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Large piezoelectric strains from polarization graded ferroelectrics

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The potential applications of polarization graded ferroelectrics as high performance sensors and actuators are theoretically investigated. A static bending can be expected in polarization graded ferroelectric plates, forming a vertical displacement. This is due to the built-in strain gradient that arises from the grading of the composition and concomitantly, the spontaneous self-strain. Numerical results of two compositionally graded ferroelectrics, $BaTiO_3-Ba_{1-x}Sr_xTiO_3$ and $PbTiO_3-Pb_{1-x}Zr_xTiO_3$, show a high dynamic response of the displacement under an external electric field, yielding as much as ~23% strain at 50 kV/cm in $PbTiO_3-Pb_{0.6}Zr_{0.4}TiO_3$, which is comparable to large displacement actuators formed from ceramic/ceramic and ceramic/metal multilayer mesomaterials. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2358963]

Sensors and actuators are integral components in smart systems. They can be used to both sense and respond to external stimuli, such as pressure, temperature, voltage, and electric and magnetic fields. Over the past decade, ferroelectric (FE) and piezoelectric materials and structures have been the workhorses for active and passive systems.[1,2] These materials can be used for a myriad of applications including acoustic sensors for sonar, biomedical devices, positioners, pumps, switches, linear actuators, and microelectromechanical system devices. Homogeneous FEs, such as lead zirconate titanate ($PbZr_xTi_{1-x}O_3$, PZT), in particular, have been used for mechanical actuation with the magnitude of induced strains in the direct extension mode on the order of a few tenths of a percent although higher piezoelectric response and strains ($>1\%$) have been reported in single-crystal relaxor FEs.[3] More recently, new actuator/sensor devices called reduced and internally biased oxide wafer[4–7] (RAINBOW™) and thin unimorph driver[8,9] (THUNDER™) ceramics have been developed which routinely yield displacements on the order of tens of percent at modest field levels. These are essentially composite materials systems consisting of a piezoelectric ceramic layer bonded to a cermet (RAINBOW™) or a metal (THUNDER™) layer.[10] In both cases, the high displacement is the result of a built-in nonlinear stress field due to the thermal expansion mismatch between the layers.

In this study, we present theoretical results regarding the dynamic deformation characteristics of polarization graded FEs and show that a high displacement can be expected in such constructs due to the built-in stress field that arises from the grading of the spontaneous self-strain of FE-paraelectric (PE) structures. The polarization gradient may be achieved through systematical compositional variations and by setting up a temperature or strain gradient across a homogeneous FE.[11,12] The analysis indicates that the displacement in such constructs can be comparable to RAINBOW™ and THUNDER™ actuators. This theoretical methodology may be employed to design future structures for optimum performance while taking into account the structural and mechanical limitations of the base materials.

Consider a single-crystal compositionally graded FE plate of thickness h and lateral dimensions L such that $L \gg h$. We will assume that the easy axis of polarization is along the plate thickness such that $P_1=P_2=0$ and $P_3=P(z)$, where P_i are the components of the polarization vector. The plate is homogeneous in the lateral directions (x and y). The thermodynamic potential of a graded FE can be expressed through[13]

$$F=\int_{-h/2}^{h/2}\left[\frac{1}{2}a_1P^2+\frac{1}{4}a_{11}P^4+\frac{1}{6}a_{111}P^6+\frac{A}{2}\left(\frac{dP}{dz}\right)^2 - EP-\frac{1}{2}E_DP+F_{\mathrm{el}}\right]dz, \tag{1}$$

where a_1, a_{11}, a_{111}, and A are the free energy (Landau) expansion coefficients. a_1 is given by the Curie-Weiss law: $a_1=(T-T_C)/(C\varepsilon_0)$, where T_C is the Curie temperature, C is the Curie constant, and ε_0 is the dielectric permittivity of free space. We assume that the coefficients a_{11} and a_{111} are temperature independent. The Ginzburg coefficient A can be approximated as $\delta^2|a_1|$, where δ is a characteristic length along which the polarization varies. A is positive and thus the gradient term in the above relation acts as a restoring force that serves to damp out the spatial variations in P.[14] The inhomogeneous nature of the graded FE is reflected through the position-dependent expansion coefficients with respect to the composition such that $a_1(z)$, $a_{11}(z)$, $a_{111}(z)$, and $A(z)$. E is the electrical external field. The internal electric field (E_D) arises from the inhomogeneous distribution of the polarization and can be minimized by the formation of electric domains.[15]

The last term of Eq. (1) is the elastic energy of the built-in, position-dependent stress field. This energy results from the electrostrictive coupling between the polarization and the self-strain, and has two components: (i) the biaxial elastic energy due to the variation of the self-strain along h and (ii) the energy associated with the bending of the ferroelectric due to the inhomogeneous elastic deformation. A complete

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derivation of this elastic energy incorporating both sources has been given elsewhere.[13] Accordingly,

$$F_{\mathrm{el}}(z) = \bar{C}\left[\bar{\varepsilon} + \left(z - \frac{h}{2}\right)\bar{\kappa} - Q_{12}P^2(z)\right]^2, \tag{2}$$

where Q_{12} is the electrostrictive coefficient relating the strain in the x-y plane to polarization $P(z)$, $\bar{C}=C_{11}+C_{12}-2C_{12}^2/C_{11}$ is an effective elastic modulus, and C_{ij} are the elastic moduli at constant polarization. All these terms are spatially dependent upon z due to the compositional variation. $\bar{\varepsilon}$ and $\bar{\kappa}$ are the strain and curvature of the reference plane and can be calculated according to the condition that the net force and bending moment are zero,[16–18] i.e.,

$$\int_{-h/2}^{h/2} \sigma(z)dz = 0, \quad \int_{-h/2}^{h/2} z\sigma(z)dz = 0, \tag{3}$$

such that

$$\bar{\varepsilon} = \frac{1}{h}\int_{-h/2}^{h/2} Q_{12}P^2(z)dz, \tag{4a}$$

$$\bar{\kappa} = \frac{12}{h^3}\int_{-h/2}^{h/2} zQ_{12}P^2(z)dz. \tag{4b}$$

Minimizing the free energy equation with respect to polarization yields the Euler-Lagrange equation,

$$A\frac{d^2P}{dz^2} = \bar{a}_1P + \bar{a}_{11}P^3 + a_{111}P^4, \tag{5}$$

with renormalized coefficients

$$\bar{a}_1 = a_1 - 4\bar{C}Q_{12}\left[\bar{\varepsilon} + \left(z - \frac{h}{2}\right)\bar{\kappa}\right], \tag{6}$$

$$\bar{a}_{11} = a_{11} + 4\bar{C}Q_{12}^2. \tag{7}$$

Equation (5) can be solved numerically which yields the equilibrium polarization gradient and the initial (static) curvature of the compositionally graded FE.

In this study, we consider two compositionally graded systems: $\mathrm{BaTiO_3}$–$\mathrm{Ba_{1-x}Sr_xTiO_3}$ (BT-BST) with $0 < x \leqslant 0.25$ and $\mathrm{PbTiO_3}$–$\mathrm{Pb_{1-x}Zr_xTiO_3}$ (PT-PZT) with $0 < x \leqslant 0.6$. In this composition range, these FE materials have the prototypical perovskite lattice which transform from the PE cubic to FE tetragonal crystal structure upon cooling and are FE at room temperature. The thermodynamic parameters and the electrostrictive coefficients of these materials are known,[19,20] which allows us to perform the numerical analysis. Since the elastic moduli of PZT and BST have not been measured, in our calculations we used a linear averaging of the elastic properties of the end compositions $\mathrm{PbTiO_3}$, $\mathrm{PbZrO_3}$, $\mathrm{BaTiO_3}$, and $\mathrm{SrTiO_3}$.

The graded FE plate will form a dome shape similar to the RAINBOW™ and THUNDER™ ceramics due to the built-in strain gradient. We can determine the static displacement of this dome-shaped structure according to geometry shown in Fig. 1(b) as

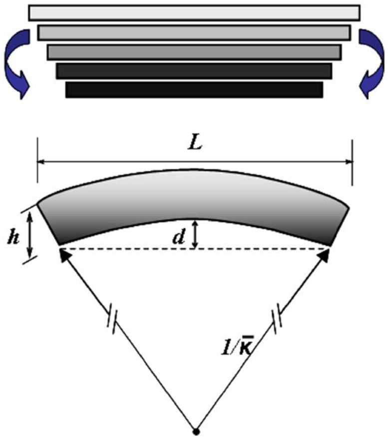

FIG. 1. (Color online) Schematic diagrams showing bending of a polarization graded ferroelectric plate. The gradient of self-strain results in a radius of curvature $\bar{\kappa}$ and a dome-shaped bending with displacement of d.

$$d(E) = \frac{1}{\bar{\kappa}} - \sqrt{\left(\frac{1}{\bar{\kappa}}\right)^2 - \left(\sin\frac{L}{2}\right)^2}, \tag{8}$$

where $\bar{\kappa}$ and L are the curvature and the lateral dimension of the plate, respectively.

The results show a large nonlinear strain along the plate thickness due to the built-in stress that results from the grading of the self-strain. For a plate with dimensions of $25.4 \times 25.4 \times 0.5\ \mathrm{mm}^3$ graded along the plate thickness, the dome that forms may have a height as much as ~101% and ~18% of the plate thickness for PT-PZT 60/40 and BT-BST 75/25, respectively. Graded BT-BST 75/25 has a lower spontaneous displacement due to an overall smaller spontaneous polarization and self-strain compared to graded PT-PZT 60/40.[21] This effect is comparable to RAINBOW™ and THUNDER™ ceramics of similar physical dimensions.[4] It should be noted that the dome shape in graded ferroelectrics is a result of the spatial variation of the self-strain while the dome shape in THUNDER™ devices is due to the thermal expansion coefficient difference between layers.

For actuator applications, the relative displacement under the external electric field is of primary interest. We can define this dynamic response as the "unblocked" displacement, ΔD, relative to the static curvature resulting from the compositionally induced strain gradient as

$$\Delta D(E) = \frac{d(E) - d(E=0)}{d(E=0)} \times 100\,\% . \tag{9}$$

Figure 2 shows ΔD for a $25.4 \times 25.4 \times 0.5\ \mathrm{mm}^3$ graded PT-PZT 60/40, PT-PZT 80/20, and BT-BST 75/25 plates, respectively. As the field increases, the relative displacement can reach as high as ~23% at 50 kV/cm for PT-PZT 60/40 though it is lower in PT-PZT 80/20 and BT-BST 75/25 due to the smaller polarization gradient. It is important to point out that the applied electric field opposes the spontaneous polarization within the graded structure. As the opposing electric field is increased, it will eventually suppress and reverse the polarization and the bending direction. In the case of the electric field along the polarization, the displacement will decrease with increasing electric field, i.e., negative ΔD. The reason for this is that the end with weaker polarization is more sensitive to the electric field, for example, PZT end and BST end, which has higher susceptibility and is thus much

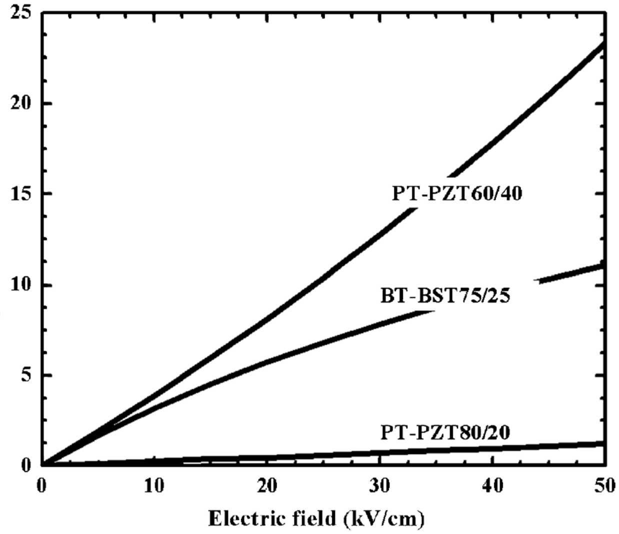

FIG. 2. Displacement relative to the static deformation caused by the compositionally induced strain gradient, ΔD, as a function of external electric field for PT-PZT 60/40, PT-PZT 80/20, and BT-BST 75/25.

easier to be polarized in the presence of the electric field. Therefore, the polarization gradient will be washed out and the resulting built-in strain will diminish.

A similar response would be expected from temperature-graded FEs. A systematic temperature variation across a homogeneous FE results in a commensurate polarization gradient and a built-in stress field.[22] A homogeneous FE can be thus be deflected either up or down depending upon the sign of the temperature gradient.

In a similar vein, flexoelectric materials have also received considerable attention wherein an induced polarization arises from a gradient in transverse strain in the transverse direction.[23,24] Here, the mechanically induced gradient plays the role of the strain gradient created in this study by a compositional variation. The flexoelectric effect peaks around the FE Curie temperature where there exists a maximum internal polarization gradient, similar to what has been found in other graded ferroelectric structures and devices.[12,13,22]

We caution, however, that the FE systems discussed in this letter are ceramic materials and are inherently brittle. Thus, the realization of such high deformations could be problematic due to cracking or delamination. The graded FEs chosen for this study should display the highest possible strain since the polarization gradient across PT-PZT and BT-BST is maximized for PT-PZT 60/40 and BT-BST 75/25. Mechanical failure could be avoided through employing smaller polarization gradients across the plate. The spontaneous strain would naturally be smaller but still significantly higher than the piezoelectric response of homogeneous FE or piezoelectric materials. Furthermore, the built-in stress might trigger the formation of structural (elastic) domains as to minimize its effect, the spontaneous displacement.

As shown theoretically, graded FEs might have a tremendous potential in sensors and actuators. We hope that these results will stimulate future experimental studies in these materials systems.

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