Enhanced electric field-induced strain and electrostrictive response of lead-free BaTiO$_3$-modified Bi$_{0.5}$(Na$_{0.80}$K$_{0.20}$)$_0.5$TiO$_3$ piezoelectric ceramics

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ABSTRACT

Lead-free (1-x)Bi$_{0.5}$(Na$_{0.80}$K$_{0.20}$)$_0.5$TiO$_3$-xBaTiO$_3$ or (1-x)BNKT-xBT (x = 0 – 0.15 mol fraction) piezoelectric ceramics have been investigated. The optimum density was found for the ceramic sintered at 1125°C, corresponding 98 – 99% of their theoretical values. All ceramics had a single perovskite structure. The coexisting mixed tetragonal-rhombohedral phases all over the entire compositional range with tetragonal phase becoming superior at higher BT. The diffuse phase transition was promoted by the addition of BT. With increasing BT content, the $T_{nr}$ and $T_{D}$ values were found to decrease. Phase transition from ferroelectric (FE) phase to ergodic relaxor (ER) phase was also induced by the changes in BT content. These changes significantly suspended long-range ferroelectric order, then correspondingly reducing the $P$, and $E_{r}$ resulting in an improvement of electrostrictive and electric field-induced strain responses. A significant increasing of electric field-induced strain response ($S_{max} = 0.37\%$ and $d_{33}^{*} = 630$ pm/V) is noted for the $x = 0.05$ ceramic. Furthermore, high electrostrictive coefficient ($Q_{33}$) of 0.0421 m$^2$/V$^2$ is observed for this composition, which was more than ~ 2 times (~108%) as compared to the pure BNKT ceramic. The studied results indicated these ceramics can be considered promising candidates for actuator applications.

1. Introduction

Lead-based oxide ceramics, such as Pb(Zr$_{1-x}$Ti$_x$)O$_3$ or PZT, have shown excellent electric field-dependent strain performance in the closeness of the morphotropic phase boundary or MPB region [1,2]. Unfortunately, lead element has a serious hazard to human and is harmful to the environment and surroundings [1–3]. As well as concerning the legislations oppose the use of toxic elements, several of the hard researches have been devoted to exploring the competitive lead-free counterparts, aiming to substitute the PZT-based oxide ceramics in the future [3–5]. Extensive researches have been carried out and search for lead-free piezoceramics over the past decade, for example, Bi$_{0.5}$Na$_{0.5}$TiO$_3$ (BNT), BaTiO$_3$ (BT), or K$_{0.5}$Na$_{0.5}$NbO$_3$ (KNN)-based systems [6,7].

As a promising candidate, bismuth-based piezoelectric ceramics, for example, bismuth sodium titanate (Bi$_{0.5}$Na$_{0.5}$TiO$_3$ or BNT), might suggest potential alternative of PZT-based oxide ceramics [8]. The Bi$^{3+}$ ion has 6s$^2$ lone pair electrons similarly to Pb$^{2+}$ ion, which can cause high ferroelectricity and good polarizability; so the BNT ceramic has attracted extensive attention [1]. The pure BNT ceramic has a typical perovskite structure with rhombohedral phase at ambient temperature and possesses a large remnant polarization ($P_r \sim 32$ $\mu$C/cm$^2$) and high Curie temperature ($T_C \sim 320\, ^\circ\, C$) [3,8–10]. However, the BNT ceramic has many drawbacks, such as: (1) it has a large coercive field ($E_c$) and high electrical conductivity, so it is easy to broken down during the polarization, (2) Na and Bi easily volatilize at high temperatures (HT), resulted to reduce the density of the material, and (3) BNT ceramic cannot satisfy the actual application of capacitors due to the dielectric constant ($\varepsilon_r$) at room temperature (RT) is only ~ 800 [5]. Moreover, BNT ceramics had a lower normalized strain coefficient ($d_{33}^{*} = S_{max}/E_{max}$), which limit its application in the electronic devices [3]. In order to overcome these drawbacks of the pure BNT ceramic, it must be necessary to increase the performance of BNT-based systems via a chemical modifications [10].

The compositional modifications are greatly desirable to reduce the $E_c$ value and improve the electrical properties, such as piezoelectric, electric field-induced strain, ferroelectric, and dielectric properties, through the construction of MPB between the rhombohedral R3c (Bi$_{0.5}$Na$_{0.5}$TiO$_3$, BNT) and tetragonal P4bm (Bi$_{0.5}$Na$_{0.5}$TiO$_3$, BKT) [11,12]. A classical of the Bi$_{0.5}$(Na$_{0.5}$K$_{0.5}$)TiO$_3$ (BNKTx) system exhibits excellent electromechanical response, piezoelectric, and ferroelectric at MPB of 16 – 20% BKT content [7,11,13]. The improvement in electrical properties (i.e. dielectric and piezoelectric properties) of the BNKTx ceramics at MPB was found to be similar to that observed in the PZT ceramic. The giant electromechanical strain has occurred when the
BNKTx ceramics at MPB was modified with other ABO₃ compounds [7].

To date, a chemical modification of the BNKT system for achieving high electric field-induced strain has been extensively investigated. A large $d_{33}$ of 617 pm/V was obtained in the lead-free CaZrO₃-modified Bi₀.₅Na₀.₇₈K₀.₂₂O₃TiO₃ ceramics, which reported by Hong et al. [14]. This result supports a prediction that a large strain at the ferroelectric-relaxor transition region can be observed when the tolerance factor of the perovskite is reduced by the substitution of ABO₃-type modifiers. The composition-dependent strain performance of the binary system 0.96Bi₀.₅Na₀.₇₈K₀.₂₂O₃TiO₃ -0.04Na₁₋ₓLiₓNbO₃ or BNKT-100xLi (x = 0 – 1.0) was investigated by Li et al. [15]. The highest unipolar strain of 0.443% and bipolar strain of 0.45% was observed at $x = 0.75$ under an applied voltage of 70 kV/cm with corresponding of the $d_{33}$ of 633 pm/V. The effects of (Fe₀.₅Nb₀.₅)O₃ or FN modified on the piezoelectric, electric field-induced strain, ferroelectric, and dielectric properties of the BNKT ceramic were studied by Lv et al. [16]. A large unipolar strain of 0.42% with corresponding $d_{33}$ value of 642 pm/V for the ceramic with $x = 0.04$ are observed (under an applied voltage of 6.5 kV/mm) because of the phase transition from ER to FE. These results indicate that BNKT modified by FN ceramics would have great potentials for electromagnetic actuator applications. The crystal structures of the lead-free Bi₀.₅(Na₀.₇₅K₀.₂₅)₀.₃(Ti₁₋ₓSnₓ)O₃ ceramics (BNKT-xSn, $x = 0 – 0.08$) were investigated together with their strain behavior by Xi et al. [17]. Both I-E and P-E loops confirmed the coexistence of ER and FE phases at the $x = 0.02$ ceramic, where a high maximum strain ($S_{max}$) of 0.37% was achieved with low degree of strain hysteresis ($H = 21.8$%). The high strain value is contributed to the phase transition from FE into ER. Moreover, the low degree of strain hysteresis is helpful from the existence of ER phase, which should pave the way for future developing high-performance actuators.

Among various lead-free materials, barium titanate (BaTiO₃ or BT) is one of the most widely studied lead-free ferroelectric material due to its superior properties, that is, high dielectric constant, high electromechanical coupling factor ($k_{33}$), and low dielectric loss characteristics [18]. The BT ceramic was the first oxide perovskite type ferroelectric material developed and thoroughly studied, ever since its discovery more than 60 years ago [19,20]. It has been well recognized as the first ferroelectric oxide that has been utilized to the manufacture electronic components and studied by three independent Japanese, Russian, and American teams since 1943 [21,22]. At temperature greater than 120°C, BT ceramic has a paraelectric with cubic phase. In the temperature ranging 5 – 120 °C, the tetragonal ferroelectric phase with the tetragonality $(c/a)$ ratio of $\sim 1.01$ was obtained. It is an important ceramic material for the electronic and memory devices [18].

Takenaka et al. [23] found that the addition of BaTiO₃ (BT) can improve the electrical properties of the Bi₀.₅Na₀.₅TiO₃ (BNT) ceramic. The MPB between BNT rhombohedral and BT tetragonal phases was found at the composition of $x = 0.06 – 0.07$, which highly enhanced dielectric ($\varepsilon_r = 580$ and $T_C = 288$ °C) and piezoelectric ($d_{33} = 125$ pC/N and $k_{33} = 55$%) properties. The depolarization temperature ($T_d$), which is the rhombohedral-tetragonal phase transition temperature, between 100 – 130 °C is reported. They suggested that the BNT-BT ceramics with the MPB composition are superior as piezoelectric properties, which this composition has a potential to be one of a promising lead-free piezoelectric candidates for the actuator applications. Solid solutions of the (1-x)BNT-xBT were also studied by Badapanda et al. [24]. They reported that high dielectric properties ($\varepsilon_{max} = 9672$, $T_m = 280$ °C), large strain performance ($S_{max} = 0.038%$, $d_{33} = 194$ pm/V), and highest remnant polarization ($P_r = 48.16 \mu C/cm^2$) were observed in the MPB composition of $x = 0.08$, which makes this composition suitable for actuator, memory, and transducer applications.

Therefore, the composition of Bi₀.₅(Na₀.₇₅K₀.₂₅)₀.₃TiO₃ or BNKT, which is the MPB composition within the tetragonal and rhombohedral phases (an increasing of the number of polar axes), was selected in this work as the base material due to its a has a good piezoelectric properties at MPB region. The BaTiO₃ (tetragonal phase) was chosen as part of the solid solution with the BNKT ceramic because of its superior dielectric and ferroelectric performances, that make it extremely important system for fundamental electroceramics research. This research thus aimed to fabricate the binary system of (1-x)BNKT-xBT ceramics. We have studied in detail of the phase structure, relaxor behavior, ferroelectric, and dielectric properties with varying concentration of the BT additive. To our knowledge, the effects of BT concentrations on the electric field-induced strain response of the BNKT ceramic have not yet been done so far. So we have focused on the study of the electric field-induced strain properties. In addition, in order to observe the possible correlation between $Q_{33}$ as a function of BT concentration in this study, we investigated the electrostrictive properties of the BNKT-BT ceramics with a large variation of $x$ from $x = 0$ to $x = 0.15$, and revealed a linear relationship between $Q_{33}$ as a function of BT concentration. Optimum composition with an enhancement electrical properties was obtained. It is expected that the new system has great potential for excellent electrical properties as compared to the pure BNKT ceramic or other systems.

2. Experimental procedure

The lead-free ceramics of (1-x)Bi₀.₅(Na₀.₇₅K₀.₂₅)₀.₃TiO₃-xBaTiO₃ or (1-x)BNKT-xBT was prepared by a conventional mixed-oxide method. The starting materials were Bi₂O₃ (99.9%), Na₂CO₃ (99.5), K₂CO₃ (99%), TiO₂ (99.9%), and BaCO₃ (98.9%). For removing
any moistures from the powders, all carbonate powders were first dried for 24 h at 120 °C. The raw materials of BNKT and BT powder were separately stoichiometrically weighed, ball-milled in high purity ethanol solution for 24 h, and dried in an oven at 120°C for overnight. The BNKT and BT powders were separately calcined at 900 °C for 2 h and 1200 °C for 2 h, respectively, in a closed Al₂O₃ crucible. After calcination, the BNKT and BT calcined powders were then weighed and mixed again to produce the mixed powders of (1-x)BNKT-xBT (x = 0 – 0.15 mol fraction). The calcined powders were sieved, and after that a few drops of 3 wt% polyvinyl alcohol (PVA) were added as a binder. The mixed powders were pressed into disks with a thickness of 1.3 mm and 10 mm in diameter. The green pellets were embedded into the powder of the same composition for minimizing the loss of the volatile elements and were then sintered in a covered alumina crucible at 1125 °C for 2 h and a heating/cooling rate of 5 °C/min.

The X-ray diffractometer (XRD-Phillip, X-pert) was used to characterize the crystal structures of all ceramics. The Raman spectra was carried out on the polished sintered samples and recorded by Raman spectroscopy. Bulk density was measured with Archimedes' method. A scanning electron microscope (SEM, JEOL JSM- 6335 F) was used to determine the microstructure of all ceramics. Grain size was calculated from the SEM micrographs by a mean linear interception method. All samples were carefully polished to obtain the parallel scratch-free surfaces (thickness of 1 mm) before being subjected to electrical characterizations. To form an electrode, the silver conducting paste was fired onto two sides of each sample at 700 °C for 15 min. Dielectric properties were measured from 25 to 500 °C with various frequencies of 1, 10, 100, 500, and 1000 kHz using a 4292A LCR meter (4276A, Hewlett Packard). The thermally stimulated depolarization current (TSDC) was used to determine the depolarization temperature or T_d of all ceramics. The current density-temperature (J-T) from the TSDC technique was also plotted. A ferroelectric test system with radiant technology (Precision High Voltage Interface) was used to measure the polarization-electric field (P-E) hysteresis loop. An AC electric field of 60 kV/cm (at 1 Hz) was applied to each ceramic, and the P-E hysteresis loops were also recorded. The maximum polarization (P_max), the maximum electric field (E_max), the remanent polarization (P_r), and the coercive field (E_c) were determined from the P-E hysteresis loops. Room temperature strain-electric field (S-E) were obtained under an electric field of 60 kV/cm (@ 0.1 Hz) using an optical displacement sensor (Fotonic Sensor, MTI-2100) combined with a radiant technology system. The negative strain (S_nag) and the maximum strain (S_max) were calculated from the bipolar S-E curve. The normalized strain coefficient (d^s_{33}) was also determined with the following equation: d^s_{33} = S_{max}/E_{max}. The electrostrictive coefficient (Q^s_{33}) was determined from the S-P curves. For piezoelectric measurements, all ceramics were poled for 15 min at 25 °C with an applied DC electric field of 5 kV/mm in a stirred silicone oil bath. The low-field piezoelectric coefficient (d^33) was then recorded after the 1-day aged ceramic at a frequency of 50 Hz using d^33-meter (KCF technologies, SS865).

3. Results and discussion

3.1. Phase evolution, Raman analysis and microstructure

Figure 1(a) displays X-ray diffraction patterns of the (1-x)BNKT-xBT ceramics with 2θ = 20 – 80°. All the XRD patterns are similar to the previous BNT-based ceramics in many literatures [10,25,26]. No extra peaks other than BNKT-based perovskite were observed, which indicated that all ceramics present a single perovskite structure. With increasing BT content, the slightly shifting of XRD peaks to lower 2θ angles was observed, which indicated the increase in the unit cell volume. This was expected due to the inclusion of Ba^{2+} (1.42 Å) on the A-site [Bi^{3+} (1.17 Å), Na^+ (1.18 Å), and K^+ (1.33 Å)] and Ti^{4+} (0.74 Å) on the B-site of the host BNKT perovskite structure [27]. The detailed view of XRD analysis at 2θ = 39° – 41° and 44° – 48° is shown in Figure 1(b) and 1(c), respectively. A marginally splitting of (111)_{B}/(111)_{A} and (200)_{T}/(200)_{T} demonstrated that the pure BNKT ceramic (x = 0) had a phase mixture of rhombohedral-tetragonal phases [4]. However, the coexistence of the mixed rhombohedral-tetragonal phases transformed into a tetragonal-rich phase at higher BT content. This can be evidenced by the existing of the splitting (200)_{T}/(002)_{T} peaks, and the merging of the (111)_{B}/(111)_{A} peak into a single (111) peak, thus indicating that the amount of tetragonal phase is higher than that of the rhombohedral phase when the BT content was increased. To check the characteristic of phase transformation in more details, the tetragonality ratio (c/a) was calculated and its values are summarized in Table 1. The c/a value increased from 1.0115 for the x = 0 ceramic to around 1.0163 for the x = 0.15 ceramic. This was corresponding with the phase transition from mixed rhombohedral-tetragonal to tetragonal-rich phase as the modifier was increased as mentioned above.

Raman spectroscopy was utilized to complement the XRD results in order to further confirm the phase transition behavior of the studied ceramics. Raman spectroscopy usually reflected the large length-scale average structures [28,29]. Based on many related literatures, the deconvolution of the ceramics was applied which was performed according to eight
Figure 1. X-ray diffraction patterns of the (1-x)BNKT-xBT ceramics where (a) $2\theta = 20 - 80^\circ$, (b) $2\theta = 39 - 41^\circ$, and (c) $2\theta = 44 - 48^\circ$.

Table 1. Physical, microstructure, and dielectric properties of the (1-x)BNKT-xBT ceramics.

| $x$ | Density (g/cm$^3$) | Relative density (%) | $c/a$ | Average grain size (µm) | $T_d$ (°C) | $T_m$ (°C) | $\varepsilon_{max}$ | tan δ | $\delta_a$ (K) |
|-----|-------------------|---------------------|-------|------------------------|----------|----------|-----------------|-------|------------|
| 0   | 5.78              | 98.88               | 1.0115| 2.54                   | 170.7    | 326.9    | 4527            | 0.0189| 60         |
| 0.05| 5.80              | 99.16               | 1.0132| 1.44                   | 152.3    | 323.5    | 4905            | 0.0184| 104        |
| 0.10| 5.81              | 99.48               | 1.0147| 1.47                   | 142.6    | 323.0    | 4908            | 0.0128| 143        |
| 0.15| 5.83              | 99.66               | 1.0163| 1.43                   | 140.1    | 312.4    | 4936            | 0.0186| 173        |

* = Data from TSDC technique.

Figure 2. Raman spectra of the (1-x)BNKT-xBT ceramics measured at room temperature (RT).
Gaussian-Lorentzian modes [29–32]. Based on Raman data as showed in Figure 2, it can be obviously detected that four main regions are consistent with the previous reports [29–32]. The first one at around 120 cm\(^{-1}\) was associated with the vibration of A-site cations such as Na, K, and Bi in the perovskite structure (Na-O, K-O, Bi-O, and Ba-O bonds). The second at around 270 cm\(^{-1}\) was assigned as Ti-O vibrations (\(A_1\) mode). The third frequency region at around 450 – 695 cm\(^{-1}\) host’s modes was noted for the vibration of the TiO\(_6\) octahedra, most likely as a superposition of longitudinal optical (LO) and transverse optical (TO) bands of \(A_1\) character. The high-frequency region greater than 700 cm\(^{-1}\) has been linked to \(A_1\) (longitudinal optical) and E (longitudinal optical) overlapping bands [33,34]. From Raman data in Figure 2, the spectrum recorded in this system is in good agreement to that of the previously reported 0.80BNT-0.20BKT ceramics [29,35], which presented a structure consisting of the tetragonal (P4bm) and rhombohedral (R3c) phases. It can be found that with higher BT content, the Ti-O mode vibration centered ~ 260 cm\(^{-1}\) becomes broader and starts splitting into 2 bands that shifted apart from each other. The TiO\(_6\) octahedra vibration in the range of 450 – 655 cm\(^{-1}\) was also separated into three distinct bands in the ceramics (three overlapping bands). This also correlated well with the XRD analysis which indicated that the crystal structure of the lead-free (1-x)BNKT-xBT ceramics transformed from the mixed rhombohedral-tetragonal (for the \(x = 0\) ceramic) into the tetragonal-rich phase (for the \(x = 0.15\) ceramic) [36,37].

Figure 3 shows the FE-SEM micrographs with as sintered surfaces mode of the (1-x)BNKT-xBT ceramics (where \(x = 0 – 0.15\)). The change of average grain size value as a function of BT content is shown in Table 1. The SEM result confirmed that all ceramics were dense with relatively high-density values of 5.78 – 5.83 g/cm\(^3\) (relative density greater than 98 – 99%), which is an increase in the density of samples observed as the BT modifier content increased. Most grains of all ceramics showed a clear grain boundary. SEM image of the BNKT ceramic displayed a cubic-like grains with an average grain size of 2.54 µm. The average grain size reduced significantly with increasing BT content and reached the minimum value of 1.43 µm for the \(x = 0.15\) ceramic. It was ascribed to the solubility limit of the additive BT in the host BNKT ceramics, the excess BT element will pinch at the grain boundary, resulted to interrupt and hinder the growth of grain [38,39].

### 3.2. Dielectric properties

Plot of temperature dependence on the dielectric constant (\(\varepsilon_r\)) and dielectric loss (\(\tan \delta\)) of the (1-x)BNKT-xBT ceramics with various frequencies of 1, 10, 100, 500, and 1000 kHz are shown in Figure 4. The related dielectric properties values are also summarized in Table 1. All ceramics revealed two distinguishing dielectric anomalies. The peak in low temperature range (below 150 °C) is denoted as the depolarization temperature (\(T_d\)), which suggested to be an indication of the stability of the ferroelectric domains [40,41]. The peak in high temperature (around 300 °C) is referred to the maximum temperature (\(T_m\)) or Curie point (\(T_c\)), which is the maximum value of dielectric constant [42]. As the temperature was increased, the dielectric constant gradually increased, jumped at \(T_d\) and then reached a maximum value at \(T_m\) and finally gradually decreased [43,44]. There are several techniques to determine the \(T_d\) value as can be found in the literatures such as the thermally stimulated depolarization.

![Figure 3](image-url)  
**Figure 3.** The FE-SEM micrographs with as sintered surfaces of the (1-x)BNKT-xBT ceramics where (a) \(x = 0\), (b) \(x = 0.05\), (c) \(x = 0.10\), and (d) \(x = 0.15\).
current (TSDC), piezoelectric resonance measurements, dielectric permittivity and dielectric loss, temperature dependent in situ $d_{33}$, and high-temperature in situ XRD, etc [45]. In this work, the TSDC technique was used to determine the $T_d$ value, due to this technique is the most accurate and is highly recommend from many researchers [46,47]. The measurements were performed on the poled ceramics. Figure 5 shows the current density as a function of temperature ($J$-T curves) of the studied ceramics, measured by the TSDC technique. The $T_d$ value was estimated from the $J$-T peak and the values are summarized in Table 1. Based on Figures 4 and 5, it can be seen that the $T_d$, $T_m$ and the maximum dielectric constant ($\varepsilon_{\text{max}}$) values of the pure BNKT ceramic in this study were found to be 170.7 °C, 326.9 °C, and 4527, respectively. The $T_d$ and $T_m$ values are found to decrease while the $\varepsilon_{\text{max}}$ slightly increased with increasing BT content (see Figure 6(a)).
and 6(b)). The decreasing of $T_d$ and $T_m$ values with further increasing BT content also indicated the reduction of the stability of ferroelectric domains. Due to the incompatibility of their crystal lattices which the crystal structure of the ceramics evolved from the mixed rhombohedral-tetragonal to more symmetric tetragonal-rich phase with increasing BT content, resulting to a reduction of the thermal stability in the long-range ferroelectric domains \cite{48,49}. The temperature-dependent dielectric study of all ceramics showed a broad peak at $T_d$ and $T_m$, indicating that the phase transition was the diffuse type.

In order to give a quantitative analysis of the diffuse phase transition behavior, many relations have been attempted to describe the dielectric permittivity of relaxors at $T > T_m$, the Lorentz-type empirical relation is one of which:

$$\frac{\varepsilon_A}{\varepsilon} = 1 + \frac{(T - T_A)^2}{2\delta_A^2}$$  \hspace{1cm} (1)

where the $\varepsilon_A$ is the extrapolated value of dielectric constant at $T = T_A$, $T_A$ is the temperature of the dielectric peak, and $\delta_A$ is the diffuseness parameter, respectively. The parameter $\delta_A$ is frequency-independent at high enough frequencies, and it reflects the diffuseness parameter of the dielectric peak \cite{50}. The $\delta_A$ increases from 60 K for the pure BNKT ceramic to around 173 K for the $x = 0.15$ ceramic. This result indicates that a higher degree of the diffuseness phase transition was produced by the BT additive \cite{50}. The substitution disordering in the arrangement of cations at one or more crystallographic sites in the lattice structure is the reason for the diffuseness or the broadening of the dielectric peak. This lead to the heterogeneous domains. In addition, the diffuseness may be due to the defect-induced relaxation at the high temperature \cite{51–53}.

### 3.3. Ferroelectric and electric field-induced strain responses

The polarization-electric field ($P$-$E$) hysteresis loop and bipolar strain-electric field ($S$-$E$) of the (1-x)BNKT-xBT ceramics, measured at room temperature (RT) and electric field of 60 kV/cm are shown in Figures 7 and 8.

**Figure 6.** Plot of (a) $T_m$ and $T_d$ values as a function of BT content, and (b) $\varepsilon_{max}$ as a function of BT content (inset: $\delta_A$ as various BT content).
respectively. Higher values of applied electric field (> 60 kV) caused the samples to undergo a dielectric breakdown. For more details, the remanent polarization ($P_r$), the coercive field ($E_c$), the maximum polarization ($P_{\text{max}}$), the maximum strain ($S_{\text{max}}$), the negative strain ($S_{\text{neg}}$), the difference between the lowest strain and zero-field strain (visible only in the bipolar curve) [46,54], and the normalized strain coefficient ($d_{33}^* = S_{\text{max}}/E_{\text{max}}$) values are also presented.

**Figure 7.** Polarization-electric field ($P$-$E$) hysteresis loop and current-electric field ($I$-$E$) of the (1-$x$)BNKT-$x$BT ceramics, measured at RT under an electric field of 60 kV/cm and a frequency of 1 Hz where (a) $x = 0$, (b) $x = 0.05$, (c) $x = 0.10$, and (d) $x = 0.15$.

**Figure 8.** Bipolar strain-electric field data ($S$-$E$) of the (1-$x$)BNKT-$x$BT ceramics, measured at RT under an electric field of 60 kV/cm and a frequency of 0.1 Hz where (a) $x = 0$, (b) $x = 0.05$, (c) $x = 0.10$, and (d) $x = 0.15$. 

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listed in Table 2. The well-saturated P-E hysteresis loops were observed for all ceramics. The pure BNKT ceramic exhibited a typical ferroelectric behavior with the square shape, which indicating the strong ferroelectric order. It had the maximum values of $P_{\text{max}}$ of 38.31 $\mu$C/cm$^2$, $P_r$ of 31.45 $\mu$C/cm$^2$, and $E_c$ of 35.06 kV/cm, which all were in good agreement with those from many literatures [46,55].

The bipolar S-E curve of the pure BNKT ceramic showed a typical butterfly-shaped loop with the $S_{\text{max}}$ of 0.22% and the calculated $d_{33}^*$ of 384 pm/V. This was related with the ferroelectric domain switching at $E_c$, where the largest $S_{\text{neg}}$ of $-0.10\%$ was observed. However, the incorporation of BT content in the based-ceramics significantly influenced on the hysteresis loop, strain shape, and the polarization responses. The obtained P-E hysteresis loop for the $x = 0.05$ ceramic showed a pinching-type characteristic. The $P_r$ of this ceramic severe decreased to from 31.45 to 14.42 $\mu$C/cm$^2$ and the $E_c$ dropped steeply from 35.06 to 13.94 kV/cm. The drop of both $E_c$ and $P_r$ values with the pinched of P-E hysteresis loops for the $x = 0.05$ ceramic indicated that the long-range ferroelectric order (as observed in the pure BNKT ceramic) was disrupted by the additive of BT. The pinched P-E hysteresis loops correspond with two current peaks in the I-E curves (denoted as “1” and “2”) (see Figure 7), which indicated a formation of ergodic relaxor phase (ER) [56]. This also corresponded to a large electric field-induced strain of 0.37% with a $d_{33}^*$ of 630 pm/V along with a slight reduction in $S_{\text{neg}}$ of $-0.09\%$ for this sample ($x = 0.05$). This may be due to a phase transition from a ferroelectric to ergodic relaxor phase (FE to ER), accompanied by the disordering of the long-range ferroelectric order with increasing the BT content [57]. Based on the obtained results, it is believed that this composition contained a mixture of FE and ER states. The obtained result is similar to those observed in the Sr(Hf$_{0.5}$Zr$_{0.5}$)O$_3$-modified Bi$_{0.96}$Na$_0$K$_2$O$_5$ TiO$_3$ [4], the Bi$_{0.96}$Na$_0$K$_2$TiO$_3$-Na$_1$.Li$_1$NbO$_3$ [15], and SrTiO$_3$-modified 0.98Bi$_{1/2}$Na$_{1/2}$TiO$_3$-0.01LaFeO$_3$ systems [58]. However, with further increasing BT content up to $x = 0.10 - 0.15$, the hysteresis loop shape transformed from a pinching-type character to a normal ferroelectric loop. The $E_c$ and $P_r$ values increased significantly as the BT content was increased up to $x = 0.10 - 0.15$. While, the bipolar S-E curve reversed back to a normal butterfly shape. The observed large strain value seemed to exist only over a narrow compositional region. When the composition deviated this narrow range, the strain would rapidly decreased [46,55,59]. A slight decreasing of the $d_{33}^*$ and $S_{\text{max}}$ was observed at higher BT content. The $d_{33}^*$ and $S_{\text{max}}$ values were then decreased to the minimum value of 0.21% and 369 pm/V, respectively, for the $x = 0.15$ ceramic (see Figure 9).

### 3.4. Electrostrictive properties

The electrostrictive properties of the lead-free (1-$x$) BNKT-xBT ceramics was studied in this section. The electrostrictive effect or ES is an important parameter for many actuator devices for the practical applications. It is a common effect for all insulators or dielectrics materials, which observed in all crystal symmetries (it has no symmetry limitation). Normally, the ES can be determined by the electrostrictive coefficient ($Q_{33}$), which can be expressed as equation [60–62]:

$$S_3 = Q_{33}P_3^2$$

where $S_3$ is the longitudinal strain, $Q_{33}$ is the longitudinal electrostrictive coefficient along the direction of

Table 2. Ferroelectric- and electric field-induced strains properties of the (1-x)BNKT-xBT ceramics.

| $x$ | $P_{\text{max}}$ ($\mu$C/cm$^2$) | $P_r$ ($\mu$C/cm$^2$) | $E_c$ (kV/cm) | $S_{\text{max}}$ (%) | $S_{\text{neg}}$ (%) | $d_{33}^*$ (pm/V) | $Q_{33}$ (m$^2$/C$^2$) |
|-----|------------------|-----------------|-------------|-------------------|-----------------|----------------|------------------|
| 0   | 38.31            | 31.45           | 35.06       | 0.22              | -0.10           | 384            | 0.0202           |
| 0.05| 32.91            | 14.42           | 13.94       | 0.37              | -0.09           | 630            | 0.0421           |
| 0.10| 32.54            | 20.73           | 21.49       | 0.27              | -0.08           | 468            | 0.0318           |
| 0.15| 30.07            | 20.55           | 27.15       | 0.21              | -0.08           | 369            | 0.0308           |

![Figure 9](image-url)  
Figure 9. Plot of the maximum strain ($S_{\text{max}}$) and normalized strain coefficient ($d_{33}^* = S_{\text{max}}/E_{\text{max}}$) as a function of BT content.
applied electric field, and $P_3$ is the polarization in the direction of the poling axis. Plots of $S$ as a function of $P^2$ of the (1-x)BNKT-xBT ceramics are shown in Figure 10 (a-d). The variation of $Q_{33}$ value of the studied ceramics as a function BT content is displayed in Figure 10(e) and the related value is summarized in Table 2. The pure BNKT ceramic had the $Q_{33}$ value of 0.0202 m$^2$/C$^2$. The highest $Q_{33}$ value of 0.0421 m$^2$/C$^2$ is noted for the $x = 0.05$ ceramic, which was more than ~2 times (or 108%) as compared to the pure BNKT ceramic. This value is high when compared to several previous works reported on lead-free electrostrictors and other BNT-based (see Table 3) [61–65]. Based on Table 3, the $Q_{33}$ value for the previous studied ferroelectric ceramics are compared with $Q_{33}$ values of the (1-x)BNKT-xBT ceramics. It was seen that the currently presented $Q_{33}$ value is highly competitive to those of known materials i.e. Bi$_{12/3}$Na$_{0.82}$K$_{0.18}$TiO$_3$-BaZrO$_3$ [61], PMN-PT [62], Bi$_{0.5}$(Na$_{0.76}$K$_{0.22}$)TiO$_3$-Bi(Ni$_{0.3}$Tio$_{0.3}$)O$_3$ [63], and BNT-BT-KNN [64,65]. In general, the $Q_{33}$ value in this work is almost twice as much as that of these systems. This suggested that the BNKT-BT system has a large electrostrictive effect over other lead-free and lead-based ferroelectric materials. Nomura and Uchino [66] pointed

![Figure 10. Plots of S as a function of P² where (a) x = 0, (b) x = 0.05, (c) x = 0.10, (d) x = 0.15, and (e) the Q₃₃ value as a function of BT content.](image)

| System | $Q_{33}$ (m²/C²) | Ref. |
|--------|-----------------|-----|
| BNKT-0.05BT | 0.0421 | This work |
| Bi$_{12/3}$(Na$_{0.82}$K$_{0.18}$)TiO$_3$-0.05 BaZrO$_3$ | 0.0250 | [61] |
| PMN-0.32PT | 0.0140 | [62] |
| PMN-0.37PT | 0.0150 | [63] |
| Bi$_{0.5}$(Na$_{0.76}$K$_{0.22}$)TiO$_3$-0.05Bi(Ni$_{0.3}$Tio$_{0.3}$)O$_3$ | 0.0300 | [63] |
| (0.94-x)BNT-0.06BT-xKNN, $x = 0.20$ | 0.0260 | [64] |
| (1-x)[0.94Bi$_{0.5}$Na$_{0.3}$TiO$_3$-0.06BaTiO$_3$]-K$_{0.1}$Na$_{0.9}$NbO$_3$, $x = 0.18$ | 0.0270 | [65] |
out that the relaxor character had influenced the electrostrictive properties. They also reported that the disordered state for the relaxors due to large rattling space caused a high electrostrictive effect. Therefore, it is inferred that in some BNT-relaxor solid solutions giant electrostrictive effect could be identified [60].

4. Conclusion

In this research, the lead-free Bi$_{0.5}$(Na$_{0.8}$KO$_{0.2})$O$_{3}$TiO$_{3}$ modified by BaTiO$_{3}$ ceramics were successfully synthesized by a conventional mixed-oxide method. The results indicated that BT enhanced the electrical properties of the pure BNKT ceramic. All samples exhibit a single perovskite structure without any impurity phases, which indicated that the BT is dissolved into the BNKT lattice structure to form a homogeneous solid solution. The incorporation of BT into the BNKT ceramic significantly affected the piezoelectric and ferroelectric properties. An enhancement of the $S_{\text{max}}$ from 0.22% for the $x = 0$ ceramic to around 0.37% for the $x = 0.05$ ceramic was achieved. The corresponding highest the normalized strain coefficient ($d_{33}^* = 630$ pm/V) and the electrostrictive coefficient ($Q_{33} = 0.0421$ m$^2$/C$^2$) was found for the $x = 0.05$ ceramic, which can be ascribed to the compositional induced disorder leading to the coexistence of the ergodic relaxor (ER) phase along with the ferroelectric (FE) phase. Besides, based on the obtained results, the $(1-x)\text{Bi}_{0.5}\text{(Na}_{0.8}\text{KO}_{0.2})\text{O}_{3}-x\text{BaTiO}_{3}$ lead-free oxide ceramics are promising candidates for the piezoelectric and actuator applications.

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No potential conflict of interest was reported by the author(s).

References

[1] Khan HU, Ullah A, Ullah M, et al. Structural, electrical and electromechanical properties of $(\text{Bi}_{0.5}\text{Na}_{0.8}\text{K}_{0.2})\text{O}_{3}$ lead-free piezoelectric ceramics. Physica B. 2020;584:121202.
[2] Li L, Fang D, Wang RX, et al. Domain evolution and improved electrical property of BiMn$_{0.2}$/Nb$_{0.1}$/O$_{3}$ doped 0.8Na$_{0.8}$Bi$_{0.5}$TiO$_{3}$-0.2K$_{0.5}$Bi$_{0.5}$TiO$_{3}$. Ceram Int. 2020;46:7947–7953.
[3] Wang C, Li Q, Zhang W, et al. Large electric field-induced strain in the novel BNKT-BNLBTZ lead-free ceramics. J Mater Sci Technol. 2020;45:15–22.
[4] Gupta SK, Quade RM, Gibbons B, et al. Electric field-induced strain in Sr(Hf$_{0.5}$Zr$_{0.5}$O$_{3}$)-modified Bi$_{0.5}$Na$_{0.8}$K$_{0.2}$O$_{3}$ piezoelectric ceramics. J Appl Phys. 2020;127:074104.
[5] Liu Y, Li Y, Zheng Z, et al. Dielectric temperature stability of Nb-modified Bi$_{0.5}$Na$_{0.8}$K$_{0.2}$O$_{3}$ lead-free ceramics. Ceram Int. 2021;47:4933–4936.
[6] Dong G, Fan H, Liu L, et al. Large electrostrain in Bi$_{1/2}$Na$_{1/2}$TiO$_{3}$-based relaxor ferroelectrics: a case study of Bi$_{1/2}$Na$_{1/2}$TiO$_{3}$-Bi$_{1/2}$K$_{1/2}$TiO$_{3}$-Bi(Ni$_{0.2}$Na$_{0.8}$BiO$_{3}$) ceramics. J Materiomet. 2021;7:593–602.
[7] Tran VDN, Vu LH, Van VL, et al. Structure evolution and electrical properties of lead-free Bi$_{0.5}$Na$_{0.4}$K$_{0.5}$O$_{3}$ piezoceramics by isovalent La doping. J Mater Sci: Mater Electron. 2021;32:4363–4371.
[8] He H, Lu W, Oh JAS, et al. Probing the coexistence of ferroelectric and relaxor states in Bi$_{0.5}$Na$_{0.5}$TiO$_{3}$-based ceramics for enhanced piezoelectric performance. ACS Appl Mater Interfaces. 2020;12:30548–30556.
[9] Chen X, Liao Y, Li G. Structure and electrical properties of [(Na$_{0.8}$Ba$_{0.2}$)$_{3/5}$Bi$_{0.5}$Li$_{0.1}$]$\times$TiO$_{3}$ piezoceramics with A-site nonstoichiometry. J Asian Ceram Soc. 2020;8:1036–1042.
[10] Ren X, Yin H, Tang Y, et al. The large electro-strain in BNKT-BST-100xTa lead-free ceramics. Ceram Int. 2020;46:1876–1882.
[11] Wang X, Gao H, Hao X, et al. Enhanced piezoelectric, electrocaloric and energy storage properties at high temperature in lead-free Bi$_{0.5}$Na$_{0.5}$K$_{0.5}$O$_{3}$ ceramics. J Mater. 2019;45:4274–4282.
[12] Kushvaha DK, Rout SK, Tiwari B. Structural, piezoelectric and high density energy storage properties of lead-free BNKT-BCTZ solid solution. J Alloys Compos. 2019;782:270–276.
[13] Basar NU, Khan MI, Ullah A, et al. Effect of Bi(Zn$_{0.5}$Ti$_{0.5}$)O$_{3}$ substitution on structural and electromechanical properties of Bi$_{0.5}$Na$_{0.75}$K$_{0.25}$O$_{3}$ lead-free piezoelectric ceramics. Mater Res Express. 2019;6:095804.
[14] Hong IK, Han HS, Yoon CH, et al. Strain enhancement in lead-free Bi$_{0.5}$Na$_{0.75}$K$_{0.25}$O$_{3}$ ceramics by CaZrO$_{3}$ substitution. J Intell Mater Syst Struct. 2013;24:1343–1349.
[15] Li Q, Ning L, Wang C, et al. Gaint electric field-induced strain and ferroelectric behavior of Bi$_{0.5}$Na$_{0.4}$K$_{0.1}$TiO$_{3}$-Na$_{0.1}$Li$_{0.1}$NbO$_{3}$ lead-free ceramics. Mater Sci Eng B. 2021;263:114819.
[16] Lv YK, Duan SF, Zhao ZH, et al. Enhanced electromechanical strain response in (Fe$_{0.2}$Nb$_{0.8}$)$_{0.37}^{+}$-modified Bi$_{0.5}$Na$_{0.8}$K$_{0.2}$O$_{3}$ lead-free piezoelectric ceramics. J Mater Sci. 2018;53:8059–8066.
[17] Xi H, Yu L, Qian H, et al. Large strain with low hysteresis in Sn-modified Bi$_{0.5}$Na$_{0.75}$K$_{0.25}$O$_{3}$ lead-free piezoceramics. J Mater Sci. 2020;55:1388–1398.
[18] Ismail FA, Osman RAM, Idris MS, et al. Structure and electrical characteristics of BaTiO$_{3}$ and Ba$_{0.97}$Sr$_{0.03}$TiO$_{3}$ ceramics. Solid State Phenom. 2018;280:127–133.
[19] Jaffe B, Cook WR, Jaffe H. Piezoelectric ceramics. London: Academic Press; 1971.
[20] Panda PK. Review: environmental friendly lead-free piezoelectric materials. J Mater Sci. 2009;44:5049–5062.
[21] Xu-Holland. Ferroelectric materials and their applications. North-Holland Elsevier Science Publishers: Amsterdam; 1991.

[22] Acosta M, Novak N, Rojas V, et al. BaTiO3-based piezoelectrics: fundamentals, current status, and perspectives. Appl Phys Rev. 2017;4:041305.

[23] Takenaka T, Maruyama K, Sakata K. (Bi1−xNa2x)TiO3-BaTiO3 system for lead-free piezoelectric ceramic. Jpn J Appl Phys. 2019;58:2326–2329.

[24] Badapanda T, Sahoo S, Nayak P. Dielectric, ferroelectric and piezoelectric study of BNT-BT solid solutions around the MPB region. IOP Conf Series: Mater Sci Eng. 2017;178:012032.

[25] Dong G, Fan H, Jia Y, et al. Strain properties of (1-x)Bi0.5Na0.5TiO3-xBi(0.252Mg0.752Ta1.25)O3 electroceramics. Ceram Int. 2020;46:12111–21215.

[26] Fan P, Zhang Y, Zhang Q, et al. Large strain with low hysteresis in Bi1−xLi0.1xTiO3 modified Bi1/2(Na0.82K0.18)1/2TiO3 lead-free piezocermics. J Eur Ceram Soc. 2018;38:4404–4413.

[27] Shannon RD. Revised ionic radii and systematic studies of interatomic distances in halides and chalcogenides. Acta Cryst A. 1976;32:751–767.

[28] Eerd BW, Damjanovic D, Klein N. Structural complexity of (Na0.5Bi0.5)TiO3-BaTiO3 as revealed by Raman spectroscopy. Phys Rev B. 2010;82:104112.

[29] Hao J, Shen B, Zhai J. Switching of morphotropic phase boundary and large strain response in lead-free ternary (Bi0.5Na0.5TiO3-K0.5Sr0.5TiO3-K0.5Bi0.5Na0.5)NbO3 system. J Appl Phys. 2013;113:114106.

[30] Bai W, Liu F, Li P. Structure and electromechanical properties in Bi0.5(Na0.5TiO3)-based lead-free piezocermics with calculated end-member Bi(Na0.5Ti0.5)O3. J Eur Ceram Soc. 2015;35:3457–3466.

[31] Parija B, Badapanda T, Rout SK, et al. Morphotropic phase boundary and electrical properties of 1-xBi0.5Na0.5TiO3-xBi(0.252Mg0.752Ta1.25)O3 lead-free piezoelectric ceramics. Ceram Int. 2013;39:4877–4886.

[32] Yao Q, Wang F, Xu F, et al. Electric field-induced giant strain and photoluminescence-enhancement enhancement in rare-earth modified lead-free piezoelectric ceramics. ACS Appl Mater Interfaces. 2015;7:5066–5075.

[33] Aksel E, Forrester JS, Kowalski B, et al. Structure and properties of Fe-modified Bi0.5Bi0.5TiO3 at ambient and elevated temperature. Phys Rev B. 2012;85:024121.

[34] Rout D, Moon KS, Kang SJL, et al. Dielectric and Raman scattering studies of phase transitions in the (100-x)Na0.5Bi0.5TiO3-xSrTiO3 system. J Appl Phys. 2012;108:084102.

[35] Hao J, Shen B, Zhai J, et al. Large strain response in 0.99(Bi0.5Na0.5K0.5)TiO3-0.01(K2Na)1/2NbO3 lead-free ceramics induced by the change of K/Na ratio in (KxNa1−x)NbO3. J Am Ceram Soc. 2013;96:3133–3140.

[36] Bhandari S, Sinha N, Kumar B. Enhanced microstructure and electrical properties of Mn-modified Bi0.5Na0.5K0.5TiO3 ferroelectric ceramics. Ceram Int. 2016;42:4274–4284.

[37] Malik RA, Hussain A, Zaman A, et al. Structure-property relationship in lead-free A- and B-site co-doped Bi0.5(Na0.5K0.5)TiO3-SrTiO3 incipient piezocermics. RSC Adv. 2015;5:96953–96964.

[38] Li Q, Wang G, Yadav AK, et al. Large electrostrictive effect and energy storage density in MnCO3 modified Na0.325Bi0.395Sn0.672Ta1.25O3 lead-free ceramics. Ceram Int. 2020;46:3374–3381.

[39] Shukla A, Choudhary RN. Study of electrical properties of La1/4Mn3/4-modified PbTiO3 nanocramics. J Mater Sci. 2012;47:5074–5085.

[40] Jo W, Schaab S, Sapper E, et al. On the phase identity and its thermal evolution of lead free (Bi1/2Na1/2)TiO3 -6mol% BaTiO3. J Appl Phys. 2011;100:074106.

[41] Anton EM, Jo W, Damjanovic D, et al. Determination of depolarization temperature of (Bi1/2Na1/2)TiO3-based lead-free piezocermics. J Appl Phys. 2011;100:094108.

[42] Chu BJ, Chen DR, Li GR, et al. Electrical properties of (Na1/2Bi1/2)TiO3-BaTiO3 ceramics. J Eur Ceram Soc. 2002;22:2115–2121.

[43] Dai YJ, Zhang XW, Chen KP. An approach to improve the piezoelectric property of (Bi6/5Na3/5)TiO3-(Bi5/3K5/3)TiO2-BaTiO3 lead-free ceramics. Int J Appl Ceram Technol. 2011;8:423–429.

[44] Jo W, Dittmer R, Acosta M, et al. Giant electric-field-induced strains in lead-free ceramics for actuator applications-status and perspective. J Electroceram. 2012;29:71–93.

[45] Anton EM, Jo W, Damjanovic D, et al. Determination of depolarization temperature of (Bi1/2Na1/2)TiO3-based lead-free piezocermics. J Appl Phys. 2011;100:094108.

[46] Jaita P, Watcharapason A, Kumar N, et al. Lead-free (Ba0.7Sr0.3)TiO3-modified Bi0.5(Na0.8K0.2)3TiO3 ceramics with large electric field-induced strains. J Am Ceram Soc. 2016;99:1613–1624.

[47] Jaita P, Manotham S, Rajiyanagul G. Influence of Al2O3 nanoparticle doping on depolarization temperature, and electrical and energy harvesting properties of lead-free 0.94(Bi0.5Na0.5)TiO3-0.06BaTiO3 ceramics. RSC Adv. 2020;10:32078–32087.

[48] Liu X, Xue S, Li F, et al. Giant electrostrain accompanying structural evolution in lead-free NBT-based piezocermics. J Mater Chem C. 2018;6:814–822.

[49] Maqbool A, Hussain A, Malik RA, et al. Evolution of phase structure and giant strain at low driving fields in Bi-based lead-free incipient piezoelectrics. Mater Sci Eng B. 2015;199:105–112.

[50] Ke S, Fan H, Huang H, et al. Lorentz-type relationship of the temperature dependent dielectric permittivity in ferroelectrics with diffuse phase transition. Appl Phys Lett. 2008;93:112906.

[51] Kitanaka Y, Noguchi Y, Miyayama M. Oxygen-vacancy-induced 90°-domain clamping in ferroelectric BiSrTiO3 single crystals. Phys Rev B. 2010;81:094114–094121.

[52] Hong KS, Park SE. Phase relations in the system of (Na1/2Bi1/2)TiO3-PbTiO3. II. Dielectric property. J Appl Phys. 1996;79:388–392.

[53] Li J, Wang F, Qin X, et al. Large electrostrictive strain in lead-free Bi0.5Na0.5TiO3-BaTiO3-KNbO3 ceramics. Appl Phys A. 2011;104:117–122.

[54] Wang XX, Choy SH, Tang XG, et al. Dielectric behavior and microstructure of Bi1/2Na1/2TiO3-Bi1/2K1/2TiO3- BaTiO3 lead-free piezoelectric ceramics. J Appl Phys. 2005;97:104101.

[55] Jaita P, Watcharapason A, Cann DP, et al. Dielectric, ferroelectric and electric field-induced strain behavior of Bi(Ti0.65Sn0.35)O3-modified Bi0.5(Na0.8K0.2)3TiO3 lead-free piezocermics. J Alloys Compd. 2014;596:98–106.

[56] Malik RA, Hussain A, Rahman JU, et al. Structural transition and giant strain induced by A- and B-site concurrent donor doping in Bi0.5(Na0.8K0.2)3TiO3-SrTiO3 ceramics. Mater Lett. 2015;143:148–150.

[57] Ge RF, Zhao ZH, Duan SF, et al. Large electro-strain response of La5+ and Nb5+ co-doped ternary 0.85Bi0.5
Na0.5TiO3-0.11Bi0.5K0.5TiO3-0.04BaTiO3 lead-free piezoelectric ceramics. J Alloys Compd. 2017;724:1000–1006.

[58] Dinh TH, Han HS, Lee JS. Large strain response under low field in SrTiO3 modified Bi1/2Na1/2TiO3-LaFeO3 piezoceramics. Mater Lett. 2020;258:126793.

[59] Li L, Zhang J, Wang RX, et al. Thermally-stable large strain in Bi(Mn0.5Ti0.5)O3 modified 0.8Bi0.5Na0.5TiO2-0.2Bi0.5K0.5TiO3 ceramics. J Euro Ceram Soc. 2019;39:1827–1836.

[60] Jin L, Huo R, Guo R, et al. Diffuse phase transitions and giant electrostrictive coefficients in lead-free Fe3+-doped 0.5Ba(Zr0.3Ti0.7)O3-0.5(Ba0.5Ca0.5)TiO3 ferroelectric ceramics. ACS Appl Mater Interfaces. 2016;8:31109–31119.

[61] Tran VDN, Han HS, Yoon CH, et al. Lead-free electrostrictive bismuth perovskite ceramics with thermally stable field-induced strains. Mater Lett. 2011;65:2607–2609.

[62] Li F, Jin L, Xu Z, et al. Electrostrictive effect in Pb(Mg1/3 Nb2/3)O3-xPbTiO3 crystals. Appl Phys Lett. 2013;102:152910.

[63] Ullah A, Gul HB, Ullah A, et al. Giant room-temperature electrostrictive coefficients in lead-free relaxor ferroelectric ceramics by compositional tuning. APL Mater. 2018;6:016104.

[64] Zhang ST, Yan F, Yang B, et al. Phase diagram and electrostrictive properties of Bi0.5Na0.5TiO3-BaTiO3-K0.5 Na0.5NbO3 ceramics. Appl Phys Lett. 2010;97:122901.

[65] Zhang ST, Kounga AB, Jo W, et al. High-strain lead-free antiferroelectric electrostrictors. Adv Mater. 2009;21:4716–4720.

[66] Nomura S, Uchino K. Electrostrictive effect in Pb(Mn1/3 Nb2/3)O3 type materials. Ferroelectrics. 1982;41:117–132.