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# STUDY OF THERMALLY AND WATER TREE AGED POLYETHYLENE BY INFRARED SPECTROSCOPY, ABSORPTION CURRENTS AND SPACE CHARGE MEASUREMENTS

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## ABSTRACT

*The paper deals with thermal and water tree ageing of polyethylene insulation used for medium voltage cables. Laboratory-made flat polyethylene samples have been aged thermally and under the effect of water and ac electric field, at high frequency (5 kHz). Absorption/resorption currents and infrared spectroscopy measurements have been performed before and after ageing. The evolution of the electric charge trapped in the water treed samples has also been monitored. The results are analyzed in view of correlating the water tree ageing of the materials with the evolution of the electric charge in the insulation, in order to elaborate a non destructive methodology for estimating the treeing degree of a cable.*

## KEYWORDS

Power cable – Ageing – Water tree – Space charge – Absorption currents

## INTRODUCTION

Power cable insulations are submitted during service to permanent and accidental stresses (electrical, thermal, mechanical, water, radiations etc.). Under the effect of these stresses, the insulations suffer different degradation processes, which lead both to the decrease of physical properties and to the reduction of lifetime [1]. As heat, oxygen and moisture are considered as main ageing agents, heat-resistant (cross-linked) polyethylene, oxygen-resistant polyethylene (with anti-oxidants) and water-tree resistant polyethylene (containing water-tree retardants) have been manufactured [2-3]. Moreover, the cable manufacturers provide the cables with barriers against moisture and water trees. However, most of the operating medium voltage cables are not equipped with barrier against water penetration, and their insulations do not contain water tree retardants. Tests allowing to detect the water trees' presence in cable insulation and to estimate the insulation ageing state and the remaining life are therefore needed. Several methods, from which ones are already used whilst the others are still being experimented, have been proposed for this purpose [4-6].
The present work is concerned with experiments carried out for analyzing the water tree growth in unaged and thermally aged low density polyethylene samples, which have also been characterized by infrared spectroscopy, space charge measurement and absorption / resorption currents. The aim is to follow the modification of the values of space charge and of the absorption/resorption currents in the water treed polyethylene samples in view of using these results for assessing the state of cable insulation.

## EXPERIMENTAL SET UP

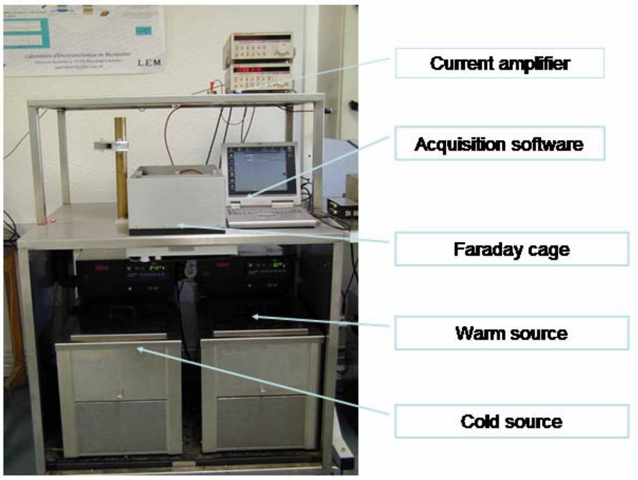


**Figure 1 : Thermal step method bench for space charge measurements in flat samples [7]**

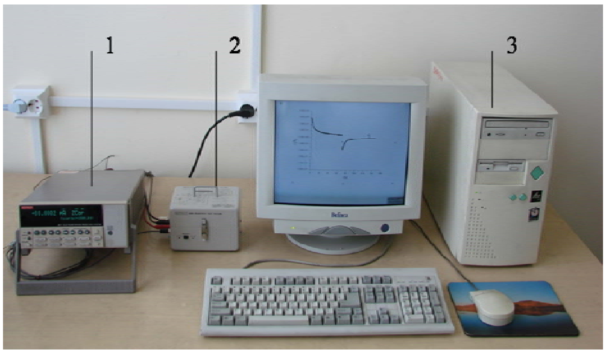


**Figure 2 : Experimental set up for measuring absorption/resorption currents : 1- Keitlhey 6517 electrometer, 2- Keithley 8009 measurement cell, 3- computer.**

Squared plaques with a surface of 150x150 mm$^2$ and of thickness $d$ = 0.5 mm have been manufactured from low density polyethylene pellets by pressmoulding at 200 bars, 145°C. The plaques have been submitted to a first thermal conditioning at 50°C during 2 days, then disks with a diameter $D$ = 60 mm have been cut from the plaques. Then, the disks have been provided with graphite electrodes (with a diameter of 50 mm) and subjected to space charge and absorption/resorption currents measurements. The space charge measurements were performed with the thermal step

method in short-circuit conditions using the installation from Figure 1. The samples have been submitted to a thermal step of -30 K and the thermal step currents have been recorded during 10 s. The absorption currents were measured with the set up from Figure 2 by applying a 1000 V dc voltage during 2 hours.
After the measurement of space charge and absorption currents, the samples were thermally aged in a pulsed air oven. The space charge and absorption currents were then again measured.
Infrared spectroscopy measurements have been carried out on virgin and thermally aged samples. The absorption bands from Table 1 have been examined in particular, as they are known as being the most important diagnostics bands for assessing the state of polyethylene insulation.

| Absorption band [cm$^{-1}$] | Presence of bonds or groups | Comment |
|---|---|---|
| 1150-1300 | C-O ester bonds | DB |
| 1575-1610 | C-O carboxylate jons | DB |
| 1630-1640 | C=C duplex bonds | weak DB |
| 1710-1720 | C=O carbonyl groups | main DB |
| 2900-2650 | C-H in methylene groups $CH_3$ | DB |
| 3100-3600 | OH stretching bonds | DB |

**Table 1 : Diagnostic absorption bands for polyethylene (DB = diagnostics band of infrared spectrum)**

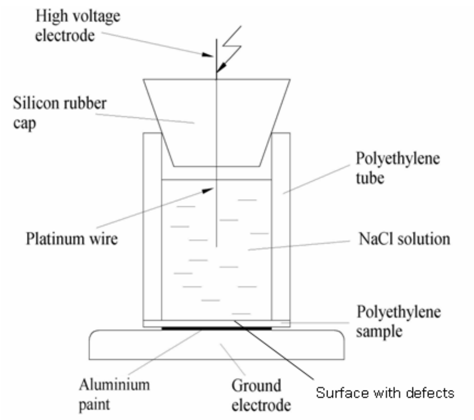


**Figure 3 : Cell used to produce water trees.**

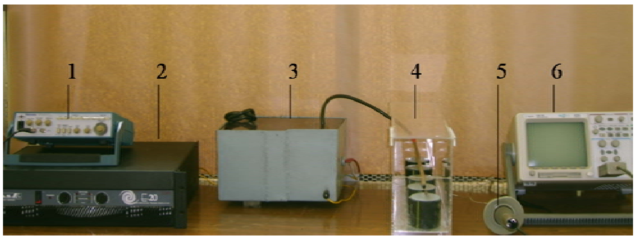


**Figure 4 : Experimental set up for laboratory development of water trees in flat samples: 1- Variable frequency power supply, 2- Amplifier, 3 – 100 V/ 5000 V ac transformer, 4 – 1/1000 high voltage probe, 5 – oscilloscope.**

In order to initiate and develop water trees in an accelerated manner, superficial defects have been made on one side of each disk by pressing it onto abrasive paper by means of a Carver press at a pressure of 10 to 22 MPa for 2 to 5 minutes. On the other side of the disk, an aluminium layer has been placed, and then the disk was glued on a polyethylene cylinder of inner diameter $D_c$ = 50 mm and of height $h_c$ = 70 mm (Figure 3). A 0.1 mol/l NaCl solution has been used to accelerate the initiation of water trees. Five cells were grouped in a cell holder and subjected to a 2kV RMS, 3-5 kHz ac voltage during 24 to 72 hours (Figure 4).
After water tree development, space charge and absorption current measurements were performed on a part of the disks. For water tree measurements, the rest of the disks have been placed in a rhodamine solution for 3 days at a temperature of 60°C. 3 slices with the thickness of 200 µm have then been microtomed from each disk (Figure 5 left). The water trees have been visualized on each slice by using a microscope and the maximum lengths and diameters $L_k$ and $D_k$ were measured (Figure 5 right) [8].

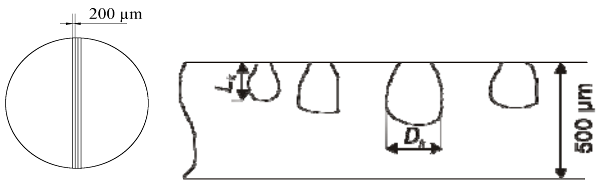


**Figure 5 : Localization and thickness of slices cut from the flat samples for water trees measurement (left) and measurement of the diameter $D_k$ and of the length $L_k$ of water trees grown in the samples (right).**

## RESULTS AND DISCUSSION

The results obtained after thermal ageing of the samples ($T_1$ = 95°C and $T_2$ = 105°C during 500 hours), followed by electrical ageing in the 0.1 mol NaCl/water solution at 2 kV RMS, 5 kHz, are presented herein after.
The infrared spectroscopy investigations made on virgin and thermally aged samples (examples in the next Figures) did not show significant changes in the infrared spectra after ageing in air at 95°C during 500 h. The analysis of the spectrograms showed slight changes in the samples aged at 105°C during 183 h. Most important (but still slight) evolutions have been observed in the samples aged at 105°C during 500 h, particularly in the 1180-750 cm$^{-1}$ band (less C-H vinyl bonds) and in the 3900-3000 cm$^{-1}$ band (presence of OH groups).

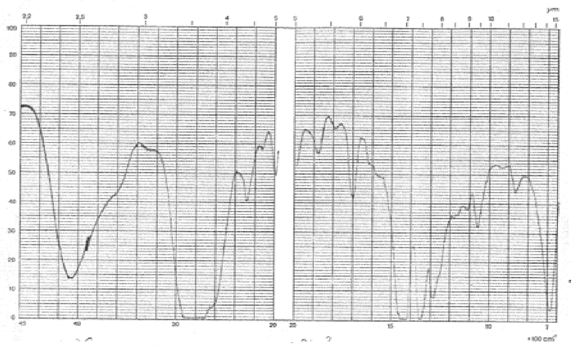

**Figure 6 : Infrared spectrogram of a virgin sample.**

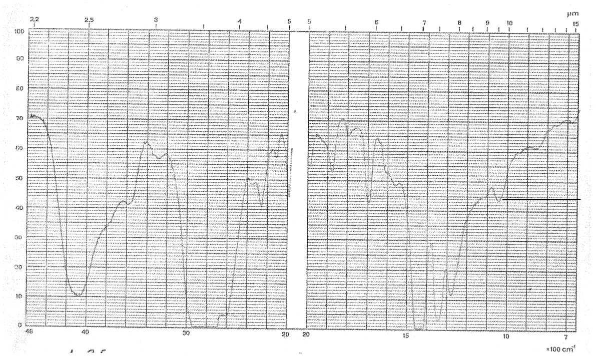

**Figure 7 : Infrared spectrogram of a sample aged at 95°C during 500 hours.**

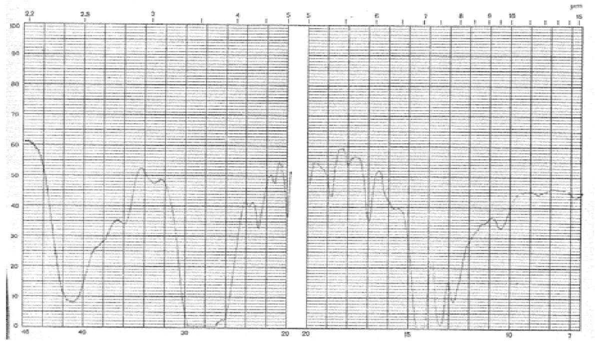

**Figure 8 : Infrared spectrogram of a sample aged at 105°C during 500 hours.**

The ac electrical ageing in aqueous environment created in the samples water trees with the densities, dimensions and volumes given in the Table below.

| Type of samples | ac ageing time [h] | Characteristics of water trees | | | |
|---|---|---|---|---|---|
| | | average diameter [μm] | average length [μm] | volume [$m^3$] | density [$mm^{-2}$] |
| Virgin | 24 | 138.54 | 151.06 | 0.85e-8 | 1.90 |
| | 48 | 226.50 | 226.19 | 7.51e-8 | 4.20 |
| | 72 | 287.94 | 265.22 | 15.3e-8 | 4.53 |
| Aged thermally | 24 | 189.39 | 184.39 | 12.1e-8 | 2.20 |
| | 48 | 210.56 | 251.03 | 22.3e-8 | 11.00 |

**Table 2 : Dimensions of water trees in virgin and thermally aged LDPE samples after ac conditioning in 0.1 mol/l NaCl/water solution (2 kV RMS, 5 kHz)**

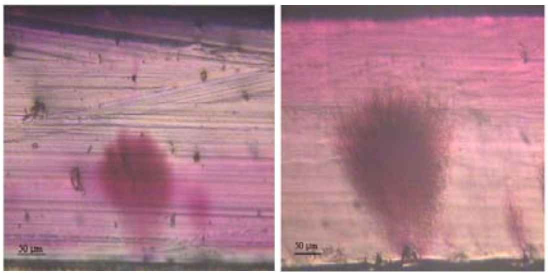

**Figure 9 : Water trees grown after submission to electrical stress in 0.1 mol NaCl solution (2 kV RMS, 5 kHz, 24 h) of thermally unaged (left) and thermally aged LDPE samples (95°C, 500 h) (right)**

In Figure 9 are presented water trees (developed after 24 hours of submission to 2 kV RMS, 5 kHz ac voltage in aqueous solution) in a virgin sample and in a sample aged thermally during 500 hours at 95°C. It can be noticed that, after thermal ageing, the dimensions (average diameter and length), the volume and the concentration of the water trees increased (Table 1). The increase of the concentration of water trees is likely due to the decrease of the mechanical properties of the polyethylene as a result of the submission to the thermal stress, and, consequently, to the increase of the concentration of the surface defects produced by the press (Figure 10).

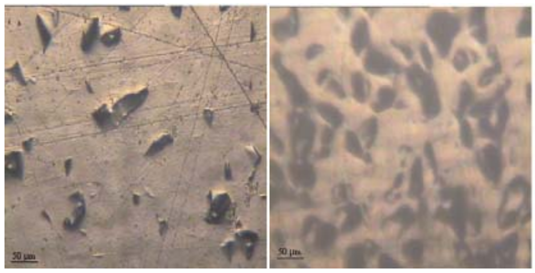

**Figure 10 : Defects after pressing in a thermally unaged LDPE sample (left) and in a LDPE sample aged thermally at 105°C during 500 h (right)**

The thermal ageing also determines a modification of the absorption currents: their amplitudes decrease and the slopes of the variation curves increase (Figure 11). This is likely due to the decrease of the initial concentration of charge carriers and to the evaporation of the water existing in the sample, without witnessing important degradation of the samples.

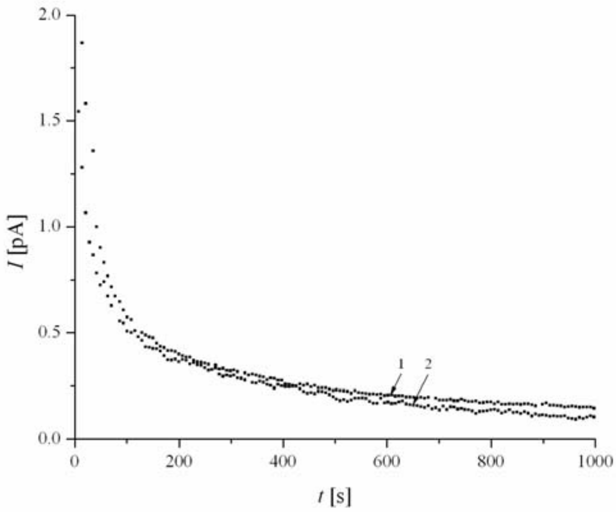


**Figure 11 : Absorption currents in a virgin LDPE sample (1) and in a thermally aged sample (105°C, 500 h) (2)**

The absorption currents measured in virgin, thermally aged, electrically aged and thermo-electrically aged samples are presented in Figure 12. We note that the presence of water trees provokes (as expected [4, 9-10]) an increase of the absorption current. Moreover, the values of the absorption current in the thermally aged water treed samples are higher than in the water treed samples unaged thermally: the corresponding ratio measured in Figure 12 reads 1.15 at 7 s, 2.4 at 60 s and 3.7at 3500 s. This is likely due to a higher amount of mobile space charge in the sample and to the change of the sample's permittivity.

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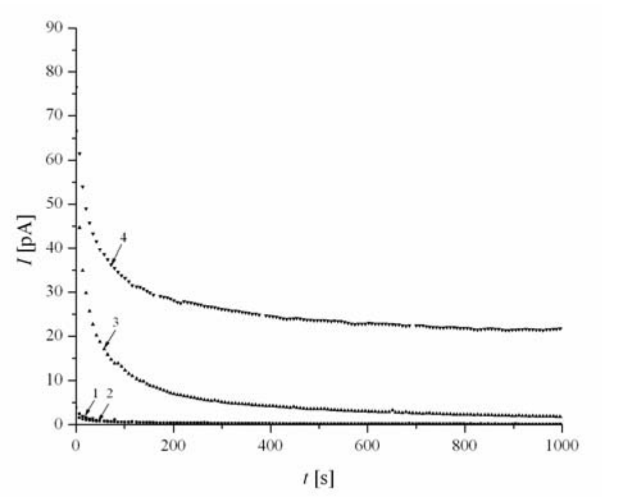


**Figure 12 : Absorption currents in different samples: virgin LDPE sample (1), thermally aged sample LDPE (2), LDPE sample ac-conditioned in 0.1 mol/l NaCl/water solution (3), thermally aged LDPE sample ac-conditioned in 0.1 mol/l NaCl/water solution (4)**

The following Figures present thermal step currents measured on thermally aged samples and on samples in which water were developed. We should recall that the amplitude of a thermal step current is directly related to the electric field across the sample and to the charge content of the sample.

The thermal step currents measured on the virgin samples are weak (< 30 pA) but not zero, witnessing a low amount of space charge present in the samples from the manufacturing. The thermal conditioning provoked modifications in the amplitude of the thermal step currents, but these changes are limited, suggesting that the thermal stress favoured redistribution and partial recombination of the carriers, but did not result in a significant increase of the charge amount. This correlates to IR spectroscopy results presented earlier and to the absorption currents presented in Figure 11.

The thermal step currents measured on the samples after submission to ac field and appearance of water trees are higher than before (100 to 150 pA). This is due to the appearance in the water treed samples of charge carriers associated to the water trees. The measurements made at different times after the ac ageing show an important decrease of the currents after 6 days (150 pA down to 60 pA), suggesting a quite important mobility of the carriers resulting in a migration and recombination of a significant part of the likely ionic charge, which is in accordance with the absorption current data from Figure 12. The dynamics of the thermal step current decrease in time let us suppose that significantly higher thermal step currents (and consequently charge amounts and internal electric fields) would have been measured just after the ac conditioning.

The comparison of thermal step currents measured on thermally aged and thermally unaged samples at the same moment after ac conditioning (which is 21 days after) shows higher signals in the thermally aged samples. These results, which are likely due to more charge present in the thermally aged samples, correlate with the higher amounts and densities of water trees developed in the thermally aged samples, as shown in Table 2. For instance, an average density of 11 $mm^{-2}$ of water trees was measured in the thermally aged sample after submission to ac field during 48 h, with respect to 4.20 $mm^{-2}$ measured in the sample unaged thermally and submitted to the same electrical stress. The thermal step results also correlate with the absorption currents shown in Figure 12, where the highest absorption current is exhibited by the thermally and electrically aged sample.

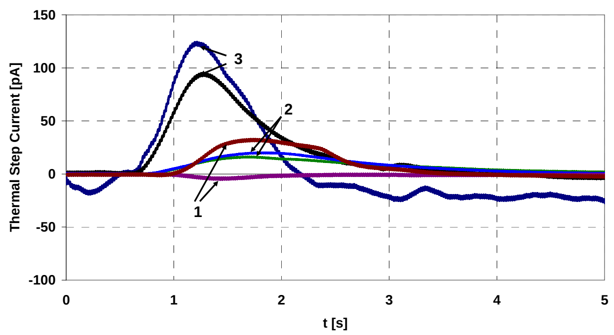


**Figure 13 : Thermal step currents measured on two LDPE samples before conditioning (1), after thermal conditioning in air at 95°C during 500 h (2) and after thermal conditioning in air at 95°C during 500 h followed by ac conditioning at 2 kV RMS, 5 kHz during 24 h in 0.1 mol/l NaCl/water solution (4).**
**The measurements were made 21 days after each conditioning by applying the thermal step to the side of the samples to which the high voltage has been applied and from which the water trees have developed.**

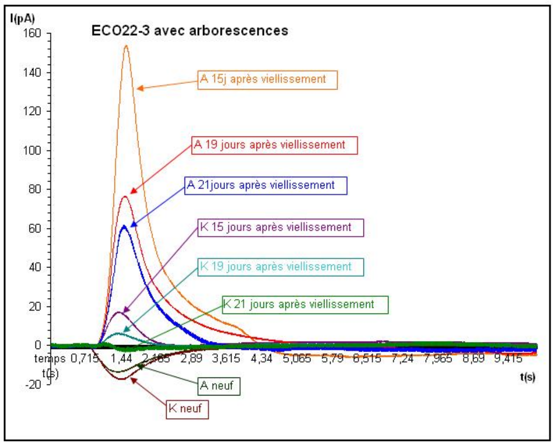


**Figure 14 : Thermal step currents measured on a virgin LDPE sample ("neuf") and on the same sample conditioned at 2 kV RMS, 5 kHz during 48 h in 0.1 mol/l NaCl/water solution. The measurements were made 15, 19 and 21 days after the conditioning.**
**<u>Legend</u> : A = thermal step applied to the "anode" side, K = thermal step applied to the "cathode" side, the "anode" being the high voltage electrode.**
**The water trees detected in the sample grown from the "anode" towards the "cathode".**

In order to determine the amount and the distribution of space charge in the water treed samples, the measured thermal step currents must undergo a numerical treatment. Several methods for processing thermal step currents recorded on homogenous materials have been set up so far

[5, 7], but they are not directly usable on samples with variable permittivity as those containing water trees. To evaluate the electric charge in a water treed sample, we have carried out simulations with the COMSOL multiphysics software by using an average length of water trees of 100 µm considered as distributed uniformly in the neighbourhood of the high voltage electrode. The relative permittivity of the water treed region was considered as equal to 3 times that of polyethylene, i.e. 6.6.
Three simple types of charge distributions were considered: uniformly charged water trees, uncharged water trees and a 40 µm uniformly charged area in front of the water treed region, and uniformly charged water trees plus a 40 µm uniformly charged water treed region in front of the water trees (Figure 15).

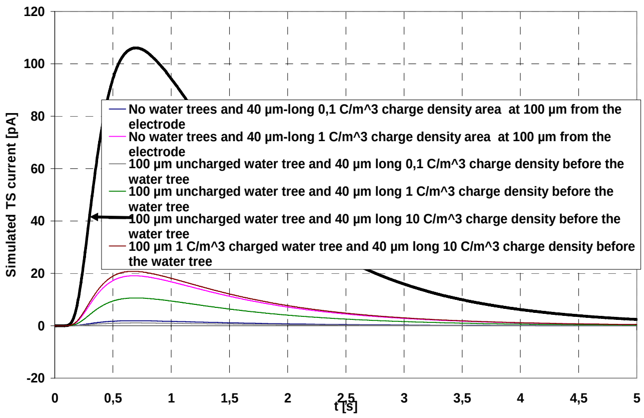


**Figure 15 : Simulated thermal step currents in a water treed sample with different locations of electric charge associated to the water trees**

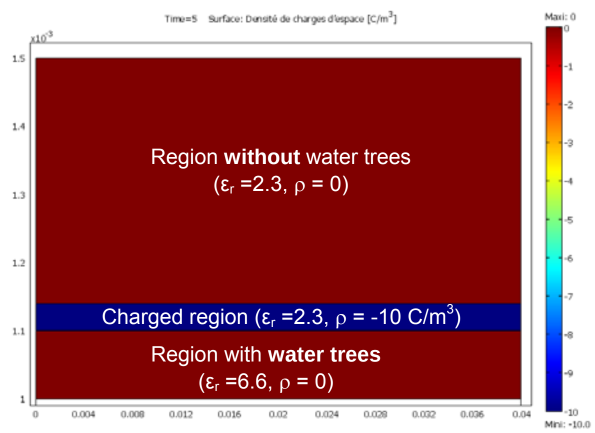


**Figure 16 : Example of a 500 µm LDPE sample containing uncharged water trees and uniformly distributed charge density. If a thermal stimulus identical to that of the measurement bench used in the work is applied in short-circuit conditions to the lower side of the sample, the thermal step current of 105 pA from Figure 15 is obtained**

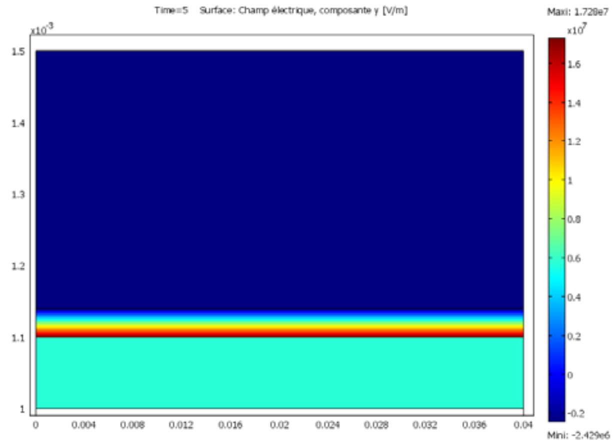


**Figure 17 : Electric field distribution in the case shown in Figure 16**

The simulations show that, for obtaining thermal step currents of the order of 100 pA as those observed experimentally, the samples must contain charge densities as high as 1 to 10 C/m$^3$, inducing electric fields that could exceed 17 kV/mm (Figure 17). Taking into account that the space charge measurements were made several weeks after the ac conditioning and that the dynamics of space charge recombination is important (Figure 14), it is very likely that the electric charge and field amplitude in the water treed regions are even higher. This means that the water treed samples have been actually submitted to electric fields exceeding by several dozens of kV/mm the applied field.

## CONCLUSION AND PROSPECTS

The results presented in the present paper show an important increase of the absorption currents and thermal step currents in the water treed polyethylene samples. These changes are related to the appearance in the material of electric charge associated to the water trees, which is likely of ionic origin. It was found that the space charge amount and water trees content are higher in the samples submitted to a thermal stress, even if significant changes in the chemical structure of the samples were not detected. Using the experimental results, the values of the electric field associated to the space charge contained in the measured water treed samples has been estimated as several times higher than the field applied during electrical conditioning.
The significant increase of the absorption currents and space charge signals may be used as an indicator for the presence of water trees in the insulation of power cables. Setting up a correlation law between the space charge amount and the length of water trees in power cable insulation, by way of controlled growth of water trees in power cables, would lead to a non destructive and easy-to-use methodology of assessing the water tree growth in the operating cables. This is the next step of the present work.

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