

Structural and electrical properties of $\text{Na}_{0.47}\text{K}_{0.47}\text{Li}_{0.06}\text{NbO}_3$ lead-free piezoelectric ceramics modified by AgSbO_3

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Abstract

$(1-x)\text{Na}_{0.47}\text{K}_{0.47}\text{Li}_{0.06}\text{NbO}_3$ (NKLN)– $x\text{AgSbO}_3$ lead-free piezoelectric ceramics were prepared using a reaction sintering method. The effects of AgSbO_3 doping on the structural and electrical properties of NKLN ceramics sintered at 1000–1040 °C were studied. The dopant affected densification, phase content, sintering temperature, microstructure and electrical properties. Variations in the relative intensity of X-ray diffraction peaks were consistent with Ag^+ and Sb^{5+} ions substituting on the perovskite lattice to produce a change in the proportions of co-existing tetragonal and orthorhombic phases. Grain growth during secondary re-crystallization was also affected. The temperature of the orthorhombic–tetragonal (O–T) phase transition and the Curie temperature (T_C) decreased as a result of AgSbO_3 modifications. The dielectric and piezoelectric properties are enhanced for the composition near the orthorhombic–tetragonal polymorphotropic phase boundary. The $0.92\text{Na}_{0.47}\text{K}_{0.47}\text{Li}_{0.06}\text{NbO}_3$ – 0.08AgSbO_3 ceramics exhibited optimum electrical properties ($d_{33}=252$ pC/N, $\epsilon_r=1450$, $\tan \delta=0.02$, and $T_C=280$ °C). These results reveal that $(1-x)\text{Na}_{0.47}\text{K}_{0.47}\text{Li}_{0.06}\text{NbO}_3$ – $x\text{AgSbO}_3$ ceramics are promising materials for lead-free piezoelectric application.

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1. Introduction

Lead zirconate titanate (PZT) has been developed over several decades to become the market-leading piezoelectric ceramic and used in a wide variety of transducer, sensor and actuator applications. However, legislation arising from health and environmental concerns has intensified research into finding suitable alternatives to lead-based piezoceramics.

A solid solution of ferroelectric KNbO_3 and anti-ferroelectric NaNbO_3 , such as $(\text{Na}_{0.5}\text{K}_{0.5})\text{NbO}_3$ (NKN), has attracted much attention as an alternative, lead-free piezoelectric materials because of their relatively high piezoelectric and ferroelectric properties, and high Curie temperature [1,2]. However, it is well known that dense and well sintered NKN ceramics are very difficult to obtain by ordinary sintering processes because of the high

volatility of alkali metal oxides at high temperatures. The hot-pressing techniques and spark plasma sintering (SPS) with high densities produce materials with excellent piezoelectric properties, but such processing techniques are not appropriate for industrial applications. To improve densification and electrical properties of NKN ceramics, many modifiers are incorporated into NKN to form new solid solution, such as NKN–BaTiO_3 [3], NKN–LiNbO_3 [4], NKN–LiSbO_3 [5] and NKN–LiTaO_3 [6].

In this study, $(1-x)\text{Na}_{0.47}\text{K}_{0.47}\text{Li}_{0.06}\text{NbO}_3$ – $x\text{AgSbO}_3$ ceramics were fabricated and sintered at lower temperature by a reaction sintering approach. The effects of AgSbO_3 content on phase structure, densification and electrical properties of $\text{Na}_{0.47}\text{K}_{0.47}\text{Li}_{0.06}\text{NbO}_3$ ceramics were then investigated.

2. Material and methods

The $(1-x)\text{Na}_{0.47}\text{K}_{0.47}\text{Li}_{0.06}\text{NbO}_3$ (NKLN)– $x\text{AgSbO}_3$ lead-free ceramics ($x=0.02$, 0.04 , 0.06 , and 0.08 mol) were

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prepared by a conventional mixed oxide method. The raw materials used in this study are K_2CO_3 (99.0%, Aldrich), Na_2CO_3 (99.9%, Sigma-Aldrich), Li_2CO_3 (> 99.0%, Fluka), Nb_2O_5 (99.9%, Aldrich), Ag_2O (99.0%, Sigma-Aldrich) and Sb_2O_5 (99.995%, Aldrich). Before weighing, the K_2CO_3 and Na_2CO_3 powders were separately dried in an oven at 150 °C for 24 h. A NKLN powder was prepared before reacting with AgSbO_3 . The starting powders were weighed according to the stoichiometric formula and then ball mixed in plastic jars with ZrO_2 balls for 24 h using ethanol as the medium. Mixtures were dried and calcined at 800 °C for 2 h. After that, the calcined powders were re-milled again for 24 h with Ag_2O and Sb_2O_5 to obtain $(1-x)\text{Na}_{0.47}\text{K}_{0.47}\text{Li}_{0.06}\text{NbO}_3-x\text{AgSbO}_3$ compositions. A reaction-sintering approach was used to produce the AgSbO_3 -modified NKLN ceramics, in that no second powder calcination step was employed. The obtained powders were subsequently pressed into green disks with a diameter of 16 mm. These compacts were sintered in air at low temperature ranging from 1000 to 1040 °C for 2 h in covered alumina crucibles. The sintered disks were polished in order to measure their electrical properties. Silver paste was painted on both surfaces of the samples to form electrode and then subsequently fired at 600 °C for 10 min. The ceramics were poled under a dc electric field of 3–5 kV/mm at 120 °C in silicone oil bath for 25 min.

The phase structures of the sintered $(1-x)\text{Na}_{0.47}\text{K}_{0.47}\text{Li}_{0.06}\text{NbO}_3-x\text{AgSbO}_3$ ceramics were determined by an X-ray diffractometer (XRD, X'Pert MPD, Philips), using a CuK_α radiation. The bulk density of ceramics was measured by the Archimedes method. The microstructures of sintered samples were observed using a scanning electron microscope (SEM, JSM-5800LV, JEOL). The piezoelectric constant d_{33} was measured by a piezo- d_{33} meter (YE2730A d_{33} Meter, APC International, Ltd.). Dielectric properties were obtained using a high precision LCR meter (LCR 821, GW INSTEK) at 1 kHz as a function of temperature under 1 V measuring voltage.

3. Results and discussion

Fig. 1 shows the XRD patterns of $(1-x)\text{NKLN}-x\text{AgSbO}_3$ samples as a function of x . The intensity ratio of the pair of peaks at $2\theta=45^\circ\text{--}46.5^\circ$ in each pattern was used as an indication of the tetragonal/orthorhombic phase content. In the case of a tetragonal NKN-type polymorph, the XRD peak multiplicities are such that the intensity of the lower angle (002) peak in the $45^\circ\text{--}46.5^\circ$ 2θ pair is around half of that higher angle (200) peak, $I_{002}/I_{200}=0.5$ [7]. For an orthorhombic NKN-LT phase, the peak intensities in the $45^\circ\text{--}46.5^\circ$ 2θ pair are reversed, giving $I_{220}/I_{002}=1.7$ [7]. A change in the relative intensities of certain main-phase peaks, for example the 002 and 200 peaks at $45^\circ\text{--}46.5^\circ$ 2θ , was apparent with changing AgSbO_3 content. It was found that addition of AgSbO_3 to NKLN had obvious influence on the phase structure of the ceramics. A mixture of orthorhombic and tetragonal

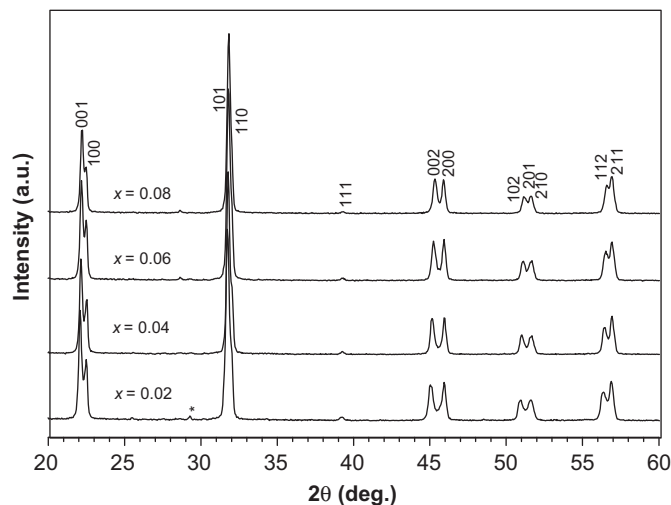


Fig. 1. X-ray diffraction patterns of the $(1-x)\text{NKLN}-x\text{AgSbO}_3$ (*= $\text{K}_6\text{Li}_4\text{Nb}_{10}\text{O}_{30}$).

Table 1

The Intensity ratios ($I_{002/200}$), orthorhombic–tetragonal polymorphic phase transition temperature (T_{O-T}), Curie temperature (T_C) and piezoelectric (d_{33}) constant of $(1-x)\text{NKLN}-x\text{AgSbO}_3$ samples.

AgSbO_3 content (mol)	$I_{002/200}$	T_{O-T} (°C)	T_C (°C)	d_{33} (pC/N)
0.02	0.95	82	380	157
0.04	0.96	44	346	208
0.06	0.97	40	322	210
0.08	1.02	30	280	252

phases is expected for all samples with $0.02 \leq x \leq 0.08$ from a measured intensity ratio (I_{002}/I_{200}) of ~ 1.0 , Table 1. This suggests that substitution of Ag^+ and Sb^{5+} ions on the perovskite lattice occurs and affects phase stability. The dopant may promote the stability of the tetragonal phase in the NKLN parent composition through a slight change in the position of the tetragonal–orthorhombic phase boundary on the NKN–LN phase diagram. In addition, faint extra peaks were present which were of similar d -spacings to a tungsten bronze phase (e.g. $\text{K}_6\text{Li}_4\text{Nb}_{10}\text{O}_{30}$). It was also found that peaks of secondary phase became lower with increasing AgSbO_3 content, indicating that the AgSbO_3 addition can reduce the formation of unstable secondary phases enhancing the stability. Moreover, it was also found that the positions of the diffraction peak of $(1-x)\text{NKLN}-x\text{AgSbO}_3$ ceramics shifted to higher angles slightly with the increase of x . This is due to the host Na^+ (ionic radius 1.39 Å) and K^+ (1.64 Å) are replaced by Ag^+ (0.67 Å) and Nb^{5+} (0.64 Å) is replaced by Sb^{5+} (0.76 Å), respectively [8]. As a result, the geometrical distortion of the ceramics was induced by the substitution of AgSbO_3 for NKLN lattice.

Fig. 2 shows the bulk density of the ceramics as a function of x and sintering temperature. The density of the ceramics increased quickly with the increase of AgSbO_3

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