SRINIVASAN, SUDARSAN. Piezoelectric, Dielectric and Ferroelectric Thin Films on Metal Substrates for Microelectronic Applications. (Under the direction of Dr. Angus I. Kingon)

The purpose of this research has been to demonstrate the possibility of integrating piezoelectric, dielectric and ferroelectric- lead and barium based oxide thin films and PVDF polymer on flexible metal substrates for microelectronic applications.

Investigations on the key processing parameters and properties relationship for lead zirconate titanate (PZT, 52/48) and barium zirconate titanate (BZT, 35/65) based thin films on Cu foils were performed and studied.

The impact of the oxygen partial pressure on the electrical properties of PZT and BZT thin films during processing has been explored, and demonstrated that high quality films and interfaces can be achieved through control of the pO₂ within a window predicted by thermodynamic stability considerations. It should be noted that the high temperature processing of barium based ferroelectric oxides can be processed on Cu foils in a wider window of pO₂ compared to that of processing lead based ferroelectric oxides. Also, the high volatile nature of lead makes the processing of lead based ferroelectric oxides difficult. Considering these issues, this work shows the processing technique undertaken to achieve high quality barium and lead based oxide thin films on Cu foils. The demonstration has broad implications, opening up the possibility of the use of low
cost, high conductivity copper electrodes for a range of Pb-based and Ba-based perovskite materials, including PZT films in embedded printed circuit board applications for capacitors, varactors, and sensors; multilayer PZT piezoelectric stacks; and multilayer lead magnesium niobate-lead titanate-based dielectric and electrostrictive devices. In the case of ferroelectric PZT films on Cu foil, the capacitors do not fatigue upon repeated switching like those with Pt noble metal electrodes. Instead they appear to be fatigue-resistant like ferroelectric capacitors with oxide electrodes. This may have implications for ferroelectric nonvolatile memories. The effect of electrodes on the electrical properties of lead zirconate titanate has been investigated and discussed.

For systematic and comprehensive analysis of the electromechanical properties, an accurate analytical modeling has been developed. The analytical modeling derived can be applied to double end clamped (under various end conditions) beam geometry that is useful for the case of compliant actuator material (eg. PVDF polymers) and applications, that involve high resonance of operation. These analytical expressions represent the first set of derivations for flexural mode actuators under dynamic conditions, which yield both the resonant frequencies and the displacement at resonance of double end clamped piezoelectric actuators. These analytical expressions allow users to optimize the materials selection and dimensions of bimorph and unimorph actuators.

The reliability of the analytical expressions derived has been investigated using experimental results performed on PVDF polymer as a piezoelectric actuator. The actuators were constructed as double end clamped beams (both bimorph and unimorph cases). They were operated in flexural mode. The two ends of the actuator were clamped
such that the slope at both the ends of the actuator bending curvature at resonance is zero. This type of end condition results in increasing the resonance frequency.
PIEZOELECTRIC, DIELECTRIC AND FERROELECTRIC THIN FILMS ON METAL SUBSTRATES FOR MICROELECTRONIC APPLICATIONS

by

SUDARSAN SRINIVASAN

A dissertation submitted to the Graduate Faculty of
North Carolina State University
in partial fulfillment of the requirements for the Degree of
Doctor of Philosophy

DEPARTMENT OF MATERIALS SCIENCE AND ENGINEERING
Raleigh
2005
APPROVED BY:

Paul D. Franzon
Mark A. Johnson

Ronald O. Scattergood

Angus I. Kingon
Chair of Advisory Committee
DEDICATION

To,

appa, amma & raji
BIOGRAPHY

Sudarsan Srinivasan was born in Madras (now, Chennai), India on May 8, 1980. He received his B.Tech from the department of metallurgical engineering, Indian Institute of Technology Madras, India in 2001. His B.Tech research and thesis was, “Magnetotransport and Doping (Magnetic Ion) Studies in CMR Manganites”. After completing his B.Tech, he started his graduate study from fall 2001 towards his PhD degree in the department of materials science and engineering from NC State University.
ACKNOWLEDGEMENT

I would like to express my sincere gratitude to Dr. Angus Ian Kingon for his continued support, encouragement, patience and valuable advice during my graduate program. I would acknowledge my committee members, Drs. Ron Scattergood, Paul Franzon and Mark Johnson for putting up with my presentation abilities.

It is my pleasure to acknowledge the contribution of group members (present and past), Dr. Dan Litchenwalner, Dr. Taeyun Kim, Dr. Dong-Joo Kim, Dr. Charles Parker, James Mulling, Brian Boyette, Tom Blair, Dwi Wicaksana, and for their valuable guide and friendship. A special thanks to Edna Deas and Jan Jackson for administrative support.

Finally, to appa, amma, raji, babu, karups, dhadee, romeo, ba, maddy, dilip, ashraf, pande, ks, sandy and myself: Sub salutes!!
### Table of contents

List of Tables ........................................................................................................ ix

List of Figures ........................................................................................................ x

1 Introduction ........................................................................................................ 1
   1.1 Introduction .................................................................................................. 1
   1.2 Research goals ............................................................................................ 5
   1.3 Overview of dissertation ............................................................................. 6
   1.4 References ................................................................................................. 7

2 Literature review ................................................................................................. 9
   2.1 Ferroelectrics ............................................................................................... 9
   2.2 Piezoelectricity ........................................................................................... 13
      2.2.1 Crystal classes ...................................................................................... 13
      2.2.2 Fundamental piezoelectric relations .................................................. 14
      2.2.3 Mathematical description of piezoelectric d-coefficient ..................... 17
   2.3 The PbTiO$_3$-PbZrO$_3$ system .................................................................. 20
   2.4 Domains ..................................................................................................... 24
   2.5 Intrinsic and extrinsic contribution ........................................................... 29
   2.6 Phenomenological theory ........................................................................... 34
   2.7 Microelectromechanical devices ............................................................... 38
      2.7.1 Piezoelectric bimorph actuators ......................................................... 38
<table>
<thead>
<tr>
<th>Section</th>
<th>Title</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>2.7.2</td>
<td>Piezoelectric unimorph actuators</td>
<td>41</td>
</tr>
<tr>
<td>2.7.3</td>
<td>Device applications</td>
<td>43</td>
</tr>
<tr>
<td>2.8</td>
<td>References</td>
<td>46</td>
</tr>
<tr>
<td>3</td>
<td>Dielectric characterization of Ba(Zr_{0.35}Ti_{0.65})O_3 thin films on copper foil</td>
<td>50</td>
</tr>
<tr>
<td>3.1</td>
<td>Introduction</td>
<td>50</td>
</tr>
<tr>
<td>3.2</td>
<td>Experimental section</td>
<td>52</td>
</tr>
<tr>
<td>3.3</td>
<td>Results and discussion</td>
<td>54</td>
</tr>
<tr>
<td>3.4</td>
<td>Conclusion</td>
<td>60</td>
</tr>
<tr>
<td>3.5</td>
<td>References</td>
<td>60</td>
</tr>
<tr>
<td>4</td>
<td>PZT thin films directly on copper foil for piezoelectric, dielectric and ferroelectric applications</td>
<td>62</td>
</tr>
<tr>
<td>4.1</td>
<td>Introduction</td>
<td>62</td>
</tr>
<tr>
<td>4.2</td>
<td>Experimental section</td>
<td>65</td>
</tr>
<tr>
<td>4.3</td>
<td>Results and discussion</td>
<td>67</td>
</tr>
<tr>
<td>4.4</td>
<td>Conclusion</td>
<td>74</td>
</tr>
<tr>
<td>4.5</td>
<td>References</td>
<td>75</td>
</tr>
<tr>
<td>5</td>
<td>Effect of copper and platinum as electrodes on PZT thin films for ferroelectric applications</td>
<td>77</td>
</tr>
<tr>
<td>5.1</td>
<td>Introduction</td>
<td>77</td>
</tr>
<tr>
<td>5.2</td>
<td>Experimental section</td>
<td>79</td>
</tr>
</tbody>
</table>
6.2 a Platinum as the substrate ......................................................... 79
6.2 b Copper as the substrate ......................................................... 79
6.3 Results and discussion ............................................................... 81
   6.3.1 Results on effect of pO₂ .................................................. 86
6.4 Conclusion ................................................................................. 92
6.5 References .................................................................................. 93

6 Modelling and characterization: Dynamic behavior of piezoelectric bimorph
actuators under various end conditions ........................................... 95
6.1 Introduction .................................................................................. 95
6.2 Bimorph actuator assembly ....................................................... 99
6.3 Discussion .................................................................................... 110
6.4 Conclusion of analytical modeling ............................................. 113
6.5 Experimental section ............................................................... 114
6.6 Results and discussion ............................................................. 117
6.7 Conclusion on experimental section ........................................... 118
6.8 References ................................................................................... 119

7 Modelling and characterization: Dynamic behavior of piezoelectric unimorph actuators under various end conditions ......................... 120
7.1 Introduction .................................................................................. 120
7.2 Unimorph actuator assembly .................................................... 122
7.3 Discussion .................................................................................... 142
8 Conclusion of analytical modeling ................................................. 138
7.5 Experimental section .................................................................. 138
7.6 Results and discussion .................................................................. 141
7.7 Conclusion on experimental section ............................................ 145
7.8 References .................................................................................. 145

8 Dynamic response of single end clamped piezoelectric actuator ........... 147
8.1 Introduction ................................................................................ 147
8.2 Bimorph actuator assembly ......................................................... 148
8.3 Unimorph actuator assembly......................................................... 152
8.4 Conclusion ................................................................................ 154
8.5 References ................................................................................ 155

9 Conclusions and future work ........................................................ 156
9.1 Conclusions .............................................................................. 156
9.2 Future work ............................................................................. 158
List of Tables

Table 2.1 The theoretical properties of PZT at 25°C .................................................. 36
Table 2.2 Comparing energy densities for magnetic, shape memory alloys, electrostatic, and piezoelectric actuation............................................................... 44
Table 5.1 PZT thin films on platinized Si substrate processed under various ambient pO$_2$.................................................................................................................. 86
Table 6.1 Analytical expression available for bimorph and unimorph actuators under different end conditions.............................................................. 98
Table 6.2 Table of eight allowed boundary conditions.................................................. 105
Table 6.3 Specifications of PVDF polymer bimorph actuators............................... 115
Table 7.1 Table of eight allowed boundary conditions.............................................. 131
Table 7.2 Specifications of PVDF polymer bimorph actuators............................... 140
Table 7.3 Table of different metal shims considered for the unimorph design with its physical constants.............................................................. 142
List of Figures

Figure 2.1 First-order (a) and second-order (b) ferroelectric phase transition ($\chi$ is inverse dielectric susceptibility) ................................................................. 10
Figure 2.2 The unit cell of perovskie crystal structure ......................................................... 11
Figure 2.3 Schematic of polarization hysteresis for a ferroelectric .............................. 12
Figure 2.4 Crystal classes of ferroelectric and piezoelectric materials ......................... 15
Figure 2.5 Two dimensional description of the origin of the piezoelectric response in a single domain PbTiO$_3$ crystal (a) situation under no field (b) shift of the Ti cation away from the equilibrium position under $E_3$ or $X_3$ (c) shift of the Ti cation back towards the cell center under $E_1$ or $X_1$ (d) tilting of the Ti position giving $\Delta x$ or $\Delta P$ under shear mode ........... 20
Figure 2.6 Phase diagram of the PbZrO$_3$-PbTiO$_3$ (PZT) system ........................................ 21
Figure 2.7 Crystal structures and their allowed polar axes. The polarization direction is based on the prototype unit cell ............................................................... 22
Figure 2.8 Dielectric (a), piezoelectric (b), and polarization (c) behavior of bulk PZT ceramics ........................................................................................................... 25
Figure 2.9 Dielectric (a), piezoelectric (b), and polarization (c) behavior of single crystal PZT expected from phenomenological theory ........................................... 26
Figure 2.10 Schematic illustration of 180° domain growth in ferroelectrics ..................... 28
Figure 2.11 Schematic illustration of 90° domain growth in ferroelectrics ...................... 28
Figure 2.12 Intrinsic contribution due to the response from lattice ................................. 31
Figure 2.13 Extrinsic contribution to the piezoelectric response due to non-180° domain wall motion with an applied field for the tetragonal ferroelectrics .................................................. 31

Figure 2.14 The intrinsic and extrinsic values of dielectric constant and piezoelectric coefficients for undoped PZT (PZT-500) and PZT (52/48) .................................................................................................................. 33

Figure 2.15 Two types of cantilever bimorph actuators. (a) series and (b) parallel-connected ........................................................................................................ 40

Figure 2.16 Schematic representation of unimorph actuator with the piezoelectric element on top of the non-piezoelectric element .............................................. 42

Figure 3.1 Thermodynamic representation of Cu-Cu₂O and Cu₂O-CuO stability conditions. Region A is the annealing condition within the regime where the oxides of copper are stable. Region B is within the stable regime of Cu₂O. Region C is within the regime where Cu is non-oxidizing ........................................................................................................ 53

Figure 3.2 X-ray diffraction pattern (Cu kα radiation) of BZT thin film on Cu foil .... 55

Figure 3.3 Two micron by two micron atomic force microscopic topography image of BZT thin film on copper foil (the difference in the color of the surface is due to the superimposed roughness of copper foil) .............. 55

Figure 3.4 Voltage dependence of dielectric constant and loss tangent of BZT thin film on copper foil at room temperature ................................................. 57

Figure 3.5 Voltage dependence of tunability of BZT thin film on copper foil at 500 KHz .............................................................................................................. 58
Figure 3.6  Frequency dependence of dielectric constant and loss tangent of BZT thin film on copper foil in the range of 10 KHz to 1 MHz........................................... 58

Figure 3.7  Temperature dependence of the dielectric constant and loss tangent of BZT thin film on copper foil at 500 KHz and zero bias................................. 60

Figure 4.1  Thermodynamic representation of Cu-Cu$_2$O, Cu$_2$O-CuO and Pb-PbO stability conditions. The dashed line gives the observed pO$_2$ during heating and cooling to the stable anneal regime, indicated as region C. Region A is the sintering annealing condition within the regime where the oxides of copper and lead are stable. Region B is within the stable regime of Cu$_2$O and PbO. Region C is within the regime where PbO is stable and Cu is non-oxidizing. Region D (D1 and D2) is within the stable regime for Cu and Pb (non-oxidizing). All regions mentioned above are at 650°C................................................................. 65

Figure 4.2  X-ray diffraction pattern (Cu k$_\alpha$ radiation) of PZT thin film on Cu foil. Films were crystallized under different pO$_2$ conditions at 650°C. The legend on the right identifies the diffraction peaks labeled in the diffraction pattern. The regions marked as region A, B, C, D1 and D2 indicated different annealing conditions as shown in Figure 4.1............. 68

Figure 4.3  Two micron by two micron atomic force microscopic topography image of PZT thin film on copper foil. Average grain sizes are less than 100 nm................................................................. 69

Figure 4.4  Voltage dependence of permittivity and loss tangent of PZT thin film on copper foil, at room temperature ................................................................. 71
Figure 4.5  Polarization-electric field hysteresis loops of PZT thin film capacitor on copper foil. Note that the PZT composition is close to the morphotropic phase boundary ................................................................. 72

Figure 4.6  Fatigue characteristics of a typical Cu/PZT capacitor (Cu as the top electrode) on Cu foil, in comparison with a Pt/PZT capacitor on Pt foil. The two types of film were processed in parallel (i.e. under identical thermal and pO₂ conditions) ............................................................... 73

Figure 4.7  Leakage current dependence on applied voltage of PZT thin film on copper foil. Leakage currents increases about one order of magnitude after 10^10 switching cycles ............................................................................ 74

Figure 5.1  X-ray diffraction pattern of crystallized PZT thin film on Pt foil with no top electrode .................................................................................................................................................. 81

Figure 5.2  Electric field dependence of polarization of PZT thin film capacitor on platinum foil .................................................................................................................................................. 82

Figure 5.3  Polarization fatigue curves for identical capacitors with Pt or Cu as top electrodes .................................................................................................................................................. 83

Figure 5.4  Electric field dependence of polarization of PZT thin film capacitor on copper and platinum foil .................................................................................................................................................. 84

Figure 5.5  Polarization fatigue curves for PZT thin films processed on copper and platinum substrates .................................................................................................................................................. 84

Figure 5.6  Comparison of leakage current dependence on applied voltage of PZT thin film on platinum and copper foils with Pt and Cu top electrodes respectively .................................................................................................................................................. 85
Figure 5.7 (a) Dielectric dependence on applied voltage of PZT/Pt/SiO$_2$/Si sintered at 650°C and pO$_2$=1 atm; (b) Dielectric dependence on frequency of PZT/Pt/SiO$_2$/Si sintered at 650°C and pO$_2$=1 atm................. 87

Figure 5.8 (a) Dielectric dependence on applied voltage of PZT/Pt/SiO$_2$/Si sintered at 650°C and pO$_2$=0.21 atm; (b) Dielectric dependence on frequency of PZT/Pt/SiO$_2$/Si sintered at 650°C and pO$_2$=0.21 atm................. 88

Figure 5.9 (a) Dielectric dependence on applied voltage of PZT/Pt/SiO$_2$/Si sintered at 650°C and pO$_2$=10$^{-5}$ atm; (b) Dielectric dependence on frequency of PZT/Pt/SiO$_2$/Si sintered at 650°C and pO$_2$=10$^{-5}$ atm................. 88

Figure 5.10 (a) Dielectric dependence on applied voltage of PZT/Pt/SiO$_2$/Si sintered at 650°C and pO$_2$=10$^{-12}$ atm; (b) Dielectric dependence on frequency of PZT/Pt/SiO$_2$/Si sintered at 650°C and pO$_2$=10$^{-12}$ atm................. 89

Figure 5.11 (a) Dielectric dependence on applied voltage of PZT/Pt/SiO$_2$/Si sintered at 450°C, pO$_2$=10$^{-6}$ atm and 650°C and pO$_2$=10$^{-12}$ atm; (b) Dielectric dependence on frequency of PZT/Pt/SiO$_2$/Si sintered at 450°C, pO$_2$=10$^{-6}$ atm and 650°C and pO$_2$=10$^{-12}$ atm........................................... 89

Figure 5.12 Dielectric tunability dependence of PZT thin films processed under different ambient pO$_2$................................................................. 90

Figure 5.13 Polarization hysteresis of PZT/Pt/SiO$_2$/Si sintered at (a) 650°C, pO$_2$=0.21 atm; (b) 650°C, pO$_2$=10$^{-5}$ atm; (c) 650°C, pO$_2$=10$^{-12}$ atm; and (d) 450°C, pO$_2$=10$^{-6}$ atm and 650°C, pO$_2$=10$^{-12}$ atm................................. 91
Figure 6.1 Two types of cantilever bimorph actuators. (a) series and (b) parallel-connected................................................................. 96

Figure 6.2 Bimorph excited by external voltage................................................................. 99

Figure 6.3 Fixture showing possible rotation at both ends and translation at one
end of the actuator ......................................................................................... 105

Figure 6.4 Figure shows multiple overtones of a double end clamped actuator of
length L ............................................................................................................ 108

Figure 6.5 Comparison of normalized dynamic performance of the actuator under
different flexure end conditions ................................................................. 111

Figure 6.6 Dependence of first resonance frequency and its dynamic center
displacement on bimorph actuator length................................................... 112

Figure 6.7 Variation of first resonance frequency and its dynamic center
displacement as a function of t/L for bimorph actuators............................ 113

Figure 6.8 Test apparatus for measuring actuator deflections at a range of applied
voltages and various end conditions............................................................ 115

Figure 6.9 Shows the resonance frequency at first and second overtone versus
length of the actuator for the same end condition ........................................ 117

Figure 6.10 Shows the center deflection at first resonance versus length of the
actuator for the same end condition ............................................................. 118

Figure 7.1 Schematic representation of unimorph actuator with the piezoelectric
element on top of the non-piezoelectric element........................................ 121

Figure 7.2 Unimorph excited by external voltage......................................................... 123

Figure 7.3 Diagram showing centroidal axis with reference line at Z=0............. 126
Figure 7.4  Fixture shows possible rotation at both the ends and translation at one end of the actuator ........................................................................................................................................ 131

Figure 7.5  Comparison of normalized dynamic behavior of actuators under different boundary conditions. The displacement refers to the maximum deflection of a double end clamped cantilever unimorph actuator ......................................................................................................................................... 136

Figure 7.6  Shows the dynamic response of the actuator with respect to the thickness ratio ........................................................................................................................................ 137

Figure 7.7  Test apparatus for measuring actuator deflections at a range of applied voltages and various end conditions ........................................................................................................ 139

Figure 7.8  Comparison of different metal shims for unimorph ........................................... 141

Figure 7.9  (a) Frequency response, (b) Displacement response ........................................ 143

Figure 7.10  Shows the resonance frequency at first and second overtone versus length of the actuator for the same end condition .................................................. 144

Figure 7.11  Shows the center deflection at first resonance versus length of the actuator for the same end condition ......................................................................................... 145

Figure 8.1  Schematic diagram of a single end clamped cantilever bimorph actuator (series-connected) .................................................................................................................. 149

Figure 8.2  Schematic diagram of single end clamped unimorph actuator ................. 153
Chapter 1 Introduction

1.1 Introduction

Ferroelectric materials have been receiving renewed attention for electronic applications since 1989. Significant progress made during last decade in processing ferroelectric thin films and in their integration with Si-based technology has brought closer the implementation of a new generation of microelectronic devices [1-5]. The requirement that ferroelectric materials be processed at high temperatures in relatively oxidizing atmospheres led to the traditional use of noble metal electrodes in conjunction with these materials for ferroelectric, piezoelectric and dielectric applications. However, there has been a strong desire to utilize base metal electrodes, primarily for cost reasons. This drive has been led by the multilayer capacitor industry, where the shear number of capacitors produced has driven the significant replacement of noble metals by Ni-based electrodes in BaTiO₃-based capacitors [6-9]. This shift has not been simple, and has necessitated significant dopant engineering, and has been accompanied by a necessary reduction in processing temperatures and changes in the sintering atmospheres.

The situation for Pb-based ferroelectrics is significantly difficult, due to the propensity of PbO to reduce under substantially less reducing conditions (ie at higher pO₂ values). This has been a source of frustration for researchers who desire to utilize
PbTiO$_3$- and Pb(Zr,Ti)O$_3$-based materials for ferroelectric, dielectric, and piezoelectric applications.

This research has addressed this issue in developing PZT-based dielectric films for various microelectronic applications [10-13]. These were produced on Cu foil, allowing the integration into low cost, high density, printed circuit boards. The previous approach adopted was to utilize a Ni/Ni-phosphide interface layer between the Cu foil and the PZT. The Ni acted as an oxygen getter and barrier by partially forming NiO. While this NiO did react with the PZT, creating a series-connected low permittivity interface layer, it resulted in a system highly desirable for the embedded capacitor application. In particular, it resulted in a linear dielectric, with the trade-off in reduced capacitance density being entirely acceptable for most embedded capacitor applications.

For other applications, in particular where higher dielectric permittivity is desired, for tunable dielectrics, or for ferroelectric switching and piezoelectric applications, this Ni interface layer is not the optimum solution, as the interface layer is deleterious to these properties. Thus, this research re-addressed the issue of whether PZT can be deposited directly on Cu electrodes, without deleterious reactions. For convenience, Cu foil has been used as both the bottom electrode and the substrate. This is also the preferred approach for the case where the resultant device (capacitor, varactor, sensor) is to be integrated directly into a printed circuit board. PZT films were prepared by chemical solution deposition techniques [14] directly on copper foils, as described later. The oxygen partial pressures were carefully controlled throughout processing, as described.
Recently, microelectromechanical systems (MEMS) have become an important area of technology, building on the success of the microelectronics industry over the past 50 years [15,16]. The success of MEMS as a key technology in the twenty-first century depends in no small part on the solution of materials issues associated with the design and fabrication of complex devices. There is strong interest in introducing ferroelectric lead zirconate titanate (PZT) materials for microelectromechanical systems, since they have large piezoelectric coefficients and electromechanical coupling coefficients [17-20], which may further enhance the performance of sensing and actuation devices. In comparison with commonly used piezoelectric materials such as ZnO, PZT has more than one order of magnitude larger piezoelectric coefficients. Although successful demonstrations of MEMS based on PZT thin films have been reported, there is still a lack of fundamental understanding regarding electromechanical behavior of PZT thin films.

In designing a piezoelectric MEMS actuator for a particular application, there are many parameters to be defined, namely choosing the appropriate piezoelectric material (piezoelectric coefficient), the geometric dimensions (such as the length of the actuator, thickness of the piezoelectric material and thickness of the non-piezoelectric material, if any). Therefore, deriving an analytical expression describing the dynamic actuator’s response would simplify the process of designing the actuator. The current approach is to utilize finite element methods.

Wang et al [21] have derived the quasi-static displacement of single end clamped unimorph and bimorph actuators. The analytical expression for calculating the quasi-static displacement cannot be applied at dynamic resonance. Smits et al [21-23] have described the dynamic behavior of single end clamped bimorph and unimorph actuators.
It should also be noted that the analytical expression given [21-23] for calculating the free end deflection of single end clamped actuators tends to infinity at resonance, and therefore does not allow displacement to be calculated at resonance. Also, these and other references have considered piezoelectric actuator with only single end clamping. This condition of single end clamped actuator is applicable only for non-compliant piezoelectric materials and does not apply to soft (compliant) materials such as PVDF polymers and very thin non-compliant piezoelectric materials. A double end clamped configuration is suitable under such circumstances.

As mentioned earlier, with the increase in demand for piezoelectric materials that offer higher dynamic response, analytical equations for dynamic displacement at resonance of a double end clamped actuator will allow optimizing the actuator design for improved performance. This research has focused on deriving the dynamic behavior of double end clamped (under various condition) bimorph and unimorph actuators/sensors. In addition, this investigation studied the effects of end conditions on the dynamic behavior of these types of actuators, which also plays an important role in the actuator design. Besides its usefulness for the case of piezoelectric material, the advantage of double end clamped beam over the single end clamped beam may be seen for the case of higher resonant frequency applications. For a given actuator of fixed geometrical dimensions and other external parameters such as the electric field applied etc, held fixed, the resonant frequencies of vibration of the actuator are considerably higher for the double end clamped configuration, though possibly compromising the magnitude of deflection produced. Thus, where high resonant frequencies are required, the double end clamped configuration should be considered. The analytical equations for dynamic
displacement of an actuator at resonance can be applied to various actuator geometries and to a variety of piezoelectric materials.

1.2 Research goals

The current research initiative is to develop and characterize the integration of lead, barium based oxide thin films (and PVDF polymers) on flexible copper (and nickel) substrate for microelectronic applications. This thesis work has focused on the following objectives.

To develop and characterize the integration of PZT thin films on flexible copper substrate for microelectronic applications.

To extend the integration of piezoelectric materials on copper to barium based thin films on copper substrate.

To understand the effect of conducting electrodes (Pt and Cu) on the ferroelectric properties of these passive devices.

To develop a robust and reliable analytical modeling for better understanding of the dynamic behavior of piezoelectric beams resonators (which includes actuator and sensor applications) for MEMS device.

Perform experiments to confirm the reliability of the mentioned analytical model.
1.3 Overview of dissertation

The basic organization of the dissertation is as follows. In Chapter 2, piezoelectricity and ferroelectricity are reviewed with an emphasis on the PZT ceramics and PVDF polymer systems. Current issues for application and proposed mechanisms are also included. In Chapter 3, the development of processing BZT on flexible copper substrate is described and the corresponding electrical characterization is explained. Chapter 4 describes the thermodynamic stability of PZT during high temperature processing on copper substrates and the corresponding electrical characterization is explained. Chapter 5 describes the effect of copper and platinum as electrodes on PZT thin films and their ferroelectric responses are compared. Chapter 6 and 7 describes the analytical derivation and experimental characterization for double end clamped piezoelectric bimorph and unimorph actuators (under various end conditions) respectively. In Chapter 8, the major results and discussion that are concerned with the directions for future work are included.
1.4 References


Chapter 2. Literature review

2.1 Ferroelectrics

Ferroelectric materials can be defined as those, which exhibit an equilibrium spontaneous polarization whose orientation can be reversed by an applied electric field [1-6]. The appearance of the spontaneous polarization coincides with a structural phase transformation from a higher symmetry to a lower symmetry crystal structure as the material is cooled below its Curie temperature ($T_c$). At temperatures above $T_c$ a centric non-ferroelectric, or paraelectric, phase is favored. Below $T_c$, a non-centric ferroelectric phase is stable. In many cases, especially the perovskites, these phases differ only in the tilting and/or distortion of oxygen octahedral. In the vicinity of the Curie temperature, thermodynamic properties (e.g., dielectric, elastic, optical, and thermal properties) of ferroelectric crystals exhibit anomalies. One such anomaly is the temperature dependence of the permittivity. In most ferroelectric crystals, the relative permittivity, $\varepsilon_r$, obeys the Curie-Weiss law at temperature above the Curie temperature:

$$\varepsilon_r = \frac{C}{T-T_o}$$

(2.1)

where $C$ is the Curie-Weiss constant, which is typically on the order of $10^3 - 10^5$ K [3] and $T_o$ is the Curie-Weiss temperature. In an ideal first order phase transition, $T_o < T_c$, while in a second order phase transition $T_o=T_c$ (figure 2.1). As mentioned above, a phase transition is a characteristic feature of a ferroelectric. Phase transitions are divided into
first- and second-order transitions. The second derivatives of the thermodynamic potential change during a second-order phase transition. These derivatives include the dielectric constant, specific heat, thermal expansion coefficient, elastic modulus, and piezoelectric coefficient. In addition to a sharp change in these quantities, the first derivatives of the thermodynamic potential such as spontaneous polarization, volume, and entropy experience a sudden change during a first-order phase transition, and the latent heat of transition is released. The examination of ferroelectric phase transition is important because it is very useful for characterizing materials and correlating the different parameters of ferroelectrics.

Figure 2.1: First-order (a) and second-order (b) ferroelectric phase transition ($\chi$ is inverse dielectric susceptibility) [3].
There are literally hundreds of materials known to be ferroelectrics. However, a group of ferroelectrics known as the “perovskite family” are perhaps the most extensively studied and technologically important. Ferroelectrics such as Pb(Zr,Ti)O₃, (Ba,Sr)TiO₃, and LiNbO₃ belong to this family and have the general formula ABO₃. The term perovskite is given to this group of materials because their crystal structure is based on that of the mineral perovskite (CaTiO₃) [1]. Figure 2.2 shows the ideal perovskite crystal structure. The atoms located at the corners of the unit cell are referred to as the A-site cations and the atoms located at the center of the unit cell are referred to as the B-site cations. The A-site cations are usually larger than the B-site cations, but they have lower charge. The coordination number of the A-site cation is twelve and that of the B-site cation is six. During the phase transition at $T_c$, the relative shift of the oxygen octahedral and the cation atoms due to the structural distortion result in the generation of an electric dipole moment in the ferroelectric phases.

![Figure 2.2: The unit cell of perovskite crystal structure.](image-url)
Another important characteristic of ferroelectrics is the polarization – electric field (P-E) hysteresis loop. This loop is the evidence showing that the spontaneous polarization can be reoriented among the possible crystallographic orientations. The hysteresis in the P-E relation results from the stability of the polarization along the preferred crystallographic orientations and its resistance to reorientation. From the P-E hysteresis loop (see figure 2.3), several distinctive characteristics can be described. The saturation polarization, defined as an extrapolated value from the high field linear portion, refers to the total polarization attributable to the reversal of the spontaneous polarization. The remanent polarization ($P_r$) refers to the amount of polarization remaining after removal of the applied electric field. This reflects the degree of alignment of the dipole moments in a given ferroelectric. In an ideal single crystal, the hysteresis loop would have a rectangular central region with $P_r=P_s$. The coercive field ($E_c$) refers to the field value required to reverse the direction of the polarization.

Figure 2.3: Schematic of polarization hysteresis for a ferroelectric.
2.2 Piezoelectricity

2.2.1 Crystal classes

The first experimental demonstration of a connection between macroscopic piezoelectric phenomena and crystallographic structure was published in 1880 by Pierre and Jacques Curie [7]. Their experiment consisted of a conclusive measurement of surface charges appearing on specially prepared crystals (tourmaline, quartz, topaz, cane sugar and Rochelle salt among them), which were subjected to mechanical stress (direct piezoelectric effect). A year later it was discovered that a geometric strain is proportional to an applied electric field (converse piezoelectric effect).

Whether a crystal exhibits the piezoelectric effect is determined by its crystal class. Of the 32 crystal classes, 21 classes do not have a center of symmetry. Twenty of 21 noncentrosymmetric classes exhibit piezoelectricity. Crystal class 432 is the exception to this classification. The piezoelectric classes can be subdivided into 10 classes, which have pyroelectricity and the remaining 10 classes, which are nonpyroelectric. A pyroelectric material has unique crystallographic polar axis, which results in spontaneous polarization along this axis. A spontaneous polarization is originated from the displacement between the centers of positive and negative charge in the unit cell. When uniformly heated, the spontaneous dipole magnitude changes in response to the small thermal strains. ZnO and AlN are examples of pyroelectric material whose dipole magnitude is also sensitive to mechanical strain. An example of a piezoelectric material that is not pyroelectric is quartz. Quartz has microscopic dipoles in the unit cell, but they sum to a net polarization of zero. When a stress or an electric field is applied, one
direction will develop a larger dipole, resulting in a net polarization. Certain crystal structures which belong to the 10 polar point groups exhibit ferroelectricity. An important characteristic of a ferroelectric structure is that it contains a spontaneous polarization, which can be switched by the application of an applied electric field. Figure 2.4 is the chart that shows the breakdown of the 32 point groups into their possible ferroelectric and non-ferroelectric classes. Ferroelectric materials exhibit strong piezoelectric properties after poling operation, because randomly oriented polar axis belongs to \( \infty \infty m \) macroscopic symmetry, which is not polar [2].

2.2.2 Fundamental piezoelectric relations (thermodynamic approach)

The thermodynamic free energy function is the most general and useful tool for the macroscopic description of the solid state. The relevant thermodynamic function depends on the choice of boundary conditions [1, 3, 7].

For a polarizable deformable anisotropic linear solid, the Helmholtz free energy function, \( \Delta A \), will be expressed in terms of temperature \( T \), strain \( x \), and polarization \( P \). Under the isothermal condition, the relations is expressed as,

\[
\left( \frac{\partial \Delta A}{\partial x_{ij}} \right)_{PT} = X_{ij} = c_{ijkl}x_{kl} - a_{mij}P_m
\]

\[
\left( \frac{\partial \Delta A}{\partial P_m} \right)_{xT} = E_m = \chi_{mn}P_n - a_{mij}x_{ij} \tag{2.2}
\]
Figure 2.4: Crystal classes of ferroelectric and piezoelectric materials.

The $\chi_{mn}$ is the reciprocal dielectric susceptibility, $c_{ijkl}$ is the elastic stiffness and $a_{mij}$ is the piezoelectric constant. Noted that subscripts $i, j, k, l, m, n, o, p = 1, 2, 3$, and the indices 1, 2, 3 refer to a set of orthogonal reference axes.
The elastic Gibbs free energy function, $\Delta G_1$, is more suitable when the variables temperature $T$, stress $X$, and polarization $P$ are given as the boundary conditions, and the relations are,

$$
\left( \frac{\partial \Delta G_1}{\partial X_{ij}} \right)_{PT} = -X_{ij} = -s_{ijkl} X_{kl} - b_{mij} P_m
$$

$$
\left( \frac{\partial \Delta G_1}{\partial P_m} \right)_{XT} = E_m = \chi_{mn} P_n - b_{mij} X_{ij}
$$

(2.3)

where $s_{ijkl}$ is the elastic compliance and $b_{mij}$ is the piezoelectric constant.

With the boundary conditions of temperature $T$, strain $x$, and electric field $E$, the electric Gibbs free energy function, $\Delta G_2$, can be used and the relations are given by,

$$
\left( \frac{\partial \Delta G_2}{\partial x_{ij}} \right)_{ET} = X_{ij} = c_{ijkl} X_{kl} - e_{mij} E_m
$$

$$
\left( \frac{\partial \Delta G_2}{\partial E_m} \right)_{xT} = -P_m = -\eta_{mn} E_n - e_{mij} X_{ij}
$$

(2.4)

where $\eta_{mn}$ is the electric susceptibility and $e_{mij}$ is the piezoelectric coefficient.

When the temperature $T$, stress $X$, and electric field $E$ are given as the independent variables, the full Gibbs free energy function, $\Delta G$, is used for the relation expressed as,

$$
\left( \frac{\partial \Delta G}{\partial X_{ij}} \right)_{ET} = -X_{ij} = -s_{ijkl} X_{kl} - d_{mij} E_m
$$

$$
\left( \frac{\partial \Delta G}{\partial E_m} \right)_{XT} = -P_m = -\eta_{mn} E_n - d_{mij} X_{ij}
$$

(2.5)

where $d_{mij}$ is the piezoelectric coefficient.
These relations expressed by the set of equations are called the fundamental piezoelectric relations. From above equations and relation \( E_m = \chi_{mn}^X P_n \) or \( P_m = \eta_{mn}^X E_n \), the equivalent relations can be derived as follows,

\[
\begin{align*}
e_{mkl} &= d_{mij} c_{ijkl}^E \\
d_{mkl} &= e_{mij} s_{ijkl}^E \\
b_{mkl} &= a_{mij} s_{ijkl}^P \\
a_{mkl} &= b_{mij} c_{ijkl}^P \\
b_{mij} &= d_{mij} \chi_{mn}^X \\
d_{mij} &= b_{mij} \eta_{mn}^X \\
e_{mij} &= a_{mij} \eta_{mn}^X \\
a_{mij} &= e_{mij} \chi_{mn}^X
\end{align*}
\]

(2.6)

Since most electromechanical measurements are made under a variable electrical field or a stress, the equation (2.5) is the most convenient one for defining materials’ piezoelectric coefficients.

### 2.2.3 Mathematical description of piezoelectric \( d \)-coefficient

If a stress is applied to certain crystals they develop an electric moment whose magnitude is proportional to the applied stress. This is known as the direct piezoelectric effect [8]. For example, if a uniaxial tensile stress \( X \) is applied along one of the diad axes of a piezoelectric crystal, the electric moment per unit volume, or the polarization charge per unit area, is mathematically expressed by the following tensor equation.

\[
P_{i} = d_{ijk} X_{jk} \quad (i,j,k=1,2,3)
\]

(2.7)

where \( P_i \) is the induced polarization, \( d_{ijk} \) is the direct piezoelectric coefficient, and \( X_{jk} \) is the applied mechanical stress.

For the converse effect, the relationship is given by
\[ x_{jk} = d_{ijk} E_i \quad (i,j,k=1,2,3) \] (2.8)

where \( x_{jk} \) is the field-induced strain, \( d_{ijk} \) is the piezoelectric coefficient, and \( E_i \) is the applied electric field. In both cases, the piezoelectric coefficients \( d_{ijk} \) are numerically identical and expressed in the units C/N and m/V for the direct and converse relationships, respectively. The proof of this equality of the coefficients is based on thermodynamical reasoning and is explained at section 2.2.2. The piezoelectric coefficient \( d_{ijk} \) forms a third-rank tensor, which has 27 coefficients. When the piezoelectric coefficient is expressed by matrix notation, the subscripts \( jk \) is replaced by one subscript ranging from 1 to 6, leaving 18 independent d-coefficients. Although the matrix notation has the advantage of greater compactness than the tensor notation, it should be noted that piezoelectric matrices in matrix notation cannot be directly transformed by standard tensorial methods, rather transformation matrices specific to this convention must be used [8].

When crystal symmetry exists, the number of independent piezoelectric coefficients is reduced. For example, quartz, which belongs to point group 32 has a piezoelectric matrix expressed as

\[
\begin{bmatrix}
  d_{11} & d_{12} & 0 & d_{14} & 0 & 0 \\
  0 & 0 & 0 & 0 & d_{25} & d_{26} \\
  0 & 0 & 0 & 0 & 0 & 0 \\
\end{bmatrix} = \begin{bmatrix}
  2.3 & -2.3 & 0 & -0.67 & 0 & 0 \\
  0 & 0 & 0 & 0 & 0.67 & -4.6 \\
  0 & 0 & 0 & 0 & 0 & 0 \\
\end{bmatrix} \text{(pm/V)}
\]

The constant \( d_{11} \) represents the expansion (or contraction) in the crystallographic 1-direction when an electric field is applied in the 1-direction. The \( d_{11} \) constant of quartz is often used as a piezoelectric standard since high quality X-cut quartz is readily available and \( d_{11} \) has only small nonlinearities.
For a crystal with point group 6mm and poled piezoelectric ceramics (≈mm symmetry), the piezoelectric matrix is expressed by

\[
\mathbf{d} = \begin{bmatrix}
0 & 0 & 0 & 0 & d_{15} & 0 \\
0 & 0 & 0 & d_{24} & 0 & 0 \\
d_{31} & d_{32} & d_{33} & 0 & 0 & 0
\end{bmatrix}
\]  

(2.9)

where \(d_{31}=d_{32}, d_{24}=d_{15}\). Assuming zero stress and for an electric field of magnitude \(E_3\) applied in the 3-direction (along the crystallographic c-axis), the strain is given by

\[
\begin{bmatrix}
x_1 \\
x_2 \\
x_3 \\
x_4 \\
x_5 \\
x_6
\end{bmatrix} =
\begin{bmatrix}
0 & 0 & d_{31} \\
0 & 0 & d_{31} \\
0 & 0 & d_{33} \\
0 & d_{15} & 0 \\
d_{15} & 0 & 0 \\
0 & 0 & 0
\end{bmatrix}
\begin{bmatrix}
0 \\
0 \\
0 \\
E_3
\end{bmatrix}
\]  

(2.10)

The applied electric field causes strains both parallel (\(x_3\)) and perpendicular (\(x_1\) and \(x_2\)) to the applied field (\(E_3\)). The \(d_{33}\) piezoelectric constant represents an expansion (or contraction) of the crystal along the crystallographic 3-direction when an electric field is applied in the 3-direction; whereas, the \(d_{31}\) piezoelectric constant represents an expansion (or contraction) of the crystal perpendicular to this applied field. The constant \(d_{31}\) is often called the transverse piezoelectric constant, and \(d_{33}\) the longitudinal piezoelectric constant.

Figure 2.5 illustrates the piezoelectric effect by the two dimensional sketches which models a polar crystal of the perovskite lead titanate in its single domain ferroelectric form. To simplify the description it is assumed that the polarization resides in the Ti\(^{4+}\) ion as in PbTiO\(_3\) and the lead ion displacements are neglected.
Figure 2.5: Two dimensional description of the origin of the piezoelectric response in a single domain PbTiO₃ crystal (a) situation under no field (b) shift of the Ti cation away from the equilibrium position under $E_3$ or $X_3$ (c) shift of the Ti cation back towards the cell center under $E_1$ or $X_1$ (d) tilting of the Ti position giving $\Delta x$ or $\Delta P$ under shear mode [9].

2.3 The PbTiO₃ – PbZrO₃ system

One of the most extensively studied ferroelectric compositional families is the lead zirconate titanate system. Lead zirconate titanate (Pb(ZrₓTi₁₋ₓ)O₃ or PZT) is the solid solution of antiferroelectric lead zirconate (PbZrO₃) and ferroelectric lead titanate
(PbTiO\(_3\)). In general, the term PZT refers to a wide range of compositions with the chemical formula Pb(Zr\(_X\)Ti\(_{1-X}\))O\(_3\) rather than an individual compound.

The crystal structure and properties of PZT strongly depend upon the Ti/Zr ratio. The phase diagram for the PZT system is shown in figure 2.6. The crystal structure of PZT above the Curie temperature (\(T_C\)) is cubic paraelectric phase (\(m\overline{3}m\)), where the B site atoms (Zr or Ti) are located at the center of the unit cell. It should be noted that the Curie temperature varies from 230 to 490 °C depending on the Zr/Ti ratio. After cooling below the Curie temperature, PZT undergoes the phase transition from a paraelectric phase to a ferroelectric phase. The crystal structure of the ferroelectric phase is determined by the Zr/Ti ratio. As the mole% PbTiO\(_3\) in PZT increases, the ferroelectric phase can be orthorhombic, rhombohedral (3\(m\) or 3\(c\)), or tetragonal structure (4\(mm\)).

Figure 2.6: Phase diagram of the PbZrO\(_3\)-PbTiO\(_3\) (PZT) system [2].
In each one of these transitions, the B-site cation (Zr or Ti) is displaced from the center of the unit cell eliminating the center of symmetry, and creating a polar axis. During a cubic to tetragonal transition the B-site cation moves in one of six \(<100>\) directions towards the corners of the oxygen octahedron that surrounds it. In the case of cubic to rhombohedral transition, the B-site cation moves in one of eight \(<111>\) directions towards the centers of the faces of the oxygen octahedron. For a cubic to orthorhombic transition, the B-site cation moves in one of twelve \(<110>\) directions towards the centers of the edges of the oxygen octahedron. Figure 2.7 represents crystal structures and their allowed polar axes.

| Tetragonal Polarization along 6 equivalent \(<100>\) | Rhombohedral Polarization along 8 equivalent \(<111>\) | Orthorhombic Polarization along 12 equivalent \(<110>\) |

Figure 2.7: Crystal structures and their allowed polar axes. The polarization direction is based on the prototype unit cell.

One of interesting features in the PZT systems is a morphotropic phase boundary (MPB) in which rhombohedral and tetragonal phases co-exist at the approximate
composition 52% PbZrO$_3$, 48% PbTiO$_3$. The existence of an MPB may be related to the instability of one ferroelectric phase against another upon a critical composition change. It has been experimentally observed that the maximum values of the dielectric constant and the piezoelectric coefficients of PZT ceramics at room temperature occur on this boundary. In addition, the maximum value of the remanent polarization is also observed near MPB (Zr/Ti = 56/44). It is suggested that the availability of eight possible directions of ferroelectric polarization in the rhombohedral phase along with six in the tetragonal phase in the phase coexistence region around MPB can substantially enhance the alignment of otherwise randomly oriented ferroelectric domains on the application of a strong dc poling field. Similar experimental results have been reported in other binary piezoelectric ceramic systems, such as the PbTiO$_3$-PbSnO$_3$ and the PbTiO$_3$-PbHfO$_3$ system [2, 10]. Figure 2.8 shows the dielectric constant, the piezoelectric coefficient, and remanent polarization of PZT bulk ceramics [2, 10]. Phenomenological study shows the enhanced dielectric constant and piezoelectric coefficients near MPB irrespective of crystallographic orientation because of large Curie constant at this composition [11], while spontaneous polarization does not exhibit maximum value near MPB [12](see figure 2.9). However, it should be noted that lack of single crystal data may lead to errors during derivation of materials’ constant through phase diagram composition in this theory.

Recent investigations regarding the structure of PZT near MPB suggest that the phase diagram of PZT is more complex than previously thought [13-15]. For example, synchrotron x-ray powder diffraction study by Noheda et al. have revealed the existence of an unexpected low temperature monoclinic phase of PZT at the MPB composition,
which provides the bridge between the tetragonal and rhombohedral phases and facilitates polarization rotation [13]. Furthermore, it has been suggested that the local (microscopic) structure of a PZT near MPB differs strongly from its average (macroscopic structure) [13-15]. Although the origin of physical and electrical phenomena near MPB is still not clear, current literature says that the maximum piezoelectric response near MPB is clearly expected for both PZT ceramics and single crystal based on the experimental observation and theoretical calculation.

2.4 Domains

In ferroelectric crystals, uniform alignment of electric dipoles only occurs in certain regions of a crystal, while in other regions of the crystal spontaneous polarization exists in different direction. Such regions with uniform polarization are called ferroelectric domains. The interface between two domains is called the domain wall [1].

Domain structures in ferroelectric materials are formed in an attempt to minimize their free energy. On cooling down through $T_c$, ferroelectric distortion of the unit cell results in both spontaneous polarization and spontaneous strain in the structure. Due to the existence of surfaces, imperfections and mechanical constraints on ferroelectric crystals, there are electrostatic and elastic energies associated with the spontaneous polarization and the spontaneous strain in the crystal. The change of electrostatic and elastic energies is believed to determine the domain structure. The importance of domains
lies in that their response to heat, stress and electric field explain many of the physical and electrical properties of ferroelectrics, their response.

Figure 2.8: Dielectric (a), piezoelectric (b), and polarization (c) behavior of bulk PZT ceramics [2, 10].
Figure 2.9: Dielectric (a), piezoelectric (b), and polarization (c) behavior of single crystal PZT expected from phenomenological theory [11, 12].
The electrostatic energy is caused by depolarization fields resulting from the uncompensated polarization at the surfaces. In another words, insufficient free carriers in a ferroelectric crystal to compensate the spontaneous polarization induce the depolarization field. The electrostatic energy can be reduced by forming an 180° domain structure in the crystal. Since the formation of 180° domain structure introduces domain walls into the crystal, which results in an additional free energy, a stable 180° domain configuration is determined by minimization of the sum of the electrostatic force and domain wall energy.

The elastic energy associated with the development of the spontaneous strain can be reduced by the formation of non-180° domains. In ferroelectric ceramics, this energy is significant since each grain is constrained by its neighbors. Therefore non-180° domain patterns are often formed to minimize the intergranular stresses in ferroelectric ceramics.

Anisotropy in a ferroelectric crystal is obvious large, and the orientation of dipoles usually coincides with the direction of spontaneous polarization. It is hard to envisage that the dipole orientation changes gradually at a domain wall (such as the Bloch wall of magnetic domains), because a domain wall has the normal tendency to decrease its width in order to reduce the electrostatic and elastic energy generated near the wall. Therefore, a ferroelectric domain wall is very thin, and its thickness is about the scale of a few lattice cells.

Figure 2.10 shows 180° domain growth in the tetragonal phase. Domains whose direction of the spontaneous polarization coincides with that of the applied field, are mainly first formed at the surface (a) and then grow through the sample thickness. They begin to expand sideways after fully growing through the sample thickness (b). When the
domains have expanded sideways sufficiently far, they begin to join together (c). This 180° domain boundary motion does not accompany the mechanical strain (deformation), because the antiparallel (180°) domains are in the same spontaneous strain. 90° domain boundary motion is shown in figure 2.11. The domains whose direction of the spontaneous polarization favors that of the electric field, will grow with similar mechanism as 180° domain growth under the applied electric field. It is clear that the 90° domain wall motion accompanies the mechanical strain, since their strain states are not the same with respect to the direction of the electric field.

Figure 2.10: Schematic illustration of 180° domain growth in ferroelectrics.

Figure 2.11: Schematic illustration of 90° domain growth in ferroelectrics.
2.5 Intrinsic and extrinsic contribution

The dielectric and piezoelectric responses at room temperature for ferroelectrics were found to originate from both intrinsic and extrinsic sources [9]. The intrinsic contribution refers to a lattice contribution from the displacements of the individual ions. This corresponds to the response, which would be observed in a single domain single crystal. For example, when the electric field is applied along the polar orientation, the unit cell will change along the field direction, resulting in an induced charge or strain in each domain as illustrated in figure 2.12. This is the origin of the intrinsic contribution to the dielectric and piezoelectric response.

The intrinsic piezoelectric coefficient can be derived based on the phenomenological theory. The strain in ferroelectrics is believed to be induced by the interaction of the spontaneous polarization with the electrostrictive effect [1, 3, 7]. To derive the piezoelectric coefficient for a ferroelectric in terms of materials parameters, we start with the strain ($\varepsilon$) in a ferroelectric material, which is assumed to be

$$\varepsilon = QP^2$$  \hspace{1cm} (2.11)

where $P$ is the spontaneous polarization and $Q$ is the electrostrictive coefficient. A ferroelectric possesses a total polarization ($P$), and assuming a linear relationship with field, the total polarization is given by

$$P = P_s + \varepsilon_o \varepsilon E$$  \hspace{1cm} (2.12)

where $\varepsilon$ is the relative permittivity and $\varepsilon_o$ is the permittivity of free space. Substituting this equation into the strain equation gives
where the first term is the spontaneous strain (field independent), the second term (linear in $E$) is the piezoelectric effect, and the third term (quadratic in $E$) is due to the electrostrictive effect. The coefficient of the linear term is commonly defined as the piezoelectric constant

$$d = 2Qe_0eP_s$$

Therefore, the intrinsic piezoelectric coefficient is expressed as a relation with electrostrictive coefficient, the relative permittivity (dielectric constant) and polarization of ferroelectric materials.

For ferroelectric polycrystalline materials, the intrinsic piezoelectric and dielectric responses in any given direction along the specific direction of interest correspond to the summation of the intrinsic properties of each domain.

On the other hand, the dielectric and piezoelectric responses that originate from sources other than the intrinsic contribution are defined as extrinsic properties of the materials. The extrinsic contributions in ferroelectric materials are mainly due to domain wall motions [9]. As discussed in the previous section, the domain wall motion in response to applied electric fields or stresses result in induced charge or strain, which gives rise to a dielectric and piezoelectric response in addition to the response from the distortion of the unit cell.

It is very important that $180^\circ$ domain walls are purely ferroelectric. They can be excited electrically, and their movements lead to only a polarization change. Therefore $180^\circ$ domain wall motion contributes only to the dielectric properties of ferroelectric
materials. Non-180° domain walls, however, are both ferroelectric and ferroelastic. These domains can be excited by both an externally applied electric field and stress. Non-180° domain wall motion can cause changes in the polarization and the strain, so that it can contribute to both dielectric and piezoelectric properties. In ferroelectric materials, the primary source of the extrinsic piezoelectric response is believed to be the non-180° domain wall motion. Schematic illustration of figure 2.13 indicates that non-180° domain wall motion induces a mechanical strain with an application of electric field.

![Diagram of non-180° domain wall motion](image)

**Figure 2.12:** Intrinsic contribution due to the response from lattice.

![Diagram of extrinsic contribution](image)

**Figure 2.13:** Extrinsic contribution to the piezoelectric response due to non-180° domain wall motion with an applied field for the tetragonal ferroelectrics.
In order to better understand the influence of domain structure and domain wall motion on the material properties of ferroelectrics, the quantitative evaluation of the intrinsic and extrinsic contribution to the dielectric and piezoelectric properties is necessary. Since domain wall motion is thermally activated, it is possible to extract the intrinsic response by freezing out the extrinsic component of the piezoelectric and dielectric response at temperatures approaching zero Kelvin [16, 17]. Therefore the piezoelectric and dielectric properties measured near zero Kelvin could be completely from the intrinsic contribution. Zhang et al. determined the relative contributions of the intrinsic and extrinsic contribution to the dielectric constant and piezoelectric coefficient through measurements of the temperature dependence [16]. Based on the assumption that domain wall motions induced by weak external field do not produce volumetric changes, and thus not contribute to the hydrostatic piezoelectric coefficient, they measured the temperature dependence of the intrinsic and extrinsic properties in PZT (52/48) and doped PZT ceramics. Their results indicated more than 60% of the room temperature dielectric and piezoelectric response in PZT ceramics is from domain wall motion, and both $180^\circ$ and non-$180^\circ$ domain wall contributions are likely present in soft PZT ceramics. The observed intrinsic and extrinsic properties of undoped and donor doped PZT are given in figure 2.14. The temperature dependence of dielectric and electromechanical properties for doped PZT ceramics was investigated from 4.2 K to room temperature [17]. Even though those values at room temperature and temperature coefficients of dielectric and piezoelectric constant were strongly dependent upon the types of dopant, the dielectric and piezoelectric constant of PZT ceramics modified with
different types of dopant exhibit similar values at 4.2 K. This result confirms that doping in PZT ceramics is much more influential on extrinsic properties than intrinsic properties.

![Graphs showing dielectric constant and piezoelectric coefficients for doped PZT and PZT (52/48)](image)

Figure 2.14: The intrinsic and extrinsic values of dielectric constant and piezoelectric coefficients for undoped PZT (PZT-500) and PZT (52/48) [16].

Another experimental method was used to investigate the intrinsic dielectric properties by means of frequency dispersion characteristics of the dielectric constant [18, 19]. At high frequency where domain wall motion cannot respond to the applied signal, i.e., in the range of GHz, the response from the materials is totally due to the intrinsic response, i.e., the ionic motion of the lattice and electronic displacement of the atoms.
This method, however, only provide information on the dielectric properties of a material.

The domain wall contribution to dielectric and piezoelectric properties of ferroelectric materials has been investigated by the characterization of the domain wall response to weak dynamic fields [20-24]. In PZT ceramics, domain wall displacement is known to lead to hysteretic and nonlinear response of dielectric and piezoelectric properties with increasing the amplitude of the driving field. By studying nonlinearity with dielectric loss, bias field dependence and aging behavior, the nonlinear behavior in ferroelectric ceramics was found to be of extrinsic nature due to domain wall motions [20]. A phenomenological theory has been extended to explain the nonlinear behavior [20, 21]. However, there are still some experimental results, which could not be explained by this approach. Nonlinear behaviors of dielectric and piezoelectric response were also observed at subcoercive electric fields or stresses. It was found that this behavior could be described by the Rayleigh law originally discovered in ferromagnetic materials. In this model, randomly distributed defects in the ferroelectric materials interact with the mobile ferroelectric-ferroelastic domain walls and act as pinning centers to limit the irreversible domain wall motions [22-24].

2.6 Phenomenological theory

For better understanding of the properties of multi-domain polycrystalline PZT ceramics of films, knowledge of the properties of single crystal single domain PZT is
required. Since the growth of good quality PZT single crystals for the composition across the phase diagram has not been accomplished, single crystal data are not available from direct measurements. Therefore, thermodynamic phenomenological theory has been developed to predict the single domain (intrinsic) properties of PZT materials.

In case of barium titanate (BaTiO$_3$), since single crystals can be grown and by simple poling procedures converted into single domain states, the properties of a single crystal single domain can be measured at any temperature or stress and a full Landau-Ginsburg-Devenshire phenomenological theory was developed to explain the intrinsic properties of BaTiO$_3$ domains under any set of electric and elastic boundary conditions [25, 26].

However, the situation of PZT system is significantly more complex. Different composition across the phase diagram exhibits antiferroelectric, oxygen octahedral tilted, and simple proper ferroelectric phases. In addition, the growth of good quality PZT single crystals for the composition across the MPB has not been successful in spite of almost 30 years of continuous effort and thus no direct measurements of single domain properties. Only compositions close to pure PbZrO$_3$ and pure PbTiO$_3$ have been grown with adequate quality, which make it to use indirect methods to deduce the thermodynamic constants for other compositions. Since thermodynamic phenomenological theory has been developed to predict single domain properties of PZT, this can be used to separate the intrinsic and extrinsic contributions to the properties of multi domain polycrystalline PZT ceramics and thin films. In addition, it is very useful to understand the behavior of ferroelectric materials by studying the effect of mechanical and electrical boundary conditions on the properties of single domain state.
Table 2.1: The theoretical properties of PZT at 25°C [12].

<table>
<thead>
<tr>
<th>Mole fraction PbTiO₃ in PZT</th>
<th>0.1</th>
<th>0.2</th>
<th>0.3</th>
<th>0.4</th>
<th>0.5</th>
<th>0.6</th>
<th>0.7</th>
<th>0.8</th>
<th>0.9</th>
<th>1.0</th>
</tr>
</thead>
<tbody>
<tr>
<td>$P_s$ ($C/m^2$)</td>
<td>0.57</td>
<td>0.66</td>
<td>0.65</td>
<td>0.50</td>
<td>0.50</td>
<td>0.57</td>
<td>0.64</td>
<td>0.70</td>
<td>0.74</td>
<td>0.75</td>
</tr>
<tr>
<td>$\theta_s$ (deg.)</td>
<td>5.42</td>
<td>7.36</td>
<td>6.64</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\eta_{33}$ or $\eta_{33}'$</td>
<td>219</td>
<td>277</td>
<td>280</td>
<td>295</td>
<td>382</td>
<td>197</td>
<td>116</td>
<td>86.4</td>
<td>72.9</td>
<td>66.6</td>
</tr>
<tr>
<td>$\eta_{11}$ or $\eta_{11}'$</td>
<td>195</td>
<td>262</td>
<td>360</td>
<td>529</td>
<td>1721</td>
<td>498</td>
<td>218</td>
<td>143</td>
<td>121</td>
<td>124</td>
</tr>
<tr>
<td>$g_{33}$ ($10^3$ Vm/N)</td>
<td>26.8</td>
<td>30.9</td>
<td>34.6</td>
<td>41.9</td>
<td>96.6</td>
<td>92.9</td>
<td>101</td>
<td>114</td>
<td>124</td>
<td>134</td>
</tr>
<tr>
<td>$g_{31}$ ($10^3$ Vm/N)</td>
<td>-8.13</td>
<td>-9.53</td>
<td>-11.2</td>
<td>-15.6</td>
<td>-46.0</td>
<td>-33.8</td>
<td>-31.8</td>
<td>-34.2</td>
<td>-37.1</td>
<td>-39.2</td>
</tr>
<tr>
<td>$g_{15}$ ($10^3$ Vm/N)</td>
<td>12.9</td>
<td>14.3</td>
<td>15.5</td>
<td>18.1</td>
<td>41.0</td>
<td>38.4</td>
<td>40.8</td>
<td>44.9</td>
<td>48.6</td>
<td>50.9</td>
</tr>
<tr>
<td>$d_{33}$ ($10^{12}$ C/N)</td>
<td>47.2</td>
<td>72.1</td>
<td>107</td>
<td>189</td>
<td>327</td>
<td>162</td>
<td>104</td>
<td>87.2</td>
<td>81.2</td>
<td>79.2</td>
</tr>
<tr>
<td>$d_{31}$ ($10^{12}$ C/N)</td>
<td>-13.3</td>
<td>-21.6</td>
<td>-38.5</td>
<td>-80.5</td>
<td>-156</td>
<td>-58.9</td>
<td>-32.6</td>
<td>-26.2</td>
<td>-23.9</td>
<td>-23.1</td>
</tr>
<tr>
<td>$d_{33}$ ($10^{12}$ C/N)</td>
<td>1.81</td>
<td>1.27</td>
<td>-7.26</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$d_{33}$ ($10^{12}$ C/N)</td>
<td>24.2</td>
<td>34.5</td>
<td>42.0</td>
<td>60.0</td>
<td>624</td>
<td>169</td>
<td>78.6</td>
<td>57.0</td>
<td>52.1</td>
<td>56.1</td>
</tr>
</tbody>
</table>

The values listed from 0.1 to 0.4 were calculated from the high temperature rhombohedral equations, except for $P_s$ and $\theta_s$ from 0.1 to 0.3 which were calculated from the low temperature rhombohedral equations. The values listed from 0.5 to 1.0 were calculated from the tetragonal equations. $P_s=P_3$ in the tetragonal state, but $P_s=\sqrt{3}P_3$ in the rhombohedral states. $\theta_s=\sqrt{3}\theta_3$ and $\eta_{33}$ were used in the tetragonal state, and $\eta_{11}'$ and $\eta_{33}'$ were used in the rhombohedral state. The piezoelectric coefficients are all based on the cubic axes.

The principles of the thermodynamic phenomenological theory for ferroelectrics is based on the free energy of a ferroelectric single crystal expressed in terms of polarization as an order parameter [3, 5]. It is convenient to treat the ferroelectric phase transition by choosing the elastic Gibbs function as a state function of the ferroelectric system and the temperature, stress, and polarization as independent variables. Amin
developed a modified Devonshire form of the elastic Gibbs free energy function for the simple proper ferroelectric phases of the PZT system [27]. The data of MPB was used to determine the polarization interaction dielectric stiffness coefficients ($\alpha_{12}$, $\alpha_{112}$, and $\alpha_{123}$) in the energy function, and the spontaneous strain determined using high temperature X-ray diffraction was used to calculate the spontaneous polarization. These data were then used to determine the remaining higher order of dielectric stiffness coefficients ($\alpha_{i1}$ and $\alpha_{i11}$), and allowed for the first time the calculation of the dielectric, piezoelectric, elastic, and thermal properties of the ferroelectric single domain state [27]. Later the Curie constant was found to be dependent upon composition by means of a combination of calorimetric and phenomenological data, which make the theory to be modified to explain the compositional dependence of the Curie constant.

A thermodynamic theory for the entire PZT system was developed by Haun et al. using an energy function including additional order parameters such as antiferroelectricity, octahedral tilt angle, and coupling terms [12]. To determine the coefficients of the energy function and their composition dependence, pure and homogeneous PZT powders across the phase diagram were prepared by the sol-gel method. In addition, the physical properties of PZT ceramics fabricated using these powders were used to evaluate the coefficients of the energy function. By solving the energy function, the phase diagram, spontaneous polarization, strain, tilt angle, entropy change, dielectric, piezoelectric, and elastic properties of the PZT system across the entire composition were calculated. Both dielectric susceptibility coefficients and the dielectric anisotropy were found to depend on the composition and reach a maximum at the PZT (50/50) composition because of the peak in the Curie constant at this
composition. In addition, the piezoelectric coefficients \( (d_{ij}) \) were also dependent upon the composition, and showed large peaks near MPB composition due to the large dielectric susceptibility. Table 2.1 summarizes the theoretical properties of PZT.

2.7 Microelectromechanical devices

2.7.1 Piezoelectric bimorph actuators

Flexural mode piezoelectric actuators can be broadly classified into two categories, namely unimorph and bimorph actuators. The typical applications of these types of actuators are loudspeakers, microphone, relays, etc. Wang et al [28] presented an overview of the literature describing the operation and application of unimorph and bimorph actuators. With the application of actuators increasing in the rapidly growing technology of microelectromechanical systems, different types of piezoelectric materials are being considered including PVDF polymers. It should be noted that the compliant nature of thin PVDF actuators implies that it is typically utilized in the double end clamped geometry.

The bimorph consists of two flat piezoelectric elements bonded lengthwise, with or without an elastic support material in between. This bimorph fabrication can be done in two different ways; series and parallel connection of the two piezoelectric elements. Figure 2.15 shows the two different types of bimorph electrical connection without the elastic support element. Figure 2.15(a) is the bimorph fabricated in series connection where the polarization of the two piezoelectric elements are opposite in direction and the actuator is driven by an electrical field between the top and bottom surface electrodes.
Figure 2.15(b) is the bimorph fabricated in parallel connection where the polarization of the two piezoelectric elements is in the same direction and driving voltage is applied across half of the actuator thickness. Assuming the thickness of each piezoelectric element is the same, only half the driving voltage is needed to obtain the same electrical field as that in the case of series.

In designing an actuator for a particular application, there are many parameters to be defined, namely choosing the appropriate piezoelectric material (piezoelectric coefficient), the geometric dimensions (such as the length of the actuator, thickness of the piezoelectric material and thickness of the non-piezoelectric material, if any). Therefore, deriving an analytical expression describing the dynamic actuator’s response would simplify the process of designing the actuator.

Wang et al [28] have derived the quasi-static displacement of single end clamped unimorph and bimorph actuators. The analytical expression for calculating the quasi-static displacement cannot be applied at dynamic resonance. Smits et al [29-31] have described the dynamic behavior of single end clamped bimorph and unimorph actuators. It should also be noted that the analytical expression given [29-31] for calculating the free end deflection of single end clamped actuators tends to infinity at resonance, and therefore does not allow displacement to be calculated at resonance. Also, these and other references have considered piezoelectric actuator with only single end clamping. This condition of single end clamped actuator is applicable only for non-compliant piezoelectric materials and does not apply to soft (compliant) materials such as PVDF polymers and very thin non-compliant piezoelectric materials. A double end clamped configuration is suitable under such circumstances.
As mentioned earlier, with the increase in demand for compliant piezoelectric materials that offer higher dynamic response, analytical equations for dynamic displacement at resonance of a double end clamped actuator will allow optimizing the actuator design for improved performance. This research focused on deriving the dynamic behavior of double end clamped bimorph and unimorph actuators. In addition, this investigation studied the effects of end conditions on the dynamic behavior of these types of actuators, which also plays an important role in the actuator design.

Besides its usefulness for the case of compliant piezoelectric material, the advantage of double end clamped beam over the single end clamped beam may be seen for the case of higher resonant frequency applications. For a given actuator of fixed geometrical dimensions and other external parameters such as the electric field applied etc, held fixed, the resonant frequencies of vibration of the actuator are considerably higher for the double end clamped configuration, though possibly compromising the magnitude of deflection produced. Thus where high resonant frequencies are required, the double end clamped configuration should be considered.

![Diagram](image)

Fig. 2.15 (a). and 2.15 (b). Two types of beam bimorph actuators. (a) series and (b) parallel-connected.
The analytical equations for dynamic displacement of an actuator at resonance can be applied to various actuator geometries and to a variety of piezoelectric materials.

### 2.7.2 Piezoelectric unimorph actuators

The unimorph is one among several types of piezoelectric actuators, and consists of one flat element of piezoelectric element bonded lengthwise with an elastic support material. The typical applications of these types of actuators are seen as loudspeakers, microphone, relays, etc. Wang et al [28] presented an overview of the literature explaining the operation and application of unimorph and bimorph actuators.

Unlike bimorph actuators, unimorph actuators are unsymmetrical along the thickness of the actuator as shown in figure 2.16. Bimorphs offer larger deflection compared to that of unimorph for the same dimensions of the actuator, but the unimorph design has its own advantage over that of bimorph. For an actuator with fixed dimensions, the resonance frequency and the beam deflection at resonance can be varied by bonding suitable elastic support material, which from a fabrication standpoint is easier than that of the bimorph. When an electric field is applied to the actuators with the field applied parallel to the polarization of the piezoelectric element, the piezoelectric element expands lengthwise, whereby the elastic material restrains the piezoelectric element, producing bend as shown in figure 2.17. From the analytical expression for the dynamic displacement of the actuator at resonance, which is discussed later in this thesis, the desired dynamic properties of the actuator can be achieved by proper choices of materials and thickness of the piezoelectric support material.
Fig. 2.16 is the schematic representation of unimorph actuator with the piezoelectric element on top of the non-piezoelectric element

In designing unimorph actuator for a particular application, there are many parameters to be defined, namely choosing the appropriate piezoelectric material, *i.e.*, piezoelectric coefficient, the geometric dimensions like the length of the actuator, thickness of the piezoelectric material and the corresponding non-piezoelectric material. Therefore, it is necessary to derive an analytical equation that assists in the design of the actuator for specific applications.

Wang *et al* [28] have derived the quasi-static displacement of single end clamped unimorph and bimorph actuators. The analytical expression for calculating the quasi-static displacement cannot be applied at the dynamic resonance. Smits *et al* [29-31] have shown the dynamic working of single end clamped bimorph and unimorph actuators. It should also be noted that the analytical expression given [29-31] for calculating the free end deflection of single end clamped actuators tends to infinity at resonance, therefore, the tip displacement at resonance becomes indeterminate. Furthermore, these and other
references have considered piezoelectric actuator with only single end clamped. This condition of the single end clamped actuator is applicable only for non-compliant piezoelectric materials and does not apply to soft (compliant) materials such as PVDF polymers and very thin non-compliant piezoelectric materials. A double end clamped configuration is suitable under such circumstances.

2.7.3 Device applications

Microelectromechanical systems (MEMS) is an emerging field that has attracted much attention due to its potential to revolutionize many commercial and industrial products such as miniature instrumentation, surgical tools, military devices, sensors and actuators [32-37]. Microelectromechanical systems (MEMS) refer integrated micro-devices or systems combining electrical and mechanical components, which are fabricated using integrated circuit (IC) batch processing techniques. The success of MEMS as a key technology in the twenty-first century depends in no small part on the solution of materials issues associated with the design and fabrication of complex devices.

The integration of functional materials such as piezoelectric materials with silicon promises to add versatility to the MEMS, which leads to new functionalities. Piezoelectric microdevices have been the field of interest since the beginning of the 1990s [32, 33], because incorporating a piezoelectric material into a MEMS device may improve the utility of the system as the moving component could provide its own actuation or sensing function through either the direct or indirect piezoelectric effect. This may result in better integration of the system and simplicity in the design.
For example piezoelectric actuators offer significant advantages over both electrostatic and electromagnetic actuators [37, 38]. With motors, the greater the energy density that can be stored in the gap between rotor and stator, the greater the potential for converting to torques or useful work. With electrostatic motors, the maximum energy density storable in the air gap is determined by $E_b$, the maximum electric field before breakdown (approximately $3 \times 10^8 \text{ V/m}$ for 1 $\mu$m gaps). Energy density for electrostatic motor can be calculated from the following equation:

$$E_e' = \frac{\varepsilon_{\text{air}} E_b^2}{2}$$

Where $\varepsilon_{\text{air}}$ represents the permittivity of free space. This results in an energy density of $4 \times 10^5 \text{ J/m}^3$. With a piezoelectric motor made from PZT, the dielectric constant is three orders of magnitude larger ($\sim 1000$) than air. Therefore, the energy density also should be three orders of magnitude larger if the maximum electric field before breakdown is same to a electrostatic motor. The magnetic energy density, $E_m'$, stored in an air gap is given by:

$$E_m' = \frac{B^2}{2\mu_o}$$

<table>
<thead>
<tr>
<th>Principle</th>
<th>Maximum energy density</th>
<th>Special drive condition</th>
</tr>
</thead>
<tbody>
<tr>
<td>Magnetic</td>
<td>$9.5 \times 10^5 \text{ J/m}^3$</td>
<td>1.5 T</td>
</tr>
<tr>
<td>Shape memory alloy</td>
<td>$10.4 \times 10^6 \text{ J/m}^3$</td>
<td>1.4 W/mm3</td>
</tr>
</tbody>
</table>

Table 2.2: Comparing energy densities for magnetic, shape memory alloy, electrostatic, and piezoelectric actuation [37, 38].
where $B$ is the magnitude of the magnetic flux density in the gap and $\mu_o$ the magnetic permeability of free space. With magnetic actuators the gap energy density by pushing $B$ into saturation (about $1.5 \, T$) leads to $9.5 \times 10^5 \, J/m^3$. The energy density stored for the three actuation types reviewed and that of shape memory alloy are compared in table 2.2. Obviously, piezoelectric motors have a distinct energy advantage. In addition, other advantages include low voltage operation, no leviation, and better holding torque [38].

Ferroelectric materials based on PZT are very attractive for MEMS applications since they have large piezoelectric coefficients and electromechanical coupling coefficients. In comparison with commonly used piezoelectric materials such as ZnO, PZT has more than one order of magnitude larger piezoelectric coefficients than those of ZnO (bulk $d_{33} \sim 12 \, pC/N$) [39]. In addition, PZT films can be poled in the direction of interest due to the reorientable polar axis, which can alleviates the requirement of oriented thin film deposition like ZnO. Even though many demonstrations of piezoelectric microdevice utilizing PZTs on silicon substrate have been reported, the usage of ferroelectric materials in MEMS devices was much delayed by integration difficulties [32, 33, 40]. The difficulties arise from less flexibility of underneath materials and non-trivial etching process. Since ferroelectric material require strong oxidation atmosphere, there is limitation in choosing metal electrodes such as Ti, Zr, and Ta. In addition, ferroelectric materials are easily attacked by most common liquid etchant,
which is commonly used for selective etching in MEMS technique. Thus additional process step is required to protect ferroelectric material. Even in physical etching technique, the properties is easily degraded, it always increase the number of process step, which may limit cost competition.

Despite of integration difficulty, during the past decade, much progress has been made to the development of piezoelectric PZT films for MEMS applications. These piezoelectric devices using PZT thin films include beam actuators [41, 42], probes for atomic force microscopy [43, 44], ultrasonic micromotors [45, 46], micropumps [47], ultrasonic transducers for medical applications [48], and IR detector arrays [49]. This list demonstrates that ferroelectric materials are promising candidates for MEMS applications.

2.8 References


[41] M. Hoffmann et al, to be published in Proc. of IEEE.


Chapter 3. Dielectric characterization of Ba(Zr$_{0.35}$Ti$_{0.65}$)O$_3$ thin films on copper foil

3.1 Introduction

Tunable dielectrics have been extensively studied in the application of RF/microwave field for potential devices such as tunable filters, phase shifters and voltage-controlled oscillators. A detailed review is given by Tagantsev et al [1]. These applications require highly tunable dielectric properties at operating temperatures and voltages. Among various materials, the perovskite-type (Ba$_{1-x}$Sr$_x$)TiO$_3$ (BST) is a popular composition of study for this application [2-4]. Introduction of strontium into BaTiO$_3$ shifts the Curie temperature below room temperature [5] offering high dielectric constant, low leakage current, and low dielectric dispersion against frequency of operation. Therefore, BST is a suitable material for applications requiring high dielectric constant and high tunability (operated at room temperature). However, it is important to understand that a high dielectric constant material cannot be used at higher frequencies of operation due to the limitations of the effective impedance. Therefore, a similar material with high tunability and low leakage, but with a low dielectric constant, will be appropriate for high frequency applications.

It has been shown [6] that by substituting Ti with equimolar Zr in the B-lattice site of the BaTiO$_3$ (ABO$_3$) family, the Zr$^{4+}$ ion being more stable and a larger ionic radius than Ti$^{4+}$, expands the perovskite lattice. Therefore, the conduction due to the electrons...
jumping from Ti$^{3+}$ to Ti$^{4+}$ (if any) will be suppressed by the substitution of Ti with Zr. It has also been observed that introduction of Zr into the BaTiO$_3$ shifts the Curie temperature below 120°C (the approximate Curie temperature of BaTiO$_3$). Interestingly, it is reported in the literature [7-9], for $x>0.27$ (Ba(Zr$_x$,Ti$_{1-x}$)O$_3$), a typical relaxor-like behavior is observed in these Zr doped BaTiO$_3$.

Recent literature [8-14] includes the investigation of Ba(Zr$_x$,Ti$_{1-x}$)O$_3$ (denoted as BZT or BTZ) on platinized silicon wafers for tunable microwave applications. Though low loss materials can be prepared on platinum surfaces, this technology is not suitable for embedded devices given the cost of platinum and the high series resistance offered by thin platinum electrodes. As an alternative approach, several groups have demonstrated the preparation of high permittivity dielectrics on thin metal foils [15-18]. For voltage tunable capacitor applications involving higher frequencies, performance is typically limited by series resistance losses of the substrates and electrodes and not by the dielectric losses. Thus, the type of the electrode is of critical importance. The series resistance loss at higher frequencies can be avoided by using copper foil as the electrode and substrate.

Kingon *et al* [19] have demonstrated the integration of lead based Pb(Zr,Ti)O$_3$ on bare copper foil for microelectronic applications. Ihlefeld *et al* [20] have shown the integration of high dielectric constant BaTiO$_3$ on copper substrate for embedded passive applications.

This work extends the integration of multifunctional oxide (BZT) on bare copper, which acts as a substrate and bottom electrode. The highly tunable dielectric BZT was
prepared using chemical solution deposition (CSD) technique. This method extends the possibility of integrating highly tunable, low dielectric constant materials into printed wiring board technology and demonstrates the processes economically and environmentally amicable with the cost competitive industry.

This discussion reports the dielectric constant, dielectric loss and tunability of Ba(Zr_{0.35}Ti_{0.65})O_3 thin films on copper foil prepared by chemical solution techniques.

### 3.2 Experimental section

BZT thin films were prepared by a chemical solution or sol-gel method. Barium acetate [Ba(CH_3COO)_2], zirconium (IV) propoxide [Zr(OC_3H_7)_4], and titanium isopropoxide [Ti(OC_3H_7)_4] were used as starting materials. Acetic acid and methanol was used as solvent. A precursor solution was prepared by dissolving Barium acetate in heated glacial acetic acid. Zirconium (IV) propoxide and titanium isopropoxide were added into the solution. 2-pentanedione was added as a chelating agent. The solution was mixed and refluxed for 3 hours. The bulk solution was diluted to 0.3 molar with methanol. The diluted solution was left for 24 hours for aging process.

The solution was spin coated onto 18 μm thick bare copper foils at 3000 rpm for 30 seconds followed by a hot-plate drying step at 250 °C for 7 minutes. Further pyrolysis was done at 500 °C for 5 minutes at controlled \( pO_2=10^{-15} \) atm. This value of partial pressure of oxygen during annealing of BZT thin films on copper foil was chosen to avoid oxidation of copper. From the thermodynamic stability conditions, copper metal is
stable without oxidizing in region C as shown in figure 3.1. Annealing the BZT thin films on copper substrates at an elevated $pO_2$ results in copper oxidation, which is the region A and B as shown in the figure. The oxygen partial pressure was determined using an oxygen sensor inserted into the furnace. Controlled amounts of forming gas with nitrogen were passed inside the furnace to lower the partial pressure of oxygen. After two layers of spin coating, the films were crystallized by annealing at 900 °C for 30 minutes in a reducing atmosphere (region C) of $pO_2=10^{-11}$ atm. This process was repeated to achieve the desired thickness. The thickness of the BZT thin film on copper foil used in this report was 0.75 microns. A final reoxidation anneal was performed at 600 °C with a controlled oxygen pressure=10$^{-7}$ atm $O_2$ for 1 hour. Platinum top electrodes ~ 10$^{-4}$ cm$^2$ and 100 nm thick, were deposited by DC magnetron sputtering through a shadow mask.

![Thermodynamic representation of Cu-Cu$_2$O and Cu$_2$O-CuO stability conditions. Region A is the annealing condition within the regime where the oxides of copper are](image)

Fig. 3.1: Thermodynamic representation of Cu-Cu$_2$O and Cu$_2$O-CuO stability conditions. Region A is the annealing condition within the regime where the oxides of copper are
stable. Region B is within the stable regime of Cu$_2$O. Region C is within the regime where Cu is non-oxidizing.

The crystalline phases of the thin films were identified by x-ray diffraction (XRD) technique using Bruker AXS D-5000 x-ray diffractometer equipped with a GADDS area detector. The grain size, as determined from the surface morphology, was measured using atomic force microscopy. Dielectric properties, including dielectric constant and loss tangent were measured with a Hewlett-Packard 4192A Impedance Analyzer and an MMR Technologies Inc. cryogenic temperature stage. Dielectric constant versus voltage curves were obtained with DC biases ranging from –10 V to 10 V with an oscillator amplitude of 0.05 V and a 100 kHz frequency. Dielectric constant versus temperature was measured in the range of 100 to 400 K at zero bias.

### 3.3 Results and discussion

Figure 3.2 shows the XRD patterns formed from the BZT thin films on copper foil substrate. The pattern exhibits narrow peak widths, random orientation, and a low flat background noise level. As evident from the XRD pattern, no observable amount of copper oxides (Cu$_2$O and CuO) were formed during the crystallization and pyrolysis processes, consistent with the thermodynamic stability criteria as shown in figure 3.1.
Fig. 3.2. X-ray diffraction pattern (Cu Kα radiation) of BZT thin film on Cu foil.

Figure 3.3 shows the surface morphology of the crystallized BZT thin film on copper foil collected from atomic force microscopy (AFM) in a 2 µm by 2 µm scan size. We can observe a uniform grain size of about 0.1 µm. The difference in brightness in the image is due to the superimposed roughness of copper foil observed while imaging from atomic force microscopy.

Fig. 3.3. Two micron by two micron atomic force microscopic topography image of BZT thin film on copper foil (The difference in the color on the surface is due to the superimpose roughness of copper foil).
The ultimate test of the device quality of the films lies in the electrical properties. The dielectric constant and dielectric loss of the BZT thin films on copper foil as a function of applied DC voltage at 100 kHz is shown in figure 3.4. The dependence of the dielectric constant on the applied DC voltage can be observed, which reveals the nonlinear nature of the BZT thin films on copper foil. The dielectric constant measured at zero bias and 100 kHz was approximately 540. The tunability of the dielectric constant can be calculated from figure 3.4 and it was approximately 51% at 133 kV/cm measured at room temperature. Zhai et al [8,9] reported the dielectric permittivity of Ba(Zr$_{0.35}$Ti$_{0.65}$)O$_3$ thin-film prepared by sol-gel method deposited on platinized Si substrates to be around 120 at zero bias measured at 1 MHz. The corresponding dielectric tunability was 42% at 600 kV/cm. The differences in the value of dielectric permittivity reported can be due to the substrate effect or the processing conditions, which will change the average grain size and thereby changing the dielectric response. Similar results on the dielectric permittivity of BZT on platinized Si substrates have been reported [13,14,21,24] in the literature.
Fig. 3.4. Voltage dependence of dielectric constant and loss tangent of BZT thin film on copper foil at room temperature.

Figure 3.5 shows the variation of tunability of the BZT thin film on copper foil with the external DC bias at room temperature. It is very useful to note that the dielectric loss of the BZT thin film maintained to be low at high electric fields. Such a behavior is useful for tunable applications.
Fig. 3.5. Voltage dependence of tunability of BZT thin film on copper foil at 500 KHz.

Figure 3.6 shows the dependence of the dielectric constant and dielectric loss on the applied frequency. It is evident that the dielectric constant remains unchanged to acceptable accuracy with varying frequency in the range of 10 kHz to 1 MHz.

Fig. 3.6. Frequency dependence of dielectric constant and loss tangent of BZT thin film on copper foil in the range of 10 KHz to 1 MHz.
Zhai et al [7] have shown the ferroelectric to paraelectric transformation for the BZT thin films to be lower than room temperature. This conclusion is consistent with our observation. Figure 3.7 shows the dielectric constant and dielectric loss dependence on the on temperature at zero bias. The approximate value of the Curie temperature for Ba(Zr$_{0.35}$,Ti$_{0.65}$)O$_3$ was measured to be 220 K at 500 kHz. The phase transition for thin film BZT grown by CSD process is higher than the bulk transition temperature [8]. The shifts of transition temperature in films can be attributed to the strain accumulated within the BZT films. We can presume an orthorhombic to tetragonal phase transition near the horizontal regime of the curve where the slope of the curve vanishes.

![Graph showing temperature dependence of dielectric constant and loss tangent for BZT thin film on copper foil at 500 KHz and zero bias.](image)

**Fig. 3.7.** Temperature dependence of the dielectric constant and loss tangent of BZT thin film on copper foil at 500 KHz and zero bias.
3.4 Conclusion

BZT thin films were prepared on bare copper foil substrate by CSD process. Appropriate crystallizing conditions that prevent oxidation of the copper foil substrate were demonstrated. The films were of the perovskite phase. From the voltage dependent dielectric measurements, very high tunability (up to 51%) was observed. The dielectric loss was noted to be very low value at the operating electric field. This was achieved without the use of oxygen barriers or any interfacial layer. The dielectric constant being much lower compared to that of BaTiO$_3$ or (Ba,Sr)TiO$_3$, BZT finds itself a promising candidates as a high tunability, low capacitance density dielectric material for high tunable/low dielectric RF/microwave frequency applications and for embedded concepts in printed wiring board.

3.5 References

Chapter 4. PZT thin films directly on copper foil for piezoelectric, dielectric and ferroelectric applications

4.1 Introduction

The requirement that ferroelectric materials be processed at high temperatures in relatively oxidizing atmospheres led to the traditional use of noble metal electrodes in conjunction with these materials for ferroelectric, piezoelectric and dielectric applications. However, there has been a strong desire to utilize base metal electrodes, primarily for cost reasons. This drive has been led by the multilayer capacitor industry, where the shear number of capacitors produced has driven the significant replacement of noble metals by Ni-based electrodes in BaTiO$_3$-based capacitors [1-4]. This shift has not been simple, and has necessitated significant dopant engineering, and has been accompanied by a necessary reduction in processing temperatures and changes in the sintering atmospheres. However, for BaTiO$_3$-based capacitors, there is advantage in the fact that the partial pressures under which reduction of TiO$_2$ and BaO occur, are many orders of magnitude lower than the conditions under which Ni oxidizes. In other words, there is a relatively large processing window available, which has assisted the development of robust manufacturing routes.

Referring now to thin films of BaTiO$_3$-based materials, Ihlefeld et al [5] have shown the possibility of integrating high permittivity BaTiO$_3$ thin films directly on Cu
foil, with these materials being aimed at embedded passive applications in high density printed wiring boards. Dawley et al [6] have undertaken analogous studies on Ni substrates. Laughlin et al [7] successfully demonstrated the preparation of BST (barium strontium titanate) thin films prepared by sputtering directly on Cu foil for tunable capacitors for RF/microwave applications. The very large values of permittivity achieved, combined with high resolution cross-sectional transmission electron microscope studies, confirmed the lack of interaction between the BST film and Cu, and the presence of a high quality interface. Srinivasan et al (in preparation) has demonstrated the integration of analogous BZT (barium zirconate titanate) thin films on Cu foil, for tunable dielectric devices where lower permittivity and greater reliability is required.

The situation for Pb-based ferroelectrics is significantly more difficult, due to the propensity of PbO to reduce under substantially less reducing conditions (ie at higher pO₂ values). This has been a source of frustration for researchers who desire to utilize PbTiO₃- and Pb(Zr,Ti)O₃-based materials for ferroelectric, dielectric, and piezoelectric applications.

For PZT in piezoelectric ceramic applications, Randall et al [8] have investigated the use of barrier coatings, for example, around the Ni particles that form the electrode.

This work addressed this issue in developing PZT-based dielectric films for embedded passive capacitors [9-12]. These were produced on Cu foil, allowing the integration into low cost, high density, printed circuit boards. In this case, the approach adopted was to utilize a Ni/Ni-phosphide interface layer between the Cu foil and the PZT. The Ni acted as an oxygen getter and barrier by partially forming NiO. While this NiO did react with the PZT, creating a series-connected low permittivity interface layer, it
resulted in a system highly desirable for the embedded capacitor application. In particular, it resulted in a linear dielectric, with the trade-off in reduced capacitance density being entirely acceptable for most embedded capacitor applications.

For other applications, in particular where higher dielectric permittivity is desired, for tunable dielectrics, or for ferroelectric switching and piezoelectric applications, this Ni interface layer is not the optimum solution, as the interface layer is deleterious to these properties. Thus, this work re-addresses the issue of whether PZT can be deposited directly on Cu electrodes, without deleterious reactions. For convenience, Cu foil was utilized as both the bottom electrode and the substrate. This is also the preferred approach for the case where the resultant device (capacitor, varactor, sensor) is to be integrated directly into a printed circuit board. PZT films were prepared by chemical solution deposition techniques [13] directly on copper foils, as described in the Methods section. The oxygen partial pressures were carefully controlled throughout processing, as described.

The crystalline phases of the thin films were identified by x-ray diffraction technique using a Bruker AXS D-5000 x-ray diffractometer equipped with a GADDS area detector. The grain size was measured using atomic force microscopy. Dielectric properties, including capacitance density and loss tangent were measured with a Hewlett-Packard 4192A Impedance Analyzer. Capacitance versus voltage curves were developed with DC biases ranging from –10 V to 10 V with an oscillator amplitude of 0.05 V and a 100 kHz frequency. Ferroelectric polarization-field hysteresis loops of the PZT capacitors were measured using a Radiant Technologies RT 66A ferroelectric tester. Polarization fatigue was characterized at 500 kHz and ±5 V.
Figure 4.1. Thermodynamic representation of Cu-Cu₂O, Cu₂O-CuO and Pb-PbO stability conditions. The dashed line gives the observed pO₂ during heating and cooling to the stable anneal regime, indicated as region C. Region A is the sintering annealing condition within the regime where the oxides of copper and lead are stable. Region B is within the stable regime of Cu₂O and PbO. Region C is within the regime where PbO is stable and Cu is non-oxidizing. Region D (D1 and D2) is within the stable regime for Cu and Pb (non-oxidizing). All regions mentioned above are at 650°C.

4.2 Experimental section

PZT (52/48) solutions were prepared by standard routes [13] using Pb-acetate trihydrate, Zr-n-propoxide and Ti-isopropoxide as starting materials. Acetic acid and n-propanol were used as chelating agent and solvent, respectively. The Zr-n-propoxide and Ti-isopropoxide were mixed and stirred for 30 minutes. Acetic acid was added to the
mixture to modify the precursors. Normal propanol was then added followed by the addition of Pb acetate trihydrate while continuously stirring the solution. Fifteen mole percent lead excess was used to compensate for the lead loss during the crystallization step. The entire solution was heated up to 85~100 °C to dissolve the Pb precursor more efficiently. After the solution was cooled down, further addition of n-propanol, acetic acid, and water were carried out to control system dilution and hydrolysis reaction. The precursor solution was mixed for 24 h to ensure improved homogeneity. The wet film was spin coated onto 18 µm thick bare copper foils at 3000 rpm for 30 seconds followed by a hot-plate drying step at 250 °C for 7 minutes. Further pyrolysis was undertaken at 400 °C for 5 minutes at a controlled pO$_2$=10$^{-6}$ atm. The exact pyrolysis conditions are extremely important in determining the resultant phase assemblage and film quality. The oxygen partial pressure was determined using an oxygen sensor inserted into the furnace. Controlled amount of forming gas with nitrogen was passed inside the furnace to lower the partial pressure of oxygen to 10$^{-6}$ atm. Although this concentration of oxygen inside the furnace is conducive to oxidation of copper at 400 °C (see Fig 4.1), the kinetics of copper oxidation is sufficiently slow at 400 °C that copper oxidation was not observed experimentally during this 5 minute pyrolysis. Further lowering the oxygen concentration during pyrolysis led to strong pyrochlore formation [13] and poor film properties. The process, including solution spin, drying, and pyrolysis was repeated once. The films were then crystallized and densified by annealing at 650 °C for 15 minutes at various atmospheres. This is referred as the ‘sintering’ step. The entire process up to and including sintering was repeated once or twice to yield films of the desired thickness. The thickness of PZT in this investigation was around 1.2 µm. Platinum top electrodes ~ 10$^{-4}$
cm² in area and 100 nm thick, were deposited by DC magnetron sputtering through a shadow mask.

4.3 Results and discussion

The conventional wisdom (see, for example, ref 8) is that the conditions for stability of copper against oxidation are incompatible with the stability of PZT against reduction. However, the pO₂ – temperature stability diagram shown in figure 4.1 suggests that this is not the case. Assuming that reduction of PZT can be represented by the conditions for the reduction of PbO, then it can be seen that there is a narrow window of partial pressure of oxygen in which both Cu and PZT should be stable during the elevated temperatures of processing. This hypothesis was tested by annealing thin films of PZT on Cu foil at different pO₂ conditions, at the 650 °C temperature required for densification. These conditions included ‘region A,’ where Cu is expected to oxidize to CuO; ‘region B,’ where oxidation to Cu₂O is expected; the predicted region of stability, ‘region C’; and ‘region D,’ where reduction to Pb is expected.
Figure 4.2: X-ray diffraction pattern (Cu Kα radiation) of PZT thin film on Cu foil. Films were crystallized under different pO₂ conditions at 650 °C. The legend on the right identifies the diffraction peaks labeled in the diffraction pattern. The regions marked as region A, B, C, D1 and D2 indicates different annealing conditions as shown in figure 4.1.
Figure 4.2 shows the XRD patterns formed from the PZT thin films on copper foil substrate crystallized at 650 °C for 15 mins in the different regions of figure 4.1. Region D2 represents lower pO$_2$ than D1. Careful inspection of the figure shows that the measured phase assemblages are fully consistent with the thermodynamic predictions of figure 4.1. This confirms that there is indeed a processing window in which PZT can be processed without oxidation of the Cu electrode. Region C yielded well crystallized, randomly oriented PZT. In region D1, pyrochlore/fluorite phase was evident, and reducing the pO$_2$ even further (region D2), resulted in no evidence of perovskite PZT formation. In that case, ZrO$_2$ was the predominant crystalline phase observed, consistent with the early phase diagrams of the PbO-ZrO$_2$-TiO$_2$ system [14]. At higher oxygen partial pressures (regions A and B), the expected copper oxide phases are observed.

![Figure 4.2 XRD patterns](image)

Figure 4.3. Two micron by two micron atomic force microscopy topography image of PZT thin film surface on copper foil. Average grain sizes are less than 100 nm.
There is a subtlety that needs to be pointed out, which also helps to explain why this simple approach has eluded researchers for decades. It can be observed from figure 4.1 that processing fully within the region of stability requires the pO$_2$ to be controlled within tight, but varying, limits during heating and cooling. This level of pO$_2$ control is not normally undertaken during thin film or ceramic processing. If one were to process in a pure mixture of N$_2$ and O$_2$, without H$_2$O impurity, the oxygen partial pressure would be essentially fixed, independent of temperature. If the oxygen partial pressure were fixed to achieve the appropriate pO$_2$ at 650 °C (region C), it is clear from figure 4.1 that during heating and cooling the sample would be exposed to an excessively high oxygen partial pressure. In our experimental procedure, the oxygen partial pressure at the sintering temperature of 650 °C was achieved using a flow-controlled mixture of constant concentration of forming gas and O$_2$. In this case the pO$_2$ was lower at the reduced temperatures of heating and cooling, as shown in figure 4.1. The figure shows that this approach kept the PZT/Cu sample within the desired pO$_2$ range for much of the processing cycle. At temperatures below about 430 °C, the sample was exposed to conditions that would be expected to result in Cu$_2$O formation. However, as heating and cooling rates were kept rapid, and because of kinetic limitations, the sample showed no evidence of oxidation at these lower temperatures.

Figure 4.3 shows the surface morphology of the crystallized PZT on copper foil. From the surface image, we can observe a small but uniform grain size, with the grain sizes apparently less than 100 nm. The larger scale variation in surface morphology
corresponds to conformal coverage over the Cu foil, which has a larger surface roughness than the PZT film itself.

The ultimate test of the quality of the films, including the Cu/PZT interface, lies in the electrical properties of the films. The dielectric constant and dielectric loss of the PZT thin films (crystallized in region C) on copper foil, as a function of applied DC voltage, is shown in figure 4.4. The dependence of the capacitance density on the applied DC voltage can be observed, from which we can infer the hysteretic and nonlinear nature of the PZT thin films on copper foil. If one were to consider these films for varactor applications [15], the tunability of the dielectric capacitance calculated from the figure 4.4 is approximately 50% at 167 kV/cm, measured at room temperature.

Figure 4.4. Voltage dependence of permittivity and loss tangent of PZT thin film on copper foil, at room temperature.
Figure 4.5 shows a typical polarization hysteresis loop of a PZT thin film on copper foil. This is extremely similar to high quality ferroelectric films of the same Zr/Ti ratios on Pt/SiO₂/Si substrates, and implies that the films display normal ferroelectric switching behavior. These properties would not be achieved with a degraded interface. Note that there was no further (oxidation) anneal after the film-sintering step.

![Polarization-electric field hysteresis loops of PZT thin film capacitor on copper foil. Note that the PZT composition is close to the morphotropic phase boundary.](image)

Figure 4.5. Polarization-electric field hysteresis loops of PZT thin film capacitor on copper foil. Note that the PZT composition is close to the morphotropic phase boundary.

Figure 4.6 shows the fatigue characteristics of a typical Cu/PZT capacitor (Cu as the top electrode) on Cu foil, in comparison with a Pt/PZT capacitor on Pt foil, processed in parallel (i.e. under identical thermal and pO₂ conditions). It can be seen that the Pt/PZT/Pt capacitor displays the normal behavior expected of Pt electrodes. However, the base metal copper does not display this characteristic fatigue. Instead, it displays no apparent fatigue, although there is a superimposed increase in the leakage current, which
is commonly observed for PZT films with oxide electrodes. The results are of interest for both technical applications and reasons of mechanistic understanding of fatigue. With respect to the latter, it provides some additional evidence that suggests it is not the present of oxygen in the electrode that is of fundamental importance, but instead it may be controlled by charge injection across the metal-ferroelectric interface [16]. With respect to the apparent increase in $P_r$ with fatigue, this has been commonly observed with oxide electrodes, in particular in the case where leakage current is observed to increase during fatigue [17-19].

Figure 4.6. Fatigue characteristics of a typical Cu/PZT capacitor (Cu as the top electrode) on Cu foil, in comparison with a Pt/PZT capacitor on Pt foil. The two types of film were processed in parallel (i.e. under identical thermal and $pO_2$ conditions).

Figure 4.7 shows the dependence of leakage current on applied voltage for a typical PZT thin film on copper foil, with a Pt top electrode defining the capacitor.
message is simply that the leakage currents are acceptably low for the majority of applications.

Figure 4.7. Leakage current dependence on applied voltage of PZT thin film on copper foil. Leakage currents increases about one order of magnitude after $10^{10}$ switching cycles.

### 4.4 Conclusion

This work has shown that it is possible to process high quality PZT films on base metal copper substrates. This was achieved without the use of dopants in the PZT, or of oxygen barriers or local oxygen getters. Also, this chapter has shown that it is necessary to control the oxygen partial pressure within the tight limits predicted by a simple thermodynamic model. In order to simplify the processing, an innovative method has been employed to keep the pO$_2$ within acceptable bounds throughout the temperature cycling.
The work has a series of implications. The fact that, processing of PZT on copper has been demonstrated, implies that a copper base metal could be incorporated in a wide variety of PZT materials and applications. For example, the approach could be used for capacitors, embedded varactors, and piezoelectric-based sensors embedded directly into printed circuit boards. Furthermore, it could be adapted to the potentially large market volume PMN-PT (lead magnesium niobate – lead titanate) relaxor solid solutions, which are utilized for multilayer actuators and particular dielectric applications. Additionally, the results suggest that a route can be engineered to achieve PZT/Cu multilayer actuator stacks as long as thermal expansion mismatch issues can be addressed. Care needs to be taken however, to avoid direct reaction between Cu and PZT that occurs at higher temperatures. (Aiying Wu et al, in preparation). An important advantage of copper as an electrode in high frequency applications is the reduction of series resistance (due to the low copper resistivity and the thickness of the foil).

Finally, the work shows possibility of the integration of functional PZT films with the copper metallization of advanced microelectronic devices. In particular, it suggests that Cu electrodes should be considered for ferroelectric nonvolatile memories. Additionally, the fatigue characteristics are similar to those of PZT capacitors with oxide electrodes.

4.5 References

Chapter 5. Effect of copper and platinum as electrodes on PZT thin films for ferroelectric applications

5.1 Introduction

Ferroelectric lead zirconate titanate (PZT) thin films are promising materials utilized in non-volatile memory device applications [1,2]. The major advantages of this PZT material over other ferroelectric materials are its high dielectric constant, lower leakage, fast switching speed and large remanent polarization. However, literatures in the past have reported considerable loss of polarization with repeated switching, which makes the material unsuitable for electronic devices such as ferroelectric capacitors in non-volatile memories [3-6]. This polarization fatigue in ferroelectric thin films is assumed to be due to electromigration of charged defects, domain wall pinning. Thus, efforts have been made to solve this particular issue with respect to the PZT ferroelectric materials. Several attempts, some successful have shown the effect of RuO$_2$ as the electrode on the polarization fatigue life of PZT thin films. However, the leakage current increased by including RuO$_2$ [7-11]. Other attempts include (for example) oxygen plasma treatment on ferroelectric PZT thin films [12]. This method resulted in switched remanent polarization of 76% of the initial value at $10^8$ switching cycles. Thus, many efforts have been focused on issues retaining long-term reliability of PZT capacitors by using oxide electrode materials, modification of standard composition by introducing dopants and developing layered perovskite materials [13-17]. However, these processes have its own
drawbacks such as difficulty in fabrication, increase in leakage current, high processing temperature and compromising on the high remanent polarization and dielectric constant of PZT capacitors. Thus, a simple method of achieving fatigue-free PZT capacitors without the loss of its excellent ferroelectric properties is desirable. Also, there has been a strong desire to utilize base metal electrodes, primarily for cost reasons. This drive has been led by the multilayer capacitor industry, where the shear number of capacitors produced has driven the significant replacement of noble metals by Ni-based electrodes in BaTiO$_3$-based capacitors [18-21]. Kingon et al [22] has shown the integration of PZT thin films directly of Cu foil for several ferroelectric and non-volatile memory applications.

In the present work described in this chapter, an increase in the polarization fatigue life and good polarization saturation of PZT thin films on Cu electrode is shown without compromising the remanent polarization. Also, the leakage current was lowered in the case of copper as the electrodes, under given process conditions. For convenience, bare Pt and Cu foils were used as the substrates in order to directly compare the effect of Cu and Pt electrodes on the ferroelectric properties of PZT.

The crystalline phase of the thin films was identified by x-ray diffraction technique using a Bruker AXS D-5000 x-ray diffractometer equipped with a GADDS area detector. Ferroelectric polarization-field hysteresis loops of the PZT capacitors were measured using a Radiant Technologies RT 66A ferroelectric tester. Polarization fatigue was characterized at 500 kHz and ±5 V.
5.2 Experimental section

PZT (52/48) solutions were prepared by standard routes [23] using Pb-acetate trihydrate, Zr-n-propoxide and Ti-isopropoxide as starting materials. Acetic acid and n-propanol were used as chelating agent and solvent, respectively. Details of the solution preparation of PZT (52/48) is discussed in chapter 4.

a. Platinum as the substrate

The wet film was spin coated onto 18 µm thick bare platinum foils at 3000 rpm for 30 seconds followed by a hot-plate drying step at 250 °C for 7 minutes. Further pyrolysis was undertaken at 400 °C for 5 minutes at atmosphere. The process, including solution spin, drying, and pyrolysis was repeated once. The films were then crystallized and densified by annealing at 650 °C for 15 minutes. In this discussion, we refer to this as the ‘sintering’ step. The entire process up to and including sintering was repeated once or twice to yield films of the desired thickness. The thickness of PZT in this investigation was around 1.2 µm. Platinum or copper top electrodes ~ 10^{-4} cm^2 in area and 100 nm thick, were deposited by DC magnetron sputtering through a shadow mask.

b. Copper as the substrate

The wet film was spin coated onto 18 µm thick bare copper foils at 3000 rpm for 30 seconds followed by a hot-plate drying step at 250 °C for 7 minutes (similar to that on platinum foils). Further pyrolysis was undertaken at 400 °C for 5 minutes at a controlled
$pO_2=10^{-6}$ atm. The exact pyrolysis conditions are extremely important in determining the resultant phase assemblage and film quality. The oxygen partial pressure was determined using an oxygen sensor inserted into the furnace. Controlled amount of forming gas with nitrogen was passed inside the furnace to lower the partial pressure of oxygen to $10^{-6}$ atm. Although this concentration of oxygen inside the furnace is conducive to oxidation of copper at 400 °C (see Fig 4.1), the kinetics of copper oxidation is sufficiently slow at 400 °C that copper oxidation was not observed experimentally during this 5 minute pyrolysis. Further lowering the oxygen concentration during pyrolysis led to strong pyrochlore formation [24] and poor film properties. The process, including solution spin, drying, and pyrolysis was repeated once. The films were then crystallized and densified by annealing at 650 °C for 15 minutes at various atmospheres. The entire process up to and including sintering was repeated once or twice to yield films of the desired thickness. The thickness of PZT in this investigation was around 1.2 µm.

The $pO_2$ – temperature stability diagram shown in figure 4.1. Assuming that reduction of PZT can be represented by the conditions for the reduction of PbO, then it can be seen that there is a narrow window of partial pressure of oxygen in which both Cu and PZT should be stable during the elevated temperatures of processing. This hypothesis was tested by annealing thin films of PZT on Cu foil at different $pO_2$ conditions, at the 650 °C temperature required for densification. These conditions included ‘region A,’ where Cu is expected to oxidize to CuO; ‘region B,’ where oxidation to Cu$_2$O is expected; the predicted region of stability, ‘region C’; and ‘region D,’ where reduction to Pb is expected.
5.3 Results and discussion

Figures 5.1 shows the XRD patterns formed from the PZT thin films on platinum substrate crystallized at 650°C in atmosphere. Figure 4.2 (chapter 4) shows the XRD patterns formed from the PZT thin films on copper substrate crystallized at 650 °C for 15 mins in various regions as indicated in figure 4.1. Region D2 represents lower $pO_2$ than D1. Careful inspection of the figure shows that the measured phase assemblages are fully consistent with the thermodynamic predictions of figure 4.1. The processing conditions and results of PZT thin films on Cu substrates have been discussed in chapter 4.

Fig 5.1: X-ray diffraction pattern of crystallized PZT thin film on Pt foil with no top electrode.

Figure 5.2 shows a comparison of polarization hysteresis loop of a PZT thin film on Pt foil with Pt and Cu as top electrodes (we use the notation X/PZT/Y for PZT processed on X substrate and Y acts as the top conducting electrode). In case of both the
capacitor type, the remanent polarization was measured to be about 28 μm/cm² and the capacitors saturates well at about 45 μm/cm² before fatigue. The polarization fatigue behavior shows no considerable difference due to the effect of top electrodes. Also, as we can see from figure 5.3, the Pt/PZT/Cu capacitor and Pt/PZT/Pt capacitor shows a similar fatigue life. The polarization fatigue was performed on these two types of capacitors at 500 kHz with ±5 V.

Fig 5.2. Electric field dependence of polarization of PZT thin film capacitor on platinum foil.

Note that the composition is close to the so-called morphotropic phase boundary, and remanent polarization values would be expected to be larger for compositions with higher Ti-content. Figure 5.2 shows the polarization hysteresis of PZT thin films on Pt foil and compares the effect of Pt and Cu as top electrode.
Fig 5.3. Polarization fatigue curves for identical capacitors with Pt or Cu as top electrodes.

Figure 5.4 shows the polarization hysteresis loop of a PZT thin films processed parallel on Pt and Cu foils under identical conditions (region C in figure 4.1). It is evident that, the ferroelectric PZT thin film saturates well with a lower hysteresis loss (area of the loop) on copper substrate compared to that on the platinum substrate. Correspondingly, the Cu/PZT/Cu shows a fatigue free behavior until $10^{10}$ cycles of frequency in figure 5.5, where as, the Pt/PZT/Pt shows the characteristic fatigue. Also, the leakage current of Cu/PZT/Cu capacitor was lower than that of corresponding Pt/PZT/Pt (see figure 5.6).
Fig 5.4. Electric field dependence of polarization of PZT thin film capacitor on copper and platinum foil.

Fig 5.5. Polarization fatigue curves for PZT thin films processed on copper and platinum substrates.
Fig 5.6. Comparison of leakage current dependence on applied voltage of PZT thin film on platinum and copper foils with Pt and Cu top electrodes respectively.

Thus, from figures 5.2 and 5.3, it is evident that replacing Pt by Cu as the top electrode has little effect on the ferroelectric properties of PZT based capacitors. However, the effect of Cu as the substrate (or bottom electrode) shows significant improvement in the ferroelectric properties of PZT based capacitors be observed in figure 5.4- figure 5.6. It should be noted that the PZT thin films processed on Pt substrates are identical to that of PZT thin films processed on Cu substrates, which is region C in figure 4.1 and this corresponds to the reducing atmosphere conditions to avoid oxidation of Cu. In general, PZT thin films on Pt substrates are processed under atmosphere or oxidizing conditions to reduce oxygen vacancies in the PZT thin films. Therefore, it is imperative to study the effect of ambient pO$_2$ during sintering on the ferroelectric properties of PZT thin films.
5.3.1 Results on effect of $pO_2$

PZT was prepared by CSD method as explained in chapter 4 followed by depositing on a standard platinized Si substrate. The deposited PZT solution on platinized Si substrate was dried at intermediate temperature prior to final sintering at 650°C under various $pO_2$ conditions. Table 4.1 shows the pO2 values measured during several intermediate processing temperature of PZT thin films on platinized Si substrate.

Table 5.1. PZT thin films on platinized Si substrate processed under various ambient $pO_2$ (see chapter 4 for processing details).

<table>
<thead>
<tr>
<th>Sample name</th>
<th>$pO_2$ at 250°C</th>
<th>$pO_2$ at 400°C</th>
<th>$pO_2$ at 650°C</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Air</td>
<td>Air</td>
<td>1 atm</td>
</tr>
<tr>
<td>2</td>
<td>Air</td>
<td>Air</td>
<td>Air (0.21 atm)</td>
</tr>
<tr>
<td>3</td>
<td>Air</td>
<td>Air</td>
<td>$10^{-3}$ atm</td>
</tr>
<tr>
<td>4</td>
<td>Air</td>
<td>Air</td>
<td>$10^{-12}$ atm</td>
</tr>
<tr>
<td>5</td>
<td>Air</td>
<td>$10^{-6}$ atm</td>
<td>$10^{-12}$ atm</td>
</tr>
</tbody>
</table>

Figures 5.7-5.11 shows the dielectric dependence on applied voltage and frequency of PZT thin films on platinized Si substrate processed under different ambient $pO_2$. Sample name 5 (see table 5.1) represents the processing condition for PZT on Cu substrate to avoid oxidation of Cu. It is clear from the figures 5.7-5.11 that by reducing
ambient pO2 during the final sintering step, the dielectric dependence of PZT does not change significantly.

Figure 5.7.a  
Figure 5.7.b  
Figure 5.7.a. Dielectric dependence on applied voltage of PZT/Pt/SiO2/Si sintered at 650°C and pO2=1 atm. Figure 5.7.b. Dielectric dependence on frequency of PZT/Pt/SiO2/Si sintered at 650°C and pO2=1 atm.
Figure 5.8.a  
Figure 5.8.b

Figure 5.8.a. Dielectric dependence on applied voltage of PZT/Pt/SiO$_2$/Si sintered at 650°C and pO$_2$=0.21 atm. Figure 5.8.b. Dielectric dependence on frequency of PZT/Pt/SiO$_2$/Si sintered at 650°C and pO$_2$=0.21 atm.

Figure 5.9.a  
Figure 5.9.b

Figure 5.9.a. Dielectric dependence on applied voltage of PZT/Pt/SiO$_2$/Si sintered at 650°C and pO$_2$=10$^{-5}$ atm. Figure 5.9.b. Dielectric dependence on frequency of PZT/Pt/SiO$_2$/Si sintered at 650°C and pO$_2$=10$^{-5}$ atm.
Figure 5.10.a
Dielectric dependence on applied voltage of PZT/Pt/SiO₂/Si sintered at 650°C and pO₂=10⁻¹² atm. Figure 5.10.b. Dielectric dependence on frequency of PZT/Pt/SiO₂/Si sintered at 650°C and pO₂=10⁻¹² atm.

Figure 5.11.a
Dielectric dependence on applied voltage of PZT/Pt/SiO₂/Si sintered at 450°C, pO₂=10⁻⁶ atm and 650°C and pO₂=10⁻¹² atm. Figure 5.11.b. Dielectric dependence on frequency of PZT/Pt/SiO₂/Si sintered at 450°C, pO₂=10⁻⁶ atm and 650°C and pO₂=10⁻¹² atm.
The corresponding dielectric tunability with respect to an applied voltage of these PZT thin films processed under different pO\textsubscript{2} conditions is as shown in figure 5.13.

![Graph showing dielectric tunability vs. voltage](image)

**Figure 5.12.** Dielectric tunability dependence of PZT thin films processed under different ambient pO\textsubscript{2}.

**Figure 5.13.a- 5.13.d** shows the ferroelectric polarization hysteresis of PZT thin films processed under different pO\textsubscript{2} conditions.
Figure 5.13. Polarization hysteresis of PZT/Pt/SiO₂/Si sintered at (a) 650°C, pO₂=0.21 atm; (b) 650°C, pO₂=10⁻⁵ atm; (c) 650°C, pO₂=10⁻¹² atm; and (d) 450°C, pO₂=10⁻⁶ atm and 650°C, pO₂=10⁻¹² atm.

Thus, it can be clearly seen that varying pO₂ during sintering in the range considered does not have significant effect on the ferroelectric properties of PZT thin
films. Studied results (S. Srinivasan et al, unpublished results) have shown that the oxygen vacancies created in the PZT thin films is dominated due to the high volatile nature of PbO rather than the ambient partial pressure of oxygen during sintering of PZT thin films. Therefore, the ferroelectric properties of PZT thin films are not significantly different by varying the pO$_2$ during sintering step.

5.4 Conclusion

PZT was prepared and deposited on platinized Si substrate using CSD method. The effect of Cu and Pt as the top and bottom electrodes on the ferroelectric properties of PZT based capacitors were studied. It has been observed that replacing Pt by Cu as the top electrode has little effect on the ferroelectric properties of PZT based capacitors because the Pt bottom electrode dominates the fatigue. The effect of Cu as the substrate (or bottom electrode) shows significant improvement in the ferroelectric properties of PZT based capacitors. The PZT thin films processed on Pt substrate shows a characteristic fatigue whereas the PZT thin films processed on Cu substrate does not fatigue on switching to $10^{10}$ cycles.

The work has a series of implications. The fact that, this work demonstrated the better performance of PZT with copper implies that a copper metal electrode could be incorporated in a wide variety of PZT materials and applications. For example, the approach could be used for capacitors, embedded varactors, and piezoelectric-based sensors embedded directly into printed circuit boards. An additional important advantage
of copper as an electrode in high frequency applications is the reduction of series resistance (due to the low copper resistivity and the thickness of the foil).

5.5 References


Chapter 6. Modeling and characterization: Dynamic behavior of piezoelectric bimorph actuators under various end conditions

6.1 Introduction

Flexural mode piezoelectric actuators can be broadly classified into two categories, namely unimorph and bimorph actuators [1]. The typical applications of these types of actuators are loudspeakers, microphone, relays, etc. Wang et al [2] presented an overview of the literature describing the operation and application of unimorph and bimorph actuators.

With the application of actuators increasing in the rapidly growing technology of microelectromechanical systems, different types of piezoelectric materials are utilized, including PVDF polymers.

The bimorph consists of two flat piezoelectric elements bonded lengthwise, with or without an elastic support material in between. This bimorph fabrication can be done in two different ways; series and parallel connection of the two piezoelectric elements. Figure 6.1 shows the two different types of bimorph electrical connection without the elastic support element. Figure 6.1(a) is the bimorph fabricated in series connection where the polarization of the two piezoelectric elements are opposite in direction and the actuator is driven by an electrical field between the top and bottom surface electrodes. Figure 6.1(b) is the bimorph fabricated in parallel connection where the polarization of the two piezoelectric elements is in the same direction and driving voltage is applied
across half of the actuator thickness. Assuming the thickness of each piezoelectric element is the same, only half the driving voltage is needed to obtain the same electrical field as that in the case of series.

In designing an actuator for a particular application, there are many parameters to be defined, namely choosing the appropriate piezoelectric material (piezoelectric coefficient), the geometric dimensions (such as the length of the actuator, thickness of the piezoelectric material and thickness of the non-piezoelectric material, if any). Therefore, deriving an analytical expression describing the dynamic actuator’s response would simplify the process of designing the actuator. The current approach is to utilize finite element methods.

![Diagram of beam bimorph actuators](1a)  
![Diagram of beam bimorph actuators](1b)

Fig. 6.1(a). and 6.1(b). Two types of beam bimorph actuators. (a) series and (b) parallel-connected.

Wang et al [2] have derived the quasi-static displacement of single end clamped unimorph and bimorph actuators. The analytical expression for calculating the quasi-static displacement cannot be applied at dynamic resonance. Smits et al [3-5] have
described the dynamic behavior of single end clamped bimorph and unimorph actuators. It should also be noted that the analytical expression given [3-5] for calculating the free end deflection of single end clamped actuators tends to infinity at resonance, and therefore does not allow displacement to be calculated at resonance. Also, these and other references have considered piezoelectric actuator with only single end clamping. This condition of single end clamped actuator is applicable only for low-compliance piezoelectric materials and does not apply to soft (compliant) materials such as PVDF polymers and very thin non-compliant piezoelectric materials. A double end clamped configuration is suitable under such circumstances. Table 6.1 summarizes the analytical expressions available for describing bimorph and unimorph actuators. Thus, clearly we see that (see table 6.1) the past literature is missing the analytical expressions for many of the actuator conditions of practical interest.

As mentioned earlier, with the increase in demand for compliant piezoelectric materials that offer higher dynamic response, analytical equations for dynamic displacement at resonance of a double end clamped actuator will allow optimizing the actuator design for improved performance. This work shows the derivation of the dynamic behavior of double end clamped bimorph actuators. The derivation of the double end clamped unimorph is discussed in chapter 7. In addition, our investigation studies the effects of end conditions on the dynamic behavior of these types of actuators, which also plays an important role in the actuator design.

Besides its usefulness for the case of compliant piezoelectric material, the advantage of double end clamped beam over the single end clamped beam may be seen for the case of higher resonant frequency applications. For a given actuator of fixed
geometrical dimensions and other external parameters such as the electric field applied etc, held fixed, the resonant frequencies of vibration of the actuator are considerably higher for the double end clamped configuration, though possibly compromising the magnitude of deflection produced.

Table 6.1. Analytical expression available for bimorph and unimorph actuators under different end conditions.

<table>
<thead>
<tr>
<th>Classification based on type of actuator</th>
<th>Actuator clamping condition</th>
<th>Analytical equations for quasi-static displacement</th>
<th>Analytical equations for dynamic displacement</th>
<th>Analytical equations for dynamic resonant frequencies</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>double end</td>
<td>--</td>
<td>This work-Chapter 6</td>
<td>This work-Chapter 6</td>
</tr>
<tr>
<td></td>
<td>double end</td>
<td>--</td>
<td>This work-Chapter 7</td>
<td>This work-Chapter 7</td>
</tr>
</tbody>
</table>

Thus where high resonant frequencies are required, the double end clamped configuration should be considered. The analytical equations for dynamic displacement
of an actuator at resonance can be applied to various actuator geometries and to a variety of piezoelectric materials.

### 6.2 Bimorph actuator assembly

This section shows the working of series connected beam bimorph actuator, and the derivation of an analytical expression for dynamic displacement at resonant frequencies. The parallel connected beam bimorph actuator differs merely by the expression for dependence of the voltage across each piezoelectric element on the applied external applied field. Otherwise, the same dynamic derivation holds true.

![Diagram of bimorph actuator](image)

**Fig. 6.2.** Bimorph excited by external voltage.

The piezoelectric bimorphs consist of two strips of piezoelectric elements, each with length, ‘L’, width, ‘w’, and thickness, ‘tₚ’, where, assumed that,
\[ L \gg w; \]
\[ L \gg t_p; \]

The two sheets of piezoelectric elements, equal in thickness, are bonded in such a way that their polarization vectors are opposite in direction. In such a situation, on application of an external applied field, \( \vec{E}_3 \), the element with its polarization parallel to \( \vec{E}_3 \) expand lengthwise, and the piezoelectric element with its polarization antiparallel to \( \vec{E}_3 \) contract lengthwise, there by resulting in a bend of the assembly of the beam of both end clamped. The bimorph bending results in a deflection along the z-axis, and this deflection is measured as ‘\( \delta \)’ (center displacement).

The electric field developed across each piezoelectric element due to applied voltage is denoted as \( E_3 \) and is given as:

\[ E_3 = -\frac{V}{2t_p} \]  
(1)

Under the presence of an external voltage, \( V \), when the piezoelectric elements bend, an internal strain is produced in each element, which is equal to in magnitude \( d_{31}E_3 \), and opposite in sign [3].

Therefore, the stress developed on each piezoelectric element is given as:

\[ Stress = -\frac{d_{31}E_3}{S^{E}_{11}} \]  
(2)

Where

\( d_{31} \): piezoelectric constant of the material
$S_{11}^E$: elastic constant.

We investigate the dynamic behavior of the bimorph actuator with no non-piezoelectric element in between the two piezoelectric elements. The bimorph assembly is symmetrical along the interface of the piezoelectric element and the centroidal axis coincides with the interface.

The centroidal axis of the bimorph actuator is given as:

$$Z_c = \frac{\int ZdA}{\int dA} = t_p$$  \hspace{1cm} (3)

The moment of inertia $I_c$ of the bimorph along its centroidal axis is given as:

$$I_c = \left( \frac{wt_p^3}{12} + wt_p(Z_c - \frac{t_p}{2})^2 \right) + \left( \frac{wt_p^3}{12} + wt_p(Z_c - t_p - \frac{t_p}{2})^2 \right)$$  \hspace{1cm} (4)

Substituting for $Z_c$,

$$I_c = \frac{2wt_p^3}{3}$$  \hspace{1cm} (5)

We can assume a superimposed external moment, ‘$M$’ (uniform), which restrains the double end clamped piezoelectric elements from bending. This moment applied has to be the same in magnitude of that produced due to the strain while bending, but opposite in direction.

The moment acting on each piezoelectric element is given as [3]

$$M = \frac{d_{31}E_pwt_p^2}{S_{11}^E}$$  \hspace{1cm} (6)
The equation for the beam curvature under the presence of an external applied moment $M_{[8-11]}$ is given as:

$$E_p I_c \frac{\partial^2 z}{\partial x^2} = M$$  \hspace{1cm} (7)

Where

$E_p$: Young’s modulus of the piezoelectric element,

$z$: deflection of the actuator in the $Z$ axis, which is a function of $x$.

The dynamic motion of a beam takes the form $[11]$

$$\frac{\partial^2 M}{\partial x^2} = -\rho A \frac{\partial^2 z}{\partial t^2}$$  \hspace{1cm} (8)

Where

$\rho$: specific density of the piezoelectric material

$A$: cross sectional area.

From the above two differential equations, it can be observed that

$$E_p I_c \frac{\partial^4 z}{\partial x^4} = -\rho A \frac{\partial^2 z}{\partial t^2}$$  \hspace{1cm} (9)

In (9), the left hand side is a variable of ‘$x$’ and the right hand side is a variable of, ‘$t$’ containing the function $z$. The function $z$ can be written as a product of two independent functions, each of which depends exclusively on one variable. Therefore, we write

$$z = z(x,t) = Z(x)\phi(t)$$  \hspace{1cm} (10)

Substituting $z$ from (10) into (9) gives,
\[ E_p I_c \frac{\partial^4 [Z(x) \Phi(t)]}{\partial x^4} = -\rho A \frac{\partial^2 [Z(x) \Phi(t)]}{\partial t^2} \]  

(11)

By our definition, \( \phi(t) \) does not depend on ‘x’ and \( Z(x) \) does not depend on ‘t’.

Therefore,

\[ E_p I_c \Phi(t) \frac{\partial^4 Z(x)}{\partial x^4} = -\rho A Z(x) \frac{\partial^2 \Phi(t)}{\partial t^2} \]  

(12)

\[ E_p I_c \frac{\partial^4 Z(x)}{\partial x^4} \frac{1}{Z(x)} = -\rho A \frac{\partial^2 \Phi(t)}{\partial t^2} \frac{1}{\Phi(t)} \]  

(13)

Thus, on simplification, it can be seen that, left hand side of (13) shows the variation of ‘z’ on ‘x’, and right hand side shows the variation of ‘z’ on ‘t’, which are mutually exclusive variables and therefore, the two sides are equal to a constant.

To describe the actuator vibration response, let

\[ \Phi(t) = \Phi_o e^{jft} \]  

(14)

Where

\( f \): frequency of vibration of the actuator.

Therefore,

\[ \frac{\partial^2 \Phi(t)}{\partial t^2} = -f^2 \Phi_o e^{jft} \]  

(15)

Hence,

\[ E_p I_c \frac{\partial^4 Z(x)}{\partial x^4} \frac{1}{Z(x)} = -\rho A f^2 \frac{\partial^2 \Phi(t)}{\partial t^2} \frac{1}{\Phi(t)} = \rho A f^2 \]  

(16)

Solving the above differential equation of the order 4,
\[ Z(x) = C_1 \cos \Omega x + C_2 \sin \Omega x + C_3 \cosh \Omega x + C_4 \sinh \Omega x \quad (17) \]

Where,

\[ \Omega^2 = \frac{f}{a} \quad (18) \]

And

\[ a^2 = \frac{E \rho I_c}{\rho A} \quad (19) \]

To solve for the four variables in (15), we need four conditions, which can be obtained from the boundary conditions. It is important to consider the several boundary conditions that could be possible with this type of flexural actuator, because, it can be expected that each and every unique boundary condition yields different dynamic behavior of the actuator, namely the resonance frequency and the dynamic displacement at resonance. In order to perceive the actuator with its possible end conditions, the actuators can be installed in a fixture designed to control whether the actuator ends are free to rotate or translate in different possible boundary conditions. Based on the design of the method of clamping the end of the actuator, the fixture blocks or allows rotation and translation on both or either side of the actuator.

The actuators fixtures are as shown in figure 6.3 allowing for several possible configurations, depending on the application [12].
Fig. 6.3. Fixture showing possible rotation at both end and translation at one end of the actuator.

From the figure 6.3 we can observe that there could be eight different permutations of boundary conditions, as explained in table 6.2.

Table 6.2. Table of eight allowed boundary conditions.

<table>
<thead>
<tr>
<th>End Condition</th>
<th>Rotation at Left end</th>
<th>Rotation at Right end</th>
<th>Translation</th>
<th>Cases considered</th>
</tr>
</thead>
<tbody>
<tr>
<td>B</td>
<td>Yes</td>
<td>No</td>
<td>Yes</td>
<td>No</td>
</tr>
<tr>
<td>C</td>
<td>Yes</td>
<td>Yes</td>
<td>No</td>
<td>Yes</td>
</tr>
<tr>
<td>D</td>
<td>No</td>
<td>Yes</td>
<td>No</td>
<td>Yes</td>
</tr>
<tr>
<td>E (same as D)</td>
<td>Yes</td>
<td>No</td>
<td>No</td>
<td>Yes</td>
</tr>
<tr>
<td>F</td>
<td>No</td>
<td>No</td>
<td>No</td>
<td>Yes</td>
</tr>
<tr>
<td>G (same as B)</td>
<td>No</td>
<td>Yes</td>
<td>Yes</td>
<td>No</td>
</tr>
<tr>
<td>H</td>
<td>No</td>
<td>No</td>
<td>Yes</td>
<td>No</td>
</tr>
</tbody>
</table>

Note: The possible freedom at the ends of the actuator can be broadly classified into rotation and translation. Since, translation at the left end and right end results in the same
boundary conditions, it results in a sum of three different boundary conditions, namely 1. rotation at the left end, 2. rotation at the right end, and 3. translation at either end. This gives a total of 8 possible boundary conditions. Also, the rotation at the left end or right end is similar. Thus, we have 6 possible boundary conditions.

Boundary conditions A, B, G and H involves free translation of the actuator at one end. This offers a higher degree of freedom for the actuator to behave under the dynamic condition. Analytical equations for these types of end conditions (A, B, G and H) have not considered in this work. We therefore consider four different possibilities of the end conditions, namely boundary conditions C, D, E and F. Typically, for MEMS design, end condition F with no translation or rotation is the most important of the possible end conditions.

**End condition C.**

Rewriting (17),

$$Z(x) = C_1 \cos \Omega x + C_2 \sin \Omega x + C_3 \cosh \Omega x + C_4 \sinh \Omega x$$

The end conditions used are:

1. $Z(x=0)=0,$
2. $Z(x=L)=0,$
3. $\frac{\partial^2 Z}{\partial x^2} (\@ x=0)=0,$
4. $\frac{\partial^2 Z}{\partial x^2} (\@ x=L)=0.$

Solving (17) with these 4 boundary conditions [11], we get the expression for the resonance frequencies given as:

$$\sin \Omega L = 0$$

(20)
This gives,

\[ \Omega L = n\pi, \quad n=1,2,3,\ldots \]

From which, the resonance frequency is found to be,

\[ f = \sqrt{\frac{E_p I_c}{\rho A}} \left( \frac{n\pi}{L} \right)^2, \quad n=1,2,3,\ldots \] \quad (21)

The driving condition used is different for odd and even overtones. The condition in the case of odd overtone is:

\[ \frac{\partial^2 Z}{\partial x^2} (x = L/2) = \frac{M_{\text{applied}}}{E_p I_c} \] \quad (22a)

This equation can be modified for an odd overtone as:

\[ \frac{\partial^2 Z}{\partial x^2} (x = L/2n) = \frac{M_{\text{applied}}}{E_p I_c} \] \quad (22b)

Where, “n” indicates the order of resonance.

Solving for the four independent constants \( C_1, C_2, C_3, C_4 \) in (17), the maximum center displacement at odd overtones of the actuator at steady state is given as:

\[ Z(x = L/2) = \frac{M_{\text{applied}}}{E_p I_c \Omega^2} \left( \frac{\cos(\Omega L/2) - \cosh(\Omega L/2)}{\cos(\Omega L/2) + \cosh(\Omega L/2)} \right) \] \quad (23)

So, this offers the time dependent dynamic displacement during one period of actuator oscillation as:

\[ z(x = L/2, t) = \frac{M_{\text{applied}} e^{\text{ift}}}{E_p I_c \Omega^2} \left( \frac{\cos(\Omega L/2) - \cosh(\Omega L/2)}{\cos(\Omega L/2) + \cosh(\Omega L/2)} \right) \] \quad (24)
A similar analytical expression of dynamic displacement at resonance can be derived for even overtones.

The resonances shown are represented in figure 6.4 the various overtones of a double end clamped actuator. It can be seen from figure 6.4 that the center deflection is present only for the odd overtones and not for the even overtones. In case of the even overtones, the maximum deflection does not occur at the center, rather occurs at both sides away from the center of the beam.

![Diagram showing resonances](image)

**Fig. 6.4.** Figure shows multiple overtones of a double end clamped actuator of length L.

**End conditions D and E.**

Assuming symmetry in the actuator assembly, end conditions D and E yields the same dynamic behavior of the actuator. By similar approach as shown for the end condition C, the end conditions for this case are,

1. \( Z(x=0) = 0, \)
2. \( Z(x=L)=0, \)

3. \( \partial Z/\partial x (@x=0)=0, \)

4. \( \partial^2 Z/\partial x^2 (@x=L)=0. \)

Solving (17) with these 4 boundary conditions, we obtain the resonance frequencies as a function given as:

\[
\tan \Omega L = \tanh \Omega L
\]  

(25)

Applying the driving condition (22a) for an odd resonance and solving for the four independent constants \( C_1, C_2, C_3, C_4 \) in (17), we get:

\[
z(x = L/2, t) = \frac{M_{\text{applied}} e^{i\Omega t}}{E_p I_c \Omega^2} \left( \frac{\cos(\Omega L/2) - \cosh(\Omega L/2)}{\cos(\Omega L/2) + \cosh(\Omega L/2)} \right)
\]

**End condition F.**

The end conditions for this case are,

1. \( Z(x=0)=0, \)

2. \( Z(x=L)=0, \)

3. \( \partial Z/\partial x (@x=0)=0, \)

4. \( \partial Z/\partial x (@x=L)=0. \)

Solving (17) with these 4 boundary conditions,

We obtain the resonance frequencies as a function given as:

\[
\cos \Omega L x \cosh \Omega L = 1
\]  

(26)

The maximum center displacement at odd overtones of the actuator at steady state is given as:
\[ Z(x=L/2) = \frac{M_{\text{applied}}}{E_i I_x \Omega^2} \left( \frac{-\cosh^2(\Omega L/2) + \cos^2(\Omega L/2) - 2 \sin(\Omega L/2) \sinh(\Omega L/2)}{\cos^2(\Omega L/2) + \cosh^2(\Omega L/2) - 2} \right) \]  \hspace{1cm} (27)

And the time dependent dynamic displacement during one period of actuator oscillation is:

\[ z(x = L/2, t) = \frac{M_{\text{applied}} e^{i\Omega t}}{E_i I_x \Omega^2} \left( \frac{-\cosh^2(\Omega L/2) + \cos^2(\Omega L/2) - 2 \sin(\Omega L/2) \sinh(\Omega L/2)}{\cos^2(\Omega L/2) + \cosh^2(\Omega L/2) - 2} \right) \]  \hspace{1cm} (28)

Comparison of the dynamic behavior of the bimorph actuator designed with different boundary conditions are shown in figure 6.5.

6.3 Discussion

The approach taken here to derive the analytical equations for resonance of a double end clamped actuator is unique and accurate compared to other methods shown earlier (see for example [3]). Since, there are four unknown constants in equation (17), which is a general form of an equation for the vibration of a beam, we have identified four boundary condition in the case of double end clamped actuator to be able to solve for the resonance frequency, unlike the case for a single end clamped actuator. Using this calculated resonance frequency and applying a driving boundary condition (22a and 22b), one can calculate the dynamic displacement of the actuator at resonance.

It is of interest to compare the dynamic behavior of the bimorph actuators under various boundary conditions. Figure 6.5 shows the trend of dynamic maximum deflection of the actuators versus the resonance frequencies for the four possible boundary conditions.
conditions. As expected, the actuator satisfying the end condition C offers higher displacement because of reduced constraint (due to the freedom of rotation at the end). This is also the reason why it resonates at lower frequency compared with the end conditions D, E and F. Following the end condition C, the end conditions D and E offers the next set of higher dynamic displacement.

Fig. 6.5. Comparison of normalized dynamic performance of the actuator under different flexure end conditions.

The resonance frequency and the dynamic displacement of the bimorph actuator at resonance are normalized with respect to the dynamic displacement of the bimorph actuator at its first resonance under the end condition C. We can observe the highest deflection produced by the actuator under the end condition C followed by the end conditions D, E and F respectively. Correspondingly, the resonance frequency value increases down the same order.
Using the analytical equations derived in the previous section, it is possible to find the dependence of the dynamic behavior of an actuator on its geometric parameters, such as the length, thickness, etc.

Figure 6.6 shows the variation of the dynamic behavior (resonance frequency and center deflection of the actuator at resonance) on the length of the actuator. Figure 6.7 shows the dependence of the resonance frequency and the center deflection of bimorph actuators on the thickness of the piezoelectric element. It is evident from the figure 6.7 that it is possible to design the actuator with respect to the length, depending on the need of the application that requires a desired resonance frequency and corresponding center displacement. Once the length of the actuator is fixed based, on the criteria mentioned, it is also possible to choose the thickness of the piezoelectric element by the similar approach as shown in figure 6.7.

Fig. 6.6. Dependence of first resonance frequency and its dynamic center displacement on bimorph actuator length.
Fig. 6.7. Variation of first resonance frequency and its dynamic center displacement as a function of $t_p/L$ for bimorph actuators.

6.4 Conclusion on analytical modeling

The dynamic behavior of a bimorph actuator under various conditions has been determined. Analytical expressions for the bending resonance and center beam deflection are derived. Using the basic principles of deriving the dynamic case as shown, similar analytical expressions can be derived for any type of geometry of an actuator. Maximum center deflection can be obtained by maximizing the expression for dynamic deflection while varying any of the variables, such as, thickness of the piezoelectric element, thickness of the elastic support, the length of the beam, etc.

It is shown that the dynamic displacement at resonance increases as the number of degrees of freedom increases. As seen in the case of end condition C, the end of the actuators are freely allowed to rotate compared to that of the case of end conditions D, E
and F, and hence, the displacement at their respective resonances are maximum among these four different end conditions. This conclusion is consistent with the experimental results obtained by Mulling et al., [12]. With the same explanation, as the number of degrees of freedom increases, the resonance frequencies are lowered. However, it should be noted that the end condition F (both sides of the beam clamped) is important, as it is the easiest to fabricate in a MEMS device.

6.5 Experimental section

It is of interest to compare the dynamic behavior of the bimorph actuators obtained by analytical modeling versus the experimental data to testify the analytical derivation.

Commercially available PVDF polymer [13] was used as a piezoelectric material in this investigation. The specifications of the actuator considered in this investigation are given in table 6.3.
Table 6.3. Specifications of PVDF polymer bimorph actuator

<p>| | |</p>
<table>
<thead>
<tr>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Piezoelectric Element</td>
<td>PVDF</td>
</tr>
<tr>
<td>Thickness</td>
<td>28 micron</td>
</tr>
<tr>
<td>Width</td>
<td>1 mm</td>
</tr>
<tr>
<td>Length</td>
<td>ranges between 5-20 mm</td>
</tr>
<tr>
<td>Non-piezoelectric Element</td>
<td>Does not exist</td>
</tr>
<tr>
<td>Conducting Electrodes</td>
<td>Au</td>
</tr>
<tr>
<td>Thickness</td>
<td>0.04 micron</td>
</tr>
<tr>
<td>Width</td>
<td>1 mm</td>
</tr>
<tr>
<td>Length</td>
<td>same as piezo element</td>
</tr>
</tbody>
</table>

The experimental apparatus is shown in figure 6.8.

Fig. 6.8. Test apparatus for measuring actuator deflections at a range of applied voltages and various end conditions.
A linear variable differential transformer (LVDT) for measuring changes in position of the actuator surface at its center is mounted in a frame positioned above the actuator fixture. The LVDT core is connected to a steel shaft constrained by a linear bearing at its lower end. Based on its design, the test fixture blocks or allows rotation in the left axle, right axle, or both axles. The means for allowing or blocking translation was available only in the right axle.

On application of an external electric field, the upper piezoelectric layer with its polarization direction parallel to the electric field expands in the direction orthogonal to its poling direction—parallel to its surface, whereas, the lower piezoelectric layer with its polarization direction anti parallel to the electric field contracts in the direction orthogonal to its poling direction. This introduces internal stresses producing an increment of deflection away from flatness. When electrical excitation is stopped, the internal stress produced is absent and hence, the actuator regains its original configuration of flatness.

The applied voltage must be limited to avoid damage to the actuator. This limits the usefulness of reversed polarity operation.

Data from each test were automatically acquired in conjunction with a LabVIEW computer program, which determined measurement range under each condition, then acquired and averaged 20,000 points for each measurement. To calibrate the relationship between LVDT signal and actuator position, a scaling factor (µm/mV or equivalently, mm/V) was determined, using a number of reference thicknesses.
6.6 Results and discussion

In this section, the experimental results were compared with that obtained by analytical modeling. Figure 6.9 shows the variation of resonance frequency at the first and second overtones with respect to the length of the actuator. The applied voltage for this case was 600 V. As it can be observed that the resonance frequency is inversely proportional to the length of the actuator. As the order of the overtone increases, the resonance frequency increases dramatically for small actuators.

![Graph showing variation of resonance frequency with length](image

Fig. 6.9 shows the resonance frequency at first and second overtone versus length of the actuator for the same end condition.

The corresponding center deflection of the actuator at first resonance is shown as in figure 6.10. As explained earlier, the center deflection of double end clamped actuator at even overtones is zero. The experimental results matched close to 10% of the...
analytical modeling whereas the center deflection was slightly away from 10% matching. This is expected due to the limitation of the experimental set up. The tip of the LVDT, which measures the deflection of the actuator, approximates the deflection in the range of 0.5 to 1 mm long. Hence, the accuracy of measuring the center deflection of an actuator decreases considerably for smaller length actuator.

Fig. 6.10 shows the center deflection at first resonance versus length of the actuator for the same end condition

6.7 Conclusion on experimental section

The dynamic behavior of double end clamped bimorph actuators has been determined. Analytical expressions for the bending resonance and center beam deflection were derived and thus used the modeling to compare with the experimental results. The
experimental data was close to the values obtained by analytical modeling. Thus, it is very useful to extract a theoretical data from the analytical modeling prior fabricating the actuators.

6.8 References

Chapter 7. Modeling and characterization: Dynamic behavior of piezoelectric unimorph actuators under various end conditions

7.1 Introduction

The unimorph is one among several types of piezoelectric actuators, and consists of one flat element of piezoelectric element bonded lengthwise with an elastic support material. The typical applications of these types of actuators are seen as loudspeakers, microphone, relays, etc. Wang et al [1] presented an overview of the literature explaining the operation and application of unimorph and bimorph actuators.

Unlike bimorph actuators, unimorph actuators are unsymmetrical along the thickness of the actuator as shown in figure 7.1. Bimorphs offer larger deflection compared to that of unimorph for the same dimensions of the actuator, but the unimorph design has its own advantage over that of bimorph. For an actuator with fixed dimensions, the resonance frequency and the beam deflection at resonance can be varied by bonding suitable elastic support material, which from a fabrication standpoint is easier than that of the bimorph. When an electric field is applied to the actuators with the field applied parallel to the polarization of the piezoelectric element, the piezoelectric element expands lengthwise, whereby the elastic material restrains the piezoelectric element, producing bend as shown in figure 7.2. From the analytical expression for the dynamic displacement of the actuator at resonance, which is shown later in this chapter, the
desired dynamic properties of the actuator can be achieved by proper choices of materials and thickness of the piezoelectric support material.

Fig. 7.1 is the schematic representation of unimorph actuator with the piezoelectric element on top of the non-piezoelectric element

In designing unimorph actuator for a particular application, there are many parameters to be defined, namely choosing the appropriate piezoelectric material, *i.e.*, piezoelectric coefficient, the geometric dimensions like the length of the actuator, thickness of the piezoelectric material and the corresponding non-piezoelectric material. Therefore, it is desirable to have an analytical equation that assists in the design of the actuator for specific applications.

Wang *et al* [1] have derived the quasi-static displacement of single end clamped unimorph and bimorph actuators. The analytical expression for calculating the quasi-static displacement cannot be applied at the dynamic resonance. Smits *et al* [2-4] have
shown the dynamic working of single end clamped bimorph and unimorph actuators. It should also be noted that the analytical expression given [2-4] for calculating the free end deflection of single end clamped actuators tends to infinity at resonance, therefore, the tip displacement at resonance becomes indeterminate. Furthermore, these and other references have considered piezoelectric actuator with only single end clamped. This condition of the single end clamped actuator is applicable only for non-compliant piezoelectric materials and does not apply to soft (compliant) materials such as PVDF polymers and very thin non-compliant piezoelectric materials. A double end clamped configuration is suitable under such circumstances. The dynamic displacement at resonance of double end clamped piezoelectric bimorph actuators was derived (chapter 6) [5]. This chapter explains the dynamic behavior of double end clamped unimorph actuators by deriving analytical equations for the resonance frequency and the dynamic center displacement at resonance.

7.2 Unimorph actuator assembly

This section shows the working of a piezoelectric beam unimorph actuator and the derivation for calculating an analytical expression for dynamic displacement at the resonant frequencies.

The piezoelectric unimorphs consist of one strip of piezoelectric element and another strip of non-piezoelectric element (or elastic support) each with length, ‘L’,
width, ‘w’, thickness of piezoelectric element, ‘tₚ’, and the non-piezoelectric element with thickness, ‘tₘ’, where, assumed that,

\[ L >> w; \quad L >> tₚ, tₘ \]

The piezoelectric element and the non-piezoelectric element are bonded along their length. In such a situation, on application of an external applied field \( \overline{E}_z \), the piezoelectric element with its polarization parallel or antiparallel to \( \overline{E}_z \) expand or contract lengthwise respectively, and the non-piezoelectric element restrains the physical change of the piezoelectric element resulting in a bend of the assembly of the beam actuator of double end clamped as shown in figure 7.2.

Fig. 7.2. Unimorph excited by external voltage.
The thickness of the piezoelectric element (top element) is \( t_p \) and the thickness of the non-piezoelectric element (bottom element) is \( t_m \). The length of the actuator is \( L \) and the external voltage applied is \( V \).

The unimorph bending results in a deflection along the z-axis, and this deflection is measured as ‘\( \delta \)’ (center displacement). The electric field developed across the piezoelectric element due to the applied voltage is denoted as \( E_3 \) and is given as:

\[
E_3 = -\frac{V}{t_p} \tag{1}
\]

Under the presence of an external voltage, \( V \), when the piezoelectric element bends, an internal strain is produced in both the piezoelectric element and the non-piezoelectric element. We can assume to superimpose an external moment, ‘\( M \)’ (uniform), which restrains the actuator from bending. This moment applied has to be the same in magnitude of that produced due to the strain while bending, but opposite in direction. To obtain the various considerations to solve the dynamic case, the strains in the piezoelectric and the elastic support at the interface are equal in magnitude and opposite in sign.

\[
\left( \frac{F_p S_{11}^p}{wt_p} + \frac{t_p M_p}{2 E_p I_p} - d_{31} E_3 \right) + \left( -\frac{F_m S_{11}^m}{wt_m} + \frac{t_m M_m}{2 E_m I_m} \right) = 0 \tag{2}
\]

Where

- \( d_{31} \): piezoelectric constant of the material
- \( S_{11}^p \): elastic constant of the piezoelectric material
- \( S_{11}^m \): elastic constant of the non-piezoelectric material
\( E_p \): Young’s modulus of the piezoelectric material

\( E_m \): Young’s modulus of the non-piezoelectric material.

Here, we assume a positive sign for \( F_p, F_m, M_p \), and \( M_m \) and due course of derivation, the actual sign could be achieved.

Displacement along x-axis is zero.

Therefore,

\[
F_p + F_m = 0
\]  \hspace{1cm} \text{(3)}

Sum of all the moments along the interface is zero.

\[
(M_p + M_m) + \left( -\frac{F_p t_p}{2} + \frac{F_m t_m}{2} \right) = 0
\]  \hspace{1cm} \text{(4)}

Assuming the thickness of the unimorph assembly \( \ll R \), radius of curvature for the bending, then

\[
R = \frac{M_p}{E_p I_p} = \frac{M_m}{E_m I_m}
\]  \hspace{1cm} \text{(5)}

The unimorph assembly is not symmetrical along the interface of the piezoelectric element and the elastic support, like a bimorph assembly, so the centroidal axis does not coincide with the interface, and it has to be calculated. The approach for deriving the resonance frequency and the center displacement of beam unimorph actuator at resonance is similar to that, performed for bimorph [5]. The critical difference in the derivation here is that, the centroidal axis for unimorph does not coincide with the interface layer between the piezoelectric element and the non-piezoelectric element as it does for the
bimorph of two similar piezoelectric elements. Hence, the moment of inertia of a unimorph actuator assembly will be considerably different to that of a bimorph actuator assembly.

\[ Z_c = \int ZdA = E_m wt_m^2 + E_p wt_p \cdot (2t_m + t_p) \]
\[
2(E_m wt_m + E_p wt_p)
\]

(Fig. 7.3. Diagram showing centroidal axis with reference line at Z=0.)

The centroidal axis of the unimorph actuator is given as:

\[
Z_c = \int ZdA = \frac{E_m wt_m^2 + E_p wt_p \cdot (2t_m + t_p)}{2(E_m wt_m + E_p wt_p)}
\]

(6)

The moment of inertia \( I_c \) along its centroidal axis [10] of the unimorph is given as:

\[
I_c = \left( \frac{wt_m^3}{12} + wt_m(Z_c - \frac{t_m}{2})^2 \right) + \left( \frac{wt_p^3}{12} + wt_p(Z_c - (t_m + \frac{t_p}{2})^2) \right)
\]

(7)

Substituting for \( Z_c \),
\[
I_c = \left( \frac{\text{wt}_m^3}{12} + \text{wt}_m \left[ \frac{E_{p} t_p (t_p + t_m)}{2(E_m t_m + E_{p} t_p)} \right]^2 \right) + \left( \frac{\text{wt}_p^3}{12} + \text{wt}_p \left[ \frac{E_{m} t_m (t_p + t_m)}{2(E_m t_m + E_{p} t_p)} \right]^2 \right)
\] (8)

The total strain produced in the piezoelectric element as a function of Z:

\[
S^p = \left( \frac{d_{31} E_{3} S_{11}^p t_m (S_{11}^m t_p^3 + S_{11}^p t_m^3)}{K} \right) - \left( \frac{6d_{31} E_{3} S_{11}^p S_{11}^m t_p t_m (t_p + t_m) [Z - \frac{t_p}{2}]}{K} \right) - d_{31} E_{3} ;
\] (9)

\begin{align*}
0 & \leq Z \leq t_p \\
\end{align*}

And the total strain in the elastic support as a function of Z:

\[
S^m = -\left( \frac{d_{31} E_{3} S_{11}^m t_m (S_{11}^m t_p^3 + S_{11}^p t_m^3)}{K} \right) - \left( \frac{6d_{31} E_{3} S_{11}^p S_{11}^m t_p t_m (t_p + t_m) [Z - \frac{t_m}{2}]}{K} \right) ;
\] (10)

\begin{align*}
0 & \leq Z \leq t_m \\
\end{align*}

Where,

\[
K = 4S_{11}^p S_{11}^m t_p^3 + 4S_{11}^p S_{11}^m t_m^3 + (S_{11}^p)^2 t_m^4 + (S_{11}^m)^2 t_p^4 + 6S_{11}^p S_{11}^m t_p t_m^2
\] (11)

Solving for \( M_p \) and \( M_m \) from the four equations (2), (3), (4) and (5), we get:

\[
\sum M = M_p + M_m \quad \text{(since } F_p \text{ and } F_m \text{ equals zero at resonance)}
\]
\[
\sum M = \frac{d_{31}E_3w(t_p + t_m)(S_{11}t_p^3 + S_{11}^p t_m^3)}{2K}
\]  \hspace{1cm} (12)

\[
M_{\text{applied}} = \sum M
\]  \hspace{1cm} (13)

The equation for the beam curvature [7-10] under the presence of an external applied moment \( M \) is given as:

\[
E_p I_c \frac{\partial^2 z}{\partial x^2} = M
\]  \hspace{1cm} (14)

Where

\( z \) : deflection of the actuator in the Z axis which is a function of \( x \).

The dynamic motion of a beam takes the form [10]:

\[
\frac{\partial^2 M}{\partial x^2} = -\rho^* A \frac{\partial^2 z}{\partial t^2}
\]  \hspace{1cm} (15)

Where

\( \rho^* \): density of the piezoelectric unimorph actuator and can be calculated as:

\[
\rho^* = \frac{\rho_m t_m + \rho_p t_p}{t_m + t_p}
\]  \hspace{1cm} (16)

\( A \): area of the piezoelectric unimorph actuator given as:

\[
A = w(t_m + t_p)
\]  \hspace{1cm} (17)

From the above two differential equations, it can be observed that:

\[
E_p I_c \frac{\partial^4 z}{\partial x^4} = -\rho^* A \frac{\partial^2 z}{\partial t^2}
\]  \hspace{1cm} (18)
In (18), the left hand side is a variable of ‘x’ and the right hand side is a variable of ‘t’ containing the function z. The function z can be written as a product of two independent functions, each of which depends exclusively on one variable. Therefore, we write

\[ z = z(x, t) = Z(x) \phi(t) \]  \hspace{1cm} (19)

Substituting z from (19) into (18) gives,

\[ E_p I_c \frac{\partial^4 [Z(x), \Phi(t)]}{\partial x^4} = -\rho^* A \frac{\partial^2 [Z(x), \Phi(t)]}{\partial t^2} \]  \hspace{1cm} (20)

By our definition, \( \phi(t) \) does not depend on ‘x’ and \( Z(x) \) does not depend on ‘t’.

Therefore,

\[ E_p I_c \Phi(t) \frac{\partial^4 Z(x)}{\partial x^4} = -\rho^* A Z(x) \frac{\partial^2 \Phi(t)}{\partial t^2} \]  \hspace{1cm} (21)

\[ E_p I_c \frac{\partial^4 Z(x) / \partial x^4}{Z(x)} = -\rho^* A \frac{\partial^2 \Phi(t) / \partial t^2}{\Phi(t)} \]  \hspace{1cm} (22)

Thus, on simplification, it can be seen that the left hand side of (22) shows the variation of ‘z’ on ‘x’, and right hand side shows the variation of ‘z’ on ‘t’, which are mutually exclusive variables and therefore, the two sides are equal to a constant.

To describe the actuator vibration response, let

\[ \Phi(t) = \Phi_0 e^{jft} \]  \hspace{1cm} (23)

Where
\( f \): frequency of vibration of the actuator.

Therefore,

\[
\frac{\partial^2 \Phi(t)}{\partial t^2} = -f^2 \Phi_o e^{ift} \tag{24}
\]

Hence,

\[
E_p I_c \frac{\partial^4 Z(x) / \partial x^4}{Z(x)} = -\rho^* A \frac{\partial^2 \Phi(t)}{\partial t^2} / \Phi(t) = \rho^* Af^2 \tag{25}
\]

Solving the above differential equation of the order 4,

\[
Z(x) = C_1 \cos \Omega x + C_2 \sin \Omega x + C_3 \cosh \Omega x + C_4 \sinh \Omega x \tag{26}
\]

Where

\[
\Omega^2 = \frac{f}{a} \tag{27}
\]

And

\[
a^2 = \frac{E_p I_c}{\rho^* A} \tag{28}
\]

To solve for the four variables in (26), we need four conditions, which can be obtained from the boundary conditions. It is important to consider the several boundary conditions that could be possible with this kind of flexural actuator, because, it can be expected that each and every unique boundary condition yield different dynamic behavior of the actuator, namely the resonance frequency and the dynamic displacement at resonance. If we consider the actuator with its possible end conditions, the actuators can be installed in a fixture designed to control whether the actuator end are free to rotate or
translate in different possible boundary conditions. Based on the design of the method of clamping the end of the actuator as in figure 7.4, the fixture blocks or allows rotation and translation on both or either side of the actuator.

Table 7.1. Table of eight allowed boundary conditions.

<table>
<thead>
<tr>
<th>End Condition</th>
<th>Rotation at Left end</th>
<th>Rotation at Right end</th>
<th>Translation</th>
<th>Cases considered</th>
</tr>
</thead>
<tbody>
<tr>
<td>B</td>
<td>Yes</td>
<td>No</td>
<td>Yes</td>
<td>No</td>
</tr>
<tr>
<td>C</td>
<td>Yes</td>
<td>Yes</td>
<td>No</td>
<td>Yes</td>
</tr>
<tr>
<td>D</td>
<td>No</td>
<td>Yes</td>
<td>No</td>
<td>Yes</td>
</tr>
<tr>
<td>E (same as D)</td>
<td>Yes</td>
<td>No</td>
<td>No</td>
<td>Yes</td>
</tr>
<tr>
<td>F</td>
<td>No</td>
<td>No</td>
<td>No</td>
<td>Yes</td>
</tr>
<tr>
<td>G (same as B)</td>
<td>No</td>
<td>Yes</td>
<td>Yes</td>
<td>No</td>
</tr>
<tr>
<td>H</td>
<td>No</td>
<td>No</td>
<td>Yes</td>
<td>No</td>
</tr>
</tbody>
</table>

Fig. 7.4. Fixture shows possible rotation at both the end and translation at one end of the actuator.
From figure 7.4, we can observe that there could be eight different permutations of boundary conditions, explained as shown in table 7.1.

Boundary conditions A, B, G and H involves free translation of the actuator at one end. This offers a higher degree of freedom for the actuator to behave under the dynamic condition. Deriving an analytical equation for these types of end conditions (A, B, G and H) is complicated and is not considered in this discussion.

We therefore consider four different possibilities of the end conditions, namely boundary conditions C, D, E and F.

**End condition C.**

Rewriting (26),

\[ Z(x) = C_1 \cos \Omega x + C_2 \sin \Omega x + C_3 \cosh \Omega x + C_4 \sinh \Omega x \]

The end conditions for this case are:

1. \( Z(x=0)=0 \),
2. \( Z(x=L)=0 \),
3. \( \frac{\partial^2 Z}{\partial x^2} (x=0)=0 \),
4. \( \frac{\partial^2 Z}{\partial x^2} (x=L)=0 \).

Solving (26) with these 4 boundary conditions [10], we get the expression for the resonance frequencies given as:

\[ \sin \Omega L = 0 \]  

(29)

This gives,

\[ \Omega L = n\pi , \quad n=1,2,3, \ldots \ldots \]
From which the resonance frequency is found to be:

\[
f = \sqrt{\frac{E_p I_c\left(\frac{n\pi}{L}\right)^2}{\rho^2 A}}, \quad n=1,2,3,\ldots, \tag{30}
\]

The driving condition for even overtones is:

\[
\frac{\partial^2 Z}{\partial x^2}(x = L/2) = \frac{M_{\text{applied}}}{E_p I_c} \tag{31a}
\]

And the driving condition for odd overtones is:

\[
\frac{\partial^2 Z}{\partial x^2}(x = L/2n) = \frac{M_{\text{applied}}}{E_p I_c} \tag{31b}
\]

Where, “n” indicates the order of resonance.

Solving for the four independent constants \( C_1, C_2, C_3, C_4 \) in (26), the maximum center displacement for odd overtones of the actuator at steady state is given as:

\[
Z(x = L/2) = \frac{M_{\text{applied}}}{E_p I_c \Omega^2} \left( \frac{\cos(\Omega L/2) - \cosh(\Omega L/2)}{\cos(\Omega L/2) + \cosh(\Omega L/2)} \right) \tag{32}
\]

So, this offers the time dependent dynamic displacement during one period of actuator oscillation as:
\[ z(x = L/2, t) = \frac{M_{\text{applied}} e^{\text{int}}}{E_p I_c \Omega^2} \left( \frac{\cos(\Omega L/2) - \cosh(\Omega L/2)}{\cos(\Omega L/2) + \cosh(\Omega L/2)} \right) \]  

(33)

Similarly, the equation for the dynamic displacement for even resonances can be derived. The derivation is not shown in this work.

**End conditions D and E.**

Assuming symmetry in the actuator assembly, end conditions D and E yields the same dynamic behavior of the actuator. By similar approach as shown for the end condition C, the end conditions to this case are:

1. \( Z(x=0)=0 \),
2. \( Z(x=L)=0 \),
3. \( \frac{\partial Z}{\partial x} (@x=0)=0 \),
4. \( \frac{\partial^2 Z}{\partial x^2} (@x=L)=0 \).

Solving (26) with these 4 boundary conditions,

We obtain the resonance frequencies as a function given as:

\[ \tan \Omega L = \tanh \Omega L \]  

(34)

Applying the driving condition (31a) for odd overtones for solving the four independent constants \( C_1, C_2, C_3, C_4 \) in (26), the time dependent dynamic displacement at odd overtones during one period of actuator oscillation as:
\[ z(x = L/2, t) = \frac{M_{\text{applied}} e^{int}}{E_p I_x \Omega_0^2} \left( \frac{\cos(\Omega L/2) - \cosh(\Omega L/2)}{\cos(\Omega L/2) + \cosh(\Omega L/2)} \right) \]

**End condition F.**

The end conditions to this case are:

1. \( Z(x=0)=0, \)
2. \( Z(x=L)=0, \)
3. \( \frac{\partial Z}{\partial x} (@x=0)=0, \)
4. \( \frac{\partial Z}{\partial x} (@x=L)=0. \)

Solving (26) with these 4 boundary conditions,

We obtain the resonance frequencies as an implicit function given as:

\[ \cos \Omega x \cosh \Omega L = 1 \quad (35) \]

Solving for the four independent constants \( C_1, C_2, C_3, C_4 \) in (26) using the three end conditions and one driving condition (31a), the maximum center displacement at odd overtones of the actuator at steady state is given as:

\[ Z(x = L/2) = \frac{M_{\text{applied}}}{E_p I_x \Omega_0^2} \left( -\cosh^2(\Omega L/2) + \cos^2(\Omega L/2) - 2 \sin(\Omega L/2) \sinh(\Omega L/2) \right) \right) \]

\[ \cos^2(\Omega L/2) + \cosh^2(\Omega L/2) - 2 \]

(36)

This offers the time dependent dynamic displacement during one period of actuator oscillation as:

\[ z(x = L/2, t) = \frac{M_{\text{applied}} e^{int}}{E_p I_x \Omega_0^2} \left( -\cosh^2(\Omega L/2) + \cos^2(\Omega L/2) - 2 \sin(\Omega L/2) \sinh(\Omega L/2) \right) \right) \right) \]

\[ \cos^2(\Omega L/2) + \cosh^2(\Omega L/2) - 2 \]

(37)
7.3 Discussion

This section shows the flexural performance of a unimorph actuator, by applying the dynamic expressions derived in the previous section. From (29), (34) and (35), it possible to calculate the resonance frequency of a unimorph actuator under various boundary conditions. Similarly, using the equations (33), (37) derived for the dynamic displacement at resonances, it is possible to obtain a schematic showing the dynamic displacement of a unimorph actuator at resonances as shown in the figure 7.5 for the various boundary conditions.

![Graph](image)

Fig. 7.5. Comparison of normalized dynamic behavior of actuators under different boundary conditions. The displacement refers to the maximum deflection of a double end clamped beam unimorph actuator.
Detailed explanation of the advantages of unimorph actuators is given by Wang et al. [1]. A maximum deflection of the actuator can be obtained by choosing an appropriate thickness ratio of the piezoelectric material and the elastic support. This optimization of the deflection of the actuator can be achieved by maximizing the dynamic displacement equations (33), (37). Figure 7.6 shows the dynamic center displacement and the resonance frequency of the unimorph actuator and the variation of these two dynamic parameters at first resonance with the thickness ratio.

For the same length of the actuator and other external variables held the same, such as the applied electric field etc., the deflection produced by the unimorph type of actuator is less than that produced by bimorph actuators. Furthermore, the maximum deflection produced by unimorph actuator under the optimized thickness ratio is nearly half of the bimorph actuator.

Fig. 7.6. Shows the dynamic response of the actuator with respect to the thickness ratio.
7.4 Conclusions regarding analytical modeling

The dynamic behavior of the double end clamped unimorph types of actuators under various mechanical boundary conditions has been determined. Analytical expressions for the bending resonance and beam deflection are derived. Maximum deflection can be obtained by maximizing the expression for dynamic deflection with varying any of the variables, such as the thickness of the piezoelectric element, the thickness of the elastic support, the length of the beam, etc. A similar approach could be applied to obtain an analytical expression for the dynamic behavior of additional geometries (e.g., circular unimorph disks).

7.5 Experimental section

It is of interest to compare the dynamic behavior of the unimorph actuators obtained by analytical modeling versus the experimental data to testify the analytical derivation.

Commercially available PVDF polymer [12] was used as a piezoelectric material in this investigation. The specifications of the actuator considered in this investigation are given in table 7.2.

The experimental apparatus is shown in figure 7.7.
Fig. 7.7. Test apparatus for measuring actuator deflections at a range of applied voltages and various end conditions.

A linear variable differential transformer (LVDT) for measuring changes in position of the actuator surface at its center is mounted in a frame positioned above the actuator fixture. The LVDT core is connected to a steel shaft constrained by a linear bearing at its lower end. Based on its design, the test fixture blocks or allows rotation in the left axle, right axle, or both axles. The means for allowing or blocking translation was available only in the right axle.

On application of an external electric field, the upper piezoelectric layer with its polarization direction parallel to the electric field expands in the direction orthogonal to its poling direction — parallel to its surface, whereas, the lower piezoelectric layer with its polarization direction anti parallel to the electric field contracts in the direction orthogonal to its poling direction. This introduces internal stresses producing an increment of deflection away from flatness. When electrical excitation is stopped, the internal stress produced is absent and hence, the actuator regains its original
configuration of flatness. The applied voltage must be limited to avoid damage to the actuator. This limits the usefulness of reversed polarity operation.

Data from each test were automatically acquired in conjunction with a LabVIEW computer program, which determined measurement range under each condition, then acquired and averaged 20,000 points for each measurement. To calibrate the relationship between LVDT signal and actuator position, a scaling factor (\(\mu m/mV\) or equivalently, \(mm/V\)) was determined, using a number of reference thicknesses.

<table>
<thead>
<tr>
<th>Table 7.2. Specifications of PVDF polymer bimorph actuator</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Piezoelectric Element</strong></td>
</tr>
<tr>
<td>Thickness</td>
</tr>
<tr>
<td>Width</td>
</tr>
<tr>
<td>Length</td>
</tr>
<tr>
<td><strong>Non-piezoelectric Element</strong></td>
</tr>
<tr>
<td><strong>Conducting Electrodes</strong></td>
</tr>
<tr>
<td>Thickness</td>
</tr>
<tr>
<td>Width</td>
</tr>
<tr>
<td>Length</td>
</tr>
</tbody>
</table>
7.6 Results and discussion

Using the analytical expression for the dynamic displacement of the beam of piezoelectric unimorph actuator, it is possible to optimize the expression by maximizing the dynamic displacement, which gives an idea of choosing the suitable metal shim for unimorph design. By such a process of optimization (see table 7.2), it was found that nickel as an elastic shim with PVDF polymer piezoelectric element offers maximum dynamic deflection of the actuator (see figure 7.8). To study the performance of the unimorph actuators, different thicknesses of nickel were electroplated on one side of the PVDF polymer in order to achieve the actuator assembly.

![Fig. 7.8. Comparison of different metal shims for unimorph.](image)
Table 7.3. Table of different metal shims considered for the unimorph design with its physical constants.

<table>
<thead>
<tr>
<th>Shim Metal</th>
<th>E_p (Young's modulus of piezoelectric element) x10^9 (N/m²)</th>
<th>E_m (Young's modulus of metal shim) x10^9 (N/m²)</th>
<th>ρ_m (mass density of metal shim) x10³ (Kg/m³)</th>
<th>ρ_e (mass density of piezoelectric element) x10³ (Kg/m³)</th>
<th>h (thickness of piezoelectric element) (μm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Au</td>
<td>2.3</td>
<td>7.8</td>
<td>19.3</td>
<td>1.78</td>
<td>28</td>
</tr>
<tr>
<td>Pt</td>
<td>2.3</td>
<td>16.8</td>
<td>21.1</td>
<td>1.78</td>
<td>28</td>
</tr>
<tr>
<td>Ni</td>
<td>2.3</td>
<td>20</td>
<td>8.9</td>
<td>1.78</td>
<td>28</td>
</tr>
<tr>
<td>W</td>
<td>2.3</td>
<td>41.1</td>
<td>19.3</td>
<td>1.78</td>
<td>28</td>
</tr>
<tr>
<td>Cu</td>
<td>2.3</td>
<td>13</td>
<td>8.9</td>
<td>1.78</td>
<td>28</td>
</tr>
<tr>
<td>Al</td>
<td>2.3</td>
<td>7</td>
<td>2.7</td>
<td>1.78</td>
<td>28</td>
</tr>
<tr>
<td>Si</td>
<td>2.3</td>
<td>8</td>
<td>2.3</td>
<td>1.78</td>
<td>28</td>
</tr>
</tbody>
</table>

We now, having selected the material as the elastic shim, perform analytical modeling on the actuator to choose the appropriate geometry, namely, the length of the actuator and the thickness of the elastic support for a given PVDF polymer thickness.

Using the analytical equations, it is possible to find the dependence of the dynamic behavior of an actuator on its geometric parameters, such as the length, thickness, etc.

Figure 7.9 shows the variation of the dynamic response (resonance frequency and center deflection of the actuator at resonance) on the length of the actuator and the thickness of the elastic support, which is nickel in this case.
7.9(a). Frequency response

Figure 7.10 shows the variation of resonance frequency at the first with respect to the length of the actuator. The applied voltage for this case was 600 V. As it can be observed that the resonance frequency is inversely proportional to the length of the
actuator. As the order of the overtone increases, the resonance frequency increases dramatically for small actuators.

Fig. 7.10 shows the resonance frequency at first and second overtone versus length of the actuator for the same end condition.

The corresponding center deflection of the actuator at first resonance is shown as in figure 7.11. As explained earlier, the center deflection of double end clamped actuator at even overtones is zero. The experimental results matched close to 10% of the analytical modeling whereas the center deflection was slightly away from 10% matching. This is expected due to the limitation of the experimental set up. The tip of the LVDT, which measures the deflection of the actuator, approximates the deflection in the range of 0.5 to 1 mm long. Hence, the accuracy of measuring the center deflection of an actuator decreases considerably for smaller length actuator.
Fig. 7.11 shows the center deflection at first resonance versus length of the actuator for the same end condition.

### 7.7 Conclusion on experimental section

The dynamic behavior of double end clamped bimorph actuators has been determined. Analytical expressions for the bending resonance and center beam deflection were derived and thus used the modeling to compare with the experimental results. The experimental data was close to the values obtained by analytical modeling. Thus, it is very useful to extract a theoretical data from the analytical modeling prior fabricating the actuators.

### 7.8 References


Chapter 8. Dynamic response of single end clamped piezoelectric actuators

8.1 Introduction

Chapter 6 and 7 show the dynamic behavior of double end clamped piezoelectric bimorph and unimorph actuators respectively. This type of clamping is utilized for actuators operating at high resonance frequency. Single end clamping is utilized for high deflection or displacement actuators. Wang et al [1] presented an overview of the literature describing the operation and application of unimorph and bimorph actuators.

As described in previous chapters, in designing an actuator for a particular application, there are many parameters to be defined, namely choosing the appropriate piezoelectric material (piezoelectric coefficient), the geometric dimensions (such as the length of the actuator, thickness of the piezoelectric material and thickness of the non-piezoelectric material, if any). Therefore, deriving an analytical expression describing the dynamic actuator’s response would simplify the process of designing the actuator.

Smits et al [2-4] have described the dynamic behavior of single end clamped bimorph and unimorph actuators. However, it should be noted that the analytical expression given [2-4] for calculating the free end deflection of single end clamped actuators tends to infinity at resonance, and therefore does not allow displacement to be calculated at resonance. Thus, clearly we see that the past literature is missing the
analytical expressions for calculating the tip displacement of single end clamped actuator at resonance, which is an important parameter to design the actuators.

8.2 Bimorph actuator assembly

Extending the double end clamped bimorph actuator configuration from chapter 6 to single end clamped, the piezoelectric bimorphs consist of two strips of piezoelectric elements, each with length, ‘L’, width, ‘w’, and thickness, ‘t_p’, where, assumed that,

\[ L \gg w_p \]
\[ L \gg t_p \]

The two sheets of piezoelectric elements, equal in thickness, are bonded in such a way that their polarization vectors are opposite in direction (see figure 8.1). In such a situation, on application of an external applied field, \( E_3 \), the element with its polarization parallel to \( E_3 \) expand lengthwise, and the piezoelectric element with its polarization antiparallel to \( E_3 \) contract lengthwise, there by resulting in a bend of the assembly of the beam of both end clamped. The bimorph bending results in a deflection along the z-axis, and this deflection is measured as ‘δ’ (center displacement).

The electric field developed across each piezoelectric element due to applied voltage is denoted as \( E_3 \) and is given as:
Fig 8.1. Schematic diagram of a single end clamped cantilever bimorph actuator (series-connected).

\[ E_3 = -\frac{V}{2t_p} \]  

(1)

Under the presence of an external voltage, \( V \), when the piezoelectric elements bend, an internal strain is produced in each element, which is equal to in magnitude \( d_{31}E_3 \), and opposite in sign.

Therefore, the stress developed on each piezoelectric element is given as:

\[ \text{Stress} = -\frac{d_{31}E_3}{S_{11}^E} \]  

(2)

Where

\( d_{31} \): piezoelectric constant of the material

\( S_{11}^E \): elastic constant.

This work considers the dynamic behavior of the bimorph actuator with no non-piezoelectric element in between the two piezoelectric elements. The bimorph assembly
is symmetrical along the interface of the piezoelectric element and the centroidal axis coincides with the interface.

The moment of inertia $I_c$ of the bimorph along its centroidal axis is given as (from chapter 6):

$$ I_c = \frac{2wt_p^3}{3} \tag{3} $$

Also, the superimposed external moment can be calculated as shown in chapter 6. The moment acting on each piezoelectric element is given as

$$ M = \frac{d_{31}E_pwt_p^2}{S_{11}} \tag{4} $$

The equation for the beam curvature under the presence of an external applied moment $M$ is given as:

$$ E_p I_c \frac{\partial^2 z}{\partial x^2} = M \tag{5} $$

Where

$E_p$: Young’s modulus of the piezoelectric element,

$z$: deflection of the actuator in the Z axis, which is a function of $x$.

The dynamic motion of a beam takes the form

$$ \frac{\partial^2 M}{\partial x^2} = -\rho A \frac{\partial^2 z}{\partial t^2} \tag{6} $$

Where

$\rho$: specific density of the piezoelectric material

$A$: cross sectional area.
From the above two differential equations, it can be observed that

\[ E_p I_c \frac{\partial^4 Z}{\partial x^4} = -\rho A \frac{\partial^2 Z}{\partial t^2} \]  

(7)

Solving the above differential equation of the order 4 (see chapter 6 for details),

\[ Z(x) = C_1 \cos \Omega x + C_2 \sin \Omega x + C_3 \cosh \Omega x + C_4 \sinh \Omega x \]  

(8)

Where,

\[ \Omega^2 = \frac{f}{a} \]  

(9)

And

\[ a^2 = \frac{E_p I_c}{\rho A} \]  

(10)

To solve for the four variables in (10), we need four conditions, which can be obtained from the boundary conditions.

**End conditions**

Rewriting (8),

\[ Z(x) = C_1 \cos \Omega x + C_2 \sin \Omega x + C_3 \cosh \Omega x + C_4 \sinh \Omega x \]

The end conditions used are:

1. \( Z(x=0)=0 \), (clamped end)
2. \( \frac{\partial Z}{\partial x} (x=0)=0 \), (clamped end)
3. \( \frac{\partial^2 Z}{\partial x^2} (x=L)=0 \), (no moment at free end)
4. \( \frac{\partial^3 Z}{\partial x^3} (x=L)=0 \). (no shear force at free end)
Solving (8) with these 4 boundary conditions, we get the expression for the resonance frequencies given as:

\[ 1 + \cos \Omega L x \cosh \Omega L = 0 \]  
\[ \text{(11)} \]

The driving condition is (the moment is built up at the fixed end):

\[ \frac{\partial^2 Z}{\partial x^2} (x = 0) = \frac{M_{\text{applied}}}{E_p I_c} \]  
\[ \text{(12)} \]

Solving for the four independent constants \( C_1, C_2, C_3, C_4 \) in (8), the maximum tip displacement of the actuator at steady state is given as:

\[ Z(x = L) = \frac{M_{\text{applied}}}{E_p I_c \Omega^2} \left( \frac{\cos \Omega L \sinh \Omega L - \sin \Omega L \cosh \Omega L}{\sin \Omega L + \sinh \Omega L} \right) \]  
\[ \text{(13)} \]

So, this offers the time dependent dynamic displacement during one period of actuator oscillation as:

\[ Z(x = L, t) = \frac{M_{\text{applied}} e^{i \Omega t}}{E_p I_c \Omega^2} \left( \frac{\cos \Omega L \sinh \Omega L - \sin \Omega L \cosh \Omega L}{\sin \Omega L + \sinh \Omega L} \right) \]  
\[ \text{(14)} \]

**8.3 Unimorph actuator assembly**

Similar to the section 8.2, the dynamic tip displacement can be calculated for single end clamped unimorph actuators.

The piezoelectric unimorphs consist of one strip of piezoelectric element and another strip of non-piezoelectric element (or elastic support) each with length, ‘L’, width, ‘w’, thickness of piezoelectric element, ‘\( t_p \)’, and the non-piezoelectric element with thickness, ‘\( t_m \)’, where, assumed that,
The piezoelectric element and the non-piezoelectric element are bonded along their length as shown in figure 8.2. In such a situation, on application of an external applied field \( E_z \), the piezoelectric element with its polarization parallel or antiparallel to \( E_z \) expand or contract lengthwise respectively, and the non-piezoelectric element restrains the physical change of the piezoelectric element resulting in a bend of the assembly.

![Fig. 8.2. Schematic diagram of single end clamped unimorph actuator.](image)

The thickness of the piezoelectric element (top element) is \( t_p \) and the thickness of the non-piezoelectric element (bottom element) is \( t_m \). The length of the actuator is \( L \) and the external voltage applied is \( V \).

The unimorph bending results in a deflection along the z-axis, and this deflection is measured as ‘\( \delta \)’ (center displacement). The electric field developed across the piezoelectric element due to the applied voltage is denoted as \( E_z \) and is given as:
\[ E_3 = -\frac{V}{t_p} \] \hspace{1cm} (15)

Under the presence of an external voltage, \( V \), when the piezoelectric element bends, an internal strain is produced in both the piezoelectric element and the non-piezoelectric element. We can assume to superimpose an external moment, ‘\( M \)’ (uniform), which restrains the actuator from bending. This moment applied has to be the same in magnitude of that produced due to the strain while bending, but opposite in direction. To obtain the various considerations to solve the dynamic case, the strains in the piezoelectric and the elastic support at the interface are equal in magnitude and opposite in sign.

Considering the same boundary conditions as discussed in section 8.2, we obtain the time dependent tip displacement of single end clamped unimorph actuator as

\[ Z(x = L, t) = \frac{M_{\text{applied}}}{E_p I_c \Omega^2} e^{i t} \left( \frac{\cos \Omega L \sinh \Omega L - \sin \Omega L \cosh \Omega L}{\sin \Omega L + \sinh \Omega L} \right) \]

The superimposed external moment and the moment of inertia can be obtained from chapter 7.

### 8.4 Conclusion

The dynamic behavior of single end clamped bimorph and unimorph actuators has been determined. Analytical expressions for the bending resonance and tip deflection were derived.
8.5 References


Chapter 9 Conclusions and Future work

9.1 Conclusions

This study has contributed by demonstrating the possibility of integrating piezoelectric, ferroelectric and dielectric thin films (ceramic oxides and polymers) with flexible copper substrates, which has proved to be critically important in the advanced microelectronic device applications.

As explained in chapter 3, BZT thin films were prepared on bare copper foil substrate by CSD process. Appropriate crystallizing conditions that prevent oxidation of the copper foil substrate were demonstrated. From the voltage dependent dielectric measurements, very high dielectric tunability (up to 51%) was observed. The dielectric loss was noted to be very low value at the operating electric field. This was achieved without the use of oxygen barriers or any interfacial layer. The dielectric constant being much lower compared to that of BaTiO$_3$ or (Ba,Sr)TiO$_3$, BZT finds itself a promising candidates as a high tunability, low capacitance density dielectric material for high tunable/low dielectric RF/microwave frequency applications and for embedded concepts in printed wiring board. From chapter 4, it is wise to conclude that processing high quality PZT films on base metal copper substrates can be performed with ease. In order to simplify the processing, an innovative method has been employed to keep the pO$_2$ within acceptable bounds throughout the temperature cycling.
The work has a series of implications. The fact that, processing of PZT on copper has been demonstrated, implies that a copper base metal could be incorporated in a wide variety of PZT materials and applications. For example, the approach could be used for capacitors, embedded varactors, and piezoelectric-based sensors embedded directly into printed circuit boards. Furthermore, it could be adapted to the potentially large market volume PMN-PT (lead magnesium niobate – lead titanate) relaxor solid solutions, which are utilized for multilayer actuators and particular dielectric applications. Additionally, the results suggest that a route can be engineered to achieve PZT/Cu multilayer actuator stacks as long as thermal expansion mismatch issues can be addressed. An important advantage of copper as an electrode in high frequency applications is the reduction of series resistance (due to the low copper resistivity and the thickness of the foil).

Finally, the work opens up the tantalizing possibility of the integration of functional PZT films with the copper backend metallization of advanced microelectronic devices. In particular, it suggests that Cu electrodes should be considered for ferroelectric nonvolatile memories. Additionally, the fatigue characteristics are similar to those of PZT capacitors with oxide electrodes. Thus the system will provide useful information for a mechanistic study of ferroelectric fatigue.

The dynamic behavior of a bimorph and unimorph actuator under various conditions has been studied. Analytical expressions for the bending resonance and center beam deflection were derived, as explained in chapter 6 and 7. Using the basic principles of deriving the dynamic case as shown in these chapters, similar analytical expressions can be derived for any type of geometry of an actuator. Maximum center deflection can be obtained by maximizing the expression for dynamic deflection while varying any of
the variables, such as, thickness of the piezoelectric element, thickness of the elastic support, the length of the cantilever beam, etc. Analytical expressions for the bending resonance and center beam deflection derived were compared with the experimental results. The experimental data was close to the values obtained by analytical modeling. Therefore, it is very useful to extract a theoretical data from the analytical modeling prior fabricating the actuators.

9.2 Future work

Although the results of this work has opened up the feasibility of using copper as the electrode for several microelectronic applications, it is important to address several issues.

First, the thermal coefficients of expansion of copper and PZT should be understood to avoid any cracking of the ferroelectric films.

Second, it would be beneficial to improve the precursor solution chemistry to reduce the crack formation of the ferroelectric films processed on flexible copper substrates.

Third, it would be useful to extend the integration of copper as the electrodes with other extensively used ferroelectric materials, such as SBT, PLZT etc.

Fourth, effect of partial pressure of oxygen during high temperature processing of PZT should be understood for improving the ferroelectric properties and lower loss required in embedded passive application.