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Electric-field-induced structural changes in multilayer piezoelectric actuators during electrical and mechanical loading

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ABSTRACT

The effects of electrical and mechanical loading on the behavior of domains and phases in Multilayer Piezoelectric Actuators (MAs) is studied using *in situ* high-energy X-ray diffraction (XRD) and macroscopic property measurements. Rietveld refinement is carried out on measured diffraction patterns using a two-phase tetragonal (*P*4*mm*) and rhombohedral (*R*3*m*) model. Applying an electric field promotes the rhombohedral phase, while increasing compressive uniaxial pre-stress prior to electric field application favors the tetragonal phase. The competition between electrical and mechanical energy leads to a maximal difference between electric-field-induced phase fractions at 70 MPa pre-stress. Additionally, the available volume fraction of non-180° domain reorientation that can be accessed during electric field application increases with compressive pre-stress up to 70 MPa. The origin for enhanced strain and polarization with applied pre-stress is attributed to a combination of enhanced non-180° domain reorientation and electric-field-induced phase transitions. The suppression of both the electric-field-induced phase transitions and domain reorientation at high pre-stresses (>70 MPa) is attributed to a large mechanical energy barrier, and alludes to the competition of the electrical and mechanical energy within the MA during applied stimuli.

1. Introduction

Piezoelectric actuators serve as system-enabling components in many applications. For example, they are the core element of modern energy-efficient fuel injection systems in automotive engines, and are also used in power harvesting, micro actuation, and vibration suppression [1]. One of the leading types of piezo actuators in the market today are multilayer piezoelectric actuators (MA(s)) with interdigitated metallic electrodes. MAs allow the fabrication of large ceramic components that are operated at lower voltages compared to bulk monolithic structures, *i.e.*, applied voltage is in the volt range versus kilovolt to achieve the desirable strain. This is accomplished by increasing the number and decreasing the thickness of individual piezoceramic layers. The majority of piezoelectric actuators are composed of lead zirconate titanate (PZT), $Pb(Zr_xTi_{1-x})O_3$, a binary compound that exhibits enhanced dielectric and piezoelectric properties near the morphotropic phase boundary (MPB) [2].

When discussing the performance of MAs utilizing MPB PZT, domain wall motion is often discussed as a dominant mechanism, whereas phase fraction changes are less often discussed. Applying high electric fields and/or mechanical loads to PZT enables domain wall motion, which modifies the electromechanical properties of the material [3]. The existence of domain walls is dependent on the crystallographic structure of the material, and they have been widely studied using electrical property measurements, X-ray diffraction (XRD) [4,5], piezoresponse force microscopy (PFM) and transmission electron microscopy (TEM) [6]. The knowledge of domain behavior under external loads (electrical and/or mechanical) is essential for understanding macroscopic material behavior.

The scientific community has made considerable efforts to study the effects of electrical and mechanical loading on bulk PZT

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ceramics. Bipolar strain and polarization hysteresis of soft lanthanum-doped PZT (PLZT) under compressive mechanical loads, i.e. up to 60 MPa, have been reported by Lynch [7]. From this study, material depolarization, decreases in coercive field, and changes in the piezoelectric coefficient were observed in response to increasing applied compressive stress. The observed behavior in Lynch's study was attributed to ferroelastic domain reorientation. Chaplya and Carman provided a more detailed explanation of the observed response by conducting both bipolar and unipolar strain and polarization measurements with applied compressive loads on commercial PZT-5H samples [8]. Unipolar measurements showed that, at intermediate compression loads (50–60 MPa), maxima in the strain and polarization are observed. The enhanced response was explained in terms of the available volume fraction of non-180° domains and the difference in domain wall pressure created by electrical and mechanical loading. These results suggested that the peak in enhanced strain and polarization response can shift to a higher stress value upon increasing the amplitude of the electric field, thus demonstrating a dependence on the balance between the electrical and mechanical energy. Ultimately, all previous studies share a common observation in the materials response: the enhancement in electric-field-induced strain under moderate compressive pre-stresses is attributed to non-180° domain wall motion. To separate the earlier described effects of mechanical constraints during application of electric field (*i.e.* ferroelectric domain reorientation and phase composition), *in situ* XRD coupled with strain and polarization measurements are needed. XRD adequately probes the various electromechanical responses of ferroelectrics and yields insight into the material's structure during applied stimuli [4].

This work reports phase rearrangement and domain texture of commercial MAs, quantitatively measured using *in situ* high energy XRD coupled with macroscopic electromechanical response measurements. Unipolar polarization and strain measurements were synchronized to the measured XRD patterns to establish structure-property relations that describe the enhanced strain response seen in MAs under the application of compressive pre-stresses. The results show that, while domain switching does contribute to the properties, a portion of the enhanced response also originates from electric-field-induced phase transitions. Hence, this work establishes new structure-property relationships in PZT-based MAs that can be used to design actuators with enhanced electromechanical properties by tailoring electric-field-induced phase transitions and ferroelectric domain configurations.

2. Experimental

2.1. Multi-layer piezoelectric actuators

Commercially available Multilayer Piezoelectric Actuators (MAs) were supplied by the company TDK (Deutschlandsberg, Austria). The MAs belonged to the latest generation of so-called “High-Active Stacks”, whereby the passive zone is minimized down to approximately 110 μm thickness. The MAs had an original size of $3.4 \times 3.4 \times 27$ mm^3 and were constituted by a stack of ~70 μm-thick piezoceramic layers with interdigitated copper electrodes. The piezoceramic is lead zirconate titanate, PZT, with near-MPB composition with a multi-phase structure (tetragonal and rhombohedral) that appears predominantly tetragonal from X-ray diffraction. MA samples were ground down to a size of $1.8 \times 3.4 \times 27$ mm^3 by the supplier. Copper wires were soldered on each termination side and a silicon-based passivation layer was applied over the whole MA to avoid electric arcing on the ground surface during testing. The samples were supplied in the unpoled state.

2.2. Electromechanical measurements

As-received MA samples were mounted into a uniaxial screw-driven testing machine (Microstrain ME30-1, Messphysik, Fürstenfeld, Austria), which was placed inside the experimental hutch of the 11-ID-C beamline at the Advanced Photon Source (Argonne National Laboratory, Argonne, IL) (cf. Fig. 1). Samples were placed between two steel loading punches, but ceramic (Si_3N_4) discs with a thickness of 5 mm each were placed between each side of the MA and the punches to provide electric isolation and stiff contact points. Samples were aligned in a way that the X-ray beam could pass at the center of the sample. The testing machine was closed-loop controlled (EDC120 controller, DOLI, Munich, Germany) with a sampling rate of 1 kHz. The force applied to the sample was measured using an S-shaped 5 kN load cell (KAP-S, AST, Dresden, Germany). Strain (displacement of the MA during loading) was measured by three independent strain gauges (DD1, HBM, Darmstadt, Germany), each with a maximum amplitude of ± 2.5 mm, which were fixed on the lower punch at angles of 120° apart and measured the relative displacement of the upper punch with respect to the lower one. The data measured by each strain gauge was collected separately and averaged afterwards. The total voltage was applied with a Keithley 2450 Source Meter Unit. Charge flow at the MA was measured using a Sawyer-Tower circuit by connecting the sample in series to a reference capacitor (foil-capacitor) with a total capacitance of 1430 μF. Voltage was measured at the reference capacitor using a Keithley 6514 Electrometer.

Each measurement (i.e. for each value of the applied mechanical load) was performed on different MA samples, which were always loaded in their as-received, unpoled state. Before applying any mechanical load, a virgin reference XRD-pattern was acquired on each sample. Subsequently, mechanical load was applied and then three voltage ramps (between 0 V and approx. 210 V, thus representing 3 kV/mm at single layer level) were performed while keeping the mechanical load constant. The voltage was incremented by 10 V steps with a rate of 10 V/s. Each voltage value was held for the time necessary to acquire one XRD pattern (approx. 5 s).

2.3. In situ diffraction experiment

Diffraction patterns were measured *in situ* during simultaneous application of electric fields and mechanical load using high-energy X-rays at beamline 11-ID-C of the Advanced Photon Source at Argonne National Laboratory. Each individual sample was mounted in the screw-driven testing fixture described above. Electrode connections were attached to the high voltage source via alligator clips. Diffraction data were measured in transmission mode using a

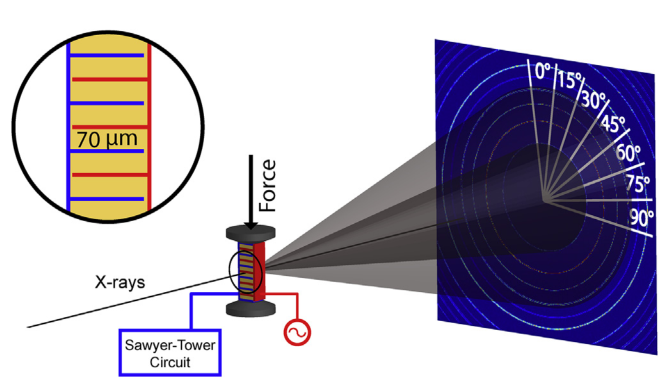

Fig. 1. Schematic of experimental setup at the beamline 11-ID-C of the Advanced Photon Source.