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Xavier Sidambarompoulé, Jean-Charles Laurentie, Petru Notingher, Thierry Paillat, Paul Leblanc, et al.. Development of an experimental setup in order to measure space charges in dielectric liquids. 11th Conference on the French Society of Electrostatics SFE 2018, Aug 2018, Grenoble, France. <hal-02007212>

HAL Id: hal-02007212

<https://hal.science/hal-02007212v1>

Submitted on 29 Jan 2025

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Development of an experimental setup in order to measure space charges in dielectric liquids

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Abstract— When a liquid is put into contact with a solid, charges appear at the interface solid/liquid, resulting in the formation of an Electrical Double Layer (EDL). Many studies have been made to measure the EDL charge, but its theoretical distribution has not been confirmed experimentally yet, as so far there has been no method allowing direct measurement of space charge profiles in dielectric liquids. In this work, we present a measurement bench designed and developed to measure space charges in liquid samples using the principle of the Thermal Step Method. Results obtained on cyclohexane in liquid and solidified state are shown and commented.

Keywords— Electrostatic, space charges

I. INTRODUCTION

Accumulation of space charge in dielectric liquids is often related to reliability issues in power components such as power transformers. Indeed, when a liquid is put into contact with a solid, two charge layers of opposite signs appear at the interface between the two materials. This Electrical Double Layer (EDL) is the result of electrochemical reactions occurring at the interface between the liquid and the solid. The effects of the EDL may be beneficial or may lead to electrostatic hazard. In both cases, studying the EDL is of great interest in term of industrial applications. Due to the ionic mobility, some of the charges diffuse from the interface toward the liquid and an equilibrium is reached after a time related to the diffusion velocity and to the duration of the reactions occurring at the interface.

In order to determine the amount of charge in the liquid, the technique of flow electrification is mainly used. When the liquid is set into movement, the charges of the diffuse layer move with the liquid flow and lead to the electrification phenomenon and to the streaming current. The study of the streaming current versus the flow velocity allows estimating the EDL properties, particularly the value of the charge. However, such analysis is likely to underestimate the space charge density near the interface.

In the last decades, several non-destructive methods have been set up for measuring space charge in solid dielectrics [1]. Among them, the Thermal Step Method (TSM) has been widely used on a variety of insulating materials and structures, providing, as most of the thermal methods, significant measurement sensitivity and easiness.

In the present paper, we will present an experimental set-up developed for measuring electric charge in insulating liquids using the principle of the TSM. Theoretical aspects concerning the EDL and the TSM are first considered, then the design and the set-up of the experimental bench is described, and several results obtained with the bench are presented and discussed.

II. PROBLEMATICS AND THEORETICAL ASPECTS

A. Electrical Double Layer (EDL)

Stern [2] describes the Electrical Double Layer as two charged areas of opposite sign, one in the solid and one in the liquid. The one in the liquid is subdivided in two layers: the compact and the diffuse layer. It is widely believed and accepted that the compact layer has a thickness of the order of the ionic radius and it is composed of charges unable to move, but so far there is no experimental evidence on this matter.

The diffuse layer is developing from the compact layer toward the liquid bulk. The charge distribution in the diffuse layer is broadly modeled as a decreasing exponential with a maximum value $\rho_w = \rho_{wd}$ on the compact layer side (Figure 1). The value ρ_{wd} is considered to be reached after the physicochemical equilibrium between the liquid and the solid has been obtained. The diffuse layer thickness is related to the Debye length δ_0 :

$$\delta_0 = \sqrt{\frac{\varepsilon_0 \varepsilon_r D_0}{\sigma}} \quad (1)$$

where ε_0 and ε_r are the vacuum permittivity and the relative permittivity of the liquid, D_0 is the mean ionic diffusion coefficient of the liquid and σ its bulk conductivity.

The amount of charge in the diffuse layer is mainly estimated by flow electrification experiments [3]. However, the charges close to the interface cannot be approached with such experiments as the velocity of the

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This work is supported by the French Government program "Investissements d'Avenir" (LABEX INTERACTIFS, reference ANR-11 LABX-0017-01).

liquid near a motionless wall is nil, so these charges cannot be carried away by the liquid flow. New techniques are therefore needed to better characterize the amount and the distribution of the charges in the EDL.

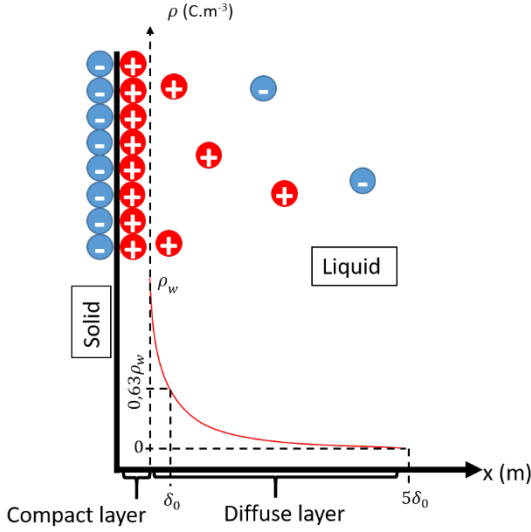


Figure 1: Schematic representation of the EDL

B. Thermal Step Method

The TSM is a non-destructive method to measure space charge, designed and proven on solid dielectrics. 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.

TSM principle for a flat sample is given in Figure 2. The sample is defined by its geometry (thickness d and surface S) and its dielectric properties (relative capacitance variation with the temperature α and permittivity ϵ). Because the sample is considered as infinitely flat and that thermal conduction is the main heat transfer mode in solids, the problem can be assumed as unidirectional.

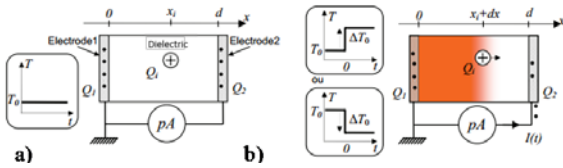


Figure 2: Principle of the TSM on a flat sample; sample at initial temperature (a) and after the thermal step (b)

The sample is short-circuited between the electrodes located at $x = 0$ and $x = d$, so when the 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. 2) measured by a current amplifier. By considering first order variations of the capacitance and by neglecting the conductivity of the dielectric, the resulting current can be written as [4]

$$I(t) = -\alpha C \int_0^d E(x) \frac{\partial \Delta T(x, t)}{\partial t} dx \quad (2)$$

where C , $E(x)$ and $\partial \Delta T(x, t)/\partial t$ are respectively the sample capacitance at the initial temperature T_0 , the

electric field distribution and the temperature distribution within the sample. In order to obtain high amplitude signals, it is preferable to have a thermal stimulus as close as possible to a thermal step. In order to obtain the space charge distribution $\rho(x)$, the electric field is extracted from eq. (2) [5], then the Poisson law is used:

$$\rho(x) = \nabla \cdot (\epsilon E(x)) \quad (3)$$

For a cylindrical sample of radii R_{int} and R_{ext} , the expression of the thermal step current is [5]:

$$I(t) = -\alpha C \int_{R_{int}}^{R_{ext}} E(r) \frac{\partial \Delta T(r, t)}{\partial t} dr \quad (4)$$

For the experimental setup, a cylindrical cell containing the liquid under test has been designed and used, as presented herein after.

III. DESIGN AND SET-UP OF THE MEASUREMENT BENCH

A. Measurement cell

In order to use the TSM on liquid samples, a measurement cell has been designed. This cell is composed of three cavities enclosed by stainless steel cylinders (Figure 3).

The inner cavity is used for the circulation of the cooling fluid that provides the thermal step and the outer one is filled with air. This outer cavity acts as a buffer to maintain the initial temperature on the outer side, avoiding heat transfer toward the environment. The middle cavity containing the sample is 3 mm wide in order to avoid interactions between the two EDL developed at the internal and external areas of the cavity. This value of the sample thickness has been set by considering that the EDL profile has reached $\rho = 0 \text{ C.m}^{-3}$ at $x = 5 \times \delta_0$, with δ_0 of the order of 200 μm .

Three temperature sensors are placed in the cell: two PT1000 probes (in the outer and the inner cylinder) and a type T thermocouple (in the middle of the liquid).

The height of the cell (125 mm) has been determined to have a sample capacitance of at least 100 pF (value known to allow obtaining measurable TSM signals in solid samples). Cylinders used to hold the sample are also used as electrodes.

B. Measurement bench

In order to have a set-up compatible with the experimental cell, an entire measurement environment has been designed (Figure 4). The TSM principle has been adapted both to solid and liquid states of the sample. For the solid state, liquid nitrogen is moved across the cavities (1) and (3) of the experimental cell to solidify very quickly the sample ($T_{liquid N_2} = -195^\circ\text{C}$). It is considered that space charges are frozen in their initial location before the solidification, as the time need for the charges to move is much longer than the solidification time, and the liquid conductivity also diminishes with the temperature. The range of working temperature was chosen between -50°C

and +60°C, allowing to work both in solid and liquid samples. The cryostat is composed of two tanks of 50 liters each, regulated in temperature.

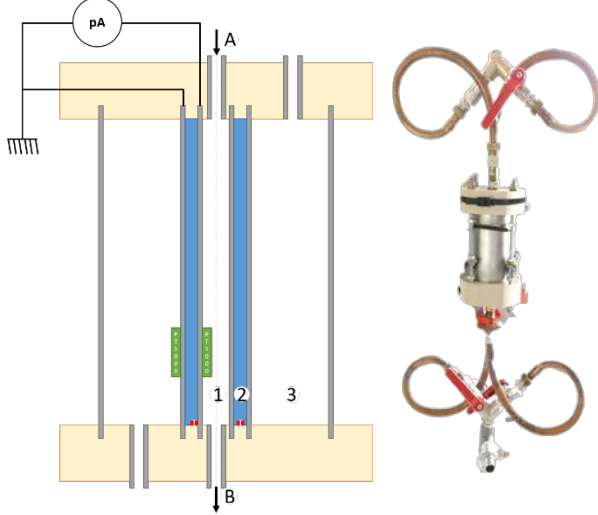


Figure 3: Representation and photograph of the TSM cell - (A) Cooling inlet, (B) Cooling outlet - (1) Cooling fluid cavity, (2) Cyclohexane cavity, (3) Air cavity – Temperature sensors: PT1000 probes in green and thermocouples in red

The tanks are connected to the cell through a hydraulic system according to the diagram from Figure 5. There are 3 functioning states. First, the tank at T_0 sends the cooling fluid to the cell, until its temperature reaches the temperature set point. In a second step, the second tank is connected to the system (disconnecting the first tank) and cool/heat pipes as close to the cell as possible: this is the free wheel step. Finally, the “temperature step” cooling fluid is sent to the cell to apply the thermal step to the sample.

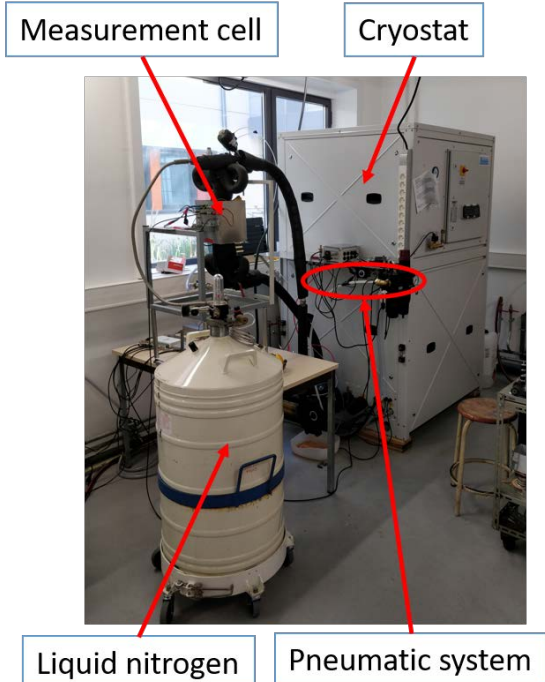


Figure 4: Photography of the measurement bench

The opening of the valves is made via a pneumatic distributor system in order to avoid as much as possible electromagnetic disturbances. A Labview program has been developed to manage the pneumatic distribution and to record signals from the temperature sensors of the cell and of the cryostat, as well as the TSM current measured by the current amplifier (Keithley 428).

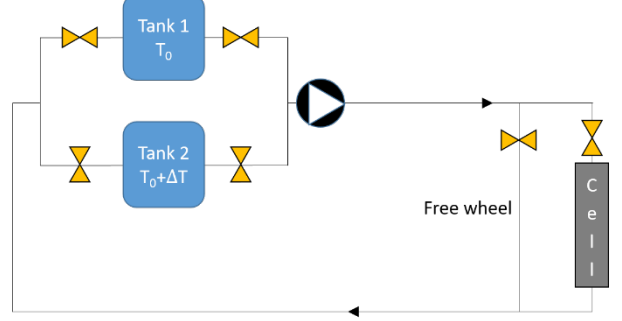


Figure 5: Hydraulic diagram of the measurement bench

IV. RESULTS AND DISCUSSION

A. Studied liquid

The liquid chosen for the study was cyclohexane (C_6H_{12}), an insulating liquid with a high melting point (6.47°C), in order to allow studies in both solid and liquid states in a reasonable range of temperatures. Its thermal conductivity worth $\lambda = 0.2 \text{ W.m}^{-1}.\text{K}^{-1}$, its specific heat $C_p = 1850 \text{ J.kg}^{-1}.\text{K}^{-1}$ [6] and its liquid state density ρ_m is defined by the following equation [7]:

$$\rho_m(T) = \frac{84.1595 \times 0.8908}{0.27396^{1 + \left(1 - \frac{T}{553.58}\right)^{0.2851}}} \quad (5)$$

The measurements that we performed with an Irlab LDTR-4 instrument gave an electrical conductivity $\sigma = 6.28 \times 10^{-13} \text{ S.m}^{-1}$ and a relative permittivity of $\epsilon_r = 2$.

B. Measurement conditions

Before filling the experimental cell with the liquid sample, the sample is cleaned with “used” sample liquid, and dried. A second cleaning is then made with “new” liquid, then the cell is dried with gaseous nitrogen to avoid the presence of water. In order to study the fully developed EDL, we left the liquid sample in contact with the cell for several hours before starting the measurements.

For solid state measurements, the sample is solidified (after the EDL has been fully developed) using liquid nitrogen and its temperature is maintained below the melting point. We wait for the sample to reach the initial temperature T_0 while the two electrodes are in short-circuit. We apply the temperature step while measuring the TSM current and temperature from the sensors in the cell. When the measurement is over, we reset to the initial temperature of the cell.

For liquid measurements, the procedure is the same except for the solidification phase and for the duration between two measurements. Indeed, it has been shown that thermal convection can occur during TSM on liquid samples and possibly affect the EDL [8]. To have a good reproducibility, several hours were left between successive measurements.

C. Solid state measurements

Figure 6 shows thermal step currents obtained with the cyclohexane solid sample with a thermal step between $T_0 = 0^\circ\text{C}$ and $T_0 + \Delta T = -30^\circ\text{C}$, by keeping the sample in solid state for all the presented measurements. The obtained signals are easily measurable (>1 nA) and have a reproducible shape. We also see that the amplitude is modified as and when the measurements are done (one hour separates each signal). By observing the temperature measurements, we notice that the temperatures are not exactly the same for each signal (Figure 7). The currents of higher amplitude are the ones with the higher temperature step, as we could expect from eq.(4).

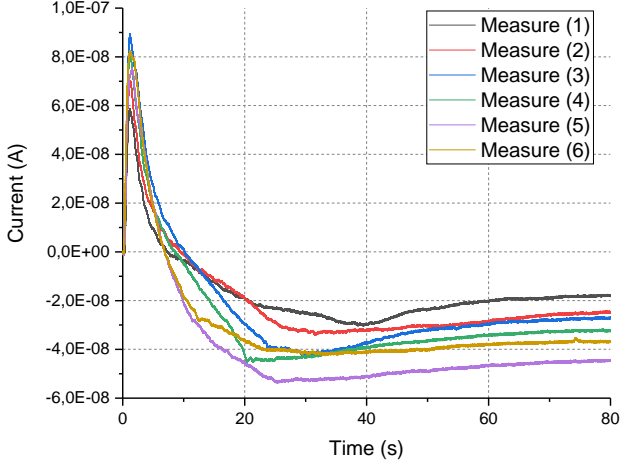


Figure 6: TSM experimental currents measured with a thermal step of -30°C on solid cyclohexane

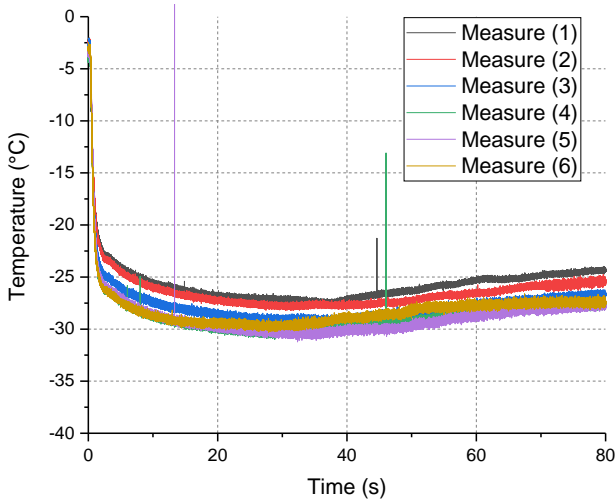


Figure 7: Temperature applied on the experimental cell for solid cyclohexane measurements

We notice that, after 10 seconds, the sign of the current is reverted, than the signals tend to go back to zero. This strongly suggests that the thermal wave has encountered an inversion of the electric field, which corresponds to expected results in terms of field behavior. However, if we compare those measures to numerical results obtained in the same conditions (temperature step, velocity of cooling fluid but with an EDL containing only

the diffuse layer) [8]), we observe a good fit at the beginning of the curves from Figure 8. On the other hand, the amplitudes do not match (a ratio of 30,000 differentiates experimental and numerical results). A hypothesis for this difference is the influence of the compact layer, which has not been taken into account in the numerical model. Indeed, the electric field within this layer is more important than the one in the diffuse layer and the local permittivity of the compact layer is possibly much higher than in the bulk.

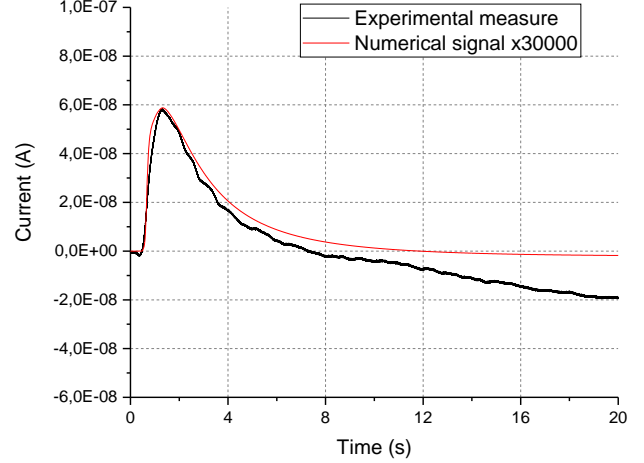


Figure 8: Comparison between a measured signal and a numerically simulated one

D. Liquid state measurements

Measurements of the same sample in liquid state have been made with an initial temperature of 40°C and a step temperature of 10°C (Figure 9). We observe signals with high amplitudes (similar to solid measurements), and we see that reproducibility is excellent.

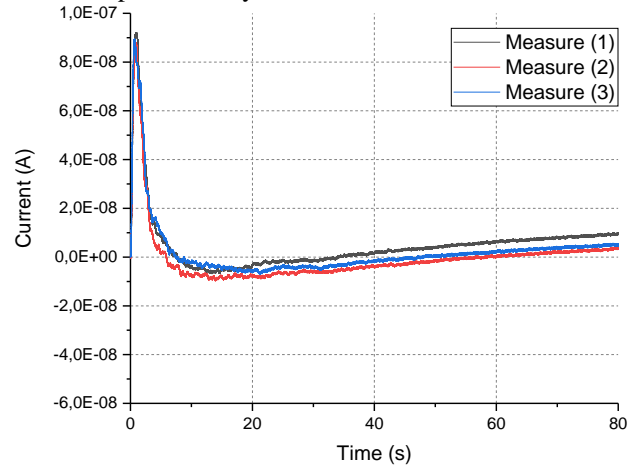


Figure 9: TSM experimental currents measured with a thermal step of -30°C on liquid cyclohexane

We notice that the amplitudes of the signals measured on the cyclohexane sample in solid and liquid states are comparable. In both cases, the sign reversal occurs after ten seconds. This tends to confirm that quickly solidifying

the sample does not modify the distribution of space charges.

This part of the signal (the first few seconds) is the more relevant part. Indeed, from the edge of the sample toward a few hundred micrometers, the thermal wave is important enough to give significant information on space charge at the interface. This will help to better understand the compact layer in terms of location and amount of space charges, but also to confirm theoretical models of the diffuse layer. By performing measurements with thermal steps applied on both edges of the sample, it should be possible to reconstruct the spatial distribution of the electric field at the interface (the model will lack precision at the middle of the sample, where the electric field should be low enough to be considered negligible).

V. CONCLUSIONS AND PERSPECTIVES

In this work, we have presented a measurement bench designed for non-destructive charge measurements in dielectric liquids, in order to study the Electrical Double Layer (EDL) in particular. The presented results, obtained on cyclohexane, have shown that the system is able to deliver measurable, high amplitude TSM signals from samples set both in liquid and in solidified state. The experimental results have been analyzed and confronted to numerical simulations in order to improve the physical modelling and to make it as relevant as possible.

An interesting way to proceed will be study liquids with different EDL (quantity of charge, polarity of charge). This will give information to complete the physical modelling and to progress toward the full comprehension and determination of the charge in the liquid.

ACKNOWLEDGMENT

The authors would like to acknowledge M. Le Gouaréguer for the measurements concerning the electrical properties

of cyclohexane made during his internship at the PPRIME Institute.

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