High piezoelectricity after field cooling AC poling in temperature stable ternary single crystals manufactured by continuous-feeding Bridgman method

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Abstract: After field cooling (FC) alternating current poling (ACP), we investigated the dielectric and piezoelectric properties of $[001]_{\text{pc}}$ -oriented $0.24\text{Pb}(\text{In}_{1/2}\text{Nb}_{1/2})\text{O}_3$ (PIN)– $0.46\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$ (PMN)– 0.30PbTiO_3 (PT) (PIMN–0.30PT) single crystals (SCs), which were manufactured by continuous-feeding Bridgman (CF BM) within morphotropic phase boundary (MPB) region. By ACP with $4\text{ kV}_{\text{rms}}/\text{cm}$ from 100 to 70 °C, the PIMN–0.30PT SC attained high dielectric permittivity ($\epsilon_{33}^T/\epsilon_0$) of 8330, piezoelectric coefficient (d_{33}) of 2750 pC/N, bar mode electromechanical coupling factor k_{33} of 0.96 with higher phase change temperature (T_{pc}) of 103 °C, and high Curie temperature (T_{c}) of 180 °C. These values are the highest ever reported as PIMN– x PT SC system with $T_{\text{pc}} > 100$ °C. The enhancement of these properties is attributed to the induced low symmetry multi-phase supported by phase analysis. This work indicates that FC ACP is a smart and promising method to enhance piezoelectric properties of relaxor-PT ferroelectric SCs including PIMN– x PT, and provides a route to a wide range of piezoelectric device applications.

Keywords: field cooling (FC) alternating current poling (ACP); PIMN–0.30PT; high piezoelectricity

1 Introduction

Lead magnesium niobate $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$ (PMN)–

lead titanate PbTiO_3 (PT) based relaxor ferroelectric single crystals (SCs) have attracted extensive attention over the last 20 years, compared to the conventional polycrystalline lead zirconate titanate (PZT) ceramics [1–3]. Owing to their high piezoelectric properties as the key component of electromechanical devices, including medical imaging, actuators, underwater acoustic ultrasound, and nondestructive testing evaluation

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transducer, their excellent performance and applications have also promoted the rapid development of various growth methods of SCs. As a conventional relaxor-PT SC growth process, one charge Bridgman (OC BM) causes nonuniformity of TiO_2 concentration along growth direction due to the compositional segregation. Consequently, it leads to a large variation of dielectric and piezoelectric properties within a whole SC ingot [1]. In order to eliminate the compositional segregation, a continuous-feeding Bridgman (CF BM) method has developed in 2011 by Matsushita and Echizenya [4,5] of JFE Mineral Co., Ltd., Japan, which has been commercialized in the last decade. Until now, they have successfully grown high homogeneous binary PMN–0.30PT and ternary $\text{Pb}(\text{In}_{1/2}\text{Nb}_{1/2})\text{O}_3$ (PIN)–PMN– x PT SCs with large size of more than 80 mm in diameter and 300 mm in length by the CF BM method [5].

There is no doubt that electric poling is an essential process to obtain proper piezoelectric properties of relaxor-PT SCs. Electric field cooling (FC) direct current poling (DCP) for $\text{Pb}(\text{Zn}_{1/3}\text{Nb}_{2/3})\text{O}_3$ (PZN)–PT and PMN–PT SCs, by applying about 10 kV/cm from 200 °C (above Curie temperature (T_C) in cubic (C) phase) to room temperature (RT), has been studied to enhance the piezoelectric properties since 1982. These binary SCs, such as PZN–PT and PMN–PT, have relatively low Curie temperature ($T_C = 120\text{--}170$ °C) and low coercive field ($E_c = 2\text{--}3.5$ kV/cm). However, these behaviors limit applications in transducers indeed, and utilization scope would be further restricted by ferroelectric phase change temperature (T_{pc}) of T_{RM}/T_{MT} (R: rhombohedral; M: monoclinic; T: tetragonal), which occurs at a significantly lower level ($T_{pc} = 50\text{--}100$ °C) than T_C . Thus, SC systems with high T_{pc} and T_C for enhanced temperature usage range are desired to meet the thermal stability requirement which could avoid the influence of thermal generated by high-power electromechanical devices, such as sonar transducers. In order to obtain high electrical properties with high T_{pc} , T_C , and E_c , ternary SCs PIMN– x PT are regarded as the best composition [6].

In 2014, Yamamoto *et al.* [7,8] reported that dynamic alternating current poling (ACP) is an effective process to further enhance electrical properties of PMN–PT and PIMN–PT SCs compared with traditional static DCP process. Since then, many ACP studies, involving compositions, domain structure, orientations, transparency, and electric field condition for relaxor-PT SCs, have been conducted in last several

years [9–34]. Among them, the effect of ACP temperature dependence of PMN– x PT SC properties was investigated by some groups [13,26]. The close relationship between the optimal ACP temperature and performance was also revealed. Meanwhile, present authors reported high temperature ACP at 90 °C for 0.24PIN–0.46PMN–0.30PT (PIMN–0.30PT) CF BM SC and obtained $\varepsilon_{33}^T/\varepsilon_0 = 7000$, $d_{33} = 2340$ pC/N, and bar mode electromechanical coupling factor $k_{33} = 0.95$ [29]. Besides, Ma *et al.* [19] reported combination poling of DCP and ACP for 0.25PIN–0.43PMN–0.32PT OC BM SC and obtained $\varepsilon_{33}^T/\varepsilon_0 = 7210$, $d_{33} = 2610$ pC/N, with $T_{pc} = 114$ °C.

However, there is no report showing $\varepsilon_{33}^T/\varepsilon_0 > 8000$, $d_{33} > 2700$ pC/N, and $k_{33} > 0.95$ with above 100 °C T_{pc} of ternary SCs. The FC ACP for PMN–0.30PT and PIMN–0.26PT SCs was firstly investigated in our previous work, and also proved as an effective process for improving the electric performance and thermal stability of PMN-based SCs [25]. Taking into account the interaction among different factors in the ACP process due to dynamic phase transition during cooling, it is particularly important to achieve enhanced electric and thermal performance through optimizing the cooling zone and electric field conditions. Therefore, in this paper, we investigated effect of FC ACP process on CF BM PIMN–0.30PT SCs to improve electrical properties with higher T_{pc} .

2 Experiments

The PIMN–0.30PT SCs (JFE Mineral Co., Ltd., Chiba, Japan) grown along $[011]_{pc}$ direction manufactured by the CF BM method were used in this research. Pseudocubic perovskite indication was used in our work. These SC samples with a rectangular plate of (W)5.0 mm $\times$ (L)5.0 mm $\times$ (T)0.4 mm were prepared along $[001]_{pc}$ with 300 nm gold (Au) electrodes as shown in Fig. 1(a). Dynamic phase transition with decreasing temperature in FC ACP process is illustrated with diagram in Fig. 1(c). The FC poling process performed in air atmosphere without silicone oil [25], has an advantage of a free of organic solvent after poling of SC plates. In order to compare the ACP voltage and DCP voltage, we applied a root mean square (RMS) voltage per centimeter (V_{rms}/cm) as all electric field condition units. The term “rms” also called the effective or heating value of alternating current

Fig. 1 PIMN–0.30PT SCs manufactured by (a) CF BM method, possessing multi-phase structure located in morphotropic phase boundary (MPB) region nearby R phase, are illustrated in (b) ternary phase diagram [15]. The entire FC ACP system of the PIMN–0.30PT SCs, through (c) phase transition path of T→M→R during cooling, was performed in air atmosphere.

(AC), is equivalent to the direct current (DC) voltage and energy that would provide the same amount of heat generation in a resistor. The AC V_{rms} was calculated by multiplying 71% of the AC sine wave peak voltage (V_{pk}). All samples were kept for 10 min at RT after poling, and started the measurement of capacitance and loss by impedance analyzer (Agilent Technology, 4194 A, CA, USA) at 19–29 °C. Meanwhile, the temperature dependence coefficients (TDC) were also calculated. The free dielectric permittivity ($\epsilon_{33}^T/\epsilon_0$) and clamped dielectric permittivity ($\epsilon_{33}^S/\epsilon_0$) were calculated from the measured capacitance with a frequency at 1 kHz and at the two times of anti-resonant frequency ($2f_a$). Bar mode electromechanical coupling factor k_{33} was obtained from $\epsilon_{33}^T/\epsilon_0$ and $\epsilon_{33}^S/\epsilon_0$ according to Eq. (1) [6,15]:

$$k_{33} = \sqrt{\frac{\epsilon_{33}^T/\epsilon_0 - \epsilon_{33}^S/\epsilon_0}{\epsilon_{33}^T/\epsilon_0}} \quad (1)$$

The electromechanical coupling factor (k_i) of SC plate was calculated according to IEEE standards [35,36]. The d_{33} was measured at RT using piezo d_{33}/d_{31} meter (Institute of Automation, Chinese Academy of Sciences, ZJ-6B, China). Furthermore, temperature dependence of $\epsilon_{33}^T/\epsilon_0$ and dielectric loss

($\tan\delta$) for SCs were obtained by a computer-controlled LCR meter (Hioki E. E. Co., IM-3570, Nagano, Japan) at 1 kHz from RT to 250 °C.

3 Results and discussion

These material constants (MCs) of $\epsilon_{33}^T/\epsilon_0$, $\epsilon_{33}^S/\epsilon_0$, and d_{33} for PIMN–PT SCs have large TDC, as reported by Qiao *et al.* [37]. To compare the poling process effects of all MCs exactly, we applied MC₂₅ at 25.0 °C calculated in Eq. (2) according to Electronic Industries Alliance (EIA) RS 128 standard:

$$MC_{25} = \frac{MC_T}{1 + TDC \times (T - 25.0)} \quad (2)$$

where MC₂₅ is the material constant at 25.0 °C, MC_T is material constant at real measured temperature, and T is measured temperature. The actual measured TDC values of $\epsilon_{33}^T/\epsilon_0$, d_{33} , and $\epsilon_{33}^S/\epsilon_0$ of the PIMN–0.30PT SC were 8500, 8800, and 6700 ppm/°C, respectively.

Table 1 shows the dielectric and piezoelectric properties of the PIMN–0.30PT SCs poled by diverse temperature and electric field conditions. To facilitate comparison, all calculated MC₂₅ are shown in Table 1.

Table 1 Poling conditions and electrical properties of PIMN–0.30PT SCs

No.	Poling	Voltage ^a	Frequency (Hz)	Cycle	Temperature (°C)	$\varepsilon_{33}^T/\varepsilon_0$	$\varepsilon_{33}^S/\varepsilon_0$	Loss (%)	k_t (%)	k_{33} (%)	d_{33} (pC/N)
1	HT DCP	8	—	—	50	5110	600	0.39	59.0	95.8	1760
2 ^b	HT ACP	4	10	12	90	7000	680	0.59	59.3	95.0	2340
3	FC ACP	4	0.06	20	120–60	7270	680	0.77	55.2	95.2	2370
4	FC ACP	4	0.2	20	100–80	6720	650	0.86	57.7	95.0	2310
5	FC ACP	6	0.1	20	100–70	7940	660	0.54	55.9	95.8	2200
6	FC ACP	6	0.1	20	100–60	8040	800	0.89	58.3	94.9	2600
7	FC ACP	4	0.1	20	100–70	8330	660	0.33	58.2	96.0	2750

^aAll voltage units are kV_{rms}/cm.

^bNo. 2 is the result of previous published work [29].

No. 3 with lower frequency (0.06 Hz) applied 4 kV_{rms}/cm shows $\varepsilon_{33}^T/\varepsilon_0$ and d_{33} of 7270 and 2370 pC/N, which increased by 4% and 1%, respectively, slightly more than those of high temperature (HT) ACP (No. 2) with the same poling voltage. It reveals that FC ACP would enhance piezoelectric properties by applying AC field through dynamic multi-phase transition during cooling procedure. However, when FC poling temperature ranges from 100 to 80 °C with 0.2 Hz (No. 4), all properties decreased to the level ($\varepsilon_{33}^T/\varepsilon_0 = 6720$, $d_{33} = 2310$ pC/N) below that of No. 2. Therefore, we optimally adjusted the conditions, including increasing poling voltage to 6 kV_{rms}/cm, decreasing frequency to 0.1 Hz, and setting end temperature point at 70 °C. Dielectric properties, which continue to rise while temperature end point decreased to 60 °C (No. 6), increased significantly by 18% comparing with those of the former (No. 4). It indicates that high voltage ACP at low poling temperature is an effective method to enhance properties. Given that moderately increasing poling temperature could decrease the E_c , and thus effectively reduce the poling voltage [29], we reduced the poling voltage to 4 kV_{rms}/cm and changed the cooling region from 100 to 70 °C. Thus, the best properties ($\varepsilon_{33}^T/\varepsilon_0 = 8330$, $k_{33} = 0.96$) including the ultrahigh piezoelectricity ($d_{33} = 2750$ pC/N) were attained as shown in Fig. 2(a). The $\varepsilon_{33}^T/\varepsilon_0$ and d_{33} were enhanced by 19% and 18%, respectively, compared to those of HT ACP (No. 2), and $k_{33} = 0.96$ is the highest ever reported as PIMN–xPT SCs as authors know. The lowest $\varepsilon_{33}^T/\varepsilon_0 = 5110$ and $d_{33} = 1760$ pC/N for the same SC after HT DCP (No. 1) may be due to over poling by high voltage [26,34]. And comparison of $\varepsilon_{33}^T/\varepsilon_0$ and d_{33} with different poling conditions of PIMN–xPT SCs is illustrated in Fig. 2(b). Compared with HT DCP, PIMNT SC after the HT ACP obtains

Fig. 2 (a) Real measured value of the ultrahigh d_{33} and (b) comparison of $\varepsilon_{33}^T/\varepsilon_0$ and d_{33} among HT DCP, HT ACP [29], FC ACP, and DCP–ACT [19] of PIMN–xPT SCs.

about 33% increment of d_{33} which continues to grow by 18% after the FC ACP. Meanwhile, $\varepsilon_{33}^T/\varepsilon_0$ of the FC ACP was 17% higher than that of DCP–ACT (ACT: alternating current treatment) [19] possessing higher $T_{pc} = 114$ °C. Therefore, PIMN–0.30PT SC can achieve significant performance improvement through multi-phase transition by ACP during FC process.

Temperature dependence of $\varepsilon_{33}^T/\varepsilon_0$ and $\tan\delta$ of the sample No. 7 in Table 1 is performed as shown in Fig. 3. FC ACP SC has two distinct dielectric peaks at around 103 and 116 °C of $\varepsilon_{33}^T/\varepsilon_0$, the same as those of $\tan\delta$ before phase changing from T to C, which can be attributed to phase transitions of R to M (T_{RM}) and M to T (T_{MT}), respectively [10,25]. Furthermore, two

Fig. 3 Temperature dependence of (a) $\epsilon_{33}^T/\epsilon_0$ and (b) $\tan \delta$ of PIMN-0.30PT SCs poled by HT DCP, HT ACP [29], and the optimized FC ACP.

peaks of HT ACP and one peak of HT DCP before T_C point also proved the existence of M phase after ACP [26,29]. Compared with the HT ACP crystal, as shown in Fig. 3(b), three dielectric loss peaks appear at 104, 107, and 116 °C, which further reflects the induced phase path of $R \rightarrow MA \rightarrow MC \rightarrow T$ [19,38] by FC ACP. Thus, we speculated that there are two kinds of distinct phase change order between R and T phase transition [39,40]. Moreover, it is worth noting that FC ACP SC not only improves performance, but also has a significant increment of T_{pc} comparing with that of HT ACP ($T_{RM} = 78$ °C) [19,31]. Therefore, the greater electric and thermal performance improvement of PIMN-0.30PT SC is attributed to the low symmetry multi-phase constructed by phase changing through dynamic ACP cooling procedure.

These $\epsilon_{33}^T/\epsilon_0$ and d_{33} of various PMN-based relaxor SCs and their corresponding T_{pc} and T_C are shown in Figs. 4(a) and 4(b), respectively. We found that the electric properties are often in inverse proportion to the thermal properties among diversified SCs. It means that relaxor SCs with higher electrical properties usually cannot have higher thermal stability (T_{pc} above

Fig. 4 (a) $\epsilon_{33}^T/\epsilon_0$ and (b) d_{33} of various PMN-based relaxor SCs, and their corresponding T_{pc} and T_C [4,11,12,14,15,17,19–21,25,26,29,39,41,42].

100 °C). However, in this work, after FC ACP PIMN-0.30PT possesses higher T_{pc} of 103 °C and T_C of 180 °C while obtaining the highest $\epsilon_{33}^T/\epsilon_0$ and d_{33} among all binary and ternary PMN-based relaxor SC systems. Thus, the ultrahigh electric performance and thermal stability, these two major advantages, are both achieved in the PIMNT SC after FC ACP successfully.

To clarify the polarization mechanism clearly, we analyzed the difference between dynamic ACP and static DCP through contrast domain size and polar direction of multiple joints during electric poling, as shown in Fig. 5. We can see that initial polar direction is randomly distributed inside the unpoled SCs, as shown in Figs. 5(a) and 5(b). As poling electric voltage increasing, most of polar directions begin to arrange along one direction, as shown in Figs. 5(c) and 5(d). Since voltage direction of AC electric field dynamically reverses, the polar direction and domain structure would also be changed, as shown in Fig. 5(e). However, after certain duration, the domain structure does not change significantly inside SC because of the single

Fig. 5 Schematic images of (a), (c), (e), and (g) dynamic ACP and (b), (d), (f), and (h) static DCP processes; lateral phase of domain patterns observed on the fresh fracture of the ternary PMN-based SC after (i) ACP and (j) DCP. The PFM images are referred from He *et al.* [14].

direction and stable value of DC electric voltage, as shown in Fig. 5(f). On the contrary, the domain structure and polar direction of SC after ACP gradually tend to be orderly as poling direction reversal repeated several times, as shown in Fig. 5(g). And with applying periodic voltage, inner structures would also be changed by the external factors, such as frequencies and number of cycles [7,8,14,30,31]. In the end, a uniform stripe-like layer 109° domain structure ranging from 1 to 2 μm , similar to the piezoelectric force microscopy (PFM) pattern shown in Fig. 5(i), forms in ACP SC instead of the dispersed domain structure after DCP in Fig. 5(j) [7,14]. Moreover, outstanding properties, such as the ultrahigh piezoelectricity and transparency, are proved to be attributed to the extrinsic contribution of larger domain size, 109° domain wall, and periodic domain configuration [17,30]. Besides, dynamic polarization is easier to utilize reverse potential to eliminate the pinning effect generated in spontaneous polarization compared to static poling [28]. Thus, we consider that the ordering domain configurations with proper domain size also contribute to excellent electric and thermal performance of PIMNT SCs [31].

Based on the current compositions and T_{pc} after JFE Mineral Co. standard poling condition [4], we proposed a composition–temperature phase diagram of PIMN–xPT system manufactured by CF BM process.

As shown in Fig. 6(a), we update schematic phase diagram after ACP according to previous work [25,29]. The various monoclinic phases diffuse into the R and T regions, as shown in Figs. 6(a) and 6(b). Thus, MPB is not a narrow compositional region containing low symmetry phases any more, but an extremely broad region with co-existing phases significantly expands into the adjacent phase regions. As PT content increasing, the T_{C} increases linearly, whereas the T_{pc} after ACP slightly increases at first, and then T_{RM} and T_{MT} decrease to RT at higher PT composition (Fig. 6(b)). With further increment of PT, low symmetry phases act as a bridge between R and T in such a broad composition region with co-existing phases. It indicates a relationship among various ferroelectric phases with different polarization vectors. And polarization vectors rotate to the $[001]_{\text{pc}}$ direction in each domain under $[001]_{\text{pc}}$ electric field resulting in large piezoelectric distortion. These differences in the orientation within plane where the polarization lies, are often designated M_{A} , M_{B} , M_{C} , and orthorhombic (O), as shown in Fig. 6(c). This model is informed by X-ray diffraction under applied field, confirming that the phase transition under increasing applied fields transforms from approximately R, through low symmetry phases to the T [43]. In the M_{B} and O phases, the polarization vector lies within a $(100)_{\text{pc}}$ plane, while the polarization vectors of the M_{A} and M_{C} phases lie within $(110)_{\text{pc}}$

Fig. 6 Schematic phase diagram of CF BM PINMT system of (a) DCP and (b) ACP; (c) possible polarization vectors of low symmetry phases (M_A , M_B , M_C , and O), R and T phases in perovskite crystals, and polarization projection on the $(110)_{pc}$ plane; and (d) illustration of lattice deformations for the two paths of phase transition from RT to HT over T_c .

plane and $(010)_{pc}$ plane, respectively. To reveal the phase rotation behaviors clearly, we proposed the polarization projection on the $(110)_{pc}$ face. As illustrated in Fig. 6(c, bottom), the extinction angle is equivalent to the angle between the projection of spontaneous polarization on the $(110)_{pc}$ face and the $[001]_{pc}$ axis. Accordingly, polarization rotation path can be tracked in the $(110)_{pc}$ plane through domain observation by using polarizing microscope (PLM) with a crossed polarizer/analyzer (P/A) pair [39]. As reported by Bell *et al.* [40] and Fu *et al.* [43], on applying an increasing electric field to a $[001]_{pc}$ -poled SC, there are two polarization rotating paths from R and T phases via low symmetry. As shown in Fig. 6(d), top path is M_B switching to O within the $(100)_{pc}$ plane, and bottom path is M_A to M_C with the polarization rotation within $(110)_{pc}$ towards $(010)_{pc}$ plane. A small electric field will induce a large strain level (thus a large piezoelectricity) in low-field region, switching via the M_A and M_C phases is more feasible compared to the other continuous path due to the minimum energy during polarization rotation [43]. Thus, more characterization methods are necessary to

clarify the specific phase transition process and phase parameters in future research.

4 Conclusions

In this paper, we studied dielectric, piezoelectric, and electromechanical properties of $[001]_{pc}$ -oriented PIMN–0.30PT SCs manufactured by CF BM method in the MPB region nearby R structure after FC ACP. We found the optimal polarization conditions, 4 kVrms/cm, 0.1 Hz, and 20 cycles from 100 to 70 °C, and attained excellent properties, $\epsilon_{33}^T/\epsilon_0$ of 8330, d_{33} of 2750 pC/N, and k_{33} of 0.96 with higher T_{pc} of 103 °C. These values are the highest among all binary and ternary relaxor-PT SCs with $T_{pc} > 100$ °C. Optimizing FC conditions based on previous experiment, the ACP process improved the electrical performance through inducing the crucial low symmetry phases inside SCs by multi-phase transition during cooling process. While significantly improving the electric performance of the SCs, the FC ACP also increases the T_{pc} compared to HT ACP SCs. It makes wider temperature application

of the SC compared to binary SC PMN–PT. This work shows that FC ACP is a convenient and effective method to enhance piezoelectricity of relaxor-PT SCs including PIMN–xPT, and promotes the development of a wide range of piezoelectric device applications.

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