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Stress-dependent ferroelectric properties of $Pb(Zr_{1/2}Ti_{1/2})O_3$–$Pb(Zn_{1/3}Nb_{2/3})O_3$ ceramic systems

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Abstract

Effects of compressive stress on the ferroelectric properties of ceramics in PZT–PZN systems were investigated. $(1 - x)Pb(Zr_{1/2}Ti_{1/2})O_3(xPb(Zn_{1/3}Nb_{2/3})O_3$ or $(1 - x)$PZT–(x)PZN ($x = 0.1$–0.5) ceramics were prepared by a conventional mixed-oxide method. The ferroelectric properties under compressive stress of the PZT–PZN ceramics were observed at stress levels up to 170 MPa using a compressometer in conjunction with a modified Sawyer–Tower circuit. It was found that with increasing compressive stress the area of the ferroelectric hysteresis (P–E) loops, the saturation polarization (P_{sat}), the remanent polarization (P_r), and the coercive field (E_c) decreased. These results were interpreted through the non-180° ferroelectric domain switching processes.

Keywords: C. Ferroelectric properties; D. PZT; Stress

1. Introduction

In many practical sensor and actuator applications, piezoelectric and ferroelectric ceramics are often subjected to mechanical loading, either deliberately in the design of the device itself or because the device is used to change shapes as in many smart structure applications or the device is used under environmental stresses such as in underwater transducers [1–3]. A prior knowledge of how the material properties change under different load conditions is therefore very crucial for proper design of a device and for suitable selection of materials for a specific application. Despite the fact, material constants used in many design calculations are often obtained from a stress-free measuring condition, which in turn may lead to incorrect or inappropriate actuator and transducer designs [4,5]. It is therefore important to determine the properties of these materials as a function of applied stress. Previous investigations on the stress-dependence dielectric and electrical properties of many ceramic systems, such as BT, PZT, PMN, PMN–PT, PZT–BT, and PMN–PZT have clearly emphasized the importance of the subject [6–13]. In this paper, the stress dependence of the ferroelectric properties of ceramics in PZT–PZN system is presented.

Recently, there have been a great deal of interest in lead zirconate titanate-lead zinc niobate $(Pb(Zr_{1/2}Ti_{1/2})O_3(Pb(Zn_{1/3}Nb_{2/3})O_3$ or PZT–PZN) systems because of their high dielectric, piezoelectric, and ferroelectric properties [14–16]. PZN is a ferroelectric material with frequency-dependent dielectric relaxation behavior, as well as diffuse phase transition at 140 °C [17]. A single crystal of PZN shows extremely high dielectric and piezoelectric constants [18]. PZT also has a high piezoelectric properties, particularly compositions (Zr:Ti ~ 52:48) close to a morphotropic phase boundary (MPB) between the tetragonal and the rhombohedral phases [19]. Therefore, PZT is the most popular ferroelectric material used in electronic devices [4,19,20]. As both PZT and PZN possess excellent electrical properties, enhanced properties can thus be expected from ceramics in PZT–PZN system. There have been many previous reports on the electrical properties of PZT–PZN ceramics [14–16]. Earlier report on the stress-dependent dielectric properties of PZT–PZN also showed very interesting results [21]. However, there has been no systematic study on the influence of an applied stress on the ferroelectric properties of the PZT–PZN ceramics. Therefore, it is the aim of

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this study to determine the ferroelectric properties of the $(1-x)$PZT–(x)PZN ceramics as a function of compressive stress.

2. Experimental procedure

$(1-x)Pb(Zr_{1/2}Ti_{1/2})O_3$–$(x)Pb(Zn_{1/3}Nb_{2/3})O_3$ (with $x = 0.1$–0.5) ceramics were prepared from starting PZT and PZN powders by a conventional mixed-oxide method via vibro-milling technique [22]. Perovskite-phase PZN powders were obtained from the columbite method with zinc niobate ($ZnNb_2O_6$) selected as the precursor, while PZT powders were prepared by a simple mixed-oxide method [23]. For optimization purpose, sintering was carried out at temperatures between 1200 and 1250 °C for 2 h. Details of the processing and characterizations were provided elsewhere [22–24].

Before studying the dielectric properties, the specimens were lapped to obtain parallel faces. After coating with silver paint as electrode at the faces, the specimens were heated at 750 °C for 12 min to ensure the contact between the electrode and surface of ceramic. The ferroelectric hysteresis (P–E) loops were characterized by using a computer controlled modified Sawyer–Tower circuit. The electric field was applied to a sample by a high voltage AC amplifier (Trek, model 610D) with the input sinusoidal signal with a frequency of 100 Hz from a signal generator (Goodwill, model GAG-809). The detailed descriptions of this system were explained elsewhere [9,10]. To study the effects of the compressive stress on the ferroelectric properties, the uniaxial compressometer was constructed for simultaneous and parallel applications of the mechanical stress and the electric field [12]. Measurements were performed as a function of mechanical stress applied discretely between 0 and 170 MPa. During the measurements, a desired stress was first applied to the sample and then the electric field was applied. The ferroelectric hysteresis (P–E) loop was recorded at room temperature (25 °C) for both loading and unloading conditions. The parameters obtained from the loops were the saturation polarization (P_{sat}), the remanent polarization (P_r), and the coercive field (E_c), which were defined as the points where the loops reach the maximum polarization, cross the zero field, and cross the zero polarization, respectively. The measurements reported were for the samples during their first mechanical stress cycle. It should also be noted that the reported ferroelectric parameters were obtained after a total of 10 cycles of the electric field were applied to the sample at each constant stress level.

3. Results and discussion

The polarization vs. electric field (P–E) hysteresis loops of the 0.9PZT–0.1PZN, 0.7PZT–0.3PZN, and 0.5PZT–0.5PZN ceramics under different compressive stresses are shown in Figs. 1–3, respectively. It should first be noticed that the area of the P–E loops decreases with increasing compressive stress. The changes follow similar trend for all the ceramics. The P–E loop area indicates the polarization dissipation energy of a ferroelectric material under one full cycle of electric field application [23]. This amount of the energy loss is directly related to the amount of domain participating in the switching process during the application of electric field [10]. Unlike in the PMN–PZT system, in which the stress-free loop area decreased with increasing PMN content, as the ceramics gradually change from ferroelectric PZT with macro-domain states to electrostrictive PMN with nano-domain states [13,17], the stress-free loop areas of all the ceramics are generally very similar. For a given composition, the decrease in the loop area with increasing compressive stress is a result of the stress-induced domain wall suppression [10].

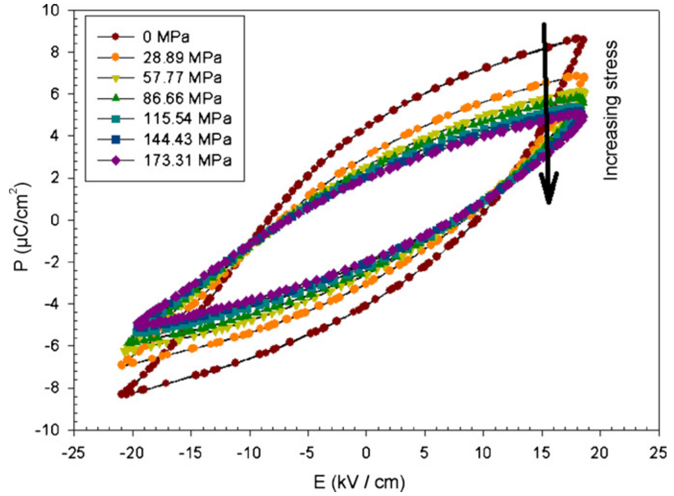

Fig. 1. Polarization vs. electric field (P–E) hysteresis loops as a function of compressive stress for 0.9PZT–0.1PZN ceramic (loading cycle).

The changes in the saturation polarization (P_{sat}), the remanent polarization (P_r), and the coercive field (E_c) with the compressive stresses are plotted in Figs. 4–6, respectively. It should be noted here that the P_{sat} and P_r values are calculated from average of the positive (P_+) and negative (P_-) values, e.g. $P = \{[(P_+) - (P_-)]/2\}$. It should first be noticed that though the values are not significantly different, the stress-free values of the P_{sat} and P_r are generally the largest (with E_c nearly minimal)

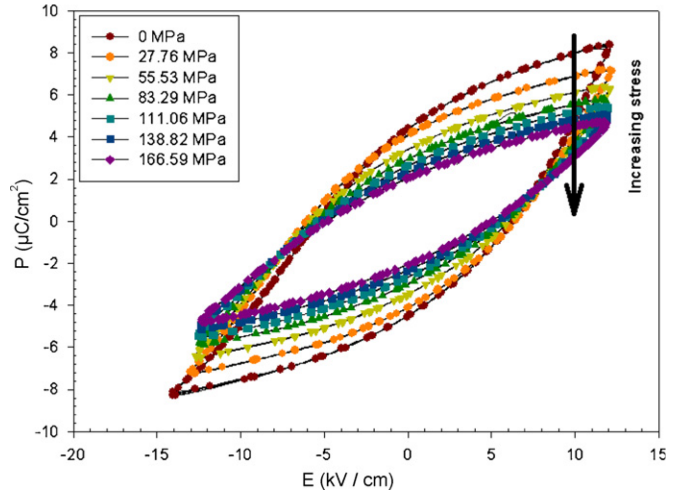

Fig. 2. Polarization vs. electric field (P–E) hysteresis loops as a function of compressive stress for 0.7PZT–0.3PZN ceramic (loading cycle).

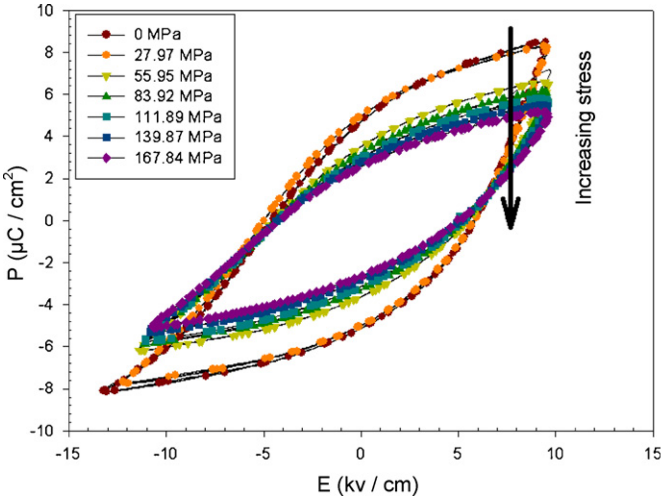

Fig. 3. Polarization vs. electric field (P–E) hysteresis loops as a function of compressive stress for 0.5PZT–0.5PZN ceramic (loading cycle).

for compositions near MPB ($x = 0.3$–0.4). In addition, the P_r value for tetragonal compositions, i.e. 0.9PZT–0.1PZN and 0.8PZT–0.2PZN, is lower than that of the rhombohedral compositions (0.6PZT–0.4PZN and 0.5PZT–0.5PZN). The E_c, however, shows the reverse trend. These observations are anticipated because the ceramic systems gradually change from the compositions with tetragonal structure ($x = 0.1$ and 0.2) with high E_c and low P_r through the near MPB composition with highest polarization values to the compositions with rhombohedral structure ($x = 0.4$ and 0.5) with low E_c and high P_r, as mentioned in previous investigation [14–17]. This clearly clarifies the existence of the MPB near $x = 0.3$ and 0.4, as reported earlier [21]. Figs. 4 and 5 clearly show that both the saturation and remanent polarizations decrease as the compressive stress increases. The decrease is generally very similar with the polarization values at 170 MPa and approximately 50–75% of the stress-free values. In addition, noticeable decreases in the polarization values of all the ceramic are also observed after a complete cycle of the mechanical stress. This observation suggests a significant stress-induced de-poling in PZT–PZN ceramics resulting in the observed decrease of the polarization values under high stress levels [11,25]. Interestingly, as opposed to the polarizations, the applied compressive stress shows little or no influence on the coercive field, as demonstrated in Fig. 6.

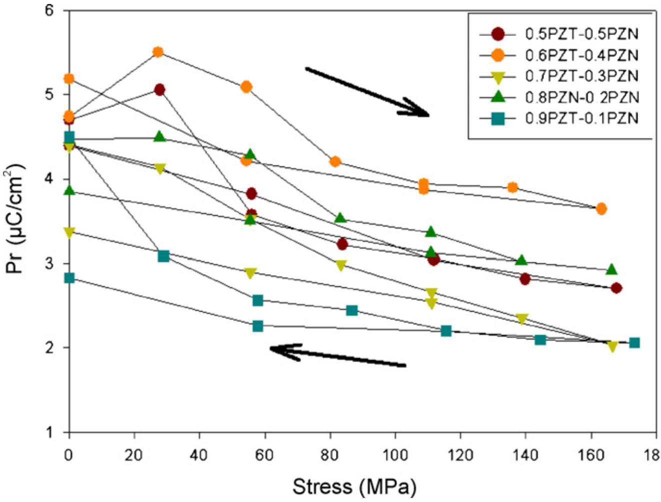

Fig. 5. Changes in remanent polarization (P_r) with compressive stress for $(1 - x)$PZT–xPZN ceramics (loading and unloading).

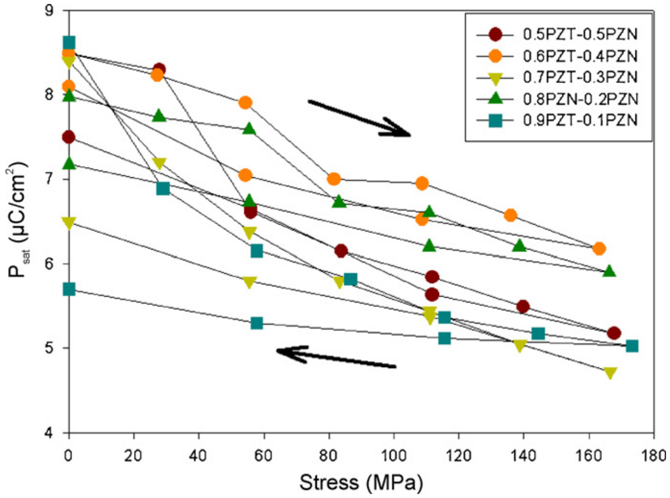

Fig. 4. Changes in saturation polarization (P_{sat}) with compressive stress for $(1 - x)$PZT–xPZN ceramics (loading and unloading).

To understand, at least qualitatively, these experimental results on ferroelectric PZT–PZN ceramic, one can interpret the changes in terms of domain-reorientation processes. When the compressive stress is applied in the direction parallel to the polar axis (poling) direction, the applied stress tends to keep the ferroelectric domains aligned with their polar axes away from the stress direction through the non-180° ferroelastic domain switching processes [6,7,11,25]. Therefore, it takes larger than usual applied electric field to reorient the domains along the stress direction, resulting in lower value of the saturation polarization (P_{sat}), as shown in Figs. 1–4. When the electric field is reduced to zero, the domains tend to rotate back away from the applied compressive stress direction, resulting in

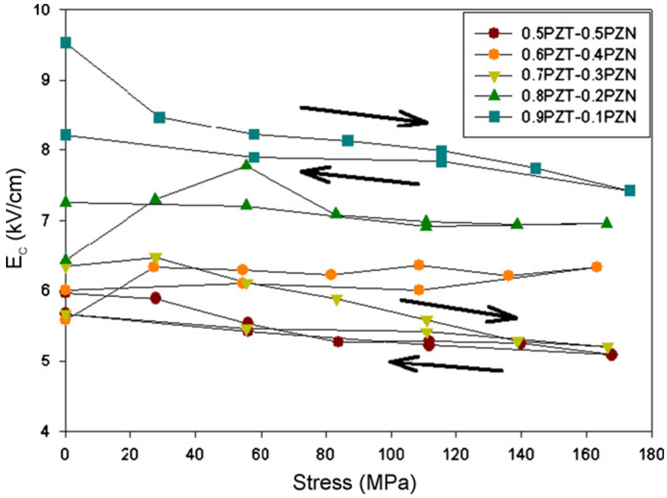

Fig. 6. Changes in coercive field (E_c) with compressive stress for $(1 - x)$PZT–xPZN ceramics (loading and unloading).

lower than usual remanent polarization (P_r), as depicted in Figs. 1–3 and 5 [7,25]. Furthermore, considering the decrease of the hysteresis loss with increasing compressive stress, it indicates that more and more ferroelectric domains are constrained by the applied stress and cannot be re-oriented by the electric field so as to participate in the polarization reversal. Consequently, both the saturation and remanent polarizations become lower with increasing compressive stress [12]. The results of the changes of the hysteresis parameters of the ferroelectric ceramics with increasing compressive stress are in agreement with the previous investigations on many ferroelectric ceramics [5,7,12,26,27].

Finally, it should also be mentioned that in this paper $Pb(Zr_{1/2}Ti_{1/2})O_3$ is selected as a base composition for PZT–PZN system, and ceramic systems gradually change from compositions with tetragonal to rhombohedral as increased the amount of modified PZN to PZT. In this case, the symmetric phase (tetragonal and rhombohedral phases) is changed by the amount of modified PZN, and the amount of PZN itself is also changed by PZN content in PZT–PZN system. From our observation in Figs. 4 and 5, the 0.5PZT–0.5PZN ceramic with rhombohedral phase showed the smallest changes in P_{sat} and P_r. However, the content of PZN in 0.5PZT–0.5PZN ceramic is too much to obtain high piezoelectric properties in PZT–PZN system, as compared with 0.2PZN–0.8PZT to 0.4PZN–0.6PZT. Therefore, it is possible that the best compositions for obtaining both large ferroelectric properties and small changes by the compressive stress in P_{sat} and P_r should be in the range of (0.2–0.4)PZN–(0.8–0.6)PZT with the rhombohedral phase. Therefore, it is likely that there are two competing mechanisms of PZN content and symmetric phase in controlling the changes of the properties under the influence of applied stress.

4. Conclusions

Effects of compressive stress on the ferroelectric properties of ceramics in PZT–PZN system were investigated. The ferroelectric properties under compressive stress are observed at stress levels up to 170 MPa using a compressometer in conjunction with a modified Sawyer–Tower circuit. The results show that the area of the ferroelectric hysteresis (P–E) loops, the saturation polarization (P_{sat}), the remanent polarization (P_r), and the coercive field (E_c) decrease with increasing compressive stresses. The non-180° ferroelectric domain switching processes are responsible for the changes observed. Furthermore, noticeable decreases in the polarization values of all the ceramic are also observed after a complete cycle of the mechanical stress, indicating a significant stress-induced depoling in PZT–PZN ceramics. Finally, this study shows that the applied compressive stress has significant influences on the hysteresis properties of the PZT–PZN ceramics.

Acknowledgments

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