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Supplementary Information

for

Relaxor induced performance tuning around morphotropic phase boundary in

 $Ba_{0.86}Sr_{0.14}Ti_{0.94}Sn_{0.06} \ modified \ BNT-based \ ceramics$

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Experimental procedure

(1-x) Bi_{0.51}Na_{0.5}TiO₃-xBa_{0.86}Sr_{0.14}Ti_{0.94}Sn_{0.06}O₃ [abbreviated as (1-x) BNT-xBSTS, x =0, 0.02, 0.04, 0.06, 0.08, and 0.10] ceramics were synthesized by the conventional solid stated reaction method. Raw materials including Bi₂O₃ (99.0 %), Na₂CO₃ (99.8 %), TiO₂ (98 %), BaCO₃ (99.0 %), SrCO₃ (99.0 %), and SnO₂ (99.5 %) were produced by Sinopharm Chemical Reagent Co., Ltd. (China). First, raw materials were weighed according to the corresponding formula, and then they were ball milled for 24 h with both ZrO₂ ball and ethyl alcohol as the media. Next, the dry powders were calcined at 850 °C for 6 hours in a sealed alumina crucible. The calcined powders were pressed into disks with a diameter of about 10 mm and a thickness of about 1.0 mm under a pressure of 10 MPa after being mixed with a binder of 8 wt% polyvinyl alcohol (PVA). After the removal of PVA, the disks were sintered at 1150-1200 °C for 3 h in the air atmosphere. To characterize their electrical properties, the silver paste was covered on both sides of the samples and fired at 600 °C for 10 min to form the electrodes. All samples were poled under a dc voltage of 3 kV for 15 min at room temperature in a silicone oil bath.

The crystal structures of the sintered samples were determined by X-ray diffraction (XRD) using Cu K_{α} radiation (XRD, Bruker D8 Advanced XRD, USA). The microstructure of the sintered samples was characterized by field emission scanning electron microscopy (FE-SEM, Zeiss Supra 55, Germany). A ferroelectric tester (TF Analyzer 2000E, Germany) was used to measure the polarization versus electric field hysteresis loops(*P-E*) and the strain-electric field curves (*S-E*). The d_{33} values were

measured by a d_{33} tester (ZJ-3A, China) for the poled samples. The curves of dielectric constant (ε_r) and dielectric loss (tan δ) against different temperatures (-150 °C ~ 500 °C) were measured by an LCR analyzer (DMS-2000, China).



FIGURE S1 XRD patterns of (1-x) BNT-xBSTS ceramics.



FIGURE S2 The diffusive factor fitting curve of (1-x) BNT-*x*BSTS ceramics: (a) x =

0, (b) x = 0.02, (c) x = 0.04, (d) x = 0.06, (e) x = 0.08, and (f) x = 0.10.



FIGURE S3 P_{max} of the (1-x) BNT-xBSTS ceramics as a function of temperature.



FIGURE S4 Bipolar *S*-*E* curves of the (1-*x*) BNT-*x*BSTS ceramics as a function of temperature: (a) x = 0, (b) x = 0.06, (c) x = 0.08, and (d) x = 0.10; (e) S_{pos} , (f) S_{pol} , (g) D_{s} .



FIGURE S5 P_{max} of the (1-x) BNT-xBSTS ceramics as a function of the electric field.



FIGURE S6 Bipolar *S*-*E* curves of the (1-*x*) BNT-*x*BSTS ceramics as a function of the electric field: (a) x = 0, (b) x = 0.06, (c) x = 0.08, and (d) x = 0.10; (e) S_{pos} , (f) S_{pol} .

Composition	Space	a (Å)	b (Å)	c (Å)	Content (%)	Sig	R _w
	group						
x = 0	R3c	5.4851	5.4851	13.5169	100	1.8384	8.0505
<i>x</i> = 0.02	R3c	5.4935	5.4935	13.5225	87.28	1.9233	7.6391
	P4bm	5.4963	5.4963	3.8857	12.72		
<i>x</i> = 0.04	R3c	5.4998	5.4998	13.5491	84.51	1.8799	7.2169
	P4bm	5.5139	5.5139	3.9266	15.49		
<i>x</i> = 0.06	R3c	5.5162	5.5162	13.5112	61.18	1.9321	8.1161
	P4bm	5.5222	5.5222	3.8973	38.82		
<i>x</i> = 0.08	R3c	5.5419	5.5419	13.6792	7.32	1.7027	6.5607
	P4bm	5.5254	5.5254	3.9055	92.68		
<i>x</i> = 0.10	P4bm	5.5306	5.5306	3.9122	96.18	1.9262	8.0414
	P4mm	3.9579	3.9579	3.9396	3.82		

TABLE S1 Refinement results in (1-x) BNT-xBSTS ceramics