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Space Charge Dynamics in Polypropylene

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Abstract : We investigated the space charge dynamics in polypropylene, M-PP and Z-PP which were polymerized using metallocene and Ziegler catalysts, respectively. Space charge distributions depended upon temperature, polarity and electrode materials. Carriers injected from the semiconducting electrode played an important role in the formation of space charge in PP at high temperatures. The amount of space charge was the largest around 40 °C for both M-PP and Z-PP. They showed similar space charge dynamics in spite of their difference in morphology. Z-PP showed larger current than M-PP. These results were also compared with those of polyethylene and they were discussed considering the difference in morphology among M-PP, Z-PP and polyethylene.

Introduction

Cross-linked polyethylene has been widely used as insulating material for electric power cables because of its excellent electrical and mechanical properties. The cross-linking improves the thermal property of polyethylene (PE) which has a low melting point and, as a result, it improves the performance of PE-extruded power cable. However, cross-linked polymers are not suitable for the recycle of polymeric materials. Recently, polypropylene (PP) which has a higher melting point than PE has attracted much attention as an insulating material for power cables. Especially syndiotactic polypropylene prepared by metallocene catalyst has been reported to have excellent properties compared with cross-linked PE [1]. It is also suitable for recycle because it is not cross-linked.

The space charge sometimes affects the breakdown of an insulating material because it enhances a local electric field [2]. Therefore, the performances of DC power apparatus and cables also strongly depend upon the space charge [3]. However, the space charge behavior in polypropylene is not well understood yet. In this paper, we investigated space charge dynamics in polypropylene, M-PP and Z-PP which were polymerized using metallocene and Ziegler catalysts, respectively. These results were compared with those of polyethylene.

Experimental

Specimens : We used 100 μm-thick films of M-PP and Z-PP which were polymerized using metallocene and Ziegler catalysts. They contain 3.0 - 3.5 % of ethylene which was randomly copolymerized. We also used 100 μm-thick films of two kinds of low-density polyethylene, M-PE and LDPE which were polymerized with metallocne catalyst and with a conventional high-pressure method, respectively. The properties of films used are listed in Table 1.

Table 1: Specimen

Sample	Density (g cm^{-3})	Melting point (°C)
M-PP	0.886	125
Z-PP	0.885	143
M-PE	0.9225	116.8
LDPE	0.9185	109.4

Space charge measurement: Space charge distributions in specimens were measured with the PEA method [3,4] as shown in Fig. 1. A specimen was set in

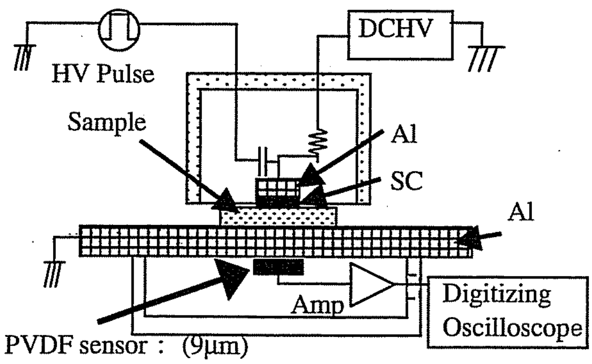


Fig. 1: Setup for space charge measurement.

the Al / Semiconducting (SC) electrode system. A positive or negative DC voltage was applied to the SC electrode and the Al electrode was grounded. In this paper, 'positive polarity' and 'negative polarity' mean that positive voltage and negative voltage were applied to the SC electrode, respectively. The DC field of 50 MVm^{-1} was usually applied to a specimen for 90 minutes and then the electrodes were short-circuited for 30 minutes.

Results and discussion

Space charge in M-PP and Z-PP : The space charge distributions in M-PP at 23, 40 and 60 °C are shown in Fig. 2. The DC field of +50 MV/m was applied to the SC electrode for 90 minutes (positive polarity) and the changes in space charge are shown in Figs. 2(a), (c) and (e). Then, the specimen was short-circuited and the resultant space charges are shown in Figs. 2(b), (d) and (f). The amount of space charge is the largest around 40 °C. Positive carriers injected from the SC electrode are moving to the counter Al electrode (see Fig. 2(c)). At 60 °C, the space charge increases with time up to about 10 minutes and then it decreases. The space charge after the short-circuit is stable at 23 °C (see Fig. 2(b)).

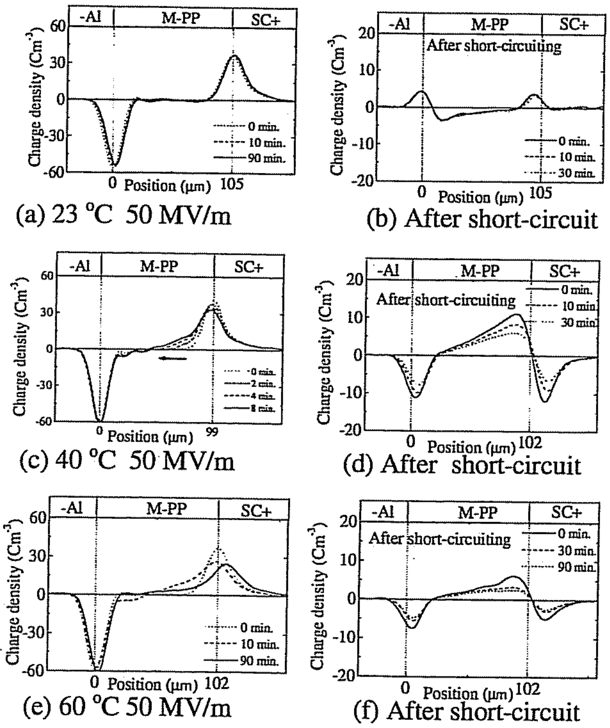


Fig. 2: Changes in space charge in M-PP at 23, 40 and 60 °C under DC field of +50MV/m for 90 minuutes and after the short-circuit for 30 minutes.

Figure 3 shows the space charge distribution in M-PP under DC field of –50 MV/m (negative polarity) at 40 °C. One can see the difference between positive polarity (Figs. 2 (c) and (d)) and negative polarity (Fig. 3). In the negative polaity, positive and negative carriers are injected from the Al and SC electrode, respectively, and they move to the counter electrode. The amount of space charge is the largest around 40 °C.

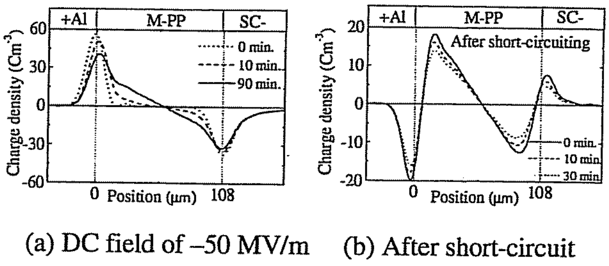


Fig. 3: Changes in space charge in M-PP at 40 °C under DC field of -50MV/m for 90 minuutes and after the short-circuit for 30 minutes.

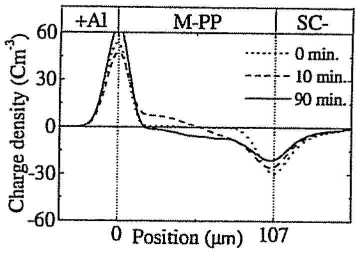


Fig. 4: Changes in space charge in M-PP at 60 °C under DC field of -50MV/m for 90 minuutes.

Figure 4 shows the space charge distribution in M-PP at 60 °C and DC field of –50 MV/m. Both positive and negative space charges are formed at the beginning (see 10 min) and then negative charge from the SC electrode becomes dominant (see 90 min).

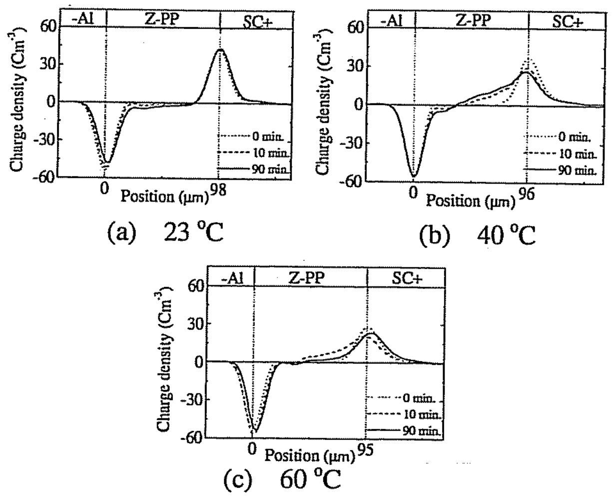


Fig. 5: Changes in space charge in Z-PP under DC field of +50 MV/m at 23, 40 and 60 °C.

Figure 5 shows the changes in space charge in Z-PP at DC field of +50 MV/m (positive polarity). From the comparison between Figs. 2 and 5, one can see the following points. (1) Z-PP and M-PP show similar behaviors of space charge. (2) Z-PP has larger space charge at 23 °C than M-PP. (3) Carriers move a little faster in Z-PP than in M-PP.

In order to check the difference in space charge between M-PP and Z-PP, the amounts of space charge after the application of DC field of 50 MV/m for 90 minutes are plotted in Figs. 6 and 7. The amounts of space charges were calculated from the space charge profiles just after the short-circuit.

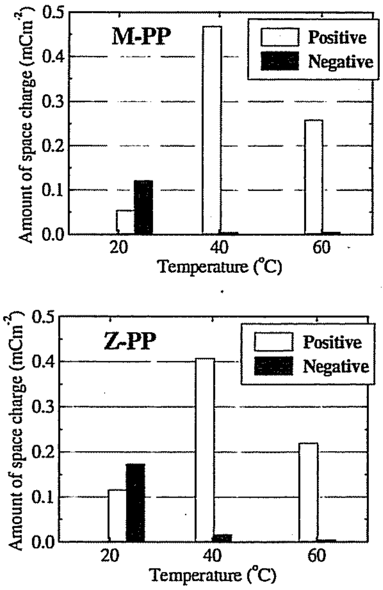


Fig. 6: Total amounts of space charges in M-PP and Z-PP after the DC field application of +50 MV/m (positive polarity) at 23, 40 and 60 °C.

In the case of positive polarity (SC anode and Al cathode), the positive charge injected from the SC anode is dominant in both M-PP and Z-PP at 40 and 60 °C, although the negative charge injected from the Al cathode is larger at 23 °C. (see Fig. 6) In the case of negative polarity (SC cathode and Al anode), on the other hand, large positive and negative charges are in M-PP and Z-PP at 40 °C and the negative charge injected from the SC cathode is dominant at 60 °C. (see Fig. 7) Figures 6 and 7 lead to the following conclusions;

(1) Z-PP and M-PP show similar behaviors of space charge in spite of the difference in morphology.
(2) Space charge is the largest around 40 °C.
(3) Charge injected from the SC electrode is dominant at 60 °C.
(4) The negative charge injected from the Al cathode plays an important role at 23 °C.
(5) In negative polarity, the positive charge injeced from the Al anode plays an important role at 40 °C, but it is surpassed at 60 °C by the negative charge from the SC cathode.

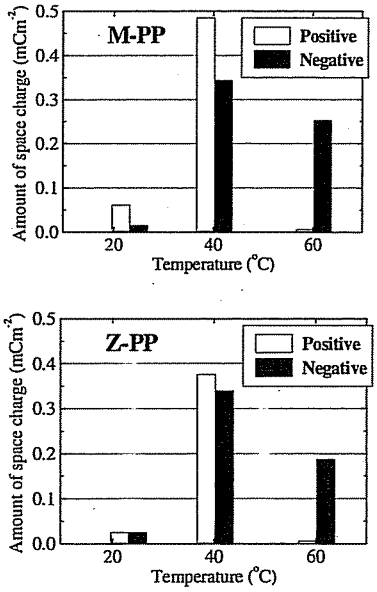


Fig. 7: Total amounts of space charges in M-PP and Z-PP after the DC field application of -50 MV/m (negative polarity) at 23, 40 and 60 °C.

Current in M-PP and Z-PP : In order to measure DC current, M-PP or Z-PP specimen (100 μm in thickness) was set between SC and Al electrodes (50 mmϕ). The charging curent under DC field of +50 MV/m and the dischaging current after the short-circuit were measured for 90 and 30 minutes, respectively. Figure 8 shows charging currents in M-PP and Z-PP at 60 °C and Figure 9 shows the temperature dependence of charging

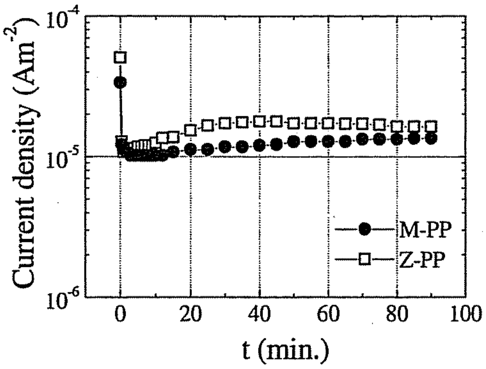


Fig. 8: Charging currents for M-PP and Z-PP at 60 °C under DC field of +50 MV/m (positive polarity).

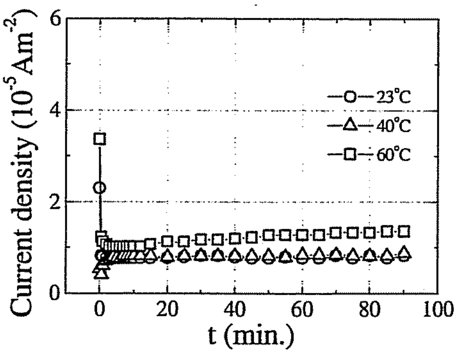
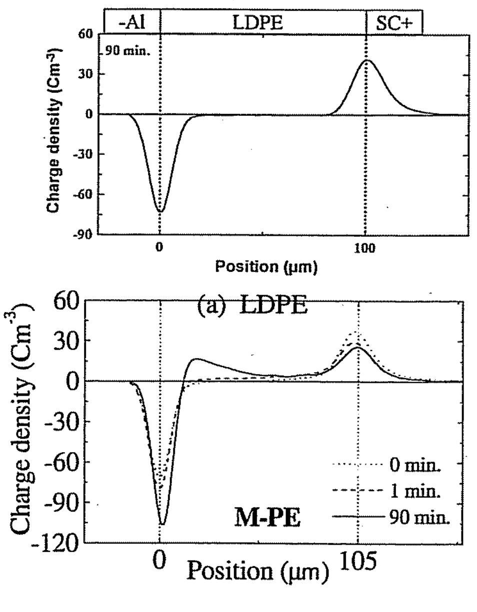


Fig. 9: Temperature depndence of charging current in M-PP at various temperatures under DC field of +50 MV/m (positive polarity).

current density in M-PP at +50 MV/m. The charging current increases with temperature and Z-PP shows a little larger current than M-PP. These are consistent with the results of space charge as shown in Figs. 2 and 5 where charge carriers move a little faster in Z-PP than in M-PP.

Space charge in LDPE and M-PE : The space charge distributions in LDPE and M-PE under the same condition are shown in Fig. 10 [5]. There ia almost no



(b) M-PE

Fig. 10: Space charge distributions in LDPE and M-PE at 60 °C, +50 MV/m.

space charge in LDPE during the application of DC 50 MV/m. In M-PE, positive charge injected from the SC anode reached the counter Al electrode and form the hetero space charge in one minute. Carriers can move much faster in polyethylene than in polypropylene. The space charge dynamics are quite different between polypropylene and polyethylene.

These results will be discussed considering the difference in morphology and so on.

Conclusions

We investigated the space charge dynamics in M-PP and Z-PP. The main conclusions are as follows.

(1) Z-PP and M-PP show similar behaviors of space charge whose amount is the largest around 40 °C.
(2) Charge injected from the SC electrode is dominant at high temperatures (40 and 60 °C).
(3) Polypropylene show different space charge dynamics from polyethylene.

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