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Comparative studies of dynamic hysteresis responses in hard and soft PZT ceramics

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Abstract

Lead zirconate titanate (PZT) is a ferroelectric material with very interesting and useful dynamic hysteresis properties. Normally, PZT is doped with donors or acceptors to yield better electrical properties. Soft and hard PZT ceramics are respectively donor- and acceptor-doped PZT, which are commercially available and widely employed in various applications. Previous investigations have mainly been focused on the dynamic hysteresis at room temperature and under stress-free condition. However, when used, these ceramics are normally subjected to stress. More importantly, the ambient temperature is usually not at room temperature. Therefore, this study was to investigate dynamic hysteresis behavior of both hard and soft PZT ceramics with varying compressive stress and temperature. The results clearly revealed the influence of external stress and temperature on the dynamic hysteresis of both types of PZT ceramics. Increasing stress and temperature resulted in a decrease of the hysteresis area of the two types of PZT ceramics.

Keywords: A. Dynamic hysteresis; B. Hard PZT; C. Soft PZT

1. Introduction

Lead zirconate titanate ($Pb(Zr_{1-x}Ti_x)O_3$ or PZT) ceramics are among the lead-based complex perovskites that have been investigated extensively, from both academic and commercial viewpoints. They are employed extensively in sensor and actuators applications, as well as smart systems [1–6]. The most widely studied and used PZT compositions are in the vicinity of the morphotropic phase boundary (MPB) between the tetragonal and rhombohedral ferroelectric phases [1,4,6–8]. However, to meet the requirements for specific applications, PZT ceramics are usually modified with dopants [1,4,9,10]. Generally, donor (higher valency) additives induce 'soft' piezoelectric behaviors with higher dielectric and piezoelectric activities suitable for sensor and actuator applications. On the other hand, acceptor (lower valency) additives result in 'hard' piezoelectric behaviors particularly suitable for ultrasonic motor applications [1,2,4–6].

However, in these applications, PZT ceramics are often subjected to mechanical loading under different temperatures, either deliberately in the design of the device itself or the use in changing shapes for many smart structure applications or the uses under environmental stresses or environmental heats [1–7]. A prior knowledge of how the material properties change under different conditions is therefore crucial for proper design of a device and for suitable selection of materials for a specific application. Despite that fact, material constants used in many design calculations are often obtained from a stress-free measuring condition or room temperature condition, which in turn may lead to incorrect or inappropriate designs of actuator and transducer. It is therefore important to determine the properties of these materials as functions of applied stress and temperature. Previous investigations on the stress dependence of dielectric and electrical properties of many ceramic systems, i.e. undoped-PZT, PLZT, BT, PMN–PT, PZT–BT and PMN–PZT, have clearly emphasized the importance of the subject [11–19]. The present study aims at studying technically important and commercially available soft and hard PZT ceramics. Many investigations have already revealed interesting results on the dielectric and piezoelectric properties of the

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soft and hard PZT ceramics under stress [13–15,19]. However, there has been no work on the stress and temperature dependence of the ferroelectric properties of the ceramics. Therefore, this study is undertaken to investigate the influences of the uniaxial compressive stress and temperature on the ferroelectric properties of the soft and hard PZT ceramics.

2. Experimental procedure

A commercially available soft PZT (PKI-552, Piezo Kinetics Inc., USA) and hard PZT ceramics (APCI-840, USA) were used in this study. Soft PZT properties presented (measured by the supplier) a longitudinal charge coefficient d_{33} = 550 pm/V; a planer coupling factor k_p = 0.63, a dielectric constant (1 kHz) ε_r = 3400, a Curie temperature T_c = 200 °C and a bulk density = 7.6 g/cm^3. Hard PZT properties presented d_{33} = 290 pm/V, k_p = 0.59, ε_r = 1250, T_c = 325 °C and a bulk density = 7.6 g/cm^3. The disc-shaped samples with a diameter of 6.27 mm and a thickness of 1 mm were pre-poled by the supplier.

The ferroelectric hysteresis (*P–E*) loops were characterized 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 50 Hz from a signal generator (Goodwill, model GAG-809). To study the effects of the uniaxial stress and temperature on the ferroelectric properties, the uniaxial compressometer was constructed with heating system. The compressometer with heater were developed for simultaneous applications of the mechanical stress, the temperature and the electric field. The compressometer cell with heating system consisting of a cylindrical brass cell with a heavy brass base, a brass ram, precisely guided loading platform provides true uniaxial stress during mechanical loading, while the heater provides desired heat to increase the temperature up to 200 °C. The prepared specimen was carefully placed between the two alumina blocks and the electric field was applied to the specimen via the copper shims attached to the alumina blocks. With this setting, the uniaxial compressive stress was applied parallel to the electric field direction and heat was applied to the brass and passed to sample through silicone oil. During the measurements, the specimen was immersed in silicone oil to prevent high-voltage arcing during electric loading. The uniaxial compressive stress was supplied by the servohydraulic load frame and the applied stress was monitored with the pressure guage of the load frame. Heating of the sample was applied by 300 W heater, which was tightened to brass and the applied heat was monitored and controlled by multimeter and computer interface. Measurements were performed as functions of mechanical compressive stress and temperature. Mechanical stress was applied discretely between 0 and 324 MPa and temperature was applied discretely between 25 and 160 °C. The ferroelectric hysteresis (*P–E*) loop was recorded at discrete temperature for each mechanical loading condition. The measurements reported were for the samples during mechanical loading. 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 constant stress and temperature.

3. Results and discussion

The polarization versus electric field (*P–E*) hysteresis loops of the soft and hard PZT ceramics under different compressive stress during loading are shown in Figs. 1 and 2, respectively. It should first be noticed that the area of the *P–E* loops decreases steadily with increasing the compressive stress in both soft and hard PZT ceramics. The *P–E* loop area indicates the polarization dissipation energy of a ferroelectric material subjected to one full cycle of electric field application. This amount of energy loss is directly related to volume involved in the switching process during the application of electric field [2]. Therefore, the decrease in the loop area with increasing stress is a result of the stress-induced domain wall motion suppression [19]. The polarization dissipation energy is consequently found to decrease with increasing the applied stress, indicating that the sample volume contributing to polarization reversal decreases with the increasing compressive stress.

The change in the saturated polarization (P_{sat}), the remanent polarization (P_r), and the coercive field (E_c) with the uniaxial

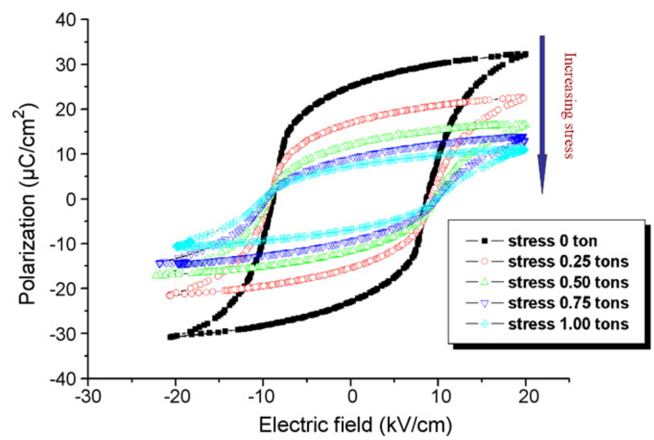

Fig. 1. Polarization vs. electric field (*P–E*) hysteresis loops as a function of compressive stress for soft PZT ceramic during loading.

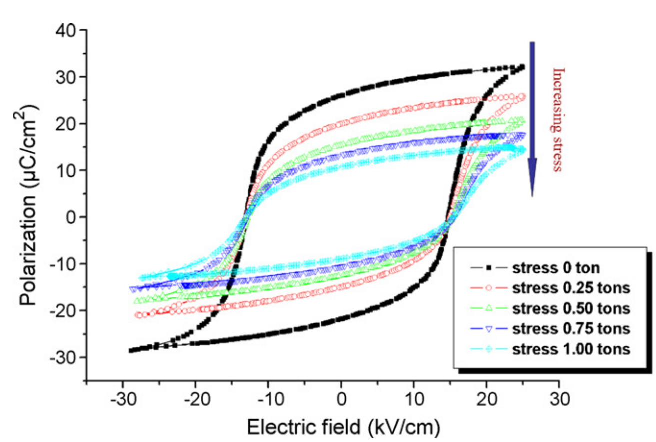

Fig. 2. Polarization vs. electric field (*P–E*) hysteresis loops as a function of compressive stress for hard PZT ceramic during loading.

compressive stress is similar to the trend observed in the dissipation energy (loop area). Both the saturated and remanent polarization decrease as the compressive stress increases. This suggests a significant stress-induced decrease in the switchable part of the spontaneous polarization of the soft and hard PZT ceramics resulting in the observed decrease in the polarization values, as well as the dissipation energy, under high stress [15,23]. In contrast, the applied stress shows little or no influence on the coercive field (E_c). It should also be noted that previous investigations on other ceramic systems, such as BT, PLZT, PMN–PT and PMN–PZT, showed a similar tendency [11,12,19–22,24].

To understand, these experimental results on the soft and hard PZT ceramics, one can interpret the changes in term of domain-reorientation processes [13–19]. When uniaxial compressive stress is applied in the direction parallel to the poling direction, the applied stress tends to keep the ferroelectric domain aligned with their polar axes away from the stress direction through the non-180° ferroelectric domain switching processes. Therefore, it takes a larger than usual applied electric field to reorient the domains along the stress direction, resulting in a lower value of the saturated polarization (P_s). When the electric field is reduced to zero the domain tend to rotate back away from the applied stress direction, resulting in a lower than usual remanent polarization (P_r). Furthermore, the decrease in the dissipation energy with increasing compressive stress indicates that more and more ferroelectric domains are constrain by the stress and cannot be reoriented by the electric field so as to participate in the polarization reversal. Consequently, both the saturated and remanent polarizations become lower with increasing compressive stress [19]. The results of changes in the ferroelectric characteristics of the soft and hard PZT ceramics with increasing compressive stress are in agreement with the previous investigations of many ferroelectric ceramics [11,12,18–25].

The (P–E) hysteresis loops of the soft and hard PZT ceramics under different temperatures are shown in Figs. 3 and 4, respectively. In both cases, area of the P–E loops decreases steadily with increasing the temperature, indicating that polarization dissipation energy decreases when the temperature increases. This is caused by that higher temperature provides

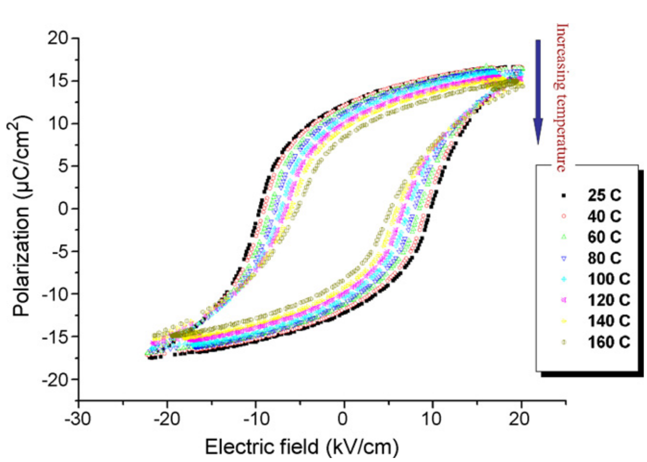

Fig. 3. Polarization vs. electric field (P–E) hysteresis loops as a function of temperature for soft PZT ceramic.

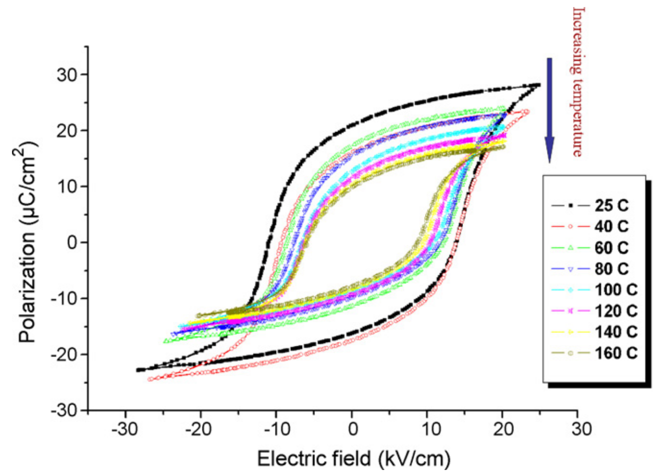

Fig. 4. Polarization vs. electric field (P–E) hysteresis loops as a function of temperature for hard PZT ceramic.

higher thermal fluctuation to the polarization order parameter, which reduces the ferroelectric interaction among the dipoles. Therefore, even E_0 is fixed, the polarization direction is easily turned with the electric field at higher temperatures due to the smaller ferroelectric interaction providing a reduction in both remanent polarization (P_r) and coercivity (E_c). For this reason, the hysteresis loop area is reduced with increasing the temperature. Additionally, slightly asymmetric P–E loops are also observed in the hard PZT, as displayed in Figs. 2 and 4. The observation could be attributed to the presence of the complex defects, which provide the internal field-bias. Therefore, the ferroelectric interaction between these defect dipoles which are trapped near domain walls, and the other dipoles in the domain makes the polarization switching and domain reorientation processes, as well as the domain wall motions, more difficult in one field direction than the other direction. This results in asymmetric loops, and the differences between E_c^+ and E_c^-, and P_r^+ and P_r^-, as observed in Figs. 2 and 4.

4. Conclusions

In this study, the effects of stress and temperature on the ferroelectric properties of soft and hard PZT ceramics are investigated. The results show that in both types of PZT ceramics the ferroelectric characteristics, i.e. the area of the ferroelectric hysteresis (P–E) loops, which corresponds to the energy dissipation, the saturated polarization (P_{sat}) and the remanent polarization (P_r), decrease with increasing compressive stress, while the coercive field (E_c) is virtually unaffected by the applied stress. The non-180° ferroelectric domain switching and stress-induced domain wall suppression processes are responsible for the changes observed. Similar decrease of the ferroelectric parameters is also observed with increasing temperature. The cause for the thermal effect is believed to reduce ferroelectric interaction due to higher thermal fluctuation. More importantly, this study undoubtedly shows that the applied stress and the temperature have significant influences on the ferroelectric properties of the soft and hard PZT ceramics.

Acknowledgments

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