

Gigantic Strain Induced by Electric Fields in Core/Shell Structured Alkaline Niobates Polycrystals

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In spite of intensive studies last decade as well reviewed in recent articles, the electric field-induced strain (EFIS) in the non-Pb-based actuator materials, e.g. alkaline-niobates ($K_{0.5}Na_{0.5}NbO_3$ (KNN) or bismuth-alkali titanates ($Bi_{0.5}Na_{0.5}TiO_3$) is not yet satisfactory enough to replace the Pb-based counterparts. Inspired by the PZT case, wherein a temperature-insensitive morphotropic phase boundary (MPB), many of the non-Pb based researches focused on realization of the MPB by chemical modification to the parent ferroelectrics like ($K_{0.5}Na_{0.5}NbO_3$ (KNN). However, such modified, non-Pb based piezoelectrics near the MPB mostly produce $d_{33}^* \approx 600$ pm/V at best, still inferior to the commercial PZT ($d_{33}^* \approx 400-700$ pm/V). In 2004, Saito *et al.*¹ used a pioneering way of texturing grains in a ceramic by use of crystallite templates to achieve $d_{33}^* = 750$ pm/V in the Li, Ta, and Sb-modified KNN. This method also produced the enhanced temperature stability over the non-textured one while $T_c \sim 250$ °C of the modified KNN was inevitably lower than that of KNN ($T_c \approx 400$ °C). The normalized strain d_{33}^* thus achieved still marks the largest among non-Pb based actuator materials. On the other hand, fabrication of a textured ceramic from a template control requires multiple processing efforts and a higher production cost, thereby becoming less competitive than the commercial PZT ceramics.

In this work, we have successfully fabricated (K,Na)(Nb,Ta)O₃-based polycrystalline ceramics, EFIS of which is superior to that of the textured KNN ceramics. Scanning transmission electron microscopy revealed that a core-shell configuration in each individual grain was favorably observed and thus such a duplex structure composed of a polar core and a nonpolar shell, as shown in Fig. 1. The giant electro-strain (Fig. 2) was thus explained by the core-shell model wherein electric field-induced strain can be enhanced by field-induced polarization propagations from the polar core to nonpolar shell region, as described in Fig. 3. Our results offer a new mechanism to achieve large electromechanical coupling in non-Pb based ceramics without resorting to complex domain texturing process¹. The concept of the core-shell structural model in principle is not restricted to the KNN based actuator materials but can be applied to broader class of non-Pb based piezoelectric materials. Therefore, in our CZ-modified KNLNT with giant strains, there exist a high possibility of producing low cost, high-performance lead-free piezoelectrics for various electromechanical applications

References

[1] Y. Saito et al., Nature 432 (2004) 84.

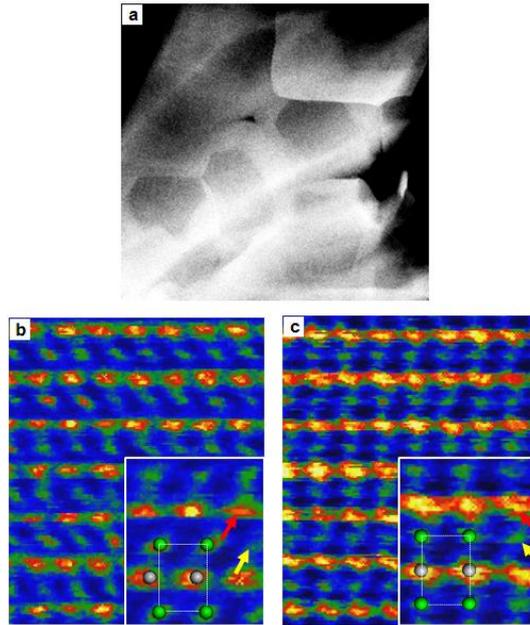


FIG. 1. (a) DF-STEM image for the core-shell structured grains in the sample. ABF-STEM images of (b) core region and (c) shell region in a CZ-modified KNLNT. The brighter spots denoted by the red arrow indicate the heavy *B*-site atomic column of Nb in the core and Nb & Ta in the shell; the less bright spots, the yellow arrow, indicate light *A*-site atomic column of K, Na and Li.

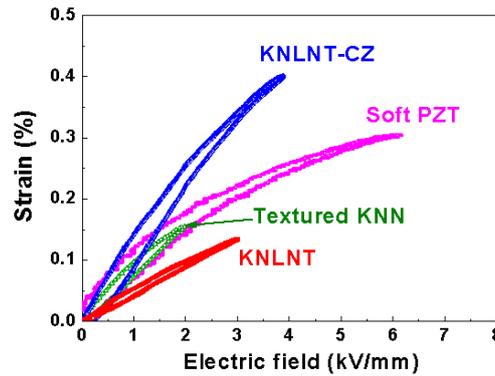


FIG. 2. Unipolar *S-E* loops of KNLNT and KNLNT-CZ in comparison with those of soft PZT and textured KNN ceramics.

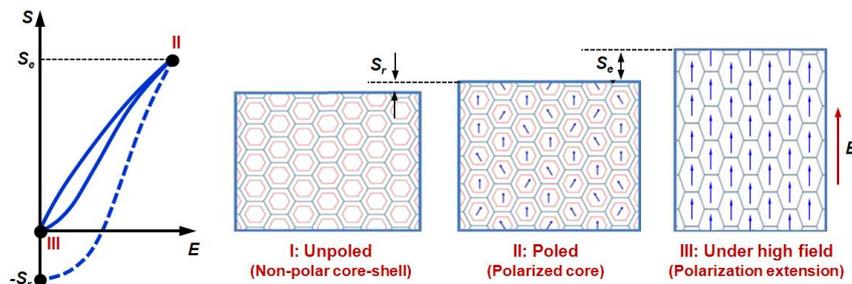


FIG. 3. Unipolar *S-E* curve for our KNLNT-CZ ceramic, which shows unpoled core-shell structures (state I), generation of polarization in the cores by poling (state II), and polarization propagation to the nonpolar shell region (state III) under applied fields. The dashed line in the *S-E* curve represents a poling process by electrical as well as thermal activation.