

## Charge storage and relaxation process of polymer film by thermally stimulated discharge current technique

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**Abstract.** Charge storage and relaxation process of the polyimide film electret were investigated using the thermally stimulated discharge current technique. Homo- and heterocharges were mainly observed up to and above the polarization temperature.

**Keywords.** Electret; thermally stimulated discharge current; homocharge, heterocharge; residual polarization; relaxation process; polarization temperature; conductivity.

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### 1. Introduction

Over the years permanently electrified materials or “Electrets” have gained importance in several areas such as acoustic transducers, switches, electrophotography, electrostatic recordings, air filters, motors, generators, dosimeters and in biophysics (Sessler 1980). This is mainly due to the development of polymer films which have good electrical, thermal and chemically stable properties.

The charge-storage and charge decay processes in electrets may be best understood by applying the well-known thermally stimulated discharge current (TSDC) technique (Van Turnhout 1975) which allows precise analysis of a single relaxation process and enables determination of the dipolar relaxation time and activation energy.

The aim of the present work is to understand the charge storage mechanism in the polyimide thermo- and corona-electret by the TSDC technique.

### 2. General features of electret

In general, the distribution of the charge  $q$  through thickness  $h$  for a flat electret may be characterized from the following parameters,

- (i) Surface charge density  $\sigma_1 = \sigma_{x=0}$ ,  $\sigma_2 = \sigma_{x=h}$ ;
- (ii) Volume charge density  $\rho = \rho(x)$ ;
- (iii) Residual polarization  $P_s = P_s(x)$ .

Considering these three factors, the value of  $q$  and the dipole moment  $M_E$  of the electret

can be obtained from the following expressions (Baricova and Koikov 1979)

$$q = \sigma_1 + \sigma_2 + \int_0^h \rho dx \tag{1}$$

$$M_E = \epsilon\epsilon_0 V_E = \frac{\sigma_2 - \sigma_1}{2} h + \int_0^h \rho \left( x - \frac{h}{2} \right) dx + \int_0^h P_s dx, \tag{2}$$

where  $V_E$  is the electret potential. The electric moment  $M_E$  is determined at the point  $x = h/2$  (if  $q = 0$ ,  $M_E$  is not dependent on the co-ordinate  $x$ ).

Experimentally the values of  $V_E$  and  $q$  may be determined by the induction method which consists of; (a) the lifting electrode, (b) the vibrating electrode and (c) the revolving electrode. In each case, the electret is placed between the two metal electrodes in which electric charges are induced depending on the potential difference between the electrodes. In this case one electrode is fixed and the other is movable i.e. lifting, vibrating or revolving.

For the first case (a), initially the electrodes are shorted, then we get the following expressions, i.e.

$$E_0 V = \sigma_{01}(d_1 + d_2 + h/\epsilon) + \sigma_1(d_1 + h/\epsilon) + \sigma_2 d_2 + \int_0^h dx \int_0^x \frac{\rho dx}{\epsilon} + d_2 \int_0^h \rho dx + \int_0^h P_s/\epsilon dx = 0, \tag{3}$$

$$\sigma_{01} + \sigma_{02} = - \left( \sigma_1 + \sigma_2 + \int_0^h \rho dx \right) = -q, \tag{4}$$

where  $\sigma_{01}, \sigma_{02}$  are the initial charge densities of the electret surfaces and  $d_1, d_2$  are the respective distances between electrodes and electrets. For simplicity, we can write the following expressions;

$$\int_0^h dx \int_0^x \rho/\epsilon dx + d_2 \int_0^h \rho dx + \int_0^h P_s/\epsilon dx = \epsilon_0 \Delta V. \tag{5}$$

Therefore,

$$\sigma_{02} = \frac{\Delta V + \sigma_1(d_2 + h/\epsilon) + \sigma_2 d_2}{d_1 + d_2 + h/\epsilon} - q. \tag{6}$$

To measure  $\sigma_{02}$ , we have to first remove the upper electrode from the electret and then find out the potential difference between the electret and the lower electrode. Due to difficulties in measuring  $\sigma_{02}$ , this method is rarely used.

In the case of (b) and (c), the compensation method (Van Turnhout 1975) is used for measuring the surface charge density. In this method, potential  $V_k$  are applied across the electrodes so that the value of  $\sigma_{02}$  will be equal to zero and as a result the alternating signal will be obtained by the vibrating or revolving electrode. The measuring value of  $V_E = M_E/\epsilon\epsilon_0$  and  $q$  will be dependent on  $V_k$  for the two positions of the electrets i.e.  $V_k = V_h = V_0$ . If the fixed electrode is on the surface of the electret, i.e.  $X = 0$  or  $X = h$

respectively, then we get the following relations,

$$V_E = (V_h - V_0)/2 \quad \text{and} \quad q = \epsilon\epsilon_0(V_0 + V_h)/(2\epsilon d - h). \tag{7}$$

Equation (7) may be used to determine the value of the total charge  $q$  and the potential difference  $V_E$  between the surfaces of the electret by the compensation method. If  $q = 0$ , i.e. the distribution of the charge on the thickness of the electret is not important, then the effective surface charge density will be obtained by the following expressions,

$$\sigma_{\text{eff}} = V_k \epsilon \epsilon_0 / h, \tag{8}$$

where  $V_k = V_h = -V_0$  (when  $q = 0$ ). The effective charge density  $\sigma_{\text{eff}}$  may be positive or negative depending on whether homo- or heterocharge was stored in the electrets.

To analyse the experimental results, theoretical models were used for time-depending charge relaxation from electret. The most appropriate model is to consider the dielectric as a heterogeneous structure with respect to thickness (Baricova and Koikov 1979) i.e. the conductivity  $\gamma$  depends upon the co-ordinate  $X$ . For a simple case, this model may be analysed in the form of a three-layer structure of dielectrics (figure 1) whose thin external layer has a higher volume conductivity ( $\gamma_1, h_1/2$ ) with respect to the volume conductivity of internal layer, i.e.  $\gamma_1 \gg \gamma_2, h_1 \ll h_2$ .

In the above cases, homo-charge may be stored between the junction of layers of thickness  $h_2$  and  $h_1/2$  (figure 1) and the relaxation process are analysed by solving the problem of condensor (Baricova and Koikov 1979) i.e. the two-layer structure of dielectrics. Then the following expressions may be written for the process of discharging condensor;

$$\begin{aligned} \frac{d\sigma}{dt} &= \gamma_1 E_1 - \gamma_2 E_2; \quad \sigma = \epsilon_0(\epsilon_2 E_2 - \epsilon_1 E_1); \\ E_1 h_1 + E_2 h_2 &= 0; \quad V_E = \sigma h_2 / \epsilon \epsilon_0. \end{aligned} \tag{9}$$

After solving the above equations, we get the expressions for current density and relaxation time as

$$\begin{aligned} J &= \sigma h_1 h_2 (\epsilon_1 \gamma_2 - \epsilon_2 \gamma_1) / \epsilon_0 (\epsilon_1 h_2 + \epsilon_2 h_1), \\ \sigma &= \sigma_0 \exp\left(-\int_0^t dt/\tau\right), \end{aligned} \tag{10}$$

$$\tau = \epsilon_0 (\epsilon_1 h_2 + \epsilon_2 h_1) / (\gamma_1 h_2 + \gamma_2 h_1) \tag{11}$$

where  $\sigma_0$  is the initial value of charge density.

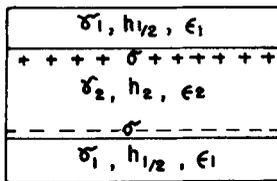


Figure 1. Three-layer structure of dielectrics.

The above equations show that the layers  $h_{1/2}$  and  $h_2$  have not only the different volume conductivity but also different dielectric constants  $\epsilon_1$  and  $\epsilon_2$ . This condition allows us the use of equation (10) for analysing the process of discharging electret when a thin polymer film of thickness  $h_{1/2} \approx h_2$  and volume conductivity  $\gamma_1 \ll \gamma_2$  is placed between the electret and electrode. For this case, it is not always necessary to consider the higher conducting upper surface of the electrets. Equation (10) may be used to analyse the relaxation process at a constant temperature (then  $\tau = \text{const}(t)$  and  $\sigma = \sigma_0 \exp(-t/\tau)$ ) or at a temperature which linearly increases with time i.e.  $T = \beta t$  (then  $\tau = f(T)$  and  $\sigma = \sigma_0 \exp[-\int_{T_0}^T (dT/\beta\tau)]$ ).

### 3. Experimental

The sample used in this investigation was polyimide ( $\pi$ M) film (Ty-72) of about  $40 \mu\text{m}$  in thickness. An aluminium electrode (28 mm diameter) was vacuum-deposited on one surface of the film. The upper free surface (non-metallized surface) was charged by exposure to a negative corona discharge for 5 s in air at atmospheric pressure and room temperature. After charging the sample was short-circuited for 5 minutes to remove frictional and stray surface charges. In this case, the samples were depolarized at a linear heating rate of  $1^\circ\text{C}/\text{min}$ .

Thermoelectrets were prepared by applying an electric field of  $32.5 \text{ kV}/\text{cm}$  and temperature varying from  $20$  to  $230^\circ\text{C}$  with a 2 h polarizing time. After 2 h, the heating source was cut off and the system allowed to cool to room temperature under ambient conditions. TSDC was observed under the open-circuit condition at a linear heating rate of about  $3^\circ\text{C}/\text{min}$ . A constant heating rate was achieved using an automatic temperature controller. An electrometer (BK2-16, USSR) and an X-Y plotter were used to measure the discharge current.

### 4. Results and discussion

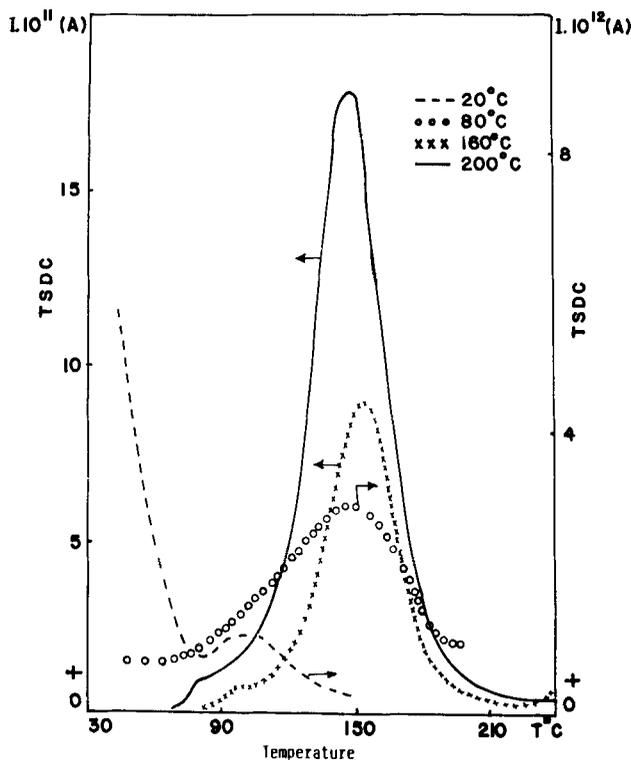
Typical TSDC spectra for thermoelectret of the polyimide film at various polarizing temperatures ( $T_p$ ), ranging from  $20$  to  $200^\circ\text{C}$  are shown in figure 2. When the electret surfaces are in direct contact with the electrodes (electrode-electret-electrode system) and if they are polarized by the electric field, then the direction of TSDC may be related to the movement of the charge carriers or the homocharge that have been stored on the upper layer of the electret or due to the destruction of the residual polarization.

Figure 2 shows that the peak of TSDC spectra increases with increase of polarization temperature. In other words the charge density stored into the polyimide film and its relaxation time increases. The relaxation time may be calculated from the TSDC spectra by using the familiar formula (Baricova and Koikov 1979);

$$\tau = Q(t)/\dot{i}(t), \quad (12)$$

where  $Q(t) = \int_0^t i dt$  charges which flows through the upper surfaces of the film at time  $t$  and  $\dot{i}(t)$  is the TSDC at time  $t$ .

To investigate the nature of the charges stored in the polyimide film, a thin polymer film (teflon) of thickness  $10 \mu\text{m}$  was placed between the electret (non-metalized surface)



**Figure 2.** TSDC spectra for thermoelectret of polyimide film at different temperatures of polarization (electrode-electret-electrode system)  $E_p = 32.5$  kV/cm.

and the electrode. Comparison between the two systems i.e. electrode-electret-electrode (EEE) system and the electrode-electret-teflon-electrode (EETE) system will indicate whether homo-charges or hetero-charges were stored in the film under the electric field. TSDC spectra of the polyimide film (EETE system) at various temperatures of polarization ranging from 20 to 120°C have two peaks (figure 3). Direction of the TSD current (figure 3) for the low temperature peak is the same as the direction of the TSDC peak of the EEE system, i.e. the low temperature peak may arise due to the residual polarization or due to the movement of the charge carriers on the upper layer of the film. The height of this peak is usually small compared to the height of the high temperature peak. Figure 3 shows that the direction of the high temperature peak of the TSDC spectra is completely opposite to the direction of the peak of TSDC spectra which have been obtained in the EEE system (figure 2). This peak may be due to the relaxation of the homo-charges through the thickness of the film.

In the interval of polarization temperature ranging from 160 to 180°C, TSDC spectra show only one peak (figure 4) whose direction is the same as the direction of the TSDC spectra shown in figure 2. This peak may arise due to the destruction of the residual polarization or due to the displacement of the charges to the electrode side.

Again the TSDC spectra at the temperature of polarization from 200 to 230°C show two peaks (figure 5) i.e. the low temperature peak may be related to the destruction of

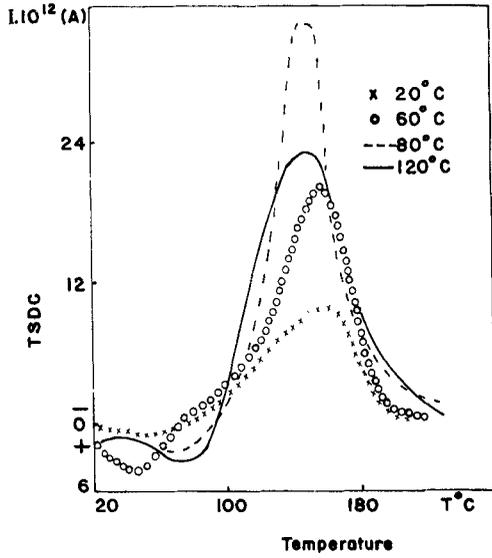


Figure 3. TSDC spectra for thermoelectret of polyimide film from the temperature of polarization 20°C to 120°C (electrode-electret-teflon-electrode system),  $E_p = 32.5$  kV/cm.

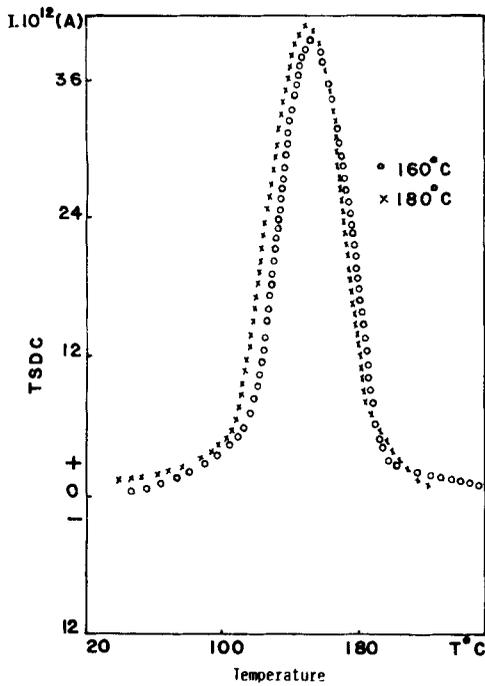


Figure 4. TSDC spectra for thermoelectret of polyimide film at the temperature of polarization 160°C and 180°C respectively (electrode-electret-teflon-electrode system),  $E_p = 32.5$  kV/cm.

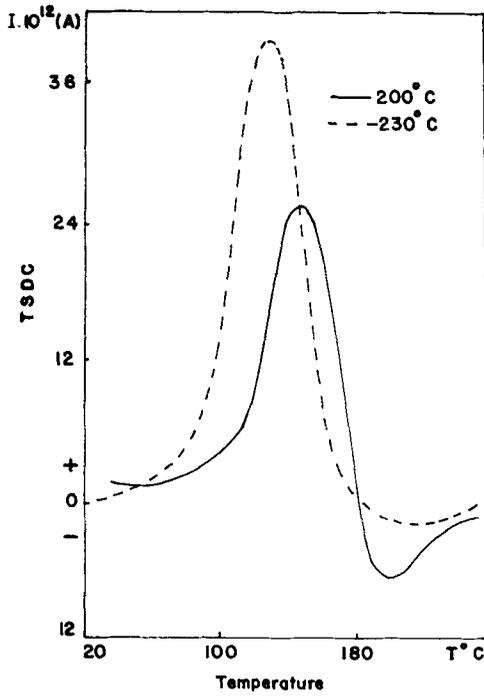


Figure 5. TSDC spectra for thermoelectret of polyimide film at the temperature of polarization 200°C respectively (electrode-electret-teflon-electrode system)  $E_p = 32.5$  kV/cm.

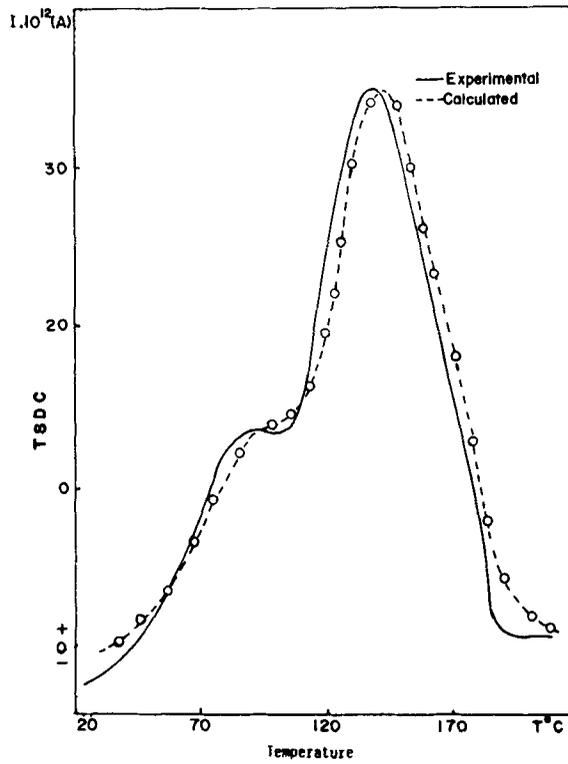


Figure 6. Experimental and calculated TSDC spectra for corona-electret of polyimide film at 20°C.

the residual polarization and the high temperature peak (200–230°C) may arise due to the relaxation of homo-charges. Thus the charge storages mechanisms in the polyimide film are different at different temperatures of polarization ranging from 20 to 230°C.

Figure 6 shows the calculated and experimental TSDC spectra for corona electret polyimide film in the EETC system at room temperature. Usually, homo-charges have been stored in the polymer film during the exposure of the corona discharging. The calculated (Baricova *et al.* 1982) TSDC spectra have been obtained by making use of equations (10) and (11). The calculated values and the experimentally obtained TSDC spectra agree (figure 6).

The results support the hypothesis that the dielectrics are heterogeneous with respect to the thickness, i.e. the conductivity of the film plays an important role in the relaxation process of the electret.

## 5. Conclusion

From the experimental results, it is clear that mainly homo-charges have been stored in the polyimide film and the role of the residual polarization is not important up to the temperature of polarization of 120°C under the electric field. Above this temperature major parts of the charges may be obtained due to dielectric polarization. Homo-charges may be stored in the polyimide film under corona discharges at room temperature.

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