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Short communication

Effect of PMN content on the phase structure and electrical properties of PMN–PZT ceramics

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Abstract

xPb($Mg_{1/3}Nb_{2/3}$)O_3–(1−x)Pb($Zr_{0.47}Ti_{0.53}$)O_3(xPMN–(1−x)PZT) ($0.15 \leq x \leq 0.30$) ceramics were prepared by the conventional oxide-mixed method. The phase structure, ferroelectric and piezoelectric properties and field-induced strain behavior were systematically investigated. It was found that 0.25PMN−0.75PZT possessed superior electrical properties due to its composition close to the MPB (Morphotropic Phase Boundary). The composition near the MPB enables the existence of multiple polarization directions and consequently facilitates the domain reorientation, which should be responsible for the superior electrical properties. A large remnant polarization (P_r) of 34.2 μC/cm^2 and a high piezoelectric coefficient (d_{33}) of 698 pC/N were obtained. Especially, a large field-induced strain of 2.2‰ (at 40 kV/cm) was attained, which shows a great promise for actuator applications.

Keywords: C. Ferroelectric; C. Piezoelectric; PMN-PZT; Field-induced strain

1. Introduction

Lead-based perovskite-type solid solutions consisting of ferroelectric and relaxor materials have attracted a growing fundamental and practical interest due to their excellent electrical properties [1–3]. Among the lead-based complex perovskites, lead magnesium niobate (PMN) and lead zirconate titanate (PZT) ceramics have been investigated extensively, both from academic and commercial viewpoints. These two kinds of ceramics possess distinct characteristics that in turn make each one suitable for different applications. With the complementary features of PMN and PZT described in many publications, the solid solutions between PMN and PZT are expected to exhibit better dielectric and piezoelectric properties than those of the single-phase PMN and PZT. However, recent literatures mainly focused on the investigations of dielectric and relaxation properties of PMN–PZT solid solutions [4–7]. There are a few literatures [8–10] about the ferroelectric and piezoelectric properties of PMN–PZT solid solutions. Especially, field-induced strain behavior of PMN–PZT is rarely reported, which is a significantly important parameter for actuator applications.

In this study, PMN–PZT ceramics with different PMN contents were prepared by the conventional oxide-mixed method. The phase structure, ferroelectric and piezoelectric properties and field-induced strain behavior were systematically investigated. It was found that 0.25PMN–0.75PZT possessed superior electrical properties due to its composition close to the MPB. Especially, a very high field-induced longitudinal strain was obtained for this composition, which shows a great potential for actuator applications.

2. Experimental procedure

The general formula of materials studied here was xPb($Mg_{1/3}Nb_{2/3}$)O_3−(1−x)Pb($Zr_{0.47}Ti_{0.53}$)O_3, where x=0.15, 0.20, 0.25 and 0.30. Reagent-grade powders of Pb_3O_4, $SrCO_3$, $BaCO_3$, $MgCO_3 \cdot 5H_2O$, Nb_2O_5, TiO_2 and ZrO_2 were weighed by mole ratio and mixed together by ball-milling for 8 h. After drying, the mixture was calcined at 650 °C for 1 h and 850 °C for 2 h.

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Thereafter, the calcined powders were ball-milled for 24 h. A polyvinyl alcohol was added to the dried powders and then the powders were pressed into disks. The green pellets were burned out at 800 °C for 1 h and then sintered at 1260 °C for 2 h. For the property measurement, samples were polished and coated with silver paint as electrodes. Afterward, the samples were heat-treated at 700 °C for 30 min to ensure the contact between the electrodes and ceramic surfaces.

The phase structures of specimens were examined by X-ray diffraction (XRD) using CuKα radiation (D/MAX-2550V, Rigaku, Tokyo, Japan). Microstructure of ceramics was observed by SEM (Model JSM-6700F; JEOL, Tokyo, Japan). An LCR analyzer (HP 4284 A; Hewlett-Packard, Kobe-shi, Japan) was used to measure the dielectric properties at a heating rate of 2 °C/min. The polarization versus electric field (*P–E*) behavior and the field-induced longitudinal strain behavior (*S–E*) were determined using a standard ferroelectric tester (TF analyzer 2000, aixACCT, Germany). The piezoelectric properties were measured after samples were poled in silicon oil and aged in air for 24 h. A quasistatic piezo d_{33} meter (Model ZJ-3A, Institute of Acoustics Academic Sinica, China) was used to measure piezoelectric constant (d_{33}). The electromechanical coupling coefficient (k_p) and electromechanical quality factor (Q_m) were determined by resonance and anti-resonance technique [11] using an impedance analyzer (Model Agilent 4294A, CA).

3. Results and discussion

Fig. 1 shows the XRD patterns of xPMN–(1−x)PZT with x=0.15, 0.20, 0.25 and 0.30. It can be seen that a perovskite phase was obtained for all the compositions. The phase structure demonstrated a transition from tetragonal to rhombohedral with the increase of PMN content. From the XRD analysis, it could be deduced that 0.25PMN–0.75PZT may be located closest to the MPB region, where the tetragonal and rhombohedral coexist. The composition near the MPB may be responsible for the subsequent excellent ferroelectric and piezoelectric properties, which could be attributed to the existence of multiple polarization directions.

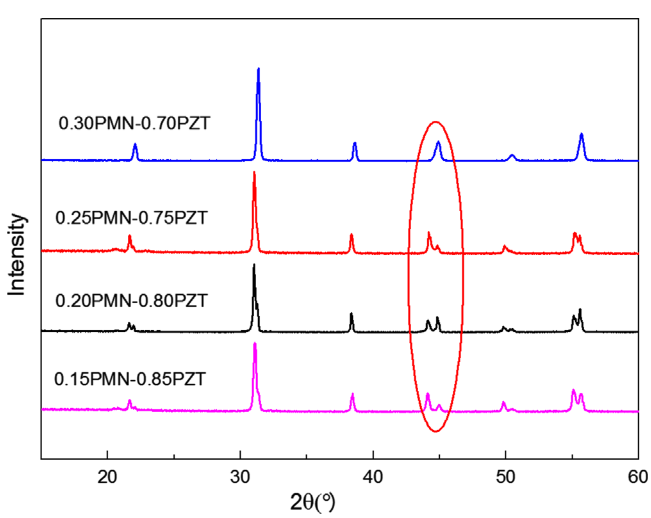

Fig. 1. X-ray diffraction patterns of the sintered xPMN–(1−x)PZT ceramics.

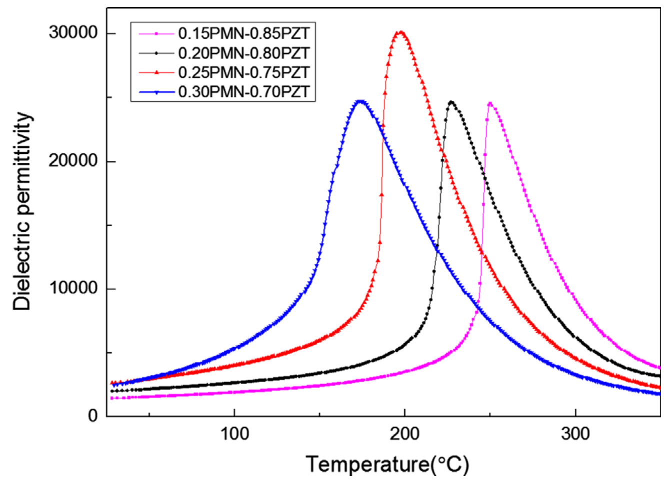

Fig. 2. Temperature dependence of dielectric permittivity at 1 kHz for the xPMN–(1−x)PZT ceramics.

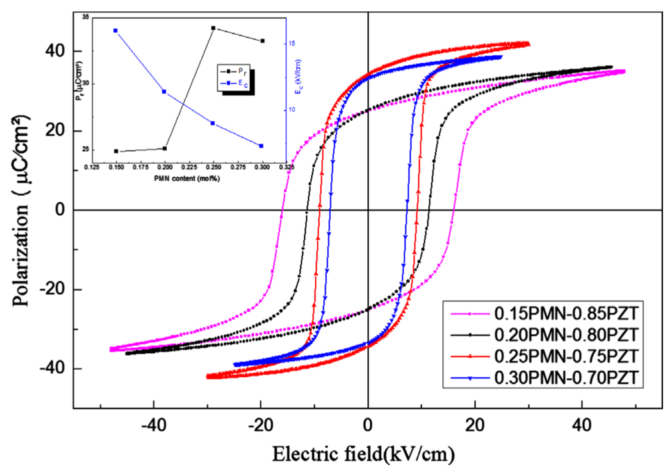

Fig. 3. Polarization–electric field (*P–E*) hysteresis loops of the xPMN–(1−x)PZT ceramics.

Fig. 2 shows the temperature dependence of the dielectric permittivity at 1 kHz frequency of PMN–PZT ceramics with different PMN amounts. It is observed that the T_{max} (the temperature that shows the maximum dielectric constant) decreases with an increase of PMN content, which could be ascribed to the low T_c (−12 °C) of PMN. The dielectric constant at T_{max} increased with an increase of PMN and the maximum value of 30,109 was obtained for 0.25PMN–0.75PZT ceramics at 1 kHz frequency. However, the value decreased significantly with the continuous increase of PMN content. The high dielectric permittivity at T_{max} of 0.25PMN–0.75PZT should be attributed to its composition close to MPB of the ternary system.

Fig. 3 shows the *P–E* curves of PMN–PZT ceramics with different PMN amounts. From the inset, it can be seen that the remnant polarization (P_r) increases and the coercive field (E_c) decreases when the amount of PMN increases. The maximum value of P_r~34.2 μC/cm^2 could be obtained for 0.25PMN–

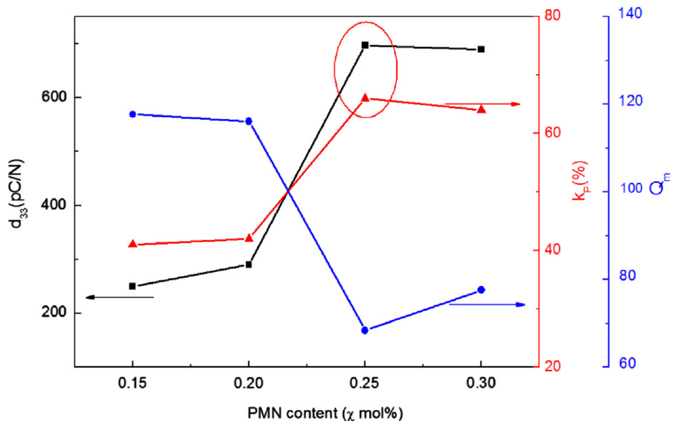

Fig. 4. Piezoelectric coefficient d_{33}, electromechanical coupling factor k_p and quality factor Q_m of the xPMN–(1−x)PZT ceramics.

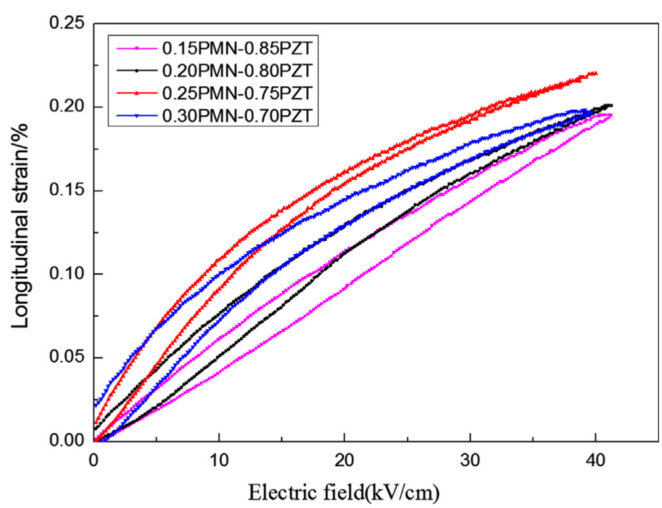

Fig. 5. Field-induced longitudinal strain behavior of the xPMN−(1−x)PZT ceramics.

0.75PZT. The superior ferroelectric properties of 0.25PMN–0.75PZT should be ascribed to the composition close to MPB [12,13]. The MPB composition with the existence of multiple polarization directions facilitates domain reorientation and consequently enables the preferred ferroelectric properties.

Fig. 4 shows the changes in piezoelectric coefficient d_{33}, electromechanical coupling factor k_p and electromechanical quality factor Q_m values as a function of the amount of PMN. When the amount of PMN was lower than 0.25 mol%, d_{33} and k_p increased rapidly with the increase of PMN content. The optimized values for d_{33} of 698 pC/N and k_p of 66% were obtained for 0.25PMN–0.75PZT, which are higher than those in the ever reported literatures [2]. The superior piezoelectric properties could be attributed to the multiple polarization directions of the composition near the MPB. However, when the amount of PMN was above 0.25 mol%, d_{33} and k_p decreased slightly. These results correspond with the phase structure transformation illustrated in Fig. 1.

Fig. 5 shows the field-induced longitudinal strain behavior of PMN–PZT ceramics with different PMN amounts. The strain increased with the increase of PMN content. Then the value tended to decrease with the continuous increase of PMN amount. The strain of 2.2‰ under the applied electric field 40 kV/cm was observed for 0.25PMN–0.75PZT specimen, which could be attributed to the appropriate composition near the MPB and the consequent superior piezoelectric properties. This large strain is the largest in the ever reported PMN–PZT systems [14,15], which shows a great promise for actuator applications.

4. Conclusion

The PMN–PZT ceramics with different PMN contents were fabricated by the conventional oxide-mixed method. The phase structure, ferroelectric and piezoelectric properties were systematically investigated. Also, the field-induced longitudinal strain behavior was studied. It was found that 0.25PMN–0.75PZT possessed superior electrical properties due to its composition close to the MPB. The composition near the MPB enables the existence of multiple polarization directions and consequently facilitates domain reorientation, which should be responsible for the superior electrical properties. The maximum dielectric constant value of 30,109 at T_{max} was obtained for 0.25PMN–0.75PZT ceramics at 1 kHz frequency. A large remnant polarization (P_r) of 34.2 μC/cm^2 and a high piezoelectric coefficient (d_{33}) of 698 pC/N were obtained. Especially, a large field-induced strain of 2.2% (at 40 kV/cm) was attained, which shows a great potential for actuator applications.

Acknowledgments

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