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Dielectric and Piezoelectric Properties of Pb Perovskite Relaxor type Single Crystals by AC poling
交流分極された鉛系ペロブスカイト型単結晶の圧電誘電特性

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1. Introduction

Introduction

Solid solution of $Pb(Mg_{1/3}Nb_{2/3})O_3$-$xPbTiO_3$ (PMN-xPT) and the other relaxor ferroelectric single crystals (SCs) have attracted great research interest over the past decades owing to their outstanding piezoelectric and electromechanical properties, and widely applicated in sensors, transducers, and actuator technology. In order to improve the performance of ultrasonic (US) medical echo probes, a higher performance piezoelectric material is required, and relaxor SCs of PMN-PT have been considered as a promising material for next generation transducers for medical echo and some of medical probes have already been commercialized with PMN-PT SCs due to relatively higher piezoelectric constant d_{33} > 1500 pC/N, which is nearly twice that (d_{33}<900 pC/N) of traditional lead zirconate titanate (PZT) ceramics.

In general, the growth method of PMN-PT SCs is one charge Bridgman (OC BM) method. To solve the compositional segregation, JFE Mineral Co. Japan developed new SC growth method named continuous-feeding Bridgman method (CF BM), and this method can control the melt composition during crystal growth process and improve the composition uniformity.1)

Recently, Yamamoto and Yamashita et al., reported dynamic AC poling (ACP) can enhanced 20 to 100% of the dielectric and piezoelectric properties of SCs compared to conventional statice DC poling (DCP).2-3) After these reports, many researchers have investigated the ACP research in the world.4-7)

When piezoelectric materials suffer the abrupt changes in external conditions such as temperature and biasing filed etc, gradual changes in its electrical and physical constants with time will occur, which called aging rate (AR) of materials.8)

For medical US echo probes, the larger AR of crystals resulted in deteriorating US imaging qualities and the shorter service life of these probes. The AR of ferroelectric ceramics have been reported by many researchers. However, there is a few studies on the AR of PMN-PT based relaxor SCs. Therefore, it is necessary to systematically investigate these AR of various PMN-PT based SCs manufactured by OC BM and CF BM process and poled by ACP and DCP.

The purpose of this study is to systematically investigate the different crystal growth methods, compositions, and poling methods of (PIN)-PMN-PT SCs and to compare with several type of PZT ceramics ($\varepsilon_{33}^T/\varepsilon_0$ > 2000) which have already been widely used in medical echo application.

2. Experimental methods

In this experiment, the [001]-growth and [001]-oriented PMN–28PT and PMN-30PT SC plates manufactured by OC BM, and the [001]-growth and [001]-oriented PIMN-26PT, PIMN-30PT and PMN-30PT SC plates manufactured by CF BM, and several $Pb(Zr,Ti)O_3$ (PZT) ceramics were used as comparisons. Figure 1 shows poling experiment set up for this experiment. In order to reduced mechanical cleaning effects by a ultrasonic cleaner after poling using organic aceton, we used air poling system as shown in Fig. 1.

The size of samples were about 0.25 to 1.0 cm^2. After various DCP and ACP conditions, AR of $\varepsilon_{33}^T/\varepsilon_0$ of 14 of SC samples were investigated from 10 to 1000 hours after each poling process. In addition, 3 types of PZT ceramics with different $\varepsilon_{33}^T/\varepsilon_0$ were investigated as comparison.

Aging rates (AR) were determined in next equation.

$$AR=(MC_{25(t0)}-MC_{25(10t0)})/\ MC_{25(t0)} \qquad (1)$$

$MC25(_{t0})$: performance parameters at 25 °C at t_0 time;

$MC25(10_{t0})$: performance parameters at 25 °C at $10t_0$ time

Here, the MC is $\varepsilon_{33}^T/\varepsilon_{0.}$

Other measurement were shown in previous report.6-7)

3. Result and Discussion.

Table I shows all $\varepsilon_{33}^T/\varepsilon_0$ and AR of 17 SCs and ceramics investigated in this study.

Figure 2 shows all $\varepsilon_{33}^T/\varepsilon_0$ and AR map of 17 SCs and ceramics. As shown in these Table I and Fig.2, AR of $\varepsilon_{33}^T/\varepsilon_0$ were varied from 0.70%/decade with $\varepsilon_{33}^T/\varepsilon_0$ of 2400 of the ACP CF BM

PIMN-30PT SC to 2.7 %/decade of the ACP OC BM PMN-30PT SC with $\varepsilon_{33}{}^{T}/\varepsilon_0$ of 14000. These results indicated higher $\varepsilon_{33}{}^{T}/\varepsilon_0$ SCs tends to show higher AR. However, AR of $\varepsilon_{33}{}^{T}/\varepsilon_0$ for PMN-(PIN)-xPT SCs were smaller than that of three types of PZT ceramics with the same $\varepsilon_{33}{}^{T}/\varepsilon_0$. The AR of $\varepsilon_{33}{}^{T}/\varepsilon_0$ for the PMN-30PT SCs manufactured by OC BM ($\varepsilon_{33}{}^{T}/\varepsilon_0 \approx 14000$) had almost same AR of $\varepsilon_{33}{}^{T}/\varepsilon_0$ with PZT ceramics ($\varepsilon_{33}{}^{T}/\varepsilon_0 \approx 5600$). This means AR of PMN-(PIN)-PT SCs are stable than PZT ceramics in spite of poling method.

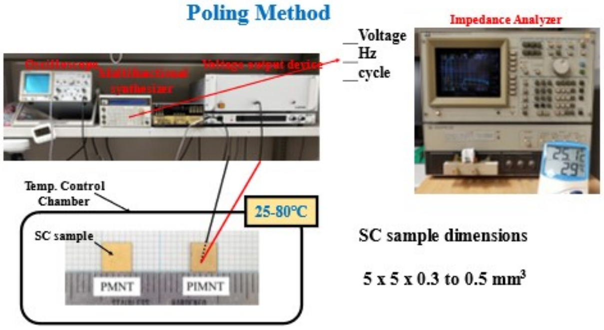

Fig. 1. High temperature poling method in air atmosphere and SCs samples.

Table I, Aging rate vs. dielectric constant of 17 SCs and ceramics.

#	Sample	Aging rate (%/decade)	dielectric constant
1	CF BM PIMN-30PT ACP	2.50	8000
2	CF BM PIMN-30PT ACP	0.96	9080
3	CF BM PIMN-30PT ACP	0.88	8250
4	CF BM PIMN-30PT ACP	0.71	2400
5	OCBM PMN-28PT ACP	2.21	6650
6	OCBM PMN-28PT ACP	1.70	7000
7	CF BM PIMN-26PT DCP	1.13	3230
8	CF BM PIMN-26PT ACP	1.18	2930
9	OCBM PMN-30PT DCP	2.11	7920
10	OCBM PMN-30PT ACP	2.73	14340
11	CFBM PMN-30PT DCP	1.5	6000
12	CFBM PMN-30PT ACP	1.58	7190
13	CFBM PMN-30PT ACP	2.2	8000
14	CFBM PMN-30PT ACP	1.4	5040
15	PZT ceramics K5600	3.00	5600
16	PZT5H ceramics K3400	2.40	3400
17	PZT5A ceramics K2500	1.40	2500

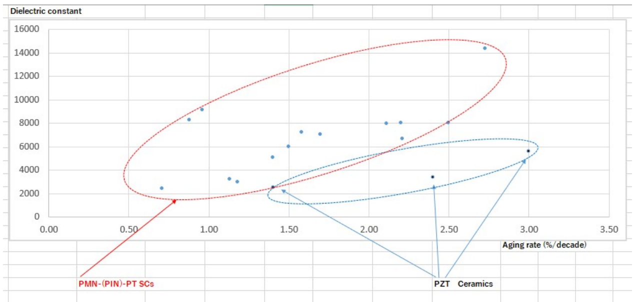

Fig.2 Aging rate vs. dielectric constant map of 17 SCs and ceramics.

4. Conclusions

Aging rates (AR) of $\varepsilon_{33}{}^{T}/\varepsilon_0$ of (PIN)-PMN-xPT SCs with different compositions, growth methods, and poling conditions have been systematically investigated and next conclusions were obtained.

(1) AR of $\varepsilon_{33}{}^{T}/\varepsilon_0$ were varied from 0.70%/decade to 2.7 %/decade. Higher $\varepsilon_{33}{}^{T}/\varepsilon_0$ SC tends to show higher AR.

(2) AR of $\varepsilon_{33}{}^{T}/\varepsilon_0$ for PMN-xPT SCs were smaller than that of PZT ceramics with the same $\varepsilon_{33}{}^{T}/\varepsilon_0$.

(3) AR of $\varepsilon_{33}{}^{T}/\varepsilon_0$ for PMN-30PT SCs manufactured by OC BM ($\varepsilon_{33}{}^{T}/\varepsilon_0 \approx 14000$) had almost same AR of $\varepsilon_{33}{}^{T}/\varepsilon_0$ with PZT ceramics (($\varepsilon_{33}{}^{T}/\varepsilon_0 \approx 5600$).

Acknowledgment

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