poor dielectric strength of eco-friendly gas compared to that of SF6 [20, 21]. This is mainly due to that the introduced dielectric coating layer can enhance the breakdown strength through modification of the capacitive electric field distribution and superposition for the electric field with charge accumulation on the coating surface. Furthermore, the electron field emission and ionization can be reduced and the metallic particle
movement can be inhibited when coating dielectric material on the surface of electrode \([22–25]\). As to dielectric coating material, epoxy resin (EP) is a good option because of its excellent characteristics, such as superior electrical insulating, high mechanical and chemical stabilities, as well as good processing ability \([26–28]\). Moreover, the introduction of inorganic fillers into epoxy matrix is an effective way to reinforce the physicochemical properties of epoxy \([29, 30]\).

In this study, filler/epoxy hybrid composites with nano-SiO\(_2\) particles are used as dielectric coating materials and N\(_2\) is applied as insulating gas. To the best of our knowledge, there is no report on the application of SiO\(_2\)/EP nanocomposites as the dielectric coating materials to improve the lightning impulse (LI) breakdown voltage of gas-coating insulation system so far. The LI breakdown voltage of gas-coating insulation system is evaluated through extensive LI breakdown tests on a rod-plane electrode configuration. For nanocomposites, the weight percentage of SiO\(_2\) filler varies between 0 and 5 wt%. In addition, a range of characterizations are carried out to acquire more information about the coating materials.

### 2. Experimental

#### 2.1. Coating material

In this work, raw material of SiO\(_2\) was purchased from Sinopharm Chemical Reagent Company, China, and the average particle size of SiO\(_2\) is about 20–30 nm. Curing agent of 4,4′-diamino diphenylmethane (DDM), curing accelerator (DMP-30, CP) and Bisphenol A epoxy resin with an epoxy equivalent weight of 184–190 g eq\(^{-1}\) were purchased from Shanghai Resin Company, China.

In order to achieve a uniform dispersion, planetary centrifugal mixing technique was employed to fabricate the epoxy composites filled with SiO\(_2\) nanoparticles in the weight percentage range of 1–5 wt%. The fabrication procedures are described as following. Firstly, adding a certain amount of EP, filler, curing agent and curing accelerator into the as-prepared vessel, which was treated in Thinky ARE-310 blender at 1000 r min\(^{-1}\) for 5 min and 2000 r min\(^{-1}\) for 10 min, and then followed by the ultrasound for 20 min. Finally, the vessel was transferred into a vacuum chamber in order to eliminate the air bubbles caused by the mixing and dispersion processes. The prepared adhesive was divided into two parts, one part was used as dielectric material coated on the surface of rod electrode, and the other part was applied to prepare specimen for characterization. For coating, an automated dip-coater (DP100-BE, Jiangyin Jiatu Company, China) was applied. The substrate was held for 30 s in the colloidal solution and vertically withdrawn with a constant rate of 100 mm min\(^{-1}\). As to the sample for characterization, the adhesive was poured into the prepared mold as quickly as possible. Then, the coating and thin film were putted into a vacuum oven with the constant temperature of 60 °C for 120 min, 100 °C for 120 min, and 150 °C for 120 min to acquire fully cured samples.

#### 2.2. Experiment set up and procedure

Figure 1 (a) displays the configuration of the selected rod-plane electrode in this study. Both the rod electrode and the plane electrode are made of stainless-steel, and the diameter of rod electrode with a hemispherical tip, which has the same diameter as the cylindrical part of the rod, is 14 mm, while the diameter of plane electrode is 50 mm. For electrical breakdown tests, the rod-plane electrode arrangement was placed inside an insulation gas tank as shown in figure 1 (b). The insulation tank is assembled by upper cover plate and side wall, which are made of polymethyl methacrylate. The plane electrode was fixed inside the enclosure and could not be easily removed or replaced, while the rod electrode with or without dielectric coating was placed at the opposite side, which
could be disassembled conveniently. The gap length between the electrodes was set at 20 mm to acquire the desired electric field distribution, which is comparable to that of a real size 40.5 kV C-GIS (Cubicle Gas Insulated Switchgear). After setting the electrodes in the insulation gas tank, atmospheric air was exhausted by vacuum pump to the ∼53 Pa level, and then followed by the introduction of the environmentally friendly gas of N2 up to a relative pressure of 0.06 Mpa. The gas and gas-coating breakdown tests were carried out by applying positive polarity impulse voltages on the rod electrode through the up-and-down method, and the 50% breakdown voltage \( U_{50} \) of the tested results was evaluated.

### 2.3. Characterization

The phase composition of epoxy composites were determined by x-ray diffraction (XRD, Bruker D2 PHASER, Germany) operated at 10 mA and 30 kV using Cu K\( \alpha \) radiation with a wavelength of 1.54 Å, and the scans were measured in the 2\( \theta \) range from 10\( ^\circ \) to 80\( ^\circ \). The chemical states of SiO\(_2\) particles were characterized using a Thermo Fisher Scientific ESCALAB Xi + x-ray photoelectron spectroscopy (XPS) instrument. The powder sample was mounted on double-sided adhesive tape and evacuated at room temperature. CasaXPS software was employed to analyze all the obtained binding energies (BEs), and the carbon (C1s) peak at 284.8 eV was used as a reference. Micromorphology of nanocomposite samples was inspected by using FEI Quanta 600 and KEYENCE TEM images of SiO\(_2\) nanoparticle and SiO\(_2\)/EP nanocomposites were collected from JEM-F200 electron microscope. For TEM operation, specimen was prepared by ultrasonically dispersing SiO\(_2\) powders in deionized water, and then dropping a few drops of the suspension onto the as-prepared copper grid. Coating Thickness Gauge (Minitest 735, EPK, Germany) was adopted to measure the thickness of coating layer of rod electrodes, and the obtained thickness value was about 75 \( \mu \)m. Thermogravimetry analyses (TGA, Mettler Toledo TGA/DSC 3, Switzerland) were carried out at a heating rate of 10 \( ^\circ \)C min\(^{-1}\) in N2. A differential scanning calorimetry (DSC, Mettler Toledo DSC 822e, Switzerland) spectrometer was used to measure the glass transition temperature (T\( g \)) of neat epoxy and epoxy composites. 8–10 mg samples were placed in a 40 \( \mu \)L aluminum pan and tested in N2. Dielectric property of the prepared nanocomposite samples was investigated using a broadband dielectric spectroscopy instrument (Concept 80, Novocontrol Technologies, Germany). A schematic representation of the dielectric spectroscopy setup can be seen in figure 2. The system includes a ZGS Alpha active cell and an automatic temperature control unit (Quatro cryosystem) with a precision of 0.1 \( ^\circ \)C. The tested sample lies in cryogenic cell, which can be cooled down to −160 \( ^\circ \)C and heated up to 400 \( ^\circ \)C with 0.01 \( ^\circ \)C temperature stability. Measurements were performed in a nitrogen atmosphere, and the frequency ranged from \( 10^{-2} \) to \( 10^{6} \) Hz.

### 3. Results and discussion

#### 3.1. Materials characterization

Figure 3(a) shows the XRD patterns of SiO\(_2\), EP and SiO\(_2\)/EP nanocomposites at room temperature. For SiO\(_2\) sample, it can be seen that a broad diffraction peak appears at ∼23\( ^\circ \) (2\( \theta \)), which is characteristic of amorphous SiO\(_2\) [31, 32]. As to SiO\(_2\)/EP nanocomposites, all the XRD profiles are similar and they can be imposed by using the two patterns of SiO\(_2\) and EP, suggesting the structure of nanocomposites may be explained by the coexistence of SiO\(_2\) in some region and EP in the other region. Moreover, since no sharp diffraction peaks are observed for any of the SiO\(_2\)/EP nanocomposites and a broad peak presents in the 2\( \theta \) range of 10\( ^\circ \)–30\( ^\circ \), the nanocomposites are considered amorphous without long range order [33].

X-ray photoelectron spectroscopy (XPS) was employed to analyze the surface state information of SiO\(_2\) nanofiller. According to the survey spectrum of SiO\(_2\) sample, as presented in figure 3(b), the presence of Si and O elements can be easily clarified. Following the survey scan, the Si 2p core level peak, as displayed in figure 3(c),
was also performed. During the fitting, the ratio of Lorentzian–Gaussian was fixed and the Shirley background subtraction method was utilized, moreover, the spectrum was calibrated by using the carbon binding energy of 284.8 eV. As shown in figure 3(c), the peak appearing at 103.5 eV was attributable to SiO$_2$, while the peaks at 102.2 and 101.1 eV were ascribed to SiO$_x$ [34]. Meanwhile, the composition ratios of 90% and 10% were obtained for SiO$_2$ and SiO$_x$, respectively.

Figure 3. (a) XRD patterns of SiO$_2$ nanofiller and SiO$_2$/EP nanocomposites. (b) XPS survey spectrum of SiO$_2$ nanofiller. (c) XPS spectrum of Si 2p for SiO$_2$ nanofiller.
3.2. Thermal properties

The glass transition temperature \((T_g)\) of SiO\(_2\)/EP nanocomposites was measured using differential scanning calorimetry (DSC) at a heating rate of 10 °C min\(^{-1}\) in N\(_2\), and the corresponding DSC curves are depicted in figure 4(a). \(T_g\) of epoxy usually associates with the cross-linking density of resin matrix [35]. Due to the crosslinking degree in epoxy matrix is not uniform, the glass transition temperature occurs in a wide temperature range. Hence, three characteristic temperatures, the onset temperature, the intermediate temperature and the end temperature, can be observed from the DSC curve [36]. Here, the glass transition temperature is attributed to the intermediate temperature. As shown in figure 4(a), the DSC curves of all samples
The phenomenon cannot be perfectly eliminated, since the large surface area and high surface energy of fillers acting as physical interlock points in the cured organic matrix, which provides a steric hindrance for curing reactions of composites and restricts the motion of macromolecular chain, resulting in the enhancement of $T_g$. Besides, for SiO$_2$/EP nanocomposites, it can be noted that $T_g$ is increased with the increase of nanoparticle content until it reaches to 3 wt%. After that, $T_g$ presents decreased tendency. The reason may be that higher filler concentration inevitably brings about the aggregation of nanoparticles with relatively larger dispersion size, which can hamper the curing reaction and reduce the curing degree, leading to the decrease of cross-linking density of the epoxy composite [37, 38].

Figure 4(b) shows the typical TGA curves of SiO$_2$/EP nanocomposites measured in the temperature range 30 °C–600 °C under nitrogen atmosphere. As displayed in figure 4(b), all the samples exhibit similar thermal behaviour and merely one-step decomposition, implying that the existence of SiO$_2$ nanoparticles did not significantly alter the degradation mechanism of epoxy matrix. Moreover, the TGA results demonstrate that the main weight loss of SiO$_2$/EP nanocomposites occurs in the temperature range between 250 and 450 °C, which is ascribed to the thermal degradation of the cured epoxy network [39, 40]. Additionally, the characteristic thermal parameters are listed in table 1. It can be found that SiO$_2$/EP nanocomposites possess a higher decomposition temperature compared to that of neat EP, suggesting SiO$_2$ nanofiller can partly enhance the thermal stability of epoxy. Moreover, the maximum degradation temperature ($T_{\text{max}}$) of SiO$_2$/EP nanocomposites is increased in comparison with that of neat epoxy, as presented in figure 4(c), this is probably attributed to the interfacial interaction between SiO$_2$ nanofiller and epoxy matrix, which can restrict the movement of the polymer molecular chains and the mobility of the polymer fragments at the interfaces of epoxy, giving rise to the retard of thermal decomposition.

| Temperature | 0 wt% SiO$_2$ | 1 wt% SiO$_2$ | 2 wt% SiO$_2$ | 3 wt% SiO$_2$ | 4 wt% SiO$_2$ | 5 wt% SiO$_2$ |
|-------------|---------------|---------------|---------------|---------------|---------------|---------------|
| $T_{\text{onset}}$ (°C) | 324.5 | 340.8 | 341.7 | 343 | 342.5 | 327.3 |
| $T_{\text{peak}}$ (°C) | 392 | 394.2 | 395.7 | 399 | 397.2 | 392.8 |
| $T_{\text{max}}$ (°C) | 395.5 | 397.2 | 398.5 | 401 | 399.7 | 396 |

are flat and do not show any endothermic and exothermic peaks, suggesting the samples were cured completely. The $T_g$ of SiO$_2$/EP nanocomposites with 0, 1, 2, 3, 4 and 5 wt% SiO$_2$ nanoparticles are 116.5, 118.2, 121.7, 126.2, 124.3 and 116.5 °C, respectively. As compared to that of neat epoxy, an increase of $T_g$ is observed for SiO$_2$/EP nanocomposites, this can be ascribed to the nanofillers acting as physical interlock points in the cured organic matrix, which provides a steric hindrance for curing reactions of composites and restricts the motion of macromolecular chain, resulting in the enhancement of $T_g$. Besides, for SiO$_2$/EP nanocomposites, it can be noted that $T_g$ is increased with the increase of nanoparticle content until it reaches to 3 wt%. After that, $T_g$ presents decreased tendency. The reason may be that higher filler concentration inevitably brings about the aggregation of nanoparticles with relatively larger dispersion size, which can hamper the curing reaction and reduce the curing degree, leading to the decrease of cross-linking density of the epoxy composite [37, 38].

Morphology and microstructure analysis

SEM was carried out to investigate the microstructure of SiO$_2$/EP nanocomposites. For observation, samples were prepared by brittle failure in liquid nitrogen and coated with gold via high vacuum sputtering coating machine to obtain a good conductance. The typical SEM images of the fracture surface of SiO$_2$/EP nanocomposites are exhibited in figure 5. The neat epoxy resin presents a very smooth fracture surface, manifesting a brittle fracture. For nanocomposites, the epoxy matrix and SiO$_2$ nanoparticles can be easily distinguished. As a whole, SiO$_2$ nanoparticles were distributed homogeneously in EP matrix, and these dispersed nanoparticles could act as the obstacle for crack propagation, thus resulting in the formation of rough surface as presented in SEM images.

Morphology of the as-prepared SiO$_2$ sample was characterized with transmission electron microscopy (TEM). As shown in figure 6(a), the micrograph reveals that SiO$_2$ particles are connected with each other intimately and the average particle size is about 20–30 nm. During TEM measurements, energy-dispersive x-ray (EDX) analysis was performed on the sample. The EDX profile (figure 6(b)) demonstrates the composition of Si and O elements, which is in good accordance with the XPS result. For SiO$_2$ nanofiller, as listed in the table of figure 6(b), the atom percentages of Si and O are 33.9% and 66.1%, respectively. Additionally, in order to get more information about the dispersion state of SiO$_2$ nanofiller in epoxy matrix, the nanocomposite sample was also investigated via TEM technique. For acquiring TEM micrographs of the nanocomposite, thin film of 20–40 nm thickness, as presented in figure 6(c), was cut out from the bulk material of the fabricated specimen by means of a diamond saw. Figure 6(d) shows the typical TEM image of 3 wt% SiO$_2$/EP nanocomposite, it can be seen that SiO$_2$ nanofiller in the epoxy matrix exhibits some traces of agglomerates. In fact, the agglomeration phenomenon cannot be perfectly eliminated, since the large specific surface area and high surface energy of nanoparticles contribute to the agglomeration [41].

3.4. Dielectric permittivity

The incorporation of nanofiller usually affects the dielectric properties of EP. A fully automated Alpha-A dielectric analyzer by Novocontrol was used to investigate the dielectric properties of the nanocomposite
samples. Measurements were carried out in N₂ atmosphere. The frequency varied between $10^{-2}$ and $10^6$ Hz, while the temperature increased from $-20$ to $120 \degree$C. For credibility, three parallel measurements were carried out for each sample.

Figure 7 displays the typical behavior of dielectric permittivity ($\varepsilon'$) of SiO₂/EP nanocomposites as a function of frequency at different temperatures. It can be easily observed that, in the whole frequency range, the
permittivity decreases with increasing frequency. Generally, the permittivity of filler/EP nanocomposite is dominated by the polarization related with epoxy and nanoparticles, and it is also affected strongly by the interfacial polarization at the interface between nanoparticles and epoxy. Hence, such a decline in permittivity is seen, since the frequency of measurement can affect the polarization process. In a typical EP system, the permittivity is governed by the number of orientable dipoles existing in the system and their ability to orient under an applied electric field. The epoxy is apt to possess higher permittivity in a lower frequency range due to most of the free dipolar functional groups in the epoxy chain are able to orientate at lower frequency of applied field. However, it will become more difficult for larger dipolar groups to orientate themselves when the frequency of applied voltage increases. Therefore, the influence of dipolar groups on the permittivity reduces continuously with the increase in frequency. Correspondingly, in the measured frequency range, the permittivity of neat EP will decrease with the increase of frequency. Similarly, the increasing frequency of applied filed will also bring about the decline of the inherent permittivity of SiO₂ nanofiller. Basing on the effective medium theories and mixing rules, which are applied to work out the permittivity in filler/polymer heterogeneous system, the combination of both effects gives rise to the decrease of permittivity for nanocomposites with the increase of frequency.
Moreover, it is noticeable that the $\varepsilon'$ increases with the increase of temperature, and there is a mutation when the temperature reaches about 120 °C. For example, at $10^{-3}$ Hz, the $\varepsilon'$ of neat epoxy rises about two times as the temperature increases from 20 to 120 °C. The prominent increase in $\varepsilon'$ of neat epoxy with temperature is related to the enhanced interfacial polarization. Basing on the Maxwell-Wanger-Sillars (MWS) effect, for heterogeneous polymer composites, space charges will be accumulated when the current flows from the interface of composite, resulting in the interfacial polarization [47, 48]. The abrupt change in $\varepsilon'$ can be ascribed to the glass transition movement, which frequently occurs in polymer materials when the chain segments begin to move and interact with each other. When temperature gets close to the glass transition temperature, the movement of polymer chain will intensify and the free charges will accumulate at the interface of material. Therefore, the sudden increase in $\varepsilon'$ with temperature is mainly attributed to the enhanced mobility of different bound charges, such as induced dipoles and interfacial space charges. This thermal excitation of different bound charges with the increase of temperature is the primary reason for the increase of $\varepsilon'$.

Another interesting observation from figure 7 is that, in most cases, there is a decline in permittivity of nanocomposites compared to that of neat EP. Moreover, it can be found that the filler content have a nonnegligible influence on the permittivity of filler/EP nanocomposites. The permittivity decreases at first and then increases with the increase in SiO$_2$ nanofiller content, and the lowest permittivity appears between 2 and 4 wt%. When a small amount of nanofiller is introduced into EP matrix, because of the interaction between epoxy chain and nanofiller, some thin immobile layers will be generated, and these layers can restrict the mobility of epoxy chain in the bulk material, thus resulting in the decrease of permittivity for filler/EP nanocomposites. After the minimum, the influence of filler’s inherent permittivity on resulting nanocomposites’ permittivity enhances as the filler content increases [49–52]. The permittivity of two phase dielectric follows the Lichteneker-Rother mixing rule:

\[
\log \varepsilon_c = x \log \varepsilon_1 + y \log \varepsilon_2
\]

where, $\varepsilon_c$ is the composite permittivity, $\varepsilon_1$ and $\varepsilon_2$ are the permittivity of filler and epoxy, and $x$, $y$ are the concentration of filler and epoxy. It is well known that SiO$_2$ nanofiller have higher permittivity compared with that of neat EP, this may be the reason to the increase of permittivity with higher filler contents.

### 3.5. Dielectric loss

Figure 8 presents the frequency dependency of the imaginary part of permittivity $\varepsilon''$ for SiO$_2$/EP nanocomposites at tested temperatures. The results show that $\varepsilon''$ increases with the increase in temperature, and when the temperature approaches to 120 °C, the $\varepsilon''$ increases abruptly, suggesting the dielectric loss is dominant in high temperature and leads to larger dissipation factor. Moreover, it can be observed that, in high frequency region, there is no obvious change in $\varepsilon''$. While in low frequency range, the $\varepsilon''$ varies significantly, suggesting the existence of nanoparticles seems to have a greater influence in lower frequency range. Since the conductivity of epoxy composites contributes to its dielectric loss, the variation of $\varepsilon''$ in lower frequency range may derive from the conductivity of nanocomposites, which can be affected by the presence of nanofillers. At each measured temperature, the applied frequency and the number of charge carriers are the dominant factors of the nanocomposites’ conductivity [53, 54]. Generally, the incorporation of nanofiller can affect the generation of mobile charge and the movement of charge carriers in polymer dielectrics, especially in lower frequency range where the conductivity will play a more important role, thus the variation in dielectric loss in low frequency range might be ascribed to the effect of electrical conductivity of inorganic fillers [47, 55, 56]. Additionally, the frequency dependence of dielectric loss tangent (tan$\delta$, tan$\delta$ = $\varepsilon''$/ $\varepsilon'$) is displayed in the insets of figure 8, and the peak appearing in low frequency range is mainly due to the $\alpha$-relaxation process, since dipoles have less time to orient themselves in the direction of the alternating field with the increase of frequency. At ~20 °C, a broad peak can be observed when the frequency increases to ~10$^4$ Hz, and this peak shifts to higher frequencies with the increase in temperature, which is ascribed to the $\beta$-relaxation process associating with the mobility of side chains in polymer.

### 3.6. Electric conductivity

The variation of AC conductivity ($\sigma_{ac}$) of SiO$_2$/EP nanocomposites as a function of frequency in the temperature range between –20 and 120 °C are presented in figure 9. AC conductivity $\sigma_{ac}$ because of localized states in dielectric material could be expressed using Jonscher’s power law [57]:

\[
\sigma_{ac}(\omega) = \sigma_{dc} + A\omega^s
\]

where $\omega$ represents the angular frequency, $\sigma_{dc}$ signifies the DC conductivity when $\omega$ closes to 0, $A$ stands for the pre factor, and $s$ is the frequency exponent. Both $A$ and $s$ have a weak dependence on temperature. As shown in figure 9, $\sigma_{ac}$ increases with the increase in frequency as well as temperature. The increasing trend of $\sigma_{ac}$ with the increase of frequency is probably related with the space charge carriers in material, while the improvement of
conductivity with temperature may be ascribed to the large heat energy absorbed in the materials, thus leading to the enhancement of dipole mobility. Moreover, the enhanced frequency independent of conductivity with the increase in temperature in low frequency region suggests the DC conductivity of the materials improve, since the random diffusion of charge carriers followed by hopping mechanism. The hopping frequency occurs at the point where the slope changes and it shifts to higher frequency with the increase in temperature, implying an enhancement in the carrier hopping rate of mobile charge carriers. The change in behavior of the $\sigma_{ac}$, from the initial gradual increase to rapid increase as the frequency increases, may be ascribed to a change in the hopping behavior of charge carriers, from long range to short range.

3.7. Gas-coating breakdown voltage

Figure 10(a) depicts the Weibull plots of insulation breakdown voltage (BDV) of the tested electrodes using SiO$_2$/EP nanocomposites with different filler concentration as dielectric coatings at a gas pressure of 0.06 MPa, and the gas gap is 20 mm. For comparison, the electrode without coating was also tested. As shown in figure 10(a), the 50% BDV $U_{50}$ of the electrode with dielectric coating of EP material is larger than that of bare electrode. Furthermore, it can be found that when applying SiO$_2$/EP nanocomposites as coating materials, the electrodes exhibit a better performance compared to that of neat EP, implying the introduction of SiO$_2$.
nanofiller into EP matrix can reinforce the property of epoxy and the nanocomposite is more suitable for the application of dielectric coating material.

The Peak BDVs of the tested electrodes coated with SiO\textsubscript{2}/EP nanocomposites are displayed in figure 10(b). It can be found that the BDV increases at first and then decreases as the filler content increases, and it varies between 83 and 93 kV. The maximum BDV occurs in the electrode coated with 3 wt\% SiO\textsubscript{2}/EP nanocomposite material, and as compared to that of bare one, the BDV is improved about 29.17\%. Basing on the obtained results, it can be concluded that the BDV of bare electrode can be significantly improved by coating with SiO\textsubscript{2}/EP nanocomposites. Moreover, the 3 wt\% SiO\textsubscript{2}/EP coating material gives rise to the highest BDV value, suggesting 3 wt\% SiO\textsubscript{2}/EP is a very promising dielectric coating material.

3.8. Electric field distribution

Figure 11(a) shows the simulation result of the electric field between rod and plane electrodes. The length of the gas gap between the tip of the rod electrode and the plane electrode is set at 20 mm. It can be found that the electric field strength is very high in the vicinity of rod electrode’s tip, where the electric field distribution is divergent. Figure 11(b) displays the electric field distribution along a straight line between the top of rod and the surface of plane. The horizontal axis value represents the distance between the local position and the top of rod,
while the local electric field strength $E$ is displayed on the vertical axis. It is well known that the $E$ value of the internal electrode is 0, therefore, the data before 80 mm are omitted for a clear observation. As can be seen in figure 11(b), compared with that of bare electrode, the electric field distribution is changed when dielectric coatings with different permittivity are applied on the surface of rod electrodes. The electric field strength inside coating is lowered significantly. Furthermore, at the coating-gas interface, the electric field strength shows a jump which is caused by the permittivity of coating. Even with the jump in the electric field strength at the coating surface, the maximum electric field strength in the gas is reduced with respect to the uncoated situation. The maximum electric field strength in the uncoated situation is roughly $1.1738 \times 10^7$ V m$^{-1}$, while in the coated situation, it is about $1.1729 \times 10^7$ V m$^{-1}$. Because the difference between the permittivity of the coating...

Figure 10. (a) Weibull plots of insulation BDV for tested electrodes using SiO$_2$/EP nanocomposites as coating materials. (b) Peak BDV and enhancement of the tested electrodes using SiO$_2$/EP nanocomposites as coating materials.

Figure 11. (a) Simulation of the electric field between rod and plane electrodes. (b) Electric field distribution in a rod-plane configuration with SiO$_2$/EP nanocomposite coatings. (c) Enlargement view of the orange rectangle area. (d) Enlargement view of the violet rectangle area.
and the gas is smaller, the step increase in the electric field strength at the interface is also smaller. Fortunately, solid dielectric materials have a higher electric breakdown strength compared to that of insulating gas. As shown in figure 11(c), the electric field strength increases with the decrease of permittivity in the coating zone.

Figure 12. The variation in peak BDV and $\varepsilon'$ of SiO$_2$/EP nanocomposites.

Figure 13. (a) The typical image of the electrode with coating. (b) Cross section view of the electrode with coating. (c) The typical surface view of the electrode with coating after LI BD tests.
Table 2. Cost of the gas-coating (N$_2$-3wt% SiO$_2$/EP) insulation system and gas (SF$_6$) insulation.

| Items           | EP          | Curing agent | Curing accelerator | Nano SiO$_2$ | N$_2$     | SF$_6$ |
|-----------------|-------------|--------------|---------------------|--------------|-----------|--------|
| Usage amount    | 2 mg        | 1.8 mg       | 0.05 mg             | 0.12 mg      | 12.94 L   | 9.71 L |
| Unit price      | 3.43 USD kg$^{-1}$ | 2.86 USD kg$^{-1}$ | 5.14 USD kg$^{-1}$ | 51.43 USD kg$^{-1}$ | 2.38 $\times$ 10$^{-4}$ USD L$^{-1}$ | 5.23 $\times$ 10$^{-2}$ USD L$^{-1}$ |
| Total price     | 6.86 $\times$ 10$^{-6}$ USD | 5.15 $\times$ 10$^{-6}$ USD | 2.57 $\times$ 10$^{-7}$ USD | 6.17 $\times$ 10$^{-6}$ USD | 3.08 $\times$ 10$^{-3}$ USD | 0.51 USD |
Meanwhile, the electric field strength reduces rapidly in the gas, and it decreases with decreasing permittivity as displayed in figure 11(d).

3.9. Influence of permittivity on BDV
The variation in peak BDV and $\varepsilon'$ of Si$_2$O$_2$/EP nanocomposites is presented in figure 12. Interestingly, it can be found that the dielectric coating material with low permittivity always gives rise to a high BDV. As mentioned in the former section, through investigating the relationship between dielectric property and electric field distribution, it was demonstrated that the permittivity of coating can lead to the altering of electric field strength. It is well known that the BDV relates intimately with the electric field distribution. Moreover, considering the homogeneous coating thickness and similar testing conditions, we can speculate that the dielectric property of coating material has a significant influence on the BDV performance.

3.10. SEM of the electrode after breakdown voltage test
SEM was performed to get an insight into the microstructure of the electrode with coating after breakdown voltage tests. For SEM observation, the tested electrode was cut into two parts, as shown in figure 13(a), and the head part was adopted to perform SEM. Figure 13(b) exhibits the cross section view of the electrode with coating, it can be clearly observed that the coating material adheres to the stainless-steel electrode intimately, suggesting there was no delamination occurred at the interface between coating layer and substrate electrode during the tests. Moreover, the SEM result reveals that the thickness of coating layer is about 75 $\mu$m, which is very close to the value measured by coating thickness gauge.

Figure 13(c) displays the typical surface view of the electrode with coating after breakdown voltage tests. Through careful observation, it can be observed that some traces, as the yellow rectangle indicated, appeared at the surface of coating, which might be caused by the surface discharge during the up-and-down tests. Moreover, some part of the coating, as the red rectangle pointed out, was penetrated. Generally, during the breakdown voltage tests, once some part of the insulator is broken down, the value of the subsequent breakdown voltage will be lower than that of the first breakdown voltage. This is mainly due to that the weak spot will be always the breakdown point. Therefore, we deduce that the initial discharge path is bridged between the surface of the coating and the plane electrode. When the voltage reaches to the threshold value of the gas-coating insulation system, the discharge penetrates the coating and goes into gas.

3.11. Cost analysis of gas-coating insulation system and gas insulation
Generally, cost is an important parameter in the practical application of GIS. Therefore, in order to compare the cost between the gas-coating (N$_2$/3wt% Si$_2$O$_2$/EP) insulation system and gas (SF$_6$) insulation only when their 50% breakdown voltages are the same, the usage amount and unit price of each raw material are summarized in table 2. Here, it is worth noting that the listed unit price for each raw material is the market price in China. After careful calculation, it can be obtained that the total amount of gas insulation only is about 165 times higher than that of gas-coating insulation system, suggesting gas-coating composite insulation system has a fairly high level of competitiveness in the application of GIS.

4. Conclusions
In this study, the rod electrodes with Si$_2$O$_2$/EP nanocomposites dielectric coatings were successfully fabricated, and the thickness of coating layer was about 75 $\mu$m. The dielectric permittivity of neat epoxy resin was changed after the incorporation of SiO$_2$ nanofiller. In most cases, there is a decline in permittivity of SiO$_2$/EP nanocomposites compared to that of neat EP within the frequency range of $10^{-2}$–$10^6$ Hz at 20 °C. Moreover, the permittivity of nanocomposites increases at first and then decreases as the filler content increases. The 50% BDV $U_{50}$ of the electrode with EP dielectric coating material is larger than that of electrode without EP coating. Furthermore, when using SiO$_2$/EP nanocomposites as dielectric coatings, a better BDV performance is obtained, suggesting the introduction of SiO$_2$ nanofiller into EP matrix can reinforce the property of epoxy. For the electrode with 3 wt% SiO$_2$/EP coating material, a peak BDV value of 93 kV was acquired, which was enhanced approximately 29.17% compared to that of bare electrode. Additionally, the electric field distribution can be altered when applying dielectric coatings with different permittivity on the surface of electrodes. For the dielectric coating materials, a lower permittivity always brings about a higher BDV. Through the discharge traces appeared at the surface of the electrode with coating after breakdown voltage tests, we speculate that, before the threshold value of the gas-coating insulation system, the discharge path is bridged between the surface of the coating and the plane electrode, and after that, the discharge penetrates the coating and goes into gas.
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