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【Technical Note】

Measurement of Flexoelectric Effect in Lead Zirconate Titanate Ceramics*

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Abstract

Lead zirconate titanate (PZT) ceramics is usually used as a piezoelectric material. However it has characteristics of not only piezoelectricity but also flexoelectricity. Piezoelectricity is a phenomenon that electric polarization is induced by strain, and flexoelectricity is the one that the polarization is induced by strain gradient. In this study, flexoelectricity in poled soft PZT ceramics is measured. In order to eliminate influence of piezoelectricity, PZT ceramic thin plates are subjected pure bending using four-point bending experimental mechanism. Strain gradient along the direction of thickness in the plate is caused by the bending motion. Electric charge between two electrodes which is set on the center of the plate surface is measured under quasi-static sinusoidal load. Even though the polarization for piezoelectric effect is eliminated by the experimental setup, the influence of piezoelectric effect still remains since polarization by piezoelectricity is much larger than the one by flexoelectricity. So the influence of piezoelectricity is eliminated using qualitative difference between piezoelectricity and flexoelectricity for poling direction. Eventually, flexoelectric coefficient of the order of 10^{-5} C/m is measured.

Key words: Flexoelectricity, Piezoelectric Ceramics, Lead Zirconate Titanate

1. Introduction

Lead zirconate titanate (PZT) ceramics is used in various industrial fields as piezoelectric materials. Sensor and electric power generation are the most familiar applications of piezoelectric materials. It is important to raise efficiency of electric polarization for external mechanical energy from performance and ecology points of view. Piezoelectric effect is polarization phenomenon proportional to strain caused in the piezoelectric material. On the other hand, flexoelectric effect is polarization phenomenon proportional to strain gradient in the material. The flexoelectric effect is paid attention usually for liquid crystal but rarely for solid materials. However it can be utilized in order to raise polarization efficiency to external mechanical energy in conjunction with piezoelectric effect. Majdoub et al. showed analytical results that nanostructures of piezoelectric material such as PZT or barium titanate (BaTiO3) can generate large polarization due to not only piezoelectric effect but also flexoelectric effect[(1),(2)].

Flexoelectric effect is a phenomenon whose concept have introduced by Meyer in 1969 as a piezoelectric effect in liquid crystals[(3)]. The terminology, flexoelectricity, was utilized by Gennes[(4)] from several years later the Meyer's introduction. As liquid crystals can be deformed large by nature, and as some sorts of the liquid crystal have permanent dipole moment and particular molecule geometry, they cause electric polarization remarkably for deformation with strain gradient by external force and shape effect. On the other hand, it is not so usual that solid piezoelectric materials are deformed with large strain gradient. And

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also piezoelectric polarization is much larger than flexoelectric polarization. They have not paid so much attention to flexoelectricity in solid materials. However when piezoelectric materials are industrially utilized, it is very important to raise efficiency of electric polarization by means of integrating various possibilities. The flexoelectricity can be utilized for it.

There are some researches to investigate amount of electric polarization of piezoelectric materials. Ma and Cross measured flexoelectric polarization of several solid materials without poling[(5)-(7)]. We aim at raising polarization efficiency of piezoelectric materials in the future, and focus on flexoelectric effect as one of important possibility for our aim. In this paper, electric charge caused by flexoelectric effect in poled PZT ceramics is measured.

2. Experimental setup

Flexoelectric effect is caused by deformation with strain gradient. In order to generate flexoelectric polarization, strain in the piezoelectric material caused by external load must be changed for position. Also when a poled piezoelectric material is deformed by load, electric polarization is induced by not only flexoelectric effect but also piezoelectric effect. Therefore in order to measure purely polarization for only flexoelectric effect caused by deformation of the measured piezoelectric element, amount of entire strain over electrodes on the element must be zero as a result of summation of tensile and compressive strain. In this study, pure bending of piezoelectric beam shape element with uniform cross section is utilized.

Figure 1 shows top view of experimental setup to measure electric polarization for flexoelectric effect. Uniform strain gradient in the direction of thickness generates by four point bending. As a thin PZT plate specimen is bent through four precision type cylindrical pins set symmetrically, constant bending moment occurs without shear force along the specimen between the inside two pins. Also bending stress along the cross section of the specimen causes symmetrically with respect to the neutral surface in the specimen with constant strain gradient. The total amount of summation of bending stress along longitudinal direction in the cross section becomes zero.

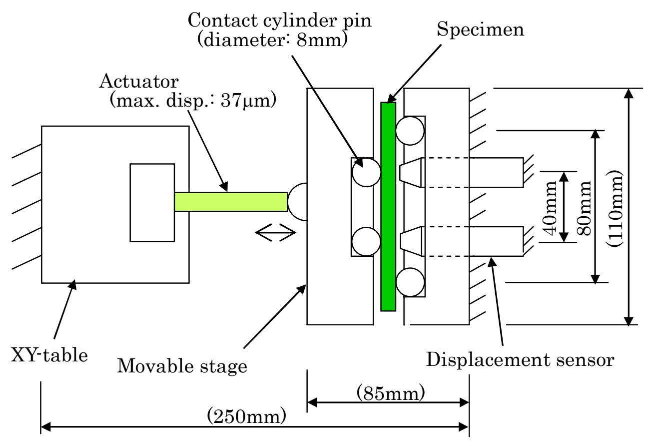

Fig. 1 Top view of the experimental setup.

Distances between the pairs of pins in the left and right hand sides of Fig.1 are 40 and 80mm respectively. The pins drawn at the left hand side are set on a translation stage guided with parallel springs, and the stage is pushed by a stacked piezoelectric actuator whose maximum displacement is 37μm. The actuator gives the stage quasi-static sinusoidal motion of 0.5Hz. The actuator is set on a precision XY-table movable with micrometer heads, and the table adjusts initial indentation of the specimen. Two capacitance type non-contact displacement sensor probes measure specimen's surfaces from the opposite side to the pushing pins.

Figure 2 is schematic view of the PZT piezoelectric specimen. Dimensions of specimen and electrodes are 90x10x1mm and 30x9.4mm respectively. Silver electrodes of the same area are printed on both sides. The figure shows distances between the pair of load line by the cylindrical pins. Since thickness of the specimen and area of the electrode influence on accuracy of measurement, surface of the PZT specimen is finished by means of lapping and the electrode is printed with 0.2mm dimensional tolerance. Poling direction of the specimen is thickness direction, i.e. parallel direction to the load. In order to avoid the influence of piezoelectric effect by the compressive load, distances between the load line and electrode ends are designed to 5mm for each side. Specifications of the PZT specimen are in Table 1. Three same specimens are prepared for experiments. All experiments are executed at temperature of 20 degrees centigrade and humidity of less than 50%.

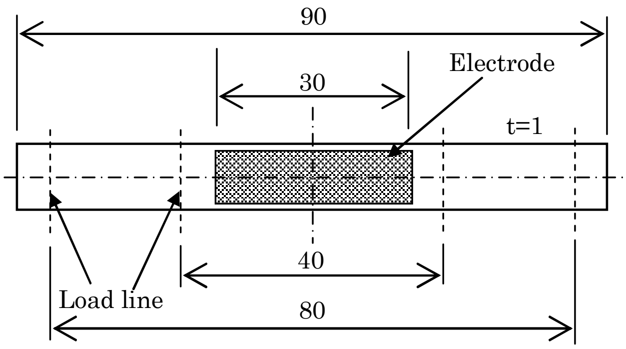

Fig. 2 Schematic view of PZT specimen.

Table 1. Characteristics of PZT specimen

Material	$Pb(Zr\text{-}Ti)O_3$
Dimensions of specimen mm	$90\times10\times1$
Dimensions of electrodes mm	30×9.4
Electromechanical coupling factor k_t %	52
Relative permittivity e_{33}/e_0	2130
Piezoelectric constant d_{31} m/V	-210×10^{-12}
Piezoelectric constant d_{33} m/V	472×10^{-12}
Piezoelectric constant d_{15} m/V	758×10^{-12}
Quality factor	80
Capacitance nF	6.245

3. Results and discussion

Figure 3 shows waves of electric charge and displacement as the specimen is bent by sinusoidal motion of 15μm amplitude and 0.5Hz frequency. The displacement is average value between the two capacitance type displacement sensor probes. Initial indent of load point is 20μm. The amount of the electric charge generated on the electrodes is measured by a charge amplifier with sensitivity of 1mV/pC. It is not an essential problem that the electric charge delays to displacement with 90 degrees phase difference. The reason of the phase difference is just characteristics of electric charge measurement with the charge detection amplifier adopted in the experiments. It is confirmed by another experiment using an electric potential detection amplifier of high input impedance that electric polarization is generated at the same phase with the 0.5Hz sinusoidally driven displacement without delay.

Electric polarization generated by flexoelectric effect is denoted as eq. (1)[6].

$$P_i = d_{ijk}\sigma_{jk} + \mu_{ijkl}\frac{\partial \varepsilon_{jk}}{\partial x_l} \tag{1}$$

where P is polarization, d is piezoelectric coefficient, σ is stress, ε is strain, x is position, and μ is flexoelectric coefficient which is a fourth-rank polar tensor. The suffixes (i, j, k, l =1, 2, 3) follow Einstein summation convention. As influence of piezoelectric effect by compressive load is eliminated by the experimental setup, and also as total amount of stress in longitudinal direction along cross section equals to zero due to the four point bending, the first term for piezoelectric effect in eq.(1) becomes zero, and then the polarization can be denoted as eq.(2).

$$P_i = \mu_{ijkl}\frac{\partial \varepsilon_{jk}}{\partial x_l} \tag{2}$$

From the experimental results shown in Fig.3, electric polarization P can be calculated by eq.(3).

$$P = \frac{Q}{s} \tag{3}$$

where Q is electric charge and s is area of a electrode. Also strain gradient along cross section for four point bending can be calculated by eq. (4)

$$\frac{\partial \varepsilon}{\partial x} = \frac{12y}{L^2} \tag{4}$$

where y is the measured displacement, i.e. the deformation at the loading points, and L is distance between the outer side two pins. From the amplitudes of the sinusoidal wave shown in Fig.3, a point can be plotted in the graph shown in Fig.4 using eqns. (3) and (4). Figure 4 shows relations between strain gradient and polarization for different amplitude sinusoidal motions. The values of both strain gradient and polarization are plotted as peak-to-peak values of sinusoidal change. The results show linear relation between strain gradient and polarization. A line in Fig.4 is drawn using least square method. Slope of the line means the flexoelectric coefficient. For the results of Fig.3, the value of flexoelectric coefficient is calculated as 32.9μC/m using eqs.(3) and (4).

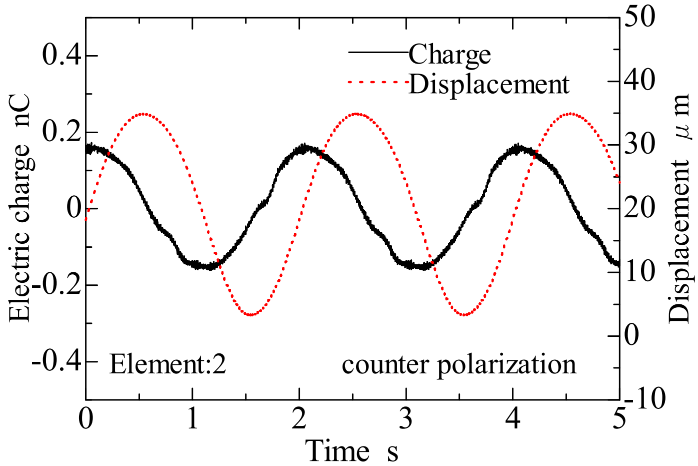

Fig. 3 An example of the experimental results for sinusoidal bending motion.

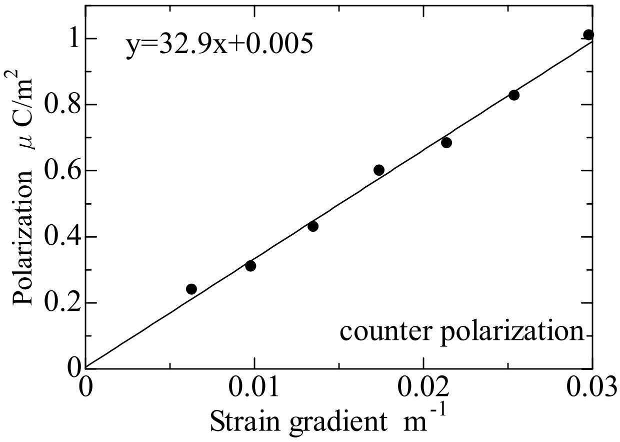

Fig. 4 An example of plots of the experimental results calculated from sinusoidal responses such as the one shown in Fig.3.

The results shown in the Figs.3 and 4 are the ones when the specimen is pushed to the opposite side from poling direction. The specimens used in this study are poled PZT thin plate. Piezoelectric polarization is much larger than flexoelectric one for macroscopic size elements. Though the experimental set up is designed to eliminate influence of piezoelectric effect, the influence of piezoelectricity still appears on the experimental results due to microscopic non-uniformity of the ceramic material. However sign of polarization by piezoelectric effect changes to opposite when bending direction changes. On the other hand, sign of polarization by flexoelectricity doesn't change by the difference of load direction(8). Therefore even though influence of the piezoelectricity remains in the results, it can be cancelled by averaging the results between different bending directions taking wave phase and sign of electric charge into account. The three specimens show a similar value of the average flexoelectric coefficient. The average value of flexoelectric coefficient μ_{12} among all three specimens is about 49μC/m. According to the references, the order of some examples of flexoelectric coefficient for unpoled PZT materials is in 10^{-1} or 100μC/m(6),(7). It is found that polarization by flexoelectricity in poled PZT is relatively larger than unpoled one.

4. Conclusions

In summary, flexoelectric effect in poled lead zirconate titanate (PZT) ceramics was investigated. Influence of piezoelectric effect is cancelled by pure bending of thin plate specimen and by taking average between the results of different bending direction. The average values are almost same for all three prepared specimens. The order of obtained flexoelectric coefficient value was 10^{-5}C/m for poled PZT. We are going to investigate detail influences of the poling treatment of the specimens on flexoelectricity in the next work.

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