

# Structure analysis and electrical evolution observation in $\text{Bi}_{0.5}(\text{Na}_{(1-x)}\text{K}_x)_{0.5}\text{TiO}_3\text{-BiAlO}_3$ + f- $\text{Bi}_{0.5}(\text{Na}_{(1-x)}\text{K}_x)_{0.5}\text{TiO}_3$ ceramics

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In recent years,  $\text{Bi}_{0.5}(\text{Na}_{(1-x)}\text{K}_x)_{0.5}\text{TiO}_3\text{-BiAlO}_3$ (BNKT-BA) ceramics are considered to be promising candidates for the Bi-based piezoelectric materials due to its large strain and high depolarization temperature ( $T_d$ )[1]. Because of the relatively low remanent ( $P_r$ ) polarization value at the zero electric-field, however, soft BNKT-BA ceramics exhibit the non-polar phase feature. This non-polar phase require large poling field with phase transition to polar phase. In order to complement these drawback, hard ferroelectric ceramics such as ferroelectric- $\text{Bi}_{0.5}(\text{Na}_{(1-x)}\text{K}_x)_{0.5}\text{TiO}_3$  (f-BNKT) were usually added to decrease poling field[2]. Despite of a great number of reports about measurements piezoelectric properties, studies of microscopic domain evolution leading high piezoelectric efficiency are still rare. In this study, we investigated the microstructure of BNKT-BA + f-BNKT and electric field induced phase transition by transmission electron microscopy (TEM). We used a LaB6 TEM (JEM-2100, JEOL) for in situ electric field inducing experiment. Before apply electric field, bright field (BF) images, centered-dark-field (CDF) images and selected area electron diffraction (SAED) pattern of the ceramics were systematically analyzed. With the SAED patterns (FIG. 1 (a)), we could know the existence of the R3c phase and the P4bm phase in the ceramics by the superlattice spots of  $1/2(000)$  and  $1/2(00e)$ , indicating anti-phase (a-a-a-) and in-phase (a0a0c+) octahedral tilting, respectively. The morphology and distributions of these tilted domains are also shown in the CDF images (FIG. 1 (b) and (c)). Although we added the f-BNKT particles which size of 5~20  $\mu\text{m}$ , most of the BNKT-BA + f-BNKT grains are consisted of ferroelectric R3c phase core region and shell region of mixed P4bm and R3c phases as typical relaxor. The real-time change about phase and domain modification could be observed in FIG. 2 by in-situ TEM during electric field applied. This observation verifies the electric field-induced behavior in the only non-polar phase of BNKT-BA, and the effect of addition f-BNKT for phase transition of core-shell grain. The outstanding properties, such as large strain and high  $P_s$  might be due to phase transition with core(polar)-shell(non-polar) structure.

## References

- [1] A. Ullah et al., J. Am. Ceram. Soc. 94 (2011) 3915.
- [2] D.S. Lee et al., J. Appl. Phys. 112 (2012) 124109.

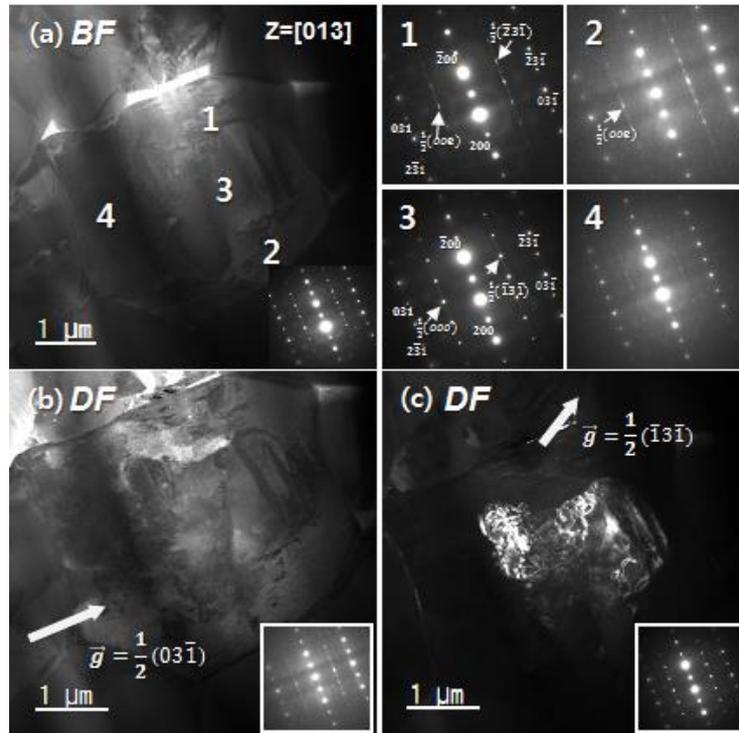


FIG. 1. TEM micrographs of core-shell grain : (a) Bright field image of [013] zone-axis grain and SAED patterns. (b), (c) Centered Dark field images using  $\vec{g} = \frac{1}{2}(03\bar{1})$  and  $\vec{g} = \frac{1}{2}(\bar{1}3\bar{1})$ , respectively.

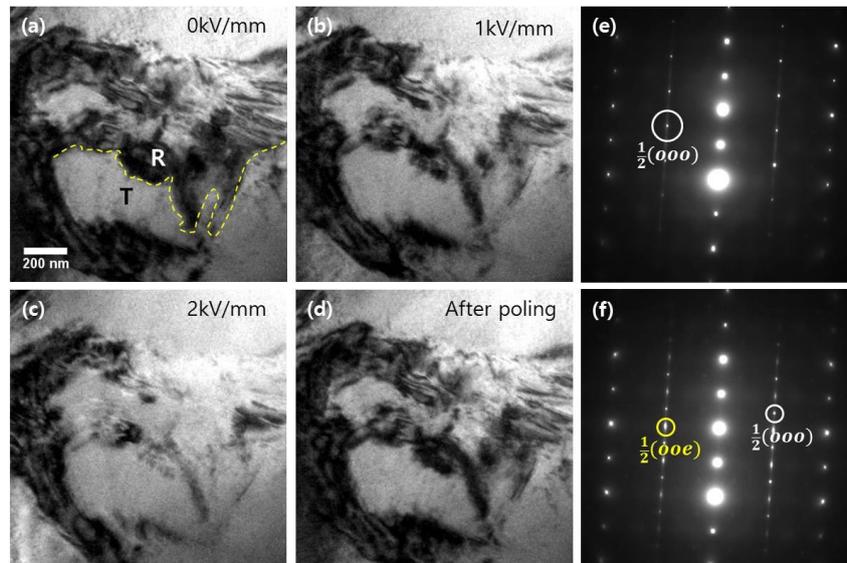


FIG. 2. TEM observations along the [013] zone axis of a core-shell grain under the electric fields. 0kV/mm. (b) 1kV/mm. (c) 2kV/mm. (d) After poling. (e), (f) SADE patterns from region R and T respectively.