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Failure Characteristics of PZT Ceramic During Cyclic Loading

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Failure characteristics of $PbZrTiO_3$ (PZT) ceramic plates are investigated under cyclic loading with rods of different diameters, i.e., different contact areas (0–20 mm). The voltage generated under loading by the rod with the smallest diameter (contact area) is higher than those for the larger contact areas. This is due to the high strain induced in the PZT ceramic. However, the opposite trend is seen when the loading exceeds 60 N, i.e., the voltage obtained for the smallest contact area is lower. This is caused by failure of the PZT ceramic. The voltage generated under cyclic loading by the 5-mm, 10-mm, 15-mm, and 20-mm rods drops by about 10% in the early cyclic loading stage, but then remains constant until 10,000 cycles. The reduction in voltage is influenced mainly by 90° domain switching. In this case, many grains (about 15% of the total) are switched: a random domain orientation is switched to the $\langle 100 \rangle$ direction perpendicular to the ceramic plate, i.e., a crystalline texture is formed. In contrast, there is significant reduction in voltage under loading by the 0-mm rod (point contact). As the extent of domain switching for the 0-mm rod is similar to that for the other rods, the reduction in electrical generation can be attributed to crack generation resulting from the high deformation.

Key words: Piezoelectric ceramic, lead zirconate titanate ceramic, electrical power generation, domain switching, cyclic loading

INTRODUCTION

Environmental issues have led to increasing interest in clean energy systems, based for example on wind, solar, wave, or geothermal power. Piezoelectric and thermoelectric materials, made of rubber and ceramics, are expected to play an important role as components of such systems for generation of electricity. Piezoelectric ceramics possess the unique property of a spontaneous polarization that is reversible in an applied electric field. Their importance lies in the induction of a voltage difference across two surfaces of a piece of piezoelectric ceramic material as it is subjected to high alternating stress. A number of lead-based and lead-free piezoelectric ceramics[1–3] have been developed for various engineering applications, including, among others, $PbZrTiO_3$ (PZT) ceramics, BaTiO piezoelectric ceramics, and $Bi_4Ti_3O_x$ ferroelectric ceramics. Because of their excellent electromechanical response, PZT ceramics are used in a wide range of applications.

Energy harvesting from vibrating structures[4] is a recently developed application in which piezoelectric materials are used for the generation of electrical energy.[5] Harvested energy systems have been employed in various locations,[6] such as ticket gates, buildings, and bridges, in which PZT ceramic plates, placed on floors, are subjected to cyclic mechanical loads by people walking on these floors. In these situations, because the PZT ceramics are loaded repeatedly for a long period of time, the possibility of their failure must be considered. Piezoelectric ceramics exhibit several failure characteristics, for example, domain switching and crack generation,[7] which lead to deterioration of their piezoelectric properties, although the detailed nature of the effect of failure characteristics on the generation of

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electricity has yet to be clarified. Domain switching or domain motion can occur under high applied stress and the consequent elastic strain.[8] It has been reported that the dielectric constant of PZT films can be attributed to 180° domain wall motion.[9] Localized domain switching adjacent to a crack tip will have a significant effect on the stress intensity and energy release rate. Because domain switching is directly related to electrical generation characteristics,[10] there have been a number of investigations of domain orientations using x-ray diffraction (XRD) and electron backscatter diffraction (EBSD).[11,12]

Because energy harvesting with PZT ceramics is not very efficient, an optimized harvesting technique is required. There have been a number of experimental and numerical studies of the efficiency of electrical power generation by PZT ceramics under loading.[13] The electrical voltage is sharply enhanced with increasing strain in the early loading stage. Following this initial increase, there is a rapid fall to zero voltage because of the elimination of electrons in the PZT ceramic. The rate of decrease depends on the flexibility of the ceramic because of the consequent variations in strain. Good estimates of voltages generated by PZT ceramics can be obtained by numerical methods using commercial software.

In order to gain a deeper understanding of the electrical generation characteristics of PZT ceramics, various experiments have been conducted under different loading conditions. However, there have been no continuous investigations of the voltage generated under cyclic loading for long periods of time. Information on the fatigue properties of PZT ceramics is especially important for energy harvesting systems, because more than 90% of component failures are caused by fatigue. Therefore, the aim of the present work is to investigate the effect of material failures on electrical power generation under various cyclic loading conditions for a high number of cycles (10,000). This failure analysis, including domain switching, is conducted using EBSD.

EXPERIMENTAL PROCEDURES

Material and Experimental Conditions

The material used in this study is a commercial $PbZrTiO_3$ (PZT)-based ceramic of tetragonal structure with a *c*/*a* ratio of 1.014, produced by Murata Manufacturing Co., Ltd. in Japan. Thin circular plates 25 mm in diameter and 0.3 mm thick were fabricated, and were fixed to a round brass plate. A silver-based electrode was coated onto one surface of each plate. The representative piezoelectric properties of these PZT ceramic plates were as follows: electromechanical coupling coefficient $k_{33} = 0.68$, piezoelectric constant $d_{33} = 603$ pm/V, elastic constant $S_{33}^{E} = 18.8$ pm^2/N, Poison's ratio $\sigma^{E} = 0.36$, density $\rho = 8.0 \times 10^3$ kg/m^3, permittivity $\varepsilon_{33} = 20.5$ nF/m, and relative permittivity $\varepsilon_{33}^{T}/\varepsilon_0 = 4720$.[14]

Figure 1 shows a schematic illustration of the test apparatus. A PZT ceramic plate, placed on the sample stand, was loaded via a circular rod. The sample stand and loading rods were made of commercial wrought aluminum alloy, e.g., JIS-A5052. Different rod diameters (contact area: 0 mm, 5 mm, 10 mm, 15 mm, and 20 mm) were used so that the stress level in the ceramic plate could be varied: the smaller the rod diameter, the greater the stress. Note that the contact area of 0 mm is related to the point-like contact, where the tip of the loading rod was designed to be spherical in shape. A bending load was applied cyclically to the ceramic plate in air using a screw-driven universal testing machine with 50 kN capacity (Shimadzu, AG-Xplus). The detailed loading conditions were as follows: the maximum load was applied at 10–100 N for either 10 or 10,000 cycles in a square-wave loading mode with R-ratio = 0.05 and loading frequency $f = 0.25$ Hz. Note that the cyclic loading was conducted slowly to reduce the risk of sample damage caused by a high loading speed.[15] The load and strain values of the ceramic plate during cyclic loading were measured using a commercial load cell and a strain gauge, respectively. The strain gauge was attached to the center of the ceramic plate for this measurement (see Fig. 1). The electrical power generated by the PZT ceramic was measured during cyclic loading using a digital multimeter (Fluke 8846A). In the present study, the mean maximum voltage was used as a parameter to evaluate electrical power generation.

Failure analysis was carried out using a scanning electron microscope (SEM) and EBSD. The EBSD analysis was conducted under the following conditions to examine the domain orientations before and after cyclic loading: accelerating voltage 15 kV, beam current 12 nA, and step size 1 μm. The sample surfaces for the analysis were polished mechanically using a cloth with alumina particles.

RESULTS AND DISCUSSION

Electrical Power Generation Under Different Loading Stresses

Figure 2 shows the variation in the mean maximum voltage as a function of the maximum load for different circular rods. Note that the mean voltage was estimated using the data obtained for a 10-cycle loading. The voltage generally increases with increasing applied load, although its actual value depends on the rod diameter. When a low load of less than 50 N is applied, a high voltage is obtained for the smaller-diameter rods, owing to the high stress level. However, a different trend in the voltage is seen at loads above 60 N: the voltages for the smaller rod diameters of 0 and 5 mm are lower than those for larger diameters. In this case, the voltages for the 15-mm and 20-mm diameters are as high as 90 V at 100 N, which is more than 10% higher than that for the 0-mm rod. The lower

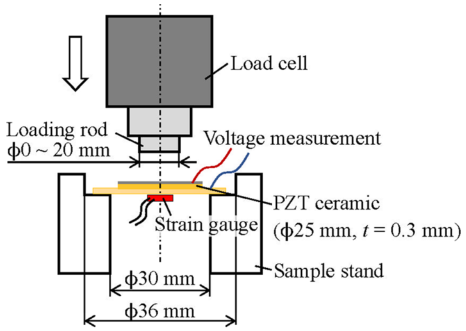

Fig. 1. Schematic diagrams of loading apparatus for investigating electrical power generation by PZT ceramic plates.

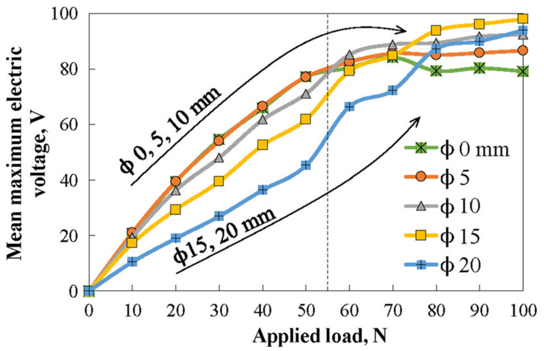

Fig. 2. Mean maximum voltage as a function of load applied by various loading rods.

voltage for the smaller rods may be attributed to damage to the PZT ceramic.

Figure 3 shows the variation in the bending strain of the PZT ceramic plates as a function of the applied load for different circular rods. It is clear that the strain increases with increasing applied load, and high strain is obtained under loading with the smaller rods. This may be associated with the generated voltage, especially at low applied loads of less than 50 N (see Fig. 2). Interestingly, the strain increases nonlinearly with increasing applied load for the 0-mm rod at the high load values, where severe failure may occur, leading to the low voltage shown in Fig. 2.

Electrical Power Generation for Cyclic Loading

Figure 4 shows the voltage as a function of the number of loading cycles up to 10,000 cycles for a PZT ceramic loaded at a maximum of 100 N using loading rods with diameters of 0 mm, 5 mm, 10 mm, 15 mm, and 20 mm. Note that the results for diameters of 0 mm and 5 mm in Fig. 4 were also presented in our previous work.[10] As can be seen, the voltage decreases in the early fatigue stage, following which it remains almost constant for all loading conditions. In our previous work, similar results were found at a lower load of 40 N.[10] The rate of decrease is clearly different for the 0-mm rod: in the early fatigue stage at less than 2000 cycles, the voltage drops significantly by about 15% for the 0-mm rod, while it drops by less than 10% for the other rods with diameters of 5–20 mm. These different electrical generation characteristics could be caused by different types of material failure. Jones and Hoffman[16] investigated the failure characteristics of piezoelectric ceramics during unipolar cycling at 1 Hz and 50% of the coercive field, where non-180° domain switching gives rise to about 34% of the measured d_{33} coefficient of 400 pm/V.

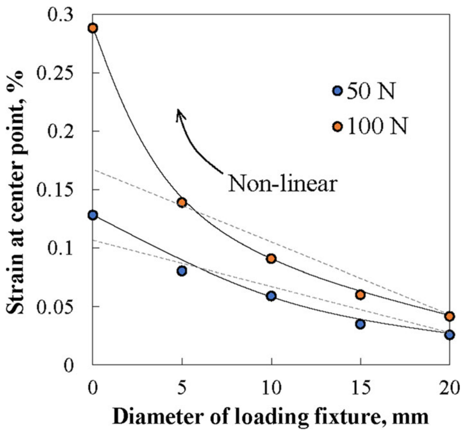

Fig. 3. Strain on a PZT ceramic plate as a function of applied load.

To understand the failure characteristics of the PZT ceramic, the profiles of ceramic plates during cyclic loading were examined. Figure 5 shows the cross-sectional shape of a ceramic plate before loading and after 1000, 5000, and 10,000 cycles. The profiles were measured using a stylus profilometer at 0.6 mm/s (SURFCOM 1500DX2, Tokyo Seimitsu Co., Ltd.). Before cyclic loading, the plate had a convex profile, which is the original shape of the PZT ceramic. This convex shape may be created by strain arising during firing of the ceramic on a brass plate. After loading for 1000 cycles using rods of 5-mm, 10-mm, 15-mm, and 20-mm diameter, the profile height was clearly reduced, although further cyclic loading for 5000 and 10,000 cycles did not cause any significant further changes to the profile. That is to say, the deformation of the PZT ceramic plate occurs in the early stage of less than 1000 cycles. Note that the deformation of the plate after loading with the 5-mm diameter rod is greater than that after loading with the 10-mm, 15-mm, and 20-mm rods, and the plate becomes almost flat. Even more severe deformation is seen for the 0-mm rod, with the initially convex shape of the plate becoming concave at 1000 cycles, and further deformation occurring as the loading is increased to 10,000 cycles. Such degrees of deformation could lead to severe deterioration of piezoelectric properties.

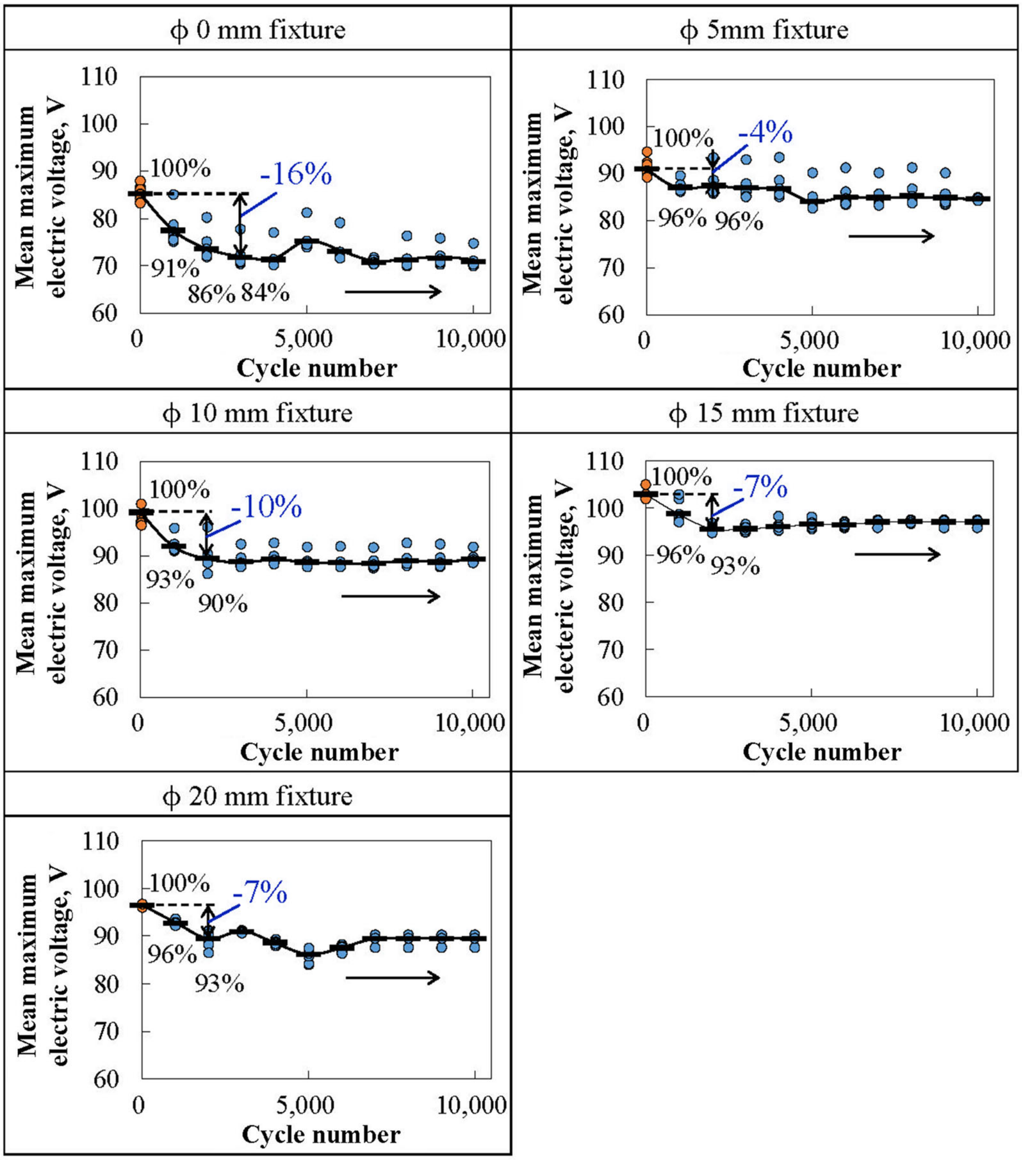

Fig. 4. Maximum voltage generated by PZT ceramic plate as a function of cycle number at a maximum load of 100 N.

To understand the decrease in generated voltage, the failure characteristics of the PZT ceramic were examined by EBSD before and after cyclic loading using the 0-mm and 20-mm rods at 100 N for 10 cycles. The observation was conducted in an area 80 × 25 μm in the center of the PZT ceramic plate. Figure 6 shows inverse pole figure (IPF) maps of the PZT ceramic at low magnification before and after cyclic loading. The same area of the ceramic was observed to enable a clear comparison of the degree of domain switching before and after loading. It is obvious from Fig. 6a and b that there are changes over a wide area that are caused by domain switching, i.e., the formation of a crystalline texture. It can be seen from the pole figures that the surface textures arising from domain switching are related to the $\langle 100 \rangle$ direction (red regions). A similar approach was adopted by Hammer et al.,[17] who found that the surface texture of PZT ceramics is dependent on the applied stress and that this cannot be completely removed by heat treatment. The switched grains are marked in yellow in the image quality (IQ) maps in Fig. 6a and b. From a domain analysis of more than 1000 grains, the number of switched grains is found to be about 15% of the total. Furthermore, it is clear from Fig. 6c that cracks are generated on the PZT ceramic plate after loading with the-0 mm rod, which could be related to severe failure of the ceramic. Note that several cracks are observed, and that these were detected only after loading with the 0-mm rod. This may be attributed to the high strain in this case, as shown in Fig. 3, and it results in the reduction in electrical power generation.[18]

The domain switching characteristics were examined in greater detail. As already noted, the rate of

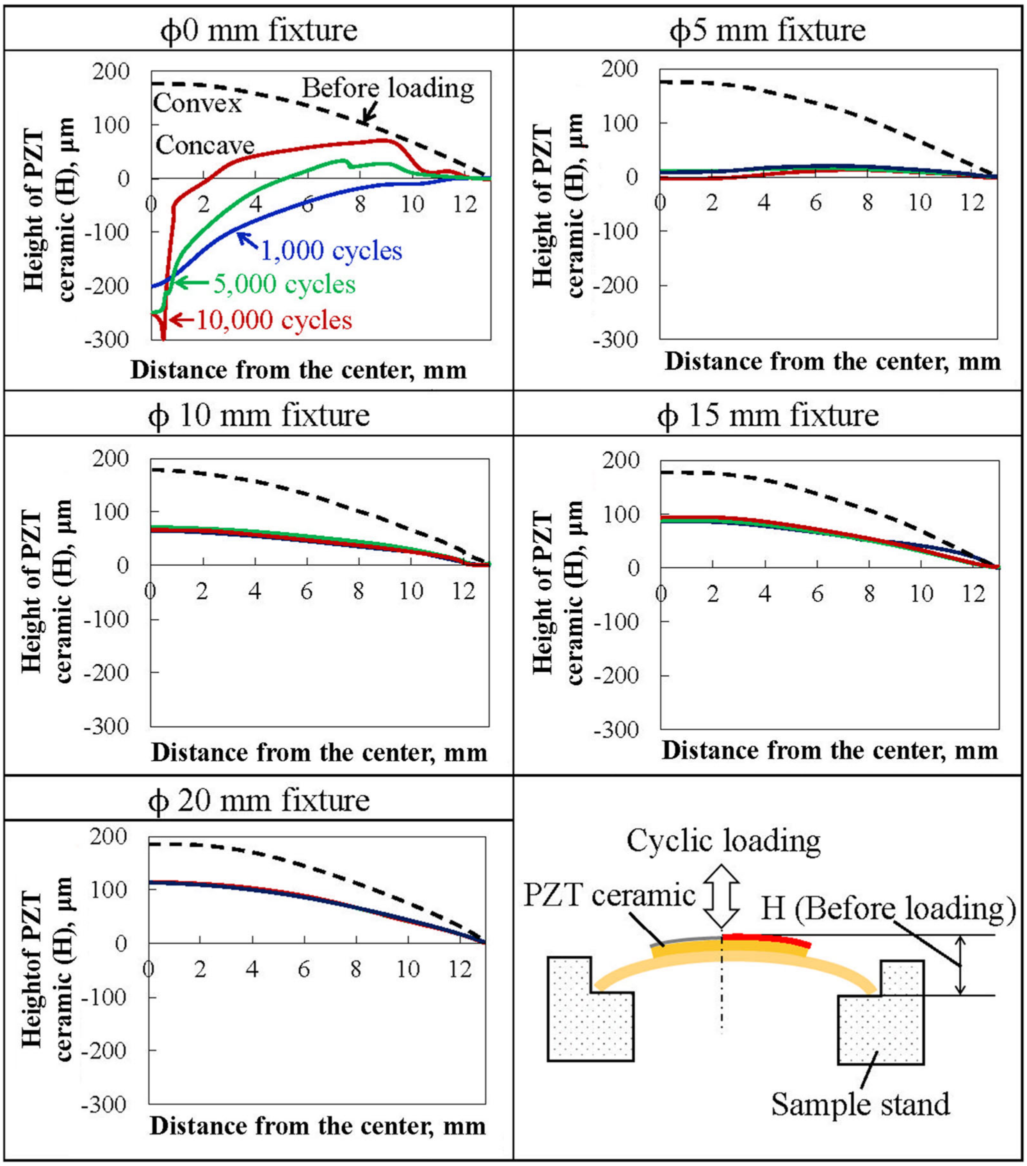

Fig. 5. Configuration of a PZT ceramic plate after loading at 0, 1000, 5000, and 10,000 cycles.

grain switching (as indicated by the red regions after loading) was similar for loading with the 0-mm and 20-mm rods. In other words, even when only a low stress was applied (20-mm rod), domain switching occurred just as in the case of high stress (0-mm rod). It also appears that almost all domains switched to the $\langle 100 \rangle$ direction (about 89.7°) can be categorized into three different patterns, as summarized in Fig. 7. A similar result has been reported previously.[19] In this case, the $\langle 100 \rangle$ direction of the tetragonal structure, i.e., the c axis, is perpendicular to the PZT ceramic plate after loading. This can be explained by the fact that the c axis ($\langle 100 \rangle$ direction), which is initially parallel to the plate, is rotated through 90° when tensile and compressive stresses are applied to the $\{100\}$ plane of the tetragonal structure (see Fig. 8). As mentioned previously, the reduction in voltage when the ceramic plate is loaded by the 0-mm rod is much greater than when it is loaded by the 20-mm rod. Because the extent of domain switching is similar for all loading rods, the low voltage generated under loading by the 0-mm rod could be due to failure caused by severe crack generation.

CONCLUSIONS

Failure characteristics of PZT ($PbZrTiO_3$) ceramic plates have been investigated under cyclic loading with rods of different diameters (0–20 mm), i.e., different contact areas. The results obtained can be summarized as follows:

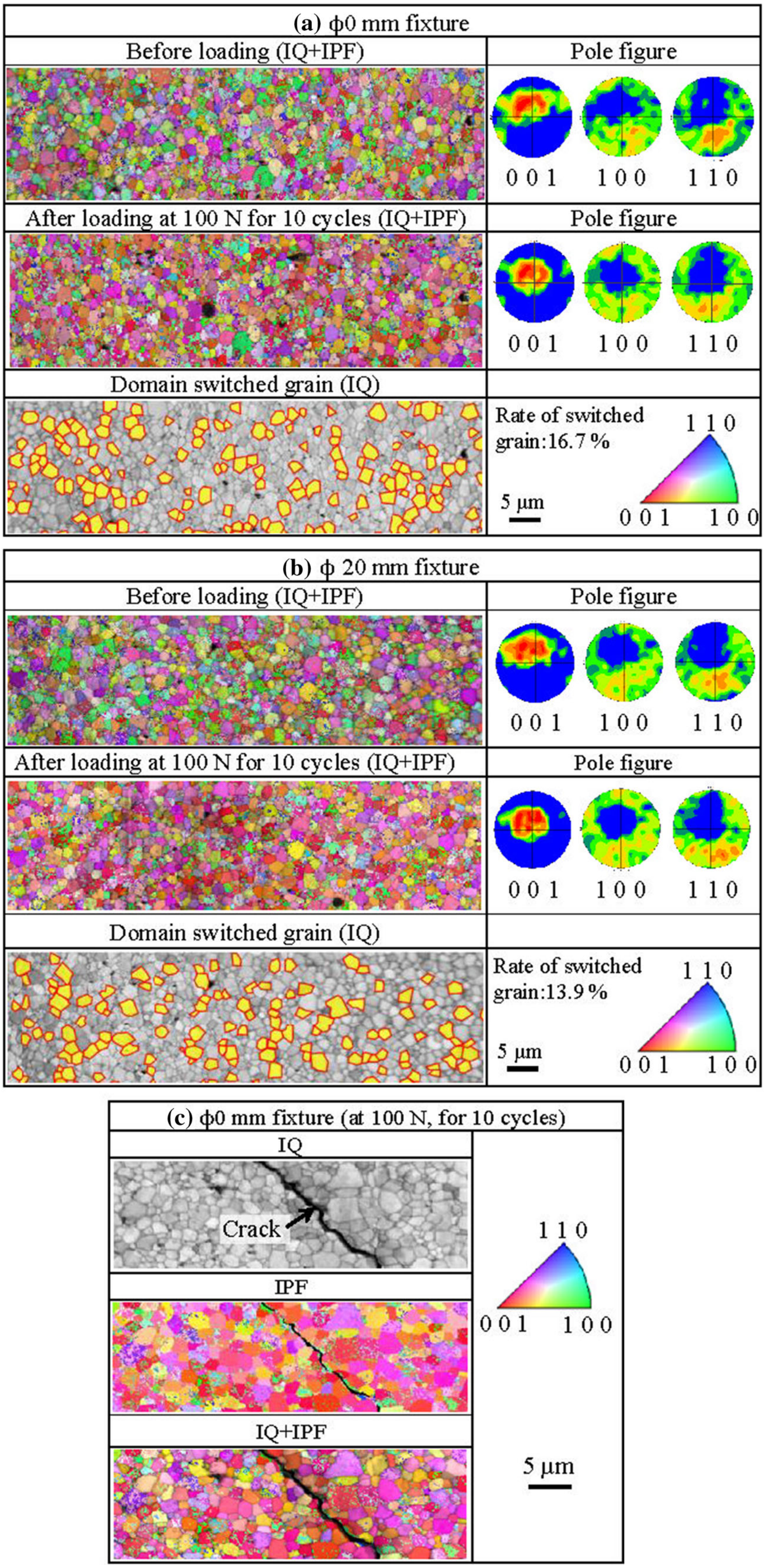

Fig. 6. EBSD analysis of PZT ceramics before and after loading at 100 N for 10 cycles: (a, b) domain orientation for 0-mm and 20-mm rods; (c) crack generation for 0-mm rod.

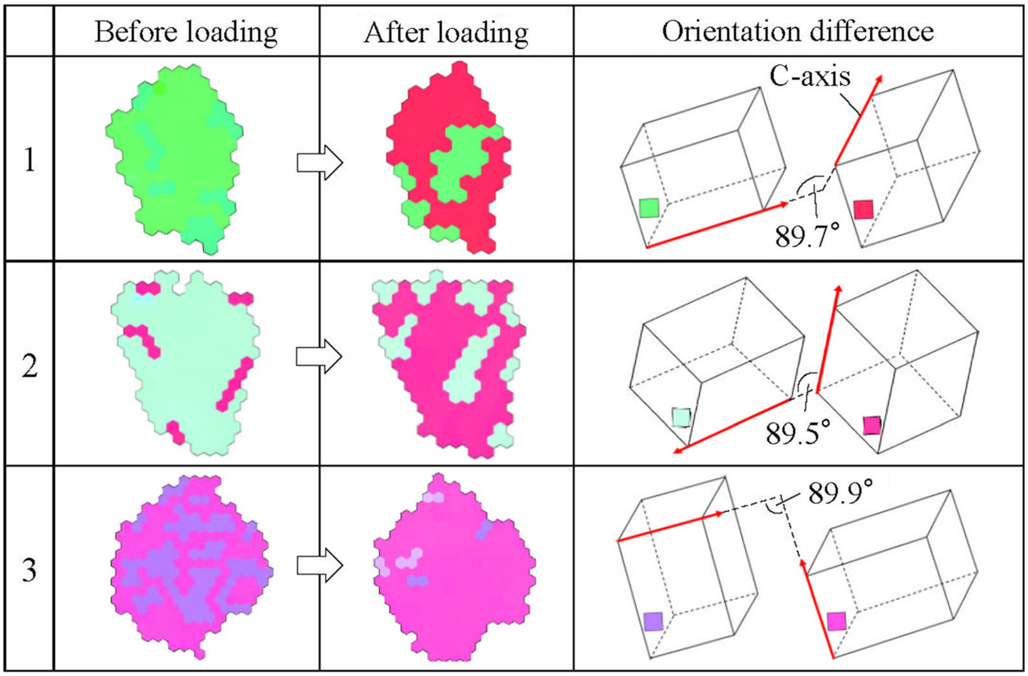

Fig. 7. Crystal orientation analysis of representative switched patterns before and after cyclic loading.

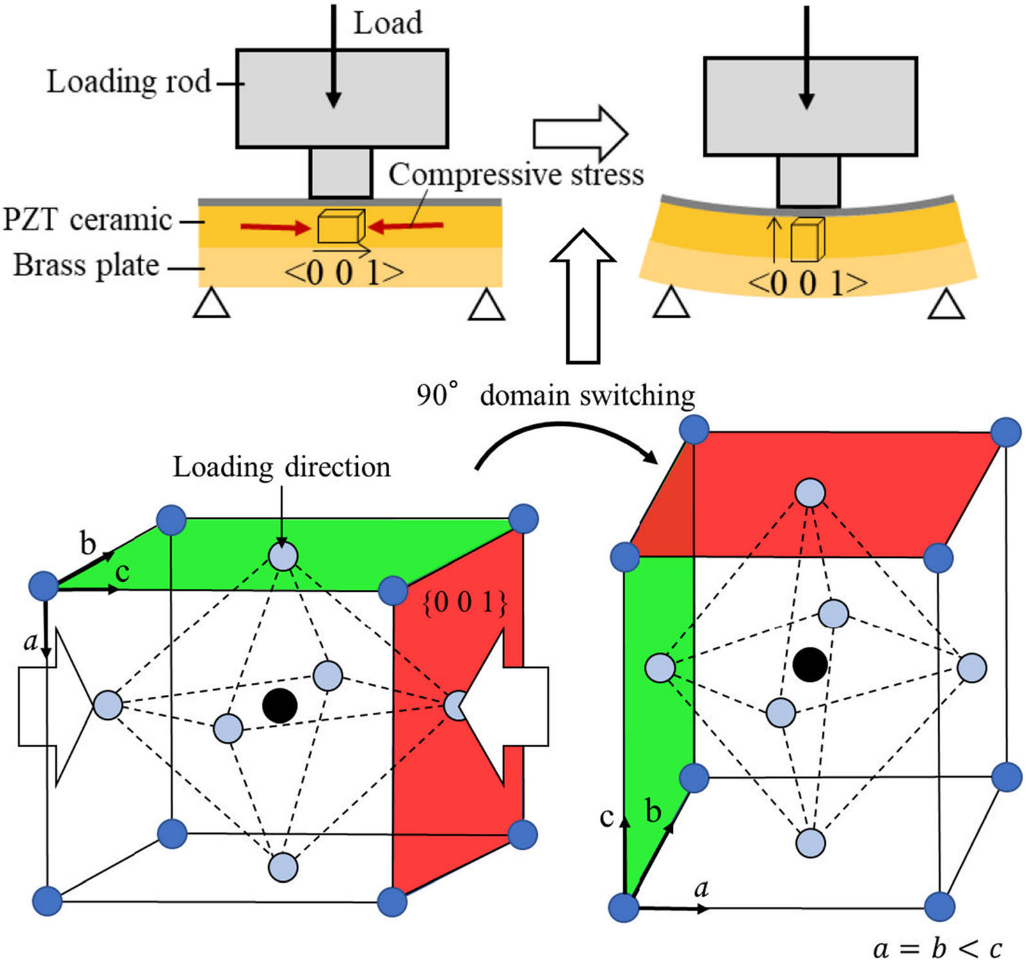

Fig. 8. Schematic diagram showing mechanism of 90° domain switching of a PZT ceramic plate.

1. A smaller contact area enhances the generated voltage, owing to the increase in strain. However, the voltages for the smallest contact areas (the 0-mm and 5-mm rods) are lower than those for larger contact areas when a high load of more than 60 N is applied. This is because of the failure of the PZT ceramic arising from the high stress concentration with the smaller-diameter rods. Under loading for 10,000 cycles, a reduction in the generated voltage occurs in the early fatigue stage (i.e., less than 1000 cycles), following a period of stable electrical generation.
2. The voltage generated under cyclic loading similarly decreases for the loading rods with diameters of 5 mm, 10 mm, 15 mm, and 20 mm. The profile height of the convex PZT ceramic plate decreases under loading by the 10–20-mm rods, and the plate becomes almost flat for the 5-mm rod. These changes in shape are caused by 90° domain switching, in which a random domain orientation is switched to the $\langle 100 \rangle$ direction perpendicular to the plate, i.e., a crystalline texture is formed. The number of switched grains is found to be about 15% of the total.
3. There is a considerable reduction in voltage generated by the PZT ceramic under cyclic loading with the 0-mm rod owing to the severe failure that occurs. The originally convex plate becomes concave after loading for fewer than 1000 cycles. In this case, the significant reduction in electrical generation is caused not only by the 90° domain switching but also by crack generation. As the extent of 90° domain switching after loading with the 0-mm rod is similar to that after loading with the 20-mm rod, the reduction in electrical generation in the former case must be due to crack generation.

CONFLICT OF INTEREST

The authors declare no conflict of interest.

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