

# Electrodes and Charge Injection in Low-density Polyethylene

## Using the Pulsed Electroacoustic Technique

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### ABSTRACT

The effects of electrode materials on space charge formation in low-density polyethylene (LDPE) have been investigated experimentally using the pulsed electroacoustic (PEA) technique. Common electrode materials used in either the laboratory or power cable industry were selected, *i.e.* aluminum, gold and carbon loaded crosslinked polyethylene (XLPE), and space charge accumulation after the application of high electric stress was monitored. Experimental results demonstrated that charge injection processes take place in all cases once the applied stress has exceeded a threshold. However the amount of charge, and the polarity of the dominant injected charges showed a significant dependence on the electrode materials under the same applied electric stress.

### 1 INTRODUCTION

POLYMERIC materials used as insulators are progressively replacing traditionally insulating materials such as impregnated paper. The electrical performance of polymeric insulating materials is a subject that has presented an increased activity in the last two decades. The major advantages are their excellent electrical properties such as high breakdown strength, low dielectric loss and extremely low conductivity. Additional inherent advantages of polymers are possible due to the ability to modify the characteristics by altering the processing, thereby changing the molecular architecture (blending and doping are also commonly used methods). Similar to other engineering materials, they are required to be extensively characterized before utilization in practical applications, consequently the high stress conductivity and electrical breakdown of insulating polymer solids have received considerable attention and much literature exists on the topic [1]. There is general agreement that their low conductivity is attributable to the small number of charge carriers, of very low mobility, and these properties in turn are associated with the high trap density in the polymer. Trapped charges in insulating materials are known to distort the local electric stress and affect the high stress conductivity and breakdown phenomena.

Charge injection and transportation have been studied extensively

and discussed over the years, based on electrical conduction of polymers between metal electrodes. Experimentally, the conduction measurement is one of the simplest to perform on a polymer. There are, however, problems in the interpretation of such measurements. A wide variety of theoretical models such as Schottky injection [2], tunneling [3], Poole-Frenkel injection [4] and space charge limited current [5–6], have been proposed and tested with experimental results. Taylor and Lewis [7] used a modified form of potential barrier rather than the Coulomb one usually chosen in the Schottky injection. Three adjustable constants were adopted to generate a 'general' potential barrier with different magnitude and slope to satisfy experimental results obtained from polyethylene terephthalate (PET) and polyethylene (PE) films.

Although the above theories partially explain actual experimental results, they rarely give a complete and unequivocal description of the observed phenomena. The major difficulty arises, in the opinion of these authors, from the fact that there is little knowledge about the charge injection at the electrodes and trapping processes in the bulk.

In theory, the conductivity in a homogeneous material is defined by the equation

$$\sigma = \sum_{i=1}^n n_i q_i \mu_i \quad (1)$$

where  $n_i$ ,  $q_i$  and  $\mu_i$  are the concentration, charge, and mobility of the

$i$ th charge carrier.  $\mu_i$  is directly related to trapping and recombination in the bulk of the material.  $n_i$  is also dependent on the charge generation process and rate, *i.e.* injection from the electrodes or ionization/dissociation in the bulk. Charge carrier mobility controls the charge carrier density that can be transported through the sample when low injection potential barriers are present [8]. The potential barrier height is strongly influenced by polymer electroaffinity and ionization potential [9] and may limit the charge transport when its value is too high.

Early experimental work [7, 10–12] indicated that the electrode materials have a significant effect on the magnitude of the conduction current, activation energy as well as space charge distribution. It is generally accepted that the difference in conduction current is due to different work functions of different metals, thus different potential barriers. Different conduction mechanisms also have been proposed based on the results obtained from different electrodes. It is obvious that a measurement of  $\sigma$  does not enable one to separate the number of charge carrier, their mobility and their sign. And even worse, the charge carrier concentration is often dependent on the electrode materials, the effective stress, temperature and charge carrier mobility.

Under laboratory conditions, electrodes such as gold or aluminum were sputter-coated or evaporated on the samples so that an external voltage could be applied to the sample. It is believed generally that metallic electrodes provide a good contact. However different results are often obtained and in some cases different conclusions are reached. In the literature, this discrepancy is generally attributed to the charge trapping in the bulk. Different electrode materials often are used in practical applications. For example, crosslinked polyethylene (XLPE) is widely used for insulation in power cables due to its excellent electrical properties, in order to 'smooth' the electric stress profile and achieve a good bonding between the XLPE and the inner conductor and the screen, XLPE loaded with carbon black (often called semicon) is used. The electrical properties derived from the laboratory measurements may be very different from those in practice. Therefore it is important to examine the semicon effect on the charge formation and trapping in the bulk of the insulation.

Recently the development in mapping the space charge in insulation materials has made measurement of the space charge distribution across the material possible. The two most important techniques utilized are the laser induced pressure propagation (LIPP) [13], and pulsed electroacoustic (PEA) methods [14]. The techniques allow the electrode effect on charge injection to be examined as well as the trapping characteristics.

It has been reported [11, 12] that space charge distributions in PE strongly depend on additives, crosslinking byproducts and oxidation products. They also show a strong influence on electrode materials, the applied field and temperature. In the present study the effect of three different electrode materials *i.e.* Au, Al and semicon (Sc) used in extruded power cables, on the charge injection has been investigated using the PEA technique. By using different combinations of three electrodes, the individual effect of electrode material on electron injection and hole injection has been examined in additive-free LDPE.

## 2 EXPERIMENTAL DETAILS

LDPE was chosen initially because its simple chemical structure and its wide applications. In order to reduce the influence of the impurities such as antioxidants and crosslinking byproducts on space charge formation, additive-free LDPE is selected in the present study so that the effect of electrode materials on charge formation can be examined. The thickness of the sample is  $\sim 200 \mu\text{m}$ . Metallic electrodes with a diameter of 8 mm were either evaporated on the sample under a vacuum  $< 2.7 \times 10^{-3} \text{ Pa}$  (Al) or sputter-coated onto the sample (Au). In the case of semicon, a thin tape was peeled off from a commercial power cable; similar size to metallic electrode was cut from the tape and attached to the LDPE sample.

The sample was stressed at different voltage levels to 10 kV at room temperature for a given time. The space charge measurements were taken using the PEA system (PEANUT™, Five Lab), which has a pulse width of 5 ns. The sensor used is  $9 \mu\text{m}$  LiNbO<sub>3</sub> that enables the system to be heated to 90°C although not applied this time. The spatial resolution of the system is determined by the pulse width, acoustic speed in the material, and sensor thickness. In our system the resolution is  $< 10 \mu\text{m}$ , which is considered adequate for the present study. In this technique, a calibration is required in order to obtain quantitative space charge distribution. This is normally carried out at a low applied stress, assuming that there is no bulk charge present. The dispersion effect of the material is not considered in the present study because its influence in a thin film is not significant. Details about the PEA technique can be found in [14].

## 3 RESULTS

Although the space charge measurements were carried out both with voltage on and off, in most of the cases the amount of space charge in the bulk is not significant compared with the charge on the electrodes due to the applied voltage. Therefore the space charge distributions with voltage off are often presented to identify charge carriers. However, in order to demonstrate the influence of electrode materials on the speed of the charge packet, charge profiles with voltage on also are presented.

### 3.1 RESULTS FROM SAMPLES WITH THE SAME ELECTRODE MATERIALS

Figure 1 shows the space charge profiles obtained using the same material for both cathode and anode. 10 kV was applied to the samples for 30 min before its removal. With Al as electrodes it can be seen that the positive charge is formed across the sample with the maximum adjacent to the anode. Since the negative peak on the electrode is much broader than it should be, it could be considered that a small amount of negative charge has been formed adjacent to the cathode. In the case of Au electrodes similar positive charge distribution to the Al electrode is observed except there is no negative charge present close to the cathode. The amount of positive charge in the bulk is fractionally smaller when compared with the Al electrodes. With semicon electrodes the charge distribution becomes very different as shown in Figure 1. Both

negative and positive charges are observed. Positive charge is adjacent to the anode while negative charge distributes itself more broadly with the maximum close to the cathode. The amount and magnitude of both negative and positive charge are significantly higher than those observed in Au and Al electrodes.

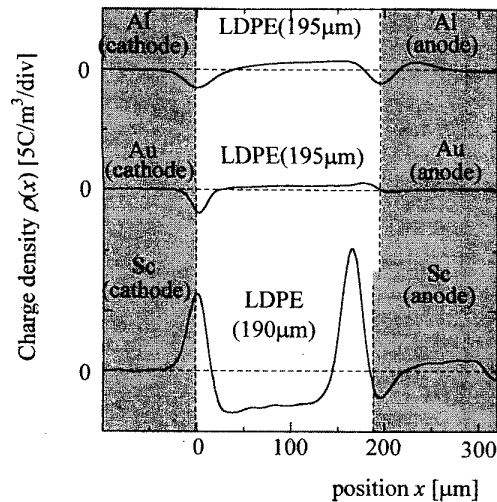


Figure 1. Charge distributions in samples with the same electrode materials after the removal of the applied voltage 10 kV.

### 3.2 RESULTS FROM SAMPLES WITH DIFFERENT ELECTRODES

Figures 2 to 7 show the space charge distribution obtained from the samples with differing electrodes. Figure 2 shows the results obtained from a sample with Al as the anode and Au as the cathode. Clearly positive charge is observed close to the anode. The magnitude is higher than those observed in the samples using either Au or Al as electrodes. On the other hand, if the electrode materials are reversed, positive charge is observed again close the anode as seen in Figure 3, but the magnitude is smaller. Another feature is a very small amount of negative charge adjacent to the cathode.

Figure 4 shows the space charge profile from the samples with Sc as the anode and Al as the cathode. A large amount of positive charge is found in the sample with its maximum in the middle of the sample. If the Al cathode is replaced by Au, the maximum shifts to the region adjacent to the cathode as shown in Figure 5. The maximum charge density in the latter is higher. If Sc and Al electrodes are reversed the charge distribution is quite different as shown in Figure 6. Here negative charge is developed close to the cathode (Sc) and positive charge close to the anode. The amount of the charge is smaller comparing with those observed with Sc as the anode. If Au replaces Al, only negative charge can be detected in the sample with its maximum close to the cathode as shown in Figure 7.

## 4 DISCUSSION

The results presented above clearly demonstrate that the electrode materials have a significant effect on charge injection. This will subse-

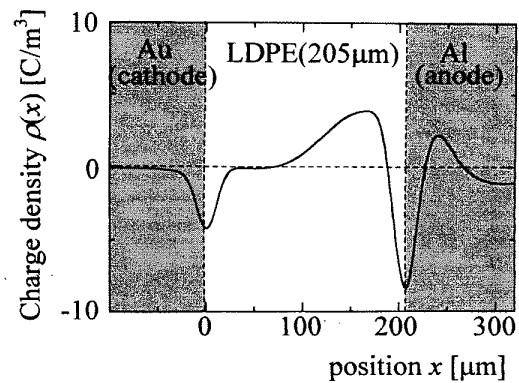


Figure 2. Charge distribution in Au(-)-LDPE-Al(+) after the removal of the applied voltage 10 kV.

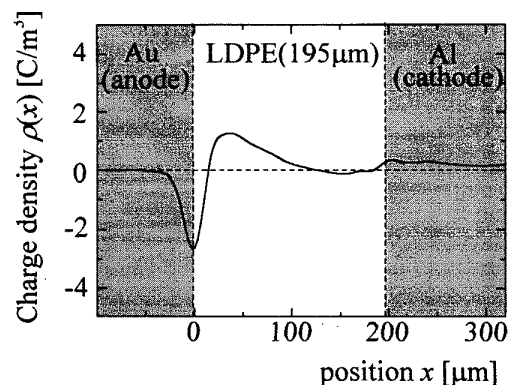
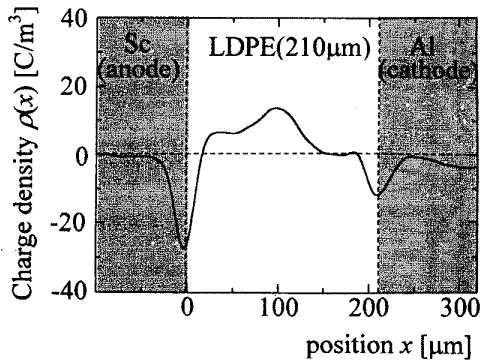


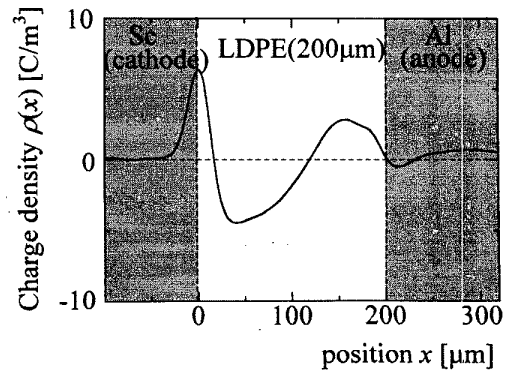
Figure 3. Charge distribution in Au(+)-LDPE-Al(-) after the removal of the applied voltage 10 kV.

quently affect charge trapping and transportation in the material. As mentioned earlier, the material used in this investigation is additive-free LDPE, and therefore the impurity effect should not be a major player in the charge generation. Bear this fact in mind; the charge formed in the sample can be attributed to charge injection from the electrodes. It is considered generally that at the electrodes, electron injection or extraction (hole injection) occurs by either thermionic emission over a stress-lowered potential barrier (so called Schottky emission) or quantum-mechanical tunneling through the barrier (so called the Fowler-Nordheim law). From the results obtained from the same electrode materials it is obvious that in the case of Al electrodes positive charge can be injected from the anode and electrons from the cathode. In the absence of ionization/dissociation in the bulk, charge distribution depends on the charge injection rates at both electrodes and the mobilities of charge carriers.

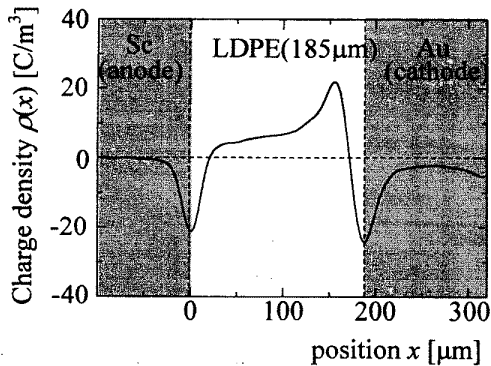
From the distribution it is evident that the process is dominated by positive charge. In the case of Au electrodes, it seems that positive charge can be injected easily from the anode as well. Once injected, the positive charge can travel through the bulk under the influence of the effective stress and distribute across the sample. It is also possible that



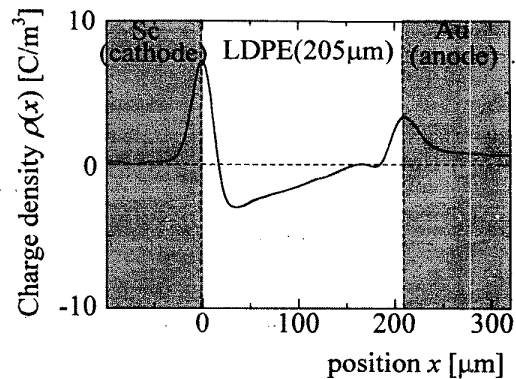
**Figure 4.** Charge distribution in Sc(+)-LDPE-Al(-) after the removal of the applied voltage 10 kV.



**Figure 6.** Charge distribution in Sc(-)-LDPE-Al(+) after the removal of the applied voltage 10 kV.



**Figure 5.** Charge distribution in Sc(+)-LDPE-Au(-) after the removal of the applied voltage 10 kV.



**Figure 7.** Charge distribution in Sc(-)-LDPE-Au(+) after the removal of the applied voltage 10 kV.

negative charge is injected into the sample from the cathode. However, the amount of negative charge is very small and is negligible compared with the amount of positive charge. Based on the amount of charge accumulated in the sample it indicates Al electrodes can inject slightly higher amount of holes than Au electrodes. In the case of Sc electrodes, two features are obvious from the charge distribution: the amount of negative charge in the bulk is larger than positive charge, indicating that electrons inject easier than holes from Sc; and second, electrons distribute broadly toward the anode while the holes are close to the anode, indicating the mobility of electron is higher than that of the hole. It is believed that the injected electrons move quickly through the bulk and meet the injected holes near the anode where they are recombined with the holes.

From the above results it clear that in the case of metallic electrodes hole injection from the anode is easier than electron injection from the cathode. It is also clear that hole injection from Al anodes is easier than that from Au anodes, because the amount of charge in the bulk is higher when the anode is Al. It may also say that the electron injection from Au cathode is extremely difficult.

In the case of Sc with either Al or Au electrodes the charge distributions are quite different from the pure metallic electrodes. In Figure 4

where the Sc is the anode and Al the cathode, the maximum positive charge occurs at the center of the sample. It is believed that the positive charge is injected from the anode. As mentioned earlier that Al cathode can actually inject electrons, the injected electrons tend to move toward the anode and neutralize with the coming holes. However, the amount of electrons is significantly lower than holes. As a consequence, the front of the positive charge approaches the cathode slowly. In order to confirm this the charge buildup during stressing is measured as shown in Figures 8 and 9 where two different stresses were used. Charge movement in a material is controlled by mobility and local electric stress or effective electric stress. The positive charges injected from the anode (Sc) move towards the cathode under the influence of the effective stress. The effective stress  $E_e(x)$  in the sample can be described using

$$E_e(x) = E_a + E_c(x) \quad (2)$$

where  $E_a = V/d$  and  $E_c(x)$  are the applied stress and the stress from the space charge distribution. The space charge distribution with voltage on, supports our earlier conclusion that electrons have a higher mobility than holes in LDPE because negative charge quickly reaches the anode. The drift velocity of charge carriers is controlled by effective stress, *i.e.* both the applied stress and the stress due to space charge.

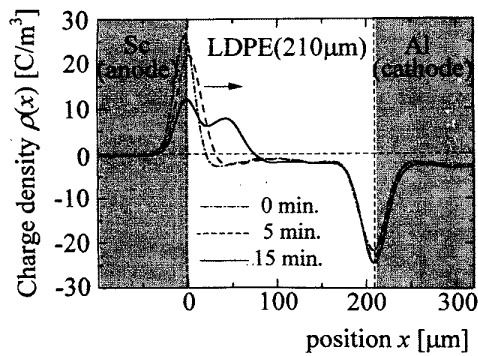


Figure 8. Charge distributions in Sc(+)-LDPE-Al(-) with the applied voltage 6 kV.

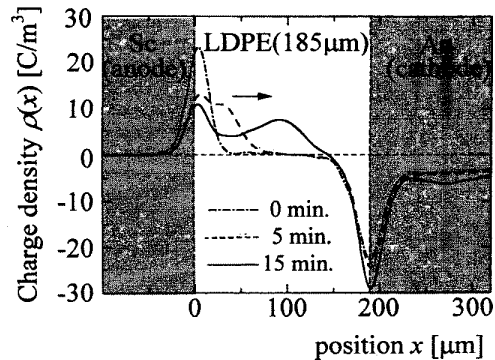


Figure 10. Charge distributions in Sc(+)-LDPE-Au(-) with the applied voltage 6 kV.

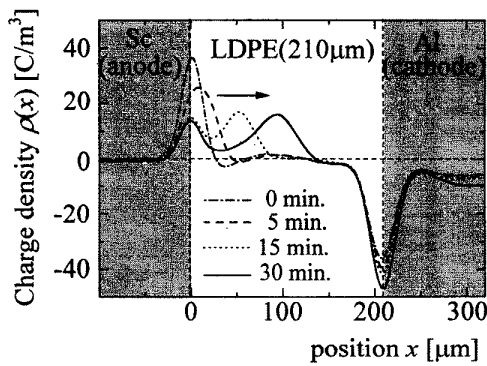


Figure 9. Charge distributions in Sc(+)-LDPE-Al(-) with the applied voltage 10 kV.

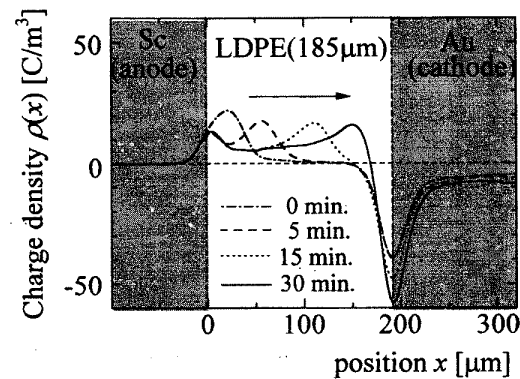


Figure 11. Charge distributions in Sc(+)-LDPE-Au(-) with the applied voltage 10 kV.

As more positive charge is injected into the bulk, the interfacial stress at the anode is reduced, and therefore the amount of the injected charge is small. Under the influence of the effective stress the injected positive charge front move towards the cathode. This is very much like the packet charge reported in the literature [15, 16]. The formation of packet charge generally is considered to occur above 100 kV/mm. However, in our case the stress is much lower than this. Our experimental results indicated the formation of packet charge could be formed at the applied stress as low as 30 kV/mm as shown in Figure 8. It is also clear that the under a lower applied stress, the movement of the positive charge front is slower than that under a higher applied stress, indicating that the speed of the positive charge packet is dependent on the electric stress. The speed of the positive charge packet depends on both injection rates at both electrodes and the mobilities of electrons and holes. The higher the stress the faster the charge carrier moves. If the Al cathode is replaced by Au, the maximum positive charge moves faster than in the case of Al cathode. This is illustrated in Figures 10 and 11. According to our previous argument, Au electrode injects very few electrons, therefore, holes injected from Sc move towards the cathode without encountering much recombination. Consequently, a higher charge packet speed is expected.

The estimation on the speed of the positive charge packet based on

Table 1. Speed of positive charge packet with different cathode electrodes and the applied stresses.

Parameter	Unit	Al cathode		Au cathode	
		6 kV	10 kV	6 kV	10 kV
$t$	s	900	900	900	900
$d$	m	46.1	49.8	91.1	107.2
$v_p=d/t$	m/s	$5.12 \times 10^{-8}$	$5.53 \times 10^{-8}$	$10.12 \times 10^{-8}$	$11.91 \times 10^{-8}$
$E_a=V/d$	V/m	$2.857 \times 10^7$	$4.762 \times 10^7$	$3.243 \times 10^7$	$5.405 \times 10^7$
$\mu=v_p/E_a$	$m^2s^{-1}V^{-1}$	n/a	n/a	$3.12 \times 10^{-15}$	$2.20 \times 10^{-15}$

the charge distributions can be carried out. From Figures 8 to 11, it is obvious that due to the presence of space charge in the bulk of the sample, the stress  $E_c$  is both position and time dependent, and so is  $E_e$ . Thus it is impossible to attempt to draw a relationship between the speed of positive charge packet,  $v_p$  and  $E_e$  from conventional measurements without knowledge of the space charge distribution. On the other hand as a first approximation, the  $v_p$  can be calculated using the applied stress and the results are summarized in Table 1.

Positive charge peaks at  $t=15$  min were used to indicate the position and the real thickness of the sample and the applied stress were adopted in the calculation. From the Table it is clear that the speed of the positive

charge packet increases slightly with the applied stress. In the case of Au cathode, since electron injection is negligible, the hole mobility can be calculated based on the mobility definition, *i.e.*

$$\mu = \frac{v}{E} \quad (3)$$

where  $v$  is the drift velocity of the charge carrier under the influence of the stress  $E$ . The calculated values also are shown in Table 1. It is noticed that the mobility actually decreases with the applied stress. Recent measurement of charge carrier mobility by Hozumi [17] in an ethylene-vinyl acetate copolymer film showed a similar trend for positive charge packets. This is contrary to expectations and further research is required. The range of mobility values obtained is in good agreement with that obtained from the transient space charge method [5] and surface charge decay method [18].

The results with voltage on confirm the analysis on space charge distributions obtained from the voltage off measurements. Compared with the results from Al cathode it suggests that the electron can be injected from Al electrode, but very little from Au electrode. The injected electron travels towards the anode and meets the injected hole on its way toward the cathode. As a result, recombination takes place. However, the amount of the injected holes from Sc is much larger than electrons from Al. Therefore un-neutralized holes keep moving towards the cathode. It appears that the speed of the positive charge packet is reduced as the recombination occurs. In the case of Au cathode, since there is very little electron injection, the positive charge packet seems to move faster.

When one examines the result from Sc to Sc the net negative charge presents in the bulk with the maximum adjacent to the cathode. Since the Sc can inject both holes and electrons, this leads to two conclusions to be drawn. First, under the same applied stresses Sc can inject more electrons than holes; second, the mobility of electrons is higher than that of holes. On the other hand the packet charge only occurs when hole injection is sufficient and dominant. In Figure 6 where Sc is the cathode and Al the anode, and comparing with the result from Sc to Sc it is clear that there is hole injection from Al but the amount of positive charge is smaller than that injected from Sc. From Figure 7 one can see that there is no positive charge in the bulk even close to the Au anode. This suggests that although there is hole injection from the anode the amount of positive charge is significantly small, holes can be recombined easily with fast electrons from the cathode.

It becomes clear now that the electrode material has a significant effect on the charge injection. When one measures the conduction current and subsequently determines the conduction mechanisms, the effect of electrode material used has to be taken into account. In our opinion the lack of understanding of electrode effect and charge trapping characteristics are the two main reasons for the discrepancy of conduction mechanisms and activation energies occurring in the literature. For example, Al can inject both electrons and holes while it is difficult to inject electrons when using Au as the cathode. The differing mobilities for electrons and holes could lead to different activation energies and even the mechanism, because in the case of Al electrodes the recombination also occurs. In the case of Sc electrodes a large amount of charge can be injected from both electrodes, leading to a significant amount of charge buildup in the material. The formation of space charge produces its

own electric stress distribution which will affect the charge injection process at the electrodes (charge injection will suppress the injecting field).

In our present investigation into the effect of electrode materials on conduction and trapping, additive-free LDPE has been selected to avoid complication introduced by additives to the conduction process. In practice polymeric materials such as XLPE antioxidants and crosslinking agents are added deliberately, this may play an important role in conduction current and trapping characteristics. Their influence on conduction can be considered in two ways: ionization of additives which will contribute to the conduction current directly; and also introducing traps which will affect the mobility of either electrons or holes injected from the electrodes. Our space charge results agree well with those reported by Mizutani [12] where similar electrode materials were used, while Sanden [11] showed different space charge results on degassed XLPE. Authors believe that the composition of the semicon may have a strong influence on charge injection. Our recent work [19] on the influence of different semicon and their combinations with different insulation on space charge formation shows a strong evidence to support our argument.

Previously the different values of conduction current of PE using different electrodes have been explained based on the work function of metals. In the case of Al and Au as electrode, it is known [20] that the work function of Au ( $4.70 \pm 0.02$  eV) is higher than Al ( $4.08 \pm 0.05$  eV) therefore the potential barrier of Al/LDPE interface is lower than that of the Au/LDPE interface. As a result, there should be more injection from Al than Au, leading to a higher conduction current. Based on this argument, the Sc used in the present study is carbon loaded XLPE and should have a very similar structure to the LDPE from the energy-band point of view. Consequently, the potential barrier between them is negligible, so the injection should be much easier than from the metal electrode. The amount of charge trapped within the bulk of the sample validates the argument.

Recently numerical techniques [21, 22] have been employed to simulate the charge distribution in polymers, qualitative agreements were obtained. We believe that by considering different potential barrier heights caused by different electrodes, different mobilities for electrons and holes and different applied voltages, it may be possible to model the observed phenomena. Work using numerical modeling is in hand and results will be reported at a future date. However, quantitative analysis of the results needs a full understanding of the energy diagram of LDPE especially the surface states of the LDPE.

## 5 CONCLUSIONS

THE effects of electrode materials on the charge trapping characteristics in low-density PE have been investigated. From the results and discussions, it is clear that the following conclusions may be drawn.

The electrode material has a significant effect on the charge injection therefore on the trapping characteristics of LDPE. From the carrier injection point of view the results indicate that for electrons the order is as follows: Sc>Al while Au injects very little; for hole the order is Sc>Al>Au. The injection rate in the case of Al electrodes is hole>electron while with Sc electrodes the injection rate is electron>hole.

From space charge profiles it is apparent that the mobility of electron is much faster than that of hole in LDPE. Positive space charge packets have been observed at a stress much lower (as low as 30 kV/mm) than previously reported. They can be observed only when the positive charge is the dominant charge carrier. The injected electrons from Al cathode can neutralize the front of the packet and as a consequence the speed of the positive charge packet is reduced.

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