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Characterization and piezoelectric thermal stability of PIN–PMN–PT ternary ceramics near the morphotropic phase boundary

Dabin Lin*, Zhenrong Li**, Fei Li, Zhuo Xu, Xi Yao

Electronic Materials Research Laboratory, Key Laboratory of Education Ministry, Xi'an Jiaotong University, Xi'an 710049, China

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The phase structure and piezo/dielectric properties of xPb($In_{1/2}Nb_{1/2}$)O_3–yPb($Mg_{1/3}Nb_{2/3}$)O_3–$z$$PbTiO_3$ (PIN–PMN–PT $x/y/z$) ternary ceramics with morphotropic phase boundary (MPB) composition were investigated. It was found that Curie temperatures of choosing composition changed from 165 °C to 293 °C and the piezoelectric properties are almost the same at room temperature in PIN–PMN–PT system near MPB. The piezoelectric coefficient d_{33} and electromechanical coupling factor k_p are higher than 440 pC/N and 60%, respectively. Temperautre and dc bias dependence of piezoelectric response for PIN–PMN–PT ceramics were measured. The usage temperature range was found to be improved, compared with PMN–PT single crystal near MPB.

1. Introduction

Relaxor-based ferroelectric crystals Pb($Mg_{1/3}Nb_{2/3}$)O_3–$PbTiO_3$ (PMN–PT) and Pb($Zn_{1/3}Nb_{2/3}$)O_3–$PbTiO_3$(PZN–PT) with morphotropic phase boundary (MPB) composition have been extensively studied due to their extraordinary large electromechanical coupling coefficient and piezoelectric coefficient [1] and [2]. The outstanding properties have stimulated the application research in transducers, sensors and actuators [3–5]. The low Curie temperature (T_c = 130–160 °C) of PMN–xPT (x = 30–35) limits their applications in high temperature environment [6–8]. Moreover, the usage temperature range of PMN–xPT ferroelectric material is also restricted by the rhombohedral to tetragonal phase transformation temperature (i.e., T_{R-T}), which is usually lower than 100 °C. Thus, it is important to improve piezoelectric thermal stability of PMN–PT crystals. The PIN–PMN–PT ternary system with relatively high T_c appears to be a promising candidate and has attracted many investigations [9–13].

Hosono et al. [8] found that the MPB composition in Pb($In_{1/2}Nb_{1/2}$)O_3–Pb($Mg_{1/3}Nb_{2/3}$)O_3–$PbTiO_3$ (PIN–PMN–PT) ternary system is an almost linear between the MPBs of PMN–32PT and PIN–37PT. The Curie temperatures of PIN–PMN–PT near MPB are changed from 160 °C to 320 °C by adjusting the composition. It is intensity to study the piezo/dielectric properties of PIN–PMN–PT ceramics. From applied viewpoint, thermal stability of piezoelectric properties and dc bias dependence of piezoelectric response are also expected to be investigated. In this work, the phase structure and electrical properties as well as their thermal stability of PIN–PMN–PT ternary ceramics near MPB were studied. The influence of dc bias dependence of piezoelectric response was investigated. These results would be helpful for developing high temperature piezoelectric materials.

2. Experimental procedure

PIN–PMN–PT ternary ceramic with MPB composition, as shown in Fig. 1, were synthesized using two steps columbite precursor method [14]. The raw materials were oxide powders with purities better than 99.9%. The $MgNb_2O_6$ and $InNbO_4$ powder were synthesized at 1000 °C and 1100 °C, respectively for 6 h. The PbO, $MgNb_2O_6$, $InNbO_4$ and TiO_2 powders were wet mixed by ball milling with ZrO_2 ball for 5 h and then the mixed powder were calcined at 850 °C for 4 h. Upon milling, the various powders were pressed using PVA binder to form pellets 12 mm in diameter and about 1.2 mm in thickness. After burning out the PVA binder at 550 °C for 4 h, the samples were sintered at 1220 °C for 6 h. To compensate for PbO loss from the pellets, a PbO-rich atmosphere was maintained by placing an equimolar mixture of PbO and ZrO_2 inside the crucible.

The structure of the sintered samples was subsequently examined by X-ray diffraction (XRD RIGAKU D/MAX-2400). Silver paste was fired on both sides of the samples at 600 °C for 10 min. The temperature dependence of dielectric properties was measured by an HP 4284 LCR meter in conjunction with a furnace. The hysteresis loop was measured by TF analyzer 2000. Before piezoelectric experiments, the PIN–PMN–PT samples were poled at 40 kV/cm dc field at 130 °C for 10 min in silicon oil. The d_{33} value was measured using a piezo-d_{33} meter (ZJ-3A). The measurement of temperature and dc bias dependent-piezoelectric coefficient d_{33} is particular described in Ref. [15]. The temperature dependence of piezoelectric coefficient k_p was measured by an HP 4294 LCR meter in conjunction with a furnace.

* Corresponding author. Tel.: +86 29 82668679; fax: +86 29 82668794.
** Corresponding author. Tel.: +86 29 82668679; fax: +86 29 82668794.
E-mail addresses: ldbin2007@live.cn (D. Lin), zhrli@mail.xjtu.edu.cn (Z. Li).

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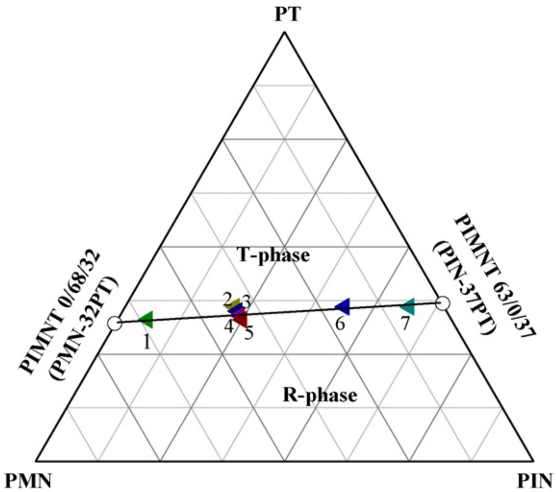

Fig. 1. Composition of samples in PIN–PMN–PT ceramic system.

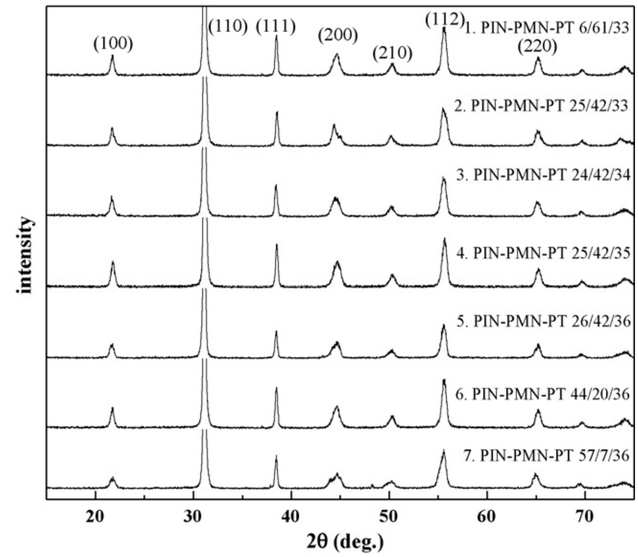

Fig. 2. XRD pattern of PIN–PMN–PT ceramics near MPB.

3. Results and discussion

XRD patterns of the PIN–PMN–PT ceramics with MPB compositions at room temperature are shown in Fig. 2 All of the patterns present pure perovskite phase without any trace of pyrochlore structure. The diffraction peak (2 0 0) shows a broden and no splitting peak, which indicates that the samples are in the MPB region. In addition, the micro-structure transitions of PIN–PMN–PT are induced from rhombohedral to tetragonal phase within the MPB region with increasing PT content.

Fig. 3 displays the SEM micrographs of the fracture surfaces of PIN–PMN–PT ceramics: (a) PIN–PMN–PT 25/42/33, (b) PIN–PMN–PT 24/42/34, (c) PIN–PMN–PT 23/42/35 and (d) PIN–PMN–PT 22/42/36. As shown in the figure, there is no presence of a second phase within the grains or at the grain boundaries. It is clearly evident that all samples are highly dense and their average grain size is about 1–3 μm.

The temperature dependence of the dielectric constant for PIN–PMN–PT ternary ceramics at 1 kHz is shown in Fig. 4. The Curie temperature varies from 165 °C to 293 °C in our choosing composition. As expected the phase transition temperatures (T_{max}) values show a strong compositional dependence within the MPB region. It suggests that T_{max} of the samples near MPB composition in PIN–PMN–PT ternary system can be adjusted. The detailed

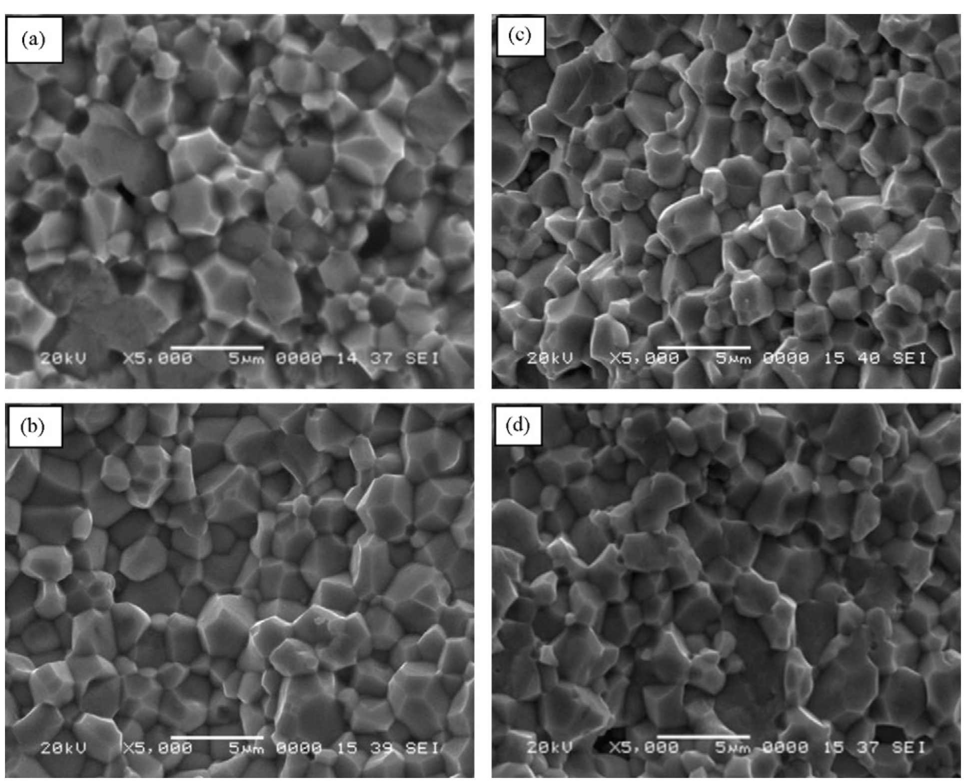

Fig. 3. Scanning electron micrographs of PIN–PMN–PT ceramics near MPB: (a) PIN–PMN–PT 25/42/33, (b) PIN–PMN–PT 24/42/34, (c) PIN–PMN–PT 23/42/35 and (d) PIN–PMN–PT 22/42/36.