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Review

A review of developments in thermal techniques for charge profile measurements in polymer electrets

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ABSTRACT

A number of methods were evolved during last three decades to understand the internal charge profile of polymer electrets. These methods essentially are based on the propagation of heat or pressure waves inside the charged samples. In both cases, electrical signals are generated due to mechanical or dielectric changes caused by heat diffusion or propagation of a pressure discontinuity in the sample. The charge distribution can be obtained from the electrical response. This paper presents detailed information on the thermal techniques to probe the charge distributions in the thickness direction of the polymer electrets as well as a comprehensive review of thermal data analysis.

1. Introduction

Due to the charge storing properties and wide applications of electrets, research in this field has grown significantly since the term 'electret' was coined by Oliver Heaviside. Early researches on electrets are well accounted in the literature [1–8].

One of the important classes of organic materials that have gained prominence in the electret research during the past 40 years are polymers. The electrets fall into the category of "charge electret" and "dipole electret" respectively depending upon the charges the electrets may have and its schematic diagram is shown in Fig. 1 [after [9]].

Charge electrets find their use in photovoltaic solar cells, acoustic transducers, air and gas filters, radiation dosimeters and micro-relay switches [10]. True polarization electrets with a (quasi)-permanent preferential dipole orientation are employed in hydrophones, ultrasound applications, infrared thermal imaging sensors, microcalorimeters due to their piezo- and pyroelectric response [11] or (for second order nonlinear optical responses) in electro-optic modulators and switches, etc. [12].

During the last 20–25 years, the understanding of charge dynamics in electrets has significantly improved because of the development of several non- destructive techniques to measure the charge profile. Space charge distribution measurement methods in polymers broadly fall into three categories- thermal methods, pressure methods and electro-acoustic methods. Essentially they are based on the non homogeneous perturbation of the electrostatic state of the sample under test. Application of thermal or mechanical stimulus creates perturbation in the electrostatic equilibrium of the sample under test giving rise to an electrical transient response which can be recorded and analyzed for the determination of charge distribution and electric field across the sample.

In particular, the application of thermal [13–22] and acoustic methods [23–31], with resolutions of the order of ~1 μm, to charged dielectrics has helped us to advance our knowledge of such phenomena in thin polymer films.

The review encompasses experimental techniques for the determination of spatial charge and field distributions in the thickness direction of the sample material and the methods of data analysis. Emphasis is laid on the direct probing such as thermal pulse and thermal wave method.

2. Space-charge and polarization measurement techniques

Charge profile measurements recently have garnered attention of many researchers all over the world. The reason for this may be attributed to the fact that measurement of charge distributions is important to understand the charge build up and decay mechanisms. Several methods have been developed to investigate 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 is the determination of mechanism of charge storage. Techniques employed to

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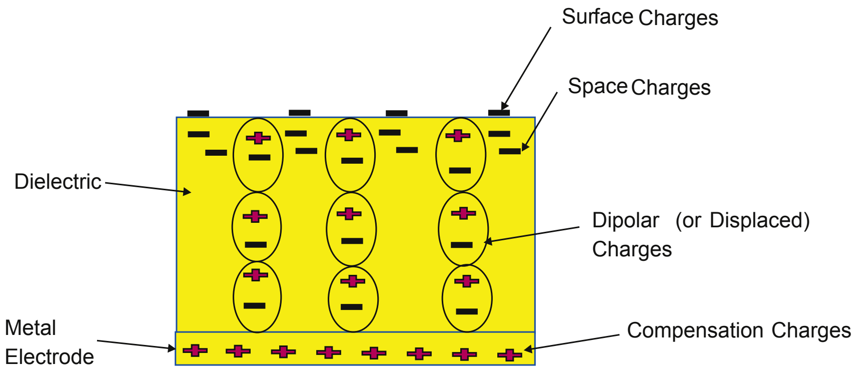

Fig. 1. Schematic cross section of a one sided metallized electret having deposited surface charges, injected space charges, aligned dipolar charges (or microscopically displaced charges), and compensated charges [after [9]].

characterize the dielectric materials have their own limitations as they yield results which are property averages over the thickness of the polymer films. 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 [32,33], measurement of electron transmission [34], and electron beam sweeping [35]. These methods and several others used for space-charge analysis were reviewed by Sessler and Gerhard-Multhaupt [36].

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.* [37] and by Walker and Jefimenko [38], both of whom sectioned carnauba wax electrets. An extension of this technique in conjunction with another method has been proposed by Collins [17]. A second general approach has been the use of multilayer or sandwich structures. Phelan [39] 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 [40] 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.

2.1. Thermal techniques

Thermal techniques are most easily applied to thin films. If used for relatively thick samples, the electric field intensity is allowed to vary only in the direction normal to the plane of the sample, otherwise the measurement becomes inaccurate. Normally the samples tested with the thermal methods require poling under DC high voltage with electric fields ranging up to 50 kV/mm [41] or even more than 100 kV/mm in some cases. The samples are poled either by using the corona discharge method or the contact poling method.

The thermal methods can be divided into three groups according to the signal used for excitation of the sample. In thermal-pulse and thermal-wave methods a short (<100 μs) light pulse or a modulated light beam, respectively, is used to illuminate (and thus heat) the absorbing sample electrode. The thermal step method generally utilizes heating of one sample side relative to the other by application of a constant temperature difference between two electrodes. Essentially the thermal step method is a kind of thermal pulse method [42–44].

The thermal pulse (TP) method is performed in time domain whereas the thermal wave method (often known as Laser-Intensity Modulation Method-LIMM) is performed in frequency domain. The measured signal, either current or voltage contains information about space-charges and knowledge of thermal diffusivity is required for data analysis.

2.2. Thermal Pulse Technique (TPT)

The thermal pulse method was proposed for the first time by Collins [13–17] and later modified by others [46–57]. A schematic diagram of the thermal pulse experiment proposed by Collins [13] is shown in Fig. 2.

The electret is aluminized on one side and mounted. When one side of the sample is heated with short duration of laser pulse the heat energy is absorbed and diffuses into the sample and causes thermal expansion as well as changes in the dielectric permittivity resulting in a variation of the surface potential in an open-circuited (e.g. one-side metallized) sample or in electrode currents in a short-circuited sample. The expression for the generated current in the case of a short circuited sample is given as [45]

$$I(t) = \frac{A}{d} \int_0^d r(z) \frac{\partial T(z,t)}{\partial t} \mathrm{d}z \tag{1}$$

with the distribution function

$$r(z) = \alpha_P P(z) - (\alpha_z - \alpha_\varepsilon)\varepsilon_0 \varepsilon E(z) \tag{2}$$

where A and d are the area of the electrode and d the sample thickness, respectively and $r(z)$ is a distribution function which contains contributions from the spatially dependent spontaneous polarization and space-charge. In Eq. (2) α_P, α_z, α_ε, ε and $E(z)$ are the relative temperature dependence of the polarization, relative

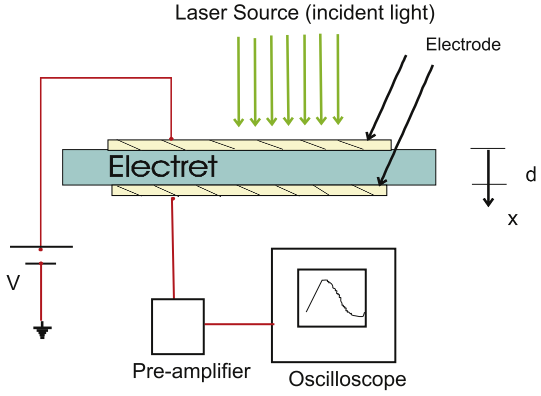

Fig. 2. Schematic diagram of the arrangement for thermal pulsing [redrawn after [13,16 and 75].