Journal of the Korean Ceramic Society (2024) 61:854–860
https://doi.org/10.1007/s43207-024-00390-0

ORIGINAL ARTICLE

Enhanced polarization retention and softening in [001]-oriented $Pb(Mg_{1/3}Nb_{2/3})$-$PbTiO_3$ single crystals through corona poling

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Received: 8 November 2023 / Revised: 27 February 2024 / Accepted: 14 March 2024 / Published online: 15 April 2024

Abstract
This study investigates the electrical properties of $Pb(Mg_{1/3}Nb_{2/3})$-$PbTiO_3$ (PMN-PT) single crystals subjected to corona poling (CorP) compared to direct current poling (DCP) and alternating current poling (ACP) methods. The results revealed the superiority of CorP in terms of polarization retention and softening. The corona-poled sample demonstrated a higher depolarization temperature (T_d ~ 100 ℃) than DCP or ACP methods (T_d ~ 90 ℃), indicating improved polarization stability at elevated temperatures. Furthermore, lowering of the coercive field (E_C) in CorP samples suggests CorP makes the materials electrically softer. These advantages stem from the noncontact nature of the CorP method, which minimizes the risk of localized dielectric breakdown, and ensures a uniform electric field distribution. This work sheds a light on the potential of CorP as a promising technique for enhancing the electrical performance of materials in piezoelectric applications.

Keywords Corona poling · Polarization retention · Depolarization temperature · Coercive field

1 Introduction

Piezoelectric materials play a critical role in a variety of applications such as sensors, actuators, transducers, and energy-harvesting devices [1, 2]. Among these materials, relaxor ferroelectric single crystals such as $Pb(Mg_{1/3}Nb_{2/3})$-$PbTiO_3$ (PMN-PT) or $Pb(Zn_{1/3}Nb_{2/3})$-$PbTiO_3$ (PZN-PT) single crystals exhibit exceptional piezoelectric coefficient (d_{33} > 1500 pC/N) and electromechanical coupling factor (k_{33} ~ 0.90) to make them highly desirable for advanced piezoelectric applications [3–9]. Recently, it has been revealed that all the functional properties of these crystals can be further enhanced only by properly chosen poling technique, meaning that the impact of poling treatment on the functional properties needs systematic investigation [10]. Conventional poling techniques, such as direct current poling (DCP) and alternating current poling (ACP) methods, have been widely used to induce the polarization in ferroelectric single crystals [11–13]. Nevertheless, these techniques often have limitations, including direct contact poling, which can lead to the occurrence of localized dielectric breakdown and degradation of material performance [14]. Therefore, the exploration of feasible poling techniques that can address these limitations and enhance the piezoelectric properties of PMN-PT single crystals is of great interest. It is interesting to note that corona poling (CorP) offers several advantages, such as noncontact poling and inducing uniform electric field distribution [15]. These advantages minimize the risk of localized breakdown and ensure uniform distribution of applied electric field strength in comparison to DCP and ACP methods [16]. The CorP technique involves applying an electric field generated by corona discharging onto the unelectroded surface of the sample, inducing polarization alignment of the ferroelectric domains [17, 18].

In this study, we systematically investigated and compared the effects of various poling techniques, including DCP, ACP, DCP followed by ACP, ACP followed by DCP, as well as CorP, on the dielectric and piezoelectric properties

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of PMN-PT single crystals. Each poling technique was conducted at both room temperature (– 25 ℃) and elevated temperature (– 90 ℃) to comprehensively understand the impact of poling temperature on the poling process. By analyzing the ferroelectric properties, such as polarization–electric field hysteresis loops, dielectric constant, depolarization temperature, and loss tangent, we aim to gain insights into the advantages and limitations of different poling techniques. The results from this study will contribute to the optimization of poling processes for PMN-PT single crystals, enabling improved performance and enhanced utilization in various piezoelectric applications. Considering the advantages of CorP including its noncontact nature and uniform electric field distribution, CorP could be a promising technique for achieving improved polarization and alignment in relaxor-PT single crystals.

2 Materials and methods

The [001]-oriented rhombohedral PMN-29PT single crystal was prepared by the Bridgman method obtained from iBULe Photonics Co. Ltd. (Incheon, South Korea). The crystal sample was cut into the 4 mm × 4 mm × 0.5 mm along the growth direction from the grown crystal boule. The two opposite sides of the crystal surface perpendicular to the [001] direction were gold coated via sputtering and subsequently fired at 300 ℃ for 30 min in the air. Figure 1a shows the configuration of conventional poling methods for ACP and DCP. The crystal sample was subjected to an electric field in a silicon oil bath within a heating mantle. The electric field was applied using a commercial apparatus, aixPES (aixACCT Systems GmbH 2013, Germany), at both room temperature (25 ℃) and elevated temperature (90 ℃). For CorP, as illustrated in Fig. 1b, the sample with surface electrode was exposed to a corona needle connected to a power supply (HAR-30P1, MATSUSADA, Otsu, Japan). We supposed no degradation of the sample after CorP based on previous results that showed approximately the same d_{33} whether the ferroelectric materials were with or without surface electrode [18]. When the high voltage is applied to the corona needle, the air is discharged, creating an electric field between the corona needle (top) and the conductive plate (bottom). The distance between the corona needle and the conductive plate was 3 cm, and the voltage was treated at 10 kV for 10 min.

Figure 2 depicts the waveform of electric field against time for four different poling techniques. For DCP, a constant electric field is applied at 1 kV/mm for 1 min as shown in Fig. 2a. In Fig. 2b, the waveform exhibits an oscillating pattern, representing the alternating nature of the applied electric field for ACP. We selected the poling condition of ± 1 kV/mm, triangular waveform, 1 Hz, and three cycles because relaxor-PT single crystals show similar piezoelectric coefficient and dielectric permittivity after three cycles of AC electric field in the previous study [19]. In the case of DCP followed by ACP (DCP@ACP), the waveform starts with a constant electric field and is then switched to an AC field with the same frequency and amplitude as the ACP waveform in Fig. 2c. Conversely, in ACP followed by DCP (ACP@DCP) waveform, an AC field is initially applied, and is changed to a constant electric field with the same amplitude as shown in Fig. 2d. To prevent the influence of sample composition, only one PMN-29PT single crystal was used for the poling experiments. We first used the pristine sample, poled the sample with specific poling technique, and finally depoled the sample with thermal annealing at 250 ℃ for 30 min.

Temperature-dependent dielectric properties, such as dielectric permittivity (ε_r) and dielectric loss (tanδ), were measured at 1 kHz using a novocontrol broadband dielectric spectrometer (Novocontrol Technology GmbH & Co. KG, Hundsangen, Germany) equipped with a temperature controller over the temperature range from 25 ℃ to 200 ℃

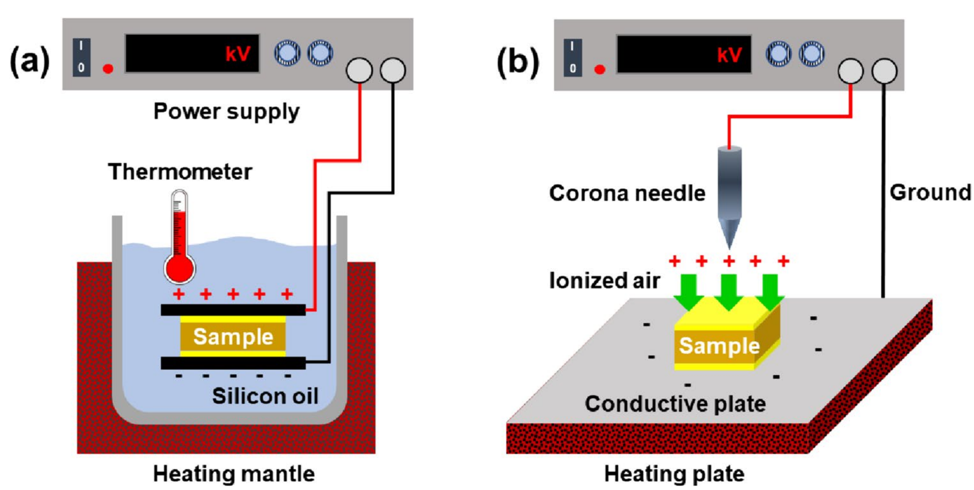

Fig. 1 Experimental configuration. **a** Conventional poling methods for DCP and ACP with gold-coated surfaces of the PMN-PT single crystal can be subjected to an electric field in a silicon oil bath using a heating mantle. **b** CorP where the sample was placed on a heating plate with a conductive layer, and the sample was exposed to a corona needle connected to the voltage source

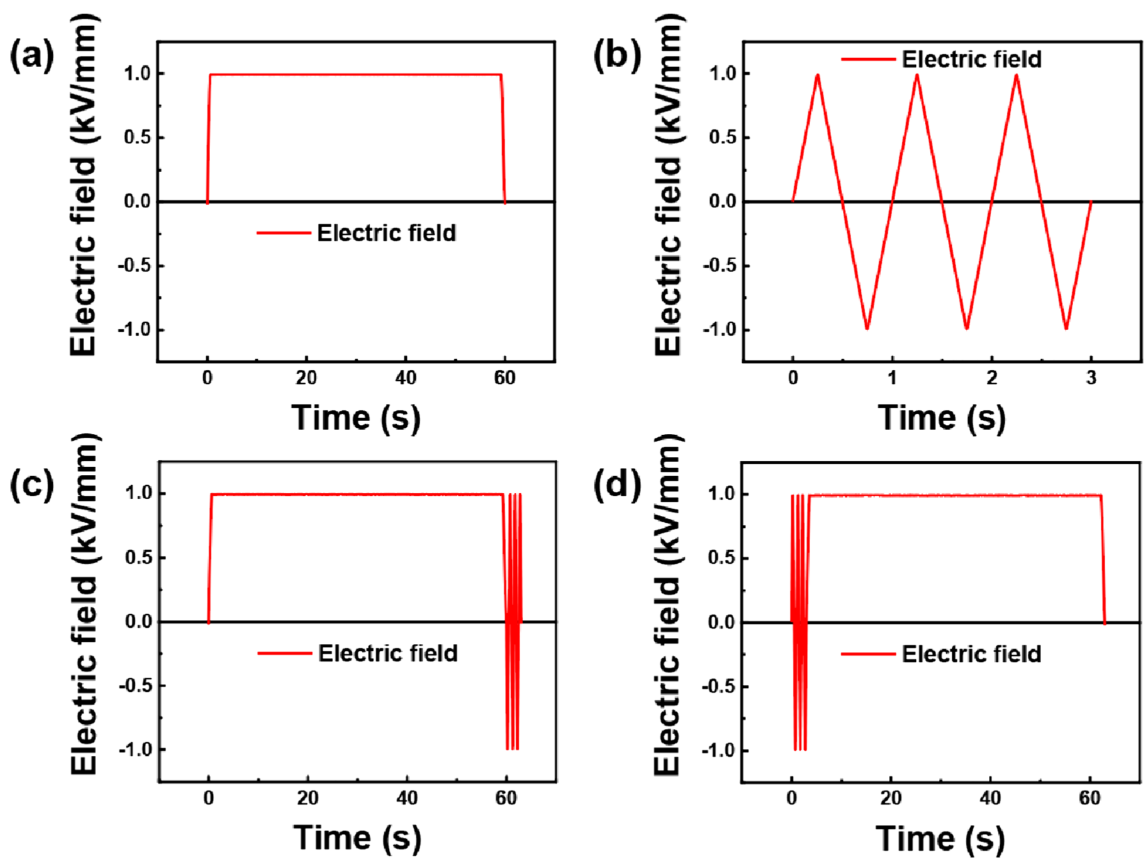

Fig. 2 Waveform of the electric field against time for four different poling techniques; **a** DCP, **b** ACP, **c** DCP followed by ACP (DCP@ACP), and **d** ACP followed by DCP (ACP@DCP)

at the heating rate of 3 ℃/min. The depolarization temperature (T_d,) was set in a first peak of tanδ from the temperature-dependent dielectric properties [20]. The piezoelectric coefficient (d_{33}) was measured using a commercial Berlincourt-type d_{33} meter (YE2730A, SINOCERA, Yangzhou, China). The impedance analyses were carried out to measure the electromechanical coupling factor (k_t) and mechanical quality factor (Q_m) using a multi-frequency LCR meter (HP 4194A, Agilent Technologies, Santa Clara, USA) with an Agilent 16034E test fixture. The polarization–electric field (P-E) loop and current–electric field (I-E) curve were measured with the poled samples at a condition of ± 1 kV/mm, 1 Hz, and 25 ℃ using the aixPES.

3 Results and discussion

3.1 Temperature-dependent dielectric properties

The temperature-dependent permittivity and tanδ for different poling techniques at 25 ℃ and 90 ℃ were examined in Fig. 3. As shown in Fig. 3a–e, there were significant fluctuations in the permittivity and tanδ curves depending on the poling conditions. This suggests that different poling methods lead to variations in the electrical properties of the PMN-PT single crystals. Each poling technique, *i.e.*, DCP, ACP, DCP@ACP, ACP@DCP, and CorP, might apply distinct electrical fields or stresses to the ferroelectric material, inducing different degrees of alignment or orientation within the internal structure [21, 22]. On the other hand, Fig. 3f-(j) indicates that the fluctuations in permittivity and tanδ curves are relatively small regardless of the specific poling methods except for the case of CorP where the permittivity at 25 ℃ slightly dropped as shown in Fig. 3j and Fig. 4a. It is noteworthy that the depolarization temperature (T_d) decreased when poling at 90 ℃, denoted by Poling(90), compared to poling at 25 ℃, denoted by Poling(25). These results indicate that the temperature at which the poling process was carried out plays a crucial role in minimizing variations in the electrical properties due to variations in poling methods [23, 24].

3.2 Poling-dependent electrical properties

Of particular importance, among all the poling methods at 90 ℃, the corona-poled sample exhibited a higher T_d, of approximately 100 ℃, whereas the other poling methods

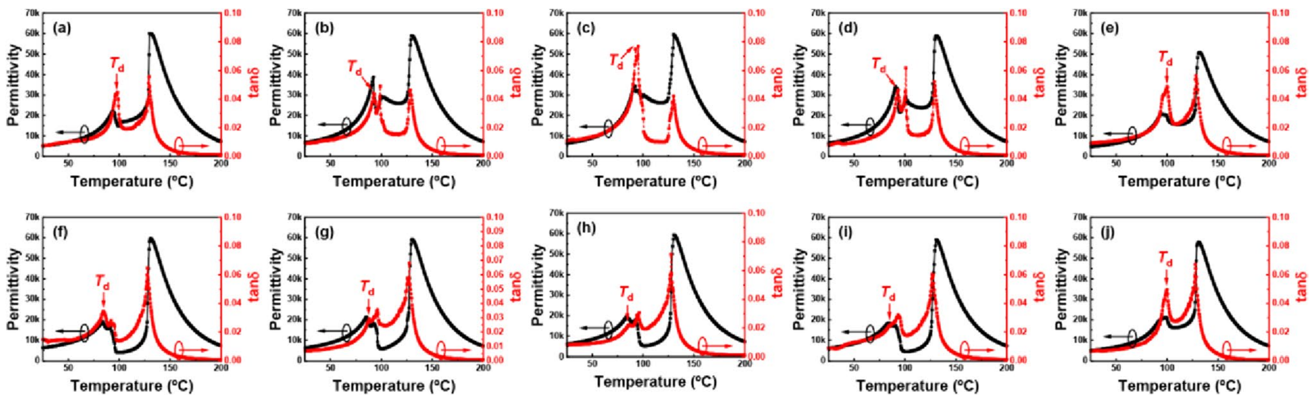

Fig. 3 Temperature-dependent dielectric properties for different poling methods, including dielectric permittivity and tanδ of **a**, **f** DCP, **b**, **g** ACP, **c**, **h** DCP@ACP, **d**, **i** ACP@DCP, and **e**, **j** CorP at **a–e** 25 ℃ and **f–j** 90 ℃, respectively

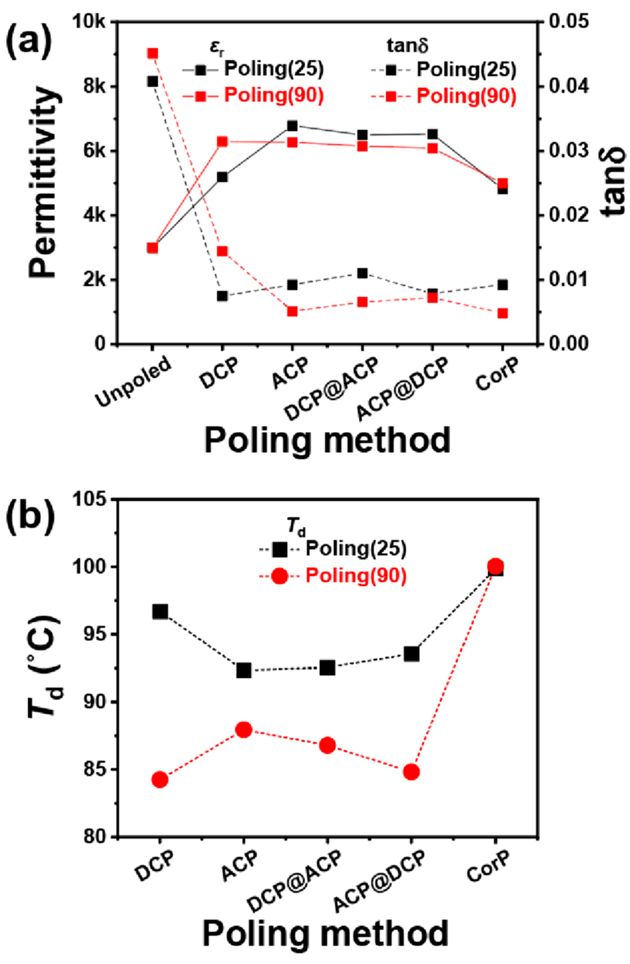

Fig. 4 Dependence of poling method and poling temperature with **a** dielectric permittivity and tan*δ* at 1 kHz and 25 ℃ and **b** depolarization temperature (*T*d) which was set in a first peak of tan*δ* from Fig. 3

showed a lower T_d below 90 ℃ as shown in Fig. 4(b). The higher T_d value for the corona-poled sample suggested that CorP is more effective in maintaining polarization at elevated temperatures. The higher T_d observed in the CorP compared to other poling methods could be attributed to several factors. First, the CorP method might result in enhanced polarization retention within the material, leading to a thermally stable polarization [25]. This improved polarization retention will contribute to a higher T_d by effectively resisting depolarization at elevated temperatures. Second, the CorP technique may have introduced a more robust structural configuration within the material [26]. This increased structural stability could be influenced by material characteristics, or the nature of the polarization induced during the CorP process. The improved structural stability enables the material to preserve its polarization even at higher temperatures, thus resulting in a higher T_d. Finally, the CorP method might decrease the tendency of the material to undergo thermal relaxation at elevated temperatures [27, 28]. Thermal relaxation refers to the rearrangement of the material to the internal structure over time, which can lead to depolarization [29]. By reducing the thermal relaxation process, the corona-poled samples can sustain their polarization state at elevated temperatures, leading to a higher T_d. In summary, the higher T_d exhibited by the corona-poled sample can be originated to enhanced polarization retention, improved structural stability, and reduced thermal relaxation within the material, which could be associated with the noncontact poling employed, providing uniform electric field strength distribution in the sample. These factors can contribute to the effective maintenance of polarization state at elevated temperatures, making the CorP method particularly advantageous in achieving a higher T_d compared to other poling methods.

Figure 5 presents the piezoelectric coefficient (d_{33}), electromechanical coupling factor (k_t), and mechanical quality factor (Q_m) depending on different poling methods. In Fig. 5a, the d_{33} of Poling(25) was remarkable in the case of ACP and combined techniques, *i.e.*, DCP@ACP and

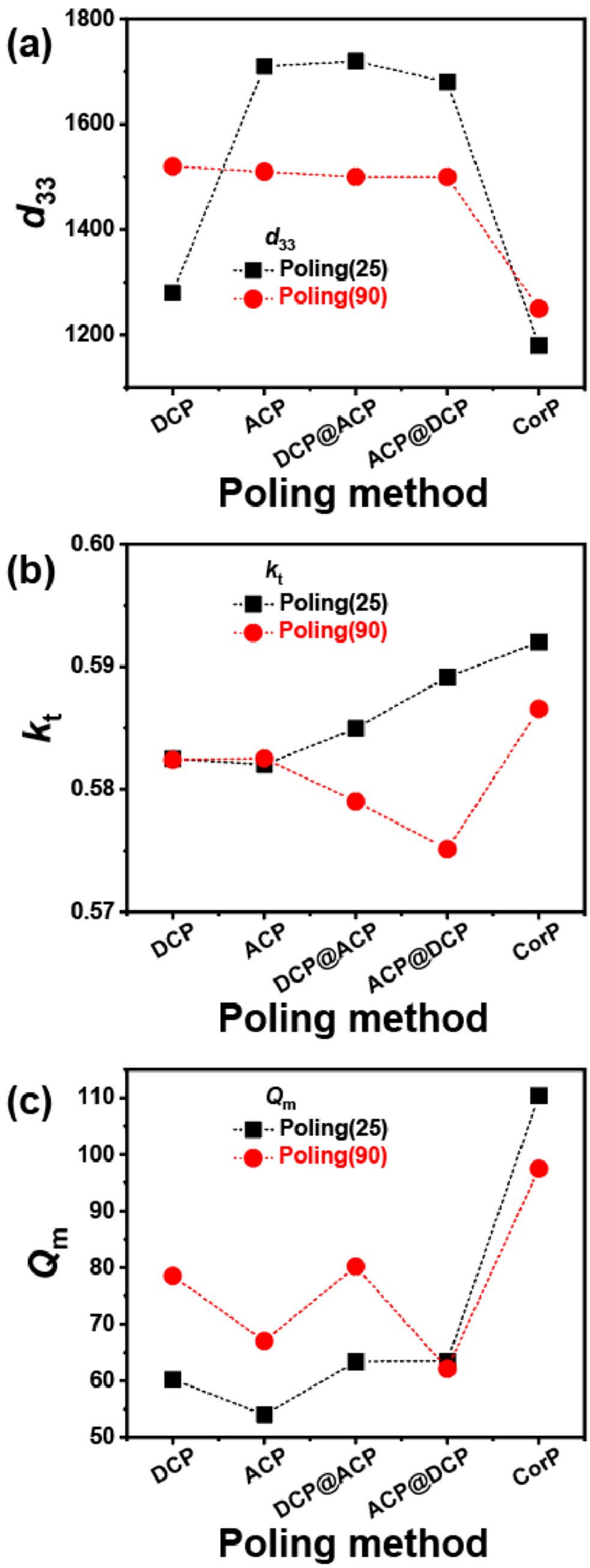

Fig. 5 **a** Longitudinal piezoelectric coefficient (*d*33), electromechanical coupling factor with thickness vibration mode (*k*t), and mechanical quality factor (*Q*m) depending on the poling method and poling temperature

ACP@DCP, which were contributed to the effect of ACP with high piezoelectric and dielectric properties. However, in Poling(90), the d_{33} of conventional poling methods were almost identical to each other, which was consistent with the suppressed variations in temperature-dependent dielectric properties as shown in Fig. 3. It should be noted that CorP showed a similar d_{33} to DCP and slight higher d_{33} in the case of Poling(90) compared to Poling(25). This is because domain switching would be facilitated at elevated temperatures, and the degree of poling can be increased. On the other hand, in Fig. 5b and c, k_t of thickness vibration mode and Q_m were calculated from impedance analyses (See Supplementary Information Fig. S1–S4). It is noteworthy that CorP showed slightly better k_t and notably high Q_m compared to other techniques. Therefore, CorP can be an effective method to pole the PMN-PT single crystals with better electromechanical properties, despite the lower d_{33} compared to ACP-containing cases.

Figure 6 illustrates the polarization versus electric field (P-E) loops and current versus electric field (I-E) curves for different poling conditions including DCP, ACP, and CorP. The coercive field (E_C) of the CorP sample was observed to be lower than that of other poling methods which indicates easier polarization reversal as shown in Fig. 6c. This can be attributed to the noncontact nature of the CorP method allowing the uniform distribution of electric field resulting in homogeneous electric field strength in the sample, which enables a synchronized and coherent reorientation of ferroelectric domains, leading to a lower E_C [18]. Given that conventional poling techniques such as DCP and ACP methods directly contact the samples, localized regions of a high electric field can cause a localized dielectric breakdown, especially in the contact region [14]. In summary, the combination of noncontact poling and uniform electric field distribution in CorP can mitigate the risk of localized breakdown and ensure a homogeneous application of the electric field, resulting in lower E_C and electrically softer.

4 Conclusion

This study revealed that corona poling (CorP) exhibited significant advantages in terms of its electrical properties compared to other poling methods. The corona-poled PMN-PT single crystal demonstrated a higher depolarization temperature (T_d), indicating improved polarization retention at elevated temperatures. In addition, the CorP sample exhibited a lower coercive field (E_C) suggesting that CorP would make it electrically softer. These benefits can be originated to the noncontact nature of CorP, which minimizes the risk of localized breakdown and ensures a uniform electric field distribution. Our findings highlight the potential of CorP as an effective poling technique for enhancing the electrical

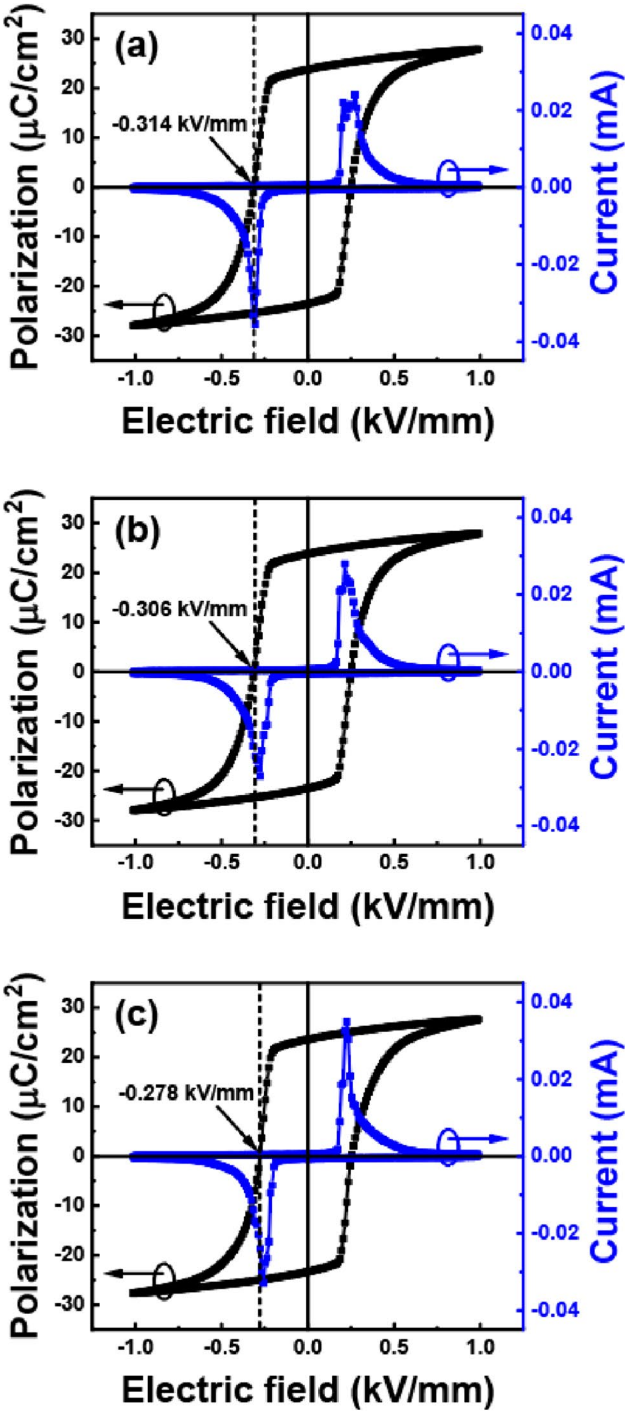

Fig. 6 Room-temperature polarization versus electric field (P-E) loops and current versus electric field (I-E) curves for **a** DCP, **b** ACP, and **c** CorP. The coercive field (*E*C) of the CorP sample showed a lower value compared to DCP and ACP. All the P-E loops and I-E curves were measured after each poling method at 90 ℃

performance of relaxor ferroelectric single crystals. Moreover, further investigations will delve into the underlying mechanisms and explore the applicability of CorP in different material systems, opening new possibilities to the piezoelectric applications.

Supplementary Information The online version contains supplementary material available at https://doi.org/10.1007/s43207-024-00390-0.

Acknowledgements This research was supported by the Material Technology Development Program (No. 1415182019) through the Korea Evaluation Institute of Industrial Technology (KEIT). A part of Sun's work was supported by the US National Science Foundation under Grant No. 2309184. Ryu was supported by the US National Science Foundation under Grant No. 2309184.

Funding This work was funded by Korea Evaluation Institute of Industrial Technology, 1415182019, Wook Jo.

Data availability The datasets used and/or analyzed during the current study are available from the corresponding author on reasonable request.

Declarations

Conflict of interest The authors declare no conflicts of interest. Wook Jo is an Associate Editor of the Journal of the Korean Ceramic Society. Associate Editor status has no bearing on editorial consideration.

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