



Dielectric properties, relaxor behavior and temperature stability of $(1-x)(K_{0.4425}Na_{0.52}Li_{0.0375})(Nb_{0.87}Ta_{0.06}Sb_{0.07})O_3-xBa_{0.4}Sr_{0.6}TiO_3$ ceramics



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ABSTRACT

In this paper, $Ba_{0.4}Sr_{0.6}TiO_3$ was added in the $(K_{0.4425}Na_{0.52}Li_{0.0375})(Nb_{0.87}Ta_{0.06}Sb_{0.07})O_3$ ceramics to improve the temperature stability. The phase structure, microstructure, dielectric, diffuse phase transition, relaxor behavior and temperature stability of $(1-x)(K_{0.4425}Na_{0.52}Li_{0.0375})(Nb_{0.87}Ta_{0.06}Sb_{0.07})O_3-xBa_{0.4}Sr_{0.6}TiO_3$ (abbreviated as $(1-x)KNLNTS-xBST$) ceramics were investigated. The phase structure of these ceramics changed from tetragonal phase to cubic phase with $Ba_{0.4}Sr_{0.6}TiO_3$ additives. A significant reduction in the temperature dependence of relative permittivity occurred at $x = 0.15$, with $\Delta C/C_{298K}$ was around $\pm 15\%$ over the temperature range 273–528 K, and loss tangent, $\tan \delta \leq 0.03$. The result indicated that this material may have great potential for a variety of high temperature applications. The diffuse phase transition and the temperature of the maximum dielectric permittivity shifting toward higher temperature with increasing frequency, which were observed in the $(1-x)KNLNTS-xBST$ solid solution. The dielectric relaxor behavior obeyed a modified Curie-Weiss law. The dielectric relaxor was attributed to the appearance of polar nanoregions owing to the formation of random fields. The ferroelectric characteristic decreased with the addition of BST. These results confirmed that the $(1-x)KNLNTS-xBST$ ceramics can be regarded as an alternative for high temperature lead-free piezoelectrics.

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1. Introduction

Recently, much attention for lead-free ceramics have been paid to $K_{0.5}Na_{0.5}NbO_3$ (KNN)-based piezoelectric ceramics because of their relatively high piezoelectric properties and Curie temperature (693 K) [1–4]. The main problems of pure KNN ceramics are difficulty of preparing dense samples and relatively low electrical performances. In order to improve the densification and enhance the electrical properties, numerous substitution approaches such as introducing various metal ions into A- and/or B- sites have been investigated for several years. Since Saito reported a breakthrough in textured KNN-based ceramics with co-dopants of Li, Ta and Sb in 2004 [3], $(K,Na,Li)(Nb,Ta,Sb)O_3$ (abbreviated as KNLNTS) ceramics have attracted much attention of researchers, and recently become one of the most popular systems of KNN-based ceramics [5–9].

The improved electrical properties of ceramics could be attributed to the existence of the polymorphic phase transition (T_{O-T})

near room temperature [10]. Therefore, in order to obtain excellent electrical properties of KNLNTS based ceramics, it is necessary to make the T_{O-T} to approach room temperature. However, it has been found that these KNLNTS ceramics possessed low temperature stabilities, which limited the practical applications. In order to enhance the temperature stability of KNLNTS ceramics, modifications (NiO, AgTaO₃, BiCoO₃, and CaTiO₃, etc.) have been used but the favorable values can be hardly simultaneously obtained [11–17].

The $Ba_{1-x}Sr_xTiO_3$ is an excellent example of lead-free relaxor ferroelectrics with good electrical property and low T_{O-T} [18]. In this work, $Ba_{0.4}Sr_{0.6}TiO_3$ was introduced to the KNLNTS ceramics to form $(1-x)(K_{0.4425}Na_{0.52}Li_{0.0375})(Nb_{0.87}Ta_{0.06}Sb_{0.07})O_3-xBa_{0.4}Sr_{0.6}TiO_3$ (abbreviated as $(1-x)KNLNTS-xBST$) ceramics in order to improve electrical properties and temperature stabilities of KNLNTS ceramics. Moreover, it is reported that the addition of CuO in KNN-based ceramics can decrease the sintering temperature and improve the densification of the ceramics effectively [19–21], hence, 0.4mol% CuO was added to improve the densification of $(1-x)KNLNTS-xBST$ ceramics. The effects of the BST content on the phase structure, microstructure, dielectric, ferroelectric, temperature stability of the $(1-x)KNLNTS-xBST$ ceramics were investigated

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systemically.

2. Experimental

The conventional ceramic fabrication technique was used to prepare $(K_{0.4425}Na_{0.52}Li_{0.0375})(Nb_{0.87}Ta_{0.06}Sb_{0.07})O_3$ ceramics using analytical-grade oxides or carbonate powders: K_2CO_3 (99%), Na_2CO_3 (99.8%), Li_2CO_3 (99.5%), Nb_2O_5 (99.5%), Ta_2O_5 (99.99%), and Sb_2O_3 (99.5%). The stoichiometric powders were mixed by ball-milling with distilled water for 4 h and then dried and calcined at 1173 K for 5 h. $Ba_{0.4}Sr_{0.6}TiO_3$ powders were prepared by the solid state reaction method using $BaCO_3$, $SrCO_3$ and TiO_2 as starting materials, wet-milled with distilled water for 4 h, dried and calcined at 1423 K for 4 h. After calcination, KNLNST, CuO and $Ba_{0.4}Sr_{0.6}TiO_3$ powders were weighted according to the formula of $(1-x)(K_{0.4425}Na_{0.52}Li_{0.0375})(Nb_{0.87}Ta_{0.06}Sb_{0.07})O_3-xBa_{0.4}Sr_{0.6}TiO_3$ ($x = 0.00, 0.025, 0.05, 0.10, 0.15, 0.20$), and 0.2 mol% CuO (99%) powders were added. The mixed powders were ball milled for 4 h and dried. The obtained powders were mixed with 3 wt% polyvinyl alcohol (PVA) solution, and then uniaxially pressed into pellets with a diameter of 10 mm under 300 MPa pressure, subsequently, sintered in the air at 1373–1453 K for 3 h.

The crystal structures were examined by using an X-ray diffraction (D/max 2200 pc, Rigaku, Tokyo, Japan) with $CuK\alpha$ radiation ($\lambda = 0.15406$ nm). The microstructure morphologies were obtained by scanning electron microscopy (JEOL JSM-6390A JEOL Ltd. Tokyo). For the measurement of dielectric and piezoelectric properties, silver paste electrodes were formed on the two circular surfaces of the disk-shaped specimens after firing at 923 K for 5 min. Dielectric and ferroelectric measurements were carried by Agilent E4980A impedance analyzer and a ferroelectric analyzer (Premier II, Radiant, USA). Polarization was carried out in silicon oil bath at room temperature under a DC electric field of 3–4 kV/mm for 10 min. The piezoelectric coefficient (d_{33}) was measured using a piezo- d_{33} meter (2J4AN, China). The polarization versus electric field (P - E) hysteresis loops of the ceramics were measured using DGS-6C (Teak, USA).

3. Results and discussion

All the samples show density over 94% at optimistic sintering

temperature, and Fig. 1 shows the X-ray diffraction XRD patterns of $(1-x)KNLNST-xBST$ ceramics at room temperature. The samples show pure perovskite structure with no secondary phase with lower BST, while with high BST concentration, the secondary phase peaks can be seen between 2θ angles 25–30° and 30–40°, which are identified to correspond to the secondary phase $K_3Li_2Nb_5O_{15}$ as it has been indicated in Fig. 1. It is significant that the relative intensity of (002)/(020) peaks around $2\theta = 45^\circ$ change obviously, as shown in Fig. 1. It is well known that KNN-based ceramics usually show orthorhombic(O) and/or tetragonal(T) symmetry at room temperature, which can be identified by the following details in the XRD patterns [22] When the material is of orthorhombic phase with cell parameters $a \approx c > b$, $I(002)/I(020)$ roughly equals 2 and the (002) peak appears at a smaller Bragg angle, but the $I(002)/I(020)$ decreases to 0.5 for a tetragonal phase with $a = c > b$ [23,24]. In the present study, the $I(002)/I(020)$ of $(1-x)KNLNST-xBST$ ($x = 0$) ceramics is about 1, indicating the coexistence of orthorhombic and tetragonal phases in $(1-x)KNLNST-xBST$ ceramics when $x = 0.00$. With the addition of BST, the $I(002)/I(020)$ of samples changed. When $x = 0.025$, the $(1-x)KNLNST-xBST$ ceramic was tetragonal structure; when $x = 0.50$, the (002)/(020) peaks turned into one broaden single peak, it indicated the coexistent of tetragonal and cubic(C) structure; when $x = 0.20$, the (002)/(020) peak changed sharp, indicated the decrease of tetragonal structure, and $(1-x)KNLNST-xBST$ ceramic was cubic structure. This can be explained that the radii of Ba^{2+} (0.161 nm, CN = 12) and Sr^{2+} (0.144 nm, CN = 12) are much larger than that of Nb^{5+} (0.064 nm, CN = 6), which are very close to those of Na^+ (0.139 nm, CN = 12) and K^+ (0.164 nm, CN = 12) [25]. Therefore, the Ba^{2+} and Sr^{2+} are suitable for being introduced in the A site of the ABO_3 perovskite structure. Conversely, Ti^{4+} is introduced in the B site owing to the small ion radius (0.074 nm, CN = 6). Therefore, the addition of BST result in the distortion of $(1-x)KNLNST-xBST$ ceramic.

The lattice parameters of $(1-x)KNLNST-xBST$ ceramics could be determined by Scherrer formula:

$$1/d^2 = (h/a)^2 + (k/b)^2 + (l/c)^2 \quad (1)$$

The results were shown in Fig. 2, c decreased with increasing x , while a and b tended to be equal and increased together. When $x = 0.025$, the lattice parameters $a = b \neq c$, the $(1-x)KNLNST-xBST$ ceramics showed tetragonal structure at room temperature, when

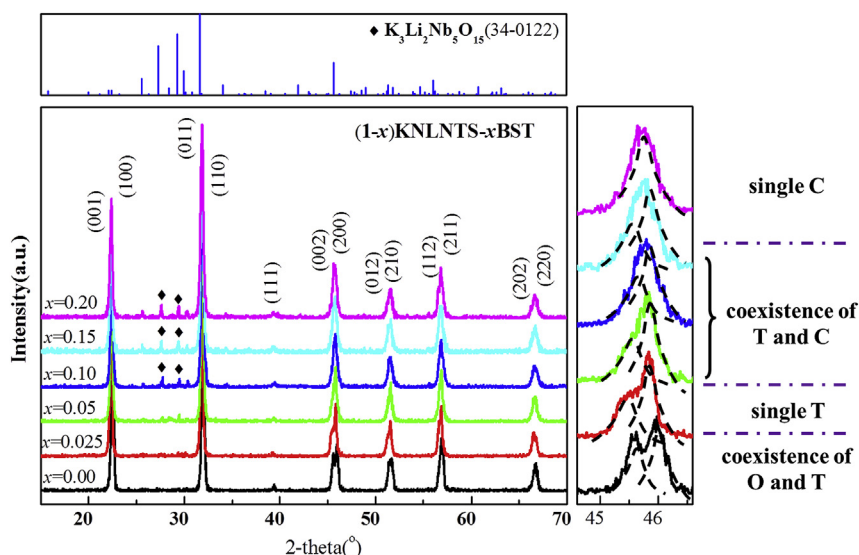


Fig. 1. X-Ray patterns of the $(1-x)KNLNST-xBST$ ($x = 0.025-0.20$) ceramics.

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