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Feature Article

Domain configuration and piezoelectric properties of $(K_{0.50}Na_{0.50})_{1-x}Li_x(Nb_{0.80}Ta_{0.20})O_3$ ceramics

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Abstract

Domain structure plays an important role in determining piezoelectric properties of ferroelectric materials. However, limited studies have been carried out on the domains of (K,Na)NbO_3-based lead free ceramics. The domain configuration, domain reversal behavior and piezoelectric properties of $(K_{0.50}Na_{0.50})_{1-x}Li_x(Nb_{0.80}Ta_{0.20})O_3$ (KNN-Li_x) ceramics with $x = 0.02$, 0.035 and 0.05, were studied in this research. It was observed that ceramics with different phases show distinctly different domain configurations and domain reversal behaviors. When compared to other two compositions, $x = 0.035$ with coexistent orthorhombic-tetragonal phases at room temperature was found to possess curved domain stripes and larger average domain width, leading to optimal piezoelectric properties with $d_{33}^* = 260$ pm/V and $k_p = 48\%$. Based on the microstructures, polarization hysteresis loops and unipolar strain curves under high electric field, it was concluded that the larger domain size and easier domain switching are due to the coexistence of orthorhombic and tetragonal phases, account for the improved properties in KNNT-$Li_{0.035}$ ceramics.

Keywords: (K,Na)NbO_3-based; Domains; Piezoelectric properties; Domain size; Domain switching

1. Introduction

The (K,Na)NbO_3-based (hereafter denoted as KNN-based) ceramics have been actively studied in last few years as lead-free alternatives to the conventional lead-based piezoelectric materials, such as Pb(Zr,Ti)O_3 (abbreviated as PZT) ceramics. PZT ceramics with morphotropic phase boundary compositions were found to possess superior dielectric and piezoelectric properties, being the mainstay materials for electromechanical devices such as ultrasonic transducers, actuators and sensors. However, due to the environmental regulations, such as RoHS, much attention has been paid to the exploration of lead-free piezoelectric ceramics. In last decade, lots of researches have been focused on the enhanced piezoelectric properties in KNN-based ceramics by optimizing sintering condition or compositional tuning.[1–15] Nevertheless, the enhanced piezoelectric properties obtained in the modified KNN ceramics at room temperature were believed to associate with the downward shift of the orthorhombic-tetragonal polymorphic phase transition (PPT) temperature[2–7,11] or the increase of the rhombohedral-orthorhombic polymorphic phase transition temperature.[15] As a consequence, some undesirable behaviors, such as thermal instability, time-aging instability and depolarization of the piezoelectric properties, were reported to exist in KNN-based ceramics. However, the property versus microstructure relationship is not yet established, leading to the question of the inherent origin of the high piezoelectricity at room temperature unresolved.

Generally speaking, the dielectric and piezoelectric properties of ferroelectric ceramics are attributed to the intrinsic and extrinsic contributions. The intrinsic contribution is related to the lattice distortion, whereas the extrinsic one is ascribed mainly to the domain wall motion.[16–20] The piezoelectric nonlinearities

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in ferroelectric ceramics under subcoercive field are believed to result from the non-180° domain wall motion.[21,22] Meanwhile, the domain instability accounts for the time-dependent and thermal instabilities of dielectric and piezoelectric properties. It is thus desirable to understand the role that the domain structure and domain stability play on the properties of ferroelectric ceramics. On the other hand, the Li, Ta-modified $(K_{0.50}Na_{0.50})NbO_3$ ceramics with the composition of $(K_{0.50}Na_{0.50})_{1-x}Li_x(Nb_{0.80}Ta_{0.20})O_3$ were reported to possess good piezoelectric properties,[7,11,23] but investigations on the domain structure have been rarely carried out so far. This article studied the piezoelectric properties, ferroelectric phases, domain patterns and domain reversal behavior reflected by P–E (polarization versus electric field) loops and S–E (strain versus electric field) curves.

2. Experimental

The $(K_{0.50}Na_{0.50})_{1-x}Li_x(Nb_{0.80}Ta_{0.20})O_3$ ceramics (with $x = 0.02$, 0.035 and 0.05, respectively) were prepared by conventional solid-state reaction method. The raw materials of K_2CO_3, Na_2CO_3, Li_2CO_3 and Nb_2O_5 were dried at 120 °C for 24 h to remove the absorbed moisture, and weighed according to the stoichiometric ratio. After ball-milling and drying, the mixed powders were calcined for 4 h at 920 °C, 900 °C or 870 °C according to the Li concentrations. The calcined powders were then ground, ball-milled and dried. The obtained powders were pressed into pellet disks with 15 mm in diameter and 1.5 mm in thickness at 100 MPa. Finally, sintering was carried out at 1145 °C, 1135 °C and 1125 °C for 2 h, respectively.

X-ray powder diffraction was performed on the ground as-sintered samples to determine the phase. Poling was conducted in silicon oil under the dc field of 3.5 kV/mm in the whole procedure, in which the temperature was first raised to 120 °C, kept for 30 min and then cooled down to room temperature. The piezoelectric measurements were carried out on the poled samples after 24 h aging. The k_p values were calculated from the resonance-antiresonance frequencies, measured by the impendence phase-gain analyzer (HP4194A). The piezoelectric coefficients d_{33} were measured by Berlincourt d_{33} meter. The electric field-induced strains were measured by a linear variable differential transducer (LVDT) driven by a lock-in amplifier (Model SR830; Stanford Research system, Sunnyvale, CA). The high field piezoelectric coefficients d_{33}^* were calculated from the unipolar S–E (strain versus electric field) curves. The polarization versus electric field (P–E) hysteresis loops were measured by Radiant precision workstation Premier II. The domain structures were revealed by chemical etching technique,[24] based on different etching rates for the positive and negative ends of the ferroelectric dipoles. The unpoled and poled ceramic specimens were polished and etched at room temperature in a mixed aqueous solution of HCl/HF acid. Domain patterns were observed by scanning electron microscopes (SEM).

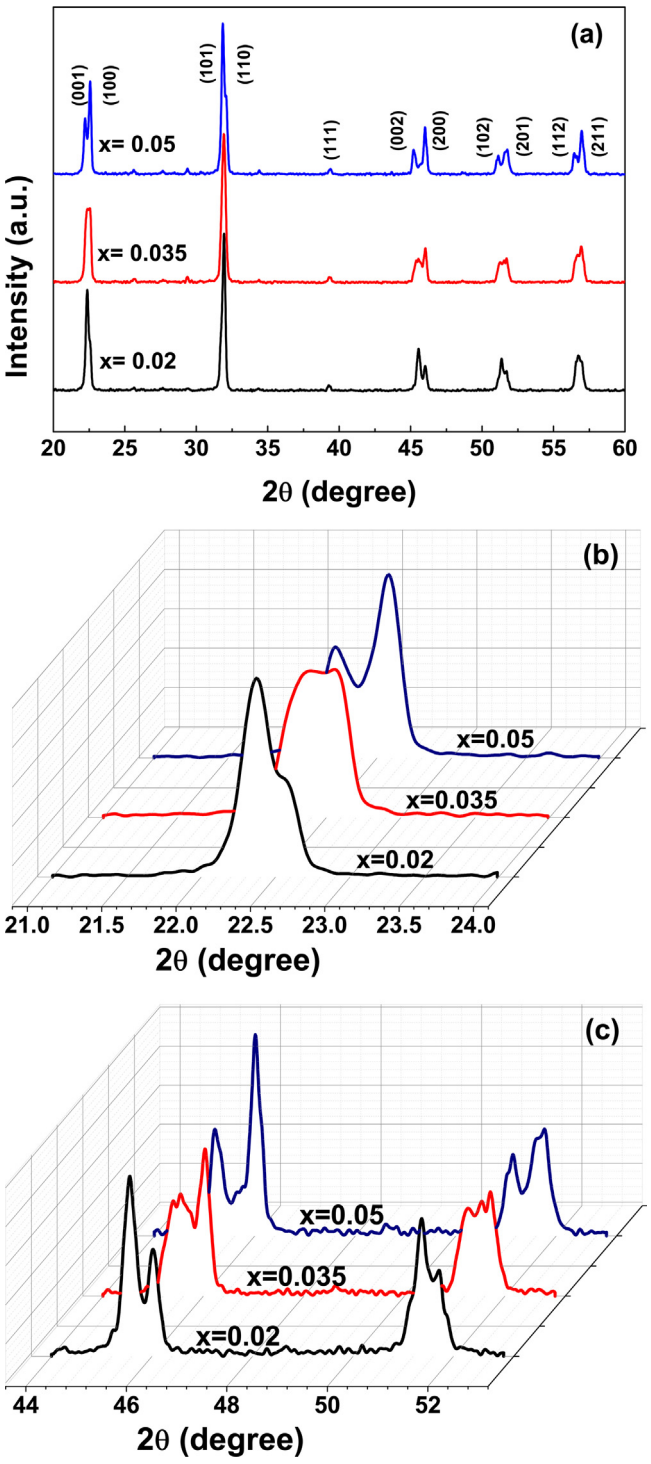

Fig. 1. XRD patterns for $(K_{0.50}Na_{0.50})_{1-x}Li_x(Nb_{0.80}Ta_{0.20})O_3$ ceramics: (b) and (c) are partly enlarged pictures of (a).

3. Results and discussion

Fig. 1(a) shows XRD patterns of the KNNT-Li_x with $x = 0.02$, 0.035 and 0.05, respectively. Fig. 1(b) and (c) are the partly enlarged views of Fig. 1(a). As can be seen, all these KNNT-Li_x ceramics exhibit the pure perovskite structure. It is believed that the relative intensities of diffraction peaks generally reflect