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# Ion generation and transport in low density polyethylene under electric stress

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**Abstract**—Synthetic materials such as cross-linked polyethylene (XLPE) are increasingly used as electrical insulation in HVDC power cables, compared to oil impregnated insulation. However, because of their dielectric properties, these materials can accumulate charges in the insulation, generated by dissociation of species or injected at the electrodes. This accumulation phenomenon can cause an increase of the electric field which can lead to the dielectric breakdown of the material, meaning thereby stopping the distribution of electricity. It is therefore important to understand these charge accumulation mechanisms for HVDC application, and particularly the role of ions, generated by dissociation of by-products of cross-linking agents, that can transport and take part to space charge accumulation. In this goal, we have considered a simple model material consisting of low-density polyethylene (LDPE) films soaked in cumyl-alcohol, a by-product often encountered in peroxide-crosslinked XLPE and thought to dissociate under field to provide ionic carriers. The impact of the soaking time, electric field and temperature on space charge density profiles recorded using the Pulsed Electro Acoustic method (PEA) and on charging current are presented and compared to those obtained on unsoaked LDPE samples.

**Keywords**—space charge, low density polyethylene, ions, transport, cumyl alcohol

## I. INTRODUCTION

The increasing demand in energy implies a fast development of the electrical transport network over long distances across different countries. In these conditions, high voltage direct current (HVDC) links are now preferred over alternative current (HVAC) ones. Thanks to its electrical and mechanical properties, cross-linked polyethylene (XLPE) has become the challenging material in electrical insulation for HVDC cables. However, this material can store charges when submitted to an electrical stress, increasing the electric field locally, and potentially leading to the ageing of the material or its dielectric breakdown [1]. These charges can originate from injection processes at the electrodes or from dissociation of species in the volume of the material. Carriers generated by ionization can be electrons or holes that can travel across the material and be extracted at one electrode. However, generated carriers can also be ions that can also travel if not too big, but cannot be extracted at the electrode. These ions will accumulate inside the dielectric, giving rise to an increase of the electric field that will disturb the applied field. Polyethylene cross-linking agent, dicumyl-peroxide (DCP), is known to lead to by-

products such as acetophenone, alphas-methylstyrene, or cumyl alcohol, that are thought to dissociate when submitted to an electric field [2]. This has already been put forward [3, 4], when polyethylene has been soaked in these different by-products in order to see the impact of their presence on space charge measurements mainly. In these works, space charge measurements were performed fields, beyond 25kV/mm, and the authors concluded that cumyl alcohol introduces the highest amount of charges. In the present paper, we focus on the role played by cumyl alcohol only on space charge accumulation. Applied fields are low compared to previous studies [3- 5], in order to better understand the way ions can be generated in the material.

## II. SAMPLE PREPARATION

Low density polyethylene (LDPE) has been chosen as it is the base material for XLPE, and it does not contain any by-products or additives able to dissociate. LDPE samples of thickness 250 to 300  $\mu\text{m}$  have been prepared, and then soaked in liquid cumyl alcohol at 70°C, the melting temperature of cumyl alcohol being of 32°C. This temperature of 70°C has been chosen as it remains low compared to the melting temperature of LDPE, which is of the order of 110°C. Hence, the chemical structure of the sample should not be changed due to the soaking temperature. Fig. 1 presents the variation of the sample weight (%) as a function of the soaking time. The absorption rate is high for the first hour of soaking, while it increases slightly from 2 to 8 hours.

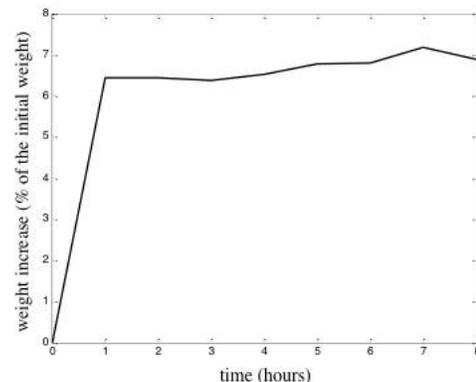


Fig. 1. Weight monitoring of a LDPE sample soaked in liquid cumyl alcohol at 70°C.

The weight increase is comparable to what has been obtained previously by different authors [3, 4], although the soaking time is longer in our case compared to what has been set in the literature. This is mainly due to the temperature during the soaking time, which is lower in our case (70 vs. 80°C).

Space charge measurements and current measurements have been performed on pure and soaked LDPE samples for different applied electric fields and different temperatures. Space charge measurements were recorded using the Pulsed Electro Acoustic method (PEA). For space charge and current measurements, no electrodes have been deposited on the sample to avoid cumyl alcohol evaporation during processing of the electrodes. Hence, for PEA measurements, the top electrode (high voltage) is a semiconductor, whereas the bottom electrode (grounded) is in Aluminum. For current measurements, brass electrodes of 25 mm diameter were used; measurements were realized at room temperature.

At the end of each measurement step (PEA or current), the sample weight has been recorded. The sample weight returns to its initial value after each measurement, meaning that the cumyl alcohol disappeared from the sample, even at room temperature.

### III. RESULTS AND DISCUSSION

#### A. Untreated Samples

Space charge measurements have been performed on untreated LDPE for different applied voltages (20 min polarization, 20 min depolarization for fields of 10, 15, 20 and 25kV/mm at 25°C), and will serve as reference in this study (Fig. 2). Positive charges are observed, coming from the anode, and travelling across the sample with time. During depolarization, positive charges remain in the dielectric. An increase of the applied voltage implies an increase of the positive charge density measured in the bulk. No negative charges are detected in these profiles, although they can be present, but hidden by the dominant positive charges. This kind of profiles is always measured for pure LDPE under low applied voltage at 25°C [6].

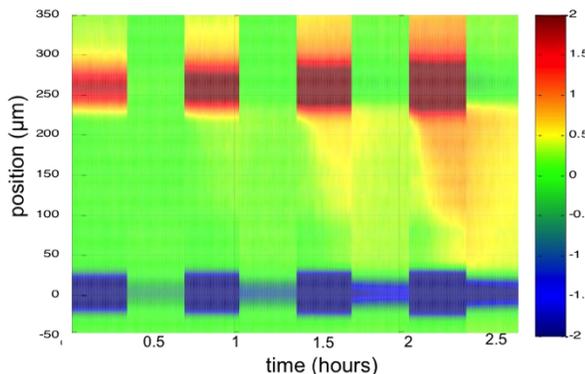


Fig. 2. Space charge distribution as a function of time and space in an untreated LDPE for a protocol of 20 min polarization, 20 min depolarization, for fields of 10, 15, 20 and 25 kV/mm, at 25°C. Colour bar represents charge density scale in C/m<sup>3</sup>.

#### B. Effect of the applied field on treated samples

The same experimental protocol of application of the voltage has been used on LDPE samples soaked for 4 hours in cumyl alcohol at 70°C (polarization/depolarization of 30 min). The space charge pattern (Fig. 3) differs from that of the untreated sample of Fig. 2. Under 10 kV/mm stress, positive charges are detected at the vicinity of the cathode (i.e. heterocharges), as soon as the voltage is applied. Negative charges are also detected next to the anode, their density being lower than that of positive charges.

Under 25 kV/mm field, positive homocharges, coming from the anode, are observed as well as positive heterocharges. The charge in the bulk remains positive during depolarization. The presence of cumyl alcohol, either at the surface or in the bulk of the dielectric, changes completely the space charge behaviour, going from a dominance of positive homocharges to the dominance of positive heterocharges. Even the charge injection detected at 25 kV/mm for the treated sample seems stronger than the one at 25 kV/mm in an untreated one. This is due to the presence of heterocharges at the vicinity of the anode, increasing the electric field locally and hence increasing the charge injection. The same phenomenon holds at the cathode, as negative charge injection can be detected.

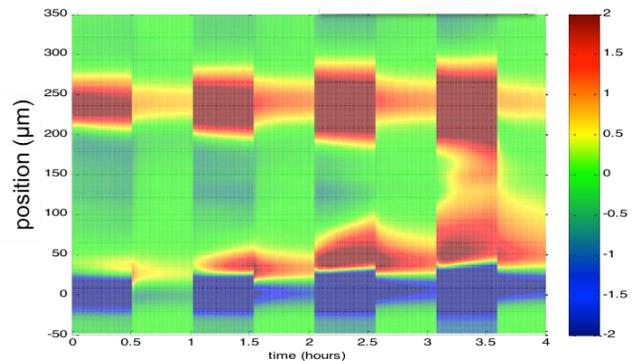


Fig. 3. Space charge distribution as a function of time and space in a LDPE soaked in cumyl alcohol for 4 hours at for a protocol of 30 min polarization/30 min depolarization. Fields of 10, 15, 20 and 25 kV/mm, at 25°C.

Current measurements have been performed on reference LDPE and samples soaked for 4 hours, under fields of 10 and 15 kV/mm (Fig. 4) during 30 min. For untreated samples, the current decreases with increasing time, and there is no stabilization. For treated samples, the current is higher than the untreated one, the difference being of more than one decade in the case of 15 kV/mm. Moreover, the shape of the current is different compared to an untreated sample, as the current seems to stabilize at long time. For 10 kV/mm, the current even seems to increase with time. Such feature has already been observed in polyethylene (LDPE and XLPE) but only when the electric field is high and charge packets can be measured [7-8].

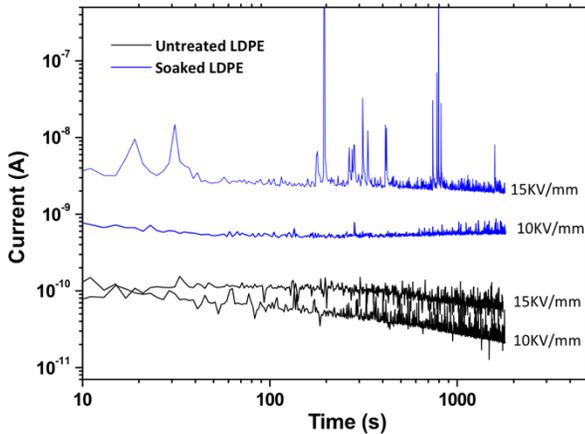


Fig. 4. Current measurements for untreated LDPE (reference) and LDPE soaked in cumyl alcohol for 4 hours, for applied fields of 10 and 15 kV/mm.

### C. Effect of the soaking time on space charges measurements

The concentration of cumyl alcohol present in the LDPE sample is directly linked to the soaking time. In order to see the impact of the by-product concentration on the space charge pattern, the soaking time has been increased from one hour to four hours. The applied electric field was set to 20 kV/mm at room temperature, and the polarization/depolarization times were 1 hour/1 hour, to observe the dynamic of charge accumulation.

Fig. 5 shows the space charge measurements for soaking times of 1, 2 and 4 hours. When the soaking time is short (1h), i.e. when the concentration of cumyl alcohol is low, or when the cumyl alcohol concentration is not homogeneous inside the dielectric, a small amount of heterocharges are detected at each electrode during polarization. These charges are present short after application of the voltage, and their density does not change to a large extent with polarization time. No charge injection is detected for this time, even if heterocharges are present, increasing the electric field at the electrodes.

When the soaking time increases, more heterocharges, positive and negative, are measured inside the dielectric. Consistently with results shown in Fig. 3, the positive heterocharges seem to spread towards the middle of the dielectric, going against the electric field. During depolarization, these charges disappear slowly from the dielectric. For a soaking time of 4 hours, the observations are the same as for 2 hours of soaking time, and positive charge injection is also detected at the anode. When the voltage is set to zero, the injected positive charges seem to disappear immediately, whereas the positive heterocharges, present in the second half of the dielectric, disappear slowly and are still present after one hour of depolarization. It seems that there are two mechanisms for charge disappearance: the first one, fast, implying charge injected from the electrode, and a second one, slower, implying heterocharges. This goes in the direction of two kinds of species, one mobile, i.e. electrons (holes), and the second, slower: ions. These ions could have been created by dissociation of the cumyl alcohol, leading to an immobile negative ion, and a mobile positive one ( $H^+$ ). A second possibility is the ionization of the cumyl alcohol, leading to the creation of a positive ion, immobile, and an electron. However, this last hypothesis is only valid if ionization provides more

electrons (and ions) than charge injection, as negative charge injection also occurs at the cathode, enhanced by the local electric field.

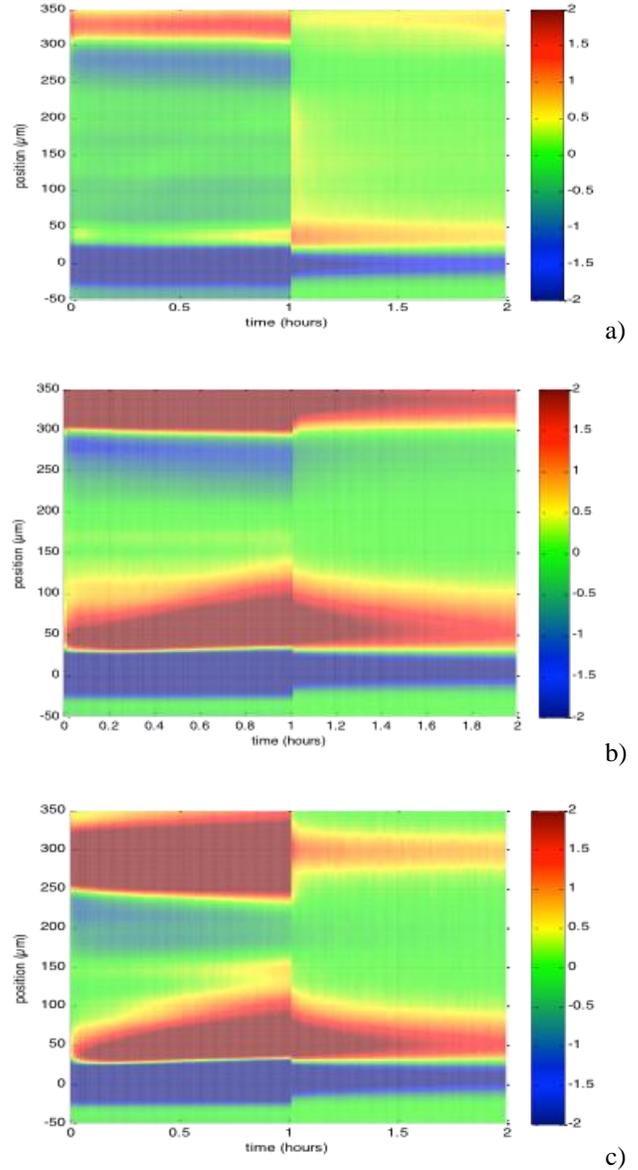


Fig. 5. Space charge distribution as a function of time and space for a protocol of 1 hour polarization/1 hour depolarization at 20 kV/mm and 25°C, for a LDPE sample soaked during a) one hour, b) two hours and c) four hours in cumyl alcohol at 70°C.

### D. Effect of temperature on heterocharge build up

Space charge measurements have been performed for LDPE samples soaked for 4 hours in cumyl alcohol, at temperatures of 10, 25 and 50°C, and for different electric fields, in order to see the impact of the temperature on the dynamics of heterocharges. The experimental protocol consists of polarization/depolarization cycles of 20 min, with increasing fields. At 10°C (Fig. 6a), the charge dynamics is comparable to what has been measured at 25°C (Fig. 3), i.e. positive and negative heterocharges are present even at low field, and there is an increase in the amount of heterocharges that spread towards the

middle of the dielectric with time. The only difference here is that the density of negative heterocharges seems higher compared to  $T=25^{\circ}\text{C}$ , and the injection of positive charges at the anode at 25 kV/mm is not observed. At  $T=50^{\circ}\text{C}$  (Fig. 6b), the space charge patterns are more difficult to interpret. Heterocharges seem to be present, with a lower density compared to  $T=25^{\circ}\text{C}$ . At high fields (25 kV/mm), charge injection is detected at the cathode.

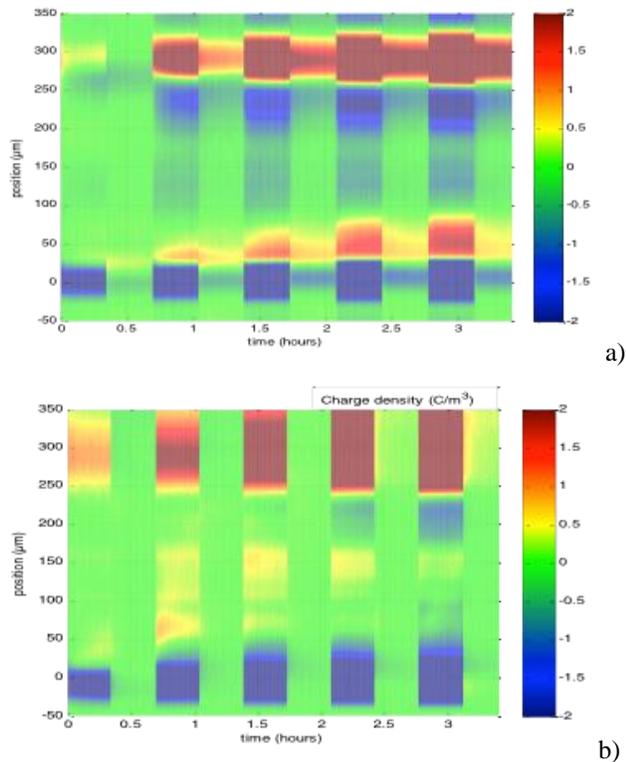


Fig. 6. Space charge distribution as a function of time and space for a LDPE sample soaked four hours in cumyl alcohol, for a protocol of 20 min polarization/20 min depolarization and for fields of 5, 10, 15, 20 and 25 kV/mm. Space charge measurements performed at a)  $10^{\circ}\text{C}$ , and b)  $50^{\circ}\text{C}$ .

### E. Outcomes for modelling purpose

Obtained results pave the way on features to consider for understanding and modelling of charge build-up in by-product containing materials. Dealing with soaked samples, the concentration profile, function of soaking time and temperature has to be built, along with the evolution of the concentration profile (outgassing) in the atmosphere where measurements are carried out. Concerning physical processes of charge generation and transport, evidence of ionic species formation has been given and the kinetics as well as the nature of ions has to be anticipated. Aside that, electronic carriers generated at the electrodes are still present and their contribution must be superposed to that of ionic phenomena. A more difficult task is

to estimate the impact by-products may have on the transport of electronic carriers. Finally, owing to the polar nature of the byproducts, orientation polarization (and related gradient) has to be considered as possible contributions to displacement current and charge density profile.

## IV. CONCLUSION

The presence of cumyl alcohol in LDPE samples has a strong impact on the dielectric behavior as probed by space charge and current measurements. It increases the charging current by more than one decade, for moderate field (15 kV/mm), and it creates heterocharges, that disturb the electric field at the electrodes, and provides more space charge in the dielectric compared to a pure LDPE sample. An increase of the soaking time in cumyl alcohol implies an increase of the heterocharge density. However, it is difficult to link the soaking time to the concentration in cumyl alcohol, as it is not sure that the absorption is homogeneous in the volume of the LDPE sample. Physico-chemical measurements need to be performed to answer to this question. The presence of heterocharges has been attributed here to ions introduced by the cumyl alcohol, that either dissociate to form an immobile negative ion and a proton, that accumulate at the cathode, or that ionize, forming an electron and an immobile positive ion. More work needs to be done to identify the type of ions that are created when introducing cumyl alcohol in a LDPE matrix.

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