

Charge Distribution in Polyethylene/Ethylene Vinylacetate Laminates and Blends

Kwang S. Suh, Jae Young Kim, Chang Ryong Lee

Department of Materials Science, Korea University, Seoul, Korea

and Tatsuo Takada

Electronic Measurement Lab., Musashi Institute of Technology, Tokyo, Japan

ABSTRACT

The formation of space charge in polyethylene/ethylene vinylacetate copolymer (PE/EVA) laminates was investigated using a pulsed electroacoustic (PEA) method. The formation of heterocharge was observed in PE while positive charge (holes) was distributed over the EVA specimen. In the laminates, however, a large amount of charge accumulates at the interface between PE and EVA, resulting in an increase of the electric field in the PE layer. When the thickness of the EVA layer is smaller than that of the PE layer, negative charge dominates in the PE layer due to enhanced charge injection caused by the increased electric field in the PE layers. When these polymers are blended, the amount of heterocharge in the PE/EVA blends varies inversely with the vinylacetate content by the enhancement of homocharge trapping at PE/EVA interfaces and/or by the enhancement of charge injection via an increased electric field in the PE component.

1. INTRODUCTION

VARIOUS types of interface are found in polymeric insulating materials or in electrical insulating systems. Typical examples are the interface between layers of power cables and the interface in polymer blends. The former is the macroscopic and sharply definable interface and the latter is the microscopic and randomly distributed interface. Since the interface can govern the performance of electrical insulation systems, their roles and effects should be well characterized for a better understanding of electrical insulation phenomena.

For the study of interfacial phenomena, the polymer blends may not be a good choice for the specimen to work with because the interface in the blends is so microscopic in size and so randomly distributed all through the specimen that the interpretation of test results may be very difficult. However, the polymer laminate may provide the interface between two or more parent polymers and presumably simulate the interface in polymer blends. Moreover, the nature of the interface in the laminates can be altered by simple treatment such as heat treatment or pasting of chemicals [1].

Some results have been reported on space charge problems in the polymer laminates. Suzuoki *et al.* [2], through their work on the thermally stimulated currents (TSC) of polyethylene/ethylene vinylacetate copolymer (PE/EVA) laminates, reported that the charge develops at the interface and the positive charge (holes)

is injected at the EVA side near the interface. A similar result was observed by other investigators [3–5]. All these works considered only double layer laminates, *i.e.*, PE/EVA laminates, which is not sufficient for the explanation of charge formation in PE/EVA blends. More complicated laminates are needed.

The aim of this paper is to investigate the roles or effects of interface on the charge formation in PE/EVA laminates. In this study, the array of laminates has been extended to triple layered structures and an attempt was made to explain the charge formation in PE/EVA blends via the results on the laminates.

2. EXPERIMENTS

2.1. SAMPLES

The parent polymers used in this study are low density PE and EVA. Density and melt index of the PE are 0.920 g/cm³ and 2.0 g/10 min, respectively. The nominal weight percentage of vinyl acetate (VA) of EVA is 15%.

PE/EVA laminates were prepared as follows. First, polymer sheets of proper thicknesses were prepared using a hot press at 120°C for 10 min for both PE and EVA. These sheets were then pressed at 80°C to prepare the laminates. The thickness of polymer sheet was adjusted to result in laminate thickness of ~ 1.0 mm. The thickness of each layer for double layer laminates is ~ 0.5 mm and that for triple layer laminates is ~ 0.35 mm.

PE/EVA blends were prepared using a two-roll mill at 120°C and their compositions were adjusted to result in the VA content of blends to be 1 and 4%, corresponding to 6.5 and 22.0%wt of EVA, respectively.

The semiconductive electrodes were used for the measurement of charge distributions. This semiconductive material is made of EVA, conductive carbon black and additives such as processing aids and antioxidant. It has been demonstrated earlier that the composition of the semiconductive material influences the formation of charge in polyethylene [6]. Therefore, the same type of semiconductive material was used for the electrodes throughout the present study. Films of $\sim 150 \mu\text{m}$ thickness were prepared by curing at 180°C using a hot press. The cured semiconductive film was vacuum degassed at 80°C for 100 h to remove the residual byproducts which may affect the formation of space charge in PE [7, 8] and then attached against the sample using a hot iron.

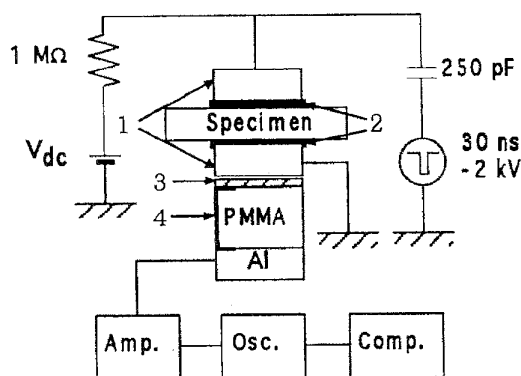


Figure 1. Schematic diagram of PEA instrument for the measurement of space charge distribution. 1: Al electrode, 2: semiconductive electrode, 3: PVDF, 4: Al film.

2.2. CHARGE AND FIELD DISTRIBUTIONS

The charge distributions were measured by a pulsed electroacoustic (PEA) method, the schematic diagram of which is shown in Figure 1 [9]. This instrument is composed of three parts: pulse generator, sample cell including a piezoelectric detector, and data processing part including electronic equipments. The electric pulses with amplitude of -2 kV and width 30 ns were used for 1.0 mm thick samples. The piezoelectric detector was made of $28 \mu\text{m}$ thick PVDF film. A PMMA block was used as an acoustic absorber behind the PVDF film in order to prevent the reflection of acoustic pressure wave. The data processing part consists of a digitizing oscilloscope, preamplifier and personal computer for data collection, display and calibration. The measured signal was calibrated with the area of calibration peak obtained at 5 kV .

The dc voltages from 5 to 40 kV at a 5 kV interval was applied across the laminates in a stepwise manner. In each step, the voltage was applied for 30 min and then the voltage was turned off. Immediately after the voltage was turned off, the space charge distribution was measured by applying the source pulses. The

voltage was then ramped up to the next test voltage. The final voltage was 40 kV , corresponding to $\sim 40 \text{ kV/mm}$.

The electric field distributions were calculated using the following equation

$$E(x) = \frac{1}{\epsilon} \int \rho(x) dx \quad (1)$$

$$\epsilon = \epsilon_o \epsilon_r$$

where ρ is the charge density, ϵ_o is permittivity in vacuum and ϵ_r the permittivity of the sample (2.2 for polyethylene, and 2.3 for EVA [10]).

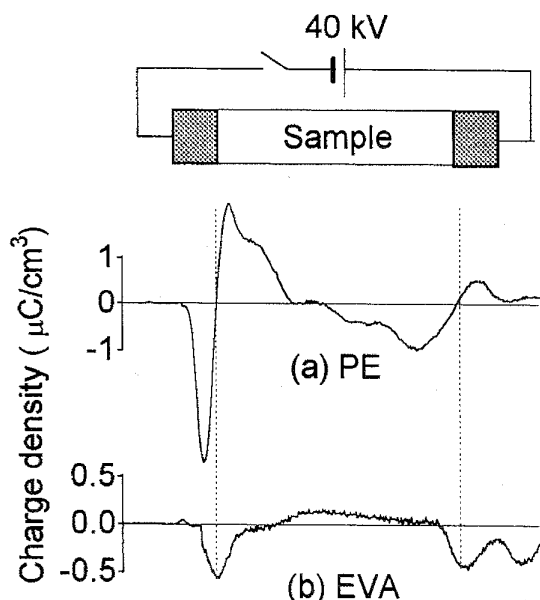


Figure 2. Space charge distribution of PE and EVA after the discharge (40 kV).

3. RESULTS AND DISCUSSION

3.1. PARENT POLYMERS

Spatial charge distributions of parent polymers, PE and EVA, are shown in Figure 2. These distributions were measured within $< 3 \text{ min}$ after the voltage was turned off.

As shown in Figure 2, the PE shows the heterocharge, i.e. the positive charge near the cathode and the negative charge near the anode. The heterocharge in PE may originate from impurities, polar groups and/or low molecular weight short PE chains [11]. On the other hand, the EVA shows the positive charge (hole) spread throughout over the specimen, which agrees well with the previous report by Suzuoki *et al.* [2]. A higher voltage results in a larger charge density for both PE and EVA.

3.2. DOUBLE LAYER LAMINATES

Figures 3 and 4 show the space charge and electric field distributions in PE (-)/EVA (+) laminates (Figure 3) and EVA (-)/PE (+) laminates (Figure 4). In these Figures, (a) shows the charge

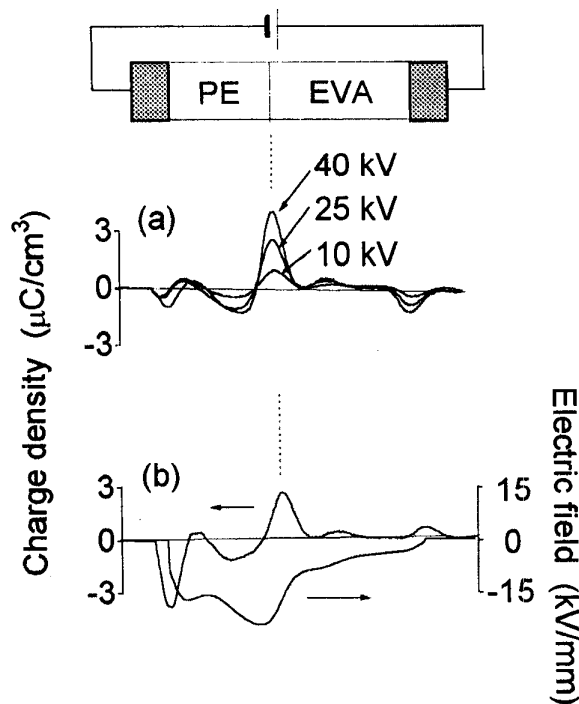


Figure 3. PE/EVA laminate. (a) space charge distribution after the discharge, (b) space charge and electric field distribution at 20 kV.

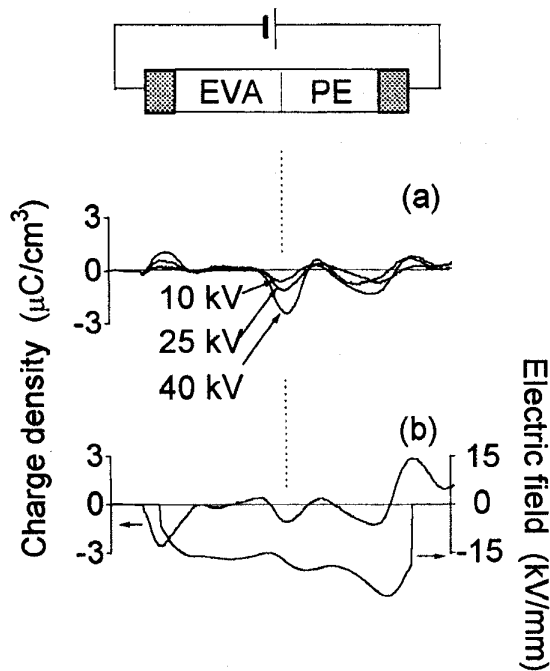


Figure 4. EVA/PE laminate: (a) space charge distribution after the discharge, (b) space charge and electric field distribution at 20 kV.

distribution at 10, 25 and 40 kV and (b) shows the charge and electric field distributions at 20 kV.

In PE (-)/EVA (+) laminates, the PE layer has positive charge

near the cathode and the negative charge near the interface, whereas the EVA layer has only a small amount of positive charge near the interface. It can be seen that the negative charge is located broadly in the PE layer. A large positive charge peak is observed at the interface between PE and EVA layers, which agrees with the previous observation [4]. As shown in Figure 4, the charge distributions at three voltages in EVA (-)/PE (+) laminates are almost the same as in Figure 3 except for the polarity of major charge peaks.

These two Figures show that the electric field in the PE layer is much higher than that in EVA, regardless of sample geometry. When the average field is $\sim 18 \text{ kV}/\text{mm}$, the maximum electric field in the PE layer becomes $\sim 24 \text{ kV}/\text{mm}$ for the PE (-)/EVA (+) laminate and $28 \text{ kV}/\text{mm}$ for the EVA (-)/PE (+) laminate.

The large charge peaks at the interface may be due to the trapping of homocharge which migrates through the EVA layer which has a much higher effective charge mobility than PE. The holes in the case of Figure 3 and the electrons in the case of Figure 4 migrate toward the counter electrode side and they are trapped near the interface which acts as a barrier for charge transport. These charges may be located at the interface in the EVA side, as pointed out by Cai [3].

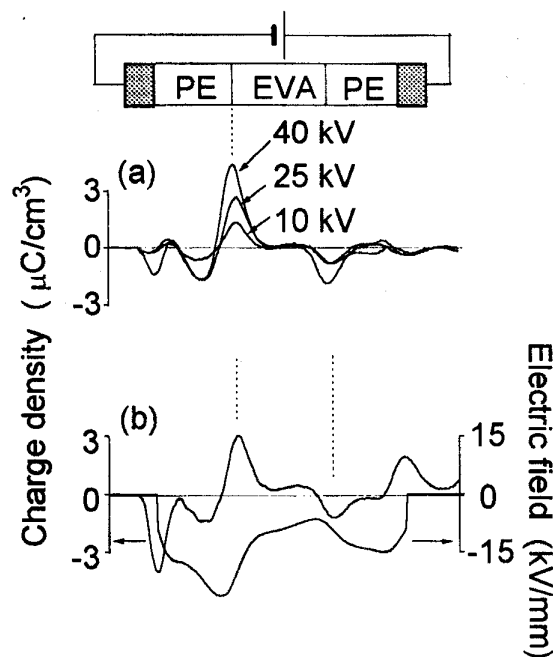


Figure 5. PE/EVA/PE laminate. (a) space charge distribution after the discharge, (b) space charge and electric field distribution at 20 kV.

3.3. TRIPLE LAYER LAMINATES

Figures 5 and 6 show the space charge and field distributions of triple layer laminates, PE (-)/EVA/PE (+) laminates (Figure 5) and EVA (-)/PE/EVA (+) laminates (Figure 6). In these Figures, (a) shows the charge distribution at 10, 25 and 40 kV and (b) shows the charge and electric field distribution at 20 kV.

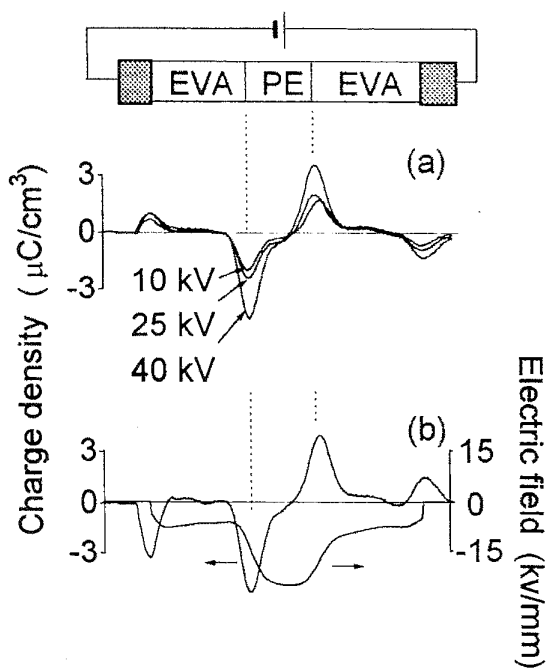


Figure 6. EVA/PE/EVA laminate. (a) space charge distribution after the discharge, (b) space charge and electric field distribution at 20 kV.

Charge distributions of PE (-)/EVA/PE (+) laminates are almost the same as in Figure 3 for the PE (-)/EVA (+) laminates except for the charge distributions in the PE layer near the anode. The charge distributions in the PE layer near the anode are not clear, probably because the signal generated in this layer might be attenuated considerably. As expected, large interfacial charge peaks are observed and almost no charge is found in the EVA layer. As in double layer laminates, the EVA (-)/PE/EVA (+) laminate shows the reversed polarity with other features being almost the same as in PE (-)/EVA/PE (+) laminates.

It was also found that, regardless of sample geometry, the electric field in the PE layer is much higher than that in the EVA layer. At an average field of 18 kV/mm , the electric field of PE layer reaches $\sim 24 \text{ kV}/\text{mm}$ in the PE (-)/EVA/PE (+) laminate and 26 kV/mm in the EVA (-)/PE/EVA (+) laminate.

The origin for the large interfacial charge can be explained in a similar way to that for double layer laminates. In the case of PE (-)/EVA/PE (+) laminates, the homocharge injected into the EVA layer might migrate toward the counter electrode side. That is, the positive charge migrates toward the cathode while the negative charge migrates toward the anode side. These charges are trapped near the interface. In the case of EVA (-)/PE/EVA (+) laminates, the charge injected from the electrodes (the negative charge from the cathode and the positive charge from the anode) migrates toward the counter electrodes and subsequently gets trapped near the interface. In this case, the EVA layers can be regarded as an extension of the electrodes.

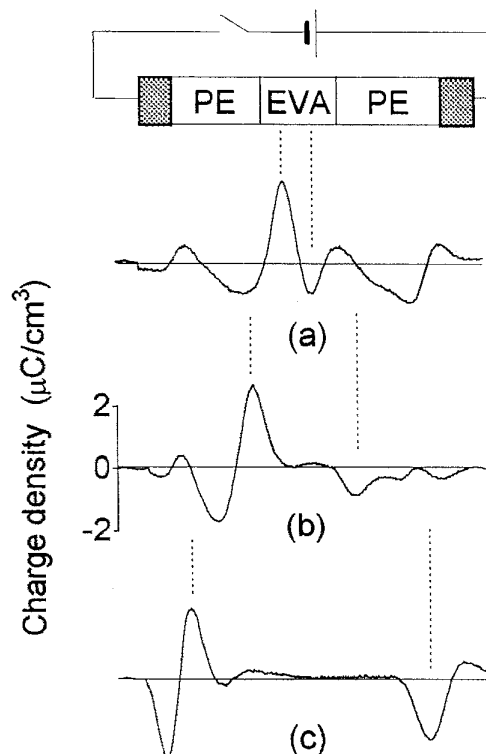


Figure 7. Thickness dependence of space charge distribution in PE/EVA/PE laminates. (a) 100 μm EVA, (b) 330 μm EVA, (c) 800 μm EVA.

3.4. EFFECTS OF LAYER THICKNESS

The results on the effects of layer thickness on the charge distributions in PE (-)/EVA/PE (+) laminates are shown in Figure 7. The thickness of EVA layer was 100, 330 and 800 μm and the thickness of PE layers was adjusted to result in laminate thickness of $\sim 1.0 \text{ mm}$.

When the thickness of EVA layer is 100 μm , two large interfacial charge peaks are found in the middle and negative charge predominates in the PE layers (Figure 7(a)). These charge peaks are separated by some distance when its thickness is 330 μm (Figure 7(b)). When the thickness of EVA layer is 800 μm , the interfacial charge peaks are separated even further and the charge peaks near the electrodes are overlapped (Figure 7(c)).

When the thickness of any layer is much smaller than the others, the charge distributions near the interface of laminates are overlapping so that the discrimination of contribution of each component is no longer trivial. This overlapping of charge peaks results from the instrumental limitation. The spatial resolution of this instrument was estimated from the calibration peaks at 5 kV to be $\sim 70 \mu\text{m}$, which is not sufficient in resolving the charge distribution developed over $\sim 100 \mu\text{m}$ distances.

3.5. PE/EVA BLENDS

In order to correlate the situation of the laminates to the situation of the blends, the PE/EVA blends were prepared and their

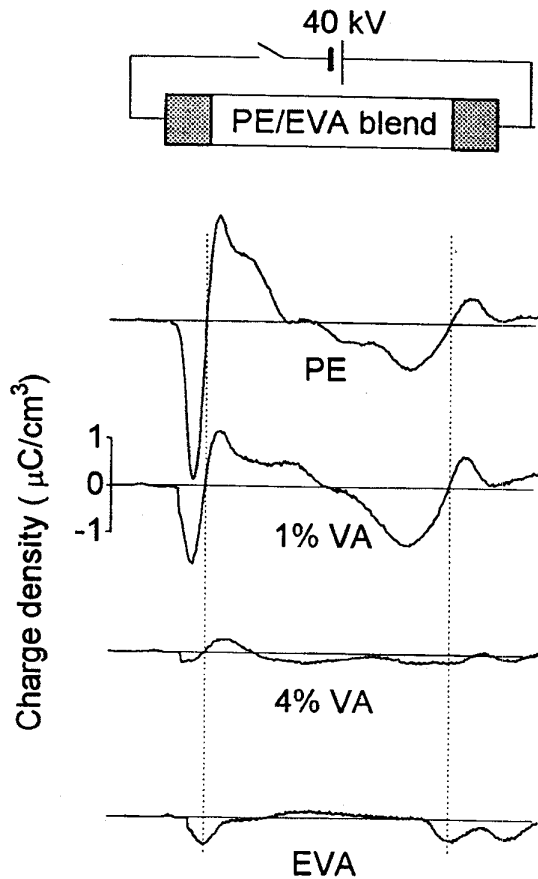


Figure 8. Space charge distribution of PE/EVA blends after the discharge (40 kV).

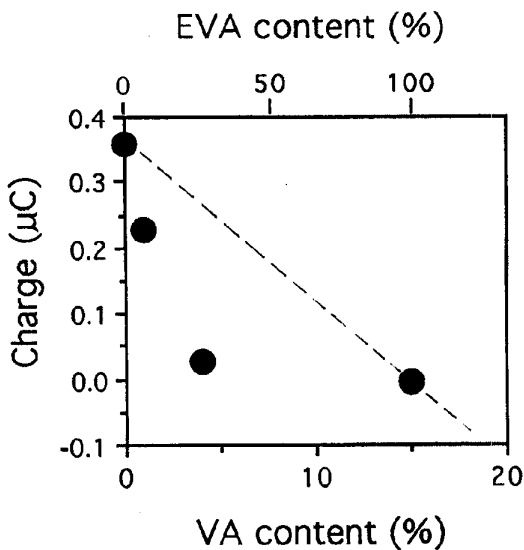


Figure 9. Charge near the cathode of PE/EVA blends: (●) experimental value, (---) additive rule.

charge distributions were measured. The results for the PE/EVA blends are shown in Figure 8 and the charge near the cathode was plotted as a function of VA content in Figure 9.

The blend containing 1% VA shows the formation of heterocharge with the magnitude of charge being small compared with that of pure PE. The shape of charge distribution of this blend resembles that of PE. The blend containing 4% VA shows only a small amount of heterocharge near the cathode and negative charge spread broadly in the bulk. A decrease of heterocharge near the cathode is seen more clearly in Figure 9, where the experimental values are compared with those expected by an additive rule which does not take the effects of interface between PE and EVA phases into account. As shown in Figure 9, the VA content dependence of charge near the cathode shows a negative deviation from a simple additivity rule based upon the values of PE and EVA.

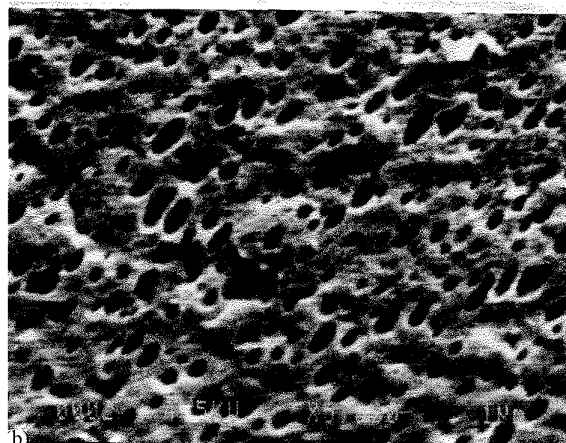


Figure 10. SEM pictures of cryofractured PE/EVA blends. (a) VA 1% (EVA 6.5%wt), (b) VA 4% (EVA 22%wt).

PE/EVA blends are known to have two morphologies depending on the EVA content. One is the 'sea-island morphology' which is found at low EVA content and the other is the 'co-continuous morphology' which is found at high EVA content. The former is also known to form the interpenetrating polymer network (IPN)

structure. Figure 10 shows SEM pictures of a cryofractured cross section of the blends. The EVA phase was removed by extracting in toluene at 50°C for 24 h [12]. It can be found that both blends, 1 and 4% VA, have the sea-island morphology, where the sea is the PE and the island is the EVA. In this situation, it is not conceivable that the charge dissipates through the EVA component because the EVA domain is surrounded by more resistive PE component. This Figure indicates that there are interfaces, acting as charge accumulating sites, between the PE and EVA phases. The interfacial charge will increase the local electric field in the PE component, as observed with the laminates.

It is assumed that the heterocharge decreases because the homocharge increases. There may be two possibilities for the increase of homocharge with the increase of VA content. The first one may be that the trapping of injected charge enhances as the VA content increases, resulting in a decrease of heterocharge. The other possibility may be that the increased electric field at the PE component in the blends due to the accumulated interfacial charge enhances the homocharge injection. This enhanced homocharge injection may neutralize the heterocharge. It was found from the laminates that the interfaces have a large amount of charge and this interfacial charge distorts the electric field in the direction to increase the field in the PE layer. The homocharge injection into the PE component may dominate due to this increased electric field. The same thing should happen inside the blend because it has interfaces between PE and EVA domains, although they are microscopic and randomly distributed. The data we see in the measured charge distributions of PE/EVA blends should be the averaged value because the PE and EVA components in the blends are located so closely.

4. CONCLUSIONS

The PE shows heterocharge while the EVA shows positive charge (holes) dispersed broadly in the bulk.

In the laminate samples, a large amount of charge accumulates near the interface, resulting in an increase the electric field in the PE layers. The electric field of the PE layer is higher when the EVA is located near the cathode than when the PE is located near the cathode.

When the thickness of EVA is much smaller than that of the PE layer, mainly negative charge develops in the PE layer, probably due to the enhanced charge injection which is again caused by the increased electric field in the PE layers.

The heterocharge in the PE/EVA blends decreases as the VA content of blends increases by the enhancement of homocharge

trapping at the interfaces and/or by the enhancement of negative charge injection via an increased electric field in the PE component.

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REFERENCES

- [1] K. S. Suh, J. J. Lee, J. H. Park and T. Takada, "Charge Formation in PE/polymer Laminates", Proc. CEIDP, Pocono Manor, PA, USA, pp. 203-208, 1993.
- [2] Y. Suzuoki, G. Cai, T. Mizutani and M. Ieda, "Effects of Interface on Electrical Conduction in Pe-eva Composites", J. Phys. D: Appl. Phys., Vol. 17, pp. 141-146, 1984.
- [3] G. Cai, "Evaluation of Internal Electric Field of PE/EVA Composite Systems by TSC-separation Methods", IEEE Trans. Electr. Insul., Vol. 25, pp. 702-706, 1990.
- [4] N. Hozumi, T. Okamoto and T. Imazo, "Space Charge Accumulation and Decay at the Interface between Polyethylene and Ethylene-vinylacetate Copolymer", Proc. 8th ISH, Yokohama, Japan, pp. 115-118, 1993.
- [5] K. S. Suh, J. J. Lee, J. Y. Kim and T. Takada, "Charge Formation in PE/ethylene-based Copolymer Laminates", Proc. 8th ISH, Yokohama, Japan, pp. 111-114, 1993.
- [6] T. Ditchi, C. Alquie, J. Lewiner, E. Favrie and R. Jocteur, "Electrical Properties of Electrode/polyethylene/electrode Structures", IEEE Trans. Electr. Insul., Vol. 24, pp. 403-408, 1989.
- [7] Y. Li, M. Yasuda and T. Takada, "Influence of Crosslinking Residues on Spatial Charge Distribution", Proc. 3rd ICPADM, Tokyo, Japan, pp. 1210-1213, 1991.
- [8] K. S. Suh, E. J. Kim, M. K. Han and T. Takada, "Charge Accumulation Characteristics in XLPE with Heat Treated Semiconductive Electrodes", Proc. 4th ICSD, Sestri Levante, Italy, pp. 418-422, 1992.
- [9] Y. Li, M. Yasuda and T. Takada, "Pulsed Electroacoustic Method for Measurement of Charge Accumulations in Solid Dielectrics", IEEE Trans. Diel. Electr. Insul., Vol. 1, pp. 188-195, 1994.
- [10] J. Brandrup and E. H. Immergut, *Polymer Handbook*, 3rd Ed., John Wiley and Sons, p. V-18, 1989.
- [11] K. S. Suh, J. H. Koo, S. H. Lee, J. K. Park and T. Takada, "Origin of Heterocharge in Polyethylene", Proc. 4th ICPADM, Brisbane, Australia, pp. 5-8, 1994.
- [12] I. Ray and D. Khastgir, "Correlation between Morphology with Dynamic Mechanical, Thermal, Physicomechanical Properties and Electrical Conductivity for EVA-LDPE Blends", Polymer, Vol. 34, pp. 2030-2037, 1993.

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