

## Piezostimulated Current. A New Approach to the Study of Dielectric Relaxation with the Pressure as the Parameter

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We propose a new method for the study of the dielectric-relaxation phenomena called piezostimulated current, analogous to the one of thermally stimulated current. A dielectric is prepolarized and subjected to a very high hydrostatic pressure. The electric field is then removed and the depolarization current recorded as the pressure is lowered. The piezostimulated-current peak obtained for 6-polyamide enables one to calculate the following characteristics: activation volume  $\Delta V$ , activation enthalpy  $\Delta H$ , and activation energy  $E$ .

The dielectric-relaxation phenomenon in solid insulators has so far been studied either by the determination of dielectric losses under alternating fields at variable frequencies,<sup>1</sup> by means of charging- or discharging-current measurements of dielectrics subjected to a direct electric field,<sup>2</sup> or else by the thermally stimulated-current (TSC) method.<sup>3</sup> These methods enable us to determine the relaxation time of dipoles as well as their activation enthalpy, or that of the traps, as a function of the temperature. The new method that we propose, analogous to the TSC method, introduces another thermodynamic parameter, the pressure.

To obtain the simplest explanation of the principle, assume that the dielectric contains one type of dipole only. (1) An electric field  $E$ , at atmospheric pressure  $P_a$  and at temperature  $T$ , is applied to the dielectric for time  $t$  sufficiently long to enable the dipoles to be completely polarized. (2) Then, the sample is subjected to a very high hydrostatic pressure  $P \gg P_a$  that keeps the dipoles in their previous configuration. (3) Thereafter, the electric field is suppressed and the pressure is decreased progressively. The rate of the pressure decrease  $b = dP/dt$  is known. The dielectric remains short circuited as the pressure decreases, and consequently, the dipoles can reorient, resulting in a depolarization current [a piezostimulated current (PSC)]. This PSC has one or several peaks depending on whether the dielectric under consideration has one or more types of dipoles.

An analysis of a single PSC peak allows one to determine the relaxation time  $\tau$  for the corresponding type of dipole as a function of the pressure only, at a given temperature,  $[\tau(P)]_T$ . Following the application of an electric field at a

temperature  $T$  and pressure  $P$ , the dielectric acquires a polarization  $\mathcal{P}$ . During the depolarization, the variation of  $\mathcal{P}$ , as a function of time, is expressed by

$$d\mathcal{P}/\mathcal{P} = dt/\tau. \quad (1)$$

If the depolarization current is defined as

$$i(t) = d\mathcal{P}/dt \text{ where } \mathcal{P}(t) = \int_t^\infty i(t) dt, \quad (2)$$

we obtain

$$\tau(t) = \mathcal{P}(t)/i(t). \quad (3)$$

The value of  $\mathcal{P}(t)$  is determined from the partial area bounded by the curve  $i(t)$  and the time-axis between time  $t$  and  $\infty$ . Knowing the relationship between the pressure and the time from the rate  $b$  at which the pressure decreases, one can find

$$\tau(P) = \mathcal{P}(P)/i(P) \quad (4)$$

for a given temperature  $T$ .

In a first experiment, we examine the influence of  $b$  on the position and the form of the peaks obtained at 35°C for 6-polyamide (glass transition temperature  $T_g = 48^\circ\text{C}$ ; crystallinity 20%; water absorption rate = 3.5% for a relative humidity of 65%). The sample was polarized with an electric field of 22 kV/cm for 5 h.

We note on the curves of Fig. 1, obtained from the curves of  $i(t)$  and  $P(t)$ , the displacement of the peaks towards the low pressures and the development of three or four relaxation peaks when the rate  $b$  decreases from 4 to 2 bar/sec. It is obvious that this method is precise enough to study the various types of dipoles separately, by methods comparable with those used for TSC.

In this paper, we shall limit ourselves to studying the highest peak; this enables us, in an initial approximation, to use the highest rate  $b = 4$

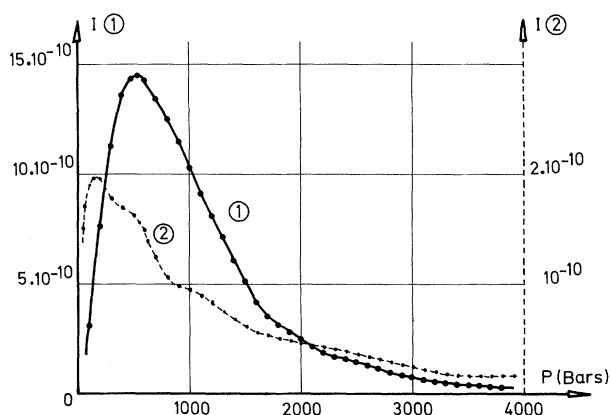


FIG. 1. Shape and the shift of the PSC for a test sample of 6-polyamide with pressure decrease rate,  $b = dP/dt$ . Curve 1 obtained with  $b = 4$  bar/sec; curve 2 obtained with  $b = 2$  bar/sec. Temperature  $T = 35^\circ\text{C}$ ; sample thickness,  $40 \mu\text{m}$ ; sample diameter,  $20 \text{ mm}$ ; crystallinity,  $20\%$ ; glass transition temperature,  $48^\circ\text{C}$ .

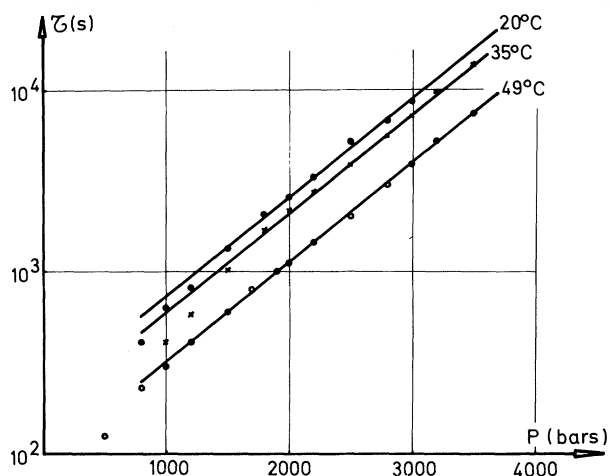


FIG. 2. Variation of relaxation time  $\tau$  with pressure  $P$  obtained from the PSC curve for temperatures  $20^\circ\text{C}$ ,  $35^\circ\text{C}$ , and  $49^\circ\text{C}$ , with  $b = 4$  bar/sec.

bar/sec.

Figure 2 shows the curves of  $\tau(P)$  computed from curves  $i(P)$  and from relationship (4) for three different temperatures,  $20^\circ\text{C}$ ,  $35^\circ\text{C}$ , and  $49^\circ\text{C}$ . They show a linear segment for  $P > 1000$  bar. For pressures less than  $1000$  bar, their curvature is perhaps due to the fact that the dielectric contains two or more types of dipoles and that at the relaxation time chosen, we partially take into account their existence. If we consider only the linear part of the curves, we can compute the activation volume  $\Delta V$ , the activation enthalpy  $\Delta H$ , and the activation energy  $E$  of the dipoles from the experimental results of Fig. 2.

We find that for a pressure of  $1500$  bar and a temperature of  $20^\circ\text{C}$

$$\Delta H = 0.23 \text{ eV},$$

$$\Delta V = 50.5 \times 10^{-30} \text{ m}^3, \text{ i.e., } 30.4 \text{ cm}^3/\text{mole},$$

$$E = 0.19 \text{ eV}.$$

These results are to be compared with those obtained by Loussier<sup>4</sup> on the absorption currents of 6-polyamide under pressure, with use of Fourier

series, which give an activation volume of  $36 \text{ cm}^3/\text{mole}$ .

This method enables us to make a rapid and precise evaluation of the characteristic values of 6-polyamide under pressure compared with other methods. Moreover, it is possible with low rates and with the washing trick analogous to that of the TSC method to separate the peaks and to study the various types of relaxation in the solids separately.

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