

Space Charge and Field Distributions in Low-Density Polyethylene

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Abstract: The space charge distributions in LDPE (low-density polyethylene) were measured by PEA (pulsed electro-acoustic) method. We used two kinds of LDPEs: m-LDPE polymerized with metallocene catalyst and LDPE polymerized under high-pressure process. The space charge in these two samples strongly depended upon the material of electrode. Semicon electrode enhanced carrier injection into LDPEs. The measurements of space charge dynamics revealed the effective mobility of positive and negative carriers. The space charge was much more stable in m-LDPE than in LDPE.

1 Introduction

Low-density polyethylene (LDPE) has been widely used as an insulator in electric power cables because of its excellent electrical and mechanical properties. In 1980's, a new method to prepare LDPE using metallocene catalyst was developed. Low-density polyethylene polymerized with metallocene catalyst (m-LDPE) has narrower composition distribution and molecular weight distribution than LDPE polymerized under high-pressure process. Furthermore the molecular structure of m-LDPE is controllable. Several reports have pointed out that m-LDPE has higher breakdown strength and higher mechanical strength[1,2]. Therefore, the application of m-LDPE to insulating material for electric power cables has attracted much attention.

In this paper, the space charge distributions in m-LDPE and LDPE were compared and space charge behaviors were discussed. The space charge results were also compared with the results of conduction current.

2 Experimental

2.1 Samples

We used two kinds of LDPEs: m-LDPE polymerized with metallocene catalyst and LDPE polymerized under a high-pressure process. The properties of their films are shown in Table 1. These LDPE films have no additives and their thicknesses are about 100 μm .

Table 1 Samples

Sample	Density (gcm^{-3})	Melting point ($^{\circ}\text{C}$)
m-LDPE	0.9227	121.4
LDPE	0.9185	109.4

2.2 Space charge and current measurements

Space charge distributions were measured by PEA (Pulsed Electro-Acoustic) method [3]. We used the Al/semicon electrode system. The conduction currents of the samples were also measured under the same conditions. Positive and negative DC voltages were applied to the semicon electrode and the Al electrode was connected to the ground. In this paper, "positive polarity" means that positive voltage was applied to the semicon electrode and "negative polarity" means that negative voltage was applied to the semicon electrode.

Field dependence of space charge and current

DC fields of 5 MVm^{-1} , 10 MVm^{-1} , 30 MVm^{-1} , 50 MVm^{-1} , 70 MVm^{-1} , 90 MVm^{-1} were applied in the way shown in Fig. 1. Field applying time and short-circuiting time were 10 min., respectively. The space charge distributions were measured just after and 10 minutes after applying electric field and after short-circuiting. The conduction currents were also measured under the same procedure.

Space charge formation under a fixed field

DC field of 50 MVm^{-1} was applied for 90 min. and then short-circuited for 90 min. Space charges were measured every one minute.

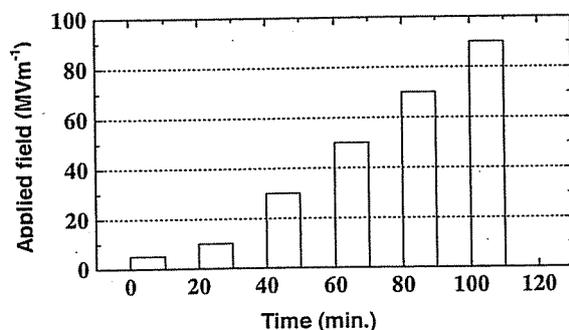


Figure 1 Process of DC field application. Field applying time and short-circuiting time are 10 min., respectively.

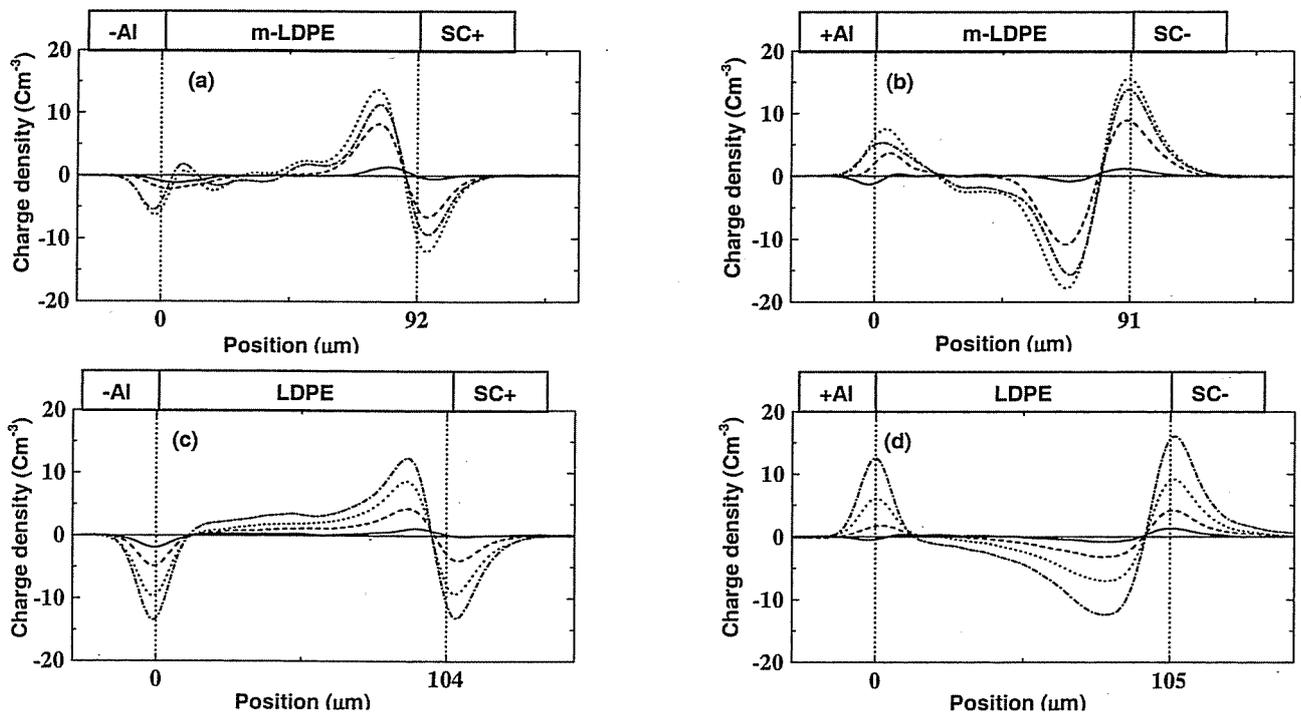


Figure 2 Space charge distributions just after short-circuiting : (a) m-LDPE (positive polarity), (b) m-LDPE (negative polarity), (c) LDPE (positive polarity), (d) LDPE (negative polarity); — 30 MVm⁻¹, 50 MVm⁻¹, 70 MVm⁻¹, -.-.- 90 MVm⁻¹

3 Results and discussion

3.1 Field dependence of space charge and conduction current

Figure 2 shows the space charge distributions just after short-circuiting in m-LDPE and LDPE, respectively. Space charges depended on the polarity of applied field. In positive polarity where positive voltage was applied to the semicon electrode, positive space charge is dominant near the anode. On the other hand, in negative polarity where negative voltage was applied to the semicon electrode, negative space charge is dominant. These results suggest that positive and negative carriers be injected from the semicon electrode. In m-LDPE, space charge is formed in the vicinity of the semicon electrode and injected carriers stay near the semicon electrode. In LDPE, on the other hand, space charge widely spreads over the sample and carriers injected from semicon electrode arrive at the opposite electrode.

We calculated the amounts of space charge formed in samples. They are plotted in Fig. 3. Space charges in both LDPE and m-LDPE start to increase around 40 MVm⁻¹ and there is not a big difference in the amount of space charge between them. However, the amount of space charge in m-LDPE shows a maximum around 70 MVm⁻¹ and then decreases at 90 MVm⁻¹, while that in LDPE increases with applied field.

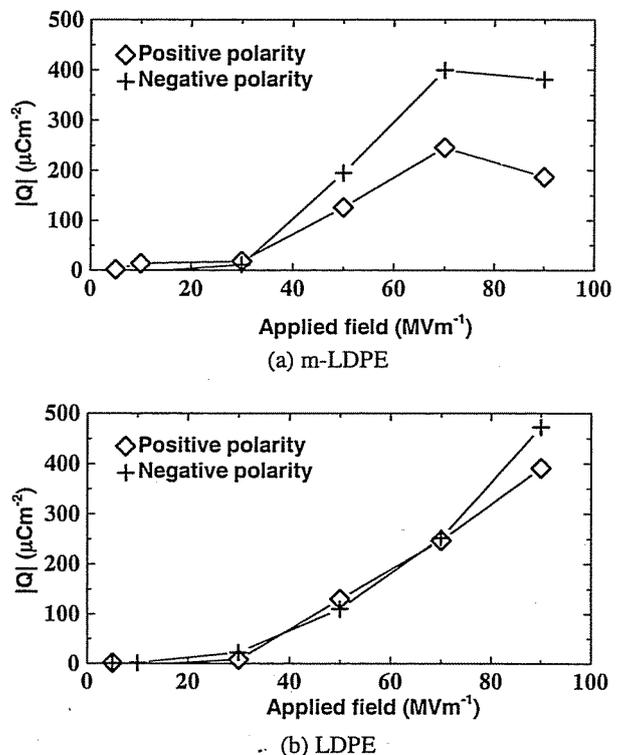


Figure 3 Accumulated space charge $|Q|$ calculated from Fig. 2.

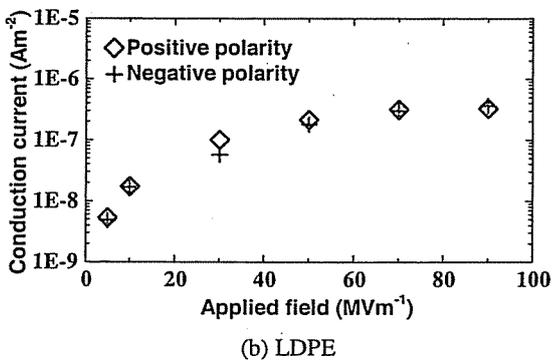
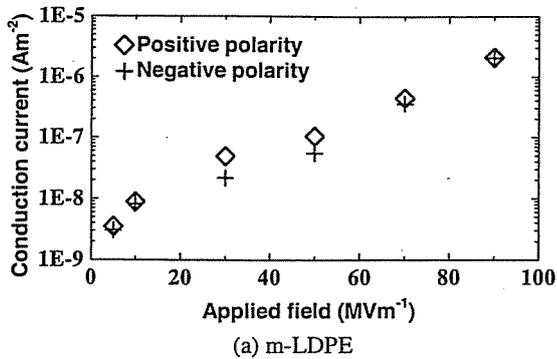


Figure 4 Current density – applied field characteristics

3.2 Conduction current

Figure 4 shows the conduction currents in m-LDPE and LDPE. In this figure, the values of currents are those 10 minutes after applying electric field. The current in LDPE tends to saturate with increasing applied electric field. This is associated with the suppression of electrode field due to homo space charge formed near the semicon electrode. On the other hand, the increasing current in m-LDPE is consistent with the decrease of the space charge at 90 MVm⁻¹ for m-LDPE.

3.3 Space charge migration

The space charge distributions at the field of 50 MVm⁻¹ for 90 min. are shown in Fig. 5. In m-LDPE, space charge injected from semicon electrode arrives at the opposite Al electrode in about 20 min. for positive polarity and about 40 min. for negative polarity. Although space charge gradually migrates with time, homo space charge is located near the semicon electrode even after 90 min. In LDPE, space charge migration is faster than that in m-LDPE. Space charge front arrives at the opposite Al electrode in about 3 min. for positive polarity and about 10 min. for negative polarity. The amount of space charge increases and spreads over the sample with time.

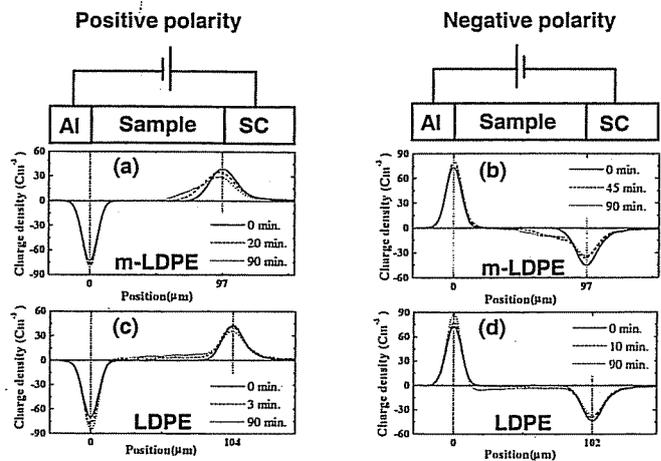


Figure 5 Space charge distributions after applying $\pm DC 50 MVm^{-1}$ at room temperature: (a) m-LDPE (positive polarity), (b) m-LDPE (negative polarity), (c) LDPE (positive polarity), (c) LDPE (negative polarity)

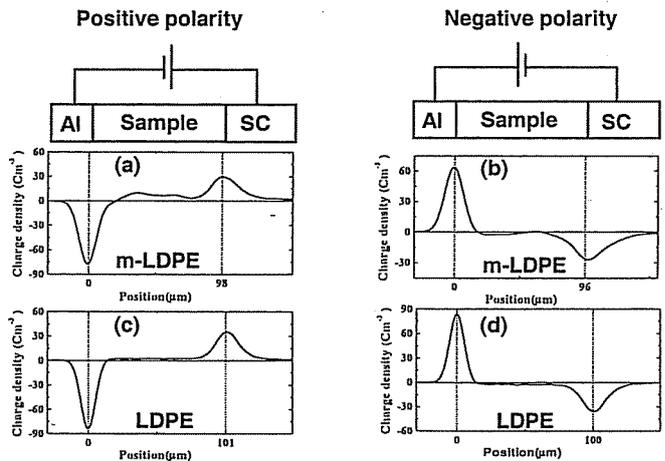


Figure 6 Space charge distributions 90 min. after applying $\pm DC 50 MVm^{-1}$ at 40°C : (a) m-LDPE (positive polarity), (b) m-LDPE (negative polarity), (c) LDPE (positive polarity), (c) LDPE (negative polarity)

Figure 6 shows the space charge distributions 90 min. after applying DC 50 MVm⁻¹ at 40 °C. At 40 °C, the polarity of space charge was also the same polarity of the semicon electrode. But the space charge migration is much faster than that at room temperature. Space charge spreaded within 1 min.

We estimated the mobility of the space charge front from Figs. 5 and 6. When space charge front migrates Δx in Δt , its mobility can be calculated from following equation.

$$\mu = \frac{\Delta x}{E \Delta t} \quad (1)$$

where E is the applied field. At 40 °C, since space charge migrates through the sample within 1 min., the mobility of

Table 2 Mobilities estimated from space charge migration

	LDPE		m-LDPE	
	μ_+ ($\text{m}^2\text{V}^{-1}\text{s}^{-1}$)	μ_- ($\text{m}^2\text{V}^{-1}\text{s}^{-1}$)	μ_+ ($\text{m}^2\text{V}^{-1}\text{s}^{-1}$)	μ_- ($\text{m}^2\text{V}^{-1}\text{s}^{-1}$)
R. T.	9.8×10^{-15}	2.2×10^{-15}	7.3×10^{-16}	5.4×10^{-16}
40°C	more than 2.7×10^{-14}			

R. T. : room temperature

μ_+ : mobility of positive charge at positive polarity

μ_- : mobility of negative charge at negative polarity

space charge is estimated at least above $2.7 \times 10^{-14} \text{ m}^2\text{V}^{-1}\text{s}^{-1}$. Estimated mobilities are listed in Table 2.

3.3 Space charge decay after short - circuiting

Figures 7 and 8 show the space charge distributions in short circuiting after applying field of 50 MVm^{-1} in 90 min. In m-LDPE, space charge is very stable even at 40 °C. On the other hand, in LDPE space charge rapidly decreases with time.

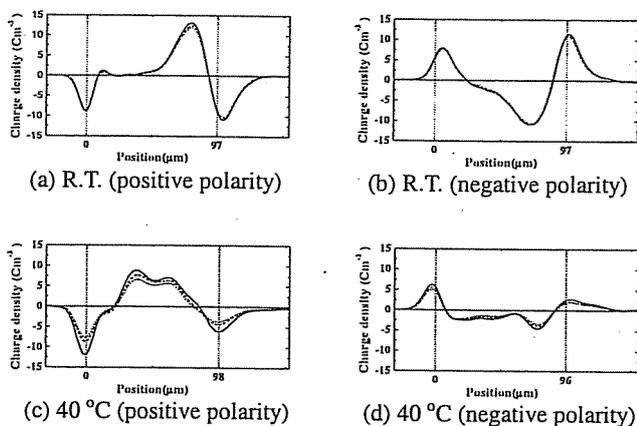


Figure 7 Space charge distributions after short-circuiting in m-LDPE: — 0 min., --- 30 min. and 90 min.

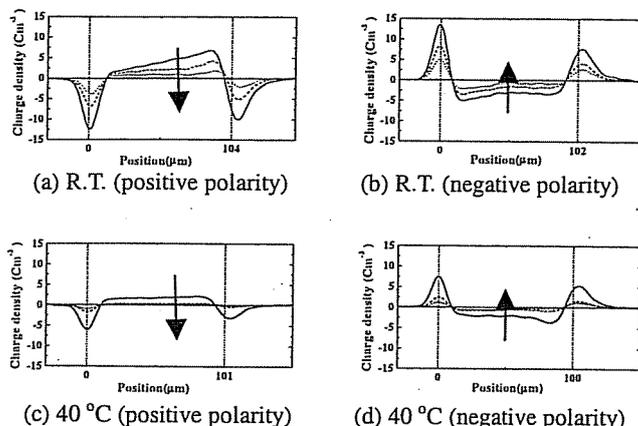


Figure 8 Space charge distributions after short-circuiting in LDPE: — 0 min., --- 30 min. and 90 min.

4 Conclusions

We investigated the space charge and conduction phenomena in low-density polyethylenes prepared by metallocene catalyst and by conventional high-pressure method. The main conclusions obtained are as follows

- (1) Space charge distributions in both LDPEs depended on the electrode materials.
- (2) The amount of space charge shows a maximum around 70 MVm^{-1} for m-LDPE, but it increases with field for LDPE.
- (3) Space charge behaviors are explained with the results of conduction current.
- (4) The mobility of space charge front is smaller in m-LDPE than in LDPE
- (5) Space charge after short-circuiting is much more stable in m-LDPE than in LDPE.

References

- [1]K. Ishimatsu, C. Banmongkol, T. Mori, T. Mizutani and M. Ishioka, "High-Field Electrical Properties of New LDPE Films Prepared using Metallocene Catalyst", *T. IEE Japan.*, Vol. 118-A, No. 7/8, 819-825 (1999).
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