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접촉에 따른 LDPE의 전기전도 현상

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Electrode Contacts and Electrical Conduction in Low Density Polyethylene

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요 약 : 폴리에틸렌과 전극사이의 접촉의 종류에 따른 폴리에틸렌의 전기전도 특성의 변화에 대하여 알아 보았다. 본 연구에서 사용한 접촉은 실리콘유, 반도전성 카본블랙-고분자 복합체 및 sputter한 알루미늄 등이다. 실험결과 접촉의 종류에 따라 LDPE의 전류밀도, 활성화 에너지 및 유효전하이동도 등의 전도현상이 다르게 나타나는 것으로 밝혀졌다. 전력케이블의 내부반도전층 원료로 사용되고 있는 반도전 필름접촉의 전도특성이 sputter한 알루미늄 접촉의 전도특성과 매우 유사한 것으로 밝혀졌으나 실리콘유를 접촉으로 사용한 경우 상당히 다른 전도현상을 갖는다. 실리콘유를 접촉으로 사용한 경우 sputter한 경우에 비하여 활성화 에너지가 월등히 낮았으며 유효전하이동도는 약 2차수 이상 높은 값을 갖는다는 사실을 알았다. 실리콘유를 사용하였을 때 일어나는 현저한 변화는 일정한 전압구간에 걸쳐 전류밀도의 증가율이 감소하는 현상인데, 이 현상은 실리콘유를 접촉으로 사용했을 경우 이종전하의 축적으로 인한 계면분극 현상과 전하 sweeping 현상에 기인하는 것으로 생각된다.

Abstract : Electrical conduction characteristics of low density polyethylene have been investigated using such contacts as silicone oil, semiconductive carbon black compound and sputtered Al. It was observed, as a result, the conduction characteristics change depending on the type of contacts. The major change was that the magnitudes of current densities, activation energies and effective charge mobilities were different depending on the type of contacts. It was also found that the conduction behavior of LDPE with the semiconductive contact is very similar to that with the sputtered Al contact. On the other hand, the silicone oil contact showed a low activation energy and a high effective charge mobility compared to those of sputtered Al contact. A more pronounced change due to the silicone-oil contact was a considerable suppression in a rate of change of current density at a certain voltage range. All observed changes due to the silicone-oil contacts were attributed to the interfacial polarization and subsequent charge sweeping effect enhanced by the accumulation of heterocharges.

INTRODUCTION

Recently the charge injection and thus formed space charge has become one of the important factors that has to be controlled for an improvement of the reliability of polyethylene for cable insulation.[1~3] Regarding this, the contact problem becomes considerably important, since the electrical behavior may be greatly influenced by the type of interface between the electrode and polyethylene. In an actual experimentation, such electrode contacts as vacuum evaporated metals, silicone-oils and semiconductive compounds are widely used. Vacuum evaporated metals are to get a good contact at the interface between the electrode and test film, silicone oils are to get a flash-over or corona-proof experimental set-up, and semiconductive contacts are found in real cables.

Different electrical behaviors such as charge injection or conduction characteristics have been observed when the electrode or contact is different. For instance, F. Chapeau et al. has found that homocharges are developed in polyethylene when the vacuum evaporated metal is used as a contact whereas heterocharges are accumulated when the conductive silicone grease is used as a contact.[4] T. Ditchi et al. has also reported that the composition in semiconductive compound for the strand shield is critical in determining the internal charge distribution in XLPE.[5] Besides, it has been previously reported[6] that the conduction characteristics in low density polyethylene with a silicone-oil contact are different from those with a sputtered Al contact. One of the major modifications in case of the silicone-oil contact is a considerable suppression in the rate of change of conduction currents at certain voltages. At this moment, no information is available on the reasons for such suppression. Also, activation energies for the electrical conduction seem to depend on the type of contact.[6,7] For examples, a graphite contact yields an activation energy of about 1.05 eV, while an evaporated metal contact (Al) about 0.85 eV. On the other hand, the silicone-oil contact yields only about 0.50 eV. In a practical sense, however, such contacts as vacuum evaporated metal electrodes, silicone-oil layer, or conductive grease cannot be found in real cable systems. Since medium voltage cables have a semiconductive layer called a "strand shield" between the metal conductor and the insulation layer, it is quite questionable whether the information obtained with the above mentioned electrodes can be directly applied to the real cables.

The present study, a continued work to understand the electrical behaviors in medium voltage power cables, was aimed to investigate the effects of contact on the electrical conduction characteristics in low density polyethylene using such contacts as sputtered Al, semiconductive compound, and silicone oil.

EXPERIMENTAL

The experimental apparatus and procedures employed in the present study are the same as those reported elsewhere.[6] It basically consists of a dc power supply, electrometer, and electrodes. For the present study, the measurement system was modified, the major one being that a general purpose interface board(GPIB)-controlled data acquisition system was used to collect the charging currents continuously right after the application of dc voltages. LDPE film under test is placed between two aluminum blocks and the steady state charging currents were obtained at 40 minutes after the application of dc voltages.[6] From measured conduction currents, changes in such conduction behaviors as conduction mechanisms, effective charge mobility and activation energy due to the type of contact were analyzed. All measurements were made at temperatures ranging from 20 to 90℃

Contacts were prepared as follows : Sputtered Al contacts were prepared by sputtering about 500 Å thick Al layer on both surfaces of LDPE film. Semiconductive compound produced by Dong Seon Material Co. Ltd. was compression molded to about 40 μm thick films and physically attached to both surfaces of LDPE film. This compound is the same material as the one which is being used as a strand shield of medium voltage power cables. For silicone-oil contacts, a drop of silicone oil was simply pasted on both sides of LDPE film and the thickness of silicone-oil layer was controlled by adjusting the tension of spring-tightening device. A thin silicone-oil layer was obtained from the high tension of the spring-tightening device and the thick one from the slight tension. The exact thicknesses of silicone-oil layers are not known at this moment.

RESULTS AND DISCUSSION

Current Densities vs. Electric Field

Typical J-E(current density-electric field) plots for the LDPE with a thick silicone-oil contact at 70 and 90℃ are shown in Fig. 1, whereas that with a thin silicone-oil layer at 70℃ is also shown for the comparison. As expected, the current density increases with the field and temperature. Three regions, Region I, II and III, are clearly seen from the J-E curves with a thick silicone-oil layer at 70 and 90℃, the average slopes being 2.03, 1.30, and 2.15, respectively. With the assistance of literature information,[8~10] Regions I and III can be assigned as the space charge limited conduction(SCLC) mechanism. In Region II, however, rates of change of conduction currents(current density in this case) were considerably suppressed at a wider range of dc fields compared to that with the thin silicone-oil layer. Such suppression seems to appear at a wider range of electric fields at higher temperatures. In addition, a d^{-3} dependence of current density to the film thickness holds true for Regions I, II and III, which suggests that the SCLC is the major conduction mechanism for these regions.[6]

Electrical conduction characteristics of LDPE with a semiconductive contact were evaluated at the temperature range from 20 to 90℃, the results of which are shown in Fig. 2. It was observed that

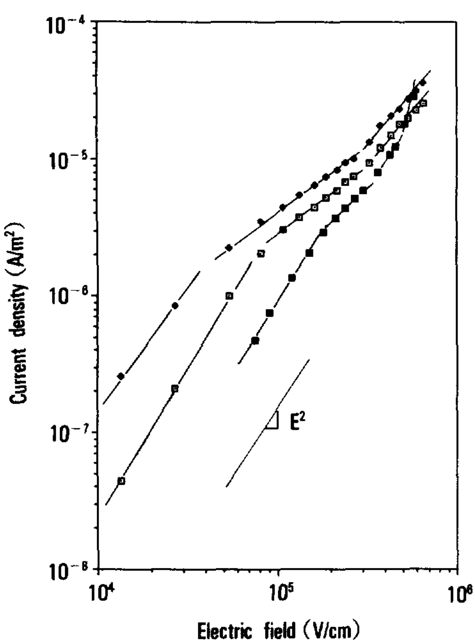

Fig. 1. J-E characteristics of LDPE with the silicone-oil contacts : Thin oil layer(■), Thick oil layer at 70℃(⊡), Thick silicone-oil layer at 90℃(◆).

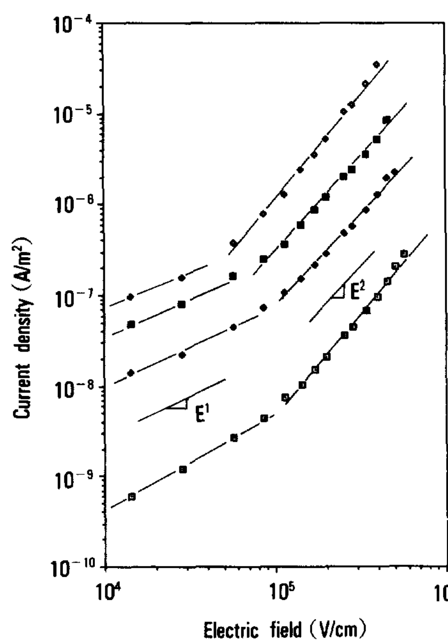

Fig. 2. J-E characteristics of LDPE with semiconductive contact : 20℃(⊡), 50℃(◆), 70℃(■), 90℃(◇).

the slope in a current density vs. electric field plot changes from about 1.0 to about 2.0 at all temperatures, which indicates that the conduction mechanisms change from the ohmic to the SCLC as the voltage increases.[6,8~10] Interestingly, however, no marked suppression in a rate of change of current density was observed when the semiconductive film was used as a contact.

Current densities at 70℃ with different contacts were compared in Fig. 3. The results at other temperatures such as 20, 50 and 90℃ showed a similar trend to that at 70℃. From the figure it can be clearly seen that the magnitude of current density changes due to the nature of contact. Sputtered Al shows the lowest current density, the silicone-oil the highest and the semiconductive film the intermediate. The shapes of J-E curve for both sputtered Al and semiconductive contacts are very much the same as each other. In this case, the conduction mechanism can be represented as the ohmic,

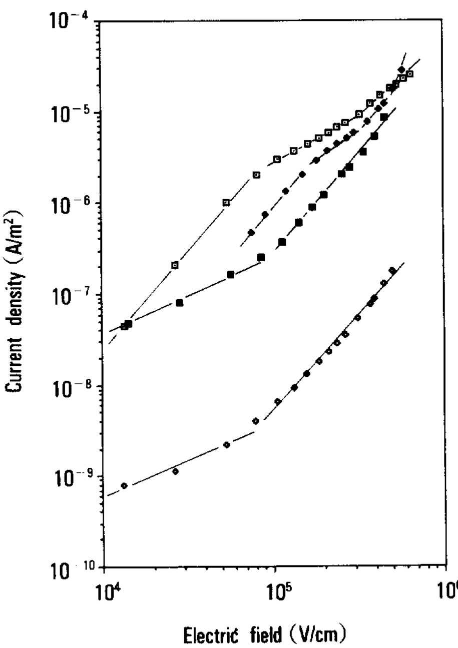

Fig. 3. Effect of various contacts on the J-E characteristics of LDPE at 70℃ : Sputtered Al contact(◇), Semiconductive contact(■), Thin silicone-oil contact (◆), Thick silicone-oil contact(⊡).

SCLC and Fowler-Nordheim conductions in order of increasing electric field. As mentioned earlier, no marked suppression in a rate of change of current density was observed with both sputtered Al and semiconductive contacts. On the other hand, the conduction behaviors in case of silicone-oil contacts are quite different. The major difference is that the rate of change of current density is considerably suppressed at a certain voltage range. Besides, for both sputtered Al and semiconductive contacts the transition from the ohmic to the SCLC occurs at about 80 kV/cm, whereas for both silicone-oil contacts only SCLC is observed at comparable electric fields. In the latter case, the transition from the ohmic to SCLC seems to occur below 10 kV/cm.

Activation Energy

Activation energies for various contacts were calculated from the slopes in σ vs 1/T plots by assuming that the temperature dependence of electrical conductivity follows the Arrhenius-type equation : $\sigma=\sigma_0\exp(-\Delta E/kT)$, where σ is the electrical conductivity, σ_0 pre-exponential factor, ΔE an activation energy for the electrical conduction, k a Boltzmann constant, and T an absolute temperature.

Calculated activation energies are plotted against the applied field for various contacts in Fig. 4. In the figure, the one pronounced feature is that the magnitude of activation energy depends on the type of contact. Sputtered Al shows the highest, semiconductive contacts the intermediate, and silicone-oil contacts the lowest activation energy. In the case of sputtered Al, the activation energy was estimated to be about 0.80 eV, which agrees well with the published value.[7] For silicone-oil contacts the thin layer shows a little higher activation energy than the thick one. It was also found that for most contacts the activation energy increases at low fields and then decreases at high fields, the rate of its change being dependent on the type of contact. For example, no or slight decrease in activation energies as a function of voltage is observed in both sputtered Al and semiconductive contacts

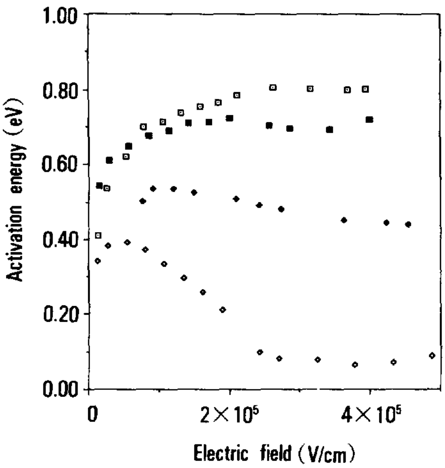

Fig. 4. Activation energy for the electrical conduction at various electrode contacts : Sputtered Al contact (⊡), Semiconductive contact(■), Thin silicone-oil contact(◆), Thick silicone-oil contact(◇).

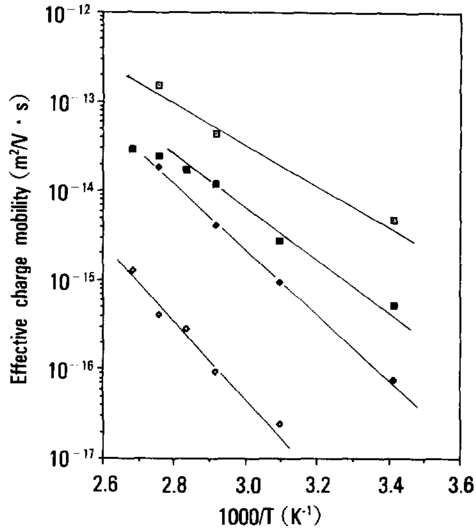

Fig. 5. Effective charge mobility of LDPE at various electrode contacts : Sputtered Al contact(◇), Semiconductive contact(◆), Thin silicone-oil contact(■), Thick silicone-oil contact(⊡).

while a large decrease in both silicone-oil contacts.

Effective Charge Mobility

Effective charge mobility was estimated from current densities exhibiting the SCLC mechanism using the following Child equation : $J=(9\varepsilon\varepsilon_0\mu_{eff}V^2/8d^3)$, where J is the current density, ε the relative permitivity, ε_0 the permitivity of free space, μ_{eff} the effective charge mobility, V the applied voltage, and d the film thickness. The value of 2.5 was used for the relative permitivity of polyethylene.[8]

The temperature dependence of effective charge mobility for various contacts is shown in Fig. 5, where the calculated effective charge mobility was plotted against 1/T. In the figure it can be found that the effective charge mobility depends on both temperature and type of contact. The effective charge mobility increases as the temperature increases and all contacts except the thin silicone-oil show a linear change of charge mobility against 1/T. In the case of thin silicone-oil contact, a rate of change of effective charge mobility decreases at high temperatures. A nonlinearity can be explained in terms of the morphological effect.[9] At high temperatures the enhanced chain motion renders the motion of charges active so that the conduction becomes easier than at lower temperatures, the net result being a decrease of a slope in a log σ vs. 1/T plot. As far as the magnitude of effective charge mobility is concerned, the sputtered Al shows the lowest ranging from 10^{-17} to 10^{-15} $m^2/V \cdot s$, the silicone-oil the highest ranging from 10^{-15}-10^{-13} $m^2/V \cdot s$, and semiconductive contacts the intermediate values ranging from 10^{-16}-10^{-14} $m^2/V \cdot s$ at all temperatures.

Discussion

It has been found throughout the present study that the type of contact alters the conduction characteristics. Semiconductive film and sputtered Al contacts showed the typical conduction characteristics similar to the ones reported.[8,9] However, the silicone-oil contacts exhibited somewhat different conduction characteristics. The major changes due to silicone-oil contacts are (1) the magnitude of conduction current densities and (2) a considerable suppression in a rate of change of current density at a certain voltage range.

It seems that the charge accumulation plays a major role in exhibiting these features. It has been reported previously that the homocharges are injected in case of evaporated metal contacts while the heterocharges are accumulated in case of silicone grease contact.[4] When the heterocharges are formed, an internal field might be distorted in such a manner that the electric field in the interfacial region becomes higher than the externally applied one, so that more charges are injected into the polyethylene.[12,13] In this situation the observed conduction current will be higher than in contacts where the homocharges are injected. Based on this concept, it can be said that the current density with silicone-oil contacts is higher than that with sputtered Al contact. The continuous accumulation of charges may keep increasing the electric field at the interfacial region. When the electric field at the interfaces becomes high enough to drive the charges toward the opposite electrode a charge sweeping effect becomes dominant so that the rate of change of current density may be suppressed.

Additional changes due to silicone-oil contacts are that (1) the activation energy for conduction decreases considerably, (2) the effective charge mobility increases, and (3) the electric field where the transition from the ohmic to the SCLC occurs is altered. Note that the transition occurs below 10 kV/cm with silicone-oil contacts and at about 80 kV/cm with both semiconductive and sputtered Al contacts.

All these features seem to result from the interfacial polarization at the additional less resistive oil layer. It seems that the heterocharge accumulated at this layer helps the accumulation of more charges by increasing the local electric field. Therefore, the activation energy for the electrical conduction decreases and the charge mobility increases. Also, due to the enhanced charge injection, the SCLC occurs at lower electric fields.

A great attention may have to be paid to the interpretation of the results obtained with silicone-oil contacts. The present study combined with our previous report[6] indicates that the most striking change due to the silicone-oil layer is the magnitude of activation energy and charge mobility. For a thick silicone-oil layer, the observed activation energy was below 0.1 eV and the charge mobility increases by an order of about 2. All these changes were attributed to the interfacial polarization enhanced by the heterocharge accumulation. This also indicates that the insulating oil used for a flash-over proof and corona-proof experimental set-up may change the conduction characteristics as well as charge accumulation characteristics.

SUMMARY

Effects of contact materials on the electrical conduction characteristics in LDPE were investigated. Such contacts as silicone-oil, sputtered Al, and semiconductive film were used in the present study. It has been observed that the type of contact modifies the electrical conduction behavior of LDPE. The major features are as follows :

(1) Magnitudes of current densities, activation energies and effective charge mobilities are different depending on the type of contacts. Silicone-oil contacts show the highest current density, lowest activation energy, and highest charge mobility, whereas sputtered Al contact the lowest magnitude of current density, highest activation energy, and lowest charge mobility. On the other hand, the semiconductive contact shows intermediate values for all three quantities.

(2) A considerable suppression in the rate of change of current density at a certain voltage range is observed in case of silicone-oil contacts, whereas no such suppression is observed in both semiconductive and sputtered Al contacts.

All observed changes due to the silicone-oil contacts were attributed to the interfacial polarization and subsequent charge sweeping effect enhanced by the accumulation of heterocharges.

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