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# Assessment of Fallouts of the Thermal Step Method for measuring Space Charge in Liquid Dielectrics

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**Abstract**—This focuses on studying the applicability of the Thermal Step Method (TSM) in liquid dielectrics. The TSM is widely used on solid dielectrics, in which thermal conduction prevails. Experimental results and numerical simulations are employed to study the use of the TSM in dielectric liquids, particularly with respect to liquid convection and permittivity variation with temperature.

**Index Terms**—liquid dielectrics, space charges, thermal step method, electrical double layer

## I. INTRODUCTION

Space charge present in dielectric liquids has been often involved in failures, but it can also bring benefits in other situations. Measuring these charges as precisely as possible is therefore important, both from a scientific and applicative point of view. The existing methods for measuring space charges in liquids do not bring information about the space charge distribution. The Thermal Step Method (TSM), which has been widely used in solid dielectrics, is studied in this paper for the case where the charges are those coming from the Electrical Double Layer (EDL) occurring at the liquid/solid interface. The EDL is used to generate a reproducible space charge distribution profile in the samples.

## II. PRINCIPLES

### A. Thermal Step Method (TSM)

Among the space charge measurement techniques, the TSM is a non-destructive method, implying no physical degradation of the sample. This method uses the thermoelectric properties of the material in order to generate a current when a temperature step is applied to the sample surface. The principle of the TSM in the case of a flat sample is given by Fig. 1. The sample is defined by its geometry (thickness  $d$  and surface  $S$ ) and its electric properties (relative capacitance variation with the temperature  $\alpha$  and permittivity  $\epsilon$ ). Because it is considered infinitely flat and that thermal conduction is the only heat transfer in solids, the problem can be assumed as unidirectional. The sample is short-circuited between the electrodes located at  $x = 0$  and  $x = d$ , so when a temperature step  $\Delta T = T - T_0$  is applied to electrode 1, the influence charges at electrodes are redistributed through the external circuit, creating a capacitive current (Fig. 1). By considering first order variations of the capacitance and by neglecting

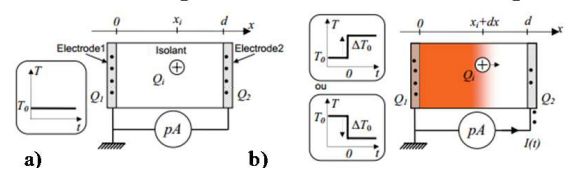


Fig. 1. Principle of the TSM on a flat sample ; sample at initial temperature (a) and after the thermal step (b).

the conductivity of the dielectric, the resulting current can be written as [1]:

$$I(t) = -\alpha C \int_0^d E(x) \frac{\partial \Delta T(x, t)}{\partial t} dx \quad (1)$$

where  $C$ ,  $E(x)$  and  $\frac{\partial \Delta T(x, t)}{\partial t}$  are respectively the sample capacitance at initial temperature  $T_0$ , the electric field distribution and the variation of the temperature distribution within the sample. The response of the dielectric to the thermal stimulus is linked to the steepness of the thermal excitation, which should be preferably close to a temperature step for obtaining high amplitude signals. In order to obtain the space charge distribution  $\rho(x)$ , the electric field is extracted from the equation (1) [2], then the Poisson law is used:

$$\rho(x) = \nabla \cdot (\epsilon E(x)) \quad (2)$$

### B. Electrical Double Layer (EDL)

The physicochemical phenomenon occurring at the solid-liquid interface and responsible of the development of the Electrical Double Layer remains poorly understood. Different scenarios have been made: it might be an adsorption or a corrosion reaction or a mix of those two, that is assumed depending on the solid/liquid couple. According to Stern's model [3], two areas of opposite signs, one in the liquid and the other one in the solid, compose the EDL. In the liquid, two layers have been identified: the compact and the diffuse layer (Fig. 2). Usually, the thickness of the diffuse layer is defined as the Debye length  $\delta_0$ :

$$\delta_0 = \sqrt{\frac{\epsilon_0 \epsilon_r D_0}{\sigma}} \quad (3)$$

where  $\epsilon_0$  and  $\epsilon_r$  are the vacuum permittivity and the relative permittivity of the liquid,  $D_0$  is the mean ionic diffusion co-

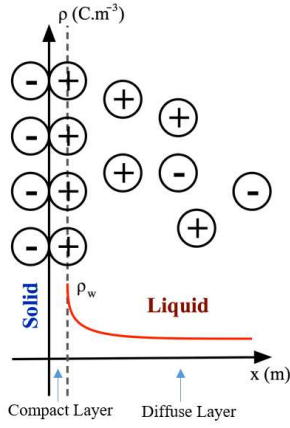


Fig. 2. Representation of the Electrical Double Layer

efficient of the liquid and  $\sigma$  its bulk conductivity. Considering an infinite contact time between the solid and the liquid, the EDL is fully developed and the space charge density at the wall  $\rho_w$  reaches its maximum value  $\rho_{wd}$ .

The EDL developed at the interface between two identical materials remains always the same, so it provides a reproducible space charge distribution in the liquid. But the finality of this study is to use the TSM to obtain information on the compact layer. Indeed, today EDL properties are estimated using the flow electrification phenomenon. This experimental measurement allows to assess the total amount of the EDL charges neglecting the compact layer charge and to extrapolate the value of  $\rho_{wd}$  [4] [5]. However, such measurements cannot be used to determine the space charge density profile.

### III. EXPERIMENTAL SETUP

The experimental set up for the study is composed of three cavities enclosed by stainless steel cylinders of height 125 mm (Fig. 3). The inner and the outer cavities can be used to circulate the cooling fluid that provides the thermal step (pure ethanol). The middle cavity contains cyclohexane, an insulating liquid with a high melting point, allowing studies both in solid and liquid state. We use the inner cavity to generate the temperature step and the outer one is filled with air, which acts as a buffer to maintain the initial temperature at the edges during measurement. We made experiments in both liquid and solid state, to compare the behavior. To solidify the sample, we circulate liquid nitrogen ( $T_{liquidN_2} = 77.36 K$ ) in the cell when it is at room temperature.

### IV. SETUP FOR NUMERICAL SIMULATIONS

In order to study several aspects concerning the application of the TSM to liquids, we made simulations in a geometry similar to the one used during the experiments. The measurement cell was represented in Comsol Multiphysics in a 2D axisymmetric configuration. The velocity of the cooling fluid was set to  $u = 2 m.s^{-1}$  and the fluid came from upside the cell. Because the inlet diameter is smaller than the diameter of the inner cavity, the initial speed in this cavity

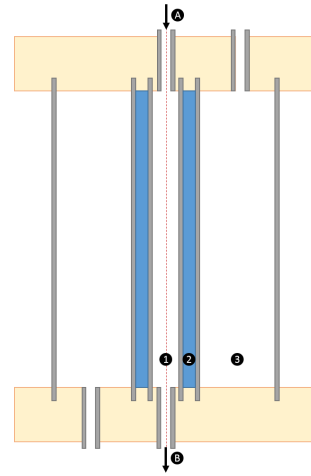


Fig. 3. Representation of the TSM cell - (A) Cooling inlet, (B) Cooling outlet - (1) Cooling fluid cavity, (2) Cyclohexane cavity, (3) Air cavity

is  $0 m.s^{-1}$  and we wait for 10 s so the flow establishes, then we apply the temperature step on the inlet edge. To study the presence of thermal convection, we include the density variation of cyclohexane with the temperature (4), so a compressible laminar flow within the sample is considered [8]:

$$\rho_m(T) = \frac{84.1595 \times 0.8908}{0.27396^{1+(1-\frac{T}{553.58})^{0.2851}}} \quad (4)$$

Concerning the EDL between 2 parallel plans, a simplified model of space charge distribution can be used when those plans are far away enough and when the radius of curvature is low comparing to the Debye length [9]. So we set a symmetric exponential decreasing profile in cyclohexane:

$$\rho(r) = \rho_{wd} \cdot e^{-\frac{r-R_{int}}{\delta_0}} \quad (5)$$

with  $\rho_{wd} = 10^{-4} C.m^{-3}$ ,  $r$  the radius and  $R_{int} = 16 mm$  the internal radius of the sample cavity. In cylindrical coordinates, when the radius of curvature is sufficiently large in front of the thickness of the sample, (1) becomes:

$$I(t) = -\alpha C \int_{R_{int}}^{R_{ext}} E(r) \frac{\partial \Delta T(r, t)}{\partial t} dr \quad (6)$$

where  $R_{ext} = 19 mm$  is the external radius of the sample cavity. From dielectric spectroscopy performed on cyclohexane with *Solartron Analytical ModuLab XM MTS*, values of  $\alpha_{liquid} = 0.02 K^{-1}$  and  $\alpha_{solid} = 0.007 K^{-1}$  have been assessed. The simulations have been made under the hypothesis that the thermal step does not affect the EDL.

### V. RESULTS AND DISCUSSIONS

#### A. Experiments

Fig. 4 shows thermal step currents obtained with the cyclohexane sample in liquid and solid form. They put into evidence that the response of the sample to the thermal stimulus is high enough to obtain measurable signals, of amplitude as high as 5 nA. However, the response to the same thermal step of

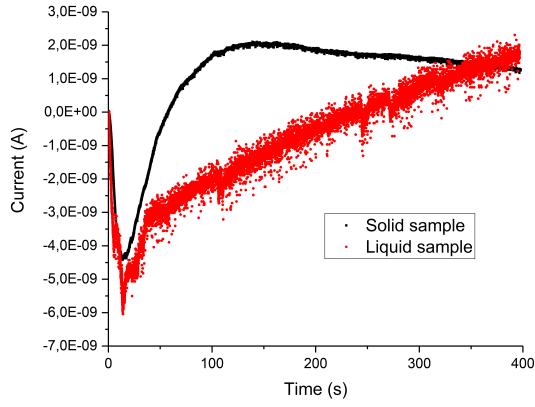


Fig. 4. TSM experimental currents measured with a thermal step of 25 K in liquid and solid state

$\Delta T = 25 K$  do not witness an identical behavior of the sample in solid and liquid state. In the solid state, the TSM response is close to those obtained classically [2]. Considering the fact that freezing the cyclohexane also froze the space charges without displacing them, the different shape of the current in liquid state could be an effect of the thermal convection moving charges during the measurement. The amplitude of the peak is superior for the liquid sample, but the value of  $\alpha$  is not the same in the two states. Moreover, the difference in behavior must be also affected by the heat transfer, which is not the same into the two states, just as the thermodynamical characteristics. This is because when the sample is solid, its initial temperature is 223.15 K, but when it is liquid, the temperature is 293.15 K. The fact that the noise is lower in solid state supports the hypothesis that the space charges can be "frozen" in their initial state.

### B. Numerical simulations

In order to study the effects of the convection in liquid cyclohexane on the thermal step response, we have used the representation from Fig. 3 and the data from section IV. The velocity of the cooling liquid has been increased in the simulations with respect to the experiment presented above. We wanted to simulate this effect for the case of a more dynamic signal, as close as possible to the application of a perfect thermal step. This is the reason why the simulated signals have a much higher dynamics than the experimental data.

First, we can notice, according to the results from Fig. 5, the presence of thermal convection within the cyclohexane, witnessed by the speed profile across the liquid sample. The convection is due to the non-uniform temperature diffusion across the liquid cyclohexane (shown in the region (2) of Fig. 6), which results in a variation of its density (according to (4)), and thus to liquid movements in the sample. These movements are similar to Rayleigh-Bénard convection, although in this case the tempered sides are not up and down edges. Similar cases have been studied, but none has been found in a con-

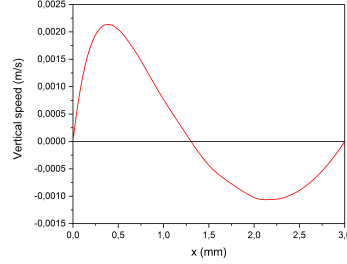


Fig. 5. Speed profile in cyclohexane - Positive step -  $t = 2$  s after the step -  $z = 75$  mm

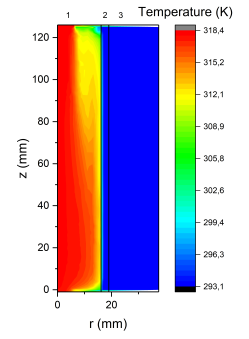


Fig. 6. Temperature profile in cyclohexane - Positive step -  $t = 2$  s after the step - (1) Cooling fluid cavity, (2) Cyclohexane cavity, (3) Air cavity

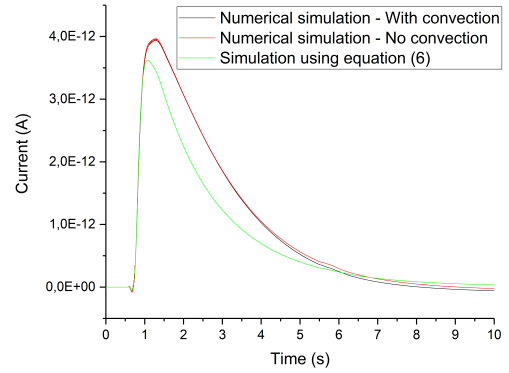


Fig. 7. TSM currents calculated with a thermal step of +25 K in liquid and solid state

figuration similar to ours, disallowing to compare our results with literature [6] [7].

In the aim of strictly studying the effect of the thermal convection on the TSM current, we used the same value of  $\alpha$  in two cases: with and without the computation of convection in the sample. Fig. 7 and 8 show currents computed with the two models for positive and for negative stimuli. We notice that the influence of the convection on the TSM

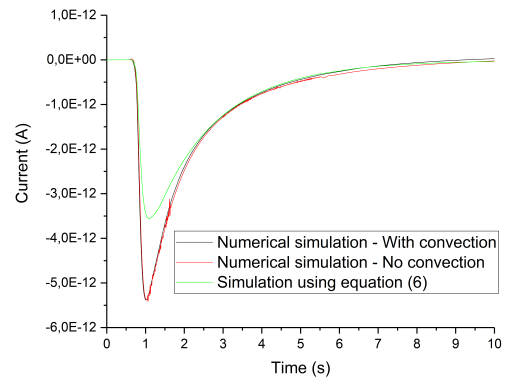


Fig. 8. TSM currents calculated with a thermal step of -25 K in liquid and solid state

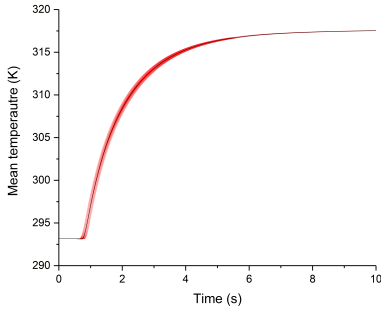


Fig. 9. Average temperature calculated at the edge of the sample

currents is indistinguishable. This means that in the considered configuration, the diffusion of the temperature is quicker than the generation of the flow movement within the sample, so, thermal conduction can be considered as the only heat transfer mode. By studying the average temperature profiles at the edge of cyclohexane extracted from simulations results (Fig. 9), we concluded that the dispersion is not as high as we could expect. It seems that the stainless steel inner cylinder helps to uniform the temperature by acting as a buffer, but it is not very efficient at the top and the bottom of the cell.

Equations (1) and (6) have been set up by considering that the coefficient of variation of the permittivity with the temperature  $\alpha_{eps} = \frac{1}{\epsilon} \frac{d\epsilon}{dT}$  is low enough to consider a first order variation of the capacitance with temperature [1]. However, the differences between numerical simulations and simulations obtained with equation (6) (Fig. 7 and 8) seem to question this assumption.

In order to check the validity of equation (1) and (6) for the studied insulating liquid, we simulated TSM responses in 1D with Comsol using 3 values of  $\alpha$  :  $2.10^{-2} K^{-1}$ ,  $2.10^{-3} K^{-1}$  and  $2.10^{-4} K^{-1}$ . In Fig. 10, two currents are shown for each value of  $\alpha$ : first, the current calculated with the analytical expression (1) and the surface integral of the displacement current density, calculated numerically with Comsol in electrostatic regime :

$$J_D(t) = \frac{d(\epsilon_0 \epsilon_r E(x))}{dt} \quad (7)$$

It comes out from Fig. 10 that, for the highest value of  $\alpha$ , the analytical current does not fit with the numerical current, while it fits perfectly when the variations are lower. This suggests that, for values of  $\alpha$  approaching  $10^{-2} K^{-1}$ , the approximation of the variation of the ratio  $\frac{dx}{\epsilon(x)}$  with temperature by  $\frac{dx}{\epsilon(x)} (1 + \alpha(T))$  [1] may lead to significant error and highest order terms should be taken into account in the TSM equations. Finally, the differences of amplitudes between numerical and experimental currents could come from the charges present in the compact layer, that has not been taken account in the simulations.

## VI. CONCLUSION

In this work, measurements and simulations have been presented in order to study the use of the Thermal Step Method

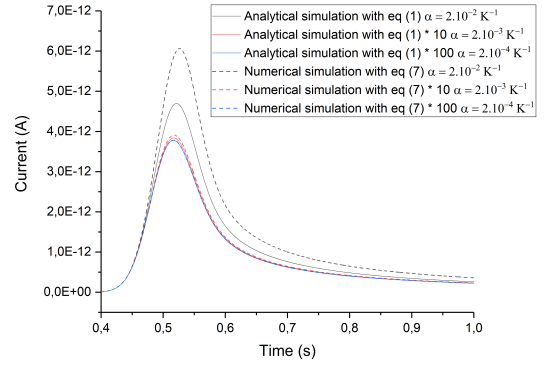


Fig. 10. Comparison of TSM responses calculated analytically with equation (1) and numerically with equation (7) currents by varying  $\alpha$

for space charge measurements in liquid dielectrics. Experimental results, obtained on cyclohexane samples with fully developed Electrical Double Layer, showed signals with amplitudes of the order of nA, easily measurable with the present instruments. Simulations results showed that the influence of convection effects in the liquid could be avoided provided that the dynamics of the thermal transfer is high enough. The computations also suggest that higher-order effects related to the variation of the permittivity in liquids should be considered in the thermal stimulus signal equation.

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