#### **ORIGINAL RESEARCH ARTICLE**



# High-Transmittance (K<sub>0.5</sub>Na<sub>0.5</sub>)NbO<sub>3</sub> Ferroelectric Ceramics Modified **by Sr(Bi**<sub>0.5</sub>Ta<sub>0.5</sub>)O<sub>3</sub>

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#### **Abstract**

(1−*x*)(K0.5Na0.5)NbO3-*x*Sr(Bi0.5Ta0.5)O3 (KNN-*x*SBT, *x*=0.018, 0.02, 0.022, 0.024, 0.026, 0.028) transparent ferroelectric ceramics were prepared using the solid-phase method. The phase structure, microstructure, light transmittance, and optical and electrical properties of KNN-*x*SBT were investigated. It was found that the pseudo-cubic phase structure, higher compactness, and smaller grains of KNN can be efectively regulated by introducing the second component, SBT, thus achieving strong light transmittance and good energy storage performance for KNN-*x*SBT ceramics. When *x*=0.022, the near-infrared band (1100 nm) transmittance of the ceramics reaches 72.2%, with energy storage density ( $W_{rec}$ ) of 0.53 J/  $cm<sup>3</sup>$ ; the maximum dielectric constant is obtained for ceramics at  $x = 0.018$ , with a value of 2935. The KNN-*x*SBT ceramic sample has relaxor ferroelectric properties and is an environmentally friendly lead-free transparent ferroelectric ceramic.

**Keywords** Transparent ceramics · lead-free · ferroelectric ceramics · energy storage

# **Introduction**

Transparent ceramics have been widely used in the feld of electronics, energy storage, and some extreme environments due to their better thermal, mechanical and excel-lent chemical stability.<sup>[1,](#page-5-0)[2](#page-5-1)</sup> As potential candidates for new electronic products, transparent ferroelectric ceramics have promising applications in emerging felds such as solid-state lighting, scintillating applications, composite armors, optical components, electro-optical devices, and even biomedi-cal materials, such as screens, etc.<sup>[3,](#page-5-2)[4](#page-5-3)</sup> To date, lead-based transparent ferroelectric ceramics such as lead lanthanum zirconate titanate (PLZT) have been widely studied and applied; however, the lead is harmful to human health and the environment,  $5.6$  $5.6$  which necessitates the active development of lead-free ceramic materials to replace lead-based ceramics.[7](#page-5-6),[8](#page-5-7)

Perovskite-type lead-free ceramics such as  $(Bi_0, Ba_0, 5)$  $TiO<sub>3</sub>$  (BNT) and  $(K<sub>0.5</sub>Na<sub>0.5</sub>)NbO<sub>3</sub>$  (KNN) have the poten-tial to replace lead-based ceramics.<sup>[9](#page-6-0)</sup> KNN ceramics have a large coupling coefficient, excellent electrical properties, high energy storage performance, and light transmittance in the visible and infrared regions, and have become the most widely studied calcium one of the ferroelectric materials with a titanite crystal structure.<sup>[10–](#page-6-1)12</sup> However, pure KNN ceramics are usually opaque and have low density due to the volatilization of alkali metal ions (such as  $K^+$  and  $Na^+$ ).<sup>13</sup> The transmittance of ceramics is afected by many factors, including phase structure, grain size, and relative density. $^{14}$  $^{14}$  $^{14}$ According to literature reports, the introduction of divalent ions into the positions A and B of the second component of KNN ceramics can achieve the effect of controlling the grain growth of the ceramics, and can also improve the density and adjust the phase structure.<sup>[15–](#page-6-5)[17](#page-6-6)</sup> Among them, doping of  $Sr^{2+}$  $(R=1.44 \text{ Å}, \text{CN}=12)$  can significantly improve the electri-cal properties of ceramic samples.<sup>[18,](#page-6-7)[19](#page-6-8)</sup> Bi<sub>2</sub>O<sub>3</sub> is usually used as a sintering aid, and it has an obvious efect on reducing the sintering temperature of ceramics, which can reduce the grain size and increase the density of ceramics.<sup>[20](#page-6-9)–22</sup> However, the high valence of Bi<sup>3+</sup> ( $R = 1.38$  Å, CN = 12) may lead to an increase in the obstacles of the ceramic structure and inhibit the growth of grains. $^{23}$  The solid solution of the second component  $Sr(Bi<sub>0.5</sub>Ta<sub>0.5</sub>)O<sub>3</sub>$  (SBT) significantly

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inhibits the growth of crystal grains, so that the grain size of the ceramic sample reaches the nanoscale, which efectively improves the optical transmittance of the ceramic sample. In addition, the grain size affects the breakdown strength (BDS) of the ceramic, where the smaller the grain size, the higher the BDS, and enhances the energy storage potential of the ceramic sample. $^{24}$  $^{24}$  $^{24}$ 

Transparent ceramics typically possess high density, few pores at grain boundaries, cubic phase structure with low anisotropy, lack of impurities, and moderate grain size. Additional attractive properties include fneness of raw powders, high uniformity of granulation, and appropriate sintering temperature. Nevertheless, special sintering techniques such as hot isostatic pressing (HIP), spark plasma sintering (SPS), and hot-pressing (HP) may impose excessive cost and the need for complicated processes.<sup>13</sup> HIP, a technique in which gas-assisted pressure is applied threedimensionally to the compact, seems to be advantageous for the fabrication of dense ceramics with homogeneous microstructure and desired complex shapes, in comparison with pressureless sintering and uniaxial HP techniques<sup>[25](#page-6-13)</sup>; spark plasma sintering (SPS), while reducing the volatility of obtained samples, is characterized by a rapid heating and consolidation rate. The use of SPS allows for dramatically reduced sintering times. Compared with traditional sintering processes, which take on the order of hours to days to complete, a typical SPS process ranges from tens of minutes to a few hours and provides overall better performance,<sup>[4](#page-5-3)</sup> whereas HP has a higher product yield and greatly enhanced specimen density, and reduced cracking and warping of the plates during the final densification phase.<sup>26</sup> But these processes have higher technological requirements, leading to problems of high cost and complex fabrication. Although it is difficult to obtain KNN-based transparent ceramics with stable composition and high density by solid-phase sintering, it can be used as a new method for the preparation of KNN-based transparent ceramics due to its advantages of simple fabrication and low cost.

In this study, a second component,  $Sr(Bi<sub>0.5</sub>Ta<sub>0.5</sub>)O<sub>3</sub>$ (SBT), is introduced into KNN to modify its phase structure, grain size, and relative density, thereby improving its optical and electrical properties.

# **Experimental Details**

 $(1-x)(K_{0.5}Na_{0.5})NbO<sub>3</sub>-xSr(Bi<sub>0.5</sub>Ta<sub>0.5</sub>)O<sub>3</sub>$  (KNN-*x*SBT, *x* = 0.018, 0.02, 0.022, 0.024, 0.026, 0.028) transparent ferroelectric ceramics were prepared using the solidphase method. Analytical-grade reagents  $Na_2CO_3$  (99.8%),  $K_2CO_3$  (99.5%),  $Nb_2O_5$  (99.99%),  $Ta_2O_5$  (99.99%),  $Bi_2O_3$ (99.999%), and  $SrCO<sub>3</sub>$  (99.9%) were used as the raw chemicals, with the error controlled within 0.0005 g, and 99% ethanol solution and zirconium balls were used as the medium. The powder was poured into a ball mill jar, and 50 ml of ethanol solution was added along with two kinds of zirconium balls with diameters of 5 mm and 8 mm in a quantity ratio of 2:1. The powder was subjected to a rolling ball mill at 410 rpm for 24 h, and then dried and sintered at 860°C with a heating rate of 4°C/min. The powder was ball milled a second time and dried, after which 5 wt.% polyvinyl alcohol (PVA) solution was added and the mixture was passed through a 100-mesh sieve. The granulated powders were pressed into a disk with a diameter of 12 mm. Firstly, the PVA in the blanks was eliminated at 600°C for 2 h, followed by sintering at 1180°C for 2 h. The blanks were polished to 0.3 mm to test the optical properties, phase structure, and surface micro-morphology of the samples.

The optical transmittance of the sample was measured in the range of 200–1100 nm using an ultraviolet–visible–infrared (UV–Vis–IR) spectrophotometer (PerkinElmer Lambda 950, Waltham, MA, USA), and the optical bandgap of the sample was calculated. The phase composition of the sintered samples was identifed by x-ray difraction (XRD, D8 Advance, Bruker) with difraction angles ranging from 20° to 80°. The microstructures of the sample surface were characterized using feld emission scanning electron microscopy (FE-SEM, Quanta 450 FEG, FEI), and the average grain size of the sample was calculated. Silver paste was fired on both sides of the samples at 600°C for 1 h as the electrodes for the dielectric and piezoelectric measurements. The samples were placed in a silicon oil bath for polarization by applying 30–200 kV to test for their electrical hysteresis lines by a ferroelectric integrated system (P-PMF, Radiant Technologies) at room temperature.

## **Results and Discussion**

The transmission spectra of the KNN-*x*SBT ceramic samples tested in the range of 200–1100 nm and the physical image of the ceramic samples are shown in Fig. [1](#page-2-0). In Fig. [1](#page-2-0)a, all KNN-*x*SBT samples have a certain light transmittance. For the KNN-*x*SBT samples with  $x = 0.022$  and  $x = 0.024$ , it can be seen that the letters below the ceramic are very clear, indicating that these two samples have higher transmittance than other samples, which is also confrmed by test results for light transmittance shown in Fig. [1](#page-2-0)b and c. Here, it can be seen that the transmittance of the KNN-*x*SBT ceramic samples frst increases and then decreases with the increase in the second component SBT. When  $x = 0.022$ , the transmittance of the KNN-*x*SBT ceramics reaches the highest transmittance of 69.36% in the visible light band (780 nm) and 72.2% in the near-infrared band. At the same temperature, the transparent ceramics with the same composition



<span id="page-2-0"></span>**Fig. 1** (a) Photograph of the KNN-*x*SBT transparent ceramics. (b) and (c) The variation curves of transmittance (%) with wavelength (nm) in the range of 200–1100 nm and content (*x*), respectively.

are subjected to other treatments and then applied pressure is the most efective practice to improve transmittance presently, the transmittance of the ceramics at 1100 nm reached the highest 78.5%, while the transmittance of the ceramic samples without pressure is 70.2%, which is a signifcant difference.<sup>[26](#page-6-14)</sup>

Figure [2a](#page-2-1) shows the difraction patterns of the KNN-SBT in the range of 20°–80°. Compared with the KNN standard card (PDF#77-1133), with the increase in the content of SBT, the difraction peaks show obvious changes. From  $x=0.018$  to  $x=0.028$ , the XRD pattern of the ceramic samples shows no impurity peaks, and the intensity changes in the difraction peaks are weak, indicating that the ceramic



<span id="page-2-1"></span>**Fig. 2** (a) XRD pattern of KNN-*x*SBT ceramics; (b) magnifed XRD pattern in the 2*θ* range at 42°–48°.

samples have the standard perovskite structure and that the second component SBT is completely integrated into the KNN unit cell to form a homogeneous solid solution without generating a second phase. To further analyze the efect of SBT on the phase structure, the (200) difraction peak of XRD in the range of 42°–48° is magnifed. Figure [2](#page-2-1)b shows that there is no splitting of the difraction peaks, which indicates that the introduction of the second component SBT has successfully regulated the phase structure of KNN ceramics, resulting in a phase structure transformation from tetragonal to pseudo-cubic phase. With the increase in SBT, the difraction peak angle shifts from high to low, which is because of the substitution between ions with diferent radii and the outermost electrons in the system. When the Bi<sup>3+</sup> ( $R = 1.38$  Å, CN=12) and Ta<sup>5+</sup> ( $R = 0.64$  Å,  $CN=6$ ) replace Nb<sup>5+</sup> (0.64 Å, CN=6) of the B-position in the KNN unit cell, oxygen vacancies will be generated in the unit cell to achieve valence balance, and as the amounts of Bi<sup>3+</sup> (*R* = 1.38 Å, CN = 12) and Ta<sup>5+</sup> (*R* = 0.64 Å, CN = 6) increase, more oxygen vacancies will be generated in the unit cell. Eventually, the unit cell structure collapses, which causes lattice distortion, thus leading to smaller cell volume and a shift of the (200) difraction peak to a higher angle. In addition, the ionic radius of  $Sr^{2+}$  ( $R = 1.44$  Å,  $CN = 12$ ) is smaller than that of  $K^+$  ( $R = 1.64$  Å,  $CN = 12$ ) and larger than that of Na<sup>+</sup> ( $R = 1.39$  Å, CN = 12). Therefore, the ionic radii of  $Sr^{2+}$  and Na<sup>+</sup> are closer, so it is easier for  $Sr^{2+}$  to replace  $Na<sup>+</sup>$ , resulting in an increase in the unit cell volume.<sup>27-[29](#page-6-16)</sup>

The unit cell parameters were calculated based on the XRD data using JADE 6.0 software and are listed in Table [I.](#page-3-0) With the increase in SBT doping content, the a, b, and c of all ceramic samples are very close, and the value of c/a is close to 1, which indicates that the crystal structure of all ceramic samples is in a pseudo-cubic phase structure. Abnormal difraction peaks appear at 0.02≤*x*≤0.024 and

shift to higher angles, which may also be caused by changes in the phase structure. Due to the optical isotropy of the highly symmetric cubic phase, only one refractive index  $n_0$  is given for the incident light that passes through the ceramic sample, and no new scattering center is generated at the grain boundary, thereby reducing the scattering of the incident light, which can signifcantly improve the optical transmittance of the ceramic sample. For this reason, KNN $x$ SBT ceramics have excellent light transmittance.<sup>[27](#page-6-15)[,28](#page-6-17)</sup>

The addition of the second component SBT signifcantly inhibits the growth of KNN-based ceramic grains during the ceramic sintering process, so that the grain size of KNN*x*SBT ceramics reaches the nanoscale, which efectively improves its transmittance to incident light. The microstructure of the KNN-*x*SBT ceramic sample at ×30,000 magnifcation is shown in Fig. [3.](#page-3-1) It can be seen that the KNN*x*SBT ceramics have fne grains and a dense microstructure. In Fig. [3](#page-3-1)a and e, when  $x=0.018$  and  $x=0.026$ , the surface grains of the ceramic samples are slightly melted, the grain

<span id="page-3-0"></span>**Table I** The lattice parameters of KNN-*x*SBT ceramics

$\boldsymbol{x}$	a(A)	b(A)	c(A)	$V(A^3)$	c/a
0.018	3.9689	3.9826	3.9922	63.10	1.0059
0.02	3.9830	3.9764	3.9741	62.94	0.9978
0.022	3.9674	3.9825	3.9840	62.95	1.0004
0.024	3.9808	3.9822	3.9787	63.07	0.9995
0.026	3.9870	3.9828	3.9728	63.09	0.9964
0.028	3.9818	3.9818	3.9832	63.15	1.0042

boundaries are blurred, and the grains are irregular in shape, which is probably due to the high sintering temperature. The high light transmittance of KNN-*x*SBT ceramics is derived from the symmetrical pseudo-cubic phase structure and the dense microstructure of the KNN-*x*SBT ceramics, which are confrmed by the XRD patterns in Fig. [2](#page-2-1) and by the SEM morphology in Fig. [3.](#page-3-1) However, when comparing the properties of these two series of ceramics with conventional sintering (CS) and HP sintering, all HP specimens are without obvious pores and have much higher density than the values for CS specimens obtained by SEM as reported in the literature,  $30$  which is also evidence that HP samples have better light transmittance. $31,32$  $31,32$ 

Figure [4](#page-4-0) shows the *P–E* hysteresis loops of the KNN*x*SBT ceramics at room temperature under an applied electric feld of 80 kV. It can be seen that all the KNN-*x*SBT ceramics have saturated hysteresis loops, and the hysteresis loops become thinner with the increase in the content of SBT. The temperature-dependent dielectric constant (*ε*<sup>r</sup> ) and loss (tan*δ*) of the KNN-*x*SBT ceramics at 1 kHz, 10 kHz, and 100 kHz test frequencies are shown in Fig. [5,](#page-4-1) where the double peaks merge into a broad peak with increasing SBT from low to high temperature in the ceramics, which is considered as evidence for the transition from ferroelectrics to relaxor ferroelectrics.<sup>28</sup> This indicates that the relaxivity of the KNN*x*SBT ceramic samples is enhanced, and the introduction of the second component SBT enhanced the symmetry of the phase structure of the KNN-*x*SBT, resulting in a transformation of the phase structure into the pseudo-cubic phase. However, the pseudo-cubic phase structure with higher symmetry makes



<span id="page-3-1"></span>**Fig. 3** (a–f) Surface micromorphology and grain size distribution of KNN-*x*SBT ceramics (*x*=0.018, 0.02, 0.022, 0.024, 0.026, 0.028).



<span id="page-4-0"></span>**Fig. 4** KNN-*x*SBT ceramics under 80 kV/cm electric feld: (a) *P–E* hysteresis loops and (b) variation trends of  $P_m$ ,  $P_r$  with content (*x*).

polarization difficult, and the ferroelectric properties weaken with the increase in  $SBT<sup>33</sup>$ . The dielectric constants of the HP specimens are slightly higher than those of the CS samples, the shape of the hysteresis loops for the HP specimens is closer to a square loop than for CS specimens, and the HP specimens more easily reach saturation polarization than the  $CS$  specimens.<sup>30</sup> With the increase in the second component, SBT,  $P_{\text{max}}$  decreases from 24.08  $\mu$ C/cm<sup>2</sup> when  $x = 0.018$  to 7.29  $\mu$ C/cm<sup>2</sup> when  $x=0.028$ , and  $P_r$  decreases from 10.42  $μC/cm<sup>2</sup>$  at *x* = 0.018 to 1.48  $μC/cm<sup>2</sup>$  at *x* = 0.028, as shown in Fig. [4b](#page-4-0).  $E_c$  decreases with increasing SBT, from 16.8 kV/ cm when  $x = 0.018$  to 14.52 kV/cm when  $x = 0.028$ . The reason for the gradual weakening of the ferroelectric properties of the KNN-*x*SBT ceramic samples is closely related to the change in the phase structure. By comparison with values from the corresponding references, for the same ceramic samples, high  $P_r$  and low  $E_c$  values are obtained by hot pressing relative to pressureless sintering.[30](#page-6-18) The energy storage density *W* of ferroelectric ceramics can be estimated according to its *P–E* hysteresis loop using Eq. [1](#page-4-2):

<span id="page-4-2"></span>
$$
W = \int_{0}^{P_{\text{max}}} E \, \mathrm{d}P \tag{1}
$$

where *E* is the electric feld and *P* is the polarization. The recoverable energy storage density  $W_{\text{rec}}$  can be calculated according to the following Eq. [2](#page-5-8):



<span id="page-4-1"></span>**Fig. 5** Temperature-dependent dielectric constant  $(e_r)$  and loss (tan $\delta$ ) of the KNN-*xSBT* ceramics.



<span id="page-5-10"></span>**Fig.** 6 Variation trends of  $W_{\text{rec}}$  and  $\eta$  with content (*x*).

$$
W_{rec} = \int_{P_r}^{P_{\text{max}}} E \, \mathrm{d}P \tag{2}
$$

Energy storage efficiency  $\eta$  is also an important parameter for measuring the energy storage performance of materials, and energy storage efficiency  $\eta$  can be calculated by Eq. [3:](#page-5-9)

$$
\eta = \frac{W_{\text{rec}}}{W} = \frac{W_{\text{rec}}}{W_{\text{rec}} + W_{\text{loss}}} \times 100\%
$$
\n(3)

According to the above equations, using the *P–E* hysteresis loop of the KNN-SBT ceramic samples, the *W* and *W*rec of the KNN-SBT ceramic samples are calculated under the critical breakdown strength. The energy storage performance of ceramic samples with diferent SBT doping content was studied.<sup>[27](#page-6-15)</sup>

According to the *P–E* hysteresis loop of KNN-*x*SBT ceramics, its recoverable  $W_{\text{rec}}$  and energy storage efficiency *η* were calculated as shown in Fig. [6](#page-5-10). The recoverable  $W_{\text{rec}}$  of KNN- $x$ SBT is up to 0.57 J/cm<sup>3</sup> when the second component  $x = 0.018$ , and then decreases as *x* increases.  $W_{\text{rec}}$  decreases to 0.24 J/cm<sup>3</sup> with  $x=0.028$ , and the  $\eta$  of KNN-SBT ceramics reaches the highest value of 66.8% when *x*=0.026. The best overall energy storage performance of the KNN-*x*SBT ceramics is achieved with values of  $W_{\text{rec}} = 0.53 \text{ J/cm}^3$  and *η*=50.09% when *x*=0.022.

### **Conclusions**

KNN-*x*SBT transparent ferroelectric ceramics can be successfully prepared using the solid-phase method. The introduction of a second component, SBT, signifcantly affects the phase structure and microstructure of KNN

ceramics, thereby affecting their light transmittance, ferroelectric relaxation behavior, and energy storage characteristics. When  $x=0.022$ , the maximum transmittance of 69.36% and 72.2% is reached in the bands at 780 nm and 1100 nm, respectively. It can be seen that the *P–E* loop changes from the typical *P–E* loops of ferroelectrics to slim loops, and the curves in the dielectric thermogram merge from double peaks into one broad peak with the increase in the SBT content, which illustrates the transition to relaxor ferroelectrics. As a result, the ferroelectric properties are

decreased and the energy storage performance is improved. When  $x = 0.022$ , the KNN-SBT ceramics have the best comprehensive energy storage performance:  $P_{\text{max}} = 15.7$  $\mu$ C/cm<sup>2</sup>,  $P_r = 2.69 \,\mu$ C/cm<sup>2</sup>,  $E_c = 12.24 \,\text{kV/cm}$ ,  $W_{\text{rec}} = 0.53 \,\text{J/m}$ cm<sup>3</sup>,  $\eta$  = 50.09%. The  $W_{\text{rec}}$  decreases with the increase in *x*, and the energy storage efficiency of all the KNN-*x*SBT ceramics is greater than 45%. The results show that the  $0.978(K_{0.5}Na_{0.5})NbO_3-0.022Sr(Bi_{0.5}Ta_{0.5})O_3$  is a potential ferroelectric multifunctional material with excellent transmittance and good energy storage properties.

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<span id="page-5-9"></span>**Conflict of interest** The authors declare no competing fnancial interests.

### **References**

- <span id="page-5-0"></span>1. S.F. Wang, J. Zhang, D.W. Luo, F. Gu, D.Y. Tang, Z.L. Dong, G.E.B. Tan, W.X. Que, T.S. Zhang, S. Li, and L.B. Kong, Transparent ceramics: processing, materials and applications. *Prog. Solid State Chem.* 41, 20 (2013).
- <span id="page-5-1"></span>2. H.T. Wu, G.B. Hu, S.Y. Shi, X. Liu, H. Wang, J.W. Xu, L. Yang, W. Qiu, and S.J. Zhou, Effect of Ho addition on the optical and electrical properties of 0.98KNN-0.02SYT ceramics. *J. Electron. Mater.* 51, 831 (2022).
- <span id="page-5-2"></span>3. Z.H. Xiao, S.J. Yu, Y.M. Li, S.C. Ruan, L.B. Kong, Q. Huang, Z.R. Huang, K. Zhou, H.B. Su, Z.J. Yao, W.X. Que, Y. Liu, T.S. Zhang, J. Wang, P. Liu, D.Y. Shen, A. Mathieu, J. Zhang, and D.Y. Tang, Materials development and potential applications of transparent ceramics: a review. *Mater. Sci. Eng. R Rep.* 139, 100518 (2020).
- <span id="page-5-3"></span>4. X.Q. Chen and Y.Q. Wu, Fabrication and optical properties of highly transparent MgO ceramics by spark plasma sintering. *Scr. Mater.* 162, 14 (2019).
- <span id="page-5-4"></span>5. P.K. Panda, Review: environmental friendly lead-free piezoelectric materials. *J. Mater. Sci.* 44, 5049 (2009).
- <span id="page-5-5"></span>6. G.H. Haertling, Ferroelectric ceramics: history and technology. *J. Am. Ceram. Soc.* 82, 797 (1999).
- <span id="page-5-6"></span>7. S.F. Wang, Q.B. Liu, E.P. Cai, F.H. Mou, and A. Xue, Relaxor behavior and superior ferroelectricity of  $Y_2O_3$ -doped  $(Ba_{0.98}Ca_{0.02})(Ti_{0.94}Sn_{0.04}Zr_{0.02})O_3$  lead-free ceramics. *J. Rare Earths.* 40, 942 (2022).
- <span id="page-5-7"></span>8. Y.B. Sun, H. Wang, G.B. Liu, H. Xie, C.R. Zhou, G.H. Chen, C.L. Yuan, and J.W. Xu, High energy storage efficiency and high

electrostrictive coefficients in BNT-BS-*x*BT ferroelectric ceramics. *J. Mater. Sci. Mater. Electron.* 31, 5546 (2020).

- <span id="page-6-0"></span>9. X.Z. Wang, Y. Huan, Z.X. Wang, X.J. Lin, S.F. Huang, T. Wei, L.T. Li, and X.H. Wang, Electrical conduction and dielectric relaxation mechanisms in the KNN-based ceramics. *J. Appl. Phys.* 126, 104101 (2019).
- <span id="page-6-1"></span>10. B. Chen, P.F. Liang, D. Wu, X.M. Zhao, X.S. Qiao, Z.H. Peng, L.L. Wei, X.L. Chao, and Z.P. Yang, High-efficiency synthesis of high-performance K<sub>0.5</sub>Na<sub>0.5</sub>NbO<sub>3</sub> ceramics. *Powder Technol*. 346, 248 (2019).
- 11. X.M. Zhao, X.L. Chao, D. Wu, P.F. Liang, and Z.P. Yang, Simultaneous realization of high transparency and piezoelectricity in low symmetry KNN-based ceramics. *J. Am. Ceram. Soc.* 102, 3498 (2019).
- <span id="page-6-2"></span>12. S.T. Li, Y. Yue, X.J. Ning, M. Guo, and M. Zhang, Hydrothermal synthesis and characterization of  $(1-x)K_0$ ,  $Na_0$ ,  $NbO_3$  $x\text{Bi}_0$ <sub>5</sub>Na<sub>0.5</sub>TiO<sub>3</sub> lead-free ceramics. *J. Alloys Compd.* 586, 248 (2014).
- <span id="page-6-3"></span>13. C. Lin, H.J. Wang, J.Z. Ma, B.Y. Deng, W. Xiao, T.F. Lin, X.H. Zheng, and Y. Xing, Efect of dwell time on cold sintering assisted sintering based highly transparent  $0.9K_{0.5}Na_{0.5}NbO_3-0.1LiBiO_3$ ceramics. *J. Alloys Compd.* 826, 154249 (2020).
- <span id="page-6-4"></span>14. Z.Y. Cen, X.H. Wang, Y. Huan, Y.C. Zhen, W. Feng, and L.T. Li, Defect engineering on phase structure and temperature stability of KNN-based ceramics sintered in diferent atmospheres. *J. Am. Ceram. Soc.* 101, 3032 (2018).
- <span id="page-6-5"></span>15. B.Y. Qu, H.L. Du, Z.T. Yang, and Q.H. Liu, Large recoverable energy storage density and low sintering temperature in potassium-sodium niobate-based ceramics for multilayer pulsed power capacitors. *J. Am. Ceram. Soc.* 100, 1517 (2017).
- 16. G.B. Hu, H.N. Liu, J.T. Wang, Y.B. Sun, H. Wang, J.W. Xu, and L. Yang, Regulating the structural, transmittance, ferroelectric, and energy storage properties of  $K_{0.5}Na_{0.5}NbO_3$  ceramics using Sr(Yb<sub>0.5</sub>Nb<sub>0.5</sub>)O<sub>3</sub>. *J. Electron. Mater.* 50, 968 (2021).
- <span id="page-6-6"></span>17. G.B. Hu, J.T. Wang, X. Liu, H.N. Liu, H. Wang, J.W. Xu, L. Yang, C.R. Zhou, and W. Qiu, Structural, transmittance, ferroelectric, energy storage, and electrical properties of  $K_0$ ,  $Na_0$ ,  $NbO_3$  ceramics regulated by Sr(Yb<sub>0.5</sub>Ta<sub>0.5</sub>)O<sub>3</sub>. *J Mater. Sci. Mater. Electron.* 32, 22300 (2021).
- <span id="page-6-7"></span>18. J. Jumpatam, B. Putasaeng, N. Chanlek, and P. Thongbai, Infuences of  $Sr^{2+}$  doping on microstructure, giant dielectric behavior, and non-ohmic properties of  $CaCu<sub>3</sub>Ti<sub>4</sub>O<sub>12</sub>/CaTiO<sub>3</sub>$  ceramic composites. *Molecules* 26, 1994 (2021).
- <span id="page-6-8"></span>19. J. Jumpatam, B. Putasaeng, N. Chanlek, J. Boonlakhorn, P. Thongbaid, N. Phromviyo, and P. Chindaprasirt, Signifcantly improving the giant dielectric properties of  $CaCu<sub>3</sub>Ti<sub>4</sub>O<sub>12</sub>$  ceramics by co-doping with Sr<sup>2+</sup> and F-ions. *Mater. Res. Bull.* 133, 111043 (2021).
- <span id="page-6-9"></span>20. Y.R. Wang, Y.P. Pu, Y.F. Cui, Y. Shi, and H.Y. Zheng, Enhanced energy storage density of  $Ba<sub>0.4</sub>Sr<sub>0.6</sub>TiO<sub>3</sub>$  ceramics with additive of Bi<sub>2</sub>O<sub>3</sub>-B<sub>2</sub>O<sub>3</sub>-ZnO glass. *Mater Lett.* 201, 203 (2017).
- 21. J.Q. Li, J.J. Wang, F.M. Wu, H. Ma, T.Y. Ma, Y. Tian, D.Q. Liu, and B. Yang, Microstructure and electric properties of  $Bi<sub>2</sub>O<sub>3</sub>$ -doped  $(K<sub>0.5</sub>Na<sub>0.5</sub>)NbO<sub>3</sub>$  lead-free ceramics. *Coatings*. 12, 526 (2022).
- <span id="page-6-10"></span>22. X.X. Wang, X.G. Tang, K.W. Kwok, H.L.W. Chan, and C.L. Choy, Effect of excess  $Bi<sub>2</sub>O<sub>3</sub>$  on the electrical properties and microstructure of  $(Bi_{1/2}Na_{1/2})TiO_3$  ceramics. *Appl. Phys. A: Mater. Sci. Process.* 80, 1071 (2005).
- <span id="page-6-11"></span>23. H.R. Liu, Q. Li, J. Ma, and X.C. Chu, Effects of Bi<sup>3+</sup> content and grain size on electrical properties of SrBi<sub>2</sub>Ta<sub>2</sub>O<sub>9</sub> ceramic. Mater. *Lett.* 76, 21 (2012).
- <span id="page-6-12"></span>24. B.B. Liu, X.H. Wang, R.X. Zhang, and L.T. Li, Grain size efect and microstructure infuence on the energy storage properties of fine-grained BaTiO<sub>2</sub>-based ceramics. *J. Am. Ceram. Soc.* 100, 3599 (2017).
- <span id="page-6-13"></span>25. K. Itatani, T. Tsujimoto, and A. Kishimoto, Thermal and optical properties of transparent magnesium oxide ceramics fabricated by post hot-isostatic pressing. *J. Eur. Ceram. Soc.* 26, 639 (2006).
- <span id="page-6-14"></span>26. L. Esposito, A. Piancastelli, and S. Martelli, Production and characterization of transparent  $MgAl<sub>2</sub>O<sub>4</sub>$  prepared by hot pressing. *J. Eur. Ceram. Soc.* 33, 737 (2013).
- <span id="page-6-15"></span>27. H.N. Liu, J.T. Wang, H. Wang, J.W. Xu, C.R. Zhou, and W. Qiu,  $\mathrm{Er}^{3+}$  and  $\mathrm{Sr(Bi_{0.5}Nb_{0.5})O_3}$ -modified (K<sub>0.5</sub>Na<sub>0.5</sub>)NbO<sub>3</sub>: a new transparent fuorescent ferroelectric ceramic with high light transmittance and good luminescence performance. *Ceram Int.* 48, 4230 (2022).
- <span id="page-6-17"></span>28. J. Zhang, J.W. Xu, L. Yang, Z.J. Cao, C.L. Yuan, C.R. Zhou, H. Wang, and G.H. Rao, Controlling light-induced dielectric response of Sr/Ni-modified  $(K_{0.5}Na_{0.5})NbO<sub>3</sub>$  ceramics by narrow bandgap method. *Mater. Sci. Semicond Process.* 143, 106521 (2022).
- <span id="page-6-16"></span>29. H.T. Wu, S.Y. Shi, X. Liu, H. Wang, J.W. Xu, L. Yang, W. Qiu, and S.J. Zhou, The Ba( $Bi_{0.5}Ta_{0.5}O_3$  modified  $(K_{0.5}Na_{0.5})NbO_3$ lead-free transparent ferroelectric ceramics with high transmittance and excellent energy storage performance. *J. Mater. Sci. Mater. Electron.* 33, 16045 (2022).
- <span id="page-6-18"></span>30. G.C. Deng, A.L. Ding, X.S. Zheng, X. Zeng, and Q.R. Yin, Property improvement of  $0.3Pb(Zn_{1/3}Nb_{2/3})O_3$ - $0.7Pb_{0.96}La_{0.04}(Zr_xTi_{1-x})_{0.99}O_3$  ceramics by hot-pressing. *J. Eur. Ceram. Soc.* 26, 2349 (2006).
- <span id="page-6-19"></span>31. J.F. Lin, Y. Zhou, Q.L. Lu, X. Wu, C. Lin, T.F. Lin, K.H. Xue, X.S. Miao, B.S. Sa, and Z.M. Sun, Reversible modulation of photoenergy in Sm-doped  $(K_{0.5}Na_{0.5})NbO<sub>3</sub>$  transparent ceramics via photochromic behavior. *J. Mater. Chem. A.* 7, 19374 (2019).
- <span id="page-6-20"></span>32. X. Wu, S.B. Lu, and K.W. Kwok, Photoluminescence, electrooptic response and piezoelectric properties in pressurelesssintered Er-doped KNN-based transparent ceramics. *J. Alloys Compd.* 695, 3573 (2017).
- <span id="page-6-21"></span>33. G.W. Yan, and Q.Q. Qiu, B.J. Fang, Z.H. Chen Correlation between phase structure and polarization of Mg doped (Ba<sub>0.98</sub>Li<sub>0.02</sub>)TiO<sub>3</sub> energy storage ceramics. *J. Mater. Sci: Mater. Electron.* 33, 20981 (2022).

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