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Fabrication and Electrical Properties of (K, Na)$Bi_5Ti_5O_{18}$ -based Bismuth Layer-structured Ferroelectric Ceramics

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Abstract

Electrical and piezoelectric properties of (K,Na)$Bi_5Ti_5O_{18}$-based bismuth layer-structured ferroelectric ceramics were examined in this study. Especially, $Sr_{0.75}Ca_{0.25}Na_{0.5}Bi_{4.5}Ti_5O_{18}$ + CeO_2 1wt% [SCNBT] ceramics were prepared by solid phase reaction and hot forging methods and their electrical, ferroelectric and piezoelectric properties were evaluated. SCNBT ceramic were found to have an electromechanical coupling factor k_{33} of 0.13 and a piezoelectric constant d_{33} of 17 pC/N. Hot-forged SCNBT ceramic was found to exhibit good ferroelectric and piezoelectric properties with an k_{33} of 0.32 and d_{33} of 41 pC/N.

1. Introduction

Bismuth layer-structured ferroelectrics (BLSFs) have attracted attention as candidates for pressure sensor materials that can be used at relatively high temperatures (~600 ºC) because of their high Curie point T_c, low dielectric loss, and excellent fatigue properties [1-8]. Some of the BLSF ceramics with layer number m = 5, $(K_{0.5}Bi_{0.5})_2Bi_4Ti_5O_{18}$ (KBT5) and $Sr_{1-x}Ca_xNa_{0.5}Bi_{4.5}Ti_5O_{18}$ (SCNBT), have relatively good piezoelectric properties with a high T_c of 540 ºC and 607 ºC, respectively. In general, BLSF ceramics with higher m indicate lower T_c and larger piezoelectric constant d_{33} as compared with those with lower m number. On the other hand, these two compositions are attractive due to high T_c above 500 ºC. Therefore, if we can clarify and enhance the piezoelectric properties of these two compositions, they are attractive candidate materials for high temperature sensor applications with high d_{33} simultaneously. In this manuscript, we focus on the SCNBT ceramics.

Randomly oriented SCNBT ceramic indicates relatively high d_{33} of 31 pC/N [9]. It has also been reported that the addition of CeO_2 to BLSF ceramics results in lower dielectric loss and improved piezoelectric properties [10-14]. Furthermore, since the spontaneous polarization axis of BLSF is limited to the a and b planes, high piezoelectricity can be expected by applying grain orientation. Hot forging samples has been reported to improve d_{33} by 10 pC/N on average for the BLSF ceramics [15-17]. In this study, we examined the electrical properties of the SCNBT ceramics. In addition, their temperature stability was evaluated.

2. Experimental procedure

We weigh according to the composition formula $Sr_{0.75}Ca_{0.25}Na_{0.5}Bi_{4.5}Ti_5O_{18}$ + CeO_2 1 wt% [SCNBT] using $SrCO_3$, CeO_2, Na_2CO_3, $CaCO_3$, Bi_2O_3 and TiO_2 as starting materials, and a sample was prepared by an ordinary fired (OF). Ordinary fired ceramics were calcined at 850ºC -2 h and sintered at 1180ºC -2 h. Grain oriented ceramics were fabricated by using the hot-forging method (HF) at sintering temperature of 1140ºC for -6 h with forging pressure for -2 h during firing. The grain orientation factor, F, was calculated using the Lotgering method. Electrodes of fired-on Ag paste were formed for electrical measurements. For the piezoelectric measurement, the samples were poled at 9 kV/mm for 5 min at 200 ºC in a heated silicone oil bath. Measurement of X-ray diffraction (Rigaku RINT 2000), resistivity (Agilent 4339), dielectric temperature characteristic (YHP 4275 A), P-E hysteresis loop (Toyo Technica 6252 Rev. C) was conducted by conventional method.

3. Results and discussion

Fig. 1 shows SEM images of the grains of (a) OF and (b) HF SCNBT ceramics. Relative densities of OF and HF SCNBT ceramics were 90% and 95%, respectively. Those grains of OF and HF-SCNBT were observed to have a dense micro structure and plate-like grains, which is typical in BLSF ceramics. The grains of the OF-SCNBT were randomly oriented. On the other hand. HF-SCNBT ceramics showed were oriented in one direction.

Fig. 2 shows the frequency dependences of the impedance $|Z|$ and phase θ in the (33) mode for the (a) OF and (b) HF SCNBT ceramics. The values of k_{33}, d_{33} and mechanical quality factor Q_m for OF-SCNBT showed 0.13, 17pC/N and 556, respectively. On the other hand, the values for HF-SCNBT exhibited 0.32, 41 pC/N, 839, respectively. Hot-forged ceramics improved d_{33} by 20 pC/N and k_{33} by about 0.2 as compared to OF ceramics. Hot-forged SCNBT ceramics have better piezoelectricity.

4. Conclusions

OF-SCNBT ceramic were found to have k_{33} of 0.13 and a d_{33} of 17 pC/N. Then, HF-SCNBT ceramic were found to exhibit good ferroelectric and piezoelectric properties with k_{33} of 0.32 and d_{33} of 41 pC/N, which were almost twice as large as those of OF ceramics. Details of temperature stability will be presented on the day.

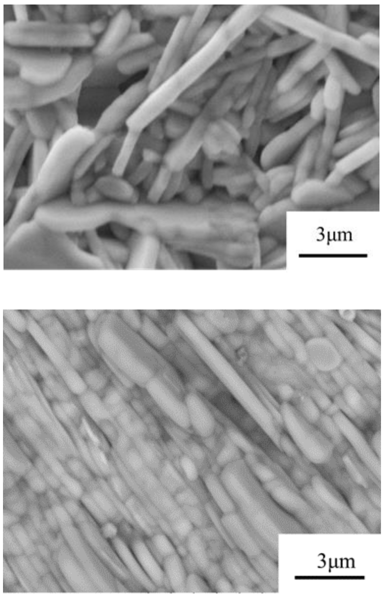

Fig. 1 SEM micrographs of (a) OF and (b) HF SCNBT ceramics.

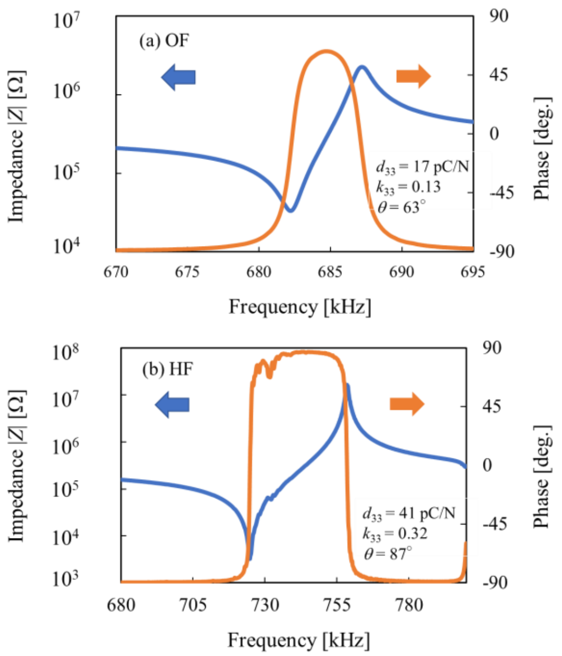

Fig. 2 Frequency dependences of the impedance $|Z|$ and phase θ in the (33) mode for the (a) OF and (b) HF SCNBT ceramics.

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