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Superior energy storage properties in $SrTiO_3$ - based dielectric ceramics through all-scale hierarchical architecture

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Characterization, electrical measurement and numerical simulation

The crystalline structures of powders and ceramics were analyzed by an X-ray diffraction (XRD, X Pert pro, PANalytical B.V.) with Cu Kα radiation. The microscopic morphologies of samples were observed by a transmission electron microscopy (TEM, Talos F200X, FEI) and a high-resolution field emission scanning electron microscope (SEM, SU8020, Hitachi). The morphologies of domain structure were observed by a piezoresponse force microscopy (PFM, Oxford, MFP-3D infinity). The UV-visible (UV-vis) absorption spectrum was meticulously examined using a UV-vis spectrophotometer (UV-3600, Shimadu). The HRMS-900 system and DMS-2000 (Partulab) were used to measure temperature dependent direct current (DC) conductivity and dielectric parameters, respectively. For determining the DC E_b, we utilized a withstand voltage test system (RK2671AM). The P-E loops were tested by a ferroelectric analyzer (TF 3000) to determine the key energy storage parameters. The charge-discharge curves were obtained using a pulsed charge-discharge system (CFD-003, Tongguo technology). For those performance measurements, we carefully prepared the ceramics to have a controlled thickness ranging from 0.03 to 0.10 mm. Additionally, we determined the Vickers hardness of the ceramic using a Vickers hardness tester (MH-500). This involved applying a precisely controlled load of 4.9033 N for a time duration of 15 s.

To compare the effect of microstructure on breakdown strength of ceramics, a finite element software (COMSOL) was used to computationally simulate the spatial distribution of electric potential and local electric fields. A rectangular initial geometry with a total size of 10.24×6.95 μm^2 was constructed for simulation, in which the spatial distributions of grains and grain boundaries were modeled according to the cross-sectional SEM images. When an electric field of 170 kV/cm was externally applied to the upper and lower sides of the ceramic, the distributions of those electrical parameters were calculated based on the electrostatic balance equation.

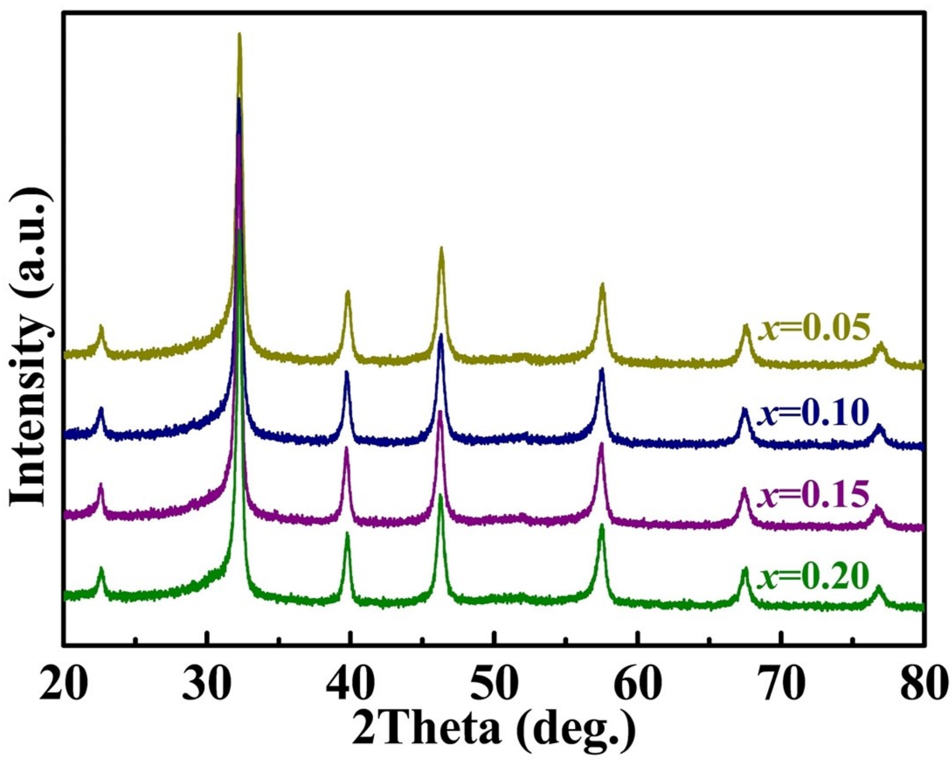

Fig. S1 XRD patterns of (1-x)SBT-xBNZT powders.

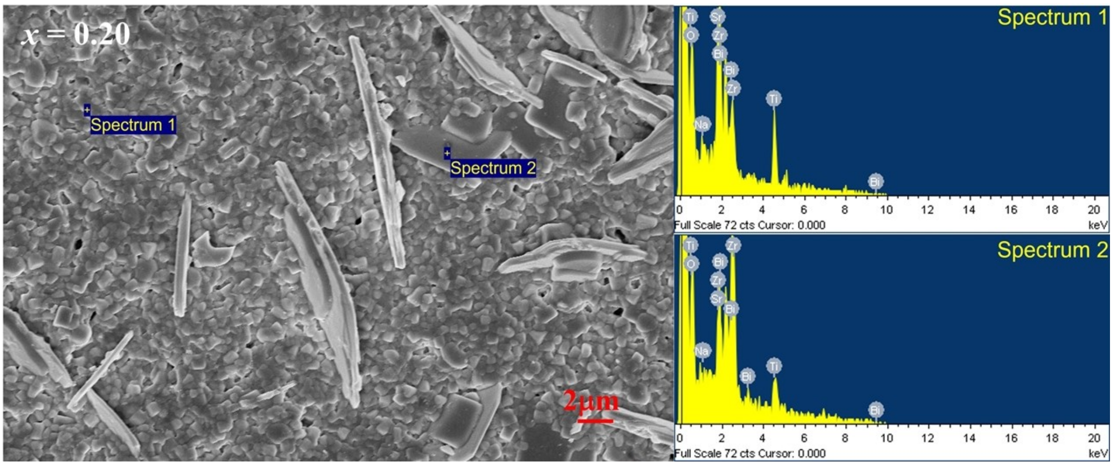

Fig. S2 Low magnification SEM image of x = 0.20 ceramic.

Table S1 The distribution of elements in spectrum 1 and 2.

Spectrum 1			Spectrum 2		
Element	Weight%	Atomic%	Element	Weight%	Atomic%
O K	26.14	63.26	O K	21.36	66.38
Na K	1.29	2.18	Na K	0.03	0.08
Ti K	19.35	15.64	Ti K	11.27	11.69
Sr L	28.17	12.45	Sr L	12.99	7.37
Zr L	7.66	3.25	Zr L	5.03	2.74
Bi M	17.39	3.22	Bi M	49.31	11.73
Totals	100%		Totals	100%	

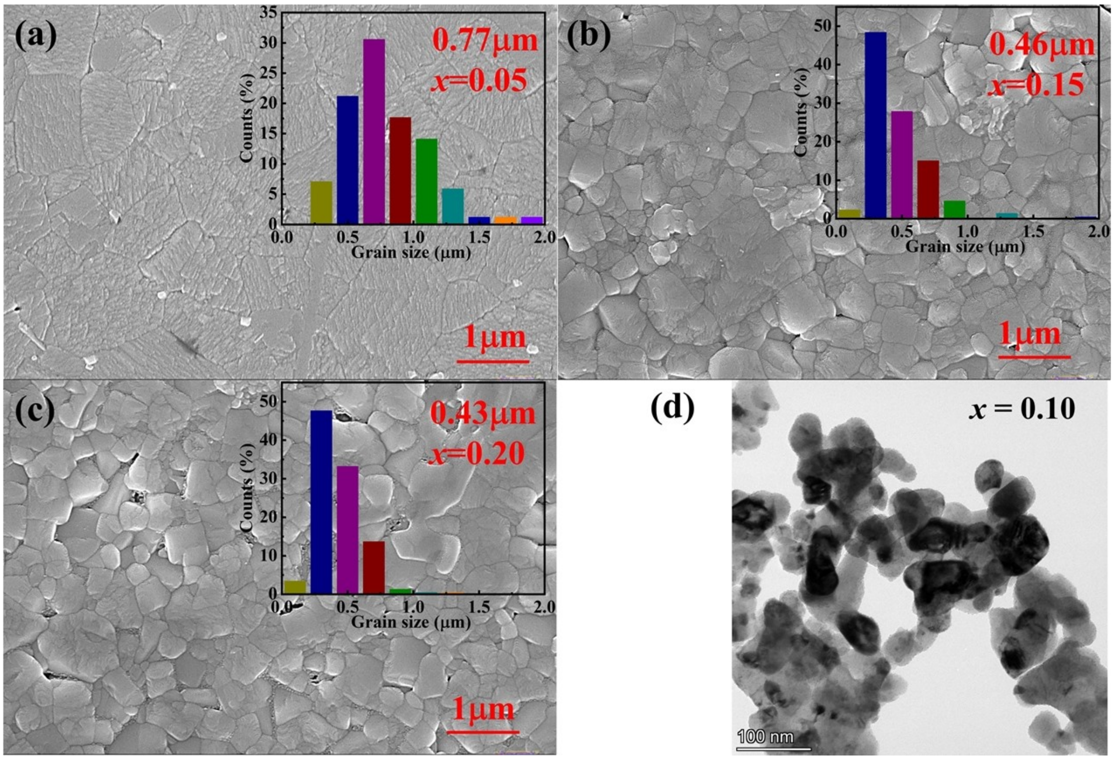

Fig. S3 Thermally etched surface SEM images of (a) $x = 0.05$, (b) $x = 0.15$, and (c) $x = 0.20$; (d) TEM image of $x = 0.10$ ceramic powders.

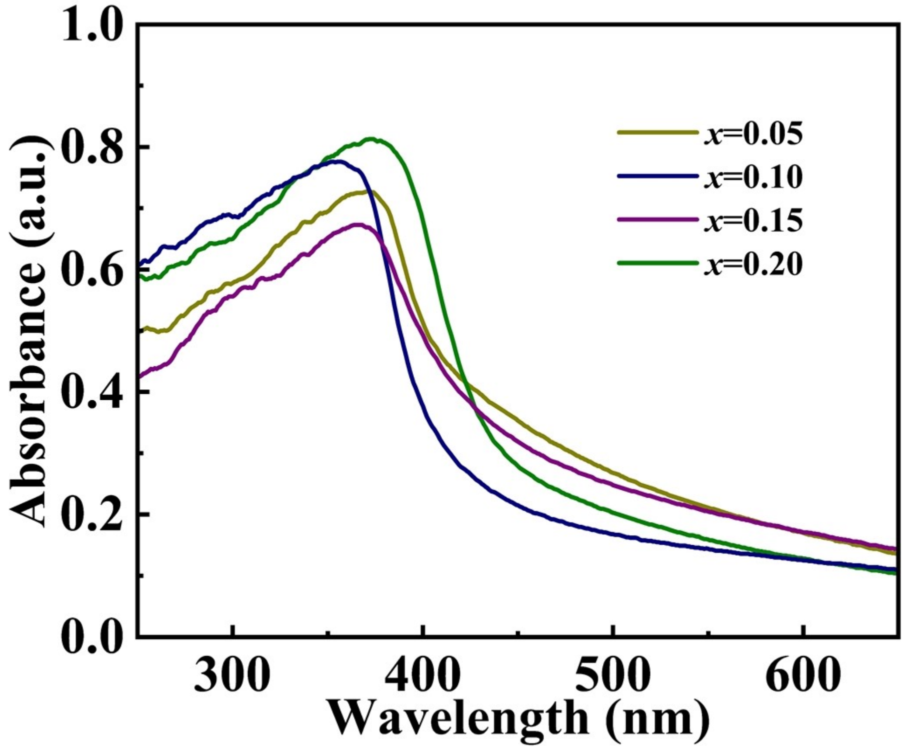

Fig. S4 UV-vis absorption spectra.

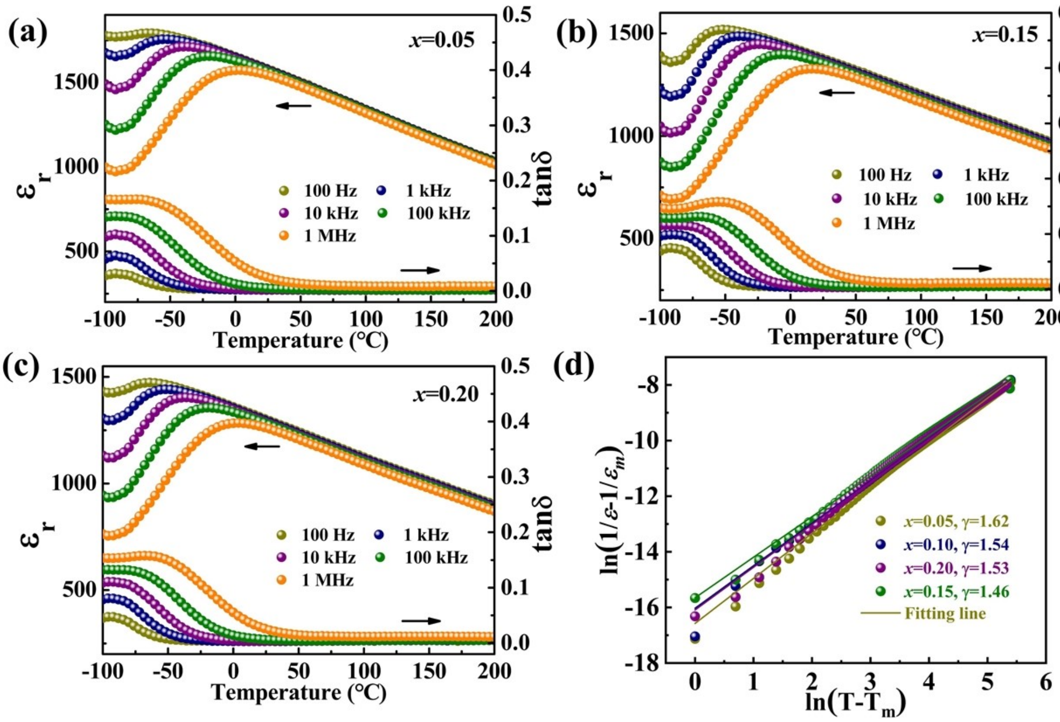

Fig. S5 (a-c) Temperature dependence of ε_r and tanδ of (1-*x*)SBT-*x*BNZT (*x* = 0.05, 0.15, 0.20) ceramics with various frequencies; (d) Plots of ln($1/\varepsilon_r$-$1/\varepsilon_m$) versus ln(T-T_m) at 1 MHz for (1-*x*)SBT-*x*BNZT ceramics.

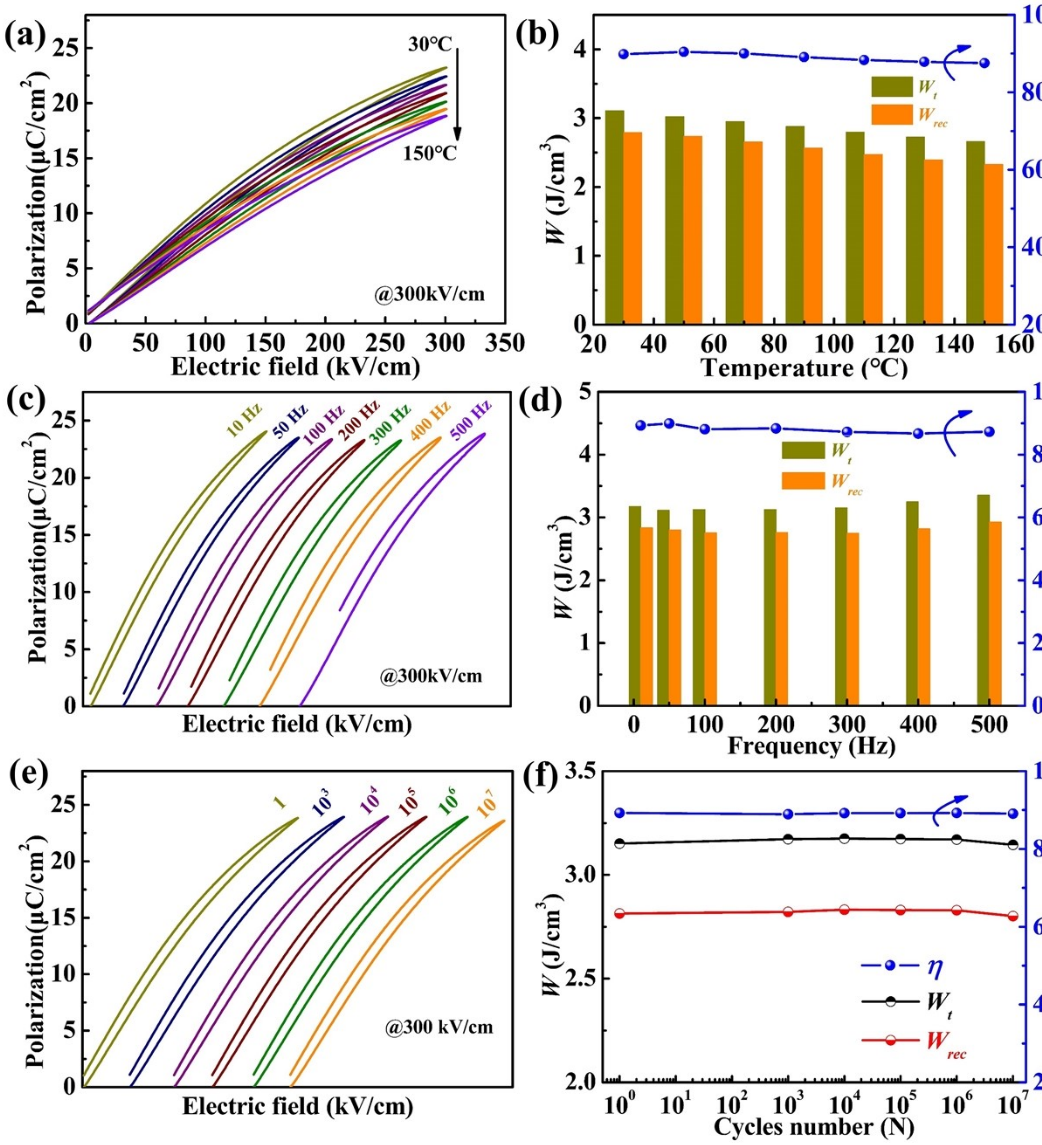

Fig. S6 Stability of energy storage of x = 0.10 ceramics: (a, c, e) Unipolar *P-E* loops at different temperatures, frequencies, and cycle numbers; The corresponding W_t, W_{rec} and η are concluded in (b), (d) and (f).

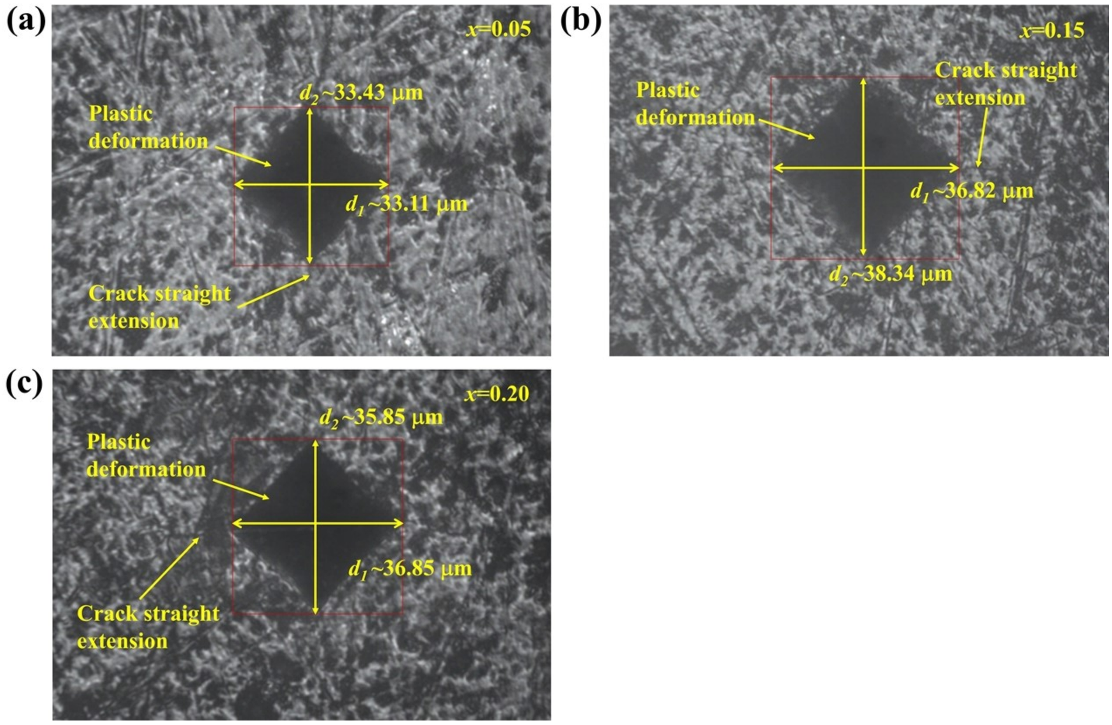

Fig. S7 The patterns of Vickers indentations and cracks of (1-*x*)SBT-*x*BNZT (x = 0.05, 0.15, 0.20) ceramics.

Table S2 The parameters of Vickers hardness test of the x = 0.05 ceramic.

Number	Load (N)	d_1 (μm)	d_2 (μm)	d_{ave} (μm)	H_v (GPa)
1	4.9033	33.11	33.43	33.27	8.21
2	4.9033	34.22	32.56	33.39	8.15
3	4.9033	33.22	34.20	33.71	7.99
					Average:8.12

Table S3 The parameters of Vickers hardness test of the x = 0.10 ceramic.

Number	Load (N)	d_1 (μm)	d_2 (μm)	d_{ave} (μm)	H_v (GPa)
1	4.9033	32.55	33.19	32.87	8.41
2	4.9033	33.03	33.73	33.38	8.16
3	4.9033	33.22	33.66	33.44	8.13
					Average:8.23

Table S4 The parameters of Vickers hardness test of the x = 0.15 ceramic.

Number	Load (N)	d_1 (μm)	d_2 (μm)	d_{ave} (μm)	H_v (GPa)
1	4.9033	36.82	38.34	37.58	6.43
2	4.9033	37.99	37.65	37.82	6.35
3	4.9033	39.33	38.19	38.76	6.05
					Average:6.28

Table S5 The parameters of Vickers hardness test of the x = 0.20 ceramic.

Number	Load (N)	d_1 (μm)	d_2 (μm)	d_{ave} (μm)	H_v (GPa)
1	4.9033	36.85	35.85	36.35	6.87
2	4.9033	35.77	37.03	36.40	6.86
3	4.9033	37.55	35.67	36.61	6.78
					Average:6.84