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Volume and surface polarization of electrets

M R BHIDAY, U GUPTA, M RANADE and S RAO
Department of Physics, University of Poona, Poona 411007

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Abstract. An attempt is made, to obtain mathematical relations correlating the volume and surface effects of the dielectric to those of the electrical impedance of the dielectric system exhibited in the presence of a quasistatic sinusoidal electrical field. A new experiment is described to differentiate clearly the two types of polarizations in an electret.

Keywords. Electret; volume charges; surface charges; thermoelectret; radioelectret.

1. Introduction

An electret is characterised by two types of charges. The heterocharges are the real volume charges, and the homocharges are charges developed on the surface due to breakdown in the electrode-electret interface, and other reasons hitherto not quite clear. The volume charges are useful for applications involving the dielectric permittivity, while, the surface charges are useful in electrostatic instruments and other applications such as electro-photography.

The methods so far used to study the polarization of an electret consist in either measuring the charges induced on a conducting plate (Gubkin 1960) or measuring the time integral of the depolarization current obtained by destroying the electret thermally (Gross 1949).

We present here a new method (Bhiday *et al* 1972) to measure the surface and volume charges independently of each other, without destroying the electret's polarization.

2. Formalism of the problem

2.1. *Resistor and capacitor*

A theory is developed in order to separate the volume and surface charges in terms of the electrical properties of the electret.

The electret is considered as a parallel plate capacitor having electrical properties under excited conditions. A rectangular slab of semimetallized dielectric (on one of its surfaces) is kept over a copper plate and excited by an alternating voltage source (figure 1a). The metallized layer provides a finite controllable surface resistance r ohms per unit area. The fringing effects at the edges of the plate can be neglected by assuming that the separation between the resistive sheet and the plate is very small, compared to the surface area of the dielectric i.e. $(a \sim b \gg d)$.

Two situations could be considered, wherein the resistive sheet and the conducting

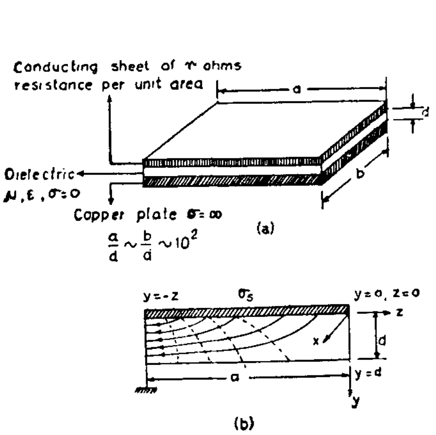

Figure 1. Resistor-capacitor device.

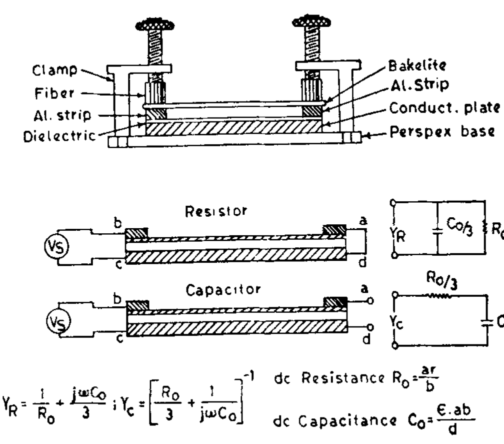

Figure 2. Sample holder for preparation of the electret and the equivalent circuits. The dielectric is kept between a conductor (bottom shaded area) and a resistive sheet top shaded area).

plate are short circuited (Resistor), or open circuited (Capacitor). The corresponding equivalent circuits are shown in figure 2.

The net effect of the surface charge manifests through the 'resistor', while the 'capacitor' represents only the volume polarization. In order to obtain the contributions of the surface charges and the volume charges separately, the admittances Y_R and Y_C are calculated for the 'resistor' and the 'capacitor' respectively as shown below.

2.2. *Mathematical analysis*

As shown in figure 1b the coordinate axes are chosen such that the resistive sheet is in the x-z plane at $y=0$. The metallic conductor of zero resistance is placed at $y=d$. A sinusoidal voltage source of amplitude V_0 is applied between the resistive sheet at $z=-a$ and the lower conducting plate. The resistive sheet is short circuited with the conducting plate at $y=0$ and $z=0$. Let the width of the dielectric along the x-axis be denoted by b, and its permittivity by ϵ. The zero order surface current in the sheet should be uniform. If σ_s is the surface conductivity of the resistive sheet, then the zero order surface current density $\mathbf{K}_0$ is given by

$$\mathbf{K}_0 = \frac{\sigma_s}{a} V_0 \mathbf{i}_z \tag{1}$$

The zero order potential ϕ_0 on the xz plane is

$$(\phi_0)_{y=0} = -\frac{z}{a} V_0 \tag{2}$$

This result depends only on the characteristic of the resistive sheet and not on the geometry of the lower plate. The zero order potential ϕ_0 over the surface at $y=0$ satisfies Laplace's equation. By imposing the boundary conditions, namely,

$$(\phi_0)_{y=d} = (\phi_0)_{z=0} = 0 \tag{3}$$

one obtains for ϕ_0 the expression

$$\phi_0 = -\frac{V_0 z}{a}\left(1-\frac{y}{d}\right), \quad \text{for } y \geqslant 0 \tag{4}$$

The corresponding zero order electric field is given by

$$\boldsymbol{E}_0 = \frac{V_0}{d}\left[-\frac{z}{a} i_y + \frac{d}{a}\left(1-\frac{y}{d}\right) i_z \right] \tag{5}$$

The y component of the zero order electric field yields for the surface charge density σ_0,

$$\sigma_0 = -\epsilon \frac{V_0}{d}\frac{z}{a} \tag{6}$$

By considering the current in the resistive sheet and using the boundary conditions one obtains for the zero-order magnetic field in the space bounded by parallel sheets, the short circuiting conductor, and the plane of the current source when the lower plate is earthed

$$\boldsymbol{H}_0 = -i_x \frac{K_0}{2} \quad \text{or} \quad \boldsymbol{H}_0 = -i_x \frac{\sigma_s V_0}{2a} \tag{7}$$

Using the equations (5) and (7), the zero order complex Poynting vector can be written as

$$\boldsymbol{S}_0 = \tfrac{1}{2}(\boldsymbol{E}_0 \times \boldsymbol{H}_0) \quad \text{or} \quad \boldsymbol{S}_0 = \frac{\sigma_s V_0^2}{4ad}\left[-\frac{z}{a} i_z + \frac{d}{a}\left(1-\frac{y}{d}\right) i_y \right] \tag{8}$$

In order to obtain the admittance, it is necessary to know the average power dissipated in the resistive sheet per unit width in the x direction and the average total electric and magnetic energy per unit width in the same direction.

The average zero order power dissipated in the resistive sheet per unit width in the x direction can be readily obtained to be

$$\langle p_{d0} \rangle = \tfrac{1}{2}\left(\frac{V_0}{a}\right)^2 \sigma_s = \tfrac{1}{2}\sigma_s \left| \boldsymbol{E}_0 \right|^2_{y=0} \tag{9}$$

This gives the total zero order power dissipated in the resistive sheet per unit width in the x direction and can be readily obtained from equation 9.

$$\langle P_{d0} \rangle = \int_{-a}^{0} \langle p_{d0} \rangle \, dz = \tfrac{1}{2}\left(\frac{\sigma_s}{a}\right) V_0^2 \tag{10}$$

The zero order average electric and magnetic energies are given by

$$\langle w_{e0} \rangle = \tfrac{1}{4}\epsilon \left| \boldsymbol{E}_0 \right|^2 = \tfrac{1}{4}\epsilon \frac{V_0^2}{d^2}\left[\frac{z^2}{a^2} + \frac{d^2}{a^2}\left(1-\frac{y}{d}\right)^2 \right] \tag{11}$$

and

$$\langle w_{m0} \rangle = \tfrac{1}{4}\mu \left| \boldsymbol{H}_0 \right|^2 = \tfrac{1}{4}\mu \left(\frac{\sigma_s V_0}{2a}\right) \tag{12}$$

By integrating over the volume enclosed by the resistive sheet and the dielectric, the total electric and magnetic energies are found to be,

$$\langle W_{e0} \rangle = \tfrac{1}{12}\epsilon V_0^2 \left(\frac{a}{d} + \frac{d}{a}\right) \tag{13}$$

$$\langle W_{m0} \rangle = \tfrac{1}{16}\mu\sigma_s^2 V_0^2 \frac{d}{a} \tag{14}$$

In the low frequency behaviour considering only the terms up to first order, the input power can be written as a sum of average and the reactive powers (Fano *et al* 1960), i.e.,

$$\langle P_0\rangle + jQ_1 = \langle P_{d0}\rangle + 2jw\,[\langle W_{m0}\rangle - \langle W_{e0}\rangle] \tag{15}$$

where $\langle P_1\rangle$ and Q_0 have been eliminated from the left hand side because they are zero in this case (Fano *et al* 1960).

The impedance per unit width in the x direction is given by

$$Z_0 + Z_1 = \frac{2}{|I_0|^2}[\langle P_0\rangle + jQ_1] \tag{16}$$

which yields, when $b/a \ll a/b$,

$$Z_0 + Z_1 = \frac{a}{\sigma_s} + \tfrac{1}{2}jw\left[\tfrac{1}{2}\mu ad - \tfrac{2}{3}\epsilon\frac{a}{d}\left(\frac{a}{\sigma_s}\right)^2\right] \tag{17}$$

The admittance per unit width to the first order is,

$$Y_0 + Y_1 = \frac{2}{|V_0|^2}[\langle P_0\rangle - jQ_1] \tag{18}$$

which gives

$$Y_0 + Y_1 = \frac{\sigma_s}{a} + jw\left[\frac{\epsilon a}{3d} - \tfrac{1}{4}\mu\frac{d}{a}\sigma_s^2\right] \tag{19}$$

Since the conductivity σ_s is small, σ_s^2 can be neglected and so,

$$Y_0 + Y_1 = \frac{\sigma_s}{a} + \frac{jw\epsilon a}{3d} \tag{20}$$

By definition, the admittance of the system is

$$Y = \frac{1}{R_0} + jwC \tag{21}$$

Comparing the real and the imaginary parts in (20) and (21), one obtains

$$\frac{1}{R_0} = \frac{\sigma_s}{a} \tag{22}$$

and

$$C = \tfrac{1}{3}\,\epsilon\frac{a}{d} \tag{23}$$

The capacitance of the system when it is open circuited is given by

$$C_0 = \epsilon\frac{a}{d} \tag{24}$$

From equations (23) and (24) one gets

$$C = \tfrac{1}{3}C_0 \tag{25}$$

Thus, the quasi-static admittance can now be expressed as

$$Y_R = \frac{1}{R_0} + jw\frac{C_0}{3} \tag{26}$$

It follows, therefore, that the capacitive reactance of the ' resistor ' reduces by one-third of its original value. A predominant effect of surface properties, should be, therefore obtained by measurement of Y_R.

A similar calculation yields the admittance of the capacitor system and it can be shown to be

$$Y_C = \left(\frac{1}{jwC_0} + \frac{R_0}{3}\right)^{-1} \tag{27}$$

where, the value of the resistance is reduced to be one-third of its original value. A major contribution is, thus, provided by the capacitive properties of the system. A measurement of Y_C, therefore, should give information regarding the volume of the dielectric. Since, the surface and the volume polarization should have different effects on the dielectric properties, the measurements of Y_R and Y_C for polarized samples would identify these polarizations separately if compared with those measured with the unpolarized sample.

3. Experimental technique

Rectangular samples of size 8 cm $\times$ 7 cm were prepared out of 2 mm thick perspex. One of their surfaces was coated with aluminium by vacuum deposition. The resistance was controlled externally, to obtain a definite predecided finite resistance. The second surface of the sample was kept in contact with a conducting plate.

The sample holder shown in figure 2, consists of a bakelite plate with four brass clamps at its two edges. The clamps were insulated at their ends with a fiber. Two finely polished thick aluminium strips were kept over the edges of the sample to serve as contacts to the resistive sheet. Terminals *b* and *c* were connected to the oscillator. The admittance Y_R was measured when terminals *a* and *d* were short circuited, and Y_C when *a* and *d* were kept open.

The sample was polarized by keeping the cell in a thermostat at 130°C and applying high electric field across *b* and *d*. After two hours the sample was cooled to room temperature. The electric field was removed and the admittance measured. This corresponds to the 'polarized' value of the admittance as plotted in figure 4.

The admittance of the system was measured with a Marconi's circuit magnification meter type 1245. The accuracy of absolute measurements of admittance with this instrument was about $\pm 5\%$. Even so, the measurement of Y as a function of time for, both, the polarized and the unpolarized samples and their comparison allowed us to clearly distinguish the changes brought about on polarization of the sample.

4. Results and discussion

4.1. *Thermoelectrets*

The measurements were made at frequencies from 40 kHz to 300 kHz. The values of Y_R and Y_C increased with increasing frequency, as expected from equations (26) and (27). Figure 3a shows the variation of Y_R and Y_C according to these equations for arbitrary values of R_O and C_O obtained theoretically. The nature of the variation remains the same for observed values of admittances (figure 3b). The rather steep increase in the experimental value of Y_R may be correlated with the variation of the internal impedance of the measuring unit. However, a modified theory is being worked out for a better fit which will be reported later.

Though the experimental results do not agree very well with the theoretical ones, the relations suggest the possibility of the independent measurements of the required

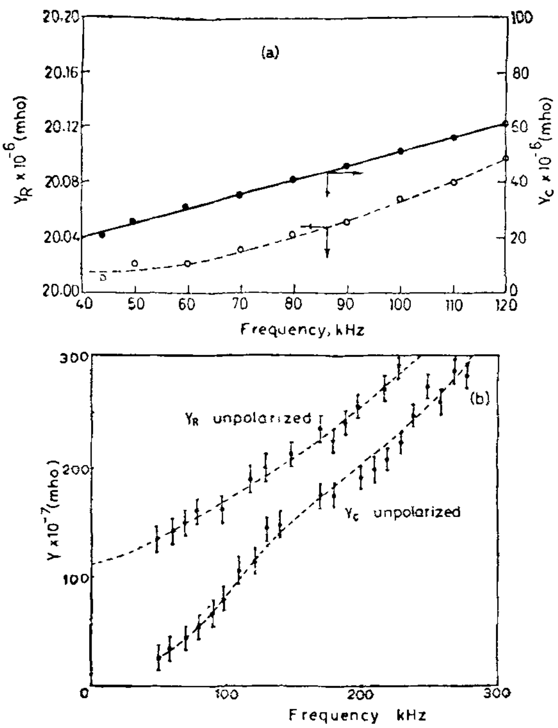

Figure 3. Variation of the admittance with the applied frequency: (a) Theoretical plot; (b) Experimental plot.

quantities. The difference in the values of Y_R and Y_C in the unpolarized dielectric obtained experimentally varied with the frequency, being larger in the low frequency region (below 100 kHz). Hence, the present experiments were carried out with the lowest frequency that could be obtained, viz. 44 kHz.

The polarization of the electret varies with time. So Y_R and Y_C were measured over a scale of time. Figure 4 presents the results obtained with a perspex electret polarized at 12·6 kV/cm. The difference Y_C (pol.) $-$ Y_C (unpol.) increased with time. It appears from the behaviour that the volume polarization builds up with time, even after removal of the electric field. This variation of the heterocharge was not resolved when measured with the vibrating electrode method of studying the storage of charges of electrets.

In another experiment the value of Y_C was measured before and after it's polarization. The difference continued to increase up to 48 hr. After 50 hr the electret was heated at 130°C for 4 hr in order to destroy its polarization. On cooling the sample the admittance resumed its original value. Similar result was obtained with the variation of Y_R. This experiment confirms that the observed differences in admittances were due to the polarization of the dielectrics.

When the electrets were polarized at 31·6 kV/cm (figure 5) Y_C increased in several similar electrets while Y_R deviated very little from its original value. In all these experiments the unpolarized samples exhibit irregular fluctuations, perhaps due to uncontrolled atmospheric conditions. The surface may be undergoing changes due to humidity.

4.2. *Radioelectrets*

Samples polarised with gamma radiations (radioelectrets) from a kilo-Curie Co^{60} source were also studied by the new method. Electrets of perspex indicated poor absorption for radiations. This was also confirmed by the measurement of the decay of the surface

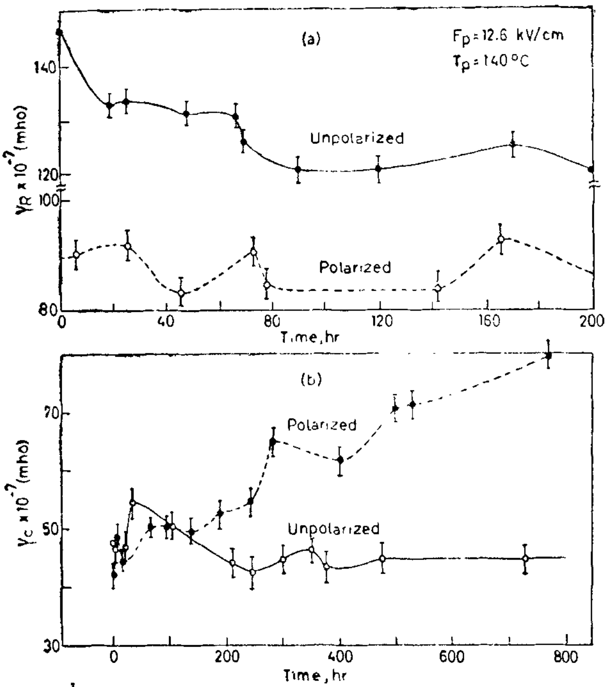

Figure 4. Results obtained with an electret polarized at 12·6k V/cm.

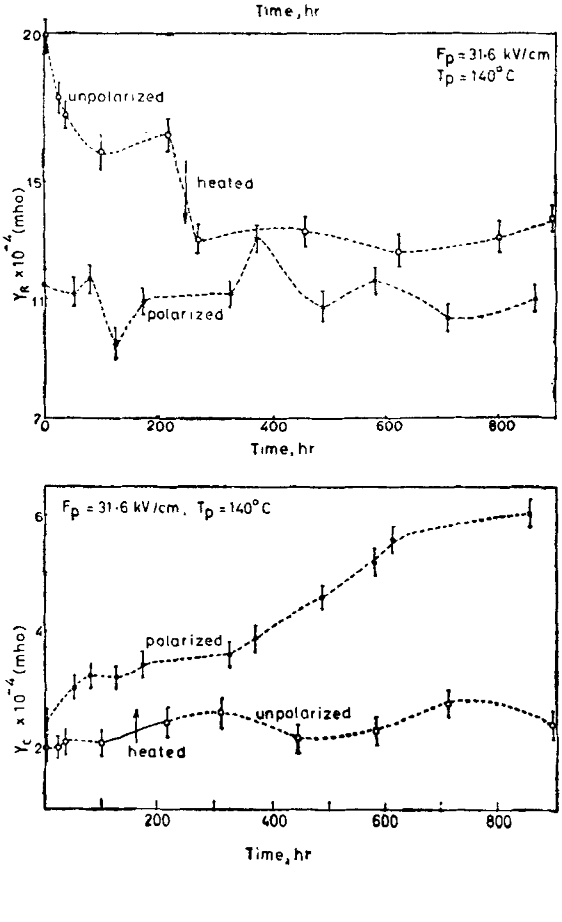

Figure 5. The variation of admittance for an electret polarized at 31·6 kV/cm.

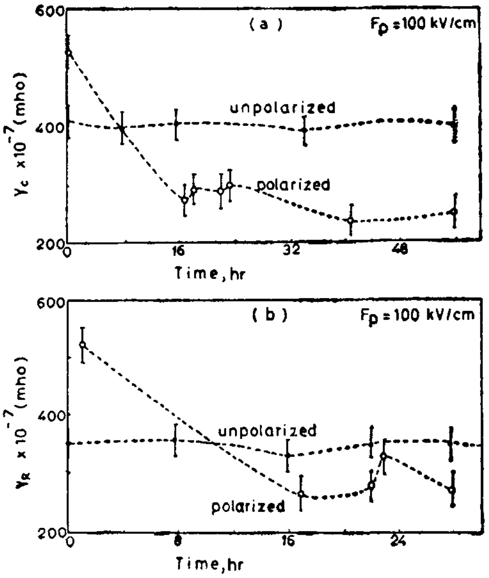

Figure 6. Variation of admittance for an electret of mica.

charge density of radioelectrets of perspex. This indicates the possibility of different mechanisms of polarization in the two methods of preparation of electrets. In thermoelectrets the molecules get enough energy and mobility for movement under the external electric stress and elevated temperatures, and form molecular dipoles, while radioelectrets are not expected to contain dipoles with long relaxation times. Radiation produces free electrons and ions which are drifted along the direction of the electric field leading to trapping and/or storage in the dielectric in several ways.

The polarization in radioelectrets of perspex is mainly confined to the surface of the sample because of the ionization of gases at the contact interface.

Radioelectrets of mica were successfully prepared for the first time and tested by the new method. Figure 6 shows the variations of Y_C and Y_R which indicate an initial increase followed by a slow decay of Y_C and Y_R. The experiment proves the existence of the polarization in mica, which can be easily produced by gamma radiations.

The electrets of perspex and mica which were prepared thermally and by gamma radiations gave different results. This shows that the admittances of electret depend on the volume and surface polarizations. The volume polarization of the electret is directly related to the applied electric field; if the field is kept very high (~30 kV/cm) the surface charges caused by bieakdown of tiny gaps at the electrode-dielectric interface mask the volume charges, thereby causing considerable changes in Y_R. The electret prepared with corona discharge contains little volume charges and therefore, it is not expected to show any change in Y_C. Such electrets are being used to design an electro-photography apparatus in our laboratory.

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