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Space Charge in Low Density Polyethylene Prepared by Metallocene Catalyst

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Abstract: We investigated the space charge behavior in low-density polyethylene polymerized by using metallocene catalyst (m-LDPE). The space charge distributions were measured by Pulsed Electro-Acoustic (PEA) method. We used the Al/semiconductive electrode system. The specimens are four types of m-LDPE; mB is polymerized with butene, and mH-1, mH-2 and mH-3 are polymerized with hexene as comonomers. The densities of mB, mH-1, mH-2 and mH-3 are 0.9228, 0.925, 0.9283 and 0.9308 g/cm^3, respectively. We discussed the effects of comonomer, density and temperature on the space charge behavior. The experimental results revealed the strong dependence of density and temperature on space charge behavior, but the types of comonomer (butene or hexene) did not affect space charge properties so much.

Introduction

Low-density polyethylene (LDPE) has been widely used as insulating material for electric power cables because of its excellent electrical and mechanical properties. Recently, much attentions have been paid to DC power cables [1]. Space charge has been pointed out to play an important role in DC insulation.

In 1980's, a new method to prepare LDPE using metallocene catalyst was developed. m-LDPE has narrower composition distribution and molecular weight distribution than LDPE polymerized by high-pressure process. Moreover, the molecular structure of m-LDPE is controllable. One of the methods is to control the type or content of comonomers. Several reports have pointed out that m-LDPE has a higher breakdown strength and higher mechanical strength in conperison with LDPE porimerized by the high-presssure process [2,3]. Therefore, the application of m-LDPE to insulating material for electric power cables has attracted much attention.

In this papaer, we investigated the effect of the types and quantity of comonomers on the space charge behavior in m-LDPE.

Experimental

Samples

We used four kinds of m-LDPE films: mB is polymerized with butene, and mH-1, mH-2 and mH-3 are polymerized with hexene as comonomers. Here, mH-1, mH-2 and mH-3 were named by the order of density. The properties of films used are listed in Table 1. The additives are two kinds of anti-oxidants and neutralizer, which are contained 0.05 wt% each. Their thickness is about 100 μm.

Table 1: Samples

Sample	Comonomer	Density (g/cm^3)	Melting point (°C)
mB	Butene	0.9228	117.5
mH-1	Hexene	0.925	122.3
mH-2	Hexene	0.9283	123.0
mH-3	Hexene	0.9308	124.0

Space charge and current measurements

We measured space charge distributions and DC currents in LDPEs. Both measurements were carried out at room temperature (about 23 °C) and 60 °C.

As shown in Fig. 1, space charge distributions were measured with the pulsed electro-acoustic (PEA) method [4]. We used the Al/semicondumtive (SC) electrode system. A positive DC voltage was applied to the SC electrode and Al electrode was grounded. The DC field of 50 MV/m was applied to a specimen for 90 min. and then the electrodes were short-circuited for 90 min.

To measure DC current under the same conditions as the space charge measurements, we used the upper electrode unit of the PEA setup as shown in Fig. 2.

Results and discussion

Space charge distributions

Room temperature: Fig. 3 shows the space charge distributions in m-LDPEs under 50 MV/m at room temperature (23 °C). All m-LDPEs have similar space

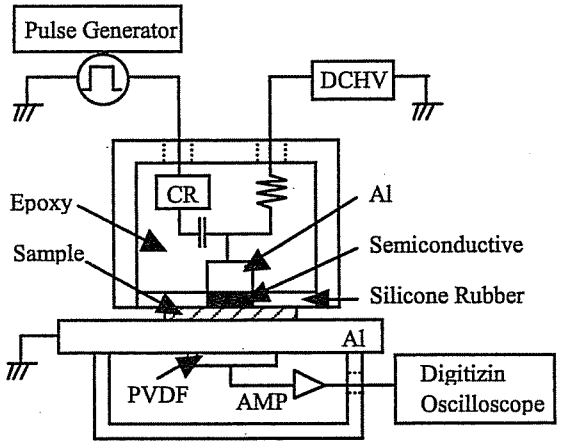

Fig. 1 Setup for space charge measurement (PEA method)

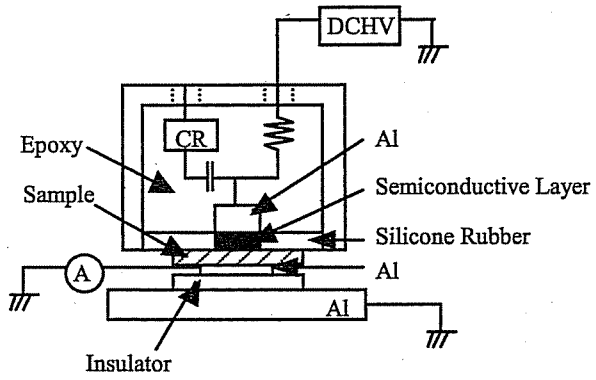

Fig. 2 Setup for DC current measurement

charge behavior at room temperature. Positive carriers seem to be injected from the SC anode and positive space charge is formed near the SC anode in mB, mH-1, mH-2 and mH-3 and negative carriers seem to be injected from the Al cathode and negative space charge is also formed near the Al cathode in mB, mH-1 and mH-2. However, negative space charge is hardly formed in mH-3.

We calculated the space charge amounts at 90 min. after applying the DC field. Fig. 4 shows the dependence of space charge amounts on film density. The space charge amounts decreases slightly with film density in m-LDPEs with hexane as comonomer. But the space charge amounts in mB is smaller than that in mH-1 in spite of its lower density.

High temperature (60 °C): Fig. 5 shows the space charge distributions in m-LDPEs under 50 MV/m at 60 °C. mB and mH-1 have similar space charge distributions as shown in Figs. 5(a) and (b). Positive carriers seem to be injected from the SC anode to form positive space charge. They arrive at the counter Al

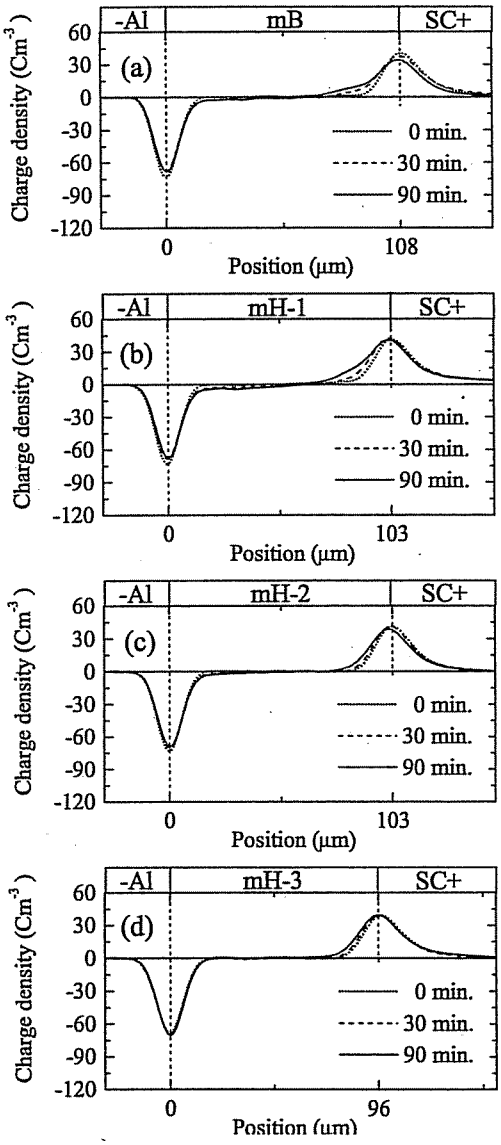

Fig. 3 Space charge distributions in mB (a), mH-1 (b), mH-2 (c) and mH-3 (d) at 23 °C (50 MV/m)

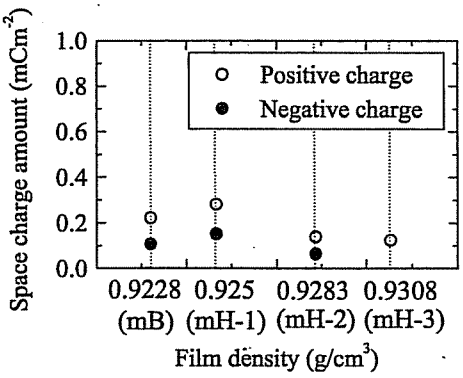

Fig. 4 Dependence of space charge amount on film density (50 MV/m , 23 °C)

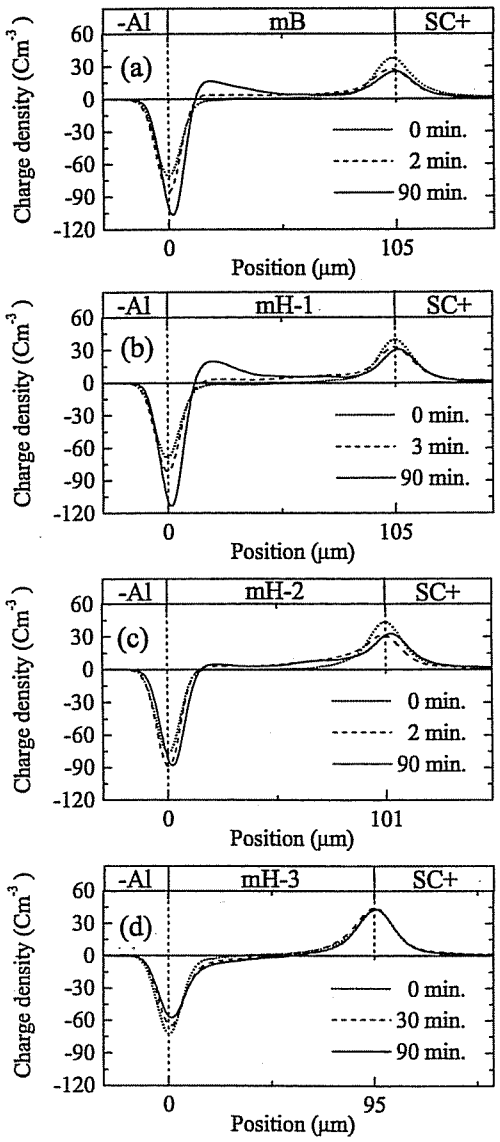

Fig. 5 Space charge distributions in mB (a), mH-1 (b), mH-2 (c) and mH-3 (d) at 60 °C (50 MV/m)

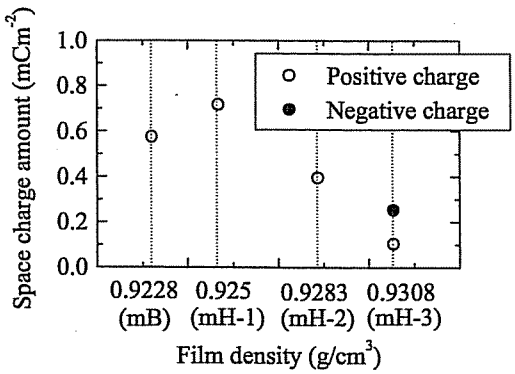

Fig.6 Dependence of space charge amount on film density (50 MV/m , 60 °C)

electrode within 2 or 3 min. after the field application. The positive space charge is accumulated throughout the bulk, especially near the Al cathode.

In mH-2 (Fig. 5(c)), positive carriers seem to be injected from the SC anode to form positive space charge. They arrive at the counter Al electrode in about 2 min. The positive space charge is accumulated throughout the bulk.

In mH-3 (Fig. 5(d)), both positive and negative carriers seem to be injected from the SC anode and the Al cathode, respectively, and homo space charges are formed.

The space charge distributions at 90 min. after the DC field application depend on film density so much. Positive hetero space charge is observed near the Al cathode in mB, mH-1 and mH-2. On the other hand, homo space charge is observed near both the SC anode and the Al cathode in mH-3. The positive hetero space charge is more remarkable in mB and mH-1 than that in mH-2. These results suggest that the migration of space charge is faster in a lower density m-LDPE [5,6].

The results of mB and mH-1 suggest that the types of comonomer (butene and hexane) don't affect the space charge distributions so much.

We calculated the space charge amounts at 90 min. after applying DC field. Fig. 6 shows the dependence of space charge amounts on film density. In mH-1, mH-2 and mH-3, the space charge amounts decreases with film density.

Temperature dependence: The results of Figs. 3 and 5 suggest that carrier transport is thermally activated. The positive space charge is staying near the SC anode at room temperature to form homo space charge, but they arrive at the Al cathode at 60 °C and form the hetero space charge in mH-1 and mH-2. The facts that the space charge amounts at room temperature is smaller than that at 60 °C as shown in Figs. 4 and 6 suggests that positive carrier injection is also thermally activated process.

DC currents

Fig. 7 shows DC charging currents at 50 MV/m. At room temperature, there is little difference in the DC charging currents among m-LDPEs. At 60 °C, the charging currents gradually increase with time except for mH-3. In mH-3, the DC charging current hardly changes. The DC charging current at room temperature is smaller than that at 60 °C in each m-LDPE.

At room temperature, the result of the DC charging currents is consistent with the result that the four m-LDPEs have similar space charge distributions. At 60 °C, the increase of DC charging currents with time may suggest a slow rise of temperature by Joule heating or

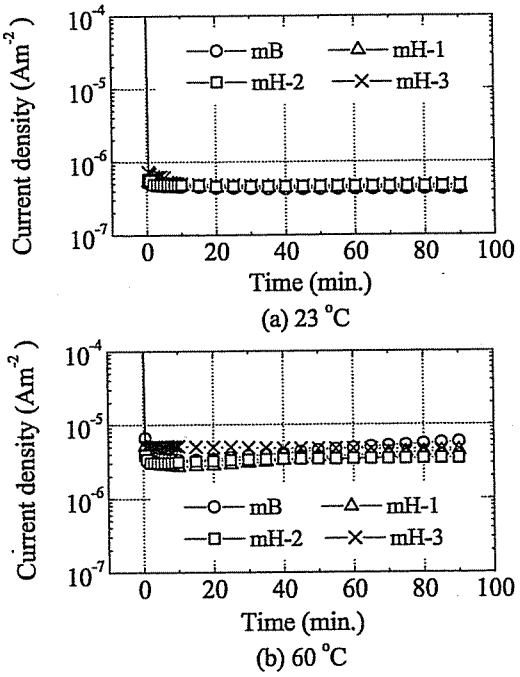

Fig. 7 DC charging current at 23 °C (a) and 60 °C (b) (50 MV/m)

the enhancement of electron injection due to the hetero space charge near the Al cathode.

Conclusion

We investigated space charge and DC charging current in various m-LDPEs. The results are summarized as follows.

(1) The space charge distributions and the space charge amounts strongly depend on film density, that is, comonomer content.
(2) The difference in space charge behavior among m-LDPEs is clear at 60 °C.
(3) The space charge amounts decreases with film density.
(4) The types of comonomer (butene and hexane) don't affect the space charge behavior in m-LDPE so much.

References

[1] F. Ruter, J. L. Parpal and S. G. Swingler, "A Review of HVDC Extruded Cable Systems", CIGRE 2000 paper P2-03, 2000.
[2] K. Ishimatsu, C. Banmongkol, T. Mori, T. Mizutani and M. Ishioka, "High field Electrical Properties of New LDPE Films Prepared using Metallecene Catalyst", *T. IEE Japan.*, Vol. 118-A, No. 7/8, pp. 819-825, 1999.
[3] Y. L. Huang and N. Brown, "Dependence of Slow Crack Growth in Polyethylene on Butyl Branch Density: Morphology and Theory", *J. Polym. Sci: Part B: Polym Phys.*, Vol. 29, pp. 129-137, 1991.
[4] T. Maeno and L. Fukunaga, "High-resolution PEA Charge Distribution Measurement System", IEEE Trans. Dielectr. Electr. Insul., Vol. 3, No. 6, pp. 754-757, 1996.
[5] T. Mizutani, H. Semi and K. Kaneko, "Space Charge Behavior in Low-Density Polyethylene", IEEE Trans. on Diel. Electr. Insul., Vol. 7, No. 4, pp. 503-508, 2000.
[6] T. Mizutani, K. Shinmura, K. Kaneko, T. Mori, M. Ishioka and T. Nagata, "Space Charge Behavior near LDPE/LDPE Interface", Annual report IEEE Conf. Electr. Insul. Dielectr. Phenomena, pp. 800-803, 2000.

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