Space Charge Behavior in Low-Density Polyethylene

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ABSTRACT

The space charge distribution in low-density polyethylene (LDPE) was measured with the pulsed electroacoustic (PEA) method. We used three types of LDPE: LDPE-L and LDPE-H were prepared by the high pressure process, and m-LDPE was polymerized with a metallocene catalyst. Space charge in LDPE strongly depended upon the electrode material. Semiconductive electrodes enhanced carrier injection into LDPE and, as a result, space charge. The density, polymerization process, applied field, temperature and so on also affected the space charge behavior. This space charge behavior was compared with the results of dc current measurements.

1 INTRODUCTION

RECENTLY, much attention has been paid to HV dc (HVDC) extruded polyethylene (PE) cable. A long-distance ac 500 kV cross-linked polyethylene (XLPE) cable power transmission line soon will be in practical use in Japan, while the development of HVDC extruded polyethylene (PE) cables is still an important target [1]. There are several differences between ac and dc insulation. In dc insulation, the electric field depends strongly upon the space charge and the conductivity of the insulating material. For example, space charge of the same polarity as the one of the closest electrode (homo space charge) reduces the field near the electrode, while space charge of opposite polarity (hetero space charge) enhances it. The breakdown strength thus depends upon space charge [2]. Moreover, in a dc power transmission line, the polarity of the dc voltage often is reversed in order to change the flow of electric power in the line. A breakdown has been caused by the voltage reversal in the case of homo space charge. Therefore, a better understanding of space charge behavior in insulating materials is very important for the successful design of HVDC cables.

However, the space charge behavior in insulating polymers, such as low-density polyethylene (LDPE) and XLPE, has not been well understood yet despite many published papers. There are two main difficulties. One is that space charge in an insulating polymer is very sensitive to various factors such as the physical and chemical structures of the insulating polymer, additives, electrode conditions, aging, applied field, temperature and so on [3, 4]. The other is related to the difficulty in measuring the space charge in an insulating solid directly and non-destructively. The development of non-destructive techniques such as the pulsed electroacoustic (PEA) method and the laser induced pressure propagation (LIPP) method has removed the latter difficulty [4, 5].

In this paper, we measure the space charge behavior in various LDPE with the PEA method and discuss its results quantitatively. These space charge results are compared also to the results of dc current measurements.

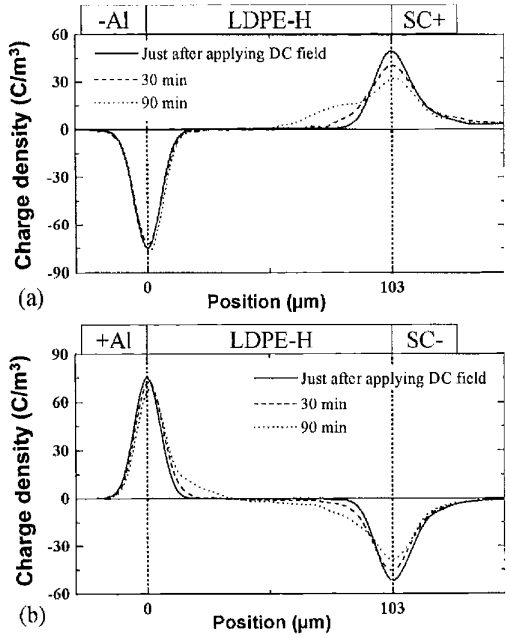

Figure 1. Space charge distributions in LDPE-H for (a) positive polarity and (b) negative polarity (applied field 50 MV/m, 23℃).

2 EXPERIMENTAL

We used three types of LDPE: LDPE-H, LDPE-L and m-LDPE. LDPE-H and LDPE-L were polymerized by high pressure process. The densities of LDPE-H and LDPE-L are 0.9255 and 0.9185 g/cm^3, respectively; thus LDPE-H has a higher density than LDPE-L. m-LDPE was polymer-

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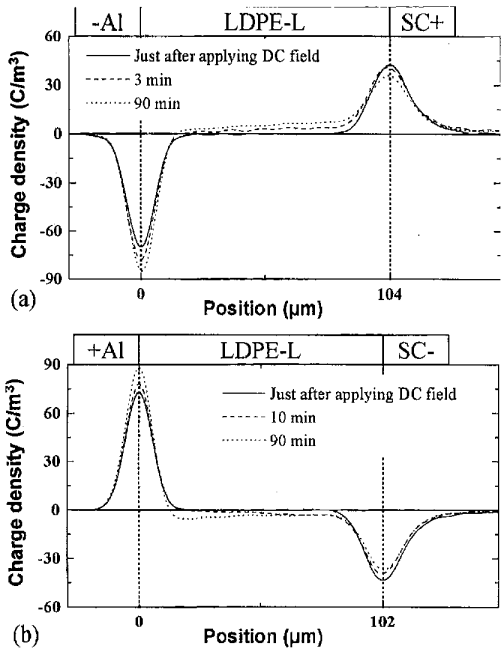

Figure 2. Space charge distributions in LDPE-L for (a) positive polarity and (b) negative polarity (applied field 50 MV/m, 23℃).

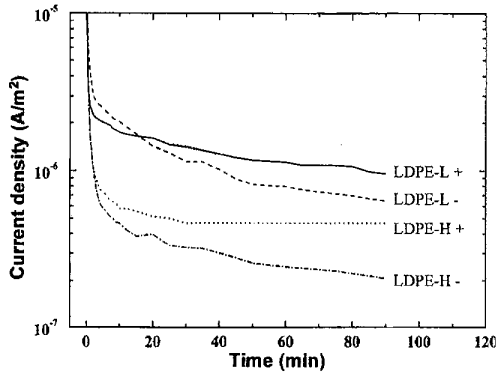

Figure 3. Current density *vs.* time characteristics (applied field: 50 MV/m, 23℃).

Table 1. Properties of LDPE films.

Sample	Process	ρ g/cm^3	T_m ℃
LDPE-L	high	0.9185	109.4
LDPE-H	pressure	0.9255	116.8
m-LDPE	metallocene	0.9227	121.4

ized with metallocene catalyst. The properties of the films are shown in Table 1. These LDPE films have no additives and their thickness is $\sim$100 μm.

We measured space charge distribution and dc current in an LDPE specimen which was set between a semiconductive electrode and an Al electrode. The space charge distribution was measured with the PEA method [4]. In order to obtain a space charge profile, a deconvolution procedure was applied to the measured signal [6]. Positive and negative dc voltages were applied to the semiconductive electrode, and the

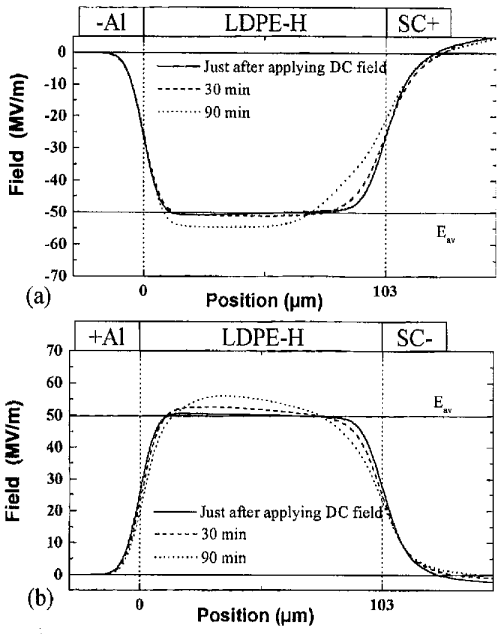

Figure 4. Field distribution in LDPE-H for (a) positive polarity and (b) negative polarity (applied field 50 MV/m, 23℃). Calculated from Figure 1.

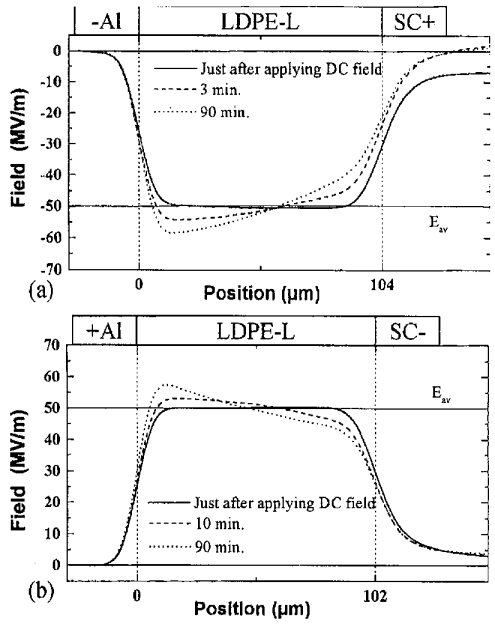

Figure 5. Field distribution in LDPE-L for (a)positive polarity and (b) negative polarity (applied field 50 MV/m, 23℃). Calculated from Figure 2.

Al electrode was connected to ground.

In this paper, 'positive polarity' means that positive voltage was applied to the semiconductive electrode and 'negative polarity' means that negative voltage was applied to the semiconductive electrode. Space charge was measured every minute under the application of a dc field and also after short-circuiting. The dc current was measured with the same electrode system as the space charge measurement.

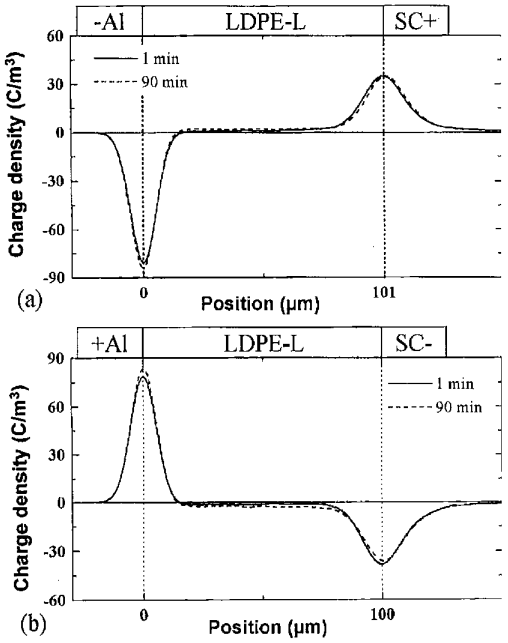

Figure 6. Space charge distribution in LDPE-L for (a) positive polarity and (b) negative polarity (applied field 50 MV/m, 40℃).

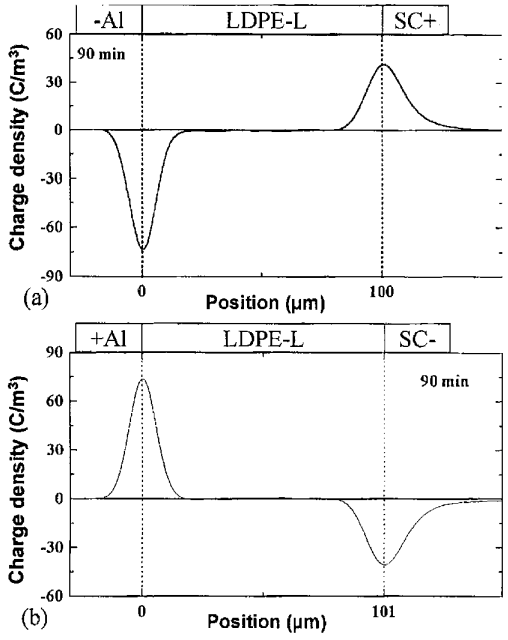

Figure 7. Space charge distribution in LDPE-L for (a) positive polarity and (b) negative polarity (applied field 50 MV/m, 60℃).

3 RESULTS AND DISCUSSION

3.1 SPACE CHARGE AND DC CURRENT IN LDPE-H AND LDPE-L

Figure 1 shows the space charge distributions in LDPE-H under the application of a dc field of 50 MV/m at 23℃. For the positive polarity (Figure 1(a)), positive carriers are injected from the semiconductive electrode to form a space charge and they stay near the semiconductive electrode even after 90 min. For the negative polarity (Figure 1(b)), negative carriers are injected from the semiconductive electrode and positive carriers are injected from the Al electrode.

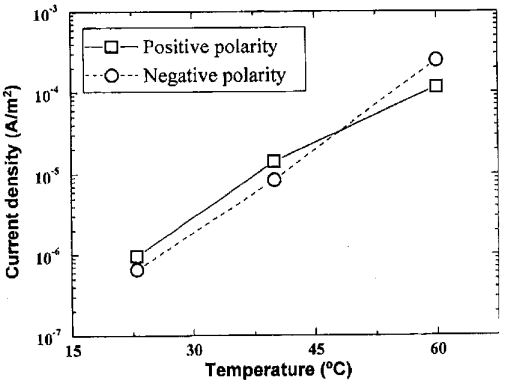

Figure 8. Temperature dependence of dc current (90 min) in LDPE-L under the application of a dc field of 50 MV/m.

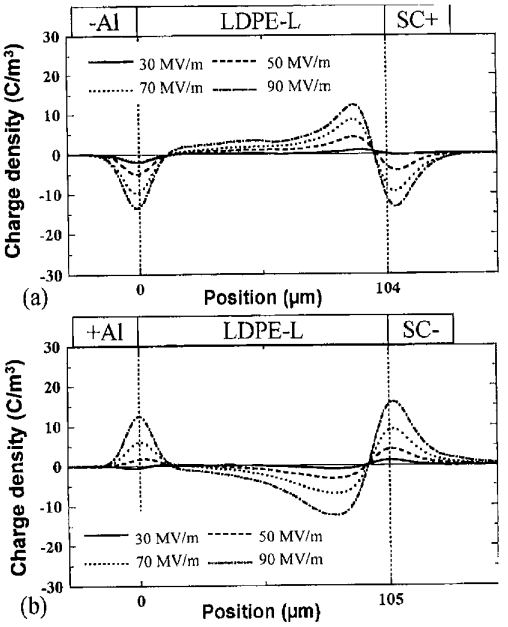

Figure 9. Space charge distribution in LDPE-L. Each dc field was applied for 10 min (23℃) and then space charge was measured just after short-circuiting.

Figure 2 shows the space charge distributions in LDPE-L. For positive polarity (Figure 2(a)), positive carriers are injected from the semiconductive electrode and the front of positive space charge arrives at the counter Al electrode in ~3 min as shown in Figure 2(a). For negative polarity (Figure 2(b)), negative carriers are injected from the semiconductive electrode and the front of negative space charge arrives at the counter Al electrode in ~10 min. These results suggest that both positive and negative carriers are injected more easily from the semiconductive electrode than the Al electrode, and that positive carriers are more mobile than negative ones. Moreover, the migration of space charge is

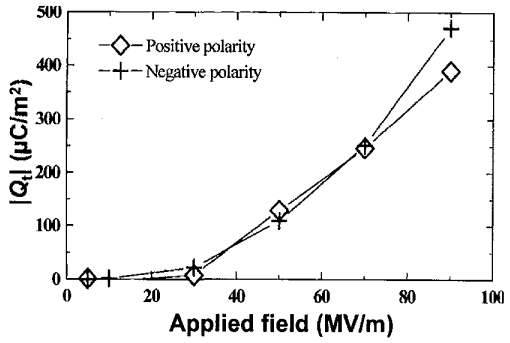

Figure 10. Total amount of space charge $|Q_t|$ in the specimen, calculated from Figure 9.

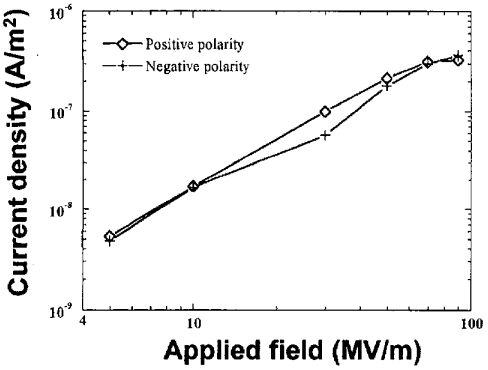

Figure 11. Current density measured 10 min after the application of dc field (23℃).

much faster in LDPE-L than in LDPE-H.

Figure 3 shows the dc currents in LDPE-L and LDPE-H under a field of 50 MV/m at 23℃. In both samples, the dc current for positive polarity is larger than that for negative polarity. It is not inconsistent with the fact that positive carriers migrate faster than negative ones, as shown in Figures 1 and 2. LDPE-L shows a higher current than LDPE-H for both polarities. This is not inconsistent with the result of the space charge experiment, because the space charge front reaches the Al counter electrode more quickly in LDPE-L than in LDPE-H.

It is interesting that LDPE-L having a lower density or a higher amorphous fraction shows a higher charge mobility or a higher current density than LDPE-H. Both LDPE-L and LDPE-H are polymerized by the high pressure process and they are nominally free from additives. Such differences might be caused by a difference in density or degree of crystallinity. However, the current density depends upon the product of charge density and mobility, both of which are sensitive to a small amount of impurities. For further discussion, we will need to have more information.

3.2 FIELD DISTRIBUTIONS IN LDPE-H AND LDPE-L

Using Poisson's equation, we can obtain the field distribution from the space charge distribution, neglecting the piezoelectric effect. Figure 4 shows the field distribution in LDPE-H, calculated from Figure 1. One can see the field reduction near the semiconductive electrode and

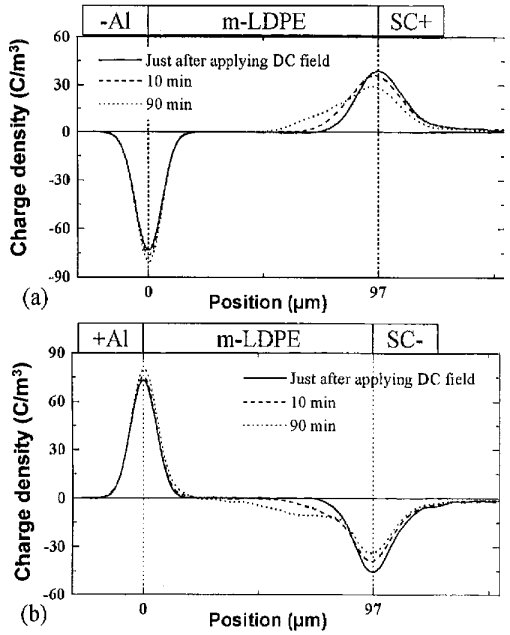

Figure 12. Space charge distribution in m-LDPE for (a) positive polarity and (b) negative polarity, under a dc field of 50 MV/m (23℃).

the field enhancement near the Al electrode, since homo space charge is formed near the semiconductive electrode.

Figure 5 shows the field distributions in LDPE-L calculated from Figure 2. For both polarities, the field is reduced near the semiconductive electrode and enhanced near the Al electrode. In this case, the field reduction near the semiconductive electrode and the field enhancement near the Al electrode are ~15% of the average applied field (E_{av} = 50 MV/m) 90 min after the application of the dc field.

3.3 TEMPERATURE DEPENDENCE OF SPACE CHARGE AND dc CURRENT

Figures 6 and 7 show the space charge distribution in LDPE-L at 40℃ and 60℃, respectively. For both polarities, space charge injected from the semiconductive electrode is spread over the sample within a minute at 40℃. One can see the space charge front arriving at the counter Al electrode in ~ 1 min in Figure 6. The amount of space charge decreases with increasing temperature, and space charge is not observed for either polarity at 60℃.

Figure 8 shows the temperature dependence of dc current in LDPE-L. The values of current are those recorded 90 min after the application of dc field (50 MV/m). The dc current increases with temperature for both polarities, while the amount of space charge decreases with temperature as mentioned above. These results suggest that charge carriers from the semiconductive electrode flow out through LDPE-L without contributing to space charge formation at 60℃.

3.4 FIELD DEPENDENCE IN LDPE-L

To investigate the field dependence of space charge and dc current, dc fields of 5, 10, 30, 50, 70 and 90 MV/m were applied for 10 min

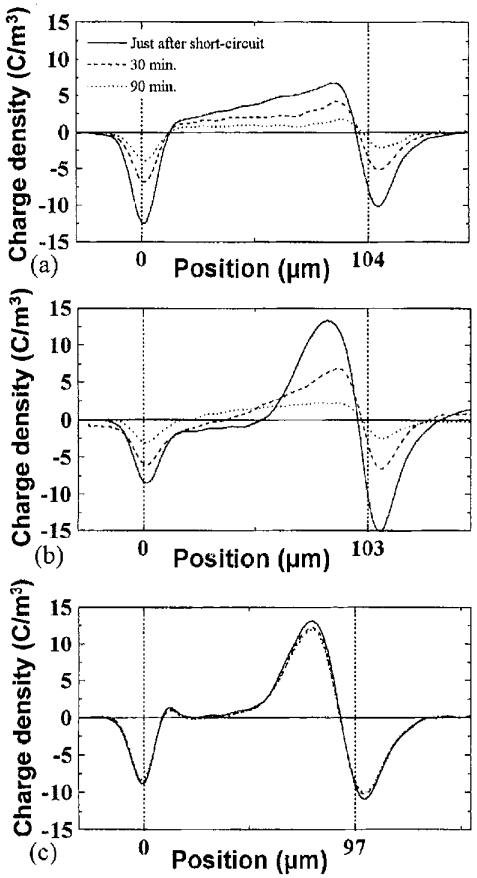

Figure 13. Space charge distribution in (a) LDPE-L, (b) LDPE-H and (c) m-LDPE after short-circuiting. A dc field of 50 MV/m was applied to the specimen for 90 min (23℃) and space charge was measured after short-circuiting.

and the specimen was short-circuited for 10 min after the application of each field. Figure 9 shows the space charge distribution in LDPE-L as a function of dc field. Each dc field was applied for 10 min at 23℃ and the space charge distribution was measured just after short-circuiting. Figure 10 shows the total amount of space charge in the specimen calculated from Figure 9. Space charge started to increase at ~ 30 MV/m for both polarities. This field is the threshold of carrier injection from the semiconductive electrode. The amount of space charge increases with applied field and there is no significant difference between positive and negative polarities.

Figure 11 shows dc current (23℃) in LDPE-L 10 min after the application of dc field. The current tends to saturate with increasing applied field. This may be associated with the suppression of electrode field due to homo space charge formed near the semiconductive electrode.

3.5 SPACE CHARGE IN m-LDPE

Figure 12 shows the space charge distribution (23℃) in m-LDPE at a field of 50 MV/m. For both polarities, charge carriers are injected from the semiconductive electrode like in LDPE-H and LDPE-L. Space charge stays near the semiconductive electrode even 90℃ after the dc field application.

Figure 13 shows the space charge distributions in m-LDPE after short-circuiting at 23℃ (positive polarity), together with those for LDPE-L and LDPE-H. The dc field of 50 MV/m was applied to the specimen for 90 min and then the specimen was short-circuited. Space charge was measured 0, 30 and 90 min after the short-circuit. The space charge is very stable in m-LDPE, whereas in LDPE-H and LDPE-L it decreases rapidly with time and its amount at 90 min drops to ~ 30% of the initial value (0 min).

The m-LDPE, polymerized with metallocene catalyst, is a new material and it has been reported to have narrower structure and molecular weight distributions than LDPE (LDPE-H and LDPE-L) polymerized by the high-pressure process [7]. Furthermore, the molecular structure of m-LDPE is more controllable. Ishimatsu *et al.* have reported that m-LDPE has a high breakdown strength [8]. The application of m-LDPE to insulating material for electric power cables has attracted much attention. However, it is not so clear how much the difference in morphology between m-LDPE and LDPE affects the electrical properties such as the space charge. Further research work in the field is required.

4 CONCLUSIONS

We investigated the space charge and the dc currents in several LDPE specimens, including the new m-LDPE. The main conclusions obtained are

(1) The space charge distribution in LDPE depends on the electrode material. A semiconductive electrode enhances carrier injection into LDPE.

(2) The experimental results suggest the effect of morphology or density on the space charge behavior in LDPE.

(3) The amount of space charge increases with applied field and decreases with temperature above room temperature. Carrier mobility increases with temperature.

(4) The space charge is very stable in m-LDPE.

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