Supporting Information

Strong relaxor enabled excellent capacitive energy storage performance in $Bi_{0.5}Na_{0.5}TiO_3$-based binary system

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Material preparation

Ceramics composed of $(1-x)Bi_{0.5}Na_{0.5}TiO_3$–$xSmFeO_3$ [(1–x)BNT–xSF] with x values of 0.05, 0.1, 0.15, and 0.2 were synthesized using the solid-state method. High-purity raw materials, namely Na_2CO_3 (≥ 99.8%), Bi_2O_3 (≥ 99%), TiO_2 (≥ 98%), Fe_2O_3 (≥ 99%) and Sm_2O_3 (≥ 99.9%), supplied by Sinopharm Chemical Reagent Co., Ltd, China, were measured in stoichiometric proportions. The powders underwent two stages of ball milling at 300 rpm for 24 hous with an intermediate calcination step at 850 °C for 6 hours. After drying, the powder mixture was granulated using 3 wt.% polyvinyl alcohol (PVA) as a binder. Subsequently, the granulated powders were compacted into pellets under a pressure of 200 MPa and then sintered at temperatures ranging from 1000 to 1100 °C for 2 hours. The thickness of the ceramic sample is around 40 μm -80 μm.

Characterization

The phase composition of the ceramics was determined using XRD analysis with a D8 ADVANCE-A25 system from Bruker, Germany. The microstructure and elemental distribution were examined using a FE-SEM (Zeiss Gemini SEM 300, Germany) and energy-dispersive spectroscopy (EDS) with an Oxford Instruments X-MaxN SN 78861 detector, respectively. Prior to analysis, the ceramic samples were polished and subjected to thermal etching at temperatures ranging from 950 to 1050 °C for 0.5 hours. The grain sizes of the ceramics were quantified using ImageJ software. Dielectric properties, specifically the dielectric constant and loss, were measured from 25 to 400 °C and across a frequency range of 1 to 1000 kHz using a DPTS-AT-600 dielectric testing system from China. Breakdown strength was assessed using a dielectric breakdown test system from PolyK Technologies, USA, with voltage increments of

500 V per second. The *P–E* loop characteristics of the ceramics were studied at 10 Hz using ferroelectric test equipment from PolyK Technologies, USA. The frequency and temperature stability were investigated using a PolyK Technologies, USA. Fatigue stability was recorded using another ferroelectric test device (FETS-2000, China). Finally, charge and discharge behavior were evaluated using a CCDM-1000 system.

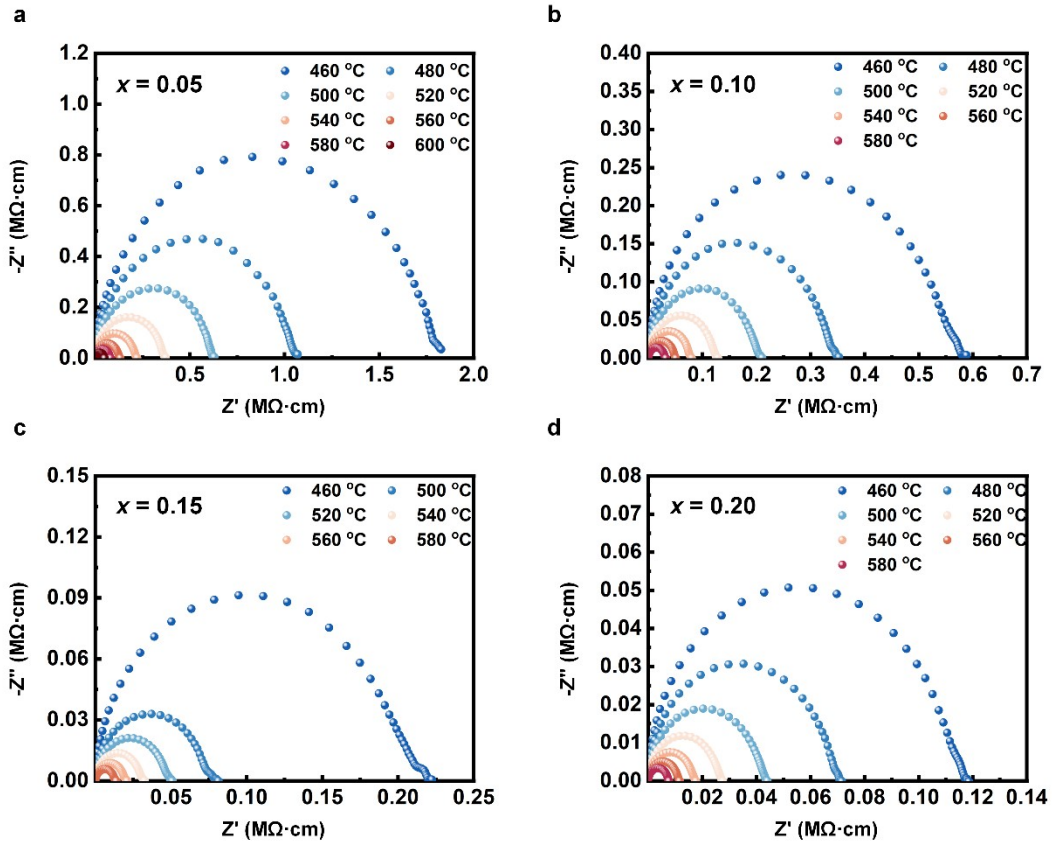

Figure S1 Impedance spectrum of (1 − *x*)BNT−*x*SF dielectric ceramics: **(a)** *x* = 0.05, **(b)** *x* = 0.1, **(c)** *x* = 0.15, AND **(d)** *x* = 0.20.

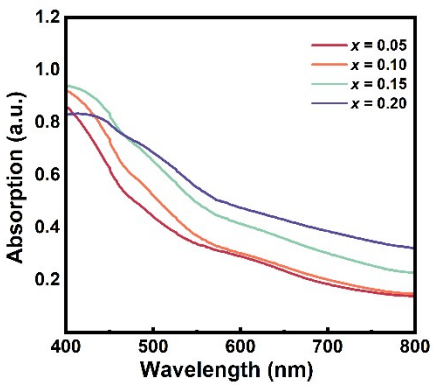

Figure S2 UV-vis absorption spectrum of the $(1-x)$BNT–xSF (x = 0.05, 0.10, 0.15, and 0.20) ceramics.

Table S1. A comparison of W_{rec} between 0.9BNT−0.1SF and other reported BNT-based lead-free bulk ceramics.

Composition	W_{rec} (J/cm^3)	η (%)	Reference
0.8BNST−0.2Ba(5M)O_3	4.89	92.1	[1]
0.8BNST−0.2Ba(5M)O_3	7.16	93.3	[2]
BNKTYN−9NN	1.6	73.1	[3]
BNT−BZT−0.04Sm	1.12	89.6	[4]
0.8BNT−0.2CT	1.26	91.3	[5]
BNBT−4LZNT	1.66	73.97	[6]
BNBT−0.2NNCS	1.86	88.23	[7]
BNT−BT−BSTN	2.07	94.5	[8]
0.88(0.85BNT−0.15BSmT)−0.12NN	2.42	81.18	[9]
0.6BNT−0.4STS	2.47	83.2	[10]
BNT−0.25CT	2.74	91	[11]
0.85BNKT−0.15SMN	3.5	86	[12]
BNST−0.15BMS	3.76	78.8	[13]
0.70BLNBT−0.30STN	4.2	89.3	[14]
BNKMN−0.3SLT	4.03	85.2	[15]
BNST−0.1AN	4.3	83	[16]
0.94(BNST−BMN) −0.06BZ	4.07	91	[17]

BNST−0.15BNH	4.11	82.1	[18]
0.6BS−0.4NN	4.44	81.8	[19]
BSNBT−0.08BBT	4.62	79.1	[20]
BS−0.15BMT	4.82	84.9	[21]
0.7BNLT−0.3NN	4.83	78.9	[22]
BNT−NN/7wt%CZT	4.93	93.3	[23]
BNT−SLnT	4.94	88.45	[24]
BNT−BCZT−0.05BS	4.97	84.4	[25]
BNT−NN−0.25SZM	5.2	85	[26]
BNT−SBT0.12SZNT	5.2	91	[27]
BNST−0.16BMN	5.5	90.1	[28]
BNT−0.33SLZT	5.09	88	[29]
NBT−SBCT−0.2BMH	5.9	85	[30]
NBT−SBT/6AlN	5.53	90	[31]
BNST−0.12BMH	5.59	85.3	[32]
BNT−BAT−0.4CT	5.81	97.8	[33]
NBCSTO	5.84	80	[34]
BNT−BT−0.12La	5.93	77.6	[35]
BNT−SST−0.03LMN	5.94	86.9	[36]
BNT−ST−0.08BMS	5.99	76	[37]
BNT−0.15ANT	6.6	72	[38]

BNBT−0.06CH	6.19	93.5	[39]
BNLT−0.16SANT	6.43	88	[40]
BNCTZ−0.5wt%LC	6.57	70	[41]
0.9NBST−0.1BMS	6.68	89.1	[42]
BNT−SBT−0.08LMT	7.3	93	[43]
BNT−CBT−0.28NBCT	7.13	83	[44]
BNTSNA	7	95	[45]
0.88BNT−0.12BT	8	86	[46]
My work	9.05	71.4	

Table S2. Comparison of W_{rec} in these Pb-free ceramics with different E_b (Except for some of the work in **Table S1**).

Composition	W_{rec} (J/cm^3)	E_b (kV/cm)	Reference
BTP3	0.41	75	[47]
$Ba(Ti_{0.9744}Mn_{0.01}Nb_{0.0125})O_3$	0.43	105	[48]
0.9BT−0.1BMN−0.3 wt. % $MnCO_3$	1.7	210	[49]
0.88BT−0.12BMS	2.25	240	[50]
0.9BT−0.1BNS	2.52	240	[51]
BT−Ta	2.85	350	[52]
0.9BST−0.1BMN	3.34	400	[53]
BT−BZ−0.4CT	3.46	590	[54]
BT−SBT−NdVPP	4.2	460	[55]

0.65BT−0.35(SBT−BMZ)	4.03	370	[56]
BT−0.16BMS	4.28	550	[57]
BT−BLN−0.072BMT	4.42	500	[58]
BT−SBT−0.015CT	4	480	[59]
BT−H(Mg)	5.18	640	[60]
BBTMT−0.1	5.97	710	[61]
BT−BMT−0.15NT	6.02	780	[62]
$(Ag_{0.92}Sr_{0.04})(Nb_{0.78}Ta_{0.22})O_3$	5.6	300	[63]
$Ag_{0.94}La_{0.02}Nb_{0.8}Ta_{0.2}O_3$	6.73	540	[64]
$Ag_{0.91}La_{0.03}Nb_{0.9}Ta_{0.1}O_3$	8.6	460	[65]
BF−BT−0.08SMZTN	2.4	190	[66]
BNFT−SBT−NT	3.44	210	[67]
BF−BT−0.15SNA	3.95	300	[68]
BF−ST−LMN	6.3	450	[69]
$BF-BHfTiO_3-0.17NT$	6.3	425	[70]
BFO−0.5(BST−BZN)	7.4	680	[71]
BF−BT−0.14SSN	8.5	470	[72]
$0.8BNST-0.2Ba(5M)O_3$	4.89	351	[1]
BNKTYN−9NN	1.6	110	[3]
0.8BNT−0.2CT	1.26	150	[5]
BNBT−4LZNT	1.66	139	[6]

BNT−ST: 0.1 wt% AlN	2.07	160	[73]
0.88(0.85BNT−0.15BSmT)−0.12NN	2.42	170	[74]
0.85BNKT−0.15SMN	3.5	300	[12]
BNKMN−0.3SLT	4.03	300	[15]
BNST−0.1AN	4.3	230	[16]
0.94(BNST−BMN) −0.06BZ	4.07	260	[17]
BNST−0.15BNH	4.11	240	[18]
0.6BS−0.4NN	4.44	260	[19]
BNKST−0.15BMT	4.82	270	[21]
BNT−SLnT	4.94	440	[24]
BNT−NN−0.25SZM	5.2	370	[26]
BNT−SBT0.12SZNT	5.2	330	[27]
NBT−SBCT−0.2BMH	5.9	260	[30]
NBT−SBT/6AlN	5.53	360	[31]
NBCSTO	5.84	430	[34]
BNT−BT−0.12La	5.93	440	[35]
BNLT−0.16SANT	6.43	505	[40]
BNCTZ−0.5wt%LC	6.57	380	[41]
0.9NBST−0.1BMS	6.68	405	[42]
BNT−SBT−0.08LMT	7.3	390	[43]
0.85(0.76BNT_3−0.04SZ−0.2NN)−0.	7.05	387	[75]

15MnO_2			
0.6BNT−0.4(0.7ST−0.3BLT)−0.5at.%Nb_2O_5	8.63	520	[76]
0.80NN−0.20BNH	6.45	380	[77]
NN−BMT−0.15BNST	6.49	540	[78]
0.6(0.96NN−0.04BF)−0.4CT	6.84	712	[79]
NN−BMT−0.15ST	6	590	[80]
NN−BNT−0.2CT	7.1	646	[81]
0.52BF−0.4ST−0.08NN	4.4	300	[82]
BF−ST−SBT	5.61	440	[83]
ST−CA−MT−BCB	1.05	287.8	[84]
BNT−SBT−0.24ST−0.06SZ	1.25	108	[85]
ST−0.4BNKTZS	1.45	100	[86]
0.9SBT–0.1BMH	3.1	360	[87]
0.9SBT−0.1Bi(M_1M_2)O_3	3.71	340	[88]
BNLST−0.4ST	3.78	360	[89]
0.5BNT−0.5$Sr_{0.7}Sm_{0.2}TiO_3$	3.81	305	[90]
0.3SNBT	3.94	390	[91]
$Dy_{0.008}Sr_{0.992}TiO_3$	4	510	[92]
BNT−ST−SNN	5.22	340	[93]
0.2SNBCT	6	440	[94]

My work	9.05	517

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