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# Effect of Graphene Coating on Space Charge Characteristic of XLPE and Semiconductive Layer at Different Temperatures

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**ABSTRACT** DC cross-linked polyethylene (XLPE) power cables are being used recently for HV/EHV applications, the reliability of which is claimed to be significantly affected by space charge accumulation in the insulating material. Many methods have been provided for limiting the injection and accumulation of space charge. In the paper, three kinds of specimens with graphene coating applied on a sandwich composed of XLPE and semiconductive layer were made. Space charge distribution of specimens was measured at 10 kV/mm and 40 kV/mm under room temperature and 60 °C. The measurement results show that positive charges are injected and accumulate in reference XLPE specimen. Negative charge accumulates in XLPE, and the quantity of positive charge injected from anode is significantly reduced, when graphene is coated on the semiconductive side. Little space charge is injected into XLPE coated with graphene, and positive charge accumulates near the interface between XLPE and electrode, which lowers electric field near the anode. Graphene layer introduced deeper trap distribution, thus making difficult negative charge injection from the cathode into XLPE.

**INDEX TERMS** Graphene coating, dc cable, XLPE, space charge, trap distribution.

## I. INTRODUCTION

Injection and accumulation of space charge in HV and EHV DC cross-linked polyethylene (XLPE) power cables are one of the prime problems leading to the degradation and aging of electrical insulation [1]–[3]. Space charge can affect also cable accessories, like e.g. joints and/or termination due to charge accumulation at the interfaces between different insulating materials. Unlike the HVDC cable body where the semiconductive layer (semicon) provides good contact [4], [5], in cable accessories a discontinuity in permittivity and conductivity is naturally present due to the different mechanical, chemical and physical properties of the layers constituting the accessory, leading to several interfaces, e.g. metal/polymer, semicon/polymer and polymer/polymer.

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It has been verified that metal/polymer or semicon/polymer interface can lead the injection of charge which results in the distortion of electric field in the insulation bulk at high fields and temperatures [6], [7].

Many methods have been researched for blocking the injection of space charge into insulation. Milliere *et al* deposited a layer of silver nanoparticles embedded in a semi-insulating organosilicon matrix on the surface of polyethylene, and found that the space charge injection was suppressed by silver nanoparticles [8], [9]. Du *et al* studied the space charge behavior of fluorinated polyimide. Their results showed that the charge injection was suppressed by fluorination [10]. Li *et al* further researched the influence of fluorination, and Cr<sub>2</sub>O<sub>3</sub> coating on the surface trap and charge injection, and provided a field-dependent charging model based on dominant charge transport behavior under different electric fields [11], [12]. An *et al* again tailored

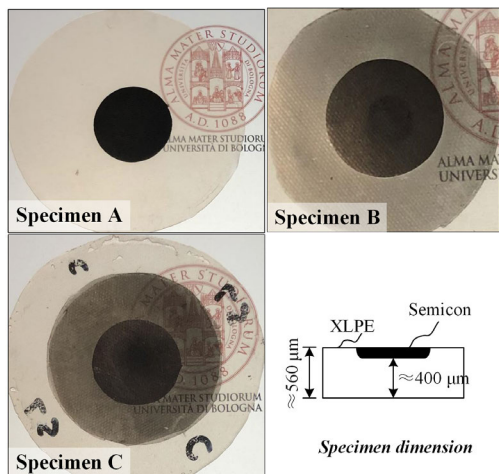


FIGURE 1. Specimens without and with graphene coating.

the surface properties of polyethylene again by fluorination technique. They found that charge injection was blocked by the fluorinated layer at the semicon electrode interface and the film/surface fluorinated plate interface [13]. These researches provide various methods for suppressing the space charge injection by modifying the interface between electrode and insulation.

In order to block the charge injection into XLPE, this paper makes use of a few-layer graphene coating layer spread over the surfaces of the specimen. Indeed, graphene electrical properties are rather unique. A single-layer graphene shows semimetal characteristics with zero bandgap, which can be slightly opened up to 0.25 eV in multi-layer graphene [14]. Therefore, the potential barrier height of junction where graphene is in contact with insulation is quite large, which may affect space charge accumulation features. For these reasons space charge distribution of specimens was measured at different electric fields and temperatures, and trap distribution of charges accumulating near both electrodes was calculated.

## II. EXPERIMENTAL SETUP

### A. SPECIMEN PREPARATION

The specimens were made by Nexans. They consisted of an XLPE block of 560 μm with a semicon layer inserted in the top of the specimen having a thickness of 160 μm (see Fig. 1). Therefore, the thickness of XLPE below the semicon was about 400 μm. For analyzing the effect of graphene coating on space charge, few-layer graphene dissolved in an appropriate solvent was mechanically applied by Nanesa S.r.l on the semicon side, and on both sides of XLPE and semicon, as shown in Fig. 1. The main characteristics of the specimens are shown in Table I. Then, all specimens were treated under vacuum at 80 °C and 50 Pa for 24 hours in order to eliminate the possible by-product and mechanical stresses during preparation.

### B. MEASUREMENT SETUP

The electrode configuration for different specimens is shown in Fig. 2. Space charge was measured by pulsed

TABLE 1. Sheet resistance of specimens.

Name	Sheet resistance of graphene layer (Ω/sq)	
	XLPE side	Semiconductive side
specimen A	No graphene	No graphene
specimen B	No graphene	3700
specimen C	5700	3000

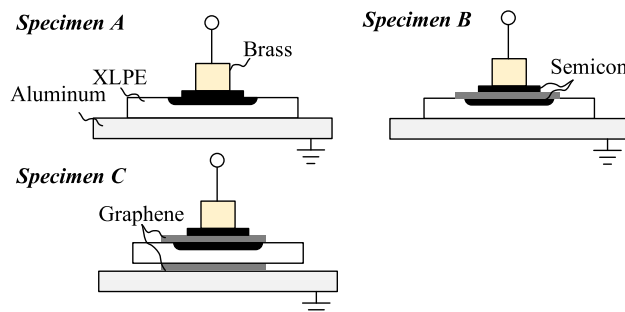


FIGURE 2. Specimen dimension and electrode configuration.

electroacoustic (PEA) technique at room temperature and 60 °C under the applied electric field of 10 kV/mm and 40 kV/mm, respectively. The anode was a semicon, and the cathode was aluminum. A voltage pulse with the amplitude of 500 V and the width of 10 ns was applied to specimen through a coupling capacitor. The specimen was firstly polarized about 10800 seconds, and then depolarized about 3600 seconds. Before the measurement at each electric field, the specimen was shorted and discharged at 50 °C for more than 2 days in order to remove residual charge.

## III. EXPERIMENTAL RESULTS

### A. SPACE CHARGE PROPERTIES AT ROOM TEMPERATURE

Fig. 3 shows the space charge distribution of specimen A without graphene coating at room temperature. It can be found from Fig. 3a that a small amount of charge is injected from both anode and cathode at room temperature and 10 kV/mm, accumulating as homocharge. Indeed, for polyethylene-based insulation the threshold field separating ohmic and space-charge limited conduction (SCLC) ranges between 10 and 20 kV/mm [2], [15]. At depolarization phase, the presence of homocharge in the vicinity of both anode and cathode provides further evidences of the observation reported above. When electric field is risen to 40 kV/mm, a small amount of negative charge moves and reaches the anode in the first part of polarization, as shown in Fig. 3b. Then, charge is injected from both electrodes: homocharge accumulation at the anode neutralizes the abovementioned negative charge from cathode, and its injection depth in the polymer is deeper than that from cathode. During depolarization phase, homocharge accumulation in the vicinity of both electrodes is clearly observable, dissipating slowly with time.

The space charge distribution of specimen B with graphene coating outside the semicon side is shown in Fig. 4.

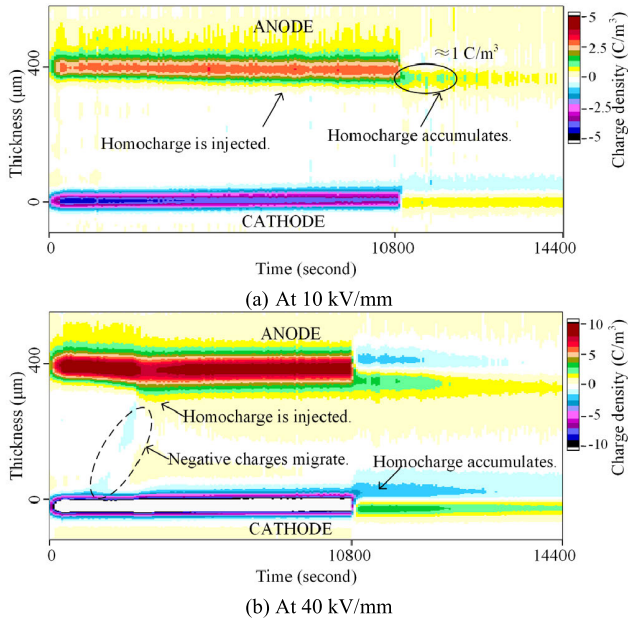


FIGURE 3. Space charge distribution of specimen A at room temperature.

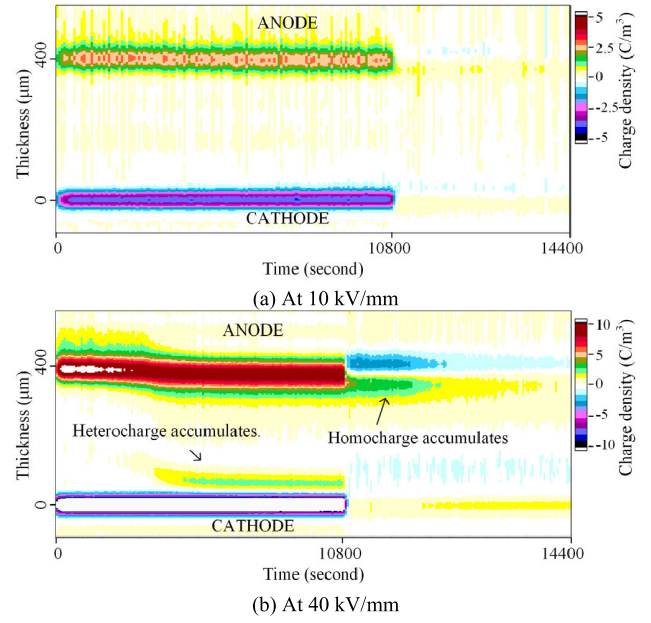


FIGURE 5. Space charge distribution of specimen C at room temperature.

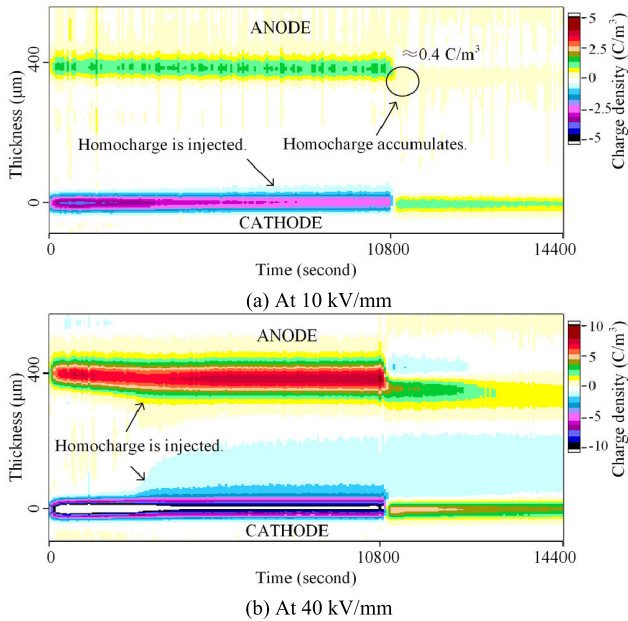


FIGURE 4. Space charge distribution of specimen B at room temperature.

Fig. 4a indicates that the space charge distribution of specimen B is similar to specimen A at 10 kV/mm. However, the homocharge density accumulating near anode in specimen B,  $0.4 \text{ C/m}^3$ , is smaller than that in specimen A,  $1 \text{ C/m}^3$ . When the electric field is risen to 40 kV/mm, an evident homocharge injection can be observed near both electrodes during poling phase, see Fig. 4b. The injection depth of homocharge from cathode is high, and negative charge dominates. The interfacial charge density of specimen B between semicon and XLPE is lower than that of specimen A during poling phase.

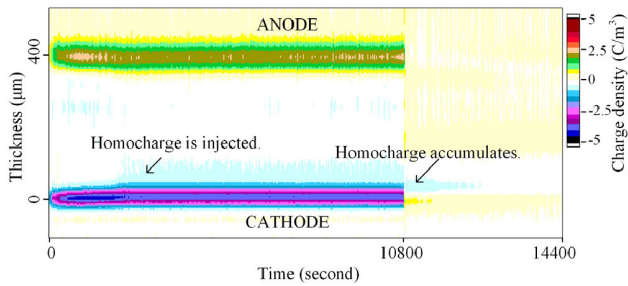
Fig. 5 shows the space charge profiles of specimen C coated with graphene on both sides of semicon layer and XLPE at room temperature. At low electric field as shown in Fig. 5a, charge injection from the electrode is prevented. When electric field rises to 40 kV/mm, as shown in Fig. 5b, during poling phase positive charge is injected from anode and accumulates close to the anode, as clearly observable during depolarization phase. Other positive charge, probably injected from anode, migrates and accumulates in the vicinity of cathode. No clear injection can be seen near the cathode. During depolarization, the heterocharge accumulated in the vicinity of cathode is fast dissipated, while homocharge close to the anode takes long time to decay completely.

Comparing with specimen A and B, when XLPE side is coated with graphene, no injected charge can be seen, and heterocharge migrating from anode accumulates. During depolarization, injected homocharge can only be seen in the vicinity of anode. Therefore, graphene coating on the surface of XLPE is seen to suppress quite well the negative charge injection and block heterocharge while it seems less efficient on preventing injection on the semicon side.

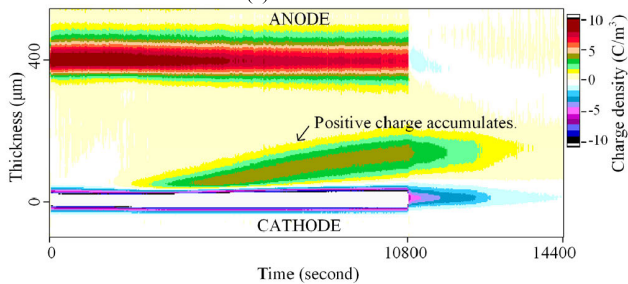
**B. SPACE CHARGE DISTRIBUTION AT 60 °C**

Fig. 6 is the space charge distribution of specimen A at 60 °C. At 10 kV/mm, a small amount of charge, accumulating as homocharge, is injected from cathode as shown in Fig 6a. When electric field is risen to 40 kV/mm as shown in Fig 6b, positive charge accumulates in the vicinity of cathode and neutralized the negative charge injected from cathode after the voltage is applied about 10 min. The density of positive charge also increases with time, which broadens the width of positive charge accumulating in XLPE.



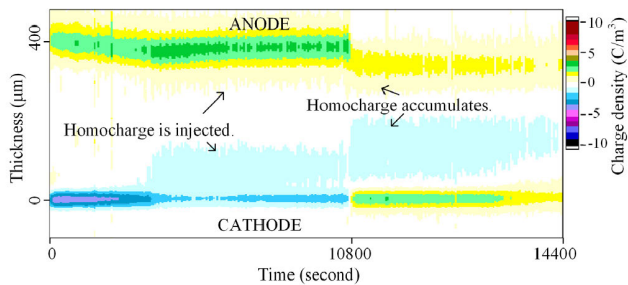


(a) At 10 kV/mm

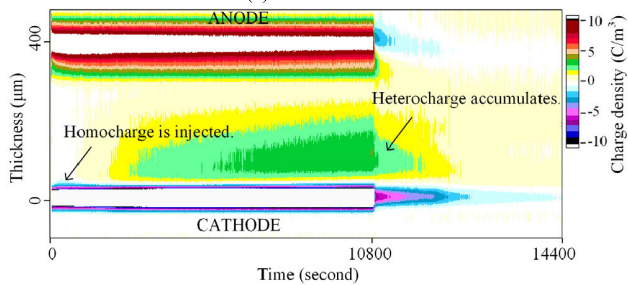


(b) At 40 kV/mm

FIGURE 6. Space charge distribution of specimen A at 60 °C.



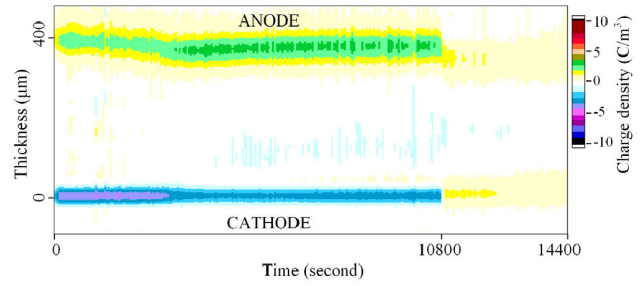
(a) At 10 kV/mm



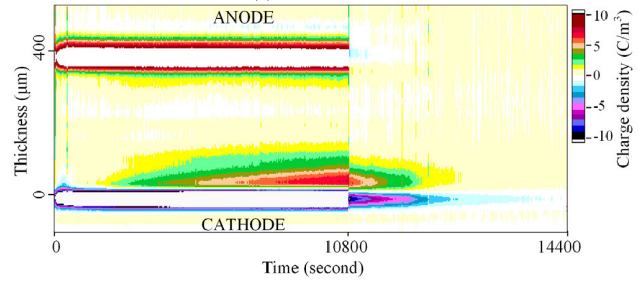
(b) At 40 kV/mm

FIGURE 7. Space charge distribution of specimen B at 60 °C.

Fig. 7 displays the space charge distribution of specimen B at 60 °C. Because of the increasing of temperature, a larger amount of charge is injected from both electrodes, with respect to room temperature, even at 10 kV/mm, with prevalence of negative charge. When the electric field is risen to 40 kV/mm the situation is reversed: a small amount of negative charge accumulates in the vicinity of cathode at the initial stage, but it will be neutralized by positive charge from anode with time. Then, a large quantity of positive charges accumulates near the cathode. At high temperature, graphene coating on the outside of semicon cannot suppress positive charge injection.



(a) At 10 kV/mm



(b) At 40 kV/mm

FIGURE 8. Space charge distribution of specimen C at 60 °C.

Fig. 8 represents the space charge distribution of specimen C at 60 °C. Comparing with space charge of specimen B, the injected charge quantity of specimen C from cathode at 10 kV/mm is much smaller. At 40 kV/mm, the charge density of heterocharge accumulating in specimen C behind cathode is larger than that in specimen B, which may be due to graphene coating preventing charge extraction from cathode too.

#### IV. DISCUSSION

##### A. TRAP DISTRIBUTION

According to the isothermal decay current (IDC) method which is a general method used to the study of trap energy of organic solid [16], an approximate method for calculating the trap distribution can be obtained. The relationship between surface potential  $V_s(t)$  and current density  $j(t)$  in depolarization process can be expressed as:

$$j_{decay}(t) = \frac{C}{A} \frac{dV_s(t)}{dt} \quad (1)$$

$$C = \epsilon_0 \epsilon_r A / d \quad (2)$$

where  $C$  is the equivalent capacitance of the sample,  $A$  and  $d$  are the surface area and thickness of sample respectively. The relationship between surface space charge density and surface potential  $V_s$  is:

$$\sigma(t) = \epsilon_0 \epsilon_r V_s(t) / r' \quad (3)$$

where  $r'$  is the mean injection depth of charges.

If  $r'$  is assumed to be a selected region  $\Delta x$ , no more than about 100  $\mu\text{m}$ , from electrode interface to insulation as shown in Fig. 9, the surface space charge density can be expressed as:

$$\sigma(t) = \int_0^{\Delta x} |\rho_{dp}(t, x)| dx \quad (4)$$

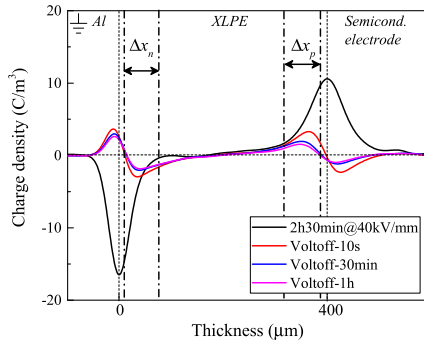


FIGURE 9. Selected region of the mean injection depth of charges in specimen A.

where  $\rho_{dp}(t)$  is the space charge volume density of insulation during depolarization phase;  $\Delta x$  can be equal to  $\Delta x_p$  or  $\Delta x_n$ , depending on which charge (positive or negative) is considered.

The decay characteristic of positive or negative space charge density can be fitted by a double exponential function, so the relationship between decay current density and charge density of space charge during depolarization can be obtained by (1) to (4) as:

$$j_{decay}(t) = \frac{\Delta x}{d} \frac{d\sigma(t)}{dt} \quad (5)$$

where  $r'$  is equal to  $\Delta x$ .

Then, the trap depth,  $E_t$ , and trap energy density,  $N(E_t)$ , can be calculated by:

$$E_t = kT \ln vt \quad (6)$$

$$N(E_t) = \frac{2t}{qdKT} f_0^{-1}(E_t) j_{decay}(t) \quad (7)$$

where  $k$  is Boltzmann's constant, which is  $8.568 \times 10^{-5} \text{ eV/K}$ ,  $T$  is absolute temperature,  $\nu$  is de-trapping attempt frequency which is  $kT/h$ ,  $h$  is Planck constant. It should be noted that the trap distribution calculated according to the above method is the trap depth of trapped charges and does not reflect the entire trap distribution in the material.

Fig. 10 shows trap distribution at room temperature with a selected region  $\Delta x$  is about  $60 \mu\text{m}$ . For the positive charge trapped in XLPE in the vicinity of the anode, the obtained trap distribution is plotted in Fig. 10a. All specimens have about two levels of deep traps. The shallowest trap depth of reference specimen, namely specimen A, is about  $0.89 \text{ eV}$ , which is slightly larger than that of specimen B and C,  $0.87 \text{ eV}$ , coated with graphene on the outside of semicon. The depth of deeper traps is about  $0.95 \text{ eV}$ . It can be concluded that graphene coating on the outside of semicon can reduce the depth of shallow trap occupied, which causes that positive charge detraps more easily and transfers into the bulk of XLPE near anode.

Fig. 10b shows the trap distribution of negative charges trapped in XLPE close to the cathode. The trap depth of specimen without graphene coating is about  $0.95 \text{ eV}$  by

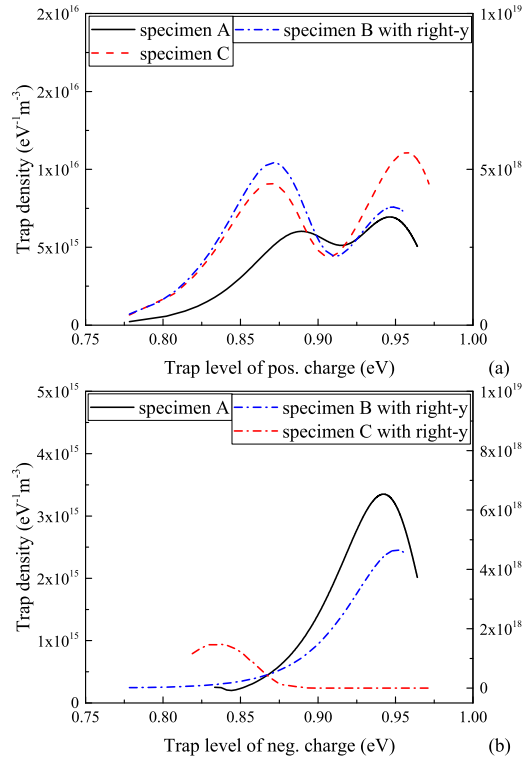


FIGURE 10. Trap distribution under room temperature near (a) anode, and (b) cathode.

contrast, the trap depth of specimen C with the graphene coating on the XLPE layer is less than  $0.85 \text{ eV}$ . This provides evidence that when graphene is directly coated on XLPE side, charges are only trapped in the shallow trap of XLPE. In this case, the trapped charges mainly originate from anode. Indeed, homocharge injection from cathode decreases, and not enough negative charge is able to neutralize the positive charge migrating from anode, which finally leads to heterocharge accumulation in the vicinity of cathode.

### B. INFULENCE OF TEMPERATURE

The space charge distribution of different specimens also depends on the temperature. At  $40 \text{ kV/mm}$ , the space charge distribution is interesting. At lower temperature, the electrode interfacial charge is significantly changed by graphene coating. As discussed in the previous section, the shallower trap level of XLPE occupied is lowered with the graphene coating, which results from a small amount of positive charge accumulation. However, when temperature is increased to  $60 \text{ }^\circ\text{C}$ , the accumulation of positive charge in both specimen B and C is evident. In addition, positive charge does not immediately accumulate in the vicinity of cathode when the electric field is applied. In this case, indeed, it takes more than  $20 \sim 30$  minutes after the electric field is applied, when the positive charge packet can be noticed. Moreover, sometimes, the space charge distribution does not reach a stable state even though the electric field is applied more than 3 hours, especially at high temperature and electric field. Therefore,

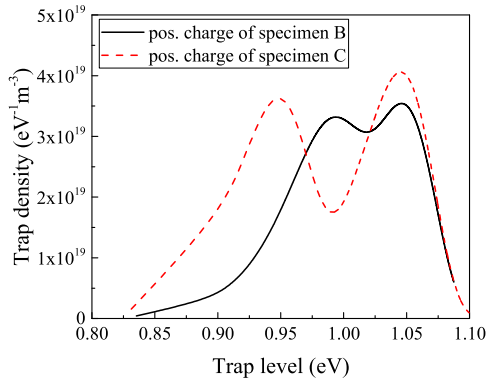


FIGURE 11. Trap distribution at 60 °C.

when one calculates the trap distribution, the specimen should be polarized as long as enough until the space charge reaches a steady-state.

The trap distribution of specimen B and C at 60 °C and 40 kV/mm with a selected region  $\Delta x$  is about 100  $\mu\text{m}$  is plotted in Fig. 11. It should be noted that only trapped positive charges in a selected region of about 100  $\mu\text{m}$  near cathode are calculated because there is no evidence of negative charge in specimens at 60 °C. It can be seen that the deepest traps of both specimen B and C is about 1.05 eV. The shallowest trap level of specimen C is about 0.95 eV, smaller than that of specimen B, 0.99eV. Due to the lower shallow trap level, more positive charges can be trapped and de-trapped during volt-on, forming charge carriers.

C. SUPPRESSION MECHANISM OF SPACE CHARGE

It is possible to explain the effect of graphene coating on XLPE side contacting with cathode in accordance with Fig. 12. Fig. 12a is the band structure of specimen before contact, where the band gap  $E_g$  of XLPE is about 8 ~ 9 eV, work function of Aluminum  $W_m$ , semicon  $W_s$  and graphene is about 4.04 eV, 5.6 eV and 4.48 eV, respectively [17], [18]. In a freestanding graphene layer the Fermi energy of conduction and valence bands meets at the conical point, namely the Dirac point.

Generally, when electric field is applied as shown in Fig. 12b, the potential barrier of hole injection from anode lowers, which results in the positive charge being injected and hopping easily between shallow traps above valance band in XLPE, so the positive charges can be injected more easily than negative charge, and form homocharge accumulation in the vicinity of anode. On the contrary, at cathode, with the rise of electric field, the Dirac point of graphene shifts upward, which provides a similar effect to the introduction of deep traps. Moreover, energy bands of XLPE also are distorted. In this case, only when negative charges from cathode jump over both the Dirac point and potential barrier of XLPE, charges can be injected into XLPE, so negative charge injection is limited at the metal electrode. At 40 kV/mm, conduction band further bends, the probability of hole passing

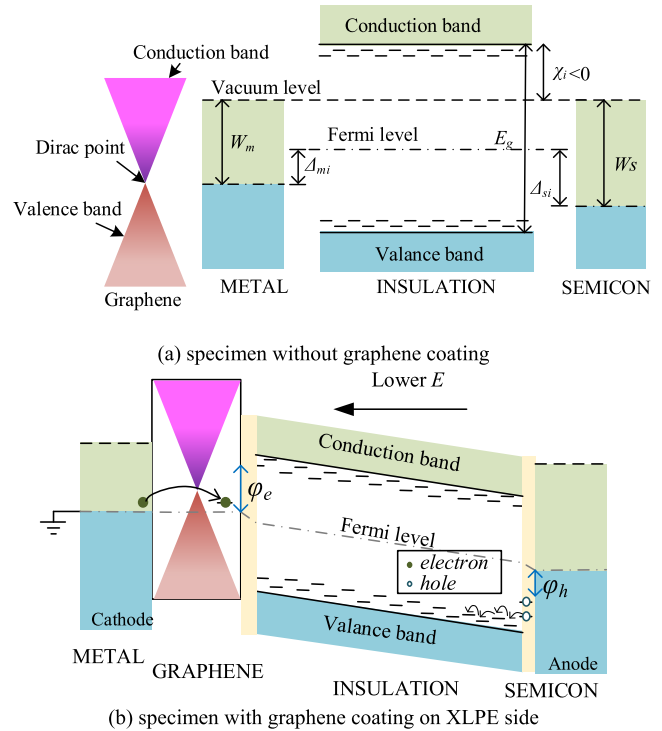


FIGURE 12. Band structure of specimen.

through barrier also increases, which leads the positive charge further accumulating on the front of cathode. If no graphene is coated on XLPE side, negative charge can more easily jump over or tunnel potential barrier at high electric field, which explains why a small amount of negative charges accumulates in specimen B (see Fig. 4b).

V. CONCLUSION

For suppressing the injection and accumulation of space charge in the insulation of HVDC cables, a thin graphene coating layer can be deposited on the insulation surface. Via PEA technique, the space charge distribution of specimens coated with graphene was tested, and the distribution of traps trapped positive and negative charges was calculated. At room temperature, graphene coating on XLPE side can effectively suppress the injection of negative charge from cathode. Moreover, when a layer of graphene was only coated on the semicon side, positive charge accumulating near the anode seems to be blocked, which means that the quantity of charges injected from semicon electrode decreases. At higher temperature, graphene coating can also prevent charge extraction from cathode, so the quantity of positive charges which is neutralized by negative charges lowers. Indeed, more positive charges are seen to accumulate in the vicinity of cathode. The analysis of energy band indicates that Dirac point of graphene on XLPE side can affect the injection of negative charge. With the raise of electric field, Dirac point moves upward, which results in reducing the negative charge injected into conduction band of XLPE.



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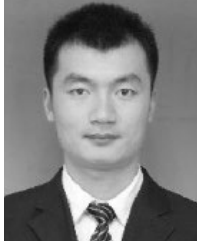
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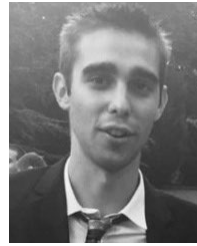


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nanofibers and their integration in smart composite laminates for energy harvesting and sensing.

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the PEA methods, charging–discharging current measurements, and life tests.

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