Equivalent failure model of force-electric load and dielectric properties on epoxy resin-based three-dimensional composites

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Abstract
Epoxy resin (EP) has excellent insulation properties, yet the toughness of pure EP is poor, therefore, MWCNTs nanoparticles are added to improve the toughness of EP. However, MWCNTs have excellent conductivity, consequently, organic montmorillonite (OMMT) nanoparticles are attempted to improve the adverse effect of MWCNTs on EP matrix. In this paper, the dielectric and impact properties of three-dimensional composites are characterized and analysed, and the results show that the mechanical properties of matrix are improved by the incorporation of MWCNTs and OMMT nanoparticles, on the other hand, the excessive incorporation of nanoparticles will lead to the increase of dielectric loss and relative permittivity, simultaneously, the increase of electric field frequency will also have a greater impact on the dielectric properties of the composites. In the meantime, the equivalent relationship between the failure caused by voltage breakdown and external loading is studied. Through the relevant research, the relevant properties of the sample can be changed and the damage degree can be reduced under the fixed external voltage.

1. Introduction
Epoxy resin (EP) has excellent insulation properties and poor toughness, therefore, it is necessary to improve the mechanical properties of EP, yet, the addition of nanoparticles will affect the dielectric properties of EP. MWCNTs is a kind of nanoparticles with high aspect ratio, on the other hand, the toughness of EP can be improved by adopted with MWCNTs, nevertheless, MWCNTs have high conductivity, which will adversely affect the insulation performance of EP, therefore, OMMT nanoparticles are added into the matrix to reduce the adverse effects of MWCNTs. Carbon nanotubes (CNTs) are widely used due to the excellent properties [1].

Many experts and scholars have studied MWCNTs in many fields, especially in the modification of EP matrix. Gojny et al found the percolation threshold of composites when the mass fraction of MWCNTs is 0.1 wt%, on the other hand, with the increase of MWCNTs content, the conductivity of composite increases, yet the thermal conductivity of that is little affected by the content of MWCNTs [2]. Ajayan et al used the method of melt mechanical mixing to make MWCNTs uniformly dispersed in EP, simultaneously, it was found that MWCNTs on the shear surface had no fracture after being acted on [3]. Cooper et al studied the deformation of MWCNTs in EP matrix and found that the load could be transferred from the matrix to the MWCNTs [4]. Maria et al modified MWCNTs and found that EP/MWCNTs could achieve excellent electrical conductivity without decreasing mechanical properties at very low impurity content [5]. Schadler et al found that the compressive elastic modulus of EP/MWCNT composites is higher than that of tensile elastic modulus, simultaneously, it was found that the outer layer of MWCNTs acts only in tension, yet the inner and outer layers of MWCNTs transmit loads together during compression [6]. Zhou et al found that MWCNTs carried external forces and weakened...
the energy of fracture, which hindered the propagation of cracks and led to the disorderly distribution of cracks [7]. Although MWCNTs can improve the mechanical properties of EP, it is necessary to modify MWCNTs for improving the dispersion in EP. Yaping Zheng et al modified MWCNTs with mixed acid, and grafted the surface of acidified MWCNTs with urethane hyperbranched polymer [8]. Gong et al used surfactants to improve the dispersion of MWCNTs in EP, and changed the interfacial energy of MWCNTs, to improve the elastic modulus of composites [9]. Weglikowska and Stevens modified single-walled carbon nanotubes (SWCNTs) with diamines and successfully attached amino groups to the wall, however, the former was prepared by acyl chloride method, yet the latter was obtained by fluorination method [10, 11]. Wang et al modified MWCNTs with amino groups, resulting in improved compatiblity of MWCNTs with EP, and more uniform dispersion, in addition, the mechanical, optical and thermal stability of the composites were significantly improved [12]. Park et al found that MWCNTs have high aspect ratio and surface energy to agglomerate easily, when the content of MWCNTs increases to a certain level, the dispersion of MWCNTs in the matrix changes from needle-like fiber structure to agglomerated winding structure, which greatly limits the excellent performance [13]. Jin et al found that the modified MWCNTs were well dispersed in EP and interfacial bonded with polymer matrix, simultaneously, the modified EP/MWCNTs nanocomposites showed higher storage and loss modulus than that of the unmodified MWCNTs [14-16]. Although MWCNTs have many advantages, in this paper, OMMT nanoparticles are adopted with matrix to observe and analyze the performance changes of epoxy resin-based three-dimensional composites. OMMT is a kind of layered silicate nanoparticles, simultaneously, and there are many studies and reports on OMMT [17-22]. Xuefang Chen et al used cobalt nanofibers as mechanical support to improve conductivity. The one-dimensional coaxial structure can stably buffer the volume change, and avoid the structural damage and improve the mechanical properties. In the composite materials, cobalt nanofibers as the mechanical support of nickel silicate nanoflakes improve the specific surface area and conductivity of the materials [23-26]. Although there are a large amount of studies on EP/MWCNTs and EP/OMMT composites, yet not three-dimensional composites, in particular, the mechanical and dielectric properties of the composites are required to be investigated, simultaneously. Therefore, the three-dimensional composites based on EP are further analyzed in this paper. Furthermore, the equivalent failure relationship is established by comparing the voltage breakdown of the sample with the damage caused by the external loading of the sample. Through the relevant research, the relevant properties of the sample can be changed and the damage degree can be reduced under the fixed external voltage.

2. Experimental section

2.1. Materials
Bisphenol A Epoxy Resins (E-44) are bought from Wuxi Resin Phoenix Co. Ltd, China, Industrial Grade. The curing agent is bought from Methyl hexahydrophthalic anhydride, which is originated from Anael Chemical Co., Ltd, China (Purity: >99%). The accelerator is bought from Pyridine, which is originated from Tianjin No.6 Chemical Reagent Factory, (Purity: >99%). Raw montmorillonite (MMT) is bought from Xi’an Comprehensive Test Center Rock Mineral Chemical Institute, China. The organic modification reagent is Octadecyl trimethyl ammonium chloride, which is bought from China Pharmaceutical Group Chemical Reagents Co., Ltd Raw multi-walled carbon nanotubes (MWCNTs), length: 5 ~ 15 μm, diameter: 40 ~ 60 nm, (Purity: >97%), are bought from Shenzhen Nano-port Co., Ltd

2.2. Preparation of OMMT
Raw montmorillonite (mass: 20 g) and abundant distilled water are laid into a three-necked flask and stirred for 30 min at 80 °C. Octadecyl quaternary ammonium salt (mass: 5.6 g) dissolved in distilled water, is poured into the three-neck flask. The mixture, which is mechanically stirred at 80 °C for 2 h, is filtered by a suction device. At last, chloride ions are not detected with silver nitrate solution by pouring distilled water. The treated montmorillonite (OMMT) is obtained.

2.3. Preparation of acidified MWCNTs
Raw MWCNTs (mass: 2.5 g), doped with H2SO4(98%) and HNO3(68%), are laid in flasks at 80 °C for 6 h under the action of ultrasonication, afterwards, the mixture is poured into the distilled water and then filtered, after that, distilled water is continuously poured to dilute the mixture until the PH value of the mixture is closely neutral. At last, the acidified MWCNTs are dissolved in acetone reagent to maintain dissolution.

2.4. Preparation of epoxy resin-based three-dimensional composites
EP is dissolved in acetone solution, then, OMMT and MWCNTs are doped with the matrix and stirred for 24 h at 80 °C, in the meantime, the residual acetone and bubbles are eliminated by vacuum evacuation. After that, the
curing agent (Methyl hexahydrophthalic anhydride) is poured into the mixture and stirred for 30 min to achieve uniform mixture. Afterwards, the accelerator (pyridine) is poured into the mixture, and the mixture is poured into the mould, then, cured by a certain heating gradient. Finally, the composites are cooled and preserved.

2.5. Characterization

The volumetric resistivity of epoxy resin-based three-dimensional composites is tested by ZC36 high resistance meter produced by Shanghai Precision Scientific Instruments Co., Ltd. The impact strength of the specimens is tested by K-12 simple-supported beam tester of Hongdu Machinery Works, simultaneously, the test standard refers to GB/T 1043–2008. Fracture surface morphology of composites are observed by scanning electron microscope (SEM). Dielectric constant and dielectric loss tester of frequency conversion electric field (type: Alpha-A) bought from Novocontrol Technologies Co., Ltd, is used to measure and analyze the dielectric constant and loss. Samples are processed into circular slices (diameter: 30 mm, thickness: 1 mm), then, aluminum electrodes are coated on both layers.

3. Results and discussion

3.1. Volumetric resistivity of epoxy resin-based three-dimensional composites

As an insulating material, the volume resistivity of EP is very high. EP must be modified to improve the toughness. In this paper, adding modified MWCNTs into EP will certainly affect the insulation performance of composite, because MWCNTs is a metallic conductive inorganic tubular nanoparticle. Compared with some data, the percolation threshold of composite in this paper is higher due to the length of original MWCNTs and conductivity decreasing after acidification treatment. Moreover, the surface structure of MWCNTs after acidification treatment is damaged, which may also affect the conductivity of MWCNTs. It is expected that the addition of OMMT can restrain the electrical properties of EP caused by the addition of MWCNTs. As shown in figure 1, compared with pure EP, the volume resistivity of composite (MWCNTs with 0.1 wt%) is decreased. When the content of MWCNTs is 0.3 wt% and 0.5 wt%, the volume resistivity of EP/MWCNTs does not change a lot, but still shows a weak downward trend. However, the volume resistivity of composite (MWCNTs with 0.7 wt%) decreases again. When the content of MWCNTs is 0.9 wt%, the volume resistivity of EP/MWCNTs composites does not change significantly, owing to the agglomeration of MWCNTs. When the organic montmorillonite materials were added, it was found that the volume resistivity of EP/MWCNTs/OMMT composites was higher than that of EP/MWCNTs as the same content of MWCNTs being added, which indicated that the addition of OMMT did improve the insulation properties of the composites, due to the addition of OMMT. The lamellar structure hinders the contact and charge transfer of tubular MWCNTs particles. At the same time, it can be observed that the volume resistivity of EP/MWCNTs/OMMT composites...
does not change regularly with the increase of OMMT content. It may be due to the agglomeration of organically treated montmorillonite nanoparticles when the content of OMMT is too high. The OMMT materials contain the migration phenomena caused by impurity ions, and the OMMT particles can be dispersed uniformly in the matrix at low content, but not prevent the ion migration on the surface of the OMMT lamellae after peeling. With the increase of MWCNTs content, the volume resistivity of EP/MWCNTs/OMMT composites shows a downward trend, however, the downward trend is relatively flat compared with the volume resistivity of EP with only MWCNTs, which indicates that the addition of OMMT has indeed played a role in slowing down the effect of MWCNTs on the electrical conductivity of EP.

3.2. Analysis of dielectric constant and dielectric loss of epoxy resin-based three-dimensional composites
MWCNTs have excellent electrical conductivity, yet, EP has excellent insulation properties, therefore, many tiny capacitors are formed between MWCNTs and EP. Under the action of electric field, the interface between MWCNTs and EP easily leads to the accumulation of charges. Figures 2(a) and (b) are principle and schematic diagrams of the equivalent capacitance formed between MWCNTs with EP, respectively. Figure 2(a) shows the voltage redistribution in series among the capacitors formed by the incorporation of nanoparticles, nevertheless, figure 2(b) shows the capture of charge by a large amount of equivalent small capacitors under the action of electric field.

The frequency of general alternating electric field is 50 Hz, so it is necessary to study dielectric constant and loss of insulating materials under the condition of constant frequency. As shown in figure 3, the dielectric constant and loss of EP cured under three curing gradients were analyzed. In figure 3, the dielectric constant and loss of EP are the smallest when the curing gradient is 100 °C (2 h)/150 °C (3 h). The curing temperature has a

Figure 2. Principle (a) and schematic diagram (b) of equivalent capacitance on epoxy resin matrix composites doped with MWCNTs.
great influence on the formation of the spatial network structure on EP, which is mainly reflected by the degree of crosslinking and the length of molecular chains among adjacent crosslinking points. As the curing gradient increases, the degree of crosslinking increases, but the length of molecular chains between adjacent crosslinking points decreases. Under the condition of applied electric field, the larger the degree of crosslinking and the smaller the length of molecular chains among adjacent crosslinking points, the more difficult it is to establish dipole polarization.

According to Clausius equation:

\[ \varepsilon_r - 1 = \frac{N\alpha E_i}{\varepsilon_0 E} \]  \hspace{1cm} (1)

\( \varepsilon_r \)—Relative permittivity;
\( N \)—Molecular Number in Unit Volume of Dielectric
\( \alpha \)—Molecular polarizability \( (F \cdot m^2) \);
\( \varepsilon_0 \)—Vacuum relative dielectric constant
\( E_i \)—Electric field strength acting on molecules \( (V/m) \)
\( E \)—Macroscopic average electric field strength \( (V/m) \)

The higher the degree of cross-linking, the more difficult it is to establish dipole polarization, resulting in a smaller relative permittivity. However, at the curing gradient of 120 °C/150 °C, the relative permittivity increases, which may be due to the excessive curing temperature, resulting in some damage about the internal molecular chains of the materials and local defects. (For example, due to local thermal aging of materials, the ability to store charges decreases, resulting in an increase in relative permittivity and dielectric loss).

For EP/MWCNTs composites, MWNTs and EP are cross-distributed. MWNTs can be regarded as a metal conductor and EP as an insulating medium. Therefore, MWNTs and EP can be regarded as many tiny capacitors. The whole composite specimen is made up by many tiny capacitors in parallel. Under the condition of external alternating electric field, many electrons are captured by the interface between MWCNTs and EP, which results in the local accumulation of space charge and the non-uniform distribution of free charge in the medium and the polarization of space charge with macroscopic dipole moment. In figure 4, relative permittivity and dielectric loss of EP/MWCNTs composites were measured under 50 Hz frequency electric field with the change of MWCNTs content. Relative permittivity and dielectric loss of the material decrease firstly and then increase with the improvement of MWCNTs content. The higher the content of MWCNTs, the more equivalent small capacitors formed by EP composites. Compared with pure EP, MWCNTs is equivalent to impurities, which affects the excellent charge storage performance of the original EP.

As shown in figure 4, with the improvement of MWCNTs content, the dielectric loss of composites decreases firstly and then increases. The more the two-phase interface is formed, and the more the interface loss will be formed, however, the dielectric loss of pure EP is higher, owing to impurities or bubbles in the sample, and the electrons polarize at the bubbles or impurities. The degree of polarization is much greater than that of EP doped with MWCNTs.

Taking three-dimensional composites with OMMT content of 6 wt% as an example, as present in figure 5(a), the dielectric constant of the composites decreases rapidly with the increase of applied electric field.
Figure 4. The Dielectric Loss and Dielectric Constant of EP/MWCNTs under the condition of different content about MWCNTs (50 Hz alternating electric field).

Figure 5. Frequency conversion dielectric constant (a) and dielectric loss (b) of three-dimensional composites with 6 wt% OMMT content.
When the electric field frequency increases to a certain extent, relative permittivity does not change any more, which indicates that with the increase of electric field frequency, the relative permittivity in the medium does not change, however, when the frequency of the electric field increases to a certain extent, the molecular polarization is unable to change synchronously with the frequency of the electric field, leading to the dielectric constant no change, simultaneously, the composites with different content of MWCNTs show the same trend.

In figure 5(b), the frequency conversion dielectric loss of EP matrix composites, doped with different MWCNTs at 6 wt% OMMT content, is analyzed, on the other hand, the dielectric loss of the composites increases with the increase of electric field frequency, due to the electronic oscillation in the samples with the change of electric field frequency. When the electric field frequency reaches a certain value, the dielectric loss will reach the maximum value.

For the three-dimensional epoxy resin matrix composites with OMMT and MWCNTs nanoparticles, the relative permittivity and dielectric loss of the composites were measured at the applied electric field frequency of 50 Hz. As present in figure 6(a), the relative permittivity of the composite is the lowest when the contents of MWCNTs and OMMT are 0.4 wt% and 2 wt%, respectively, in the meantime, when the contents of MWCNTs and OMMT are 0 wt% and 4 wt%, respectively, the relative permittivity of the composite is the highest. The higher the relative permittivity is, the stronger the ability of storing charge is, therefore, the relative permittivity of the composite can be measured, showing that the existence of MWCNTs changes the ability of EP to store charge. However, when the content of MWCNTs exceeds a certain value, the effect of MWCNTs on the ability to store charge decreases. In figure 6(b), when the content of OMMT reaches a certain level, the dielectric loss of the composites is the highest, simultaneously, when the content of MWCNTs is too high, the dielectric loss of the composite.
composites will also increase. Furthermore, when the content of OMMT is a certain value, the adverse effect of adding MWCNTs into EP will be inhibited, due to the interfacial coupling between layered silicate OMMT and MWCNTs, which hinders the conductive path formed by MWCNTs and reduces the contact between MWCNTs and EP. Excessive incorporation of nanoparticles will lead to uneven dispersion of nanoparticles, which negatively affect the epoxy resin matrix.

Relaxation polarization time, which is a parameter to investigate the dielectric loss, is the time when the medium tends to steady state. The longer the relaxation polarization time is, the longer the polarization time is, furthermore, when the applied electric field frequency is high and the polarization is unable to keep up with the change frequency of electric field, the polarization loss will be greater. In figure 7, the relaxation polarization time of EP/MWCNTs composites is shown under different MWCNTs content. When MWCNTs content is 0.1 wt%, the relaxation polarization time is the longest, however, when MWCNTs content is 0.3 wt%, the relaxation polarization time is the shortest. With the increase of MWCNTs content, the relaxation polarization time increases gradually, which indicates that the doping of MWCNTs increases. When a large amount of MWCNTs nanoparticles are adopted with EP, the polarization lag becomes serious gradually, and the time to complete polarization stabilization becomes longer and the loss increases. Because of the excellent conductivity of MWCNTs, many tiny capacitors are formed when MWCNTs are added to epoxy resin matrix, simultaneously, the interface between MWCNTs and EP will hinder the movement of electrons in alternating electric field, resulting in an increase in the time to complete polarization stabilization and loss.

The relaxation polarization time of the composites changed greatly due to the incorporation of MWCNTs and OMMT nanoparticles. Figure 8 shows the relaxation polarization time of the composites after incorporation of OMMT (2 wt%, 4 wt%, 6 wt%, 8 wt%) and MWCNT (0 wt%, 0.1 wt%, 0.3 wt%, 0.5 wt%, 0.7 wt%, 0.9 wt%). When the contents of MWCNTs and OMMT are 0.1 wt% and 2 wt%, respectively, the relaxation polarization time of the composites is the highest, yet much lower than that of the composites with only 0.1 wt% MWCNTs, indicating that a certain content of OMMT will increase the relaxation loss. Moreover, as present in figure 8, when the contents of MWCNTs and OMMT are 0.1 wt% and 6 wt%, respectively, the relaxation polarization time of composites is the lowest, and the content of MWCNTs is also 0.1 wt%. However, the different content of OMMT has a great influence on the relaxation polarization time of composites, which indicates that the higher the OMMT content, the less the effect of MWCNTs nanoparticles on the relaxation loss of epoxy resin matrix.

The relative permittivity is a complex number composed of real part and imaginary part, in which the real part represents the ability of the material to store charge, while the imaginary part represents the heat loss caused by the dipole vibration damping in the dielectric, and the dielectric loss is the ratio of the imaginary part and the real part of the relative permittivity. Due to the large imaginary part of the relative permittivity of MWNCTS, the loss caused by dipole vibration damping is large, which leads to large dielectric loss, especially with the increase of applied electric field frequency. On the other hand, when the frequency of the electric field is low, the dipole molecules can follow the alternating direction of the electric field, and the dielectric constant is large, which is close to the measured value of the dielectric constant under DC voltage. When the frequency exceeds a certain critical value, the change of the polarity molecule is incapable of keeping up with the change of the electric field, and the relative permittivity begins to decrease. With the increase of the frequency, the relative permittivity finally approaches the relative permittivity caused only by the electronic displacement polarization. Because
montmorillonite is a kind of non-conductive nano particle with lamellar structure, it will not cause EP to change from insulator to conductor and lose its proper function. However, no matter what kind of nano materials are adopted with the EP matrix, it will have a negative impact on the insulation performance, but the impact of doping non-conductive nano particles is smaller than that of doping conductive nano particles. Although MWNTs can improve the mechanical properties of the EP, in order to reduce the adverse effect of MWNTs nanoparticles on the dielectric properties of the EP, montmorillonite nanoparticles are added to achieve the purpose of mutual coating of montmorillonite and MWNTs nanoparticles, to prevent MWNTs from forming a conductive network, and to reduce the loss caused by the conductivity of MWNTs.

3.3. Fracture morphology and impact strength of epoxy resin-based three-dimensional composites

When MWCNTs and OMMT nanoparticles are adopted with EP, the fracture strength of EP will be affected, on the other hand, the toughness of composites has changed, hence, the corresponding cross-section images will be distinct. Figure 9 shows the fracture surface of three-dimensional epoxy resin matrix composites with different contents of MWNTs and OMMT. As present in figure 9, the fracture surface of pure epoxy resin is smooth, which indicates that the mechanical properties are poor, nevertheless, the fracture surface becomes rough obviously after adding nanoparticles, which indicates that the toughness of the composite is improved, simultaneously, the cross-sections of the composites with different content of nanoparticles are rough, yet not significant.

The impact properties of the composites changed when OMMT and MWCNTs nanoparticles were adopted with epoxy resin. Figure 10 shows the impact strength data of epoxy resin based three-dimensional composites. When the OMMT content is 0 wt%, the impact strength of the composites increases with the increase of MWCNTs content, but the increase of impact strength is small. When the content of OMMT is 4 wt%, the impact strength of the composites increases with the increase of the content of MWCNTs, but the range of increase is small. However, when the content of MWCNTs is the same, the impact strength of the composites increases greatly after adding OMMT, which indicates that OMMT plays a greater role in improving the impact strength of the composites. The content of MWCNTs is much lower than that of OMMT, consequently, MWCNTs are unable to affect the epoxy resin matrix as OMMT nanoparticles. When the composite material is subjected to external impact, MWCNTs are tubular structure, which will cause MWCNTs to be pulled out of EP at the fracture site, nevertheless, OMMT is layered silicate, when the composite material is subjected to external impact load, the fracture occurs between the layers of OMMT.

3.4. Equivalent failure model of force-electric load

For the voltage breakdown of the sample, the breakdown holes and cracks occur mainly in the middle of the sample, which correspond to the failure phenomena caused by external loading in the relevant region. Figure 11 is a schematic diagram of the sample after voltage breakdown, that is, besides the annular hole in the middle of the sample, there will also be carbonization and fracture region.

The relationship between polar coordinates and Descates coordinates [27]:

![Figure 8. Relaxation polarization time of epoxy resin-based three-dimensional composites with different OMMT and MWCNTs contents.](image)
Figure 9. Section morphology of epoxy resin-based three-dimensional composites (a). Pure epoxy resin (b). 0 wt% OMMT, 0.3 wt% MWCNTs (c). 0 wt% OMMT, 0.5 wt% MWCNTs (d). 0 wt% OMMT, 0.7 wt% MWCNTs (e). 4 wt% OMMT, 0.3 wt% MWCNTs (f). 4 wt% OMMT, 0.5 wt% MWCNTs (g). 4 wt% OMMT, 0.7 wt% MWCNTs.

Figure 10. Impact strength of epoxy resin-based three-dimensional composites.
and \( y \) are two arguments of stress function, so are \( \rho \) and \( \varphi \). Get the following conclusion:

\[
\frac{\partial \varphi}{\partial x} = \frac{\partial \varphi}{\partial \rho} \frac{\partial \rho}{\partial x} + \frac{\partial \varphi}{\partial \varphi} \frac{\partial \varphi}{\partial x} = \cos \varphi \frac{\partial \varphi}{\partial \rho} - \frac{\sin \varphi}{\rho} \frac{\partial \varphi}{\partial \varphi} \quad (2a)
\]

\[
\frac{\partial \varphi}{\partial y} = \frac{\partial \varphi}{\partial \rho} \frac{\partial \rho}{\partial y} + \frac{\partial \varphi}{\partial \varphi} \frac{\partial \varphi}{\partial y} = \sin \varphi \frac{\partial \varphi}{\partial \rho} + \frac{\cos \varphi}{\rho} \frac{\partial \varphi}{\partial \varphi} \quad (2b)
\]

If the \( x \)-axis and the \( y \)-axis are respectively transferred to the direction of \( \rho \) and \( \phi \), made the \( \phi \) as zero. \( \sigma_x, \sigma_y, \tau_{xy} \) become \( \sigma_\rho, \sigma_\varphi \) and \( \tau_{\rho\varphi} \). The following equations could be derived when the volume force is not considered.

\[
\sigma_\rho = (\sigma_\rho)_{\varphi=0} = \left( \frac{\partial^2 \varphi}{\partial \rho^2} \right)_{\varphi=0} = \frac{1}{\rho} \frac{\partial \varphi}{\partial \rho} + \frac{1}{\rho^2} \frac{\partial^2 \varphi}{\partial \varphi^2} \quad (3a)
\]

\[
\sigma_\varphi = (\sigma_\varphi)_{\varphi=0} = \left( \frac{\partial^2 \varphi}{\partial \varphi^2} \right)_{\varphi=0} = \frac{\partial^2 \varphi}{\partial \rho^2} \quad (3b)
\]

Figure 11. Model diagram of alternating current voltage applied to the sample (a); Model diagram of sample after voltage breakdown (b).
$$\tau_{\rho\varphi} = (\tau_{\rho\varphi})_{\varphi=0} = -\frac{\partial^2\phi}{\partial x \partial y} \bigg|_{\varphi=0} = -\frac{\partial}{\partial \rho} \left( \frac{1}{\rho} \frac{\partial \phi}{\partial \varphi} \right)$$  \hspace{1cm} (3c)

It is proved that the stress components conform the following equations when \( f_\rho = f_\varphi = 0 \) has been established.

$$\frac{\partial \sigma_\rho}{\partial \rho} + \frac{1}{\rho} \frac{\partial \tau_{\rho\varphi}}{\partial \varphi} + \frac{\sigma_\rho - \sigma_\varphi}{\rho} + f_\rho = 0; \quad \frac{1}{\rho} \frac{\partial \sigma_\varphi}{\partial \varphi} + \frac{\partial \tau_{\rho\varphi}}{\partial \rho} + 2\tau_{\rho\varphi} + f_\varphi = 0$$  \hspace{1cm} (4)

$$\frac{\partial^2 \phi}{\partial x^2} + \frac{\partial^2 \phi}{\partial y^2} = \frac{\partial^2 \phi}{\partial \rho^2} + \frac{1}{\rho} \frac{\partial \phi}{\partial \rho} + \frac{1}{\rho^2} \frac{\partial^2 \phi}{\partial \varphi^2}$$  \hspace{1cm} (5a)

$$\left( \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} \right) \phi = 0$$  \hspace{1cm} (5b)

$$\left( \frac{\partial^2}{\partial \rho^2} + \frac{1}{\rho} \frac{\partial}{\partial \rho} + \frac{1}{\rho^2} \frac{\partial^2}{\partial \varphi^2} \right) \phi = 0$$  \hspace{1cm} (5c)

The deflection \((w)\) and load \((q)\) are both regarded as functions of polar coordinates, \(w = w(\rho, \varphi), q = q(\rho, \varphi)\).

$$\frac{\partial w}{\partial x} = \cos \varphi \frac{\partial w}{\partial \rho} - \sin \varphi \frac{\partial w}{\partial \varphi} \quad \frac{\partial w}{\partial y} = \sin \varphi \frac{\partial w}{\partial \rho} + \cos \varphi \frac{\partial w}{\partial \varphi}$$  \hspace{1cm} (6a)

$$\nabla^2 w = \frac{\partial^2 w}{\partial \rho^2} + \frac{1}{\rho} \frac{\partial w}{\partial \rho} + \frac{1}{\rho^2} \frac{\partial^2 w}{\partial \varphi^2}$$  \hspace{1cm} (6b)

Differential equation of elastic surface:

$$D \nabla^4 w = qD = \frac{E\delta}{12(1-\mu^2)}$$  \hspace{1cm} (7a)

\(E\)–modulus of elasticity \(\delta\)–thickness of thin plate \(\mu\)–poisson ratio

$$D \left( \frac{\partial^2}{\partial \rho^2} + \frac{1}{\rho} \frac{\partial}{\partial \rho} + \frac{1}{\rho^2} \frac{\partial^2}{\partial \varphi^2} \right) \left( \frac{\partial^2 w}{\partial \rho^2} + \frac{1}{\rho} \frac{\partial w}{\partial \rho} + \frac{1}{\rho^2} \frac{\partial^2 w}{\partial \varphi^2} \right) = q$$  \hspace{1cm} (7b)

According to Ritz method: ignore the \(\varepsilon_x, \gamma_{xy}\) and \(\gamma_{xz}\). As follow, the equations of deformation potential energy:

$$V_\varepsilon = \frac{1}{2} \iiint (\sigma_x \varepsilon_x + \sigma_y \varepsilon_y + \tau_{xy} \gamma_{xy}) \, d\rho \, d\varphi \, dz$$  \hspace{1cm} (8a)

$$\sigma_x = -\frac{Ez}{1-\mu^2} \left( \frac{\partial^2 w}{\partial \rho^2} + \frac{\partial^2 w}{\partial \varphi^2} \right); \quad \varepsilon_x = -\frac{\partial^2 w}{\partial x^2}$$  \hspace{1cm} (8b)

$$\sigma_y = -\frac{Ez}{1-\mu^2} \left( \frac{\partial^2 w}{\partial y^2} + \frac{\partial^2 w}{\partial x^2} \right); \quad \varepsilon_y = -\frac{\partial^2 w}{\partial y^2}$$  \hspace{1cm} (8c)

$$\tau_{xy} = -\frac{Ez}{1+\mu} \frac{\partial^2 w}{\partial x \partial y}; \quad \gamma_{xy} = -2\varepsilon_x$$  \hspace{1cm} (8d)

Then put them into formula (8a), educe the following equation (9a):

$$V_\varepsilon = \frac{E}{2(1-\mu^2)} \iiint (\nabla^2 w)^2 - 2(1-\mu) \left( \frac{\partial^2 w}{\partial x^2} \frac{\partial^2 w}{\partial y^2} - \left( \frac{\partial^2 w}{\partial x \partial y} \right)^2 \right) \, d\rho \, d\varphi \, dz$$

$$= \frac{D}{2} \iint \left( (\nabla^2 w)^2 - 2(1-\mu) \left( \frac{\partial^2 w}{\partial x^2} \frac{\partial^2 w}{\partial y^2} - \left( \frac{\partial^2 w}{\partial x \partial y} \right)^2 \right) \right) \, d\rho \, d\varphi$$  \hspace{1cm} (9a)

The second integral could be transformed into the equation:

$$\iint \left( \frac{\partial^2 w}{\partial x^2} \frac{\partial^2 w}{\partial y^2} - \left( \frac{\partial^2 w}{\partial x \partial y} \right)^2 \right) \, d\rho \, d\varphi$$

$$= \iint \left( \frac{\partial w}{\partial x} \frac{\partial^2 w}{\partial x \partial y} + \frac{\partial w}{\partial y} \frac{\partial^2 w}{\partial x \partial y} \right) \, d\rho \, d\varphi$$  \hspace{1cm} (9b)
condition of the displacement boundary. The variation of deflection $w$ is realized by the variation of coefficient $C_m$. $w_m$ changes with coordinates which has nothing to do with the above variation. In order to determine coefficient $C_m$, the following equation (10) should be applied. In the bending of thin plates, the volume and surface force are all classified into the load $q$.

$$\frac{\partial V_c}{\partial C_m} = \iint f_z w_m dx dy dz + \iint f_z w_m dS = \iint q w_m dx dy$$  \hspace{1cm} (10)$$

$C_m$ is defined with $m$ linear equations. The deflection $w$ is obtained with the equation $w = \sum C_m w_m$, to solve questions of internal force.

Deformation energy formula for thin plates of equal thickness—

$$V_c = \frac{D}{2} \iint \left\{ (\nabla^2 w)^2 - 2(1 - \mu) \left[ \frac{\partial^2 w}{\partial \rho^2} \left( \frac{1}{\rho} \frac{\partial w}{\partial \rho} + \frac{1}{\rho^2} \frac{\partial^2 w}{\partial \varphi^2} \right) \right]^2 \right\} \rho d\rho d\varphi$$

(11a)

$$\frac{\partial V_c}{\partial C_m} = \iint q w_m \rho d\rho d\varphi$$

(11b)

In axisymmetric problems, transverse loads and deflections are only functions of $\rho$ that are $q = q(\rho)$, $w = w(\rho)$. The expression of deformation potential energy (11-a) and (11-b) is simplified to

$$V_c = \pi D \int \left[ \rho \left( \frac{d^2 w}{d\rho^2} \right)^2 + \frac{1}{\rho} \left( \frac{dw}{d\rho} \right)^2 + 2 \mu \frac{d^2 w}{d\rho^2} \frac{d^2 w}{d\rho^2} \right] d\rho$$

(11c)

$$\frac{\partial V_c}{\partial C_m} = 2\pi \int q w_m \rho d\rho$$

(11d)

The following equations could be educed for circular plate with an outer radius of $a$ when the entire boundary of the circular plate is fixed.

$$V_c = \pi D \int \left[ \rho \left( \frac{d^2 w}{d\rho^2} \right)^2 + \frac{1}{\rho} \left( \frac{dw}{d\rho} \right)^2 \right] d\rho$$

(11e)

In figure 12, the clamped side circular plate is uniformly distributed under the center circle area of $q_0$, with a radius of $a$. The expression of deflection is set to

$$w = \left( 1 - \frac{\rho^2}{a^2} \right)^3 \left[ C_1 + C_2 \left( 1 - \frac{\rho^2}{a^2} \right) + C_3 \left( 1 - \frac{\rho^2}{a^2} \right)^2 + \ldots \right]$$

(12a)
which could be satisfied with the condition of displacement boundary.

\[ (w)_{\rho=a} = 0; \left( \frac{dw}{d\rho} \right)_{\rho=a} = 0; \left( \frac{dw}{d\rho} \right)_{\rho=0} = 0 \]

Simplify equation (12a)

\[ w = C_i w_1 = C_i \left( 1 - \frac{\rho^2}{a^2} \right)^2 \]  \hspace{1cm} (12b)

Derive the first and the second derivative of \( w \):

\[ \frac{dw}{d\rho} = -\frac{4C_i}{a^2} \left( 1 - \frac{\rho^2}{a^2} \right) \rho \frac{d^2w}{d\rho^2} = -\frac{4C_i}{a^2} \left( 1 - \frac{3\rho^2}{a^2} \right) \]  \hspace{1cm} (12c)

**Figure 13.** Finite Element Simulations of Displacement Distribution (a) and Stress Distribution of Epoxy Resin (b).
Substitute (12c) into (11e).

\[ V_c = \pi D \int_0^a \left\{ \rho \left[ \frac{4C_1}{a^2} \left( 1 - \frac{b^2}{a^2} \right) \right]^2 + \frac{1}{\rho} \left[ \frac{4C_1}{a^2} \left( 1 - \frac{b^2}{a^2} \right) \rho \right]^2 \right\} d\rho \]

\[ = \frac{32\pi DC_1^2}{3a^2} \]

\[ \frac{\partial V_c}{\partial C_m} = \frac{\partial V_c}{\partial C_1} = \frac{64\pi DC_1}{3a^2} \]

According to (12b),

\[ 2\pi \int q_{w_m} \rho d\rho = 2\pi \int_0^b q_0 \left( 1 - \frac{\rho^2}{a^2} \right) \rho d\rho \]

\[ = \pi q_0 \frac{b^2}{3} \left( 3 - \frac{3b^2}{a^2} + \frac{a^2}{b^2} \right) \]

Substitute (12e) and (12f) into (11e) to work out \( C_1 \).

\[ w = \frac{q_0 a^4}{64D} \left( 3 - \frac{3b^2}{a^2} + \frac{b^2}{a^2} + \frac{b^2}{a^2} \left( 1 - \frac{b^2}{a^2} \right)^2 \right) \]

In this model, \( a = 5 \times 10^{-2} \text{ m}, b = 10^{-4} \text{ m}, q_0 = 220 \text{ Mpa}, D = 0.2610 \text{ N } \cdot \text{m} \).

The result of calculation: \( w_{m=0} \approx 0.9878 \text{ mm} \).

An external loading (220 MPa) is applied to the 200 \( \mu \text{m} \) area of the center diameter of the specimen, and the stress generated in the 200 \( \mu \text{m} \) area of the specimen can reach the strength of the specimen (60 MPa), as shown in the gray area in figure 13(a). Figure 13(a) shows the finite element simulation of the sample displacement, the displacement of the center position is 0.9894 mm, which is basically consistent with the theoretical calculation of 0.9878 mm. Therefore, it can be assumed that the stress failure caused by external loading is 220 MPa, which is the equivalent failure load of voltage breakdown. The diameter of the breakdown hole is generally about 200 \( \mu \text{m} \).

According to the finite element analysis as shown in figure 13, the external load can be considered to be about 220 MPa, and the area of action can be assumed to be the area of breakdown. Assuming that the volume of the breakdown channel is \( V \) (cylindrical channel with radius \( r \) and depth \( l \)), the electrostatic energy in the channel can be expressed as follows [28–30]:

\[ dV = \pi r^2 dl \]

\[ W_0 = \frac{1}{2} E_0 w = \frac{1}{2} \varepsilon E^2 dV + \frac{1}{2} \sigma \gamma dV + W_c \]

\( E_0 \)– Equivalent applied load; \( E \)–Electric field strength

\( W_c \)–Carbonization energy \( \varepsilon \)–Dielectric constants of channel cracks; \( \gamma \)–strain

\( \sigma \)–Maxwell stress induced by electrostatic field

The impairing energy \( W_c \) caused by external loading is energy equivalent to that caused by applied voltage, that is, the energy required to produce the above two equivalent failure modes is the same on the premise of producing the same size of failure holes.

4. Conclusion

In this paper, the dielectric and mechanical properties of three-dimensional epoxy resin-based composites are tested and analyzed. The volume resistivity of the composites is affected by the content of MWCNTs and OMMT, on the other hand, MWCNTs have excellent conductivity, which is disadvantageous to the insulation properties of EP. The different curing gradients of composites will affect the dielectric properties, due to different curing gradients leading to different crosslinking degree of EP. With the increase of MWCNTs content, the dielectric constant and dielectric loss of the composites decrease firstly and then increase under the condition of 50 Hz electric field. With the increase of electric field frequency, the dielectric constant of three-dimensional composites decreases rapidly until a certain value, nevertheless, the dielectric loss of three-dimensional composites increases with the increase of alternating electric field frequency. The dielectric constant and dielectric loss of epoxy resin-based three-dimensional composites are measured under 50 Hz electric field, in other words, when the content of OMMT and MWCNTs nanoparticles is too high, the dielectric constant and dielectric loss will increase. The study on relaxation polarization time of EP/MWCNTs composites shows that when the content of MWCNTs is 0.3 wt%, the relaxation polarization time is the lowest, i.e. the loss is the smallest. The study on relaxation polarization time of three-dimensional epoxy resin composites shows that the
relaxation pole of the composites is reduced by incorporating OMMT nanoparticles into EP. When the contents of OMMT and MWCNTs are 6 wt% and 0.1 wt%, respectively, the relaxation polarization time of the composites is the shortest. The impact properties of three-dimensional composites show that the content of OMMT is higher than that of MWCNTs, consequently, OMMT has a greater toughening effect on EP. The relationship between the two kinds of failure energy is established via analyzing the breakdown of the sample by voltage and the failure hole of the same size under the action of the applied load. The failure degree can be reduced via changing the characteristics of the sample material under the action of the same voltage.

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