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Simulation of reverse piezoelectricity in ferroelectric polymers by Finsler geometry model

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ABSTRACT

Finsler geometry model is applied for simulation of deformation induced by external electric fields in ferroelectric polymers. Obtained strain-field curves are in a good agreement with reported experimental data for PVDF-based polymers. Thus, we find that Finsler geometry modeling is suitable for reproducing the reverse piezoelectric effect in ferroelectric polymers.

1. Introduction

Polymer films are widely used for creating actuators to convert the electrical energy into the mechanical energy. Polymer materials have better mechanical properties comparing with low-molecular ferroelectrics, but their shape-change caused by the electric field is typically very complex and difficult to simulate. Earlier, Finsler geometry (FG) technique has been successfully applied for modeling the shape transformation of liquid crystal elastomers under external electric fields [1]. In the present work, we further extend the FG model for describing the reverse piezoelectric effect in ferroelectric polymers. This model effectively implements the role of the positional and directional degrees of freedom in polymers.

2. The model

We use 3D FG model which was reported in [2]. For the simulation of a field-induced deformation, the Hamiltonian in [2] is modified by taking into account an additional dipole interaction with the external electric field. The continuous Hamiltonian S_1 corresponding to the Gaussian bond potential is given by:

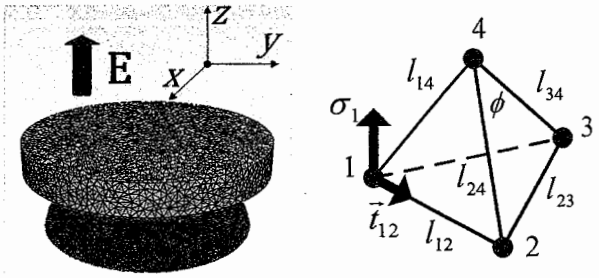
$$S_1 = \int \sqrt{g} g^{ab} \frac{\partial \vec{r}}{\partial x_a} \frac{\partial \vec{r}}{\partial x_b} d^3x, \qquad (1)$$

where $\vec{r}$ is a position vector of a three-dimensional body with coordinates $x_a (a = 1, 2, 3)$. The symbol g^{ab} is the inverse of metric tensor g_{ab} and g is its determinant.

To define the discrete Hamiltonian, we use a thin cylindrical body (Fig.1(a)). This body is discretized by tetrahedrons using the Voronoi tessellation. We introduce a variable σ_i, which corresponds to the direction of the dipole moment at the vertex i. The discrete metric tensor has the form: $g_{ab} = \begin{pmatrix} v_{12}^{-2} & 0 & 0 \\ 0 & v_{13}^{-2} & 0 \\ 0 & 0 & v_{14}^{-2} \end{pmatrix}$. This form is obtained from Euclidean metric by replacing its diagonal elements with v_{ij}^{-2}, where v_{ij} is the Finsler length given by the projection $\sigma_i \cdot \vec{t}_{ij}$ of σ_i on link $\vec{t}_{ij}$ of the tetrahedron such that

$$v_{ij} = \sqrt{1 - |\sigma_i \cdot \vec{t}_{ij}|^2} + v_0, \qquad (2)$$

where $\vec{t}_{ij}$ is an unit tangential vector of the bond ij (Fig.1(b)), and $v_0 = 0.001$ is a cutoff.



(a) (b)

Fig. 1 (a) The three-dimensional thin cylinder discretized by tetrahedrons and (b) a tetrahedron on which the discrete Hamiltonian is defined.

Replacing the integration in Eq.(1) with a sum over tetrahedrons and including the symmetric terms obtained by index replacement with the factor 1/4, we have a discrete version of S_1 [2]. With some extra terms, the discrete Hamiltonian for the FG model of the ferroelectric polymer is given by:

$$\begin{aligned} &S = S_1 + \kappa S_2 - d_1 S_3 - d_2 S_4 + U_{3D} + U_{vol}, \\ &S_1 = \textstyle\sum_{ij} \Gamma_{ij} l_{ij}^2, \ \Gamma_{ij} = 1/\left(4\bar{N}\right) \sum_{\text{tet}} \gamma_{ij}(\text{tet}), \\ &S_2 = \textstyle\sum_i (1 - \cos(\phi_i - \pi/3)), \\ &S_3 = \textstyle\sum_i \sigma_i \cdot \vec{E}, \quad S_4 = \textstyle\sum_i (\sigma_i \cdot \vec{E})^2, \\ &U_{3D} = \begin{cases} \infty & (V_{tet} \le 0) \\ 0 & (V_{tet} > 0) \end{cases}, \\ &U_{vol} = \begin{cases} 0 & |V - V_0| \le \Delta V) \\ \infty & (\text{otherwise}) \end{cases}, \end{aligned} \qquad (3)$$

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where κ, d_1 and d_2 are constants, $\vec{E}$ is the external electric field, l_{ij} denotes a length of the bond ij. The coefficients $\Gamma_{ij}(\Gamma_{ij}=\Gamma_{ji})$ are given by the sum of tetrahedrons sharing the bond ij, and γ_{ij}(tet) for the tetrahedron of vertices 1234 are determined via components of the metric tensor such that [2]:

$$\begin{aligned}\gamma_{12} &= \frac{v_{12}}{v_{13}v_{14}} + \frac{v_{21}}{v_{23}v_{24}}, \gamma_{13} = \frac{v_{13}}{v_{12}v_{14}} + \frac{v_{31}}{v_{32}v_{34}}, \\ \gamma_{14} &= \frac{v_{14}}{v_{12}v_{13}} + \frac{v_{41}}{v_{42}v_{43}}, \gamma_{23} = \frac{v_{23}}{v_{21}v_{24}} + \frac{v_{32}}{v_{31}v_{34}}, \\ \gamma_{24} &= \frac{v_{24}}{v_{21}v_{23}} + \frac{v_{42}}{v_{41}v_{43}}, \gamma_{34} = \frac{v_{34}}{v_{31}v_{32}} + \frac{v_{43}}{v_{41}v_{42}}.\end{aligned} \quad (4)$$

The second term κS_2 in Eq.(3) plays a role of deformation strength against bending and shear deformation, both of which have an influence on the material shape. The value of the parameter κ is approximately proportional to the ratio between flexural and elastic modulus of the material. ϕ_i in S_2 is an internal angle of the triangle. The third term $d_1 S_3$ is an energy of the dipole interaction with the external electric field, which causes the piezoelectric effect. $d_2 S_4$ is the potential describing the quadratic electrostrictive effect. U_{3D} is a constraint for the volume of the tetrahedron for not being negative. We assume that the material is perfectly incompressible and its value of Poisson ratio is equal to 0.5. To prevent the volume from changing, the last term U_{vol} is introduced, where ΔV is a mean value of the volume of one tetrahedron and V_0 is an initial volume of the whole cylinder, which is determined by the simulation without U_{vol} in the absence of external electric field.

3. Results

The simulation of electric field-induced deformation is carried out by Monte-Carlo (MC) method on a cylinder of size $(N, N_B, N_T, N_{tet}) = (10346, 69964, 116041, 56422)$, where N, N_B, N_T, N_{tet} denote the total number of vertices, bonds, triangles, and tetrahedrons, respectively. The MC updates of locations of vertices and σ are performed using the standard Metropolis algorithm. The ratio of the cylinder height and its diameter is equal to 0.125. The uniform external electric field is applied along the axis of the cylinder i.e. along the z axis.

The simulation results are presented in Fig.2. Fig.2(a) shows the dependence of diameter stretching ϵ_d on the external electric field E and comparison with experimental data for P(VDF-TrFE-CFE) films [3]. This dependence has a quadratic form with the plateau in the area of large deformation caused by polarization saturation. The unit of energy in our simulation is $k_B T = 4 \times 10^{-21}$[J] which is a room temperature. In Fig.2 d_1 and d_2 are pointed in units [MV^{-1}] and [MV^{-2}], respectively. In Fig.2(b) we compare our simulation results for longitudinal thickness shrinking with experimental data of highly electrostrictive networks of β-PVDF [4]. The networks are highly polar and have larger tensile strength comparing with uncrosslinked polymers. Thus, the simulation results for networks have a better fitting with experimental data for a smaller value of κ and larger values of d_1 and d_2 than in the case of P(VDF-TrFE-CFE) films.

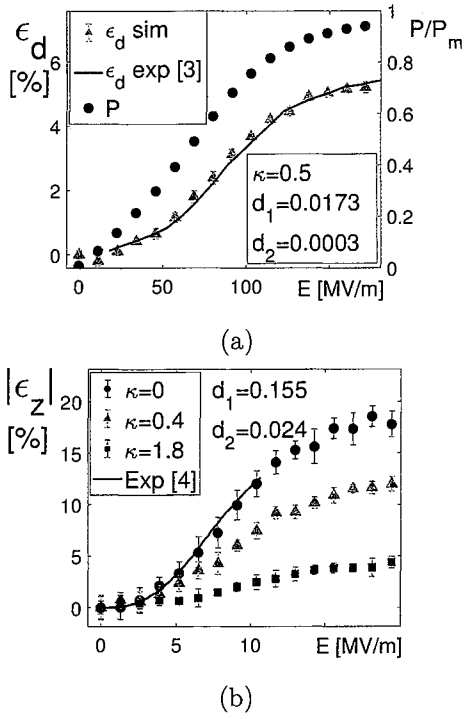

Fig. 2 (a) The diameter-strain ϵ_d (left axis) and the polarization P/P_m (right axis) vs. the external electric field E. P_m is the maximum possible value of polarization. (b) The thickness-strain $|\epsilon_z|$ vs. E with several different κ values.

4. Concluding Remarks

The model on the basis of FG is proposed for reproducing the reverse piezoelectric effect in ferroelectric polymers. The simulation results of deformation, induced by the electric field, are in good agreement with reported experimental data for both longitudinal shrinking and transverse stretching. Hence, we conclude that this model is suitable to predict the shape transformation of polymer actuators. However, compression of polymers was not taking into account in this work. Changing of polymer volume during deformation will be considered in further study.

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