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# A new strategy to realize high energy storage properties and ultrafast discharge speed in Sr<sub>0.7</sub>Bi<sub>0.2</sub>TiO<sub>3</sub>-based relaxor ferroelectric ceramic



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### ABSTRACT

Although tremendous studies have been focused on dielectric ceramics to achieve excellent energy storage and charge-discharge performance, the dielectric ceramics with high comprehensive energy storage properties for pulsed power applications are still in shortage. The large hysteresis came from domain switching process, severely weaken the energy storage and charge-discharge properties. Here, a strategy through ergodic relaxors with high dynamic polar nanoregions (PNRs) featuring with fast discharge rate and high energy storage efficiency was proposed to achieve high energy storage properties and extremely fast discharge speed in ferroelectric ceramic. The ergodic relaxors of  $Sr_{0.7}Bi_{0.2}TiO_3$  modified by  $Na_{0.73}Bi_{0.09}NbO_3$  (SBT-NBN) ceramics were selected to verify the feasibility of the strategy. Encouragingly, large  $W_{rec}$  of 2.49 J/cm<sup>3</sup> together with high  $\eta$  of 89.7% can be achieved simultaneously at 250 kV/cm with the composition of 0.96SBT-0.04NBN. The outstanding stability of energy storage characteristics for temperature (25–125 °C) and frequency (1–100 Hz) was also found in 0.96SBT-0.04NBN ceramics. Besides, 0.96SBT-0.04NBN ceramics also possessed an extremely fast discharge rate (~34 ns), remarkable power density  $P_D$  (57.3 MW/cm<sup>3</sup>) as well as high current density  $C_D$  (955.4 A/cm<sup>2</sup>) at 120 kV/cm. This work provided an original strategy for high energy storage efficiency and high power density ceramics in pulse power applications.

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# 1. Introduction

Dielectric ceramic capacitors are widely utilized in pulsed components and energy storage devices due to their apparent benefits of ultrafast charge/discharge capability, high power density ( $P_D$ ), and excellent thermal stability, showing huge advantages in weapons, vehicles and power electronics [1–6]. Yet, the relatively low energy storage density and efficiency of dielectric capacitors limit their further applications [2]. Consequently, exploring novel energy storage materials with high energy storage properties has become an urgent issue [4,7]. Generally, the energy storage properties can be calculated by employing polarization-electric field (P-E) loops, expressed by [7]:

Energy storage densityW = 
$$\int_0^{P_{max}} EdP$$
 (1)

Recoverable energy storage density 
$$W_{rec} = \int_{P_r}^{P_{max}} EdP$$
 (2)

Energy storage efficiency 
$$\eta = \frac{W_{rec}}{W} * 100\%$$
 (3)

where P,  $P_{max}$ ,  $P_r$  and E denote the polarization, maximum polarization, remnant polarization and applied electric field, respectively. Accordingly, the crucial factors to improve energy storage performance of dielectric capacitors are low  $P_r$ , large  $P_{max}$ , as well as high breakdown strength (E<sub>B</sub>).

Relaxor ferroelctrics have been extensively investigated for energy storage applications owing to the small  $P_r$ , and a relative large  $P_{max}$  can be induced under a large electric field [8,9].  $Sr_{0.7}Bi_{0.2}TiO_3$  (SBT), a typical lead-free ergodic relaxor ferroelectric with the perovskite structure, possesses excellent ferroelectric relaxor behavior together with diffused maximum dielectric constant in a wide temperature range, benefiting from  $Bi^{3+}$  off centering and Sr site vacancies [10,11]. Furthermore, a high dielectric permittivity can be

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obtained in SBT ceramic, which is attributed to dipole polarization related to dipole fluctuation of polar nanoregions (PNRs) [10–12]. However, in the past decades, studies focused on the energy storage performance of (Sr, Bi)TiO<sub>3</sub>-based ceramics were rarely reported [10,12,13]. Zhao et al. [10] reported a Sr<sub>0.7</sub>Bi<sub>0.2</sub>TiO<sub>3</sub>-based relaxor ceramics with high energy storage density of 2.1 J/cm<sup>3</sup> and high  $\eta$  of 97.6% at 290 kV/cm. Zhang et al. [12] realized Sr<sub>(1-1.5x)</sub>Bi<sub>x</sub>TiO<sub>3</sub> ceramics with energy storage density of 1.63  $I/cm^3$  and n of ~61.4% at 217.6 kV/cm. Moreover, (Sr, Bi)TiO<sub>3</sub>-based ergodic relaxor ferroelectric ceramics are also an potential candidate for pulsed chargedischarge applications. The ergodic relaxor ferroelectrics with PNRs are converted into unstable ferroelctrics with long-range polar structure under external electric field, and the long-range polar structure will guickly collapse after withdrawing the electric field [5], which lead to a fast discharge rate. Thus a large  $P_D$  can be realized due to the stored energy can be released in a short time. Zhao et al. [10] reported fast discharge rate ( $\tau_{0,9}$ ) of 124 ns and high power density of 50.1 MW/cm<sup>3</sup> in Sr<sub>0.7</sub>Bi<sub>0.2</sub>Ca<sub>0.1</sub>TiO<sub>3</sub> ceramics. Despite the great achievements made so far, it is necessary to acquire bulk ceramics with good energy storage and charge-discharge performance [14,15].

Furthermore, Na<sub>0.73</sub>Bi<sub>0.09</sub>NbO<sub>3</sub> was reported to possess good dielectric temperature stability [16–18], as well as decreases P<sub>r</sub> and improve E<sub>B</sub> [19]. Thus good energy storage performance may be obtained with Na<sub>0.73</sub>Bi<sub>0.09</sub>NbO<sub>3</sub> as dopants. Wang et al.[19] reported energy density of 1.56 J/cm<sup>3</sup> and high efficiency of 92.5% in (1-*x*) (0.94Bi<sub>0.5</sub>Na<sub>0.5</sub>TiO<sub>3</sub>-0.06BaTiO<sub>3</sub>)-*x*Na<sub>0.73</sub>Bi<sub>0.09</sub>NbO<sub>3</sub> ceramics. Hu et al. [16] fabricated Bi<sub>0.5</sub>Na<sub>0.5</sub>TiO<sub>3</sub>-Na<sub>0.73</sub>Bi<sub>0.09</sub>NbO<sub>3</sub> ceramics with excellent energy storage density (~2.41 J/cm<sup>3</sup>) and efficiency (~81.6%) successfully.

Here, the energy storage and charge-discharge properties of (1-x)Sr<sub>0.7</sub>Bi<sub>0.2</sub>TiO<sub>3</sub>-xNa<sub>0.73</sub>Bi<sub>0.09</sub>NbO<sub>3</sub> ((1-x)SBT-xNBN) ceramics were

investigated systematically. The design strategy of this work was shown in Fig. 1. The high dynamic and weak correlation PNRs were supposed to generate by SBT ergodic relaxor ferroelectric ceramics, contributing to the fast discharge rate. The modulation of NBN could effectively reduce the size of PNRs, thus reducing  $P_r$  and improving  $E_B$ . Finally, a large  $W_{rec}$  of 2.49 J/cm<sup>3</sup> and a high  $\eta$  of 89.7% were achieved simultaneously in 0.96SBT-0.04NBN ceramics, together with an extremely fast discharge rate of 34 ns and a remarkable  $P_D$  of 57.3 MW/cm<sup>3</sup>. Besides, the  $W_{rec}$  displayed good temperature stability over 25–125 °C, as well as excellent frequency stability from 1 to 100 Hz. All the results indicated that (1-*x*)SBT-*x*NBN ceramics were promising options for energy storage applications.

# 2. Experimental procedures

A series of (1-x)SBT-xNBN (x = 0, 0.02, 0.04, 0.06, 0.08, 0.1) ceramics were synthesized by conventional solid-state reaction method. Reagent-grade SrCO<sub>3</sub> (Xilong scientific, 99.00%), Bi<sub>2</sub>O<sub>3</sub> (Xilong scientific, 99.00%), Na<sub>2</sub>CO<sub>3</sub> (Xilong scientific, 99.98%), TiO<sub>2</sub> (Zhongxing electronic, 99.00%) and Nb<sub>2</sub>O<sub>5</sub> (Sinopharm, 99.50%) powders were weighted according to stoichiometric ratios, and then were mixed with ethanol alcohol and zirconia balls by ball milling for 24 h. After dying, the milled powders were calcined at 860 for 3 h in air. The formation reaction was listed as follow:

$$0.7SrCO_3 + TiO_2 + 0.1Bi_2O_3 \to Sr_{0.7}Bi_{0.2}TiO_3 + 0.7CO_2\uparrow$$
(4)

$$0.73Na_2CO_3 + Nb_2O_5 + 0.045Bi_2O_3 \rightarrow 2Na_{0.73}Bi_{0.09}NbO_3 + 0.73CO_2\uparrow$$
(5)

Then the calcined powders were granulated with 8 wt% of polyvinyl alcohol (PVA) as binders. The granulated particles were pressed



Fig. 1. Schematic diagram of the strategy to obtain high energy storage properties and extremely fast discharge speed.



**Fig. 2.** Room-temperature XRD patterns of (1-*x*)SBT-*x*NBN ceramics with 2θ ranging over (a) 20°C- 80°C (b) 42°C- 48°C. (c) Room-temperature Raman spectrum for (1-*x*) SBT-*x*NBN ceramics.

into disks with a diameter of 12 mm under an axial pressure of 100 MPa. The binders were burned out at 600 °C for 2 h, followed by sintering the samples at 1250 °C for 3 h in air. The pellets were embedded in the corresponding powders during sintering to prevent the loss of volatile bismuth and sodium.

The crystalline phase was detected by X-ray diffraction (XRD, Bruker, D8-2-advance) with the Cu K $\alpha$  radiation. Room-temperature Raman spectra were measured by a Horiba Lab-Ram iHR550 spectrometer. The microstructure and compositions of the samples were observed using a field-emission scanning electron microscopy (SEM) (FEG-450, FEI, USA) with elemental mapping. The dielectric properties were obtained by using an LCR meter (Keysight, 4980 A) with the temperature ranging from – 100–200 °C. The ferroelectric properties were measured using a ferroelectric test system (TF ANAYLZER 2000HS, aixACCT Systems GmbH, Aachen, Germany). The charge-discharge properties were investigated through a capacitor charge-discharge system (CFD-003, Gogo Instrument, China).

# 3. Results and discussion

Fig. 2(a) displays the XRD patterns of (1-x)SBT-xNBN ceramics recorded at room temperature. Clearly, the (1-x)SBT-xNBN ceramics

exhibit a single perovskite structure with no apparent trace of the impurities when x = 0-0.04. It suggests that NBN are dissolved into the matrix lattice of SBT and formed a single solid solution. However, the peaks of second phase (Titanium Oxide  $Ti_6O_{11}$ ) are found when x exceeds 0.04. There is no splitting of (200) and (111) peaks for (1-x)SBT-*x*NBN ceramics. It corresponds to a pseudo-cubic phase of all the samples (Fig. 2(b)). The room-temperature Raman spectroscopy, which can probe a local ionic configuration with short length scale, is carried out to further detect the structure evolution of (1-x)SBTxNBN ceramics [20,21]. Raman spectrum is conducted from 50 to  $1000 \text{ cm}^{-1}$  range, as shown in Fig. 2(c). The peaks from A to G are fitted by the Lorentz function for x = 0 sample. Four main vibrations can be discerned [22]. The vibration of A-site ions including Bi, Na and Sr ions in perovskite is below 200 cm<sup>-1</sup>. The modes between 200 and 400 cm<sup>-1</sup> are related to B-site and oxygen ions. It can be seen that the mode D becomes broader and mode E is gradually separated from D, the two modes shift apart from each other. The vibrations of  $BO_6$  oxygen octahedron is between 450 and 700 cm<sup>-1</sup> [22,23]. The modes become broaden and shift toward to low frequency, representing a change in the structure of perovskite with increasing NBN contents. The superposition for all vibration effects is shown over 700 cm<sup>-1</sup> [22].

Fig. 3(a)-(f) show the typical surface morphology of (1-x)SBT-xNBN ceramics, and a dense microstructure with little pores can be found. The average grain size of the (1-x)SBT-xNBN ceramics increase from  $1.8 \,\mu\text{m}$  to  $3.44 \,\mu\text{m}$ , with the *x* increase from 0 to 0.04. This phenomenon shows that NBN doping in SBT can promote the growth of grains, and can be explained as the inevitable volatilization of bismuth and sodium during the high sintering temperature, which results in  $V'_{Na}$  and  $V'''_{Bi}$ , thus the oxygen vacancies would be generated to sustain the overall electrical neutrality [24,25]. The oxygen vacancies are beneficial to mass transportation during sintering [25]. A notable characteristic of the micrographs with inhomogeneous spherical grains combined with rod-like grains is observed in the grain boundaries, with x exceeding 0.04, as shown in Fig. 3(d)-(f). The energy dispersive spectrometer (EDS) analysis of 0.9SBT-0.1NBN ceramic shown in Fig. 3 reveals that the rod-like inclusion has higher signal intensities in Ti and O, indicating the secondary phase is a Ti-enriched phase, which is consistent with the XRD results.

Fig. 4 displays the room-temperature frequency dependence of the dielectric permittivity ( $\varepsilon_r$ ) and dielectric loss (tan $\delta$ ) of (1-*x*)SBT-*x*NBN ceramics, indicating excellent frequency stability of dielectric properties in the frequency range from 100 Hz to 1 MHz. The dielectric constants of samples increase firstly and experience a decline process with the increasing contents of NBN. Furthermore, the dielectric loss is always kept at a low value (<0.01), which benefits to energy storage properties.

Fig. 5(a)-(f) present the temperature dependence of  $\varepsilon_r$  and tan $\delta$  for (1-*x*)SBT-*x*NBN ceramics. The maximum dielectric permittivity  $\varepsilon_m$  temperature (T<sub>m</sub>) for (1-*x*)SBT-*x*NBN ceramics are lower than room temperature (T<sub>room</sub>), demonstrating an ergodic relaxor behavior for all samples. Besides, a typical relaxor behavior is observed in (1-*x*)SBT-*x*NBN ceramics and proved by the broadening and frequency dispersion of dielectric peaks [11]. The modified Curie-Weiss law is conducted to confirm the relaxor behavior of (1-*x*)SBT-*x*NBN ceramics, and the equation is given below:



Fig. 3. The SEM images of fresh surface for (1-x)SBT-xNBN ceramics, accompanied with EDS mapping for 0.9SBT-0.1NBN ceramics.



$$\frac{1}{\varepsilon} - \frac{1}{\varepsilon_m} = \frac{(T - T_m)^{\gamma}}{C}, \quad T > T_m$$
(6)

where  $\varepsilon_m$  is the maximum value of dielectric constant,  $T_m$  is the temperature for  $\varepsilon_m$ , C is the Curie-Weiss constant and  $\gamma$  reveals the degree of relaxor. The  $\gamma$  value range between 1 (typical ferroelectric) and 2 (complete relaxor ferroelectric) [2]. It can be seen in Fig. 6(a)-(f), the linear relationship of  $\ln(1/\epsilon - 1/\epsilon_m)$  and  $\ln(T-T_m)$  can be found in all the contents, accompanied with  $\gamma$  ranging between 1.77 and 1.48, demonstrating a powerful relaxor trait. Nevertheless, sometimes the  $\gamma$  value may not indicates the relaxor behavior well, for its value strongly depends on extrinsic defects, such as the homogeneity of composition [7]. Therefore, another method is conducted to further confirm the relaxor behavior, and we define a parameter  $\Delta T_{relaxor}$  to quantify the frequency dispersion, which can be defined as follow [7,26].



**Fig. 5.** Temperature-dependent  $\varepsilon_r$  and tan $\delta$  of (1-*x*)SBT-*x*NBN ceramics.

(7)

$$T_{relaxor} = T_{\varepsilon_{m(1 \ MHz)}} - T_{\varepsilon_{m(1 \ kHz)}}$$

together with smaller volume of PNRs, which is favorable for chargedischarge properties.

where  $T\varepsilon_{m(1 \text{ MHz})}$  is the temperature for  $\varepsilon_m$  at 1 MHz and  $T\varepsilon_{m(1 \text{ kHz})}$  is the temperature for  $\varepsilon_m$  at 1 kHz. The  $T_m$ ,  $\gamma$  and calculated  $\Delta T_{relaxor}$ values for (1-x)SBT-xNBN ceramics are integrated into Fig. 7, from which we can find that  $\Delta T_{relaxor}$  almost unchanged when  $x \le 0.04$ , while it decreases with x > 0.04, suggesting a reducing relaxor characteristics. In addition, it can be found that the  $T_m$  of (1-x)SBTxNBN ceramics shift to lower temperature with increasing of NBN contents. Thus the correlation between PNRs became weaker, The energy storage properties of the (1-x)SBT-xNBN ceramics are investigated using P-E loops, which are conducted at 150 kV/cm and 5 Hz (Fig. 8(a)). The P-E loops become more and more pinched with the addition of NBN, which indicates the decreasing of ferroelectricity. Fig. 8(b) displays the current density (I-E) curves for (1-x)SBT-xNBN ceramics. No obvious current peaks can be detected, confirming the ergodicity [5], which is consistent with the results of temperature dependent dielectric constant (Fig. 4). The evolution of



**Fig. 6.** The curves of  $\ln(1/\epsilon - 1\epsilon_m)$  as a function of  $\ln(T-T_m)$  for the (1-x)SBT-xNBN ceramics at 1 MHz.



**Fig. 7.** (a)-(c)  $T_m$ ,  $\gamma$ ,  $\Delta_{Trelaxor}$  values as a function of x for (1-x)SBT-xNBN ceramics, respectively.

P<sub>max</sub>, P<sub>r</sub> and ΔP (P<sub>max</sub> - P<sub>r</sub>) are shown in Fig. 8(c). The P<sub>max</sub> and P<sub>r</sub> decrease with the increase of *x*. Additionally, P<sub>r</sub> decreases sharply when *x* ≤ 0.04, while it almost unchanged when *x* > 0.04. As a result, a high ΔP can be achieved when *x* = 0.04, demonstrating a promising for energy storage properties. Furthermore, it should be noticed that the 0.96SBT-0.04NBN ceramics possess moderate ε<sub>r</sub> of ~ 1360 and low tanδ of ~ 0.002 at room temperature, which are favorable for high E<sub>B</sub>.

Room-temperature unipolar P-E loops measured at various electric fields and 5 Hz are used to illustrate the energy storage properties of 0.96SBT-0.04NBN ceramics, as presented in Fig. 9(a). The 0.96SBT-0.04NBN ceramics show slim P-E loops with high  $P_{max}$  and negligible  $P_r$  under different electric fields, which are benefit to enhance energy storage properties. To qualify the energy storage properties, we summarize the  $W_{rec}$ ,  $W_{tot}$ ,  $W_{loss}$ , as well as  $\eta$  under different electric fields, as shown in Fig. 9(b). It can be seen that  $W_{rec}$  and  $W_{tot}$  are improved obviously with enhancing electric field, while  $W_{loss}$  increases slightly. Furthermore, the  $\eta$  decreases with enhancing electric field due to the large hysteresis of P-E loop at high



**Fig. 8.** (a) P-E loops of (1-x)SBT-xNBN ceramics at 150 kV/cm and 5 Hz. (b) Corresponding P<sub>max</sub>, P<sub>r</sub> and  $\Delta P$  (P<sub>max</sub> - P<sub>r</sub>) values as a function of *x*. (c) I-E curves of (1-x)SBT-xNBN ceramics at 150 kV/cm and 5 Hz.

electric field. It is well accepted that the formation of long-range polar ordering through domain switching can be induced by a high enough electric field [5]. Especially, a combination of excellent energy storage properties for 0.96SBT-0.04NBN ceramics was achieved at 250 kV/cm ( $W_{rec} = 2.49 \text{ J/cm}^3$ ,  $\eta = 89.7\%$ ).

For the practical application in dielectric capacitors, large  $W_{rec}$  and high  $\eta$ , together with their excellent thermal stability and frequency stability are required. Fig. 10(a) displays the temperature-dependent unipolar P-E loops of 0.96SBT-0.04NBN ceramics in the temperature range from 25 °C to 125 °C at 100 kV/cm and 5 Hz. It is



Fig. 9. (a) Unipolar P-E loops of 0.96SBT-0.04NBN ceramics with different electric fields at 5 Hz and ambient temperature. (c) I-E loops of 0.96SBT-0.04NBN ceramics. (b)  $W_{rec}, W_{loss}, W_{tot}$  and  $\eta$  of 0.96SBT-0.04NBN ceramics.

evident that the 0.96SBT-0.04NBN ceramics maintain slim P-E loops over the whole temperature range. When the temperature increases from 25 °C to 125 °C, the  $P_{max}$  diminishes from 11.64  $\mu$ C/cm<sup>2</sup> to 9.57  $\mu$ C/cm<sup>2</sup>, accompanied by negligible P<sub>r</sub> (~0  $\mu$ C/cm<sup>2</sup>). These characteristic are related to the relaxor behavior of 0.96SBT-0.04NBN ceramics, as mentioned above. Fig. 10(b) depicts the  $W_{rec}$  and  $\eta$  as a function of different temperature, which show sound stability in this temperature range (W<sub>rec</sub>: 0.52–0.43 J/cm<sup>3</sup>, η: 92–90%). The excellent temperature stability of the  $W_{rec}$  and  $\eta$  for 0.96SBT-0.04NBN ceramics may attribute to the neighboring PNRs coupling which can only occur under high electric field and low temperature [27]. The unipolar frequency dependence of P-E loops for 0.96SBT-0.04NBN ceramics measured at 100 kV/cm and room temperature are represented in Fig. 10(c), the corresponding energy storage properties are depicted in Fig. 10(d). The slight fluctuations of  $W_{rec}$  and  $\eta$ confirm the frequency-insensitivity characteristic of 0.96SBT-0.04NBN ceramics in this frequency range. For example, the  $W_{rec}$  is  $0.55\,J/cm^3$  at 1 Hz and  $0.53\,J/cm^3$  at 100 Hz. Besides, the  $\eta$  which always more than 85% are achieved, indicating a good frequency stability of energy storage properties.

Fig. 11(a) displays the room-temperature overdamped pulse discharge current waveforms of 0.96SBT-0.04NBN ceramics, in which the current reaches the peak in a short time, and the



Fig. 10. Unipolar P-E loops of 0.96SBT-0.04NBN ceramics at different temperature (a) and different frequencies (c) at 100 kV/cm. (b) and (d) The corresponding calculated W<sub>d</sub> and η vs temperature and frequency, respectively.

discharge process lasts less than 100 ns. According to the curves, the discharge energy density  $(W_{\rm d})$  can be obtained by the following formula:

$$W_d = R \int i(t)^2 dt / V \tag{8}$$

where R is the total load resistor (R =  $120 \Omega$ ), i(t) is the discharge current obtained by using the oscillograph, and V represents the sample volume [28,29]. As displayed in Fig. 11(b), the  $W_d$  increases from 0.20 J/cm<sup>3</sup> at 60 kV/cm to 0.77 J/cm<sup>3</sup> at 120 kV/cm. In addition,  $W_d$  reaches the maximum value in an extremely short time, and  $\tau_{0.9}$ is introduced to confirm the discharge speed, which is defined as the time to achieve 90% of the final value [29]. The remarkable  $\tau_{0.9}$  of 34 ns for 0.96SBT-0.04NBN ceramics demonstrating its high potential for fast pulse power applications. Fig. 11(c) presents the underdamped discharge curves for 0.96SBT-0.04NBN ceramics at various electric fields and room temperature. With the enhancement of electric field, the first current peak increases gradually. Based on the obtained underdamped discharge waveforms, the maximum current  $(I_{max})$ , calculated current density  $(C_D = I_{max}/S)$  and power density  $(P_D$ =  $EI_{max}/2$  S) are plotted in the Fig. 11(d) [5]. It can be found that the  $I_{max}$ ,  $C_D$  and  $P_D$  monotonously increase from 15.0 A, 477.7 A/cm<sup>2</sup> and 14.3 MW/cm<sup>3</sup> to 30.0 A, 955.4 A/cm<sup>2</sup> and 57.3 MW/cm<sup>3</sup>, respectively,

with increasing electric field from 60 kV/cm to 120 kV/cm. According to the equation:

$$C_D = \frac{I_{max}}{S} = \frac{dQ_{max}}{S dt} = \frac{dP_{max}}{dt}$$
(9)

the large  $C_D$  of 0.96SBT-0.04NBN ceramics can be attributed to the high induced polarization and short discharge time [5]. Besides, the large electric field of 120 kV/cm is also responsible for the large P<sub>D</sub> of 0.96SBT-0.04NBN ceramics. These results reveal the prospect of 0.96SBT-0.04NBN ceramics for application in pulsed power technology.

Fig. 12(a) and (b) exhibit intuitive comparisons of energy storage properties [7,14–16,19,27,28,30–40] and charge-discharge performance [8,41–47] for 0.96SBT-0.04NBN ceramics and other reported ceramics. Most of the previously reported ceramics exhibit a common minus that the trade-off between  $W_{rec}$  and  $\eta$ . Note that the brand-new 0.96SBT-0.04NBN ceramics show a large  $W_{rec}$  (2.49 J/ cm<sup>3</sup>) and a high  $\eta$  (89.7%) simultaneously, which is superior to other ceramics. Besides, 0.96SBT-0.04NBN ceramics also possess an ultrafast discharge speed (~34 ns) combined with a remarkable  $P_D$  of 57.3 MW/cm<sup>3</sup>. These results fully reveal the superiority of 0.96SBT-0.04NBN ceramics in pulse ceramic capacitors.



**Fig. 11.** (a) Overdamped pulse discharge current curves of 0.96SBT-0.04NBN ceramics under various electric fields. (b)  $W_d$  as a function of time for 0.96SBT-0.04NBN ceramics. (c) Underdamped discharge waveforms of 0.96SBT-0.04NBN ceramics at different electric fields. (d) Variation of  $I_{max}$ ,  $P_D$  and  $C_D$  as a function of electric field.



**Fig. 12.** (a)  $W_{rec}$  and  $\eta$  of 0.96SBT-0.04NBN ceramics and some previously published ceramics. (b) A comparison of  $\tau_{0.9}$  and  $P_D$  for 0.96SBT-0.04NBN ceramics and other reported ceramics.

## 4. Conclusions

In summary, the (1-*x*)SBT-*x*NBN ceramics are studied for energy storage applications. The introduction of NBN into SBT ergodic relaxors lead to the enhanced  $W_{rec}$  of 2.49 J/cm<sup>3</sup> and  $\eta$  of 89.7% at 250 kV/cm for 0.96SBT-0.04NBN ceramics. Besides, the  $W_{rec}$  and  $\eta$  of 0.96SBT-0.04NBN ceramics. Besides, the  $W_{rec}$  and  $\eta$  of 0.96SBT-0.04NBN ceramics possesse excellent thermal stability and frequency stability. More significantly, an extraordinary fast discharge rate of 34 ns and remarkable  $P_D$  of 57.3 MW/cm<sup>3</sup> are obtained, which can be attributed to the high dynamic and weak correlation PNRs in ergodic relaxor ferroelectric state. The notable improvements in energy storage and charge-discharge properties indicate that 0.96SBT-0.04NBN ceramic is a promising candidate for advanced pulsed power applications.

#### **CRediT** authorship contribution statement

Kai Yao: Investigation, Methodology, Writing - original draft. Changrong Zhou: Funding acquisition, Supervision, Resources, Writing review & editing. Jiang Wang: Investigation, Methodology. Qingning Li: Investigation, Methodology. Changlai Yuan: Investigation, Methodology. Jiwen Xu: Investigation, Methodology. Guohua Chen: Investigation, Methodology. Guanghui Rao: Supervision, Resources, Writing - review & editing.

#### **Declaration of Competing Interest**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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