Enhanced recoverable energy density of $0.94\text{Bi}_{0.50}(\text{Na}_{0.78}\text{K}_{0.22})_{0.50}\text{Ti}_{1-x}(\text{Fe}_{0.50}\text{Nb}_{0.50})_x\text{O}_3-0.06\text{BaZrO}_3$ lead-free relaxor ceramics doped by $(\text{Fe}_{0.50}\text{Nb}_{0.50})^{4+}$ complex ions

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Abstract

The innovative $0.94\text{Bi}_{0.50}(\text{Na}_{0.78}\text{K}_{0.22})_{0.50}\text{Ti}_{1-x}(\text{Fe}_{0.50}\text{Nb}_{0.50})_x\text{O}_3-0.06\text{BaZrO}_3$ (BNKTBZ-xFN) lead-free relaxor ceramics were designed and fabricated by the conventional solid-state technique. The dielectric, ferroelectric, energy storage, and impedance performance of these ceramics were investigated systematically. XRD patterns revealed that the $(\text{Fe}_{0.50}\text{Nb}_{0.50})^{4+}$ complex ions had completely dissolved in the lattice and constructed a single solid-solution. The addition of $(\text{Fe}_{0.50}\text{Nb}_{0.50})^{4+}$ complex ions had an obvious impact on the grain size and crystallinity of BNKTBZ-xFN ceramics, which led to a large dielectric breakdown strength. In addition, the degree of diffusivity (γ) for BNKTBZ-xFN ceramics were increased from ~ 1.72 to ~ 1.93 , indicating an enhanced relaxor behavior with the doping of $(\text{Fe}_{0.50}\text{Nb}_{0.50})^{4+}$ complex ions. The highest recoverable energy density (W_{rec}) $\sim 1.20 \text{ J/cm}^3$ and energy-storage efficiency (η) $\sim 72\%$ were attained at 120 kV/cm for BNKTBZ-2FN ceramic with the excellent fatigue resistance. Therefore, the BNKTBZ-2FN ceramic might be a promising candidate for energy-storage electronic devices.

Keywords BNT-based ceramics · Energy density · AC impedance · Fatigue properties

1 Introduction

As the rapid update of electronic devices, the requirement for novel materials with small size and large energy density becomes more and more urgent [1, 2]. Dielectric ceramics as a new promising capacitor material get much attention due to its large W_{rec} and η [3–5]. It is known that the value of W_{rec} for dielectric ceramics could be obtained by the formula: $W_{\text{rec}} = \int_{P_r}^{P_{\text{max}}} E dP$, where P_{max} , P_r , and E are maximum polarization, remanent polarization, and applied electric field [6–8], respectively. It is worth noting that the difference of $(P_{\text{max}} - P_r)$ and large dielectric breakdown strength (DBS) are beneficial to enhance the recoverable energy density [1, 9, 10]. At present, relaxor ferroelectric (RF) and anti-ferroelectric (AFE) ceramics are widely applied in

dielectric electronic products, due to their excellent energy-storage properties. Compared with RF ceramics, AFE ceramics dominate the major market for commercial ceramics due to its typical double hysteresis and larger W_{rec} . However, a large proportion of AFE materials are lead-based ceramics, and the lead element is unfriendly to the environment and human health [11–13]. As a result, lots of effort have been devoted to replacing lead-based AFE ceramics with lead-free RF ceramics.

Original $\text{Bi}_{0.50}\text{Na}_{0.50}\text{TiO}_3$ (BNT) ceramic is a promising lead-free ceramic used for energy-storage material. Pure BNT ceramics have large P_r ($= 38 \mu\text{C/cm}^2$) [14], P_{max} ($= 43 \mu\text{C/cm}^2$), and high DBS ($= 120 \text{ kV/cm}$) [15]. As described above, the high DBS and P_{max} of pure BNT ceramics will enhance the W_{rec} , but the large P_r would hinder the improvement of recoverable energy-storage

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density. In the literature for BNT-based ceramics [6, 16–18], it is reported that the chemical doping, such as A-site ions substitution, B-site ions substitution or complex ions substitution, is an effective procedure to decrease the P_r and induce an obvious relaxor characteristics for pure BNT ceramics. It would be beneficial to improve the W_{rec} . In 2007, Yao et al. [19] discovered that $(\text{Bi}_{0.5}\text{Na}_{0.5})_{0.94}\text{Ba}_{0.06}\text{Zr}_{0.04}\text{Ti}_{0.96}\text{O}_3$ ceramics presented a typical relaxor characteristic with unchanged P_{max} but reduced $P_r \sim 7.38 \mu\text{C}/\text{cm}^2$, which was helpful to the enhancement of W_{rec} . In the meantime, Shen et al. [20] found that $0.72\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3-0.18\text{Bi}_{0.5}\text{K}_{0.5}\text{TiO}_3-0.10\text{SrTiO}_3$ thin films modified by Fe^{3+} ions attained a large $W_{rec} \sim 20.34 \text{ J}/\text{cm}^3$, indicating that Fe^{3+} ions will optimize the energy-storage density performance of BNT-based ceramics. In 2016, Zhao et al. [21] found that $(\text{Bi}_{0.5}\text{Na}_{0.5})_{0.94}\text{Ba}_{0.06}\text{TiO}_3$ ceramics doped by $(\text{Al}_{0.50}\text{Nb}_{0.50})^{4+}$ complex ions would enhanced electrical properties significantly. Therefore, it is rational to substitute the B-site ions of BNT-based ceramics by complex ions to improve energy-storage density [22, 23]. $0.94\text{Bi}_{0.5}(\text{Na}_{0.78}\text{K}_{0.22})_{0.5}\text{TiO}_3-0.06\text{BaZrO}_3$ system doped by $(\text{Fe}_{0.50}\text{Nb}_{0.50})^{4+}$ complex ions were prepared in the present work and their structure, microstructure, AC impedance, energy-storage density, and fatigue resistance were studied systematically.

2 Experimental procedure

$0.94\text{Bi}_{0.50}(\text{Na}_{0.78}\text{K}_{0.22})_{0.50}\text{Ti}_{1-x}(\text{Fe}_{0.50}\text{Nb}_{0.50})_x\text{O}_3-0.06\text{BaZrO}_3$ (abbreviated as BNKTBZ-xFN, where $x = 0, 0.01, 0.02, 0.03$ and 0.04) ceramics were fabricated using the traditional solid-state reaction method. Raw powders of Bi_2O_3 (99%), Na_2CO_3 (99.8%), K_2CO_3 (99%), Nb_2O_5 (99.5%), TiO_2 (98%), BaCO_3 (98%), Fe_2O_3 (99%) and ZrO_2 (99%) were used and weighed in their calculated stoichiometric ratios. The preparation of these ceramics was similar to our previous work [6, 24]. At first, the weighed powders were mixed and ball-milled for 12 h with alcohol as the medium. Secondly, drying in the oven for 12 h and sieving, the mixtures were calcined at 840°C for 4 h and re-milled for 12 h. The re-milled powders were dried and pressed into small pellets under 150 MPa by the cold isostatic pressing technique and then sintered at 1160°C for 2 h in air atmosphere. In order to prevent the evaporation of Na^+ , K^+ , and Bi^{3+} ions in the sintering process, all pellets were embedded in the same composition powders. The sintered ceramics were polished using silica carbide sheets to a thickness of $\sim 0.50 \text{ mm}$, and silver paste was covered on both sides and then heated at 550°C for 30 min.

The phase purity and structure of the BNKTBZ-xFN ceramics were characterized using X-ray powder diffractometer (XRD; D/Max-RB, Rigaku, Tokyo, Japan) with Cu-K α radiation. The measurement was done using the

Bragg angle (2θ) ranging from 10° to 90° with $3^\circ/\text{min}$ scan rate. The surface morphology of the sintered ceramics was studied by scanning electron microscope (SEM; JSM EMP-800, JEOL, Tokyo, Japan). The ferroelectric property analyzer (TF-2000, aixACCT, Aachen, Germany) was used to test ferroelectric properties. The dielectric properties of all ceramics were investigated by the precision impedance analyzer (4294A, Agilent, Santa Clara, CA). The temperature-dependent ac-impedance spectra were recorded by an impedance analyzer (1260A, Solartron, Hampshire, UK).

3 Results and discussion

Room-temperature XRD spectra of BNKTBZ-xFN ceramics are displayed in Fig. 1a. All the ceramics display the perovskite-type structure with no obvious secondary phase, indicating that the Fe^{3+} and Nb^{5+} ions have completely dissolved into the pure BNKTBZ ceramic and form a solid-solution. In addition, the zoomed XRD patterns of (111) and (200) peaks are shown in Fig. 1b. It is noticed that (111) and (200) peaks present a single peak without any peak splitting phenomenon, suggests the pseudo-cubic symmetry of BNKTBZ-xFN ceramics [25]. Also, the characteristic peaks shift toward a low diffraction angle, suggesting the expansion of crystal lattice. This might be related to the difference between Ti^{4+} ($0.605 \text{ \AA}$), Fe^{3+} ($0.645 \text{ \AA}$), and Nb^{5+} ions ($0.64 \text{ \AA}$) [26, 27].

The surface microstructure of BNKTBZ-xFN ceramics is displayed in Fig. 2a–e. It is revealed that all ceramics show a dense, and homogeneous surface microstructure without any obvious pores. In addition, all of the ceramics present cubic and cuboid like shape with the distinct grain boundaries. It is reported that the fine grain size and less pore are beneficial to obtained high DBS and as a result

Fig. 1 The XRD patterns of BNKTBZ-xFN ceramics for 2θ of **a** 20° – 70° and **b** 39° – 47.5°

Fig. 2 SEM images of BNKTBZ-xFN ceramics, where **a** BNKTBZ, **b** BNKTBZ-1FN, **c** BNKTBZ-2FN, **d** BNKTBZ-3FN, **e** BNKTBZ-4FN, and **f** the average grain size as the function of composition

excellent energy-storage properties in the literature [21, 28]. The average grain size distribution plots are presented in inset Fig. 2a–e. As illustrated in Fig. 2f, the average grain size firstly increases and then decreases with the increasing $(\text{Fe}_{0.50}\text{Nb}_{0.50})^{4+}$ complex ions. In addition, the maximum value of $\sim 0.93 \mu\text{m}$ is attained for BNKBT-2FN ceramic.

Figure 3 presents the temperature-dependent dielectric properties of BNKTBZ-xFN ceramics for the temperature range (30–400 °C) at the frequencies of 1, 10, and 100 kHz. All of the ceramics present two typical permittivity peaks T_s and T_m . The first peak T_s refers to the phase transition from ferroelectric to relaxor ferroelectric phase

Fig. 3 Temperature-dependent dielectric permittivity (ϵ') and loss tangent ($\tan \delta$) of BNKTBZ-xFN ceramics, where **a** BNKTBZ, **b** BNKTBZ-1FN, **c** BNKTBZ-2FN, **d** BNKTBZ-3FN, **e** BNKTBZ-4FN composi-

tion, and **f** plots of $\ln(1/\epsilon' - 1/\epsilon'_m)$ versus $\ln(T - T_m)$ for all ceramics at the frequency of 1 kHz

and exhibit a distinct frequency-dispersion phenomenon. This is attributed to the thermal evolution of *R3c* and *P4bm* polar nanoregions (PNRs) [29]. The second peak T_m is corresponding to the relaxation process of *R3c-P4bm* PNRs and thermal evolution of *P4bm* PNRs [30]. It is revealed that T_s decreases slightly with the increasing composition. The peak of T_m does not show any significant variation. In addition, the peak T_m exhibits more diffused characteristics. It indicates that the doping of $(\text{Fe}_{0.50}\text{Nb}_{0.50})^{4+}$ complex ions induce charge disorder and form a random electric field [31]. The random field can destroy the long-range ferroelectric order and increase the weakly polar phase, which leads to a significant relaxor characteristic [32, 33]. In addition, the degree of diffusivity (γ) is used to describe the relaxor ferroelectric using the following equation [34]:

$$\ln \frac{1}{\epsilon'} - \frac{1}{\epsilon'_m} = \gamma \ln(T - T_m) + C \quad (1)$$

where ϵ'_m and T_m are the maximum dielectric permittivity and the corresponding temperature, respectively. The γ varies from 1 for a typical ferroelectric ceramics to 2 for a complete relaxor ferroelectrics [30, 35]. The plots of

$\ln \frac{1}{\epsilon'} - \frac{1}{\epsilon'_m}$ versus $\ln(T - T_m)$ data and corresponding linear fitting results are presented in Fig. 3f. The calculated γ value increases from ~ 1.72 for pure BNKTBZ to ~ 1.93 for BNKTBZ-4FN ceramics with the addition of $(\text{Fe}_{0.50}\text{Nb}_{0.50})^{4+}$ complex ions. It suggests that the $(\text{Fe}_{0.50}\text{Nb}_{0.50})^{4+}$ complex ions can improve the relaxation for all of the ceramics remarkably.

To clarify the relaxation mechanism and defects for BNKTBZ-xFN ceramics, the impedance spectroscopy is used to analyze as shown in Fig. 4. The impedance Z and modulus M are usually applied to describe the influence of grain and grain boundary effect in BNT-based ceramics [1, 36, 37]. Complex AC impedance curves of the BNKTBZ-xFN at 873 K and BNKTBZ-2FN ceramic at various temperatures are presented in Fig. 4a and b, respectively. It is noticed that only one single semicircle for all curves, indicating a single conduction mechanism arising from the grain contribution [38]. The only grain effect contribution can be further demonstrated by Fig. 4c. It is revealed that the peak of $Z''/Z_{\max}''$ and $M''/M_{\max}''$ coincide at a similar frequency. It is generally assumed that if the two typical peaks coincide at the same frequency, which suggests a long-range

Fig. 4 **a** AC impedance curves of the BNKTBZ-xFN ceramics at 873 K, **b** impedance plots of BNKTBZ-2FN composition at various temperatures, **c** plots of $Z''/Z_{\max}''$ and $M''/M_{\max}''$ for BNKTBZ-2FN

ceramic as the function of frequency, and **d** the Arrhenius curves of bulk conductivity for all ceramics

grain conduction mechanism, otherwise the short-range grain boundary conduction in BNT-based ceramics [30, 39]. Therefore, the long-range grain conduction mechanism plays a dominant role in the relaxation behavior of the BNKTBZ-2FN ceramic. The activation energy (E_a) in the BNKTBZ- x FN ceramics is obtained by the Arrhenius equation [40]:

$$\ln \sigma = \sigma_c \exp \left(\frac{-E_a}{KT} \right) \quad (2)$$

where σ , σ_c , and k are the direct-current conduction, the pre-exponent constant, and the Boltzmann constant, respectively. The value of E_a varies from ~ 0.86 to 0.91 eV attained by the fitting line of $\ln \sigma$ versus $1000/T$ as shown in Fig. 4d, which corresponds to the E_a of oxygen vacancies [29]. The E_a of oxygen vacancy is associated with the concentration of oxygen vacancies. Therefore, a small E_a value of BNKTBZ-4FN ceramic implies that the more significant oxygen vacancies induced relaxation compared to other ceramics.

The P - E loops of BNKTBZ- x FN ceramics are presented in Fig. 5a. The BNKTBZ- x FN ceramics display evident relaxor characteristics with slim and slanted loops. The J - E curves for all ceramics are exhibited in Fig. 5b. The leakage current density decreases significantly by the introduction of $(\text{Fe}_{0.50}\text{Nb}_{0.50})^{4+}$ complex ions, indicating to the enhancement of DBS and W_{rec} . In addition, only one current peak P_1 is observed in the J - E curves. It indicates the phase transition of ferroelectric-relaxor ferroelectric [41]. As plotted in Fig. 5c, with the addition of $(\text{Fe}_{0.50}\text{Nb}_{0.50})^{4+}$ complex ions,

the value of P_{max} does not show an obvious change. However, the P_r decreases gradually, and the $(P_{\text{max}} - P_r)$ attains the maximum value of $25.87 \mu\text{C}/\text{cm}^2$ for BNKTBZ-2FN ceramic, suggesting that the BNKTBZ-2FN ceramic might possess the large W_{rec} . The total energy density is calculated by the formula $W = \int_0^{P_{\text{max}}} E dP$, and η can be achieved using the formula $\eta = W_{\text{rec}}/W$ [42]. The change of W , W_{rec} , and η is presented in Fig. 5d. With the doping of $(\text{Fe}_{0.50}\text{Nb}_{0.50})^{4+}$ complex ions, the calculated W and W_{rec} are found to increase firstly and then after the decrease. BNKTBZ-2FN ceramic achieves the highest $W_{\text{rec}} \sim 0.88 \text{ J}/\text{cm}^3$ with $\eta \sim 70\%$, which is consistent with the analysis of Fig. 5c.

As described above, the BNKTBZ-2FN ceramic presents high energy-storage performance with the addition of $(\text{Fe}_{0.50}\text{Nb}_{0.50})^{4+}$ complex ions. Therefore, it is reasonable to make a comparison between BNKTBZ and BNKTBZ-2FN ceramics. As plotted in Fig. 6a and b, the BNKTBZ-2FN ceramic exhibits more pinched loops and large DBS. The BNKTBZ-2FN ceramic has the larger DBS $\sim 120 \text{ kV}/\text{cm}$ compared to $90 \text{ kV}/\text{cm}$ for the BNKTBZ ceramic, which indicates that $(\text{Fe}_{0.50}\text{Nb}_{0.50})^{4+}$ complex ions have a significant effect in the DBS. In addition, the W_{rec} increases from $0.75 \text{ J}/\text{cm}^3$ at $90 \text{ kV}/\text{cm}$ for pure BNKTBZ to $1.2 \text{ J}/\text{cm}^3$ for BNKTBZ-2FN ceramic at $120 \text{ kV}/\text{cm}$ as presented in Fig. 6c and d. Meanwhile, the energy-storage efficiency η also increases significantly from 56% for BNKTBZ ceramic to 72% for BNKTBZ-2FN ceramic. The comparison of W_{rec} for BNKTBZ-2FN ceramic with different BNT-based systems are presented

Fig. 5 **a** The polarization (P) and **b** leakage current density (J) versus the applied electric field (E) for all ceramics; **c** The P_{max} , P_r , $P_{\text{max}} - P_r$ and **d** the total energy density (W), the recoverable energy density (W_{rec}), the energy-storage efficiency (η) as the function of composition

Fig. 6 The P - E loops for (a) BNKTBZ and (b) BNKTBZ-2FN ceramics; the change of W , W_{rec} and η for (c) BNKTBZ and (d) BNKTBZ-2FN ceramics

Fig. 7 The comparison of W_{rec} for BNKTBZ-2FN ceramic with other BNT-based ceramics

Fig. 8 The fatigue performance of BNKTBZ-2FN ceramic measured at 60 kV/cm from 10^0 to 10^5 switching cycles

in Fig. 7 [7, 13, 43–49], and BNKTBZ-2FN ceramic present an excellent recoverable energy density.

The fatigue property of dielectric ceramic is an important characteristic for the application in electronic devices. Therefore, the fatigue resistance of the BNKTBZ-2FN ceramic is carried out and presented in Fig. 8. P - E loops of the BNKTBZ-2FN ceramic are almost unchanged up to 10^5 switching cycles. It is observed that the estimated P_{max} , P_r and W_{rec} do not decrease at all. It suggests excellent fatigue stability for BNKTBZ-2FN ceramic.

4 Conclusion

A series of novel lead-free BNKTBZ- x FN relaxor ceramics were investigated for energy-storage applications in detail. XRD patterns indicated that the $(\text{Fe}_{0.5}\text{Nb}_{0.5})^{4+}$ complex ions had dissolved in the BNKTBZ lattice and formed a stable solid-solution. ϵ' - T curves presented an

obvious relaxor characteristic, and J - E loops showed a decreased leakage current density with the addition of $(\text{Fe}_{0.5}\text{Nb}_{0.5})^{4+}$ ions. In addition, the degree of diffusivity (γ) of BNKTBZ- x FN ceramics increased from 1.72 to 1.93 with the increasing composition. High W_{rec} of $\sim 1.20 \text{ J/cm}^3$ was achieved for BNKTBZ-2FN composition with the applied electric field of 120 kV/cm. The AC impedance suggested the oxygen vacancy conduction mechanism and grain effect induced relaxation behavior in BNKTBZ- x FN relaxor ceramics. In summary, the BNKTBZ-2FN ceramic with high fatigue resistance might be a promising candidate for energy-storage devices.

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Compliance with ethical standards

Conflict of interest The author(s) declare that they have no competing interests.

References

- Wang C, Li Q, Zhang W, Yan B, Yadav AK, Peng H, Fan H (2020) $[\text{Bi}_{0.5}(\text{Na}_{0.4-x}\text{Li}_x\text{K}_{0.1})]_{0.96}\text{Sr}_{0.04}\text{Ti}_{0.975}\text{Ta}_{0.025}\text{O}_3$ lead-free RELAXOR ceramics with the enhanced recoverable energy density. *Ceram Int* 46:715–721
- Liu X, Yang H, Yan F, Qin Y, Lin Y, Wang T (2019) Enhanced energy storage properties of BaTiO_3 - $\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3$ lead-free ceramics modified by $\text{SrY}_{0.5}\text{Nb}_{0.5}\text{O}_3$. *J Alloy Compd* 778:97–104
- Xu Q, Li T, Hao H, Zhang S, Wang Z, Cao M, Yao Z, Liu H (2015) Enhanced energy storage properties of NaNbO_3 modified $\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3$ based ceramics. *J Eur Ceram Soc* 35:545–553
- Li Q, Li M, Wang C, Zhang M, Fan H (2019) Enhanced temperature stable dielectric properties and energy-storage density of BaSnO_3 -modified $(\text{Bi}_{0.5}\text{Na}_{0.5})_{0.94}\text{Ba}_{0.06}\text{TiO}_3$ lead-free ceramics. *Ceram Int* 45:19822–19828
- Wang G, Lu Z, Li J, Ji H, Yang H, Li L, Sun S, Feteira A, Yang H, Zuo R, Wang D, Reaney IM (2020) Lead-free $(\text{Ba}, \text{Sr})\text{TiO}_3$ - BiFeO_3 based multilayer ceramic capacitors with high energy density. *J Eur Ceram Soc* 40:1779–1783
- Wang C, Li Q, Yadav AK, Peng H, Fan H (2019) $\text{Bi}_{0.48}(\text{Na}_{0.84}\text{K}_{0.16})_{0.48}\text{Sr}_{0.04}(\text{Ti}_{1-x}\text{Ta}_x)\text{O}_3$ lead-free ceramics with enhanced electric field-induced strain. *J Alloys Compd* 803:1082–1089
- Hao J, Xu Z, Chu R, Li W, Juan D, Peng F (2015) Enhanced energy-storage properties of $(1-x)[(1-y)(\text{Bi}_{0.5}\text{Na}_{0.5})\text{TiO}_3-y(\text{Bi}_{0.5}\text{K}_{0.5})\text{TiO}_3]-x(\text{K}_{0.5}\text{Na}_{0.5})\text{NbO}_3$ lead-free ceramics. *Solid State Commun* 204:19–22
- Luo L, Wang B, Jiang X, Li W (2014) Energy storage properties of $(1-x)(\text{Bi}_{0.5}\text{Na}_{0.5})\text{TiO}_3$ - $x\text{KNbO}_3$ lead-free ceramics. *J Mater Sci* 49:1659–1665
- Ma W, Fan P, Salamon D, Kongparakul S, Samart C, Zhang T, Zhang G, Jiang S, Chang J, Zhang H (2019) Fine-grained BNT-based lead-free composite ceramics with high energy-storage density. *Ceram Int* 45:19895–19901
- Chen J, Qi H, Zuo R (2020) Realizing stable relaxor antiferroelectric and superior energy storage properties in $(\text{Na}_{1-x/2}\text{La}_{x/2})(\text{Nb}_{1-x}\text{Ti}_x)\text{O}_3$ lead-free ceramics through A/B-site complex substitution. *ACS Appl Mater Interfaces* 12:32871–32879
- Zhao L, Liu Q, Gao J, Zhang S, Li J (2017) Lead-free antiferroelectric silver niobate tantalate with high energy storage performance. *Adv Mater* 29:1701824
- Liu Z, Chen X, Peng W, Xu C, Dong X, Cao F, Wang G (2015) Temperature-dependent stability of energy storage properties of $\text{Pb}_{0.97}\text{La}_{0.02}(\text{Zr}_{0.58}\text{Sn}_{0.335}\text{Ti}_{0.085})\text{O}_3$ antiferroelectric ceramics for pulse power capacitors. *Appl Phys Lett* 106:262901
- Ding J, Liu Y, Lu Y, Qian H, Gao H, Chen H, Ma C (2014) Enhanced energy-storage properties of $0.89\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3$ - 0.06BaTiO_3 - $0.05\text{K}_{0.5}\text{Na}_{0.5}\text{NbO}_3$ lead-free anti-ferroelectric ceramics by two-step sintering method. *Mater Lett* 114:107–110
- Yanamandra R, Kandula KR, Bandi P, Reddy HSK, Asthana S, Patri T (2018) Enhanced energy storage density in lead free $(\text{Na}_{0.5}\text{Bi}_{0.48}\text{Eu}_{0.02})\text{Ti}_{1-x}\text{Nb}_x\text{O}_3$ ($x=0.00, 0.01$ & 0.02) ceramics. *AIP Conf Proc* 1953:050063
- Zhang L, Pu Y, Chen M, Li R, Guo X, Cui Y (2018) Enhanced energy-storage properties of $(1-x)\text{Na}_{0.5}\text{Bi}_{0.5}\text{TiO}_3$ - $x\text{BaSnO}_3$ ceramics. *Ceram Int* 44:S207–S210
- Li T, Fan H, Long C, Dong G, Sun S (2014) Defect dipoles and electrical properties of magnesium B-site substituted sodium potassium niobates. *J. Alloys Compd.* 609:60–67
- Fu P, Xu Z, Chu R, Li W, Xie Q, Zhang Y, Chen Q (2010) Effect of Dy_2O_3 on the structure and electrical properties of $(\text{Bi}_{0.5}\text{Na}_{0.5})_{0.94}\text{Ba}_{0.06}\text{TiO}_3$ lead-free piezoelectric ceramics. *J Alloys Compd* 508:546–553
- Hao J, Xu Z, Chu R, Li W, Du J (2015) Large electric-field-induced strain in SrZrO_3 modified $\text{Bi}_{0.5}(\text{Na}_{0.80}\text{K}_{0.20})_{0.5}\text{TiO}_3$ lead-free electromechanical ceramics with fatigue-resistant behavior. *J Alloys Compd* 647:857–865
- Yao Y, Tseng T, Chou C, Chen HHD (2007) Phase transition and piezoelectric property of $(\text{Bi}_{0.5}\text{Na}_{0.5})_{0.94}\text{Ba}_{0.06}\text{Zr}_y\text{Ti}_{1-y}\text{O}_3$ ($y=0-0.04$) ceramics. *J Appl Phys* 102:094102
- Wu S, Chen P, Zhai J, Shen B, Li P, Li F (2018) Enhanced piezoelectricity and energy storage performances of Fe-doped BNT-BKT-ST thin films. *Ceram Int* 44:21289–21294
- Zhao Y, Xu J, Zhou C, Yuan C, Li Q, Chen G, Wang H, Yang L (2016) High energy storage properties and dielectric behavior of $(\text{Bi}_{0.5}\text{Na}_{0.5})_{0.94}\text{Ba}_{0.06}\text{Ti}_{1-x}(\text{Al}_{0.5}\text{Nb}_{0.5})_x\text{O}_3$ lead-free ferroelectric ceramics. *Ceram Int* 42:2221–2226
- Sun Y, Zhao Y, Xu J, Yang L, Zhou C, Rao G, Wang H (2020) Phase transition, large strain and energy storage in ferroelectric $(\text{Bi}_{0.5}\text{Na}_{0.5})\text{TiO}_3$ - BaTiO_3 ceramics tailored by $(\text{Mg}_{1/3}\text{Nb}_{2/3})^{4+}$ complex ions. *J Electron Mater* 49:1131–1141
- Pang S, Yang L, Qin J, Qin H, Xie H, Wang H, Zhou C, Xu J (2019) Low electric field-induced strain and large improvement in energy density of $(\text{Lu}_{0.5}\text{Nb}_{0.5})^{4+}$ complex-ions doped BNT-BT ceramics. *Appl Phys A Mater Sci Process* 125:119
- Li Q, Gao S, Ning L, Fan H, Liu Z, Li Z (2017) Giant field-induced strain in Nb_2O_5 -modified $(\text{Bi}_{0.5}\text{Na}_{0.5})_{0.94}\text{Ba}_{0.06}\text{TiO}_3$ lead-free ceramics. *Ceram Int* 43:5367–5373
- Yao M, Pu Y, Zhang L, Chen M (2016) Enhanced energy storage properties of $(1-x)\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3$ - $x\text{Ba}_{0.85}\text{Ca}_{0.15}\text{Ti}_{0.9}\text{Zr}_{0.1}\text{O}_3$ ceramics. *Mater Lett* 174:110–113
- Velu S, Swamy CS (1997) Effect of substitution of $\text{Fe}^{3+}\text{Cr}^{3+}$ on the alkylation of phenol with methanol over magnesium-aluminium calcined hydrotalcite. *Appl. Catal. A: General.* 162:81–91
- Yan F, Yang H, Lin Y, Wang T (2017) Dielectric and ferroelectric properties of SrTiO_3 - $\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3$ - $\text{BaAl}_{0.5}\text{Nb}_{0.5}\text{O}_3$ lead-free ceramics for high-energy-storage applications. *Inorg Chem* 56:13510–13516

28. Wang T, Jin L, Li C, Hu Q, Wei X (2015) Relaxor ferroelectric BaTiO₃-Bi(Mg_{2/3}Nb_{1/3})O₃ ceramics for energy storage application. *J Am Ceram Soc* 98:559–566
29. Li Q, Wang C, Zhang W, Fan H (2019) Influence of compositional ratio K/Na on structure and piezoelectric properties in [(Na_{1-x}K_x)_{0.5}Bi_{0.5}]Ti_{0.985}Ta_{0.015}O₃ ceramics. *J Mater Sci* 54:4523–4531
30. Li Q, Ning L, Hu B, Peng H, Zhao N, Fan H (2019) Large strain response in (1-x) (0.94Bi_{0.5}Na_{0.5}TiO₃-0.06BaTiO₃)-xSr_{0.8}Bi_{0.1}□_{0.1}Ti_{0.8}Zr_{0.2}O_{2.95} lead-free piezoelectric ceramics. *Ceram Int* 45:1676–1682
31. Wang XX, Kwok KW, Tang XG, Chan HLW, Choy CL (2004) Electromechanical properties and dielectric behavior of (Bi_{1/2}Na_{1/2})_(1-1.5x)Bi_xTiO₃ lead-free piezoelectric ceramics. *Solid State Commun* 129:319–323
32. Yang Z, Hou Y, Pan H, Chang Y (2009) Structure, microstructure and electrical properties of (1-x-y) Bi_{0.5}Na_{0.5}TiO₃-xBi_{0.5}K_{0.5}TiO₃-yBi_{0.5}Li_{0.5}TiO₃ lead-free piezoelectric ceramics. *J Alloy Compd* 480:246–253
33. Rahman JU, Hussain A, Maqbool A, Ryu GH, Song TK, Kim W, Kim MH (2014) Field induced strain response of lead-free BaZrO₃-modified Bi_{0.5}Na_{0.5}TiO₃-BaTiO₃ ceramics. *J Alloys Compd* 593:97–102
34. Li X, Jing Q, Xi Z, Liu P, Long W, Fang P (2018) Dielectric relaxation and electrical conduction in (Bi_xNa_{1-x})_{0.94}Ba_{0.06}TiO₃ ceramics. *J Am Ceram Soc* 101:789–799
35. Liu L, Chen X, Jing R, Lian H, Wu W, Mou Y, Liu P (2019) Electrical and photoluminescence properties of (Bi_{0.5-x/0.94}Er_{x/0.94}Na_{0.5})_{0.94}Ba_{0.06}TiO₃ lead-free ceramics. *J Mater Sci: Mater in Electro* 30:5233–5239
36. Hajra S, Sahoo S, Das R, Choudhary RNP (2018) Structural, dielectric and impedance characteristics of (Bi_{0.5}Na_{0.5})TiO₃-BaTiO₃ electronic system. *J Alloys Compd* 750:507–514
37. Li Q, Zhang W, Wang C, Ma J, Ning L, Fan H (2018) Ag modified bismuth ferrite nanospheres as a chlorine gas sensor. *RSC Adv* 8:33156–33163
38. Peng B, Zhang Q, Lyu Y, Liu L, Lou X, Shaw C, Huang H, Wang Z (2018) Thermal strain induced large electrocaloric effect of relaxor thin film on LaNiO₃/Pt composite electrode with the coexistence of nanoscale antiferroelectric and ferroelectric phases in a broad temperature range. *Nano Energy* 47:285–293
39. Liu Z, Zhang A, Geng X, Lu J, Mao Y, Ouyang S (2019) A novel Na_{0.5}K_{0.5}NbO₃-La(Zn_{0.5}Ti_{0.5})O₃ ceramics with excellent dielectric properties at wide temperature range. *Ceram Int* 45:16842–16847
40. Lian H, Shao X, Chen X (2018) Structure and electrical properties of Ca²⁺-doped (Na_{0.47}Bi_{0.47}Ba_{0.06})TiO₃ lead-free piezoelectric ceramics. *Ceram Int* 44:11320–11330
41. Zhao Z, Ye M, Ji H, Li X, Zhang X, Dai Y (2018) Enhanced piezoelectric properties and strain response in <001> textured BNT-BKT-BT ceramics. *Mater. Design*. 137:184–191
42. Pang S, Yang L, Qin J, Qin H, Xie H, Wang H, Zhou C, Xu J (2019) Low electric field-induced strain and large improvement in energy density of (Lu_{0.5}Nb_{0.5})⁴⁺ complex-ions doped BNT-BT ceramics. *Appl Phys A* 125:119
43. Xie H, Yang L, Pang S, Yuan C, Chen G, Wang H, Zhou C, Xu J (2019) The evolution of phase structure, dielectric, strain, and energy storage density of complex-ions (Sr_{1/3}Nb_{2/3})⁴⁺ doped 0.82Bi_{0.5}Na_{0.5}TiO₃-0.18Bi_{0.5}K_{0.5}TiO₃ ceramics. *J Phys Chem Solid* 126:287–293
44. Li Q, Yao Z, Ning L, Gao S, Hu B, Dong G, Fan H (2018) Enhanced energy-storage properties of (1-x) (0.7Bi_{0.5}Na_{0.5}TiO₃-0.3Bi_{0.2}Sr_{0.7}TiO₃)-xNaNbO₃ lead-free ceramics. *Ceram Int* 44:2782–2788
45. Yao M, Pu Y, Zheng H, Zhang L, Chen M, Cui Y (2016) Improved energy storage density in 0.475BNT-0.525BCTZ with MgO addition. *Ceram Int* 42:8974–8979
46. Yu Z, Liu Y, Shen M, Qian H, Li F, Lyu Y (2017) Enhanced energy storage properties of BiAlO₃ modified Bi_{0.5}Na_{0.5}TiO₃-Bi_{0.5}K_{0.5}TiO₃ lead-free antiferroelectric ceramics. *Ceram Int* 43:7653–7659
47. Li Q, Zhang W, Wang C, Ning L, Wang C, Wen Y, Hu B, Fan H (2019) Enhanced energy-storage performance of (1-x) (0.72Bi_{0.5}Na_{0.5}TiO₃-0.28Bi_{0.2}Sr_{0.7}□_{0.1}TiO₃)-xLa ceramics. *J Alloys Compd* 775:116–123
48. Hu B, Fan H, Ning L, Gao S, Yao Z, Li Q (2018) Enhanced energy-storage performance and dielectric temperature stability of (1-x) (0.65Bi_{0.5}Na_{0.5}TiO₃-0.35Bi_{0.1}Sr_{0.85}TiO₃)-xKNbO₃ ceramics. *Ceram Int* 44:10968–10974
49. Wang B, Luo L, Jiang X, Li W, Chen H (2014) Energy-storage properties of (1-x) Bi_{0.47}Na_{0.47}Ba_{0.06}TiO₃-xKNbO₃ lead-free ceramics. *J Alloys Compd* 585:14–18

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