Space charge and conduction in LDPE- polypropylene copolymer blends

Chao Zhang,[1] Teruyoshi Mizutani,[1] Kazue Kaneko,[1] Tatsuo Mori,[1] and Mitsugu Ishioka[2]
[1]Department of Electrical Engineering, Nagoya University, Nagoya 464-8603, Japan
[2]Japan Polychem Co., Kawasaki 210-0865, Japan

Abstract: In the previous paper, the electrical breakdown properties of blend polymer of LDPE and polypropylene copolymer were investigated. The impulse breakdown strength in the high temperature region (90 °C) was improved by blending. In the present paper, the space charge behavior and electrical conduction of the blends were studied with the electrode system of aluminium/sample/semiconductive layer. The specimen with 90 wt% LDPE and 10 wt% polypropylene copolymer (B10) had a larger amount of space charge than pure LDPE (PE) at 30 °C. On the other hand, the DC current of B10 was less than that of PE in this temperature region. It was found that the anode field was related to the current in both PE and B10, which suggests that the carrier injection from the anode is dominant to the electrical conduction. The DC currents of both PE and B10 showed good straight lines on the Schottky plot with the same Schottky coefficients.

Introduction

Low-density polyethylene (LDPE) has been widely used as power cable insulation. Further improvement of electrical properties of LDPE is required to develop higher-performance cables. Blend is one of ways to get higher-performance insulating polymers [1, 2]. In our previous work, the effects of blend of the random copolymer of ethylene and propylene (EP) on electrical breakdown of LDPE were investigated [2, 3]. We found that the impulse breakdown strength of LDPE in the high temperature region was improved by blending with EP. It increased with an increasing content of polypropylene copolymer. In the present study, the space charge behavior and the DC current properties of blend polymers of LDPE and EP were investigated.

It is well known that the space charge strongly affects electrical conduction and other electrical properties of insulating polymers [4, 5]. In general, the existence of space charge can distort the local electrical field, and thus affects the conduction and breakdown phenomena. Therefore, to know the correlation between space charge behavior and DC current properties of the insulating polymers may be helpful to understand the conduction process more deeply. In this study, the space charge distribution was directly observed with pulsed electro-acoustic method (PEA) [6]. The mechanism of conduction was discussed on the basis of the experimental results of both space charge behavior and current properties. The formation of space charge in blend polymer of LDPE and EP were also discussed.

Experimental

Specimens

LDPE and EP used in this study were free from any additives. LDPE was produced with the high-pressure process. EP was copolymer of ethylene and propylene with the ethylene content of 4.5 wt%. They were originally in the form of extruded pellets. The densities of pellet were 0.920 g/cm^3 and 0.898 g/cm^3 for LDPE and EP, respectively. Two kinds of specimens were measured in this study, one was pure LDPE, the other was the blend polymer of 90 wt% LDPE and 10 wt% EP. They were films with thickness of about 0.1 mm, and prepared by T-die method. They were called PE and B10, respectively.

Measurements of space charge and DC current

Space charge distribution in specimen was measured by PEA method [6]. Figure 1 shows the schematic diagram of PEA used in this study. The film specimen is placed between a semiconductive electrode (upper electrode) and an aluminium electrode (lower electrode, ground).

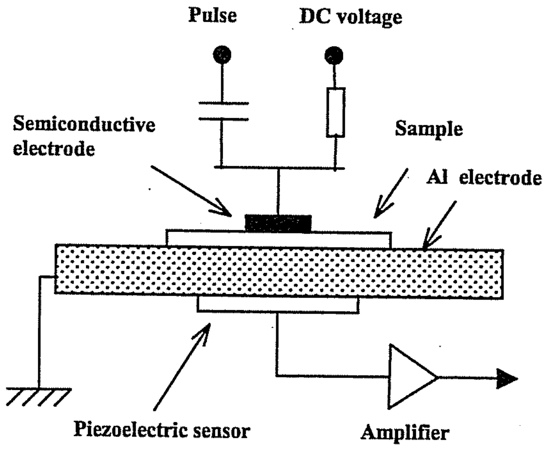

Figure 1: Schematic diagram of PEA for the measurement of space charge distribution.

0-7803-7053-8/1/$10.00 2001 IEEE

The polyvinylidenefluoride film is employed as a piezoelectric transducer to convert acoustic wave to electrical signal. In this study, a positive DC voltage was applied to the semiconductive electrode, and two experimental procedures were carried out: (1) The DC average field (E_{av}) of 50 MVm^{-1} is applied to a sample for 90 minutes, and then the electrodes are short-circuited for another 90 minutes, the change in space charge distributions and charging currents are measured; (2) One DC field is applied to a sample for 20 minutes and then another higher DC field is successively applied after the short-circuited for 60 minutes. Space charge distributions and DC currents are measured. All the measurements were carried out at 30 °C.

Results and Discussion

Space charge distribution and current under E_{av} of 50 MVm^{-1}

Typical results of space charge distribution in specimens under E_{av} of 50 MVm^{-1} at 30 °C are shown in Figure 2. Positive carriers are injected from the semiconductive electrode in PE (see Fig. 2a), and the amount of positive space charge increases with time in the first 20 min after the application of voltage, and then it tends to constant as shown in Fig. 2b. In the case of B10, positive carriers are also injected from the semiconductive electrode, and the amount of positive space charge increases with time in the first 10 min as shown in Fig. 2c. Then, the positive space charge decreases except near the aluminium electrode (see Fig. 2d). B10 has a greater amount of positive space charge than PE. This can be easily seen from Fig. 3 in which the average space charge densities were calculated from Fig. 2.

The electrical field in a specimen can be calculated by Poisson's equation with the result of space charge distribution. Figure 4 shows the time dependence of the field for PE and B10 under E_{av} of 50 MVm^{-1} at 30 °C. Both of PE and B10 have the cathode field higher than 50 MVm^{-1} and the anode field lower than 50 MVm^{-1} after the application of DC voltage. The field at the middle part is approximately equal to applied E_{av} of 50 MVm^{-1} for both PE and B10. The distortion of field near the electrode is attributed to space charge and it is about 30 % of applied field for B10.

Figure 5 shows the typical time dependence of charging current densities for PE and B10 at 30 °C. PE

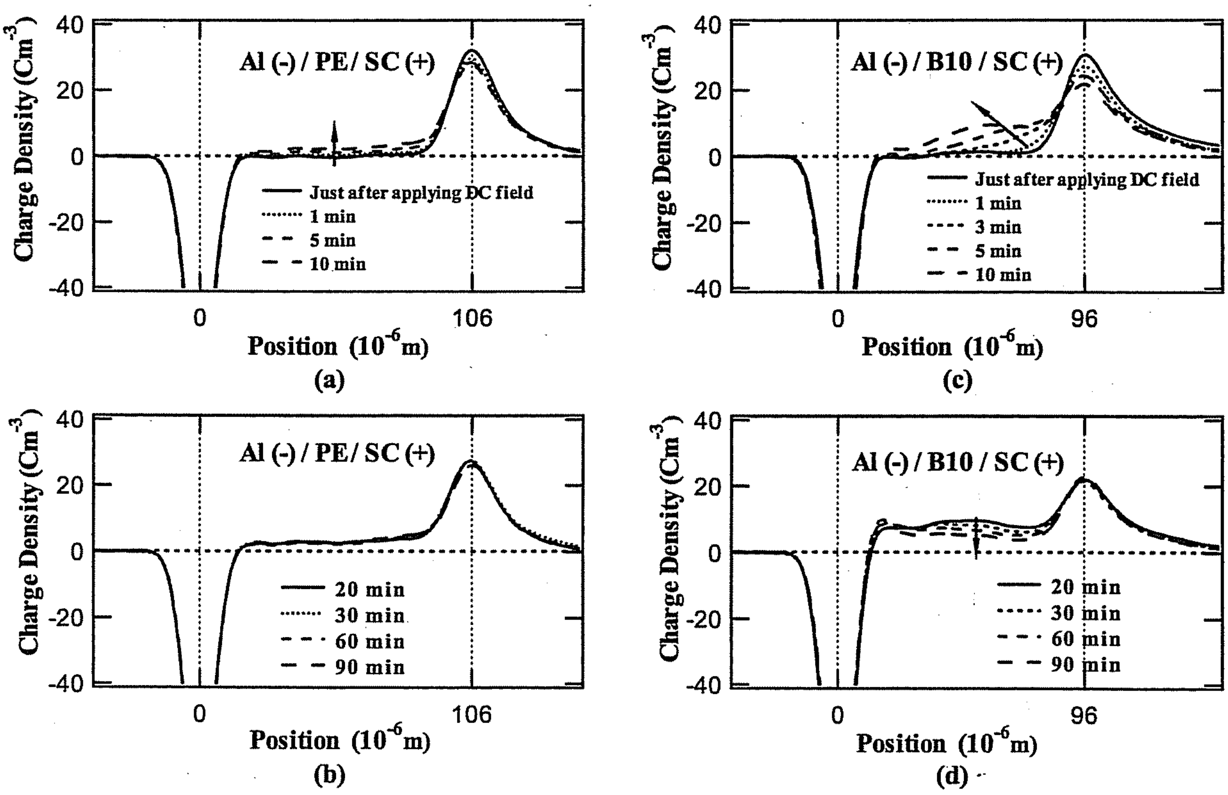

Figure 2: Space charge distribution under E_{av} of 50 MVm^{-1} at 30 °C. (a) PE (0-10 min), (b) PE (20-90 min), (c) B10 (0-10 min), (d) B10 (20-90 min).

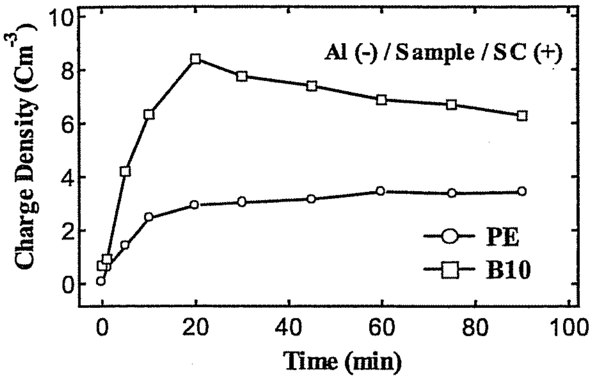

Figure 3: Average space charge density calculated from Fig. 2.

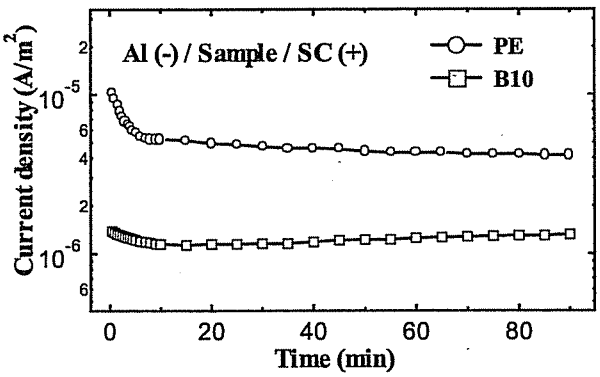

Figure 5: Time dependence of charging current under E_{av} of 50 MVm^{-1} at 30 °C.

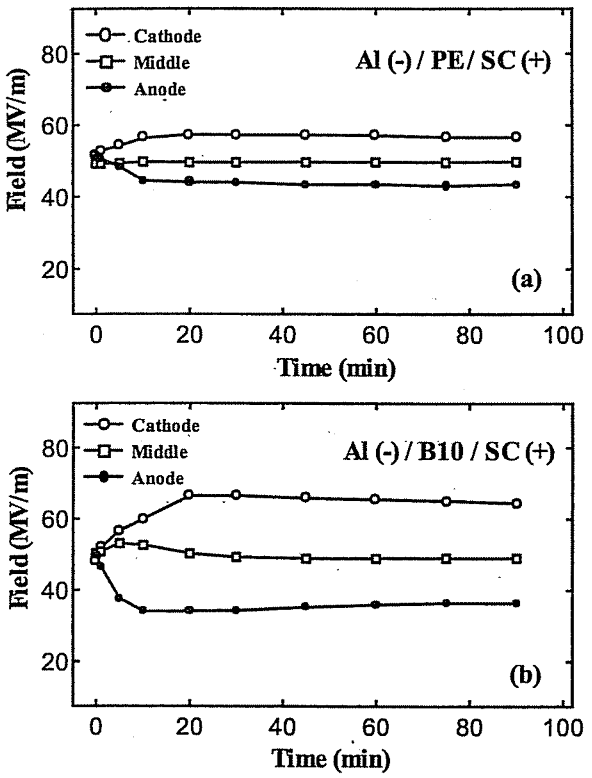

Figure 4: Field vs. time under E_{av} of 50 MVm^{-1} at 30 °C. (a) PE, (b) B10.

has a higher current than B10 under E_{av} of 50 MVm^{-1}. The current density of PE decreases with time during the voltage application while that of B10 decreases in the first 15 min, and then turns to increase slowly with time.

The anode field shows the similar tendency as the current. Both the anode field and current density of PE are higher than those of B10 while the cathode field of PE is less than that of B10. Moreover, the anode field and current of B10 increase with time from about 10-15 min as shown in Figs. 4b and 5. They suggest that the current is related to the anode field.

Figure 6 shows the logarithm of the current density (ln*J*) against the square root of the anode field ($E_a^{1/2}$) of B10 obtained from Figs. 4 and 5 (30-90 min). It shows a good straight line whose slope gives the relative permittivity of 4.0. It suggests Schottky conduction for B10.

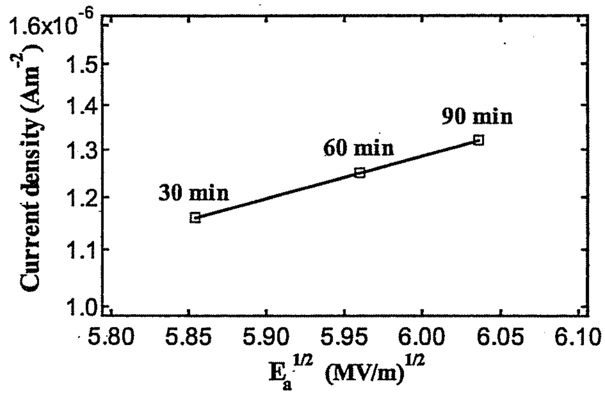

Figure 6: ln*J* against $E_a^{1/2}$ for B10 under E_{av} of 50 MVm^{-1}.

Current under different DC field

Since the changes in space charge and current are small 20 minutes after the application of DC voltage (see Fig. 2 and 5), the current and space charge were measured for 20 minutes at different DC voltages with the procedure (2) described in the preceding experimental section. The resultant currents are plotted against E_{av} in Figure 7. The current of PE is higher than that of B10 in the range lower than 70 MV/m, and the difference between them is small in the range higher than 70 MV/m.

The anode field (E_a) was calculated from the space charge profile measured at 20 minutes (under E_{av} of 20, 50, 60, 80 MVm^{-1}). The plots of ln*J* against $E_a^{1/2}$ for PE

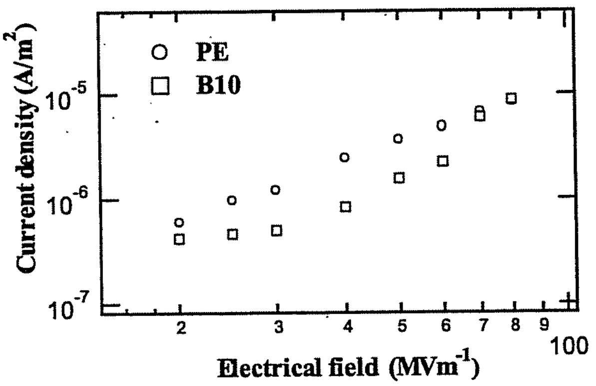

Figure 7: Field dependence of charging current.

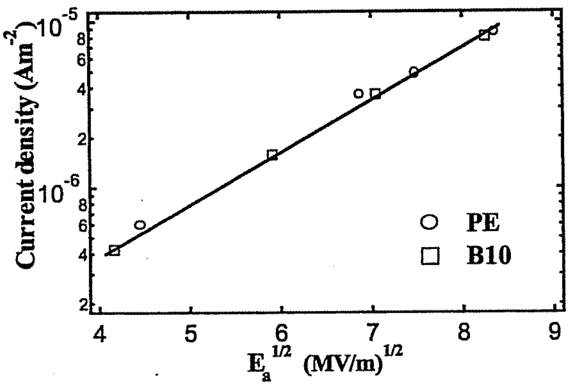

Figure 8: ln*J* against $E_a^{1/2}$ for B10 under different E_{av}.

and B10 are shown in Figure 8. The ln*J* has an approximately linear relationship with $E_a^{1/2}$ regardless of PE and B10. The slope of a straight line is 0.72 [(MV/m)$^{-1/2}$] which corresponds to the relative permittivity of 4.3. This value agrees well with the one obtained from Fig. 6. These results strongly support Schottky conduction for the DC currents in PE and B10.

Effect of blend on space charge distribution and current

According to the experimental results mentioned above, the amount of space charge in B10 is larger than that in PE. This can be explained by trap sites introduced by blending EP with LDPE. Some papers [7, 8] treat with the relationship between carrier trapping and space charge behavior although the nature of traps remain ambiguous. In blend polymer of LDPE and EP, trap sites may be located in the interface region between LDPE and EP.

The increase of trap sites by blending results in the increase of the amount of space charge, and leads to the more serious distortion of the electric field. The DC current is also affected by it.

Conclusions

We have studied the space charge behavior and DC current of blend polymers of LDPE and EP. The amount of space charge increased by blending, and thus the electrical field distribution changed. The charging current depended on the anode field, and the plot of ln*J* vs. $E_a^{1/2}$ for both PE and B10 showed a straight line, suggesting Schottky conduction.

References

[1] H. Kato, S. Simoura and M. Kawahigashi, "Morphology and Dielectric Strength of Some New Blended Polymeric Materials", Proc. 4th ICPADM, Paper4247, Brisbane, Australia, pp. 172-174, 1994.

[2] Zhang Chao, Tatsuo Mori, Teruyoshi Mizutani and Mitsugu Ishioka, "Effects of Blend of Polypropylene Copolymer on Electrical Breakdown of Low Density Polyethylene", Trans. IEE Japan, Vol. 121-A, pp. 218-224, No. 3, 2001.

[3] C. Zhang, T. Mori, T. Mizutani, M. Ishioka and Y. Chen: "Morphology and Electrical Breakdown Properties of LDPE-Polypropylene Copolymer Blends", J. Polym. Sci. Part B: Polym. Phys., Vol. 39, pp. 1741-1748, No. 15, 2001.

[4] T. Mizutani, "Space Charge Measurement Techniques and Space Charge in Polyethylene", IEEE Trans. Dielectr. Electr. Insul., Vol. 1, pp. 923-932, No. 5, 1994.

[5] Y. Suzuoki, H. Muto, T. Mizutani and M. Ieda, "Effects of Space Charge on Electrical Conduction in High-Density Polyethylene", J. Phys. D, Vol. 18, pp. 2293-2302, 1985.

[6] T. Takada, "Acoustic and Optical Methods for Measuring Eletric Charge Distributions in Dielectrics", IEEE Trans. Dielectr. Electr. Insul., Vol. 6, pp. 519-547, 1999.

[7] T. Mizutani, "Space Charge Distributions in Insulating Polymers", J. Electrostatics Japan, Vol. 19, pp. 350-357, No. 5, 1995.

[8] G. C. Montanari, "The Electrical Degradation Threshold of Polyethylene Investigated by Space Charge and Conduction Current Measurements", IEEE Trans. Dielectr. Electr. Insul., Vol. 7, pp. 309-315, No. 3, 2000.

Author address: Chao Zhang, Department of Electrical Engineering, Nagoya University, Nagoya 464-8603, Japan, E-mail: c-zhang@ieee.org.