

Determination of the polarization distribution in poled ferroelectric polymer by the thermal pulse method

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Abstract

Three thick films of P(VDF-TrFE)(80/20) copolymer have been poled under different conditions. The polarization distributions in these films have been evaluated by least-square fitting the charge response induced by a thermal pulse.

I. Introduction

The determination of the polarization distribution from the observed charge response in a thermal pulse experiment requires the solution of an integral equation [1-5]. DeReggi et al. [3-5] showed that the charge response could be analyzed in terms of the Fourier coefficients of the polarization distribution in the sample.

An alternative procedure is used in the present work. The polarization distributions in three poled P(VDF-TrFE) films have been evaluated by applying a least-squares fitting procedure to the charge response as described below.

II. Method for Evaluating Polarization Distribution

DeReggi et al. [3-5] have given an expression for the short-circuit charge $Q(t)$ flowing in an external circuit that connects the electrodes on opposite surfaces of the sample. The charge response per unit area of a thermally pulsed sample with a polarization distribution $p(x)$ is given by

$$\frac{Q(t)}{A} = \frac{\alpha_c}{L} \int_0^L p(x) T(x, t) dx \quad (1)$$

where x is the depth measured from the thermally pulsed surface, t is the time lapsed, T is the temperature rise induced by the thermal pulse, L is the sample thickness, and α_c is the temperature coefficient of the capacitance.

If we define the relative polarization as $P(x) = \alpha_c A p(x) / L$, the numerical form of (1) at time t_j can be written as

$$Q(t_j) = \sum_{i=1}^n P(x_i) \Delta x_i T(x_i, t_j) \quad (j = 1 \text{ to } m) \quad (2)$$

By applying the least-square fitting procedure to $Q(t_j)$, we let

$$R = \sum_{j=1}^m [Q(t_j) - \sum_{i=1}^n P(x_i) \Delta x_i T(x_i, t_j)]^2 \quad (3)$$

$$\text{and take } \frac{\partial R}{\partial P(x_i)} = 0$$

We then obtain

$$\sum_{j=1}^m T(x_k, t_j) [Q(t_j) - \sum_{i=1}^n P(x_i) \Delta x_i T(x_i, t_j)] = 0 \quad (k = 1 \text{ to } n) \quad (4)$$

(4) can be rewritten in a matrix form

$$[T][Q] = [T][T]' [P\Delta x] \quad (5)$$

where $[T]'$ is the transpose of $[T]$. Therefore $P(x_i)$ can be evaluated by multiplying the left hand side of (5) by the inverse matrix of $[T][T]'$.

In the evaluation, the temperature rise $T(x_i, t_j)$ has been calculated using a well-known expression and the thermal diffusivity directly determined by a flash radiometry method [6]. For the calculation of $P(x_i)$ at any given x_i , 100 $Q(t_j)$ data points have been used.

III. Experiment

Three vinylidene fluoride-trifluoroethylene P(VDF-TrFE) (80/20) copolymer samples A, B and C each of thickness about 0.8mm, were studied. Sample A was poled under 16kV for one hour and then under 22kV for 2 hours at 110°C. Sample B was poled under

16kV for 2 hours at 115°C. Sample C was poled under 18kV for 2 hours at 90°C and reversely poled under 20.5kV for 3 hours at 110°C. Thin layers of silver paint were deposited on the two surfaces of the sample to serve as electrodes and also as absorbing layers for laser pulse.

One surface of the sample was irradiated by a pulsed beam (wavelength 1064nm, pulse energy 15mJ) of a Nd/YAG laser. Under a non-Q-switched mode, the pulse width is about 50μs. A digital storage oscilloscope was used to record the short-circuit charge response signal. Fig.1 a and b show the charge

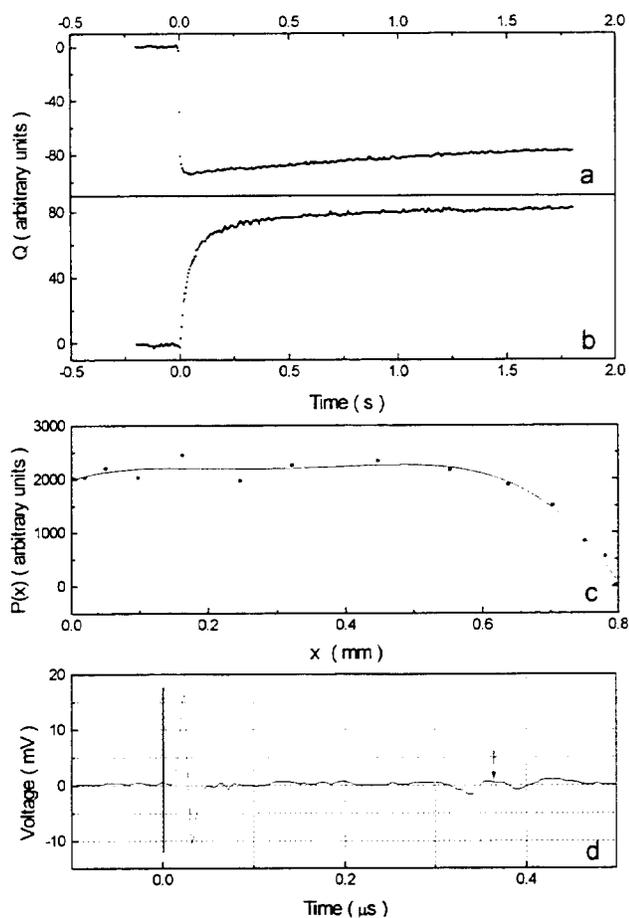


Fig.1. Experimental results for sample A. (a) and (b) are the thermal pulse response signals $Q(t)$ when the anode and cathode sides were irradiated by the pulsed laser, respectively. The square symbols in (c) show the polarization distribution evaluated using (5). The side $x=0$ corresponds to the anode on poling. (d) LIPP signal when the anode side was irradiated. The arrow shows the exit of the pressure wave at the cathode.

responses of sample A when irradiated by the laser at the anode and cathode sides, respectively.

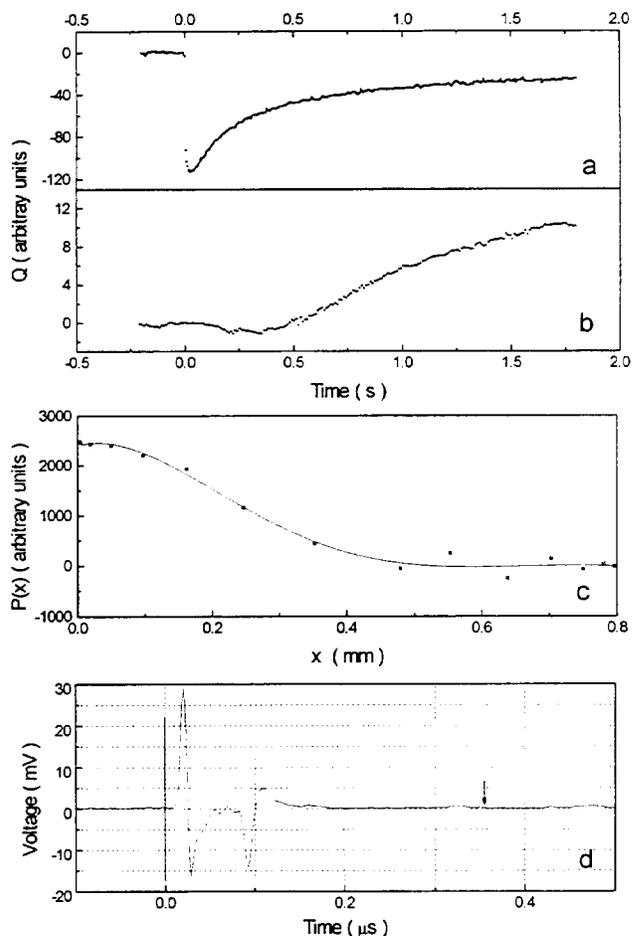


Fig.2. Experimental results for sample B. (a) and (b) are the thermal pulse response signals $Q(t)$ when the anode and cathode sides were irradiated by the pulsed laser, respectively. The square symbols in (c) show the polarization distribution evaluated using (5). The side $x=0$ corresponds to the anode on poling. (d) LIPP signal when the anode side was irradiated. The arrow shows the exit of the pressure wave at the cathode.

The charge response in Fig.1a reaches a minimum within 10ms, corresponding to a thermal diffusion depth of about 30μm, and then changes slowly with time. On the other hand, the charge response shown in Fig.1b rises slowly and becomes steady after 100ms, which corresponds to a diffusion depth of about 100μm. These results indicate that sample A is well poled at the anode side, but has an unpoled region at the cathode side. Fig.1c shows the polarization distribution evaluated using (5). The first half of $P(x)$

is calculated from the response data shown in Fig.1a while the rest of $P(x)$ is calculated from the data shown in Fig.1b. It is seen from Fig.1c that the sample has low polarization in a region near the

cathode occupying about 10% of the thickness, but rather uniform polarization in the remaining region. This agrees with the results of the laser-induced pressure pulse (LIPP) measurements performed using laser pulses of wavelength 1064nm and pulse width 10ns. (see Fig.1d).

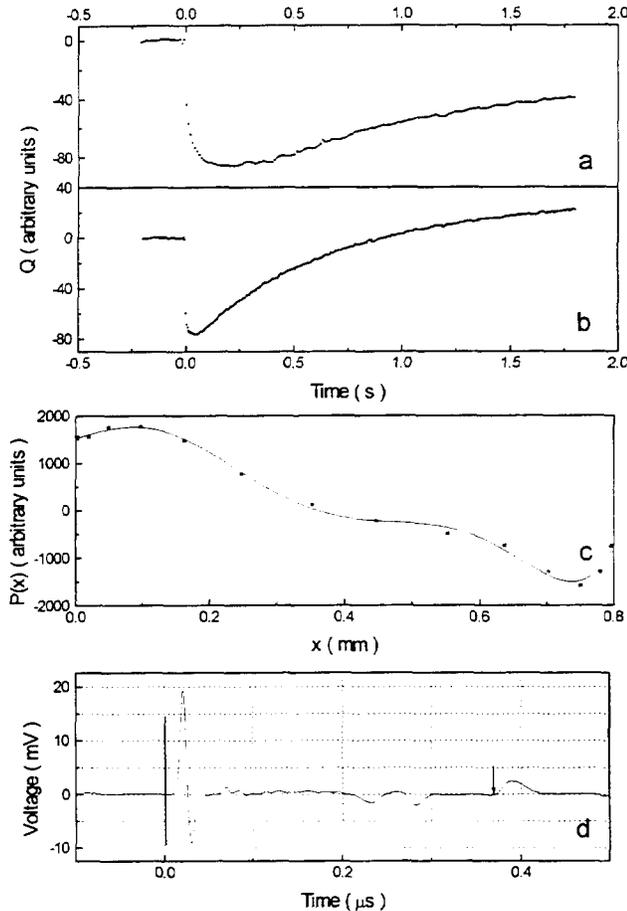


Fig.3. Experimental results for sample C. (a) and (b) are the thermal pulse response signals $Q(t)$ when the anode and cathode sides were irradiated by the pulsed laser, respectively. The square symbols in (c) show the polarization distribution evaluated using (5). The side $x=0$ corresponds to the anode on poling. (d) LIPP signal when the anode side was irradiated. The arrow shows the exit of the pressure wave at the cathode.

Fig.2 and Fig.3 are the results for sample B and C, respectively. It is clearly shown in Fig.2c that not more than 30% of the thickness of sample B is polarized. Since sample C is well poled on both sides, Fig.3c shows a positive polarization in the first half of the sample, but a negative polarization of almost the same magnitude in the second half. The LIPP results shown in Fig.2d and Fig.3d also agree with these thermal pulse observations.

IV. Conclusion

We have shown that the polarization distribution in a poled polymer can be evaluated by a least-squares fitting of the charge response induced by a thermal pulse. The calculated distributions are consistent with those deduced from laser-induced pressure wave measurements.

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