Fabrication of textured $(Bi_{0.5}K_{0.5})TiO_3$ piezoelectric ceramics via conventional sintering method

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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 [$(Bi_{0.5}K_{0.5})TiO_3$ or BKT] piezoelectric ceramic is one of the promising candidates because of its relatively high Curie temperature (T_c~400°C), high depolarization temperature (T_d~300°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-$BaTiO_3$, BKT-$(Bi_{0.5}Na_{0.5})TiO_3$, BKT-$BaTiO_3$-$(Bi_{0.5}Na_{0.5})TiO_3$, and so on. However, few studies on the fabrication of BKT and BKT-based ceramics by conventional sintering have been reported owing to their difficult densification due to narrow sintering window. In this study, we focused on optimization of preparation conditions for the synthesis of <100>-oriented BKT ceramics via conventional sintering, using HTO-Bi_2O_3-$KHCO_3$ and HTO-TiO_2-Bi_2O_3-$KHCO_3$ reaction system, where HTO refers to plate-like $H_{1.08}Ti_{1.73}O_4.nH_2O$ template particles with uniform-morphology and high aspect ratio. 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-orientation imparted by the <100>-oriented BKT mesocrystal particles to other BKT-based solid solutions, with crystal symmetry other than tetragonal, could yield significant properties enhancement due to domain engineering.

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 HTO templates were added and mixed with a binder solution by ball-milling. The slurry was tape-cast to form green sheets, which were cut, stacked, and pressed at 61 MPa for 10 min at 80 °C to make green compacts. After organic components were removed by heating, the compacts were sintered by conventional sintering method with or without weight-pressing and atmosphere powder. Meanwhile, the effect of heating program used during the sintering on the crystal structure, orientation, relative density, and microstructure was investigated. The relative densities of the sintered samples were measured by the Archimedes method. The crystal structures and microstructures were investigated by Cu K_α X-ray diffraction (XRD) and scanning electron microscopy, respectively. 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 three-step-temperature program embedded and weighted sintering (EWS). However, the obtained relative density and F_{100} is not sufficiently high. The F_{100} value of 68% in $(Bi_{0.5}Na_{0.5})TiO_3$ piezoelectric ceramics was increased to 95% with optimum 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 monotonically 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%.

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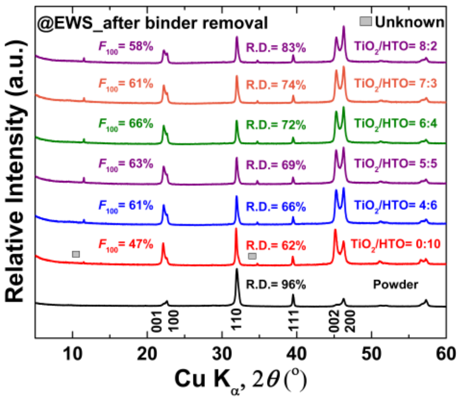

Fig. 1. XRD patterns of the as-sinterd BKT ceramics prepared by three-step EWS.