

Comparison of high-temperature DC poling and AC poling for lead perovskite relaxor-PbTiO₃ single crystals

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1. Introduction

Lead-based perovskite piezoelectric single crystals (SCs) such as binary Pb(Mg_{1/3}Nb_{2/3})O₃-PbTiO₃ (PMN-PT) and ternary Pb(In_{1/2}Nb_{1/2})O₃-PMN-PT have been utilized for ultrasonic sensors, actuators and imaging devices. Among these applications, ultrasound probes have been taking a center position since early 2000.¹⁾ For SC devices, poling procedures for these SCs are essential to obtain high piezoelectric and dielectric properties such as piezoelectric constant (d_{33}) and dielectric permittivity ($\epsilon_{33}^T/\epsilon_0$). Conventional direct current poling (DCP) with high temperature field cooling (HT-FC) has been used since the first discovery of these SCs in 1982.²⁻⁴⁾ Lin *et al.* reported that outstanding d_{33} (2200 pC/N) can be obtained by DCP with HT-FC from 260 °C to room temperature (RT) at 6 kV/cm, which is superior to DCP at RT ($d_{33} = 1300$ pC/N for PMN-0.28PT SC).⁴⁾ Recently, alternating current poling (ACP), a novel and readily-controllable domain engineering method has attracted many researchers' interests owing to enhanced d_{33} and $\epsilon_{33}^T/\epsilon_0$.⁵⁻¹¹⁾ However, there is no report on systematic comparisons regarding the effectiveness of DCP with HT-FC and ACP processes for PMN-PT SCs grown by high composition and properties uniformity of SCs. Therefore, the purpose of this study is to compare electrical properties of DCP, FC-DCP and ACP poling conditions for PMN-0.30PT SCs grown by continuous feeding Bridgeman (CF-BM) process which has verified for composition and piezoelectric properties uniformity.^{11,12)}

2. Experimental procedure

[011]-seeded and [001]-oriented CF-BM PMN-0.30PT SC plates (L13 mm × W3 mm × T0.48 mm³, coercive field (E_c) = 2.2 kV/cm at RT) were sputtered with NiCr/Au (30/200 nm) electrodes on their (001) -surfaces. DCP with HT-FC was performed from 180°C to RT by applying 3 kV/cm (LV), and 6 kV/cm (HV) in a temperature controlled electric oven. At 60 °C, the middle temperature (MT)-ACP processes were performed with a bipolar sine wave of 3 kVrms/cm at 0.2 Hz with 12 cycles, based on Wang and Sun *et al.*'s study.^{9,10)} The standard (STD)-DCP process was conducted at 5 kV/cm for 1 min according to the IEEE standard as

comparisons.¹³⁾ Aging for 96 hours after each poling process. these piezoelectric and dielectric properties were evaluated with an impedance analyzer (Agilent Technology, 4194 A), and piezo d_{33} meter (ZJ-6B).

3. Results and discussion

Table I shows poling conditions and the averaged materials constants of PMN-PT SC with four different poling processes of (a) STD-DCP, (b) HT LV FC-DCP, (c) HT HV FC-DCP, and (d) MT-ACP. These material constants were measured at 25 °C. Figure 1 shows the temperature dependent $\epsilon_{33}^T/\epsilon_0$ of PMN-PT SCs poled by (a) STD-DCP. The SCs showed phase change temperature (T_{R-T}) at 86 °C, loss peak temperature (T_c) at 140 °C, and maximum $\epsilon_{33}^T/\epsilon_0$ temperature (T_m) at 143 °C, which is consistent with the JFE Mineral Co. report.¹²⁾ Figure 2 shows the $\epsilon_{33}^T/\epsilon_0$ of PMN-PT SCs by four poling processes. The highest $\epsilon_{33}^T/\epsilon_0$ of 10000 was obtained by (d) MT-ACP, followed by 7500 of (b) HT LV FC-ACP, 6250 of (c) HT LV FC-ACP and 6200 of (a) STD-DCP. Figure 3 shows the d_{33} of four poling processes of PMN-PT SCs. The highest d_{33} of 3000 pC/N was obtained by (d) MT- ACP, followed by 2500 pC/N of (b) HT LV FC-DCP (c) HT HV FC-DCP, and 1900 pC/N of STD-DCP. The (b) HT LV FC-DCP showed a 21% and 32% increase in $\epsilon_{33}^T/\epsilon_0$ and d_{33} compared to the (a) STD DCP, respectively, however, not as much as the 62% and 58% improvement of the (d) MT-ACP. However, (c) HT HV FC-DCP SCs showed decreased properties caused by an over-poling effect of PMN-PT SCs.¹⁴⁾

Table I. Material constants of PMN-0.3PT SCs by four poling processes, (a) STD-DCP, (b) HT LV FC-DCP, (c) HT HV FC-DCP, and (d) MT-ACP.

Poling process	STD-DCP	HT LV FC-DCP	HT HV FC-DCP	MT-ACP
Voltage (kV/cm)	5	3	6	3
Temperature (°C)	25	200-30	200-30	60
Free dielectric constant	6190	7500	6250	10000
Clamped dielectric constant	1140	1080	980	1350
Dielectric loss (%)	0.38	0.25	0.3	0.27
Calculated k_{33} (%)	90	92.8	91.8	93.3
kt (%)	59.5	60	58	60.5
Nt (Hz•m)	1915	1910	1680	1920
k_{31} (%)	45	53.5	56	47
N_{31} (Hz•m)	770	605	630	570
Sound velocity (m/sec.)	4580	4580	4380	4560
d_{33} (pC/N)	1900	2500	1910	3000

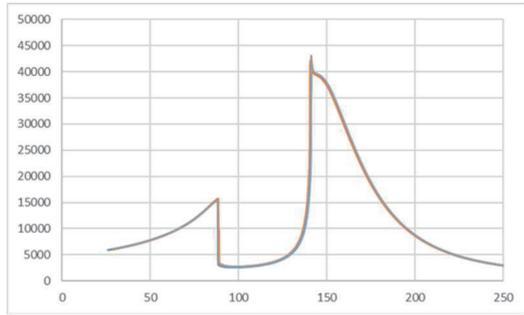


Fig. 1. Temperature dependence of free dielectric constant of STD-DCP PMN-0.3PT SC grown by CF-Bridgman process.

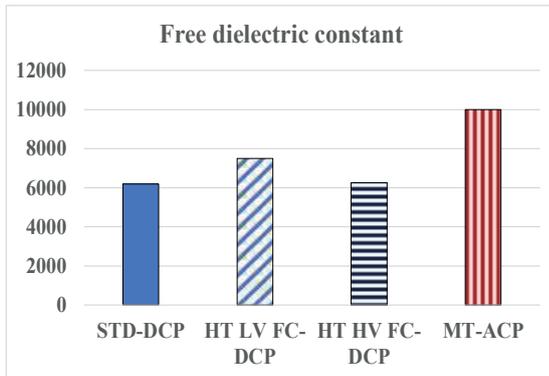


Fig. 2. Free dielectric constant $\epsilon_{33}^T/\epsilon_0$ of PMN-0.3PT SC by different poling process, STD-DCP, HT LV FC-DCP, HT HV FC-DCP, and MT-ACP.

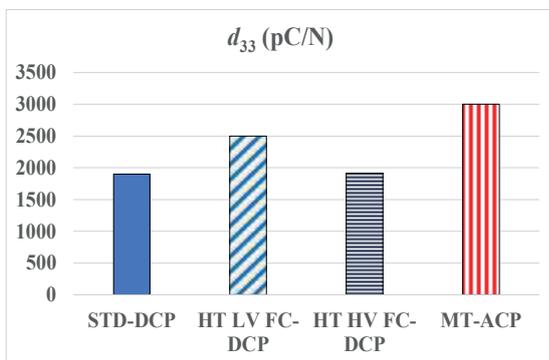


Fig. 3. Piezoelectric strain constant d_{33} of PMN-0.3PT SC by different poling process, STD DCP, HT LV FC-DCP, HT HV FC-DCP, and MT-ACP.

4. Conclusions

We have investigated the effectiveness of poling processes and systematic comparisons for PMN-0.3PT SCs prepared by the CF-BM method with high compositional and properties uniformity by four processes (a) STD DCP, (b) HT LV FC-DCP, (c) HT HV FC-DCP, and (d) MT-ACP. The highest $\epsilon_{33}^T/\epsilon_0$ and d_{33} were obtained by the MT-ACP ($\epsilon_{33}^T/\epsilon_0 = 10000$, $d_{33} = 3000$ pC/N), followed by HT LV FC-DCP ($\epsilon_{33}^T/\epsilon_0 = 7500$, $d_{33} = 2500$ pC/N), HT HV FC-DCP ($\epsilon_{33}^T/\epsilon_0 = 6250$, $d_{33} = 1910$ pC/N), and STD-DCP ($\epsilon_{33}^T/\epsilon_0 = 6200$, $d_{33} = 1900$ pC/N). The HT LV FC-DCP showed a 21% and 32% increase in $\epsilon_{33}^T/\epsilon_0$ and d_{33} compared to STD-DCP, respectively, however, not as much as the 62% and 58% improvement of the MT-ACP. These results provide a good guidance for the design of various piezoelectric devices based on PMN-PT SCs.

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