

# Measurement of electric charge in solid insulators by influence non destructive methods. The thermal step method

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Non-destructive methods for measuring electric charge and polarization in solid dielectric materials have been set up in recent years. Their common principle is related to the influence charges which appear at the electrodes of a sample containing space charge. When the material is submitted to an external disturbance, the influence charges vary, giving birth to a transient current or voltage signal, from which charge density and electric field distributions across the sample can be calculated. The use of adapted stimuli, which do not alter the electrical state of the sample, allows monitoring the time evolution of the charge. The most employed methods, based on pressure or thermal stimuli, are briefly reviewed in this paper. A thermal technique, namely the Thermal Step Method, is brought in sharper focus. Results obtained on materials and structures for electrical and electronic applications are presented.

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## 1. Introduction

Measuring electric charge in insulating materials has been an important research field for the last decades. Indeed, various studies addressing organic and mineral dielectrics during the 1960's and 1970's, which investigated the mechanisms and applications of electric charge accumulation and transport, have brought in focus the need for techniques allowing to quantify and localize the charge in materials and structures used in electrical and electronic engineering. This encouraged the development of several charge characterization techniques, which can be classified in "destructive" and "non destructive". The "destructive" techniques are so called because they change the electrical state of the sample by evacuating the charge or by modifying the charge amount. A widely-known method in this category is the thermally stimulated current technique [1-2], which consists of linearly heating a sample and measuring the "thermally stimulated" current appearing in the external circuit under the effect of charge release and dipolar disorientation. Generally, this type of method do not supply information about charge localization, but allows estimating the amount of charges and the energies needed to evacuate the different type of carriers and/or to disorient the dipolar species. As the samples are discharged or electrically modified after the measurements, destructive techniques do not allow surveying charge evolution with time.

"Non destructive" techniques do not evacuate the charges trapped within the material and allow determining their distribution across the sample. They present the advantage of allowing time monitoring of the charge. Used in conjunction with destructive techniques, they allow a

complete characterization of charge-related processes and phenomena. They are the subject of the present paper.

## 2. Principle of non destructive techniques for electric charge measurement in solid dielectrics

Developed since the beginning of the 1980's, most of these techniques are based on the application of a non uniform external stimulus to the insulating device and on the recording of current or voltage responses, which depend on the amount and distribution of the charge in the sample.

The physical principle of such a method is shown in Figure 1. Figure 1a) presents a flat insulating specimen containing a bulk charge noted  $Q_i$ , in the absence of any external stimulus. Because the sample is in electrostatic equilibrium, influence charges (noted  $Q_1$  and  $Q_2$ ) appear at electrodes, under the law of electrostatic influence. The application of a stimulus to one electrode (e.g. a pulse or step of mechanical or thermal origin to the left electrode, Figure 1b) results in a relative displacement of the trapped charge with respect to electrodes combined to a non homogenous variation of the permittivity while the wave induced by the stimulus crosses the sample. This leads to a redistribution of the influence charges at electrodes, resulting either in the appearance of a transient current in the external circuit (if the sample is short-circuited, as in Figure 1) or in a transient modification of the voltage across the sample (in open circuit conditions). The transient signal lasts until the system has reached a new state of electrostatic equilibrium, i.e. until the stimulus

wave has crossed the sample. The analysis of the signal (which is recorded externally) allows calculating the electric charge and field distribution across the material.

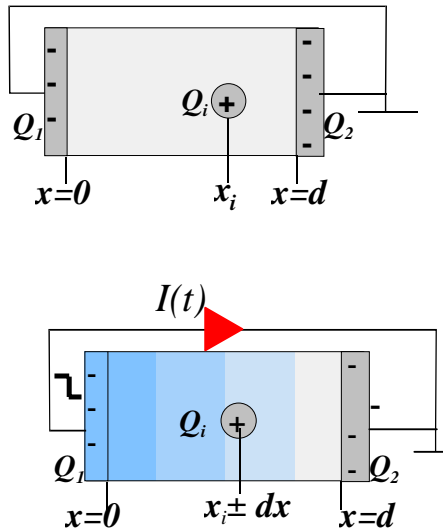


Fig. 1. Principle of non destructive influence techniques (example for a short-circuited specimen) a) Insulating sample containing an electric charge  $Q_i$  in the bulk, in the absence of the stimulus (electrostatic equilibrium); b) Insulating sample after the application of the stimulus on the left electrode.

Because the measured signal is due to the redistribution of influence charges, these techniques are also called “influence methods”. The most popular ones are based on thermal or mechanical stimuli. A Kerr effect technique using the birefringence induced by an electric field in an insulator was also set up [COE 93], but the application of this method is limited to transparent dielectric materials, particularly to those presenting a high electro-optical birefringence. The sensitivities of the presently available techniques are in the range  $0.001\dots 1 \text{ C/m}^3$  and their resolutions between 100 nm and 25  $\mu\text{m}$ , depending on the features of each technique, on the nature and of the thickness of the studied sample.

### 3. Thermal techniques

In this case, one surface of the sample is submitted to a sharp temperature variation, leading to a non uniform dilation (or contraction) of the material and to a local (inhomogeneous) variation of its permittivity. The thermal permittivity variation coefficients of common materials are very low (of the order of  $10^{-4}$  to  $10^{-6}$ ), but enough to provide the thermal techniques the best sensitivities ( $1 \text{ mC/m}^3$ ) and resolutions ( $< 0.1 \mu\text{m}$ ), the reason being an efficiency of the thermal transfer much better than those of energy transfers of mechanical origin. Thermal methods are usually “slow”, the duration of the measured signals

being greater than 1 s. Consequently, the time resolution of these techniques is limited to several dozens of seconds. In contrast, thermal methods provide significant signal-to-noise ratios and excellent reproducibility.

The so-called “thermo-expansion” signal which is measured and recorded has an expression of the form (for a short-circuit measurement):

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

where  $S$  is the surface of the sample,  $d$  is the thickness of the sample,  $E(x)$  the electric field distribution across the sample in the thickness direction (which is a depth coordinate:  $x \in [0, d]$ ),  $\alpha(x,t)$  is a function containing the laws of variation of thickness and permittivity with temperature, and  $\partial \Delta T(x,t)/\partial t$  is the law of variation of the temperature with time at the abscissa  $x$ . Using the recorded current and knowing the material parameters and the temperature repartition, the electric field (and charge) distribution can be calculated. Solving the above integral equation may be a delicate task, but the available mathematical techniques have improved significantly in recent years and allow obtaining remarkable results.

The essential difference between the thermal methods lies in the way of generating the localized temperature variation, which has a direct impact both on the amplitude of the thermal perturbation (and consequently on the measurable thickness) and on the measurement devices to be set up. According to the technique used for producing the “thermal wave”, one can distinguish:

- the Thermal Pulse Method (TPM) [3-4], which is based on the application of a thermal pulse (obtained by laser heating) on a side of the sample with electrodes in open circuit. The pulse diffuses through the sample by local dilation of the material. This method provides an excellent resolution ( $< 1 \mu\text{m}$ ) and is particularly suited for studying thin samples ( $< 100 \mu\text{m}$ ), as the heat pulse dampens in thick materials;
- the Laser Intensity Modulation Method (LIMM) [5] consists of heating one face of the sample with a laser beam whose intensity is modulated at variable frequency, and to measure the resulting pyroelectric current. For the same reasons as the TPM, the LIMM is suited for thin samples ( $< 100 \mu\text{m}$ ), with a resolution below  $10 \mu\text{m}$ ;
- the Focused Laser Intensity Modulation Method (FLIMM) and the Focused Laser Acoustic Modulation Method (FLAMM) [6-7] use the same principle as the LIMM, except that the laser beam is focused and can be moved, thus providing a three-dimensional cartography of the sample. Their resolutions and thickness ranges are of the same order as for the LIMM;
- the Thermal Step Method (TSM) [8-9] consists of measuring the thermo-dilation current response of the sample after applying a temperature step on

one of its faces. Because of the significant thermal energy transferred to the material, this last method is suited for both thin and thick samples (100 nm to 25 mm) of flat or cylindrical shape. As for the other techniques, its resolution depends on the thickness of the specimen and can be as high as 100 nm (and even better) for the case of very thin samples as the ones used in microelectronics. A version of this technique called the Alternative Thermal Wave Method (ATWM) [10] has been set up more recently and uses a sinusoidal thermal perturbation for analyzing samples with thicknesses of the order of 100  $\mu\text{m}$ .

#### 4. Pressure techniques

Pressure techniques use an elastic wave for quantifying the charges trapped within the material. Because of the high sound velocity in solid insulators, pressure methods are “fast” methods: the duration of the response signal does not exceed several microseconds. This makes them particularly attractive for surveying the dynamics of the charges, especially under periodical electrical stress and under thermal stress. In contrast, the energetic efficiency of the stimulus being lower than that of thermal methods, the signal-to-noise ratio is usually low. Consequently, obtaining reproducible results requires quite often an averaging of several dozens (to hundreds) successive measurements. The spatial resolution of pressure techniques can be as high as 1  $\mu\text{m}$ , with a sensitivity of the order of 10  $\text{mC/m}^3$ . Nowadays, the most popular techniques are:

- methods using a pressure wave [11-12], which have as principle the compression of the material by a transient wave with a straight front. Different means are employed for generating the pressure wave (shock wave tubes, piezoelectric transducers, laser pulses). The most developed technique is the Laser Induced Pressure Pulse (LIPP) method [12], where a laser pulse of several nanoseconds is applied for creating the elastic wave. The equations relating the measured signal to the electric field across the sample are similar to those of the thermal methods. For instance, the current measured from a short-circuited sample reads:

$$I(t) = \lambda CG(\varepsilon) \int_0^d E(x) \frac{\partial P(x,t)}{\partial t} dx \quad (2)$$

where  $d$  is the thickness of the sample,  $\lambda$  is the compression ratio of the material,  $C$  is the capacitance of the sample before compression,  $G(\varepsilon)$  is a coefficient related to the variation of the permittivity with pressure,  $E(x)$  is the electric

field distribution across the sample in the thickness direction and  $\partial \Delta P(x,t)/\partial t$  is the law of variation of the pressure with time at the abscissa  $x$ . From the measured current, if the material parameters and the pressure repartition are known, the electric field (and charge) distribution can be calculated. If the attenuation of the pressure wave is negligible (e.g., in thin samples), a reading of the charge distribution can be obtained without mathematical treatment, fact that constitutes a significant advantage. However, taking into account the problems related to the reflection of the pressure wave at interfaces requires often a complex signal analysis and the use of deconvolution techniques.

- the Pulsed Electroacoustic Method (PEA) [13] stands on a slightly different principle, consisting of placing a piezoelectric transducer on one side of the sample and on exciting the specimen with a short high voltage pulse. The electrostatic forces generated by the charges under the effect of this stimulus give birth to a pressure wave which reaches the transducer after a time  $t_A$ . The value of  $t_A$  allows to determine the position of the charges. The PEA method is a widespread technique and seems to be well suited to specimens of thickness exceeding 100  $\mu\text{m}$ .

Positive comparisons have been made between thermal methods, as well as between the LIPP and the PEA. Comparing thermal and pressure techniques was found to be a quite difficult task, particularly because of different sensitivities and dynamics that each class of methods possesses.

#### 5. The thermal step method (TSM): theory and set up Governing equations in short-circuit conditions

Let us come back to the insulating sample from Figure 1a, of thickness  $d$  and of surface  $S$  and provided with two conducting electrodes of abscissa  $x = 0$  and, respectively,  $x = d$ . The material is considered as homogeneous and infinitely flat ( $d \ll S^{1/2}$ ); thus, the electric field is assumed as constant in a plane parallel to electrodes. The sample is placed in short circuit at a temperature  $T_0$ .  $Q_i$  is the electric charge contained in the bulk of the sample.

As explained earlier, because the system consisting of sample, electrode and conducting wire is in electrostatic equilibrium, the charge  $Q_i$  situated at the abscissa  $x_i$  induces at electrodes charges, noted  $Q_1$  and  $Q_2$ , under the principle of electrostatic influence. The dependence of the induced charges on the space charge  $Q_i$  and on the geometrical dimensions of the sample can be obtained

with the aid of the short circuit condition  $\int_0^d E(x) dx = 0$ ,

of the electric charge conservation law  $Q_1 + Q_2 + Q_i = 0$  and of Gauss' theorem:

$$Q_1 = -\frac{d-x_i}{d}Q_i, \quad Q_2 = -\frac{x_i}{d}Q_i \quad (3)$$

Let us now consider that one face of the specimen suffers a sharp temperature variation, i.e. a temperature step  $\Delta T = T - T_0$  is applied to the left face of the specimen (Figure 1b). The diffusion of the thermal front  $\Delta T(x, t) = T(x, t) - T_0$  across the sample will result in local variations of the permittivity and of the abscissae (expansion or contraction), which can be written (in first order):

$$\varepsilon = \varepsilon_{T_0} (1 + \alpha_\varepsilon \Delta T), \quad dx = dx_0 (1 + \alpha_d \Delta T) \quad (4)$$

where  $\alpha_\varepsilon$  is the coefficient of variation of the material permittivity with the temperature and  $\alpha_d$  its thermal expansion coefficient,

$$\alpha_\varepsilon = \frac{1}{\varepsilon} \frac{d\varepsilon}{dT}, \quad \alpha_d = \frac{1}{x} \frac{dx}{dT} \quad (5)$$

These variations of the abscissae and permittivity provoke a modification of the influence charges, which can be written as:

$$Q_1(t) = -Q_i \frac{\int_{x_i}^d \frac{dx}{\varepsilon(x, t)}}{\int_0^d \frac{dx}{\varepsilon(x, t)}}, \quad Q_2(t) = -Q_i \frac{\int_0^{x_i} \frac{dx}{\varepsilon(x, t)}}{\int_0^d \frac{dx}{\varepsilon(x, t)}} \quad (6)$$

With the previous notations, and by putting:

$$\alpha = -\frac{1}{C} \frac{dC}{dT} \approx \frac{1}{x} \frac{dx}{dT} - \frac{1}{\varepsilon} \frac{d\varepsilon}{dT} = \alpha_d - \alpha_\varepsilon, \quad (7)$$

we get for  $Q_2(t)$  the expression :

$$Q_2(t) = -Q_i \frac{x_i}{d} \left[ 1 + \frac{\alpha}{x_i} \int_0^{x_i} \Delta T(x, t) dx - \frac{\alpha}{d} \int_0^d \Delta T(x, t) dx \right] \quad (8)$$

The variation of the induced charges is reflected in the external circuit by the appearance of a current  $I(t)$ , called **thermal step current** :

$$I(t) = -\frac{dQ_2(t)}{dt} \quad (9)$$

If the value of the thermal step current and the spatial distribution of the temperature are known for every instant  $t$ , the value of the charge  $Q_i$  and its position  $x_i$  can be determined.

In general, if we define by  $\rho(x) = \partial D(x)/\partial x = \varepsilon \partial E(x)/\partial x$  the volume charge density at the abscissa  $x$  in a flat layer of thickness  $dx$ , it can be shown by integration on the entire sample [8] that the total current in the external circuit has the expression:

$$I(t) = -\alpha C \int_0^d E(x) \frac{\partial \Delta T(x, t)}{\partial t} dx \quad (10)$$

where  $E(x)$  is the electric field at the abscissa  $x$  and  $C$  the capacitance of the sample before the application of the thermal step.

As shown by equation (10), the thermal step current amplitude is a function of the amount of charge stored in the sample, of its distribution and of the material parameters. In practice, the value of the thermal step current has a value between 1 pA and 1  $\mu$ A.

It is to note that, in practice, the imperfect thermal contact between the sample and the radiator which produces the temperature variation attenuates and delays the thermal step, a fact which cannot be neglected while processing the current. To simplify the calculations, this effect can be taken into account by a so-called “equivalent-added thickness”  $x_0$ : the thickness  $x_i$  of the different interfaces, of mean reciprocal thermal diffusivity  $W_i$ , is considered as the equivalent of a layer  $x_0$  having the reciprocal thermal diffusivity  $W_0$  of the insulator:  $W_i x_i^2 = W_0 x_0^2$ . The limits of the thermal step current integral (0 and  $d$ ) can be replaced by  $x_0$  and  $x_0 + d$  (the electric field across  $x_0$  is assumed to be zero):

$$I(t) = -\alpha C \int_{x_0}^{x_0+d} E(x) \frac{\partial \Delta T(x, t)}{\partial t} dx \quad (11)$$

**Experimental set up.** In practice (Figure 2), the thermal step is created by a cold liquid circulating within a radiator in contact with the sample (or by a transient current which heats a resistive material under the sample). The electrodes are connected through a current amplifier. The latter is connected to a computer, which records the current and does the numerical data processing. After performing a measurement, the sample can be reheated using a warm liquid, and the experiment can be repeated.

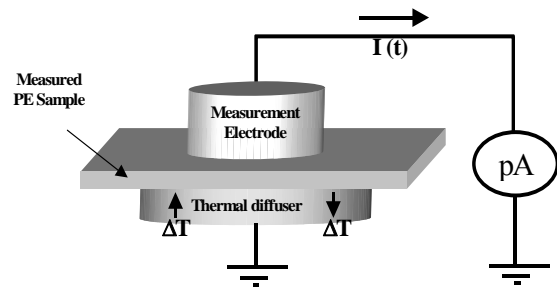


Fig. 2. Experimental set up of the TSM in short-circuit conditions

**Measurements under applied dc field.** The principle of the method also stands if a dc voltage is applied to the sample (“under field” measurement). For low applied voltages and low conduction or polarization current across the sample, an experimental set up using an electronic circuit (often integrated in commercially available current amplifiers) can be employed to polarize the sample during the measurement. This is particularly suitable for thin samples with high resistivity (e.g. insulating layers in electronic components, as it will be shown herein after).

However, if a high voltage is to be applied to the sample, the current amplifier must not be in contact with the high voltage source; and if a set up with the current amplifier placed between the sample submitted to the high voltage and the ground is used (as for, e.g., conduction current measurements), the conduction and the polarization currents are likely to mask the thermal step current. A solution to these problems is a “compensation sample” [9], of identical dimensions as the measured specimen and placed oppositely to the latter (Figure 3). By connecting one side of the compensation sample to the current amplifier and the other to the measured specimen via an electrode, a “double capacitor” is obtained. A voltage can then be applied to the middle electrode, and the thermal step to the measured sample. The TSM current will be acquired via the “compensation sample”. Using two identical specimens offers the considerable advantage of compensating the polarization and conduction currents which can occur under high fields; the measured current is due solely to the internal field of the measured sample.

The measurement “under applied field” is made in two steps (Fig. 4):

- during conditioning, the voltage is applied to the middle electrode and the current amplifier is short circuited. Thus, the two samples constitute two identical capacitances placed in parallel with respect to the voltage source (Fig. 4a).

- in order to prevent the transport of induced charges at electrodes via the HV generator rather than via the current amplifier, the power source has to be disconnected during the measurement (Figure 4b). The thermal step current is then measured by exciting thermally the studied specimen (in contact with the thermal diffuser), while the current amplifier is connected to the compensation sample. This time, the two samples are in series with respect to the current amplifier, and the short-circuit conditions are fulfilled.

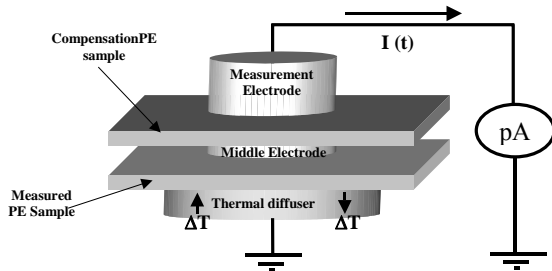


Fig. 3. Principle of the “double capacitor”.

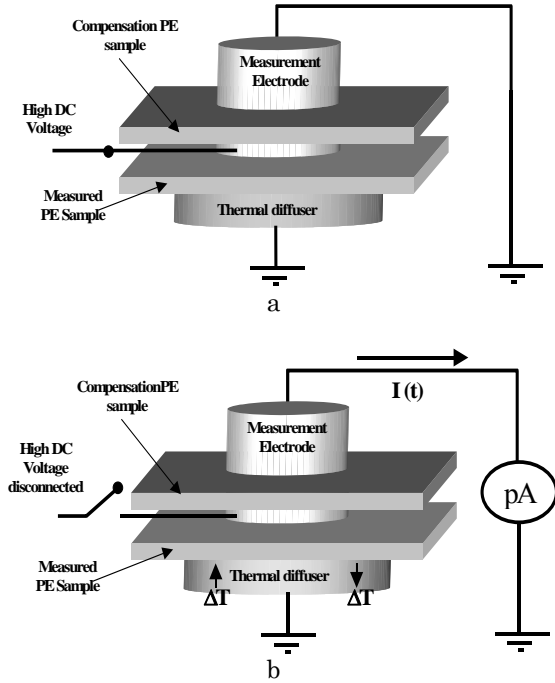


Fig. 4. Thermal step method under applied dc field; a) Application of the high voltage, b) Thermal step measurement

The thermal step current in the external circuit is:

$$I(t) = -\alpha C_2 \int_{x_0}^{x_0+d} E(x) \frac{\partial \Delta T(x,t)}{\partial t} dx \quad (12)$$

where  $C_2$  is the capacitance seen by the current amplifier. During the measurement, the two samples are in series, and, if they are identical,  $C_2=C/2$ . If no voltage is applied to the samples (“voltage off”), the thermal step current should be the half of that given by a measurement made on the thermally excited sample in short-circuit conditions, where  $C_2=C$ .

A particular and interesting case is that of low applied fields, when there is no charge injection in the sample, the electric field can be considered as constant throughout the material:  $E=V/d$ , where  $V$  is the applied voltage. In this case, the thermal step current (12) becomes directly proportional to the applied voltage:

$$I(t) = -\alpha C_2 \frac{V}{d} \int_{x_0}^{x_0+d} \frac{\partial \Delta T(x,t)}{\partial t} dx \quad (13)$$

This current can be used for calibration purposes.

The calibration can also be performed if the measured sample already contains space charge giving a residual field  $E_c(x)$ . Considering the externally applied electric field  $E_e=V/d$  (which do not modify the space charge distribution) externally, the total field in the sample

becomes  $E(x)=E_r(x)+E_e$ . Equation (12) can then be written as:

$$\begin{aligned} I(t) &= -\alpha C_2 \int_{x_0}^{x_0+d} [E_r(x) + E_e] \frac{\partial \Delta T(x,t)}{\partial t} dx \\ &= -\alpha C_2 \int_{x_0}^{x_0+d} E_r(x) \frac{\partial \Delta T(x,t)}{\partial t} dx - \alpha C_2 E_e \int_{x_0}^{x_0+d} \frac{\partial \Delta T(x,t)}{\partial t} dx \\ &= I_0(t) - \alpha C_2 \frac{V}{d} \int_{x_0}^{x_0+d} \frac{\partial \Delta T(x,t)}{\partial t} dx = I_0(t) + I_f(t) \end{aligned} \quad (15)$$

where  $I_0(t)$  is the current due to the residual space charge and  $I_f(t)$  the current due to the external field. In other words, the total current should be the sum of the "voltage off" current and the "voltage on" current.

$$I_D(t) = I_+(t) - I_-(t) = -\alpha C \int_{x_0}^{x_0+d} E_e \frac{\partial \Delta T(x,t)}{\partial t} dx = -\alpha C E_e \int_{x_0}^{x_0+d} \frac{\partial \Delta T(x,t)}{\partial t} dx, \quad (18)$$

which is twice the value from equation (13). Thus, by measuring this calibration current and by knowing the parameters before the integral and the sample thickness  $d$ , the last equation can be used for estimating  $x_0$  and the thermal diffusivity of the sample, on which the temperature diffusion depends.

**Temperature variation across the sample.** To determine  $E(x)$ , the thermal step current must undergo numerical treatment, which requires a perfect knowledge of the temperature derivative  $\partial \Delta T(x,t)/\partial t$  at every moment and for every abscissa. The geometry of the plate sample involves heat transfer taking place mainly perpendicularly to the surface of the sample (in the  $x$ -direction). Then  $\partial \Delta T(x,t)/\partial t$  is given by the heat equation in plane geometry :

$$\frac{\partial^2 \Delta T(x,t)}{\partial x^2} = W \frac{\partial \Delta T(x,t)}{\partial t} \quad (19)$$

where  $W = \mu c/\lambda$  is the reciprocal of the thermal diffusivity of the material, with  $\mu$  the volume density of the insulator,  $c$  its specific heat and  $\lambda$  its thermal conductivity. Boundary conditions must be associated with the heat equation (19) in order to obtain a solution for  $\Delta T(x,t)$ .

**Calculation of electric field and charge distribution.** The TSM was one of the first techniques to give electric field and space charge values in polymeric samples and in insulated power cables. The fundamental problem, as for all the others influence methods using a

Let us now note  $I_+$  the current from (14) obtained by applying  $E_e = V/d$ :

$$I_+(t) = -\alpha \frac{C}{2} \int_{x_0}^{x_0+d} [E_r(x) + E_e] \frac{\partial \Delta T(x,t)}{\partial t} dx \quad (16)$$

The application to the specimen of an external field with the same value as  $E_e$  but of opposite polarity ( $-V/d$ ) will give a thermal step current  $I_-$  such as:

$$I_-(t) = -\alpha \frac{C}{2} \int_{x_0}^{x_0+d} [E_r(x) - E_e] \frac{\partial \Delta T(x,t)}{\partial t} dx \quad (17)$$

The difference between the currents obtained by the two "under field" measurements will then be:

thermal or pressure stimulus, is solving the integral equation of the acquired signal. During the last years, several calculation techniques have been developed, according to the boundary conditions than can be associated to the heat equation (one or two thermal sources). These processing techniques, based on Fourier series, successive differentiations or matrix inversion, are detailed in [8-9, 14-15]; we will only present here the main lines of several approaches.

A general approach consists of writing the thermal step current equation in a matrix form, which allows obtaining the field distribution by inverting the temperature matrix. This often involves treating an ill-conditioned matrix, which may require the use of regularization techniques, or reducing the error resulting after numerical inversion by iterations [14].

In the case of flat samples with thicknesses from several tenths of microns to several millimeters, the thermal source (due to sharp arrival of a cold or warm liquid in a small thermal diffuser) is supposed to supply a thermal step at  $x=0$  (origin). Solving the heat equation with this one-source model (infinite semi-plane) gives:

$$\frac{\partial \Delta T(x,t)}{\partial t} = \Delta T \frac{x}{t} \left( \frac{W}{4\pi t} \right)^{\frac{1}{2}} \exp \left[ -\frac{Wx^2}{4t} \right] \quad (20)$$

with  $W$  the reciprocal of the thermal diffusivity of the sample. This stimulus, which has the appearance of a wave, moves slowly across the insulator. Its most advanced position is noted  $A(t)$  in Figure 5.

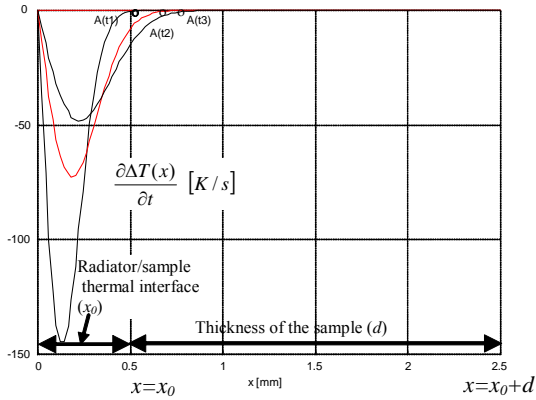


Fig. 5. Progression of the thermal wave across the insulating sample.

By approximating the expression of the temperature wave with a polynomial function of order  $m$ , an expression of  $E(A)$  can be obtained by deriving  $m$  times the thermal step current. This approach has the advantage of simplicity and fast processing, on the expense of sensitivity to measurement noise which limits the value of  $m$  to 2 or 3.

If we consider now that there is a second temperature source at the other side of the sample (corresponding to a massive electrode), placed at the abscissa  $x_0+d$  and maintained at room temperature during the experiment, the solution of the heat equation is:

$$\frac{\partial \Delta T(x,t)}{\partial t} = \left( \frac{2\Delta T_0 \pi D}{L^2} \right) \sum_{n=1}^{\infty} n \exp\left(-t/\tau_n\right) \sin(n\pi x/L) \quad (21)$$

where  $L = x_0+d$  and  $\tau_n = L^2/(D n^2 \pi^2)$ . This form of the stimulus leads to putting the electric field under the form of a Fourier series:

$$E(x) = \sum_{n=1}^{\infty} A_n \sin(n\pi x/L) \quad (22)$$

Unfortunately, this natural form can give the electric field only across a fraction of the sample, as  $E(x) = 0$  for  $x = L$ , which is not realistic. A “pseudo-Fourier” series, non nil at  $x = L$ , can be used [15]. In this approach,  $E(x)$  is developed in a co-sinus and sinus series, by taking the center of the sample as origin for the overall thickness:

$$E(x) = \sum_{n=0}^{\infty} A_n \cos(n\pi(x-l)/d) + \sum_{n=0}^{\infty} B_n \sin(n\pi(x-l)/d) \quad (23)$$

where  $l = x_0+d/2$ . By introducing this field expression in the thermal step equation, then by integrating, it is possible to obtain by regression 7 to 14 terms of the series. An example of resolution is shown in Figure 6. Good results have also been obtained in cylindrical geometry

(application to cables), with a significant noise level (up to 5%) [15].

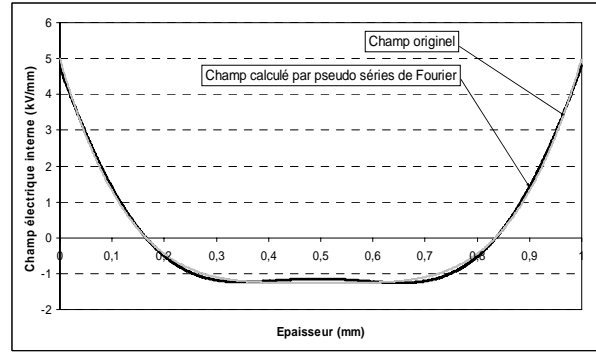


Fig. 6. Example of field calculation by pseudo-Fourier series (grey line: original field, black line: calculated field, x-axis: thickness of the sample (mm), y-axis: electric field (kV/mm)).

In all the quoted cases, the space charge distribution across the sample is obtained from the electric field via the Poisson equation:  $\rho(x) = dD(x)/dx = \varepsilon dE(x)/dx$ .

## 6. Applications

**Influence of molar weight and cooling rate on the presence of space charge in polymeric samples.** In order to study the appearance of space charge at manufacturing, several samples were press-molded from extruded tapes of polyethylene base resins. Four flat samples of same density ( $\sim 960\text{kg/m}^3$ ) and thickness (2 mm), but with different molar weights  $M_w$  were evaluated. We notice from Figure 7 the space charge distribution is symmetrical for all the samples and that the space charge seems directly related to molecular weight: the higher the molecular weight, the more space charges. On the other hand, microscopic observation of samples having the same density but different molecular weight (Figure 8) leads to the conclusion that molecular weight acts on the size of spherulites: the higher the molecular weight, the smaller the spherulites are. In can be concluded that the smaller the spherulites, the greater the space charge. This conclusion is in agreement with the theory of *Ikezaki et al.* [16], who believes space charge is mainly trapped in the core of spherulites and in the amorphous inter-spherulitic regions, which contain many defects and impurities.

Fig. 9 shows the space charge measured on two samples with  $M_w = 100000$  g/mol cooled differently. The solid line corresponds to the charge distribution measured in a sample cooled symmetrically, whilst the dashed line corresponds to a sample cooled asymmetrically. For the latter sample, one side was rapidly cooled to room temperature by a water circuit (circa 40 K/min), whereas the other side was cooled significantly slower due to the thermal inertia of the press-molding system (1 K/min). It comes out that an asymmetrical cooling induces an asymmetrical distribution of space charges: the side cooled

rapidly exhibits more space charge than the one cooled slowly. A microscope inspection also shown that that a rapid cooling led to small spherulites.

These results show that press-molded polyethylene plates exhibit space charge even without prior application of an electric field. This has also been observed on most polymers. Thus, the appearance of charges regions likely occurs during the solidification of the material, the thermal gradient existing between the bulk and the sides of the sample during cooling being probably responsible for the drift of the negative charges toward the bulk.

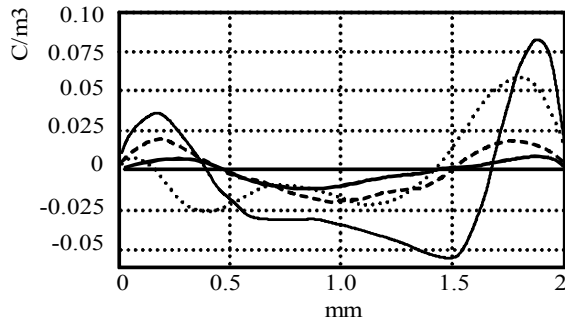
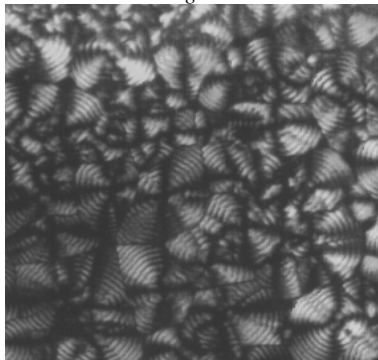
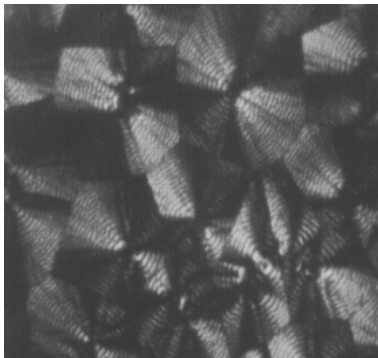


Fig. 7. Space charge distribution in polyethylene samples with different molecular weights ( $M_w$ ) after manufacturing. Solid line:  $M_w = 100000$  g/mol, dotted line:  $M_w = 71000$  g/mol, dashed line:  $M_w = 59000$  g/mol, bold solid line:  $M_w = 48000$  g/mol.



a



b

Fig. 8: Photomicrograph of the sample with  $M_w = 100000$  g/mol (a) and of the sample with  $M_w = 48000$  g/mol (b) (slow cooling).

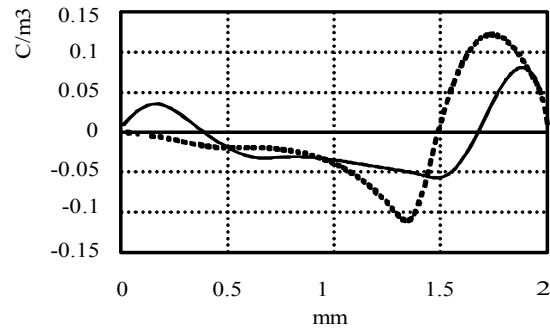


Fig. 9. Influence of the cooling rate on the space charge distribution in polyethylene samples (Solid line: symmetrical cooling, dashed line: asymmetrical cooling)

**Charge accumulation under ac and dc field.** The following figures show electric field and space charge distributions obtained in cross-linked (XLPE) polyethylene samples submitted to high dc and ac fields at different temperatures (45°C, 60°C and 90°C). It comes out that high internal fields establish in the samples submitted to high dc stress, especially at 60°C. This means that the overall field (applied + remnant) can exceed locally the applied field in a significant manner, resulting in local overstress which accelerates material degradation and may lead to early breakdown. Thus, the space charge accumulation phenomenon is one of the major present challenges concerning the design of high voltage dc materials and accessories, and receives significant attention from cable manufacturers and utilities. An important research direction in this area is producing insulating materials with field and temperature-controlled conductivity, able to prevent space charge accumulation by evacuating the charge periodically.

The accumulated space charge exhibited by the ac-conditioned samples is much lower, even if the conditioning time is much higher than in dc at a similar constraint [18]. This is due to the fact that in ac most of the carriers injected at electrodes during a half cycle are ejected during the next half cycle. The observed charge corresponds to the fraction of the carriers trapped at levels deep enough to retain them when the field is inverted: they are the image of deep traps ( $> 1\text{eV}$ ), corresponding to important structural defects. On one hand, this charge can contribute to ac degradation (e.g. by increasing the local energy of the dielectric and/or by promoting localized fatigue mechanisms), and, on the other hand, its presence can be a marker of the aging state of the insulation. As accumulation of steady charge in ac is related to the weakening of the dielectric structure, its survey can help to assessing the state of the insulator.



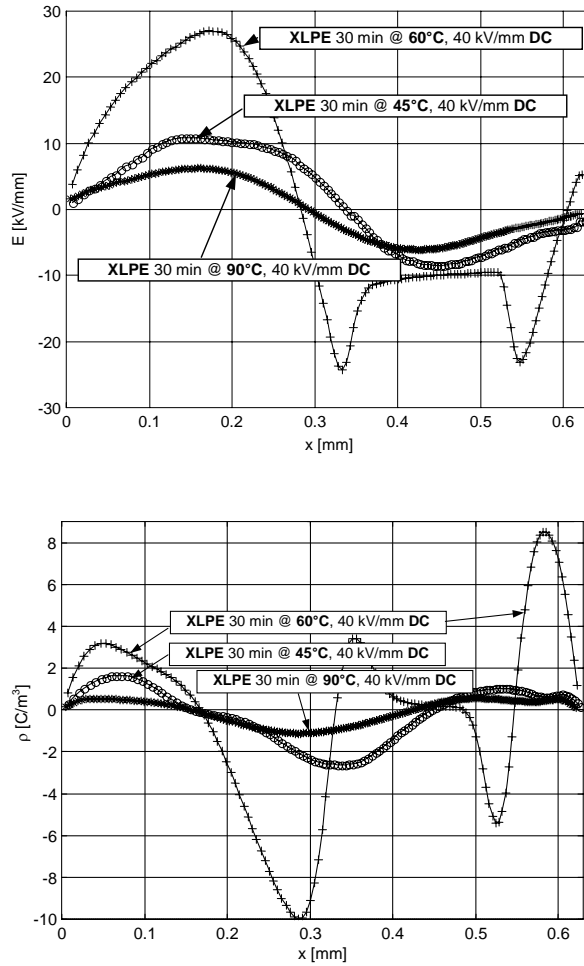


Fig. 10. Distribution of electric field and space charge in dc-conditioned XLPE samples

Both in ac and dc, there is a maximum charge accumulation for a particular value of the temperature (near 60°C, in the discussed case). This indicates a balance between carrier injection and conduction; for certain values of the temperature, the amount of injected charge can exceed significantly the charge released by conduction. As discussed earlier, this “critical temperature”, which is material-dependent and may change during operation, can be a factor for increasing insulating systems reliability, as periodic increases of the operating temperature of the material above the “critical value” facilitate the removal of previously accumulated charge.

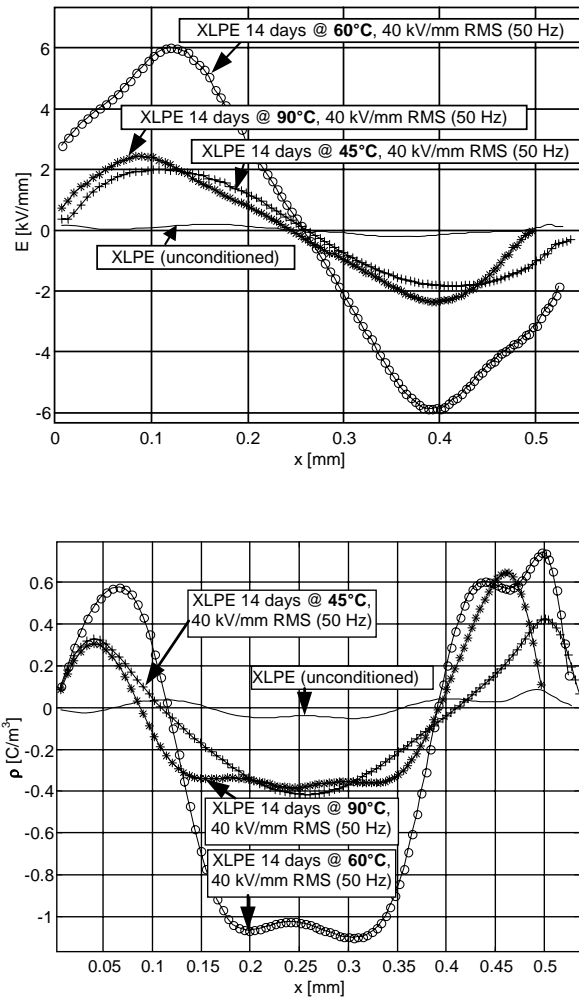


Fig. 11. Distribution of electric field and space charge in ac-conditioned XLPE samples.

**Charge measurement in thin layers used microelectronics.** Recent work [19-20] have shown the possibility of using the TSM for characterizing thin layers and structures for microelectronics, from 100 nm to 2 nm. The aim is to bring more information on the materials and structures by using the high sensitivity of the TSM. An example of application of the TSM to a planar metal-oxide-semiconductor biased structure is presented below (Figure 12). There is charge in the structure at the Si substrate level, at the Si/SiO<sub>2</sub> interface and in the SiO<sub>2</sub> insulating oxide.

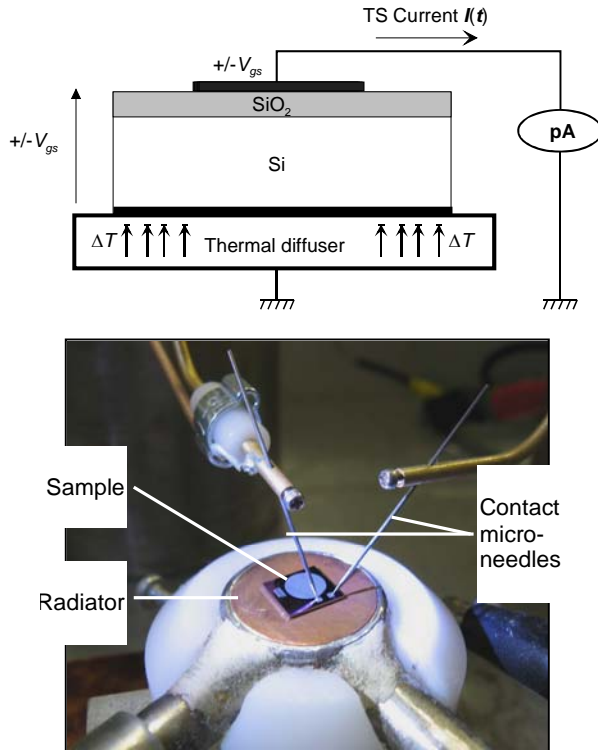


Fig. 12. Experimental set up for TSM on a biased metal-oxide-semiconductor structure

In this case, the measured thermal step current varies with respect to the applied voltage, as the space charge region at the Si/SiO<sub>2</sub> interface is strongly dependent of the gate voltage (Figure 13). By using the measured thermal step current, possibly in conjunction with capacitance voltage measurements, the different charges in the structure can be obtained (Figure 14).

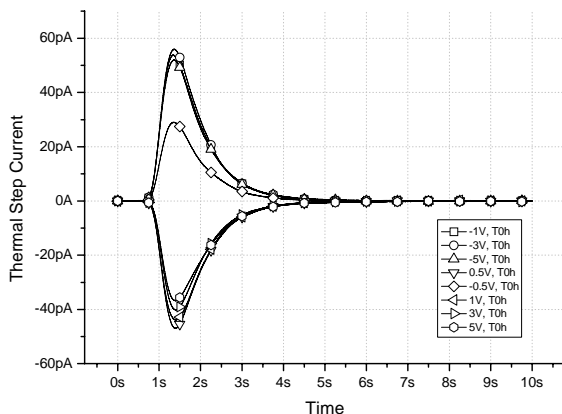


Fig. 13. Thermal step currents measured on a flat MOS capacitor with p-type Si substrate and 100 nm SiO<sub>2</sub> layer under different gate voltages

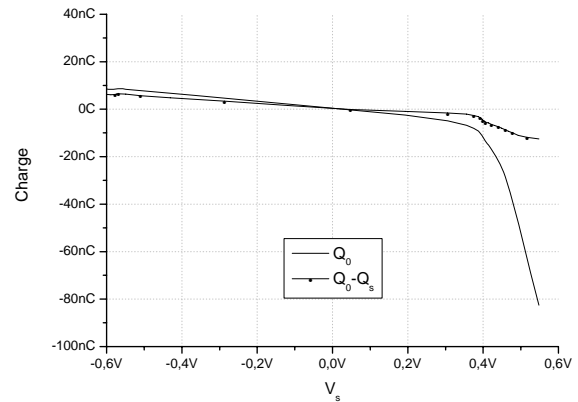


Fig. 14. Total charge of the MOS structure ( $Q_0$ ) and oxide + interface charge ( $Q_0-Q_s$ ) versus Si substrate potential calculated with TSM

## 5. Tendencies and prospects

The influence non destructive methods for measuring electric charge in solid insulators have played and will continue to play a significant part, as well in scientific research as in the development domain. The “low energy” thermal methods as LIMM, FLIMM and ATWM can provide results on thicknesses from 10 to 100 microns, with a material or component orientation. The high voltage “under field” TSM is installed in industrial facilities and contributes to material choice for dc power transport applications. On the other hand, the TSM has shown its possibilities as a characterization technique for micro and nano-electronics. Due to their high dynamics, the pressure methods can be used directly for measurement under alternative stress.

To conclude, because space charge is an image of the microstructure of the materials, it has been an important matter since 30 years to set up non destructive methods for measuring this parameter related to the history of a material or component. These methods – under the form of two distinct schools – have significantly progress in different domains, leading today to a protocol [21] which may serve as a basis for more and more numerous scientific and industrial studies.

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