

Mechanism of bending electrostriction in thermoplastic polyurethane

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The mechanism of bending electrostriction in polyurethane films is discussed and elucidated through a numerical calculation. The simulations are carried out on a model in which charge carriers are assumed to be electrons injected from the cathode by the Schottky effect, and the positive charges are immobile. Under a dc field, our simulation results show that the electrons go out of the anode, leaving behind a large quantity of positive charge around the anode. As a result, the electric field near the anode eventually becomes much larger than that near the cathode. The asymmetrical electric field distribution leads to an asymmetrical stress distribution through the electrostriction effect and thus to bending of the polyurethane film under the application of a dc electric field. The results can also explain the gradual change in bending direction after reversing the polarity of the electric field. © 2004 American Institute of Physics. [DOI: 10.1063/1.1760837]

I. INTRODUCTION

The application of an electric field to some materials can displace charge and lead to field-induced strain along the thickness direction. The field-induced strain in thickness is proportional to the square of the applied electric field and this property of the materials is called electrostriction. Bending electrostriction was reported by Kawai in 1967 and 1970.^{1,2} He reported that a polyurethane film would bend by applying an electric field and its deformation was proportional to the square of the electric field. Thus, bending electrostriction is different from normal electrostriction. In recent years elastomeric polyurethane was found to generate large electric field-induced strain. However, the mechanisms leading to bending are not yet well understood and the details remain uncertain. In order to utilize polyurethane as a practical actuator, it is necessary to clarify its origin.

In their recent papers, Watanabe *et al.* reported that the bending deformation might result from an asymmetric distribution of space charge formed by a space-charge-limited current.³ They measured the space-charge distribution using the pulsed electroacoustic method⁴ and suggested that a possible cause of the bending was the repulsion between space charges. However, in our opinion this may not be the complete story because the bending disappears quickly once the applied field is removed, while the space charge distribution can only evolve more slowly. The bending mechanism of the polyurethane film has still not been studied in more detail. There has not been any theoretical model that satisfactorily explains the mechanism of the bending electrostriction of the polyurethane film in a quantitative way.

In this paper, a one mobile carrier model has been proposed to investigate the time dependence of current as well as the charge and electric field distributions across the polyurethane film during the application of electric field. A mechanism for bending based on electrostriction considerations is discussed. Numerical calculation shows that this

model can explain some of the bending features of the polyurethane film under an applied electric field.

II. THEORETICAL CONSIDERATIONS AND NUMERICAL CALCULATION

When an on/off series of dc voltages of 1200 V [Fig. 1(a)] is applied to the polyurethane film, the current passing through the polyurethane film behaves as shown in Fig. 1(b) and the bending displacement of the polyurethane film is shown in Fig. 1(c).⁵ The thinner lines in Fig. 1(a) represent a brief duration in which the voltage is turned off and then turned on again to its former value. In Figs. 1(b) and 1(c) spikes occur at corresponding times. A memory effect is noted here: in bending as well as in the current, the response returns to roughly its former value after the “off-on” spike. We shall have an explanation about this memory effect later on. It would be neater for the present discussion to discount all the voltage off-ons and therefore the response spikes, and simply consider a voltage input history as shown by the thicker lines in Fig. 1(a) and the broad responses in Figs. 1(b) and 1(c) without the spikes. Then we can divide the current-time (J - t) characteristic into three regions. In region I, the current initially decreases quite sharply with time and then tends to a near steady state. Upon voltage inversion in region II, the negative current increases first and then decreases with time, tending also to approach a steady value. In region III a current-time characteristic similar to that in field region II is obtained following another voltage inversion. Thus it suffices to limit the discussion to field regions I and II. As the polyurethane film is sufficiently thin, one-dimensional equations are applicable. We take the x axis in the direction of the thickness from the cathode $x=0$ to the anode $x=a$, where a is the sample thickness. Note in our convention, x is always measured from the cathode in each region, whether I or II.

We assume there are two kinds of charges in the polyurethane film. The positive and negative charge densities are $ep(x,t)$ and $-en(x,t)$, where e is electronic charge and p, n are number densities. The positive charges are immobile and

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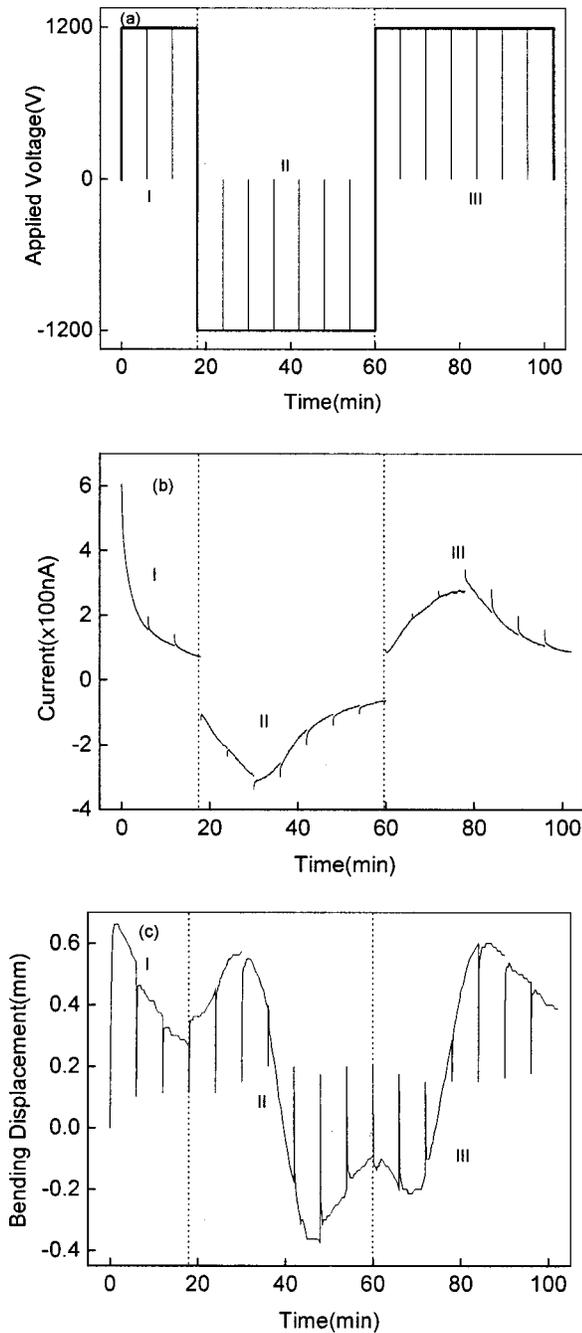


FIG. 1. (a) Time profile of the application of voltage across the polyurethane (PU). (b) The charging and reverse charging currents under the application of the on/off electric field. (c) The bending displacement of PU film under the application of the on/off electric field.

the mobile charge carriers are negative (electrons). The negative charge carriers' mobilities μ are assumed to be uniform across the film. The diffusion constant of electrons is assumed to be zero and any time-dependent polarization of the film material is neglected for simplicity.

When a positive field, whose average value is \bar{E} , is applied, the current flowing in the film is

$$J(t) = j(x,t) + \epsilon \frac{\partial}{\partial t} E(x,t), \quad (1)$$

$$j(x,t) = \mu en(x,t)E(x,t), \quad (2)$$

where ϵ is the permittivity of polyurethane. The electric field $E(x,t)$ is calculated from Poisson's equation

$$\epsilon \frac{\partial}{\partial x} E(x,t) = e[p(x,t) - n(x,t)] \quad (3)$$

and the circuit condition

$$\int_0^a E(x,t) dx = \bar{E}a, \quad (4)$$

where a is film thickness. The cathode is at $x=0$ and the anode at $x=a$.

Since the applied average electric field \bar{E} is constant throughout the charging (region I) or reverse charging (region II) process, the spatial integration of Eq. (1) gives

$$J(t) = \frac{1}{a} \int_0^a j(x,t) dx. \quad (5)$$

The negative charge carriers (electrons) are assumed to be injected from the cathode ($x=0$) by Schottky emission and drift to the anode ($x=a$) when the internal field $E(x,t) \leq 0$. As the boundary condition, the Schottky current is taken as

$$j(0,t) = -A_s \{ \exp(B_s \sqrt{|E(0,t)|}) - 1 \}, \quad (6)$$

where $A_s \propto T^2 \exp(-e\phi_B/kT)$ and $B_s = (e/2kT)(e/\pi\epsilon)^{1/2}$. The constant ϕ_B is the barrier height for electron injection at $E(0,t) = 0$.

The time derivative of the positive charge density is expressed as

$$\begin{aligned} \frac{\partial}{\partial t} p(x,t) = & -\frac{1}{\tau} n(x,t) + [N - p(x,t)] \\ & \times A_{PF} \exp(B_{PF} \sqrt{|E(x,t)|}), \end{aligned} \quad (7)$$

where $A_{PF} \propto \exp(-e\phi_D/kT)$, $B_{PF} = (e/kT)(e/\pi\epsilon)^{1/2}$, and ϕ_D is the energy depth of positive charge carriers at zero field. On the right-hand side of Eq. (7), the first term represents recombination with characteristic time τ , and the second term denotes Poole-Frenkel charge generation from sites of number density N . Then the rate of change of $n(x,t)$ follows from Eqs. (1) and (3).

For numerical calculation, we divide the sample into $m = 600$ equal parts in the direction of thickness, and x is replaced by $x_k = (k-1)\Delta x$, where $\Delta x = a/m$ and $k = 1, 2, \dots, m+1$. The densities $n(x,t)$ and $p(x,t)$ are approximated by the average values n_k and p_k in the k th division, respectively. The field E and the current j are the values at each division boundary. The derivatives are replaced by the difference quotients while integrations are approximated with the trapezoidal rule. Quantities at time $t + \Delta t$ are calculated by incrementing corresponding values at time t by the first order Taylor expansion terms in Δt .⁶⁻⁸

Now we are in a position to discuss bending of the film under an applied field. We assume that it is an electrostriction effect arising from the development of a nonuniform distribution of $E(x,t)$ across the film thickness. $E(x,t)$ produces stresses in x direction as well as in other directions with magnitudes proportional to $E^2(x,t)$. Let the y direction be

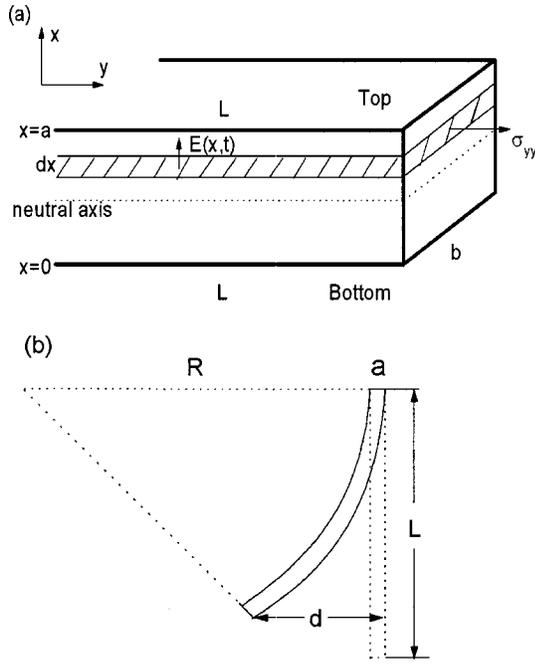


FIG. 2. (a) Diagram of the polyurethane film. (b) The schematic bending diagram.

pointing along the length of the film and the stress acting per unit area on a strip of area $b dx$ normal to the x direction and located at x [Fig. 2(a)] is expressed as

$$\sigma_{yy}(x,t) = \xi E^2(x,t). \tag{8}$$

Here ξ is an electrostriction coefficient of the film material and is a positive constant, and b is film width. The system of forces $\sigma_{yy}(x,t)b dx$ acting on various locations of the film cross section may be integrated into a resultant force $F(t)$ and a moment $M(t)$

$$F(t) = \int_0^a \sigma_{yy}(x,t)b dx, \tag{9}$$

$$M(t) = \int_0^a x\{\sigma_{yy}(x,t) - \bar{\sigma}_{yy}(t)\}b dx, \tag{10}$$

where $\bar{\sigma}_{yy}(t) = F(t)/ab$ is the mean stress acting on the cross section of area ab . $F(t)$ stretches the film while $M(t)$ provides the bending moment leading to the bending behaviors here investigated. In this paper we assume for simplicity that the pure bending theory is applicable. Thus the action of $M(t)$ bends the film to a curvature $R(t)$ given by

$$M(t) = \frac{Y}{R}I, \tag{11}$$

where I is the second moment of area and Y is Young's modulus. In calculating I we take the neutral plane at mid thickness [the dotted line in Fig. 2(a)]. Figure 2(b) is the film bending's schematic diagram in which L is the film length, a is the film thickness, and d is the bending displacement of the film. Because experimentally $L \gg d$, it is reasonable to neglect the expansion of the film for simplicity. Thus the relationship between R and displacement d can be expressed as

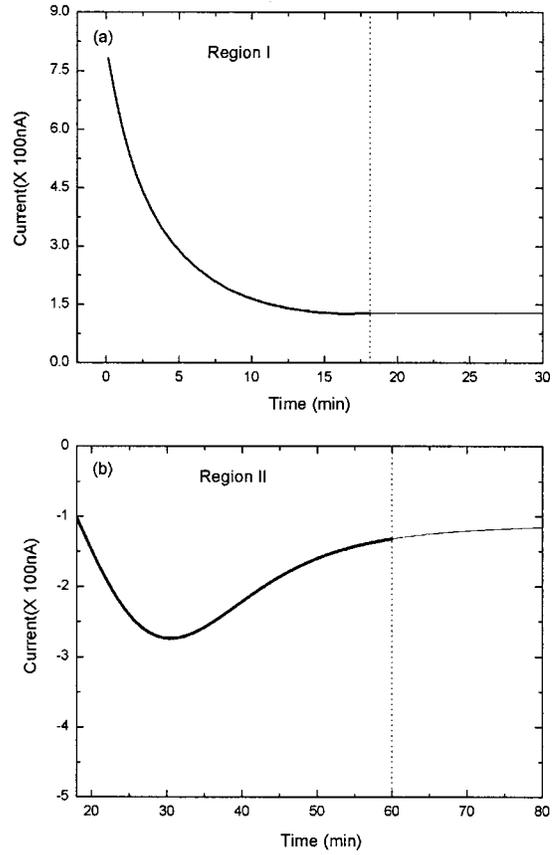


FIG. 3. The calculated $J-t$ curves (a) in region I and (b) region II.

$$d = \frac{L^2}{2R}. \tag{12}$$

From Eqs. (11) and (12), we get the displacement

$$d = \frac{L^2}{2YI}M(t), \tag{13}$$

which is the final equation we use to calculate the film displacement in the simulation.

Finally we discuss the memory effect displayed by the film's response before and after an off-on action in either region I or II. Because the charge density distribution in the polyurethane film evolves only very slowly, we assume that it remains the same before and after the off-on action of the applied voltage. Then from Eqs. (1)–(5), we can see that the electric field distribution and the total current flowing through the film will maintain the same values. The bending displacement of the film is calculated from Eqs. (9), (10), and (13) in our calculation; it will also maintain the same values as before the off-on interruption. This explains why the currents and the bending exhibit a memory effect.

III. RESULTS AND DISCUSSION

The values of some parameters which we use in the calculation are listed as follows: $\epsilon = 6.20 \times 10^{-11} \text{ C}^2\text{N}^{-1}\text{m}^{-2}$ ($\epsilon/\epsilon_0 \approx 7.0$), $a = 1.30 \times 10^{-4} \text{ m}$, $T = 300 \text{ K}$, $\bar{E} = 9.23 \times 10^6 \text{ V/m}$, $\mu = 5 \times 10^{-9} \text{ m}^2\text{V}^{-1}\text{s}^{-1}$, and $N = 10^{22} \text{ m}^{-3}$. By

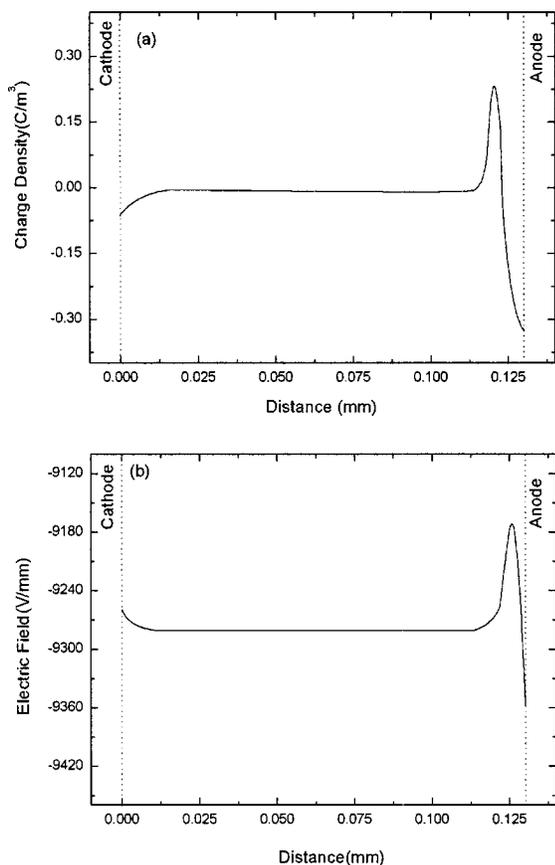


FIG. 4. Calculated space-charge distribution curve (a) and calculated electric field distribution curve (b) in the polyurethane film in region I at $t = 1$ min.

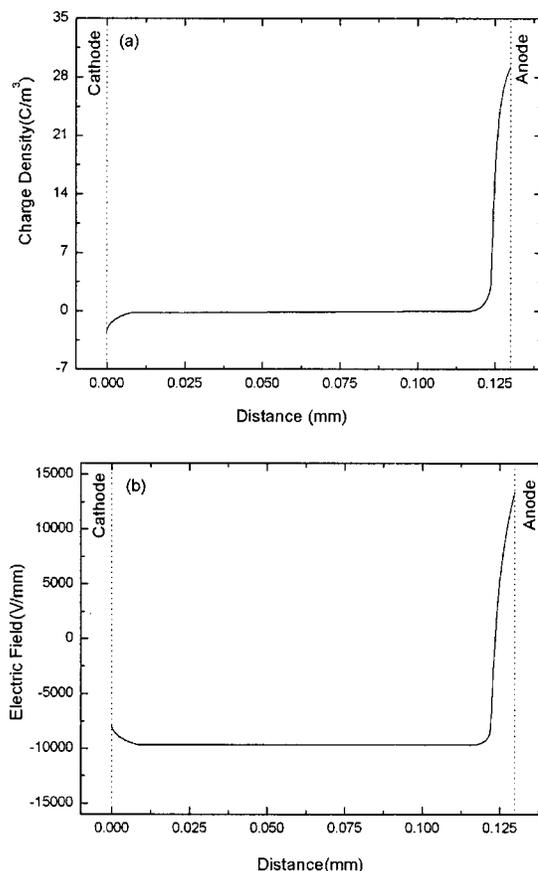


FIG. 5. Calculated space-charge distribution curve (a) and calculated electric field distribution curve (b) in the polyurethane film in region I at $t = 18$ min (before reversing the polarity).

comparing the experimental current with the injection current, we are led to choose 2.00×10^{-7} A/m² as the value of A_S .

The simulated $J-t$ curves in field regions I and II are shown as thicker lines in Figs. 3(a) and 3(b), respectively. They are roughly similar to the experimental ones and have the same order of magnitude (see Fig. 1). The thinner lines show a continuation of the theoretical response, should the applied voltage remain the same for a longer time.

When the dc voltage of $U = 1200$ V is applied to the polyurethane film at $t = 0$ min, the negative and positive charge densities are assumed to be equal across the film due to charge neutrality. The charge distribution $\rho(x,0) = e[p(x,0) - n(x,0)] = 0$ and the electric field $E(x,0) = -(U/a)$ is homogeneous, equal to the average electric field across the film. We take charge injection to occur in the next time step. Figure 4 shows the charge and electric field profiles at $t = 1$ min. Comparing the initial charge and electric field distributions with those at $t = 1$ min, we can see that there are some negative space charge accumulated near the cathode along with both positive and negative charges accumulated near the anode [Fig. 4(a)] and that the electric field near the anode is noticeably larger than the one near the cathode [Fig. 4(b)]. According to the bending electrostriction equation [Eq. (8)], the field-induced stress perpendicular to the thickness direction of the polyurethane film will be inhomogeneous.⁹ The stress near the anode will be somewhat

larger than the stress near the cathode and the part near the anode of the film will elongate more than the part near the cathode; hence the film will bend from the anode to the cathode. This conclusion agrees with the experimental results. Figures 5(a) and 5(b) are the calculated charge and electric field distributions at $t = 18$ min. From Fig. 3(a), we can see that the current has reached a near steady state at $t = 18$ min. At this time, positive charge accumulates near the anode along with a small amount of negative charge near the cathode [Fig. 5(a)]. The electric field near the anode is shown in Fig. 5(b). It is not easy to see by using a similar intuitive analysis as before that the film will also bend from the anode to the cathode. One has to rely on a calculation of bending moment from Eq. (11) to affirm that it has the same sign as at $t = 1$ min and hence the same bending direction.

Because the space-charge distribution can only evolve slowly, we use the charge distribution at $t = 18$ min of field region I as the initial charge distribution for field region II in our simulation [Fig. 6(a)]. In Fig. 6 note that the cathode is still labeled as $x = 0$ and the anode $x = a$ as before. According to the equation $\int_0^a E(x,t) dx = -U$ and the initial charge distribution, the initial electric field distribution [Fig. 6(b)] is obtained. From Fig. 6 we can see that the electric field near the cathode is much larger than the one near the anode. It explains why the film bends to anode side (the former cathode) first when the polarity of the electric field is reversed. Figure 7 shows the charge and electric field distributions in

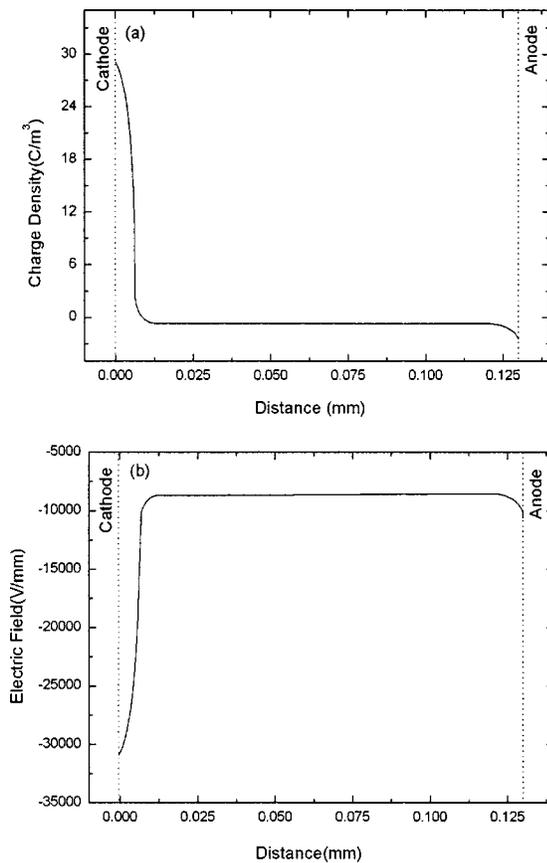


FIG. 6. Space-charge distribution curve (a) and calculated electric field distribution curve (b) in the polyurethane film in field region II at $t = 18$ min (after reversing the polarity).

field region II at an intermediate time $t = 28$ min. The positive charge density near the cathode has decreased while the positive charge density near the anode has increased. The electric field near the anode has increased from negative to positive. During the process, the electric field magnitude near the cathode (the former anode) is always larger than the one near the anode (the former cathode), and a calculation of the bending moment shows that the bending is in the same direction as before the polarity of the electric field is reversed at $t = 18$ min. The positive charges near the anode will continue to increase while the positive charges which are formerly accumulated near the cathode decrease. As time goes on, a new charge layer forms at the anode and the former charge layer diminishes at the cathode. Eventually the film will change its bending direction and bend from the anode to the cathode. Figure 8 shows the charge and electric field distributions in field region II at $t = 60$ min. From Fig. 3(b), we can see that the current has nearly reached a steady state at this time. At this state, there is only a small quantity of negative charge near the cathode but a larger amount of positive charge near the anode. The polyurethane film will thus bend from the anode to the cathode due to the asymmetric field distribution in the film. The profiles of the charge and electric field distributions are similar to the ones in field region I at $t = 18$ min (see Fig. 5). A similar bending response will be repeated in field region III.

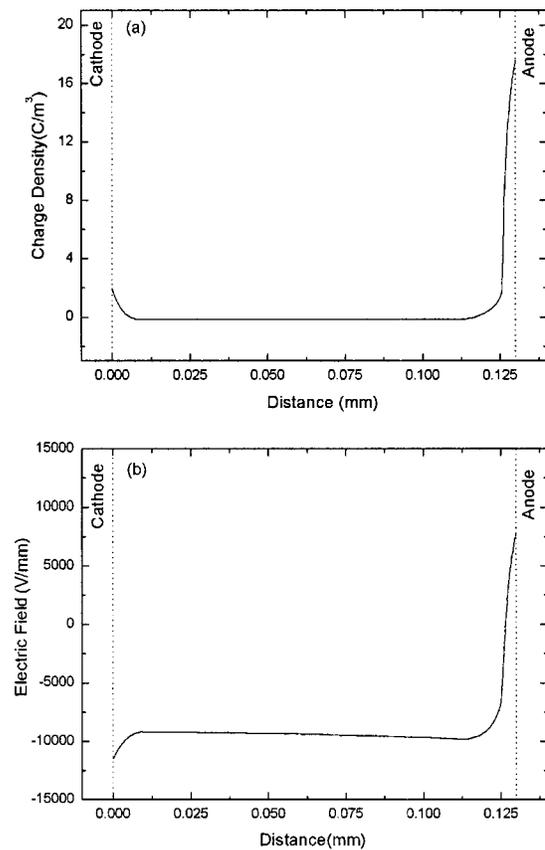


FIG. 7. Calculated space-charge distribution curve (a) and calculated electric field distribution curve (b) in the polyurethane film in field region II at $t = 28$ min.

Since it is our belief that the polyurethane film's bending results from the asymmetric stress in different parts of the film, we have calculated the bending displacement to test whether it can reach agreement with the experimental measurements [Fig. 1(c)]. In the quantitative calculation, we treat the polyurethane film's bending as a pure bending problem. We calculate the stress $\sigma_{yy}(x, t)$ in different divisions of the film according to Eq. (9) since $E(x, t)$ are known from the above. Then we calculate the moment $M(t)$ caused by the inhomogeneous stress by Eq. (10) and finally the bending displacement of the film's free end by Eq. (13). The results are shown in Fig. 9; again only the thicker lines are to be compared with the experiment. The shapes and bending magnitudes agree roughly with the experimental results except that the maximum bending displacements are a little larger than measured in both regions I and II. We think the discrepancy may come from the pure bending treatment, among others. Since we only aim to validate the bending mechanism of the film, we think it is reasonable for the simplified calculation.

In our simulation, the key features as observed in experiments are reproduced, namely (a) the tendencies of the current in regions I and II, (b) positive charges accumulate near the anode and a small amount of negative charge near the cathode at near steady state,¹⁰ (c) and furthermore, the calculated temporal change of the displacement of the film also agrees fairly well with the experimental results. The discrepancies between simulation and experiment may be attributed

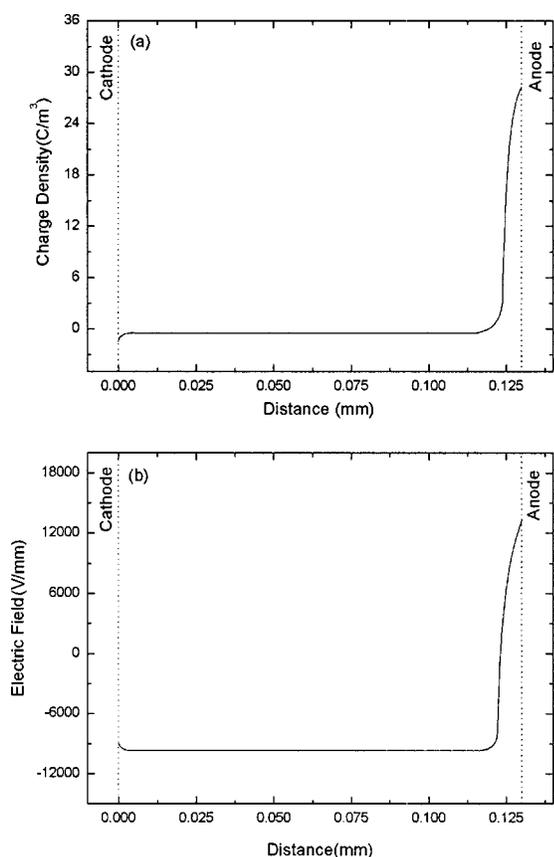


FIG. 8. Calculated space-charge distribution curve (a) and calculated electric field distribution curve (b) in the polyurethane film in field region II at $t = 60$ min.

to our assumption of negligible polarization and diffusion effect, of zero mobility of the positive charge, as well as the use of pure bending theory. Thus it is not unreasonable to suppose that the inhomogeneous charge distribution and hence inhomogeneous electric field distribution in the film is a possible origin of the bending behavior.

IV. CONCLUSIONS

We have considered a model for the electric field-induced bending behavior of polyurethane film. This model consists of negative charge carriers and immobile positive charges with Schottky injection and Poole-Frenkel charge generation. The simulation results show that the inhomogeneous charge distribution in the film causes an asymmetric electric field distribution which leads to inhomogeneous stress in the cross section of the polyurethane film, resulting in the bending behavior. We have presented an analysis of the evolution of the measured current and also quantitatively

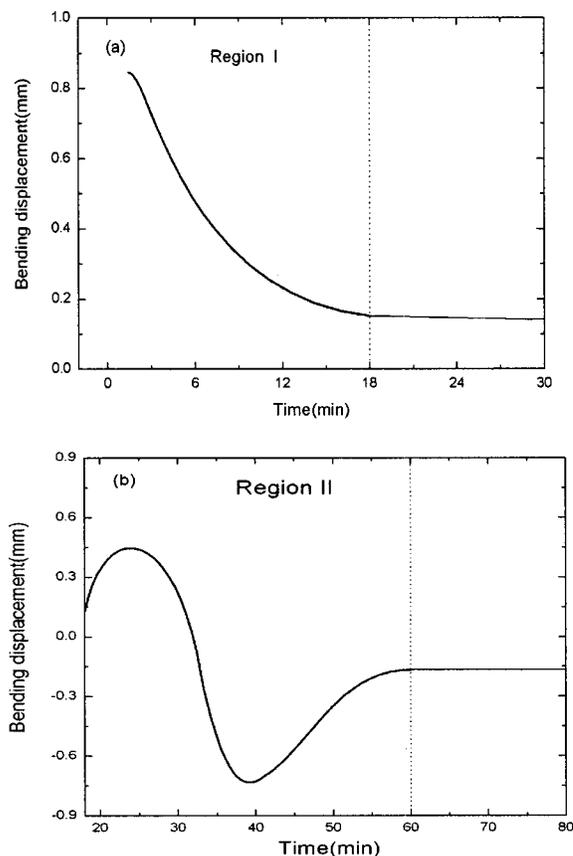


FIG. 9. Calculated bending displacements (a) in region I and (b) in region II.

calculated the bending displacement during the application of the electric field. The simulation results agree well with the experiment. The model may be further improved by considering nonzero mobility of positive charge carriers, diffusion of charge carriers, and polarization of the film material.

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