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Converse electrostrictive effect in dielectric polymers

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a r t i c l e i n f o

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A B S T R A C T

This paper aims at exposing a physical model for converse dielectric electrostrictive effect using Debye/Langevin formalism through the use of Boltzmann's statistics. The principle of the proposed approach consists of considering the electric field orientability of dipoles within the polymer matrix as a function of the applied strain, allowing the derivation of the polarization or electric displacement as a function of the electric field and strain or stress. The proposed model also permits taking into account the saturation effect that occurs in the polarization and that limits the mechanical to electrical conversion in electrostrictive dielectric polymers.

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Thanks to their flexibility, low weight, low cost and processability, electroactive polymers have been of great interest in the research community for sensing and actuation applications (for instance artificial muscles, micropumps and low frequency, large strain systems $[1,2]$). In particular, electrostrictive polymers have received much attention because oftheir very low Young's modulus (down to a few MPa) that allows the design of large strain systems [\[3–7\].](#page--1-0) Because of these advantages, the use of electrostrictive devices has also attracted the attention of the research community for energy harvesting systems [\[8–12\].](#page--1-0) However, linearized electrostrictive effect is always used for the derivation of harvesting abilities, and no physical parameters are linked to the electrostrictive coefficients, preventing any optimization possibility in the material elaboration. Two classes of electrostrictive polymers are usually considered, owing to their ionic [\[13,14\]](#page--1-0) or dielectric origin [\[15–18\].](#page--1-0) Although ionic polymers exhibits higher strain abilities, their low frequency limit and low force may not be suitable for actuation, while dielectric polymers have a much higher cut-off frequency.

Nevertheless electrostrictive effect is still the subject of controversy in the scientific community. Phenomenological analysis such as the use of free energy is traditionally used to derive the constitutive equations of electrostriction $[19-24]$, but such an approach does not give significant physical insights to have a comprehensive understanding of such a coupling. In particular, a saturation in the response of electrostrictive actuators that

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appears in the high electric field region cannot be explained in a physical way using such an approach, although some models considered that such an effect lies in a hyperelastic behavior [\[25\],](#page--1-0) or pre-defined polarization saturation functions [\[26\]](#page--1-0) that have no direct link with the underlying physical mechanisms in the system.

However, Li et al. [\[27\]](#page--1-0) expose a physical model allowing the expression of the polarization using Brillouin and Langevin functions, hence relating the saturation effect from local considerations. However, their study is limited to the actuation case (i.e., direct electrostriction), and low-field behavior yielding almost linear relationship is assumed through the paper.

Meanwhile, Capsal et al. proposed the use of Debye/Langevin formalism to explain the origin of direct electrostriction in dielectric polymers from dipolar orientation for actuation modeling [\[28\],](#page--1-0) even in the high-field region, allowing the demonstration and validation of polarization saturation effect that limits the actuation abilities as well. In this model, the expression of the polarization P_3 along x_3 axis as a function of the electric field E_3 follows a Langevin function as [\[28\]:](#page--1-0)

$$
P_3 = N\mu_0 \mathcal{L}\left(\frac{\mu_0 E_3}{k_b \tau}\right),\tag{1}
$$

with N the dipole density, μ_0 the strain-free mean dipole moment magnitude, k_b the Boltzmann's constant and τ the temperature. $\mathcal{L}(x)$ denotes the Langevin function of the variable x $(\mathcal{L}(x)) =$ $\coth(x) - 1/x$).

Although the Langevin function describing the polarization saturation is well-known in the field of nonlinear optics [\[29\],](#page--1-0) these works provided its application to electrostriction and the effect on the strain that was still not well documented up to now.

Fig. 1. Modeling of dipole orientation.

The purpose of this paper is to extend such a model to converse electrostrictive effect, where the electric field-induced polarization is a function of the mechanical strain or stress [\[19\].](#page--1-0) While direct electrostriction is generally defined as a quadratic dependence of the strain to the electric field (contrary to piezoelectric direct coupling), converse electrostriction requires an activation mechanism in order to allow mechanical to electrical energy conversion, as dipoles need to be pre-oriented. Here, it is proposed to explain such an effect by the change in the macromolecule segment orientations caused by a compressive strain which changes the dipole angle with respect to the x_3 axis (Fig. 1). However the dipole orientation probability with respect to the macromolecule segment without any applied external electric field is still symmetric with respect to the x_1 axis, yielding a null global polarization.

Hence, without any applied strain, the mean dipolar moment magnitude μ_{0} with respect to the x_{3} axis along which the electrical solicitation is applied is given as $\mu_0 = \tilde{\mu} \langle \cos(\varphi) \rangle_0$, with $\langle \cos(\varphi) \rangle_0$ the mean cosine value of the macromolecule segment angle φ with the x_3 axis and $\tilde{\mu}$ the true dipole moment. In other words, as the dipole has only the rotation around the macromolecule segment as a degree of freedom, it would never be perfectly oriented along the x_3 axis. Under compressive strain $-S_3$, the macromolecule segments tend to become perpendicular to this axis, and thus the dipole may be better aligned with the x_3 axis yielding a higher polarizability. In this case, the expression of the dipolar moment becomes $\mu = \tilde{\mu} \langle \cos(\varphi) \rangle$, and the mean value of the cosine may be linked to the macroscopic compressive strain by considering the mean relative displacement difference along the x_3 axis as:

$$
-S_3 = \left(\frac{L_0(\cos(\varphi)) - L_0(\cos(\varphi))_0}{L_0(\cos(\varphi))_0}\right) \tag{2}
$$

where L_0 refers to the mean macromolecule segment length. Hence, it is therefore possible to express the dipolar moment as a function of the applied strain $-S_3$ and unstretched dipolar moment μ_0 as:

$$
\mu = \tilde{\mu}[(1 - S_3)\langle \cos(\varphi) \rangle_0] = (1 - S_3)\mu_0 \tag{3}
$$

Inserting this into Eq. (1) thus yields the expression of the polarization as a function of the applied strain:

$$
P_3 = N(1 - S_3)\mu_0 \mathcal{L}\left(\frac{\mu_0(1 - S_3)E_3}{k_b \tau}\right)
$$
 (4)

Hence, the application of a compressive strain results in a better orientation and alignment of dipoles with the electric field. This therefore permits increasing the macroscopic polarization of the material, as it has been empirically reported by some groups [\[30\].](#page--1-0)

Fig. 2(a) shows the comparison between experimental measurements of the polarization versus the electric field as well as theoretical predictions using the previously exposed model in the case of a polyurethane elastomer (PU) submitted to a longitudinal strain perpendicular to the electric field. Theoretical parameters have been obtained from manufacturer's datasheet and preliminarymeasurements giving a strain-free relative susceptibility value of 5.7 and a Poisson's ratio linking the strain along the perpendicular axis to the electric field to the compressive strain $-S_3$ parallel to the electric field of 0.49. Experimental data were recorded using the setup illustrated in [Fig.](#page--1-0) 3, which consists in a polymer sample clamped at one end to a Newport XMS50 ironless linear motor attached to a Newport SA series breadboard, at to the other end to clamps attached to the same breaboard. The linear motor is driven by an Agilent 33220A function generator through a Newport XPS motor controller. For the electrical part, the polymer was subjected to an AC or DC electric field applied by a Trek 10/10B high voltage amplifier driven by an Agilent 33220A function generator. A Keithley 428 current amplifier was used to monitor the current on an Agilent DS07034A oscilloscope. Displacement and voltage were also monitored through the oscilloscope. Hence, from the displacement, voltage and current and from the preliminary identification in terms of dimensions and material properties, it was possible to respectively derive the strain, electric field and polarization. These results clearly demonstrate a good agreement between experimental data and predicted polarization value from Debye/Langevin formalism, although this model does not relate the losses shown by the measurements.

Furthermore, the linearization of Eq. (4) considering small electrical and mechanical excitations permits expressing the

Fig. 2. Evolution of the polarization and low-electric field susceptibility for polyurethane polymer with several pre-strain values (measurement frequency: 1 kHz). Measurements have been done considering nominal quantities as the electric measurements were performed before and after stretching.

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