

Review Article

Current Development in Lead-Free $\text{Bi}_{0.5}(\text{Na,K})_{0.5}\text{TiO}_3$ -Based Piezoelectric Materials

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The lead-free piezoelectric ceramics display good piezoelectric properties which are comparable with $\text{Pb}(\text{Zr,Ti})\text{O}_3$ (PZT) and these materials overcome the hazard to the environment and human health. The $\text{Bi}_{0.5}(\text{Na,K})_{0.5}\text{TiO}_3$ (BNKT) is rapidly developed because of good piezoelectric, ferroelectric, and dielectric properties compared to PZT. The origin of giant strain of BNKT piezoelectric materials was found at morphotropic phase boundary due to crystal change from tetragonal to orthorhombic and/or precipitation of cubic phases, in addition to domain switching mechanism. The dopants or secondary phases with ABO_3 structure as solid solution are expected to change the crystal structure and create the vacancies which results in enhancement of the piezoelectric properties. In this work, we reviewed the current development of BNKT by dopants and secondary phase as solid solution. Our discussion will focus on role of dopants and secondary phase to piezoelectric properties of BNKT. This result will open the direction to control the properties of lead-free piezoelectric materials.

1. Introduction

The piezoelectric phenomena were discovered by Nobel laureates Pierre and Jacques Curie in 1880 during measurement of surface charges appearing under stress of some crystals such as tourmaline, quartz, and Rochelle salt [1]. During the World War I, Paul Langevin invented the underwater sonar for submarine detection [1]. The detecting device was fabricated by thin quartz crystal glued on two steel plates with resonant frequency of 50 kHz which could detect a submarine at the depth of 1500 meters. However, his achievements were not overlooked by any industrial nation. During World War II, the huge dielectric constants of piezoelectric ceramics materials were isolatedly discovered in the United States, Japan, and the Soviet Union by researching the materials to improve capacitor of devices [1]. The discovery of easily manufactured piezoelectric ceramics with astonishing performance characteristics naturally touched off a revival of

intense research and development into piezoelectric devices which led to their widespread applications. Among them, barium titanate (BaTiO_3) ceramic was discovered with the giant dielectric constant of 1100, ten times higher than that of rutile TiO_2 , the highest value at that time [2]. The first commercial device made from BaTiO_3 was a phonograph pickup and was produced in 1947 [2]. The productions which used the piezoelectric materials were rapidly developed in the late 1950s because a number of other piezoelectric ceramics were found; in particular, lead niobate piezoelectric ceramics were discovered in 1952, and lead zirconate titanate (PZT) compositions were found in 1955 [1, 2]. Several piezoelectric ceramics have been commonly used today such as BaTiO_3 , PZT (and modified composition such as $(\text{Pb,Lu})(\text{Zr,Ti})\text{O}_3$, etc.), and lead magnesium niobate ($\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$) or lead zinc niobate $\text{Pb}(\text{Zn}_{1/3}\text{Nb}_{2/3})\text{O}_3$. To date, PZT is one of the most widely exploited and extensively used piezoelectric materials for piezoelectric actuators, sensors, transducers,

and so forth, even though lead is a very toxic substance as it can cause damage to the kidneys, brain, the nervous system, and, especially, the intelligence of the children [1]. In addition, the volatilization of PbO during the high temperature sintering process not only causes environmental pollution but also generates instability in the composition and electrical properties of the production. According to the recent developments, the European Union (EU) is planning to restrict the use of hazardous substances such as lead as well as other heavy metals [3, 4]. Asia, China, Japan, and Republic of Korea have also enacted similar policies and legislations to control the usage of lead-containing materials [5–7]. In order to circumvent the drawback of lead toxicity, extensive research is focused on the quest for alternate piezoelectric materials. Therefore, there has been a growing research interest in developing alternative lead-free piezoelectric materials that can eventually replace the current lead-based ones. There is no equivalent substitution for PZT till now; therefore, its use still continues. This may be a temporary respite, but the legislation certainly impressed the researchers to develop alternative lead-free piezoelectric materials in order to replace lead-based materials [8–10]. In this paper, we overviewed current developments in $\text{Bi}_{0.5}(\text{Na,K})_{0.5}\text{TiO}_3$ (BNKT) lead-free piezoelectric ceramics and effects of various dopants on their piezoelectric properties.

2. $\text{Bi}_{0.5}(\text{Na,K})_{0.5}\text{TiO}_3$ -Based Lead-Free Piezoelectric Materials

Among lead-free piezoelectric materials perovskite-based type, bismuth sodium titanate $\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3$ (BNT), bismuth potassium titanate $\text{Bi}_{0.5}\text{K}_{0.5}\text{TiO}_3$ (BKT), and solid solution based on these compounds seem to be considered as the most promising materials choice [11–14]. In the following section a brief introduction of these materials is given.

2.1. Bismuth Sodium Titanate and Bismuth Potassium Titanate

2.1.1. $\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3$ Material. BNT is one of the most important lead-free materials discovered by Smolensky et al. in 1960, which has an ABO_3 distorted perovskite structure [10, 15]. The structure of BNT can be considered in two ways: one way is that the bismuth and sodium cations occupy the corners of a cubic unit cell, oxygen anions occupy the face centers, and titanium cations occupy the center of the oxygen octahedra that are formed; the other way is a three-dimensional cubic network of 8 corner-sharing TiO_6 octahedra with bismuth and sodium cations at the center of the cube formed by the octahedra [16]. In the ABO_3 perovskite structure, A cations localize at the corners, B cations localize at the body center, and oxygen anions localize at the face centers. In case of BNT, the bismuth and sodium ions are on the A -site and titanium ions are on the B -site of the structure; this is only to show the stoichiometry that is present in an ideal mixture. However, the real material does not exhibit any long-range ordering. The BNT was rhombohedral structure with $a = 3.98 \text{ \AA}$ and $\alpha = 89.67^\circ$ at room temperature [17]. The BNT

undergoes three phase transitions as crystallographic structure changes. Pronin et al. and Zvirgzds et al. obtained that the first phase transition tetragonal-cubic phase occurred at 320°C whereas the second phase transition temperature of rhombohedral-tetragonal phase was 540°C as they determined the Curie temperature [18, 19]. It reveals an interesting dielectric which is anomaly with low phase-transition temperature ($\sim 200^\circ\text{C}$) from ferroelectric to antiferroelectric phase. Suchanicz et al. reported that the electrostrictive strain increased upon heating when dielectric permittivity (ϵ') grew with maximum near 320°C ; result from attribution of ferroelectric (FE)/antiferroelectric (AFE) transition [20]. Above 320°C , the electrostriction coefficient (Q_{11}) was equal to $(0.4-0.3) \times 10^{-2} \text{ C}^{-2} \text{ m}^4$ and was typical for materials with diffused phase transitions [20]. The piezoelectric module d_{33} was 100–120 pC/N at $\epsilon' = 500$ which belongs to [100] direction of the BNT single crystal [21]. Emelyanov et al. characterized the piezoelectric properties of (001) plane in the rhombohedral BNT single crystals phase which were of piezoelectric coefficients (d_{31}, d_{33}) $d_{31} = 160 \text{ pC/N}$, $d_{33} = 60 \text{ pC/N}$, $k_{31} = 0.55$, and $k_{33} = 0.40$ [22]. The remnant polarization (P_r) and the coercive field (E_c) of BNT single crystal are $38 \mu\text{C/cm}^2$ and 73 kV/cm , respectively [10]. Zhao et al. reported that the values of d_{33} and coupling factor (k_t) were 102 pC/N and 0.58 , respectively, and they were strongly influenced by the grain size [23, 24]. Piezoelectric properties of the BNT ceramics, polarized by electric field of 40 kV/cm at 200°C , were $k_{31} = 0.10$, $d_{31} = 15 \text{ pC/N}$, $d_{33} = 70 \text{ pC/N}$, $\epsilon' = 300$, and $\tan \delta = 0.011$ [25].

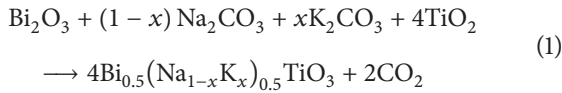
2.1.2. $\text{Bi}_{0.5}\text{K}_{0.5}\text{TiO}_3$ Material. BKT was also first fabricated by Smolenskii and Agranovskaya which has a perovskite type ferroelectric structure belonging to tetragonal crystal at room temperature [11]. The BKT has been investigated much less than BNT because it was not easy to prepare high-dense ceramics due to the fact that secondary phases such as $\text{K}_2\text{Ti}_6\text{O}_{13}$ easily formed during sintering at high temperature, even though synthesis of the compound was not difficult [10, 26]. BKT is a ferroelectric material with the Curie temperature (T_C) of 380°C [27]. Ivanova et al. reported that BKT has a tetragonal structure with $a = 3.913 \text{ \AA}$ and $c = 3.993 \text{ \AA}$ at room temperature, which does not show any sign of ordering, whereas a phase transition into a pseudocubic phase was observed at about 270°C , and transition into a cubic phase was at 410°C [28]. Hiruma et al. obtained the remnant polarization $P_r = 22.2 \mu\text{C/cm}^2$ and coercive field $E_C = 52.5 \text{ kV/cm}$, electromechanical factor $k_{33} = 0.28$ and $d_{33} = 69.8 \text{ pC/N}$ [29].

Literature survey indicates that BKT is studied weaker than BNT; its behavior is clearly indicated that it does not show such unusual phenomena as BNT (isotropic points and “disappearance” of phase transitions). It is clear that both phases of BKT are ferroelectric that they both are diffused and, most probably, overlapped each other. There is, of course, a problem: coexistence of a paraelectric (PE) and two ferroelectric (FE) phases within a crystal lattice. In addition, the BKT single phase is not easy to fabricate in high-dense structure. However, such a problem is not new for ferroelectric

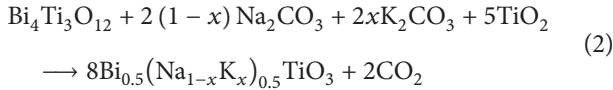
perovskites. It is clear that many properties are still not studied. However, by combining the sol-gel and conventional solid-state reaction method, Zhu et al. obtained the high compact density of ~91.2%, which overcame the low density of ~70% of ceramics prepared by only traditional solid state synthesis [30]. The result was promoted to further investigate the properties of BKT.

2.2. $B_{0.5}Na_{0.5}TiO_3$ - $B_{0.5}K_{0.5}TiO_3$ Solid Solutions Systems

2.2.1. $B_{0.5}(Na,K)_{0.5}TiO_3$ Fabrication Methods. The BNKT ceramics were first fabricated by Bührer by conventional ceramics method via starting materials with metal oxide Bi_2O_3 and TiO_2 and alkali carbonate powder Na_2CO_3 and K_2CO_3 [12]. The BNKT powder was obtained through ball milling and solid state reaction by following equation:



The single crystals $(1-x)BNT$ - $xBKT$ ($0 < x < 0.14$) were fabricated by flux method [31]. The grain oriented and textured BNKT ceramics were first fabricated by Tani, through reactive template grain growth method, using plate like $Bi_4Ti_3O_{12}$ (BiT) particles as a template [32]. The BiT platelets were aligned parallel to the tape casting direction, and grain oriented ceramics were prepared from Bi_2O_3 and TiO_2 using molten salt synthesis. Additional amounts of Na_2CO_3 , K_2CO_3 , and TiO_2 to stoichiometry were included in the mixing batch to react with the BiT according to following equation:



Recently, BNKT powders were prepared by the sol-gel process [33]. The starting materials as analytical-grade chemical: bismuth nitrate ($Bi(NO_3)_3 \cdot 5H_2O$), sodium acetate ($CH_3COONa \cdot 3H_2O$) or sodium nitrate ($NaNO_3$), potassium acetate (CH_3COOK) or potassium nitrate (KNO_3), and tetrabutyl titanate ($Ti(OC_4H_9)_4$) or titanium isopropoxide ($Ti(OC_3H_7)_4$) were used to prepare a BNKT precursor solution. Then, the sol was heated to get dried gels. Finally, the dried gels were calcined and annealed to remove the organic ingredients and to promote crystallization, respectively. In addition, the BNKT thin films and nanofibers were also fabricated by a sol-gel method and electrospinning technique as reported by Chen et al. [34]. After preparing the sol, the thin films and nanofibers were prepared on Pt/Ti/SiO₂/Si substrate by spin coating and electrospinning, respectively. The Li-doped BNKT thin films have been grown by pulsed laser deposition (PLD) using a krypton fluoride (KrF) excimer laser with a wavelength of 248 nm [35].

2.2.2. Crystal Structure of $B_{0.5}(Na,K)_{0.5}TiO_3$. Bührer reported that the lattice parameters of $B_{0.5}Na_{0.5}TiO_3$ increased with BKT concentration addition [12]. Pronin et al. indicated that the $(1-x)Bi_{0.5}Na_{0.5}TiO_3$ - $xBi_{0.5}K_{0.5}TiO_3$ solid solution was

rhombohedral at $x < 0.18$, pseudocubic at $x = 0.18$ – 0.40 , and tetragonal at $x > 0.40$ at the room temperature [18]. On the morphotropic phase boundary at $x = 0.18$, the lattice parameters and unit cell volume change by jump [18]. However, Kreisel et al. obtained the structural change in solid solution $(1-x)BNT$ - $xBKT$ by using the Raman scattering. Results showed existence of a phase transition at x between 0.4 – 0.5 . However, it also indicated that the phase transition in range $x = 0.6$ – 0.8 due to existence of nanosize $Bi^{3+}TiO_3$ and $(Na_{1-2x}K_{2x})^+TiO_3$ clusters [36]. This region is considered as a morphotropic phase boundary (MPB) where the MPB described the boundary that separates regions of difference symmetries and can be crossed through a change in composition. However, the MPB was not reported clearly [37].

2.2.3. Some Physical Properties at Morphotropic Boundary Diagram. The Curie temperature of $(1-x)Bi_{0.5}Na_{0.5}TiO_3$ - $xBi_{0.5}K_{0.5}TiO_3$ solid solution was found to go through a minimum at $x = 0.1$ – 0.2 [17]. Sasaki et al. studied this system and obtained a maximum of $d_{31} = 42$ pC/N with $k_p = 0.23$ which was observed on the first morphotropic boundary [14]. Elkechai et al. obtained $d_{33} = 96$ pC/N, $k_p = 0.21$, and $N_p = 2800$ Hzm at $x = 0.16$ [38]. Yoshii et al. investigated the piezoelectric properties of a solid solution of the binary system, $xBi_{0.5}Na_{0.5}TiO_3$ - $(1-x)Bi_{0.5}K_{0.5}TiO_3$ [39]. Fine piezoelectric properties in lead-free piezoelectric ceramics were obtained near MPB composition between the rhombohedral and tetragonal structures, and the highest electromechanical coupling factor, k_{33} , and piezoelectric constant, d_{33} , were 0.56 pC/N for $Bi_{0.5}(Na_{0.84}K_{0.16})_{0.5}TiO_3$ and 157 pC/N for $Bi_{0.5}(Na_{0.8}K_{0.2})_{0.5}TiO_3$, respectively. However, the T_d of $Bi_{0.5}(Na_{0.8}K_{0.2})_{0.5}TiO_3$ was low at $174^\circ C$. The T_d of the MPB composition was low, and the T_d near the MPB composition was sharply decreased. It is thought that $Bi_{0.5}(Na_{0.7}K_{0.3})_{0.5}TiO_3$ is a candidate composition for lead-free actuator applications owing to its relatively large piezoelectric constant, d_{33} of 126 pC/N, dynamic d_{33} of 214 pm/V, and high depolarization temperature, T_d of $206^\circ C$. Recently, Izumi et al. reported that a small amount of BKT substitution suppressed the remnant polarization from $38 \mu C/cm^2$ at $x = 0$ to about $15 \mu C/cm^2$ at $x = 0.02$; piezoelectric strain constant (d_{33}) is enhanced by increasing x up to 297 pm/V at $x = 0.14$ during studying the $(1-x)BNT$ - $xBKT$ single crystals ($0 < x < 0.14$) [31].

BNT-BKT solid solutions are interesting because of three phenomena: (i) existence of two morphotropic boundaries, (ii) neighborhood of the antiferroelectric (AFE) phase of BNT and high-temperature ferroelectric (FE) phase of BNT, and (iii) complicated coexistence of several phases within one perovskite lattice because of the phase-transition diffusion.

2.2.4. Mechanism Electric-Field-Induced Giant Strain

(1) Electric-Field-Induced Phase Transition. Ferroelectric crystals are characterized by their asymmetric or polar structures. In an electric field, ions undergo asymmetric displacement and result in a small change in crystal dimension, which is

proportional to the applied field [40, 41]. However, the effect is generally very small and thus limits its usefulness.

In the search for lead-based materials with large electric-field-induced phase transition (EFIS), an alternative and applicable approach for ceramics was reported by Uchino et al. and Pan et al. based on the work of Berlicourt et al. [5–7]. They observed a large strain due to a change of the unit cell in $(\text{Pb},\text{La})(\text{Sn},\text{Zr},\text{Ti})\text{O}_3$ ceramic because of a transition from antiferroelectric to ferroelectric phase which was induced by the electric field.

Zhang et al. proposed that the high strain in lead-free $\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3\text{-BaTiO}_3\text{-K}_{0.5}\text{Na}_{0.5}\text{NbO}_3$ system came both from a significant volume change caused by the field-induced antiferroelectric-ferroelectric phase transition and from the domain contribution caused by the induced ferroelectric phase [42, 43]. Jo et al. suggested that origin of the large strain in $(\text{K}_{0.5}\text{Na}_{0.5})\text{NbO}_3$ -modified $(\text{Bi}_{0.5}\text{Na}_{0.5})\text{TiO}_3\text{-BaTiO}_3$ lead-free piezoceramics is due to the presence of a nonpolar phase that brings the system back to its unpoled state once when the applied electric field is removed, which leads to a large nonpolar strain [44]. In addition, Lee et al. reported that the giant EFIS was attributed to the transition from nonpolar to ferroelectric phases in BNKT-BiAlO_3 small grains with ferroelectric BNT large grains during external electric field execution [45]. Recently, Lee et al. suggested a model on the basis of the coexistence of polar nanoregions and a nonpolar matrix which can reversibly transform into a polar ferroelectric phase under cyclic fields via observation of giant EFIS in Sn doped BNKT [46]. Ullah et al. suggested that the origin of the large electric-field-induced giant strain is an inherently large electrostrictive strain combined with an additional strain introduced during electric-field-induced phase transition [47]. However, the origin of phase transition from polar to nonpolar due to doping is unclear because the explanation based on the distorted tolerance factor results from difference radius of dopants, but the tolerance factor just estimated the phase stability and could not predict the stability of structural type. The further mechanism understating needs to be further investigated.

(2) *Domain Switching.* The volume regions of the material with the same polarization orientation are referred to as ferroelectric domains [48]. When the sample is under zero field and strain-free conditions, all the domain states have the same energy; but if an electric field is applied, the free energy of the system is lowered by aligning the polarization along the electric field. Thus, large applied electric fields can permanently reorient the polarization between the allowed domain states, which are restricted by crystallography. As a result, polycrystals random orientation can be electrically poled to produce net piezoelectric coefficients. Recently, Ren pointed out that the large EFIS in ferroelectric crystals is caused by point-defect-mediated reversible domain switching [49]. It is noted that the defect dipoles tend to align along the spontaneous polarization direction which was suggested by electron paramagnetic resonance experiments [50, 51] and theoretical modeling [52, 53]. The domain switched and aligned by the applied electric field. The defects symmetry and defects dipole moment cannot be rotated in such diffusionless

process resulting in restoring force or reversing internal field that favored reverse domain switching when electric field is removed. However, theoretical calculation indicated that the ultrahigh electromechanical response in single-crystal piezoelectrics resulted from polarization rotation during poling processing [54].

3. Role of Dopants in $\text{Bi}_{0.5}(\text{Na},\text{K})_{0.5}\text{TiO}_3$

3.1. Role of Substitution in A-Site and B-Site in $\text{Bi}_{0.5}(\text{Na},\text{K})_{0.5}\text{TiO}_3$. The $\text{Bi}_{0.5}(\text{Na},\text{K})_{0.5}\text{TiO}_3$ is considered as a typical ABO_3 perovskite where Bi^{3+} , K^+ , and Na^+ ions localize at A-site and Ti^{4+} ions localize at B-site. In this part, the current studies of the effect of dopants on BNKT's properties have been presented.

3.1.1. Rare-Earth Doped BNKT. The rare earth elements are multivalent when they were doped in BNKT which resulted in interesting and complicating phenomena. Li et al. reported that electromechanical coupling factor (k_p) of $\text{Bi}_{0.5}\text{Na}_{0.44}\text{K}_{0.06}\text{TiO}_3$ was increased from 25.4 to 27.8% with 0.2 wt% CeO_2 dopant and then decreased with higher CeO_2 content [55]. Liao et al. also obtained enhancement in piezoelectric properties of $\text{Bi}_{0.5}(\text{Na}_{1-x-y}\text{K}_x\text{Li}_y)_{0.5}\text{TiO}_3$ via CeO_2 doping [56]. The $\text{Bi}_{0.5}(\text{Na}_{0.725}\text{K}_{0.175}\text{Li}_{0.1})_{0.5}\text{TiO}_3$ ceramics doped with 0.1 wt% CeO_2 show good performance with high piezoelectric constant ($d_{33} = 220$ pC/N) and high coupling factor ($k_p = 39.3\%$) [56]. Wang et al. reported the effects of La substitution at Bi-site in $(\text{Bi}_{1-x}\text{Na}_{0.8}\text{K}_{0.2}\text{La}_x)\text{TiO}_3$ which were k_p maximum of 28% with 0.5 wt% La doping and bipolar maximum strain of 0.16% with 2 wt.% La doping [57]. Yuan et al. reported the strain enhanced up to $k_p = 35\%$ in La-doped $[\text{Bi}_{0.5}(\text{Na}_{0.75}\text{K}_{0.15}\text{Li}_{0.10})_{0.5}]\text{TiO}_3$ [58, 59]. Yang et al. obtained the electromechanical coupling factor $k_p = 27\%$ by substitution of 0.0125 wt.% Nd_2O_3 in $0.82\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3\text{-}0.18\text{Bi}_{0.5}\text{K}_{0.5}\text{TiO}_3$ ceramics [60]. The 0.3 wt.% Sm_2O_3 substitution in $0.82\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3\text{-}0.18\text{Bi}_{0.5}\text{K}_{0.5}\text{TiO}_3$ ceramics exhibited the high planar coupling factor ($k_p = 22.4\%$) which were reported by Zhang et al. [61]. This group also obtained the enhancement of electromechanical coupling factor $k_p = 24.63\%$ with 0.2 wt.% Gd_2O_3 doping in $0.82\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3\text{-}0.18\text{Bi}_{0.5}\text{K}_{0.5}\text{TiO}_3$ ceramics [62]. Zhi-Hui et al. reported optimum value of 0.15 wt.% Dy_2O_3 added in $\text{Bi}_{0.5}(\text{Na}_{0.82}\text{K}_{0.18})_{0.5}\text{TiO}_3$ for enhancement of electrical properties [63]. Fu et al. obtained the effect of Ho_2O_3 and Er_2O_3 dopants in $0.82\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3\text{-}0.18\text{Bi}_{0.5}\text{K}_{0.5}\text{TiO}_3$ where the k_p was 24.26 and 23.82% for 0.1 wt% Ho_2O_3 and 0.6 wt.% Er_2O_3 dopants, respectively [64, 65]. Following this work, Fu et al. reported the effects of Eu_2O_3 on the structure and electrical properties of $0.82\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3\text{-}0.18\text{Bi}_{0.5}\text{K}_{0.5}\text{TiO}_3$ lead-free piezoelectric ceramics where the optimum doping of 0.2 wt.% Eu_2O_3 has displayed the highest planer coupling factor $k_p = 25.1\%$ [66].

3.1.2. Transition Metal Doped $\text{Bi}_{0.5}(\text{Na},\text{K})_{0.5}\text{TiO}_3$. Han et al. first reported that the adding CuO in $\text{Bi}_{0.5}(\text{Na},\text{K})_{0.5}\text{TiO}_3$ ceramics resulted in decreasing the sintering temperature [67]. Do et al. reported that the k_p of $\text{Bi}_{0.5}(\text{Na}_{0.82}\text{K}_{0.18})_{0.5}\text{TiO}_3$

decreased when it was added with CuO [68]. The S_{\max}/E_{\max} was 214 pm/V for added 0.02 mol CuO, but it increased to 427 pm/V for added 0.02 mol Nb_2O_5 [68]. Jiang et al. obtained the best piezoelectric properties with $k_p = 30\%$ for 0.2 wt.% Mn^{2+} doped $(\text{Na}_{0.5}\text{K}_{0.2})_{0.5}\text{Bi}_{0.5}\text{TiO}_3$ [69, 70]. In addition, the $(\text{Na}_{0.5}\text{K}_{0.2})_{0.5}\text{Bi}_{0.5}\text{TiO}_3$ -0.5 wt.% Mn exhibited strong ferroelectricity with remnant polarization $P_r = 38 \mu\text{C}/\text{cm}^2$ [69]. Mn doping restrained the ferroelectric to antiferroelectric phase transition because of oxygen vacancy [69]. Hu et al. obtained the optimal electric properties in 0.16 wt.% MnCO_3 -added $74\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3$ -20.8 $\text{Bi}_{0.5}\text{K}_{0.5}\text{TiO}_3$ -5.2 BaTiO_3 which displayed the piezoelectric strain $d_{33} = 140 \text{ pC}/\text{N}$, mechanical coupling $k_p = 18\%$, and mechanical quality $Q_m = 89$ while the depolarization temperature (T_d) stays relatively high at 175°C [71]. The effect of Mn and Co on electrostrains of $\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3$ - BaTiO_3 - $\text{Bi}_{0.5}\text{K}_{0.5}\text{TiO}_3$ has been investigated by Shieh et al. [72, 73]. It was remarkable that Mn doping with an electrostrain of about 0.1% can be maintained when the Mn doping amount is in between 0.5 and 1.5 mol%, which were contrast for the codoped $\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3$ - BaTiO_3 - $\text{Bi}_{0.5}\text{K}_{0.5}\text{TiO}_3$; the values of electrostrain and d_{33} stay relatively constant regardless of the Co-doping level [72, 73]. The 0.30 wt.% MnO-added $\text{Bi}_{0.485}\text{Na}_{0.425}\text{K}_{0.06}\text{Ba}_{0.03}\text{TiO}_3$ solid solutions were found to be with optimal electrical properties of $d_{33} = 109 \text{ pC}/\text{N}$ and $k_p = 32\%$ [74]. The highest k_p values for various dopants element substitution in BNKT has been shown in Figure 1. The highest k_p of 39.3% was reported for rare-earth Ce codoped with Li-modification BNKT ceramics.

3.1.3. Other Metal-Doped $\text{Bi}_{0.5}(\text{Na},\text{K})_{0.5}\text{TiO}_3$. Do et al. reported that $\text{Bi}_{0.5}(\text{Na}_{0.82}\text{K}_{0.18})_{0.5}\text{TiO}_3$ ceramics had the value of S_{\max}/E_{\max} of 566 pm/V when 2 mol.% of Ta^{5+} substituted on Ti^{4+} site which were compared to without dopants with value of S_{\max}/E_{\max} of 233 pm/V [75]. Pham et al. obtained the enhancement S_{\max}/E_{\max} up to 641 pm/V due to 3 mol.% Nb^{5+} substitution on Ti^{4+} ions [76]. Hussain et al. reported the piezoelectric coefficient of 641 pm/V for Zr^{4+} 0.43 mol.% concentration substitution in Ti^{4+} site [77]. In addition, Hussain et al. found that Hf substitution with 3 mol.% at Ti-site resulted in enhancement of the electric-field-induced strain up to 475 pm/V with corresponding strain of 0.38% at an applied electric field of 80 kV/cm [78]. Binh et al. reported the EFIS of 278 pm/V for 0.7 wt.% Y-doped $\text{Bi}_{0.5}(\text{Na}_{0.82}\text{K}_{0.18})_{0.5}\text{TiO}_3$ which were higher than without dopant of EFIS of 228 pm/V [79]. At this moment, the highest S_{\max}/E_{\max} was 727 pm/V by codopant Li and Ta in BNKT which was reported by Nguyen et al. [80]. The $[\text{Bi}_{0.5}(\text{Na}_{1-x-y}\text{K}_x\text{Li}_y)]_{0.5}\text{TiO}_3$ ceramics show excellent piezoelectric and ferroelectric properties, and the optimum properties were reported as follows: piezoelectric constant $d_{33} = 231 \text{ pC}/\text{N}$, planar and thickness electromechanical coupling factors $k_p = 41.0\%$ and $k_t = 50.5\%$, remanent polarization $P_r = 40.2 \mu\text{C}/\text{cm}^2$, and coercive field $E_c = 2.47 \text{ kV}/\text{mm}$ [81]. Recently, Lee et al. obtained S_{\max}/E_{\max} of 585 pm/V for 5 mol.% Sn doped in $\text{Bi}_{0.5}(\text{Na}_{0.82}\text{K}_{0.18})_{0.5}\text{TiO}_3$ [46]. Furthermore, Nguyen et al. reported the enhancement EFIS in $\text{Bi}_{0.5}(\text{Na}_{0.82}\text{K}_{0.18})_{0.5}\text{Ti}_{0.95}\text{Sn}_{0.05}\text{O}_3$ by additives Li where

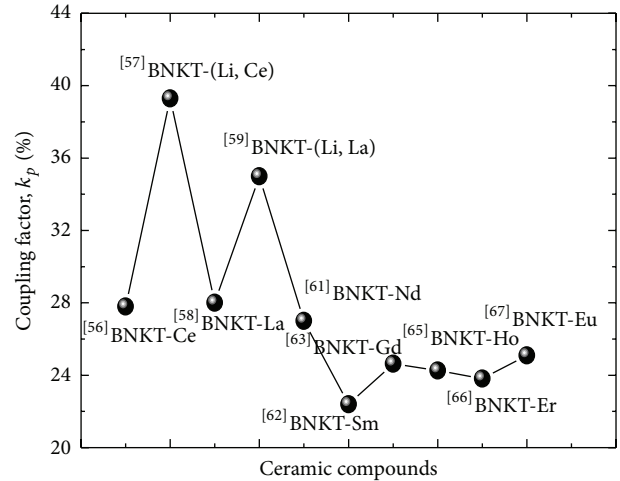


FIGURE 1: The effect of electric coupling factor in case of various dopants in BNKT.

S_{\max}/E_{\max} increased up to 646 pm/V when 4 mol.% Na was replaced with Li [82]. Liao et al. reported the effect of K and Ag dopant concentrations in $\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3$ ceramics. K- and Ag-doped ceramics exhibited good performances with piezoelectric constant $d_{33} = 189 \text{ pC}/\text{N}$, electromechanical coupling factor $k_p = 35.0\%$, remnant polarization $P_r = 39.5 \mu\text{C}/\text{cm}^2$, and coercive field $E_c = 2.3 \text{ kV}/\text{mm}$ [83]. Isikawa et al. cosubstituted Ag into A-site and Ba into B-site in BNKT and obtained enhancement in piezoelectric properties [84]. Moreover, this group also pointed out that the addition of $\text{La}_2\text{O}_3/\text{MnO}$ to BNKAT-BT specimens displayed a very large strain dynamic constant S_{\max}/E_{\max} of 415 pm/V which results from the field-forced phase transition from the paraelectric phase to the ferroelectric phase [84].

3.1.4. Nonstoichiometric Effects in $\text{Bi}_{0.5}(\text{Na},\text{K})_{0.5}\text{TiO}_3$. Ni et al. obtained the effects of A-site vacancy on the electrical properties in lead-free nonstoichiometric ceramics $\text{Bi}_{0.5+x}(\text{Na}_{0.82}\text{K}_{0.18})_{0.5-3x}\text{TiO}_3$ and $\text{Bi}_{0.5+y}(\text{Na}_{0.82}\text{K}_{0.18})_{0.5}\text{TiO}_3$ [85, 86]. The generation of A-site vacancy leads to a random defect field which results in destroying the long-range order phase induced by point field and makes the domain move easier. However, the effects of B-site vacancy or oxygen vacancy were not well-reported for BNKT system.

3.2. Role of Solid Solution of Secondary $A'B'O_3$ Dopants in $\text{Bi}_{0.5}(\text{Na},\text{K})_{0.5}\text{TiO}_3$. At the MPB of BNK-BKT binary system, an electric-field-induced strain and dynamic piezoelectric coefficient were 0.23% and 291 pm/V, respectively, at an applied electrical field of 80 kV/cm which are the considered value for application in electromechanical devices [87]. These values were low as compared with PZT-based materials; therefore it is impossible to apply for real electronic devices. The recent researches tried to enhance the dynamic piezoelectric coefficient by dopants via solid solution with other $A'B'O_3$ perovskites. In fact, the solid solution with small amount (~several mole percents) of $A'B'O_3$ perovskite

to BNKT resulted in strong enhancement the S_{\max}/E_{\max} values. Ullah et al. obtained large electric-field-induced strain in BiAlO₃-modified Bi_{0.5}(Na,K)_{0.5}TiO₃ with S_{\max}/E_{\max} of 592 pm/V at 3 mol.% BiAlO₃, near the tetragonal-pseudocubic phase boundary [87]. The S_{\max}/E_{\max} was slightly decreased to 579 pm/V via Bi_{0.5}La_{0.5}AlO₃-modified Bi_{0.5}(Na_{0.78}K_{0.22})_{0.5}TiO₃ [88]. Tran et al. modified Bi_{0.5}(Na_{0.82}K_{0.18})_{0.5}TiO₃ ceramics with Sr(K_{1/4}Nb_{3/4})O₃ and got the significant temperature coefficient of 0.38 pm/V/K [89]. Wang et al. fabricated 5 mol.% SrTiO₃-modified Bi_{0.5}(Na_{0.8}K_{0.2})TiO₃ lead-free piezoceramic which had a large unipolar strain of 0.36% (S_{\max}/E_{\max} = 600 pm/V) at a driving field of 60 kV/cm at room temperature [90]. The Bi(Zn_{0.5}Ti_{0.5})O₃ ceramic was found to enhance the electromechanical strain with S_{\max}/E_{\max} of 547 pm/V and 500 pm/V for 5Bi(Zn_{0.5}Ti_{0.5})O₃-40(Bi_{0.5}K_{0.5})TiO₃-55(Bi_{0.5}Na_{0.5})TiO₃ and 2 mol.% doped, respectively [91–93]. Hussain et al. obtained the S_{\max}/E_{\max} of 434 pm/V with 3 mol.% K_{0.5}Na_{0.5}NbO₃-modified Bi_{0.5}(Na_{0.78}K_{0.22})_{0.5}TiO₃ ceramic [94]. The 82Bi_{0.5}Na_{0.5}TiO₃-16Bi_{0.5}K_{0.5}TiO₃-3KNbO₃ ceramics had good performances with piezoelectric constant d_{33} = 138 pC/N and electromechanical coupling factor k_p = 38% [95]. Do et al. obtained the EFIS values of 443 pm/V in 4 mol.% LiTaO₃-modified 78Bi_{0.5}Na_{0.5}TiO₃-18Bi_{0.5}K_{0.5}TiO₃ [96]. Ngoc et al. reported that maximum strain of 2 mol.% BaZrO₃-modified Bi_{0.5}(Na_{0.82}K_{0.18})_{0.5}TiO₃ ceramics was two times higher than undoped case [97]. In addition, they pointed out that the modification EFIS of BaZrO₃ was better than that of BaTiO₃ [98]. The electric-field-induced strain was significantly enhanced by the CaZrO₃-induced phase transition and reached the highest value of S_{\max}/E_{\max} of 617 pm/V when Bi_{0.5}(Na_{0.78}K_{0.22})_{0.5}TiO₃ was doped with 3 mol.% CaZrO₃ dopant [99]. Kang et al. obtained S_{\max}/E_{\max} of 333 pm/V with 4 mol.% additive Bi_{0.5}(Na_{0.82}K_{0.18})_{0.5}TiO₃, and it was further increased to 363 pm/V for 2 mol.% CuO-added specimen [100]. Recently, we obtained the enhancement of S_{\max}/E_{\max} up to 668 pm/V due to modify A-site by Li in lead-free BNKT-modified with CaZrO₃ [101]. Zaman et al. obtained S_{\max}/E_{\max} of 500 pm/V via codoped Zr and LiSbO₃ [102]. The Ba_{0.85}Ca_{0.15}Ti_{0.90}Zr_{0.10}O₃, BaTiO₃, NaSbO₃, LiNbO₃, BiGaO₃ and Ba_{0.85}Ca_{0.15}Ti_{0.90}Zr_{0.10}O₃ were also found to enhance the electromechanical properties when those were solute in Bi_{0.5}(Na,K)_{0.5}TiO₃ [103–109]. Interestingly, the multiferroics materials such as BiFeO₃, BiMnO₃, and BiCrO₃-modified BNKT result in enhancement in electromechanical properties [110–112]. However, solution and/or composite BNKT with ferromagnetic such as BNKT-CoFe₂O₄ and BNKT-Fe₃O₄ were not reported [113].

4. Discussion

The mechanism of giant electric-field-induced strain could be considered as (i) electric-field-induced phase transition and/or (ii) point-defect-mediated reversible domain switching. At room temperature, the BNT system is in rhombohedral structure, and BKT is in tetragonal structure.

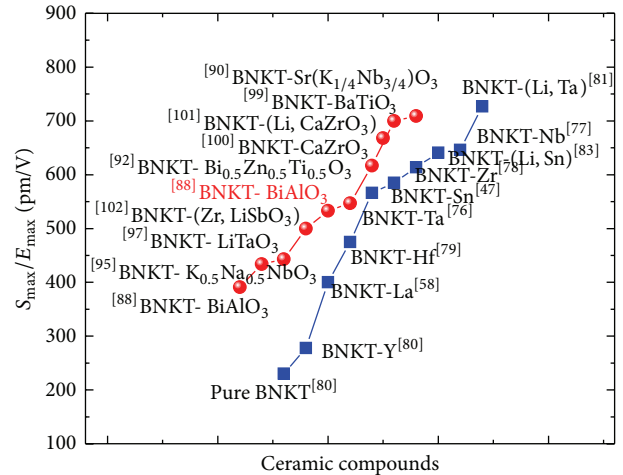


FIGURE 2: The effect of dopants and solid solution perovskite ABO₃ to BNKT.

Their solid solutions have rhombohedral-tetragonal morphotropic phase boundary near 0.16–0.20 of BKT amount [14]. The effects of elements dopants or codopants at A- or B-site of BNKT were found to be effective the piezoelectric properties by two mechanisms based on the difference radii point of view. Firstly, the dopants distorted the tolerance radii factor due to the difference the radii of dopant in comparing with Na⁺, K⁺, Bi³⁺, or Ti⁴⁺. The distortion results in phase transition from tetragonal to pseudocubic or tetragonal development from pseudocubic which was strongly related to electric-field-induced phase transition mechanism. Secondly, the dopants created the oxygen vacancy due to difference the valence state when the radii were performed which were related to point-defect-mediated reversible domain switching mechanism. The effect of dopants and solid solution A'B'O₃ to BNKT has been summarized in Figure 2. The ceramics compositions displayed the highest S_{\max}/E_{\max} values which were shown in details in Table 1. The highest S_{\max}/E_{\max} of each year was plotted at Figure 3. The result indicated that electric-field-induced giant strain of BNKT was comparable with that of PZT (such as commercial PZT (PIC151)) and therefore these modified BNKT can become a promising piezoelectric ceramics to replace the lead-based piezoelectric materials. Actually, the S_{\max}/E_{\max} values of BNKT ceramics were increased when the element at B-site and/or A-site of BNKT ceramics were modified. However, the S_{\max}/E_{\max} values quickly increased with several percent of dopants and then decreased when further dopants were added. These phenomena were also obtained while A'B'O₃ modified BNKT as solid solution because A' and B' diffused as codopant at both A- and B-sites of BNKT with similar concentration. The observation of enhancement of dynamic piezoelectric coefficient was related to (i) distorted structure, (ii) phase transition from polar to nonpolar phase, and (iii) polar phase growth inner nonpolar phase. The recent explanation of phase transition due to dopant based on distorted tolerance factor because radius between host and dopants differed and oxygen vacancy

TABLE 1: The detailed composition of BNKT-modified ceramics with highest electric-field-induced strain.

Ceramic compounds	S_{\max}/E_{\max} (pm/V)	References
$0.86\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3 - 0.14\text{Bi}_{0.5}\text{K}_{0.5}\text{TiO}_3$	297	Izumi et al. (2008) [31]
$\text{La}_2\text{O}_3/\text{MnO}$ -added $(1-y)(\text{Bi}_{0.5}\text{Na}_{0.49-x}\text{K}_x\text{Ag}_{0.01})\text{TiO}_3 - y\text{BaTiO}_3$	415	Isikawa et al. (2009) [84]
$\text{Bi}_{0.5}(\text{Na}_{0.82}\text{K}_{0.18})_{0.5}\text{Ti}_{0.97}\text{Nb}_{0.03}\text{O}_3$	641	Pham et al. (2010) [76]
$\text{Bi}_{0.5}(\text{Na}_{0.78}\text{K}_{0.22})_{0.5}\text{TiO}_3$	296	Ullah et al. (2010), [87]
$0.97\text{Bi}_{0.5}(\text{Na}_{0.78}\text{K}_{0.22})_{0.5}\text{TiO}_3 - 0.03\text{BiAlO}_3$	592	Ullah et al. (2010) [87]
$0.95\text{Bi}_{0.5}(\text{Na}_{0.8}\text{K}_{0.2})_{0.5}\text{TiO}_3 - 0.05\text{BiAlO}_3$	391	Ullah et al. (2010) [115]
$0.99(\text{K}_{0.5}\text{Na}_{0.5})_{0.95}\text{Li}_{0.05}\text{NbO}_3 - 0.01\text{Bi}_{0.5}(\text{K}_{0.15}\text{Na}_{0.85})_{0.5}\text{TiO}_3$	330	Chen et al. (2010) [33]
$\text{Bi}_{0.5}(\text{Na}_{0.82}\text{K}_{0.18})_{0.5}\text{TiO}_3 - 0.7 \text{ wt.}\% \text{Y}_2\text{O}_3$	278	Binh et al. (2010) [79]
$\text{Bi}_{0.5}(\text{Na}_{0.78}\text{K}_{0.22})_{0.5}\text{Ti}_{0.97}\text{Zr}_{0.03}\text{O}_3$	614	Hussain et al. (2010) [77]
$\text{Bi}_{0.5}(\text{Na}_{0.78}\text{K}_{0.22})_{0.5}(\text{Ti}_{0.97}\text{Hf}_{0.03})\text{O}_3$	475	Hussain et al. (2010) [78]
$\text{Bi}_{0.5}(\text{Na}_{0.82}\text{K}_{0.18})_{0.5}\text{Ti}_{0.98}\text{Ta}_{0.02}\text{O}_3$	566	Do et al. (2011) [75]
$\text{Bi}_{1/2}\text{Na}_{1/2}\text{TiO}_3 - \text{Bi}_{1/2}\text{K}_{1/2}\text{TiO}_3 - 0.02\text{Bi}(\text{Zn}_{1/2}\text{Ti}_{1/2})\text{O}_3$	500	Dittmer et al. (2011) [93]
$\text{Bi}_{0.5}(\text{Na}_{0.75}\text{K}_{0.25})_{0.5}\text{TiO}_3 - \text{BiAlO}_3$	~900	Lee et al. (2011) [45]
$0.09\text{Bi}_{0.5}(\text{Na}_{0.78}\text{K}_{0.22})_{0.5}\text{TiO}_3 - 0.01\text{Bi}_{0.5}\text{La}_{0.5}\text{AlO}_3$	579	Ullah et al. (2012) [88]
$0.975\text{Bi}_{0.5}(\text{Na}_{0.78}\text{K}_{0.22})_{0.5}\text{TiO}_3 - 0.025\text{BiAlO}_3$	533	Ullah et al. (2012) [116]
$0.97\text{Bi}_{0.5}(\text{Na}_{0.78}\text{K}_{0.22})_{0.5}\text{TiO}_3 - 0.03\text{K}_{0.5}\text{Na}_{0.5}\text{NbO}_3$	434	Hussain et al. (2012) [94]
$\text{Bi}_{0.5}\text{Na}_{0.385}\text{Li}_{0.025}\text{K}_{0.09}\text{Ti}_{0.975}\text{Ta}_{0.025}\text{O}_3$	727	Nguyen et al. (2012) [80]
$\text{Bi}_{0.5}(\text{Na}_{0.82}\text{K}_{0.18})_{0.5}\text{Ti}_{0.95}\text{Sn}_{0.05}\text{O}_3$	585	Lee et al. (2012) [46]
$0.94\text{Bi}_{1/2}(\text{Na}_{0.8}\text{K}_{0.2})_{1/2}\text{TiO}_3 - 0.06\text{Bi}(\text{Mg}_{1/2}\text{Sn}_{1/2})\text{O}_3$	633	Pham et al. (2012) [117]
$\text{Bi}_{0.5}(\text{Na}_{0.74}\text{Li}_{0.8}\text{K}_{0.18})_{0.5}\text{Ti}_{0.95}\text{Sn}_{0.05}\text{O}_3$	646	Nguyen et al. (2012) [82]
$\text{Bi}_{1/2}(\text{Na}_{0.82}\text{K}_{0.18})_{1/2}(\text{Ti}_{0.97}\text{Nb}_{0.03})\text{O}_3$	641	Pham et al. (2012) [118]
$0.97\text{Bi}_{0.5}(\text{Na}_{0.78}\text{K}_{0.22})_{0.5}\text{TiO}_3 - 0.03\text{CaZrO}_3$	617	Hong et al. (2013) [99]
$0.95\text{Bi}_{0.5}(\text{Na}_{0.8}\text{K}_{0.2})_{0.5}\text{TiO}_3 - 0.05\text{SrTiO}_3$	600	Wang et al. (2012) [90]
$0.99\text{Bi}_{0.5}(\text{Na}_{0.82}\text{K}_{0.18})_{0.5}\text{Ti}_{0.980}\text{Zr}_{0.020}\text{O}_3 - 0.01\text{LiSbO}_3$	500	Zaman et al. (2012) [102]
$0.97\text{Bi}_{0.5}(\text{Na}_{0.82}\text{K}_{0.18})_{0.5} - 0.03\text{Bi}(\text{Zn}_{0.5}\text{Ti}_{0.5})\text{O}_3$	385	Ullah et al. (2012) [119]
$5\text{Bi}(\text{Zn}_{0.5}\text{Ti}_{0.5})\text{O}_3 - 40(\text{Bi}_{0.5}\text{K}_{0.5})\text{TiO}_3 - 55(\text{Bi}_{0.5}\text{Na}_{0.5})\text{TiO}_3$	547	Patterson et al. (2012) [91]
$\text{Bi}_{0.5}(\text{Na}_{0.82}\text{K}_{0.18})_{0.5}\text{TiO}_3 - 0.02\text{CuO}$	214	Do et al. (2012) [68]
$\text{Bi}_{0.5}(\text{Na}_{0.82}\text{K}_{0.18})_{0.5}\text{TiO}_3 - 0.02\text{CuO} - 0.02\text{Nb}_2\text{O}_5$	427	Do et al. (2012) [68]
$0.98\text{Bi}_{0.5}(\text{Na}_{0.78}\text{K}_{0.22})_{0.5}\text{TiO}_3 - 0.02\text{LaFeO}_3$	~500	Han et al. (2012) [120]
$\text{Bi}_{0.5}(\text{Na}_{0.82}\text{K}_{0.18})_{0.5}\text{Ti}_{0.95}\text{Sn}_{0.05}\text{O}_3$	~600	Han et al. (2013) [121]
$0.98\text{Bi}_{0.5}(\text{Na}_{0.82}\text{K}_{0.18})_{0.5}\text{TiO}_3 - 0.02\text{BaZrO}_3$	437	Lee et al. (2013) [122]
$0.97\text{Bi}_{0.5}(\text{Na}_{0.82}\text{K}_{0.18})_{0.5}\text{TiO}_3 - 0.03\text{CaZrO}_3$	603	Lee et al. (2013) [122]
$0.98\text{Bi}_{0.5}(\text{Na}_{0.82}\text{K}_{0.18})_{0.5}\text{TiO}_3 - 0.02\text{Ba}_{0.8}\text{Ca}_{0.2}\text{ZrO}_3$	549	Lee et al. (2013) [122]
$0.94\text{Bi}_{0.5}(\text{Na}_{0.75}\text{K}_{0.25})_{0.5}\text{TiO}_3 - 0.06\text{BiAlO}_3$	930	Lee et al. (2013) [123]
$0.975\text{Bi}_{0.5}(\text{Na}_{0.80}\text{K}_{0.20})_{0.5}\text{TiO}_3 - 0.025\text{LiNbO}_3$	475	Hao et al. (2013) [124]
$0.96\text{Bi}_{0.5}(\text{Na}_{0.78}\text{K}_{0.22})_{0.5}\text{TiO}_3 - 0.04\text{Bi}(\text{Mg}_{0.5}\text{Ti}_{0.5})\text{O}_3$	636	Ullah et al. (2013) [125]
$40\text{Bi}_{0.5}\text{K}_{0.5}\text{TiO}_3 - 59\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3 - 1\text{Bi}(\text{Mg}_{1/2}\text{Ti}_{1/2})\text{O}_3$	422	Kumar and Cann (2013) [126]
$[\text{Bi}_{1/2}(\text{Na}_{0.82}\text{K}_{0.18})_{1/2}]_{0.97}\text{La}_{0.03}\text{TiO}_3$	715	Dinh et al. (2013) [127]
$0.95\text{Bi}_{0.5}(\text{Na}_{0.80}\text{K}_{0.20})_{0.5}\text{TiO}_3 - 0.05\text{Ba}(\text{Ti}_{0.90}\text{Sn}_{0.10})\text{O}_3$	649	Jaita et al. (2014) [128]
$0.97\text{Bi}_{0.5}(\text{Na}_{0.80}\text{K}_{0.20})_{0.5}\text{TiO}_3 - 0.03\text{SrZrO}_3$	617	Hussain et al. (2014) [129]
$0.99\text{Bi}_{0.5}\text{Na}_{0.4}\text{K}_{0.1}\text{Ti}_{0.98}\text{Nb}_{0.02}\text{O}_3 - 0.01(\text{Ba}_{0.7}\text{Sr}_{0.3})\text{TiO}_3$	634	Ullah et al. (2014) [130]
$\text{Bi}_{0.5}(\text{Na}_{0.80}\text{K}_{0.20})_{0.5}\text{TiO}_3 - (\text{K}_y\text{Na}_{1-y})\text{NbO}_3$	413–575	Hao et al. (2014) [131]
$0.99\text{Bi}_{0.5}(\text{Na}_{0.82}\text{K}_{0.18})_{0.5}\text{Ti}_{0.987}\text{Ta}_{0.013}\text{O}_3 - 0.01\text{LiSbO}_3$	650	Zaman et al. (2014) [132]
$0.99[(\text{Bi}_{0.5}\text{Na}_{0.4}\text{K}_{0.1})_{0.980}\text{La}_{0.020}\text{TiO}_3] - 0.02[\text{Ba}_{0.7}\text{Sr}_{0.3}\text{TiO}_3]$	650	Ullah et al. (2014) [133]

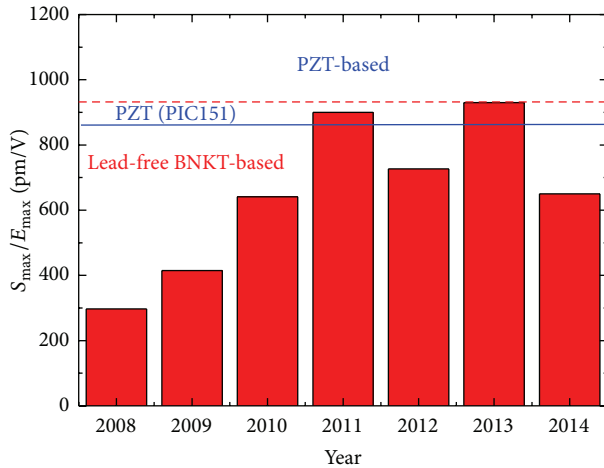


FIGURE 3: The highest values S_{\max}/E_{\max} observation in years (from 2008 to 2014) in lead-free BNKT-based ceramics.

caused difference of valence states. However, the tolerance factor has only estimated the range for stability of perovskite but it was not showed for crystal symmetries. Thus, these explanations were unreliable. The mechanism of electrical field-induced strain was not well understood which still required to further investigate. Consequently, the roles of *A*- and *B*-site on the electrical field-induced strain were unclear till now. However, we expected that the S_{\max}/E_{\max} values were further increased by using multidopant with optimal concentration of dopants for each position *A*- and/or *B*-site in BNKT ceramics.

In case of rare-earth doped BNKT, the enhancement of piezoelectric properties mostly resulted from the point-defect-mediated because rare-earth elements have various valences which was displayed by various radii of ion, depending on the valence stable state substitution. For example, the mechanism for the effect of CeO_2 doped BNKT is complicated because Ce ion possibly exists in the BNKT structure in two valence states: Ce^{4+} with radius of 0.92 Å and Ce^{3+} with radius of 1.03 Å. In view of the radius, Ce^{3+} is possible to fill in Bi^{3+} vacancies and Ce^{4+} can enter into the Bi-site. In this case, Ce^{4+} functions as a donor dopant leading to some vacancies of *A*-site in the lattice, which facilitates the movement of domain wall to improve the piezoelectric properties. Another one is that Ce^{3+} and Ce^{4+} ions occupy the *A*-site of Na^+ ($r_{\text{Na}^+} = 0.97\text{Å}$) of BNKT composition which are the same as the Ce^{4+} entering into Bi^{3+} [55, 56]. In these cases, existing vacancies bring defect in lattice which results in an increasing in the dielectric loss and grain size and have some effects of piezoelectric properties. In addition, the experiments demonstrated that rare-earth substitution in Na^+ first leads to creation of oxygen vacancies and lattice contraction, but when the dopants concentration is over a critical value then it begins to substitute for Bi^{3+} , thus it leads to lattice expansion [57–59].

In case of transition metal dopant in BNKT, the most findings confirmed that dopants resulted in the lower sintering

temperature. It is valuable in market due to reduction the cost of electronic devices.

In case of other metal dopant in BNKT, there were mixed point-defect-mediated and electric-field-induced phase transition mechanisms which depend on the valence and site-prefer to substitution. For example, the Ta^{5+} which replaced Ti^{4+} at *B*-site leads to creation of *A*-site vacancies which significantly contributed to the destabilization of ferroelectric phase in the Bi perovskite [75, 80, 114]. These factors were similar to a case of nonstoichiometric at *A*- and *B*-site of BNKT where the vacancies were occurred.

In case of solid solution between BNKT with other $A'B'O_3$ perovskites, it is quite interesting because the ideal was simple like tailor. Some of $A'B'O_3$ perovskites were unstable or hardly fabricated such as BiAlO_3 , but the theory predicted that they should have good piezoelectric properties. However, interestingly, they were stabled in solid solution in BNKT matrix and strong enhancement piezoelectric properties. Similarly, the $\text{Bi}_{0.5}\text{Zn}_{0.5}\text{TiO}_3$ (BZT) was a good ferroelectric material and it was tailed their properties in BNKT-BZT as solid solution. In addition, BNKT-modified with $\text{Bi}_{0.5}\text{Li}_{0.5}\text{TiO}_3$ has displayed the excellent piezoelectric constant d_{33} . Recently, the multiferroic materials have rapidly developed because they exhibited the electric field controlled ferromagnetism and magnetic field controlled the electric polarization. However, multiferroic properties of BNKT-based materials have not been reported. Therefore, we proposed that the solid solution and/or composite of BNKT with insulator ferromagnetism materials such as BiFeO_3 and CoFeO_4 could be a good display of multiferroics properties which will promise candidate to develop new class of multiferroics materials.

5. Conclusion

The current status of lead-free piezoelectric materials has been introduced. The lead-free BNKT-based ceramics were reviewed based on the fabrication method, effect of dopants, and solid-solution with other $A'B'O_3$ perovskite. Most findings confirmed that giant strain obtained in lead-free BNKT-based ceramics is due to phase transition from polar to nonpolar phase, and the highest S_{\max}/E_{\max} values were found at the MPB. However, the origin of phase transition was still debated. The S_{\max}/E_{\max} values of lead-free BNKT-based ceramics are comparable with soft lead-based PZT-based ceramics, which could be promising materials to readily replace the lead-based PZT-based materials in devices. In addition, the effects of secondary phase $A'B'O_3$ as solid-solution tailed the properties of BNKT matrix. Our work reviewed current developments in lead-free BNKT-based materials which were expected to understand current status of researching about BNKT-based ceramics, therefore, to be guidance for designing new class materials and applications.

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

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