

The Effect of Hygrothermal Aging on the Properties of Epoxy Resin

Youyuan Wang*, Yu Liu[†], Kun Xiao*, Can Wang* and Zhanxi Zhang*

Abstract – Because of excellent electrical properties, epoxy resin is widely used in packaging and casting power equipment. Moisture and temperature in the environment are inclined to seriously affect the insulation tolerance of epoxy resin. This work focuses on the aging characteristics of epoxy resin in hygrothermal environment. Scanning electron microscopy images show that there are micro-crack, micro-slit and holes inside aged samples. The moisture absorption process undergoes three equilibrium stages and it does not follow the Fick's second law. Observing the change of hydrogen bonds in the infrared spectra of the dried samples, it is found that chemically moisture absorption immerses when the physical moisture absorption entered the third equilibrium stage. By Debye equation to fit the imaginary part of the dielectric constant, it is concluded that the uniformity of water molecule has a great influence on the electrical conductivity loss. Furthermore, the polarization loss can be more easily affected by water molecules than small free molecules. After the aged samples being dried, their real and imaginary part of the dielectric constant descend, but their original electrical properties cannot completely restored. After chemical moisture absorption appears inside the material, the residual space charges increase significantly and the charge dissipation rate slow down obviously.

Keywords: Epoxy resin, Hygrothermal aging, Equilibrium stage, Debye equation, Space charge

1. Introduction

Epoxy resin matrix composites boast high insulation strength, excellent chemical stability, strong environmental adaptability, and thus they are widely used in electrical equipment as molding and sealing materials [1-4]. In some specific application areas, the moisture in the operating environment is inclined to affect the insulation properties of epoxy resin severely. Additionally, because of its poor thermal conductivity, the operating equipment continues withstanding the effect of heat and the reliability of equipment is seriously affected [5-8]. In this work, the influence of hygrothermal aging on the epoxy resin is mainly investigated.

At present, some theoretical and experimental researches has been done to investigate the effect of hygrothermal aging on material properties. The diffusion of water molecules is often accompanied by the relaxation of glassy polymers, so the moisture absorption can be divided into two stages. Only the first stage is in accordance with Fick's second law, and the hydrolysis of epoxy resin is most likely to lead to chain scission [9, 10]. Hydrogen bonds are formed between a part of the water entering the matrix resin and the hydrophilic groups in resin [11]. The glass transition temperature T_g of the epoxy resin is measured by DSC. The measurement result shows that the T_g of the

resin after absorbing moisture is lower about 25°C than that of the dry resin, which is caused by the plasticization of the resin [12]. Absorbed water associates with the polar hydroxyl groups, and there are two possible hydrogen-bond configurations. The O...HO con-figuration is an intramolecular hydrogen bond between the hydroxyl hydrogen and the closest ether oxygen. The second possibility is an OH...OH hydrogen bond between two hydroxyls [13]. With the increase of moisture absorption T_g gradually decreases, and the particles with hydrophobic functional groups in epoxy resin reduce the amount of moisture absorption [14, 15]. When the water molecules diffuses through the resin to the internal quartz, interface is produced between quartz and epoxy resin [16]. Water molecules cause serious degradation of the interface between fiber and epoxy resin [17]. When permeating into the carbon fiber/epoxy surface, water leads to interface delamination and matrix plasticization [18]. A breakdown model based on a water-filled cavity is proposed and discussed [19]. The epoxy resin is degraded after hygrothermal aging, transverse elastic modulus is the most easily affected and the shear modulus is the least easily affected in the mechanical properties [20]. The water absorption of the dried material at 50°C is reversible [21]. The Fourier transform infrared spectroscopy shows that the oxidation of the material results in significant changes in the absorption peaks of carbonyl and hydroxyl groups [22]. After absorbing water, the C-O and N-CO-N bonds are generated inside epoxy and fracture occurs in the main chain [23].

At present, the process of moisture absorption is not

[†] Corresponding Author: Dept. of Electrical Engineering, Chongqing University, China. (20151102030t@cqu.edu.cn)

* Dept. of Electrical Engineering, Chongqing University, China. ({y.wang, 20141102051, 20094589, zhx.zhang} @cqu.edu.cn)

Received: July 10, 2017; Accepted: November 30, 2017

clearly studied and the analysis of moisture absorption content after hygrothermal aging does not take into account the impact of thermal mass loss. The study on the effect of physical and chemical moisture absorption on the property of epoxy resin are also lacked. In order to solve the above problems, after hygrothermal aging the moisture absorption process of samples is analyzed in detail, and related moisture absorption contents are gained after considering thermal mass loss. Additionally, the research of hygrothermal aging properties mainly focuses on the influence of physical and chemical absorption on each aging characteristic. And the changes of material properties caused by chemical absorption is particularly concerned. This work is expected to be helpful in understanding the effect of hygrothermal aging on the long term durability of epoxy resin in harsh operating environment.

2. Experimental

2.1 Sample preparation

First of all, the molds are cleaned and dried. After the molds cooling, release agent is coated on molds. Epoxy resin (E51), curing agent (MTHPA), dibutyl phthalate (DBP) and accelerator (Imidazole) are mixed with the volume ratio of 100:90:10:1. The mixture is heated in incubator with 50°C for 10 minutes to make the epoxy resin viscosity decrease. With blender mixing it 1 hour, then mixture is vibrated in the ultrasonic oscillator for half an hour to make it mix evenly. Finally mixture is put into a vacuum tank to remove the bubble. After importing mixture into the molds, these samples are cured in a constant temperature chamber for 2 hours at 90°C and post-cured for 2 hours at 110°C. Finally circular specimens are produced whose diameters are 30mm and thicknesses are 0.163±0.002mm.

2.2 Hygrothermal aging

In this paper, epoxy resin is subjected to hygrothermal aging experiment (Case 1) with the relative humidity (RH) of 95% and the temperature of 80°C. In order to obtain the thermal mass loss of the material, the thermal aging experiment (Case 2) with the RH of 0% and the temperature of 80°C is carried out at the same time. Twelve kind of aging time of 0, 14, 24, 72, 96, 120, 134, 144, 158, 168, 182, 336h for Case 1 and Case 2 are carefully characterized and analyzed in this paper.

2.3 Aging characteristics test

After hygrothermal aging, the tensile fracture surface morphology, gravimetric measurement, infrared spectrum, the dielectric properties and space charge characteristics of the aged samples are investigated.

2.3.1 SEM (Scanning Electron Microscope)

The tensile fracture surfaces of specimens aged in case 1 after 14, 24, 72, 120, 144 and 182h were observed by a scanning electron microscope (Tescan MAIA3 XMH, Czech). Prior to the characterization of SEM, the samples were sputter coated with a thin layer of gold in vacuum to improve the electrical conductivity. The observed SEM images were magnified 30000 times.

2.3.2 Gravimetric measurement

Ten specimens (30mm×30mm×0.163mm) were used for each case and aging time. Before aging, the mass of each sample W_0 was measured by electronic scale and recorded. Electronic scale was with a precision 0.01mg. The average weight change of ten specimens is calculated as weight change. The weight change η after aging was defined in (1):

$$\eta = \frac{W - W_0}{W_0} \times 100\% \quad (1)$$

Where W and W_0 were the sample weights after aging and initial state, respectively.

During hygrothermal aging process, high temperature led to weight decreasing, thus the weight loss caused by thermal aging η_{ta} should be taken into account. Thus the total moisture absorption η_{tma} could be written in (2):

$$\eta_{tma} = \begin{cases} \left(\frac{W - W_0}{W_0} + |\eta_{ta}| \right) \times 100\% \\ \eta_1 (\eta_2 = 0) \end{cases} \quad (2)$$

Where η_{tma} was the total moisture absorption. η_{ta} was the weight loss rate caused by thermal aging, which was calculated by Eq. (1). η_1 and η_2 were the physical and chemical moisture absorption, respectively, which were calculated in Eq. (3) and Eq. (4).

2.3.3 Drying reprocess

The aged samples were placed in a beaker with a moisture absorbing function of dried silica particles and the beaker was put into a constant temperature blast oven at 37°C after 24h. Then the infrared spectroscopies of these samples were tested.

2.3.4 FTIR(Fourier Transform Infrared Spectroscopy)

The structural changes in epoxy resin during aging were analyzed with FTIR spectrometer. Each sample mentioned above was tested in the wave number range of 500 - 4000 cm^{-1} . One computer was connected to the spectrometer in order to receive and analyze data.

2.3.5 Dielectric properties

The dielectric properties of the materials were measured in the frequency domain from 10^0Hz - 10^6Hz at room temperature using a Novocontrol ALPHA-A high resolution dielectric analyzer. Prior to dielectric test, gold was deposited onto both surfaces of the specimens by sputtering. The diameter of the sputtered gold was 15 mm.

2.3.6 Space charge

Space charge of solid medium was measured by PEA (Pulsed Electro-acoustic) system with a pulse width of 2-5ns, the pulse amplitude of 200V, and the output voltage of 0-20kV. During the experiment, the samples aged 168h were broke down under the 15kV/mm electric field, so the DC electric field applied in the experiment was set to 10kV/mm. A DC electrical field was applied for 30min, then the electrical field was removed and the space charge dissipation was confirmed after the depolarization 10s, 360s, 960s and 1800s.

3. Result and Discussion

3.1 Properties of moisture absorption

3.1.1 SEM image

Fig. 1 is SEM image showing tensile sections of the samples aged for 14h, 24h, 72h, 120h, 144h and 182h under Case 1. It can be seen that during the initial stage of hygrothermal aging, there are not cracks and slits inside the material, whose tensile section is relatively smooth and has no defects. With the increase of aging time, the number of cracks increases and the crack becomes longer and wider. At early aging period, the distribution of cracks and slits is uneven that in the local area some long and wide cracks and slits emergence. With aging time increasing, the distribution of cracks and slits tends to be more uniform. And there are a lot of defects and holes inside material. This is because expansion, hydrolysis and deterioration act inside epoxy resin which causes its structure has been destroyed in hygrothermal environment.

Water molecules cause the material to expand. If the distribution of absorbed water molecules is uneven, the internal expansion stress is also uneven, which will lead to uneven distribution of cracks and slits. In particular, it can be seen from Fig. 1 that with the increase of aging time, formation rate of new cracks and slits becomes slower and slower, and the distribution of water molecules and cracks becomes more and more uniform. In the later period of hygrothermal aging, cracks and slits are mainly deepened on the basis of the original cracks and slits.

3.1.2 The content of moisture absorption

Unlike physical moisture absorption, chemical moisture

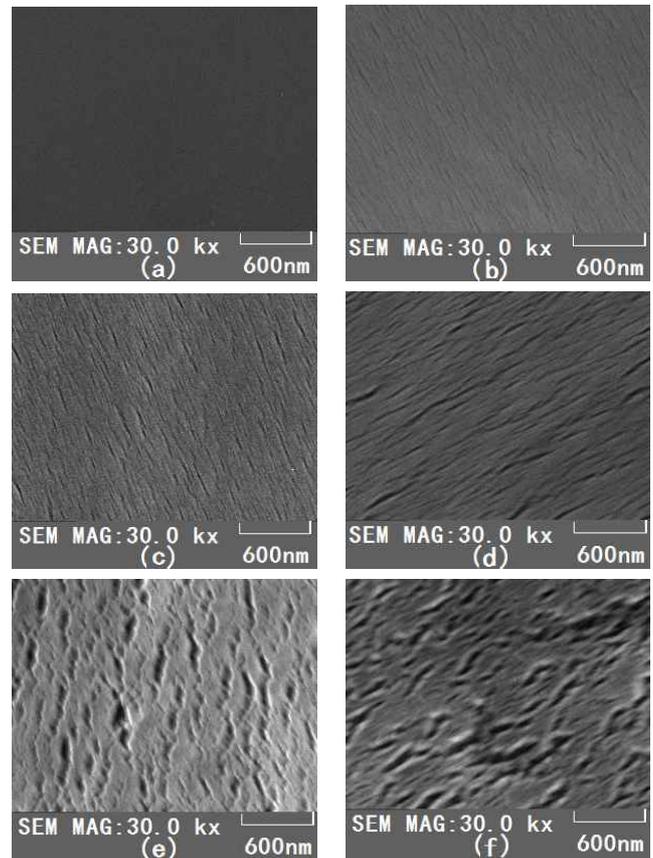


Fig. 1. SEM images of specimens aged for 14/24/72/120/144/182h

absorption is an irreversible chemical reaction. In order to clearly analyze the effect of different moisture absorption methods on the properties of the material, they are separated from total moisture absorption. The way through the physical diffusion (Way 1), crack and slit (Way 2) absorbing moisture are called physical moisture absorption. The other method through the combination of hydrogen bond (Way 3) absorbing moisture is called chemical moisture absorption. They are calculated by equations as shown below:

$$\eta_1 = \frac{W_1 - W_1'}{W_{10}} \times 100\% \quad (3)$$

$$\eta_2 = \begin{cases} \left(\frac{W_1' - W_{10}}{W_{10}} + \frac{W_{20} - W_2}{W_{20}} \right) \times 100\% \\ 0 \quad (\eta_2 < 0) \end{cases} \quad (4)$$

Where η_1 and η_2 are physical and chemical moisture absorption, respectively. W_{10} and W_{20} are the weights of unaged samples. W_1 is the weight of sample after hygrothermal aging. And W_1' is dried weight of W_1 . W_2 is the weight of sample after thermal aging. After thermal aging, the weight loss of drying aged samples are very small, thus its weight loss of caused by drying is not considered.

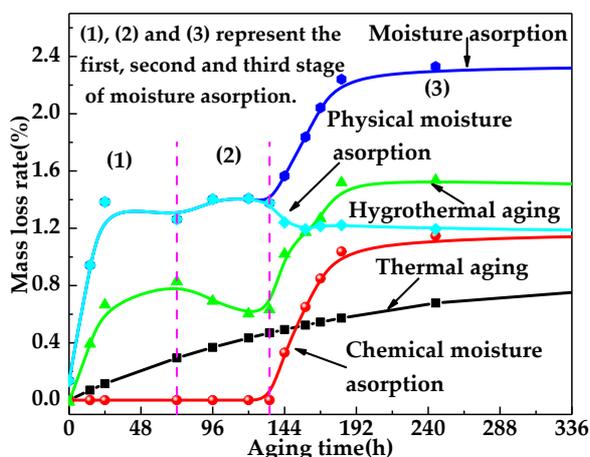


Fig. 2. The moisture absorption after hygrothermal aging

The mass loss of thermal aging η_{ta} and the mass change of hygrothermal aging η_{hta} could be calculated by Eq. (1). The parameters related to moisture absorption are gained by Eq. (2), Eq. (3) and Eq. (4). Results are shown as Fig. 2.

It can be seen that after hygrothermal aging, the weights of samples show increase-decrease-increase-decrease trend. It is found that moisture absorption behavior does not follow the Fick's second law. At the initial hygrothermal aging stage, the physical moisture absorption is the only way to absorb water molecules. After 144 hours of hygrothermal aging, the physical moisture absorption reaches equilibrium and decreases slowly. At this time, chemical moisture absorption begins. The chemical moisture absorption increases rapidly to saturation, and then slowly. As shown in Fig. 2, moisture absorption process can be divided into three stages.

In the first stage, the physical moisture absorption increases rapidly and it is much higher than that of the thermal mass loss during the aging period of 0-72 h. The first absorption peak appears and then the physical moisture absorption reaches the first stage of balance. With the increase of moisture absorption, cracks and slits increase. Thus the moisture absorption rate is accelerated, and then the water molecules begins to enter the material quickly. During the aging period of 72-134 h, the thermal mass loss increases and it is greater than the increase of physical moisture absorption, which leads to the weights of samples decreasing. Moisture absorption results in uneven internal stress, so cracks and slits continue deepening. The amount of moisture absorption is constantly increasing until reaching the second stage of the moisture absorption balance. In the third stage, after aging 134h, the internal damage of the material is serious, and the cracks and slits are gradually evenly distributed. At this stage, chemical moisture absorption appears and physical moisture absorption decreases, which indicates that a part of the physical moisture absorption is converted to chemical moisture absorption [24]. Moisture absorption are higher than that of thermal mass loss so the weights of samples

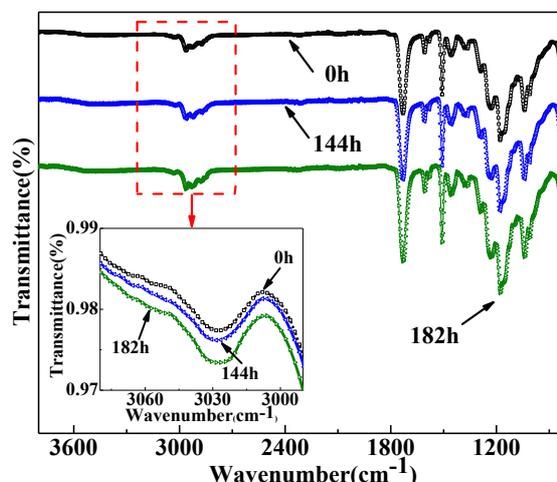


Fig. 3. The FTIR Spectra of aged specimens after drying

Table 1. The CI of samples aged after 0h, 144h and 182h

Aging time	0h	144h	182h
CI	0.4580	0.4825	0.5382

increase, then the water molecules, cracks and slits evenly distribute in the material. Hygrothermal aging reaches the third stage of moisture absorption balance.

3.1.3 FTIR of dried sample after hygrothermal aging

Fig. 3 is the FTIR Spectra of aged specimens after drying.

From Fig. 3, it can be seen that the O-H absorption peak of the dried sample in the range of 3000-3300 cm^{-1} increases after aged 144 hours in hygrothermal environment, which indicates more hydrogen bonds and more chemical moisture absorption. The absorption peak at 1750 cm^{-1} has an increasing tendency, which indicating that the C=O bond is increased.

For clearly presenting carbonyl group changing trend, carbonyl index (CI) [25, 26] is used to calculate the content of carbonyl group shown as Eq. (5).

$$CI = \frac{A_{1750}}{A_{721}} \quad (5)$$

Where A_{1750} and A_{721} are the absorbance of carbonyl group (-CO-) and methylene group (-CH₂-) with deformation vibration, respectively.

Shown in Table 1, the CI of samples aged after 0h, 144h and 182h are 0.4580, 0.4825 and 0.5382 calculated by Eq. (5) respectively, which verifies with aging time increasing, the content of carbonyl group increases. It is possible that the hydroxyl group is oxidized. The reaction equation is shown in Fig. 4. In particular, during the process of hygrothermal aging, the samples are deformed and hydrolyzed to precipitate brown matter. The hydrolysis of the material is the reverse reaction of the polymerization

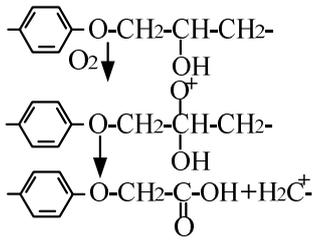


Fig. 4. The oxidation reaction of O-H group

reaction, so the brown matter contains epoxy resin, curing agent, catalyst and other raw materials [27].

3.1.4 The way of absorbing moisture

The moisture is firstly by absorbed by physical diffusion (Way 1) for that the tensile section of material is smooth and has no slit. After the presence of cracks, slits and other moisture absorption channels, the moisture absorption through the Way 1 and 2. When the moisture gradually spreads to the entire sample, the physical moisture absorption achieves balance stage and chemical moisture absorption appears. Finally, the way of absorbing moisture concludes Way 1, 2 and 3.

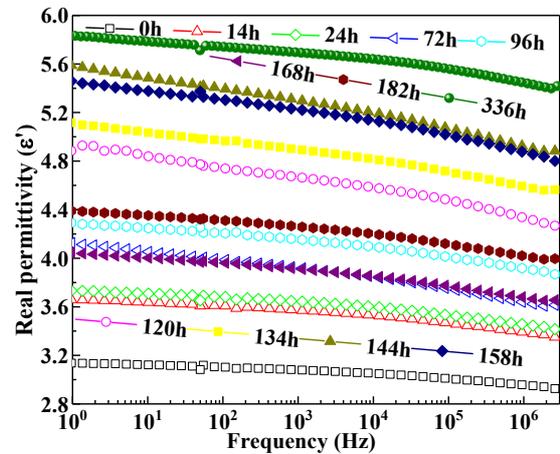
3.2 Dielectric properties

3.2.1 The effect of aging time on the real part of dielectric constant ϵ'

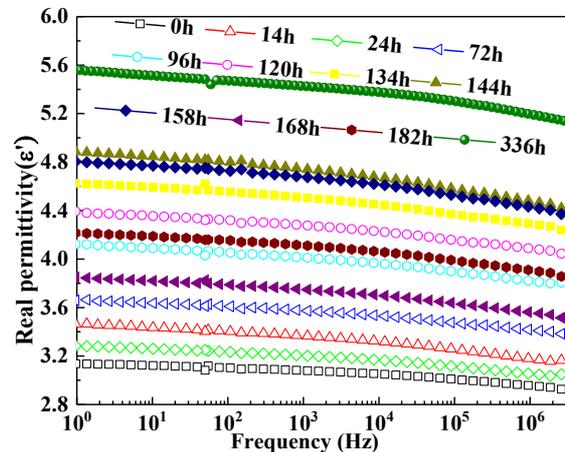
Fig. 5 shows the frequency dependence of the real part of dielectric constant ϵ' . After hygrothermal aging, ϵ' has an increase-decrease-increase trend. Water molecule as a polar molecule and free small molecule produced by the hydrolysis and oxidation of epoxy resin as an impurity, they are positively correlated with ϵ' .

Compared Fig. 5(a) with (b), it can be seen that after removing the physical moisture absorption, ϵ' has an increase – decrease – increase – decrease - increase trend. Chemical moisture absorption is beginning to appear after aged the 144h, its contribution is even, and it is positively correlated with ϵ' . Thus the amount of small free molecules has increase-decrease-increase-decrease-increase trend, and the corresponding time periods are 0-14-24-144-168-182h.

Total moisture absorption is constantly increasing with the increase of time. Before aged 144h, high temperature making the material decomposed and hydrolyzed, a part of the small free molecules are accumulated and fluctuated dynamically. ϵ' increases which indicated that the effect of water molecules absorbed by physical way on the ϵ' is greater than the role of small free molecules. After samples being aged between 144 and 168h, small free molecules get released, thus its amount decreases significantly and ϵ' begins to fall. In the third stage, the rate of thermal loss weakens thus the free small molecule increases. Finally, ϵ' increases. ϵ' is affected by the water molecules and small free molecules which include ϕ -O- ϕ (ϕ represents



(a) ϵ' of samples aged after different times



(b) ϵ' of aged samples after being dried

Fig. 5. The change rule of ϵ' after hygrothermal aging and dry reprocessing

benzene ring), R-CH₂ (R represents organic group) and so on[28].

3.2.2 The effect of aging time on the imaginary part of dielectric constant ϵ''

Fig. 6 shows the frequency dependence of the imaginary part of dielectric constant ϵ'' . After hygrothermal aging, ϵ'' has an increase-decrease-increase-decrease-increase trend, but there are no obvious changes in the positions of relaxation peaks. The value of ϵ'' at low frequency is inversely proportional to the frequency of the aging voltage, which is mainly caused by the dc conductivity. ϵ'' can be described by a modified Debye equation as follows [29]-[31]:

$$\epsilon'' = k_0 \omega^{\alpha_0} + \sum_{i=1}^n \frac{k_i (\omega \tau_{0i})^{1-\alpha_i} \cos \frac{\pi \alpha_i}{2}}{1 + 2(\omega \tau_{0i})^{1-\alpha_i} \sin \frac{\pi \alpha_i}{2} + (\omega \tau_{0i})^{2(1-\alpha_i)}} \quad (6)$$

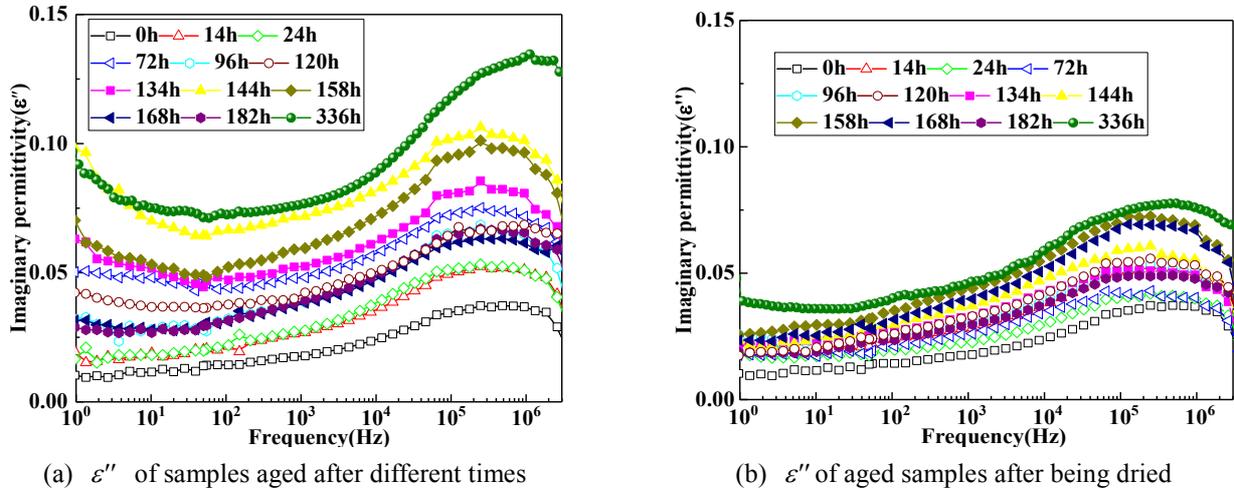


Fig. 6. The change rule of ϵ'' after hygrothermal aging and dry reprocessing

Table 2. The fitting parameters of ϵ'' by Eq. (6)

Aging time	α_0	α_i	k_0	k_i	τ_{0i}
0h	-0.0668	1.6105	0.0100	-0.0537	4.6642E-6
72h	-0.0403	1.4831	0.0507	-0.2233	3.3456E-6
96h	-0.1427	0.6870	0.0289	0.4857	3.7829E-6
144h	-0.1551	0.6544	0.0927	0.6413	3.3409E-6
158h	-0.0681	0.5896	0.0593	0.4396	2.5223E-6
336h	-0.0887	0.6521	0.0876	0.7687	6.5591E-7

Here, α_0 and α_i have relationship with the relaxation time. The case $i=1$ corresponds to the Debye model that has a single relaxation time. ω is the angular frequency $2\pi f$, and τ_{0i} is the relaxation time.

The first part of Eq. (6) is related to temperature-dependence electrical conductivity and the second part to the thermal polarization. The corresponding parameters by fitting Eq. (6) are listed in Table 2. The fitting results of ϵ'' based Debye theory are as shown in Fig. 7. The ϵ'' of unaged and aged samples has one relaxation peak in the 10^0 - 10^6 Hz. Loss of electrical conductivity and polarization increase after hygrothermal aging.

Before aged 72h, water distribution is uneven, water molecules has higher density in some local area which leads to sample conductivity loss increasing sharply. So even if the accumulation amount of small free molecules fluctuates, the amount of water molecules increases greater than that of small free molecules, which causes electrical conductivity and polarization loss to increase. At the same time, ϵ'' increases, which indicated the polarization loss can be more easily affected by water molecules. Aged between 72 and 96h, physical moisture absorption and small free molecules increase, thus polarization loss increases. Compared with before, water molecules are more uniformly distributed and they diffuse from higher density region to lower density region. Thus conductivity loss and ϵ'' reduce. It can be seen that the density of water molecule affects electric conductivity loss and the amount of molecules affects polarization loss.

Aged between 96 and 134h, the accumulation of small

free molecules increases inside samples. Physical moisture absorption decreased slightly. Thus polarization loss slightly decreased. Electrical conductivity loss increases. Aged between 134-144h, the amount of small free molecules rises. Furthermore, the content of total moisture absorption increased slightly, thus electrical conductivity loss increases. These water molecules absorbed by Way 3 have worse polarization ability than those by Way 1 and 2, for that they have hydrogen bonds with hydrophilic group. Thus polarization loss increases and ϵ'' increases. The fourth stage of aging between 144-158h, the total moisture content increases greatly. Thus polarization loss increases. The number of small free molecules decreases. Thus electrical conductivity loss decreases. The fifth stage, total moisture absorption are balance and small free molecules continue to accumulate, resulting in the loss of conductivity and polarization loss and ϵ'' increase.

Especially, the polarization loss can be more easily affected by water molecules than small free molecules. The density of water molecule affects electric conductivity loss and the amount of molecules affects polarization loss. The water molecules absorbed by Way 3 have worse polarization ability than those by Way 1 and 2, for that they have hydrogen bonds with hydrophilic group.

3.3 Space charge behavior

3.3.1 Space charge density distribution of aged samples during depolarization

Fig. 8 shows that the space charge distribution density of epoxy resin samples during depolarization after applied -20 kV/mm DC field with 30 min.

From Fig. 8, it can be seen that the space charge dissipation law of aged samples with different aging stages is basically similar that the residual space charge in samples gradually decreased with time increasing. In particular, after the emergence of chemical moisture

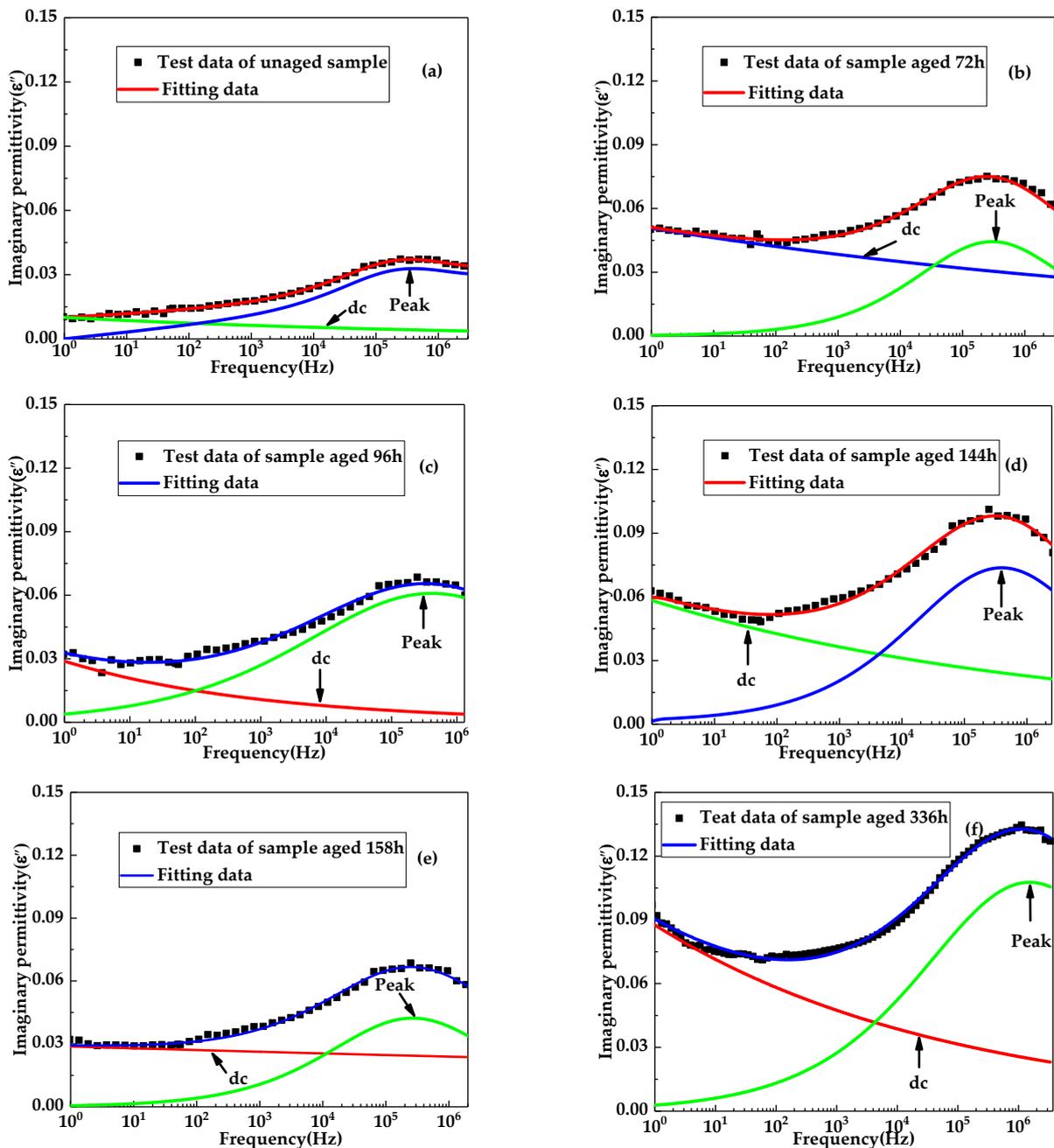


Fig. 7. The fitting results of ϵ'' based Debye theory

absorption, the density of residual space charge increases with the decay rate of residual space charge decreases significantly.

3.3.2 The decay property of total space charge during depolarization

In order to further investigate the space charge characteristics of aged samples, the total amount of space charge is used to quantitatively describe the space charge accumulation phenomenon, which can be calculated based on the charge density distribution, as shown in Eq. (7).

$$Q(t) = \int_0^d |\rho(x,t)| S dx \tag{7}$$

Where $Q(t)$ refers to the total amount of space charge into samples, d refers to the thickness of samples, $\rho(t)$ refers to the density of space charge, S refers to the electrode area.

Fig. 9 shows that the total space charge of epoxy resin samples during depolarization after applied -20 kV/mm DC field with 30 min. It can be seen that the residual charge increases with the increase of aging time. In addition, the decay rate of space charge becomes slow with aging time

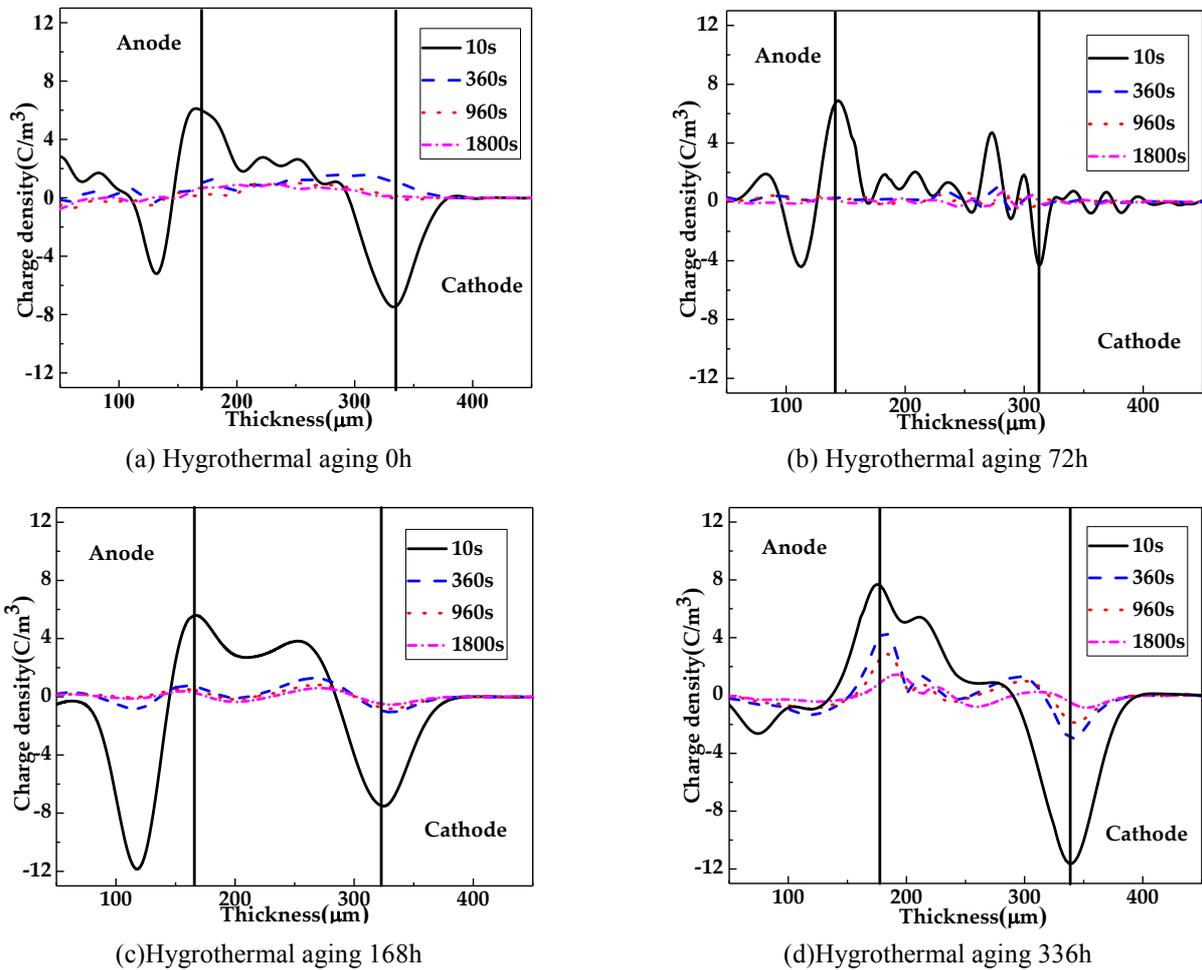


Fig. 8. Space charge density distribution of aged samples during depolarization

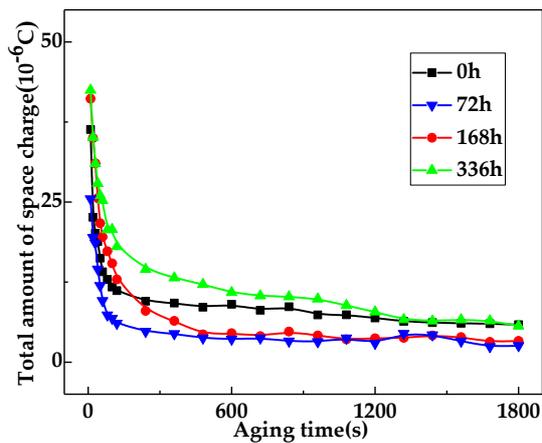


Fig. 9. The decay property of total space charge during depolarization

increasing. The result of SEM images and FTIR show that after hygrothermal aging, cracks and holes are continuously generated and new chemical bonds have been produced, which indicates internal traps of material have increased causing the decay rate of space charge to slow

down. Thus with the increase of aging time, the decay rate of total space charges declines. Especially, when the chemical moisture absorption occurs inside the material, the residual space charges increase obviously and the dissipation rate of the charges is slowed down obviously. The hygrothermal aging has slowed down the movement of the space charge.

4. Conclusion

The work presented in this paper concerns with the properties of epoxy resin under hygrothermal aging. Several significant findings are concluded. The moisture absorption of epoxy resin mainly concludes physical and chemical moisture absorption. When the physical absorption moisture reaches third equilibrium stage, chemical absorption moisture begins to appear. With the aging time increasing, the distribution of water molecules, cracks and slits tend to be uniform. The moisture absorption process undergoes three equilibrium stages and it does not follow the Fick's second law. With aging time

increasing, ε' has an increase-decrease-increase trend and ε'' has an increase-decrease-increase-decrease-increase trend. The polarization loss can be more easily affected by water molecules than small free molecules. The density of water molecule affects electric conductivity loss. The number of traps increases after hygrothermal aging, thus the decay rate of space charge slows down. The residual space charge in samples gradually decreased and the decay rate of space charge becomes slow with aging time increasing.

Acknowledgments

The work is supported by the National Natural Science Foundation (51637004).

References

- [1] Lin Y. C., Chen X. and Wang. Z. P., "A study of degradation of ACF joints in chip-on-glass assemblies," Thermal and Thermomechanical Phenomena in Electronics Systems, 2006.
- [2] Park C. H., Lee S. J. Lee, T. H. and Kim H. J., "Characterization of an acrylic pressure-sensitive adhesive blended with hydrophilic monomer exposed to hygrothermal aging: assigning cloud point resistance as an optically clear adhesive for a touch screen panel," *Reactive and Functional Polymers*, vol. 100, pp. 130-141, 2016.
- [3] Liang Mu and Wong K. L., "Electrical performance of epoxy resin filled with micro particles and nanoparticles," *Energy Procedia*, vol. 110, pp. 162-167, 2017.
- [4] Jiang Z., "Simulation and analysis of leakage magnetic field in epoxy resin cast dry-type transformer with typical structure," *Transformer*, 2017.
- [5] Zhang Y. L., Shi, X. Q. and Zhou W., "Effect of hygrothermal aging on interfacial reliability of flip chip on board (FCOB) assembly," *Electronics Packaging Technology Conference*, 2004.
- [6] Grammatikos S., Mark E. and Ball R. J., "Effects of hygrothermal aging on pultruded glass fibre reinforced polymers: a complimentary study," *International Conference on Advanced Composites in Construction Acic*, 2015.
- [7] Chhanda N. J., Suhling J. C. and Lall P., "Effects of Moisture Exposure on the Mechanical Behavior of Polymer Encapsulants in Microelectronic Packaging," *ASME 2013 International Technical Conference and Exhibition on Packaging and Integration of Electronic and Photonic Microsystems*, 2013.
- [8] Bhatti I. S. and Yan Z., "Comparative hygrothermal ageing analysis of glass and carbon fiber polymer composites: Numerical and experimental study," *International Bhurban Conference on Applied Sciences and Technology. IEEE*, 2015.
- [9] Bao L. R. and Yee A. F., "Effect of temperature on moisture absorption in a bismaleimide resin and its carbon fiber composites," *Polymer*, vol. 43, pp. 3987-3997, 2002.
- [10] Bao L. R. and Yee A. F., "Moisture diffusion and hygrothermal aging in bismaleimide matrix carbon fiber composites-part I: uni-weave composites," *Composites Science & Technology*, vol. 62, pp. 2099-2110, 2002.
- [11] Adamson M. J., "Thermal expansion and swelling of cured epoxy resin used in graphite/epoxy composite materials," *Journal of Materials Science*, vol. 15, pp. 1736-1745, 1980.
- [12] Zheng Q. and Morgan R. J., "Synergistic Thermal-Moisture Damage Mechanisms of Epoxies and Their Carbon Fiber Composites," *Journal of Composite Materials*, vol. 27, pp. 1465-1478, 1993.
- [13] Soles C., and Yee, A. F., "A discussion of the molecular mechanisms of moisture transport in epoxy resins," *Journal of Polymer Science Part B Polymer Physics*, vol. 38.5, pp. 792-802, 2000.
- [14] Zou C., Fothergill J. C. and Rowe S. W., "The effect of water absorption on the dielectric properties of epoxy nanocomposites," *IEEE Transactions on Dielectrics & Electrical Insulation*, vol. 15, pp. 106-117, 2008.
- [15] Jefferson G. D., Farah B., Hempowicz M. L. and Hsiao K. T., "Influence of hygrothermal aging on carbon nanofiber enhanced polyester material systems," *Composites Part B Engineering*, vol. 78, pp. 319-323, 2015.
- [16] Rain P., Brun E., Guillermin C. and Rowe S. W., "Experimental model of a silica/epoxy interface submitted to a hygrothermal aging: a dielectric characterization," *IEEE Transactions on Dielectrics & Electrical Insulation*, vol. 19, pp. 343-351, 2010.
- [17] Li Y., Li R., Huang L., Wang K. and Huang X., "Effect of hygrothermal aging on the damage characteristics of carbon woven fabric/epoxy laminates subjected to simulated lightning strike," *Materials & Design*, vol.99, pp.477-489, 2016.
- [18] Jr J. H. S. A., Souza S. D. B., Botelho E. C. and Amico S. C., "Carbon fiber-reinforced epoxy filament-wound composite laminates exposed to hygrothermal conditioning," *Journal of Materials Science*, vol. 51, pp. 4697-4708, 2016.
- [19] Rain P., Brun E., Guillermin C. and Rowe S., "Hygrothermal ageing of a filled epoxy resin: measurements of the insulation properties and qualitative modelling," *2010 International Conference on Solid Dielectrics*, Potsdam, Germany, 4-9 July, 2010.
- [20] Dogan A. and Atas C., "Variation of the mechanical properties of e-glass/epoxy composites subjected to

hygrothermal aging,” *Journal of Composite Materials*, vol. 50, no.1, 2016.

- [21] Zhao Y., Wang Z., Keey S. L. and Chai G. B., “Long-term viscoelastic response of e-glass/bismaleimide composite in seawater environment,” *Applied Composite Materials*, vol. 22, pp. 693-709, 2015.
- [22] Haghghi-Yazdi M. and Lee-Sullivan P., “FTIR analysis of a polycarbonate blend after hygrothermal aging,” *Journal of Applied Polymer Science*, vol. 132, no. 3, 2015.
- [23] Xiao G. Z., Delamar M. and Shanahan M. E. R., “Irreversible interactions between water and dgeba/dda epoxy resin during hygrothermal aging,” *Journal of Applied Polymer Science*, vol. 65, pp. 449-458, 2015.
- [24] Adamson M J., “Thermal expansion and swelling of cured epoxy resin used in graphite/epoxy composite materials,” *Journal of Materials Science*, vol. 15, pp. 1736-1745, 1980.
- [25] Pan Hong and Zhao Tao, “Antibacterial property of fabric treated with a fiber-reactive chitosan derivative,” *Journal of Textile Research*, vol. 32.2, pp. 96-101, 2011.
- [26] Barbes, L., Radulescu, C. and Stihl, C., “Atr-ftir spectrometry characterisation of polymeric materials,” *Romanian Reports in Physics*, vol. 66.3, 2014.
- [27] Pan Zuren, *Polymer Chemistry, Enhanced Edition*, Chemical Industry Press, 2007, p. 362-363.
- [28] Wang M. Xu X., Ji J., Yang Y., Shen J. and Ye M., “The hygrothermal aging process and mechanism of the novolac epoxy resin,” *Composites Part B Engineering*, vol. 107, pp. 1-8, 2016.
- [29] Zhao X., Liao R., Liang N., Yang L. and Li J., “Role of defects in determining the electrical properties of ZnO ceramics,” *Journal of Applied Physics*, vol. 116, pp.014103-014103-10, 2014.
- [30] Jia R., Zhao X., Li J. and Tang X., “Colossal breakdown electric field and dielectric response of Al-doped $\text{CaCu}_3\text{Ti}_4\text{O}_{12}$ ceramics,” *Materials Science & Engineering B*, vol. 185, pp.79-85, 2014.
- [31] Rhaïem A. B., Hlel F., Guidara K. and Gargouri M., “Dielectric relaxation and ionic conductivity studies of $[\text{N}(\text{CH}_3)_4]_2\text{Cu}_{0.5}\text{Zn}_{0.5}\text{Cl}_4$,” *Journal of Alloys & Compounds*, vol. 463, pp. 40-445, 2008.



Youyuan Wang He received the M.S. and Ph.D. degrees in electrical engineering in 2003 and 2008, respectively from Chongqing University, Chongqing, China. He is currently a professor of High Voltage and Insulation Technology Department at Chongqing University. His major research interests include dielectric properties and space charge behavior of nano-

composites, online detection of insulation condition in electrical devices, and insulation fault diagnosis for high voltage equipment. He is an author and coauthor of more than 50 journal papers and 30 papers published in proceedings of international conferences.



Yu Liu She was born in Shandong Province, China in 1993. She received the B.S degree in 2015 from Guangxi University, China. Now she is studying for M.S. degree in the School of Electrical Engineering, Chongqing University. Her major research interests include the micromorphology, space charge behavior, dielectric properties in the insulation materials and the molecular dynamic stimulation of polyethylene.



Kun Xiao He received B.S and M.S. degree in 2014 and 2017 respectively from Chongqing University. His research interests focus on self-healing materials and fault diagnosis of power equipment.



Can Wang He was born in Chongqing Province, China in 1991. He received the B.S degree in 2013 from Chongqing University, China. He is now a Ph.D. candidate in the School of Electrical Engineering, Chongqing University. His major research interests include nanomaterials modification, space charge behavior, dielectric properties in the insulation materials and the molecular dynamic stimulation of polyethylene.



Zhanxi Zhang He received B.S degree in 2016 from North China Electric Power University. He is now a new Ph.D. candidate in the School of Electrical Engineering, Chongqing University. His research interests are self-healing materials and dielectric properties in the insulation materials.