Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A. This journal is © The Royal Society of Chemistry 2024 Table S1 Energy storage parameters of BF-, BF-BT-, BNT- and NN-based ceramic systems.

| Ceramics | $W_{\rm rec}$ (J/cm ³) | η (%) | $E_{\rm b}({\rm kV/cm})$ | ho (J/kV·cm ²) | reference |
|--|------------------------------------|-------|--------------------------|----------------------------|-----------|
| $Bi_{0.83}Sm_{0.17}Fe_{0.95}Sc_{0.05}O_3$ | 2.21 | 76 | 230 | 9.6×10^{-3} | [41] |
| $\Box 0.67Bi_{0.9}Sm_{0.1}FeO_3-0.33BaTiO_3$ | 2.8 | 55.8 | 200 | 14.0×10^{-3} | [41] |
| $\Box 0.56BiFeO_{3}\text{-}0.3BaTiO_{3}\text{-}0.14AgNbO_{3}$ | 2.11 | 84 | 195 | 10.8×10^{-3} | [15] |
| $\Box 0.75BiFeO_3$ - $\Box 0.25Ba(Zn_{1/2}Ta_{2/3})O_3$ | 2.56 | 75 | 160 | 16.0×10^{-3} | [41] |
| □0.75BiNdFeO ₃ -0.12SrBaTiO ₃ | 1.74 | 74 | 150 | 16.6×10^{-3} | [41] |
| $0.62BiFeO_3 - 0.3BaTiO_3 - 0.08Nd(Zn_{0.5}Zr_{0.5})O_3$ | 2.45 | 72 | 240 | 10.2×10^{-3} | [45] |
| $0.61BiFeO_{3}\text{-}0.33(Ba_{0.8}Sr_{0.2})TiO_{3}\text{-}0.06La(Mg_{2/3}Nb_{1/3})O_{3}$ | 3.38 | 59 | 230 | 14.6×10^{-3} | [42] |
| $0.65BiFeO_3$ - $0.3BaTiO_3$ - $0.05Bi(Zn_{2/3}Nb_{1/3})O_3$ | 2.1 | 55.7 | 180 | 11.6×10^{-3} | [42] |
| $0.56BiFeO_3$ - $0.3BaTiO_3$ - $0.14Ba(Zn_{1/3}Nb_{2/3})O_3$ | 1.61 | ~65 | 180 | 8.9×10^{-3} | [42] |
| 0.61BiFeO3-0.33BaTiO3-0.06Ba(Mg1/3Nb2/3)O3 | 1.56 | 75 | 125 | 12.4×10^{-3} | [42] |
| 0.7(0.67BiFeO ₃ -0.33BaTiO ₃)-0.3(Sr _{0.7} Bi _{0.2})TiO ₃ | 2.4 | 90.4 | 180 | 13.3×10^{-3} | [42] |
| $0.75(Bi_{0.85}Nd_{0.15})FeO_3$ - $0.25BaTiO_3$ | 1.8 | 41.3 | 170 | 10.5×10^{-3} | [42] |
| $\Box 0.52Bi_{0.98}La_{0.02}FeO_{3}\text{-}0.48BaTiO_{3}$ | 1.22 | 58 | 140 | 8.71×10^{-3} | [42] |
| $0.65Bi_{0.5}Na_{0.5}TiO_{3}\text{-}0.35BaTiO_{3}\text{-}SrZr_{0.5}Ti_{0.5}O_{3}$ | 4.32 | 93.5 | 302 | 14.3×10^{-3} | [43] |
| $\square \square 0.8BNT-0.2SrNb_{0.5}Al_{0.5}O_{3}$ | 6.5 | 89 | 480 | 13.5×10^{-3} | [44] |
| $\square 0.16BNT-0.8NaNbO_{3}-0.04CaZrO_{3}$ | 3.7 | 82.1 | 400 | 9.2×10^{-3} | [44] |
| $(Na_{0.73}Bi_{0.08}Sm_{0.01})(Nb_{0.91}Ta_{0.09})O_3$ | 1.66 | 83.6 | 214 | 7.7×10^{-3} | [45] |
| $0.85(0.92 NaNbO_3 - 0.08 Bi(Mg_{0.5}Ti_{0.5})O_3) - 0.15 SrTiO_3$ | 6 | 81 | 300 | 20.0×10^{-3} | [46] |
| $0.2 NaNbO_3$ - $0.8 Sm(Mg_{0.5}Zr_{0.5})O_3$ | 4.3 | 85.6 | 560 | 7.6×10^{-3} | [47] |
| □0.85(0.7BiFeO ₃ -0.3BaTiO ₃)-0.15NaNbO ₃ | 8.2 | 70 | 325 | 25.2×10^{-3} | [14] |
| $\Box \Box Bi_{0.595}Ba_{0.255}Na_{0.15}Fe_{0.595}Ti_{0.255}Ta_{0.15}O_{3}$ | 8.7 | 65 | 350 (1Hz) | 24.9×10^{-3} | This work |
| $Bi_{0.595}Ba_{0.255}Na_{0.15}Fe_{0.595}Ti_{0.255}Ta_{0.15}O_3$ | 9.6 | 77 | 350 (10 Hz) | 27.4×10^{-3} | This work |
| $\Box Bi_{0.56}Ba_{0.24}Na_{0.2}Fe_{0.56}Ti_{0.24}Ta_{0.2}O_{3}$ | 8.7 | 66 | 550 (1 Hz) | 15.8×10^{-3} | This work |
| $\Box Bi_{0.56}Ba_{0.24}Na_{0.2}Fe_{0.56}Ti_{0.24}Ta_{0.2}O_{3}$ | 10.3 | 68 | 550 (10 Hz) | 18.7×10^{-3} | This work |

| <i>x</i> - NT | $W_{\rm rec}$ (J/cm ³) | ΔE (kV/cm) | ρ (J/kV·cm ²) | Measuring frequency |
|---------------|------------------------------------|--------------------|--------------------------------|---------------------|
| 0 | 2 | 175 | 11.4×10^{-3} | 1 Hz |
| 0.05 | 5.5 | 275 | 20.0×10^{-3} | 1 Hz |
| 0.10 | 7.4 | 350 | 21.1 × 10 ⁻³ | 1 Hz |
| □0.15 | 8.7 | 350 | 24.9×10^{-3} | 1 Hz |
| 0.15 | 9.6 | 350 | 27.4×10^{-3} | 10 Hz |
| 0.20 | 8.7 | 550 | 15.8×10^{-3} | 1 Hz |
| 0.20 | 10.3 | 550 | 18.7×10^{-3} | 10 Hz |
| 0.30 | 4.6 | 425 | 10.8 ×10 ⁻³ | 1 Hz |

Table S2 \Box Recoverable energy storage intensity (ρ). The calculated ρ values were obtained using
the relation $\rho = W_{\rm rec} / \Delta E$, based on Fig. 5(b,c,e,f).

$$(1-x)(0.7BiFeO_3 - 0.3BaTiO_3) - xNaTaO_3$$

$$S_{config} = -R \left[\sum_{(A-site\ cations)} x_i ln(x_i) + \sum_{(B-site\ cations)} x_j ln(x_j) + \sum_{(anions)} x_k ln(x_k) \right]$$
$$at\ x = 0$$

$$(Bi_{1.0}Fe_{1.0})_{0.7}(Ba_{1.0}Ti_{1.0})_{0.3}O_3$$

$$S_{config} = -R[2 * 0.7 \ln(0.7) + 2 * 0.3 \ln(0.3) + 1.0 \ln(1.0)]$$

$$S_{config} = -R[2 * 0.7(-0.356) + 2 * 0.3(-1.203) + 0]$$

$$S_{config} = R[0.498 + 0.721] = 1.21R$$

$$at x = 0.30$$

$$(Bi_{0.70}Fe_{0.70})_{0.7}(Ba_{0.70}Ti_{0.70})_{0.3}Na_{0.30}Ta_{0.30}O_3$$

$$\begin{split} S_{config} &= -R[2*0.49\ln(0.49) + 2*0.21\ln(0.21) + 2*0.30\ln(0.30) + 1.0\ln(1.0)] \\ S_{config} &= -R[2*0.49(-0.713) + 2*0.21(-1.560) + 2*0.30(1.203) + 0] \\ S_{config} &= R[0.698 + 0.655 + 0.721] = 2.07R \end{split}$$

Figure S1 The calculation process of configuration entropy.

Side view



Figure S2 Electrode's configuration for the P-E loop experiment.



Figure S3 Identification of the cubic Fd-3m Na₂Ta₂O₆ phase in the NaTaO₃-contained specimens.



Figure S4 Variations of different phase percentages (wt.%) of BBNFTT-*x* ceramics.



Figure S5 (a) P_{max} and P_r measured at E_b and (b) E_b , W_{rec} , and η vs. x of BBNFTT-x ceramics.



Figure S6 Temperature coefficients of capacitance (TCC) for BBNFTT-*x* from x = 0 to x = 0.30 for measuring frequency of 1 kHz.



Figure S7 EDX spectra of (a-c) BBNFTT-0.1 and (d-f) BBNFTT-0.2.