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High electric-field-induced strain of $Pb(Mg_{1/3}Nb_{2/3})O_3-PbTiO_3$ crystals in multilayer actuators

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Abstract

An optimum composition range ($29\% \leq x \leq 31\%$) of ⟨001⟩ oriented $(1-x)Pb(Mg_{1/3}Nb_{2/3})O_3-xPbTiO_3$(PMNT) crystals was ascertained for multilayer actuator applications, which exhibited high-strain and low-hysteresis behavior. A −1.5 kV/cm negative *E*-field can be applied to PMNT ferroelectric samples with low hysteresis. Forty layer actuators with individual element sizes of $7 \times 7 \times 0.7$ mm^3 were fabricated under identical processing conditions using two different materials: (1) single crystal PMNT and (2) commercial PZT-SF ceramics. Under free-load conditions, 48 μm displacements can been achieved in PMNT actuators at electric fields ranging from −1.5 to 10 kV/cm, which is more than twice the displacement of the PZT-SF actuators driven from −10 to 10 kV/cm. Under 4 kg loading, the displacements in PMNT stain actuators are decreased to 42.5 μm.

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Keywords: A. PMNT crystals; A. PMNT multilayer actuators; E. Electric-field-induced strain

1. Introduction

In recent years, relaxor-based ferroelectric single crystals $(1-x)Pb(Mg_{1/3}Nb_{2/3})O_3-xPbTiO_3$ (PMNT) and $(1-x)Pb(Nb_{1/3}Nb_{2/3})O_3-xPbTiO_3$ (PZNT) have attracted considerable attention due to their extremely high piezoelectric properties. $\langle 001\rangle_{cub}$-oriented single crystals of PMNT and PZNT in the rhombohedral phase near their morphotropic phase boundaries (MPBs) have been reported to possess ultrahigh piezoelectric response ($k_{33} \sim 94\%$, $d_{33} \sim 2500$ pC/N) and large electric field strain values (1.7%) with low-hysteresis [1–4], which are markedly superior to those of conventional Pb(Zr, Ti)O_3 (PZT) ceramics. Recent experimental and theoretical studies have indicated that this strong piezoelectric response could be driven by polarization rotation induced by an external electric field in these single crystals [5,6].

Because PMNT is more stable than PZNT in melting, it is easier and more effective to grow large, high quality single crystal PMNT by the Bridgman method [7,8]. For this reason, the high performance and availability of PMNT ferroelectric single crystals herald their wide application for piezoelectric devices, such as ultrasonic transducers and strain actuators. One of the applications for PMNT is the development of multilayer actuators for electromechanical systems. In this article, high electric-field-induced strain of ⟨001⟩-oriented PMNT crystals with different compositions (PT content) is studied and the achievements for 40 layer PMNT and PZT-SF actuators are investigated. It is believed that this research will be useful to advance the understanding of the properties of the PMNT system and promote its practical application in solid-state strain actuators.

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2. Experimental

PMNT crystals were grown directly from the melt by a modified Bridgman technique [9], and cut into ⟨001⟩-oriented $7 \times 7 \times 1.2\ mm^3$ wafers. Silver paste was painted on the samples plates and fired at 600 °C for 1 h. The dielectric maximum temperature (T_m) was determined from the maximum of permittivity using a HP4192A impedance analyzer from room temperature to 180 °C at 1 kHz. For pseudocubic PMNT crystals, T_m, which approximately equals the Curie temperature (T_c), is used to estimate the composition of the PMNT crystals with an approximate phase diagram [10]. The samples were poled in silicon oil under an applied field of 10 kV/cm for fifteen minutes near the dielectric maximum temperature (T_m) and cooled down to room temperature. The piezoelectric coefficients (d_{33}) were measured by a Berlincount-type quasistatic meter at about 55 Hz. The samples were finally grinded into a thickness of 0.7 mm and copper electrodes were deposited by vapor deposition method. Multilayer PMNT and PZT-SF actuators were made up of 40 plates of $7 \times 7 \times 0.7\ mm^3$, which were contacted by a nonconductive epoxy. The average d_{33} values for these PMNT and PZT-SF plates had directly been measured 1560 and 380 pC/N, respectively. Silver paste was used to make electrical contact to the alternate edges of the electrodes. Polarization and strain were measured using a modified Sawyer–Tower circuit and a linear variable differential transducer (LVDT). Electric fields were applied using an amplified triangle wave form at a drive frequency of 0.1 Hz.

3. Results and discussions

3.1. Dependence of strain versus composition of ⟨001⟩ oriented crystals

In strain actuator design, it is important for the material to exhibit high strain values with low nonlinear hysteresis, which determines the actuator performance. Due to the segregation during the growth of PMNT crystals by a modified Bridgman method, the compositions (PT content) of PMNT crystals fluctuate in the same boule, resulting in variations of property in the crystals. Thus it is of significant importance to determine the PMNT optimum compositional range for its application in strain actuators. The piezoelectric response is maximized for crystals with ⟨001⟩-oriented structure. So we investigated the dependence of strain on composition in ⟨001⟩-oriented PMNT crystals.

Fig. 1(a) shows strain as a function of E-field (unipolar, $E \leq 15$ kV/cm) in ⟨001⟩ oriented PMNT crystals. Strain and hysteresis as a function of composition for ⟨001⟩ oriented crystals are presented in Fig. 1(b) and (c), respectively. Due to the complexity of the domain configuration and phase composition near MPB in PMNT crystals [10–13], the domain motion and phase transformation can easily be induced by an E-field, which causes abnormally high strain and large hysteresis. In the rhombohedral ferroelectric phase, high strain values and small hysteresis can be achieved with the engineered-domain stability [3]. The solid line in Fig. 1(c) represents the gauss fits to the strain hysteresis for different compositional PMNT crystals, which show small hysteresis ($<5\%$) in the rhombohedral ferroelectric phase for 15 kV/cm. In view of the strain values in Fig. 1(b), we may select ⟨001⟩-oriented PMNT crystals of optimum composition range ($29\% \leq x \leq 31\%$) for strain actuator application. In addition, PMNT crystals in this composition range have also considerable high temperature of phase transitions from rhombohedral to tetragonal (~100 °C) [14].

For a long time the MPB of PMN–xPT crystals between the rhombohedral and tetragonal phases has been regarded to lie in the range $0.26\% \leq x \leq 0.36\%$ [15]. Nevertheless, recent works [16,17] in unpoled ceramics of PMN–xPT with $27\% \leq x \leq 35\%$ have found a monoclinic M_C (and even a second M_B) phase by diffraction experiments. Very recently, the width of the MPB region ($30\% \leqslant x \leqslant 35\%$) and an irreversible rhombohedral-monoclinic M_A-monoclinic M_C-tetragonal phase transition have been proposed by Guo et al. [18] for ⟨001⟩-poled crystals from the dielectric and piezoelectric properties. So in ⟨001⟩-oriented PMNT crystals of the optimum composition range ($29\% \leq x \leq 31\%$), an M_C phase may exist or can easily be induced. According to the Devonshire theory [19], there exists little difference between the free energies of the rhombohedral (or monoclinic) and tetragonal phases near the MPB. The observed monoclinic phases are believed to be responsible for the high piezoelectric response of these materials [16–19]. As shown in Fig. 1, samples with $x > 35\%$, which are in the tetragonal phase, show very low strain values. The hysteresis also increases as one moves from the rhombohedral into the monoclinic phase and the tetragonal phases, from $x = 0.29$ to 35%.

Strain characteristics for PMN–30%PT crystals and PZT-SF ceramics at the same drive E-field are shown in Fig. 2. Both show approximately linear behavior with low hysteresis. In contrast to PZT-SF ceramics, the strain values for PMNT crystals are more than five times at 10 kV/cm (0.18% compared to 0.035%). Commonly, the $S(E)$ relations in PMNT crystals can be well described as follows [20]:

$$S = d_{33}E + M_{11}E^2$$

Where $d_{33}E$ is the piezoelectric effect and $M_{11}E^2$ is the electrostrictive effect. However, in moderate (or low) electric field, the main contribution for strain is the piezoelectric effect. Effective piezoelectric coefficients of 1800 and 350 pC/N are calculated from the slope of strain curve, approximately consistent with their measured values (1790 and 360 pC/N).

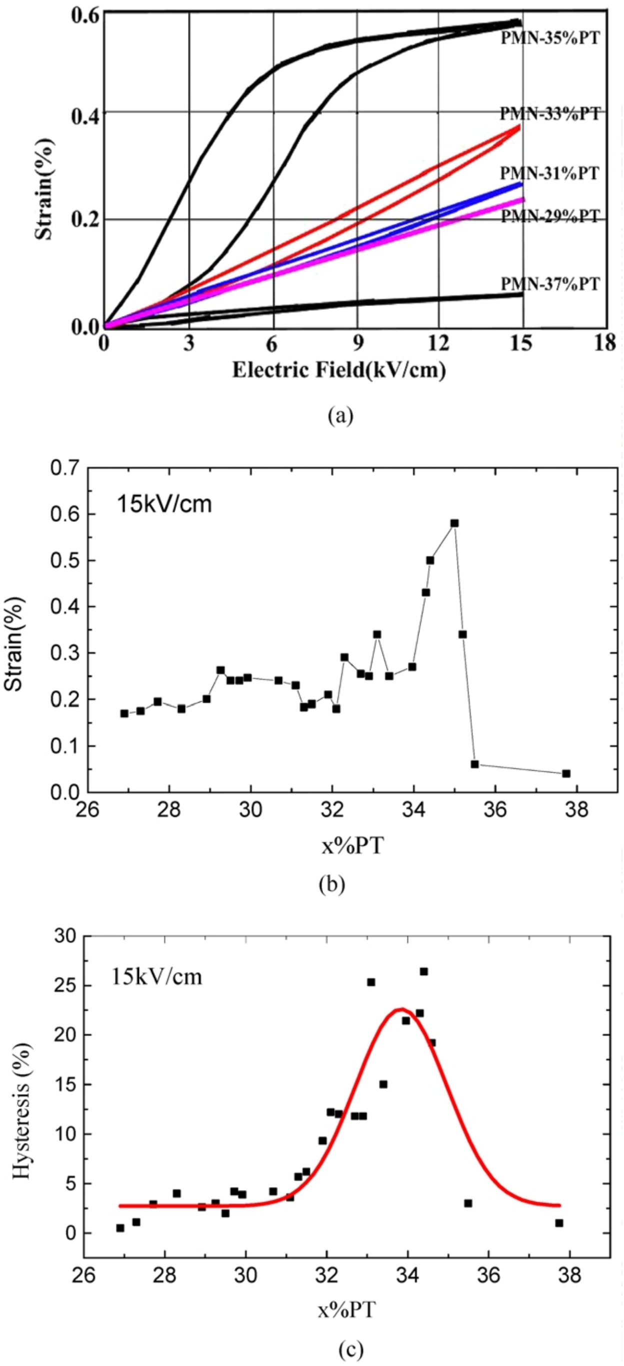


Fig. 1. (a) Strain versus E-field (unipolar, $E \leq 15$ kV/cm) curve for ⟨001⟩ oriented PMNT crystals. Strain and hysteresis as a function of PT content (x%) for ⟨001⟩ oriented PMNT crystals in (b) and (c), respectively. Solid line in (c) represents the gauss fits to strain hysteresis.

3.2. The strain behavior in bipolar electric fields

Fig. 3(a) depicts the strain behavior of ⟨001⟩ oriented PMN–30%PT crystals in a bipolar electric field. For strain actuator, it is desired that equivalent positive–negative electric field can be utilized. However, as presented in Fig. 3(a), due to ferroelectric domain switching in bipolar electric fields, the strain curve of PMNT crystals has a typical butterfly shape (nonlinear), which goes against a strain actuator design. How much negative-electric-field can be applied to a new pattern strain actuator? It is indispensable to investigate the relation of negative electric

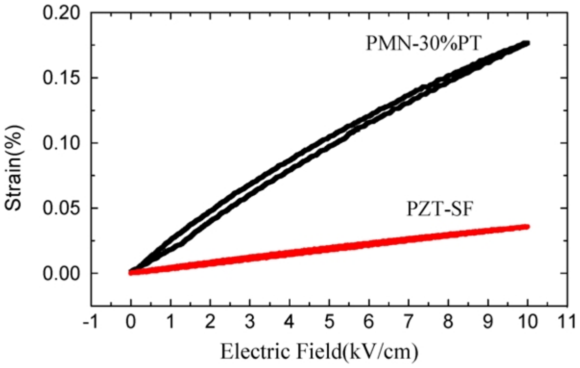
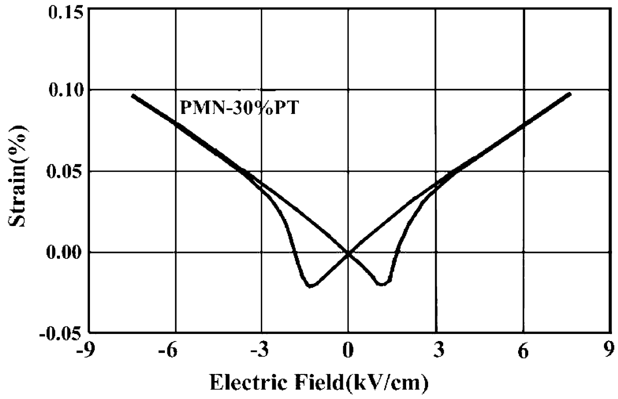


Fig. 2. The strain behavior for PMN-30%PT crystals and PZT-SF ceramics at the same drive *E*-fields.

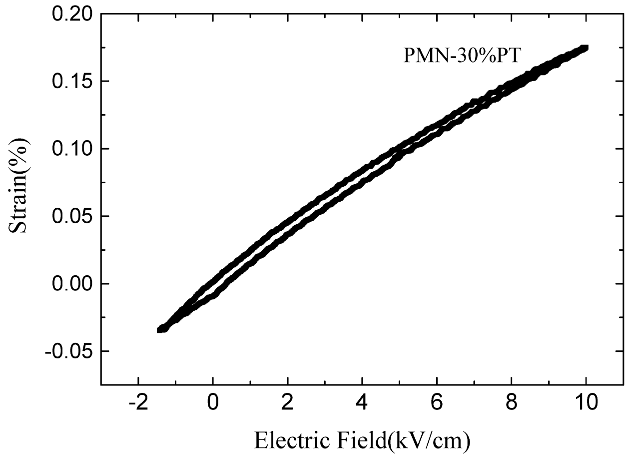
field and strain curve. As Fig. 3(b) shows, when a negative electric field is added to −1.5 kV/cm, the strain curve of the crystals still can exhibit low hysteresis (<5%) and approximate linearity. As a result, the strain values can be increased approximately 0.03% compared to those of a unipolar electric field (from 0 to 7 kV/cm), which is beneficial to the application of PMNT as strain actuator in moderate electric fields. When $E < -1.5$ kV/cm, due to ferroelectric domain switching in some crystals, the strain curve is nonlinear accompanied with large hysteresis.

3.3. Strain behavior in multilayer actuators

Strain versus *E*-field characteristics for PMNT multilayer actuators and PZT-SF multilayer actuators are shown in Figs. 4 and 5, respectively. Both show approximately linear behavior with low hysteresis. For PZT-SF ceramics, due to their large coercive fields, domain switching is difficult and equivalent positive–negative electric field can be applied to their strain actuators (Fig. 5). As presented in Fig. 4(a), under free-load condition, 48 μm displacements in 40 layer PMNT actuators can been achieved at the electric field from −1.5 to 10 kV/cm, which is more than twice



(a)



(b)

Fig. 3. (a) The strain behavior of ⟨001⟩-oriented PMN-30%PT crystals in bipolar electric fields. (b) Strain versus *E*-field curve for ⟨001⟩-oriented PMN-30%PT crystals.

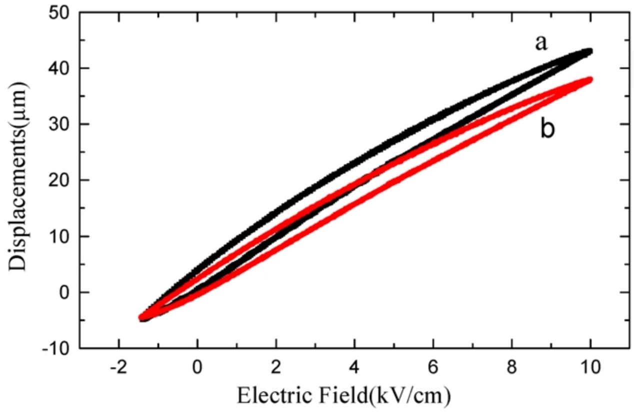


Fig. 4. Strain versus electric field characteristics for PMNT multilayer actuators (a) free-loading (b) 4 kg loading.

those of PZT-SF actuators (about 22 μm) that can been driven from −10 to 10 kV/cm. From the slope of strain, effective piezoelectric coefficients of PMNT and PZT-SF are 1500 and 390 pC/N, respectively. When a 4 kg load is added to PMNT multiplayer actuators, as shown in Fig. 4(b), the displacements are decreased to 42.5 μm.

4. Conclusions

An optimum composition range (29% ≤ x ≤ 31%) was obtained for ⟨001⟩ oriented $(1-x)Pb(Mg_{1/3}Nb_{2/3})O_3$–$xPbTiO_3$ crystals, which exhibited high-strain and low-hysteresis behavior. At the same drive E-field, the strain values in the single crystals are more than five times compared to PZT-SF ceramics. A −1.5 kV/cm negative E-field can be utilized with linear strain curve (low hysteresis), which is beneficial to the application of PMNT strain actuators in moderate electric fields, while equivalent positive–negative electric field can be utilized in commercial PZT-SF strain actuators.

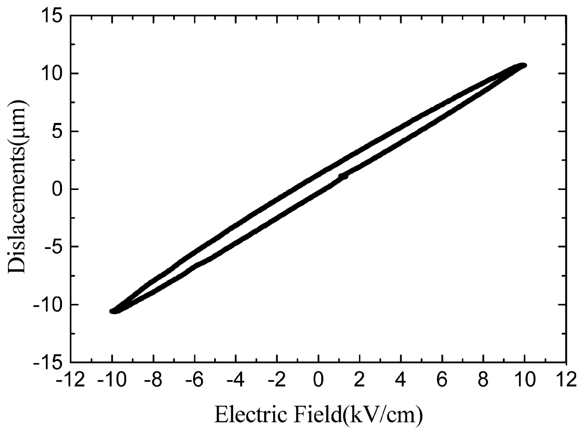


Fig. 5. Strain versus electric field characteristics for PZT-SF multilayer actuators.

Forty layer PMNT strain actuators were fabricated from their optimum compositions, with displacements of 48 μm at E-fields from −1.5 to 10 kV/cm, while 22 μm displacements were observed for PZT-SF actuators at E-fields from −10 to 10 kV/cm. When a 4 kg load was added to the PMNT multiplayer actuators, the displacements were decreased to 42.5 μm. In moderate (small) E-field, the main contribution for strain is the piezoelectric effect, so high piezoelectric constants (d_{33}) and large coercive field for the materials are desired.

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