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Evaluation of space charge accumulation characteristics in double-layered samples to simulate the behaviour of transmission cable joints under DC voltage

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ABSTRACT

In this paper we investigated the space charge accumulation characteristics in single and double-layered samples composed of EPR laminated on top of LDPE. As a result, even though only a small amount of charge accumulated in the single-layered EPR and LDPE, an extremely large amount of charge accumulation was observed in the double-layered EPR/LDPE, resulting in the electric field to significantly increase locally just after polarity reversal. We also tried to reveal this peculiar phenomenon using Quantum chemical calculation and confirmed that there is a possibility that we can explain the charge accumulation characteristics in double-layered samples.

KEYWORDS

Space charge, Pulsed electro-acoustic method, Double-layered dielectric, DC transmission cable joints, Quantum chemical calculation

INTRODUCTION

Recently, AC transmission which can be easily transformed, is the mainstream in urban areas. However, the increase of demand for long distance DC transmission is rising. This is due to the rising demand to transmit electricity from power plants using renewable energy in land and ocean faraway from urban areas, since it is more efficient to use DC transmission. As for the insulating material used for DC transmission cables, it is ideal to use maintenance free polymer materials, such as polyethylene based insulating materials. However, even though polyethylene based insulating materials show a high insulating performance for AC transmission, in DC transmission, space charge accumulates inside the insulator which may cause a dielectric breakdown. There have been reported that in LDPE (Low Density Polyethylene), under a high electric field, a large amount of packet like positive charge would accumulate and the electric field would be emphasized locally, resulting in a breakdown of the insulator [1]. Moreover, in long distance DC transmission cables, there are cable joints. These joints are composed of different dielectric layers. Therefore, space charge accumulation caused by laminating different types of insulating materials may distort the electric field. Especially, when inverting the direction of the power flow using LCC (Line Commutated converter) system, there is a need to invert the voltage polarity to change the roles of the rectifier and the inverter, which has a high possibility of leading to the malfunction upon cable joints. However, there have been few reports on this phenomenon. Therefore, in this paper we investigated the influence of polarity reversal on space charge accumulation and electric field behaviors, using the PEA (Pulsed Electro-Acoustic) method [2] when applying DC voltage upon double-layered samples composed of different insulating materials. Moreover, we focused upon the interface charge accumulation characteristics in a double-layered sample composed of EPR (Ethylene Propylene Rubber) laminated on top of LDPE ("EPR/LDPE"), as a model of cable joints for normal AC transmission line, both commercially available, and we used Quantum chemical calculation to identify issues occurred in cable joints for DC transmission.

MEASUREMENT RESULTS

Measurement condition

For the measurements in this paper, the space charge distribution in the samples were observed under various dc stresses. The polarity of the applied voltage is described as "positive" and "negative" voltage depending on the polarity of the high voltage electrode in the system. The material of the high voltage electrode and the grounded electrode are a semi-conductive layer (SC) and aluminium (Al) respectively. Therefore, when a positive voltage is applied to the sample, the SC side would be the "anode" and the Al side would be the "cathode".

Figure 1 shows the voltage application procedure. After applying a positive voltage corresponding to an electric field of 40 kV/mm for 30 min, the polarity of the applied voltage was reversed and a negative voltage corresponding to an electric field of -40 kV/mm was applied for 30 min. We conducted this procedure for 2 cycles.

Single-layered EPR and LDPE

Figures 2 and 3 show the results of the voltage polarity reversal upon single layered LDPE and EPR, respectively. In each Fig, (a) and (b) show the time dependent space charge and electric field distributions respectively, and (c) shows the time dependent maximum electric field.

As shown in Fig. 2(a), a small amount of positive charge was injected from the anode, and this positive charge

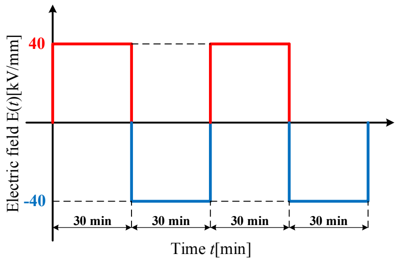


Fig. 1: Voltage application procedure

accumulated thinly inside the sample by time then it finaly reached the cathode. This phenomena was observed in all the voltage application procedures, regardless of the polarity of the applied voltage. From Fig. 2(b) we can see that when this positive charge reached the cathode (Al), the electric field near the cathode (Al) slightly increased. Therefore the electric field would always increase near the cathode(Al) after a few minutes, every time the applied voltage was reversed. However, from Fig. 2(c). it can be seen that maximum electric field fell into the extent of less than 55 kV/mm at most. On the other hand, as shown in Fig. 3(a), almost no charge accumulation was observed in EPR, regardless of the polarity of the applied voltage nor the times of polarity reversal. Therefore, as shown in Fig. 3(b) and (c), the maximum electric field hardly changed and stayed at around the applied average electric field throughout the measurement.

Double-layered EPR/LDPE

Figure 4 shows the results of double-layered EPR/LDPE when the polarity of the applied voltage was reversed. In each Fig, (a) and (b) show the time dependent space charge and electric field distributions respectively. As for (c) and (d), these Figs show the time dependent maximum electric field in the EPR side and the LDPE side respectively.

As shown in Fig. 4(a), when a positive voltage was applied, positive charge appeared from the interface between the two samples and it moved into LDPE towards the cathode (Al). After this positive charge drifted, an extremely large amount of negative charge accumulated at the interface of these two samples in the place of this positive charge. From the electric field distribution as shown in Fig. 4(b), after 30 minutes of voltage application the electric field in EPR had increased up to around 70 kV/mm although it had decreased to around 20 kV/mm in LDPE. Next, when the polarity of the applied voltage was reversed, a large amount of packet like positive charge was injected form the anode (Al) and this time, the maximum electric field in LDPE increased significantly (70 kV/mm). However, after a few minutes, the negative charge accumulated at the interface of the two layers was replaced by a large amount of positive charge. At this state, the electric field in EPR increased (70 kV/mm) although it decreased in LDPE (20 kV/mm). In the second cycle of voltage application, a similar phenomenon was also observed.

From these results, although a slight amount of charge accumulation was observed in LDPE, in single-layered EPR and LDPE, altogether only a small amount of charge accumulation was observed. Therefore, the maximum electric field only slightly increased. On the other hand, in the double-layered EPR/LDPE, an extremely large amount of charge accumulation was observed at the interface of the two layers and when the polarity reversal was conducted, a packet like positive charge which was not observed in the single-layered samples were injected from the anode or interface depending on the polarity of the applied electric field. When this happened, the maximum electric field at the LDPE side increased significantly, reaching almost twice the applied average electric field (around 70 kV/mm).

Resistance sharing

When a DC voltage is applied to a two-layer dielectric,

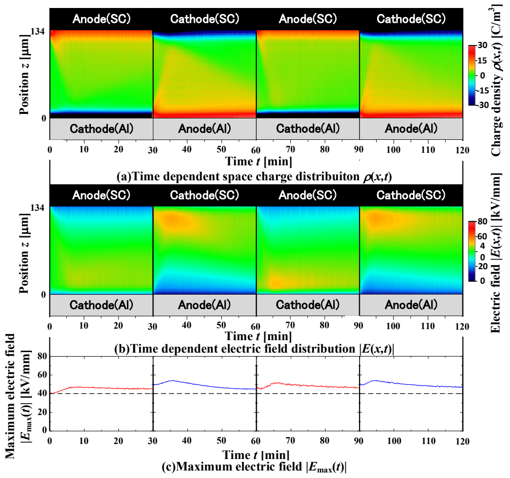


Fig. 2: Space charge distribution in LDPE

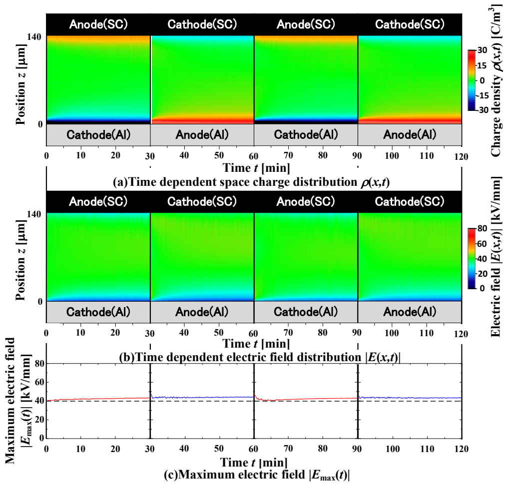


Fig. 3: Space charge distribution in EPR

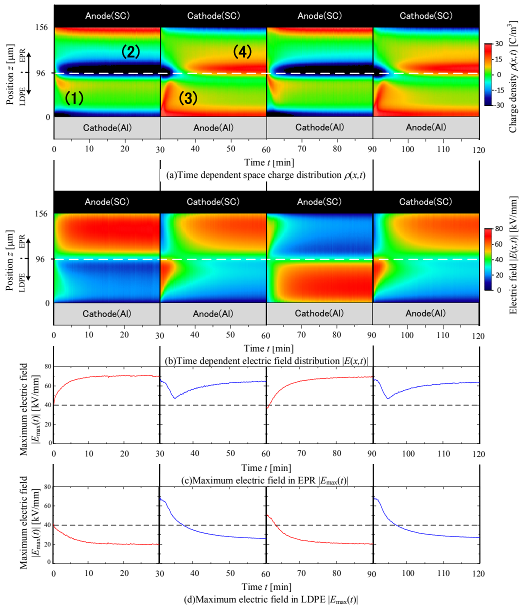


Fig. 4: Space charge distribution in EPR/LDPE

Table. 1: Various quantities of LDPE and EPR

Sample	HOMO level[eV]	LUMO level[eV]	Band gap Φ_g[eV]	Fermi level E_F[eV]	Electron Affinity X[eV]	Ionization energy Φ_i[eV]
LDPE	-7.51	1.48	8.99	3.02	-1.48	7.51
EPR	-5.89	0.86	6.75	-2.52	-0.86	5.89

theoretically, space charge accumulates at the interface of these two dielectrics according to the difference in relative permittivity or conductivity. This depends on the relation between DC voltage application time t and the whole of the dielectric relaxation time τ. When the DC voltage application time t is (t<<τ), this will be a capacitor sharing. On the other hand, when the relation between t and τ is (τ<<t), it becomes a resistance sharing. However, as shown in Fig 4, many types of space charge accumulation occur in the actual two-layer dielectric, which makes it impossible to explain by only using the above theory. The following equation explains the conductivity κ in dielectrics.

$$\kappa = \mathrm{e}n\mu \qquad [1]$$

e is the elementary charge, n is the density of carrier, and μ is the carrier mobility. From this equation, the conductivity κ is proportional to the number of charge. This means that if the material which makes contact with the dielectric changes, the conductivity κ also changes as will be described later. This means that we cannot measure the conductivity of a single-layered sample and apply it to a double-layared sample. Therefore, a different theory would be needed to explain the charge accumulation characteristics in double-layered samples.

QUANTUM CHEMICAL CALCULATION

Calculation condition and results

The calculation targets were LDPE and EPR. By solving the Schrődinger equation approximately, we achieved the "electron energy level", "electron density distribution"and "electrostatic potential distribution". From these results, we evaluated where and how charge accumulates in the molecular structure. The calculation procedures are written elsewhere [3]-[5].

Figure. 5 shows the energy band structure of LDPE and EPR. The vertical axis is a positive direction for the electron's energy. Therefore, the positive direction for hole is the opposite. Table 1 shows the various quantities of EPR and LDPE. The HOMO (Highest Occupied Molecular Orbital) level is the highest level of the valence band. On the other hand, the LUMO (Lowest Unoccupied Molecular Orbital) level is the lowest level of the conduction band. The band gap(forbidden band) Φ_g stretches between the HOMO level and LUMO level, where electron cannot exist. The Fermi level is set in the middle of the band gap. This is where the existence probability of electron is 50 %. The ionization energy is the energy needed to extract electron from neutral molecules and to become a cation. Moreover, the electron affinity X is the energy needed to take in electron and be an anion. In these calculation for LDPE and EPR, we assumed the molecular structures as will be shown later in Figs. 7 and 8.

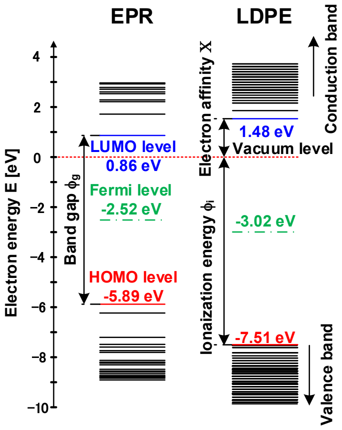


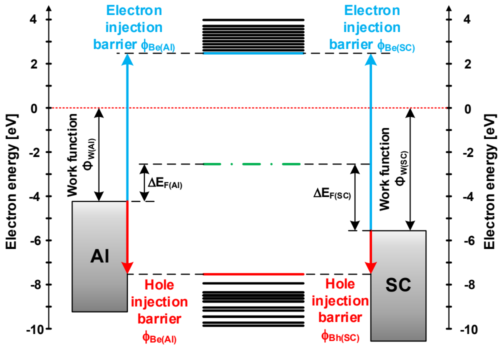
Fig. 5: Band diagram of LDPE and EPR

From Fig 5 and Table 1, LDPE has a wider band gap compared to EPR. This means that electron is harder to be excited in LDPE than in EPR. However, both LDPE and EPR have a wide band gap (10.03 eV and 6.75 eV respectively). Therefore it is unlikely that electron is excited even under a high electric field.

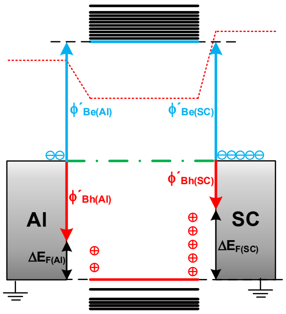
Single-layered EPR and LDPE

Charge injection barrier

Figures 6(a) and (b) show the charge injection barrier before and after an insulating material has made contact with the SC and Al electrodes respectively. When measuring the space charge distribution using the PEA system, we sandwich samples with the semi-conductive electrode (SC) and the aluminium electrode (Al).

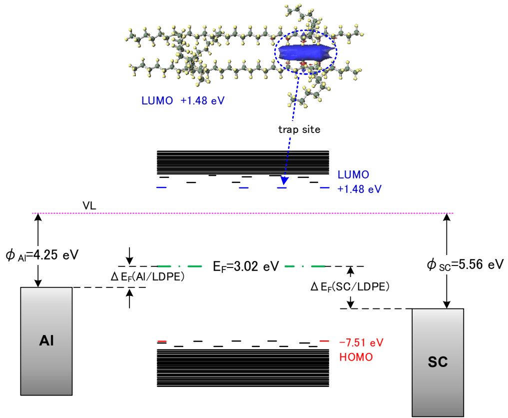


(a): Before contact

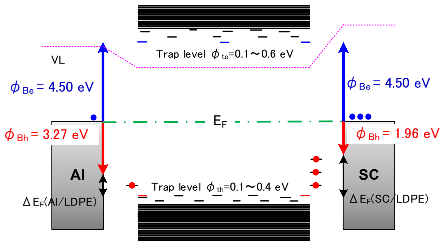


(b): After contact

Fig. 6: The generation of electric double layer caused by contact charging

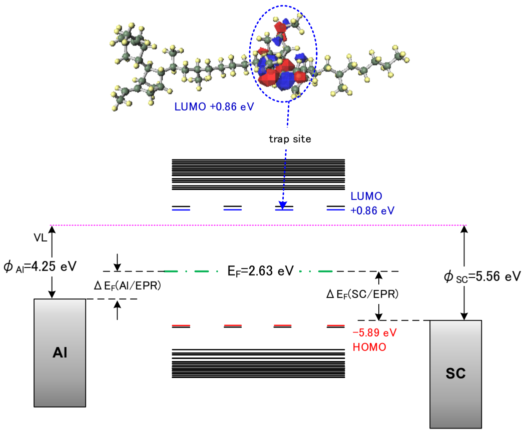


(a): Before contact

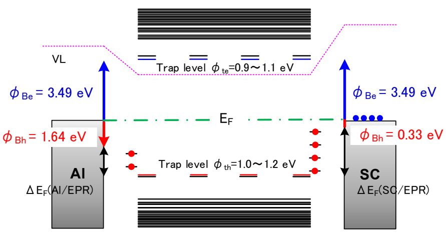


(b): After contact

Fig. 7: Localized band structure in LDPE



(a): Before contact



(b): After contact

Fig. 8: Localized band structure in EPR

Therefore, we have drawn the electron energy level when the sample has made contact with these two electrodes. When two materials contact, electron moves from the material with the higher Fermi level to the material with a lower Fermi level until the Fermi level of these two materials have coincided, creating a electric double layer [6]. Therefore, since LDPE has a higher Fermi level than both the SC and Al electrode, as shown in Fig. 6(b), negative charge from the LDPE layer accumulates in both electrodes and positive charge accumulates at either of the surface of LDPE and a hole level is fomed in the band gap forming a electric double layer. This means that positive charge is already injected into LDPE even before voltage application. As a result, the vaccum level distorts and the positive charge injection barrier decreases by ΔE_F.

Localized energy bands in LDPE and EPR

Figures 7 and 8 show the localized energy band before and after contact in LDPE and EPR respectively and Table 2 shows the charge injection barrier and trap depths in LDPE and EPR. The energy band structures of these are calculated using the molecular structure model shown at the top of each figure. In the case of LDPE, we assumed two straight chains sith several propyl groups as the model of branches of the chain. The localized band was made by drawing where each energy level shown in Fig. 5 exists in the molecular orbital. For example, if the energy level was distributed throughout the molecular chain, then we have drawn the energy level along the molecular chain. However, if it locally exists then we have only drawn it where it locally exists.

As shown in Fig. 7, in LDPE, the electron density distribution spreads around the branched points or where the molecular structure is close together. This becomes traps for electrons and holes. The trap depths were 0.1~0.4 eV for holes and 0.1~0.6 eV for electrons. After contact, an electric double layer was formed and positive charge was seen accumulated at the surface of LDPE. This means that LDPE has shallow traps. Therefore, once charge accumulates inside the sample, it can move easily in accordance to the electric field. On the other hand, LDPE has a higher barrier against electron injection (4.50 eV) compared to the barrier against hole injection (1.96 eV) as shown in table 2. Therefore, in LDPE, hole can be easily injected into the sample and once it has, it can move easily. However, electrion is extremely hard to be injected into the sample. This explains the reason why in LDPE, as shown in Fig. 2, only positive charge accumulation can be observed and why this positive charge had accumulated thinly throughout the sample in a short time. Moreover, this explains the large amount of packet like positive charge being injected just after polarity reversal inEPR/LDPE as described as (3) shown in Fig. 4. However the details for the phenomenon in EPR/LDPE are described later.

On the other hand, in EPR, as shown in Fig. 8, diene

Table. 2: Charge injection barrier and trap depths in LDPE and EPR

Sample	SC electrode side		Al electrode side		Hole trap depthΦ_{th}[eV]	Electron trap depthΦ_{te}[eV]
	Hole injection barrier$\Phi_{Bh(SC)}$[eV]	Electron injection barrier$\Phi_{Be(SC)}$[eV]	Hole injection barrier$\Phi_{Bh(Al)}$[eV]	Electron injection barrier$\Phi_{Be(Al)}$[eV]		
LDPE	1.96	4.50	3.27	4.50	0.1 ~ 0.4	0.1 ~ 0.6
EPR	0.33	3.49	1.64	3.49	1.0 ~ 1.2	0.9 ~ 1.1

(ethylidene norbornene) which have double bonds which form localized energy levels causing traps for electrons and holes (0.9~1.1 and 1.0~1.2 eV respectively). After contact, an electric double layer was formed and positive charge was seen accumulated at the surface of EPR and the hole injection barrier as shown in the same figure is extremely low (0.33 eV). However, as shown in Fig. 3, no clear charge accumulation was observed in EPR. This can also be explained by Fig. 8. Even though the positive charge injection barrier is low (0.33 eV), it can be confirmed that unlike LDPE, EPR has a deep positive and negative charge trap (0.9~1.2 eV). Therefore, even if charge is injected into the sample from the electrode, this trap prevents charge to move any deeper into the sample.

Double layered EPR/LDPE

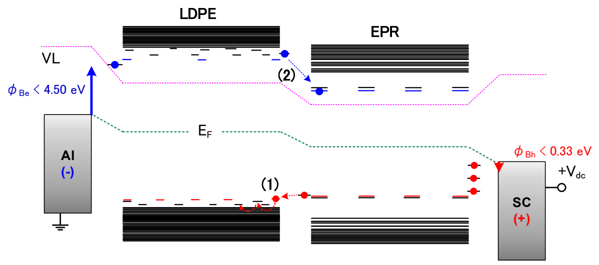
Charge injection barrier at EPR/LDPE interface

Here, we try to explain where the interface charge which caused the local electric field increase after voltage reversal in EPR/LDPE shown in Fig. 4, appeared from. For this, we have especially focused upon the charge movement which we have numbered (1)-(4) in the same figure.

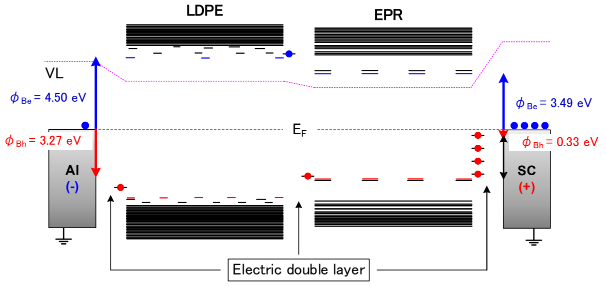
Figure. 9(b) shows the charge injection barrier and trap sites under a short circuit after contact in double-layered EPR/LDPE. Since the Fermi level of EPR (-2.52 eV) is slightly higher than LDPE (-3.02 eV), after contact, positive and negative charge accumulates in EPR and LDPE respectively, causing a electric double layer. At this point, the difference in LUMO and HOMO level between EPR and LDPE are both 1.12 eV, becoming a trap for positive charge. As shown in Table. 2, this trap has around the same depth as EPR, which was already enough to stop charge from moving into the sample, as shown in Fig. 3. Therefore, it is not easy for positive charge to overcome this trap and move from EPR to LDPE.

Figures. 9(a) and 9(c) show the charge injection barrier and trap sites when a positive or negative voltage is applied to EPR/LDPE respectively, after contact. High voltage is applied to polymeric materials via electrodes when used as electrical insulating materials. Therefore, it is important to consider how the charge injection barrier changes under voltage application from the viewpoint of insulation design. From Fig. 9(a), when a positive voltage is applied via the SC electrode, the Fermi level becomes a potential distribution of a downward slope to the right. Since the vertical axis is a positive direction for electron, the direction of the positive voltage becomes the opposite of this. However, since the diagram will become complicated, in this paper, we have left the localized band inside the sample as a horizontal line.

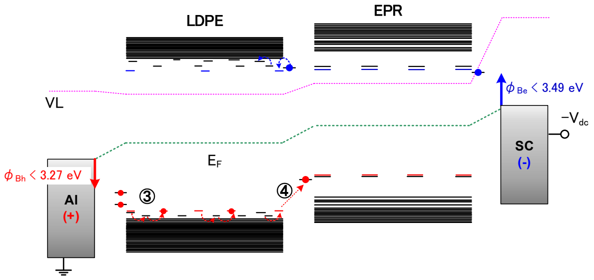
Firstly, EPR's HOMO level draws closer to the anode's (SC) Fermi level causing positive charge to be injected easier into EPR. The same goes for the interface of EPR and LDPE. Since the LDPE's HOMO level draws closer to EPR's HOMO level, this trap depth becomes shallower, resulting in the positive charge to be injected easier from EPR to LDPE. On the other hand, negative charge would move from LDPE to EPR. Moreover, as for the cathode, the negative charge injection barrier decreases and the vaccum level increases. This causes the "reversal of the electric double layer". Since the cathode's (Al) Fermi level becomes higher than LDPE's Fermi level, in addition to the positive charge which had already accumulated by contact, negative charge would accumulate by voltage application. The same goes for the positive charge at LDPE and the negative charge at EPR at the interface of EPR/LDPE, which have accumulated for the same reason. Consequently, both positive and negative charge from the anode and cathode are easily injected into EPR and LDPE respectively compared to before voltage application. On the other hand, from Fig. 9(c), when a negative voltage was applied, this time EPR's LUMO level draws closer to LDPE's LUMO level resulting in the negative charge to move from EPR to LDPE easily. As for the positive charge, since EPR's HOMO level has moved further away, the interface of the two layers have become an even deeper trap than before voltage application. Moreover, we can see that the reversal of the electric double layer has caused more charge to accumulate at the interface of EPR/LDPE.



(a): positive voltage application



(b): Short circuit (After contact)



(c): negative voltage application

Fig. 9: Space charge accumulation characteristics in EPR/LDPE

Positive voltage application to EPR/LDPE band

Next we explain the interface charge characteristics shown in Fig. 4 using this band theory we have just explained. When we take a look at the space charge distribution one more time, firstly, (1) positive charge is injected into LDPE from the interface between the two layers. After this has occurred, (2) a large amount of negative charge accumulated in its place at the interface. When we take a look at Fig. 9(a), we can see that positive charge had already accumulated at the interface of both EPR and LDPE by contact. As shown in Fig. 7, LDPE has only shallow traps. Therefore, by positive voltage application, the positive charge at the surface of LDPE would move towards the cathode (Al) and the positive charge at the interface of EPR would move into LDPE and also move towards the cathode (Al). This explains the positive charge being injected from the interface of EPR/LDPE into LDPE as seen in (1). As for the negative charge , we can see that the negative charge accumulated at LDPE would move straight to EPR by voltage application. However, from Fig. 8, EPR has a deep trap for electron (0.9 ~ 1.1 eV) which means that the negative charge which have been injected from LDPE or which have already been accumulated at the interface of EPR cannot move into EPR and would most likely accumulate at the interface of EPR/LDPE as seen in (2). This explains the large amount of negative charge accumulated at the interface of EPR/LDPE.

Negative voltage application to EPR/LDPE band

As shown in Fig. 9(c), when a negative voltage is applied to the EPR/LDPE band, positive charge accumulated at the anode (Al) side can move straight through LDPE and accumulate at the interface of EPR. However, since EPR has a deep trap for positive charge too (1.0 ~ 1.2 eV), this positive charge stays accumulated there as seen in (3). As for the negative charge accumulated in LDPE, this would move straight towards the anode (Al). This explains the space charge characteristics in Fig. 4 showing (4) a large amount of positive charge accumulated at the interface of EPR/LDPE, after (3) a large amount of positive charge being injected from the anode (Al) was observed. As for the large amount of negative charge, which had accumulated at the interface of EPR/LDPR before polarity reversal, being injected into LDPE, this can also be explained by the band theory. From the EPR/LDPE band shown in Fig. 9(a), before polarity reversal, a large amount of negative charge had accumulated at the interface of EPR. At this moment when the polarity is reversed, all of this negative charge would move towards the anode (Al). Since the LUMO level of EPR and LDPE have drawn close due to the negative voltage application, this will succeed and as a result, positive charge would accumulate in its place.

In any case, we revealed that by using Quantum chemical calculation, we may have a possibility of being able to explain the charge accmumulation characteristics at the interface of mult-layered samples. However, we have not considered any impurity which may have a role upon the interface charge. Therefore, this is a subject for future work.

CONCLUSION

In this paper, to identify issues when using cable joints which have an interface of different insulating materials for DC transmission, we used insulating materials for AC transmission and measured the space charge accumulation characteristics under an DC voltage corresponding to an electric field of ±40 kV/mm. Moreover, we used Quantum chemical calculation to consider the space charge accumulation properties in a double-layered sample and achieved the following results.

(1) In single-layered EPR and LDPE, only a small amount of charge accmulation was observed even after polarity reversal and the maximum electric field stayed rather flat.

(2) In double-layered EPR/LDPE, an extremely large amount of charge with the opposite polarity of the applied voltage accumulated at the interface of EPR/LDPE and at the stable state, maximum electric field in EPR increased significantly.

(3) In double-layered EPR/LDPE, after polarity reversal, the maximum electric field reached almost twice (around 70 kV/mm) the average applied electric field (40 kV/mm).

(4) From the Quantum chemical calculation results, we were able to explain the charge accumulation characteristics in double-layered EPR/LDPE

From these results, we made it clear that it is necessary to suppress space charge accumulation near the interface when using cable joints in application for DC transmission.

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