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Dielectric and piezoelectric properties of $Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ single crystal ultrasonic transducers with AC low-temperature poling

低温で交流分極した $Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ 単結晶
超音波振動子の誘電・圧電特性

Yiqin Sun[1‡], Tomoaki Karaki[1*], Fujii Tadashi[1], and Yohachi Yamashita[1,2]
([1]Toyama Prefectural Univ.; [2]North Carolina State Univ.)
孫 億琴 [1‡], 唐木 智明 [1*], 藤井 正 [1], 山下 洋八 [1,2] ([1] 富山県大院 工, [2] ノースカロライナ州立大 工）

1. Introduction

Currently, lead magnesium niobate $Pb(Mg_{1/3}Nb_{2/3})O_3$ (PMN) and lead titanate $PbTiO_3$ (PT) based solid solution single crystals (SC) are widely used in medical ultrasound (US) imaging system probe transducers. Generally, the [001]-oriented SC transducers are used for this application because their piezoelectric, electromechanical, and dissipation properties are superior to those of conventional $Pb(ZrTi)O_3$ ceramics. In 2014, it was reported that AC poling (ACP) of relaxor-PT SCs can significantly improve piezoelectric and electromechanical properties compared to traditional DC poling (DCP).[1,2] Although the PMN-PT ACP SC transducers are suitable for commercial applications where a high resolution quality is to be expected[3-5], there is still little concern about signal-to-noise ratio of ACP treatment and its low temperature (LT) poling. In this work, the [001]-oriented rhombohedral (R) 0.72PMN-0.28PT (PMN-28PT) SC samples of L12 × W4 × T0.3 mm grown by one charge Bridgman (OC BM) method were applied with ACP treatment at series LT, and their comprehensive properties and unwanted spurious-mode vibration (SMV) were compared and evaluated.

2. Experimental procedure

The ACP PMN-28PT SC samples were poled 40 cycles with 20 Hz bipolar sine waveform of 5 kVrms/cm along [001] direction under cool-air atmosphere at 20, 15, 10, 5, 0 and -5 °C, respectively. The DCP PZT ceramics with the same dimensions as PMN-28PT SCs were poled under 5 kV/cm (IEEE standard) at RT for 5 min. After 24 h for aging, the electrical impedance and dielectric properties were analyzed at RT by an impedance analyzer. The piezoelectric coefficient (d_{33}) values were measured by a S5865 d_{33} meter. The measurement methods of other material constants were established in our previous studies.[6-9]

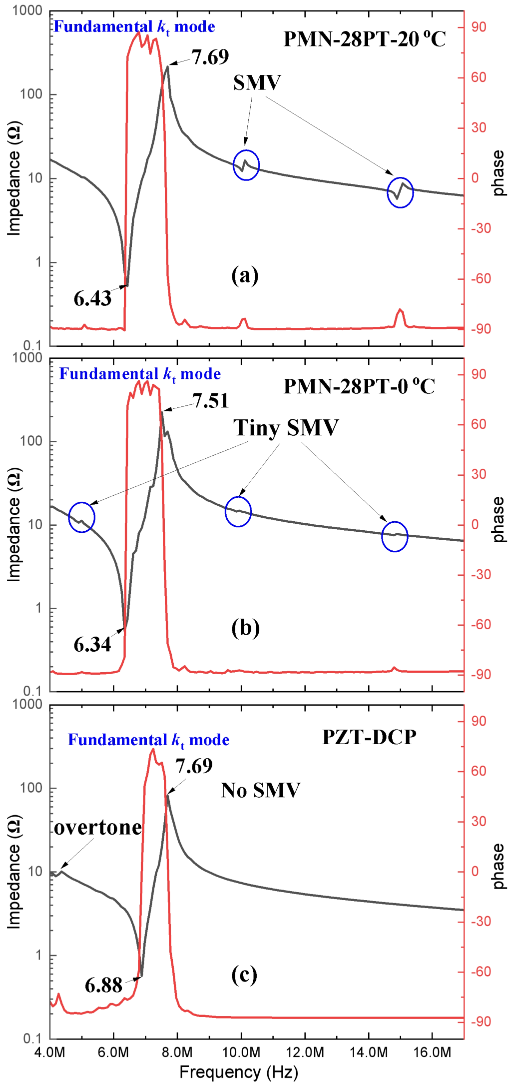

Fig. 1 Impedance and phase spectrum of (a) ACP SC poled at 20 °C; (b) ACP SC poled at 0 °C; (c) DCP PZT ceramic poled at RT (25 °C). Noted that (a) shows spurious-mode vibrations (SMV), while (b) shows tiny SMV and (c) shows no SMV near the fundamental thickness longitudinal vibration mode of L12 × W4 × T0.3 mm samples.

*E-mail address: chen@pu-toyama.ac.jp

2. Results and discussion

Table I summarizes the results obtained from PMN-28PT ACP SCs of poling at series LT compared to DCP ones, as well as the standard DCP PZT ceramics: As poling temperature decreased, the ACP SCs had a trend to show lower dielectric constant ($\varepsilon^T_{33}/\varepsilon_0$), clamp dielectric constant ($\varepsilon^S_{33}/\varepsilon_0$), as well as d_{33} values. Compared to the DCP samples, whether they were SC or PZT ceramics, the ACP SCs overall presented more outstanding dielectric and piezoelectric properties. The bar mode electromechanical coupling factor (k_{33}) calculated from $\varepsilon^T_{33}/\varepsilon_0$ and $\varepsilon^S_{33}/\varepsilon_0$ of ACP SCs showed over 92.5% which were obviously higher than that of DCP ones. The ACP SCs exhibited ultra low tan δ values of less than 0.35%. When the poling temperature of the ACP SCs dropped below 0 ℃, the SMV were significantly improved and eventually completely suppressed. The DCP PZT ceramics totally showed no SMV but only overtones at around 4.3 MHz extended from k_{31} or k_{32} vibration modes, as shown in **Fig. 1**. The SMV responses near the fundamental thickness mode showed not so large as high temperature ACP treatment[8,9] for PMN-28PT SCs. Impedance and phase spectrum of (b) ACP SC poled at 0 ℃ suggested very tiny SMV responses than (b) of ACP SC poled at 20 ℃. The (c) DCP PZT ceramic poled at RT (25 ℃) showed a clean impedance and phase spectrum, but a highest resonance frequency (f_r) of 6.88 MHz than the SC samples. The ACP SC poled at 0 ℃ gave a lower f_r of 6.34 MHz and anti-resonance frequency (f_a) of 7.51 MHz than those of samples poled at 20 ℃ (f_r = 6.43 MHz, f_a = 7.69 MHz), suggesting a more completely polarization switching. Therefore, it could be concluded that ACP SCs were more likely to excite unwanted SMV responses than DCP ones resulted from their violent vibrations when applied with high frequency AC fields. However, there is no doubt that the new technology of ACP treatment has more promising application prospects.

4. Conclusions

The [001]-oriented PMN-28PT SCs were applied with ACP treatment at series LT, and the SMV of impedance properties were investigated. We confirmed that the LT ACP treatment was also an effective way to improve the properties for PMN-28PT SCs compared to DCP SCs and also the traditional PZT ceramics. Under the same ACP conditions, decreasing poling temperature could avoid exciting unwanted SMV responses which make achieving higher axial and lateral resolution of echo US images difficult. This remarkable feature will be helpful to realize a high k_{33} and d_{33} of ACP PMN-PT SC for high-end ultrasonic imaging probe transducers.

References

1) N. Yamamoto, et al.: U.S. Patent 0062261 A1 (2014).
2) Y. Yamashita, et al.: U.S. Patent 0372219 A1 (2015).
3) J. Xu, et al.: Appl. Phys. Lett. **112**, 182901 (2018).
4) W.-Y. Chang, et al.: Mater. Res. Lett. **6**, 537 (2018).
5) C. Qiu, et al.: Nature **577**, 350 (2020).
6) Y. Sun, et al.: Jpn. J. Appl. Phys. **58**, SLLC06 (2019).
7) Y. Sun, et al.: Jpn. J. Appl. Phys. **59**, SPPD08 (2020).
8) Y. Sun, et al.: to be published in J. Materiomics, [https://doi.org/10.1016/j.jmat.2021.05.002].
9) Y. Sun, et al.: to be published in Jpn. J. Appl. Phys. [https://doi.org/10.35848/1347-4065/ac127b]

Table I Piezoelectric/dielectric/SMV properties measured at 25 ℃ of PMN-28PT SCs poled from 20 to -5 ºC by ACP compared to DCP ones, as well as the standard traditional DCP PZT ceramics.

Sample Code #	Poling Temp. (℃)	$\varepsilon^T_{33}/\varepsilon_0$	$\varepsilon^S_{33}/\varepsilon_0$	tan δ (%)	k_{33} (%)	d_{33} (pC/N)	SMV level
1	20	8260	1110	0.28	93.0	1990	****
2	15	8150	1020	0.34	93.5	1970	***
3	10	8130	1170	0.19	92.5	1990	***
4	5	8000	1130	0.16	92.7	1960	**
5	0	7610	970	0.19	93.4	1930	*
6	-5	7740	970	0.30	93.6	1900	No
7-DCP	25	5320	1400	0.26	90.0	1340	*
8-PZT	25	5420	1890	1.5	80.7	870	No