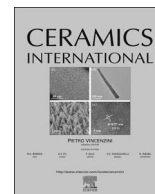




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Phase structure and electromechanical behavior of Li, Nb co-doped $0.95\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3-0.05\text{BaZrO}_3$ ceramics

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ABSTRACT

In this work, the effect of Li, Nb co-doping on the structural phase, dielectric, ferroelectric and field induced strain behavior of $0.95\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3-0.05\text{BaZrO}_3$ (BNT-BZ5) ceramic was investigated. X-ray diffraction patterns revealed the formation of single phase perovskite structure in the studied composition range. However, with increasing Li, Nb co-doping concentration, the maximum dielectric constant decreased and the dielectric maximum temperature (T_m) slightly shifted towards higher temperature. The field induced strain response increased from 0.18% for pure BNT-BZ5 to 0.38% for 1 mol.% Li, Nb modified BNT-BZ5 sample. The corresponding dynamic piezoelectric coefficient for these composition were ($S_{\text{max}}/E_{\text{max}} = 257 \text{ pm/V}$) and ($S_{\text{max}}/E_{\text{max}} = 542 \text{ pm/V}$), respectively. These results suggest that the BNLTN-BZ ceramic can be considered as a promising candidate material for piezoelectric application.

1. Introduction

Piezoelectric materials developed from the lead-based oxides are employed in various devices because of their outstanding response [1,2]. Nevertheless, lead-containing materials are toxic and face global environmental concerns [3,4]. Therefore, intensive research efforts have been focused on the improvement of lead-free materials to replace the existing lead-containing materials [5–12]. Among lead-free candidate materials, bismuth sodium titanate ($\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3$, BNT) gained much research interest because of its easy fabrication process and high polarization response [13]. Nonetheless, pure BNT has large coercive field and difficult to pole, therefore cannot be applied in practical applications.

Isovalent and/or aliovalent modifications on either A- or B-site of the BNT ceramics significantly affects its phase structure as well as the electromechanical properties [14–18]. Sasaki et al. investigated $\text{Bi}_{0.5}(\text{Na}_{1-x}\text{K}_x)_{0.5}\text{TiO}_3$ ceramics and reported their structural, dielectric and piezoelectric properties. Their structure analysis show a morphotropic phase boundary (MPB) of rhombohedral $\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3$ and tetragonal $\text{Bi}_{0.5}\text{K}_{0.5}\text{TiO}_3$ in the composition range of $x = 0.16 - 0.20$, where high electromechanical coupling factor, dielectric constant, piezoelectric constant and elastic compliance were observed [14]. In follow up research, Hiruma et al. [15] investigated the effect of divalent (Ca^{2+} , Sr^{2+} and Ba^{2+}) and trivalent (La^{3+} , Nd^{3+} , Ho^{3+} and Yb^{3+}) ions

and determined the depolarization temperature (T_d), the rhombohedral-tetragonal phase transition temperature (T_{R-T}), and the temperature of the maximum dielectric constant (T_m) on the basis of the temperature dependences of dielectric and piezoelectric properties and established a relationship between phase transition temperature and substituted ions on the A-site of the BNT ceramics. They demonstrated that T_m and T_d depend on the ionic radius of ions substituted in the A-site of BNT, T_{R-T} increases with increasing x in BNT-Ln100x, and that decreases with increasing x in BNCT100x, BNST100x, BNBT100x and BNPT100x, and form an MPB for large ions such as Sr^{2+} , Pb^{2+} and Ba^{2+} substitution on the A-site of BNT ceramics [15]. Besides A-site modification, BNT has been successfully modified on its B-site. Watcharapasorn et al. [16] studied the effect of Fe on BNT ceramics and reported that Fe^{3+} ions act as acceptor which induced the oxygen vacancies in BNT lattice that help to promote the densification and reduced its sintering temperature. Jaiban et al. [17] examined the influence of the isovalent Zr on the structure, dielectric and ferroelectric properties of the BNT ceramics. Their study describes three phase region: the orthorhombic phase ($0 \leq x \leq 0.2$), the mixed-phase ($0.3 \leq x \leq 0.4$) and the rhombohedral phase ($0.5 \leq x \leq 0.6$). The results from the high and low field dielectric responses indicated that the dielectric properties of both BNZ and BNZT ceramics were dominantly attributed to the reversible contribution. Yeo *et al.* reported Nb^{5+} and Ta^{5+} ($\leq 1.0 \text{ mol\%}$) diffuse into BNT lattice, decrease its average grain

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size and improve polarization, piezoelectric constant d_{33} while decrease its mechanical quality factor Q_m [18].

In addition to elemental doping, the solid solution of BNT with other perovskite materials such as BaTiO₃ (BT) [19], Bi_{0.5}K_{0.5}TiO₃ (BKT) [20], SrTiO₃ [21–23], and BaZrO₃ [24] exhibit good electromechanical response at certain compositions and therefore widely investigated by many researchers. Tetragonal BT as well as BKT forms an MPB with rhombohedral BNT, respectively at 5–7 and 15–20 mol. %. Both BNT-BT and BNT-BKT exhibit high electromechanical properties at MPB composition. Moreover, ST and BZ do not make a clear MPB with BNT [21,24], however, their electromechanical properties significantly enhanced at certain compositions. Nevertheless, the current performance of the BT, BKT, ST and BZ modified ceramics is not enough for device applications. It is well documented that multi-component systems of the piezoelectric materials provide better response than their binary or single component systems.

This work reports synthesis and electromechanical properties of Li, Nb co-doped BNT-BZ5 ceramics. All compositions were produced by traditional mixed oxide route and their structure, microstructure, phase transitions and strain properties were investigated a function of different Li, Nb concentration. The results suggest that selection of base composition and appropriate chemical modifier play important role in improving the field induced strain properties of the BNT-BZ ceramics.

2. Experimental procedure

Conventional mixed oxide route was followed to prepare bulk 0.95[Bi_{0.5}Na_{0.5-x}Li_xTi_{1-y}Nb_yO₃]-0.05BaZrO₃ (BNLTN-BZ5, with $x, y = 0-0.03$ mol.%) ceramics. Aldrich chemicals: Bi₂O₃ (99.99%), Na₂CO₃ (99.95%), Li₂CO₃ (99.99%), TiO₂ (99.99%), BaCO₃ (99.99%), Nb₂O₅ (99.99%) and ZrO₂ (99.98%) were used as starting raw materials. All powders were weighed according to their stoichiometric formula and then milled for 24 h in polyethylene bottles containing anhydrous ethanol and yttria-stabilized zirconia balls. After drying at 90 °C overnight, calcination was performed at 850 °C for 2 h. The calcined powders were milled and dried again. For pelletizing, polyvinyl alcohol (PVA) binder was added and passed to a 150 μm mesh sieve. The fine powders were pressed uniaxially into pellets of dimension 10 mm diameter and 0.5 mm thickness. The pellets were embedded in their corresponding powder and sintered at 1150 °C for 2 h in air.

X-ray diffraction machine (X'pert MPD 3040, Philips, The Netherlands) was used to check the crystal structure and phase purity. Scanning electron microscope (SEM, JP/JSM5200, Japan) was employed to observe the surface morphology and grain size of the samples. For electrical properties investigation, silver palladium paste was applied on both surfaces of the pellets and heat treated at 700 °C for 0.5 h to form electrodes. Impedance analyzer (HP4192A, USA) was utilized to measure the dielectric response at different frequencies. The P-E hysteresis loops were measured in silicon oil with the aid of a ferroelectric test system (Precision LC, Radian Technologies Inc., Albuquerque, NM) at 20 Hz. Field-induced strain response was measured using a contact-type displacement sensor (Millitron; Model 140) at room temperature.

3. Results and discussion

Fig. 1 presents the XRD plots of the Li, Nb co-doped BNT-BZ5 ceramics. All samples exhibited a single phase perovskite structure. No indications of impurity phases were observed for all compositions suggesting that Li, Nb ions successfully dissolved in BNT-BZ5 to form a complete and homogenous solid solution. Si powder was used as an internal standard for calibrating the XRD patterns and peak positioning. Close inspection of the XRD pattern suggests that pure BNT-BZ5 sample has a pseudo-cubic symmetry evident by its broad intensity peaks profile. However, Li, Nb modification in BNT-BZ5 results a slight

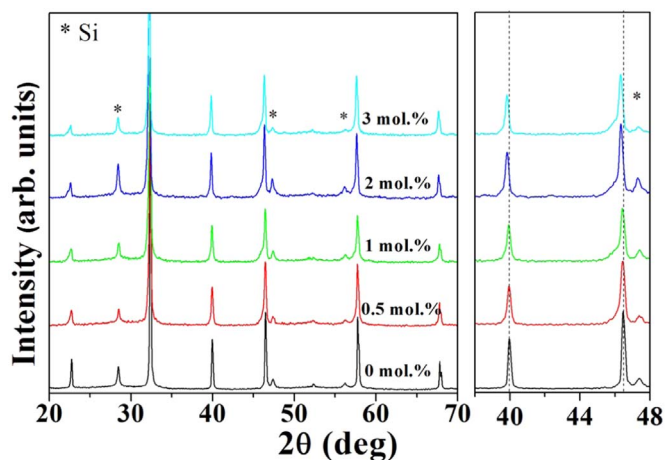


Fig. 1. X-ray diffraction profile of Li, Nb co-doped BNT-BZ5 ceramics.

hump at 2θ angle 46° indicating the appearance of a tetragonal phase. Moreover, a slight shift towards lower angle can be observed with increasing amount of Li, Nb, which can be attributed to BNT-BZ lattice expansion caused by the higher ionic size Nb⁵⁺ substitution for smaller ionic size Ti⁴⁺.

The SEM micrographs of the BNT-BZ5 ceramics with different Li, Nb contents are shown in Fig. 2. The micrographs reveal that grain sizes of the BNLTN-BZ5 ceramics continually decreased with the increasing amount of Li, Nb addition. In Li, Nb co-doped BNT-BZ5 system, it is expected that Li will occupy Na on the A-site and Nb will enter the B-site to occupy Ti. When Li⁺ (0.92 Å) enter A-site, it will substitute Na⁺ (1.18 Å) and when Nb⁵⁺ (0.64 Å) enter B-site, it will replace Ti⁴⁺ (0.605 Å) [25]. As Nb⁵⁺ and Ti⁴⁺ has charge difference, therefore Nb⁵⁺ substitution for Ti⁴⁺ will generate cation vacancies in BNT-BZ5 ceramics. Yeo et al. [18] suggested that in BNT ceramics, cation vacancies exist at grain boundaries and prohibit the mobility of grain boundaries that results in small grains. This behavior of grain size reduction is consistent with the Nb-doped PZT ceramics which has structural resemblance with the BNT ceramics [26].

The temperature dependent dielectric properties of the BNT-BZ5 ceramics co-doped with Li, Nb at different frequencies is provided in Fig. 3. Two distinct dielectric anomalies can be seen in dielectric curves of the samples: a strong frequency dependent hump around 150 °C commonly known as depolarization temperature (T_d) and relatively weak frequency dependent peaks at 300 °C called dielectric maximum (T_m), which suggest relaxor behavior of the materials. Jo et al. [27–29] suggested that the two dielectric anomalies could result from convolution of three different processes: a low temperature region where polar nanoregions (PNRs) of different symmetries coexists, intermediate temperature, where diffuse phase transition from low to higher symmetry PNRs occurs, and the high temperature regime attributed to the remaining high symmetry PNRs. Li, Nb co-doping in BNT-BZ5 significantly decrease the T_m values. Pure BNT-BZ5 has a T_m value of ~2800 which reduced to 2400 for 2 mol. % Li, Nb modified BNT-BZ5 sample. Moreover, the dielectric curves become more diffused and show lower temperature dependence with increasing amount of Li, Nb content. The T_m values decrease however, the room temperature dielectric constant increase with increasing amount of Li, Nb. At measuring frequency of 100 kHz, the room temperature dielectric constant of undoped BNT-BZ5 is 900 which enhanced to 1200 for the 2 mol% BNT-BZ5. Furthermore, the T_d gradually shifts to lower temperature and T_m move towards higher temperature. The T_d curves become more visible with doping higher Li, Nb content. It is only a hump for BNT-BZ5, however, becomes a distinct peak for 2 mol% Li, Nb modified BNT-BZ5 specimen.

Fig. 4 shows the P-E hysteresis loops of Li, Nb co-doped BNT-BZ5 ceramics at a measuring of 20 Hz at an applied electric field of 70 kV/

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