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Improved DC Conductivity and Space Charge Characteristics of XLPE for HVDC Cable Application: Effect of Voltage Stabilizers

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ABSTRACT Electric field distortion caused by the DC conductivity variation and space charge accumulation under temperature gradient in the cross-linked polyethylene (XLPE) insulation is the most serious problem in HVDC cable application. In this paper, three different voltage stabilizers, Bis(4-fluorophenyl)-methanon, Bis(4-hydroxyphenyl) ketone, and Bis[4-(dimethylamino)phenyl] diketone, denoted as A, B, and C, respectively, were mechanically dispersed into the XLPE material with an addition of 0.5 wt%, aiming at improving the DC conductivity characteristics and space charge behaviors of the XLPE insulation at different temperature. The DC conductivity under different temperature and the electric field was tested by a three-electrode system. Meanwhile, space charge properties at 30 °C and 90 °C were investigated using PEA method, and the trap level distribution was estimated by the surface potential decay (SPD) process. The experimental results indicate that all the three voltage stabilizers have improved the DC conductivity characteristics of the XLPE, especially the voltage stabilizers B and C. In addition, the XLPEs modified by the voltage-stabilizers B and C with higher deep trap level demonstrate an excellent ability on space charge suppression at different temperatures. It is concluded that owing to the excellent voltage stabilizing effect, the voltage stabilizers B and C show much potential in HVDC cable application.

INDEX TERMS HVDC insulation, cross-linked polyethylene, dc conductivity, space charge, dielectric breakdown.

I. INTRODUCTION

Due to the good physicochemical and insulation properties, XLPE is widely used in HVDC cable insulation [1]. Researches show that the electric field distortion induced by the temperature gradient and the space charge accumulation inside the XLPE insulation are the toughest problems in the operation of HVDC cables [2].

In HVDC cables, the distribution of electric field in the XLPE insulation depends mainly on the DC conductivity of the XLPE and the distribution of space charge, which is closely related to the operation temperature and the application voltage [3], [4]. The temperature gradient effect occurred during the full load operation of HVDC cable would cause the reversal of electric field distribution inside the

XLPE insulation, that is, the electric field intensity of external insulation of the cable is higher than that of the insulation close to the conductor. At the same time, space charges inside the XLPE insulation will lead to the further distortion of the electric field under the temperature gradient, which seriously affects the reliability of the long-term operation of the cables [5]. Therefore, an excellent insulation material with high DC breakdown electric field, high insulation resistance while insensitive to temperature change, as well as less space charge injection and accumulation is in great request.

For a long time, nanocomposites have been considered as an effective way to improve the performance of XLPE insulating material and a lot of work has been done [6], [7]. However, nanoparticles are easily aggregated and are difficult to be dispersed uniformly in XLPE. It is difficult for nanocomposite to achieve mass industrial production, which greatly increases the difficulty in research and development of XLPE

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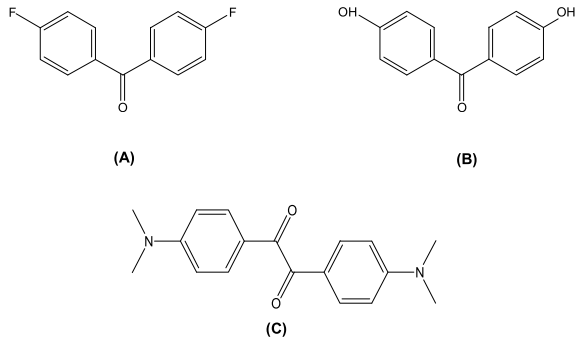


FIGURE 1. Chemical structural formula of the chosen voltage stabilizers. (a) 4,4'-Difluorobenzophenone (b) 4,4'-Dihydroxybenzophenone (c) 4,4'-Bis(dimethylamino)benzil.

cable insulation. Therefore, voltage stabilizers are put forward to solve the problem of XLPE insulation once again in recent years, which can reduce the energy of high energy electrons that injected into the materials from electrodes under high temperature (thermal assisted emission) and high electric field, prevent the energy from these electrons being transferred to the polyethylene chains and ensure that the polymer matrix is not damaged [8].

The related researches on voltage stabilizers have lasted for a long time, and it is a few decades earlier than the research on nanocomposites. While due to its poor compatibility with polymeric matrix, it didn't cause much attention and application until the voltage stabilizers with good compatibility were obtained recently [9]. Several new voltage stabilizers have been found and their stabilizing effect on inhibition of electrical tree growth, improvement of breakdown strength was reported [10]. Meanwhile, theoretical studies of voltage stabilizers based on Quantum Chemistry have been launched already [11]. Nevertheless, there are few reports on influence of the voltage stabilizer on DC conductivity and space charge inhibition for XLPE insulation materials, especially under the condition of different temperature.

In this research, the DC conductivity characteristics with the temperature change and the space charge behaviors under different temperature of XLPE insulation modified by the voltage stabilizers are studied. Three different kinds of voltage stabilizer were mechanically mixed with XLPE with an additive content of 0.5 wt%. The DC conductivity under the temperature from 30 to 90 °C, DC breakdown strength at 90 °C and space charge behaviors at 30 °C and 90 °C were measured. Besides, SPD method was employed to estimate the trap level distribution.

II. EXPERIMENTAL ARRANGEMENT

A. SAMPLE PREPARATION

Three voltage stabilizers produced by J&K Scientific Ltd, Bis(4-fluorophenyl)-methanon, Bis(4-hydroxyphenyl) ketone, Bis[4-(dimethylamino)phenyl] diketone, are shown in Figure 1 and denoted by A, B, C, respectively. No decomposition or reaction of the voltage stabilizers would occur during sample preparation [12].

The neat XLPE purchased from Borealis Company is a mixture of low-density polyethylene (LDPE), dicumyl peroxide (DCP) and antioxidant. The LDPE mixtures were desiccated at 80 °C for 24 hours using a vacuum drying oven before sample preparation. Next, the LDPE mixtures and the voltage stabilizers with an addition of 0.5 wt% were thoroughly mixed by mechanical blending at 110 °C. Finally, XLPE sample with a thickness of $250 \pm 3 \mu\text{m}$ and diameter of $\sim 10 \text{ cm}$ was obtained by stainless steel die shaping at 180 °C and 30 MPa for 30 minutes. No metallic electrodes were coated on the surface of the XLPEs during the sample preparation. The prepared XLPE samples were put into a vacuum oven for exhaust treatment for 48 hours before testing to eliminate cross-linking by-products.

B. DC ELECTRICAL EXPERIMENTS

A three-electrode system was employed to test the DC conductivity of the XLPEs, which was shown in our previous study [13]. The three-electrode system is placed in a temperature control oven, of which the temperature can be controlled from 20 to 150 °C. The charging current I was metered by an electrometer (Keithley 6517B) for 12 hours until it reached a quasi-steady state [14]. To prevent the interference of the residual charge from previous tests, new samples with a thickness of $250 \pm 3 \mu\text{m}$ were replaced in each repetitive experiment.

DC breakdown strength of XLPE samples were measured by two copper-ball electrodes at the temperature of 90 °C, which were immersed in pure insulating oil [15]. The DC voltage source with a boost rate of 0.5 kV/s is employed in the experiment. Each material was tested for 20 times to obtain the Weibull distribution.

Space charge properties of the XLPEs were tested by PEA method at 30 °C and 90 °C. In order to reduce experimental error, the uniform temperature is used instead of the temperature gradient in this study as the sample used in the space charge experiment is too thin. The experiment arrangement is detailed in our previous report [16], while the experimental temperature was controlled by an oil bath system. The upper electrode is made of semiconductor material and the lower electrode is an aluminum electrode. The polarization measurements were performed under a negative DC voltage with an electric field of 30 kV/mm for 30 minutes. Then, the depolarization tests were placed for 15 minutes. Although the polarity of voltage may affect the dynamic process of space charge, it can fully reflect the influence of the voltage stabilizer on space charge characteristics in this research.

The trap level of the samples was measured by SPD method using a surface charge measurement system, of which the schematic has been shown in the reference [17]. The experiment was conducted at room temperature with a humidity of $\sim 30 \%$. The sample was charged by DC voltage for 10 min at +5 kV. To detect the surface potential of the XLPEs, samples were moved instantly to a probe (Trek 6000B-5C), which connected to the electrostatic voltmeter (Trek 347-3HCE). The trap level distributions of XLPEs were obtained by calcu-

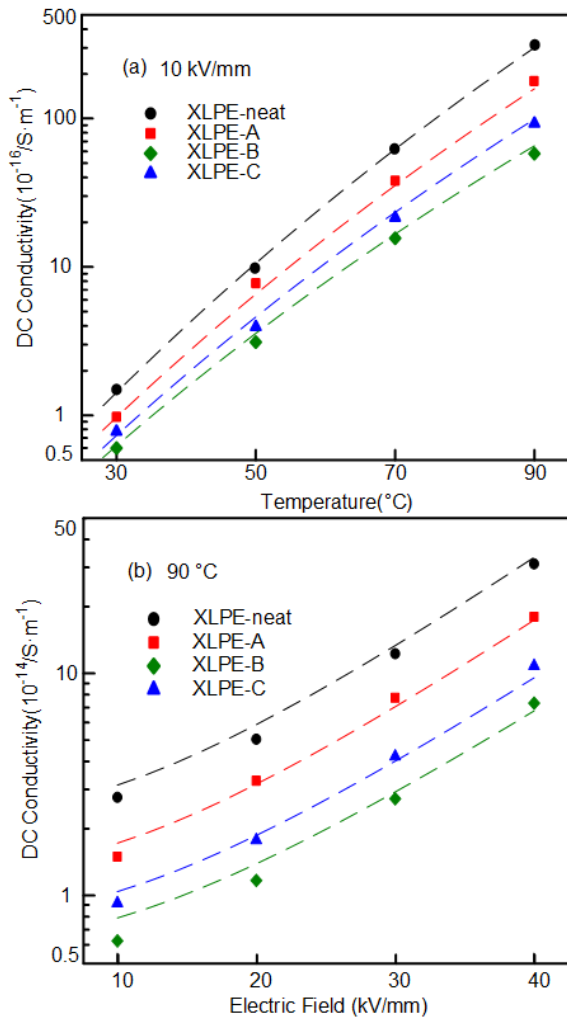


FIGURE 2. DC conductivity of the different voltage stabilizers modified XLPE as a function of (a) temperature under 10 kV/mm and (b) electric field at 90 $^{\circ}\text{C}$.

lating on the basis of SPD information, which acquired by the surface charge acquisition system. The specific calculation method can be found in our previous paper [17].

III. RESULTS AND DISCUSSION

A. DC CONDUCTIVITY

Figure 2a shows the relationship between DC conductivity and temperature from 30 to 90 $^{\circ}\text{C}$ under an electric field of 10 kV/mm. The DC conductivity of all the samples increased with the increase of temperature. It is clearly revealed that with the temperature increase from 30 to 50 $^{\circ}\text{C}$, the increase of DC conductivity of the neat XLPE is very small. While with the further temperature increase from 50 to 70 $^{\circ}\text{C}$, DC conductivity of the neat XLPE shows a notable increase. Besides, the DC conductivity of the neat XLPE increase even more dramatic with the temperature increase from 70 to 90 $^{\circ}\text{C}$. That means, the DC conductivity of neat XLPE shows an approximate exponential growth

TABLE 1. Parameters of temperature and electric field coefficients of different XLPE materials.

	A ($\text{V}/(\Omega \cdot \text{m}^2)$)	φ (eV)	B (mV)
XLPE-neat	1.65×10^5	0.85	1.38×10^{-7}
XLPE-A	2.05×10^4	0.81	1.36×10^{-7}
XLPE-B	9.42×10^2	0.74	1.29×10^{-7}
XLPE-C	4.52×10^3	0.78	1.32×10^{-7}

with the increase of temperature. The DC conductivity of the XLPE-A and XLPE-C samples show the same characteristics with that of the XLPE-neat samples under the condition of temperature increase. However, for XLPE samples modified by the voltage stabilizer B, the DC conductivity growth is less than that of the XLPE-neat with the increase of temperature, which shows a good temperature dependent DC conductivity characteristic.

The relationship between DC conductivity and electric field from 10 to 40 kV/mm at the temperature of 90 $^{\circ}\text{C}$ is shown in Figure 2b. It is indicated that with the increase of electric field from 10 to 30 kV/mm, the DC conductivity of the neat XLPE shows an obvious increase. While with the further increase of electric field from 30 to 40 kV/mm, the growth of DC conductivity of the neat XLPE slows down. For the voltage stabilizers modified samples, DC conductivity of the XLPEs is much smaller than that of the neat XLPE, especially for the XLPE-B.

The DC conductivity is a function of temperature and electric field [18]:

$$\sigma(E, T) = A \exp\left(-\varphi \frac{q}{k_B T}\right) \frac{\sinh(B|E|)}{|E|} \quad (1)$$

where A is a constant related to materials, $\text{V}/(\Omega \cdot \text{m}^2)$; φ is related to the temperature coefficient, eV; q represents the electron charge, C; $q \cdot \varphi$ represents the activation energy; k_B is Boltzmann constant, J/K; T is absolute temperature, K; B is electric field coefficient, m/V; E represent electric field, V/m. According to Equation 1 and the experimental data in Figure 2, the related parameters of the XLPE material can be calculated, which is shown in Table 1. The fitting curves for DC conductivity of different samples are also given by dotted line in Figure 2. It is obviously that the temperature coefficient φ of the voltage stabilizers modified XLPE samples is smaller than that of the XLPE-neat, while the electric field coefficient B decreases with the different voltage stabilizers. These results are consistent with the simulation results in [18], which pointed out that “good materials” have a smaller temperature coefficient of DC conductivity.

B. SPACE CHARGE CHARACTERISTICS

Figure 3 shows the space charge distributions of the voltage-stabilizer modified XLPEs at 30 $^{\circ}\text{C}$ and 90 $^{\circ}\text{C}$, respectively. Two-dimensional color drawings with a defined color bar are

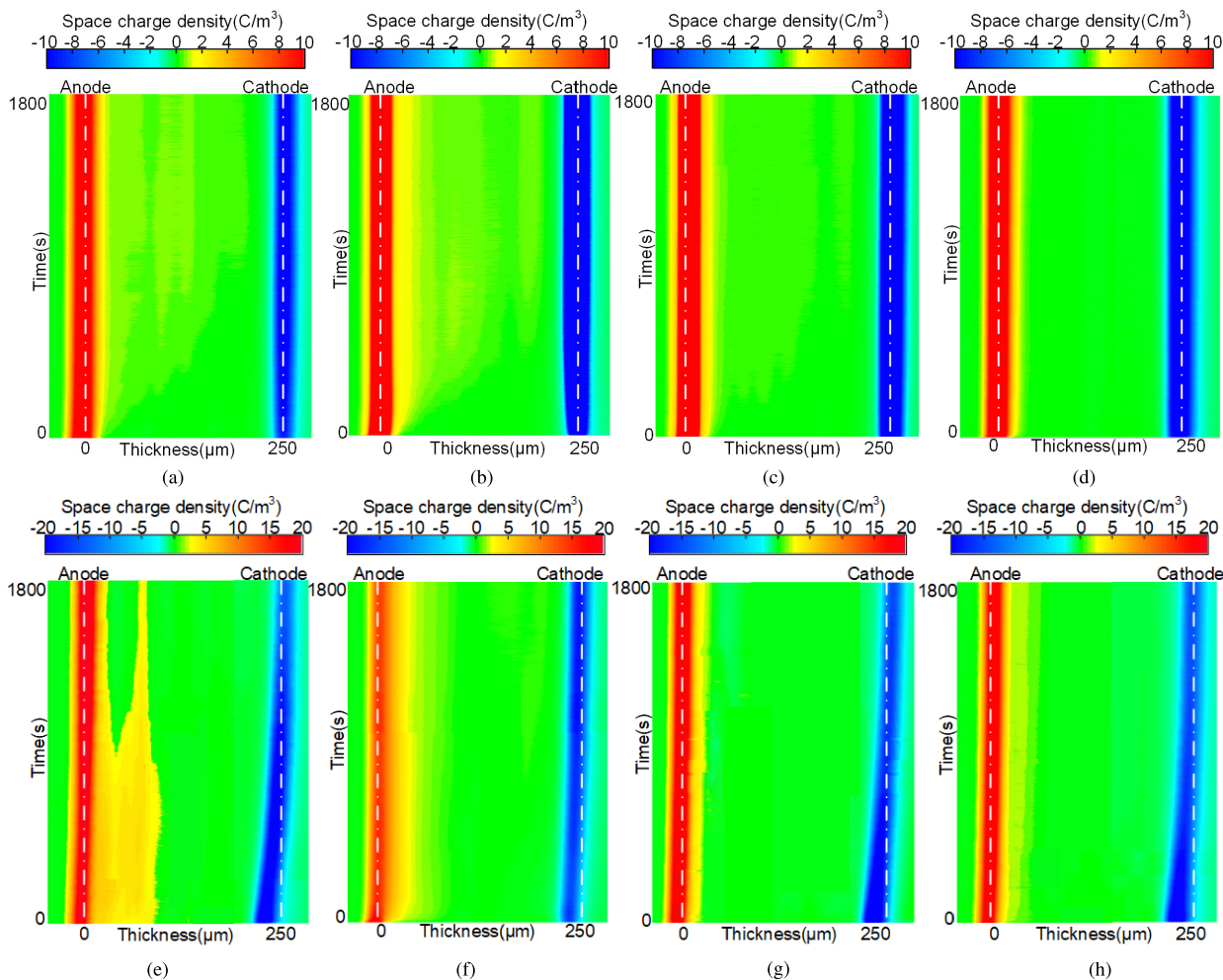


FIGURE 3. Space charge dynamic behaviors voltage-stabilizer modified XLPE samples as a function of polarization time: (a) XLPE-neat; (b) XLPE-A; (c) XLPE-B; (d) XLPE-C at 30 °C and (e) XLPE-neat; (f) XLPE-A; (g) XLPE-B; (h) XLPE-C at 90 °C.

employed to present both the polarity and amount of space charge accumulated in samples. The horizontal axis of each drawing stands for the thickness dimension of the samples, while the vertical axis stands for the time dimension.

As shown in Figure 3a, positive charges are injected from the anode and move slowly towards the cathode over time in the neat XLPE samples at 30 °C. Comparing the space charge characteristics of the XLPEs modified by different voltage stabilizers in Figures 3b, 3c and 3d at 30 °C, it clearly shows that the voltage stabilizer A didn't show obvious effect on suppressing space charge injection. Space charges that injected inside the XLPE-A samples are even a little more than that of the pure XLPEs. While in Figures 3c and 3d, only very few charges were injected into the interior of the samples XLPE-B and XLPE-C. Space charges mainly accumulate near the electrode, which shows great ability of voltage stabilizers B and C on suppressing space charges in XLPE insulation at 30 °C.

When the measurement temperature rises to 90 °C, a large amount of positive charges is injected from the anode into the neat XLPEs, as shown in Figure 3e. These injected positive

charges accumulated rapidly near the anode, forming an obvious positive charge packet. With the polarization time, the positive charge packet moves continuously inward the sample. In this process, the charge gradually dissipates, and the charge packet becomes smaller inside the sample. In the process of space charge measurement at 90 °C, the cathode is slightly shift due to the slight deformation of the sample.

For XLPE samples modified with the voltage stabilizers, space charge characteristics at 90 °C shown in Figures 3f, 3g and 3h reveals that the inhibition effect of all the three voltage stabilizers on space charge accumulation is evident at high temperature. The positive charges injected from the anode are only accumulated near the electrode. The depth of charges injected into the sample is relatively shallow, and the amount of the injected charges is much less than that of the neat XLPE. That means all the three voltage stabilizers have good ability to inhibit space charge accumulation in XLPE materials at high temperature. As the difference in DC conductivity between different XLPEs shown in Figure 2 is small, the space charge behavior of the voltage-stabilizer modified XLPEs is mainly affected by the stabilization effect

of the voltage stabilizer and the trap level distribution (will be discussed in details later) of the XLPE. Due to the voltage stabilizing effect and the trap level change introduced by the voltage stabilizer, space charge injected into the XLPE-B and XLPE-C samples is less than that of the neat XLPEs.

The above experimental results show that the effect of the voltage stabilizer on suppressing space charges is related to temperature, and the voltage stabilizers are more active at high temperature. It is worth noting that the effect of three different voltage stabilizers on the space charge inhibition in XLPE materials is different, which may be related to the type of voltage stabilizers and the types of functional groups it contained.

To reveal the influence of the voltage stabilizer on space charge characteristics further, space charge dissipation characteristics at short-circuit condition after poling of the XLPE samples at 90 °C are shown in Figure 4. In the depolarization process of XLPE samples, negative charges locate at the electrode-dielectrics interfaces, while positive charges accumulate in the bulk of samples. This is consistent with the polarization processes as shown in Figure 3. It can be observed that, after a polarization time of 30 min, space charges accumulated in sample bulks are mainly positive charges at 90 °C. The amount and depth of the injected positive space charge in all the voltage stabilizer modified XLPEs is much less and shallower than those of the neat XLPEs. In the depolarization process, space charge in the bulk of XLPEs dissipate gradually. In neat XLPEs, space charges in the vicinity of electrode dissipate rapidly, while the space charge dissipation rate in the bulk of the samples is very slow. Compared with the neat XLPEs, all three voltage stabilizers modified XLPEs show a good influence on promoting space charge dissipation at 90 °C. At the end of the depolarization, only very few space charges remain in the bulks of the XLPEs.

C. DC BREAKDOWN STRENGTH

The Weibull plots of DC breakdown strength of XLPE samples modified by different voltage stabilizers at the temperature of 90 °C is shown in Figure 5. The characteristic breakdown strength of the XLPE-neat, XLPE-A, XLPE-B, XLPE-C is 184.4, 215.2, 278.1 and 250.9 kV/mm, respectively. It reveals that the DC breakdown strength of the XLPE insulation is clearly enhanced by the addition of the voltage stabilizer especially the voltage stabilizer B. This is because under the condition of high temperature and high electric field, electrons injected from electrodes would gain more energy and could be accelerated effectively. Then, electrons are injected into the XLPE and may captured by the voltage stabilizers in XLPE samples, reacted, and became a low energy electron, which does less harm to the XLPE molecular chain. This way, DC breakdown strength of XLPEs is strengthened.

The improvement of DC breakdown strength of the voltage stabilizers modified XLPEs is also related to both DC conductivity and space charge characteristics. On the one hand,

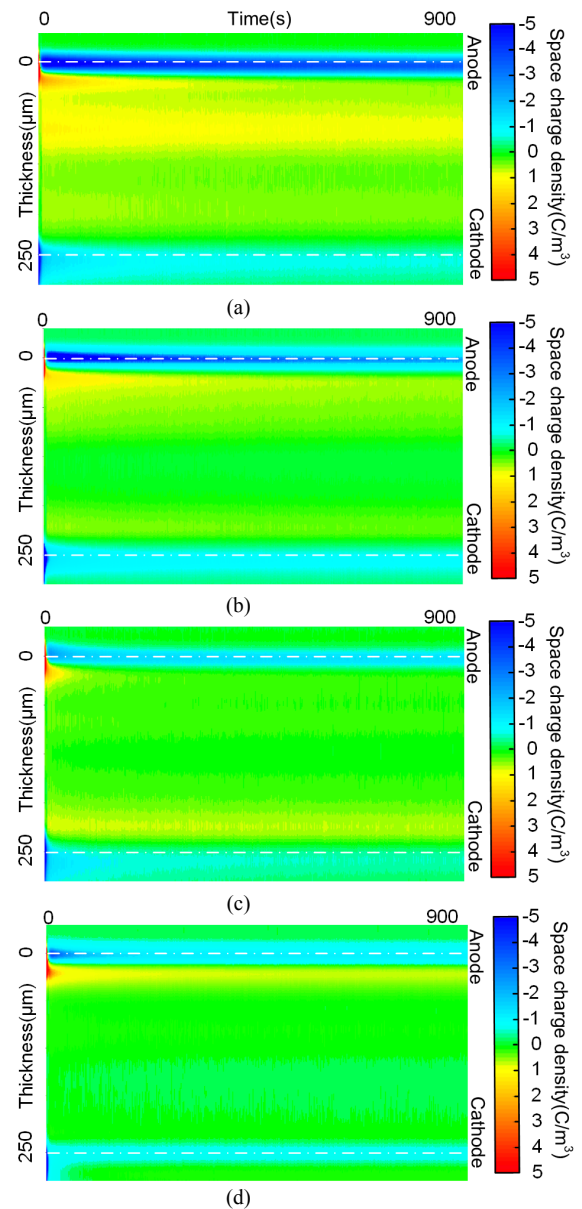


FIGURE 4. Space charge dissipation in voltage-stabilizer modified XLPEs as a function of depolarization time at 90 °C: (a) XLPE-neat; (b) XLPE-A; (c) XLPE-B; (d) XLPE-C.

voltage stabilizers weaken the energy of injected electrons and reduce the DC conductivity of the XLPEs, which slows down the carrier transport in the XLPEs; on the other hand, the space charge accumulation in the voltage stabilizers modified XLPEs is decreased and the depth of charge injection becomes shallower than that of the neat XLPEs. The electric field near the electrode is weakened due to the accumulation of homo-charges, which prevents the further injection of charges and improves the breakdown strength of XLPEs. Due to the significantly improved DC conductivity characteristics and space charge properties, the XLPE samples modified by the voltage stabilizer B shows the best DC breakdown resistance.

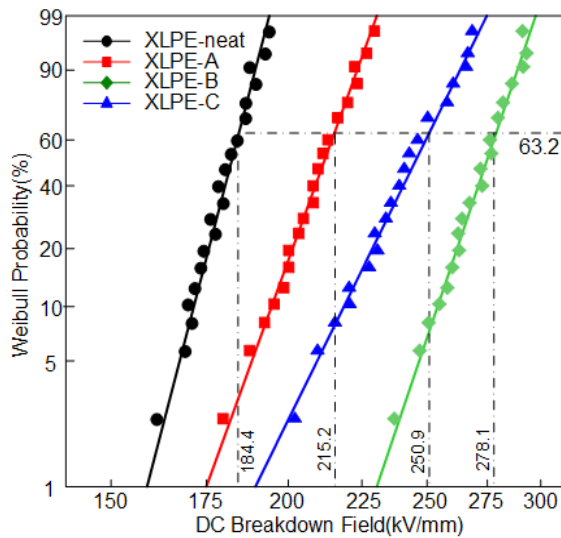


FIGURE 5. Relationship between the weibull probability and the DC breakdown field of the different voltage stabilizers modified XLPE at 90 °C.

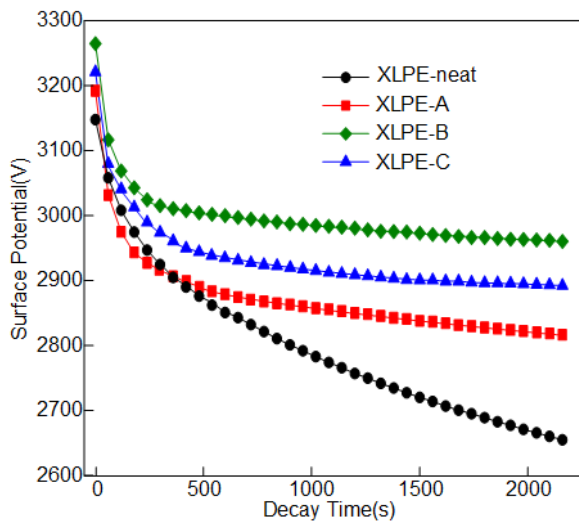


FIGURE 6. SPD curve of the different voltage-stabilizer modified XLPEs at room temperature.

D. TRAP LEVEL DISTRIBUTION

The surface charge decay processes of the XLPEs modified by the different voltage stabilizers are shown in Figure 6. The samples were charged for 10 min at +5 kV before the surface potential is measured. The initial potential of the voltage-stabilizer modified XLPEs is higher than that of the pure XLPEs. Comparing with the DC conductivity of the samples in Figure 2, it revealed that the XLPEs with higher conductivity show a faster charge decay rate in the decay process.

Figure 7 shows the trap level distributions of the different voltage-stabilizer modified XLPEs. The two-peaks distribution of trap level are observed in all samples which represent the relative shallow and deep traps locating at different trap levels. This two-peak trap level distribution has also

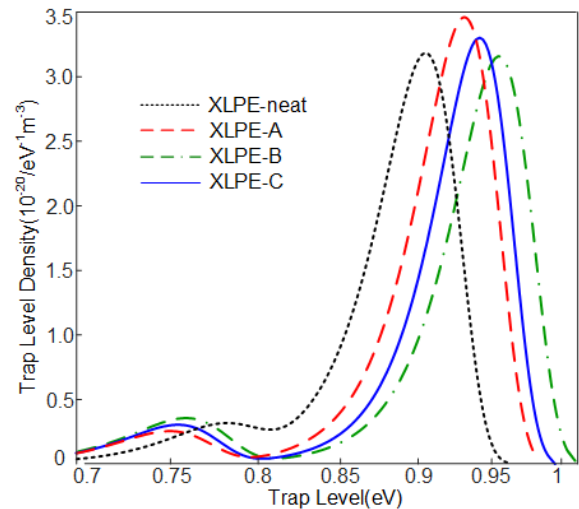


FIGURE 7. Trap distribution of the different voltage-stabilizer modified XLPEs.

been reported by previous research [19]. The neat XLPE has a deep trap peak at 0.92 eV and a shallow trap peak at 0.77 eV. The voltage stabilizer molecule has strong electronic affinity and is easier to capture carriers, which make it possible for voltage stabilizer to show the role of charge trap. Besides, the addition of voltage stabilizers can affect the trap level distribution by changing the microstructure of polyethylene. Compared with the neat XLPE, the deep trap peak of all three voltage-stabilizer modified XLPEs move towards a higher energy level; meanwhile, the shallow trap peak of voltage-stabilizer modified XLPEs move towards a lower energy level. The relationship between trap level distribution, DC conductivity characteristics and space charge behaviors will be discussed in details later.

IV. DISCUSSIONS

The stabilizing function of the voltage stabilizers used in this study is the key in XLPE modification. The ionization potential of voltage stabilizer is much lower than that of the polyethylene molecules. Hence, it is more easily for the voltage stabilizer molecule to ionize than that of the polyethylene under electric fields. The energy of resulting secondary electron is also lower and unlikely to cause additional ionization of the dielectric [8]. In addition, the energy required for the reaction of cationic radicals in voltage stabilizer molecule with hot electrons to create free radical products is much higher than that required for simple recombination to regenerate the original voltage stabilizer. This ensures that voltage stabilizer could reduce the energy of high-energy electrons under high electric field and realizes the reuse of the voltage stabilizer in theory.

For the voltage stabilizers used in this study, it can slow down injected electrons due to its stabilizing function; at the same time, the voltage stabilizer also changes the trap level distribution of the XLPE samples which has been shown

in Figure 7. In the process of polarization, electrons are more easily trapped by deep traps, thus preventing space charge injection; moreover, because of the voltage stabilization effect of the voltage stabilizer, energy of charges injected into XLPE samples from the electrode is weakened by the voltage stabilizers, the injected low-energy space charge is easier to be trapped by shallower traps, which is easier to de-trap and dissipate in the depolarization process.

The improved DC conductivity characteristics of XLPEs can be interpreted by the stabilization effect of the voltage stabilizers. The energy of the hot electrons injected from the electrodes is reduced by the introduced voltage stabilizers in XLPEs, which results in the inhibition of charge carriers' migration, leading to the decrease of conduction current and DC conductivity.

For the space charge characteristics of the voltage-stabilizer modified XLPEs, due to the voltage stabilizing effect and the trap level change introduced by the voltage stabilizers, space charge injected into the XLPEs is less than that of the neat XLPE. Meanwhile, the DC conductivity of XLPEs modified by voltage stabilizers is lower than that of the neat XLPEs, combined with the effect of voltage stabilizers and the deepened trap level, space charge injection and accumulation in the XLPE is reduced substantially. For DC breakdown strength test in Figure 5, on the one hand, voltage stabilizers weaken the energy of injected electrons and make the electrons easier to trap; on the other hand, voltage stabilizers deepen the deep trap level of XLPE and can capture space charge more efficiently to suppress the further injection of electrons. The interaction of these two factors lead to the marked increase in breakdown strength of XLPE samples.

It is worth noting that the DC breakdown strength of XLPEs is obviously improved by the voltage stabilizers at 90 °C. Comparing the performance of voltage stabilizer under different electric fields (DC conductivity measurement and space charge measurement under low electric field, DC breakdown test under high electric field) and different temperature, it can be deduced that the stabilizing effect of voltage stabilizer is more effective under high temperature and high electric field.

V. CONCLUSIONS

In summary, the effect of the voltage stabilizers on DC conductivity characteristics and space charge behaviors of XLPE insulation at different temperature were investigated. The conclusions could be summarized:

1. The voltage stabilizers could improve the DC conductivity characteristics of XLPE material. The voltage-stabilizer modified XLPEs show smaller temperature coefficient and bigger electric field coefficient of DC conductivity than that of the neat XLPEs.
2. The voltage stabilizers show good ability to inhibit space charge accumulation at different temperatures, especially at 90 °C.

3. The DC breakdown strength of the XLPE samples is improved by voltage stabilizers at 90 °C, which is related to both the DC conductivity and space charge characteristics of the XLPEs.
4. The selected voltage stabilizers could change the trap level distribution of XLPE. Coupled with its stabilizing effect, the improved electrical properties of the XLPE samples can be explained.

The above researches show that voltage stabilizers B and C have great application value in modification of polyethylene insulation materials for HVDC cables.

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