Effect of Uniaxial Stress on Hysteresis Properties of 0.1PMN–0.9PZT Ceramic

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ABSTRACT

PMN-PZT ceramic composite with formula $0.1Pb(Mg_{1/3}Nb_{2/3})O_3$–$0.9Pb(Zr_{0.52}Ti_{0.48})O_3$ or 0.1PMN–0.9PZT was prepared by a conventional mixed-oxide method. A planarelectromechanical coupling coefficient (kp) value of the ceramic was measured by a resonance method. The kp value of the ceramic was calculated to be 0.47. In addition, the uniaxial stress dependence of hysteresis properties of 0.1PMN–0.9PZT ceramic was investigated by measuring the ferroelectric parameters, i.e., remanent polarization (P_r), spontaneous polarization (P_s) and coercive field (E_c) as a function of applied stress. It has been shown that sizes of hysteresis loop of the ceramic change with increasing stress. The P_r, P_s and E_c values also vary with the applied stress. The P_r and P_s decrease significantly, while Ec increases, with increasing stress.

Key words: PMN-PZT, Planar electromechanical coupling coefficient (k_p), Hysteresis properties, Uniaxial stress

INTRODUCTION

Lead magnesium niobate ($Pb(Mg_{1/3}Nb_{2/3})O_3$ or PMN) and lead zirconate titanate ($Pb(Zr_xTi_{1-x})O_3$ or PZT) ceramics have been widely used in actuator and transducer applications (Cross, 1996; Veihland et al., 2001). PMN is a well-established relaxor-type ferroelectric of perovskite structure with excellent dielectric properties. It has advantages of having broader operating temperature range, low loss and non-hysteretic characteristics. PZT ceramics have relatively-high electromechanical coupling coefficients as compared to PMN (Zhao et al., 1999). However, PZT ceramics are fairly lossy as a result of their highly-hysteretic behavior. With these complementary features, ceramics in PMN-PZT system are expected to have a combination of excellent properties of both ceramics.

The electromechanical coupling coefficient (k) is an indicator of the effectiveness with which a piezoelectric material converts electrical energy into mechanical energy or vice versa (Haertling, 1999). A high k is usually desirable for efficient energy conversion. For a thin disc of piezoelectric ceramic, the planar electromechanical coupling coefficient (k_p) expresses a radial coupling, i.e., the coupling between an electric field parallel to the direction in which the ceramic element is polarized (direction 3) and mechanical effect that produces radial vibrations, relative to the direction of the polarization (direction 1 and direction 2).

In many actuator and transducer applications, the ceramics are subjected to high mechanical-stress field. A prior knowledge of how the material properties change under different load conditions is crucial for proper design of a device and for suitable selection of materials for a specific application (Zhang et al., 1997). Therefore, this study was carried out to measure the planar electromechanical coupling coefficient (k_p) of a binary system of the two ceramics (0.1PMN–0.9PZT) and to investigate the influences of the uniaxial stress on the hysteresis properties of the ceramic. The 0.1PMN–0.9PZT was then chosen, based on the fact that this composition is near the morphotropic phase boundary (MPB) of the binary system (Koval et al., 2003). Therefore, it was expected to possess better properties than other compositions.

MATERIALS AND METHODS

The $0.1Pb(Mg_{1/3}Nb_{2/3})O_3$–$0.9Pb(Zr_{0.52}Ti_{0.48})O_3$ ceramic composite was prepared from the starting PMN and PZT powders, synthesized by the columbite (Swartz and Shrout, 1982) and a conventional mixed-oxide methods, respectively. Initially, the PMN and PZT powders for a given composition were weighed and then ball-milled in ethanol for 24 hours. After drying process, the mixed powders were pressed hydraulically to form disc-shape pellets 10 mm in diameter and 1 mm thick, with 5 wt.% polyvinyl alcohol (PVA) as a binder. The pellets were stacked in a covered alumina crucible, filled with PZ powders to prevent lead loss. Finally, the sintering was carried out at a sintering temperature of 1275°C for 4 hours with 2 min/°C heating and cooling rates. The firing profile included a 1-hour dwell time at 500°C for binder burn-out process to complete.

The phase formations of the sintered specimens were studied by an x-ray diffractometer (SIEMENS, D500) with CuK_α radiation (λ = 0.15405 nm) at room temperature. For the electrical properties measurements, the pellets were lapped to obtain parallel faces and the faces were then coated with silver paint as electrodes. The samples were heat-treated at 750°C for 12 min, then poled in a silicone oil bath at the temperature of 110°C with applied DC field of 40 kV/cm for 30 min and field-cooled to room temperature.

The electromechanical coefficient was measured by a standard resonance-antiresonance method. In this case, a thin disc-shape sample with diameter approximately 10 times of thickness was used to determine the planar electromechanical coupling coefficient (k_p). This system is uniquely equipped to measure the AC drive amplitude dependence of the resonance-antiresonance curves, allowing for the determination of nonlinearity in the material. Plot of reactance versus frequency for a piezoelectric ceramic was obtained. The resonant frequency, f_r, at the point of minimum impedance and the anti-resonant frequency, f_a, at the point of maximum impedance were then extracted from the plot. Both of those frequencies were defined at zero reactance (IEEE Standard Definitions, 1986).

The measurement set-up, shown in Fig. 1, consisted of the test cell attached to a HP-4194A Impedance/Gain-Phase Analyzer using coaxial wires and a HP-16047C test fixture. A personal computer was used for data acquisition. In this case, test frequency ranged between 50 kHz and 5 MHz.

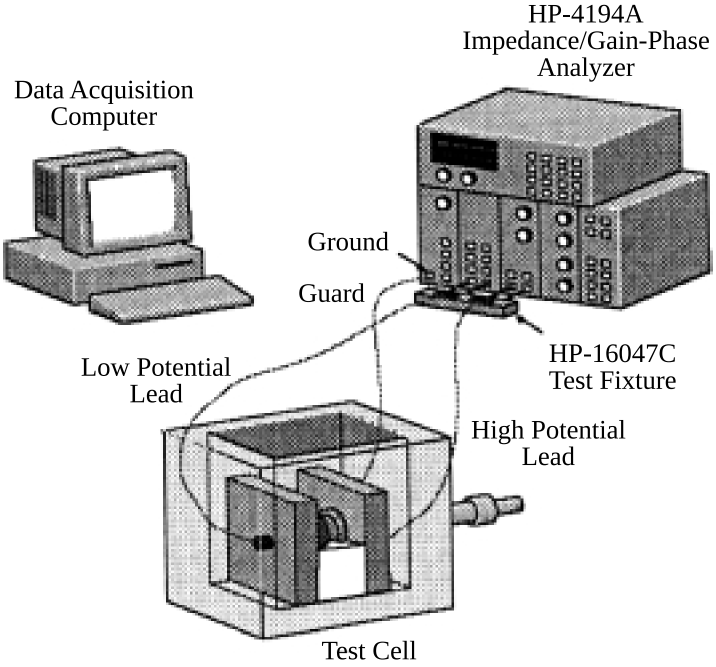

Figure 1. A schematic diagram of the experimental set-up consisting of a HP-4194A Impedance/Gain-Phase analyzer, a test cell, and a data acquisition computer.

The ferroelectric hysteresis (P-E) loops were measured by using a Sawyer-Tower circuit as shown in Fig. 2. A sinusoidal field of 2500 V_{rms} and 50 Hz was applied to the sample with fixed R_1 R_2 and C_0. The voltage across the ceramic (C_s) was recorded on the horizontal axis (X-axis) of the oscilloscope while the voltage across the capacitor (C_0), which was proportional to the polarization on the sample C_s, was recorded on the vertical axis (Y-axis) of the oscilloscope. The measurement was carried out at room temperature.

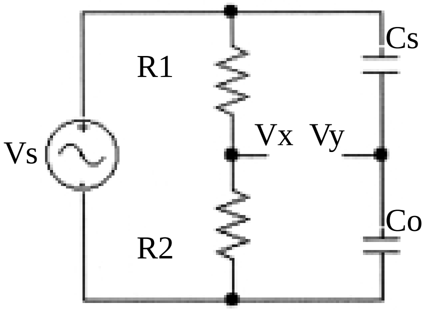

Figure 2. Sawyer-Tower circuit used for measuring hysteresis loop (Wongsaenmai et al., 2003) (R_1 = 6.8 MΩ, R2 = 10kΩ, C_0 = 0.1 μF, C_s = Sample, $C_0 >> C_s$).

To study the uniaxial stress effect on the hysteresis properties of the ceramic, a specially-designed press was used. In this apparatus, force generated by a hydraulic press was transmitted to the sample through a carefully-aligned cylinder steel. The stress on the sample was applied until the maximum stress had been reached and then reduced slowly to zero.

RESULTS AND DISCUSSION

The x-ray patterns, shown in Fig.3, show that the sintered ceramics are mainly in perovskite phase. From the XRD pattern, PZT ceramic is identified as a single-phase material with a perovskite structure, having tetragonal symmetry while PMN ceramic is a perovskite material with a cubic symmetry. The 0.1PMN–0.9PZT can be identified as a single-phase material with a perovskite structure, having tetragonal symmetry similar to PZT.

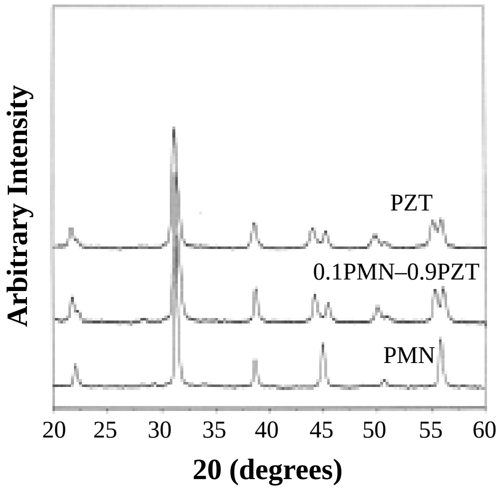

Figure 3. XRD patterns of the sintered PZT, PMN and 0.1PMN-0.9PZT ceramics.

For the planar electromechanical coupling coefficient measurement, a reactance-frequency curve of 0.1PMN-0.9PZT ceramic is shown in Fig.4. From this curve, the resonant frequency (f_r) and the anti-resonant frequency (f_a) are determined. Using Equation (1), one can calculate the planar electromechanical coupling coefficient (Murata Manufacturing),

$$k_p = \sqrt{2.51\left(\frac{f_a - f_r}{f_r}\right)} \quad \text{................} (1)$$

where k_p is the planar electromechanical coupling coefficient
f_r is the resonant frequency (Hz)
f_a is the anti-resonant frequency (Hz)

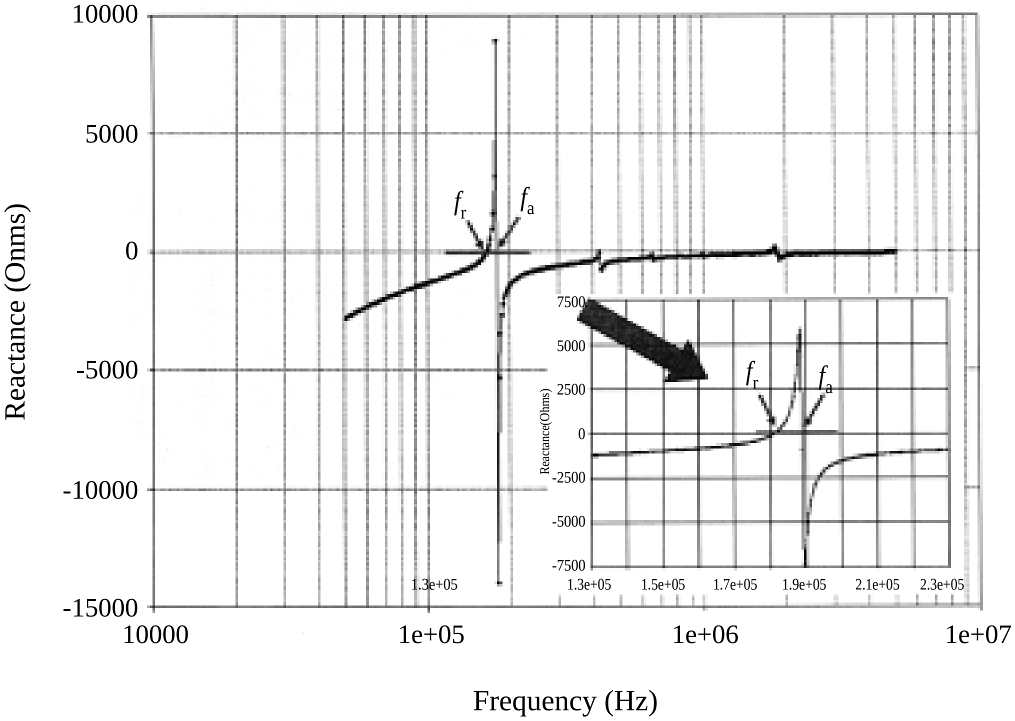

Figure 4. Reactance response of 0.1PMN-0.9PZT ceramics.

From the resonant frequency (f_r) and anti-resonant frequency (f_a), determined from reactance-frequency curve, the planar electromechanical coupling coefficient (k_p) is calculated to be 0.47.

A sequence of polarization-electric field (P-E) hysteresis loops for the 0.1PMN-0.9PZT ceramic at various levels of the uniaxial stress is illustrated in Fig.5 for increasing stress and in Fig.6 for decreasing stress. It is evident that the shapes of P-E loops change with the applied stress. The size of loop is gradually decreased with increasing stress (Fig.5) and the size of loop is, on the other hand, increased with reducing stress (Fig.6).

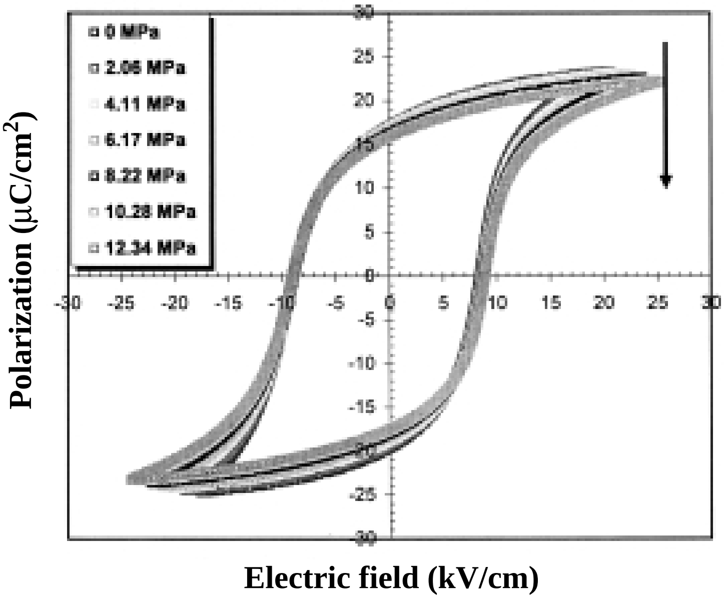

Figure 5. Hysteresis loop of 0.1PMN-0.9PZT ceramic under increasing stress.

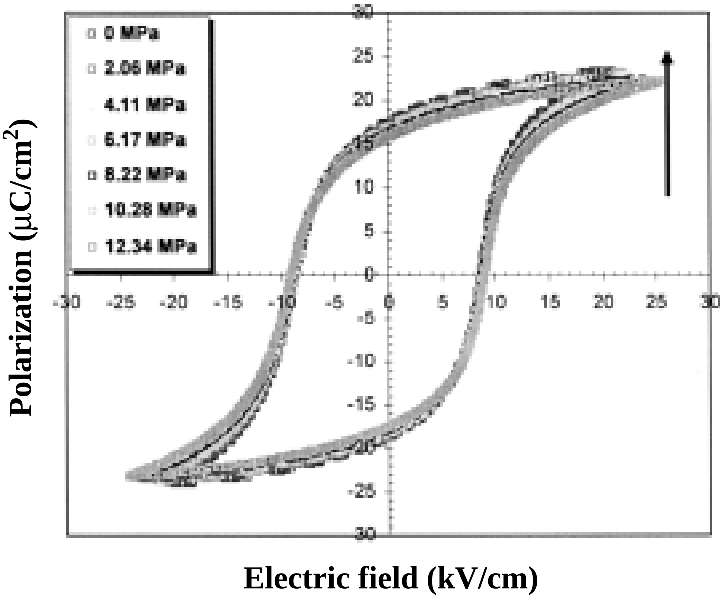

Figure 6. Hysteresis loop of 0.1PMN-0.9PZT ceramic under decreasing stress.

From the hysteresis loops, the remanent polarization (P_r), the spontaneous polarization (P_s) and the coercive field (E_c) are determined. The variations of these parameters with the applied stress are shown in Fig.7. Clearly, these values vary with the applied stress. Pr and Ps decrease slightly with increasing stress while E_c increases.

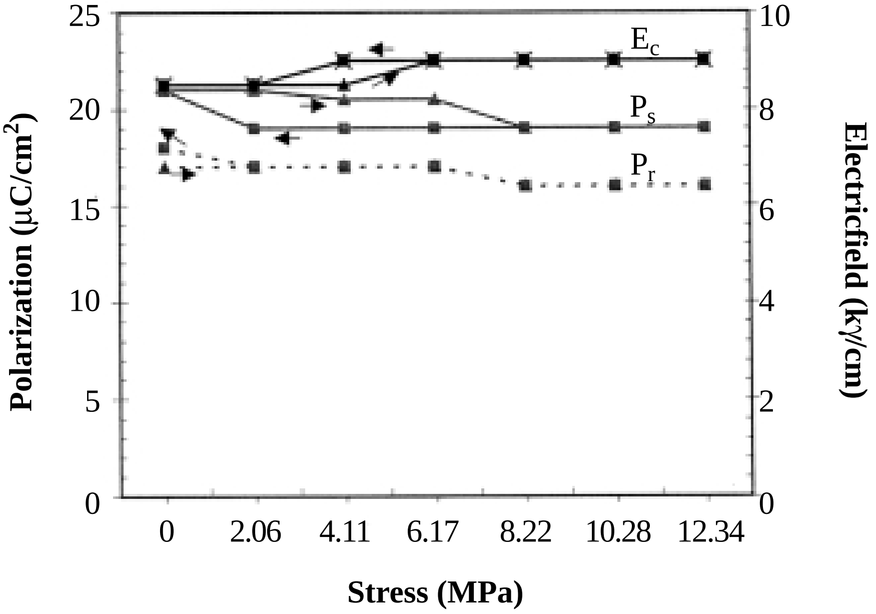

Figure 7. The Pr, Ps and Ec values that change with uniaxial stress.

These results show the effects of the domain wall motion mechanism in this ceramic (Zhang et al., 1997; Veihland et al., 2001). The applied stress imposes a frustration of the domain wall motion under applied field. The frustration of domain wall motion leads to reduction of the size of hysteresis loop. Therefore, this indicates that the stress can reduce the hysteresis loss (proportional to the area of the loop). Figure 7 clearly reveals the characteristic of hysteresis parameters P_r, P_s and E_c under the uniaxial stress. After a stress cycle, the final value of each parameter returns to its original one. This indicates that the degradation and depoling mechanisms in 0.1PMN–0.9PZT ceramic under the uniaxial stress are rather insignificant. However, it is expected that these mechanisms will become more apparent at higher stress levels.

CONCLUSION

The $0.1Pb(Mg_{1/3}Nb_{2/3})O_3$–$0.9Pb(Zr_{0.52}Ti_{0.48})O_3$ ceramic composite was successfully prepared by a conventional mixed-oxide method. The value of planar electromechanical coupling coefficient (k_p) was 0.47. With the Sawyer-Tower circuit and the uniaxial stressing system, the effects of uniaxial stress on the hysteresis properties of the ceramic were investigated. It was found that the sizes of hysteresis loop of the ceramic decreased with increasing stress. The P_r, P_s, and E_c values varied with the applied stress. P_r and P_s decreased slightly while E_c increased with increasing stress. These results indicate the significance of the domain wall motions under applied stress.

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