

Effects of Sample Preparation Conditions and Short Chains on Space Charge Formation in LDPE

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ABSTRACT

The effects of chemical change during sample preparation and low molecular weight chains of low density polyethylene (LDPE) have been investigated in order to clarify the origins for heterocharge in LDPE which does not contain antioxidants. The carbonyls formed at the surface of polyethylene (PE) sheet prepared at 180°C with polyethylene terephthalate backing films were found to contribute to the increase of heterocharge in LDPE. All aspects of the processes used for the preparation of samples must be studied carefully to ascertain possible effects on the properties of samples. The type of charge accumulated in LDPE was also found to depend on the concentration of short PE chains. Details of experimental results and their possible causes are described.

1. INTRODUCTION

POLYETHYLENE (PE) is one of the most extensively studied polymeric dielectrics. As far as space charge behavior is concerned, there is general agreement among workers in this field that the presence of volatile residuals generated by the thermal decomposition of dicumyl peroxide causes crosslinked polyethylene (XLPE) to accumulate heterocharge [1, 2]. Here, heterocharge is the real charge, the polarity of which is opposite to that of the electrode. In contrast, homocharge is the real charge, the polarity of which is the same as that of the electrode. In low density polyethylene (LDPE), however, the type of charge reported in the literature is not always consistent. For example, Ditchi *et al.* [3] and Mahdavi *et al.* [4] reported the observation of both homocharge and heterocharge in LDPE depending on the samples and test conditions. Suzuoki *et al.* [5] observed the heterocharge in LDPE at fields < 1.2 MV/cm. Li *et al.* [6] observed heterocharge in LDPE while Tanaka *et al.* [7] observed homocharge.

This discrepancy may originate from two sources. One is the material related, and the other is a process related source. The factors known to encourage the formation of heterocharge are impurities and additives. There may be organic and inorganic impurities in LDPE, which may be generated either during the polymerization process or in the post-polymerization process. Antioxidants are also known to affect the formation of space charge in LDPE.

There are some factors which can affect the formation of space charge in LDPE. The low molecular weight chain of LDPE is one of the material-related sources for heterocharge and should take some role in forming space charge. There may also occur some chemical changes during sample preparation, because slab-type samples are prepared usually at elevated temperatures ranging from 120 to 190°C with backing films such as polyethyleneterephthalate (PET) and polytetrafluoroethylene (PTFE). These temperatures may be high enough for the polymer chains to be oxidized or react with the backing polymer film.

In this study, the effects of low molecular weight parts of PE (short PE chains) on the formation of space charge in LDPE were investigated. Chemical changes which may occur during sample preparation have been examined and their effects on charge distributions in LDPE were studied also.

2. EXPERIMENTAL PROCEDURES

The effects of sample preparation conditions were studied using LDPE with density and melt index (MI) of 0.920 g/cm³ and 2.2 g/10 min, respectively. The conditions evaluated for this study were the molding temperature (120 and 180°C) and the molding time (2 to 40 min). Two backing films, polyethyleneterephthalate (PET) and polytetrafluoroethylene (PTFE), were evaluated in terms of their effects on charge distributions. PET films of

0.25 mm thickness and PTFE films of 1 mm thickness were obtained from a local supplier.

Short PE chains having low molecular weight of this LDPE were removed by immersing in xylene at various temperatures ranging from 50 to 72°C. Xylene treated LDPE was washed again with hot fresh xylene and then dried in a vacuum oven at 60°C for 50 h to remove the residual xylene. The same LDPE as for the effects of sample preparation conditions was used for this study.

Average molecular weights of xylene extracted samples were measured using a gel permeation chromatography (GPC, Waters 150C) at 170°C using a differential refractometry detector and the solvent was 1,2,4-trichlorobenzene. 100 ppm of antioxidant was added to the solution to prevent thermal degradation during a GPC experiment. An analysis of molecular structure such as concentration of structural irregularities was made by a FTIR. A surface analysis was done using FTIR-ATR technique.

The effects of MI on the charge formation were studied using the LDPEs having different melt indices ranging from 0.25 g/10 min. These samples were designated as LDPE A (MI = 0.25 g/10 min), LDPE B (MI = 2.2 g/10 min), LDPE C (MI = 1.8 g/10 min), LDPE D (MI = 6.0 g/10 min), LDPE E (MI = 24 g/10 min) and LDPE F (MI = 45 g/10 min). All samples contain ~ 30 ppm of antioxidant (BHT) mixed before the pellets are exposed to air after the polymerization process. From a preliminary study, no change in charge distributions was found in as-received and chloroform extracted LDPE B samples.

To measure charge distributions, slab-type samples were prepared by compression molding at 120°C for 10 min. However, the samples for the study of effects of sample preparation conditions were compression molded at different conditions, details of which will be described in the text. All samples have the thickness of ~ 1.0 mm. Semiconductive electrodes of ~ 150 μm thickness were also compression molded at 180°C for 10 min from raw material used to shield medium voltage power cables. Semiconductive electrodes were vacuum degassed at 80°C for 100 h to eliminate volatile residuals known to influence the formation of space charge in XLPE [1]. Vacuum degassed semiconductive electrodes were then bonded on the surface of samples using a hot iron.

The raw material for semiconductive electrodes is a compound of olefinic polymer, conductive carbon black, processing aids and antioxidants. These components are known to affect the charge distribution in LDPE [3], so that the type of semiconductive compound was maintained to be the same throughout the study.

Space charge distributions were measured by a pulsed electroacoustic (PEA) method, the details of which have been described elsewhere [1, 8]. Measurements of charge distributions were carried out as follows: First, the voltage was applied for 30 min across a ~ 1 mm thick sample with semiconductive electrodes. After voltage application for 30 min, the voltage was turned off and then immediately the spatial charge distribution was measured. After all these processes had been gone through, the voltage was ramped up to the next test voltage. The final voltage was 40 kV, which corresponds to an electric field of

~ 400 kV/cm. In this study, the charge distribution during the voltage application was not measured. Therefore, all charge distributions described in this paper represent the residual charge remaining in the sample after the voltage was removed.

One can notice that charge peaks near the anode are small compared to charge peaks near the cathode. This is because the signals generated near the anode are attenuated while traveling through the sample. Because of this reason, the charge near the cathode was considered to be significant. Besides, since sample thicknesses are slightly different from sample to sample, the charge distributions shown in the figures were aligned for the induced charge peak at the cathode to start at the same position. The geometry, such as sample thickness, voltage and electrodes, of the test sample is indicated on the top of the Figure.

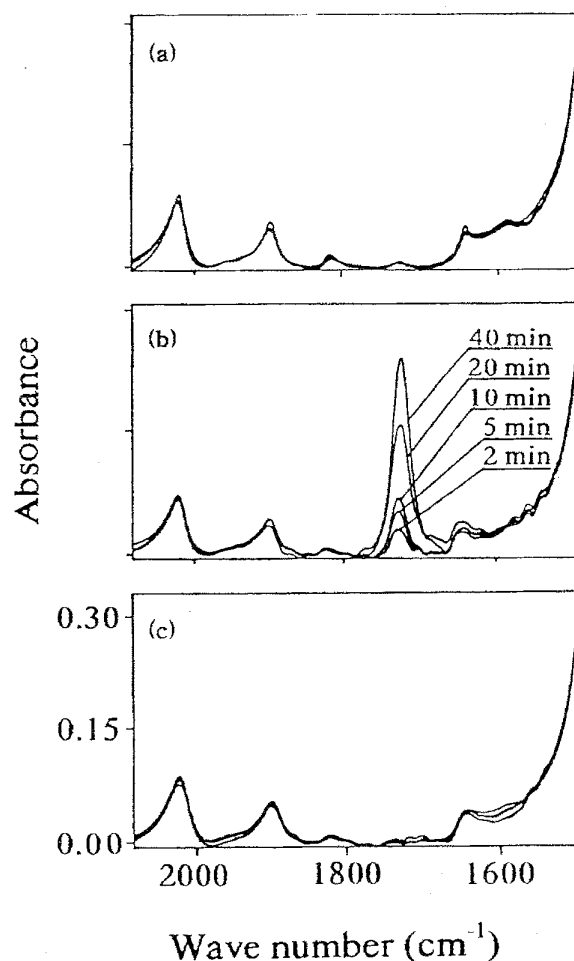


Figure 1. FTIR-ATR spectra of LDPE B with PET and PTFE backing films at 120 and 180°C. (a) LDPE B pressed at 120°C with PET backing film, (b) LDPE B pressed at 180°C with PET backing film, (c) LDPE B pressed at 180°C with PTFE backing film; molding times 2 to 40 min.

3. EXPERIMENTAL RESULTS

3.1. CHEMICAL CHANGE DURING SAMPLE PREPARATION

Figure 1 shows the evolution of carbonyl groups at the surface of compression molded LDPE B as a function of compression molding time from 2 to 40 min at 120 and 180°C. FTIR-ATR was used for this analysis. It should be mentioned that transmittance-type FTIR spectra showed no appreciable carbonyl peaks.

Several peaks are observed in FTIR-ATR spectra. Of these, the peak at 2018 cm^{-1} is an internal reference peak representing a stretching mode of methylene group of PE and the peak at 1727 cm^{-1} is a characteristic peak for the carbonyl group in PET [9]. The absorbance of this peak is lower in a 120°C result than in a 180°C result. The intensity of the carbonyl peak increases as the molding time increases at a molding temperature of 180°C while it remains relatively unchanged at a molding temperature of 120°C. The peak at 2018 cm^{-1} also remains unchanged. When the sample is prepared with PTFE backing films, no carbonyls were detected at a molding temperature of 180°C.

Figure 2 shows the charge distributions measured at 30 kV for 10 and 40 min molding times at 120 and 180°C. All data show the heterocharge. When the sample is prepared with PET backing films, the sample prepared at 180°C and fixed molding time yields a larger heterocharge peak than the sample prepared at 120°C, whereas the sample prepared for 40 min at a fixed molding temperature yields slightly smaller heterocharge peak than the sample prepared for 10 min. Figure 3 shows the molding temperature dependency of heterocharge at 30 kV in this LDPE. In this Figure, we can see that the heterocharge increases as the molding temperature increases. When the sample is prepared with PTFE backing films, heterocharge is also observed. In this case, however, the type and magnitude of heterocharge remain relatively unchanged. The magnitude of heterocharge prepared by PTFE backing films is smaller than the magnitude of heterocharge prepared by PET backing films.

The results showed a large difference in charge distribution as well as structure at the sample surface, depending on sample preparation temperature. Since no considerable change in surface structure as a function of molding time was observed in samples prepared at 120°C, this condition was adopted as a standard procedure to prepare the samples for this study. Other conditions were also employed for specific purposes, which are specified in the text.

3.2. XYLENE EXTRACTION OF LDPE

LDPE B has been extracted in xylene at temperatures of 52 to 72°C and their characteristics have been studied.

GPC curves of xylene extracted LDPE are shown in Figure 4. One can see that the low molecular weight part is removed by xylene extraction, while the high molecular weight part remains unchanged. Average molecular weights are listed in Table 1 and number average molecular weights are plotted as a function of

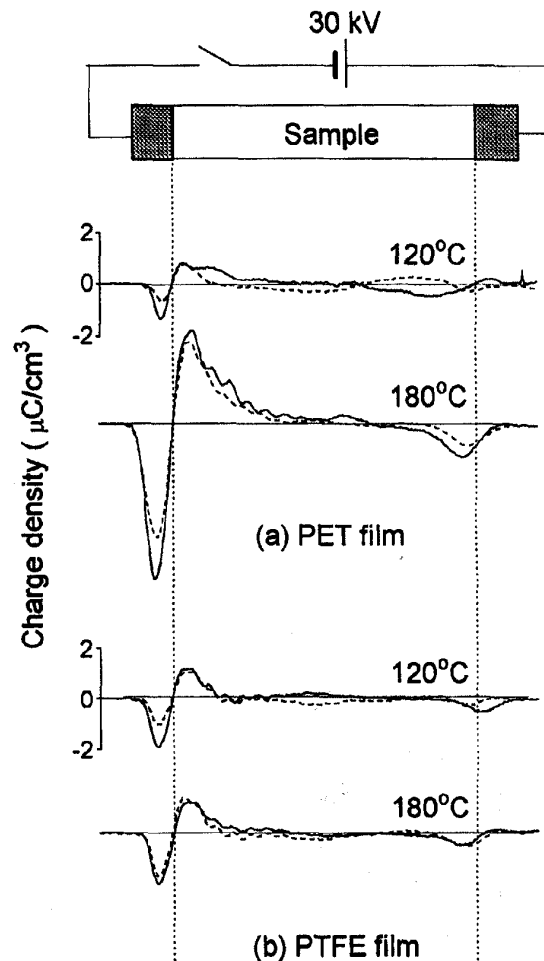


Figure 2. Charge distributions at 30 kV for 10 and 40 min molding times at 120 and 180°C 10 min (solid line), 40 min (dashed line).

Table 1. Average molecular weights of xylene-extracted LDPE B. M_n number average molecular weight, M_w weight average molecular weight.

Extr. T °C	M_n g/mole	M_w g/mole
control	18000	137000
52	26000	147000
61	34000	162000
66	37000	168000
69	41000	177000
72	44000	182000

extraction temperature in Figure 5. As shown in Table 1 and Figure 5, as the extraction temperature increases, the average molecular weight increases and polydispersity decreases, because more low molecular weight material is removed from LDPE.

Structural irregularities such as carbonyl, methyl group and unsaturation were analyzed using FTIR, the results of which are summarized in Table 2. As shown the concentration of structural

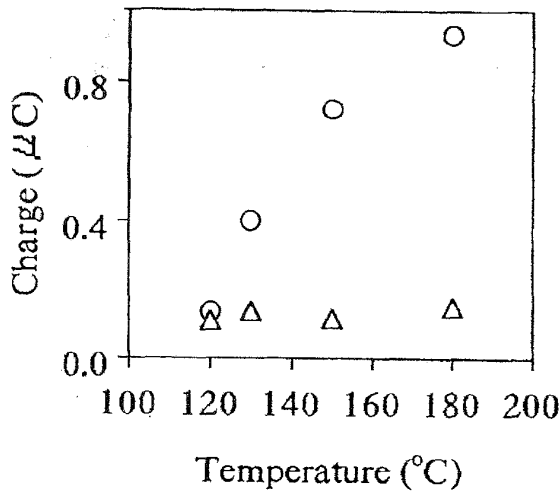


Figure 3. Molding temperature dependence of heterocharge near the cathode. Molding time 10 min, PET film (O), PTFE film (Δ).

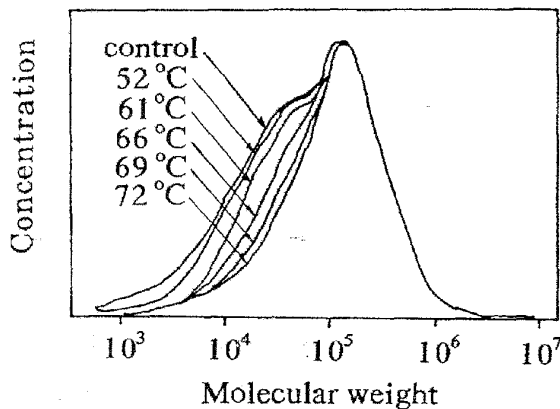


Figure 4. GPC curves of xylene treated LDPE at temperatures of 52 to 72°C.

Table 2. Concentration per 1000 carbon atoms of structural irregularities in xylene-extracted LDPE B. $RR'C=O < 0.02$ for all samples, $RCH=CHR' < 0.07$ for all samples.

Extr.T °C	RCH ₃	RCH=CH ₂	RR'C=CH ₂
control	30.79	0.295	0.346
50	31.10	0.258	0.311
60	29.82	0.230	0.295
63	29.47	0.217	0.286
67	29.21	0.197	0.287
70	27.42	0.186	0.256

irregularities of LDPE were found to decrease as extraction temperatures increased. This result indicates that both oxidation and chain scission reactions have not occurred extensively during the extraction process and that the low molecular weight PE chains are removed by xylene extraction. Otherwise, the concentration of unsaturation and methyl group should increase.

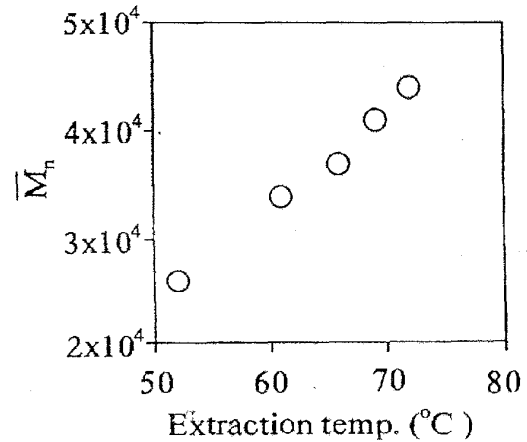


Figure 5. Number average molecular weights as a function of extraction temperature.

Figure 6 shows the charge distributions at 40 kV of control and xylene extracted LDPE. The amount of charge near the cathode is plotted in Figure 7 as a function of number average molecular weight. Heterocharge was found in all samples except the one extracted at 72°C (Figure 6). It was also found in Figure 7 that the amount of charge near the cathode decreased as the number average molecular weight increased. A very small homocharge peak is seen in the 72°C result.

3.3. LDPE SAMPLES WITH DIFFERENT MI

The effects of short PE chains on the formation of space charge in LDPE were examined further by selecting the LDPE having different MI from 0.25 to 45 g/10 min (LDPE A to LDPE F). All samples were prepared at 120°C using PET backing films.

Figure 8 shows charge distributions at 20 and 40 kV. As shown, both heterocharge and homocharge are observed in PE and a higher voltage produces a higher charge peak. The LDPE having MI of 0.25, 2.2 and 6.0 g/10 min show heterocharge whereas LDPE with MI of 1.8, 24 and 45 g/10 min show homocharge. Charge distributions obtained with LDPE A, B and D show that the width of charge distribution becomes narrower and closer to the electrode/sample interfaces as the melt index increases. These results will be discussed in the Section 4 in terms of effects of short PE chains.

3.4. BLENDS OF VARIOUS MOLECULAR WEIGHT PARTS

The role of low molecular weight parts in LDPE has been investigated further using blends of low and high molecular weight parts of LDPE B. Blends were prepared as follows. The low molecular weight parts of LDPE were removed by immersing in xylene at 67°C. The extracted wax-like material (LWP) and the residual high molecular weight parts (HWP) were separately collected and dried. These two materials were blended using an internal mixer (kneader) at 160°C for 5 min. 100 ppm antioxidant was added during the blending process. The compositions

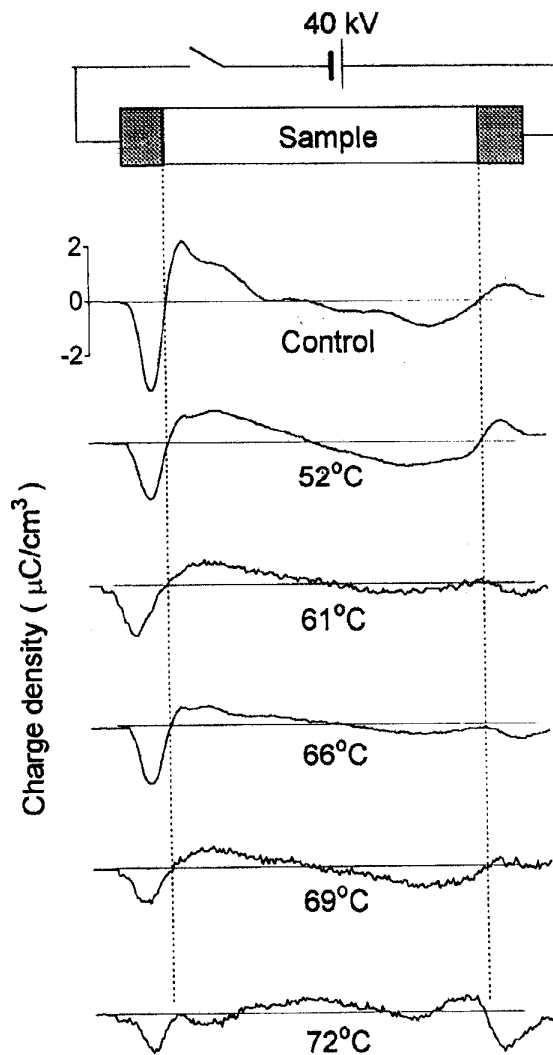


Figure 6. Charge distributions at 40 kV of control and xylene-extracted LDPE.

of LWP were 5, 10 and 20 parts per hundred resin (pphr) which corresponds to 4.8, 9.1 and 16.7% by weight, respectively.

Figure 9 shows the GPC curves of LWP/HWP blends. The LWP is the low molecular weight chain which has a number average molecular weight of ~ 6000 g/mole. The peak position of LWP component in LWP/HWP blends shifts to the HWP side when the LWP is blended with the HWP. One could expect a bimodal shape of GPC curves in these blends if the short PE chains behave independently. A shift of peak molecular weight of LWP component to the HWP side may imply that the short PE chains were recombined with the HWP chains.

Figure 10 shows the charge distributions at various voltages up to 40 kV in the blend having 10 pphr LWP. This blend shows heterocharge at all voltages. The amount of heterocharge increases first and then decreases as the voltage increases. These features are common to all blends except the blend containing 20 pphr of

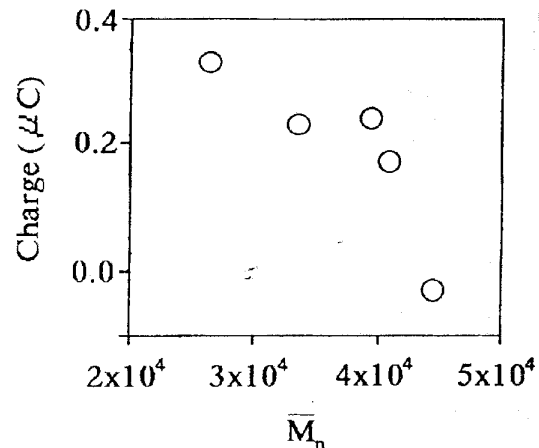


Figure 7. Charge near the cathode as a function of number average molecular weight.

LWP, as shown in Figure 11. In Figure 11, one can see that the blend with 5 pphr LWP shows the largest heterocharge at low voltages and then the heterocharge decreases as the LWP content increases. The LWP did not induce homocharge.

4. DISCUSSION

4.1. CHEMICAL CHANGE DURING PREPARATION

It was found that the chemical change can occur during the sample preparation process when the sample is prepared with PET backing films.

The important chemical change is the formation of carbonyl in LDPE. Although a carbonyl peak is observed at the surface of sample, no considerable oxidation occurs during compression molding at 180°C for up to 40 min. Evidence to support this is that no carbonyl peaks were observed when the sample was prepared with PTFE backing films, and that 1727 cm^{-1} is a characteristic peak for the carbonyl group in PET. If there were oxidation at the sample surface, the carbonyl peak at 1720 and 1740 cm^{-1} should be detected regardless of the type of backing film.

The observation of a 1727 cm^{-1} peak implies that a part of chain in PET is present at the surface of LDPE. This can be attributed to the incorporation of short PET chains on the surface of LDPE samples. However, the possibility of some chemical reaction between LDPE and PET during molding may not be excluded. This chemical reaction may occur between the functional groups of LDPE, such as carbonyl and unsaturation, and the functional groups of PET such as carbonyl. Since no appreciable carbonyl peaks are observed in a transmittance-type FTIR spectrum, it follows that this chemical reaction is confined only to the surface region of LDPE sample.

Charge distributions were found to differ depending on the type of backing polymer layer for the preparation of LDPE samples. For this, see Figures 2 and 3. Li *et al.* [10] reported that heterocharge is accumulated in LDPE when the sample was prepared

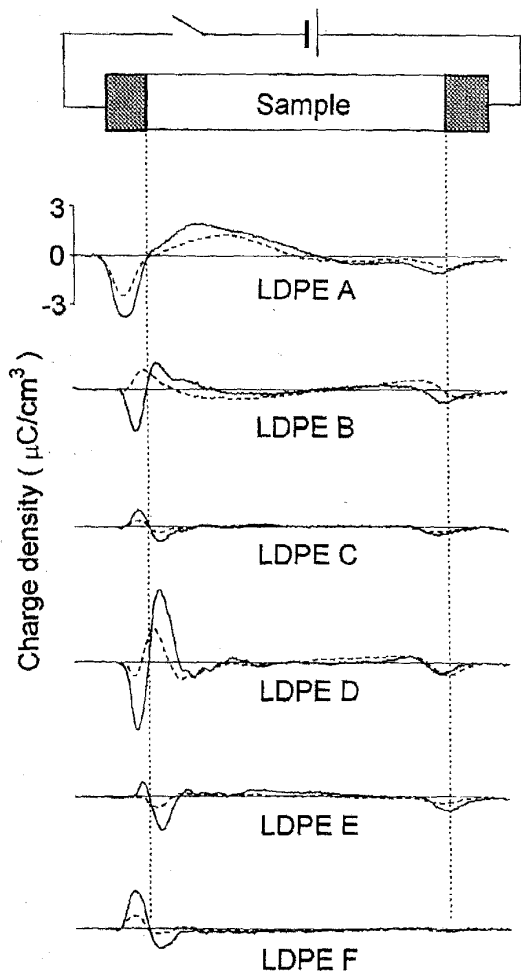


Figure 8. Charge distributions at 20 kV and 40 kV in LDPE with different MI. From the top: LDPE A (0.25 g/10 min), LDPE B (2.2 g/10 min), LDPE C (1.8 g/10 min), LDPE D (6.0 g/10 min), LDPE E (24 g/10 min), LDPE F (45 g/10 min).

with PET backing films while homocharge is accumulated when the sample was prepared with PTFE backing films. No such dramatic change was observed in this study. A close comparison of these results cannot be made at this moment because Li *et al.* did not specify sample preparation conditions. It can be said that our results are comparable to their results because we found a considerable reduction in heterocharge in PTFE backed samples. A decrease of heterocharge can be said to reflect an increase of homocharge.

The observation of carbonyl peaks at 1727 cm^{-1} with the PET film at 180°C is a clear indication of material transfer from PET. The possibility of material transfer has been addressed previously by Azrak [11], although he did not see any appreciable carbonyl peaks in LDPE with melt index 400 g/10 min by reflectance infrared measurements. Our results cannot be directly compared

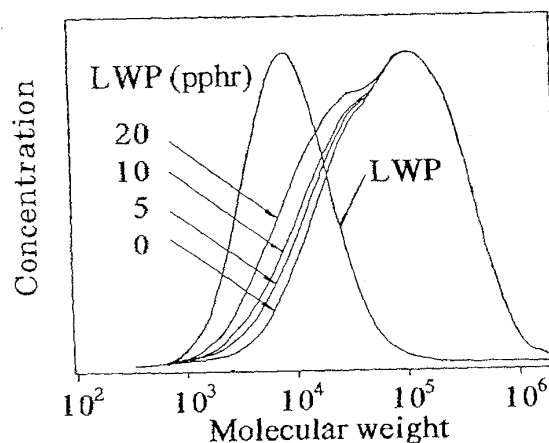


Figure 9. GPC curves of LWP/HWP blends.

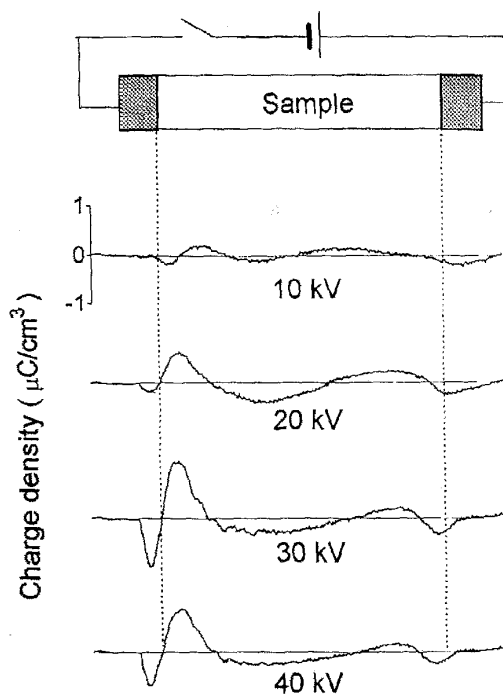


Figure 10. Charge distributions at voltages up to 40 kV in LWP/HWP blend: Composition of LWP: 10 pphr.

to his result because molding conditions were not specified in his report and also the samples were different. It is possible, as he suggested in his paper, that the polar groups can orient during the sample preparation process, which can enhance the formation of heterocharge due to the orientation of polar groups near the surface. There may be some polar components in LDPE chains because this polymer is polymerized by initiators such as oxygen or peroxides, although no information on their concentration is available at this moment. These polar components may contribute to the formation of heterocharge. This will be mentioned

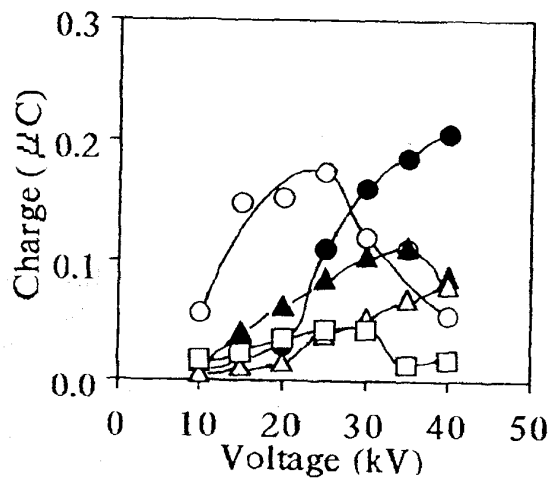


Figure 11. Voltage dependence of charge in LWP/HWP blends: Composition of LWP: 0 pphr (●), 5 pphr (○), 10 pphr (filled triangle), 20 pphr (△), LWP (□).

again in Section 4.2 on the role of short PE chains.

Heterocharge found in all PET backed samples increased as molding temperature increased from 120 to 180°C. This is contradictory to the previous report by Suzuoki *et al.* [5]. They reported that the carbonyl groups, formed by the thermal oxidation, in PE enhances the formation of homocharge due to the enhanced charge injection. If this is the case in our samples, the heterocharge peak in the 120°C test should be much larger than that in the 180°C test. Regarding the effects of carbonyls, two expectations are possible. One is that, as claimed by Suzuoki *et al.*, the carbonyls enhance charge injection or charge trapping. The other may be that the carbonyls are polarized under dc fields so that they encourage the formation of heterocharge. The latter may be the case in our samples. Probably, PET chains present in LDPE surface are polarized to form heterocharge, which increases the heterocharge peak in LDPE.

4.2. ROLE OF SHORT PE CHAINS

Some results on the roles of short chains on space charge formation in LDPE were observed. These are the heterocharge decreases as more short PE chains are removed from the LDPE; the amount of heterocharge in various polyethylene does not depend on their MI values; and the heterocharge decreases as the LWP content in LWP/HWP blends increases.

The concentration of short PE chains can be estimated by a xylene extraction. Six LDPE samples were extracted in xylene at 67°C, the results of which are shown in the Table 3. As shown, the PE with a higher MI results in a higher extractable fraction. However, the concentrations of short PE chains in LDPE B and C were quite different, although their MI are similar to 2.0 g/10 min: 4.4% for LDPE C and 12.7% for LDPE B. The extractable fraction of LDPE A is 7.0%, although its MI is 0.25 g/10 min. These results suggest that the melt index itself is not a good measure of the concentration of short PE chains.

Table 3. Concentration of short chains and charge of LDPE samples. MI at 2.16 kg; LWP extractable at 67°C.

Sample	MI g/10 min	LWP %	Q at 40 kV μC
LDPE A	0.25	7.0	0.614
LDPE B	2.2	12.7	0.366
LDPE C	1.8	4.4	-0.065
LDPE D	6.0	16.2	0.280
LDPE E	24	89.1	-0.117
LDPE F	45	95.7	-0.080

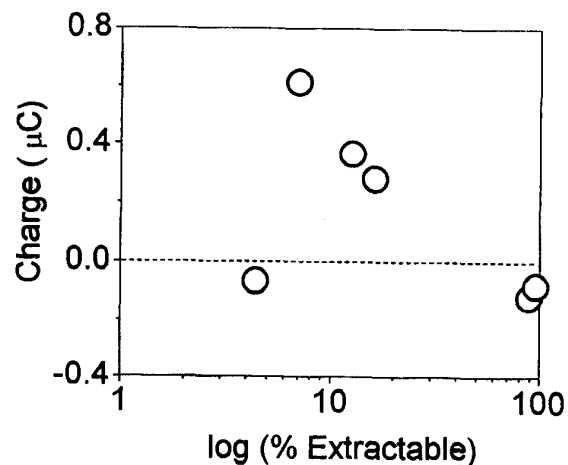


Figure 12. Extractable fraction dependence of charge near the cathode in LDPE samples having different MI.

The amount of charge near the cathode was plotted as a function of the extractable fraction in Figure 12, where one can see that the variation of charge can be represented by a single curve. Its implication is that the concentration of short chains in LDPE may be the factor controlling the type and amount of charge in LDPE samples. Homocharge develops when the short chain content is very low and very high while heterocharge develops when the short chain content is intermediate.

The development of heterocharge in LDPE is quite possibly based upon the polymerization process. The PE is polymerized by initiators such as oxygen or peroxides. This allows PE chains to possess oxygen containing sites, although its concentration should be very low. These polar groups may behave like ionic species and thus contribute to the formation of heterocharge. It is not clear whether these short PE chains were polarized or migrated under dc stresses.

Space charge may be influenced by the chain mobility of PE. As far as the heterocharge is concerned, as chains are more mobile, orientation of polar groups and migration of ionic species become easier, resulting in an increase of heterocharge. However, it may be possible that the accumulated heterocharge induces charge injection from the electrode because heterocharge will increase local electric fields. This concept may explain the present observations. Short chains should be more mobile than long chains. When the

concentration of short chains is very low ($< 2\%$), both migration and polarization should be very difficult, resulting in development of homocharge. On the other hand, as the concentration of short chains increases, both polarization and migration become easier, resulting in development of heterocharge (maybe $\sim 20\%$). As the heterocharge develops closer to the electrode/sample interfaces, the local field near the interfaces becomes higher and consequently the barrier height for charge injection becomes lower. In this situation, the decrease of heterocharge (or an appearance of homocharge in an extreme case) by enhanced charge injection is expected. This may be the case for a short chain content $> 30\%$.

The present results were explained in terms of concentration of short PE chains having low molecular weight. However, other molecular characteristics such as molecular weight, density, crystallinity and molecular weight distributions may affect the formation of space charge. Effects of solvent extraction on charge distributions with other LDPE may be of great interest. A further study on these factors may be needed.

4.3. REASONS FOR INCONSISTENT RESULTS OF THE REPORT

In this study, different charge distributions were observed depending on sample preparation conditions and material characteristics such as concentrations of short chains in LDPE. Of these, both sample preparation temperature and type of backing film were found to be the most critical factors to determine the charge distributions in LDPE. This may be one of the major reasons for inconsistent results of the reports. These facts suggest that details on sample preparation and material characteristics have to be specified in any report. In order to obtain consistent results on the charge distribution, the samples may have to be prepared carefully and high temperatures should be avoided to prevent the chemical interaction between sample and backing polymer film. The preparation of samples at low temperatures by the use of PTFE films is recommended. However, the preparation of samples at high temperatures by the use of PET backing films may be acceptable when the conditions of sample preparation are not altered during a series of experiments.

5. CONCLUSIONS

A chemical change can occur during the sample preparation process when the PET film is used as a backing layer. The important chemical change is the formation of carbonyl at the LDPE surface. No oxidation reaction occurs even at the molding conditions of 180°C and 40 min. These carbonyls contribute to the increase of heterocharge in LDPE.

The type of charge accumulated in the LDPE is governed by the concentration of short PE chains. Homocharge develops when the concentration of short PE chains is very low whereas heterocharge develops when the concentration of short PE chains increases. In this case, the short PE chains behave like an ionic species.

These short PE chains reduce the heterocharge eventually at very high concentration. The decrease of heterocharge at high content of short PE chains was tentatively attributed to the enhancement of local field near the electrode/sample interfaces and subsequent lowering of the injection barrier.

It is concluded that the different results on the formation of space charge in LDPE originates from the different nature of LDPE samples.

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