A Review of Pressure Pulse Measurement Techniques for Space-Charge and Polarization in Dielectrics

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Abstract: This review encompasses a detailed account of pressure pulse methods for measurement of space charge profiles in thickness direction of polymer electrets.

Keywords: Space-Charge, Pressure Pulse, PPS, LIPP, PWP

I. INTRODUCTION

Space charges recently have garnered attention of many researchers all over the world. The reason for this may be attributed to the fact that measurement of space charge distributions is important in understanding of the charge build up and decay mechanisms. Also in such polymer electrets where the property of the electret results from irradiating the polymers with a partially mono-energetic electron beam, information about the depth and profile of the charge distribution in such polymer electrets is important in studying the dynamics of the charging process and of subsequent (slow) discharging effects. Several methods have been developed to investigate space charge distributions in dielectrics.

In the study of polymer electrets an important problem is the determination of the spatial variation of polarization and space-charge distributions. A major unsolved problem in the study of piezoelectric and pyroelectric polymers is the determination of mechanism of charge storage. Techniques employed to characterize the dielectric materials have their own limitations as they yield results which are property averages over the thickness of the polymer films. Three major approaches have been used in the development of techniques for analysis of either polarization or space-charge distribution. Several specialized methods are available which are only suitable for the determination of space-charge distributions. The specialized techniques include the split-Faraday-cup method, [1, 2] measurement of electron transmission [3], and electron beam sweeping [4]. These methods and several others used for space-charge analysis were reviewed by Sessler and Gerhard-Multhaupt [5].

The first general technique for either polarization or space-charge utilized sectioning techniques in which successive layers of poled electrets were removed and analyzed for their charge account. Some early examples of this approach are used by Theissen *et al.* [6] and by Walker and Jefimenko [7], both of whom sectioned carnauba wax electrets. An extension of this technique in conjunction with another method has been proposed by Collins [8].

A second general approach has been the use of multilayer or sandwich structures. Phelan [9] poled sandwich structures consisting of four layers of polyvinyl fluoride (PVF) or polyvinylidene fluoride (PVF_2 also known as PVDF) and then measured the pyroelectric coefficient in each layer. Marcus [10] constructed eight-layer sandwiches and measured the piezoelectric coefficient achieving moderately high resolution. Both of these approaches have the serious drawback that the analytical technique is a destructive one.

A review of the several experimental techniques for the determination of spatial charge and field distributions in the thickness direction of the sample material is presented here. Emphasis is laid on the nondestructive methods of direct probing such as pressure-pulse methods, and methods that use waves which propagate through the charged sample.

A number of acoustic techniques are available to probe space charge and polarization profiles in the thickness direction of polymers. In principle all the methods depend upon the common physical process of ultrasonic pulse propagating through the sample.

These methods are named as

a) The Pressure-Wave Propagation Method (PWP)
b) Laser-Induced Pressure Pulse Method (LIPP)
c) Piezoelectrically Generated Pressure Pulse/Step (PPS) Method

Assuming that the considered material is homogeneous and insulating, the current $I(t)$ flowing due to the material's reaction to the external pressure is given as [11]

$$I(t)=\mathrm{X}C_0G(\varepsilon_r)\int_0^{z_f}E(z,0)\frac{\partial}{\partial t}p(z,t)dz \qquad (1)$$

where X is the compressibility of the material, $G(\varepsilon_r)$ is a function of the relative permittivity (dielectric constant) ε_r, which in turn depends on the pressure distribution in the sample. $E(z,0)$ is the electric field distribution at the time $t=0$, $p(z,t)$ is the pressure distribution and C_0 is the capacitance of the non-compressed sample:

$$C_0=\varepsilon_0\varepsilon_r.S/d \qquad (2)$$

where d is the thickness of the sample, S is the sample area.

Further the voltage difference $V(t)$ across the sample is described by:

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$$V(t) = X.G(\varepsilon_r)\int_0^{z_f} E(z,0)p(z,t)dz \qquad (3)$$

The information about charge distribution can be extracted using Poisson's equation:

$$\nabla^2 V = \frac{\rho}{\varepsilon_0 \varepsilon_r} \qquad (4)$$

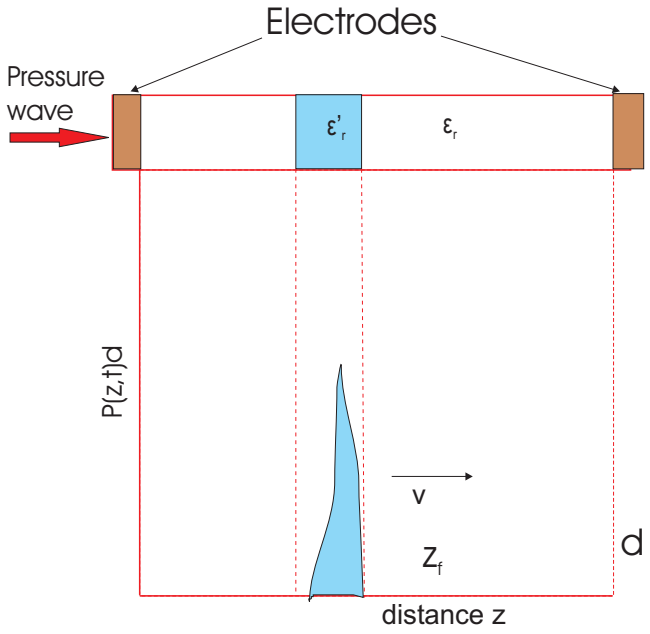

Fig.1: Principle of Pressure Pulse Methods [11]

Techniques that belong to this family share the same measurement principle, the difference is in the method of initiating the pressure pulse that travels through the measured sample.

II. PRESSURE PULSE METHODS

Pressure Wave Propagation Method (PWP)/Laser Induced Pressure Pulse Method (LIPP) and Piezoelectric ally Generated Pressure Pulse Method (PPS) A distinct advantage of some of the acoustic techniques is their ability to obtain the charge distributions in a polymeric sample without deconvolution, with a spatial resolution of at worst 10% of the sample thickness (provided that the thickness of the sample is not less than 100 µm). This depends on the time constants of the exciting pulse, the speed of the detection circuit and the sample thickness. However, for a 10 µm film, a 10% resolution is hard to achieve without a picosecond laser. Methods falling in the same category are pressure wave propagation (PWP), laser-induced pressure-pulse and piezoelectrically generated pressure pulse/step (PPS). PWP and LIPP methods are nowadays more or less synonymous, implying the use of laser-generated pressure pulses [12, 13]. After the manuscript was submitted by Sessler *et al.* [14], simultaneously a related technique and result was reported by Alquié *et al.* [15]. The technique reported by Alquié is known as pressure wave propagation (PWP) method whereas the method reported by Sessler is known as laser-induced pressure pulse [LIPP] method. LIPP and PWP methods use the same principle but differ in the generation of the pressure pulse. In both the methods a pressure pulse in the form of a longitudinal sound wave is used to produce a temporary displacement of space charge in polymeric materials. In case of PWP method reported by Alquié *et al.* [15] a wave can be generated by the impact of a pulsed CO_2 laser beam (having a rise time of about 10 ns and a duration of more than 100 ns) on a metal target bonded to the dielectric plate under investigation (which led to the straightforward visualization of electric field distributions in solid dielectrics) whereas in case of LIPP [14] a pressure pulse (~2ns) was generated due to recoil of the ablation of specially coated surface of the material which propagates through the sample. The LIPP method uses a short laser-light pulse to irradiate one side of the sample containing space charges. As the very narrow compressed region travels through the sample, a current is induced in the external circuit due to non-uniform changes in dimension and permittivity reflecting the space charge distribution. When the thickness of the compressed region is small, the profile of the current response is directly proportional to the charge distribution, including the charges on the electrodes. The principle of the pressure wave propagation (PWP) is shown in fig. 2 and the experimental setup for LIPP method is shown in fig. 3. Eisenmenger and Haardt [18-20] used a piezoelectrically generated pressure step to cause a nonuniform deformation of the sample volume behind the propagating step. The method was later termed as piezoelectrically generated pressure step Eisenmenger and Haardt [18-20] used a piezoelectrically generated pressure step to cause a nonuniform deformation of the sample volume behind the propagating step. The method was later termed as piezoelectrically generated pressure step method (PPS) [21]. In the PPS method (see fig. 4) a non-uniform driving force is used to elicit an electrical response from an electret. As an example, a nonuniformly distributed mechanical force will interact with spatially distributed polarization or space charge to produce a piezoelectric response. A shock wave tube has also been used for first pressure-step experiments which were performed on 1mm thick polyethylene (PE) film [22-24].

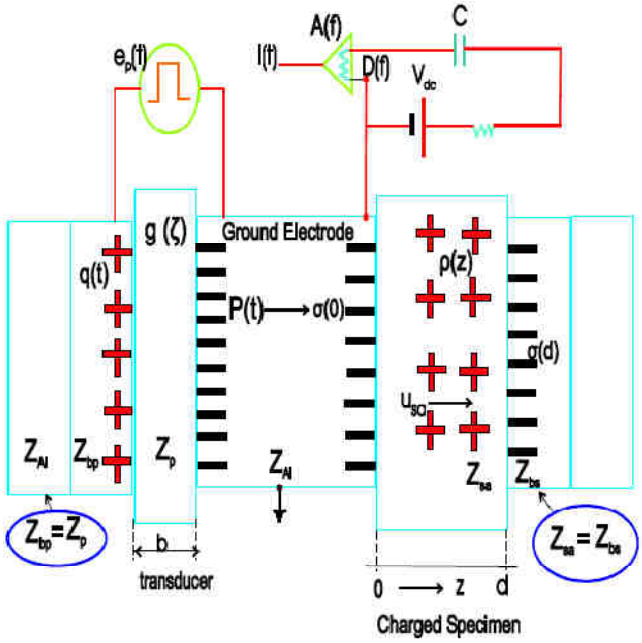

Fig. 2: PWP space charge measurement system [16].

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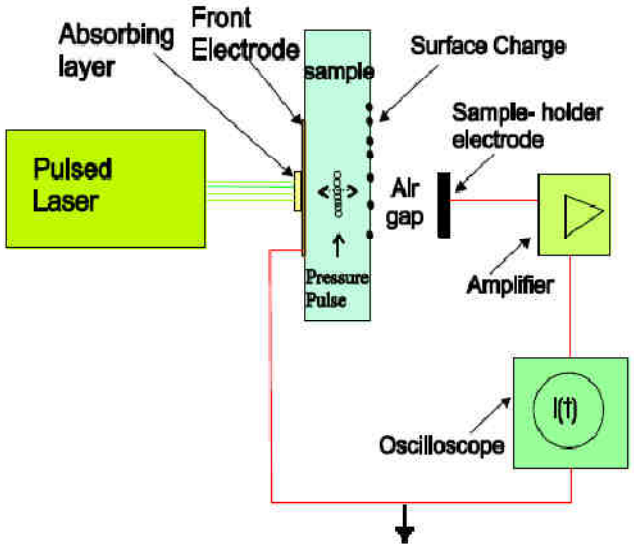

Fig. 3: Experimental setup for laser-induced pressure pulse (LIPP) method on one sided metallized samples [14, 17]

$\sigma(0)$ is the surface charge on the ground electrode, space charge $\rho(z)$ in the sample and $\sigma(d)$ on the high voltage electrode. Z_{sa} and Z_{Al} is the impedance of sample and electrode respectively. Z_{bs} is the impedance of the high voltage electrode, Z_p is the impedance of the transducer. $p(t)$ is the pulsed acoustic wave and $u_{sa}\Delta t$ is the sampling time interval which is determined by the sampling time of the high voltage pulse applied to a piezoelectric transducer.

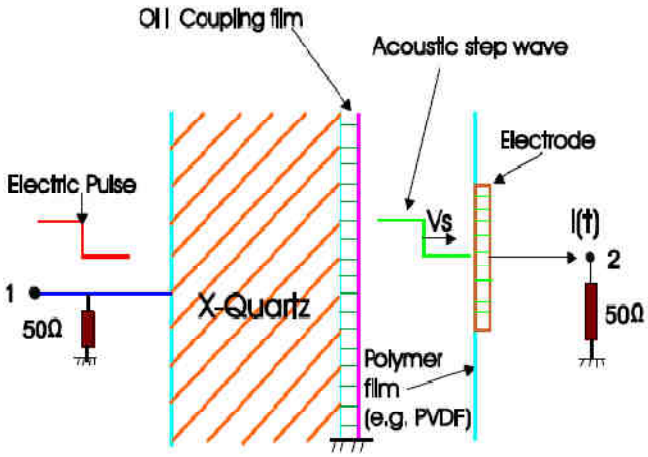

Fig. 4: Schematic diagram of the experimental technique of PPS. (1) Connection to voltage pulse generator (2) Connection to wide band amplifier [18, 19]

III. APPLICATIONS

LIPP has been extensively used by Sessler and Gerhard-Multhaupt [26-30] in their investigations of space charge and polarization profiles in thin film polymer materials FEP, PI (Polyimide)(Kapton™), PET (Polyethylene terepthalate) PVDF and Silicon oxide charged with electron beams, corona discharges or liquid contacts. A description of the application of PVDF was published by Gerhard-Multhaupt et al. [31]. With the help of these experimental technique field distributions in PVDF foils [18] and charge distribution in Mylar polyethylene terephthalate films [14] could be measured directly. Suzuoki *et al.* have utilized LIPP in their study of space charge distribution in 0.1, 0.6 and 1 mm thick slabs of low density PE [32]. A study of static electricity using the laser-induced pressure pulse has been made by Malec [33]. In this static charges accumulated on a polyethylene slab (1 mm) induced after the sliding friction upon a cotton sheet and in other case caused due to peeling off an adhesive tape placed on the aforesaid PE slab has been studied. With the help of this method it has been shown that static charges on a surface submitted to friction or gas/liquid flow may be measured using the pressure pulse method. Relationships between induced signals and surface charges were established taking into account the charge penetration depth which earlier could not be estimated using traditional methods.

The LIPP method has been proposed to detect adhesion defects in bilayer structures [34] and to measure the amount of surface charge accumulated at the vicinity of a void embedded in a solid insulator [35,36]; which are the new applications of this method. Charge distributions in thermally poled silica glass were mapped by using laser induced pressure pulse technique [37]. The PWP method has been used to measure the space charge and polarization distributions in thick slabs (50 to 200 µm) of polymer electrets [38-40] and in PE used in HV cables [41, 42]. The PWP method has been used to determine the surface charge distributions [43]. The PWP also allows the study of the revolution of the charge distribution in a sample during the polarization process during the HV stress and the investigation of the phenomena causing a reversal of the polarity of the applied voltage [44]. PPS was used by Gerhard-Multhaupt *et al.* [45] to measure electric-field distributions in the thickness direction of thin fluoropolymer-electret films from polytetra fluoroethylene (Teflon TFE) and its copolymers with hexafluoropropylene (Teflon FEP) and perfluoropropoxyethylene (Teflon PFA). They obtained charge densities as high as 2500 C/m^2 in positively and negatively charged Teflon TFE, FEP and FPA fluoropolymers under suitable charging conditions.

IV. CONCLUSIONS

The experimental resolution of all the above-mentioned methods is not high enough for thinner samples. However, this limitation can be overcome by using a different experimental setup having a Q-switched Nd: YAG laser system for generation of 4-5 ns long pressure pulses [17] or by using a quartz crystal to produce step waves of about 4-5-ns rise time [18]. Since all the methods referred to here [14-22] depend on the same physical process, namely a pressure wave propagating through the sample, their theoretical description is supposed to be identical. This however is not true since different approaches have been used for the derivation of the equations which govern the response of a charged sample to a pressure wave [14, 15, 18, 20, 21]. A detailed analysis of the response equations for pressure step, pressure-pulse, and arbitrary pressure-profile experiments

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was derived by R. Gerhard- Multhaupt [25]. The advantage of the pressure pulse method is that no mathematical deconvolution is required to interpret the data.

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