Dielectric and piezoelectric properties of AC poled relaxor

single crystals grown by solid state crystal growth method

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1. Introduction

Relaxor base piezoelectric single crystals (SC) transducers have been applied into probes for medical ultrasound diagnostic equipment, and more than 1 million SC probes have been employed in total since 2000.¹⁾ The Pb(Mg_{1/3}Nb_{2/3})O₃ (PMN)-PbTiO₃ (PT) based c SCs are generally fabricated by a melting-involved growth such as the Bridgman (BM) method around 1300 °C. However, large compositional segregation inside SC ingots by the BM method has adversely hampered reproducibility.²⁾ On the other hand, solid state single crystals growth (SSCG) is a growth process without melting of ceramic precursors, and hence it has several advantages such as high compositional uniformity, moderate machinability, low acoustic impedance (Z₃₃), and self-poling (SEP) property.³⁻⁸⁾ The SEP is piezoelectric properties without any poling (NOP) process which have been well known in many thin films with substrate. Ceracomp Co. Ltd., Korea manufactured high Qm > 400 Mn doped SSCG SC PMN- 0.3Pb(Zr,Ti)O₃ (PZT) -based SCs $(d_{33} = 1000 \text{ pC/N})$ with a product name of the CSH-40.9) Recently Maiwa et al. have studied the electrical properties of NOP and DCP for acceptor of Mn doped PMN-0.30PZT SCs with low density (7.4 g/cm³) grown by SSCG.¹⁰ After technological improvement, high-density (7.99 g/cm³) Mn doped SSCG SCs have developed recently. Therefore, the purpose of this study is to investigate the electrical properties of ACP of the high-density SSCG SCs.

2. Experimental procedure

The Mn doped PMN-PZT SC (CSM-11HD) with the Curie temperature of 150 - 160 °C was grown by

the SSCG method using a [011]-seed. After cutting to [001] plates of $4 \times 4 \times 0.5$ mm³, silver (Ag) electrode with thickness of 0.01 mm each side was fired at 650 °C for 30 min. These NOP SC properties were first measured without any poling process. After the first measurement for NOP SC, DC poling and AC poling with different cycles, voltages and frequencies were performed. Electrical impedance and dielectric properties were analyzed by an impedance analyzer (4194 A. Piezoelectric constants of d_{33} were measured piezo d_{33} meter (ZJ-4B).

3. Results and discussion

Figure 1 (a), shows that the dielectric constant of ACP SC below 125 °C was higher than DCP SC and NOP SC, At the same time, Fig.1 (b) shows the dielectric loss of ACP SC below 125 °C was lower than that of DCP SC and NOP SC. These also shows phase change temperature of ACP SC was 10 °C higher than DCP SC.

Figure 2 shows figure of merit (FOM) of $d_{33} \times g_{33}$ for the SSCG SCs by different poling conditions, the highest FOM of ACP was 52.7 pm²/N which is similar to that of DCP SC (54.2 pm/N).

Table I shows materials constants of NOP, DCP, and ACP of high density SSCD PMN-PZT SCs. It is noteworthy that the NOP SC showed very high $d_{33} =$ 740 pC/N without poling. The d_{33} of SEP SSCG SC is higher than that of PZT 5H ceramics ($d_{33}=650-700$ pC/N) And in most cases, ACP SC showed better electrical properties than DCP SC. The dielectric loss of ACP SC was 0.4% which is lower than that of DCP SCs (0.7%).

However, the current experimental data did not show a significant enhancement between ACP and DCP SCs, but according to the current results, with the change of several poling conditions, more detail study of optimal ACP condition research is conducting now and these results will be reported in a near future.





Fig. 1. (a) Temperature dependence of dielectric constant, and (b) dielectric loss of NOP, DCP, and ACP. Note: The phase change temperature of ACP SC was 10 °C higher than DCP SC.

Table I. Materials constants of NOP, DCP, and ACP of high-density SSCD PMN-PZT SCs.

Material constants	NOP	DCP	ACP
Free dielectric constant	2410	3320	3630
Clamped dielectric constant	1190	750	741
Dielectric loss (%)	0.9	0.7	0.4
Calculated k 33 (%)	71.2	88.1	89.2
Acoustic impedance (MRayles)	31.9	34.9	34.4
d 33 (pC/N)	740	1260	1300
g 33 (x10 ⁻¹² Vm/N)	34.8	43	40.5
FOM (d ₃₃ x g ₃₃ , pm ² /N)	25.8	54.2	52.7
Phase change temp. (°C)	86	76.5	86.5



Fig. 2. Figure of merit (FOM) of $d_{33} \times g_{33}$ for SSCG SC by different poling conditions.

4. Conclusions

When optimal AC poling condition was applied to the high-density SSCG SCs, it shows phase transition temperature by 10 °C higher than DCP SC, at the same time, ACP SC can improve the free dielectric constant by 10% compared to DCP SC. In addition, the ACP SCs showed lower loss than DCP SCs. However, the highest FOM of ACP SC was 52.7 pm²/N which is similar to that of DCP SC. These results show that the properties of the SCs after ACP shows some improved properties compared with DCP SCs.

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