Ferroelectric Thin-Film Active Sensors for Structural Health Monitoring

Bin Lin¹, Victor Giurgiuțiu¹, Zheng Yuan², Jian Liu², Chonglin Chen², Jiechao Jiang³, Amar S. Bhalla⁴, Ruyan Guo⁴
¹Department of Mechanical Engineering, University of South Carolina, Columbia, SC 29208
²Department of Physics and Astronomy, University of Texas at San Antonio, San Antonio, TX 78249
³Department of Materials Science and Engineering, University of Texas Arlington, Arlington, TX 76019
⁴Materials Research Laboratory, Penn State University, University Park, PA 16802

ABSTRACT

Piezoelectric wafer active sensors (PWAS) have been proven a valuable tool in structural health monitoring. Piezoelectric wafer active sensors are able to send and receive guided Lamb/Rayleigh waves that scan the structure and detect the presence of incipient cracks and structural damage. In-situ thin-film active sensor deposition can eliminate the bonding layer to improve the durability issue and reduce the acoustic impedance mismatch. Ferroelectric thin films have been shown to have piezoelectric properties that are close to those of single-crystal ferroelectrics but the fabrication of ferroelectric thin films on structural materials (steel, aluminum, titanium, etc.) has not been yet attempted. In this work, in-situ fabrication method of piezoelectric thin-film active sensors arrays was developed using the nano technology approach. Specification for the piezoelectric thin-film active sensors arrays was based on electro-mechanical-acoustical model. Ferroelectric BaTiO₃ (BTO) thin films were successfully deposited on Ni tapes by pulsed laser deposition under the optimal synthesis conditions. Microstructural studies by X-ray diffractometer and transmission electron microscopy reveal that the as-grown BTO thin films have the nanopillar structures with an average size of approximately 80 nm in diameter and the good interface structures with no inter-diffusion or reaction. The dielectric and ferroelectric property measurements exhibit that the BTO films have a relatively large dielectric constant, a small dielectric loss, and an extremely large piezoelectric response with a symmetric hysteresis loop. The research objective is to develop the fabrication and optimum design of thin-film active sensor arrays for structural health monitoring applications. The short wavelengths of the micro phased arrays will permit the phased-array imaging of smaller parts and smaller damage than is currently not possible with existing technology.

Keywords: Nanopillar, Thin-film, Active sensors, Pulsed laser deposition, Structural health monitoring

1. INTRODUCTION

1.1 Background

Piezoelectric wafer active sensors have been proven a valuable tool in structural health monitoring. Piezoelectric wafer active sensors are able to send and receive guided Lamb/Rayleigh waves that scan the structure and detect the presence of incipient cracks and structural damage. These inexpensive and unobtrusive active sensors can be used in many applications spanning from aerospace to civil infrastructure. However, the current methods for fabrication and installation of these sensors on metallic structures are rather “primitive”: pre-manufactured piezoelectric wafers are adherively bonded to the structural surface. The bonding layer is susceptible to environmental ingestion that may lead to loss of contact with the structural substrate. The bonding layer may also induce acoustic impedance mismatch with detrimental effects on damage detection. To overcome these shortcomings we are proposing to use ferroelectric thin-film active sensors...
Ferroelectric thin films have been shown to have piezoelectric properties that are close to those of single-crystal ferroelectrics, which are an order of magnitude better than common piezoceramics. In addition, the thin films require much smaller poling voltage/ power. They have been successfully integrated in micro/nano electromechanical systems, tunable wireless communication elements, and other modern devices. However, major challenges exist in extending this technology to structural health monitoring since the fabrication of ferroelectric thin films on structural materials (steel, aluminum, titanium, etc.) has not been yet attempted.

1.2 Motivation

The research objective is to develop the fabrication and optimum design of piezoelectric thin-film active sensor arrays for structural health monitoring applications. This interdisciplinary research crosses the engineering and science boundaries and addresses the problem in a coordinated approach focused on understanding the fundamentals aspects of fabricating and using thin-film active sensors on typical structural materials.

2. STATE OF THE ART

2.1 State of the art in structural health monitoring using piezoelectric wafer active sensors

Structural health monitoring (SHM) determines the health of a structure by monitoring over time a set of structural sensors. The mounting costs of maintaining our aging infrastructure can be addressed through SHM systems that will reduce unscheduled repairs while increasing safety and reliability. Built-in SHM system capable of detecting and quantifying damage would increase the operational safety and reliability, would conceivably reduce the number of unscheduled repairs, and would bring down maintenance cost.

The type and efficiency of the SHM sensors play a crucial role in the SHM system success. Ideally, SHM sensors should be able to interrogate the structure and find out its state of health, its remaining life, and the effective margin of safety. Essential in this determination is to find out the presence and extend of structural damage. Currently, structural damage is determined during scheduled inspections with sophisticated nondestructive evaluation (NDE) equipment and extensive labor costs. The challenge of SHM initiative is to develop inexpensive active sensors that can be permanently placed on the monitored structure and assess, continuous or on-demand, the state of structural health. In addition, for air and space vehicle applications, such SHM sensors must be lightweight and miniaturized such as not to compromise the in-flight vehicle performance.

In recent years, considerable progress has been achieved in developing SHM sensors that actively interrogate the structure using guided Lamb and Rayleigh waves. Of particular interest are the piezoelectric wafer active sensors that can emulate the common NDE methods (pitch-catch, pulse-echo, phased array) while being sufficiently small and inexpensive to be permanently left in place inside the monitored structure. Piezoelectric wafer active sensors seem to be the enabling technology for active SHM implementation. However, their durability and survivability is still under investigation. The adhesive bonding currently used to attach them to the structure is a potential weak link. There is an acute need for (a) seamless atomic bond between the piezoelectric active sensor and the structure; and (b) piezoelectric materials enhanced piezoelectric response (coherent crystalline structure and well-oriented domains). This research addresses these needs by developing ferroelectric thin film sensor arrays that will be impervious to environmental attacks. Improved piezoelectric properties will be attained through an ordered crystalline structure with quasi single-domain orientation. In addition, solutions for the miniaturization and integration of the active sensors with the electronic modules in a chip-size unit (such as through layered thin-film technology) will be considered as long term objectives of this research.

2.2 State of the art in piezoelectric thin-films fabrication/characterization

Barium titanate (BaTiO₃) is one of the most important ferroelectric materials, and has attracted much attention for its remarkable properties such as high dielectric constant, good ferroelectric properties, and large electro-optic and non-linear optic coefficients. Especially, BaTiO₃ has excellent piezoelectric properties resulting in the broad applications in the control systems. BTO thin films have been deposited on various substrates using a broad spectrum of techniques.
to provide a platform for many device applications as ferroelectric random access memories, optical modulators, waveguides and microelectromagnetic systems (MEMS)\textsuperscript{5-6}. On the other hand, ferroelectric BTO thin film displayed excellent piezoelectric properties, similar to its single crystal ferroelectrics\textsuperscript{7}. These unique properties enable the BTO thin films to be great candidates for the development of the unobtrusive piezoelectric wafer active sensor arrays for structural health monitoring\textsuperscript{8-9}.

Pulsed laser deposition (PLD) is considered as one of the most powerful and attractive techniques for thin-film growth. PLD has been employed for fabricating: single crystalline ferroelectric (BaSr)TiO\textsubscript{3} thin films used in microwave wireless communication applications, highly ionic conductive (La,Sr)CoO\textsubscript{3} thin films for solid state fuel cell applications, high temperature superconductive YBa\textsubscript{2}Cu\textsubscript{3}O\textsubscript{7} films, ferromagnetic (La,Ca)MnO\textsubscript{3} thin films for magnetoresistance systems, etc. Several epitaxially-grown perovskite-structured films were fabricated and characterized by Chen and Jiang: ferroelectric (BaSr)TiO\textsubscript{3} and (PbSr)TiO\textsubscript{3} thin-films with excellent dielectric tunability and microwave phase shift; giant dielectric constant CaCu\textsubscript{3}Ti\textsubscript{4}O\textsubscript{12}; PbTiO\textsubscript{3}/SrTiO\textsubscript{3} superlattices; self-assembled ferromagnetic (La,Sr)MnO\textsubscript{3} (LSMO) nanorod thin films; SnO\textsubscript{2} thin films for gas sensors.

### 3. SPECIFICATION FOR THIN FILM SENSOR

A simplified 1-D analysis of the piezoelectric wafer active sensor (PWAS) is performed next. This simplified analysis will be then used to predict the perceived benefits of the nano-fabricated ferroelectric thin-film multilayer PWAS.

Consider a piezoelectric wafer of length $l$, width $b$, and thickness $h$ that is undergoing longitudinal expansion ($u_1$) induced by the thickness polarization electric field, ($E_3$) (Figure 1). The electric field is produced by the application of a harmonic voltage, $V(t) = V \cos(\omega t)$, between the top and bottom surfaces (electrodes). Assume that the length, width and thickness have widely separated values ($h \ll b \ll l$) such that the length, width and thickness motions are practically uncoupled. Under the one-dimensional assumptions, the general constitutive equations reduce to the simpler expressions

\begin{align*}
S_1 &= s_{11} \cdot T_1 + d_{31} \cdot E_3 \quad (1) \\
D_3 &= d_{33} \cdot T_1 + e_{33} \cdot E_3 \quad (2)
\end{align*}

At zero mechanical stress $T_1$, the actuated strain is,

\begin{equation}
S_1 = d_{31} \cdot \frac{V(t)}{h} \quad (3)
\end{equation}

From Equation(3), for a given strain, actuating voltage is proportional to thickness. The thinner thickness, the less voltage need to used to actuate the sensor. Figure 2 illustrates the relation between thickness and voltage for a given strain.

The power required to drive the PWAS device is calculated as follows

\begin{equation}
P = \omega C_0 V^2 \quad (4)
\end{equation}
where \( \omega \) is the input frequency, capacitance \( C_0 = N \varepsilon_{33} \frac{A}{h} \), \( N \) is the number of layer, \( A \) is the PWAS surface area \( A = l \cdot b \).

The detail comparison of thickness with voltage, capacitance, charge, power is in the Table 1.

![Figure 2 – Relation of thickness and actuating voltage of ferroelectric PWAS (PZT APC 850 material properties and \( S1= 87.5 \) microstrain were used).](image)

From simulation, the piezoceramic wafer technology must provide high voltage input to achieve a sizable strain effect. This fact is due to the inverse relationship between electric field and wafer thickness for a given voltage. Thin-film can get the same strain with a much lower voltage. Through layering, the power of the device can be amplified many times.

The work on the thin-film multilayer deposition consists of several steps. The first step is to deposit a one-layer ferroelectric film with electrodes. The second step is to deposit a two-layer film with electrodes. The third step is to develop a method for multi-layer ferroelectric film deposition. After that, we can use the multi-layer film array in order to take advantage of the reduced array size. The final step is using the multi-layer thin film to controls the system's data acquisition and communication wirelessly like the radio frequency antenna. The long-term perspective of the proposed project is to develop the scientific basis for a wireless miniaturized system-on-a-chip.

![Figure 3 – Blueprint of multi-layer thin film PWAS array](image)
Table 1 Comparison of thickness and multilayer effects based on PZT APC 850 material properties and S1= 87.5 microstrain at frequency of 100kHz

<table>
<thead>
<tr>
<th>Thickness (µm)</th>
<th>200µm</th>
<th>3µm (100 layers, 30 nm each)</th>
<th>0.06µm (Two layers, 30nm each)</th>
<th>0.03µm (One layer)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Voltage (V)</td>
<td>100</td>
<td>1.5</td>
<td>0.015</td>
<td>0.015</td>
</tr>
<tr>
<td>Capacitance (nF)</td>
<td>3.794</td>
<td>253</td>
<td>2.53×10^6</td>
<td>50600</td>
</tr>
<tr>
<td>Charge (C)</td>
<td>3.974×10^-7</td>
<td>3.974×10^-7</td>
<td>3.974×10^-5</td>
<td>7.589×10^-7</td>
</tr>
<tr>
<td>Power (W)</td>
<td>23.8</td>
<td>0.358</td>
<td>0.358</td>
<td>0.00716</td>
</tr>
</tbody>
</table>

4. THIN FILM FABRICATION

Recently, we have developed a unique technique for the first time to achieve in-situ fabrication of BTO thin films on the typical structural material Ni using PLD system. Ultra-thin interfacial buffer layer NiO was created by in-situ oxidation treatment of Ni tape at high temperature, which would facilitate film growth by bonding the BTO layer onto the Ni substrate.

A KrF excimer PLD system with a wavelength of 248 nm was used to perform the fabrication of ferroelectric BTO thin films on Ni substrates. A layer of NiO with its thickness of 80 nm was synthesized via in situ oxidation treatment of the Ni tape in oxygen atmosphere of 300 Torr at 800°C for 3 min. NiO is rock salt structure with good electrical conductivity, which can be used as conductive electrodes and avoiding the formation of a dead layer formed between the ferroelectric film and metal substrate. It should be pointed out that the rock salt structural NiO has good crystallographic compatibility to the perovskite microstructure of BTO growth. The optimal growth conditions for ferroelectric BTO thin film growth on Ni tape were found to be an energy density of about 2.0 J/cm² with a laser repetition rate of 5Hz in an oxygen pressure of 300 mTorr at 800°C. Stoichiometric BTO target was used for the BTO growth. The typical growth rate and the film thickness were about 6 nm/min and 400 nm, respectively. The experimental setup is shown in Figure 4(a).

5. THIN FILM CHARACTERIZATIONS

5.1 X-Ray Diffraction (XRD)

X-ray diffraction θ – 2θ scan (XRD), transmission electron microscopy (TEM) (both plan-view and the cross-section), and Scanning Probe Microscope (SPM) were employed to characterize the microstructures and crystallinity of the as-grown BTO films. As seen in Figure 4 (b), the XRD pattern from the as-deposited BTO thin film on Ni shows all the
peaks from the polycrystalline BTO phases and Ni substrate. These peak positions suggest that the Ni substrate is cubic phase and the BTO film belongs to the tetragonal phase. The relative stronger intensity from the (002) diffraction in the BTO film indicates that the BTO film has a slightly preferred c-axis orientation rather than the randomly oriented grains.

5.2 Transmission Electron Microscope

The microstructure of the as-deposited BTO film was further investigated by TEM. Figure 5(a) is a bright-field TEM image with an inset of the selected-area electron diffraction (SAED) pattern of a plan-view BTO/NiO/Ni sample, showing a crystalline grain structure of the film. The size of the grains in Figure 5(a) varies from 35 nm to 160 nm in diameter, while the majority grains have a size of about 90 nm in diameter. The inset SAED pattern of the film shows sharp diffraction rings, indicating that all the grains are in plan randomly oriented. The structure of the as-grown BTO film was identified as a tetragonal structure with a space group of p4mm and lattice parameters of a=3.992 Å and c=4.036 Å. For example, the inner 6 diffraction rings 1, 2, 3, 4, 5 and 6 have a lattice spacing of 4.0 Å, 2.8 Å, 2.3 Å, 2.0 Å, 1.8 Å, 1.64 Å and 1.4 Å, respectively, which can be identified as the (001), (101), (111), (002), (102) and (112) reflection of the tetragonal BTO. Figure 5(b) is a cross-sectional TEM image showing the interface structures of the BTO films on NiO buffered Ni tapes and the inset is the low magnification image of the films. The BTO film has a thickness of about 500 nm and consists of nanopillar structures. Most of the nanopillars extend from the film/substrate interface to the film surface with a length of about 500 nm and show a lateral dimensions from 30 nm to 100 nm (inset), which is close to the value obtained from the plan-view TEM. An intermediate layer that can be identified as NiO was observed between the BTO film and Ni substrate (Fig. 2b) indicating that a NiO oxidized layer was successfully produced prior to the deposition of BTO film. The BTO film is found to be very well bound the NiO layer with a sharp interface in between. The NiO layer has a thickness of about 80 nm and a clear interface with respect to the Ni substrate.

5.3 Piezoelectric Responsive

To further understand the physical property of the as-grown BTO films, Piezoelectric Responsive Microscopy (PRM) was employed to study the multi-domain structures and domain distributions. As seen from Figure 6(a), ferroelectric domains are mainly perpendicular to the film surface with the uniform polarization, although there are about 15% ferroelectric domains polarized along in-plane direction. The PRM results further confirm that the BTO film has a preferred c-axis orientation.

The ferroelectric property measurements were also performed at room temperature, as seen Figure 6(b). It is surprisingly found that the as-grown BTO films on Ni metal tapes with a NiO buffered layer exhibit very high resistivity value of $10^{10} \text{Ω}\cdot\text{cm}$. The ferroelectricity of the BTO films was evidenced from the hysteresis loop. The room temperature spontaneous polarization, remnant polarization, and coercive field from the as-deposited BTO layer
can be obtained to be about 2.0 µC/cm² and 1.0 µC/cm², respectively, with a coercive field of 25 kV/cm. It is known that the lattice dipole along the c-axis for a tetragonal perovskite structure is the origin of the ferroelectric properties associated with BTO. In other words, the ferroelectric dipole originates from ionic displacement in the c-axis direction, only c-axis oriented BTO thin films exhibit ferroelectricity. The a-axis oriented BTO film cannot show ferroelectric hysteresis due to the randomly oriented polarization, the large spontaneous polarization obtained in the as-deposited film is consistent with the result of the microstructure measurement that the film has highly c-axis oriented texture structure. The piezoelectric response of the as-deposited BTO film was surprisingly found to be 130 (x 10⁻¹² C/N) which is about 30% larger than the values (90 – 100 x 10⁻¹² C/N) of BTO single crystalline and polycrystalline bulk materials. The large piezoelectric response might result from the uniform nanodomain structures. The nature of the mechanisms is under investigation and will be reported later on.

![Figure 6 (a) multi-domain structures and domain distributions measured by PRM; (b) Hysteresis loop measure of ferroelectric BaTiO₃ thin films.](image)

6. CONCLUSION AND FURTHER WORK

In conclusion, piezoelectric thin-film active sensors characteristic simulation has been done to give the right work plan in this paper. The c-axial preferred oriented ferroelectric BaTiO₃ thin films have been successfully fabricated on Ni metal tapes with a thin NiO buffered layer by pulsed laser ablation. Microstructure studies reveal that the as-deposited BTO films have a nanopillar structure with a preferred c-axis orientation. TEM studies of BTO/Ni interface have demonstrated that the BTO/NiO/Ni layered films have sharp interfaces with no inter-diffusion or reaction. Ferroelectric polarization measurements have shown the hysteresis loop at room temperature in the film with a large remanent polarization, indicating that the ferroelectric domains have been created in the as-deposited BTO films.

These excellent properties in this piezoelectric thin-film indicate that the as-fabricated BTO films are promised for the development of the structural health monitoring systems. The next step is to design the electrode pattern on top of BTO film to monitor the electromechanical response of the piezoelectric film.

7. ACKNOWLEDGMENTS

This material is based upon work supported by the National Science Foundation under Grant # CMS-0408578 and CMS-0528873, Dr. Shih Chi Liu Program Director and by the Air Force Office of Scientific Research under Grant # FA9550-04-0085. Any opinions, findings, and conclusions or recommendations expressed in this material are those of the authors and do not necessarily reflect the views of the National Science Foundation or the Air Force Office of Scientific Research.
8. REFERENCES


