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Characteristics and magnetic field orientation of Eu-substituted $Sr_2NaNb_5O_{15}$-based lead-free piezoelectric ceramics

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1. Introduction

Piezoelectric components commonly use PZT ($Pb(Zr,Ti)O_3$). However, using lead-based piezoelectric materials poses serious health and environmental risks. It can cause pollution and threaten ecosystems. Therefore, searching for lead-free alternatives for SDGs is a must.

It was clarified that *c*-axis crystal-oriented $(Sr,Ca)_2NaNb_5O_{15}$(SCNN) ceramics have outstanding high-power properties than hard-PZT and also was shown superior performance as ultrasonic motor using the ceramics for the high-power ultrasonic devices in our previous studies[1,2]. However, to fabricate *c*-axis crystal-oriented SCNN ceramics, a rotating magnetic field of 2–3 T or higher is required, facilitated by a superconducting magnet (SCM), Reducing the magnetic field of the permanent magnet to 1 T or less with the *c*-axis oriented in the direction of the magnetic field is required.

In practice, we experimentally observed a change of crystal orientation under the magnetic field for the rare earth substituted SCNN[3]. We focused on SNN which is base of SCNN and investigated the crystal orientation behavior of rare earth substituted SNN ceramics under high magnetic field. The nonmagnetic ions (Eu^{3+}) changed from the *a*, *b*-axis orientation to the *c*-axis orientation in the direction of the magnetic field[4]. The Eu-substituted SNN ceramics were obtained with the degree of orientation of the *c*-axis of 0.9[4]. Eu substitution into SNN decreased the piezoelectric properties[4].

In this study, we investigated the crystal orientation behavior and ferroelectric properties of Eu-substituted SNN ceramics fabricated using high magnetic fields and the improvement of the ferroelectric properties by substituting Ca ions[5].

2. Experimental Procedure

The powders of SNN and Eu-substituted SNN (Eu content: x = 0-0.06 mol, denoted as SNN-Eux) were synthesized by a conventional solid-phase reaction. The SNN and Eu-substituted SNN powders were mixed to prepare a slurry with deionized water and dispersant, respectively.

Furthermore, the Ca-substituted SNN-modified powders were synthesized with Eu content of 0.06 mol (Ca content: x = 0.02-0.1 mol, denoted as SCNNx-Eu0.06).

The crystal-orientated behavior was evaluated by dropping the slurry dispersed powder into the water on the XRD grass holder and putting in the holder in the magnetic field of 5T by SCM (JASTEC JMTD-5T52M), after drying, measuring XRD. The crystal-oriented SNN-based ceramics were fabricated to sinter a green body after forming under a standing magnetic field of 5T. The green body was formed to put the container filled the slurry under the magnetic field until dry with a disk shape of ϕ10 x 2 mm. After sintering, the ceramic samples were prepared with a disk shape of ϕ8 x 0.5 mm.

The ferroelectric properties were evaluated by the oriented SNN-based ceramics compared with randomly oriented SNN-based ceramics.

3. Results and Discussion

The relationship between the Eu content and degree of orientation is shown in Fig.1[4]. Figures 2 and 3 show the hysteresis curves of the polarization applied electric field for *c*-axis oriented and randomly oriented SNN-Eu0.03 ceramics, respectively. The remnant polarization of oriented ceramics increased compared with that of randomly ceramics. On the other hand, the coercive electric field of oriented ceramics decreased than that of randomly ceramics. It represents easy to rotate the domains for *c*-axis oriented ceramics.

Figure 4 shows the XRD profiles of Ca-substituted SNN-modified powders with a standing magnetic field of 5T. The profiles represent *c*-axis orientation parallel to the magnetic field. The degree of orientation intends to increase with Ca substitution slightly.

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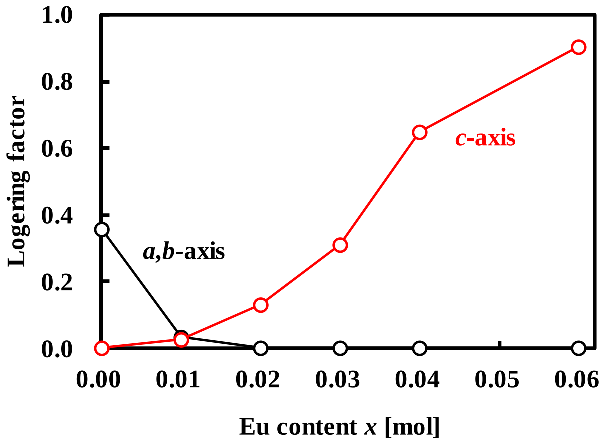

Fig.1. The degree of orientation of Eu-substituted SNN ceramics.

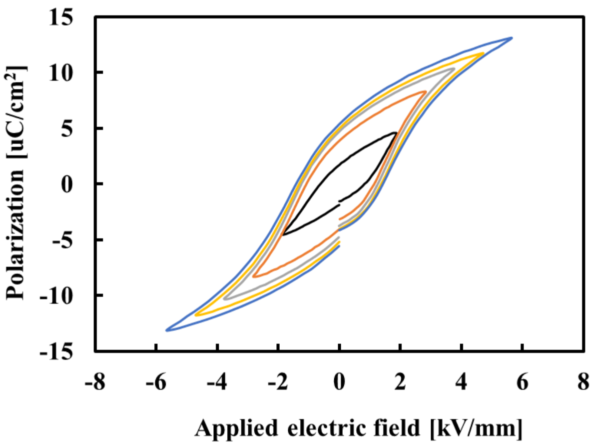

Fig.2. Hysteresis curves of *c*-axis-oriented SNN-Eu 0.03 ceramics.

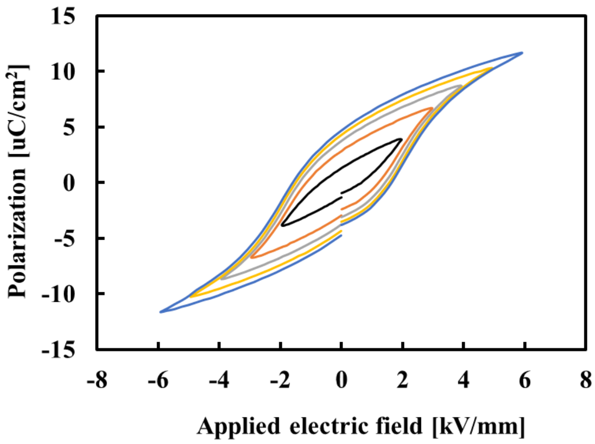

Fig.3. Hysteresis curves of randomly oriented SNN-Eu0.03 ceramics.

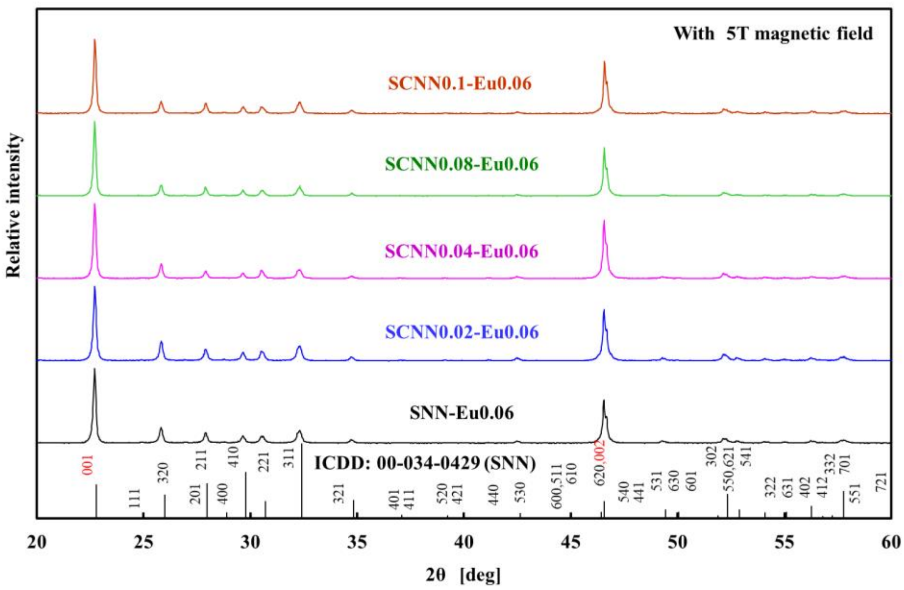

Fig. 4. XRD profiles of Ca-substituted SNN-Eu0.06 powders with magnetic field of 5T.