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Development and Electromechanical Properties of Multimaterial Piezoelectric and Electrostrictive PMN-PT Monomorph Actuators

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Abstract. Monolithic multimaterial monomorphs, comprised of varying ratios of piezoelectric $0.65Pb(Mg_{1/3}Nb_{2/3})O_3$-$0.35PbTiO_3$ to electrostrictive $0.90Pb(Mg_{1/3}Nb_{2/3})O_3$-$0.10PbTiO_3$, have been co-fired at 1150°C. The relative permittivity, displacement, and polarization hysteresis were investigated for varying ratios of piezoelectric to electrostrictive material. The permittivity of the 1:1 multimaterial monomorphs followed the dielectric mixing laws, showing a dielectric constant of 5,500 at room temperature. The P-E hysteresis loop of the 1:1 sample exhibited a maximum and remnant polarization slightly less than the piezoelectric PMN-PT 65/35, but higher than the electrostrictive PMN-PT 90/10. Displacement was found to be higher for the 3:1 monolithic monomorph actuators, reaching 76 μm at 6 kV/cm. The results indicate that by minimizing the electrostrictive layer thickness the tip displacement can be substantially increased while maintaining a lower hysteresis than that of the purely piezoelectric counterpart.

Keywords: piezoelectric and electrostrictive PMN-PT, monomorph actuators, permittivity, hysteresis, displacement

1. Introduction

Piezoelectric bending actuators are utilized as microscale positioners in precision displacement devices. For such applications, it is imperative to choose a piezoelectric material with high piezoelectric charge coefficients (d_{ij}), low dielectric permittivity (K_{ij}), and a high mechanical quality factor (Q_m) to ensure high displacement and a long lifetime. Soft piezoelectric materials are desirable for actuator applications because of their lower coercive fields (E_c), relatively low modulus of elasticity (Y_{ij}), and high piezoelectric charge coefficients (d_{ij}), and low dielectric loss ($\tan\delta$) [1]. For example, piezoelectric ceramics such as $0.65Pb(Mg_{1/3}Nb_{2/3})O_3$-$0.35PbTiO_3$ possess high remnant polarization (~40 μC/cm^2), low dielectric loss ($\tan\delta$ ~2.5%), relatively high longitudinal piezoelectric charge coefficient (650 pC/N), and are both electrically and elastically soft. Furthermore, low mechanical loss by way of low hysteresis is needed to achieve durability of the component [2]. In order to concurrently accomplish high displacement and positioning accuracy, electrostrictive materials have also been proposed for bending actuator applications [1]. Electrostrictors have theoretically no hysteresis, which is ideal to achieve high precision microscale positioning and low mechanical loss. Moreover, electrostrictive materials can produce large field-induced strains (x_i) since it is a second order (nonlinear) effect, i.e. $x_{ij} = M_{ijkl}E_kE_l$ $(i, j, k, l = 1, 2, 3)$ [1].

Conventional bender type designs of current interest include bimorph, unimorph, and functionally graded monomorph (FGM) actuators. Bimorph actuators typically consist of two piezoelectric active layers that are mechanically in parallel and electrically in series or parallel; and are bonded together by a non-active layer such as epoxy. The discontinuity of elastic properties across the thickness of bimorph actuators render the bonding interface strength weak and makes it susceptible to untimely failure [3]. Alternatively, a unimorph is comprised of a thin metal layer bonded to a ceramic

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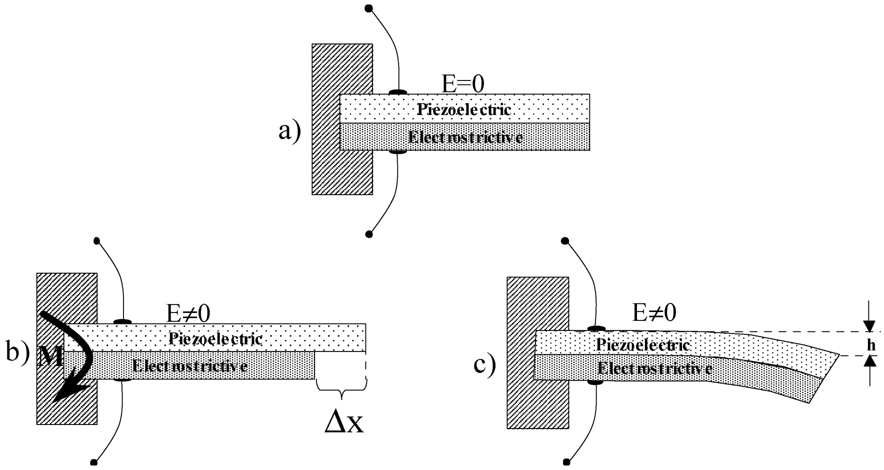

Fig. 1. (a) Typical bender configuration used in this study (b) Application of an electric field causes a transverse differential strain Δx (c) Tip deflection caused by transverse differential strain.

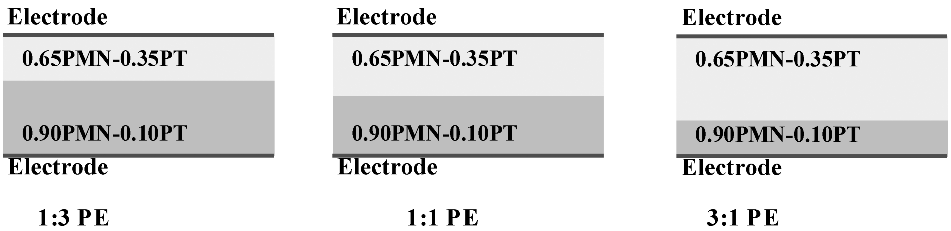

Fig. 2. Multimaterial piezoelectric and electrostrictive configurations.

plate. When an electric field is applied, a bending deformation is generated due to the constraint at the interface between the piezoelectric layer and the metal elastic layer [4].

Functionally graded monomorph (FGM) materials are designed to have a gradual change in the composition and properties and to help alleviate the sharp discontinuity at the interface. The FGM configuration allows for functionally graded materials to incorporate reliability. In order to create bending deformation, it is required that the piezoelectric material on opposite sides of the actuator generates different in-plane strains under an electric field in the thickness direction [5]. The cyclic fatigue of the FGM actuators have improved the durability compared to the traditional bimorph actuators despite the internal stress that makes the cyclic fatigue difficult to quantify [5–8].

In the present study, we have fabricated a co-sintered monomorph bending actuators that is based on electrostrictive and piezoelectric materials, and report their electromechanical properties. For this type of actuator, transverse strains opposite in sign can be generated similar to that of a bimorph when operated under an electric field oriented anti-parallel to the poling direction as shown in Fig. 1. Piezoelectric $0.65Pb(Mg_{1/3},Nb_{2/3})$-$0.35PbTiO_3$ (hereafter PMN-PT 65/35) and electrostrictive $0.90Pb(Mg_{1/3},Nb_{2/3})$-$0.10PbTiO_3$ (hereafter PMN-PT 90/10) are the materials of choice since they have compatible thermal expansion coefficients and sintering behavior. The purpose of this paper is to quantify the electromechanical properties of co-sintered multimaterial monomorph actuators as a function of piezoelectric volume fraction (Fig. 2), and to provide a phenomenological description of their dynamic behavior.

2. Experimental Procedure

The piezoelectric and electrostrictive compositions used in fabricating multimaterial monomorph actuators were PMN-PT 65/35 and PMN-PT 90/10, respectively. The ceramic powders (TRS Ceramics, PA, USA) were of ~2 to 3 μm in average particle size, and were used to synthesize filament feedstock to be stacked layer-by-layer for warm pressing. The processing steps appertaining to preparation of filament feedstock included coating of the powders with stearic acid surfactant, mixing the powder with a proprietary binder, and extrusion of the powder-binder mixtures by employing a single screw extruder [9]. The solids content of the two separate filament feedstock were 57 and 54.5% by volume for 0.65PMN-0.35PT and 0.9PMN-0.1PT, respectively. Further details of filament feedstock preparation can be found in refernece [9, 10].

Multimaterial monomorph actuators with piezoelectric to electrostrictive volume ratios 1:1, 1:3, and 3:1 were studied. The volume ratios were equivalent to the layer thickness ratios since the surface areas are the same for the piezoelectric and the electrostrictive layers as shown in Fig. 2. These monomorph actuators have been fabricated by the warm pressing method at 85°C, utilizing a Carver Laboratory Press Hot Platens (Fred S. Carver Inc., Menomonee Falls, WI, USA). The green samples were subjected to a binder burnout cycle at 550°C. The resultant performs were then co-sintered at 1150°C for 2 h in a controlled PbO atmosphere. The average final dimensions of monomorph actuators were 30 mm in length (l) by 8 mm in width (w), and 1.3 mm thickness (t). The relative sintered density of the samples was typically 97% of theoretical density.

The samples were electroded with silver epoxy, and then poled along the thickness direction in a silicon oil bath with 20 kV/cm electric field strength for 20 minutes at 110°C. The electromechanical properties were characterized after aging the samples for 24 h.

The dc-mode displacement of the actuators was determined as a function of applied electric field strength using a differential variable reluctance transducer (Microstrain Inc., Burlington, VT, USA). The dielectric constant and loss tangent of the samples were measured at 1 kHz from the temperature range of −100 to 210°C using an environmental chamber (Delta Design, Model 9023, Poway, CA, USA) in conjunction with an impedance gain/phase analyzer (HP 4194, Hewlett Packard, Palo Alto, CA, USA). The polarization versus electric field traces were determined at 10 Hz with a Radiant Precision LC system (Radiant Technologies, Albuquerque, NM, USA). A Berlincourt d_{33} piezometer (Channel Products Inc., CA, USA) was used to measure the effective piezoelectric charge coefficient of the composites at 100 Hz.

3. Results and Discussion

To establish a baseline for the dielectric response of the composite actuators, the permittivity and dielectric loss of the constituent systems were assessed quantitatively as a function of temperature. Figures 3(a) and 3(b) show the variation of relative permittivity (K_{33}) and loss tangent (tan δ) as a function of temperature for 0.65PMN-0.35PT and 0.9PMN-0.1PT at 1 kHz, respectively. The PMN-PT 65/35 system exhibits paraelectric (PE) m3m to ferroelectric (FE) 4 mm transition at 174°C, at which the relative permittivity (K_{33} @ T_{tr}) reaches 41,000. The room temperature (RT) K_{33} is ~3500, while the loss tangent is ~2.5% in the same range (see Fig. 3(a)). On the other hand, the PMN-PT 90/10 system is paraelectric at RT. The PE-FE phase transition takes place at 20°C at which K_{33} @ T_{tr} is ~23,000. The dielectric loss is 1% from 200°C down to 50°C, and increases to 11% at the onset of the PE-FE phase transition. The K_{33} for PMN-PT 90/10 at RT is ~19,000.

Figure 4 depicts the relative permittivity vs. temperature at 1 kHz for the 1:1 monomorph, which is 50% piezoelectric and 50% electrostrictive by volume. The maximum relative permittivity for this monomorph is 7000 at 65°C, while the RT value of the same is ~5500. A slight shoulder is revealed at 174°C, which is attributed to the PE-FE phase transition of the piezoelectric PMN-PT 65/35 composition of the monomorph. The dielectric loss (tan δ) for the 1:1 monomorph ranged from 2% to 4%, as the temperature is increased from 0 to 50°C.

To gain insight into the evolution of the dielectric response in the 1:1 monomorph, the measured dielectric properties were compared with the permittivity computed using the harmonic mixing law given by [11, 12]

$$\varepsilon_{\Sigma}(T) = \frac{\varepsilon_1(T)\varepsilon_2(T)}{V_1\varepsilon_2(T) + V_2\varepsilon_1(T)}, \qquad (1)$$

where ε_{Σ} is the permittivity of the monomorph, ε_1 is the relative permittivity of the piezoelectric, ε_2 is the relative permittivity of the electrostrictor, and T is the temperature, respectively. In Eq. (1), the volume fraction of the constituents are given by V_1 (piezoelectric) and V_2 (electrostrictor), with $V_1 + V_2 = 1$. Using this equation, the permittivity of the 1:1 monomorph is computed

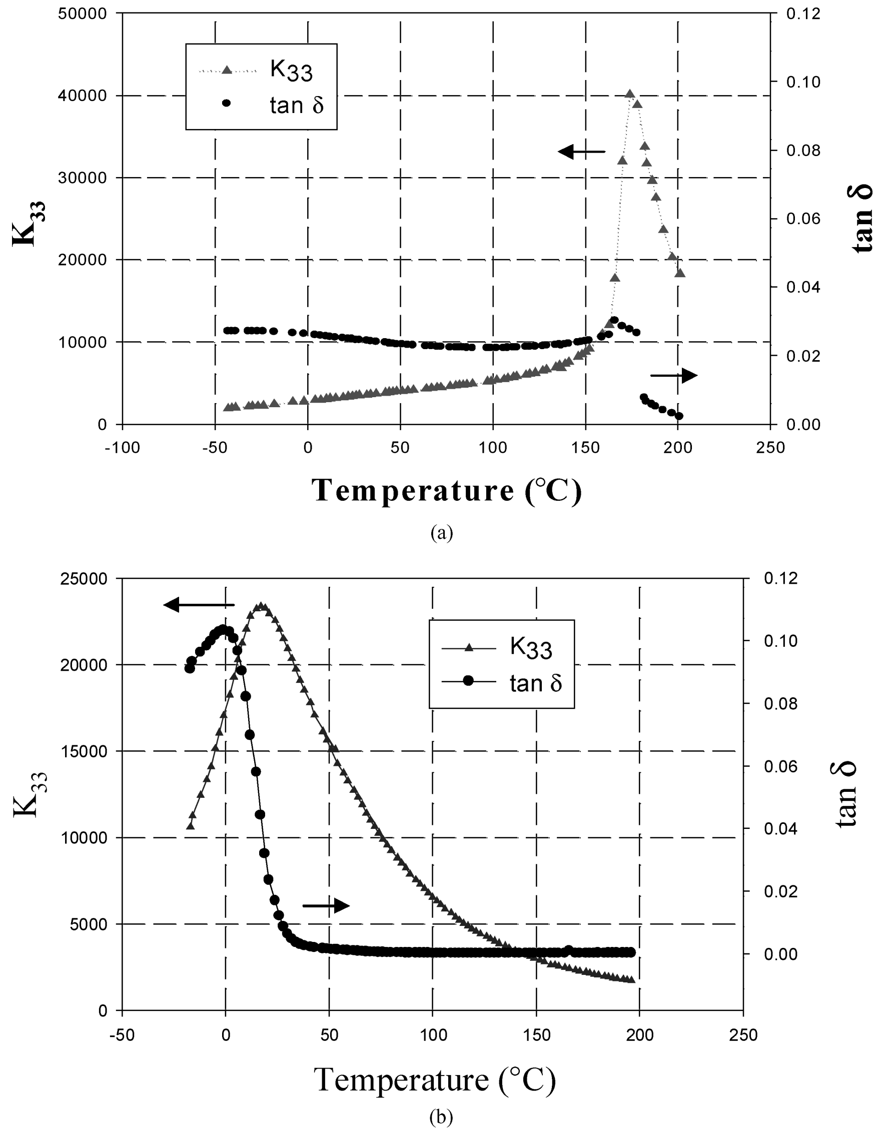

Fig. 3. Relative permittivity and loss tangent vs. temperature traces for (a) 0.65 PMN-0.35PT (b) 0.9PMN-0.1PT at 1 kHz.

and shown in Fig. 4. The computed value is 5,800 at RT, and is in conformity with the measured value of 5,500.

The temperature variation of the composite's permittivity, as computed from Eq. (1), also is in reasonable agreement with the experimentally measured composite trace. It seems that the measured trace is a superposition of the measured traces of PMN-PT 65/35 and PMN-PT90/10, respectively.

The temperature dependence of the relative permittivity and loss tangent for the 1:3 monomorph is shown in Fig. 5, where it is seem that $K_{\max}$ is 11,500 at RT. The loss tangent varies from 1 to 7.5% over the range 0–50°C. At the phase transition temperature, the dielectric loss is at its maximum just below T_c, and then falls off. This behavior clarifies the pronounced range of tan δ for the 1:3 monomorph from 0–50°C. In this composite, the measured permittivity trace is dominated by the dielectric behavior of the electrostrictive PMN-PT 90/10. In this case, there is no trace of a shoulder in the vicinity of 174°C as seen in the 1:1 composite. This behavior is indeed in conformity with harmonic

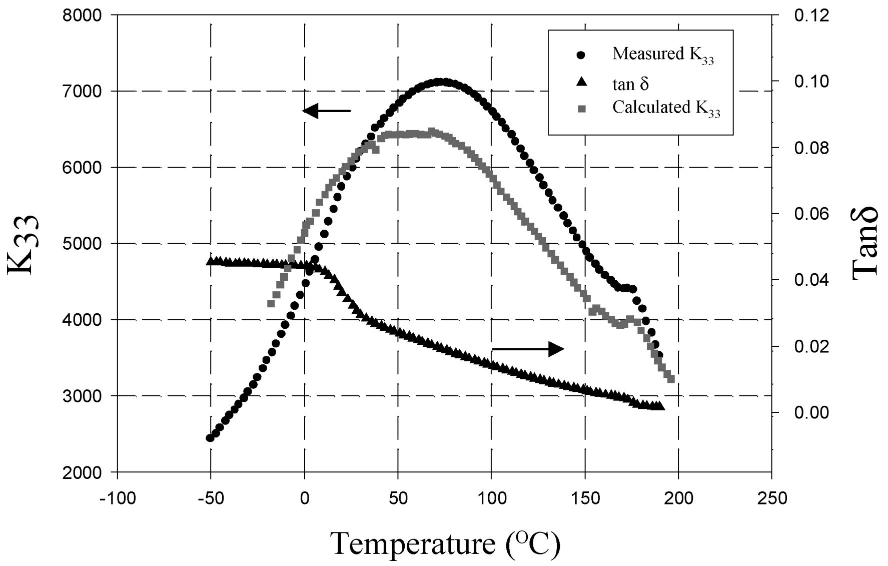

Fig. 4. Experimental and calculated variation of relative permittivity with temperature for 1:1 monomorph. Harmonic rule of mixing used in the calculation.

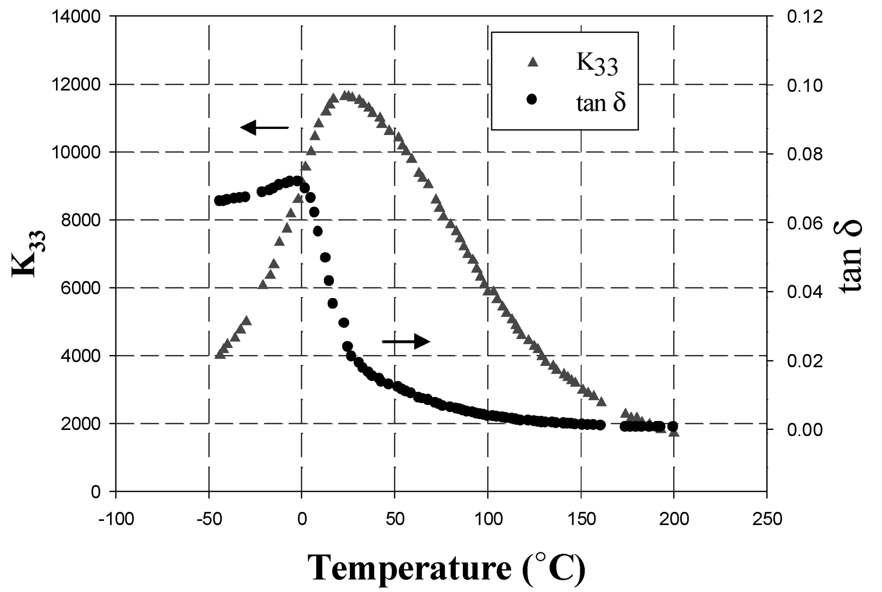

Fig. 5. Relative permittivity and loss tangent vs. temperature for 1:3 PE monomorph.

mixing, considering the fact that the piezoelectric and the electrostrictive layers are electrically in parallel [12].

As shown in Fig. 6, the 3:1 monomorph actuator exhibits a phase transition at 174°C. The observed dielectric response is reminiscent to that of PMN-PT 65/35, indicating that the piezoelectric material dominated the trace by virtue of its higher volume fraction in the composite. There is a noticeable decrease in K_{max} (∼11,000) at T_c. For temperatures ranging form 0–50°C, the tan δ spans from 2.8 to 3.5%. It should be noted that the dielectric loss is substantially lower for the 3:1 monomorph in comparison to the 1:3 in the temperature range 0–50°C.

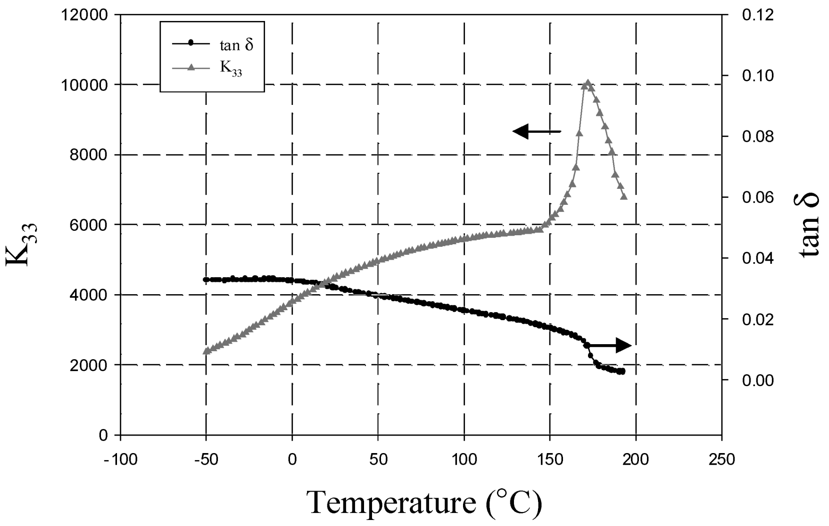

Fig. 6. Relative permittivity and loss tangent vs. temperature for 3:1 PE monomorph.

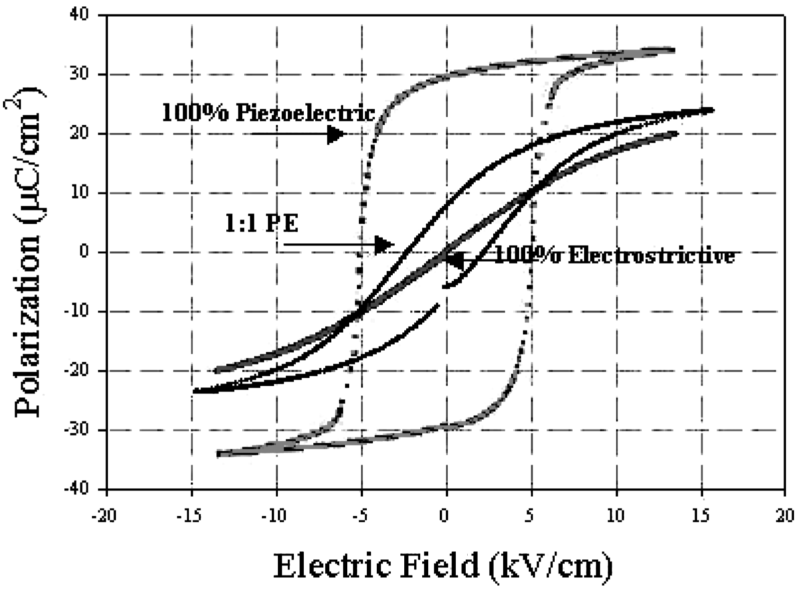

Fig. 7. P-E Hysteresis Loops for 65/35 PMN-PT-single material, 90/10 PMN-PT single material, and 1:1 PE monomorph.

In Fig. 7, the hysteresis loop of the 1:1 monomorph is compared with the ones for piezoelectric PMN-PT 65/35 and electrostrictive PMN-PT 90/10, respectively. The PMN-PT 65/35 exhibits typical ferroelectric behavior with $P_r = 30\ \mu C/cm^2$, $P_{max} = 33\ \mu C/cm^2$, and a coercive field E_c of 5kV/cm. The PMN-PT 90/10, on the other hand, shows typical paraelectric behavior with a $P_{max} = 20\ \mu C/cm^2$ at 13.0 kV/cm. The 1:1 monomorph has a $P_{max} = 24\ \mu C/cm^2$, a remnant polarization $P_r = 9\ \mu C/cm^2$, and a coercive field, E_c of 2.0 kV/cm. The P_{max} for the 1:1 composite is very close to that of the pure piezoelectric phase, meanwhile the P_r is drastically reduced. Neither parameter (P_{max} and P_r) obeys the harmonic mixing law, which indicates that the evolution of the monomorph properties is much more complex, and is most likely controlled by the interface

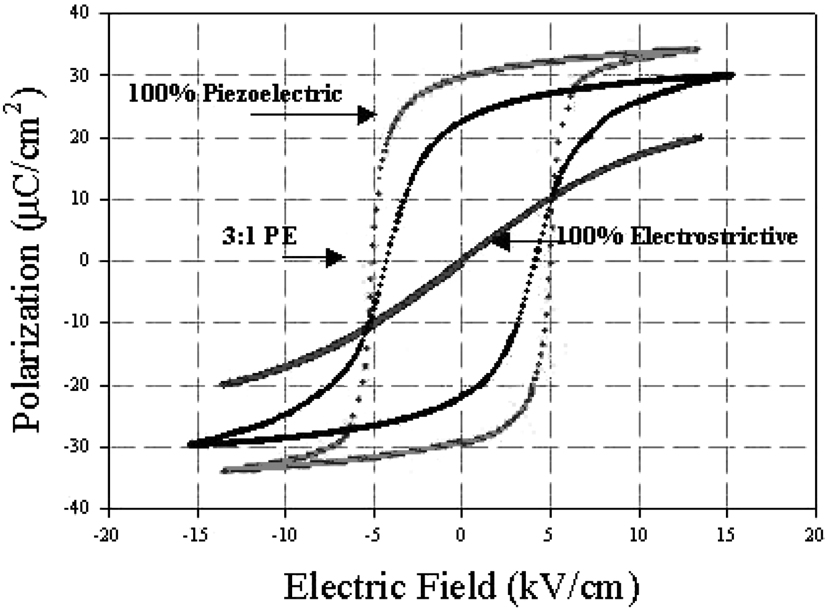

Fig. 8. P-E Hysteresis Loops for 65/35 PMN-PT-single material, 90/10 PMN-PT single material, and 3:1 PE monomorph.

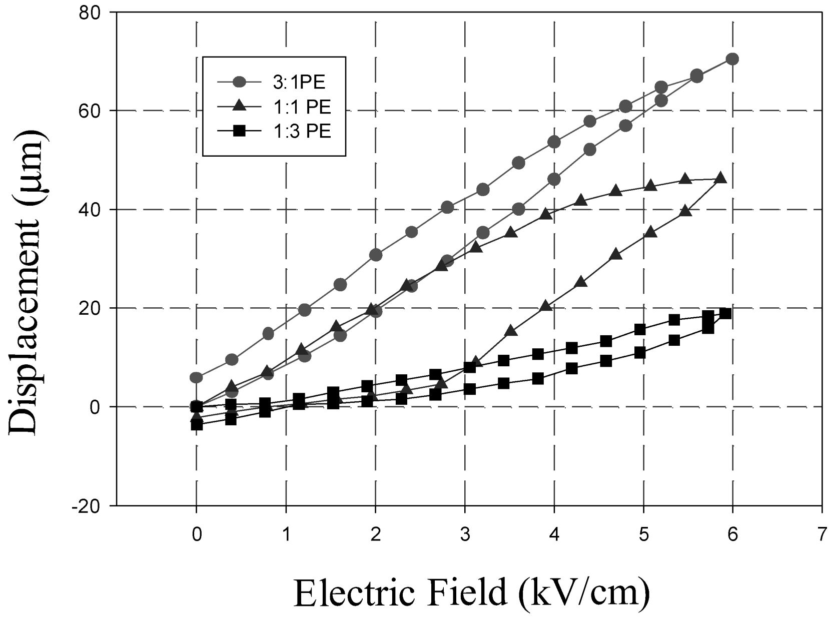

Fig. 9. Displacement vs. Electric Field for 1:1, 1:3, and 3:1 PE monomorph (total thickness is 1.26 mm).

separating the piezoelectric and the electrostrictor [13, 14]. In Fig. 7, it is also seen that the presence of the electrostrictive phase reduces the hysteretic behavior substantially, while reducing the field-induced polarization (P_{max}) as well.

The 1:3 monomorph possesses a $P_r = 5$ μC/cm^2, a $P_{max} = 20$ μC/cm^2, and a $E_c = 1.0$ kV/cm. Since the 1:3 monomorph is 75% by volume electrostrictive PMN-PT 90/10, the P-E hysteresis loop has an electrostrictive characteristic more similar to the electrostrictive composition. On the other hand, the 3:1 monomorph is 75 % by volume of piezoelectric 0.65PMN-0.35PT and exhibits a ferroelectric characteristic, with $P_r = 23\ \mu$C/cm^2 and $P_{\max} = 30\ \mu$C/cm^2, as depicted in Fig. 8.

Figure 9 depicts the dc-displacement of 1:1, 3:1, and 1:1 monomorph actuators, respectively. The maximum tip def1ection of the 1:1 monomorph was found to be 44 μm at 6 kV/cm bias field strength. The maximum tip displacement of the 3:1 monomorph actuator was 76 μm at 6 kV/cm, which corresponds to a 50% increase with respect to the 1:1 monomorph. It is interesting to note that the hysteresis of the 3:1 monomorph is also much less in comparison to the 1:1 despite its higher piezoelectric volume fraction, indicating that it would provide for a better positioning accuracy [8]. The tip displacement of the 1:3 monomorph is the smallest of the three, with a maximum at 20 μm at 6 kV/cm. The aforementioned comparison indicates that the tip displacement in such monomorph actuators can substantially be increases by clamping the transverse displacement of the piezoelectric with an electrostrictive layer that is lower in volume fraction. The electrostrictive layer also imparts lower hysteresis to the actuators, which is desirable in regard to positioning precision. In principle, one should be able to further increase the tip displacement, by decreasing the thickness of the electrostrictive layer and still maintaining lower hysteresis than that of the purely piezoelectric counterpart.

4. Conclusion

Multimaterial piezoelectric and electrostrictive PMN-PT monomorph actuators were co-sintered, and their dielectric and electromechanical properties were characterized. The experimental results indicated that maximum displacement along with minimal hysteresis could be obtained in monomorphs with 75% piezoelectric by volume (3:1 monomorph). The highest tip displacement for this composite was found to be 76 μm at 6 kV/cm. We have shown that the choice of materials and tuning of their thicknesses can be used to tailor the displacement in conjunction with the hysteresis in a bending actuator.

Acknowledgments

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