Supplementary Material

A superparaelectric design with structure optimization enables superior energy-storage performances and stabilities in $(Na_{0.5}Bi_{0.5})TiO_3$-based ceramics

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1. Experimental section

***Fabrication*:** High-purity (> 98%) Na_2CO_3, Bi_2O_3, $SrCO_3$, $CaCO_3$, Ta_2O_5, and TiO_2 powders were selected as raw materials to synthesize (1-x)NBST-xCTT (x = 0.0, 0.1, 0.2, 0.3, and 0.4) SPE-RFE ceramics. More specifically, the mixture of these powders was wet-milled stoichiometrically with zirconia balls in alcohol for 6~18 h and then dried at 80~100 °C for 3~6 h. Subsequently, the dried mixture was calcined at 800~1000 °C for 2~6 h and re-milled for 6~18 h to synthesize (1-x)NBST-xCTT powder. Then, the mixture of (1-x)NBST-xCTT powder with 40~50 wt.% solvent (toluene/ethanol), 0.6~1.5 wt.% dispersant (phosphate ester/glycerol trioleate), 2~3 wt.% plasticizer (polyethylene glycol/benzyl butyl phthalate), and 4~6 wt.% binder (Polyvinyl butyral) was milled to obtain a homogeneous slurry to fabricate the corresponding green tapes by a tape-casting method. After drying, the (1-x)NBST-xCTT green tapes were cut into the desired shape and laminated at 10~30 MPa at 50~80°C for green samples. Finally, the (1-x)NBST-xCTT SPE-RFE ceramics were prepared by burning the binder and sintering at 1150~1250°C for 2~5 hours in a sealed box furnace.

***Characterization*:** The microstructure of (1-x)NBST-xCTT SPE-RFE ceramics was detected by a scanning electron microscope (SEM, Phenom ProX, Phenom-World). The phase structures were analyzed using an X-ray diffractometer (XRD, PANalytical, X' Pert^3 Powder), and the GSAS–EXPGUI program carried out the refinement process. The select area electron diffraction (SAED) patterns, high-resolution (HR) images, and bright-field (BF) micrographs were obtained using transmission electron microscopy (TEM, FEI Tecnai G2 F30 STWIN, USA). A precision LCR meter (Agilent, E4980A) with a temperature controller measured the dielectric properties and complex impedance spectra. The electric breakdown strength was evaluated by a withstand voltage tester (Meiruike, RK2671A) under a DC voltage mode, and COMSOL Multiphysics simulated the breakdown behavior inside the ceramic. The polarization–electric field (*P*–*E*) hysteresis loops were obtained by a ferroelectric tester (Premier II, Radiant Technologies Inc., USA). The charge-discharge properties were characterized by a pulsed charge-discharge platform (CFD-003, Tongguo Technology).

2. Results and discussion

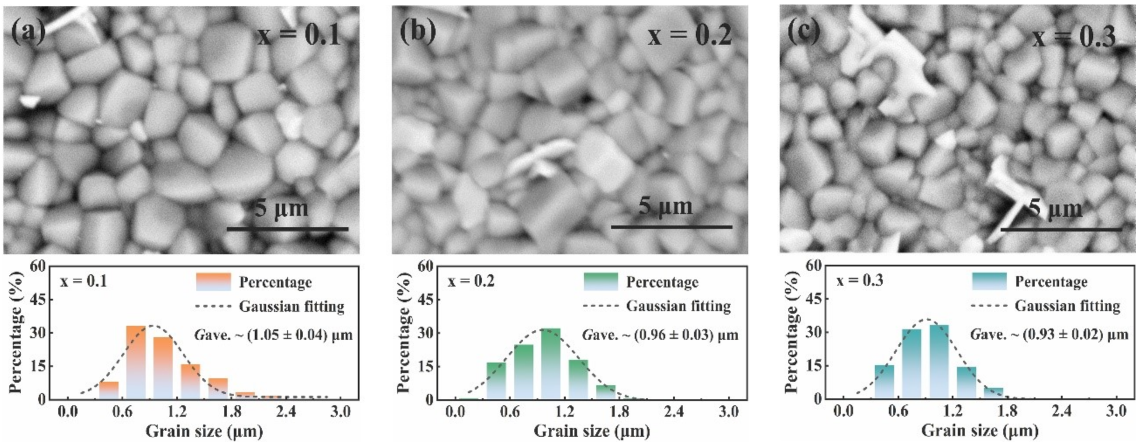

Fig. S1 (a-c) SEM images and grain size distributions of (1-x)NBST-xCTT ceramics.

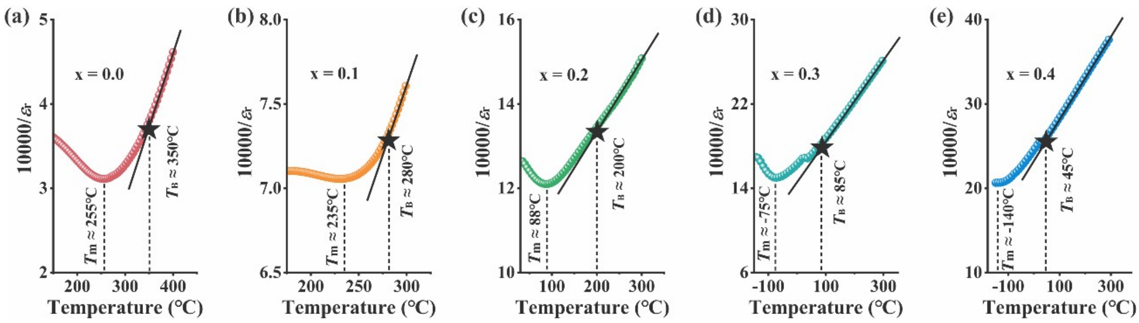

Fig. S2 (a-e) The $1/\varepsilon_r$ versus temperature curves of (1-x)NBST-xCTT ceramics.

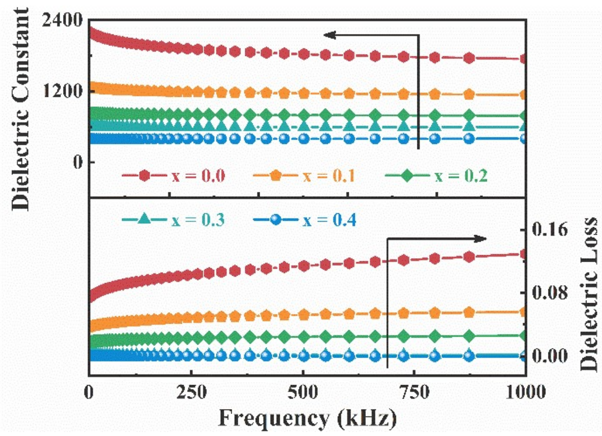

Fig. S3 (a) The frequency-dependent ε_r and tanδ of (1-x)NBST-xCTT ceramics.

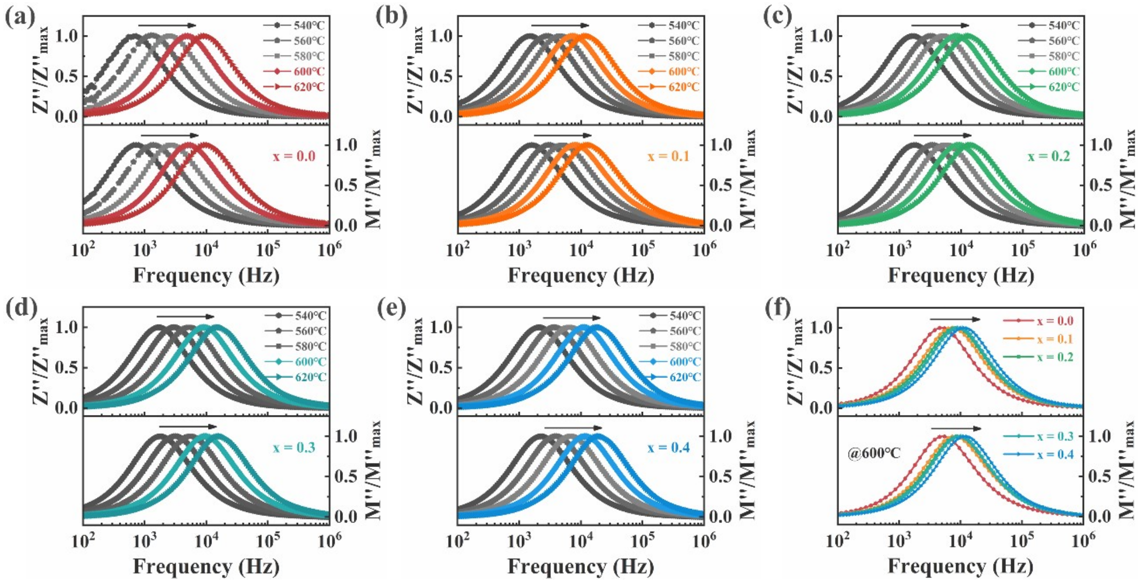

Fig. S4 (a-f) The normalized Z′′ and M′′ versus frequency at various temperatures for (1-x)NBST-xCTT ceramics.

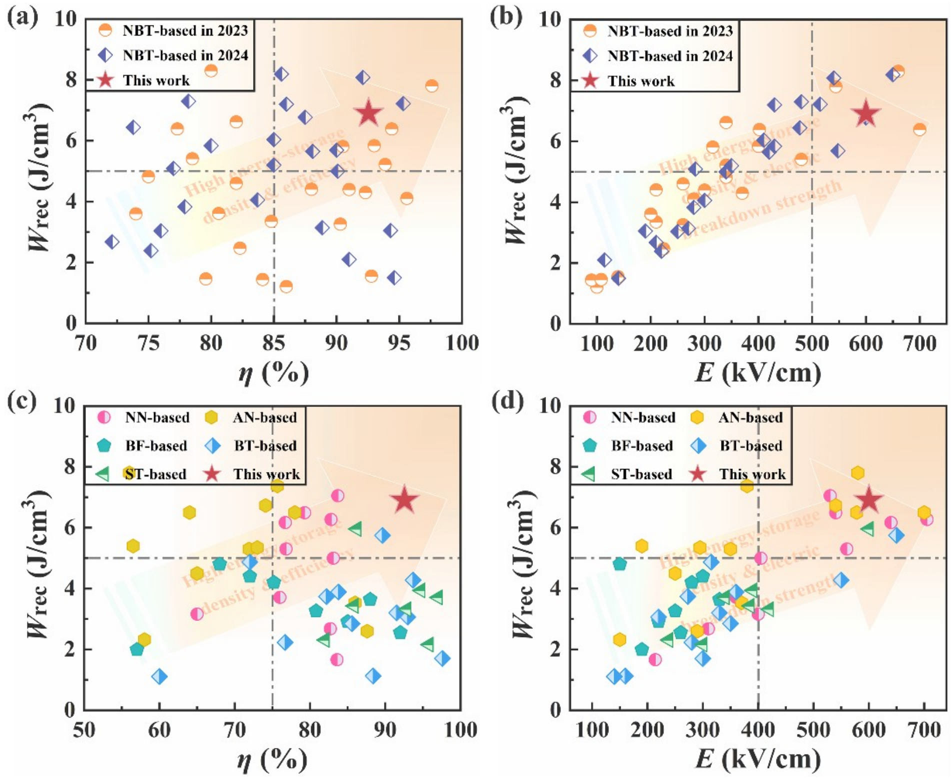

Fig. S5 Comparison of ESPs for 0.6NBST-0.4CTT in this work with other NBT-based (a-b)[1-47] and other lead-free (c-d)[48-96] ceramics.

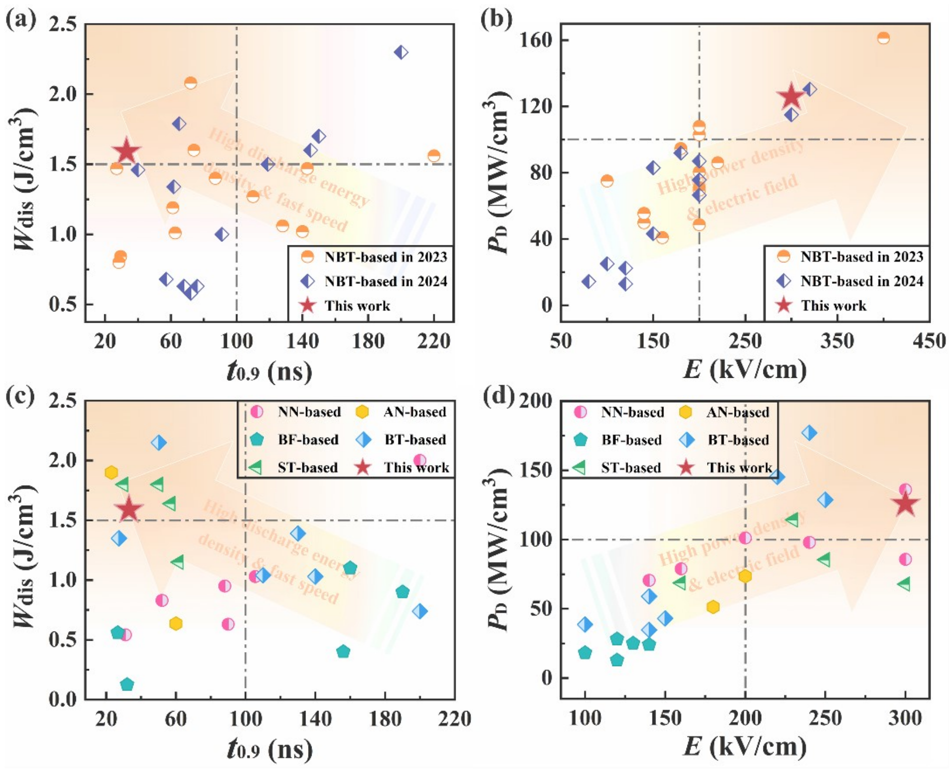

Fig. S6 (a-f) Comparison of CDPs for 0.6NBST-0.4CTT in this work with other NBT-based (a-b)[1, 2, 4, 7, 8, 10, 11, 18-21, 23, 24, 27, 29, 30, 32, 33, 38-40, 42, 44, 45, 47] and other lead-free (c-d)[49-52, 54-56, 61, 63, 69, 73-80, 83, 85-87, 92-96] ceramics.

Table S1 The fitted resistances of (1-x)NBST-xCTT ceramics at various temperatures.

	x = 0.0	x = 0.1	x = 0.2	x = 0.3	x = 0.4
	R_b (kΩ)	R_b (kΩ)	R_b (kΩ)	R_b (kΩ)	R_b (kΩ)
540°C	207.8	232.7	304.1	439.4	488.6
560°C	116.2	131.8	166.0	256.4	283.0
580°C	63.8	83.3	105.4	148.7	163.9
600°C	35.2	54.4	65.6	88.0	96.1
620°C	19.7	34.5	45.1	53.3	57.7

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