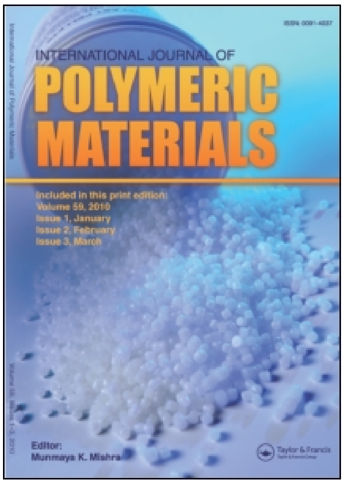

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Influence of Iodine on the Distribution of Space Charge in LDPE

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Influence of Iodine on the Distribution of Space Charge in LDPE

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The effect of iodine doping on space charge distribution in LDPE has been investigated using a probe technique. The results indicate that iodine dramatically reduces the amount of space charges formed and changes their distribution pattern when compared with plain LDPE. Whereas in plain LDPE the distribution is of the homocharge type, in iodine-doped LDPE the distribution is of the heterocharge type with negative charges predominating. X-ray diffraction patterns show that the iodine has no influence on the crystallinity of the polymer.

INTRODUCTION

When insulating polymers are subjected to an electric field charges become trapped in the bulk material. Such space charges are long-lived and cause a redistribution of the potential in the gap between the electrodes so that the actual field to which the polymer is subjected will differ markedly from the geometric field. This will influence both the conduction and breakdown properties of polymers especially when measurements are carried out using direct voltages.

Indirect evidence of the existence of space charges in PE has been obtained from the effect which dc prestressing has on its impulse breakdown strength[1] as well as on thermally stimulated currents.[2] From a practical point of view Sekii[3] has reported that the direct breakdown voltage of XLPE cables after polarity reversal is about 70% of the breakdown value without prestressing. For the direct experimental determination of the distribution and type (homo or hetero) of charges two methods have been used, that of dust figures[4] and that using an electrostatic probe.[5] The latter method was used in the present work.

In order to gain further insight into the nature and origin of charge carriers as well as the charge-transport mechanism in polymers, several workers[6,7,8] have used iodine as a dopant and determined its effect on conduction, mobility and breakdown using different techniques. The probe space charge measurements reported in the present work indicate that the presence of iodine in PE produced a significant change in the space charge distribution. X-ray diffraction patterns showed that iodine has no effect on the structure of PE.

EXPERIMENTAL DETAILS

Test specimens were of LDPE of 0.92 g/cm^3 solid cylinders of 36 mm diameter. Two hemispherically tipped stainless steel electrodes of 20 mm diameter were inserted from both ends in such a way that a distance of about 4 mm existed between their tips. Doping with iodine was carried out by pressing resublimed iodine at the electrode sites and allowing it to diffuse into the polymer for 30 hs at 60°C. The concentration of iodine was estimated to be about 20% by weight.

Specimens were placed in a perspex test vessel and immersed in silicone oil. A 100 kV direct voltage was applied between the electrodes for 48 hs. The voltage was then removed and the electrodes were short circuited before the specimen was taken out and the electrodes removed. Immediately after, specimens were cut along the cylinder axis into two identical halves using a clean sharp-edged knife. The cut surfaces were scanned using a capacitive probe; the probe voltages recorded are a measure of the effective surface-charge density.[9] Measurements on both cut halves gave similar results; moreover, for a given specimen there was no appreciable change in the measured potentials for periods of more than three hours at room temperature after voltage removal. Different specimens tested under identical conditions gave highly reproducible results. Details of the probe technique used have been described elsewhere.[10]

RESULTS AND DISCUSSION

Figure 1 shows the variation of the probe potential along the electrodes axis after application of the voltage for 48 hs. Previous measurements[10] have indicated that the space charge distribution attains a steady state after almost 24 hs. The results indicate the formation of homocharges at both electrodes. The symmetry of the distribution shows that almost equal number of positive and negative charges are formed at both electrodes so that the region between the electrodes as a whole is neutral. The maximum density of space charges occurs at some distance from the electrodes indicating that the charges move away from the source electrode.

Figure 2 shows the variation of the probe potential along the electrode axis in iodine-doped LDPE obtained under exactly the same experimental conditions as those of Figure 1. It is evident that the charge distribution is completely different from that in undoped LDPE. There is now a net negative space charge extending from the anode well into the gap and a small region of positive charges in the immediate vicinity of the cathode. Also the density of the charges is almost seven times smaller than in the case of undoped LDPE.

Although the origin and nature of charge carriers in PE have not been unequivocally determined, conduction, mobility and TSC measurements by several workers[2,6,11] have led to a number of postulates which may be summarized as follows: (a) Injection of electrons at the cathode and of holes at the anode; (b) charge generation in the polymer itself by electron or hole activation from donor- or acceptor-like states within the forbidden gap; (c) the existence of shallow traps and of deep traps capable of long-term charge storage. The deep traps are possibly associated with the crystalline/amorphous interface and the existence of terminal vinyl (unsaturated) groups lying in the amorphous region.

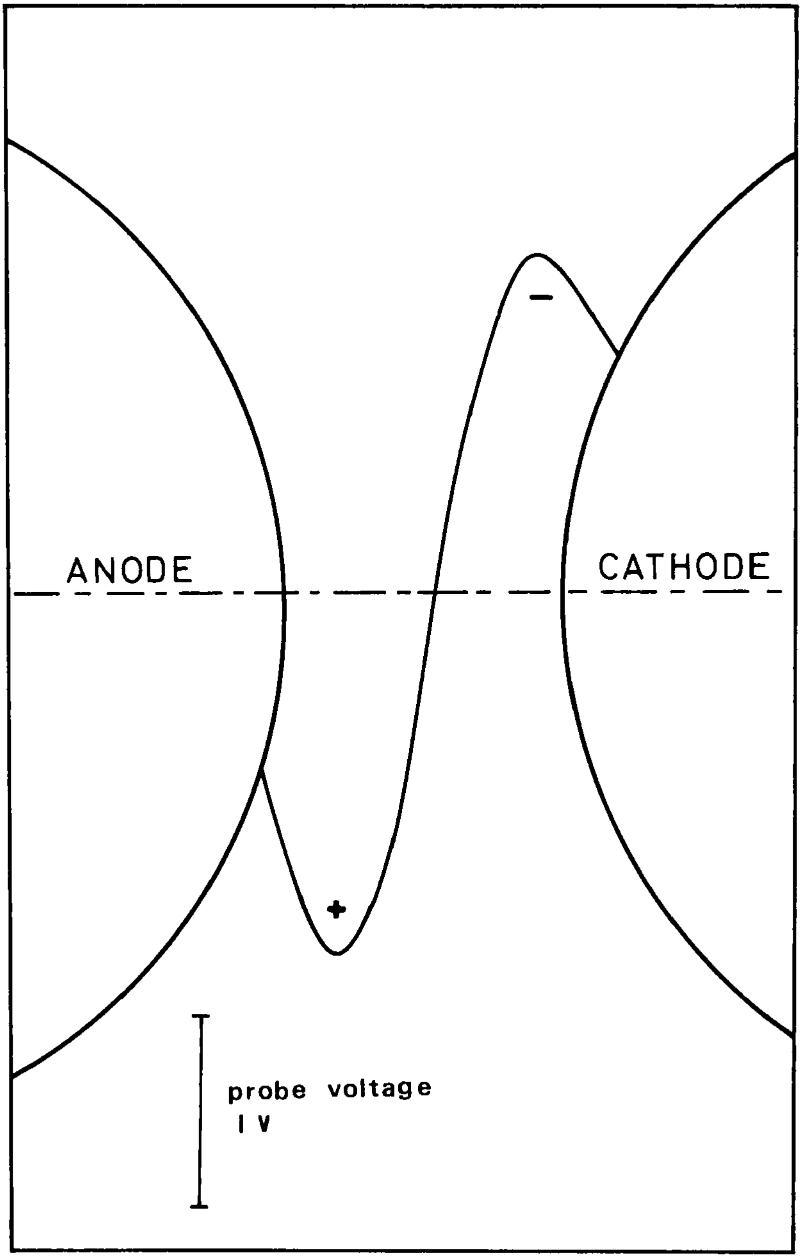


FIGURE 1 Variation of probe potential for plain LDPE. Stressing voltage, 100 kV.

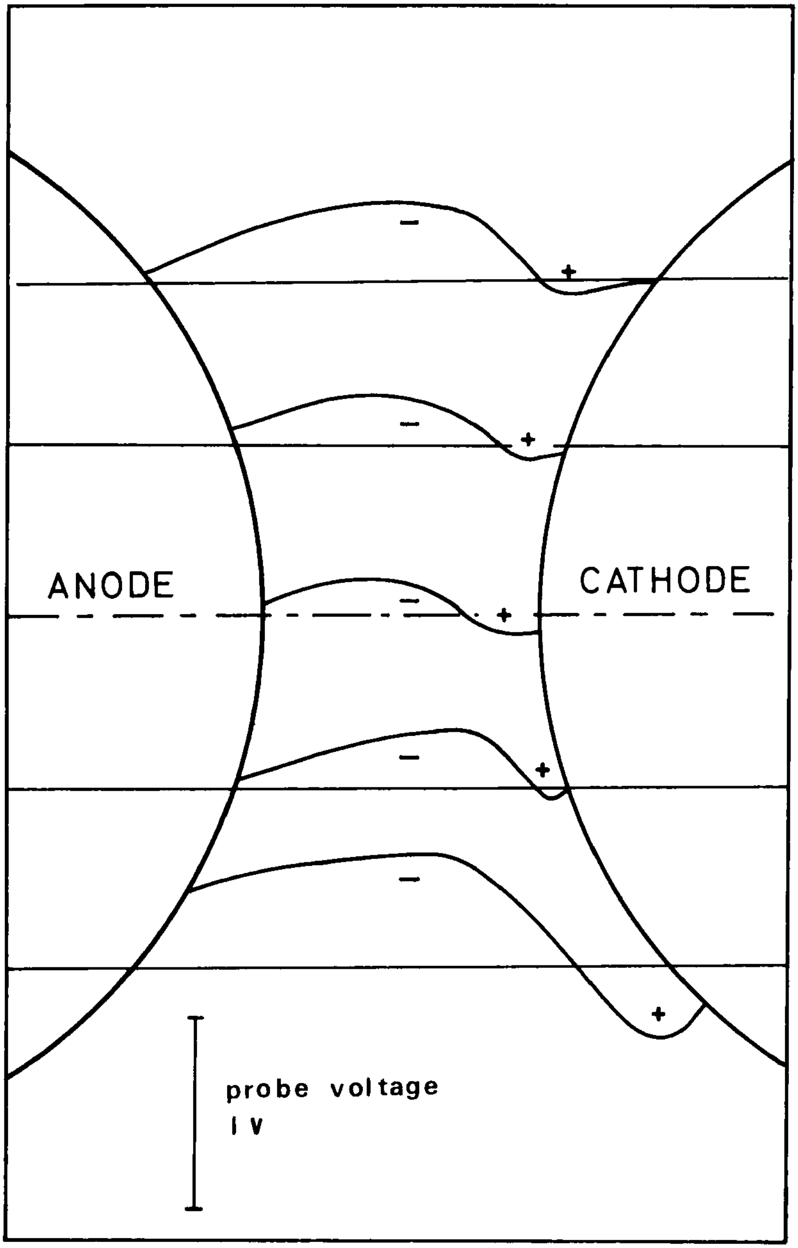
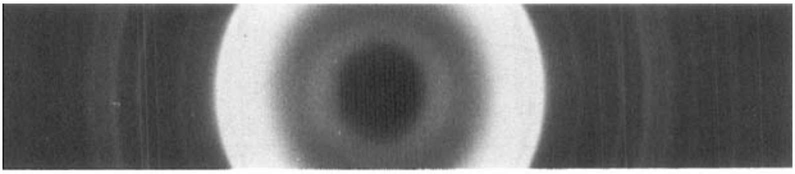


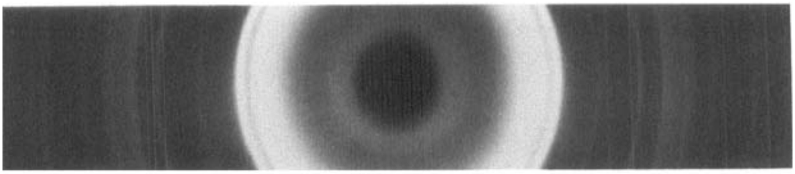
FIGURE 2 Variation of probe potential for iodine-doped LDPE. Stressing voltage, 100 kV.

Recent probe measurements by Lewis *et al.*[12] on charge transfer at metal-polyethylene interfaces have shown that the charges injected in the polymer have the same sign as that of the injecting electrode (homocharges) indicating electron transfer either to acceptor or from donor molecular states by quantum mechanical tunneling. It is evident from Figure 1 that the existence of homocharges will greatly reduce the field at both electrodes, but the field in the bulk material will be increased. However, in spite of this increased field the space charge distribution remains 'frozen in' indicating that the positive and negative charge carriers (whatever their origin) are captured in deep traps. It must be emphasized that space charge measurements give a resultant charge distribution in the sense that although positive space charge may be neutralized by a negative charge, recombination need not necessarily occur[12] and both types of charges may coexist.

X-ray diffraction of undoped and iodine-doped LDPE (Figure 3) show that the addition of iodine has no effect on the crystallinity of the material. Since it is well-known that polymers consist of crystalline regions and amorphous regions, the iodine will lie mainly in the amorphous region. Doping of PE with iodine has been found to increase the conductivity[6] and the charge mobility[8,11] by several orders of magnitude. Yoshino *et al.*[8] found that whereas in undoped



(a)



(b)

FIGURE 3 X-ray diffraction patterns. (a) plain LDPE; (b) iodine-doped LDPE.

polyethylene the electron mobility was higher than the hole mobility, in iodine-doped PE the hole mobility was larger than the electron mobility. Moreover they were able to differentiate between a slow carrier mobility associated with the amorphous region and a fast carrier mobility associated with the crystalline region. Doping with iodine was found to greatly increase only the slow carrier mobility which led them to conclude that the iodine enters mainly in the amorphous region. The present measurements indicate that the presence of iodine causes the space charge to change from a homocharge distribution to a heterocharge one, with the region between the electrodes dominated by negative charges.

Because of the electronegativity of iodine, electrons injected at the cathode will be preferentially attached to iodine molecules instead of being trapped at acceptor molecular states (traps) in the PE itself. At the anode the presence of iodine will reduce the net amount of positive charge by capturing electrons from donor molecular states. The overall decrease in the net amount of space charges formed in the presence of iodine may be attributed to an increase in the charge carrier mobility. With the iodine lying mainly in the amorphous region, two possibilities are envisaged for such an increase: (a) Iodine enters the polymer as neutral iodine molecules some of which form charge-transfer complexes with the polymer at sites of unsaturation (terminal vinyl groups) lying outside the crystalline region.[11] (b) Iodine helps to bridge the carrier transport between crystalline parts by reducing the number of deep traps associated with the crystalline/amorphous interfaces.[13]

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