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Self-poling and DC poling of Mn doped $Pb(Mg_{1/3}Nb_{2/3})O_3$-$Pb(ZrTi)O_3$ single crystals grown by solid state crystal growth process

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1. Introduction

Relaxor base piezoelectrisingle 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 $Pb(Mg_{1/3}Nb_{2/3})O_3$ (PMN)-$PbTiO_3$ (PT) based c SCs are generally fabricated by a melting-involved growth such as the Bridgman (BM) method around 1300 ºC. 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 (Z_{33}), and self-poling (SEP) property.[3-9)] The SEP is piezoelectric properties without any poling (NOP) process which have been well known in many thin films with substrate. The SEP of Sn doped bulk PMN-PT SC was also reported by He et al .[8)] Ceracomp Co. Ltd., Korea manufactured high Qm>500 Mn doped SSCG SC doped PMN-0.3Pb(Zr,Ti)O_3 (PZT) -based SCs (d_{33} = 1000 pC/N) with a product name of the CSH-40.[10)] G. J. Lee at al also reported these SEP properties for Mn-doped PMN-PT SSCG SCs.[11)] Therefore, the purpose of this study is to investigate electrical properties of NOP and DCP for acceptor of Mn doped PMN-0.30PZT SCs grown by SSCG which has verified for composition and piezoelectric properties uniformity.

2. Experimental procedure

The Mn doped PMN-PZT SC (CSH-40) with the Curie temperature of 150 - 160 ºC was grown by the SSCG method using a [011]-seed. After cutting to [001] plates of 10 × 3 × 0.5 mm^3, silver (Ag) electrode with thickness of 0.01 mm each side was fired at 650 ºC for 30 min. These NOP SC properties were first measured without any poling process. After the first measurement for NOP SC, DC poling with different voltages and directions was performed. Electrical impedance and dielectric properties were analyzed by an impedance analyzer (Agilent Technology, 4194 A, CA, USA). Piezoelectric constants of d_{33} were measured piezo d_{33} meter (ZJ-4B).

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3. Results and discussion

These NOP SC sample showed a clear impedance even though it was not polarized, and for the coupling factors k_{31}, k_t modes and d_{33} were clearly observed as shown in Fig.1. These values were 0.3 for k_{31} and 0.4 for k_t, and 520 pC/N for d_{33}, respectively. The d_{33} = 520 pC/N obtained for the SEP of the SSCG SC was superior to that of Mn doped PZT-based ceramics, such as PZT4, PZT8 and almost non-lead (Pb) piezoelectric materials. Figure 2 shows piezoelectric d_{33} for the SSCG SCs by different DC poling voltages and directions. By applying a forward DC voltage without reverse polarization rotation, high d_{33} values, 850 - 920 pC/N were obtained compared to negative DC voltage directions (620 - 810 pC/N) as shown in Fig. 2.

Figure 3 shows figure of merit (FOM) of $d_{33} \times g_{33}$ for the SSCG SCs by different DC poling voltages and directions. By applying a forward DC voltage without reverse polarization rotation, high FOM values of 30 - 32 pm^2/N were obtained compared to negative DC voltage directions (14 - 27 pm^2/N) as shown in Fig. 3

However, obtained d_{33} and FOM values of DCP SCs in this experiment were still smaller than catalogue values of the CSH-40 (1000 pC/N, 37 pm^2/N). The excellent heat resistance of the piezoelectric properties of the Mn doped PMN-PZT SSCG SC may overturn the common practice of selecting materials with a high Tc which has been regarded as a selection criterion for high heat-resistant piezoelectric materials.

Further poling and de-poling processes improvement and microstructure observation are conducted and better piezoelectric properties and their microstructures will be reported in a near future.

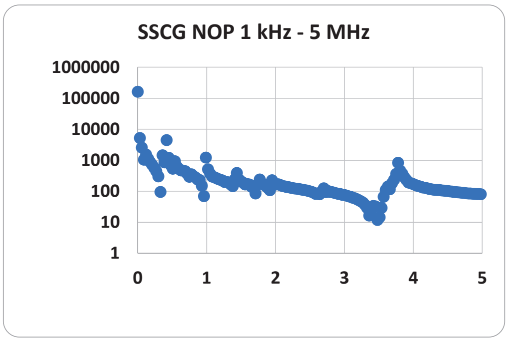

Fig. 1. Impedance vs. frequency of NOP SSCG PMN-0.3PZT SC. Note: k_{31}, k_t, and d_{33} were 0.3, 0.4, and 520 pC/N without any poling.

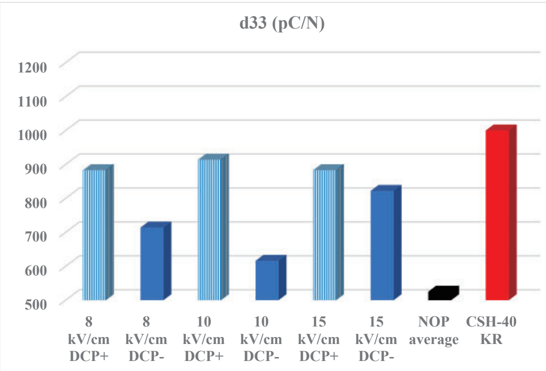

Fig. 2. Piezoelectric constants of d_{33} for SSCG SC by different DC poling voltages and directions. Note: forward (+) direction DC poling SC bringing about higher FOM values.

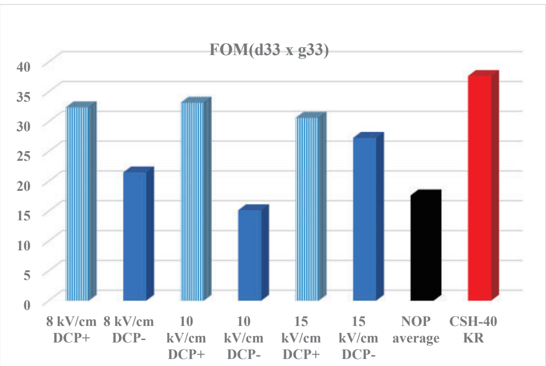

Fig. 3. Figure of merit (FOM) of $d_{33} \times g_{33}$ for SSCG SC by different DC poling voltages and directions. Note: forward (+) direction DC poling SC bringing about higher FOM values.

4. Conclusions

Acceptor of Mn-doped PMN-PZT SC grown by SSCG process after Ag electroding at 650 °C showed a high d_{33} of 520 pC/N at 25°C without any poling process which called "self-poling". Furthermore, the SC with a Curie temperature of 150 °C showed high electro mechanical coupling factors of thickness mode k_t = 0.40 and calculated bar mode coupling factor of k_{33} = 0.7. The d_{33} obtained for the self-poling of the SSCG SC was superior to that of Mn doped PZT-based ceramics and almost non-lead piezoelectric materials. When 10 kV/cm DC at 80 ºC was applied to the SCs in the forward and reverse directions of spontaneous polarization, the former showed d_{33} of 910 pC/N and the latter 620 pC/N, indicating that the ease of polarization reversal was distinctly different. The excellent heat resistance of the piezoelectric properties of the SSCG SC may overturn the common practice of selecting materials with a high Tc which has been regarded as a selection criterion for high heat-resistant piezoelectric materials.

Acknowledgment
This work was supported in part by Murata Science Foundation 2023.

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