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Ferroelectric domain structures in lead free piezoelectric ceramics composed of Bi-layer structured $SrBi_4Ti_4O_{15}$

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Abstract

The domain switching and rotation in Bi-layer structured ceramics were evaluated by investigating the poling field and pulse response dependences of the ferroelectric properties. The effect of domain clamping on electromechanical coupling factor and dielectric constant at the coercive field was confirmed in Bi-layer structured ceramics as well as in the cases of soft and hard tetragonal lead zirconate titanate, lead titanate and barium titanate ceramics. The minimum frequency constant due to domain rotations was also observed in Bi-layer structured ceramics. The remanent polarization and coercive field increased linearly with increasing the pulse field. Although relatively high DC fields up to 10.0 kV mm^{-1} could be applied to the ceramics, the P–E hysteresis loop showed a round figure. Furthermore, the oriented polarization by DC poling was highly stable in the ceramics, even though a high bipolar pulse was applied.

Keywords: Bi-layer structured $SrBi_4Ti_4O_{15}$; Bipolar pulse; Ferroelectric domain structures; Lead free piezoelectric ceramics; Poling field

1. Introduction

Domain switching and rotation in lead zirconate titanate (PZT), lead titanate (PT) and barium titanate (BT) ceramics can be caused by either or both mechanical stress and electric fields, because the ceramics possess ferroelastic and ferroelectric properties.[1] Both 90° and 180° domains exist in the tetragonal phase of PZT, PT and BT, in which ferroelastic deformation occurs only by 90° rotation. In the cases of tetragonal PZT,[2–5] PT[6] and BT[7] ceramics, we had reported on the poling field dependence of the planar coupling factor, dielectric constant and frequency constant which are affected by both 90° rotation and 180° switching.

In this pursuit, we investigated the dependence of ferroelectric properties on poling field and bipolar pulse in Bi-layer structured ceramics to clarify their domain structures. Furthermore, we discuss the difference in the effect of domain switching and rotation, when DC field and bipolar pulse were applied to the ceramics.

2. Experimental

The Bi-layer structured ceramics are composed of manganese modified $SrBi_4Ti_4O_{15}$ (SBT). Two kinds of SBT ceramic disks were prepared; the ones with 14.6 mm in diameter and 1.0 mm thick and the others with 0.42 mm thick. Poling was conducted using the former disk samples at 150° for 15 min while varying the poling field (E) from $0 \to +0.5 \to +1.0 \to +1.5 \to +2.0 \to \ldots +10.0 \to 0 \to 0 \to +2.0 \to \ldots +10.0 \to 0 \to -10.0 \to 0$ to $+10.0$ kV mm^{-1}. After each poling, the dielectric and piezoelectric properties were measured at room temperature using an LCR meter (HP4263A), an impedance/gain-phase analyzer (HP 4194A) and a d_{33} meter (Academia Sinica: ZJ-3D). The P–E hysteresis loops and bipolar triangle pulse responses were investigated regarding the latter disk samples at 150° by a high voltage test system (Radiant: RT6000HVS).

3. Results and discussion

3.1. Poling field dependence of dielectric and piezoelectric properties

Fig. 1 (a)–(d) show the effect of E on electromechanical coupling factor for thickness mode (k_t), piezoelectric d constant (d_{33}), dielectric constant (ε_r) and

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frequency constant for thickness mode (fc_t) in SBT ceramics, when E was varied from 0 to ± 10.0 kV mm^{-1}. From the plot of k_t and d_{33} vs E, the fields of $E = \pm 3.0$ kV mm^{-1}, at which k_t was minimum and d_{33} approached zero, were determined to be coercive fields (*E*c) under the poling conditions in SBT ceramics. We believe that 180° switching was the dominant factor affecting k_t and d_{33} after having poled the virgin (as-fired) ceramics.[4–7] Furthermore, the *E*c values in SBT ceramics were larger than those in PZT, PT and BT ceramics because of the high Curie point of 520°C. The minimum k_t and d_{33} owing to electrical domain clamping, such as ↑↓, occurred at the coercive field, because the domain clamping canceled out the polarization in the ceramics. From the plot of E dependence of ε_r, ε_r was found to be minimum at *E*c, because of electrical domain clamping. These phenomena have been already observed in soft and hard tetragonal PZT,[4,5] PT[6] and BT[7] ceramics. From the plot of fc_t vs E, fc_t was found to be minimum at $E = \pm 3.0$ kV mm^{-1}, the fields of which corresponded to the fields required to obtain the minimum of k_t and ϵ_r. Since the decrease in fc_t means that the ceramics have become mechanically soft by means of the change in the crystal orientation, it was thought that the domain rotations occurred at the E corresponding to the minimum fct. Consequently, it could be estimated that the poling fields of ± 3.0 kV mm^{-1} corresponded to the thresholds of the domain switching and domain rotations under the poling conditions.

3.2. Bipolar pulse dependence of ferroelectric properties

Figs. 2(b) and (c) show the effect of bipolar triangle pulse field (E) on the remanent polarization (Pr) and coercive field (*E*c) measured at 150°C when the pulse was applied during 800 ms [Fig. 2 (a)]. The Pr and *E*c increased with increasing E and reached to 10 μC cm^{-2} and 6.0 kV mm^{-1} at the E of 8.0 kV mm^{-1}, respectively. The k_t and d_{33} became 6.9% and 10 pC N^{-1} after applying the pulses up to 8.0 kV mm^{-1}. In contrast, these values by bipolar pulse poling were smaller than 20.8% and 22 pC N^{-1} obtained by the conventional DC poling.

Fig. 3 (a)–(d) show a typical *P–E* hysteresis loop and bipolar pulse cycle dependence of k_t, d_{33} and ε_r in SBT ceramics when the pulses ($E = \pm 8.0$ kV mm^{-1}, $T = 800$ ms) were applied as-fired (virgin) and DC poled ceramics up to 10 times. At the moment, the DC poling conditions were as follows; poling field: 10.0 kV mm^{-1}, poling temperature: 150°C and poling time: 15 min. It was thought that certain charges such as space charge existed in SBT ceramics independent of with or without DC poling because of the round figure of the *P–E* loops.

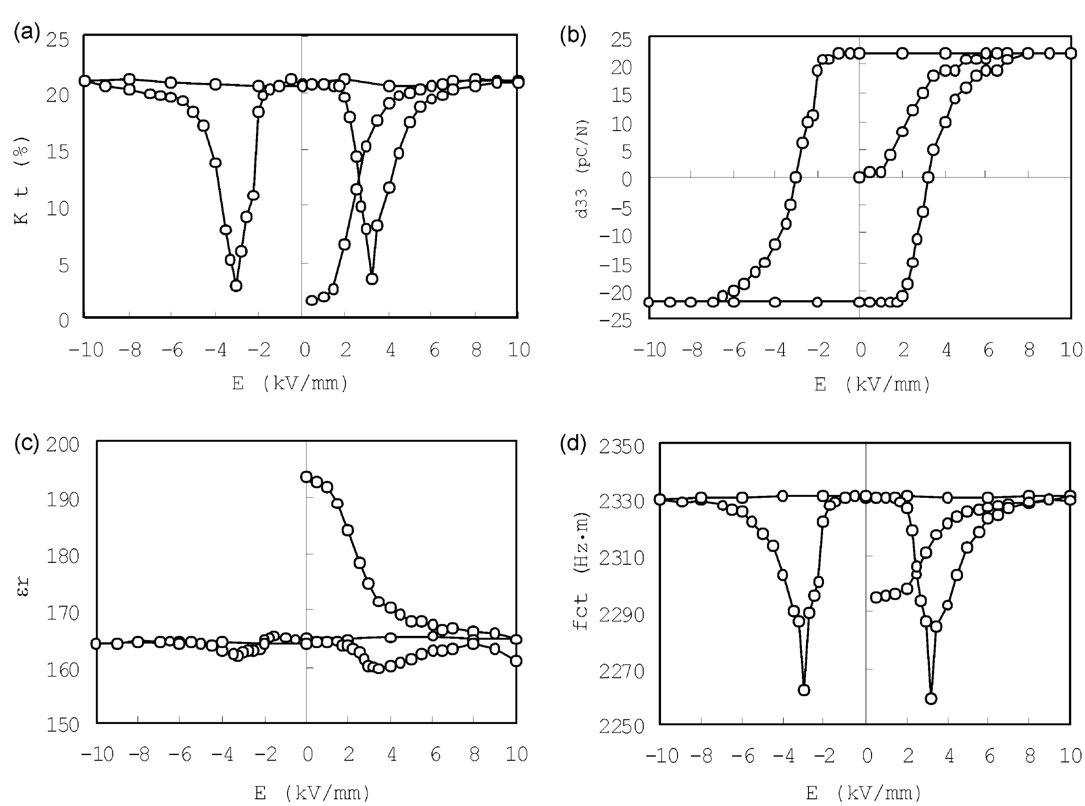

Fig. 1. Poling field (E) dependence of (a) coupling factor (k_t), (b) d_{33} constant, (c) dielectric constant (ε_r) and (c) frequency constant (fc_t) in SBT ceramics.

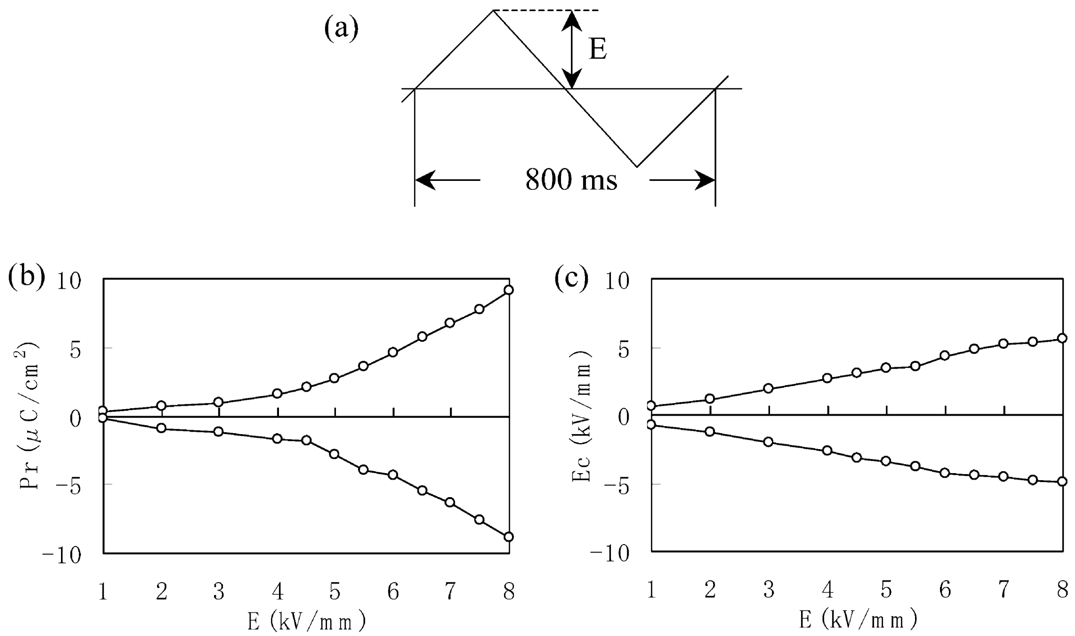

Fig. 2. (a) Bipolar pulse field (*E*) dependence of (b) remanent polarization (Pr) and (c) coercive field (*E*c) at 150°C; bipolar pulse width: $T = 800$ ms.

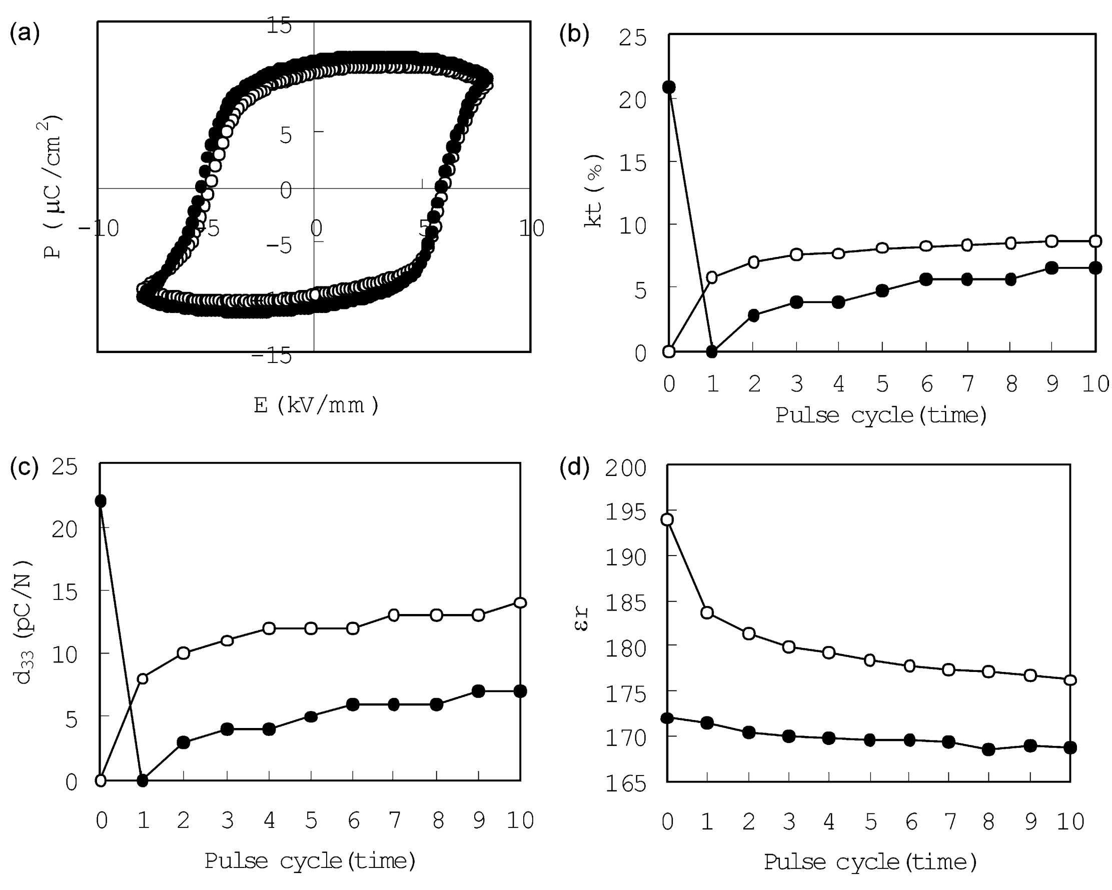

Fig. 3. (a) *P*–*E* hysteresis loops and bipolar pulse cycle dependence of (b) k_t, (c) d_{33} and (d) ε_r applying to as-fired (○) and DC poled (●) SBT ceramics.

Table 1
Bipolar pulse poling and DC poling on as-fired and DC poled SBT ceramic samples

	Poling method		
	Bipolar pulse		DC
Sample	As-fired	DC poled	As-fired
k_t (%)	8.8	6.6	20.8
d_{33} (pC N^{-1})	14	7	22

Increasing pulse cycle, the k_t and d_{33} increased, on the other hand, the εr decreased in the case of the as-fired ceramics. However, in the case of the DC poled ceramics, the k_t and d_{33} showed the minimum while the bipolar pulse was applied in one cycle, after that, these values increased with the increase of the cycle. We believe that the minimum was due to the 180° domain clamping by applying the bipolar pulse.

Table 1 shows the poling efficiency regarding k_t and d_{33} between the bipolar pulse poling and DC poling on the disk samples of as-fired and DC poled SBT ceramics. Since there was a big difference in poling time between pulse poling (400 ms$\times$10 cycles) and DC poling (30 min), the k_t and d_{33} values by bipolar pulse poling could not reach to those by DC poling. In the case of bipolar pulse poling, there were differences in k_t and d_{33} between as-fired and DC poled samples. It was thought that the oriented polarization by DC poling in SBT ceramics became more stable against the disturbance by applying the bipolar pulses.

4. Conclusions

Poling field and pulse response dependences of ferroelectric properties were investigated in SBT ceramics in comparison with those in soft and hard tetragonal PZT, PT and BT ceramics. The relationships between 180° switching accompanied with 180° domain clamping, domain rotations and dielectric and piezoelectric properties were evaluated. It was also clarified that the effect of bipolar pulses on the domain structures and the difference between DC poling and bipolar pulse poling.

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References

1. Jaffe, B., Cook, W. R. JR and Jaffe, H., *Piezoelectric Ceramics*. Academic Press, New York, 1971 pp. 162–163.
2. Ogawa, T., Takeshita, Y., Miyamoto, T. and Chun, D. I., Poling field dependence of tetragonal PZT ceramics. *Ferroelectrics*, 1996, **186**, 119–122.
3. Yamada, A., Chung, Y. K., Takahashi, M. and Ogawa, T., Poling field dependence of ferroelectric domains in tetragonal lead zirconate titanate ceramics. *Jpn. J. Appl. Phys.*, 1996, **35**, 5232–5235.
4. Ogawa, T., Yamada, A., Chung, Y. K. and Chun, D. I., Effect of domain structure on electrical properties in tetragonal PZT ceramics. *J. Korean Phys. Soc.*, 1998, **32**, S724–S726.
5. Ogawa, T. and Nakamura, K., Effect of domain switching and rotation on dielectric and piezoelectric properties in lead zirconate titanate ceramics. *Jpn. J. Appl. Phys.*, 1999, **38**, 5465–5469.
6. Ogawa, T., Poling field dependence of crystal orientation and ferroelectric properties in lead titanate ceramics. *Jpn. J. Appl. Phys.*, 2000, **39**, 5538–5541.
7. Ogawa, T., Poling field dependence of ferroelectric properties in barium titanate ceramics. *Jpn. J. Appl. Phys.*, 2001, **40**, 5630–5633.