Space charge properties of EPDM under different electric field and thermal ageing

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ABSTRACT

Ethylene Propylene Diene Monomer (EPDM) has been widely used as the insulating material of power cables and accessories. Due to the formation and injection of space charge under DC electric field, the distortion of electric field strength will influence the dielectric properties of EPDM. In the paper, the space charge profiles at different electric field and ageing temperature were carried out. The total charge quantity and trap distribution of aged EPDM were analysed. The results show that the charge injection from electrodes was intensified with the increase of electric field. The behaviour characteristics of the space charge in EPDM during polarization can be divided into four stages: (I) no charge injection, (II) bipolar injection, (III) charge transformation and recombination, and (IV) anode injection. Space charge properties of EPDM aged at different temperature are also different. With the ageing time increasing, homocharge injected from anode increases, and heterocharge accumulates in the vicinity of cathode. When EPDM is aged at 120 °C, the total charge quantity at the initial ageing stage increases with ageing time. Then it decreases. However, the total charge quantity of EPDM aged 2 days at 160 °C reaches to a higher value. The space charge properties of aged EPDM can be explained by degassing process and the theory of trap filling.

KEYWORDS

Ethylene Propylene Diene Monomer, space charge, dielectric strength, thermal ageing

AUTHOR NAMES & AFFILIATIONS

Ethylene Propylene Diene Monomer (EPDM), being of excellent moisture resistance, thermal resistance, corona resistance and high tensile strength, has been widely used as the insulating material of MV/HV power cables and accessories. Both physical and chemical defects from production process or operating ageing would introduce traps into the material. Under DC electric field, the internal trap of EPDM can capture carriers from electrode injection or impurity dissociation, which will form the space charge, and cause local electric field distortion in the insulation. Moreover, space charge accumulation will accelerate the insulation ageing, reduce the insulation life and even cause breakdown [2].

Therefore, after the technique of space charge measurement was developed in 1980s [2], many researchers focused on space charge properties of insulation under thermal and electrical stress. The space charge distribution in EPR and XLPE were measured under thermal gradient, which reveals the field distortion were enhanced with the thermal gradient [3]. The effect of electrical stress on the space charge characteristics of EPDM was studied by measuring and analyzing the space charge distribution and conductive current, which reveals

the energy loss dynamics caused by space charge [4].

For understanding the long-term ageing properties of EPDM and the effects on the space charge characteristics, EPDM specimens were thermally aged at different temperature in the paper. The space charge properties of aged EPDM were analyzed. The total charge quantity during depolarization and the trap energy distribution was calculation for explaining the charge transportation in aged EPDM.

EXPERIMENTAL ARRANGEMENT

Sample preparation

The DCJ30M type EPDM, mixed and filtered in the manufactory, was vulcanized for 15 min under 160 °C and 14 MPa in laboratory. Cured specimens were naturally cooled down to room temperature. After taken off from mould, the specimens were put into the vacuum oven with 80 °C and 50 Pa for 24 hours in order to eliminate the byproduct and mechanical stress during vulcanization. The thickness of specimen was 390 μ m. Specimens were cut into discs with a diameter of 60 mm. Before the test, all specimens' surface was cleared by absolute alcohol.

Space charge measurement

Space charge was measured by the PEA device, produced by Peanuts Five Lab. The measurement sensitivity of PEA device is about 0.2 C/m³. The upper and lower electrode of the test device is semiconductor layer (SC) and aluminium (AI) respectively. During the PEA test, negative DC voltage was applied to specimen, so AI was anode and SC was cathode. The amplitude of pulse voltage was 400 V. Firstly, the space charge profiles of EPDM at different electric field, including 5, 10, 20, and 40 kV/mm were test. The polarization time was 2 hours, and the depolarization time was 1 hour. Then, for different thermal ageing specimens, the space charge profiles at each ageing stage were measured under 20 kV/mm within 30 min. After that, the voltage was removed, and the depolarization characteristics within 60 min were measured.

Thermal ageing

For analysing the long-term effect of temperature on EPDM, the thermal ageing tests were conducted in an electro-thermostatic blast oven. The ageing temperature were 120 °C, 140 °C and 160 °C. Before space charge test, the specimens were taken off the oven and cooled down to room temperature. After space charge test, the specimens were wrapped with aluminium foil, shorted with ground and placed back to the oven.

RESULT AND DISCUSSION

Space charge at different electric field

The space charge of EPDM at different electric field for polarization about 120 min as shown in Figure 1. It can be found that the charge density in the interface between electrode and polymer increased with electric field rising. On the anode, the interfacial charge density decreases with time, and homocharge is injected into EPDM. When electric field is no more than 10 kV/mm, the space charge on the cathode hardly changes. Heterocharge accumulates near the cathode interface when electric field rises to 20 kV/mm. At 40 kV/mm, the density of heterocharge increases to about 3.4 C/m³. Therefore, the space charge exists in the vicinity of interface in most case, and the space charge distribution on the anode and cathode is different. When electric field is high enough, there is the positive charge observed in EPDM.

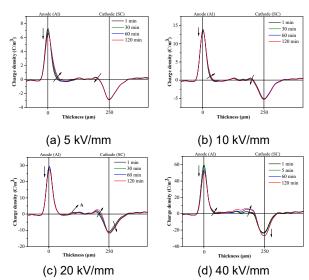


Fig. 1: Space charge of EPDM at different electric field

Figure 2 shows the space charge distribution of EPDM during depolarization after polarization for 120 min at different electric field. The accumulation of positive charge, namely homocharge, can be found near the anode during volt-off, which indicates the occurrence of charge injection. On the cathode, a small amount of negative charge can be observed and disperses quickly after poling at 5 kV/mm. At 10 kV/mm, the amount of negative charge on cathode decreases, which may result from the neutralization of positive charge from the anode. When the electric field rises to 20 kV/mm, a certain amount of positive charges accumulating in the vicinity of cathode can be found, which lead to the disappear of negative charge. The positive charge packet can also be found in EPDM. At 40 kV/mm, there are more positive charges accumulating near the cathode.

Space charge distribution at different electric field can be explained by the origination and migration of space charge. Generally, the space charge in EPDM comes from the injection and impurities. When the electric field is lower than the threshold of charge injection, the interface between EPDM and electrode is ohmic contact. With electric field rising, potential energy barrier becomes low and the amount of carriers increase due to the schottky injection. On the other hand, ionic charges from the impurity dissociation are polarized at high electric field,

which will increase the heterocharge density. The charge mobility depends on the electric field. The injected charge can easily move to inverse electrode with the increase of electric field. All in all, the space charge in EPDM can be divided as four phase.

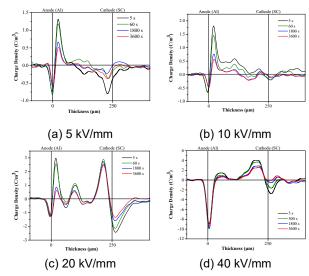


Fig. 2: Space charge of EPDM after poling at different electric field

No injected charge phase. When electric field is no more than 5 kV/mm, no charge is injected into EPDM. A small amount of space charge in EPDM is from the impurity ions.

Bielectrode injection phase. At about 10 kV/mm, injected charge can be trapped near the electrode, and cannot move into EPDM because the electric field is not high enough.

Charge migration and neutralization phase. Both charge injection and impurity dissociation are enhanced. Charges begin to drift towards reverse electrode.

Charge injection from anode. Positive charge injection will maintain a dominant position, which leads to the heterocharge accumulation near the cathode.

Space charge after different thermal ageing Aged at 120 °C

The space charge distribution of EPDM aged at 120 °C is measured at 20 kV/mm and room temperature, as shown in Figure 3. It can be found in Figure 3a that the interfacial charge at anode decreases with time, which indicates the homocharge is injected and is more clearly shown in depolarization phase. At cathode, there is no obvious charge injection. After EPDM is aged 80 days and 120 days, the injecting rate of homocharge accelerates with ageing. A small amount of heterocharge accumulates in the vicinity of cathode. For EPDM, it is difficult for SC injection, even though the sample is aged for a long time.

Aged at 140 °C

The space charge distribution of EPDM aged at 140 °C is measured at 20 kV/mm and room temperature, as shown in Figure 4. Before EPDM is aged about 30 days, the homocharge injection at anode cannot be observed. After that, homocharge can be injected into EPDM from anode. Moreover, the number of heterocharge accumulating near the cathode increases with ageing, and interfacial

homocharge at cathode also increases.

Aged at 160 °C

The space charge distribution of EPDM aged is shown in Figure 5. At anode, the interfacial charge increases with time, which is different from the charge of EPDM aged at 120 $^{\circ}$ C and 140 $^{\circ}$ C. A positive charge packet, with the maximum charge density larger than 1 C/m³, appears about 100 μ m away from the anode, more evident in the depolarization phase. Similar to lower ageing temperature, heterocharge accumulates in the vicinity of cathode.

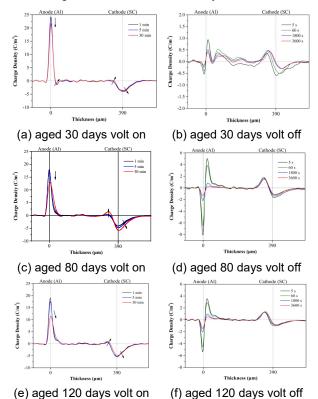


Fig. 3: Space charge distribution of EPDM at 20kV/mm after aged at 120 °C

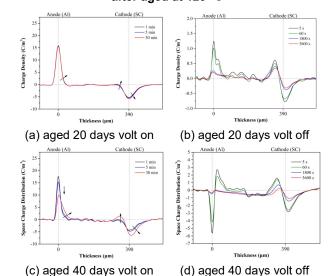


Fig. 4: Space charge distribution of EPDM at 20kV/mm after aged at 140 °C

Space charge properties

For further analysing the transportation property of charge accumulating in the material, the charge quantity in the depolarization phase is calculated. The total charge quantity, q(t), depending on time, t, can be expressed as

$$q(t) = \int_0^d |\rho(x,t)| S dx$$
 [1]

where d is the thickness of specimen, $\rho(x, t)$ is the charge density during depolarization, S is the area of specimen.

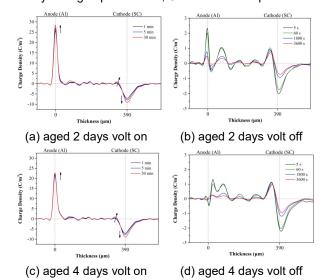


Fig. 5: Space charge distribution of EPDM at 20kV/mm after aged at 160 °C

Figure 6 shows the total charge quantity in EPDM after poled by different electric field. It can be found that the charge quantity decreases fast, and tends to a stable value after about several ten seconds. At lower electric field, 5 and 10 kV/mm, the charge quantity decreases to a same value finally. With electric field rising, the residual charges, trapped by deeper trap, increase and de-trap laboriously. The charges dissipate fast, because the quantity of charges trapped by low trap is high when the specimen is polarized at high electric field. Therefore, the increase of electric field will increase the quantity of charge in both deep and low trap.

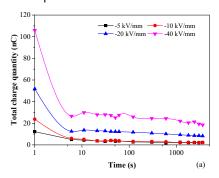


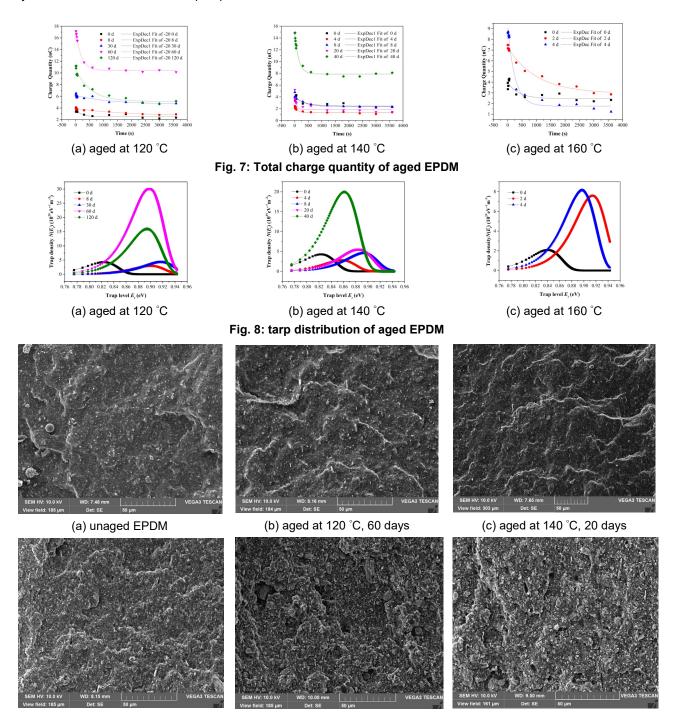
Fig. 6: Total charge quantity of EPDM after polarized at different electric field

Figure 7 shows the total charge quantity of EPDM aged at different temperature. The charge quantity of EPDM is not the linear dependence on ageing time. For well explaining the phenomenon of charge transportation in aged EPDM, the space charge is divided as fast charges and slow charges. Fast charges are the charges will dissipate during

initial several ten seconds and can denote the charges trapped by lower trap. Slow charges are the charges left in the specimen and denotes the charges trapped by deeper trap. At the first half stage of 120 °C and 140 °C ageing, the quantity of fast charges is low and almost equal to the slow charge. With the ageing, the fast charge quantity rises to a high value. The quantity of fast charges of EPDM aged at 160 °C depends on the ageing temperature, and increases with ageing.

Both fast charges and slow charges are influenced by the trap distribution in EPDM as shown in Figure 8. the trap energy distribution was test on the method of Isothermal decay current ^[5]. For EPDM aged about 30 days and 20 days at 120 °C and 140 °C, the trap depth increased with

ageing time. On the contrary, the charge density is nearly stable. This phenomenon is due to the degassing process during which the dissociation and volatilization of internal residuals and sulfurized by-products results in the increase of trap depth ^[6]. With the ageing, the antioxidant is gradually consumed to empty, and the molecular chain of EPDM begins breaking, which increases the number of ions and polar groups in EPDM, and leads to increase both trap density and the number of trapped charges. In addition, it looks like that the trap depth also decreases, which may be due to deeper traps filled firstly. When the deeper traps are filled up, the mobility increases and the trap depth decreases at this stage.



(d) aged at 120 °C, 120 days

(e) aged at 140 $^{\circ}$ C, 40 days

(f) aged at 160 °C, 6 days

Fig. 9: SEM images of EPDM

Compared with EPDM aged at no more than 140 °C, the trap depth of EPDM aged only 2 days at 160 °C reaches to about 0.92 eV. The trap density also increases to a high value. Therefore, 160 °C may be a critic temperature for EPDM, and results in the quick deterioration of EPDM.

Figure 9 show SEM images of the cross section of EPDM fractured in liquid nitrogen. It can be seen from Figure 9a that the surface of unaged EPDM is smooth, and there is no wrinkle and cavity. With ageing, the surface of EPDM becomes uneven as shown in Figure 9b and 9c. It seems that the delamination will occur, evidencing the loss of mechanical toughness. At the end of ageing, EPDM becomes rough and forms a mica-like structure as shown in Figure 9d~9f. Lots of cavities with the order of micrometre also form in EPDM, which indicates the dissociation and deterioration of EPDM. All in all, the defects forming in EPDM with ageing will lead to the increase of trap and the charge density.

CONCLUSION

The space charge of EPDM at different electric field was test firstly. Then space charge measurement of EPDM aged under different temperature was also carried out. At electric field less than 10 kV/mm, no charge is injected into EPDM. When electric field is higher than 10 kV/mm, charge begins to inject from anode and cathode. After electric field rises to 20 kV/mm, heterocharge accumulates in the vicinity of cathode, and suppresses the homocharge from cathode. The space charge distribution of EPDM aged at different temperature depends on the trap depth and density. At the initial ageing stage, the trap depth of EPDM aged at 120 °C and 140 °C increased due to the degassing process, so the total charge quantity is also low. With the increase of ageing time, the deterioration of EPDM causes the increase of trap density and the number of trapped charges.

Acknowledgments

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REFERENCES

- [1] D. Fabiani, G. C Montanari, C. Laurent, G. Teyssedre, P. H. F. Morshuis, R. Bodega, L. A. Dissado, A. Campus and U. H. Nilsson, 2007, "Polymeric HVDC Cable Design and Space Charge Accumulation. Part 1: Insulation/Semicon Interface", IEEE Electr. Insul. Maga., vol. 23, 11-19.
- [2] T. Takada, T. Maeno and H. Kushibe, 1987, "An Electric Stress-pulse Technique for the Measurement of Charges in a Plastic Plate Irradiated by Electron Beam", IEEE Tans. on El, Vol. 22, 497-501,
- [3] H. Uehara, G. C. Montanari, Q. Chen, Z. Li, and Y. Cao, 2016, "Comparison of space charge behavior in XLPE and EPR with thermal gradient", IEEE International Conference on Dielectrics, ICD, vol. 1, 143-146.
- [4] W. Wang, Y. Tanaka, T. Fujitomi, T. Takada, and S. Li, 2017, "Study on conduction current and space charge accumulation simultaneously in Ethylene Propylene

- Diene Monomer film", International Conference on Electrical Materials and Power Equipment, ICEMPE, vol. 1, 464-467.
- [5] Z. Lei, C. Li, R. Men, and J. He, 2018, "Mechanism of bulk charging behavior of ethylene propylene rubber subjected to surface charge accumulation", J. Appl. Phys., vol.124, 244103.
- [6] M. Fu, G. Chen, L. A. Dissado and J. C. Fothergill, 2007, "Influence of thermal treatment and residues on space charge accumulation in XLPE for DC power cable application", IEEE Trans. Electr. Insul., vol. 14, 53-64.