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Domain switching characteristics of lead zirconate titanate piezoelectric ceramics during mechanical compressive loading

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Abstract

To better understand the domain switching characteristics of lead zirconate titanate (PZT) ceramics, the orientations of domains have been directly investigated during loading and unloading using various experimental techniques. Upon loading, linear and non-linear fracture mechanics of the PZT ceramics are observed. The slope of the stress–strain response is attributed mainly to lattice strain and domain switching strain. During the loading process, electrical activity also occurs several times in the PZT ceramics. This activity is related to a lightning-like phenomenon and consists of a bright flash with a click. This electrogenerative event is caused by severe domain switching. The characteristics of domain switching and reverse switching are detected during the loading and unloading processes. The amount of domain switching depends on the grain, due to different stress levels. In addition, two patterns of 90° domain switching systems are characterized, namely (i) 90° turn about the tetragonal *c*-axis and (ii) 90° rotation of the tetragonal *a*-axis.

Keywords: PZT; Piezoelectric properties; Mechanical properties; Failure analysis; X-ray methods

1. Introduction

Piezoceramics have been increasingly utilized in numerous applications, e.g., sensors and actuators. Specifically, lead zirconate titanate piezoelectric ceramics (PZT) are widely employed, since the PZT ceramic has excellent electromechanical responses,[1] a high Curie temperature, high material strength and low sintering temperature.[2] Because the sensor and actuator must be reliable and durable, the quality of the piezoceramic in these engineering applications is important.[3] In addition, the material response, in particular the matching of the piezoelectric coefficients to the application, is extremely important, as PZT ceramics in smart structures are required to have high piezoelectrical performance. The material properties of PZT ceramics are influenced by the material characterization, such as lattice structure, domain orientation and domain switching. In recent articles, it has been shown that the occurrence of domain switching and/or domain wall formation reduces the material properties of PZT ceramics.[4] To provide direct evidences of this, the domain switching characteristics have been investigated using various methods, including electron-back-scatter diffraction,[4] Moiré interferometry,[5] neutron and X-ray diffraction,[6] scanning force microscopy,[7] piezo-force microscopy,[8] polarized light microcopy[9] and finite element analysis.[10] There are a number of related papers. The Scopus searching system reveals that more than 420 "domain switching" and "PZT" academic papers have been published to date. Using neutron and X-ray diffraction, Jones et al. have conducted quantitative assessments of reflection geometry and spherical harmonic texture analysis, measuring change parallel to and tilted from the poling axis.[11] Using this approach, the relative intensity ratio of the (2 0 0) and (0 0 2) reflections in PZT ceramics was expressed by a domain switching fraction or a multiple of a random distribution (MRD).[12] The XRD method was employed in the study by Hall et al., where a high-energy synchrotron XRD was used to examine domain switching behavior in bulk PZT ceramics.[13] Tanaka et al. have also measured X-ray diffraction to evaluate the amount of domain switching for tetragonal and rhombohedral lead zirconate titanate ceramics, where the amount of domain switching was assessed by the change of the intensity ratio,[14,15] e.g.,

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$I_{222}/I_{22\bar{2}}$, of the (2 2 2) to (2 2 $\bar{2}$) diffraction for rhombohedral PZT ceramics. The intensity ratio for normal diffraction, i.e., $\psi = 0°$, decreased with increasing the applied strain, since the spontaneous polarization direction, the (2 2 2) direction, turned towards the loading direction. To measure in-plane domain switching and lattice structure in the region inside PZT ceramics, high-energy synchrotron XRD with energy 80.8 keV and wavelength $\lambda \sim 0.154$ Å was used.[16] Several researchers have recently observed in-plane 90° domain switching of ferroelectric ceramics using the Moiré interferometry technique[5] and electron back scatter diffraction (EBSD).[4,17] From the above information, the domain switching characteristics of PZT ceramics after applied loading have been clarified. Further study is required to describe the details of domain switching characteristics under static and cyclic loading processes.

The loading applied to PZT ceramics produces permanent strain by the irreversible switching of 90° domains, which gives rise to high anisotropic deformation. It has been clarified in a previous study that the switching occurs even under small applied load. Thus, stress vs. strain curves could provide important information about the complex constitutive non-linear behavior in piezoelectric ceramics. In the study by Calderon-Moreno and Popa, the domain switching strain was interpreted using stress–strain curves.[18] Li and Rajapakse have successfully incorporated a switching assumption into the constrained domain switching model to account for non-symmetry of their stress–strain curves.[19,20] The stress–strain information for PZT ceramics is always required in order to design the PZT ceramic for a particular engineering application. The aim of this work was therefore to investigate the effect of domain switching on the material strain in PZT ceramics during static and cyclic loading processes. In this study, the measurement of domain orientation (or domain switching) was carried out using several novel experimental techniques.

2. Experimental procedures

In the present work, a commercial soft tetragonal PZT ceramic, of nominal composition $PbZrTiO_3$, was used.[21] The PZT ceramics were well sintered at 1200 °C with an average grain size of 5 μm, a density of 7.65 g/cm^3 and Curie point of 295 °C. A silver-based electroplated layer was applied to two sides of the specimens by a firing process in atmosphere, before the electrical poling process, using an electric field of 2 kV/mm in silicon oil, was carried out. The PZT ceramics adopt a perovskite structure with aspect ratio $c/a = 1.014$ ($a = b = 0.4046$ nm and $c = 0.4103$ nm). The material properties of the PZT ceramics after polarization measured by an impedance analyzer (Agilent Technologies, 4294A) were (i) effective elastic constant (c^E_{33}) 53 GPa, (ii) electromechanical coupling coefficient (k_{33}) 0.71, (iii) piezoelectric constant (d_{33}) 472 pm/v and (iv) permittivity (ε_{33}) 22.7 nF/m (or dielectric constant ($\varepsilon_{33}/\varepsilon_0$) 2502). Fig. 1 shows a schematic illustration of a specimen material. This specimen was made by the following process: the round surface of the cylindrical sample with length 12.5 mm and diameter 5 mm was polished to make four flat faces. The polishing process was conducted about 0.33 mm in depth using alumina paste down to 1 μm size, where the 12.5 × 3 mm^2 face is spread. The reason for the polishing process is to measure easily the material properties of the ceramics. Domain orientations were investigated in the flat areas (Fig. 2) under applied loading and unloading. Two systems of X-ray diffraction and EBSD analysis were employed in this investigation. In the X-ray analysis, an X' Pert Pro system (Panalytical Inc.) with a Cu tube source was utilized. An X-ray (8 keV, wavelength $\lambda \sim 1.54$ Å) was used to measure domain switching and lattice structure. The EBSD analysis was conducted by high resolution electron microscopy, using a JEM-200EX (JEOL Ltd.) with an orientation imaging microscopy (OIM) system. In the examination of EBSD, the sample surface for the observation was coated with carbon, after which the surfaces for observation were etched using a solution of 5 ml hydrochloric acid, 2 ml hydrogen fluoride and 250 ml water. Details of the EBSD technique are described in Ref. 22.

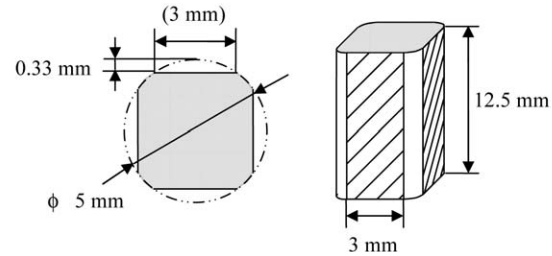


Fig. 1. Schematic illustration of the test specimen.

The static compressive loading and cyclic loading were performed in air with stroke control at 1 mm/min for the examination of the effect of domain switching on the stress–strain characteristics. The applied loading was performed on the PZT ceramics using a screw driven type EZ-Graph universal testing machine with 10 kN capacity (Shimazu Co.). The resolutions of the load and displacement in this testing machine are 0.01 N and 1 μm, respectively. Fig. 2 shows the testing

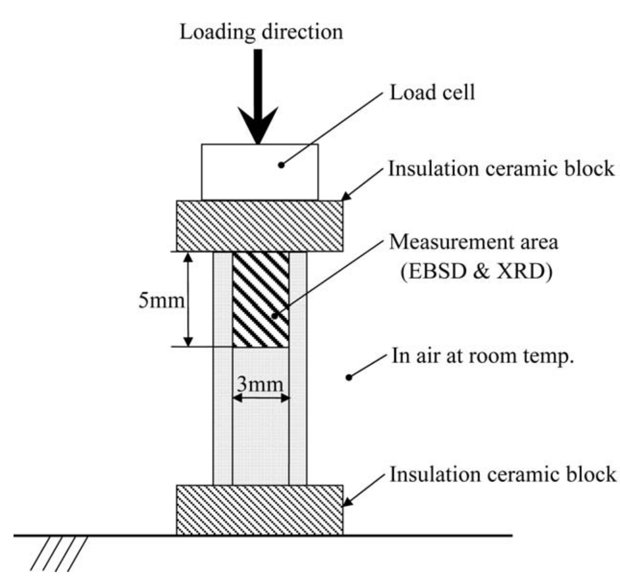


Fig. 2. Testing setup for compression and measurement area for the EBSD and XRD (open circuit).

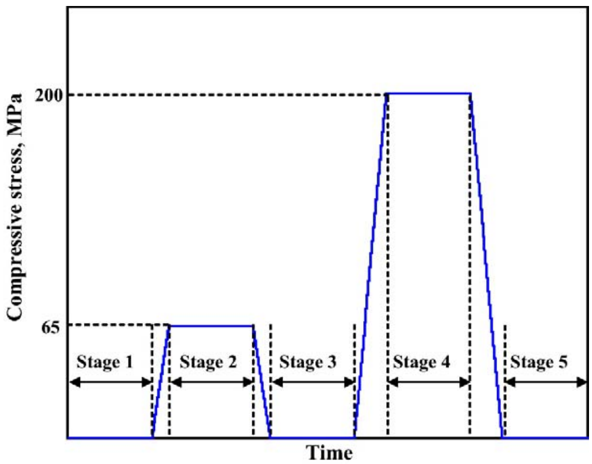


Fig. 3. Schematic illustration of the loading cycle.

setup for compression loading in open circuit.[23,24] The specimen was held between two high strength insulation ceramic blocks ($\phi25.0 \times 15.0\,mm^2$), so as to minimize diffraction interference from the specimen during the loading process. The stress and strain values were measured using a commercial load cell and strain gauges, respectively. Four strain gauges were glued on the four flat faces of the specimen. The domain orientation was examined continuously during the following five different loading stages using EBSD and XRD analysis: (1) 0 MPa; (2) 65 MPa; (3) 0 MPa; (4) 200 MPa; (5) 0 MPa, see Fig. 3. It should first be noted that the maximum applied stress of 200 MPa (Stage 4) is more than 85% of the compressive strength of the PZT ceramic. In addition, the applied stress of 65 MPa (Stage 2) was determined on the basis of the stress level at which the domain switching in the PZT ceramic begins. In the previous works, it was reported that domain switching occurs even under a small applied load of approximately 50 MPa.[25]

3. Results and discussion

3.1. Stress–strain characteristics

Fig. 4(a) and (b) shows the stress–strain curves for the PZT ceramics with and without poling, respectively. For the poled sample (Fig. 4(a)), the beginning of the loading process, i.e., the lower portion of the loading phase, exhibits a small amount of linear deformation occurring up to a relatively low stress of about 45 MPa, above which extensive non-linear deformation occurs. This is comparable with the results of previous work.[18] Similar non-linear and linear stress–strain relationships are also observed in the second loading portion (Stages 4 and 5). In this case, the linear relationship will be affected by the lattice strain of the PZT ceramics, and the non-linear relationship is caused by the permanent strain arising from the domain switching. Similar results were observed in the study by Jones et al. They examined the strain characteristic of a soft lead zirconate titanate ceramic, in which the macroscopic strain in the 0.30% ceramic consists of 0.22% domain switching strain and 0.08% lattice strain.[26] Several researchers have described

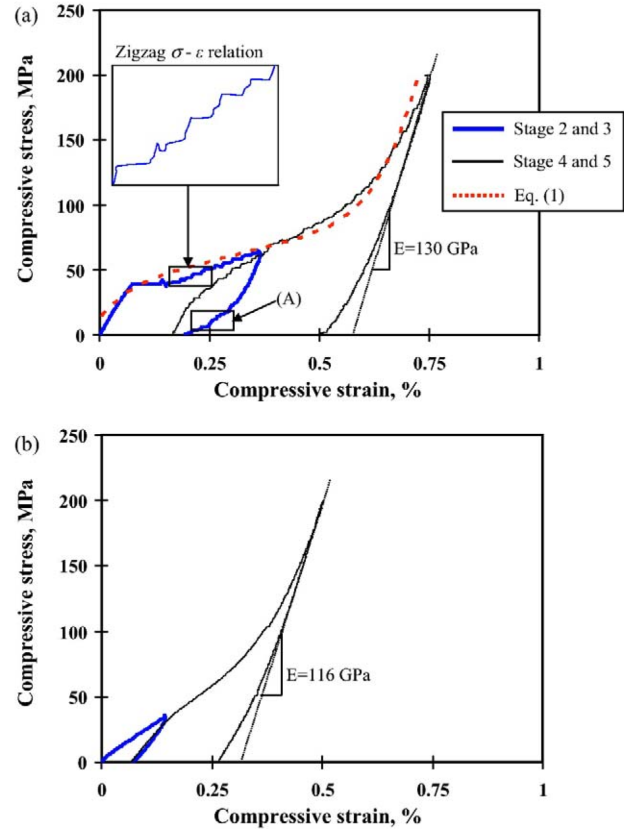


Fig. 4. Compressive stress vs. strain relationships for the PZT ceramics (a) with and (b) without the poling process.

stress–strain curves similar to the results shown in Fig. 4, where the large strain in the PZT ceramic is strongly influenced by the domain switching.[18,27] In recent years, excellent work has been carried out by Li et al. They estimated the electric and compression strains in the three types of perovskite ferroelectric ceramic, e.g., with tetragonal, rhombohedral and orthorhombic structures.[28] Their estimated strain levels were also in good agreement with previously reported analytical and numerical results.[29] In their approach, the maximum compression strain for un- and poled tetragonal PZT ceramics were calculated to be $0.269 \times (c/a - 1)$ and $0.637 \times (c/a - 1)$, respectively. Because $c/a = 1.014$ for our tetragonal PZT ceramic, the permanent strain for the un- and poled ceramics after maximum compression is calculated to be less than 0.38% and 0.89%, respectively. Both strain levels may be relatively close to our strain data shown in Fig. 4.

The initial slope of the unloading stress–strain relationships can be associated with the Young's modulus of the material free of domain switching,[30] which are 130 GPa for the poled PZT ceramic and 116 GPa for the unpoled one. The Young's modulus for un- and poled samples are in good agreement with the related ceramics reported in Ref. 30.

It is interesting to mention that Fig. 4(a) clearly shows a zigzag stress–strain relationship in the non-linear portions of the loading and unloading phases. In this case, the zigzag stress–strain curve, created in Fig. 4, can be interpreted using the domain switching models. Fig. 5 shows a schematic diagram of

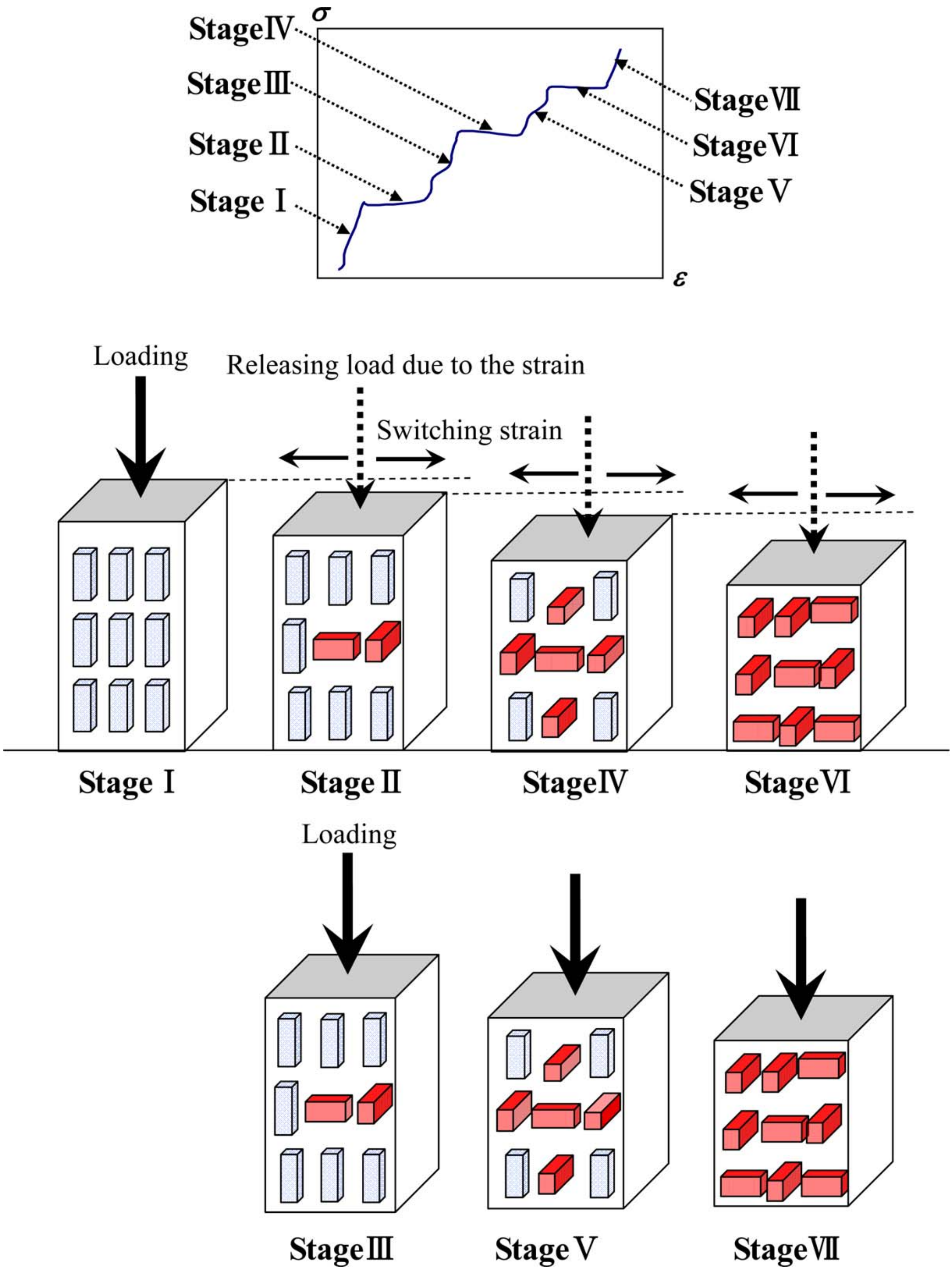


Fig. 5. Domain switching model in the zigzag compressive stress vs. strain relations.

the switching models incorporating a zigzag $\sigma - \varepsilon$ relationship. In Stage I, the stress level increases during the loading process, which is also related to Stages III, V and VII. On the contrary, the plateau $\sigma - \varepsilon$ relationships in Stages II, IV and VI might be caused by the strain acceleration due to the domain switching during the loading, i.e., the long *c*-axis of the tetragonal lattice turning to the direction perpendicular to the loading direction.[4] It is interesting to mention that the zigzag stress–strain in Fig. 4(a) can be seen even in the unloading portion, as indicated by (A). This may suggest that some domain switching or reverse switching occurred in the unloading process.[33] This will be discussed in a later section of this paper.

When PZT ceramics take on a permanent strain because of the irreversible switching of 90° domains, work hardening may occur.[4] This permanent strain can be expressed as a power law. From Fig. 4(a), the deviation from the stress–strain curves has a form characteristic of that found for phase transformations brought about by domain switching, consisting of an elastic strain region where the hardening rate diminishes as the stress increases, followed by a regime of accelerated hardening, as a saturation permanent strain is approached. Thus, the stress σ' and permanent strain ε'_P can be described by a bifunctional power low formula.[31]

$$\frac{\sigma'}{\sigma'_0} = \left(\frac{\varepsilon'_P}{\varepsilon'_0}\right)^a + \left(\frac{\varepsilon'_P}{\varepsilon'_0}\right)^b \qquad (1)$$

where a and b are empirical power law coefficients, σ'_0 is a reference stress and ε'_0 is a reference strain. The magnitudes of the coefficients a and b and the reference stress σ'_0 and strain ε'_0 values for the poled sample are $a = 0.4$, $b = 8$, $\sigma'_0 = 85$ MPa and $\varepsilon'_0 = 1.4$, which are relatively close to those for the soft PZT ceramics reported in the study by Cao and Evans.[31] The stress–strain relationship assessed by Eq. (1) is indicated by the dashed line in Fig. 4(a). As can be seen, the estimated stress vs. strain is in good agreement with our experimental result (Stages 2 and 4).

Non-linear stress vs. strain relationships can be observed for the unpoled samples, as in Fig. 4(b), which shows results similar to the poled sample. However, no clear zigzag relationships are detected. This may be caused by weak domain switching compared to the poled sample. The strain level at 200 MPa (Stage 4) for the unpoled sample is about 1.0, which is about 30% lower than that for the poled one. The different strain level between the un- and poled ceramics could be attributed to the different severity of domain switching. This result is consistent with prior work, in which a four-point bending fatigue test was performed using a soft PZT ceramics with and without poling process.[32]

In previous work, it was reported that the ferroelectric material sheds some light on the dependence of the switching of individual domains on the electric field loading rate.[27] Because the zigzag stress–strain relations are associated with more severe domain switching, as mentioned above, the zigzag relationship might also be attributed to the lightning phenomenon. To confirm this, a direct observation was carried out with the PZT ceramics during compressive loading to 200 MPa using a video camera. In addition, the sound from the ceramic was recorded during the loading process using a frequency analyzer, AS-78, (RION Co., Ltd.). It was found from this experiment that electrical activity (a lightning-like phenomenon, consisting of a bright flash with a click) in the poled PZT ceramics occurs several times although such electrical activity cannot be observed for the unpoled samples. Representative pictures of electrical activity in the poled PZT ceramic are displayed in Fig. 6, where clear electrogenesis is seen between the lower and upper sides of the specimen. The sound wave data of the click sound from the ceramic during the loading process is identified in Fig. 7(a). An interesting result

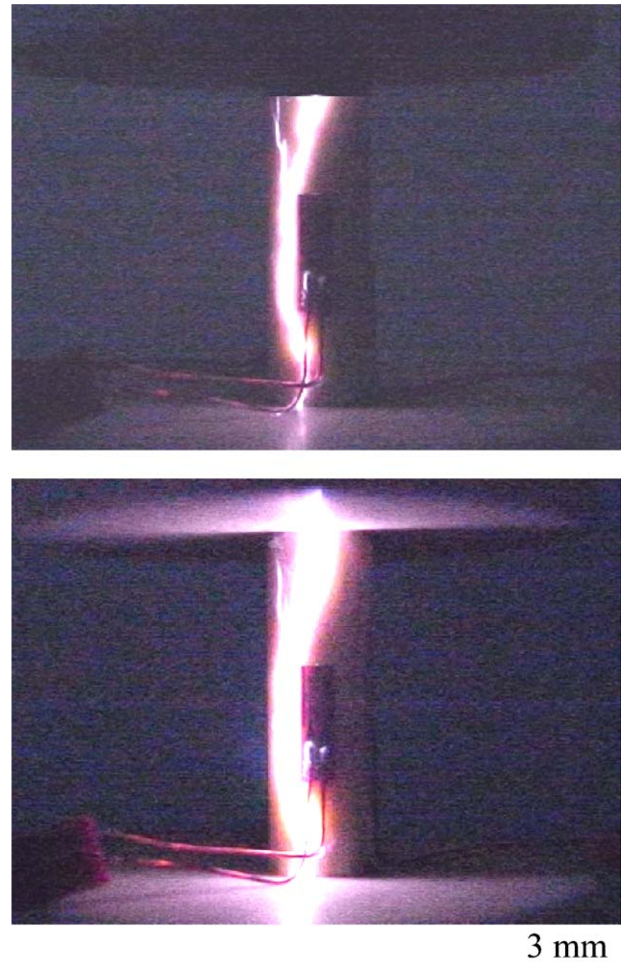


Fig. 6. Pictures of the specimen with electrogeneration during compressive loading.

in Fig. 7(a) is that the incidence of the click sounds (electrogenerative event) occur many times during the loading process and correspond to the step of the zigzag strain variation shown in Fig. 7(b). From this result, it is clear that the electrogenerative event is affected by domain switching.

To further understand the strain characteristics, the variation of strain of the PZT ceramic was measured during cyclic loading for five cycles. Fig. 8 displays the results of strain variations for the un- and poled PZT ceramics. In the 1st cycle for the poled sample (Fig. 8(a)), compressive strains of 0.73 and 1.5 were obtained in Stage 2 and Stage 4, respectively. In the unloading process of the 1st cycle, residual strains of 0.33 (Stage 3) and 0.99 (Stage 5) are observed. With increase of the cycle number, the strain level decreases suddenly, e.g., 1st cycle vs. 2nd to 5th cycle, and the strain values are almost constant following the 2nd cycle in all stages, which is a similar result to that seen in previous work.[33] In addition, although it seems that the overall strain level for the poled sample is high compared to the unpoled one (Fig. 8(b)), the strain values after the 2nd cycle for both samples are almost the same. This result suggests that the sample strain in the 1st cycle is mainly attributed to severe domain switching strain, and that lattice strain is the dominant feature in the following cycle. On the basis of the strain characteristics, the

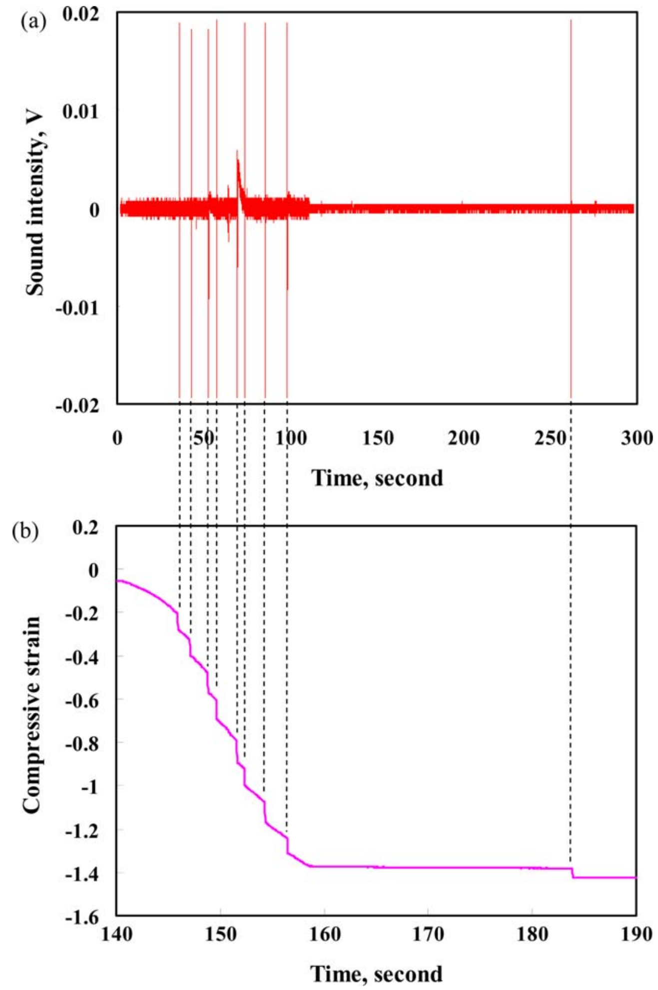


Fig. 7. Variation of (a) the sound intensity and (b) compressive strain during the loading test.

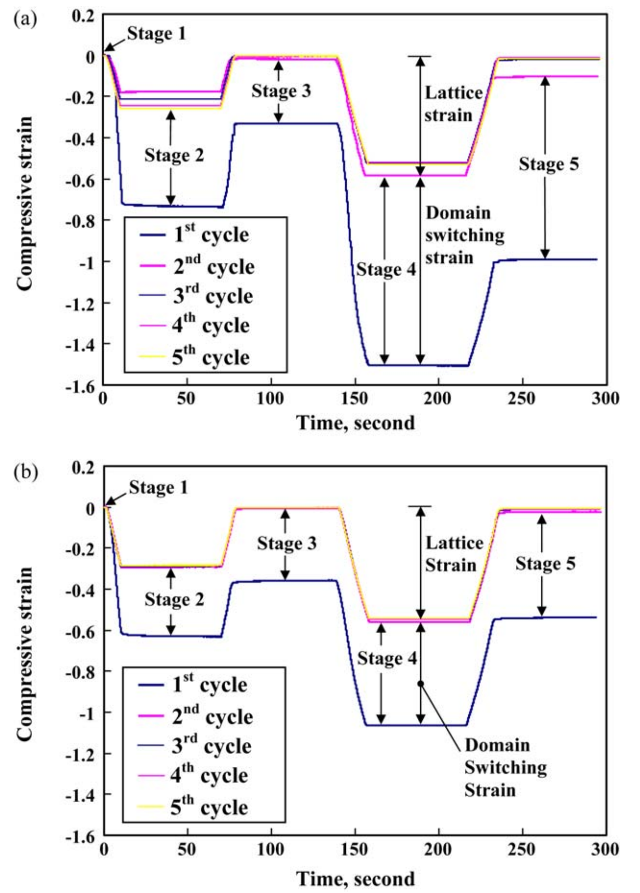


Fig. 8. Variation of compressive strain during the loading test: (a) poled sample and (b) unpoled sample.

ratio between the lattice strain (*LS*) and the domain switching strain (*DS*) can be estimated. In this case, the ratios of *LS*/*DS* for the unpoled and poled samples are 0.5 and 0.33, respectively, in which the lower the *LS*/*DS*, the stronger the domain switching. The ratio of 0.33 for the poled sample is close to the related sample (*LS*/*DS* = 0.36).[26]

3.2. Domain switching characteristics

To explain the domain switching characteristics more clearly, X-ray diffraction on the proportion of *c*- and *a*-oriented domains parallel to a given sample direction was examined. In this case, the long tetragonal *c*-axis can form parallel to any of the three ⟨0 0 1⟩ directions. The reorientation of domains by 90° domain switching from the *c*- and *a*-direction can therefore be identified and is given by the diffraction intensity ratio at the 2θ positions 43.627° and 44.880°.[11] This associated approach has been used by many researchers. Fig. 9 shows the X-ray diffraction analyses for our PZT ceramics during the loading and unloading process. With increase of applied load, the (2 0 0) peak decreases and the (0 0 2) peak increases. Such a change of the peak levels suggests 90° domain switching in the PZT ceramics.[14] Moreover, the (2 0 0) peak rises with decrease of the applied load, i.e., Stages 3 and 5. This occurrence may be related to the domain switching back to its original orientation.

In recent years, several researchers have quantitatively analyzed the amount of domain switching and domain texture using the X-ray diffraction pattern. Tanaka et al. have examined the tetragonal domain texture using the peak levels of (2 0 0) and (0 0 2) after waveform separation.[14] Pojprapai et al. have

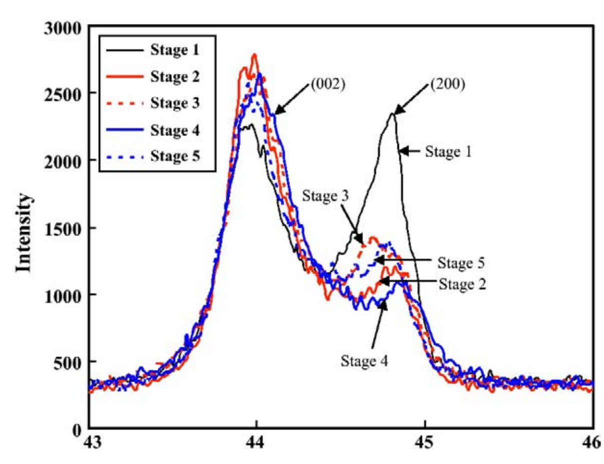


Fig. 9. X-ray diffraction patterns for the poled PZT ceramic.

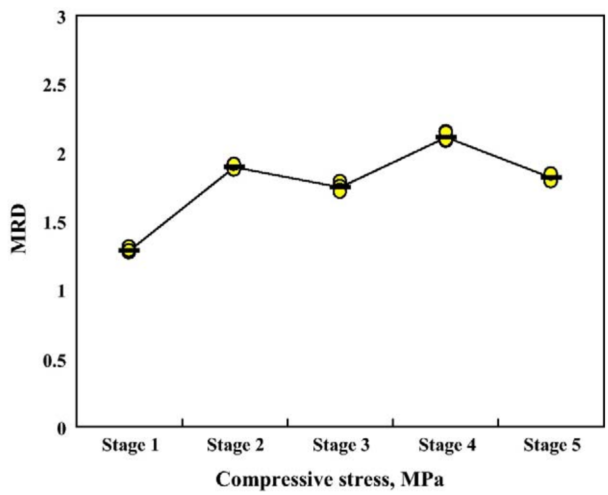


Fig. 10. MRD values obtained at different loading stages for the poled sample.

reported the domain switching behavior and shown that domain texture can be quantitatively analyzed using the unit multiple of a random distribution (MRD), where the MRD is equal to 1 when random domain orientation is obtained and the MRD becomes 3 as *c*-domains are all oriented parallel to the poling direction.[12] The formula to obtain the value of MRD can be expressed as follows:

$$MRD = \frac{3(I_{002}/I^R_{002})}{(I_{002}/I^R_{002}) + 2(I_{200}/I^R_{200})} \tag{2}$$

where I_{002} and I_{200} are the integrated intensities of the (0 0 2) and (2 0 0) peaks after mechanical loading. Details of the method of MRD estimation can be found in Ref. 12. On the basis of the X-ray diffraction analyses, the MRD values obtained for our samples at various loading stages are shown in Fig. 10. Before the loading process (Stage 1), the MRD value is approximately 1.28, and that value alters during the loading and unloading processes. As can be seen, the MRD level increases with increasing applied load, e.g., MRD = 1.89 (Stage 2) and MRD = 2.11 (Stage 4), and the MRD value drops with decrease of the loading level, e.g., MRD = 1.74 (Stage 3) and MRD = 1.81 (Stage 5). It should be pointed out that the MRD level cannot decrease to the initial MRD level of 1.3 (Stage 1) even after the applied stress is reduced to 0 MPa. This result implies that domain switching is reversed but it does not occur completely, resulting in residual strain.

To further understand the domain switching characteristics, the crystal orientation in the PZT ceramics was analyzed using EBSD analysis. This analysis was carried out at five different loading stages, as indicated in Fig. 3. In this examination, the domain orientation of 25 grains was characterized in advance. Fig. 11(a) and (b) displays the image quality maps and crystal orientation maps for the sample at the five different stages. The color level of each pixel in the crystal orientation map is defined according to the deviation of the measured crystal orientation with respect to the ND direction. The three orientations in the grains, for instance the areas with red, cyan and green, correspond to the three possible orientations (1 0 0), (0 0 1) and (1 1 0).[34] In this case, the (0 0 1) plane is related to the *c*-axis within a grain. The phases not considered in the representation are in black. From Fig. 11(b), the crystal orientation is apparently different for each individual grain. In addition, it is clear that different patterns of domain orientation can be seen, depending on the loading stage. With the analysis of the 25 grains as indicated in Fig. 11(a), three different switching systems can be distinguished. Representative switching patterns are shown in Fig. 12. As seen in Pattern I (Fig. 12(a)), 90° domain switching and reverse switching occurs during the loading and unloading processes, respectively. The amount of 90° domain switching increases with increase of the applied load, e.g., Stage 2 vs. Stage 4. In Pattern II, the amount of 90° domain switching increases with increase of the applied load, but further domain switching occurs even after removing the applied load (see Stage 3 Fig. 12(b)). This cannot be explained at the moment and will be discussed in the future. Unlike Patterns I and II, a unique 90° domain switching pattern is detected (Pattern III), in

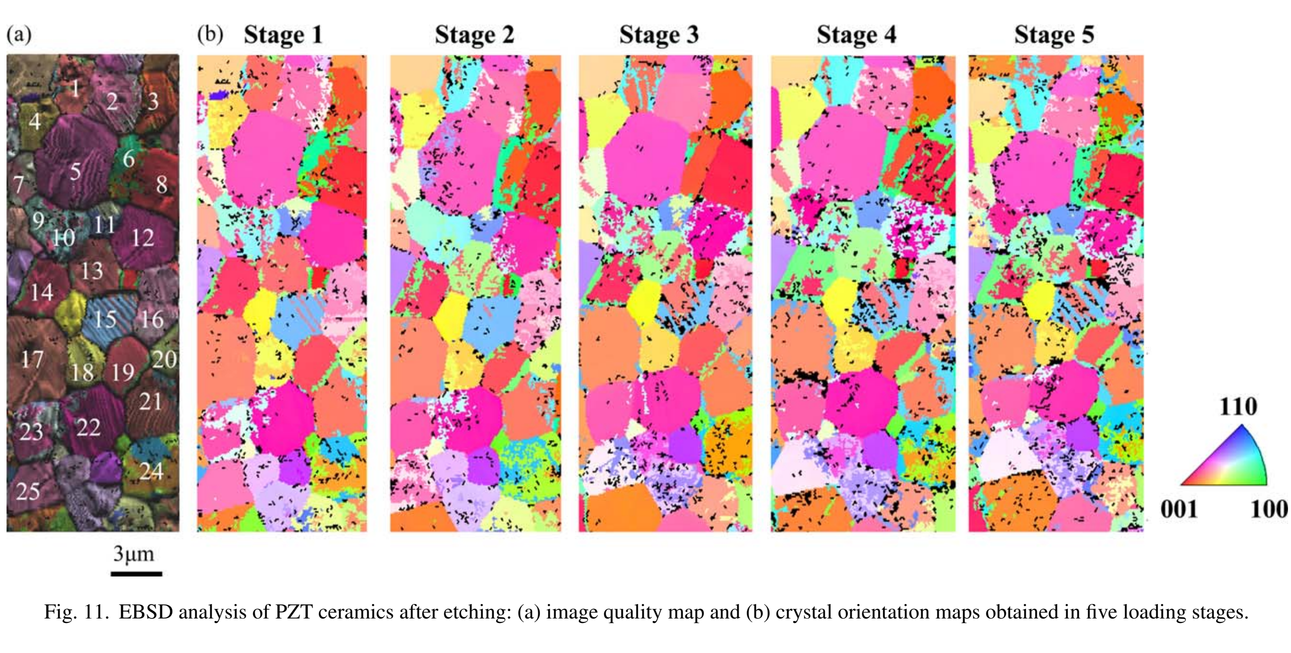
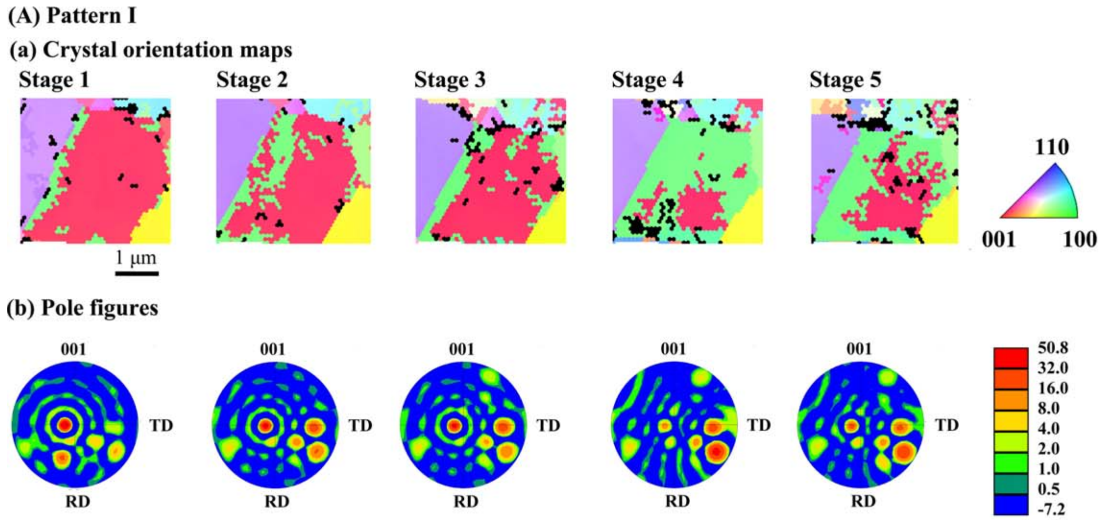
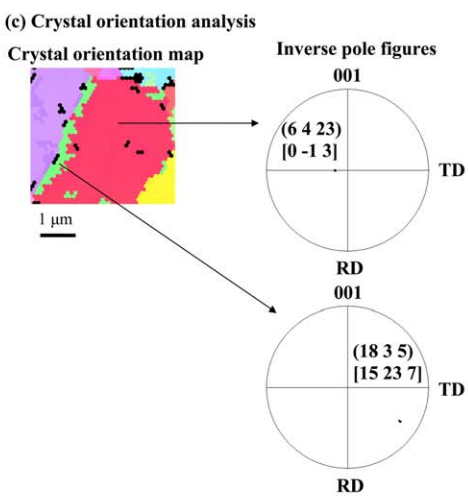


Fig. 11. EBSD analysis of PZT ceramics after etching: (a) image quality map and (b) crystal orientation maps obtained in five loading stages.





Rate of the crystal orientation (%)

	(6 4 23)	(18 3 5)
Stage 1	88.77	11.23
Stage 2	72.68	27.32
Stage 3	75.32	24.68
Stage 4	21.43	78.57
Stage 5	41.37	58.63

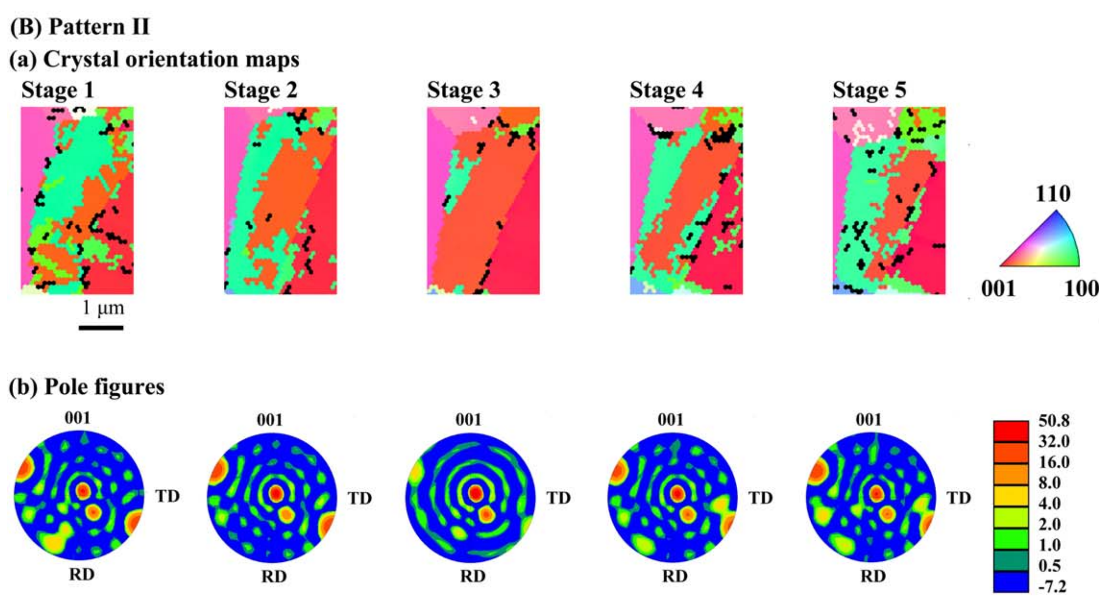
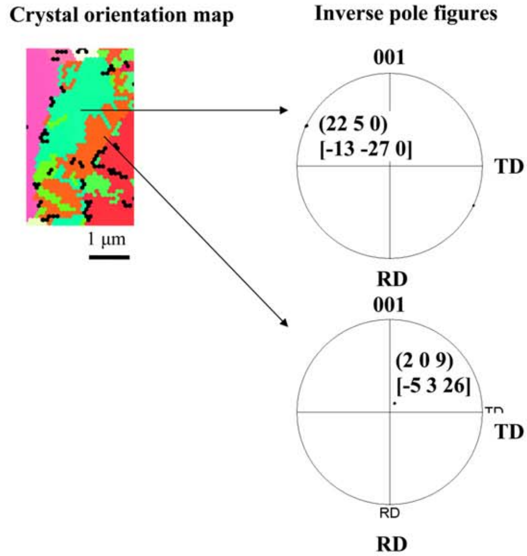


Fig. 12. EBSD analysis of PZT ceramics after etching: (a) image quality map and (b) crystal orientation maps and (c) crystal orientation analysis.

(c) Crystal orientation analysis

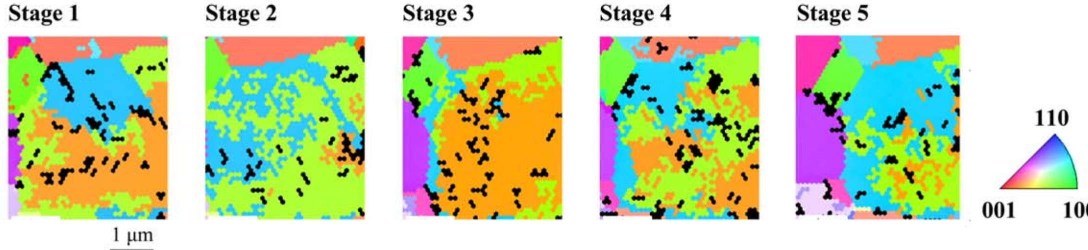


Rate of the crystal orientation (%)

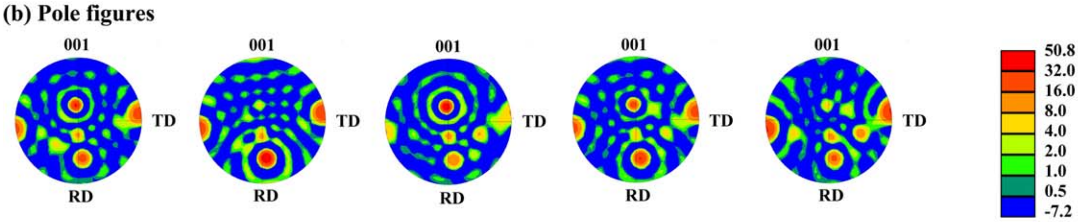
	(22 5 0)	(2 0 9)
Stage 1	57.61	42.39
Stage 2	49.04	50.96
Stage 3	9.69	90.31
Stage 4	46.48	53.52
Stage 5	60.15	39.85

(C) Pattern III

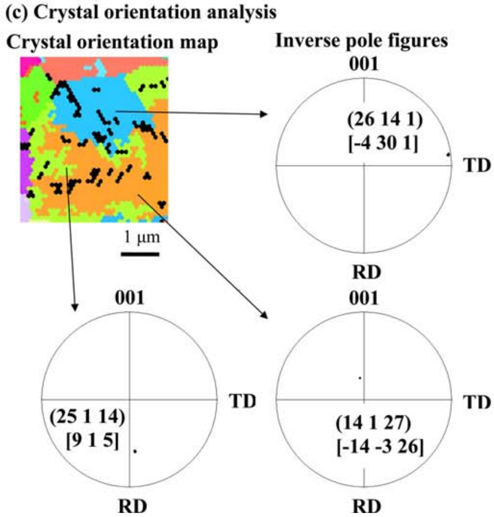
(a) Crystal orientation maps



(b) Pole figures



(c) Crystal orientation analysis



Rate of the crystal orientation (%)

	(26 14 1)	(14 1 27)	(25 1 14)
Stage 1	57.61	42.73	28.19
Stage 2	49.04	3.85	61.41
Stage 3	9.69	69.05	21.43
Stage 4	46.48	29.73	42.27
Stage 5	60.15	9.91	33.15

Fig. 12. *continued*

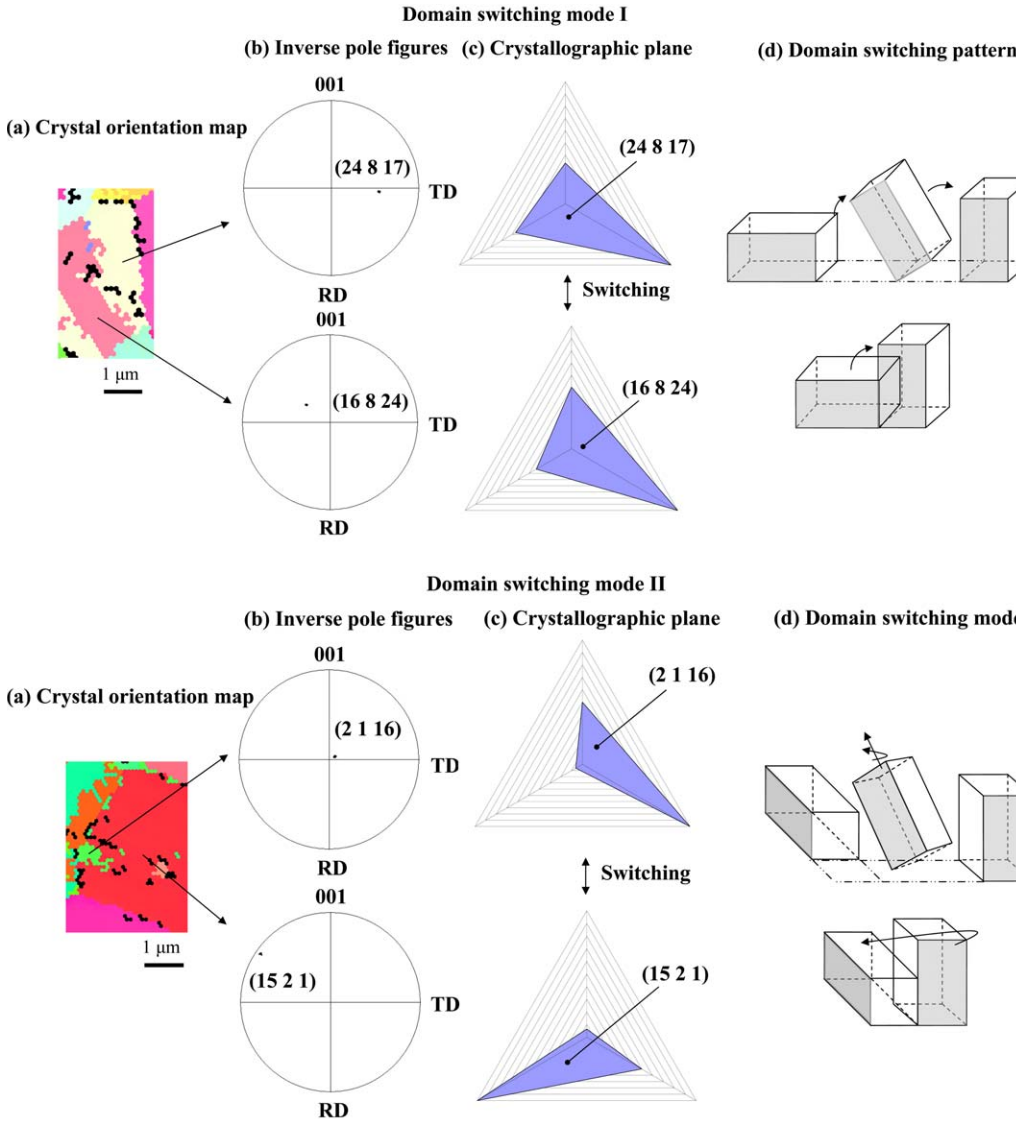


Fig. 13. EBSD analysis of poled PZT ceramics: (a) crystal orientation map, (b) inverse pole figure, (c) crystallographic plane and (d) domain switching mode.

which the tetragonal lattice turns to 90° with two split directions (Fig. 12(c)), e.g., (26 14 1) to (25 1 14) and (14 1 27). In addition, there are no clear domain switching grains even if a compression load has been applied. From the switching characteristics in 25 grains, the rate of switching systems can be summarized in Table 1. As seen, half of switching systems are associated with Pattern I and more than 25% of the system with Pattern II. The other systems showed low values of about 10%.

On the basis of the switching system in Table 1 (Pattern I, II and III), the 90° switching characteristics in each tetragonal structure were further analyzed as a function of crystal orientation before and after compression load. From this analysis, two different switching modes were observed. Fig. 13 displays the crystal orientation map, inverse pole figure, crystallographic plane and domain switching pattern. In this case, the crystallographic plane is indicated by a three-axis coordinate system (Miller-Bravais). From this, it is clear that there are two representative tetragonal lattice systems after 90° domain switching, as shown in Fig. 13(d). One is the crystal structure formed by 90° switching, and the other is the 90° switching with 90° rotation. Table 2 indicates the rate of domain switching mode obtained from the 25 grains. It is clear that about 60% and 30% of the switching modes were dominated by Mode II and Mode

Table 1
Rate of domain switching from three switching patterns shown in Fig. 11.

Pattern I	Pattern II	Pattern III	No switching
50%	28%	11%	11%

Table 2
Rate of two domain switching mode distinguished from three switching patterns in Table 1.

Mode I	Mode II	Others
31%	59%	9%

I, respectively. It should be noted that other switching modes (approximately 10% of grains) was also detected.

4. Conclusions

The domain switching characteristics of lead zirconate titanate (PZT) ceramics have been examined using various experimental approaches. On the basis of the experimental results and discussion, the following conclusions can be drawn.

(1) With compressive loading of PZT ceramics, the stress–strain curves have a form characteristic of that found for phase transformations, where the lattice strain and domain switching strain are affected. The domain switching strain forms the permanent strain of PZT ceramics. In the unloading process for the poled sample, some domains switch, and reverse switching may occur.
(2) The strain level for the poled sample at the maximum applied load of 80% fracture stress is about 30% higher than that for the unpoled one. The high degree of strain for the poled samples is caused by the greater extent of domain switching.
(3) Electrical activity in the PZT ceramics occurs several times, related to a lightning-like phenomenon and consisting of a bright flash with a click. This electrogenerative event is related to severe localized domain switching.
(4) With the X-ray and EBSD analysis, the domain switching characteristics during the unloading and loading processes have been clarified. (i) 90° domain switching and reverse domain switching occurs during the loading and unloading processes, respectively; (ii) the amount of 90° domain switching increases with increasing applied load, and further increases even after the unloading process; (iii) 90° domain switching occurs with two different patterns (the *c*-axis tetragonal crystal rotates in two different directions).
(5) The 90° switching characteristics in each tetragonal structure were further analyzed, and two different switching modes were detected. One is the crystal structure formed by 90° switching, and the other is the 90° switching with 90° rotation.

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