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Electric Loading- Induced Cracking Behavior at Electrode Edges in PZT Ceramics

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Abstract. Fatigue damage behavior under repeated electric loading was investigated on two kinds of PZT ceramics with discontinuous electrodes. Intergranular cracking was observed at the electrode edge in soft PZT under electric fields greater than ±400 V/mm. However, under the same loading conditions, no damage was observed in hard PZT. When cracking occurred, permittivity of specimens decreased with the number of cycles corresponding to the amount of mechanical damage. FEM analyses of the electroelastic field of the specimens showed that cracking due to cyclic electric loading was related to 180˚ domain switching caused by concentrated electroelastic field.

Introduction

Lead Zirconate Titanate (PZT), a perovskite-type ferroelectric ceramic, can exhibit macroscopic piezoelectric effect and inverse effect after a poling process. PZT has been used extensively for sensors and actuators due to its high piezoelectric performance. Recently, it is expected to be used as a material for intelligent systems.

However, caution must be exercised when using this material because it is brittle. When used in piezoelectric devices, micro-cracks or defects in the material may cause the device to fail or fracture. In general, voltage is applied on PZT through electrodes. A discontinuous electrode causes a concentration of electric field. It has been reported that numerous micro-cracks are initiated at the electrode edge in multilayer actuators under the repeated application of electric fields [1]. In order to increase the reliability of the material, it is important to understand its behavior under repeated electric loading. In this study, mechanical damage such as formation of micro-cracks at electrode edges and permittivity degradation behavior caused by electric loading were investigated on two kinds of PZT ceramics having discontinuous electrodes.

Materials and Experimental Procedure

This study examined soft and hard PZT ceramics (Furuuchi Chemical Co., Ltd., Japan). The properties of materials after poling are listed in Table 1. Specimens were cut from a pellet, with a diamond cutter, to a size of 5 × 5 × 1 mm. The poling direction coincided with the direction of the thickness. One major surface (5 × 5 mm) was polished with 1 μm diamond paste and spattered with a 2 mm width of Au/Pd as a discontinuous electrode. The opposite surface was used as the full electrode as shown in Fig. 1.

A sine wave alternating voltage was applied between the electrodes with electric field ranges,

Table 1 Material properties of specimens.

		Soft PZT	Hard PZT
Elastic modulus s_{33} ($\times 10^{-12}$ m^2/N)		10	14
Piezoelectric coefficient d_{33} ($\times 10^{-12}$ m/V)		1700	510
Dielectric permittivity ε_{33} ($\times 10^{-10}$ C/Vm)		215	112
Mean grain size (μm)		3.4	6.0
Density (g/cm^3)		8.0	8.0
Fracture toughness (MPa·m$^{1/2}$)	C//P	1.17	1.4
	C⊥P	0.31	0.57
Zr:Ti mol ratio		42:58	44:56

ΔE of ±100, ±200, ±400 and ±800 V/mm at frequencies of 200, 1000 and 2000 Hz. The laser microscope was used to observe the surface, and the capacitance between the electrodes was measured using an LCR meter at suitable cycle intervals.

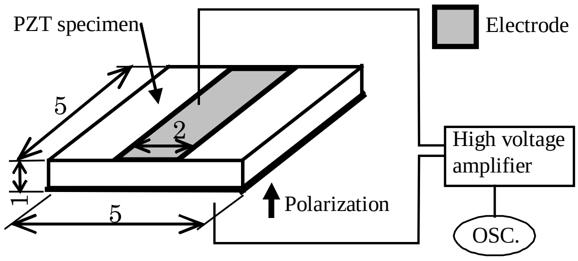

Fig.1 Specimen and testing system.

Results and Discussion

Behavior of Mechanical Damage. Fig. 2 shows successive observations near the electrode edge of a soft PZT specimen under ΔE = ±400 V/mm. The intergranular cracks were initiated along the edge of the electrode. As the number of cycles increased, the breadth of cracked region spread further. Fig. 3 shows SEM micrographs of the cracked region after testing. After intergranular cracks initiated, the rise of the grain with a cracked boundary became prominent due to cyclic deformation. However, there was no cracking at field strengths (ΔE) below ±200 V/mm. For the hard PZT, there was no damage even when the specimen was applied high ΔE of ±800 V/mm.

The relationships between the breadth of a cracked region and the number of cycles under ΔE = ±400 and ±800 V/mm are shown in Fig. 4. It can be seen that higher ΔE shortened the crack initiation life. Rate of increase of cracking breadth increased rapidly at 10^7 cycles regardless of ΔE.

Behavior of Permittivity Change. The capacitances between the electrodes of specimens were measured as a standard for material damage. Fig. 5 compares the permittivity normalized by an initial unit prior to testing and number of cycles under ΔE = ±400 V/mm. For the soft PZT, in which intergranular cracking appeared, the permittivity decreased remarkably as number of cycles increased. After 10^8 cycles, the permittivity declined by about 6%. However, there was no notable decrease in permittivity for hard PZT. The value remained constant for up to 10^8 cycles.

Fig. 6 shows the effect of the electric field range on the permittivity change. When ΔE was less than ±200 V/mm, the permittivity did not decrease, because there was no cracking. Therefore, there is a correlation between mechanical damage such as cracking and electrical damage such as a decrease in permittivity.

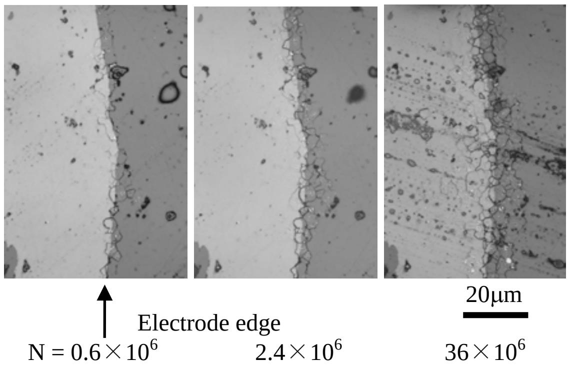

Fig.2 Observations at electrode edge of soft PZT under ΔE = ±400 V/mm.

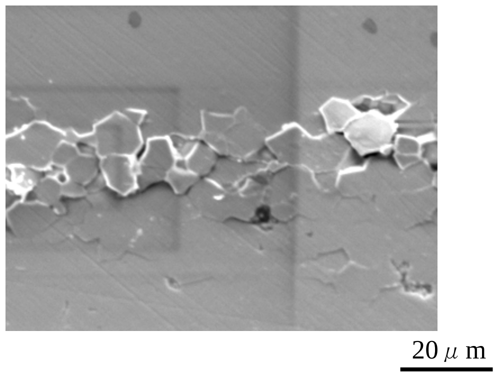

Fig. 3 SEM micrograph of cracked region in soft PZT under ΔE = ±400 V/mm.

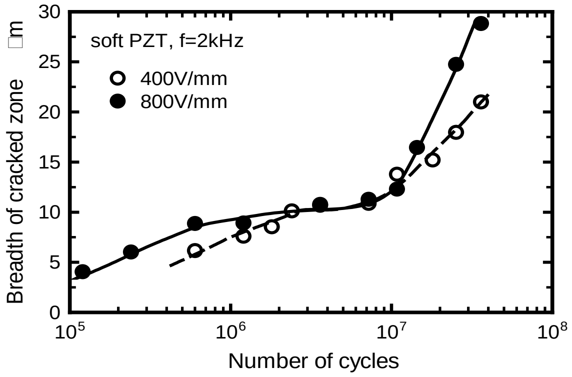

Fig.4 Relationship between the breadth of cracked zone and number of cycles.

The relationship between frequency and decreasing behavior of permittivity is shown in Fig.7. For

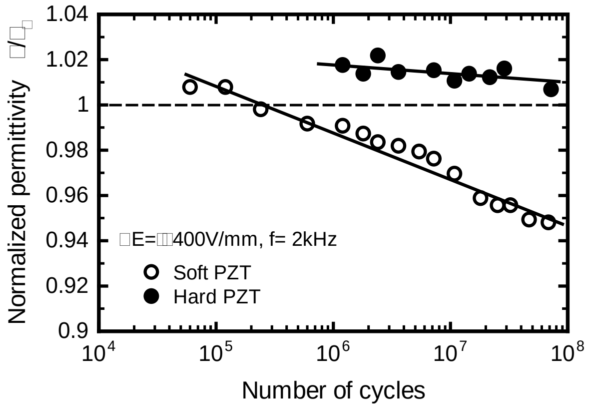

Fig.5 Relationship between normalized permittivity and number of cycles.

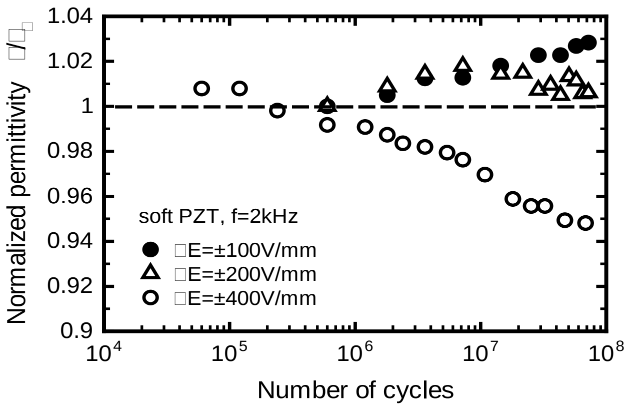

Fig.6 Effect of ΔE on permittivity change.

same ΔE, a lower frequency accel- erated the permittivity decrease. As described later, such damage behavior is concerned with domain switching caused by electric field concentrations, and is time- dependent.

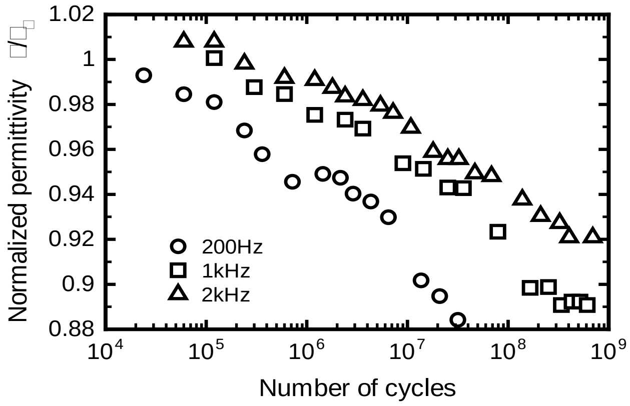

Fig.7 Effect of frequency on permittivity change.

FEM analysis on Domain Switching. Since piezoelectric ceramics are ferroelectrics, polar- ization switching is induced by either mechanical stress or an electric field. The local electroelastic field is concentrated at the edge of an electrode; therefore, switching caused by concentrated fields can affect the fatigue damage of the piezoelectric body under cyclic field [2]. In order to simulate the switching initiation at the electrode edge, finite element method (FEM) analyses were carried out on two-dimensional model of specimen. Fig. 8 shows the mesh pattern of the analysis using the FEM package, ANSYS®. The material constants used in the analysis were typical of those for a soft PZT [3]. Several researchers have proposed criteria for 180˚ domain switching [4]. Sun *et al.* have proposed a criterion (Eq. 1) that considers total strain and total electric displacement, rather than spontaneous strain and spontaneous polarization to calculate the work done.

$$\left(\sigma_{ij}\Delta\varepsilon_{ij}+E_i\Delta D_i\right)\geq 2P_S E_S\,. \tag{1}$$

where P_S is the spontaneous polarization and E_S is the coercive field, whose values in this analysis were 0.94 C/m^2 and 0.7 MV/m, respectively.

Fig. 9 shows the distribution of energy density (Eq. 2) near the electrode edge calculated in the FEM. Linear piezoelectricity is assumed.

$$\frac{1}{2}\left(\sigma_{ij}\varepsilon_{ij}+E_i D_i\right)\,. \tag{2}$$

Thus, the electroelastic field is concentrated and the maximum energy density appears at the edge. Considering the cyclic field, the energy density is twice the value of Eq. 2. Thus, the switching criterion (Eq. 1) is satisfied when the electric field is greater than 400 V/mm. Therefore, when the cyclic field exceeds ±400 V/m, the switching caused by electric field concentrations can damage the material. This is in agreement with the experimental results.

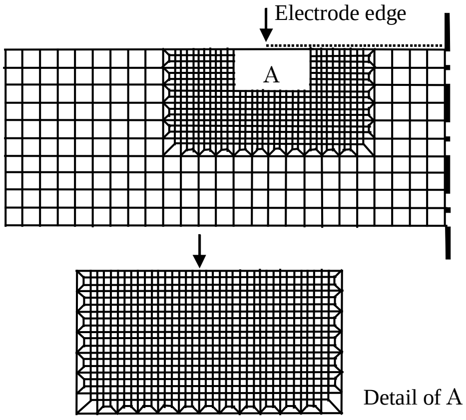

Fig. 8 Mesh pattern of FEM analysis.

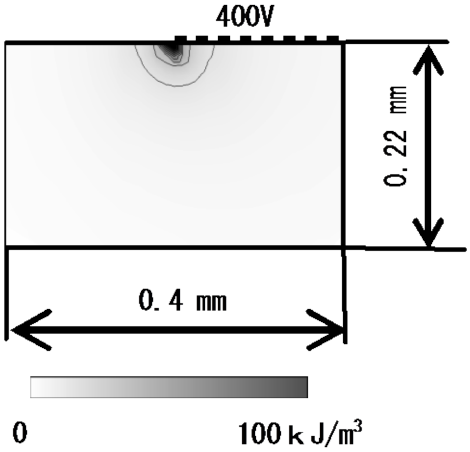

Fig.9 Distribution of energy density near the edge of electrode.

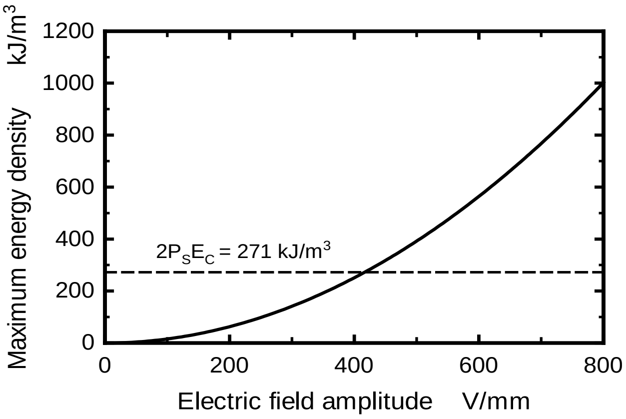

Fig.10 Relationship between maximum energy density and applied electric field.

Summary

(1) Intergranular cracking occurred due to application of repeated electric fields greater than ±400 V/mm in soft PZT. No cracking was observed in hard PZT even under high electric fields.
(2) The permittivity of specimens decreased with the number of loading cycles. Once cracking started, the rate of decrease increased remarkably.
(3) FEM analyses show that domain switching, due to concentrated electric-elastic field at electrode edge, affected the fatigue damage.

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