Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

Supplementary information

Enhanced energy storage properties of Hafnium-modified ($0.7Ba_{0.55}Sr_{0.45}$-$0.3Bi_{0.5}Na_{0.5}$)TiO_3-based relaxor ferroelectric ceramics via regulating polarization nonlinearity and bandgap

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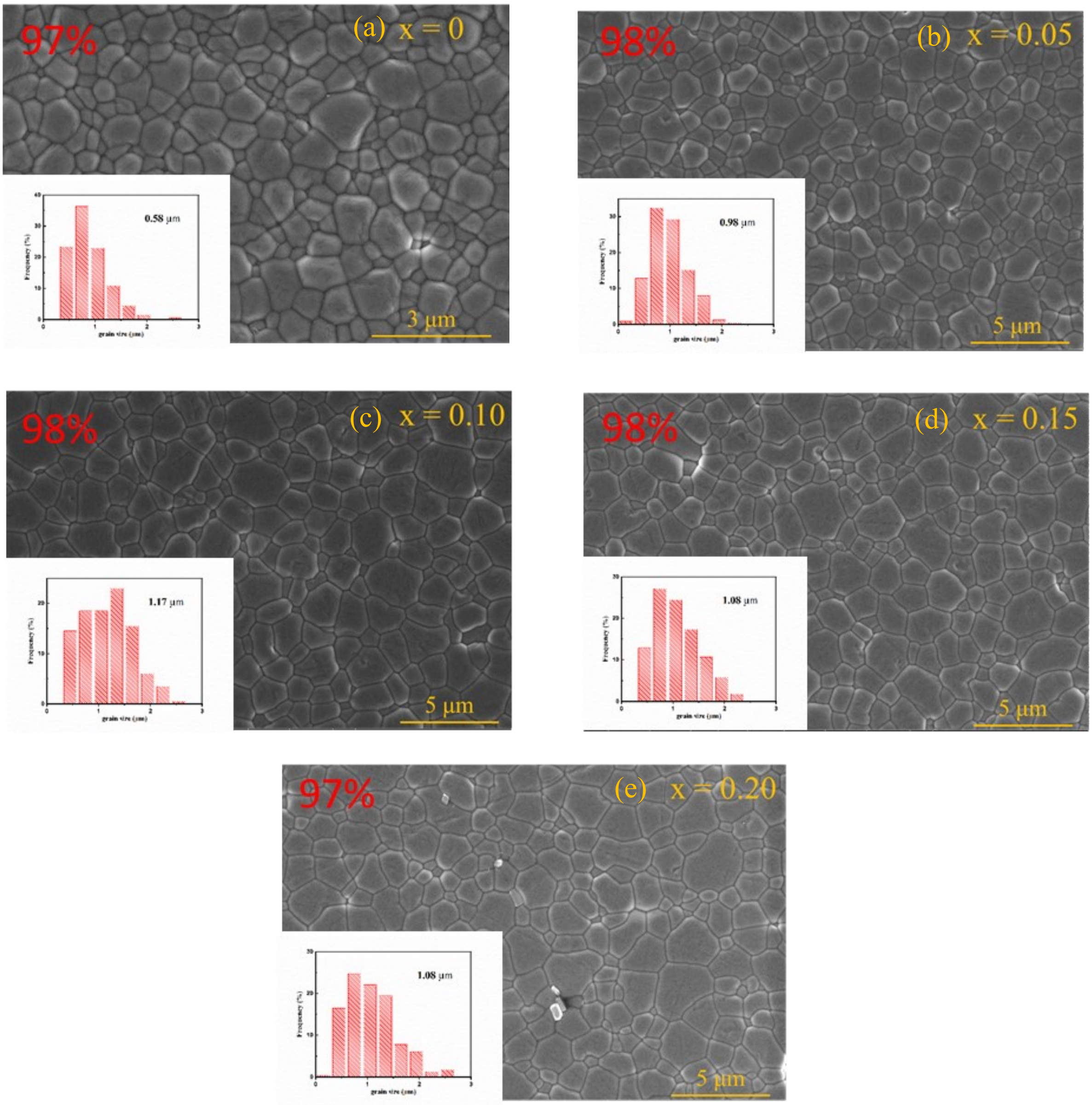

Fig. S1 The SEM micrographs of the polished and thermal-etched surfaces: (a) x=0, (b) x=0.05, (c) x=0.10, (d) x=0.15, (e) x=0.20.

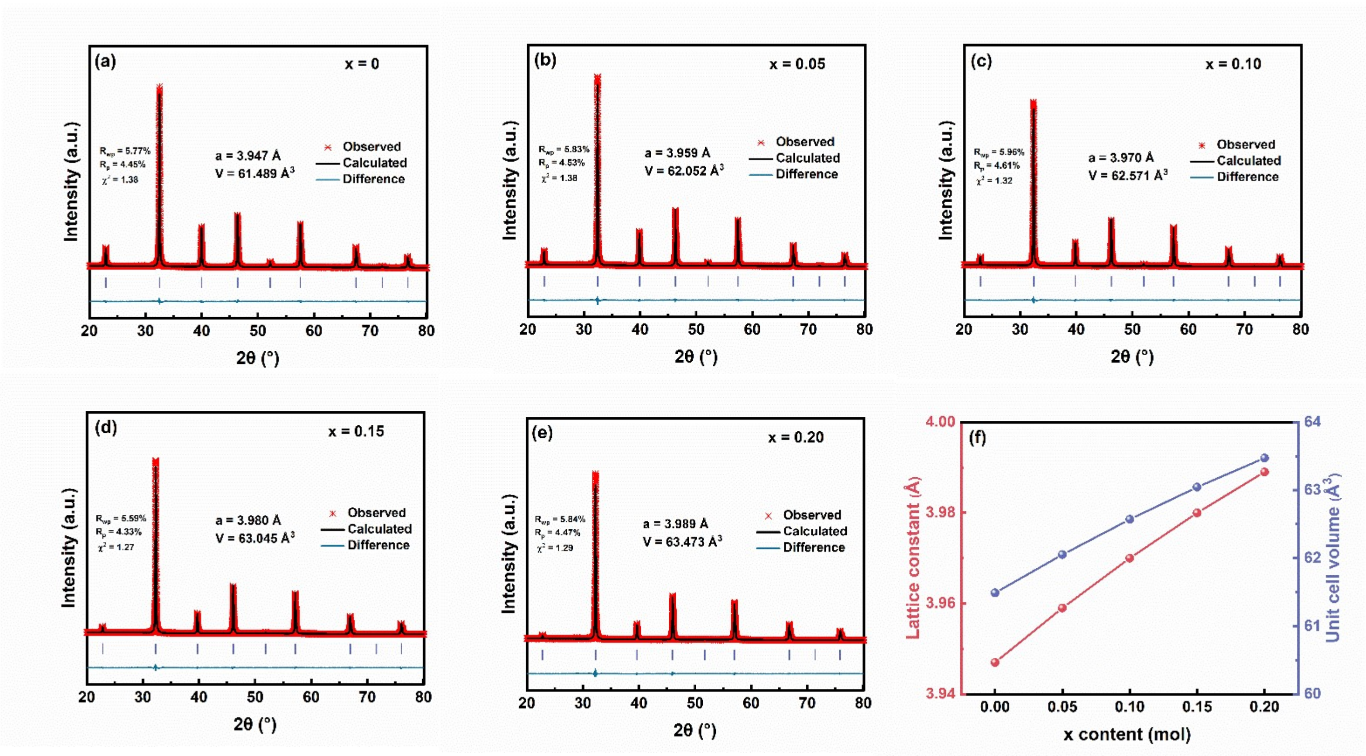

Fig. S2 Refinement XRD patterns of BSBNTH ceramics: (a) x=0, (b) x=0.05, (c) x=0.10, (d) x=0.15, (e) x=0.20; (f) The lattice constant and unit cell volume as functions of Hf content.

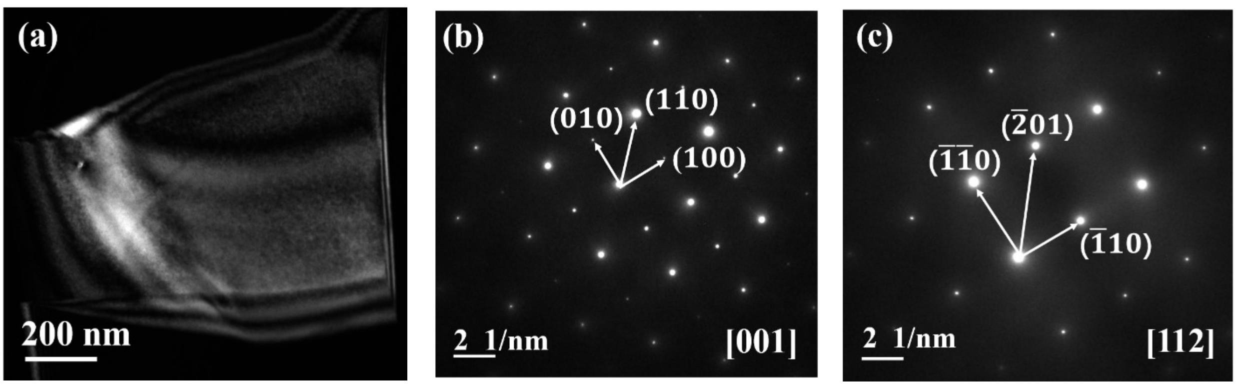

Fig. S3 (a) Dark-field TEM image; The SAED spots along (b) [001] and (c) [112] for BSBNTH-15 ceramics.

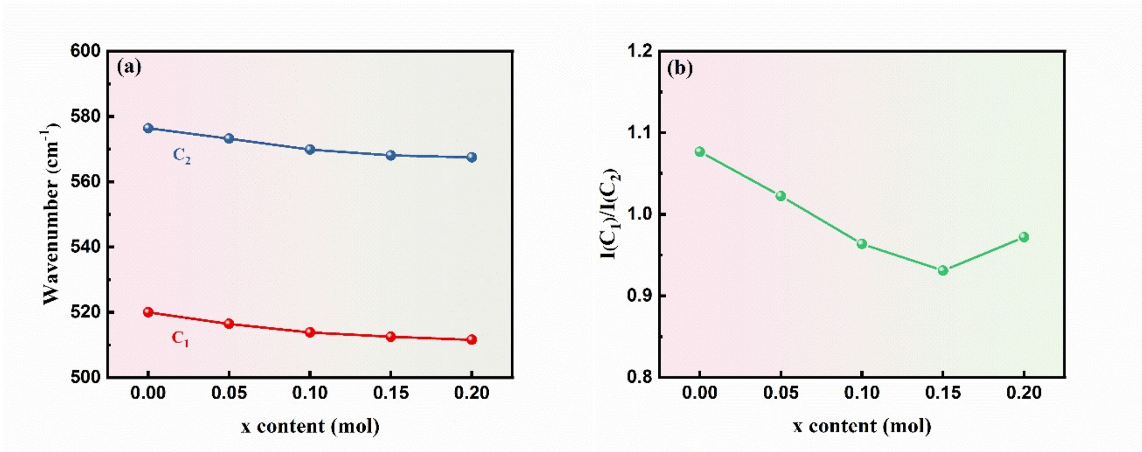

Fig. S4 The evolution of the (a) wavenumber and (b) intensity ration of the C_1 and C_2 as a function of Hf content in BSBNTH ceramics.

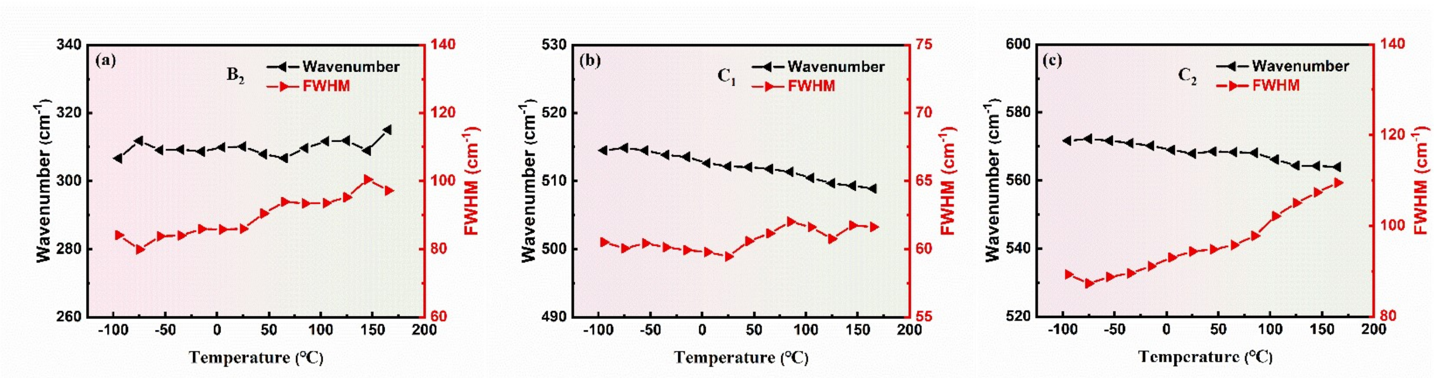

Fig. S5 (a) The peak positions of Raman spectra from − 95 ℃ to 165 ℃ for x=0.15 samples; the variance of wavenumber and full width at half maximum (FWHM) with increasing temperature for (b) B_2, (c) C_1, and (d) C_2 Raman vibration modes.

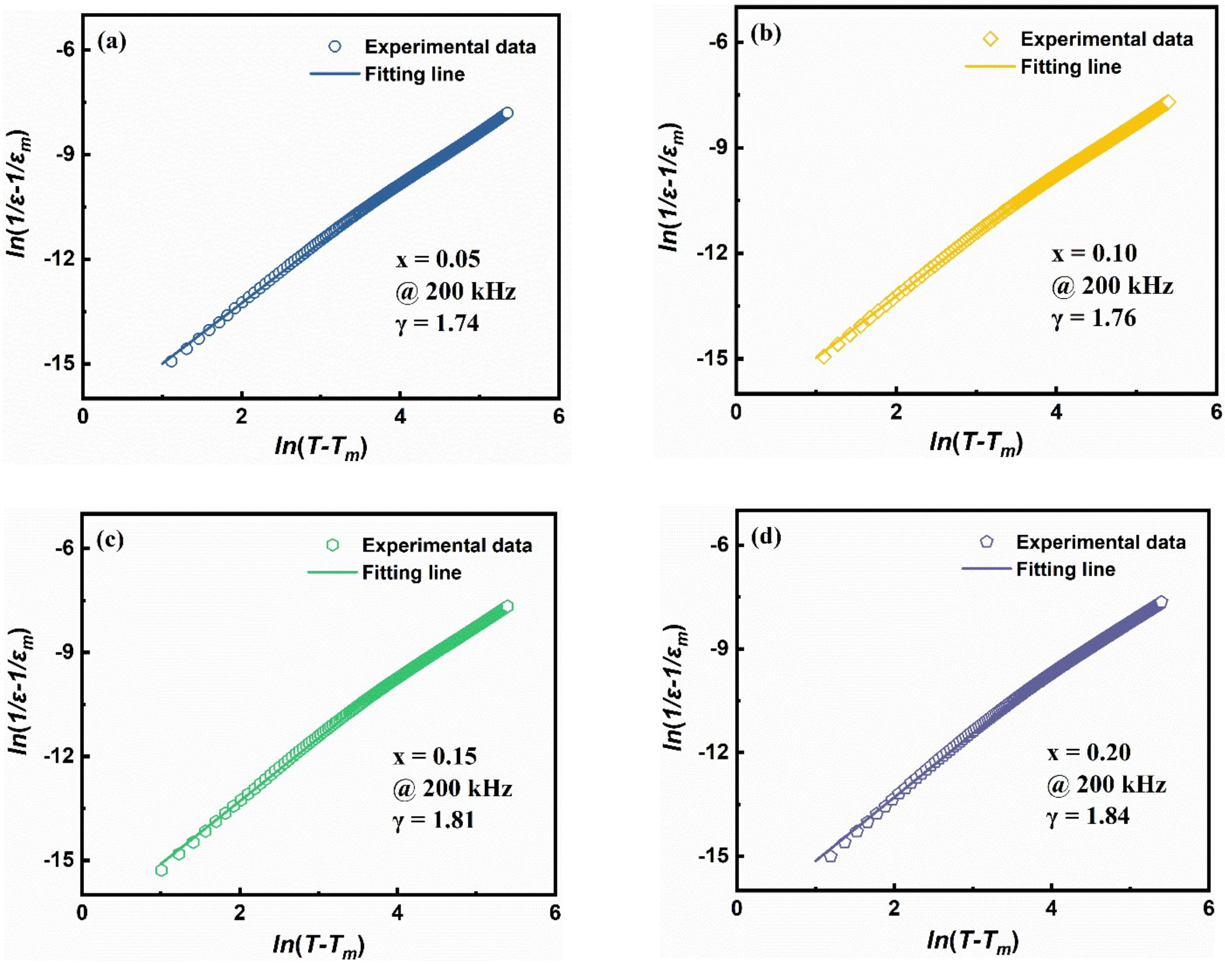

Fig. S6 ln(1/ε-1/ε_{m}) versus ln(T-T_{m}) for BSBNTH ceramics: (a) x=0.05, (b) x=0.10, (c) x=0.15, (d) x=0.20.

Table S1

The relative dielectric constant and dielectric loss of BSBNTH ceramics measured at 25 °C and 1 kHz.

25 °C，1 kHz	x=0	x=0.05	x=0.10	x=0.15	x=0.20
ε_r	3427	2218	1505	1019	754
tan δ	0.05227	0.00492	0.00229	0.00131	0.00124

Table S2

The value of R and C is interpreted on an equivalent circuit with a parallel resistance capacitance (RC) element.

650°C	x=0	x=0.05	x=0.10	x=0.15	x=0.20
R (Ω)	10991	20439	33275	34612	55363
C (F)	5.51×10^{-10}	5.79×10^{-10}	4.83×10^{-10}	4.18×10^{-10}	3.74×10^{-10}

Compounds	E (kV/cm)	W_{rec} (J/cm^3)	η (%)	ref
0.87BT-0.13Bi($Zn_{2/3}$(Nb,Ta)$_{1/3}$) O_3	**218**	**1.44**	**92.5**	**[17]**
0.88($Ba_{0.8}Sr_{0.2}$)TiO_3−0.12Bi($Zn_{2/3}Nb_{1/3}$)O_3	**225**	**1.62**	**99.8**	**[60]**
0.88BT−0.12Bi($Li_{0.5}Nb_{0.5}$)O_3	**270**	**2.03**	**88.0**	**[58]**
0.9BT−0.1Bi($Li_{0.5}Ta_{0.5}$)O_3	**280**	**2.2**	**88.0**	**[59]**
0.85BT–0.15Bi($Zn_{1/2}Sn_{1/2}$)O_3	**280**	**2.41**	**91.6**	**[12]**
0.85BT−0.15Bi($Mg_{0.5}Zr_{0.5}$)O_3	**280**	**2.9**	**86.8**	**[56]**
0.9$Ba_{0.65}Sr_{0.35}TiO_3$−0.1BMN	**400**	**3.34**	**85.7**	**[57]**
BaTiO ▯ -Bi(Mg ▯.▯ Zr ▯.▯)O ▯	**345**	**3.41**	**85.1**	**[61]**

BaTiO ▯ -Bi(Mg ▯.▯ Zr ▯.▯

)O

?

$BaTiO_3$-$Bi(Mg_{0.5}Zr_{0.5})O_3$@SiO_2				
$0.65(Ba_{0.98}Li_{0.04})Ti_{0.98}O_3$–$0.35(Sr_{0.7}Bi_{0.2})TiO_3$	**410**	**3.54**	**77.0**	**[54]**
0.65BT-0.35(SBT-BMZ)	**370**	**4.03**	**96.2**	**[53]**
$0.6(Ba_{0.75}Sr_{0.25})TiO_3$-$0.4Bi(Mg_{0.5}Hf_{0.5})O_3$	**390**	**4.3**	**92.0**	**[14]**
0.6BT−$0.4Bi(Mg_{0.5}Ti_{0.5})O_3$	**340**	**4.49**	**93.0**	**[16]**
BT−$0.06Bi_{2/3}(Mg_{1/3}Nb_{2/3})O_3$	**520**	**4.55**	**91.0**	**[15]**
$(Ba_{0.65}Sr_{0.245}Bi_{0.07})_{0.99}Nd_{0.01}TiO_3$	**460**	**4.2**	**76.0**	**[55]**
This work	**460**	**5.23**	**89.7**	
	481	**5.47**	**90.6**	

Table S3

Energy storage properties of BT-based ceramics

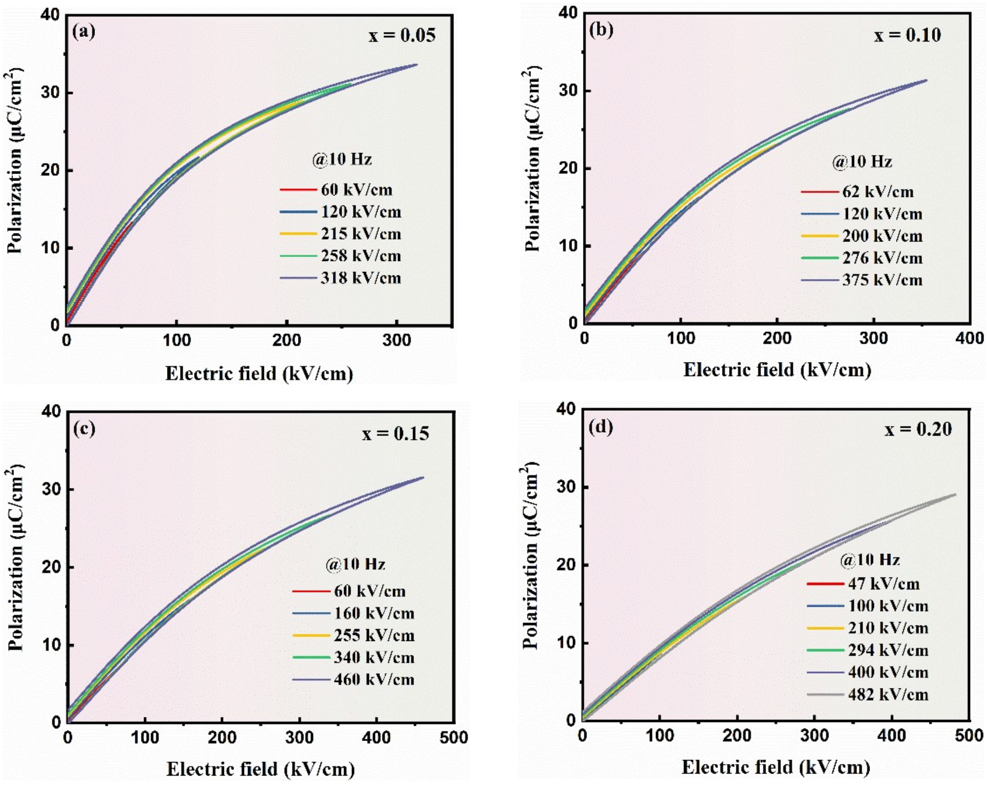

Fig S7 Unipolar P–E loops of the BSBNTH ceramics at different electric fields: (a) x=0.05, (b) x=0.10, (c) x=0.15, (d) x=0.20.

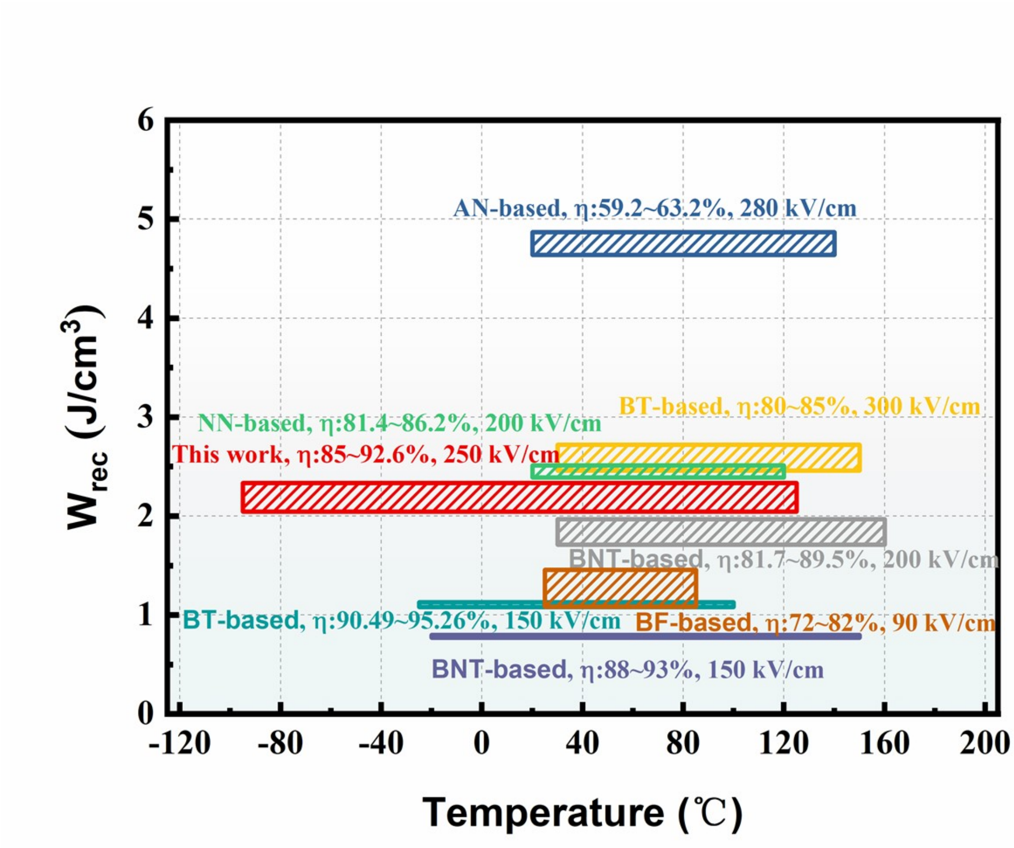

Fig. S8 Temperature-dependent W_{rec} and η of the x = 0.15 sample and recently reported lead-free ceramics (BT: $BaTiO_3$, BF: $BiFeO_3$, BNT: $Bi_{0.5}Na_{0.5}TiO_3$, NN: $NaNbO_3$, and AN: $AgNbO_3$).

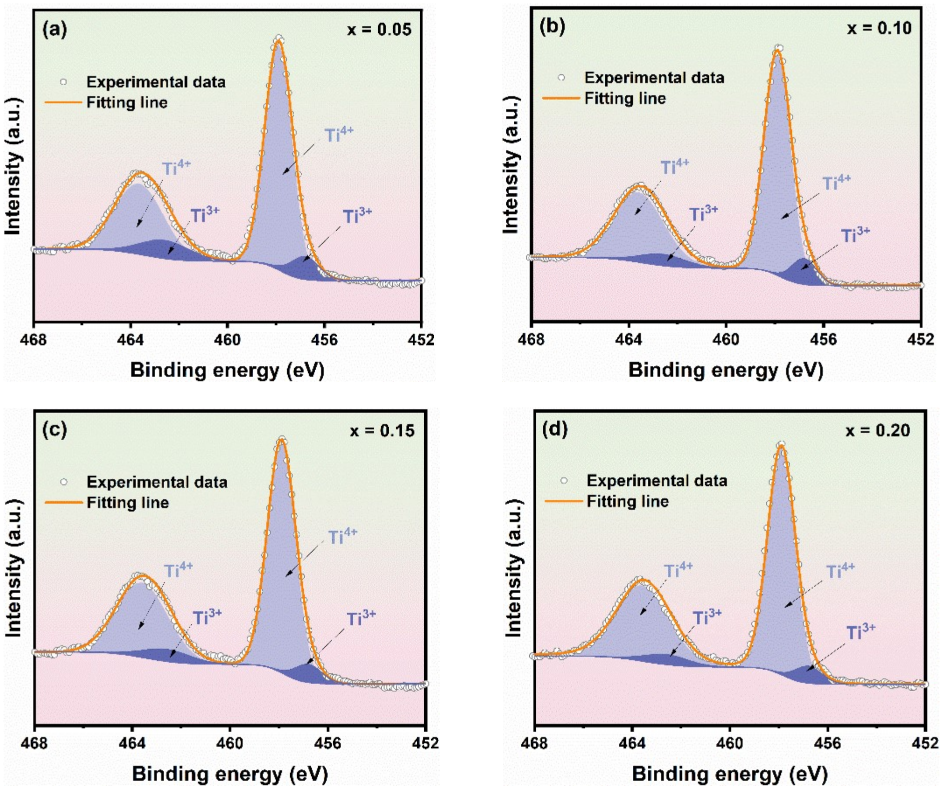

Fig. S9 XPS data of Ti 2p for BSBNTH ceramics: (a) x=0.05, (b) x=0.10, (c) x=0.15, (d) x=0.20.

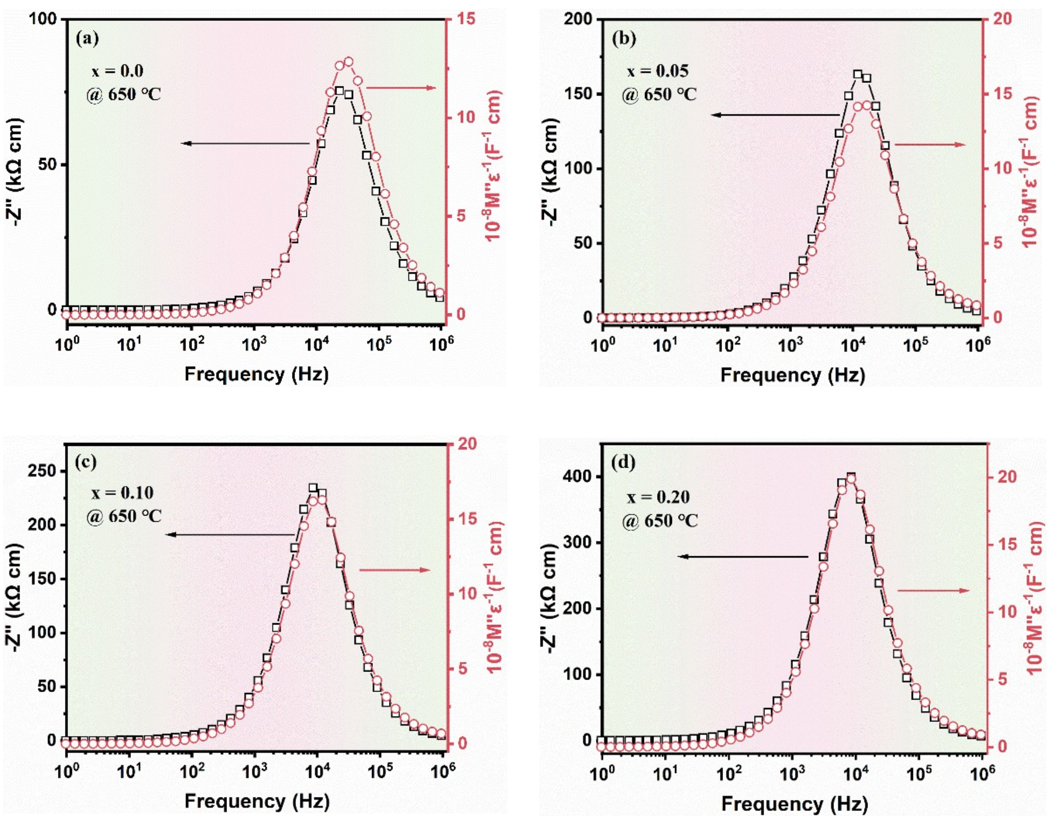

Fig. S10 Combined Z'' and M'' spectroscopic plots for BSBNTH ceramics: (a) x=0, (b) x=0.05, (c) x=0.10, (d) x=0.20.