Effect of Crystallographic Texture on the Field-Induced-Phase Transformation Behavior of Bi$_{0.5}$Na$_{0.5}$TiO$_3$ – 7BaTiO$_3$ – 2K$_{0.5}$Na$_{0.5}$NbO$_3$

Chris M. Fanchera$^{a,*}$, Thanakorn Iamsasrib, John E. Blendella$^a$ and Keith J. Bowmana,c

$^a$School of Materials Engineering, Purdue University, West Lafayette, IN 47907, USA; $^b$Department of Materials Science and Engineering, University of Florida, Gainesville, FL 32611, USA; $^c$Mechanical, Materials & Aerospace Engineering, Illinois Institute of Technology, Chicago, IL 60616, USA

(Received 19 February 2013; final form 24 April 2013)

Supplementary Material Available Online

The electric-field-induced phase transformation of {001}pc oriented Bi$_{0.5}$Na$_{0.5}$TiO$_3$ – 7BaTiO$_3$ – 2K$_{0.5}$Na$_{0.5}$NbO$_3$ bulk ceramics with a 8.3 multiples of a random distribution fiber texture was investigated using in situ diffraction. Field-dependent diffraction suggests that the high strain is a result of an electric-field-induced pseudo-cubic to tetragonal phase transformation, where the induced tetragonal phase has a strong domain texture. The effect of electric field direction was investigated by applying the electric field parallel and perpendicular to the fiber texture axis. Applied field direction affected the lattice spacing of the induced phase, domain texture, and increased poling field to induce the phase transformation.

Keywords: Preferred Orientation, Electric-Field-Induced Phase Transformation, Bismuth Sodium Titanate

Piezoelectric materials are used in a variety of applications such as actuators, sensors, and transducers.[1] Because of the high strain response, systems that exhibit an electric-field-induced pseudo-cubic to polar transformation have received strong interest as a potential alternative to lead-based ceramics.[1–4] One promising system is the solid solution Bi$_{0.5}$Na$_{0.5}$TiO$_3$–xBaTiO$_3$–yK$_{0.5}$Na$_{0.5}$NbO$_3$ (BNT–xBT–yKNN).[3] At room temperature, BNT–6BT–2KNN exhibits a $S_{\text{max}}/E_{\text{max}}$ of 567 pm/V at 8 MV/m with a low d$_{33}$ of 30 pm/V.[3] The origin of the high strain has been attributed to reversible field-induced transformation, where the addition of KNN into the BNT–xBT decreased the depolarization temperature below room temperature.[2]

In recent years, in situ diffraction experiments have been used to investigate field-induced phase transformations [5–10] and domain texture evolution.[5,6,11] Recent in situ studies have suggested BNT–7BT undergoes an oriented electric-field-induced transformation from pseudo-cubic to tetragonal.[5,6] It has also been reported that BNT–6BT–2KNN undergoes a reversible field-induced transformation from pseudo-cubic to a tetragonal and rhombohedral phase mixture.[9] While in situ studies have investigated the phase evolution of single crystals [12] and bulk ceramics,[5–8] the effect of crystallographic texture on the electric-field-induced phase transformation behavior of bulk BNT-based ceramics has only recently been investigated.[13] Here, we investigate the effect crystallographic texture and electric field direction has on the electric-field-induced phase transformation behavior of BNT–7BT–2KNN using in situ X-ray diffraction.

In situ scattering experiments were performed at beamline 5-BM-D at the Advanced Photon Source at Argonne National Laboratory using 65 keV (0.1907 Å) X-rays. Tungsten beam slits were used to limit the incident beam to a 500 × 500 μm spot size. A Mar 345-image plate positioned approximately 1.6 m from the sample was used to collect diffraction images. Diffraction data were collected while a static electric loading was applied perpendicular to the incident X-rays to emulate the electric poling process. Detailed reports of the experimental setup have been previously published.[14,15]

Samples with a {001}pc fiber texture were prepared using the tape cast method as reported elsewhere.[16] Two sets of bulk ceramics were prepared for in situ

$^*$Corresponding author. Email: cfancher@purdue.edu

© 2013 Chris M. Fancher
Figure 1. Schematic of the (a) processing coordinate system and a 2D projection of the processing coordinate system from the prospective of the incident X-rays for a field applied along (b) ND and (c) TD, where the angle from applied field, (b) and (c), are defined in terms of Bunge Euler rotation angles ($\phi_1$, $\Phi$, $\phi_2$).

diffraction; 20 layer stacks, and 80 layer stacks. Samples used for in situ diffraction were rectangular bars with of $xyz = 5 \times 1 \times 1$ and $4 \times 1 \times 1$ mm for and electric field applied along the sample normal direction (ND) and transverse direction (TD), respectively. Sintered ceramics developed a 8.3 multiples of a random distribution (MRD) {001}$_{pc}$ fiber texture. The crystallographic texture was calculated by refining zero field diffraction using the software package materials analysis using diffraction with a harmonic orientation distribution function model.\[17\] See supplementary information for details of the texture refinements. To investigate the effect of the applied field direction, relative to the fiber texture axis, the electric field was applied parallel and perpendicular to the fiber axis (processing ND). Applying the electric field perpendicular to the fiber axis probes a different distribution of crystallites because all {00l}$_{pc}$ are equally probable at all orientations in the plane perpendicular to the fiber axis.

A coordinate system can be defined relative to the initial processing axis with; ND, tape-cast direction, and TD, as shown in Figure 1(a), with the direction of the applied field is defined relative to this coordinate system, Figure 1(b) and 1(c). By projecting the sample coordinate system along the axis of the incident X-ray, the angle between the applied field and the diffraction vector ($\hat{k}$) is defined in terms of Euler angles ($\phi_1$, $\Phi$, $\phi_2$), where the rotation between the scatter vector and electric field is defined as ($\phi_1$, 0, 0) and (0, $\Phi$, 0) for an applied poling field along TD and ND, respectively.

Prior to the application of the electric field, the diffraction signal, Figures 2(a) and 3(a), showed no peak splitting, shoulders, or superlattice reflections associated with a distorted or mixed phase structure; henceforth, this zero-field structure will be referred to as pseudo-cubic. Diffraction signal as a function of angle from the applied field, Figures 2(c) and 3(c), highlights the difference in the {00l}$_{pc}$ distribution relative to the applied field, where the {200}$_{pc}$ diffraction signal has either two maxima ($\Phi = 0$ and 90) or remains constant for all angles, for a field applied along ND and TD, respectively. The large grain size, $\sim 25$ $\mu$m, gives rise to intensity variation in the {111}$_{pc}$ and {200}$_{pc}$ diffraction signal, Figure 3(c) and 3(d), due to poor sampling.

Increasing the applied poling field above 1 MV/m induces peak splitting in the (002) diffraction signal, while the {111}$_{pc}$ diffraction signal shows no splitting, as shown in Figures 2(a) and 2(b) and 3(a) and 3(b). A lack of {111}$_{pc}$ splitting suggests the observed peak splitting is consistent with a pseudo-cubic to tetragonal transformation, but the diffraction data cannot definitively determine the field-induced transformation to be a pseudo-cubic to a single tetragonal phase. Field-dependent diffraction data show the pseudo-cubic to tetragonal transformation proceeds across a wide electric field range (1–3 MV/m), and the resulting tetragonal phase shows a strong domain texture where minimal (002) diffraction signal was measured perpendicular to the applied poling field, as shown in Figures 2(d) and 3(d). Close inspection of the (002) diffraction signal at 4 MV/m shows the pseudo-cubic to tetragonal transformation results in a stronger domain texture when the electric field is applied along ND, as
Without comparable in situ studies for randomly oriented BNT–7BT–2KNN, it is not possible to determine if the \{001\}_\text{pc} texture promotes the field-induced transformation to the tetragonal phase. In situ diffraction of BNT–5.6BT single crystals has shown that the field-induced-phase transformation is only dependent on the field direction if the field material is above the relaxor transition temperature.\cite{19} The modification of BNT–BT with KNN has been reported to decrease the relaxor transition temperature below room temperature.\cite{4} Suggesting a \{001\}_\text{pc} crystallographic texture can affect the field-induced phase transformation, potentially by promoting polarization extension/rotation along the field direction.\cite{20} Sub-coercive field in situ neutron scattering experiments have shown textured BNT–7BT maintains a pseudo-cubic structure while randomly oriented ceramics develops peak splitting in both the 200 and 111 peaks.\cite{13} The authors suggested the suppression of the transformation could result from a difference in the coherence length of the polar nano regions or a microstructural effect. Without in situ data collected for a stronger applied electric loading, it is unclear if the final field-induced phase or phase transformation electric field is affected by the \{001\}_\text{pc} texture.

The \{111\}_\text{pc} diffraction signal shows distinctly different field-dependent behaviors parallel, Figure 2(a) and (3(a), and perpendicular, Figures 2(b) and 3(b), to the applied field. Perpendicular to the applied field, both peak shape and position show minimal variation with increasing field, while the diffraction signal parallel to the applied field shows two distinct inflection points with increasing field at (1 and 2 MV/m) and (1 and 2.25 MV/m) for an applied field along ND and TD, respectively. These inflection points correspond well to the electric field range where BNT–7BT–2KNN transforms from pseudo-cubic to tetragonal. The higher electric field inflection point occurs at a lower electric field than inflection points measured in polarization and strain measurements, suggesting the observed strain increase is the result of further domain alignment with increasing field, a polarization and strain loop is reported in the supplementary information.

The lattice parameters of the induced tetragonal phase were measured by fitting the diffraction data using three peaks,\cite{5} two split pseudo-Voigt functions to model the (002) and (200) diffraction peaks and a pseudo-Voigt function to fit the pseudo-cubic fraction. Prior to applying the electric field the lattice parameter of the pseudo-cubic structure was 3.89702 (5)Å for both applied field directions. The lattice parameters of the induced tetragonal phase are graphed in Figure 4 for an applied field range of 2–4 MV/m. The larger error in the measured a and c lattice parameter is the result of peak overlap between the induced phase and the initial pseudo-cubic phase. For both applied field directions, the lattice spacing of the induced tetragonal phase, Figure 4(a) and 4(b) shows a trend of c-axis expansion and a-axis contraction with
Figure 4. Electric field-dependent tetragonal lattice parameters (a) c-axis and (b) a-axis measured parallel (open) and perpendicular (solid) to the applied field, and (c) angular-dependent tetragonal lattice parameters for a 3 MV/m applied field along TD. Error bars represent the 99% confidence band from the respective peak fit.

In summary, in situ diffraction of {00l}pc oriented BNT–7BT–2KNN bulk ceramics show an oriented field-induced pseudo-cubic to tetragonal transformation for both applied field directions. The direction of the applied electric field, with respect to the processing coordinate system, affects the field-induced phase transformation behavior. Applying a poling field along TD imposes grain constraints that results from applying the electric field inside the tape-cast plane where all {001}pc are equally probable, resulting in a shorter c-axis parallel to the applied field, lower domain texture, and increased poling field to induce the phase transformation.

Supplementary online material. A more detailed information on experiments is available at http://dx.doi.org/10.1080/21663831.2013.800606.

Acknowledgements The authors in this paper acknowledge the help of Jacob L. Jones. This work was supported by the US National Science Foundation Grant No. DMR 0805022. The use of the Advanced Photon Source was supported by the US Department of Energy, Office of Science, Office of Basic Energy Sciences, under Contract No. DE-AC02-06CH11357. TI acknowledges support from the Development and Promotion of Science and Technology Talents Project, Royal Thai Government.

References

[1] Jo W, Dittmer R, Acosta M, Zang J, Groh C, Sapper E, Wang K, Rödel J. Giant electric-field-induced strains in lead-free ceramics for actuator applications—status and perspective. J Electroceram. 2012;29:71–93.

[2] Jo W, Granzow T, Aulbach E, Rödel J, Damjanovic D. Origin of the large strain response in K0.5Na0.5NbO3-modified Bi0.5Na0.5TiO3–BaTiO3 lead-free piezoceramics. J Appl Phys. 2009;105:094102-5.

[3] Zhang ST, Kounga AB, Aulbach E, Granzow T, Jo W, Kleebe HJ, Rödel J. Lead-free piezoceramics with giant strain in the system Bi0.5Na0.5TiO3–BaTiO3–K0.5Na0.5NbO3. I. Structure and room temperature properties. J Appl Phys. 2008;103:0314108-7.

[4] Zhang ST, Kounga AB, Aulbach E, Ehrenberg H, Rödel J. Giant strain in lead-free piezoceramics with giant strain in the system Bi0.5Na0.5TiO3–BaTiO3–K0.5Na0.5NbO3. II. Structure and room temperature properties. J Appl Phys. 2008;103:0314108-7.

[5] Daniels JE, Jo W, Rödel J, Jones JL. Electric-field-induced phase transformation at a lead-free morphotropic phase boundary: case study in a 93% (Bi0.5Na0.5)
TiO$_3$7%BaTiO$_3$ piezoelectric ceramic. Appl Phys Lett. 2009;95:032904-3.

[6] Daniels JE, Jo W, Rödel J, Honkimaki V, Jones JL. Electric-field-induced phase-change behavior in Bi$_{0.5}$Na$_{0.5}$TiO$_3$–BaTiO$_3$–K$_{0.5}$Na$_{0.5}$NbO$_3$: a combinatorial investigation. Acta Mater. 2010;58:2103–2111.

[7] Simons H, Daniels JE, Jo W, Dittmer R, Studer A, Avdeev M, Rödel J, Hoffman M. Electric-field-induced strain mechanisms in lead-free 94%Bi$_{1/2}$Na$_{1/2}$TiO$_3$–6%BaTiO$_3$. Appl Phys Lett. 2011;98(8):082901-3.

[8] Kling J, Tan XL, Jo W, Kleebe HJ, Fauss H, Rödel J. In situ transmission electron microscopy of electric field-triggered reversible domain formation in bi-based lead-free piezoceramics. J Am Ceram Soc. 2010;93:2452–2455.

[9] Hinterstein M, Knapp M, Holzel M, Jo W, Cervellino A, Ehrenberg H, Fauss H. Field-induced phase transition in Bi$_{0.5}$Na$_{0.5}$TiO$_3$-based lead-free piezoelectric ceramics. J Appl Crystallogr. 2010;43:1314–1321.

[10] Jones JL, Hoffman M, Vogel SC. Ferroelastic domain switching in lead zirconate titanate measured by in situ neutron diffraction. Mech Mater. 2007;39:283–290.

[11] Daniels JE, Jo W, Rödel J, Rytz D, Donner W. Structural origins of relaxor behavior in a 0.96Bi$_{0.5}$Na$_{0.5}$TiO$_3$–0.04BaTiO$_3$ single crystal under electric field. Appl Phys Lett. 2011;98:252904-3.

[12] Daniels JE, Jo W, Donner W. High-energy synchrotron X-ray diffraction for in situ diffuse scattering studies of bulk single crystals. JOM-US. 2012;64:174–180.

[13] Maurya D, Pramanick A, An K, Priya S. Enhanced piezoelectricity and nature of electric-field induced structural phase transformation in textured lead-free piezoelectric Na$_{0.5}$Bi$_{0.5}$TiO$_3$–BaTiO$_3$ ceramics. Appl Phys Lett. 2012;100(17):172906-5.

[14] Jones JL, Pramanick A, Daniels JE. High-throughput evaluation of domain switching in piezoelectric ceramics and application to PbZr$_{(0.6)}$Ti$_{(0.4)}$O$_3$ doped with La and Fe. Appl Phys Lett. 2008;93:152904-3.

[15] Daniels JE. Determination of directionally dependent structural and microstructural information using high-energy X-ray diffraction. J Appl Crystallogr. 2008;41:1109–1114.

[16] Fancher CM, Blendell JE, Bowman KJ. Poling effect on $d_{33}$ in textured Bi$_{0.5}$Na$_{0.5}$TiO$_3$-based materials. Scripta Mater. 2013;68:443–446.

[17] Lutterotti L. Total pattern fitting for the combined size-strain-stress-texture determination in thin film diffraction. Nucl Instrum Methods B. 2010;268:334–340.

[18] Jo W, Daniels JE, Jones JL, Tan X, Thomas PA, Damjanovic D, Rödel J. Evolving morphotropic phase boundary in lead-free Bi$_{1/2}$Na$_{1/2}$TiO$_3$–BaTiO$_3$ piezoceramics. J Appl Phys. 2011;109:014110-7.

[19] Lou C, Ge W, Zhang Q, Li J, Luo H, Viehland D. Crystallographic direction dependence of direct current field induced strain and phase transitions in Na$_{0.5}$Bi$_{0.5}$TiO$_3$–x%BaTiO$_3$ single crystals near the morphotropic phase boundary. Appl Phys Lett. 2012;101:141912-4.

[20] Damjanovic D. A morphotropic phase boundary system based on polarization rotation and polarization extension. Appl Phys Lett. 2010;97:062906-3.