EFFECT OF ELECTRODES ON SPACE CHARGE IN CROSS-LINKED POLYETHYLENE UNDER DC FIELD

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Abstract— Space charge behaviour at physical interfaces in cross-linked polyethylene (XLPE) films submitted to DC field has been investigated as a function of the nature of the interfaces using the pulsed electro-acoustic technique. Aluminium, gold and carbon black-loaded polyethylene were used as electrodes to investigate space charge built-up in a single dielectric layer. Charge injection rather than ionic species migration is seen to control space charge distribution in agreement with the storage conditions of the XLPE films. It is shown that the sign and distribution of space charge depend drastically on the nature and polarity of the electrodes

Index Terms — Space charge, dielectric interface, pulsed electro acoustic technique.

1 INTRODUCTION

THERMOPLASTIC materials are more and more used in Electrical Engineering as high voltage insulation. They are highly insulating media and exhibit excellent physical and chemical properties, with the possibility to be processed to comply with complex system geometry. Among them, polyethylene is the most well-known and is widely used as insulation in power cable industry. Although its formulae appears very simple, polyethylene is a generic term to name insulating materials with very different characteristics through different base resins (resin type, chemically modified or not, cross-linked or not) and formulation (antioxidants, additives, etc.) [1]. Severe constrains still limit its use in HVDC applications where electrical charges can be injected at electrode/insulation contact or generated within the bulk by dissociation of species and other electro-chemical reactions. These charges disturb the field distribution and could lead to critical field enhancement especially when the polarity of the line needs to be inverted [2]. Another important point is the need of junctions and terminations in cable systems. They have been identified as weak points with a failure probability much higher than the cable itself due to the presence of dielectric/dielectric and electrode/dielectric interfaces [3,4].

Whatever is the nature of interfaces, they play a fundamental role in the dynamic of space charge because they have specific electrical properties due to the physical and chemical disorders inherent to their nature. In the case of metal/dielectric interfaces, one generally considers that charge carriers of both signs can be injected and/or extracted from the contact depending on its nature [5, 6]. However, a correct description of charge injection from a metal into insulation using the classical injection laws (Schottky emission for example) has not been provided yet [5, 7].

Our objective in this study is to investigate the time dependent spatial distribution of space charges at metal/dielectric interfaces submitted to DC field, using the pulsed electro-acoustic method.

2 MATERIALS AND EXPERIMENTAL

2.1 SPECIMEN CHARACTERISTICS

Peelings of cross-linked polyethylene of the series described in reference [8] were used for this study. They were peeled from a medium voltage (63/90 kV) cable having a 14 mm thick insulation using a lathe equipped with a special designed knife to get optimum surface smoothness. They have a nominal width of 8 mm and a thickness of 150 μm. A thermal treatment was carried out at a temperature of 50 °C in order to get rid of the by-products of the cross-linking reactions. Investigations were made of the surfaces of the peelings since these might affect measurements. Because of the cutting procedure, the surfaces were not as smooth as if compression-moulded specimen had been used. Measurements using an atomic force microscope (AFM) indicated the two surfaces of the peelings have a different surface roughness. The typical and maximum peak-to-trough heights were measured and the RMS roughness estimated as

indicated in table 1. It was verified that the difference in surface roughness does not affect charge injection and charge storage at field value < 40 kV/mm. A full characterization of the physical, chemical and electrical characteristics of the samples can be found in [8].

Tableau 1 : Atomic force microscopy measurements of roughness of the peel faces

PEEL FACE	TYPICAL PEAK TO TROUGH HEIGHT (μM)	MAXIMUM PEAK TO TROUGH HEIGHT (μM)	RMS ROUGHNESS (NM)
SMO OTHER	~0.3	~1	~150
MAT	~1	~2	~350

2.2 EXPERIMENTAL SET-UP

The Pulsed Electro-Acoustic (PEA) apparatus for space charge measurement is particularly appropriate for investigating electrical properties such as injection, trapping and transport of electrical charges [9] because measurements can be easily performed under voltage and in volt off. The PEA method relies on disturbing the equilibrium between electrostatic and elastic forces in the material. The application of a rapidly varying voltage to the sample upsets this equilibrium causing elastic forces to react, which in turn generates elastic waves. By placing a transducer adjacent to the sample, some of these elastic waves are converted into an electric signal. The time dependence of this signal is directly related to the distribution of charges. The method has been applied to many different shapes (planar structures, coaxial geometry), and different systems have been developed with specific aims (3D imaging, fast temporal measurement, etc).

In this study, we used a conventional system where the sample is sandwiched between a thick aluminum electrode in contact with the piezoelectric sensor and a semi-conductive (SC) electrode (carbon black-loaded polyethylene film) to which is applied the voltage pulse of the PEA and the DC voltage for poling the sample under test. Measurements were performed at temperature (25 °C). The sensitivity and the spatial resolution of the measurement are of the order of 0.1 C/ m^3 and μm respectively.

2.3 MEASUREMENT PROTOCOL

The influence of the nature of the electrode/dielectric interface on charge built-up was investigated using samples with symmetrical and asymmetrical electrode systems.

Au/XLPE/Au and SC/XLPE/SC structures were prepared by sputtering gold on both sides of the peel and by pressing two semi-conductive electrodes in contact with the film, respectively. The Al/XLPE/SC structure corresponds to the usual PEA measurement configuration. A 31 kV/mm field was applied to the samples for 5 min. PEA measurements were performed during polarization and under short-circuit conditions where the charge accumulation is evidenced more easily taking into account the small space charge density (of the order of 1 C.m^{-3}) and the high value of the capacitive charge during voltage application. At this field level, charge injection is known to dominate the charge accumulation in XLPE when by-products of the cross-linking reaction have been eliminated (hetero charge is usually observed in XLPE in the presence of by-products versus homo charge after thermal treatment –see reference [10] for example). Moreover, space charge accumulation within a sample always results from a three steps process, being charge generation, transport and extraction. We have therefore chosen a short poling time of 5 min in order to reduce charge transport. In such conditions, the accumulated charge is thought to picture charge injection processes.

3 RESULTS AND DISCUSSION

Charge distribution in polarization in and depolarization are shown in Figures 1 to 3 for the three different electrode arrangements. Due to the small amount of injected charges (in the rage of C.m^{-3}), the profile under polarization does not evidenced space charge clearly due to the predominance of the capacitive charge on the electrodes (except in Figure 2-a where negative injection is stronger) . The space charge is evidenced in depolarization. The following comments can be made

-Au/XLPE/Au structure (see Figure 1): negative charges are injected from the cathode and accumulate up to 50 μm away from this electrode. There is no evidence of positive injection from the anode.

-SC/XLPE/SC (see Figure 2) : negative charges are injected from the cathode and accumulate up to the anode forming a hetero charge layer with reference to the later. There is no evidence of positive charge injection from the anode, although it could be hidden by the negative charge.

-Al/XLPE/SC (see Figure 3): when the cathode is the Al electrode, negative charge are seen to be injected and migrate up to the anode forming a hetero charge layer in the vicinity of the later. When the anode is the Al electrode, two homo charge domains are observed in the vicinity of both electrodes (not shown).

A quantification of the negative space charge q(t) accumulated in the bulk at t=5 min (end of the polarization phase) and of its decay time in depolarization has been done integrating the value of the measured charge q(x,t) over insulation thickness, as follows :

$$q(t) = \frac{1}{L} \int_0^L |q_p(x,t)| dx \qquad (1)$$

where 0 denotes the cathode position and L the distance of negative charge penetration. Induced or capacitive charges at the electrodes are not taken into account. Results are shown in Figure 4. As regards electronic injection, the results give the following ranking among different electrodes: SC > Al > Au.

As regards holes injection, the ranking is as follows: Al > Au. For SC electrode, it was not possible to conclude due to the extension of the negative charge that can hide positive injection. However, it can be noted that SC(-) is much more injecting than SC(+).The general trend indicates that Au electrodes are weakly injecting, but are able to inject electrons and holes. This has been reported in several papers [11] and could be in relation with the high work function of gold (≈ 4.7–[12]) located mid-gap within the band gap of polyethylene (8.8 eV –[13]).

The Al electrode (≈ 4.08 eV –[12]) has a lower work function which can explain a higher injection for negative carriers as compared to gold. However, Al is also more injecting for positive carriers than gold [11] which is somehow surprising.

Note than a direct relationship between electron work function and space charge has not been demonstrated so far for organic materials. Anode material was shown to influence hole injection in charge transport layers but a proportionality relationship between the two was not observed [12]. Intense work has been done in semi-conducting organics used in light-emitting diodes [14].

It was demonstrated that complex formation at the polymer-metal interface causes the barrier height not to be highly sensitive to the work function of the metals used as electrodes.

Defects states at the interfaces could pin the Fermi level. In large band gap materials all these factors can explain that the work function is not highly influential in charge injection. Interfacial states due to physical and chemical disorders at the contact could be the controlling factors, without a straightforward relationship with the work function itself [5-7]. Another explanation can be that the injection is through localized states in the band gap, not over the barrier at the contact. This also would smear out the relation between space charge accumulation and work function.
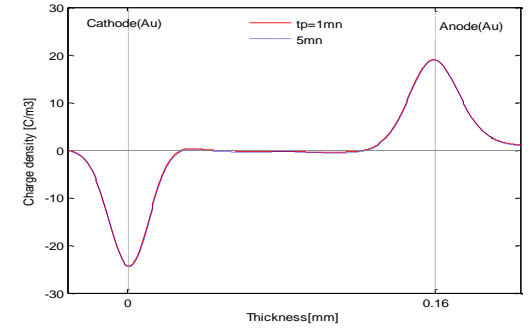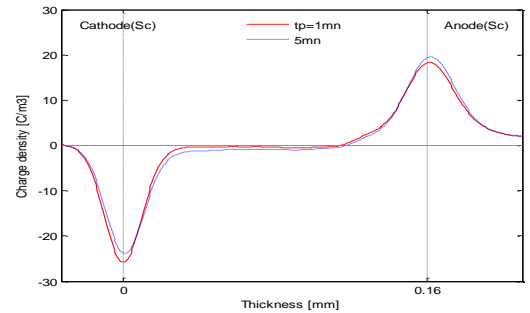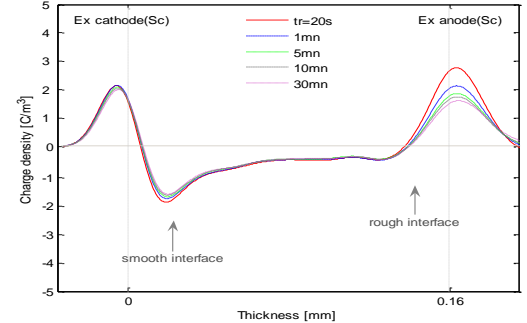

(a)-Polarization (volt on)
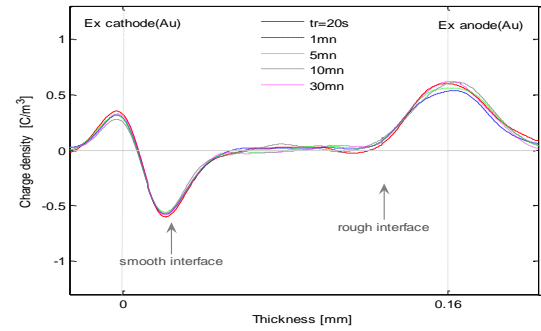
(b)-Depolarization (volt off)

Fig.1. Space charge profiles in Au-XLPE-Au (a)-Polarization (31 kV/mm, 5 min) (b)-Depolarization (starting at time t=5min)
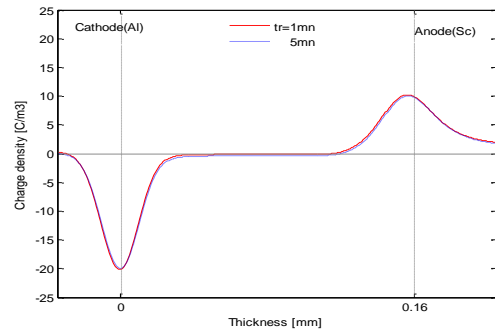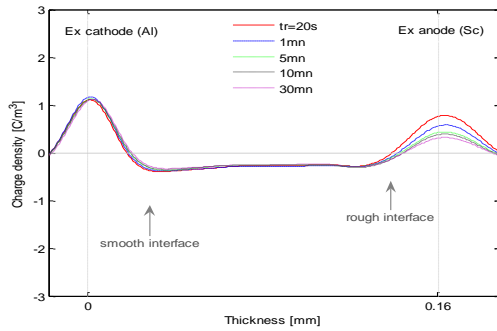

(a)-Polarization (volt on)

(b)-Depolarization (volt off)

Fig.2. Space charge profiles in SC-XLPE-SC (a)-Polarization (31 kV/mm, 5 min) (b)-Depolarization (starting at time t=5min)

(a)-Polarization (volt on)

(b) Depolarization (volt off)

Fig .3. Space charge profiles in Al-XLPE-SC (a)-Polarization (31 kV/mm, 5 min) (b)-Depolarization (starting at time t=5min)
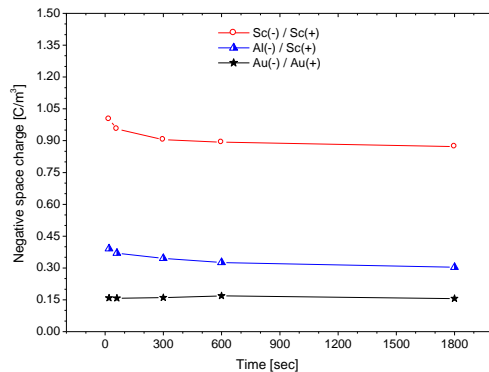

Fig.4. Amount of negative charge within the bulk of an XLPE sample submitted to a field of 31 kV/mm for 5 min for different electrode systems as indicated. The accumulated charge was calculated according to equation (1).

Finally, SC electrodes are injecting electrodes in comparison with Au electrodes which has also been reported [11]]. Since the semi-conductive electrodes are PE-based electrodes, one can think that the good matching between the molecular network help charge injection from the carbon black particles. A recent report underlines the extreme sensitivity of space charge accumulation versus the nature of both semi-

conductive electrode and polyethylene formulation [14].This advocates for the importance of the contact on charge injection but prevents simple generic explanation.

CONCLUSION

We have investigated the charging behavior of electrode/XLPE interfaces using the pulsed electroacoustic technique. XLPE peels were free from cross-linking by-products. Accordingly, the space charge distribution was dominated by injection from the electrodes. The charge accumulated in the dielectric depends on the nature of the electrode, being gold, aluminium and carbon-loaded polyethylene. Aluminum and gold electrodes were able to inject electrons and holes but the injection rate is much lower for gold. Semi-conductive electrodes were the most efficient injector for electronic carriers. Although some relationship can exist between the metal work function and the injection rate for electrons, it does not provide an explanation for hole injection. Interfacial states are probably taking over the work function in controlling charge injection.

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