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Evolution of remnant state variables and linear material properties in ferroelectric ceramics during compressive loading and unloading

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Abstract

A pre-poled lead titanate zirconate (PZT) cube specimen was subjected to impulsive compression stress with increasing amplitude. Linear material properties were evaluated graphically from the measured responses. The material properties were plotted versus relative remnant polarization and fitted by quadratic curves. Then the specimen was subjected to compressive creep loading and unloading of various constant magnitudes. The linear material properties obtained from impulsive stress loading were used to calculate the evolutions of remnant state variables and material properties during the compressive creep loading and unloading. The magnitudes of polarization and strains that were induced and recovered during the creep loading and unloading were plotted and discussed.

Keywords: Ferroelectric; Compressive stress; Switching; Remnant; Linear moduli

1. Introduction

Thanks to strong electromechanical coupling, prompt response, low power consumption, and many other excellent characteristics, piezoelectric ceramics such as lead titanate zirconate (PZT) have been used for various types of actuators and sensors. In these applications, the materials are often subjected to compressive stress large enough to cause unnecessary domain switching in the materials. The domain switching changes the internal structure of the materials and thus the characteristics of the materials, deteriorating the performance of piezoelectric devices. Therefore, it is very important to take the nonlinear behavior of the materials by strong compressive stress into consideration when selecting piezoelectric materials.

Constitutive modeling for nonlinear behavior of ferroelectric ceramic materials has been investigated by researchers [1–3]. In addition to the theoretical study, much experimental work has been made to understand the nonlinear behavior of the materials. Nonlinear electromechanical coupling behavior at room temperature had been measured and discussed by Cao and Evans [4] and Lynch [5]. Tensile creep behavior of ferroelectric ceramics at room temperature had been measured and reported [6,7]. Room-temperature ferroelastic switching and creep behavior under electric field and compressive stress had been investigated [8,9]. The change of Young's modulus of initially unpoled and pre-poled soft PZT material under compressive stress was studied using the partial unloading method, and it was found that Young's modulus increased with compressive stress [10]. In electric field and compressive stress loading experiments on non-poled and poled PZT ceramic, Zhou et al. [11] separated reversible and irreversible parts of total measured responses by evaluating the evolution of linear material properties, and they found that longitudinal and transverse irreversible strains changed significantly during both loading and unloading processes. Selten et al. [12] observed constant permittivity and linear dependency of piezoelectric coefficient on remnant polarization in a PZT ceramic under unipolar electrical loading; Liu and Huber [13] calculated the evolutions of linear elastic, dielectric, and piezoelectric moduli in a ferroelectric ceramic under electro-mechanical loading at room temperature. Webber et al. [14] measured and characterized the nonlinear switching behavior of soft lead zirconate titanate induced by compressive stress at elevated temperatures, observing strong temperature dependency of ferroelastic switching. The evolutions of linear

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materials and remnant state variables had been calculated from measured total responses at high temperatures as well as room temperature. Thermal expansion and pyroelectric coefficients had been observed to depend linearly on remnant polarization in a PZT wafer [15]; piezoelectric coefficients were given as a function of remnant polarization and temperature [16,17]. Comparison of polarization hysteresis and strain butterfly loops at room and high temperatures had been made by calculating remnant polarization and remnant strain [18]. Temperature-dependent ferroelastic behavior of ferroelectric ceramics at room and high temperatures was investigated and the evolutions of linear material properties at different temperatures were analyzed by introducing so-called relative remnant polarization [19]. A poled PZT cube was subjected to constant compressive stress of various magnitudes at four different room and high temperatures, and the evolutions of linear material properties and remnant state variables were calculated. Longitudinal and transverse remnant strains were shown to depend linearly on relative remnant polarization [20].

In present study, a poled PZT cube specimen is subjected to impulsive compression stress with increasing amplitude, and polarization and strain responses are measured during the impulsive loading. From the measured electric displacement and strains, piezoelectric coefficient and compliance coefficients are evaluated by a graphical method and plotted versus relative remnant polarization. Quadratic functions are proposed to fit the dependence of linear material properties on relative remnant polarization. Next, constant compressive stress of various magnitudes is applied to the specimen for a period of time. The stress is then removed and the specimen is left unstressed for the same period of time. During the total creep loading and unloading, electric displacement and strains are measured. Using the quadratic dependence of linear material properties on relative remnant polarization, the evolutions of remnant state variables and linear material properties during compressive creep loading and unloading are calculated. The behavior of recovered polarization and strains during the unloading period of compressive stress is calculated and discussed.

2. Experiment

The specimen that is used for experiments is a commercially available soft PZT rectangular parallelepiped (PZT5H1, Morgan Technical Ceramics, UK) of dimensions10 mm × 10 mm × 12 mm. It has been developed for applications that require high electromechanical activity and high dielectric constant, such as hydrophones, sound detectors, accelerometers, flow detectors and flow meters. The specimen PZT ceramic is electroded on the 10 mm × 10 mm faces and poled along the 12 mm direction. The 12 mm direction is referred to as longitudinal direction and designated as x_3; two mutually orthogonal directions perpendicular to x_3 are called transverse directions and designated as x_1 and x_2, respectively. The three mutually orthogonal directions x_1, x_2 and x_3 are also often called as 1, 2, and 3 directions, respectively. According to manufacturing company, the Curie point of the material is 200 °C, the density is 7400 kg m^{-3}, the coupling factor is $k_p=0.60$, the piezoelectric charge coefficients are $d_{31}=-250\times10^{-12}$ mV^{-1} and $d_{33}=620\times10^{-12}$ mV^{-1}, and the elastic compliance coefficients at constant electric field $s_{33}=21.9\times10^{-12}$ m^2 N^{-1}, $s_{11}=17.7\times10^{-12}$ m^2 N^{-1}, and $s_{12}=-5.7\times10^{-12}$ m^2 N^{-1}.

In present paper, two different types of compressive stress are applied to a poled PZT specimen at room temperature. First, impulsive compression stress with gradually increasing amplitude is applied to the specimen. A typical impulsive stress is shown in Fig. 2(a), where the amplitude of stress impulse increases from −10 MPa to −500 MPa. As will be explained below, a preload of −5 MPa is included in the stress amplitudes. In the impulse loading and unloading, stress increases and decreases at the fastest loading rate of 10 MPa s^{-1} available for the Instron 5869 machine. After both loading and unloading are finished, stress remains constant for 5 s. Longitudinal electric displacement and longitudinal and transverse strains are measured and recorded, which are used to evaluate the evolution of linear material properties during the ferroelastic switching by a graphical method. Next, constant compressive stresses of various magnitudes are applied to the specimen in the longitudinal direction of the specimen. Eight magnitudes of applied compressive stress are 20, 30, 50, 70, 100, 150, 300, and 500 MPa. A typical stress loading–unloading cycle is as follows. In the loading part, the magnitude of compressive stress increases at a slow rate of 0.5 MPa s^{-1}. Then the stress remains constant at the target level for 500 s. In the subsequent unloading part, stress decreases at the same slow rate of 0.5 MPa s^{-1}. After complete unloading, the specimen is left at zero stress for another 500 s. Because of a preloaded stress, the zero stress here is actually −5 MPa. During the whole loading and unloading cycle of compressive stress, electric displacement in x_3 direction D_3 and extensional strains in x_1 and x_3 directions S_1 and S_3 are measured and recorded. For all the eight compressive experiments, only one specimen is used. After each experiment, electric field of magnitude 1.0 MV m^{-1} is applied in $-x_3$ direction for 600 s to restore the same initial poled state. Then, after removing the electric field, the specimen is under no electric field for another 600 s. The restored initial poled state is the state represented by the polarization value of −.52 C m^{-2}. A pre-stress of −5 MPa is then applied to stabilize the initial polarization state. After another 600 s under the preload of −5 MPa, polarization is reduced to −0.50 Cm^{-2}. Thus the eight stress levels from −20 MPa to −500 MPa include the preloaded stress −5 MPa. Then, strains are set to be zero, and now it is ready to begin compressive creep loading and unloading experiments. To verify the restored poled state, several experiments at the same stress level are repeated. No significant history dependence of material responses is observed, implying that the restored poled state is sufficiently close to the initial poled state.

A schematic diagram of an experimental setup is shown in Fig. 1. The bottom face of a PZT specimen that is poled downward is in contact with the top face of steel fixture to which applied high voltage is connected. A Teflon jig with a square hole in its center, which has excellent insulation, is used as an insulating material between high voltage sources and strain gauges. Electric displacement in longitudinal direction is measured using a Sawyer–Tower bridge, where a reference capacitor (metalized polypropylene capacitor) of capacitance 100 μF is connected to the specimen in series. This is 1000