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# Study of Compatibility Between Epoxy Resin and C<sub>4</sub>F<sub>7</sub>N/CO<sub>2</sub> Based on Thermal Ageing

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**ABSTRACT** Fluorinated nitrile ( $C_4F_7N$ ) is a potential SF<sub>6</sub>-alternative gas in Gas-Insulated Transmission Line (GIL) due to its lower impact to global warming and excellent dielectric strength. To ensure the long-term physical and chemical stability at the interface between  $C_4F_7N/CO_2$  gas and epoxy resin assembly in GIL, laboratory thermal ageing tests on the material compatibility were carried out. In order to evaluate the compatibility, the authors set up a gas-solid compatibility test platform in this paper. Thermal ageing tests of  $C_4F_7N/CO_2$  and epoxy resin were set up at different temperatures. Also, the groups of SF<sub>6</sub> and He gas were adopted to evaluate the influence of temperature. The compatibility between the gas and solid insulating materials in contact was evaluated in terms of the changes of insulation performance after over long period of tests. The changes of gas compositions and solid contents were also assessed. Dielectric spectroscopy, surface flashover voltages, surface morphology, and chemical elements of epoxy resin and gas composition before and after the ageing tests were compared. The surface insulation of epoxy resin decreased only after ageing reached 160 °C, when a small amount of  $C_3F_6$  and trimer of  $C_4F_7N$  were found. Under the normal operation of GIL (below 90 °C), the compatibility of epoxy resin with 9 %  $C_4F_7N/91$  % CO<sub>2</sub> is as good as epoxy resin with SF<sub>6</sub>.

**INDEX TERMS** C<sub>4</sub>F<sub>7</sub>N/CO<sub>2</sub>, epoxy resin, compatibility, SF<sub>6</sub>-alternative gas.

#### I. INTRODUCTION

In 2015, the 3M Company introduced the environmentally friendly insulating gas fluorinated nitrile (C<sub>4</sub>F<sub>7</sub>N) which has stable chemical properties, great dielectric (1.9 times that of SF<sub>6</sub>), environmental properties (Global Warming Potential value of 2100) and about 30 years of atmospheric life. Therefore, it has superior environmental performance over  $SF_6$  [1]. Since  $C_4F_7N$  has a high liquefaction temperature (-4.7 °C) [2], it is necessary to mix with buffer gases with lower liquefaction temperature such as CO2, N2 or air to meet the operational requirements of the equipment in winter. Alstom and Grid Solution of GE developed the mixture of C<sub>4</sub>F<sub>7</sub>N and CO<sub>2</sub> "g3 (green gas for grid)" [3]. The results showed that 9 % C<sub>4</sub>F<sub>7</sub>N/91 % CO<sub>2</sub>, under uniform electric field and equal pressure, reached the same insulation strength as pure  $SF_6$  [4]. As a result, it has become one of the potential alternative gases for SF<sub>6</sub>.

To ensure the stability of  $C_4F_7N/CO_2$  mixture in gas insulation equipment, it is necessary to ascertain the compatibility of solid materials in equipment after long period of contact with the gas. Previously the authors carried out research on the compatibility of gas and solid insulating materials in Gas-Insulated Transmission Line (GIL) to determine the changes of insulation performance caused by long-term contact between the two materials, and the changes of gas composition and solid material [5] over time.

The study of gas-solid compatibility in theory and experiment was successful [6]. In reference [7], the dissociation adsorption of  $C_4F_7N$  on copper and aluminum surfaces was studied theoretically by using a material studio simulation software. The results showed that  $C_4F_7N$  had good compatibility with copper and aluminum, and the compatibility of  $C_4F_7N$  with aluminum was better than that copper, which explained the reason why  $C_4F_7N$  had good compatibility with metal theoretically. F. Kessler and other scholars studied the experimental compatibility between  $C_4F_7N$  and desiccant. By making full contact between the two materials in a sealed container, they found that  $C_4F_7N$  could react with desiccant

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to produce a small amount of decomposition products, such as  $C_3F_6$  and  $C_3H_2F_6$  [8].

It should be noted that, the compatibility between insulating materials and gas is still not fully understood. If adjacent insulating materials are incompatible with  $C_4F_7N/CO_2$ , flashover fault may occur even under normal operating conditions. EPRI Co. of the USA found that hydantoin epoxy resin showed physical and chemical reactions with a variety of insulating gases (including SF<sub>6</sub>), and the compatibility between them was poor [9]. However, the temperature of the ageing test was as high as 220 °C to 225 °C, which is very different from the actual operating conditions of electrical equipment (90 °C). Therefore, the self-decomposition of hydantoin epoxy resin due to high temperature cannot be excluded. The compatibility between epoxy resin and  $SF_6$ decomposition products in GIS was studied in [10] and [11]. It was found that the surface resistance of epoxy resin insulator with silicon filler decreased by 5 orders in magnitude  $(10^{14}-10^9)$ , while that of aluminum filler did not change significantly  $(10^{14})$ . It showed that the formula of epoxy resin filler had some influence on gas-solid compatibility.

Therefore, previous research has the following problems. First, the tests were not consistent with the actual operational condition of gas insulated equipment. Second, previously reported researches were only concerned about the change of gas purity or the change of material properties after the tests. No considerations were given to the changes caused by gas-solid interaction at the same time. Finally, the physico-chemical properties of new C<sub>4</sub>F<sub>7</sub>N/CO<sub>2</sub>, including material compatibility, have gradually become the research hotspot, while the effect of thermal ageing characteristics of epoxy resin in C<sub>4</sub>F<sub>7</sub>N/CO<sub>2</sub> have rarely been studied.

Concerning the limitation of current research, the authors built a thermal acceleration test platform to study the compatibility of epoxy resin and  $C_4F_7N/CO_2$  under 90 °C, 125 °C, and 160 °C. The composition of mixed gas was analyzed by gas chromatography-mass spectrometry (GC-MS), and the surface chemical elements of solid samples was analyzed by Fourier transform infrared spectroscopy (FTIR). The surface morphology of solid samples was observed by scanning electron microscopy (SEM). The dielectric spectroscopy measurement and surface flashover voltages were used to characterize the insulation properties of the samples.

#### **II. EXPERIMENTAL METHOD**

#### A. EXPERIMENTAL MATERIALS

Recently, gas insulated equipment such as GIL or GIS mostly used Bisphenol-A epoxy resin as the solid insulating materials. Because of its readily available raw material and low cost, it has been widely used, accounting for 90 % of all epoxy resins [12]. The Bisphenol-A epoxy resin material industrial used in the test was provided by the electrical equipment manufacturer Pinggao Group Co. Ltd, shown in Table 1. Sample A was mainly used for surface flashover voltage test.

#### TABLE 1. Roperties of Bisphenol-A Epoxy Resin.

Properties	Values
Epoxy equivalent (g/eq)	380±20
Density (g/cm <sup>3</sup> ) (120 °C)	1.1~1.3
Dielectric strength (kV/mm)	22~25
Bending strength(MPa)	110~120
Impact strength (kJ/m <sup>2</sup> )	10~12
Tensile strength (MPa)	75~85
Fillers( $\alpha$ -Al <sub>2</sub> O <sub>3</sub> , pbw)	320~330
Viscosity (mPa/s) (120 °C)	600~900
Flash point(°C)	>200
Glass transition temperature (°C)	124.2
Thermal decomposition temperature	
(Heating speed 10K/min) (°C)	350

**TABLE 2.** Comparison of Physicochemical Properties of  $C_4 F_7 N$ , 9 %  $C_4 F_7 N/91$  %  $CO_2$ , and  $SF_6$  [13].

Properties	$C_4F_7N$	9 % C <sub>4</sub> F <sub>7</sub> N /91 % CO <sub>2</sub>	$SF_6$
GWP (100 years)	2100	42	23500
Ozone depletion potential (ODP)	0	0	0
Liquefaction temperature (°C)	-4.7	-25	-64
Insulation ability (relative to SF <sub>6</sub> )	1.9	0.80	1

Its height was 10mm and diameter was 15 mm. Sample B was used for dielectric spectroscopy measurement. Its thickness was 1 mm and its diameter was 40 mm.

Previous research showed that when the proportion of  $C_4F_7N$  was 9 %, the 9 % $C_4F_7N/91$  % $CO_2$  mixture had the same insulation strength as SF<sub>6</sub> under power frequency [4]. Therefore, to practically replace SF<sub>6</sub>, the authors chose 9 % $C_4F_7N/91$  % $CO_2$  gas mixture to carry out the test. In order to compare the interaction between different gases and epoxy resin, two groups of inert gas were set up: 100 %SF<sub>6</sub> and 100 %He, among which SF<sub>6</sub> is the gas insulating medium used in GIL currently and He was set to distinguish the effect of temperature on the performance of epoxy resin since He could not react with epoxy resin.

#### **B. EXPERIMENTAL PLATFORM**

The thermal acceleration test platform in this paper, designed to study the compatibility, includes an oven and sealed tanks. The sealed tank is made of 304 stainless steel with inner diameter of 50 mm, height of 200 mm, and volume of about 400 ml. It can withstand the gas pressure of 0.8 MPa. A metal frame is placed in the tank body to fix the epoxy sample and to fully contact with the gas in the tank.

#### C. EXPERIMENTAL CONDITIONS

Glass transition of epoxy resin is the process of changing from glass state to high elastic state. When epoxy resin begins glass transition, the corresponding temperature is called glass transition temperature ( $T_g$ ). When epoxy resin is in the glass



FIGURE 1. Samples of epoxy resin.

transition zone, the mechanical properties change greatly, the movement of macromolecular chain of epoxy resin is more free and active, and the interaction with gas molecules is more obvious. This paper focuses on the changes of the epoxy resin properties and gas composition when the gas temperatures are below, above, and in the glass transition range.

The maximum allowable temperature of the directly buried GIL shell is 40 °C - 50 °C, the GIL installed in the tunnel is 60 °C - 70 °C, the maximum temperature in GIL can reach 90 °C [14] under normal operation, and the glass transition temperature of the epoxy resin used in GIL is 124.2 °C. Therefore, the thermal acceleration temperature is set to 90 °C, 125 °C, and 160 °C in order to investigate the different changes of the epoxy resin properties and gas composition, which the temperatures are below, above, and in the glass transition range. The ageing experiments were performed for a duration of 696 hours (29 days) according to international standards IEC 60811-1.

In order to avoid the influence of impurities on the test results, the epoxy resin samples and test equipment were cleaned with anhydrous ethanol before the test, and then dried in an oven at 100 °C for 24 hours and cooled to room temperature. After that, the sealed tank was filled with the test gas to 0.1 MPa for each test and then pumped out. The process was repeated three times to remove the gas impurities inside the tank. The absolute pressure of the insulating gas in GIL was 0.4 MPa - 0.8 MPa. Considering the thermal expansion effect of the gas, the absolute pressure of the test was set to 0.5 MPa.

#### **D. TEST METHODS**

#### 1) DIELECTRIC PERFORMANCE TEST

The dielectric spectroscopy measurements of epoxy resin samples before and after ageing were measured in the frequency range of between  $10^{-1}$  Hz and  $10^{6}$  Hz at room temperature (24.7 °C-25.4 °C), using a NOVOCONTROL GMBH-GEARMY high resolution dielectric analyzer. The diameter of brass plate electrodes was 40 mm with a spring so as to get a perfect contact between the sample and electrode. The relative humidity of the measurements was 40 %. Under each compatibility test condition, the average value of three samples were taken for further analysis.

#### 2) SURFACE FLASHOVER VOLTAGE TEST

The power frequency flashover voltage of epoxy resin samples in C<sub>4</sub>F<sub>7</sub>N/CO<sub>2</sub> mixture gas was measured. The YDTW-100/300 PD-free test transformer was selected and the flashover voltage test chamber is shown in Figure 2. The epoxy resin sample was placed between two Rogowski electrodes. The chamber was filled with 9 %  $C_4F_7N/91$  % CO<sub>2</sub> mixed gas, and the air pressure was gradually increased from 0.1 MPa to 0.3 MPa at an interval of 0.05 MPa. Surface flashover tests were implemented in a sealed chamber with the temperature 10 °C -15 °C and humidity less than 10%. Three samples after ageing under the same condition were taken, and the flashover tests were repeated five times for each sample in different gas pressure.



FIGURE 2. The chamber for surface flashover voltage tests.

#### 3) SEM TESTS

Scanning electron microscope (SEM) was used to observe the changes of surface morphology of epoxy resin samples at 500, 1000, and 2000 times. The instrument model was Fei quanta 200, and the model of energy spectrum accessory was EDAX Genesis 2000.

#### 4) FTIR TESTS

The surface composition of epoxy resin was analyzed by Fourier transform infrared spectroscopy (FTIR). Since it was not easy to directly measure the infrared spectrum of polymer samples by the transmission method, the attuned total reflection (ATR) accessory was used for testing [15]. The model of the instrument used was Bruker vertex 70, with a spectral range of  $4000 \text{ cm}^{-1}$ -  $400 \text{ cm}^{-1}$ .

#### 5) GC-MS TESTS

Gas chromatography mass spectrometry (GC-MS) was used to analyze the test gas and its decomposition products after ageing. The model of the instrument was HP 7890A + 5975C MSD. The injection mode was manual needle injection with the volume of 100  $\mu L$ . The carrier gas was helium with purity greater than 99.99 % and the air inlet temperature was 200 °C. The temperature was kept at 35 °C for 15min, then rose to 250 °C at the speed of 10 °C/min and was kept for 5 min.

#### **III. EXPERIMENTAL RESULTS**

#### A. DIELECTRIC SPECTROSCOPY RESULTS

The real and imaginary relative permittivity of the epoxy resin aged in different gases are plotted in Figures 3 to 5, respectively. There was a clear reduction in  $\varepsilon'$  with the increase of frequency in the range of this study. It is known that  $\varepsilon'$  of the epoxy resin and its micron composites are found to be governed by the polarization [16]. In the low-frequency range, the orientable dipoles rotate in the same direction at the same frequency, and dipoles can orient well under the applied electric field. Therefore,  $\varepsilon'$  is high when the polarization loss is small. In the high-frequency range, the directional rotation of dipoles lags behind the applied electric field, so  $\varepsilon'$  decreases with the increase of frequency.



FIGURE 3. The dielectric spectroscopy of epoxy resin after ageing in 9 %  $C_4F_7N/91$  %  $CO_2$  (a) Real part, (b) Imaginary part.

With the increase of ageing temperature, the mobility of molecular chains increased, which led to the decrease of dipole binding force, enhancement of the polarization and the increase of polarization loss. Samples after ageing all



**FIGURE 4.** The dielectric spectroscopy of epoxy resin after ageing in 100 % SF<sub>6</sub> (a) Real part, (b) Imaginary part.

showed higher  $\varepsilon'$  than original one. At the same time, in the test frequency band, the change trend of  $\varepsilon'$  was similar after ageing in three kinds of gas atmosphere.

In the low-frequency range, the dielectric loss is all contributed by the conductivity loss [17].  $\varepsilon''$  of epoxy resin before and after ageing under different temperatures showed different levels of increase from  $10^{-1}$  Hz -  $10^{2}$  Hz. It is noted that the original sample experienced more obvious increase compared with the samples after ageing. This may be caused by shallow traps on the original sample, leading to an increase in the electrical conductivity [18]. After ageing in different kinds of gases, shallow traps became deep traps partly. Deep traps can trap charges easily, inhibit the injection of bulk charges, and limit the directional movement of carriers [19], which leads to the decrease of electrical conductivity and result in decrease of  $\varepsilon''$ . According to Figure 3 (b), Figure 4 (b), and Figure 5 (b), after ageing at 90 °C, the increasing trend of  $\varepsilon''$  aged in C<sub>4</sub>F<sub>7</sub>N/CO<sub>2</sub> was less obvious than that in SF<sub>6</sub> and He, which indicates that the



**FIGURE 5.** The dielectric spectroscopy of epoxy resin after ageing in 100 % He (a) Real part, (b) Imaginary part.

dc conductivity of epoxy resin aged in C<sub>4</sub>F<sub>7</sub>N/CO<sub>2</sub> is smaller. When the motion of molecular chain was close to the thermodynamic equilibrium, the frequency corresponding to the minimum value of  $\varepsilon''$  may move to lower value [20]. At the same time,  $\varepsilon''$  increased with the improvement of ageing temperature at power frequency (50 Hz). It implies that the ac conductivity of aged sample will increase with the increase of ageing temperature. With the ageing time further extended, the ac conductivity will finally be greater than that of the original sample.

In the high-frequency range,  $\varepsilon''$  is mainly related to polarization [21]. When the polarization degree of dipoles after the ageing test was enhanced, the polarization loss was higher than that of the original sample in the high frequency range. When the ageing temperature was lower than T<sub>g</sub>, the molecular chain segments of epoxy resin were frozen, and only the chain link, branch or side base could move. With the increase of temperature, the main chain of epoxy resin in frozen changed to moveable state in the environment above  $T_g$ , and the molecular polarization ability increased [22]. Therefore, in the range of  $10^2$  Hz  $-10^6$  Hz,  $\varepsilon''$  of the sample aged in the environment of 125 °C and 160 °C was higher than that of 90 °C. In the high-frequency range, it showed a typical  $\beta$ relaxation of local motions of epoxy resin chains. The higher mobility caused by ageing test led to the movement of peak to high-frequency range [23].

**TABLE 3.** Minimum Corresponding Frequency of  $\varepsilon''$  (Hz).

Temperature	9 % C <sub>4</sub> F <sub>7</sub> N/ 91 % CO <sub>2</sub>	100 %SF <sub>6</sub>	100 %He
90 °C	26.11	33.94	50.00
125 °C	11.88	11.88	11.88
160 °C	9.14	11.88	11.88

#### **B. SURFACE FIASHOVER VOLTAGE TESTS**

When a solid dielectric is brought into gas insulation space, a distortion arises in the electrostatic field due to the permittivity difference. This distortion particularly enhances the field strength in the thin slit at the contact between the solid dielectric and metal electrode, which may also reduce the flashover [24]. Where the solid dielectric and the electrode contacting is called triple junction point, field singularity or enhancement occurs at the point since it consists of three media: the electrode, the solid spacer and a gaseous dielectric [25]. The surface flashover along the solid insulation-gas interface should be avoided in the design of gas-insulated equipment. The insertion of spacers not only distorts the electric field because of the difference of permittivity, but also accumulates and stores charge [26].

For the 50-Hz AC voltage tests, the voltage was raised to 75% of the expected flashover voltage at a rate of 10 kV/s, and then raised until flashover at a rate of 5 kV/s. The peak value of voltage was recorded. The time interval between two successive tests was 3 minutes. During the test, when the gas pressure was lower than 0.25 MPa, the surface flashover voltage of epoxy resin increased by about 20 kV for every 0.05 MPa rise of gas pressure. However, when the gas pressure continued to increase, surface flashover voltage increased to a saturation trend and the dispersion improved. When the gas pressure in the test chamber was less than 0.2 MPa, corona may produce at triple junction point, and the accumulated ions made the electric field more uniform. There was no obvious difference of flashover voltage value between the aged sample and the original sample under low gas pressure. With the increase of gas pressure, the development of discharge was faster, and the influence of charge accumulation on the electric field decreased, which caused the increase of inhomogeneity of electric field at triple junction point. Therefore, the differences of flashover voltage values became more and more obvious under high gas pressure.

By comparing Figure 6(a), Figure 6(b), and Figure 6(c), it is noted that after ageing in the environment of 90 °C and 125 °C, the surface flashover voltages epoxy resin did not



FIGURE 6. The results of surface flashover voltage tests 9 %  $C_4F_7N/91$  %  $CO_2$ , (b) 100 %  $SF_6$ , (c) 100 % He.

decrease. While the flashover voltages decreased obviously after ageing at 160 °C. Since the absolute pressure of the insulating gas in GIL was 0.4 MPa - 0.8 MPa, once the flashover fault occurred, it resulted in the ambient temperature being

higher than  $T_g$  for a long duration, and the surface insulation performance of epoxy resin became significantly reduced. At the same time, it was found that there were no obvious differences in the surface flashover voltages of epoxy resin aged in different gases, indicating that the decrease of surface insulation performance was caused by temperature rather than gas-solid reaction. After ageing at high temperature, the molecular chains of epoxy resin broke, more and more free radicals were produced by degradation, and internal defects were caused by impurities or air gap in the material, reducing the surface flashover voltage of aged samples.

#### C. SEM TESTS

The surface morphology of epoxy resin was measured by scanning electron microscope after the thermal ageing test. A range of sample surface was selected and magnified to 500, 1000 and 2000 times, respectively. The results of SEM tests before and after ageing at 160 °C magnified to 2000 times is shown in Figure 7. Comparing the aged sample with the original sample, no obvious precipitated crystals or fault sections were found on the surface of the aged epoxy resin, indicating that short-term thermal ageing test in different gas environment did not make a great impact on the surface morphology of epoxy resin.

#### D. FTIR TESTS

The FTIR spectrum of samples before and after ageing at 160 °C is shown in Figure 8. It is found that the peak time and absorbance of the sample after the ageing test in different gas environments were consistent with that of the original sample, and they were highly matched, indicating that no new substance was found on the surface of epoxy resin. The filler of epoxy resin was  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> micron powder, and the four characteristic peaks [27] of  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> are 826 cm<sup>-1</sup>,  $642 \text{ cm}^{-1}$ ,  $601 \text{ cm}^{-1}$  and  $453 \text{ cm}^{-1}$ , respectively. Compared to the spectrum of the original sample, the four characteristic peaks of Al<sub>2</sub>O<sub>3</sub> were more obvious after the test, indicating that more Al<sub>2</sub>O<sub>3</sub> was detected on the surface layer of the sample after ageing, due to the high temperature in the ageing test results in  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> fillers closer to the surface of epoxy resin [15]. After comparison, it seemed that the characteristic peak of  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> after ageing in 9 % C<sub>4</sub>F<sub>7</sub>N/91 % CO<sub>2</sub> became more obvious than that in 100 % SF<sub>6</sub> and 100 % He.

#### E. GC-MS TESTS

The authors did the gas chromatography mass spectrometry test of all the gas compositions after ageing tests. No gas impurities were detected in GC-MS test after 90 °C and 125 °C thermal ageing tests. When the ageing temperature was increased to 160 °C, the impurity gas was detected in 9 % C<sub>4</sub>F<sub>7</sub>N/91c% CO<sub>2</sub> group, as shown in Figure 9. The air in Figure 9 was mixed when taking gas from air bag. Hexafluoropropylene (C<sub>3</sub>F<sub>6</sub>) was detected in the retention time of 9 min - 10 min and 2,4,6-tri (heptafluoropropyl) - 1,3,5-triazine (C<sub>12</sub>F<sub>21</sub>N<sub>3</sub>) was detected in the retention time of 27 min - 28 min. Since there was no impurity gas in



**FIGURE 7.** The results of SEM tests before and after ageing at 160 °C (X2000) (a)Original sample, (b) 9 %  $C_4F_7N/91$  %  $CO_2$ , (c) 100 %  $SF_6$ , (d) 100 % He.

the inert gas control group at the same temperature, it was speculated that  $C_4F_7N$  reacts with epoxy resin at 160 °C, the C-N triple bond in  $C_4F_7N$  broke and polymerized into  $C_{12}F_{21}N_3$ , as shown in Figure 10. The C-C bond and C-F bond in  $C_4F_7N$  broke and generated  $C_3F_6$ .



FIGURE 8. The results of FTIR tests before and after ageing at 160°C.



FIGURE 9. The results of GC-MS test after ageing at 160  $^\circ C$  in 9 %  $C_4F_7N/91$  %  $CO_2.$ 

#### **IV. DISCUSSION**

The transition process of epoxy resin molecular chain due to  $CO_2$  was analyzed in Figure 11. When the epoxy resin was heated in the mixed gas environment, the epoxy group ring opened firstly. Under further tests, the weak bonds in the main chain such as the C-O bond and C-H bond broke and free radicals were produced.

At last, the hydroxyl was oxidized by carbon dioxide to carboxyl group [28]. It can infer that the existence of carbon

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**FIGURE 10.** The equation of generating  $C_{12}F_{21}N_3$ .



FIGURE 11. The interaction between CO<sub>2</sub> and epoxy resin.

dioxide will accelerate the oxidative decomposition and ageing process of epoxy resin.

As the  $\varepsilon''$  in the low-frequency range of epoxy resin decreased after thermal ageing, it can be explained that  $\alpha$  - Al<sub>2</sub>O<sub>3</sub> was closer to the surface of epoxy resin, which may cause  $\alpha$  - Al<sub>2</sub>O<sub>3</sub> particles to migrate to the surface, forming deep traps at the interface with epoxy resin. Thus, the volumetric conductivity of epoxy resin was reduced, which inhibited the accumulation of surface charge and improved the dc volumetric resistivity of epoxy resin.

The experimental results can also be explained by Xue's model [29]. Before thermal acceleration test, it can be seen that  $\alpha$  - Al<sub>2</sub>O<sub>3</sub> particles in the original samples were evenly distributed in the epoxy resin in the form of fillers, as shown in Figure 12(a). Traps can be divided into electron traps and hole traps. Electron traps attract electrons and hole traps attract positive charges. As the thermal ageing progresses, more Al<sub>2</sub>O<sub>3</sub> particles moved to the surface layer of the epoxy resin (Figure 12(b)). Entanglements at the end of the molecular chain, the interface between the epoxy matrix and Al<sub>2</sub>O<sub>3</sub> fillers, chemical bonding and free volume in amorphous regions together constitute the heterogeneous regions and form electron traps [30] on the surface of the epoxy resin. The carriers were easily trapped on the surface layer of epoxy resin, which could not easily escape (Figure 12(c)). This was the characteristic of deep traps. Deep traps could trap charges easily and form a dense electron region, prevented charge injection from the electrodes, and limited the directional movement of carriers [31], which leaded to the decrease of dc volumetric conductivity of epoxy resin.



FIGURE 12. The process of Al<sub>2</sub>O<sub>3</sub> particles movement (a) Surface layer of original sample, (b) The formation of deep traps, (c) The formation of trapped charge.

TABLE 4. Measurements of Dc Volumetric Resistance and Standard Deviation of Samples ( $10^{14} \Omega \cdot m$ ).

T (°C)	9 % C <sub>4</sub>	F <sub>7</sub> N/91 % CO <sub>2</sub>	100 % SF <sub>6</sub>		100 % He	
. ,	Average	Standard deviation	Average	Standard deviation	Average	Standard deviation
90	5.93	0.85	4.86	0.93	4.94	1.87
125	6.19	2.37	6.45	1.64	6.74	3.55
160	6.85	1.42	6.61	2.58	6.59	2.64

In order to verify the above conjecture, the authors used a Keithley 6517B and a standard 8009 three-electrode test kit to measure the resistivity of the samples. The applied voltage was 1 kV and the duration of polarization was 300 seconds. Each test point was measured ten times and averaged. The sample of type A and the test box were placed in a blast drying oven and the dc volumetric resistivity of the sample was measured at 25 °C. The average volumetric resistivity of the original sample was  $4.02 \times 10^{14} \Omega \cdot m$ , and the standard deviation was  $0.77 \times 10^{14} m$ . The test results of aged samples are shown in Table 4. It can be found that the dc volumetric resistivity of the sample, and the resistivity at 125 °C and 160 °C was slightly higher than 90 °C, which was consist with the results of

dielectric spectroscopy measurements. At the same time, after ageing at 90 °C in C<sub>4</sub>F<sub>7</sub>N/CO<sub>2</sub>, the dc volumetric resistivity of epoxy resin was higher than that in 100 % SF<sub>6</sub> and 100 % He, indicating that more deep traps are introduced into epoxy resin during ageing in C<sub>4</sub>F<sub>7</sub>N/CO<sub>2</sub> rather than in inert gases under 90 °C.

#### **V. CONCLUSION**

In this paper, the authors proposed the evaluation method of compatibility between gas and epoxy resin by considering the performance change and mutual interaction. A gas-solid compatibility test platform was also built. The thermal ageing tests of epoxy resin and  $C_4F_7N/CO_2$  were carried out at different temperatures. The insulation performance, surface morphology, changes in surface composition of epoxy resin, and gas composition are tested after the ageing, and the following conclusions are obtained:

- 1) The thermal ageing tests and test methods in this paper can effectively reflect the degree of interaction between epoxy resin and  $C_4F_7N/CO_2$  after contact. This paper provides the experimental basis for evaluating the compatibility between epoxy resin and  $C_4F_7N/CO_2$ .
- 2) After the ageing test, the dc volumetric conductivity of epoxy resin decreases with the increase of ageing temperature, and the ac conductivity increases with the increase of ageing temperature. Compared with the original samples, the surface flashover voltages of the samples decreased only after ageing at 160 °C.
- 3) Due to the compactness of epoxy resin, short-term thermal ageing test did not make a great impact on its surface morphology and could not reflect changes in compatibility. Furthermore, no adhesion products have been found on the surface of epoxy resin samples after the ageing test.
- 4) Under the normal operation of GIL (below 90 °C), C<sub>4</sub>F<sub>7</sub>N could not react with epoxy resin, the compatibility of epoxy resin with 9 % C<sub>4</sub>F<sub>7</sub>N/91 % CO<sub>2</sub> is similar to SF<sub>6</sub> with epoxy resin. When GIL is locally overheated (above 160 °C), C<sub>4</sub>F<sub>7</sub>N may react with epoxy resin and produce C<sub>3</sub>F<sub>6</sub> and C<sub>12</sub>F<sub>21</sub>N<sub>3</sub> (trimer of C<sub>4</sub>F<sub>7</sub>N). The impact of the by-products on the insulation performance of GIL remains to be further studied.

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