PMN-PT PIEZOELECTRIC-ELECTROSTRICTIVE BI-LAYER COMPOSITE

ACTUATORS

by

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ABSTRACT OF THE DISSERTATION PMN-PT PIEZOELECTRIC-ELECTROSTRICTIVE BI-LAYER COMPOSITE ACTUATOR

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In the past few decades, significant advances have been achieved to replace the conventional actuators, including hydraulic, shape memory alloy, electromagnetic and linear induction, with piezoelectric actuators since they are light weight and small in size, have precision positioning capabilities, offer a wide range of generative force, consume less power, and provide higher durability and reliability. The strain produced by bulk polycrystalline piezoelectric ceramics and single crystals are typically in the range of 0.1 to 1 %, respectively, which is still low for many applications. Therefore, various strain amplification designs including multilayer, bimorph, unimorph, flextensional actuators (Moonie and cymbal), co-fired and functionally graded ceramics have been proposed to enhance the displacement.

In this investigation, Piezoelectric/Electrostrictive Bi-Layer Monolithic Composites (PE-MBLC) were fabricated by co-pressing and co-sintering of the piezoelectric (PMN-PT 65/35: P) and electrostrictive (PMN/PT 90/10: E) powders. Flat and dome shaped of PE-MBLCs were obtained by optimizing processing conditions such as pressing pressure and sintering temperature. In addition, poling conditions of bilayer

composite actuators were thoroughly studied to maximize their electromechanical properties. It was found that composites had lower d_{33}^{eff} and K^{eff} values than the calculated values. This was attributed to a significant difference between relative permittivities of P and E materials as well as the presence of induced stresses in both P and E layers after sintering that hindered domain switching within piezoelectric layer during poling.

The shape change (planar to dome), electromechanical properties, and actuation performance of PE-MBLC actuators were examined as a function of volume percent of piezoelectric phase. The highest displacement ~15 μ m was obtained from PE-MBLC actuator with 50 volume % piezoelectric phase due to the transverse strain response of piezoelectric and electrostrictive layers, the geometry of the composites, and the domain reorientation.

Two designs of the flextensional actuators were also fabricated. In the first design, two truncated thin metal caps were attached to a flat PE-MBLC. In the second design, dome-shaped PE-MBLC actuators with various volume fraction of piezoelectric phase were attached to a flat metallic plate. The actuation evaluation showed that ~ 21 to 70 μ m displacement could be achieved by such designs.

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actuators with piezoelectric (PMN-PT 65/35), electrostrictive (PMN-PT 90/10), and PE-MBLC 5:5 driving elements.

Chapter 1 Introduction

Different types of actuators including hydraulic, shape memory alloy, electromagnetic, linear induction and piezoelectric actuators have been used in various applications such as optics, precision machinery and motors, etc [1]. This chapter will only review the development of piezoelectric materials and designs for actuator applications only.

Piezoelectricity was discovered by J. and P. Curie in 1880. A piezoelectric material can generate electrical charges under stress, or changes its dimension when is subjected to an electric field. These phenomena are referred to as direct and converse piezoelectric effects, and are utilized for sensor and actuator application respectively [2]. The induced piezoelectric strain is given by $S = d_{ij}E_{j}$, where S, d, and E are the displacement, piezoelectric charge coefficient, and electric field, respectively. There are three piezoelectric charge coefficients in a 4mm point group (tetragonal), namely the transverse-d₃₁, longitudinal-d₃₃, and the shear-d₁₅ coefficients. The use of these coefficients is governed via the actuator geometry and the direction of polarization with respect to the electric field applied. For instance, displacement in a stacked ceramic or multilayer thin ceramic is related to the d₃₃-coefficient and tip bending in a bimorph exploits the d₃₁-coefficient. Strain amplification in flextensional actuators, including Moonie and Cymbal, is achieved through the combination of lateral contraction and longitudinal expansion of ceramic disks and transforming them into longitudinal displacement of metallic endcaps [3].

Lead zirconate titanate ceramics (PZT) have been widely used in many applications such as transducers, generators, sensors and actuators due to their high electromechanical properties. The drawback of such piezoelectric ceramics is their low level of strain generated, which is on the order of 0.1%. Electrostrictive compositions of PMN, and its solid solutions (PMN-PT) have also been utilized as actuator materials. They generate strains up to 0.15%, which is slightly higher than that of bulk PZT's. Single crystal relaxor ferroelectrics (PMN/PT and PZN-PT) exhibit superior electromechanical properties and strains up to 1 % compared to PZT [4]. However, high cost is a major factor for single crystal. The strain levels produced in single crystals or polycrystalline ceramics are still modest and thus do not meet the requirements (more than 1000 microns displacement) in some applications such as linear mirrors, cavity pumps, loudspeakers, variable focus mirrors and laser deflection transducers [5;6].

In the past two decades, various actuator designs have been developed to improve actuation parameters such as high displacement, blocking force, fast response time, small size, and low power consumption. A multilayer actuator is comprised of a stack of several electroded ceramic discs. The displacement such actuator depends on the number of ceramic layers. Furthermore, the multilayer actuators show relatively high generative force as well as low driving voltage. Another more commonly employed design is the bender actuator (unimorph or bimorph). A bimorph consists of two layers of poled piezoelectric ceramics boned together with a conductive layer. The opposing strains (from d_{31} -coefficient) induced during the application of an electric field create a bending moment and cause the bender to deflect. Bimorphs are classified into series and parallel configurations based on the relation between the direction of polarization in each ceramic

plate and the direction of E-field applied. In series bimorph, poling directions of piezoelectric plates are anti-parallel to the electric field whereas in parallel bimorph both plates are poled in the same direction, and a bias voltage is applied to a parallel bimorph. High deflection is obtained based upon the long length and small thickness of bimorph plate. However, the drawback of this desired design is its low generative force. A unimorph is constructed of one active layer (piezoelectric layer) and one inactive layer (metal) bonded together with a thin layer of epoxy [7]. This design has been applied in Thunder [8-10], Cerambow [11] and Crescent [12] actuators. To bond the two layers, different types of bonding agents, including glue or epoxy adhesive or adhesive film, have been used in a high temperature and pressure process. Due to the curved shape and induced stress developed by thermal expansion mismatch of active and inactive layers, high displacement and moderate load-bearing capability can be obtained [13]. A stressbiased wafer called Rainbow has been developed [14-17] by reducing one side of a ceramic wafer with carbon (graphite) at 975 °C. In the resultant monolithic wafer, the reduced layer (cermet) is intimately bonded to the unreduced layer (piezoelectric layer). The difference in thermal expansion coefficients between the two layers during cooling step after heat treatment generates different level of compressive and tensile stresses giving rise to a dome shape wafer called Rainbow. Such tensile stresses on the surface of the unreduced layer contributed to high displacement due to enhanced domain reorientation when an electric field is applied. The unique dome-like structure allows Rainbow to sustain large loads. Flextensional actuators like Moonie [18;19] and Cymbal [20-24] have been invented to compensate the drawbacks of low displacement in

multilayer actuators and low generative force in bimorphs. In these flextensional actuators, a piezoelectric disc is sandwiched between two metallic caps.

The major concern in bimorph and unimorph actuators is the ceramic/metal interface. As mentioned above, bonding agents (epoxy resin/conductive epoxy) are utilized to attach ceramics and metallic layers in these two actuators. The failure of bonding layer between ceramic/metal is the main cause of crack and/or delamination after a long period of operation or use in severe environments such as high pressure, high temperature and high mechanical loading. Such failure of bonding layer degrades the actuation, lifetime and reliability. To overcome the bonding failure during long usage, a new bender design called QuickPack actuators has been introduced by ACX Inc [25]. This actuator is comprised of two piezoelectric ceramic wafers, encapsulated in a protective polyimide insulation material which prevents the ceramics from the high humidity and harsh contaminants. Thus, the actuators perform high functionality and durability. The interdigitated electrode design is another factor to improve the failure resistance. Another type of smart actuator design is active fiber composite (AFCs) [26;27]. In this structure, piezoelectric fibers are embedded between two polyimide sheets with interdigitated electrode patterns. An AFC actuator has high flexibility, robustness, as well as high in-plane actuation (d_{33}) along the fiber length.

From the mid 1990's until now, there have been many attempts to develop new types of actuators and fabrication methods to solve the bonding problems associated with unimorph and bimorph actuators. Functional Gradient Material (FGM) [28-33] and co-fired ceramic [34-37] concepts have also been introduced to improve the ceramic/metal interface problem. In FGM technique, compositionally different ceramic layers are

stacked and then sintered together to form a monolithic structure. After the poling process, the chemical composition gradient forms a gradient of d₃₁ piezoelectric coefficient in the total thickness of the actuator. The gradients such as piezoelectric coefficient, conductivity, resistivity, and permittivity between layers can generate d₃₁ gradient after poling. Like the FGM actuator, co-fired actuators have a quasi-continuous interface after sintering. A variety of materials have been studied as co-fired actuators, including piezoelectric (PMN/PT-65/35) and electrostrictive (PMN/PT-90/10) [38], co-extruded PZN-PZT and PZN-PZT/Ag [35;37] and pre-stressed thin PZT disc with dielectric low temperature co-fire ceramic (LTCC) [34].

The aim of this study is to develop piezoelectric-electrostrictive monolithic bilayer ceramic composites and flextensional actuators; to quantify their electromechanical and actuation properties; to investigate their mechanism of displacement amplification and displacement loop behaviors. In this study, piezoelectric (PMN/PT-65/35) and electrostrictive (PMN/PT-90/10) materials will used due to their chemical compatibility, high piezoelectric charge coefficients (d_{31} and d_{33}), and small thermal expansion coefficient (TEC) mismatch. The latter allows co-sintering these two materials without de-lamination. Through optimization of processing conditions, control of geometry (flat or dome structure) after sintering might be possible, which it is useful to exploit in actuator fabrication. Furthermore, two different designs of the flextensional actuators will be constructed by using the monolithic bi-layer composite. In the first design, a dome monolithic bi-layer composite will be bonded to a thin metallic plate. In another design, two truncated thin brass caps will be attached to a flat monolithic bi-layer composite. The electromechanical properties and actuation performances of such actuators will be intensively investigated.

In this thesis, chapter 2 includes the background of piezoelectric and electrostrictive effects, materials used for piezoelectric actuators, actuator designs and the actuator applications. Chapter 3 states the objectives and methods of attack to achieve this work. Chapter 4 discusses the processing, sample preparations, and evaluation of electromechanical properties of single ceramics (piezoelectric and electrostrictive ceramics). Chapter 5 explores the optimum poling conditions on the electromechanical properties of bi-layer P/E composites (stacked, laminated, co-fired bi-layer composites). Chapter 6 addresses the physical and electromechanical properties of the co-fired bi-layer composites as a function of piezoelectric phase. Chapter 7 explains the state of induced stress developed in the co-fired bi-layer composites. Chapter 8 and 9 discuss the fabrication of flextensional actuator designs fabricated from the co-fired bi-layer composites (electrically active elements) and the metal (inactive layer). Chapter 10 and 11 are devoted to the conclusions and future work.

Chapter 2

Literature survey

2.1 Piezoelectricity

Crystal structure of a solid material is made up of the ions that are constrained to occupy positions in the lattice. The smallest repeating unit of the lattice is called unit cell. The symmetry of the operation in a unit cell such as center of symmetry, axes of rotation, mirror planes, and combinations of these results in 32 point groups [39]. Among these, eleven point groups are centrosymmetric and do not possess polarization. Excluding 432 point group, the remaining twenty point groups are non-centrosymmetric giving rise to piezoelectric phenomenon. Ten point groups out of twenty are pyroelectric, and have spontaneous polarization. Among pyroelectric crystals, only those in which the spontaneous polarization can be reversed by a DC electric field are called ferroelectric [40].

Being discovered by Pierre and Jacques in 1880, piezoelectricity is a phenomenon where a material can generate an electric charge under the stress applied [2]. Quartz (SiO₂) is one of the most important natural piezoelectric single crystals. However, its piezoelectric properties are much lower than those of ferroelectric ceramics such as barium titanate (BT), lead titanate (PT), lead zirconate titanate (PZT) and lead magnesium niobate- lead titanate (PMN-PT).

Lead zirconate titanate (PZT) is one of the most important piezoelectric materials used as transducers for actuator and sensor applications. This is due to its large charge piezoelectric coefficients and coupling factors, relatively high Curie temperature, ease of tailoring its properties by composition modification and cost effectiveness. PZT is composed of a solid solution of PbZrO₃ and PbTiO₃ in which pure PbTiO₃ and PbZrO₃ have tetragonal and orthorhombic structures, respectively. Excellent properties of PZTs are originated from the co-existence of the tetragonal and rhombohedral phases at the morphotrophic phase boundary (MPB) of x = 0.52 : Pb(Zr_xTi_{1-x})O₃.

In general, the presence of the piezoelectric effects in a piezoelectric material arises from the orientation of dipoles by poling process. The direct piezoelectric effect is defined as the development of charges under a mechanical stress applied (Fig. 2.1 a). In converse effect, the application of an electric field induces changes in the dimension of a piezoelectric material (Fig. 2.1 b). The direct and converse effects are described by [2]:

$$\mathbf{D} = \mathbf{d}\mathbf{E} + \boldsymbol{\varepsilon}^{\mathrm{T}}\mathbf{E} \tag{1}$$

$$\mathbf{S} = \mathbf{s}^{\mathrm{E}} \mathbf{T} + \mathbf{d} \mathbf{E} \tag{2}$$

where D, T, E, S, d, s, ϵ are the dielectric displacement, stress, electric field, strain, piezoelectric charge coefficient, compliance, and dielectric constant, respectively. The superscripts indicate a quantity is held constant: for example ϵ^{T} means permittivity at constant stress and, in the case of s^E which represents elasticity at electric field constant.

Equation 3 and 4 determine the direct and converse piezoelectric effect in the absence of electric field and external stress, respectively. The piezoelectric coefficients are directional quantities and are normally specified by subscripts.

For instance, for the d_{33} , polarization is induced in the direction 3 while stress is applied in the direction 3.

$$D_3 = d_{33}T_3$$
 (3)

$$S_3 = d_{33}E_3$$
 (4)



Figure 2.1. Schematic representation a) direct piezoelectric effect b) converse piezoelectric effect.

2.2 Electrostriction

Electrostriction is another electromechanical phenomenon that exists in ferroelectric ceramics. The electrostriction is somewhat analogous to the converse piezoelectric effect in term of the deformation of the material induced by the application of an electric field. In the converse piezoelectric effect, the deformation is linear with respect to the applied field and change to contraction/expansion when the field is reversed [40]. In contrast to the piezoelectric, the deformation of the electrostrictor is independent upon the polarity of the field and is the square of the electric field. This means that an electrostrictor will expand under applied field regardless of its polarity. The electrostriction coefficients are defined by [41]:

$$S_{ij} = M_{ijkl} E_k E_l \tag{5}$$

$$S_{ij} = Q_{iikl} P_k P_l \tag{6}$$

where S_{ij} are the strain and M_{ijkl} and Q_{ijkl} are the electrostrictive coefficients related to the electric field (E) and polarization (P). Eliminating symmetrical components, it is obtained that M_{ijkl} or $Q_{ijkl} = M_{mn}$ or Q_{mn} (m, n = 1, 2, 3). The strains corresponded to direction of the applied E-field can be expressed as [41]:

$$S_{33} = M_{11}E_3^2$$
 or $S_{33} = Q_{11}P_3^2$ (7)

$$s_{11} = M_{12} E_3^2$$
 or $s_{11} = Q_{12} P_3^2$ (8)

where S_{33} and S_{11} are longitudinal and transverse strain, respectively. M_{11} (Q_{11}) and M_{12} (Q_{12}) are the longitudinal and transverse electrostrictive coefficients, respectively.

Electrostriction exists in all dielectric materials, which they are crystalline or amorphous, and centrosymmetric or polar. In most materials, the electrostrictive strain is extremely small. However, the electrostrictive generated in some relaxor ferroelectrics are comparable or higher than piezoelectric ceramics [42]. Those relaxor ferroelectric materials are lead magnesium niobate (PMN), lead zinc niobate (PZN) and lead lanthanum lead zirconate titanate (PLZT 9.5/65/35) [41]. These materials have the distinctive properties such as a broad transition peak between ferroelectric and paraelectric phases, strong frequency dispersion of relative permittivity and a shift of their maximum relative permittivity with frequency. The advantages of the electrostrictive materials over the piezoelectric ceramics are that hysteresis in the strainfield dependence is minimal, no ferroelectric domains in these materials hence, no poling is required. Therefore, these materials do not suffer from aging effect. Nevertheless, these materials have a limitation of the usable temperature range due to the strong temperature dependence of the electrostrictive effect.

The distinct feature of the electrostrictor is that the dimension change during the applied electric field can go back to the original size when the electric field is removed. As a consequence, the electrostrictive materials (relaxor ferroelectrics) have been widely used in the actuator applications such as micropositioners which requires very precise and reproducible motion control [3].

2.3 Materials for actuator

Magnetostrictive and shape memory alloys whose displacement are controlled by application of a magnetic field and temperature, respectively, have been used in sensor and actuator applications. However, these actuators are generally inferior to piezoelectric/electrostrictive ceramic actuators. This is because the magnetostrictive actuators are very bulky and generate magnetic noise while shape memory alloys have slow response time with very low energy efficiency. Nowadays due to miniaturization of electronic components and the need for lower driving power consumption, ceramic actuators are preferred in most of the electronic devices due to their relatively compact size [5]. In addition, piezoelectric actuators have become more popular in smart actuator systems due to their broad operating bandwidths, generating large forces. However, their drawback is small displacements. Presently, various amplified designs of the actuators have been proposed to improve the actuation performance.

2.3.1 Piezoelectric materials

The piezoelectric ceramics are classified into two groups: normal ferroelectrics and relaxors. Piezoelectric ceramics with prominent electromechanical properties possess perovskite structure with the general formula of ABO₃ shown in Fig 2.2. The A site is Na, K, Rb, Ca, Sr, Ba or Pb and the B site is occupied by Ti, Sn, Zr, Nb, Ta, or W. Several examples of piezoelectric ceramics with this structure are barium titanate (BaTiO₃:BT), lead titanate (PbTiO₃:PT) and lead zirconate titanate (PbZr_xTi_{1-x}O₃: PZT). These materials are perovskite structures and have a sharp dielectric constant peak between the ferroelectric/paraelectric transition phases at Curie temperature (T_c) as shown in Fig 2.3. Among them, PZT has been widely used as a piezoelectric transducer because of its superior piezoelectric properties at MPB phases. Figure 2.4 shows the phase diagram and tetragonal/rhombohedral morphotrophic phase boundary of PZT [43]. Modification of its properties by addition of acceptor and donor ions results in hard and soft PZTs, respectively.

- Hard PZT ceramics are doped by acceptor ions e.g. K^+ , Na^+ in the A-site, or Fe³⁺, Al³⁺ in the B-site creating oxygen vacancies. This gives rise to the increase of mechanical quality factor (Q_m) and coercive field (E_c) at the expense of reducing piezoelectric properties. Moreover, Hard PZTs such as PZT-8 shows relatively narrow displacement hysteresis loop and small strain [4].

- Soft piezoelectric ceramics are formed by the substitution of donor ions such as Nb^{5+} , Ta^{5+} , W^{6+} into the B-site or La^{3+} into the A-site. This results in the creation of Pb vacancies, reduction of Curie temperature and increase of piezoelectric properties at room temperature. Although the field induced strain (0.1%) of soft PZT is much higher than that of hard PZT, it represents higher dielectric loss and a wider displacement hysteresis loop due to domain motion [4].



Figure 2.2. The perovskite structure (ABO₃).



Figure 2.3. Phase transformation in barium titanate (typical ferroelectric perovskite materials)



Figure 2.4. Phase diagram of PZT as a function mole% PbTiO₃

2.3.2 Relaxor ferroelectrics

Lead based relaxor materials such as lead magnesium niobate (PMN) or solid solution of PMN-PT and solid solution of PZN-PT, have been utilized in transducer and actuator applications due to their high piezoelectric coefficients and relative permittivity. Two different forms of these materials, polycrystalline and single crystal relaxor ferroelectric have been used [44;45].

Due to their complex perovskite structure, $A(B_1B_2)O_3 : (B_1 = Mg^{2+}, Zn^{2+}, Ni^2)$ and $B_2 = Nb^{5+}$, Ta^{5+} , W^6), Relaxors exhibit a broad transition peak in the dielectric constant. The distinguishable characteristic of relaxor ferroelectrics compared to normal ferroelectrics is the frequency dependence of their maximum dielectric constant. The increase of frequency reduces their permittivity and shifts their T_m towards higher temperatures. This phenomenon is called dielectric relaxation behavior (Fig. 2.5) [3].

Lead magnesium niobate (PMN) is a well-know relaxor ferroelectric which exhibits strong electrostrictive behavior with the maximum dielectric constant at T_m equal to -10 °C and field induced strain > 0.1% [46]. Solid solutions of PMN with normal ferroelectric materials such as PbTiO₃ (PT) have been studied by many researchers [46-49]. PMN-PT shows both piezoelectric and electrostrictive properties based on the compositional ratio of PMN:PT. This makes them attractive for many actuator applications.



Figure 2.5. The temperature and frequency dependence of the a) permittivity and b) dissipation in PMN-PT electrostrictive ceramic [50].

The piezoelectric 0.65PMN-0.35PT is a composition on the morphotropic phase boundary (MPB) between the tetragonal and rhombohedral phases. It shows piezoelectric properties comparable to soft PZT (-5H) with d_{33} , k_t and T_c equal to 700 pC/N, 0.55 and 150 °C, respectively. The electrostrictor 0.9PMN-0.1PT can develop strain >0.1% under applied DC bias field with no hysteresis due to no spontaneous polarization at room temperature. The strain vs. electric field of piezoelectric and electrostrictive material is depicted in Fig. 2.6 a and b. The strain-E-field response of piezoelectric shows the butterfly loop due to the effect of domain reorientation while the positive strain is found in the electrostrictor due to its deformation dependent upon the square of the electric field.



Figure 2.6. Field-induced strain in a) piezoelectric and b) electrostrictive material [3].

2.4 Actuator designs

As mentioned earlier, piezoelectric/electrostrictive ceramics and/or single crystals are of interested for solid-state actuators employed in many applications such as nanoand micro-positioner, pumps, acoustic noise canceling devices and vibration suppression. Therefore, a wide range of displacement and generative force are required. Bulk piezoelectric actuators possess a low level of strain, which is on the order of 0.1%. In the past two decades, various actuator designs have been developed to amplify strain and improve actuation performances, including displacement, blocking force, durability and reliability. These designs will be discussed in details in the following sections.

2.4.1 Multilayer actuator

A multilayer actuator is comprised of a stack of several electroded ceramic discs/layers (20-40 μ m thick), shown in Fig. 2.7. The improvement of displacement depends on the number of ceramic layers ($\Delta L_{Multilayer} = N\Delta L_{single layer}$). Thus, high displacements in multilayer actuators can be achieved even at low driving voltages (40-100 V) [51]. Another characteristic feature of the stacked ceramic actuator is its high blocking force. Nevertheless, the crack/delamination of the ceramic and the internal electrodes are the main problem of this actuator design which reduces its reliability and durability. To eliminate these issues, many attempts have been made to optimize ceramic compositions [52;53] and electrode materials [54;55] used for the multilayer actuator.
2.4.2 Bender Actuator

The bender actuators are the most commonly used and consist of two or more layers of piezoelectric ceramics glued together with thin layer of conductive epoxy. The piezoelectric ceramics have parallel or opposite polarization directions depending on their series or parallel connected as shown in Fig. 2.8 [56-58]. The opposing strains of two layers were developed by the transverse piezoelectric charge coefficient (d_{31}) during the application of an electric field creating an internal bending moment and causing the bender to flex.

Two types of bimorph configurations are called series and parallel bimorph. In the series bimorph, the electric field is applied on the two external top and bottom electrodes. Therefore, this electric field is equal to the source voltage divided by the total thickness (2t) between the electrodes. While in parallel bimorph, each ceramic plate is individually connected to the source voltage. Hence, the electric field across each ceramic layer is twice the field in each layer in a series bimorph. As a result, the bending displacement of a parallel bimorph is twice that of a series type (when all other parameters are held constant). The bending displacement equations for series and parallel bimorph are respectively defined as:

$$\delta L = 3d_{31}((L^2)/2t) E$$
 (2.3.1)

$$\delta L = 3d_{31}((L^2)/t) E$$
 (2.3.2)

where d_{31} , L, t and E are the transverse piezoelectric coefficient, the length of the bimorph, thickness of the bimorph and the electric field, respectively.



Figure 2.7. Multilayer actuator [59].



Figure 2.8. a) Parallel Bimorph b) Series Bimorph

The main problem of bimorph actuators is their short life time due to delamination at the interface between piezoelectric ceramic/bonding layers. An improvement of failure resistance has resulted in a new bender design called QuickPack actuators, manufactured by ACX Inc.[25]. The QuickPack actuator is composed of two piezoelectric ceramic elements encapsulated in a protective polyimide insulation material which can eliminate gluing problem and give durability. Another design of benders called Active Fiber Composites (AFCs) has also been developed [60;61]. AFC actuators consist of unidirectional piezoelectric fibers composites which are poled along fiber direction, embedded between two polyimide sheets with interdigitated electrode design. This actuator design is more flexible and offer higher in-plane actuation due to utilization of d_{33} coefficient.

2.4.3 Unimorph actuator

A unimorph consists of a non-piezoelectric (thin metal sheet) and an electroded piezoelectric plate bonded together with epoxy. Two types of the epoxy, high temperature cure [10;12;62] and room temperature cure [7], have been used. The type of epoxy affects the geometry and the internal stress in the unimorph after curing process. The mechanism of strain in a unimorph arises from constraining the piezoelectric layer by the metal sheet during the application of an electric field which causes displacement in the axial direction at the center of piezoelectric ceramic.

Thunder developed at NASA Langley Research Center is the most useful device and manufactured by FACE international Corporation [10]. This device is comprised of a ceramic wafer attached by polyimide adhesive (LaRC-SITM) as a bonding agent to a metallic backing layer such as aluminum, stainless steel, brass, copper, etc., as shown in Fig. 2.9 a. Thunder is cured in an autoclave at high temperature (up to 320 °C and 30 psi pressure). Upon cooling down to room temperature, a curved structure is obtained due to the thermal expansion coefficient (TEC) differences of the ceramic and metal plate. Many parameters have been studied including the geometry of Thunder [10], number and type of the metals [10], thickness ratio of ceramic to metal layer [10;63] in order to improve the actuation performance of Thunder,

Another type of the unimorph is LIghtweight Piezo-composite Curved Actuator (LIPCA) [62;64]. Similar to Thunder, LIPCA exploits the TEC mismatch between the ceramic and fiber-reinforced composite layers to obtain the curved shape and to induced residual stress in the actuator after curing process. The structure of LIPCA consists of a ceramic wafer sandwiched between a top composite layer with low TEC and the base layers with high TEC as shown in Fig 2.9 b. LIPCA has been designed to replace the heavy metal layers in Thunder by the use of a lightweight fiber reinforced plastic layer including Kevlar/epoxy, carbon/epoxy and glass epoxy.

Cheng [65] has introduced novel fabrication of a curve-shaped unimorph with internal stress by electroplating process. In this process, a PZT wafer is bonded on one side to the stainless steel substrate. A curved sample is obtained after curing process. Another side of stainless steel is electroplated by nickel. This causes additional curvature as well as a higher level of internal stress inside the sample. This can enhance the loadbearing capability and improve the displacement in the unimorph.



Figure 2.9. Unimorph actuator a) Thunder [66] b) LIPCA [67].

2.4.4 Rainbow actuator

The monolithic stress-biased dome-shaped actuator named RAINBOW (**R**educed And Internally Biased Oxide Wafer) has been invented by G.H. Heartling in 1994 [68]. As shown in Fig. 2.10, one side of the sintered PZT ceramic wafer which can be piezoelectric, electrostrictive [68;69] and antiferroelectric [70] is placed on the carbon graphite block and heated to high temperature. Then, the oxidation process of the carbon leads to the loss of the oxygen in the piezoceramic side in contact with the graphite block. This results in generation of a conductive layer enriched with metallic Pb and cermet phases (ZrTiO₄, ZrO₂, La₂O₃, etc.) in the contact side of the ceramic with graphite block [71]. The causes of the dome-shaped in RAINBOW are believed to be due to the reduction in volume of reduced layer due to the oxygen loss compared to the unreduced layer, the differential thermal contraction between the reduced and unreduced layer during cooling process, and the change in volume due to the phase transformation as the material is cool down through the Curie point.

It is believed that the presence of non-uniform internal stress through the planar and thickness direction as well as the non-uniform distribution of transverse piezoelectric effect (d_{31}) contribute to the enhancement of domain switching and thus the improvement of displacement in Rainbow [15;72]. Extensive studies have investigated the effects of parameters such as resonance characteristic [73] and young's modulus of the reduced layer [14] on the actuation performance of the RAINBOW.



Figure 2.10. Reduced and Internally Biased Oxide Wafer (RAINBOW) [74].

2.4.5 Flextensional actuator

A flextensional actuator is comprised of a piezoelectric ceramic and a shell structure. There are five types of flextensional transducers that operate in the low frequency less than 10 kHz [75]. In the mid 1990s, the ceramic-metal composite, named Moonie, was developed by R.E. Newnham's research group at Penn State University. Moonie is a class V of flextensional transducer consisting of a ceramic disk sandwiched between two moon-shaped metals with shallow cavities shown in Fig 2.11 a. Originally designed for the hydrophone application [76], Moonie has been used for actuator applications. This is because the shallow cavities of the metal endcaps in moonie convert and amplify the radial displacement of the ceramic into a large axial motion of the endcaps. The displacement amplification of Moonie arises from ceramic itself and the flexural motion of the metal derived from the radial motion or transverse piezoelectric coefficient (d_{31}) of the ceramic. Several parameters have been tailored to improve the electromechanical properties of Moonie including: ceramic material [18;19;77], types of bonding agents [18], dimensions of the endcaps such as cavity depth, cavity diameter, thickness and type of the metal [18;19;77], and design of single or stacked structures [18;19;77].

The best actuation displacement in a single Moonie is ~22 μ m and the maximum generative force is 350 gf using 12.5 mm diameter and 1 mm thick of a PZT disk, 0.3 mm thick brass endcaps, 0.9 mm cavity depth and 9 mm cavity diameter [78]. To reduce driving voltage and improve displacement, a multilayer multistacked Moonie shows ~105 μ m displacement by stacking 5 multilayer Moonie together [19]. However, Moonie shows the position dependence of displacement where the highest displacement is

obtained at the center of the endcap and dramatically decreases towards the edge. Finite element analysis has shown a high level of stress concentration in the metal endcaps in the area above the bonding layer between ceramic-metal interface [76]. This lowers the effective force transfer from the ceramic to the endcaps resulting in a low displacement. To eliminate the high stress concentration and enhance the displacement, a ring-shaped groove on the exterior surface of the endcaps has been introduced. It has been found that displacement can be improved by moving the groove toward to the edge of the actuator as well as using deeper and wider grooves [20]. Nonetheless, the stress concentrations at the groove may still appear and causes the failure under long period of operation. In addition, forming grooves into the endcaps is a complicated process. Therefore, the new endcap design with truncated conical shape has proposed. This new type of flextensional actuator is called Cymbal.

The basic structure of a cymbal is the same as that of a moonie except the ceramic disk is sandwiched between two truncated conical metal endcaps as shown in Fig. 2.11 b. Unlike moonie, the transversal motion of ceramic disk is converted into a combination of flexural and rotational motion in the metal endcaps. In comparison with Moonie, this results in enhancement of displacement, increase of generative force, and reduction of position-dependency of displacement [20].



Figure 2.11. Flextensional actuator a) Moonie b) Cymbal

Finite element analysis (FEA) has been conducted to predict the effect of various parameters on performances of cymbal actuators [23;79;80]. These include the dimensions of the endcaps and cavity [81], types of the epoxy resins [23;82;83], types of metal endcaps [23;24], types of piezoelectric ceramics [20;84] and the cymbal designs by attaching the stud on the endcaps [85] as well as bonding asymmetrical endcaps on a ceramic [86]. It has been shown that the material selection of cymbal including ceramic driving elements, metal endcaps, and the epoxy resin plays a crucial role in actuation performance. Maximum displacement and small strain-electric field (S-E) hysteretic loop (low mechanical loss) can be achieved using single crystals as driving piezoelectric elements due to their high transverse piezoelectric coefficients (d_{31}) [21]. However, single crystals are expensive and thus soft PZT ceramic is the most commonly used as driving element. To obtain a moderate displacement, metals with low modulus of elasticity such as brass or titanium [22;24] with 0.25-0.3 mm thickness, 9 mm cavity diameter, 0.2-0.3 mm cavity depth are required. On the other hand, metallic caps with high modulus of elasticity provide high generative force with small displacement [84].

In the last decade, cymbal has received considerable attention for actuator applications due to its high displacement and moderate generative force (≈ 30 N) to fill the gap between bimorphs and multilayers [24].

2.4.6 Co-fired ceramic and Functionally gradient material

Novel actuator designs presented above are in the form of composites whose active piezoelectric ceramics are bonded to metallic pieces by traditional methods such as gluing. Therefore, the electromechanical response in such composites is interface-limited, often resulting in failure due to delamination under applied external load and operation under many cycles.

As a consequence, a single-plate bending actuator called monomorph has been developed by co-pressing and co-sintering of a semiconductive piezoelectric ceramic such as (K_{1/2}Bi_{1/2})ZrO₂ and PZT [87;88]. This method eliminates the use of bonding agent at the interface of two materials. The bending displacement occurs from the non-uniform distribution of electric field due to semiconductor-metal electrode contact. It has been found that displacement on the order of a few hundred microns can be obtained from a 20mm x 10mm x 0.4mm monomorph. Afterward, a variety of materials have been used to fabricate monomorphs or co-fired actuators, including piezoelectric 0.65(PMN)-0.35PT and electrostrictive 0.90(PMN)-0.10PT [89;90], co-extruded PZN-PZT and PZN-PZT doped with silver [35;37], thin PZT discs pre-stressed by co-sintering with low temperature co-fired ceramic (LTCC) dielectrics [34;91] and co-fired of PZT/Ag composite [92]. The co-fired ceramic is shown in Fig. 2.12 a.

Hall [90] has fabricated co-fired multimaterial monomorhps using piezoelectric PMN/PT (65/35) and electrostrictive PMN/PT (90/10) via the fused deposition technique. The multimaterial monomorph exhibits a good interface without cracks between two layers. In comparison with epoxy bonded piezoelectric/electrostrictive bimorphs, the tip displacement of multimaterial monomorph is smaller. However, multimaterial

monomorph shows narrow displacement hysteresis loop (reduced mechanical loss) due to non-hysteretic characteristics of its electrostrictive layer.

Yoon [35] has developed bi-layered PZN-PZT flextensional actuators by coextrusion. Due to thermal expansion coefficient mismatch between the active layer (low sintering temperature PZN-PZT ceramic) and passive layer (conductive layer PZN-PZT/Ag), a dome structure is created after sintering. High residual stress developed in a 50 mm x 20mm x 0.84 mm sample improves the axial displacement to 160 µm at 1.4 kV/mm

Juuti [34] has introduced new actuator design called pre-stressed piezoelectric unimorph (PRESTO) using PZT-5H and PZT 5A with 250 and 375 µm thicknesses as piezoelectric layers, respectively. A pre-stressing material such as AgPd paste and/or a dielectric low temperature co-fired ceramic (LTCC) tape is applied on one side of the sintered bulk piezoelectric. Due to the thermal expansion coefficient mismatch between bulk ceramic and pre-stressing material, a domed, saddle-like structure is created. The displacement-load outcome depends on the piezoelectric layer and type of electrode used. The highest displacement (118 µm) has been obtained using LTCC as a pre-stressing layer on PZT-5H. A AgPd pre-stressed PZT-5H monomorph achieves 63 µm displacement under 1 N load.

The concept of functionally gradient material (FGM) in piezoelectric actuator has been introduced to reduce the uneven distribution of stress between the ceramic/metal interface in bimorph and unimorph. FGM are fabricated by continuously varying the ceramic compositions from one surface to another within ceramic layers that are stacked and then sintered together to form a monolithic structure as shown in Fig. 2.12 b. There are several fabrication methods used to form the FGM such as powder pressing [28;93-96], tape casting [31;97-99], co-extrusion [100], fused deposition technique [89;90], centrifugal casting [101] and electrophoretic deposition [102;103]. The gradient of the compositions are derived by doping the base piezoelectric materials (PZT) for example, PZT-doped with zinc borate (ZB) [99], PZT doped with La³⁺ and Fe³⁺ [28], PZT mixed with Pt [95;104] or by using different piezoelectric compositions like PNN-PZT [96;105], PZT-BT [100]. After poling, the composition gradient transforms to a gradient of the electrical and piezoelectric properties including resistivity [28;97;99], piezoelectric coefficient [31;96;97], permittivity [96;101], and coercive field [36]. These electrical and/or piezoelectrical gradients give rise to the bending motion in FGM under the application of an electric field. In addition, the gradient of the microstructure (porosity) [106] and structural thickness gradient [107] have been reported.

Zhu [108] has developed FGM by dry pressing and co-firing of the PNN-PZ-PT system in which 0.6 PNN-0.4PZT has high dielectric permittivity and 0.5 PNN-0.5 PZT possesses high piezoelectric properties. A 20mm x 8mmx 0.7 mm bending actuator shows 20 μ m tip displacement with a very large hysteresis displacement loop attributed from 90° domain wall movements under an electric field.





Figure 2.12. a) Co-fired ceramic [37] and b) Functional graded piezoelectric ceramic.

Adikary [28] has investigated the resistivity gradient across poling direction of a piezoelectric actuator by introducing two different dopants (La^{3+} and Fe^{3+}) in PZT system. A bi-layered actuator has been developed by pressing and co-firing of 1.5% mole La^{3+} and 1% mole Fe^{3+} doped PZT. In comparison with Fe^{3+} , La^{3+} doped PZT showed more than two orders of magnitude higher resistivity, higher transverse piezoelectric coefficient (d_{31}) and permittivity. The differences of transverse piezoelectric coefficients and relative permittivities lead to the bending deflection (up to 140 micron at 1000 V/mm) of the bender under E-field excitation.

Vartuli [109] has developed the d₃₁-gradient flextensional actuator. Such actuators were fabricated by the multilayer ceramics (PZT) laminated with zinc oxide (ZnO) layers. The zinc oxide layers doped with 4-8 wt % Sb₂O₃ served as the conductor layer in the actuator. Flat or dome shape actuators with the PZT on the inside (concave) or the outside (convex) of the dome could be developed by varying the Sb₂O₃ content in the ZnO layer or changing the thickness ratio between the PZT and the ZnO layers. It was found that the highest displacement could be obtained in convex PZT dome actuators with 4 wt % Sb₂O₃ doped ZnO layer . He believed that the observed displacement was attributed to the domain switching in the PZT layer under E-field.

Li [31] have designed a new FGM actuator type utilizing d_{31} -gradient to improve axial displacement. This actuator uses the same principle as Rainbow, Thunder and Cymbal, in which the lateral-strain gradient is induced due to piezoelectric d_{31} gradient in the thickness direction and converted into the axial displacement. The bi-layered PZT/PZT actuators with different d_{31} coefficients are fabricated by tape casting and then co-firing. Due to the difference in compositions and piezoelectric properties of PZTs, the dome structure is formed. When an electric field is applied across the thickness parallel to the polarization of a sample which has dimension of 25 mm diameter and 1mm thickness, axial displacement ~16 μ m is generated at 1000 volts. Furthermore, it has been found that the highest displacement is obtained when the thickness ratio between low d₃₁ and high d₃₁ layers is at 1:1.

2.5 Applications of piezoelectric actuators

In the last three decades, a variety of smart piezoelectric actuator architectures have been proposed to improve the displacement and generative force throughout a broad frequency range. The superiority of piezoelectric actuators to conventional actuators such as electromagnetic actuators is due to their higher energy density, significantly smaller size, no electromagnetic noise, less heat due to lower frequency operation, and lower power consumption. As a result, the piezoelectric actuators have been widely used in many areas such as information technology, robotics, energy harvesting, and biomedical engineering [110] which will be addressed in this section.

2.5.1 Information technology

The miniaturized electronic devices are at high demand for customers in this era. Multifunctional cellular phones are now used for communication in conjunction with a photo camera. Therefore, the miniaturization of the automatic focus with the actuation of 0.2 mm can be produced using a bimorph. In addition, the zoom systems are developed by the use of two micro rotary motors which are comprised of a metal hollow cylinder and two PZT rectangular plates [111;112] as shown in Fig 2.13 a.

Piezoelectric actuators are used in the inkjet technologies as shown in Fig 2.13 b. Four different deformation modes to eject ink droplets including squeeze, bend, push and shear modes are developed to obtain high efficiency in printing as well as low cost manufacturing. Specific shape of piezoelectric actuator for example, a radially polarized ceramic tube, piezoelectric ceramic rods and multi-channel ceramic plate are used in a specific deformation mode [113]. A co-fired multilayer of PZT with ZrO₂ substrates developed by Epson has considerably improved the stability in vibration during printing operation and reduced the manufacturing cost.

In the field of optical fiber alignment, the optical couplers are produced from the co-firing between the base package (low temperature Co-fire Ceramic, LTCC) and a Λ -shape linear motor. Two optical fibers are inserted into the package where the Λ -shape linear motor is used to align the optical fibers into the position [110].

2.5.2 Robotics

The monolithic piezoelectric ceramics for structural actuation have been used in a wide variety of applications. However, the brittle nature of the ceramics makes them impractical in the handling, bonding, and integration into the device. Therefore, the development of the piezoelectric fiber composites composed of an active piezoelectric fibrous phase embedded in a polymeric matrix phase has been introduced. Presently, four different types of active piezoelectric fiber composites have been fabricated including 1-3 composites manufactured by Smart Material Corp [114], Active Fiber Composite (AFC) actuators developed by MIT [115], Macro Fiber Composite (MFC) developed by NASA Langley research center [116] and active composites with hollow cross-section fibers developed from the University of Michigan [117]. The 1-3 composite is used as a smart composite panel on an airplane fuselage to reduce the internal noise levels or to serve as pressure sensors to measure conditions outside the plane. AFC is also used for the structural control by suppressing the noise and vibration, and adjusting the twist of the blade in helicopter or by reducing the buffet load vibrations on the tails of F-18 [118] as shown in Fig. 2.14 a.



Figure 2.13. a) Metal tube motor b) Piezoelectric actuator for inkjet printer [110].

Monolithic multilayer piezo-stacked actuator and co-fired multilayer actuator exhibit displacement resolution on the order of 0.1 μ m or higher. With this resolution, these actuators are suitable for the applications in micro and nanopositioning devices such as x, y, and z stages, focusing microscope objective lenses, and steering mirrors shown in Fig. 2.14 b.



b)



Figure 2.14. a) Macro fiber composite in the tail fin [119] b) Precision stage. [110]

2.5.3 Energy

The field of power harvesting has experienced significant growth in the past five years due to the high demand for wireless electronics. The electrochemical battery used as a power supply in these electronics has disadvantages due to its limited lifespan and the need for periodic replacement. Thus, the self-powered devices have been developed through the concept of power harvesting. Many designs of piezoelectric actuator are incorporated to transform the external signals such as load, heat, vibration, into the electric charges as shown in Fig. 2.15. The most common piezoelectric actuator configurations used to capture the ambient energy are the fiber-based piezoelectric composites [120;121]. This is due to their ability to generate large amounts of strains. Higher strains provide more mechanical energy available for conversion into electrical energy. Other actuator designs are a unimorph cantilever beam [122], bimorph [123], thin monolithic multilayer [124] and piezoelectric cymbal [125]. High energy harvesting is obtained by using piezoelectric fiber composite, high power harvested under low excitation and loads by using unimorph and improvement of efficiency through load distribution as well as capability of withstanding high loads by using cymbal.



 $\boldsymbol{\varphi}:29mm, \boldsymbol{\varphi}_{c}:17mm, \boldsymbol{\varphi}_{d}:5mm, \, \boldsymbol{d}_{c}:1mm$

a)

Figure 2.15. a) Unimorph in the insole of a running shoe b) Cymbal for dynamic environment harvesting [125].

2.5.4 Bio/Medical applications

As shown in Fig. 2.16 a, high frequency vibrating piezo-actuator attached on the needle shows less deformation of the egg during artificial fertilization compared to the conventional method by hydraulic oil-pressure system. The other study is related to the noninvasive ultrasonic transdermal insulin delivery by using the high power ultrasonic cymbal piezo-actuators as demonstrated in Fig. 2.16 b [126].





Figure. 2.16. a) Piezo-robot for artificial fertilization [110] b) Cymbal for transdermal insulin drug delivery system. [110]

Chapter 3

Statement of Problem and Plan of Attack

A piezoelectric actuator is a device which converts electrical energy into mechanical displacement through the converse piezoelectric effect. In the past few decades, significant advances have been made with piezoelectric actuators to replace the conventional actuators such as hydraulic, shape memory alloy, electromagnetic and linear induction actuators. Piezoelectric actuators are also the most suitable devices in micropositioning, communications, and fluid control. This is because these applications require miniaturized actuators that are light weight/smaller in size and be able to offer a wide range of displacement (µm to cm), moderate generative force, low power consumption, high durability and reliability. The piezoelectric longitudinal charge coefficient values of bulk piezoelectric ceramics and single crystals are about 600-700 and 2500 pC/N, respectively. Therefore, piezoelectric ceramics and single crystals yield a strain level in the range of 0.1-1%. However, this strain level is often small for some applications in which a few hundred microns of displacement is required. To amplify displacement and generative force, various actuator designs such as unimorph, bimorph, RAINBOW, THUNDER, Moonie, and Cymbal have been proposed. In most of the design, the active piezoelectric element is attached to a metallic layer by a thin layer of conductive epoxy. Going through millions of cycles of electrical loading during operation, this bonding layer may crack at low temperatures and undergo creep or peel off at high temperatures. This degrades their performance, shortens their lifetime, and diminishes their reliability. To improve mechanical reliability and to remove the

ceramic/metal interface problem, the concept of co-fired ceramics and Functionally Graded Materials (FGM) has been introduced.

In this work, a promising method was utilized to eliminate the interface issue by co-firing ceramics of piezoelectric and electrostrictive PMN/PT compositions with different PT mole percentage. Piezoelectric PMN-PT composition exhibits large piezoelectric charge coefficients (d_{31} and d_{33}). Also, electrostrictive PMN-PT exhibits large piezoelectric coefficient under the applied electric filed. Higher piezoelectric charge coefficients positively affect the strain amplification. In addition, their thermal expansion coefficient (TEC) mismatch is reasonably small (~4 × 10⁻⁶/°C), which eliminates delamination issue during sintering process. Moreover, they show different strain characteristics upon the application of an electric field. This consequently gives rise to an asymmetric displacement behavior (under bi-polar field) based on the principle of symmetry superposition when two different materials are used in a co-fired actuator.

Therefore, the objectives of this work were:

- To fabricate the piezoelectric/electrostrictive monolithic bi-layer composite (PE-MBLC) by co-pressing and co-sintering process.
- 2. To optimize the poling conditions of the PE-MBLC.
- 3. To evaluate the electromechanical properties of PE-MBLC for actuator application.
- To design and fabricate flextensional actuators such as capped PE-MBLC and PE-MBLC unimorph using either flat or dome-shaped PE-MBLC.
- 5. To evaluate the electromechanical properties and actuation performances of the flextensional actuators.

To achieve the objectives, the following approaches were employed:

- First, processing conditions such as pressing pressure, sintering temperature, and poling conditions were optimized to obtain piezoelectric (P), electrostrictive (E) and PE-MBLC ceramics with maximum electromechanical properties and to fabricate flat and dome-shaped PE-MBLC.

- Second, the effect of the various volume ratios of P and E materials (from 0% to 100 % piezoelectric) on the physical and electromechanical properties was investigated.

- Third, the state of stress induced in the PE-MBLC after sintering was studied using X-ray diffraction on selected composites.

- Fourth, the actuation amplification was explored by developing flextensional actuators with different geometries of active elements and inactive element (metal). The first design was a Cymbal made of a flat PE-MBLC attached to two truncated conical caps. In the second design, a dome-shaped PE-MBLC was bonded to a flat metallic plate. The strain amplification and S-E loop of these flextensional actuators were extensively investigated.

The following presents the organization of this thesis.

Chapter IV will discuss the effect of ceramic consolidation and sintering temperatures on the physical properties of piezoelectric (P) and electrostrictive (E) ceramics. The optimization of poling conditions of piezoelectric ceramics will be also presented.

Chapter V will present the effect of poling parameters such as the electric field, temperature, and poling time on the electromechanical properties of PE-MBLC ceramics. The effect of P/E interface of PE-MBLC ceramics on the domain reorientation of piezoelectric layer during poling process is discussed.

Chapter VI will report the effect of various volume ratios of P/E materials of PE-MBLC on the dome height, electromechanical properties and displacement amplification.

Chapter VII will compare the state of induced stress in PE-MBLCs for different P/E volume ratios.

Chapter VIII will present a new flextensional design named PE-MBLC unimorph. The effect of the bonding area on the displacement will be discussed. Moreover, the effects of P/E volume ratios and the metal thicknesses on the improvement of actuation performance will be explained.

Chapter IX will discuss the actuation performance of Cymbal flextensional actuators fabricated by attaching flat PE-MBLC (P/E: 1/1) ceramics to two truncated conical brass endcaps.

Chapter 4

Processing study of single PMN/PT-65/35 (P) and PMN/PT-90/10 (E)

4.1 Introduction

In this chapter, the processing-property relationships of single piezoelectric and electrostrictive ceramics will be discussed. The effects of ceramic processing parameters such as consolidation pressure, sintering temperature, and poling conditions on the electromechanical properties were investigated. In addition, optimized processing conditions were used to develop co-fired piezoelectric/electrostrictive monolithic ceramics with various desired geometries.

4.2 Experimental procedure

4.2.1 Material Selection

The solid solution PMN-PT possesses both piezoelectric and electrostrictive effects depending on the compositional ratio of PMN/PT. In this study, commercially available piezoelectric composition 0.65PMN-0.35PT (PMN/PT-65/35) and electrostrictive composition 0.9PMN-0.1PT (PMN/PT-90/10) both from TRS technologies Inc. (State College, PA) were used. Table 4.1 represents the physical and electromechanical properties of these compositions [127]. Both compositions have high longitudinal (d₃₃) and transverse (d₃₁) piezoelectric charge coefficients which are desirable for actuation performance.

Property	0.65PMN-0.35PT (TRSHK1-HD)	0.9PMN-0.1PT (PMN15)
Density (g/cm ³)	7.95	7.9
Grain size (µm)	3-5	3-6
Coefficient of Thermal Expansion (10 ⁻⁶ /C)	10	≈ 6
E _c (kV/cm)	5	-
$P_s (\mu C/cm^2)$	35	26 *
$P_r (\mu C/cm^2)$	28	-
$T_{c}(^{o}C)$	150	15
d ₃₃ (pC/N)	750	700 **
d ₃₁ (pC/N)	-360	-230**
Dielectric constant @ 1kHz, 25 °C	6000	20000
Dielectric loss (%) @ 1kHz, 25 °C	1.7	<0.8
k ₃₃	0.74	0.6•
k ₃₁	0.39	0.22•
k _p	0.66	0.4•
k _t	0.5	0.5•

Table 4.1. The physical and electromechanical properties of piezoelectric (0.65PMN-0.35PT) and electrostrictive (0.9PMN-0.1PT) ceramics from TRS technologies Inc [127].

*25 °C, 35 kV/cm, ** 25 °C, 6.5 kV/cm, * 25 °C, >10 kV/cm,

4.2.2 Powder preparation

As received piezoelectric (P) and electrostrictive (E) ceramic powders were dried in an oven at 120 °C for 2 hours to eliminate their moisture. A PVA binder solution was prepared by dissolving 20 grams of polyvinyl alcohol (PVA) in 80 grams of water. Then PVA solution (6 wt %) was gradually added to the powders and the mixtures were gently mixed in mortars and pestles. These mixtures were then heated in an oven at 120 °C for 15 minutes, reground, and sieved through a 100 mesh stainless steel sieve. The sieving process was repeated for 5-10 times to obtain a homogeneous mixture.

4.2.3 Forming and heat treatment

Piezoelectric and electrostrictive powders were individually dry-pressed at 86, 114 and 229 MPa into pellets with 14.7 mm diameter and 1.15-1.3 mm thickness using a uni-axial hydraulic machine. Once the dimensions and masses of green pellets were recorded, the binder removal and pre-sintering were performed at 550 °C for 3 hours and 780 °C for 1 hour, respectively. Then the same parameters were recorded before and after the sintering step. The pellets were sintered in closed alumina crucible with 3 grams of PbZrO₃ source, at 1150, 1200 and 1250 °C for 1 hour. The heating and cooling rate were set at 3 °C/min during sintering.

4.2.4 Electrode and Poling

Sintered pellets were polished to 0.5 mm thickness and then their surfaces were cleaned with acetone. All samples were electroded with a high temperature brush paint silver paste Du Pont 7095 (W.K Robeson, Newark, DE) and heat treated at 550 °C for 15

minutes with 5 °C/min heating rate. Poling studies of piezoelectric ceramics were carried out by varying E-field, and dwell time at a constant temperature of 60 °C. The electric field and dwell time varied in the range 1-40 kV/cm and 5-45 minutes, respectively. The poling fixture (Neslab Instruments Inc., Newington, NH), silicone oil (Dow Corning 561 from EIS Inc., Philadelphia, PA) and the power supply from a Trek High Voltage Amplifier Controller Model 610C (Trek Inc., Medina, NY) were used in this experiment.

4.3 Characterization

4.3.1. Physical properties

Planar shrinkage was calculated by measuring the diameter of the samples after pressing and after sintering. Mass of the samples was weighed after binder removal as well as after sintering to calculate the Pb content. The density of the samples measured by Archimedes method calculated from the following equation:

% Density =
$$\frac{D}{S-I} \times \frac{100}{D_{th}}$$
 (4.1)

where D, S, I and D_{th} are dried weight, saturated weight, immersed weight and theoretical density, respectively. (D_{th} : PMN/PT 65/35 = 7.95 g/cm³, D_{th} : PMN/PT 90/10 = 7.9 g/cm³)

4.3.2 Electromechanical properties

- The piezoelectric charge coefficient, d_{33} , was measured with a Berlincourt piezometer (Channel Products, Inc.) at 100 Hz. At least ten points on each sample were recorded and averaged.

- The capacitance and dielectric loss were measured at room temperature and 1 kHz using an HP 4194A impedance/gain-phase analyzer (HP. Palo Alto, CA). Then the relative permittivity was calculated by:

$$K = \frac{Ct}{\mathcal{E}_o A} \tag{4.2}$$

where K, C, t and A are the relative permittivity, capacitance, thickness between the electrodes, and electrode area, respectively. The ε_0 is the permittivity of free space (8.854 x 10^{-12} F/m).

- The impedance analyzer was used to measure the frequency of maximum and minimum impedance (f_m and f_n) of the disks in order to compute the electromechanical coupling coefficients including planar (k_P), thickness (k_t), transverse (k₃₁) and longitudinal (k₃₃) coupling coefficients as well as Poisson's ratio (σ^P) of the samples according to the IEEE standards [128]. For the material in this study, it might be assumed that $f_r \approx f_m \approx f_s$ and $f_p \approx f_a \approx f_n$. The equations of the coupling coefficients are presented below:

$$\frac{k_p^2}{1-k_p^2} = \frac{\Delta f\left[\left(\boldsymbol{\sigma}^p\right)^2 + \boldsymbol{\eta}^2 - 1\right]}{f\left(1+\boldsymbol{\sigma}^p\right)}$$
(4.3)

$$\left(\mathbf{k}_{t}^{2}\right) = \frac{\pi}{2} \frac{f_{s}}{f_{p}} \tan\left(\frac{\pi}{2} * \frac{\Delta f}{f_{p}}\right)$$
(4.4)

$$(k_{31})^2 = \frac{k_p^2}{2} (1 - \sigma^p)$$
 (4.5)

$$(k_{33}^2) = k_p^2 + k_t^2 - k_p^2 k_t^2$$
 (4.6)

Where f_s and f_p are the measured fundamental series and parallel resonance frequency, respectively. Δf is $f_p - f_s$, σ^p is the Poisson's ratio and η is the frequency constant of a disk resonator. The Poisson's ratio was measured from the ratio of the first overtone to fundamental resonance frequency $(f_s^{(2)}/f_s)$ of the planar mode.

4.4.1 Effect of compaction pressures and sintering temperatures on physical properties of single piezoelectric and electrostrictive materials

The SEM micrographs of piezoelectric PMN-PT after sintering at 1150, 1200 and 1250 °C are demonstrated in Fig. 4.1. The observed micrographs showed irregularshaped grains with trans-granular fracture characteristics. The grain size increased with increasing sintering temperature (from 1150 to 1250 °C). Figure 4.2 depicts the SEM micrographs of electrostrictive PMN-PT at sintering temperature of 1150, 1200 and 1250 °C. The corner-shaped grains were observed with the inter-granular fracture along grain boundaries. The larger grain sizes were obtained by increasing sintering temperatures. In comparison with piezoelectric PMN-PT, electrostrictive PMN-PT had higher degree of packing and were denser. However, larger pore sizes and a large amount of porosity (Figure 4.1c and 4.2c) were found in the samples sintered at 1250 °C. The reasons were attributed from grain growths creating the pores as well as PbO content during high sintering temperature affecting to sinterability which will be discussed later.

The relative bulk density of both compositions vs. sintering temperature for various compaction pressures is depicted in Fig. 4.3. The electrostrictive ceramics showed slightly higher relative bulk densities than their piezoelectric counterparts (97-99 % for the former and 93.5-96.5 % for the latter). Nevertheless, the sintering temperature of 1250 °C yielded lower relative bulk density in both compositions. This was attributed to higher porosity in samples (Fig. 4.1 c and 4.2.c) and also to the higher PbO evaporation at elevated sintering temperature. Figure 4.4 shows PbO loss/gain vs. sintering temperature in piezoelectric PMN-PT and electrostrictive PMN-PT. Higher PbO

loss was found by increasing sintering temperature in piezoelectric PMN/PT (Fig. 4.4 a). While in the case of electrostrictive PMN-PT, PbO gain was observed (Fig. 4.4 b). The weight gain in electrostrictive ceramics caused from either the deficient PbO content during synthesis the electrostrictive powder as also observed by Swartz [129] or the excessive PbO source used during sintering as reported by Paik [130]. The PbO gain caused to higher density and degree of sinterability in electrostrictive PMN-PT. This is because excess PbO in the PMN system promoted the liquid phase sintering reported by Costa [131]. At 1250 °C sintering temperature, however, the lead loss was higher for piezoelectric ceramics (Fig. 4.4a) and the lead gain was lower for electrostrictive samples (Fig. 4.4 b). These results were in a good agreement with the lowest bulk densities obtained as shown in Fig. 4.3.

The relationship between the compaction pressures, sintering temperatures and planar shrinkages of both PMN/PT compositions was studied in order to successfully fabricate piezoelectric/electrostrictive monolithic bi-layer composites (PE-MBLC) with different geometries (flat or dome). The planar shrinkage data at three different compaction pressures (86, 114 and 229 MPa) vs. three different sintering temperatures (1150, 1200, 1250 °C) is shown in Fig. 4.5. In general, the planar shrinkages of piezoelectric and electrostrictive materials fell in the range of 13-17 %. In both materials, compaction pressure had a more noticeable effect on planar shrinkage than sintering temperatures. It must be noted that the samples sintered at 1250 °C showed the lowest shrinkage because the highest PbO loss in piezoelectric PMN-PT and lowest PbO gain in electrostrictive PMN-PT (Figure 4.4) resulted in lower sinterability as described earlier. The result from planar shrinkage was utilized to control the geometry of PE-MBLC. For
instance, the flat PE-MBLC was fabricated using 86 MPa pressure and 1150 °C sintering temperature since piezoelectric and electrostrictive samples had similar planar shrinkage of 16.16 %. The dome-shaped PE-MBLC was fabricated using 86 MPa pressure and 1200 °C sintering temperature. At this temperature, piezoelectric and electrostrictive ceramic showed different shrinkage of 16.6 and 16.3 %, respectively.



Figure 4.1 SEM micrographs of fractured piezoelectric PMN-PT after sintering at a) 1150 $^{\circ}$ C b) 1200 $^{\circ}$ C and c) 1250 $^{\circ}$ C.



Figure 4.2 SEM micrographs of fractured electrostrictive PMN-PT after sintering at a) 1150 $^{\circ}$ C b) 1200 $^{\circ}$ C and c) 1250 $^{\circ}$ C.



Figure 4.3. Percent bulk density of piezoelectric PMN-PT and electrostrictive PMN-PT at three pressures (86, 114, 229 MPa) and temperatures (1150, 1200, 1250 °C).



Figure 4.4. % PbO loss/gain of a) piezoelectric PMN-PT and b) electrostrictive PMN-PT vs. sintering temperatures (1150,1200,1250 °C) at three pressures (86,114,229 MPa).



Figure 4.5. Percent planar shrinkage of piezoelectric PMN-PT and electrostrictive PMN-PT at three pressures (86,114,229 MPa) and temperatures (1150,1200,1250 °C).

4.4.2 Effect of poling conditions on electromechanical properties

To optimize the poling field of piezoelectric ceramic, the poling condition at 60 ^oC for 20 minutes under 1-40 kV/cm E-field with 5 kV/cm increments was investigated. Figure. 4.6 illustrates the electromechanical properties vs. E-field. As shown, the d_{31} values were reached to highest value at 5 kV/cm applied electric filed and did not increase at elevated voltage. The values of d₃₃ and all coupling coefficients (k_p, k_t and k₃₁) were stable after 25 kV/cm. However, it was observed that the dielectric constant decreases slightly with higher applied voltage. For future polarization condition, E-field at 30 kV/cm was chosen to assure that piezoelectric ceramics were fully poled. Next step in optimization of the poling duration, while poling temperature and applied electric field were chosen constant at 60 °C and 30 kV/cm, the dwell time from 5 to 40 minutes was investigated. Figure 4.7 shows that all electromechanical properties were reached their maximum values after 5 minutes poling. However, the value of relative permittivity decreased with increasing dwell time. The slight decrease in the relative permittivity at higher voltage (Fig 4.6 a) or longer poling period applied (Fig 4.7 a) caused from the micro-cracks generated in the sample. Tai also observed the decreasing of relative permittivity due to micro-cracks after cyclic loading [132]. At this stage, the optimized poling condition was set to be 30 kV/cm, 60 °C and 5 minutes throughout the rest of this study.

The effect of the sintering temperature on piezoelectric charge coefficients for the samples prepared under different compaction pressures is shown in Fig. 4.8. The measured d_{33} and d_{31} values were achieved and were in good agreements with the values reported by TRS technologies Inc shown in Table 4.1.



Figure 4.6. Electrical properties of piezoelectric PMN/PT poled at 60 °C, 20 minutes and varying applied voltage.



Figure 4.7. Electrical properties of piezoelectric PMN/PT poled at 30 kV/cm, 60 °C and varying poling time.



Figure 4.8. d_{33} and d_{31} of piezoelectric PMN/PT poled at 30 kV/cm, 60 $^{\circ}$ C, 5 minutes

4.5 Conclusions

The processing and poling conditions were studied for the PMN-PT ceramics used. The measured planar shrinkages of both materials (piezoelectric and electrostrictive) were found to be about 13-17 % for the compaction pressures (86, 114 and 229 MPa) and sintering temperatures (1150, 1200, 1250 °C) employed in this study. Under these conditions, the piezoelectric and electrostrictive ceramics had 97-99 % and 93.5-96.5% relative bulk densities, respectively. Electrostrictive ceramics gained 0.4-0.8 % PbO while their piezoelectric counterparts had 0.2-0.6 % lead loss. It is interesting to note that the relative bulk density and planar shrinkage decreased at 1250 °C sintering temperature. Considering the planar shrinkage, flat and dome-shaped PE-MBLCs were fabricated using 86 MPa compaction pressure and 1150 °C and 1200 °C sintering temperatures, respectively. The optimum poling condition for a single piezoelectric ceramic was found to be 30 kV/cm, 60 °C and 5 minutes.

Chapter 5

Processing and poling study of bi-layer Piezoelectric/Electrostrictive

5.1 Introduction

This chapter will discuss the effect of poling conditions on the properties of stacked, laminated and co-fired bilayer piezoelectric/electrostrictive PMN-PT ceramics. Specifically, these bilayer ceramics were constructed in order to compare the effect of the different piezoelectric and electrostrictive interface on the dipole moment realignment during poling process. In the stacked bilayer, the individual sintered and electroded piezoelectric and electrostrictive disks were placed on top of one another. While the laminated bilayer was fabricated by bonding the individual sintered and electroded piezoelectric (P) and electrostrictive (E) disks together using a thin layer of room temperature cured epoxy (Epotek-301, Epotek technologies, MA). The co-fired bilayer was fabricated by co-pressing and co-sintering of piezoelectric and electrostrictive powders. Two different geometries, flat and dome-shaped co-fired bilayer ceramic actuators, were fabricated.

The poling is an important step to align the dipoles in piezoelectric and ferroelectric materials. Thus the optimum poling conditions which provided the highest electromechanical properties in co-fired bilayer composites should be investigated. The poling of co-fired bilayer composites is more complex than a single piezoelectric ceramic because of the difference in relative permittivity (K) of the piezoelectric and electrostrictive materials (4000 and 20000, respectively) at room temperature. Therefore, considerable voltage drop across the electrostrictive layer due to relative permittivity

difference effecting on poling of piezoelectric layer. This chapter aims to present an in depth study of poling conditions including applied E-field, temperature, and dwell time of bilayer ceramics with respect to the relationship between permittivity of P and E ceramics and the induced stress from P/E interface. Therefore, the following five bilayer ceramics have been studied:

- 1. Stacked P/E composite: electroded piezoelectric and electrostrictive poled together
- Laminated P/E composite: electroded and poled piezoelectric bonded to electroded electrostrictive
- 3. Laminated P/E composite: electroded piezoelectric and electroded electrostrictive bonded and poled together
- 4. Co-fired P/E composite : flat PE-MBLC poled at once
- 5. Co-fired P/E composite : dome-shaped PE-MBLC poled at once

5.2 Fabrication of bi-layer Piezoelectric/Electrostrictive (P/E) composite

5.2.1 Stacked and laminated bi-layer P/E composite

Individually sintered piezoelectric and electrostrictive ceramic disks were polished with 1200 grit sandpaper down to 0.5 mm in thickness to obtain 1:1 P/E thickness ratio. In the stacked bilayer composite, the individual sintered and electroded piezoelectric and electrostrictive ceramics were placed on top of one another. While the laminated bilayer composite was fabricated by bonding the individual sintered and electroded piezoelectric and electrostrictive disks together using a thin layer of room temperature cured epoxy (Epotek-301, Epotek technologies, MA). Figure.5.1 a-c schematically shows stacked and laminated bi-layer P/E composite designs.

5.2.2 Co-fired bi-layer P/E composite

Co-fired bi-layer P/E composite were made by pressing piezoelectric and electrostrictive materials sequentially in a single pressing step. First, the piezoelectric powder was put into the die, and then the punch was used to flatten this layer to form a uniform thickness. Then, the electrostrictive powder was added on top of the flattened piezoelectric material and co-pressed at 86 MPa. The pressed powders were removed binder at 550 °C for 3 hours and then were sintered in closed alumina crucible with PbO source, at 1150 °C and 1200 °C to obtain flat and dome-shaped bi-layer samples, respectively. Figure.5.1 d-e also depicts the schematic representation of flat and dome co-fired bi-layer P/E composite designs.

5.3 Characterizations

- The piezoelectric charge coefficient, d_{33} , was measured at 100 Hz using a Berlincourt piezometer (Channel Products, Inc.). At least ten points on each sample were recorded and averaged.

- The capacitance and dielectric loss were measured at 1 kHz and room temperature using an HP 4194A impedance/gain-phase analyzer (HP. Palo Alto, CA).

- The impedance analyzer was used to measure the frequency of maximum and minimum impedance (f_m and f_n) of the disks in order to compute the electromechanical coupling coefficients (k_p , k_t , and k_{31}) of the samples according to the IEEE standard [128].

- The hysteresis loops (P-E) of the samples were measured at various temperatures and electric fields using all three components of precision high voltage interface, the Trek high voltage power supply and vision software (Radiant Technologies Inc.,). The measurements were performed with samples immersed in a silicone oil bath.

- The displacement measurements were carried out by placing the samples in a high voltage displacement fixture and using a photonic fiber optic sensor probe (MTI2000 Instruments, Latham, NY), high voltage power supply (Trek 610 C from Trek Inc, Medina, NY), and vision software (Radiant Technologies Inc.).



Case 1 Stacked P/E composite: piezoelectric and electrostrictive poled together

Case 2 Laminated P/E composite: poled piezoelectric bonded to electrostrictive







Case 4 Co-fired P/E composite: Flat PE-MBLC





Poled flat PE-MBLC and measured d_{33} of composite

Case 5 Co-fired P/E composite: Dome-shaped PE-MBLC



Figure 5.1. Different types of bi-layer P/E composite designs.

5.4.1 Optimization of poling condition for bi-layer piezoelectric/electrostrictive (P/E)

As per the laws of electromagnetism, the following relations related to the dielectric displacements of individual electrostrictive and poled piezoelectric ceramics [133].

$$D^{p} = P_{s} + K_{1}^{p}E^{p} + K_{2}^{p}(E^{p})^{3} \quad \text{with } K_{2}^{p} < 0,$$
(1)

$$D^{e} = K_{1}^{e}E^{e} + K_{2}^{e}(E^{e})^{3} \qquad \text{with } K_{2}^{e} < 0, \qquad (2)$$

where the superscripts p and e denote piezoelectric and electrostrictive ceramics, respectively. P_s , K_1 , K_2 , and E are the spontaneous polarization, linear permittivity, third order permittivity, and electric field, respectively. The P_s will be equal to zero for an unpoled piezoelectric ceramic.

The P/E bi-layer composites represent two capacitors in a series model (Figure 5.2) where under steady state conditions

$$\mathbf{D}^{\mathbf{p}} = \mathbf{D}^{\mathbf{e}} \tag{3}$$

The equality $D^p = D^e$ cannot hold for $K_1^p = K_1^e$ as in the case for a linear dielectric since a piezoelectric and electrostrictor constitute a nonlinear dielectric media.



Figure 5.2. Model of two capacitors connected in series.

The electric field variable E^p and E^e clearly indicate that the electric field magnitude in each layer is by no means equal to the applied field but obey an empirical relation of the form $E = xE^P + (1-x)E^e$ where x is the electric field partitioning parameter $(0 \le x \le 1)$ [133]. This parameter is most critical as the electrostrictive layer causes a large voltage drop, reducing the voltage across the piezoelectric layer. Therefore, poling need to be carried out at optimized condition (high fields and/or high temperatures) to ensure saturate poling of the piezoelectric layer in the bi-layer composite.

The effective piezoelectric charge coefficients (d_{33}^{eff}) in bi-layer P/E composites have been calculated as a function of piezoelectric layer thickness (Fig. 5.3) using the equation (4) [134], in which $K_{33}^{E} = 18500$, $K_{33}^{P} = 4130$, $d_{33}^{P} = 570$ pC/N, t^{P} = thickness of piezoelectric layer, and t^{E} = thickness of electrostrictive layer.

$$d_{33}^{eff} = \frac{t^{p} d_{33}^{p} K_{33}^{E}}{t^{p} K_{33}^{E} + t^{E} K_{33}^{p}}$$
(4)

Figure 5.3 compares the calculated d_{33}^{eff} values with those of different actuator designs and poling conditions from Hall [134]. As shown, the measured d_{33}^{eff} values of P/E composites with 1:1 thickness ratio were lower than the calculated d_{33}^{eff} . This was

attributed to inefficient domain switching along applied field in the piezoelectric layer of the P/E monomorph or bimorph during the poling process which may be due to one or both of the following reasons:

- 1. The voltage drop across the electrostrictive layer is due to the difference in permittivity which could cause insufficient domain switching in the piezoelectric layer
- 2. The effect of induced stress produced in P/E monomorph and/or bimorph



Figure 5.3. Relationship between calculated effective d_{33} and piezoelectric layer thickness (%) comparing with the bimorph and monomorph P/E composites from Hall [134].

To investigate these hypotheses and find the optimum poling condition to achieve saturated poling of piezoelectric layer co-fired P/E composite, five different designs of bi-layer composites (Fig. 5.1) were studied intensively. According to difference in relative permittivity of piezoelectric and electrostrictive, the distribution of the E-field in the two layers was investigated. Thus, the dielectric displacement vs. E-field of single piezoelectric, single electrostrictive and stacked P/E bi-layer composite (case 1) have been examined as shown in Fig. 5.4. The tests were performed at 25 °C (Fig. 5.4.a) and 60 °C (Fig. 5.4.b).

As shown, D values of stacked P/E bi-layer at both temperatures fell between the D values of two single ceramics. Considering $D^p=D^e$ for two capacitors connected in series, the electric field magnitude experienced by each layer at a given value of dielectric displacement could be estimated. For example, at D = 25 μ C/cm² (25 °C), E-field distribute across single piezoelectric and electrostrictive layers were about 7 and 28 kV/cm, respectively. As predicted, the E-field applied on piezoelectric layer was much smaller than that of electrostrictive layer which could cause piezoelectric layer to be only partially poled. In addition, these two plots indicated that dielectric displacements of single piezoelectric, electrostrictive and stacked P/E bi-layer composite were slightly higher at 25 °C than those at 60 °C. The results indicated that 25 °C was the suitable temperature for poling of the stacked P/E bi-layer composite at various E-fields.



Figure 5.4. Dielectirc displacement (D) and E-field of single PMN/PT-65/35, PMN/PT-90/10 and stacked bi-layer P/E at a) 25 $^{\circ}$ C and b) 60 $^{\circ}$ C

Figure 5.5.a shows the results of d_{33}^{eff} of stacked composite (case 1) and d_{33} of piezoelectric layer versus applied field poled at 25 °C for 5 minutes in each applied field. The d_{33}^{eff} and d_{33} increased with increasing E-field to 15 kV/cm, became stable at 15-25 kV/cm and 20-30 kV/cm for stacked composite and single piezoelectric, respectively. And then both d_{33} and d_{33}^{eff} were decreased at applied field of over 30 kV/cm. It is interesting to note that, the d₃₃ of single piezoelectric ceramic poled at 20-25 kV/cm from stacking design was close to the d_{33} value for a piezoelectric ceramic poled alone (d_{33} = 580). Therefore, it could be concluded that the optimum poling field to pole the stacked P/E composite was 25 kV/cm. Figure 5.5.b shows the effect of poling duration when stacked P/E composite poled at 25 $^{\circ}$ C and 25 kV/cm. With increasing time, both d₃₃ eff and d₃₃ increased and saturated beyond 25 minutes. Form this set of experiment, one could conclude that the optimal poling condition for stacked P/E composite was 25 °C and 25 kV/cm and 30 minutes. This poling condition was then applied to different actuator designs and their d_{33}^{eff} were recorded as shown in Table 5.1. The actuator designs in Case 1 and 2 achieved calculated d_{33}^{eff} (475 pC/N), while other designs (laminated P/E composite in case 3 and co-fired P/E composite in case 4 and 5) showed lower d_{33}^{eff} values. This was attributed the development of induced stress at the P/E interfaces. The stacked P/E (without bonding layers and hence with no interface layer) had no induced stress and thus the domains in piezoelectric layer could freely switch to the direction of applied E-field during poling. As a result, d₃₃ of piezoelectric layer reached about 580 pC/N and the d_{33}^{eff} of stacked P/E reached the calculated value. The presence of epoxy layer in laminated composite and P/E interface in the co-fired composite created a stiff and the stiffest interface between the P and E layers,

respectively. These resulted in inducing stresses in the piezoelectric layer which could act against the domain alignment during poling.

From this results, one could conclude that a large difference of the relative permittivity of piezoelectric and electrostrictive ceramics as well as induced stress from the P/E interface affected to domain alignment in the piezoelectric layer during poling of the bi-layer P/E composite.

The optimum poling condition of the laminated (case 3) and co-fired composite (case 4 and 5) was needed further investigation to improve d_{33}^{eff} of the composites. Initially, the laminated bi-layer composite was poled at 25 °C, 30 minutes, under 20 kV/cm giving rise to d_{33}^{eff} of 290 pC/N. Due to a slight increase of d_{33}^{eff} it was decided to further investigate the effect of poling duration on the saturated polarization which is shown in Fig. 5.6. The d_{33}^{eff} gradually increased with poling time and reached its saturation value of 475 pC/N after 90 minutes poling. A good agreement between the experimental and calculated values (475 and 460pC/N, respectively) was attained.



Figure 5.5. d_{33} and d_{33}^{eff} of stacked bi-layered P/E by varying a) E-field b) time.

Case	Actuator design	d ₃₃ ^{eff}
1	- Stacked : piezoelectric and electrostrictive	475
2	- Laminated : poled piezoelectric bonded to electrostrictive	460
3	- Laminated : bonded piezoelectric and electrostrictive	230
4	- Co-fired : flat PE-MBLC	156
5	- Co-fired: dome-shaped PE-MBLC	200

Table 5.1 Various bi-layer P/E designs and d₃₃^{eff} poled at 25 kV/cm, 25 °C and 30 mins

Figure 5.7 shows the combined effect of the temperatures and the electric fields on the d_{33}^{eff} of the co-fired composite (cases 4 and 5). At first, both samples were poled at 25 kV/cm, 25 °C for 60 minutes (both lines, step 1-2) which yielded the d_{33}^{eff} about 210 pC/N. Then the samples were poled at higher temperature at 150 °C (upper line, step 3) resulting in increase d_{33}^{eff} from 210-270 pC/N, with the poling duration of 10 minutes. On the other hand, increase of electric field from 25 to 40 kV/cm showed slight increase in d_{33}^{eff} from 200-225 pC/N (lower line, step 3). This indicated that the temperature had a more pronounced effect on the d_{33}^{eff} than the E-field applied. Further poling to 120 minutes, with the same applied electric field (25 kV/cm) and then cooled the temperature down to room temperature (upper line, step 5), increased the d_{33}^{eff} 380 pC/N. This experiment indicated that poling at high temperature can provide enough energy to effectively switch the domains in co-fired composite.



Figure 5.6. Effect of poling time on laminated P/E sample at 20 kV/cm, 25 °C



Figure 5.7. Effect of E-field, temperature and time on d_{33} effective of co-fired P/E.

Based on the previous poling study of co-fired composite, it was concluded that high temperature poling facilitated domain switching and lowering poling temperature to room temperature with the same application of an electric field could lock the switched domain in the direction of an applied E-field and hence providing high d_{33}^{eff} in co-fired composite. This two step poling condition of : 25 kV/cm, 150 °C, 10 minutes then cooling down to 25 °C in 40 minutes at the same field applied, and poling at 25 °C for additional 30 minutes was applied to co-fired P/E composites. With this poling condition, an average d_{33}^{eff} of 320 ± 20 pC/N was obtained in dome co-fired P/E composites.

Although, the d_{33}^{eff} was substantially improved, the measured value of and 320 pC/N was still lower than the calculated value of 460 pC/N. This was attributed to

- A large difference of relative permittivity of the piezoelectric and electrostrictive ceramics which caused incomplete poling in piezoelectric layer.
- Presence of residual stresses in the piezoelectric layer which was imposed by the electrostrictive layer. Such residual stress could be generated after the sintering as well as during poling.

In sintering process, thermal expansion coefficient and sintering shrinkage mismatches between two materials could induce stress in the piezoelectric layer. This has been observed in Rainbow where the reduced layer has higher thermal expansion coefficient (~10 x 10⁻⁶ C⁻¹) than the unreduced layer (~5 x 10⁻⁶ C⁻¹) [135]. Thus, the dome or saddle Rainbows with the tensile/compressive stress at interface of the reduced layer and unreduced layer were developed [68]. In present study, piezoelectric (PMN-PT-65/35 : TEC \approx 10 x 10⁻⁶ C⁻¹) and electrostrictive (PMN-PT-90/10 : TEC \approx 6 x 10⁻⁶ C⁻¹) were used

[127]. Considering TEC mismatch between these two materials, it could be concluded that the electrostrictive layer constrained the piezoelectric layer (Figure 5.8). The piezoelectric layer at interface was under tensile force hindering the domain realignment during poling.

In poling process, different strain-E-field response could be expected in piezoelectric and electrostrictive material. Piezoelectric has more contraction than electrostrictive material when subjected to E-field. So electrostrictive layer acted as constrain layer at the P/E interfaces and prohibited the contraction of the piezoelectric layer. As a result, tensile stress could be generated in piezoelectric layer and obstructed to switching of the domains in the poling process. This tensile stress was more dominant in the co-fired P/E than laminated P/E composite due to the fact that co-fired P/E composites possessed a stronger bond from the monolithic ceramic interface which strongly affected the d_{33}^{eff} , resulting in lower than calculated value.



Figure 5.8. Schematic representation of shrinkage and induced force on piezoelectric and electrostrictive materials a) before sintering b) after sintering (no P/E interface)c) after sintering (with P/E interface)

5.4.2 Actuation behavior of bi-layer piezoelectric/electrostrictive (P/E)

Uni-polar actuation performance of the following actuator designs were investigated using a non-contact photonic fiber optic sensor probe (MTI 2000) together with a high voltage power supply and Vision Software. The schematic displacement setup is shown in Fig. 5.9.

- Case 1 : Laminated : poled electroded piezoelectric ceramic bonded to electroded electrostrictive ceramic
- Case 2 : Laminated : bonded electroded piezoelectric and electrostrictive ceramics and then poled
- Case 3 : Flat PE-MBLC
- Case 4 : Dome-shaped PE-MBLC

Since there were flat and dome-shaped actuator geometries, two different types of supports (planar and ring support) were used for displacement measurement, as shown in Fig. 5.10 a and b. The purpose of using these two different supports was to have comparable contact area of flat and dome samples to the support. Figure 5.10.c also shows the set up without support which was used only for flat bi-layer P/E actuators.

Table 5.2 compares the results of d_{33}^{eff} and displacements of all actuators. The cofired P/E bi-layer actuators gave comparable or more displacement than the laminated actuators, even through the d_{33}^{eff} of co-fired actuators was significantly lower than that of laminated actuators. Also, the actuator geometry had a more pronounced effect on the displacement. The highest displacement $\approx 15 \ \mu m$ was obtained from dome co-fired P/E actuator, which was poled with two step process. It appeared that residual stress in dome shape actuator enhanced the field-induced displacement due to domain reorientation, which was similar to what was observed in Rainbow actuator [15].

The effect of support on displacement of flat P/E actuators is shown in Table 5.2. By using planar and ring support for flat samples (Case 1, 2 and 3), a displacement range of 6-11 μ m was observed. Without using support (Fig. 5.10 c), the displacement about 3 μ m was obtained. This is because the use of support could add up the actuating force to the actuators during excitation.

From this experiment, all geometries (flat and dome) of the bilayer composites and the displacement set-up have intensively studied. One could be summarized that the planar or ring support was suitable for flat P/E actuators resulting in higher displacement compared to without support. For dome P/E actuators, only planar support was suitable.



Figure 5.9 The schematic representing the displacement measurement.



Figure 5.10. Support configuration for displacement testing a) planar support for dome and flat sample b) ring support for flat sample. c) no support for flat sample.

Sample Designation	Feature	d_{33}^{eff}	Displacement (µm)		
		(pC/N)	Planar	Ring	Without
			support	support	support
Case 1-Laminated : poled electroded piezoelectric bonded to electroded electrostrictive ceramic - 25kV/cm, 25°C,30 mins	Flat	460±55	10.5±0.5	11±1.31	2.8±0.29
Case2- Laminated : bonded electroded piezoelectric and electroded electrostrictive ceramics poled together - 20kV/cm, 25°C, 90 mins	Flat	475±15	9±2.91	9.8±1.57	3.1±0.47
Case 3- Flat PE-MBLC - Single step poling* - Two steps poling **	Flat	180±10 270±40	6.75±1.13 9.36±0.89	10.23±0.64 11.65±0.33	2.65±0.21 3.2±0
Case 4- Dome-shaped PE-MBLC - Single step poling* - Two steps poling**	Dome (≈0.2mm)	184±15 350±20	9.2±0 15±1.5	-	-

Table 5.2. Electromechanical properties and displacement of four different types bilayer P/E actuator designs measured at 10 kV/cm and 0.5 N loading.

* At 25 kV/cm, 25 °C, 30 mins

** At 25 kV/cm, 150 °C,10 mins---->25 kV/cm, cool down to 25 °C, 40 mins ---->25 kV/cm,25 °C,30 mins.

5.5 Conclusions

In this chapter, the processing and poling conditions related to electromechanical properties and actuation were studied in bi-layer P/E composites. Poling of various types of bi-layer P/E designs including stacked, laminated, and co-fired were studied to achieve efficient poling in bi-layer P/E composites. The experimental results were summarized as follows:

- 1. The co-fired ceramics had lower d_{33}^{eff} than the calculated value due to:
 - the difference in permittivity of piezoelectric and electrostrictive layers leading to impede domain alignment in piezoelectric layer during poling.
 - the induced stress generated between the P and E interfaces affecting the domain switching.

2. The optimal poling conditions of flat and dome co-fired bi-layer P/E composites were followed at 25 kV/cm, 150 °C 10 minutes, then cooling down to 25 °C for 40 minutes with electric field applied and further poling at 25 °C, for 30 minutes. With such poling condition, the d_{33}^{eff} of flat and dome composites were improved to about 270 and 350 pC/N, respectively.

3. The dome co-fired composite generated the highest displacement of 15 μ m under 10 kV/cm applied.

Chapter 6

Electromechanical displacement of piezoelectric-electrostrictive monolithic bi-layer composites

6.1 Introduction

Piezoelectricity is a first order electromechanical phenomenon which relates the mechanical displacement or strain (x) to electric filed (E) through the converse piezoelectric effect, i.e. $x_i = d_{ij}E_j$ where d_{ij} is piezostrain coefficient and i=1,...,6; j=1,2,3 [2]. This phenomenon is exploited in actuator applications including variable focus mirrors, positioners, noise vibration dampers, etc.[3]. Owing to its high piezoelectric properties in the vicinity of morphotropic phase boundary, lead zirconate titanate systems (PZT) and solid solution PMN-PT have been the most preferred materials as electromechanical actuators [3;44]. The major drawback of bulk ceramic piezoelectric actuators is their low level of strains, which is on the order of 0.1%. Electrostriction, on the other hand, is a second order electromechanical phenomenon, which is typically expressed as $x_i = M_{ij}E_j^2$, where M_{ij} is the electrostriction coefficient, and i=1,...,6; j=1,2,3. Electrostrictive compositions in the Pb(Mg_{1/3}Nb_{2/3})O₃ system (PMN), and its solid solutions with PbTiO₃ (PMN-PT) have also gained popularity in solid state actuators since strains up to 0.15% are achievable without any hysteresis in their strain-E field responses [3;44].

In this chapter, the physical and electromechanical behaviors of piezoelectricelectrostrictive monolithic bi-layer composites (PE-MBLC) as a function of piezoelectric phase volume fraction have been investigated. The objective was to experimentally determine the macroscopic behavior of a bi-layer monolithic nonlinear elasto-dielectric system under a unipolar applied field. The materials used in this study were 0.65PMN-0.35PT as the piezoelectric and 0.9PMN-0.1PT as the electrostrictor, respectively. The details of fabrication steps of monolithic bi-layer composites of piezoelectric (P) and electrostrictive (E) via co-firing were discussed in Chapter 5.
6.2 Characterization techniques

6.2.1 Optical Microscope

The thicknesses of the piezoelectric and electrostrictive layers of the monolithic composite were determined by optical microscopy (Olympus America Inc., Melville, NY).

6.2.2 Dome height

The dome height referred to as the distance from the base to the center of the inner dome surface was measured by a caliper with 0.01 mm resolution (Mitutoyo, U.S.A).

6.2.3 Electromechanical properties

- The effective piezoelectric charge coefficients (d_{33}^{eff}) were measured and calculated as described in section 5.4.1. The capacitance values were measured at room temperature and 1 kHz using an HP 4194A impedance/gain-phase analyzer. The relative permittivity was calculated using the equation shown in section 4.3.2. The effective relative permittivity of the PE-MBLCs as a function of the piezoelectric volume was calculated by:

$$K_{33}^{eff} = \frac{K_{33}^{P}K_{33}^{E}}{t^{P}K_{33}^{E} + t^{E}K_{33}^{P}}$$
(1)

where K_{33}^{E} and K_{33}^{p} represent the relative permittivity of electrostrictive and piezoelectric ceramics, respectively. The t^E and t^p are the thickness of electrostrictive and

piezoelectric layers, respectively. Also, the effective coupling coefficients of PE-MBLCs were measured and calculated as described in section 4.3.2.

The uni-polar axial displacements of PE-MBLC actuators were measured by an MTI-2000 photonic sensor in conjunction with a 10 kV (DC) power supply (Trek 610C) and a special displacement fixture. The concave side (piezoelectric layer) of PE-MBLC was placed on a planar metallic support as shown in Fig. 5.10.a. The applied electric field was parallel to the poling direction in all measurements. The same E-field strength of 10 kV/cm was applied to the composite actuators during displacement testing.

6.3 Results and discussions

The optical micrographs of the thicknesses of piezoelectric and electrostrictive layers of the PE-MBLCs after sintering are shown in Fig. 6.1. All sintered PE-MBLCs had fully dense and clear interfaces between the P and E layers with no apparent porosity. However, the tolerance between the designed and actual thicknesses after sintering fell in the range of 1-6 %. A large difference between the designed and actual thicknesses was more pronounced in the PE-MBLC with P:E 9:1 (Fig. 6.1c) compared to P:E 5:5 (Fig.6.1a). The main reason for the thickness variation was due to the powder loading issue into the mold during pressing. For example PE-MBLC (P:E 9:1), a small amount of the electrostrictive powder could not be spread over the cross sectional area of the die during the loading of powder. The second reason could be due to the diffusion of atoms (Pb, Mg, Nb and Ti) in piezoelectric and electrostrictive materials around their interface during sintering as observed by Hall [134]. She reported that the interface thickness of a co-sintered piezoelectric and electrostrictive materials was about 25 µm. Nevertheless, the moderate TEC mismatch and planar shrinkage differences of the two materials resulted in no crack/delamination after sintering. Additionally, the obtained interfaces and the shape change (planar to dome shape) of PE-MBLC composites with various piezoelectric and electrostrictive thickness ratios indicated that P and E materials were compatibly sintered. The densities of sintered PE-MBLC samples were found to be >97 % of theoretical density.

The measured dome height vs. volume percent piezoelectric material in PE-MBLCs is depicted in Fig. 6.2. All samples were sintered at 1200 °C for 1 hour. The dome height gradually increased as the volume percent piezoelectric (piezoelectric thickness) increased. The maximum dome height (~0.5 mm) was obtained from PE-MBLC with 80 vol % piezoelectric phase. The shape change from planar to dome was attributed to the difference in the planar shrinkages (16.47 % for PMN-PT 65/35 and 16.33 % for PMN-PT 90/10) and the TEC mismatch (~4x10⁻⁶ °C⁻¹) of both materials. Since the piezoelectric ceramic had more planar shrinkage and higher TEC than the electrostrictive ceramic, the piezoelectric layer was in the concave side and the electrostrictive was in the convex side of PE-MBLC. From the shape change (dome structure), it was expected that the stress was developed in the PE-MBLC samples after sintering. Therefore, the shape change-state of the stress relationship in PE-MBLCs will be intensively investigated and discussed in the next chapter.



Figure 6.1. Optical cross sectional micrographs of representative piezoelectricelectrostrictive monolithic bi-layer composites with: a) 50 volume percent, b) 70 volume percent and c) 90 volume piezoelectric phase.



Figure 6.2. The variation of dome height with volume percent piezoelectric phase in piezoelectric-electrostrictive monolithic bi-layer composites co-sintered at 1200 $^{\circ}$ C for 1 hour.

The following section presents the effective piezoelectric charge coefficients (d_{33}^{eff}) , effective relative permittivity (K^{eff}), and effective coupling coefficients of the PE-MBLC with various piezoelectric volume percent (from 0 to 100 % with 10 % increments). All samples were poled at 25 kV/cm electric field strength in a heated silicone oil bath for 10 minutes at 150 °C, then cooled down to 25 °C under the applied electric field for 40 minutes and further poled at 25 °C for 30 minutes. The samples were aged for 1 day prior to measuring their electromechanical properties.

The calculated and measured d_{33}^{eff} with various volume percent piezoelectric are shown in Fig. 6.3. The results showed that the measured d_{33}^{eff} of all PE-MBLC composites were lower than the calculated d_{33}^{eff} values.

The calculated and measured effective permittivity (K^{eff}) values as a function of piezoelectric volume percent are also depicted in Fig. 6.4. In contrast to the d_{33}^{eff} values, the calculated and measured K^{eff} decreased as the piezoelectric phase increased. The measured K^{eff} values of PE-MBLC (all piezoelectric volume percent) were also lower than the calculated K^{eff} . The discrepancy between the measured and calculated values for both piezoelectric charge coefficient and relative permittivity could be due to incomplete poling of the piezoelectric layer. A large difference between the relative permittivity of piezoelectric and electrostrictive layers resulted in a voltage drop across the P/E interfaces which hindered the switching of domains in the piezoelectric layer during poling.



Figure 6.3. A comparison of the calculated and measured effective piezoelectric charge coefficients as a function of volume percent piezoelectric phase in PEMBLC.



Figure 6.4. A comparison of the calculated and measured effective relative permittivity as a function of volume percent piezoelectric phase in PEMBLC.

As demonstrated in Fig. 6.5, the planar and thickness coupling coefficients of pure single piezoelectric (100 % piezoelectric) were 0.65 and 0.56, respectively. The coupling coefficients were gradually decreased as the volume percent of electrostrictive layer in PE-MBLC increased. The reduction in both coupling coefficients was attributed to the fact that the piezoelectric layer in PE-MBLCs was clamped by the electrostrictor layer.

Figure. 6.6 depicts the uni-polar displacement vs. piezoelectric volume percent under 10 kV/cm applied electric field. This experiment was performed by placing the concave side (piezoelectric ceramic) on the planar support (thick metal sheet). The application of an electric-field was coincided with the poling direction during testing. It was found that the uni-polar displacements of PE-MBLC composites had a Gaussian distribution with the maximum displacement of $\sim 15 \ \mu m$ at 50 vol% piezoelectric. The strain amplification was about 25 times and 40 times higher than that of a single piezoelectric (0.6 μ m) and a single electrostrictive (0.3 μ m) ceramics, respectively. Lower displacements were obtained when the amount of piezoelectric or electrostrictive layers dominated. It must be noted that PE-MBLC with the highest dome height did not generate the maximum displacement as compared the results of Figures 6.2 and 6.6. This meant that not only the stress distribution was a factor to improve the displacement but also the effect of the piezoelectric and electrostrictive strain responses influenced the strain amplification in PE-MBLC composites. In addition, the constraining interfaces of piezoelectric and electrostrictive layers allowed PE-MBLC to bend rather than to contract/expand along the thickness of the sample found single as in piezoelectric/electrostrictive ceramic.



Figure 6.5. A comparison of the calculated and measured effective coupling coefficients as a function of volume percent piezoelectric phase in PEMBLC.



Figure 6.6. Variation of axial displacement with volume percent piezoelectric phase in PE-MBLC. 0% represents a single PMN-PT 90/10 layer, and 100 % designates a single PMN-PT 65/35.

6.4 Conclusions

From this study, it was found that the shape change from planar to dome structure in co-fired piezoelectric and electrostrictor was governed by the volume ratio of the piezoelectric and electrostrictive materials. It was also realized that the effective response in d_{33}^{eff} and K^{eff} were limited by the electrostrictive layer, which acted as a nonlinear capacitor in series to the piezoelectric layer, causing incomplete poling. The effective coupling coefficients of PE-MBLCs were lower than those of a single piezoelectric ceramic since the piezoelectric layer in the composite was clamped by the electrostrictive layer. The observed amplification of displacement was shown to be strongly independent of the dome height. The maximum displacement under uni-polar excitation was observed in the PE-MBLC with 50 volume percent piezoelectric.

Chapter 7

State of stress in piezoelectric-electrostrictive monolithic bi-layer composites

7.1 Introduction

In this chapter, the states of induced stresses in the PE-MBLC actuators after sintering are presented. XRD technique was utilized to observe the intensity ratio of (200) and (002) peaks associated with the "a" and "c" domains present in the piezoelectric layer. The c-domains with their c-axes perpendicular to the XRD planes reflected (002) diffraction peak, while the a-domains with their c-axes parallel to the XRD planes reflected (200) diffraction peak. The populations of the c- and a-domains could be defined by the intensities of the (002) and (200) peaks, respectively. Therefore, the use of the intensity changes of (002) and (200) correlated to the change of the c- and a-populations [136] could determine the state of induced stress developed in the samples upon the cooling step during the sintering process.

Li [15] determined the state of stress and its distribution on the unreduced layer surface of the PLZT Rainbow actuators. In general, the $I_{(200)}/I_{(002)}$ ratio was equal to 2 with the stress-free sample due to the random orientation of the c- and a- domains. When the samples were under a tensile or compressive stress in the planar directions, the $I_{(200)}/I_{(002)}$ ratios were greater or smaller than 2, respectively. Figure 7.1 schematically represents the alignment of a- and c-domains under the induced stress and its corresponding $I_{(200)}/I_{(002)}$ ratio. He also concluded that the dome-shape configurations in Rainbow actuators were caused by the internal stress developed due to the dimension and thermal expansion coefficient mismatches of the reduced and unreduced layers. The change of dimension was attributed to the volume change of the reduced layer during the reduction process and the phase transitions during cooling through the Curie temperature of the unreduced layer. Li also found that the states of induced stress (tensile or compressive) on the surface of unreduced layer was a function of the ratio of reduced layer to the total thickness of Rainbow, as depicted in Fig. 7.2.

In obtaining dome shaped PE-MBLCs after sintering, one would anticipate that various thickness ratios of the piezoelectric to electrostrictive material could develop internal stress in the bi-layer composites. The objective of this work was to determine the state of stress generated on the surface of piezoelectric layer in the PE-MBLC actuators and to observe the domain switching behavior of PE-MBLC actuators after poling by using X-ray diffractrometry. PE-MBLC samples with P:E ratio of 8:2, 5:5 and 2:8 were chosen as the representative samples in this study.



Figure 7.1 Schematic diagram of the preferential domain orientation under the influence of the planar stresses and the corresponding intensity ratio of the (200) to (002) by X-ray diffractions [15].



Figure 7.2 The (200) and (002) diffraction profiles for Rainbow actuators with different thickness ratios. The number on the top of each profile represents the value thickness ratio (reduced layer to total thickness of the Rainbow) [15].

7.2 Experimental procedure

Single piezoelectric and/or electrostrictive ceramics and PE-MBLC samples were fabricated as described in chapters 4 and 5, respectively. The phases of sintered piezoelectric and electrostrictive ceramics were examined using the X-ray source from CuK_{α} (Siemens D500) where the scan condition of 2-theta = 20-60°, a step-scan at 0.03° and with 1 second dwell time. To determine the stress, only the surface of piezoelectric layer in all PE-MBLC samples was exposed to the x-ray. The close-up of the 2-theta of 44-46° was collected to monitor the intensities of (002) and (200) peaks by using a step-scan of 0.01° and 2 seconds dwell time.

To investigate the domain alignments of piezoelectric ceramic and PE-MBLC actuators after poling, all samples were electroded with Au by sputtering method. The XRD peak profile of Au did not appear in the 2-theta of interest. Then, the electroded piezoelectric ceramic were poled at 60 °C for 5 minutes using a 30 kV/cm poling field. The electroded PE-MBLC samples were poled at 25 kV/cm, 10 minutes in a heated silicone oil bath of 150 °C, then cooled temperature down to 25 °C under the applied electric field for 40 minutes and continued poling at 25 °C for 30 minutes. All samples were aged for 1 day prior to observe the (002) and (200) peaks. The intensities of (002) and (200) peaks were obtained, which were then used to analyze the domain switching in the piezoelectric layer.

7.3 Results and Discussions

7.3.1 XRD results of sintered piezoelectric and electrostrictive ceramics

The XRD results of piezoelectric and electrostrictive ceramics sintered at 1200 ° are depicted in Fig 7.3. Both ceramics consisted of PMN-PT perovskite together with pyrochlore and MgO phases. As known, it is difficult to obtain pure perovskite phase in these materials. Many attempts have been conducted to eliminate pyrochlore phase through a prolonged calcination step, using excess MgO and PbO during synthesis of the powders, or pre-calcination of MgO and Nb₂O₅ oxides to form the columbite structure [137]. Therefore, it was possible to observe the MgO phase in the sintered PMN-PT ceramics.

From the XRD patterns in Fig 7.3, it was found that the sintered electrostrictive ceramic was matched with the JCPDS card number 97-015-3901, a rhombohedral structure with the Pb(Mg_{0.303}Nb $_{0.607}$ Ti_{0.09})O₃ composition. The piezoelectric ceramic was matched with the JCPDS card number 97-015-5884 which represented the composition of Pb(Mg_{0.123}Nb $_{0.427}$ Ti_{0.36})O₃ and belonged to tetragonal structure. Therefore, the intensities of (002) and (200) peaks were used to determine the induced stress on the surface of the sample after sintering process as well as the domain alignment after poling.



Figure 7.3 XRD pattern of a) PMN/PT 65/35 and b) PMN/PT 90/10 sintered at 1200°C.

7.3.2 The state of induced stress on the surface of piezoelectric layer of PE-MBLC composites

In general, the heat-treatment processes such as curing of different types of materials (ceramic/metal) or co-sintering of different ceramic compositions results in the development of residual stress accompanied by a shape change (curve or dome) when the samples are cooled. The shape changes originate from the difference between thermal expansion coefficients and shrinkages of materials. Such behavior was observed in the PE-MBLCs because of ~ $4x \ 10^{-6} \ C^{-1}$ and ~ 0.12 % thermal expansion coefficient and shrinkage mismatches, respectively. As a consequence, different dome heights as well as induced stresses could result due to the volume ratios of piezoelectric to electrostrictive materials in the composites. In this experiment, the stress developed on the outer surface of piezoelectric layer of PE-MBLC samples was examined. Since X-ray source could only penetrate 60-80 µm [138] into the sample surface, the state of stress observed was roughly a surface characteristic.

Figure 7.4 shows the intensity ratio of (200) and (002) of sintered piezoelectric and three different PE-MBLCs with P:E-8:2, 5:5 and 2:8. The $I_{(200)}/I_{(002)}$ of single piezoelectric ceramic was found to be 2. This indicated that this ceramic was in stressfree state and a- and c-domains were randomly aligned. In case of PE-MBLC (P:E-8:2), $I_{(200)}/I_{(002)}$ was equal to 2.25 indicating that the piezoelectric surface was under a tensile stress (compared to Fig. 7.1). Whereas $I_{(200)}/I_{(002)}$ of PE-MBLC 5:5 and 2:8 were 1.76 and 0.85, respectively, and thus they were under compressive stresses (compared to Fig. 7.1). The state of induced stress developed in the PE-MBLC samples was influenced by the volume ratios of piezoelectric to electrostrictive materials. The induced stress-shape change relationship of the two materials schematically shows in Fig 7.5. Figure 7.5 a represents the P and E layer according to volume ratios in all three PE-MBLC samples. Figure 7.5 b shows that piezoelectric ceramic had shrunk more than the electrostrictive ceramic when piezoelectric and electrostrictive ceramics were sintered separately. Due to the co-sintering of these two layers, piezoelectric layer was expanded and was under the tension force to match the interface of the electrostrictive layer which was in compressive force as shown in Fig. 7.5 c. After the composites were cooling down to room temperature, all composites were in dome shape with the piezoelectric layers were in the concave while the electrostrictive layer was in the convex as depicted in Fig 7.5 d.



Figure 7.4 X-ray diffraction patterns showing the intensity ratio $I_{(200)}/I_{(002)}$ of sintered PMN-PT 65/35, PE-MBLC 8:2, PE-MBLC 5:5 and PE-MBLC 2:8.



b) Sintering of piezoelectric and electrostrictive ceramics without P/E interfaces



Figure 7.5 Schematic representation of the state of the stress to the change of the dimension of PE-MBLCs P:E 8:2, 5:5 and 2:8 a) showing the P and E layers b) showing the shrinkage of P and E ceramics without interface c) sintering of PE-MBLC with P/E interfaces d) shape change after cooling

The neutral axis (stress =0) of PE-MBLC samples with respect to the radius of curvature and the dome height of the composites (Figure 7.6) were calculated form the equation 5.1 and 5.2 [139].



Figure 7.6 Schematic representative of the location of P/E interface and neutral axis of PE-MBLC P:E 8:2.

$$R_n = \frac{t}{\ln\left(\frac{R_0}{R}\right)}$$
(5.1)

$$\mathbf{Y} = \mathbf{R}_{\mathbf{n}} - \mathbf{R} \tag{5.2}$$

Where R, R_o and R_n are radii of inner surface, outer surface and neutral axis, respectively. Also t, a and b are the sample thickness, dome height and radius of the dome, respectively. Y is the distance from the bottom surface of PE-MBLC to the neutral axis.

It was found that the neutral axis (Y) of PE-MBLC P:E 8:2, 5:5 and 2:8 were located at 0.4980, 0.4986 and 0.4996 mm, respectively. So, the areas below and above neutral axis were subjected to compressive and tensile stress, respectively. In the mechanical point of view, the outer surface of piezoelectric layer was subjected to

compressive force and the outer surface of electrostrictive layer was in the tensile force for all three composites (Fig 7.5 d).

The discrepancy of the state of the stress developed in piezoelectric layer of PE-MBLC (P:E 8:2) by XRD method and structural perspective may be due to:

- a large beam width of the XRD exposed on the curved surface which could affect to the intensities of (200) and (002).

- the tilt of the XRD plane on the curved surface of PE-MBLC (P:E 8:2) (0.4-0.5 mm dome height).

Figure 7.7 a and b show the domains with their c-axis perpendicular and parallel to the XRD plane on the flat surface sample which gave to the reflected peaks of (002) and (200), respectively. In figure 7.7 c, the XRD plane was not perpendicular or parallel to the axis of the domains owing to the curved surface. This may affect to intensities of (200) and (002) peaks. This phenomenon would not take place in the case of flat surface samples (piezoelectric ceramic) or could be found less affect in the PE-MBLC 5:5 and 2:8 owing to their small dome height (0.1-0.2 mm).



Figure. 7.7 A schematic of the domain orientations and their XRD planes a) c-domains (c-axis perpendicular to XRD plane) b) a-domains (c-axis parallel to XRD plane) and c) tilted of XRD plane due to the curved surface.

7.3.3 Domain switching in piezoelectric ceramic and PE-MBLC

As mentioned, the populations of a- and c-domains could be determined by the intensities of (200) and (002) diffraction peaks, respectively. Therefore, an investigation of the domain switching in piezoelectric layer of PE-MBLC corresponding to the change of $I_{(200)}$ and $I_{(002)}$ before and after poling was carried out. The intensities of $I_{(200)}/I_{(002)}$ of piezoelectric ceramic and PE-MBLCs with P:E-8:2, 5:5, 2:8 before and after poling are presented in Table 7.1. The results showed that $I_{(200)}/I_{(002)}$ of piezoelectric ceramic was equal to 2 indicating that the domains of piezoelectric ceramic before poling were randomly aligned. After poling, an application of the electric field switched the adomains to c-domains resulting in the decrease of $I_{(200)}$ and the increase of $I_{(002)}$. A $I_{(200)}/I_{(002)}$ ratio equal to 0.5 after polarization indicated that complete domain switching had taken place in the piezoelectric ceramic, as shown in Fig 7.8 a (unpoled and poled piezoelectric ceramic). Similar to piezoelectric ceramic, poled PE-MBLC (P:E-8:2) showed a decrease in $I_{(200)}$ and an increase in $I_{(002)}$. Nevertheless, observed $I_{(200)}/I_{(002)}$ ratio of poled PE-MBLC (P:E-8:2) was found to be 0.61 (Table. 7.1). This pointed out that most of the a-domain populations were switched to c-domains after polarization. Lower switching of a-domains to c-domains was observed in poled PE-MBLC with P:E 5:5. In Fig 7.8 b, $I_{(002)}$ did not enhance corresponding to a small decrease of $I_{(200)}$ for PE-MBLC (P:E-5:5). In the case of unpoled PE-MBLC (P:E-2:8), low $I_{(200)}$ and $I_{(002)}$ were observed to begin with. This was attributed to the possibility that the piezoelectric and electrostrictive phases were mixed during pressing as well as the diffusion of the two materials into each other during sintering in this composite as mentioned in chapter 6.

Table 7.1 The ratios of $I_{(200)}/I_{(002)}$ (before and after poling) and the difference values of calculated and measured d_{33}^{eff} of piezoelectric and PE-MBLC ceramics with P:E 8:2, 5:5 and 2:8.

Sample	I ₍₂₀₀₎ /I ₍₀₀₂₎ before poling	$I_{(200)}/I_{(002)}$ after poling	% difference on calculated and measured d ₃₃ ^{eff}
Single piezoelectric ceramic	2	0.5	0
PE-MBLC (P:E-8:2)	2.25	0.61	6
PE-MBLC (P:E-5:5)	1.76	1.35	39
PE-MBLC (P:E-2:8)	0.85	1.07	60



Figure 7.8 Comparison of $I_{(200)}/I_{(002)}$ intensity ratio between PMN/PT 65-35 and a) PE-MBLC with P:E a) 8:2 b) 5:5 c) 2:8.

Therefore, the observed $I_{(200)}$ and $I_{(002)}$ peaks were not significantly different from those observed in the tetragonal piezoelectric ceramic. These observed peaks might be the combination of the tetragonal and rhombohedral phases owing to the mixed phases of the electrostrictive to the piezoelectric phases. Additionally, it was found that $I_{(200)}/I_{(002)}$ were not much different before and after poling, as depicted in Fig 7.8 c and Table 7.1. One could deduce that some a-domains had switched to c-domains in this composite. Considering the % difference between the calculated and measured d_{33}^{eff} , (($\Delta d_{33}^{\text{eff}}/d_{33}^{\text{eff}}$) x 100) in PE-MBLC (P:E-2:8) was found to be ~60%, the highest value in Table 7.1. Therefore, PE-MBLC (P:E-2:8) was not effectively poled. From this experiment, one could conclude that lower domain switching in actuators such as P:E-5:5 and 2:8 was attributed to the presence of higher volumes of the electrostrictive phase.

Throughout the study in chapter 6 and 7, it could conclude that three possible factors play important roles in the displacement enhancement of PE-MBLCs. Those factors were related to 1) the transverse strain response of piezoelectric and electrostrictive layers in the PE-MBLCs, 2) the geometry (almost flat or dome) of the composites, and 3) the ratio of $I_{(200)}/I_{(002)}$ after polarization. The first factor was discussed in details in chapter 6. It was shown that the PE-MBLC (P:E-5:5) yielded the highest displacement due to its optimum transverse strain response in comparison with the other PE-MBLC actuators having a dominant either piezoelectric or electrostrictive phase. As for the second factor, it is shown that the stiffness of a dome shaped structure increases with the increase of the dome height [140]. Therefore, a less rigid dome composite could offer a larger E-field induced displacement. As seen, PE-MBLC (P:E-5:5) (~0.2 mm dome height) exhibited higher axial displacement than that of PE-MBLC (P:E-8:2) (~

0.45 mm dome height). Nevertheless, PE-MBLC (P:E-2:8) which was almost flat (~ 0.1 mm dome height), still its displacement was low (See Fig. 6.6). As discussed before, the displacement of PE-MBLC actuators was the result of transformation of the transverse contraction of piezoelectric and electrostrictive layers into an axial motion. Since the electrostrictive was the major phase in PE-MBLC (P:E-2:8), it governed the transverse contraction of this composite giving rise to the lowest displacement observed. The last factor which was the reason to obtain the highest displacement in PE-MBLC (P:E-5:5) was originated from the domain reorientation. The existence of a-domains in a piezoelectric actuator after polarization has a significant effect on the improvement of its displacement. As reported, $I_{(200)}/I_{(002)}$ ratio of PE-MBLC (P:E-5:5) was the largest value, as shown in Table 7.1. This implied that a large population of a-domain still existed in PE-MBLC (P:E-5:5) and could switch to c-domain under a bias field. This mechanism has also been observed in Rainbow actuators [15].

7.4 Conclusions

In this chapter, X-ray technique was used to identify the state of induced stress developed from the thermal expansion mismatch and shrinkage difference of the piezoelectric and electrostrictive ceramics in the PE-MBLC composites. This technique could only identify the induced stress on the surface of a sample due to limited penetration of the X-ray source. Furthermore, the $I_{(200)}/I_{(002)}$ after poling of PE-MBLC composites suggested that domain switching of piezoelectric layer was incomplete. Finally, with this technique, the $I_{(200)}/I_{(002)}$ ratio after polarization of PE-MBLC actuators could explained the effect of domain reorientations on the enhancement of displacement in PE-MBLC actuators.

Chapter 8

Bi-Layer Piezoelectric/Electrostrictive (P/E) Dome Unimorph

8.1 Introduction

In this chapter, a new flextensional actuator design named Bi-Layer Piezoelectric/Electrostrictive Dome Unimorph (BIPEDU) is introduced. The BIPEDU actuator was fabricated by attaching a piezoelectric-electrostrictive monolithic bi-layer composites (PE-MBLC) actuator to a metallic plate using an epoxy resin. The PE-MBLCs with three different volume ratios of piezoelectric to electrostrictive (P:E) of 6:4, 7:3 and 8:2 were fabricated by co-pressing and co-sintering of piezoelectric and electrostrictive powders. The electromechanical properties as well as actuation performances were evaluated for both PE-MBLC and BIPEDU actuators.

8.2 Experimental procedure

8.2.1 Fabrication of piezoelectric-electrostrictive monolithic bi-layer composites (PE-MBLC)

The fabrication of PE-MBLC samples were described in section 5.2.2 in details. The PE-MBLCs (P:E-6:4, 7:3 and 8:2) after sintering exhibited dome-shaped structure with the dome height (Figure 8.1 a and Table 8.1) of 0.3-0.45 mm. The concave side (piezoelectric layer) of PE-MBLC was then lightly polished with SiC sandpaper (1200 grit) to create a 1 mm wide flat rim where the metal plate was attached. The top and bottom surfaces of PE-MBLC samples were electroded with fire-on silver paste and heat treated at 550 $^{\circ}$ C for 15 minutes. Then, the samples were poled under 25 kV/cm electric

field strength in a heated silicone oil bath for 10 minutes at 150 °C, followed by cooling down to 25 °C during 40 minutes period under the same electric field applied and further poled at 25 °C for 30 minutes.

8.2.2 Fabrication and characterizations of bi-layer piezoelectric/electrostrictive dome unimorph (BIPEDU)

A flat metallic plates (76 μ m thick stainless steel), with identical diameter (~12.6 cm) to the PE-MBLC actuators, were punched out by a punching tool. The PE-MBLC and metallic plate were bonded together by an insulating epoxy resin applied onto the polished circumferential area. Then, they were aligned in a fixture and placed into an oven to cure the epoxy resin at 65 °C for 3 hours. The insulating epoxy resin (Eccobond 15 LV) and catalyst (Catalyst 15 LV) from Emerson and Cuming Inc. were used as the bonding layer in this study. The viscosity of epoxy resin was high enough to prevent flowing out or filling the cavity between the metal and PE-MBLC. In this study, the Eccobond/Catalyst mixing ratio of 3/1 (by weight) was used to obtain a bonding layer with moderate stiffness. In addition, its low curing temperature (25-70 °C) could prevent cracking of the ceramic due to thermal expansion mismatch between the ceramic and the metal. The thickness of the cured epoxy was about 20 μ m. Figure 8.1.a schematically shows the BIPEDU actuators.



Figure 8.1. Schematic representation of bi-layer piezoelectric/electrostrictive dome unimorph (BIPEDU) (a) no electric-field applied and (b) Under electric field.

8.3 Characterization techniques

- The effective coupling coefficients and the effective relative permittivities (K^{eff}) were measured and calculated as described in section 4.3.2. The effective piezoelectric charge coefficients (d_{33}^{eff}) were measured and calculated as described in section 5.4.1.

- The unipolar axial displacements of PE-MBLC and BIPEDU actuators were measured by an MTI-2000 photonic sensor in conjunction with a 10 kV (DC) power supply (Trek 610C) and a special displacement fixture. The applied electric field was parallel to the poling direction in all measurements. The concave side (piezoelectric layer) of PE-MBLC and the metallic plate of BIPEDU actuators were placed on a planar metallic support between the two copper electrodes as shown in Fig. 8.2 a and b, respectively.

- For the displacement-load measurements, the samples were placed between the two copper electrodes which were the same setup as the uni-polar displacement mentioned above. Additionally, the external load cells from brass and/or steel (up to 11 N with 1 N interval) were applied on top of the upper copper electrode. The contacted area of the load cells was larger than that of the top copper electrode. Then, the fiber optic probe was placed on the load cells which has a reflective paper on. The axial displacements as a function of load applied were measured under the application of an electric field of 10 kV/cm to the actuators. The displacement-load setups of PE-MBLC and BIPEDU actuators are shown in Fig. 8.3 a and b, respectively.



Figure 8.2. Schematic representation of displacement setup for (a) PE-MBLC (b) BIPEDU.



Figure 8.3. Schematic representation of displacement-load setup for (a) PE-MBLC (b) BIPEDU.

8.4 Results and discussions

8.4.1 The electromechanical properties of PE-MBLC actuator

Table 8.1 shows the dielectric and electromechanical properties of piezoelectric, electrostrictive, and PE-MBLC ceramics at three different volume ratios of piezoelectric to electrostrictive (P:E-6:4, 7:3, 8:2). The results were average of three samples. It was found that the effective piezoelectric charge coefficient (d_{33}^{eff}) values increased while the effective relative dielectric constant (K^{eff}) values decreased as the piezoelectric volume percent in PE-MBLC increased. The measured d₃₃^{eff} and K^{eff} values were slightly lower than that of the calculated values for all piezoelectric volume ratios. The difference between the calculated and measured d_{33}^{eff} was more pronounced in the PE-MBLC with P:E of 6:4 (33 %) while it was in the range of 10 % and 8 % for P:E 7:3 and P:E 8:2, respectively. In addition, the calculated and the measured K^{eff} differences in all three ratios felt in range of 25-30%. This was due to the dielectric displacements (D) of the two materials which were not equal along the thickness direction and hence affected domain switching in the piezoelectric layer during poling, as explained in chapter 5. As a result, the measured d_{33}^{eff} and K^{eff} values were slightly lower than the calculated ones. The electromechanical coupling coefficients of piezoelectric (PMN-PT 65/35), electrostrictive (PMN-PT 90/10) and PE-MBLC actuators are also displayed in Table 8.1. It was found that the effective coupling coefficients decreased by decreasing the piezoelectric volume ratio in the composites. This was attributed to the fact that the piezoelectric layer in PE-MBLC was clamped by the electrostrictive layer and could not freely vibrate. As a result of lower elastic deformation of piezoelectric layer, the PE-MBLCs had lower coupling coefficients.
Designation	Dome height (mm.)	Calculated d ₃₃ ^{eff} (pC/N)	Measured d ₃₃ ^{eff} (pC/N)	Calculated K ^{eff}	Measured K ^{eff}	tan δ (%)	k _p eff (%)	k _t eff (%)	k ₃₃ ^{eff} (%)	k ₃₁ ^{eff} (%)
Piezoelectric (PMN-PT 65/35)	-	-	630 ±10	-	4000 ±150	2.0	62	59	77	40
Electrostrictive (PMN-PT 90/10)	-	-	-	-	20000 ±150	1.9	-	-	-	-
PE-MBLC 6:4	0.3±0.02	556	370 ±20	5882	4370 ±150	2.26	35	39	50	15
PE-MBLC 7:3	0.4±0.006	580	520 ±25	5263	3715 ±70	1.93	52	47	66	27

4761

550 ±10

600

PE-MBLC 8:2

0.46±0.038

3370 ±100

57

1.72

49

70

32

Table 8.1. Comparison of dome height, dielectric and electromechanical properties of piezoelectric, electrostrictive, and PE-MBLC at P:E 6:4, 7:3, and 8:2.

8.4.2 The effect of bonding area on the displacement of BIPEDU

As mentioned in the experimental procedure, the epoxy resin was used as the bonding layer between the composite and the metallic plate. The uniform bonding was required ensuring that the induced force due to the radial contraction of composites could be transferred to the metallic plate. Therefore, the effect of bonding area on the displacement of BIPEDU actuators was examined. Two BIPEDU actuators, with and without uniform distribution of epoxy, were prepared. In the first actuator, referred to as uniformly and completely bonded BIPEDU, the epoxy resin was applied onto the whole area of polished flat rim. In the second actuator, called partially bonded BIPEDU actuator, the epoxy resin was applied only onto opposite four small areas (1-mm width and 4-mm length each area) of the polished flat rim (1mm-width and ~40-mm circumference) shown in Fig. 8.3. Then the displacements of these actuators were measured under 10 kV/cm of applied E-field. Figure 8.3 shows the displacements of PE-MBLC, completely and partially bonded BIPEDU actuators fabricated from PE-MBLC (PE : 82) with 76 µm thick stainless steel. The result showed that the PE-MBLC generated 6 µm displacement. As expected, higher displacement about 65 µm was obtained from the completely uniform bonded BIPEDU actuator and a decrease of the displacement to 40 µm yielded from partially bonded BIPEDU actuator. The results could be explained by the fact that a complete bonding area could more effectively transfer the force generated by the radial contraction of PE-MBLC actuator to the metallic plate than partially bonded BIPEDU. This study indicated that the bonding area between the PE-MBLC and the metallic plate had a notable effect on the displacement of BIPEDU actuators.



Figure 8.4. Comparison displacement of the piezopelectric-electrostrictive monolithic bilayer composites (PE-MBLC) and bi-layer piezoelectric/electrostrictive dome unimorph (BIPEDU) at P:E 8:2 with complete and partial bonding areas.

Table 8.2 compares the field-induced displacements of PE-MBLC and BIPEDU with P:E ratios of 6:4, 7:3, 8:2. All actuators were driven under 10 kV/cm electric field. It was found that the average displacements of PE-MBLCs with three different piezoelectric and electrostrictive volume ratios were in range of 5-10 μ m. The PE-MBLC actuators with higher piezoelectric volume yielded smaller displacement. As discussed in chapter 6, the contribution of transverse displacement of PE-MBLC with P:E of 6:4 was more pronounced and thus provided a higher axial displacement. The maximum piezoelectric/electrostrictive P:E 5:5 yielded the highest displacement about 15 µm. However, this P:E volume ratio could not produce a dome shape ceramic, an essential shape factor for BIPEDU design. After displacement evaluation of PE-MBLCs, the very same samples were used to fabricate BIPEDUs by attaching the PE-MBLC to a metallic plate by using epoxy resin. As shown in Table 8.2 the displacements were remarkably amplified by a factor of 7-12 for all BIPEDU actuators. The displacement of the PE-MBLCs (P:E 6:4) was 10 µm while the displacement of its BIPEDU actuator was 73 µm. Similar trends were observed for other PE-MBLC and BIPEDU actuators. The improvement of actuation performance could be attributed to the flextensional structure of the metallic plate. The force transferred through the bonding area from the ceramic to the metallic plate resulted in the contraction of metallic sheet in radial direction and caused it to deform downward, generating more displacement in BIPEDU actuators. The schematic displacement mechanism of the BIPEDU under an application of the E-field is shown in Fig. 8.1.b.

Additionally, the effect of E-field on the displacement of BIPEDU actuators was examined. Figure 8.4 depicts the displacement of BIPEDUs fabricated from PE-MBLCs with P;E - 6:4, 7:3, 8:2. The E-field ranged from 2 to 10 kV/cm with 2 kV/cm intervals. It was found that the displacements were increased by increasing the applied E-field.

Designation of P:E volume ratio	Displacement (µm) PE-MBLC	Displacement (µm) BIPEDU		
6:4	10.38±0.48	73±1.98		
7:3	8±0.4	69±4.75		
8:2	5.5±0.28	65±3.89		

Table 8.2. The displacement of PE-MBLC and BIPEDU of P:E 6:4, 7:3, and 8:2



Figure 8.5. The displacement of as a function of applied E-field of bi-layer piezoelectric/electrostrictive dome unimorph (BIPEDU) fabricated from PE-MBLC P:E-6:4, 7:3 and 8:2 attached with 76 µm thick stainless steel.

8.4.4 The displacement-load behavior of PE-MBLC and BIPEDU actuators

The displacements of PE-MBLC and BIPEDU actuators with various piezoelectric volume ratios were investigated under various loads (up to 11 N). Figures 8.6 a and b show the displacement vs. load of PE-MBLC and BIPEDU actuators under 10 kV/cm E-field. In Fig 8.5.a, a small fluctuation and slight decrease in displacement (~1 μm) was observed in the PE-MBLC actuators. On the other hand, the displacements of all BIPEDU actuators sharply decreased by increasing the applied load <5 N, as shown in Fig 8.5.b. Further increase of load gradually reduced the displacement. The difference between the displacement-load behaviors of PE-MBLCs and BIPEDUs could be explained by the fact that PE-MBLC actuators were able to freely contract in the transverse direction under the applied load limit used in this work. In BIPEDU actuator, the PE-MBLC was attached to the metallic plate, which could deform and generate more displacement under no load as discussed earlier. However, when loads were applied on the BIPEDU, their metallic plates were pressed by the external loads diminishing their deformation. As a result, lower displacement in BIPEDU actuators were obtained by increasing external loads.



Figure 8.6. The displacement-load behavior of (a) the piezopelectric-electrostrictive monolithic bi-layer composites (PE-MBLC) and (b) bi-layer piezoelectric/electrostrictive dome unimorph (BIPEDU). The ratios of P:E in PE-MBLC and BIPEDU were 6:4, 7:3 and 8:2.

8.4.5 The effect of metallic plate thickness on the displacement of BIPEDU actuators

In this study, the BIPEDU actuators were fabricated by attaching a PE-MBLC (P:E-8:2) to a flat stainless steel sheets with various thicknesses (50, 60, 76, 100, 125 and 177µm). Prior to attach the circular stainless steel to the PE-MBLC samples, the displacements of all PE-MBLC samples were evaluated under 10 kV/cm uni-polar applied E-field. The observed displacements in all PE-MBLC samples were consistence with the displacement values of $5 \pm 0.3 \,\mu\text{m}$. Then, the PE-MBLC samples were attached to the stainless steel sheets with different thicknesses. Figure 8.7 shows the displacement results of the BIPEDU actuators as a function of the stainless steel thicknesses. As seen, the displacements were increased from 52 to 65 µm as the stainless steel thickness increased from 50 to 76 μ m. Then the displacements decreased to 36, 15 and 5 μ m when thicker stainless steels of 100, 125 and 177 µm were used, respectively. It was observed that thin stainless steel sheets (50 and 60 µm) were very soft and flexible and could not uniformly deform when the PE-MBLC was radially contracted under applied E-field. Whereas thicker stainless steel sheets (100, 125 and 177 µm) were found to become stiffer and thus the induced force from contraction of the PE-MBLC under E-field excitation was not large enough to effectively deform them. From this study, the optimum stainless steel thickness was about 76 µm for the BIPEDU actuator made of PE-MBLC (P;E 8:2).



Figure 8.7. The displacement as a function of the thickness of stainless steel (50, 60, 76, 100, 125 and 177 μ m) of bi-layer piezoelectric/electrostrictive dome unimorph (BIPEDU) of P:E 8:2.

8.5 Conclusions

A new type of actuator named bi-layer piezoelectric/electrostrictive dome unimorph (BIPEDU) was fabricated by attaching the piezoelectric-electrostrictive monolithic bi-layer composites (PE-MBLC) to metallic plates. It was shown that completely bonded BIPEDU yielded higher displacement than partially bonded BIPEDU. This was attributed to the fact that a complete bond area could more effectively transfer the induced force generated by the contraction of PE-MBLC to the metallic plate. The BIPEDU actuators with 76 µm thick stainless steel yielded 7-12 times higher strain amplification than PE-MBLC actuators. The displacement-load behaviors were found to be different in PE-MBLC and BIPEDU actuators. The displacement of the PE-MBLC actuators was independent of the load, while for the BIPEDU actuators, the displacement was decreased with applied load.

Chapter 9

Piezoelectric-Electrostrictive Monolithic Bi-Layer Composite Flextensional Actuator

9.1 Introduction

Moonie and cymbal are categorized as class-V flextensional actuators which are composed of a ceramic disk sandwiched between two metallic endcaps. The induced force from the lateral contraction and longitudinal expansion of the ceramic disk is transformed into the longitudinal displacement of metallic endcaps, in which the displacement amplification is achieved. Several studies have been conducted to improve the actuation of the cymbal. Important parameters affecting the strain behavior of the cymbal are metallic endcaps (materials and dimensions), type of epoxy used for bonding the ceramic to the metal, and type of ceramic element [23].

Various types of ceramics and single crystals have been used as driving elements including lead-based systems such as soft PZT, hard PZT, relaxor ferroelectric PMN-PT, antiferroelectric-to-ferroelectric phase switching ceramic PLSnZT, PZN-PT single crystal [21;23] and lead-free systems such as Barium Strontium Titanate (BST) [141], and $(Bi_{1/2}N_{1/2})TiO_3$ - $(Bi_{1/2}K_{1/2})TiO_3$ -BaTiO_3 (BNT-BKT-BT) [22]. Cymbal actuators fabricated by such active elements have resulted in remarkable strain amplifications. Furthermore, their electric field induced displacements (u_{33}) have exhibited symmetry in regard to the polarity of applied E-field, i.e. $|u(-E_{max})| = |u(E_{max})|$. In this study, the piezoelectric-electrostrictive monolithic bi-layer composite (PE-MBLC) comprised of 50 volume percent of piezoelectric and electrostrictor each was incorporated in cymbal as

shown in Fig. 9.1. The flat composite bi-layer element was obtained by co-pressing and co-sintering the piezoelectric and the electrostrictive ceramics. The purposes of using PE-MBLC as driving element were to study its role in strain amplification, asymmetric (directional) displacement $|u(-E_{max})| \neq |u(E_{max})|$, and hysteretic behavior of displacement under applied field. Thus, this study compares the electromechanical properties of cymbals with various types of driving elements including single piezoelectric, single electrostrictive ceramics, and flat PE-MBLC (P:E-5:5). First, single ceramics and PE-MBLC composite were fabricated and their electromechanical properties and displacement were evaluated. Afterwards, the cymbals were assembled by bonding driving elements to brass endcaps using an insulating epoxy resin. Next, their strain-electric field responses under uni-polar and bi-polar conditions were examined.

9.2 Experimental procedure

9.2.1 Fabrication of ceramic driving element

Lead magnesium niobate-lead titanate (PMN-PT) powders with the compositions of piezoelectric 0.65PMN-0.35PT (P) and electrostrictive 0.9PMN-0.1PT (E) were used. The single ceramics and PE-MBLC with 50 % volume of piezoelectric material (P:E-5:5) were formed by the pressing method. The pressing pressure at 86 MPa was applied to all three types of ceramics. The binder was removed at 550 °C, with a soak time of 3 hours. The sintering process performed at 1150 °C with 1 hour dwell time. Under such processing conditions, flat PE-MBLC sample were obtained. To obtain flat and parallel surfaces, single piezoelectric and electrostrictive ceramics were polished with 1200 grit SiC sand paper. The electrodes were formed by applying silver paste on top and bottom surfaces of samples and followed by heat-treatment at 550 °C for 15 minutes. The electroded PE-MBLC samples were poled at 25 kV/cm electric field strength in a heated silicone oil bath for 10 minutes at 150 °C, then cooled down to 25 °C under the applied electric field for 40 minutes and further poled at 25 °C for 30 minutes. On the other hand, piezoelectric samples were poled at 60 °C for 5 minutes using a 30 kV/cm poling field. The electromechanical properties and displacements of all samples were measured before and after fabrication of the cymbals.

9.2.2 Fabrication of flextensional actuator (Cymbal)

In order to form Cymbal-type flextensional actuators, endcaps were prepared by die punching thin sheets of brass. The dimensions of endcaps were 0.25 mm cavity depth (d_c), 9 mm cavity diameter (Φ_c), 0.25 mm thickness (t_m), and 3 mm apex diameter (Φ_A) shown in Fig. 9.1. Brass was chosen in this study because the cymbal fabricated from brass (modulus of elasticity ~110 GPa) could yield higher displacement compared to low carbon-steel and tungsten endcaps (high modulus of elasticity) [23]. The electrostrictive, poled piezoelectric and poled PE-MBLC elements with 12.5 mm diameter (Φ) and 1 mm thickness (t) were bonded to the brass endcaps by Eccobond epoxy (three parts by weight of Eccobond 45 LV epoxy resin and one part by weight of Catalyst 15 LV resin), and then clamped in a fixture and heat-treated at 65 °C for 3 hours to cure and harden the epoxy. The advantages of the Eccobond epoxy used in this study were explained in section 8.2.2.



Figure 9.1. Schematic representative of cymbal actuator using a piezoelectricelectrostrictive monolithic bi-layer composite driving element.

9.3 Characterization techniques

9.3.1 Evaluation of ceramics and cymbals

- The interface and microstructure of sintered PE-MBLC samples were examined by field emission scanning electron microscopy (FESEM, Leo-Zeiss 982, Carl Zeiss Microimaging, Inc., Thornwood, NY).

- The piezoelectric charge coefficients (d_{33}), relative permittivity (K) and electromechanical coupling coefficients (k_p , k_t , k_{33} and k_{31}) were measured as described in section 4.3.2. The calculated piezoelectric charge coefficient (d_{33}^{eff}) and calculated relative permittivity (K_{33}^{eff}) were shown in section 5.4.1 and 6.2.3, respectively.

- The axial displacements of the uncapped and capped (cymbal) samples were measured by an MTI-2000 photonic sensor in conjunction with a 10 kV DC power supply. Measurements were carried out under both DC unipolar and bipolar modes. The displacement setup of uncapped composite and cymbal actuators are depicted in Fig. 9.2 a and b, respectively. The polarizations of single piezoelectric ceramic and piezoelectric layer of PE-MBLC were kept in the same direction of the applied electric field in all measurements.

- The degree of hysteresis (H) (Fig. 9.3) or the mechanical losses in actuator under uni-polar excitations was calculated by [3]

$$\% H = \left(\frac{\Delta X}{X_{\text{max}}}\right) \times 100 \tag{1}$$

where X_{max} is the axial displacement at maximum electric field (E_{max}) and ΔX is the width of the unipolar curve at 0.5E _{max} ($E_{max} = 10 \text{ kV/cm}$).

- The asymmetry (ξ) in u_{33} of actuators under bipolar excitations was determined using [142]

$$\xi = \begin{pmatrix} u_{33}^{+E_{\text{max}}} \\ u_{33}^{-E_{\text{max}}} \end{pmatrix}.$$
 (2)



Figure 9.2. Schematic representation of displacement setup for (a) PE-MBLC (b) PE-MBLC cymbal.



Figure 9.3 Schematic representative of degree of hysteresis (H).

9.4 Results and discussions

Scanning electron micrographs of a representative fracture surface in a sintered PE-MBLC is shown in Fig. 9.4. The interface was fully sintered with no delamination between the layers. The monolithic layers were obtained from solid state diffusion of piezoelectric and electrostrictive ceramics with possible composition variation along the P/E interfaces. A closer look revealed subtle differences between the electrostrictive and piezoelectric layers in the PE-MBLC (Figs. 9.4.b-c). The electrostrictive layer had denser microstructure with smaller and tightly packed corner-shaped grains (Fig. 9.4.b). On the other hand, the piezoelectric layer consisted of irregular-shaped grains. The relative densities of individually sintered electrostrictive and piezoelectric samples were found to be 98.2±0.3 % and 96.5±0.12 %, respectively. The average relative densities of individual single ceramics. Above all, the PE-MBLC samples remained flat after sintering at 1150 ^oC, which was crucial in the fabrication of flextensional actuators.

Table 9.1 shows the dielectric and electromechanical properties of piezoelectric, electrostrictive and PE-MBLC used in this study. The relative dielectric constant of PMN-PT 65/35 and PMN-PT 90/10 was found to be 4020 ± 130 and $18,670\pm200$, respectively. The calculated effective relative dielectric constant (K^{eff}) for the PE-MBLC was 6600, whereas the measured relative permittivity was found to be 5830 ± 150 (11 % deviation from the calculated value). The effective d_{33} of PE-MBLC was about 270 pC/N compared to the calculated d_{33}^{eff} (490 pC/N). The discrepancy (44 %) between the calculated and measured d_{33}^{eff} values was attributed to the induced stress at P/E interfaces after sintering and the permittivity difference between P/E layers. These caused

incomplete poling of the piezoelectric layer as discussed in chapter 5. Table 9.1 shows the electromechanical coupling coefficients of single piezoelectric ceramic and PE-MBLC. It was found that the electromechanical properties of PE-MBLC were substantially lower than that of a single phase piezoelectric. This was because the elastic deformation associated with the fundamental radial and thickness resonance modes in piezoelectric layer were clamped by the electrostrictive layer.



Figure 9.4. Scanning electron microscope fracture surface images of a) interface of the PE-MBLC, b) close-up view of the electrostrictive layer, and c) close-up of the piezoelectric layer.

Table 9.1. Comparison of dielectric and electromechanical properties of PMN-PT 65/35, PMN-PT 90/10 to piezoelectric-electrostrictive monolithic bi-layer composites (PE-MBLC)

Material	d ₃₃ (pC/N)	К	tan δ (%)	k _p (%)	k _t (%)	k ₃₃ (%)	k ₃₁ (%)
Piezoelectric PMN-PT 65/35	600 ±10	4020 ±130	2.2	61	58	76	39
Electrostrictive PMN-PT 90/10	-	18670 ±200	1.8	-	-	-	-
PE-MBLC 5:5	270 ±40	5830 ±150	2	24	22	32	14

Figure 9.5.a compares the axial displacement (x₃₃) of electrostrictive, piezoelectric and PE-MBLC under unipolar electric field. At 10 kV/cm, the maximum axial displacement of the PE-MBLC was significantly higher than those of the electrostrictor and piezoelectric ceramics. PE-MBLC resembled a monomorph and thus the difference between transverse strains of piezoelectric and electrostrictive layers created a bending movement that was transformed into an axial displacement. While the piezoelectric and electrostrictive ceramics only contracted and/or expanded in axial direction along their thickness. Therefore, they yielded lower axial displacement than PE-MBLC actuator. As mentioned, the mechanical losses could be estimated from the degree of hysteresis (H). The observed hysteretic loss of electrostrictive was negligibly small (~0%) while of piezoelectric was found to be 7 % (mainly caused by domain switching). For PE-MBLC, a H value of 4.6 % was obtained. The reason for a lower mechanical loss in PE-MBLC compared to its piezoelectric counterpart could be that the electrostrictive layer impeded domain reorientation in the piezoelectric layer of PE-MBLC.

Figure 9.5.b shows the unipolar electric field induced axial displacements (u_{33}) of capped (cymbal) electrostrictor, piezoelectric and PE-MBLC. At 10 kV/cm, the maximum axial displacements produced by the capped electrostrictor and piezoelectric were 9 and 20 µm, respectively, while this value for the capped PE-MBLC was 21 µm. As seen, capping increased the axial displacements of electrostrictor and piezoelectric ceramic by two orders of magnitude. On the other hand, capping only increased the axial displacement of PE-MBLC by a factor of two (from 11 to 21 µm). In a piezoelectric and a single electrostrictive ceramics, the induced force generated by their radial contraction under an applied E-field symmetrically moves end-caps (Cymbal) along the axial

direction, as shown in Fig. 9.6 a. While in case of PE-MBLC actuator, the induced force was generated by its bending movement as discussed above. Therefore, the induced force was un-symmetrically transferred to upper and lower endcaps. Consequently, this gave rise to un-symmetrical movements of endcaps along the axial direction resulting in a modest amplification in displacement, as shown in Fig. 9.6 b. The degree of hysteresis (H) of the capped piezoelectric, electrostrictive and PE-MBLC were 9, 10 and 11 %, respectively. The H values of all three capped actuators were larger than those of their corresponding uncapped ceramics. The mechanical losses in the capped ceramics were originated from the losses in the epoxy bonding layer and the mechanical losses in the end caps.



Figure 9.5. Unipolar field dependence of displacement of a) piezoelectric, electrostrictive and PE-MBLC 5:5, and b) capped (cymbal) actuators based on piezoelectric, electrostrictive and PE-MBLC 5:5



Figure 9.6. Deformation modes of a) single piezoelectric or single electrostrictive ceramic cymbal b) flextensional PE-MBLC actuator when E>0, and c) flextensional PE-MBLC actuator when E<0.

The bi-polar electric field induced axial displacements (x_{33}) of an piezoelectric and electrostrictive ceramics are shown in Fig. 9.7. a. The bi-polar displacement of piezoelectric ceramic showed a symmetrical butterfly loop while electrostrictor showed a symmetrical parabolic displacement loop with no hysteresis. Figure 9.7 b depicts the axial displacement response of the PE-MBLC under bipolar excitation. The maximum axial displacement at + E_{max} was 11.6 µm while the same for - E_{max} was 6.6 µm. Thus, the PE-MBLC exhibited an asymmetic displacement under bipolar excitation (with ξ =1.75). Fig. 9.7 b also suggests that PE-MBLC offers significantly smaller hysteresis displacement loop under bipolar excitation than a single piezoelectric ceramic.

The axial displacement-E field responses of capped piezoelectric, electrostrictive and PE-MBLC actuators under bipolar excitation are shown in Fig. 9.8. The maximum axial displacement of capped piezoelectric and electrostrictive actuators were 20 μ m and 8 μ m at $\pm E_{max}$ with symmetrical butterfly and parabolic shaped displacement loops, respectively $[u_{33}(-E_{max})=u_{33}(E_{max})]$. The maximum axial displacement at $+E_{max}$ for the PE-MBLC was 19 μ m while the same for $-E_{max}$ was 8 μ m, which resulted in an asymmetry factor of $\xi=2.4$. In other words, $u_{33}(-E_{max})\neq u_{33}(E_{max})$. Furthermore, a reduction in hysteresis was also found for capped PE-MBLC in comparison with the capped piezoelectric ceramic.



Figure 9.7. Bipolar field dependence of displacement of a) uncapped piezoelectric (PMN-PT 65/35) and electrostrictive (PMN-PT 90/10) and b) uncapped piezoelectric-electrostrictive monolithic bi-layer composite (PE-MBLC 5:5).



Figure 9.8. Bipolar field dependence of displacement in flextensional actuators with piezoelectric (PMN-PT 65/35), electrostrictive (PMN-PT 90/10), and PE-MBLC 5:5 driving elements.

In a PE-MBLC, a first order effect of piezoelectricity ($x_{ij} = d_{ijk}E_k$; i, j, k = 1, 2, 3) was superimposed on a second order effect of electrostriction ($x_{ij} = M_{ijkl}E_kE_l$; i, j, k, l = 1, 2, 3). For the configuration considered in this study (two co-fired disks), the axial displacement x_{33} of the PE-MBLC was governed by the differential strain Δx_{31} in the transverse direction under circular symmetry (Fig. 9.6 b-c). The transverse strains were defined in the piezoelectric (Eq. 3) and electrostrictive (Eq. 4) layers as follows:

$$x_{31}^{P} = -|d_{31}|E_{3}^{P} - |M_{12}^{P}|[E_{3}^{P}]^{2} , \qquad (3)$$

$$x_{31}^{E} = -\left|M_{12}^{E}\right| \left[E_{3}^{E}\right]^{2},$$
(4)

where x was the transverse strain, d was the piezoelectric charge coefficient, and M was the electrostriction coefficient in electric field notation. By subtracting Eqs 3 and 4, the following expression for the differential transverse strain ($\Delta x_{31} = x_{31}^P - x_{31}^E$) in a PE-MBLC could be obtained:

$$\Delta x_{31} = \begin{cases} \left| M_{12}^{E} \right| \left[E_{3}^{E} \right]^{2} - \left(\left| d_{31} \right| E_{3}^{P} + \left| M_{12}^{P} \right| \left[E_{3}^{P} \right]^{2} \right) \text{ if } E > 0 \\ \left(\left| d_{31} \right| E_{3}^{P} + \left| M_{12}^{E} \right| \left[E_{3}^{E} \right]^{2} \right) - \left| M_{12}^{P} \right| \left[E_{3}^{P} \right]^{2} \text{ if } E < 0 \end{cases}$$
(5)

where $d_{31}<0$ and $M_{12}<0$. According to Eq. 5, $\Delta x_{31} < 0$ when E>0, which results in an effective positive axial strain, i.e. $\tilde{x}_{33} > 0$. On the other hand, $\Delta x_{31} > 0$ when E<0, which results in an effective negative axial strain, i.e. $\tilde{x}_{33} < 0$. More importantly $|\tilde{x}_{33}^{E>0}| > |\tilde{x}_{33}^{E<0}|$, hence, $|\Delta x_{31}^{E>0}| > |\Delta x_{31}^{E<0}|$, which results in an asymmetry in the displacement vs. E response under bipolar excitation as reported in Fig. 9.7 b.

When the PE-MBLC was capped, its u_{33} -E response still remained asymmetric with respect to the polarity of the electric field (Fig. 9.8), i.e. $|u_{33}^{max}|_{-E_{max}} < |u_{33}^{max}|_{+E_{max}}$. However, $u_{33}>0$ for capped PE-MBLC actuators even when E<0 (Fig. 9.8), in contrast to the behavior of uncapped PE-MBLC actuator (shown in Fig. 9.7 b). When E>0 and parallel to the polarization direction of piezoelectric layer (Fig. 9.6.b), the effective transverse strain of PE-MBLC $\tilde{x}_{31} < 0$, or both layers were contracted. Then, the electrostrictive layer was in radial tension and the piezoelectric layer was in radial compression (Fig. 9.6.b). As a result of this elastic deformations, the upper endcap deformed inward while the bottom one deformed outwards, ultimately producing $u_{33}>0$ for E>0. Oppositely applied E-field resulted in the radial contraction of electrostrictive layer while the piezoelectric layer radially expanded, which resulted in a positive effective transverse strain ($\tilde{x}_{31} > 0$) (Fig. 9.6.c). The $\tilde{x}_{31} > 0$ induced a concave (upward) shape in the PE-MBLC, which in turn, put the upper endcap in compression and the lower one in tension. This still gave rise to a positive axial displacement.

9.5 Conclusions

The piezoelectric/electrostrictive monolithic bi-layer composite was formed by co-pressing and co-sintering to amplify the axial displacement. This also substantially reduced the hysteresis or mechanical loss. In addition, asymmetric axial displacements with respect to the polarity of applied field were obtained. A flextensional actuator was developed by sandwiching the monolithic bi-layer composite between two metallic endcaps for further amplification of axial displacement. Such an actuator also offered asymmetric displacement under an applied bipolar electric field.

Chapter 10

Conclusions

In this thesis, the piezoelectric-electrostrictive monolithic bi-layer composite (PE-MBLC) was successfully developed by co-pressing and co-sintering of piezoelectric PMN/PT 63/35 (P) and electrostrictive PMN/PT 90/10 (E) powders. The flat and dome shaped geometries of the composites were obtained by variation in the volume ratios of P and E materials and controlling of the processing parameters. The PE-MBLC ceramics were used in developing the flextensional bi-layer piezoelectric/electrostrictive dome unimorph (BIPEDU) and cymbal actuators. The processing parameters. electromechanical properties and actuation performances of the PE-MBLC and flextensional actuators have been investigated. The main objectives of this thesis were to successfully develop the monolithic bi-layer piezoelectric and electrostrictive composite and to fabricate the flextensional actuators utilizing PE-MBLCs as the driving elements with the goals of enhancing the displacement, reducing the mechanical loss and showing asymmetry behavior under bi-polar electric field.

The following conclusions were drawn from this research:

1. The processing and poling conditions of piezoelectric and electrostrictive PMN-PT ceramics:

- The measured planar shrinkages of both piezoelectric and electrostrictive ceramics were found to be about 13-17 % at different compaction pressures and sintering temperatures.

- Flat and dome-shaped PE-MBLCs were successfully fabricated at a compaction pressure of 86 MPa and sintering temperature of 1150 and 1200 °C, respectively.
- The optimum poling condition for a piezoelectric ceramic was found to be 30 kV/cm, 60 $^{\circ}$ C and 5 minutes. This provides the d₃₃ and d₃₁ of ~650 and ~300 pC/N, respectively.

2. The processing and poling conditions related to electromechanical properties and actuation of various types of bi-layer P/E composite designs:

- The optimal poling conditions to obtain high piezoelectric properties in the flat and dome co-fired bi-layer P/E composites were found to be 2 steps poling: 150 °C, 25 kV/cm for 10 minutes, then cooling down to 25 °C for 40 minutes with the same electric field applied and further poling at 25 °C, for 30 minutes.
- The d_{33}^{eff} of flat and dome composites were found to be 270 and 350 pC/N, respectively.
- The co-fired flat and dome-shaped ceramics had lower d_{33}^{eff} than the calculated value due to a large difference in permittivity of piezoelectric and electrostrictive layers and induced stress at the interface of piezoelectric and electrostrictive layers.
- The dome P/E co-fired bi-layer composite generated the highest displacement of 15 μ m (under 10 kV/cm).

- 3. The PE-MBLC actuators with varying volume percent of piezoelectric phases:
- The shape change from the planar to the dome structure in PE-MBLC ceramics was governed by the volume ratio of the piezoelectric and electrostrictive phases.
- The PE-MBLC actuator with 80 volume percent piezoelectric phase produced the largest dome height of 0.5 mm.
- The d_{33}^{eff} increased and K^{eff} decreased as the volume percent of piezoelectric increased.
- The effective coupling coefficients of PE-MBLCs were lower than those of a piezoelectric ceramic.
- The maximum displacement (~15 μ m) under uni-polar excitation was observed in the PE-MBLC with 50 volume percent piezoelectric phase.
- The observed amplification of displacement was shown to be independent of the dome height.
- 4. The induced stress developed in the PE-MBLC composites after sintering:
- The $I_{(200)}/I_{(002)}$ of PE-MBLC ceramics after polarization were used to explain the effect of the domain reorientations on the enhancement of the displacement in PE-MBLC actuators.
- The $I_{(200)}/I_{(002)}$ of PE-MBLC ceramics after poling suggested that incomplete domain switching of piezoelectric layer was observed.
- The combination of transverse strain response in piezoelectric and electrostrictive layers, the geometry of the composites, and the ratio of

- 5. Bi-layer piezoelectric/electrostrictive dome unimorph actuator:
- A novel design actuator named bi-layer piezoelectric/electrostrictive dome unimorph (BIPEDU) was developed by attaching a dome-shaped PE-MBLC ceramic to a metallic plate.
- The BIPEDU actuators yielded displacement in the range of 65-73 μ m, which is 7-12 times higher than the displacement of PE-MBLC ceramics.
- The uniform bonding of the metal to a PE-MBLC ceramic was essential in obtaining the highest displacement.
- The displacement-load behaviors were found to be different in PE-MBLC and BIPEDU actuators. The displacement of the PE-MBLC actuators was independent of the load. However, the displacement of the BIPEDU actuators decreased with the application of the loads.
- 6. Flextensional cymbal actuator:
- Flextensional cymbal actuator was developed by attaching the two truncated thin brass caps to a flat PB-MBLC ceramic with 50 volume percent piezoelectric phase.
- Higher displacement with minimum hysteresis and asymmetric axial displacements with respect to the polarity of applied field were obtained in PE-
MBLC actuator compared to the piezoelectric or electrostrictive cymbal actuators.

Chapter 11

Suggestions for future work

1. A large displacement under a low applied voltage is one of the requirements for piezoelectric actuators. Therefore, fabrication of monolithic bi-layer composites by tape casting is suggested in order to obtain thin ceramic layers. In addition, tape casting process will eliminate any variations observed in the thickness of layers prepared by dry pressing method.

2. The BIPEDU flextensional actuators yielded the highest displacement. Such BIPEDU actuators were fabricated from a PE-MBLC and 76 μm thick stainless steel. Utilization of other metals with lower and higher young elastic modulii as well as different thicknesses is suggested. This would further improve the actuation performance Finite element analysis (FEA) needs to be performed to predict the effects of various metallic sheets and their thicknesses on the actuation performance (displacement and blocking force) of the BIPEDU.

3. Reliability is a crucial parameter for actuators as it governs their life time in service. As a result, fatigue resistance is an important property which should be studied to determine polarization and strain degradation after driving them through millions of cycles.

4. Recently, environmental and safety concerns due to the use of lead-based ferroelectric materials have surged the development of lead-free ferroelectrics. As a consequence, the co-fired composites, functional gradient ceramics, and flextensional actuators fabricated from lead-free ferroelectric materials should be explored

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PUBLICATIONS

- P. Ngernchuklin, and A. Safari, "Bilayer Piezoelectric/Electrostrictive (P/E) Dome Unimorph", IEEE Transactions of Ultrasonic Ferroelectric and Frequency Control (UFFC), vol. 56, No. 12, pp. 2744-49, 2009.
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PATENT DISCLOSURE

• A. Safari, P. Ngernchuklin, "Bi-layer Piezoelectric/Electrostrictive (P/E) Dome Unimorph," filed March 2009.