

Optimization of conditions of the synthesis of textured $(\text{Bi}_{0.5}\text{K}_{0.5})\text{TiO}_3$ piezoelectric ceramics by a reactive templated grain growth method

Univ. of Yamanashi¹, NIMS² (D)Gopal Prasad Khanal¹, Sangwook Kim¹, Ichiro Fujii¹,
Shintaro Ueno¹, Tohru Suzuki², and Satoshi Wada^{1,*}

*E-mail: swada@yamanashi.ac.jp

Piezoelectric ceramics have been widely used in various applications such as sensors, actuators, and transducers. Owing to the environmental concerns for the toxicity of lead, lead-free piezoelectric ceramics have been extensively studied, among which bismuth potassium titanate $(\text{Bi}_{0.5}\text{K}_{0.5})\text{TiO}_3$ (BKT) piezoelectric ceramic is one of the promising candidates because of its relatively high Curie temperature ($T_c \sim 400^\circ\text{C}$), high depolarization temperature ($T_d \sim 300^\circ\text{C}$), and a relatively high piezoelectric constant d_{33} of 125 pC/N.¹⁾ However, the d_{33} value is not sufficient for practical actuators and the d_{33} needs to be enhanced. Besides, BKT itself being the promising lead-free candidate, it is also important as end member for a variety of solid solutions like BKT-BaTiO₃, BKT- $(\text{Bi}_{0.5}\text{Na}_{0.5})\text{TiO}_3$, BKT-BaTiO₃- $(\text{Bi}_{0.5}\text{Na}_{0.5})\text{TiO}_3$, and so on. The grain-orientation imparted by the (001)/(100)-oriented BKT ceramics to the BKT-based solid solutions with crystal symmetry other than the tetragonal system could yield significant properties enhancement due to domain engineering. And, for the preparation of textured BKT ceramics, plate-like $\text{H}_{1.08}\text{Ti}_{1.73}\text{O}_{4.7}\cdot n\text{H}_2\text{O}$ (HTO) were used as template particles owing to their uniform-morphology with high aspect ratio. In this study, we focused on optimization of preparation conditions for the synthesis of (001)/(100) oriented BKT ceramics, using HTO- Bi_2O_3 - KHCO_3 and HTO- TiO_2 - Bi_2O_3 - KHCO_3 reaction system. The matrix TiO_2 particles were used in the second reaction system beside the HTO template particles because the ratio of the matrix to the template particles strongly affects the grain orientation of the RTGG-processed ceramics.²⁾

The grain-oriented BKT ceramics were prepared by a reactive templated grain growth (RTGG) method. To ensure the well-crushing, raw powders of Bi_2O_3 and KHCO_3 were weighed according to the chemical formula and they were ball-milled in an ethanol medium. The ball-milled slurry was completely dried and raw powders of HTO were added according to the chemical formula, and they were ball-milled with a binder solution. The slurry was tape-cast to form green sheets, which were cut, piled, and pressed at an elevated temperature to make green compacts. After organic components were removed by heating, the compacts were sintered with different sintering conditions such as normal, weighted, and embedded and weighted sintering (EWS) by employing one- to three-step heating programs. The relative densities of the sintered samples were measured by the Archimedes method. The crystal structure and microstructure was investigated by Cu K_α X-ray diffraction and scanning electron microscopy, respectively. Meanwhile, the grain-orientation factor (F_{100}) was calculated by the Lotgering method.

BKT ceramics, from HTO- Bi_2O_3 - KHCO_3 reaction system, with a relative density of 62% and F_{100} of 47% were fabricated by embedded and weighted sintering at 600°C (5 h)- 950°C (5 h)- 1050°C (5 h). However, the obtained relative density and F_{100} is not sufficiently high from which the appreciable piezoelectric properties enhancement could hardly be expected. The F_{100} value of 68% in $(\text{Bi}_{0.5}\text{Na}_{0.5})\text{TiO}_3$ piezoelectric ceramics was increased to 95% with optimizing the TiO_2 /HTO molar ratio.³⁾ Thus, the effect of various molar ratios of Ti-source from matrix TiO_2 and template HTO particles (TiO_2 /HTO ranging from 0:10-8:2) on F_{100} was investigated, using HTO- TiO_2 - Bi_2O_3 - KHCO_3 reaction system, as shown in Fig. 1. Relative density was found to increase with increasing the amount of TiO_2 matrix particles, while the F_{100} value was increased until the molar ratio of 6:4 and then decreased with further increasing content of TiO_2 matrix particles. With the optimum concentration of TiO_2 matrix and HTO template particles (molar ratio of 6:4), the density was increased from 62 to 72% with a increment of 16% than that for the BKT ceramics fabricated from the HTO- Bi_2O_3 - KHCO_3 reaction system, while the increment for F_{100} is from 47 to 66% exhibiting the enhancement of 40%.

- 1) H. Nagata *et al.*, Jpn. J. Appl. Phys. **49**, 09MD08 (2010).
- 2) E. Fukuchi *et al.*, J. Am. Ceram. Soc. **85**, 1461 (2002).
- 3) D. Hu *et al.*, J. Eur. Ceram. Soc. **34**, 1169(2014).

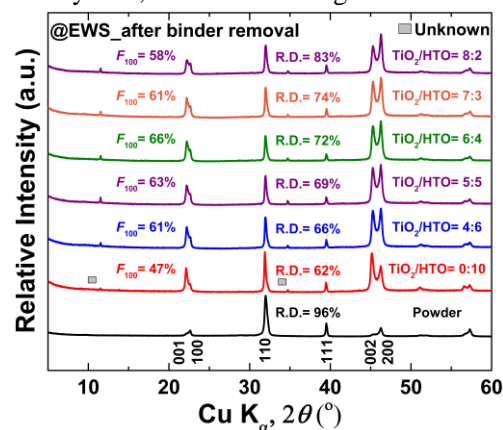


Fig. 1. XRD patterns of BKT ceramics prepared by EWS.