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To cite this article: Wei-Yi Chang , Ching-Chang Chung , Chengtao Luo , Taeyang Kim , Yohachi Yamashita , Jacob L. Jones & Xiaoning Jiang (2018) Dielectric and piezoelectric properties of 0.7 Pb(Mg_{1/3}Nb_{2/3})O₃-0.3 PbTiO₃ single crystal poled using alternating current, Materials Research Letters, 6:10, 537-544, DOI: [10.1080/21663831.2018.1498812](https://doi.org/10.1080/21663831.2018.1498812)

To link to this article: <https://doi.org/10.1080/21663831.2018.1498812>



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Published online: 24 Jul 2018.



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Dielectric and piezoelectric properties of 0.7 Pb(Mg_{1/3}Nb_{2/3})O₃-0.3 PbTiO₃ single crystal poled using alternating current

Wei-Yi Chang ^{1a}, Ching-Chang Chung ^{1b}, Chengtao Luo ^{1a}, Taeyang Kim ^{1a}, Yohachi Yamashita ^{1a}, Jacob L. Jones ^{2b} and Xiaoning Jiang ^a

^aDepartment of Mechanical and Aerospace Engineering, North Carolina State University, Raleigh, NC, USA; ^bDepartment of Materials Science and Engineering, North Carolina State University, Raleigh, NC, USA

ABSTRACT

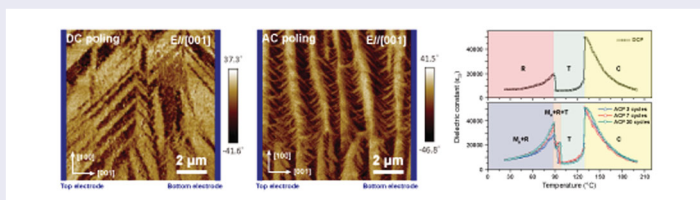
In this paper, 0.7Pb(Mg_{1/3}Nb_{2/3})O₃-0.3PbTiO₃ (PMN-30%PT) single crystal samples were poled using an alternating current (electric field) poling (ACP) method. Compared to the traditional poling method, the piezoelectric coefficient, free and clamped dielectric constants were improved more than 21%. X-ray diffraction result suggests the existence of monoclinic phase (*M_A*) in ACP samples and piezoresponse force microscopy (PFM) result further depicts the finer engineered domain structures. The ACP sample also showed the unique phase transition sequences during the depoling process. Our work could provide a novel domain engineered method to enhance piezoelectric properties of PMN-PT single crystal.

ARTICLE HISTORY

Received 20 March 2018

KEYWORDS

PMN-PT single crystal; Dielectric and piezoelectric properties; Field-induced phase transition; Aging rate



IMPACT STATEMENT

Piezoelectric and dielectric properties of relaxor-PT single crystals can be significantly enhanced by employing the new alternating current poling method, attributing to the unique heterogenous domain structure containing unprecedented domain wall density.

Introduction

Solid solution of (1-x)Pb(Mg_{1/3}Nb_{2/3})O₃-xPbTiO₃ (PMN-xPT) and the other relaxor ferroelectric single crystals have attracted intense research interest over the past decades because of their outstanding piezoelectric electromechanical properties, and the associated applications in sensors, actuators, and transducers [1–4]. In particular, their high piezoelectric coefficient and dielectric permittivity are critically important in advanced transducers [3,5,6]. There are several ways to manipulate dielectric and piezoelectric properties of PMN-xPT single crystal, such as modifying the compositions [7], applying nanocomposite electrodes [8,9], and utilizing different poling methods [9,10]. Among these methods, using different poling

methods has the greatest potential from its advantages of low cost and high efficiency. In general, direct current (electric field) poling (DCP) method is widely used in academia and industry [11]. DCP-based domain engineering study by Wada *et al.* revealed that piezoelectricity of domain engineered BaTiO₃ crystals can be enhanced from higher domain wall densities [12,13]. Recently, Yamamoto and Yamashita *et al.* reported an alternating current (electric field) poling (ACP) method that could further enhance the dielectric and piezoelectric properties of PMN-PT crystals [10,14]. While ACP may be a promising method for piezoelectric material properties enhancement, it has not been reported whether or not the domain engineering mechanism realized in DCP is similarly derived through the ACP,

CONTACT Xiaoning Jiang ✉ xjiang5@ncsu.edu Department of Mechanical and Aerospace Engineering, North Carolina State University, 3282 Engineering Building III, 911 Oval Dr. Raleigh, NC, 27606, USA

Supplemental data for this article can be accessed here. <https://doi.org/10.1080/21663831.2018.1498812>

and this question needs to be answered for crystal applications.

One important stability concern in the piezoelectric materials is aging, which can be observed in most of the ferroelectrics such as barium titanate (BaTiO_3) and lead zirconate titanate (PZT) [15–20]. Devonshire and Plessner described that aging is a gradual transformation from the free state to the clamped state, which means the domain boundary migrates from high energy polarizability to lower energy polarizability [15,21]. Aging strongly affects the application of ferroelectrics, and many investigations have been devoted to elucidating the aging phenomena in relaxor ferroelectrics [22,23]. However, there are only a few reports that mention the aging characteristics of PMN-PT single crystals [24], not mentioning the influence brought from the new ACP method.

This study reports the different poling conditions of DCP and ACP for PMN-30%PT single crystals, and their aging characteristics. The piezoelectric and dielectric properties of DCP and ACP crystal samples were measured. X-ray diffraction (XRD) was used to mark the field-induced phase transition from rhombohedral to monoclinic phase (M_A) in ACP samples, while piezoresponse force microscopy (PFM) was utilized to find the domain structures in ACP samples. The temperature dependent dielectric constant curves of both ACP and DCP samples were measured and their phase transition sequences during the depoling process were compared.

The [001]-grown PMN-30%PT single crystal (CTS Corporations, IL, USA) was cut, diced and polished to obtain samples with dimension of $9.2 \times 9.2 \times 0.6 \text{ mm}^3$. Titanium/gold electrodes (5/100 nm) were sputtered on the top and bottom surfaces of the samples. For the DCP samples, an external electric field (E-field) of 10 kV/cm was applied to pole the sample at room temperature for 30 min [11]. Its piezoelectric coefficients and dielectric constants were then measured using a Berlincourt-type d_{33} meter (YE2730A d_{33} meter, APC International Ltd., Mackeyville, PA, USA) and an LCR meter (KEYSIGHT technologies, E4980A, Santa Clara, CA, USA). The free dielectric permittivity (ϵ_{33}^T) and clamped dielectric permittivity (ϵ_{33}^S) were calculated from the measured capacitance by using an impedance analyzer (Agilent 4294A, Palo Alto, CA, USA) at 1 kHz and the frequency at two times of anti-resonant frequency ($2f_a$), respectively. The clamped dielectric permittivity was measured because of the fact that electrical impedance of a transducer is determined by ϵ_{33}^S and dimensions of the piezoelectric material [4]. For ACP samples, a bipolar triangular voltage with a designated frequency (e.g. $f = 1 \text{ Hz}$) was applied to the samples to generate an alternating E-field (e.g. with the peak-to-peak 10 kV/cm). The ACP samples were poled for up to 20 cycles or 20 seconds. After each

poling process, the piezoelectric coefficient, free (ϵ_{33}^T) and clamped (ϵ_{33}^S) dielectric permittivity were measured by the d_{33} meter and impedance analyzer mentioned earlier, respectively. Structure properties of PMN-30%PT single crystal with different poling conditions were studied using an X-ray diffractometer (SmartLab, Rigaku, Tokyo, Japan) with monochromatic $\text{CuK}\alpha$ radiation. The domain morphology was observed and characterized by a piezoresponse force microscopy, PFM (Dimension Icon, Bruker, Santa Barbara, CA, USA). The temperature dependent dielectric properties of PMN-PT single crystal were acquired by an LCR meter at every 1°C in a temperature range of 25°C to 200°C . During the measurement, the sample was heated in a silicone oil bath with a heating rate of $1^\circ\text{C}/\text{min}$ to ensure a homogeneous heating process. For repeating the experiments, the five samples were fully depoled in silicone oil at 250°C for 30 min, with top and bottom electrodes shorted during the depoling process. Each sample was tested three times to collect data.

The piezoelectric coefficients of PMN-30%PT single crystals with different poling conditions (ACP and DCP) are shown in Figure 1(a). The inset is an illustration showing the poling conditions for ACP and DCP methods. In this study, poling field is 10 kV/cm (peak to peak in ACP) and along [001] direction, and the ACP frequency is 1 Hz. The poling time is 30 min for DCP samples and less than 1 min (ACP-cycle dependent) for ACP samples. Five samples were prepared, and each sample was tested three times. The error bar in Figure 1 represents these measured data variations.

The applied E-field used in this study is 10 kV/cm. The reason why 10 kV/cm is chosen is first because it is about four times of the coercive field (E_c) of PMN-PT crystals ($\sim 2.3 \text{ kV/cm}$) [25] to make the completely poling, also the further test result (shown in Figure 1(d)) shows both ACP and DCP samples have highest dielectric constant at 10 kV/cm poling field. In Figure 1(a), the piezoelectric coefficient of the sample poled with the ACP method (ACP sample) is $2000 \pm 10 \text{ pC/N}$ which is 21% higher than the sample poled with the conventional DCP method (DCP sample), $1650 \pm 20 \text{ pC/N}$. It should be noted that high piezoelectric coefficients are observed in ACP samples which are poled for less than 1 min. The poling process of DCP samples takes 30 min with E-field of 10 kV/cm. With 2 alternative cycles (ACP-cycle) or 2 seconds, the ACP sample has exhibited a piezoelectric coefficient of $1850 \pm 10 \text{ pC/N}$, which is already higher than that of the DCP samples. Free and clamped dielectric constants of the PMN-30%PT single crystal with different poling conditions are shown in Figure 1(b). Free and clamped dielectric constants of ACP sample were 8500 ± 100 and 970 ± 70 , respectively. These values are

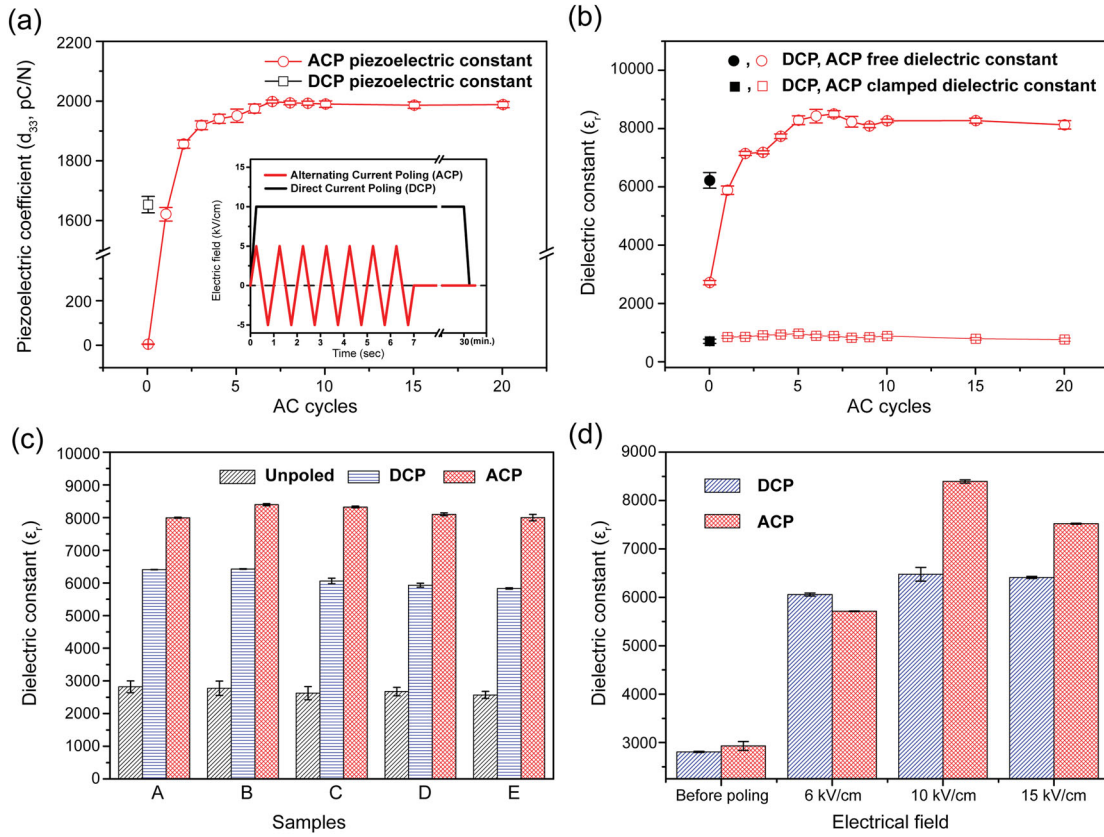


Figure 1. The piezoelectric and dielectric properties of ACP and DCP samples: (a) the piezoelectric coefficient of DCP and ACP with different number of cycles; the inset shows the poling electric field and duration time; (b) the free and clamped dielectric constant of DCP and ACP with different number of cycles; (c) the dielectric constant of five samples under unpoled, DCP and ACP conditions, where the error bar shows the deviation of three tests from each sample; (d) the dielectric constant of DCP and ACP samples at different poling electric fields.

larger than the DCP samples, which is 6120 ± 270 , and 720 ± 70 , respectively. Compared to the PMN-PT with DCP method, d_{33} , ϵ_{33}^T , and ϵ_{33}^S were improved 21%, 38% and 35%, respectively. Both Figure 1(a,b) also show that piezoelectric coefficient and dielectric constant of the ACP samples increase with the ACP-cycle up to 7 (or 7 seconds), and these values saturated after 7 cycles. The saturated piezoelectric coefficients and dielectric constants were at around 1980 ± 5 pC/N and 8140 ± 150 , respectively. To confirm the stability of the ACP method, Figure 1(c) shows good dielectric constant consistency ($< 10\%$) among five ACP samples, and the detailed standard deviations are provided in supplementary Table S1. Thus, the ACP single crystals showed clearly enhanced piezoelectric and dielectric properties over the DCP samples. In addition, electromechanical coupling coefficient (k_t) of ACP and DCP samples are calculated according to the IEEE standards, using the following equations [26].

$$k_t^2 = \frac{\pi}{2} \times \frac{f_r}{f_a} \times \tan \frac{\pi}{2} \frac{\Delta f}{f_a} \quad (1)$$

$$\Delta f = (f_a - f_r) \quad (2)$$

where f_r and f_a are the resonance and antiresonance frequency, respectively. The DCP and ACP samples both showed the similar coupling coefficient $k_t = 0.57$.

Figure 2. shows (400) peaks of PMN-30%PT single crystal with different poling conditions. XRD reference patterns are also listed in the bottom of the Figure 2. According to the phase diagram and electric field-temperature diagram [27,28], the field-induced phase transition for PMN-30%PT single crystal under the electric field along [001] direction will be rhombohedral (R) $\rightarrow$ monoclinic (M_A) $\rightarrow$ monoclinic (M_C) $\rightarrow$ tetragonal (T). (The notation M_A/M_C follows definitions of Vanderbilt and Cohen [29]). Thus, the unpoled sample peak marks the R phase. Since the peak center of the DCP sample is almost at the same 2θ position as the unpoled sample peak, it shows that DCP sample is still in the same R phase. In contrast, the peak center of ACP is obviously shifted, showing the likely existence of an M_A phase based on the reference pattern.

In addition to the average crystal structure characterization from the XRD result, the local domain structure was observed from the PFM result. Figure 3 shows the different domain structure from samples unpoled

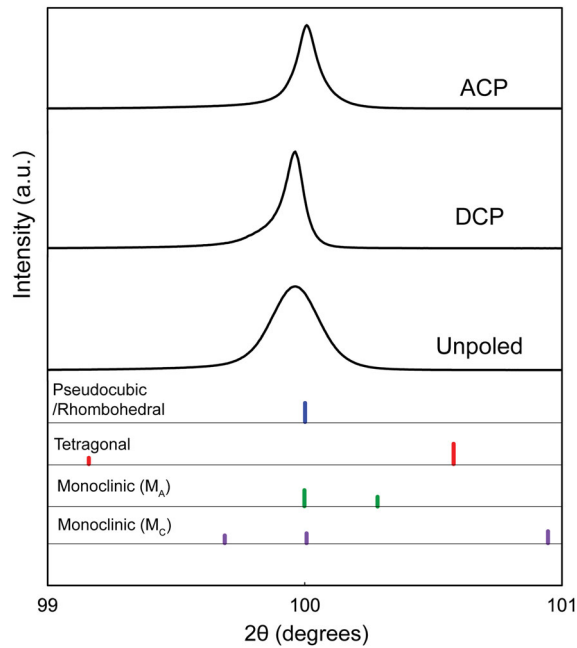


Figure 2. The XRD (400) reflection of PMN-PT single crystal resulting from different poling methods, including ACP, DCP and unpoled sample in the room temperature. The peak intensities have been normalized and the reference patterns are shown in the bottom of the figure.

and poled using DCP and ACP methods. Note that the scanned plane is (010) plane and electric field is along the horizontal [001] direction as it is marked. The original unpoled sample in Figure 3(a) hardly provides any domain information since the phase signal is weak and its amplitude followed the surface topography ($R_q < 10$ nm). After poling, new R phase domains were formed and both [101]-oriented 71° domain wall and [100]-oriented 109° domain wall from R phase domains can be observed in Figure 3(b,c). Comparingly, M phase observed in XRD is not shown in PFM results and it may be related to the skin effect [30,31]. Figure 3(b) shows clear lamellar domain structures with uniform 71° domain walls (920 ± 80 nm in width)

and non-uniform 109° domain walls (2100 ± 770 nm in width), which are engineered by conventional DCP method [32]. In Figure 3(c), the ACP sample depicted different R phase domain with backswitched domain structure. First, the vertical 109° domain wall becomes more uniform (700 ± 10 nm in width). Secondly, the 71° domain wall density (160 nm in width) is significantly increased compared with the DCP sample. Additionally, in Figure 3(c), these tiny 71° domain walls have two orientations, [101] and [10-1], between two 109° domain walls, while there is only one orientation in DCP sample shown in Figure 3(b). The supplementary PFM result, provided in Fig. S1, proves these 71° domains in ACP sample share the same in-plane $<100>$ polarization component. Thus, if the unswitched (same as DCP) $<001>$ polarization component is marked as [001] and the backswitched one is marked as [00-1], the rhombohedral $<111>$ polarizations of 71° domains can be marked as unswitched ones along [111]/[1-11] and backswitched ones along [11-1]/[1-1-1], where their $<010>$ polarization component is the out-plane PFM result shown in Figure 3. Under the ACP electric field, the domain wall nuclear, leading to the growth of both unswitched and backswitched domain walls. As a comparison, such backswitched domain structure is not observed in the DCP samples. Since the AC field used in ACP is amplitude symmetric (± 5 kV/cm), it may cause the backswitched 71° domain walls having the similar length and width to those unswitched ones. Generally, the domain structures in ACP sample are more uniform and it is corresponding to the lower full width half maximum (FWHM) in the XRD result shown in Figure 2. From the view of domain engineering, the significant domain wall density increase in ACP samples is likely the origin of the piezoelectric property enhancement.

As it is discussed in XRD results above, DCP samples remain R phase and ACP samples experience $R \rightarrow M_A$ field-induced phase transition. Here, engineered ACP domain structure still demonstrates R phase domain

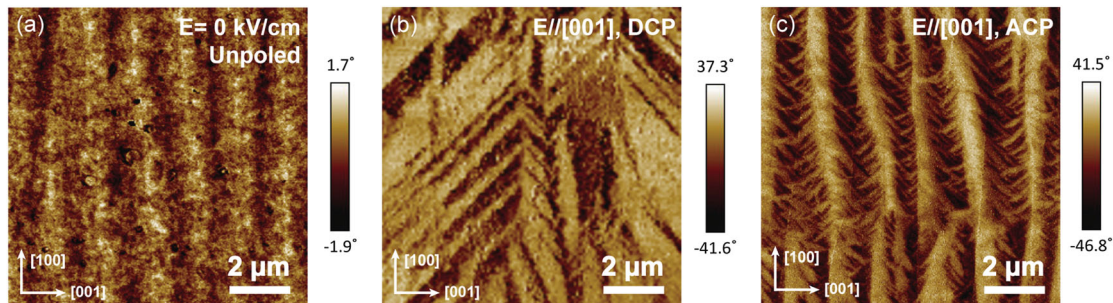


Figure 3. The PFM images of domain morphology for PMN-30%PT in different poling conditions: (a) unpoled ($E = 0$ kV/cm), (b) using DCP method, (c) using ACP method (1 Hz, 7 cycles). The poling field is 10 kV/cm and along [001] direction. The scanned plane is (010) cross section plane.

configuration (4R)[3], showing that such phase transition was not complete, and the phase transition caused by ACP is $R \rightarrow M_A + R$. The dielectric and piezoelectric properties in ferroelectrics are related to both intrinsic and extrinsic factors. In our work, ACP is proved to engineer the PMN-30PT in both ways. XRD measurement shows the intrinsic lattice symmetry is affected by the existence of M_A phase and PFM measurement shows the extrinsic domain wall density is increased significantly. Both factors lead to the outstanding dielectric and piezoelectric properties after ACP [33–35].

Further temperature-dependent dielectric constant measurement result is shown in Figure 4(a,b). Before the phase transition, the ACP samples poled with 20 AC cycles has a maximum dielectric constant value ($\epsilon_{33,max} \sim 40,000$), which is much higher than the DCP sample ($\epsilon_{33,max} \sim 20,000$) at the same temperature. It has been reported that the poled rhombohedral PMN-x%PT has a phase transition sequence of $R \rightarrow T \rightarrow C$ during the zero field heating [36]. In this work, the similar dielectric constant curve in Figure 4(a) shows the phase transition temperatures of DCP PMN-30%PT single crystal are 90°C (363 K) for $R \rightarrow T$ and 130°C (403 K) for $T \rightarrow C$. Comparingly, ACP samples have the same phase transition temperature near 90°C and 130°C, while extra anomalies of dielectric constant drops at 92°C $\sim$ 96°C (365 K $\sim$ 369 K) are shown in dielectric constant curves of ACP samples, as shown in Figure 4(b). It demonstrates that the different poling conditions lead to the different phase transition sequences during the process of depoling.

As the XRD and PFM results discussed above, in ACP samples $M_A + R$ phase exists at room temperature. So, the extra anomalies of dielectric constant appear between the $M_A + R$ and T phase. Additional in-situ XRD tracking on (004) peak was made to further study this unique phase transition sequence, and the ACP sample poled with 7 AC cycle was measured in the condition of zero-field heating. The in-situ XRD results with temperatures around the appearance of dielectric constant anomalies are shown in Figure 4(c), while the result with whole temperature range (from 25°C to 250°C) is provided in Fig. S2. As the arrows pointing out, both T phase (004) and R phase (004) are observable and indicate the existence of phase coexistence. At the first phase transition temperature around 90°C, the T phase (004) peak begins to rise and R phase peak drops a little bit. While at the temperature between 92°C and 96°C, T phase (004) peak promptly rises and R phase peak disappears at the same time. In conclusion, the dielectric constant for ACP samples dropings at 90°C and 92°C to 96°C are caused by the appearance of T phase and the completely transformation of T phase, respectively. In the meantime, Figure 4(c) cannot

determine the existence of M_A phase at this temperature range, since the M_A phase peak is covered by the intense new T phase peak. Thus, the phase transition sequence of ACP samples should be $M_A + R \rightarrow M_A + R + T \rightarrow T \rightarrow C$ (used in Figure 4(b)) or $M_A + R \rightarrow R(M_A) + T \rightarrow T \rightarrow C$ and such a phase coexistence state is not shown in DCP sample. In addition, Figure 4(b) shows that the temperatures of extra anomalies ($M_A + R + T \rightarrow T$ phase transition) are ACP-cycle-number-dependent. In other words, ACP sample with higher ACP-cycle-number has higher $M_A + R + T \rightarrow T$ phase transition temperature. And the little difference between 7-cycle and 20-cycle shows such increment is also limited. Interestingly, the lower $M_A + R \rightarrow M_A + R + T$ phase transition temperatures are ACP-cycle-number-independent, which is also shown in Figure 4(b). According to the Landau theory[37,38], the split of the T phase transition temperature indicates that the bulk free energy of R (or $R + M_A$) phase is increased nonuniformly after ACP. In details, the unchanged $M_A + R \rightarrow M_A + R + T$ phase transition temperatures between ACP and DCP samples indicate that some part of the ACP sample is not influenced by the backswitching field of ACP, so free energy change is the same as DCP sample. On the other hand, the higher $M_A + R + T \rightarrow T$ phase transition temperature from ACP sample indicates that the free energy from the rest part of ACP sample is higher than the level of DCP sample and higher ACP-cycle-number can help ACP sample approaching the free energy maximum.

Considering the new domain structure shown in PFM result, the property stability of such phase coexistence heterogenous structure was tested by the following aging experiment. In the experiment, the aging characteristics of the piezoelectric coefficient and dielectric constant were collected right after the poling process and after a certain period of time at room temperature. The properties of aging data were measured using a d_{33} meter and an impedance analyzer at room temperature. Figure 5(a) shows the aging characteristics of the piezoelectric coefficient with different poling methods. The aging behaviors of poled PMN-30%PT single crystals follow the logarithmic law similar to BaTiO₃ and PZT [39,40], and the aging equation can be written as follows.

$$P_t = P_i - A_p \log(t) \quad (3)$$

where P_t is the piezoelectric or dielectric constant after aging time t , P_i is the initial value of the property. A_p is the absolute aging rate which is the absolute decrease for one decade. The relative aging rate is expressed as

$$R_p = \frac{A_p}{P_i} \times 100 \quad (\%/decade) \quad (4)$$

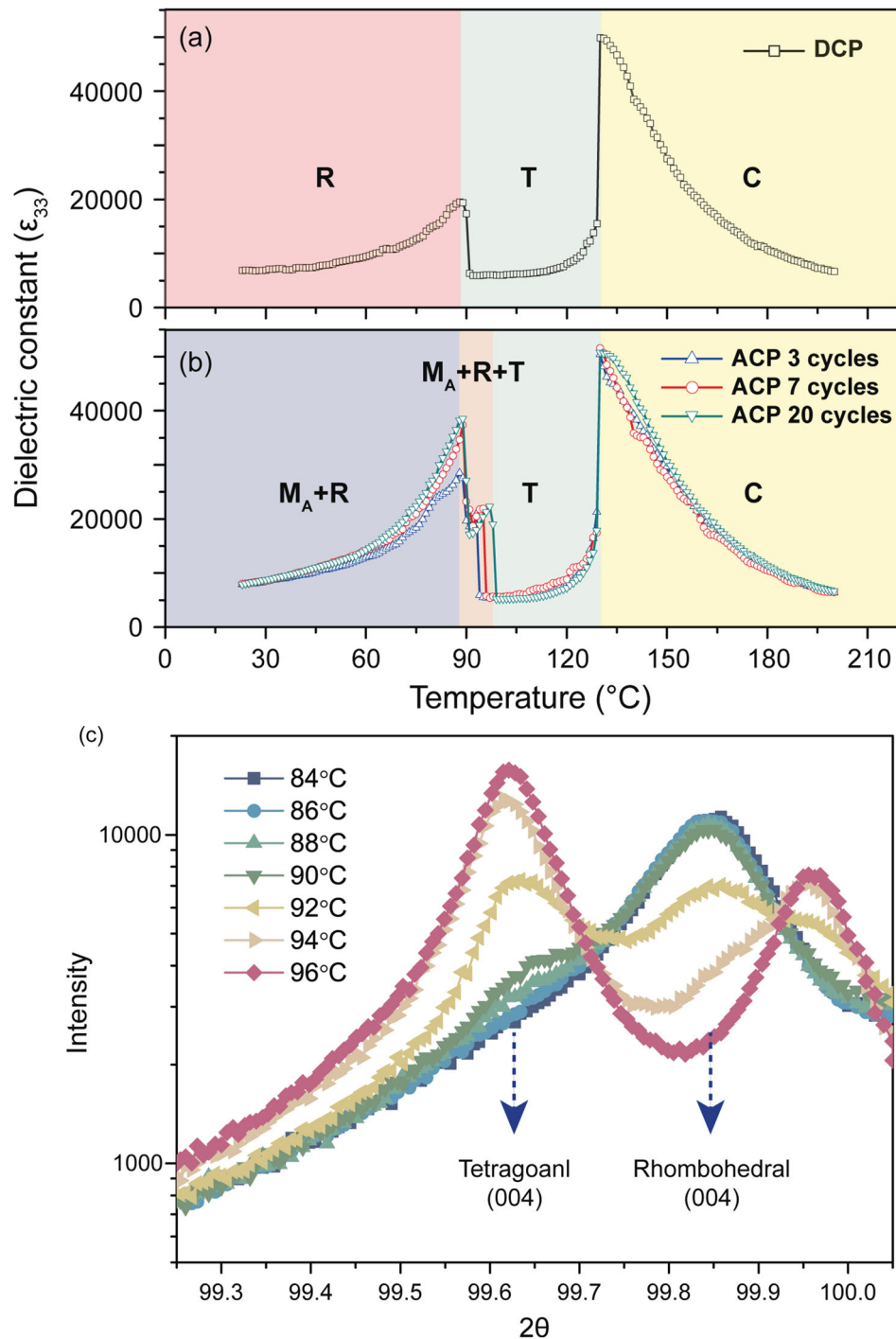


Figure 4. Dielectric constant as function of temperature for PMN-PT single crystal after (a) DCP and (b) ACP processes (including 3 cycles, 7 cycles, and 20 cycles for the ACP process). (c) (004) peaks showing *T* and *R* phase coexistence during the phase transition near 90°C in ACP sample (7 cycles).

It was observed that the piezoelectric aging of the ACP and DCP samples follow linear, logarithmic time dependence with aging rate (R_p) of 0.5%/decade and 0.6%/decade, respectively. The aging of free and clamped dielectric constant is shown in Figure 5(b). The aging rate of free dielectric constant around 1.3 ~ 2.0%/decade by DCP and ACP method, which is similar to the Mn-doped

Pb(In_{1/2}Nb_{1/2})O₃-Pb(Mg_{1/3}Nb_{2/3})O₃-PbTiO₃ (~1.5%/decade) with the dielectric constant of 4000 [41]. The ACP and DCP samples also have a similar aging rate of clamped dielectric constant. Therefore, the ACP method can be used to enhance the dielectric and piezoelectric properties with reduced time of poling process without any aging concerns.

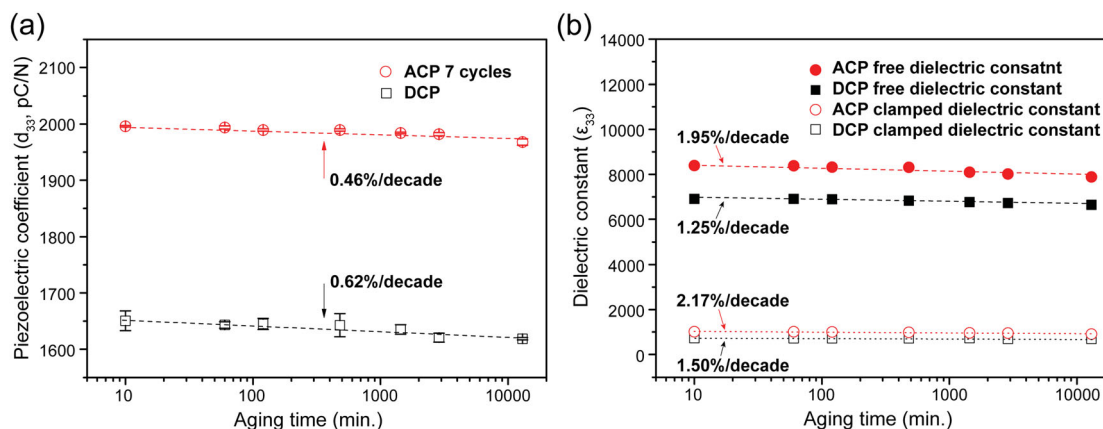


Figure 5. The aging rate of DCP and ACP. (a) The aging rate of piezoelectric coefficient, (b) The aging rate of free and clamped dielectric constant.

In summary, the ACP single crystal samples (10 kV/cm alternating current E-field with 1 Hz 7 cycles) exhibit a higher piezoelectric coefficient of 2000 ± 10 pC/N and dielectric constant of 8500 ± 100 that those of their DCP counterparts (continuities 10 kV/cm E-field for 30 min) at the room temperature. Further XRD and PFM results depict that ACP method engineers the lattice symmetry and domain structure at the same time. The enhanced piezoelectric coefficient and dielectric constant in ACP samples may originate from the appearance of field-induced M_A phase. Also, the finer R phase domain structures generated by ACP method provide the other important origin of the better piezoelectric performance from the view of domain engineering. Based on these XRD and PFM results, the unique phase transition sequences of ACP samples are marked in the temperature dependent dielectric constant measurement. In addition, the aging rate of the piezoelectric coefficient and dielectric constants of the ACP samples are 0.5%/decade and 2.0%/decade, respectively. Thus, the reported ACP method exhibits a great potential to enhance the piezoelectric and dielectric properties of relaxor-PT single crystals without aging concerns.

Acknowledgments

This work was primarily supported by ONR under Grant # N00014-15-1-2418. This work was performed in part at the Analytical Instrumentation Facility (AIF) at North Carolina State University, which is supported by the State of North Carolina and the National Science Foundation (award number ECCS-1542015). The AIF is a member of the North Carolina Research Triangle Nanotechnology Network (RTNN), a site in the National Nanotechnology Coordinated Infrastructure (NNCI).

Disclosure statement

No potential conflict of interest was reported by the authors.

Funding

This work was supported by National Science Foundation: [Grant Number ECCS-1542015]; Office of Naval Research: [Grant Number N00014-15-1-2418].

ORCID

Wei-Yi Chang <http://orcid.org/0000-0003-1000-8121>
 Ching-Chang Chung <http://orcid.org/0000-0002-6490-6827>
 Chengtao Luo <http://orcid.org/0000-0003-1207-2960>
 Taeyang Kim <http://orcid.org/0000-0002-8162-2973>
 Yohachi Yamashita <http://orcid.org/0000-0003-4432-646X>

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