Supplementary Material

Thermal depolarization and electromechanical hardening in Zn^{2+}-doped $Na_{1/2}Bi_{1/2}TiO_3$-$BaTiO_3$

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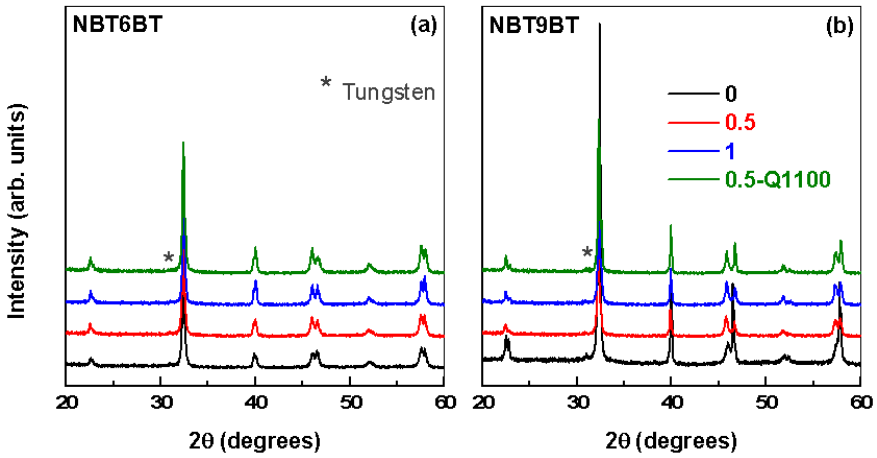

Fig. S1 Laboratory x-ray diffraction profiles of doped (0-1) and 0.5-Q1100 samples of
a) NBT6BT and b) NBT9BT

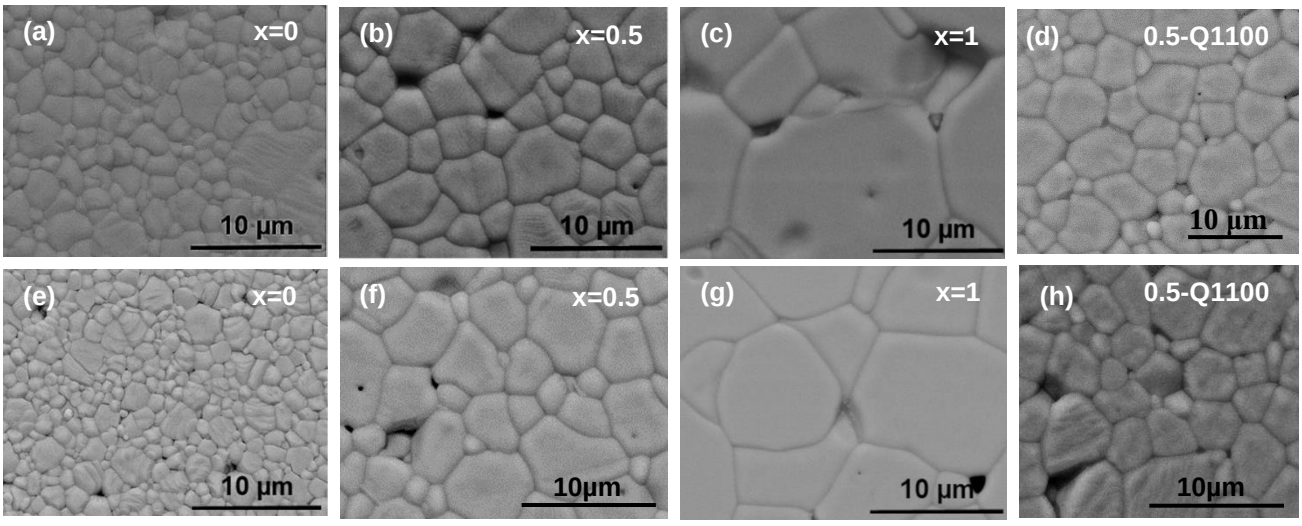

Fig. S2 SEM micrographs of doped (0-1) and 0.5-Q1100 samples of (a-d) NBT6BT and (e-h) NBT9BT

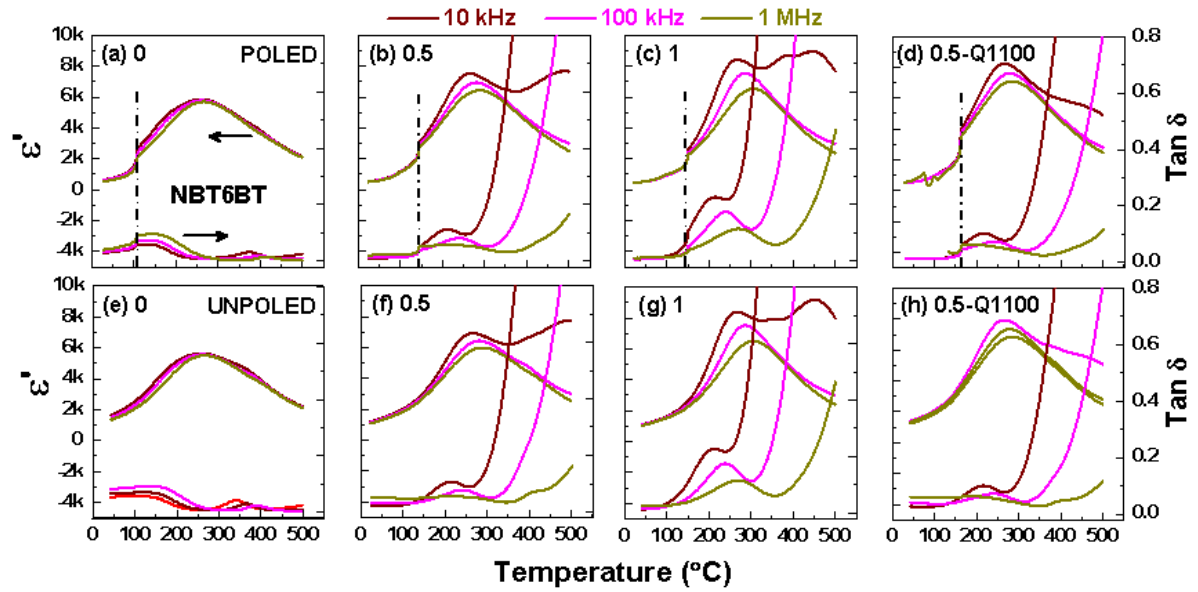

Fig. S3 Temperature- and frequency- dependent permittivity for doped and 0.5-Q1100 NBT6BT samples in the poled (a-d) and unpoled (e-h) state

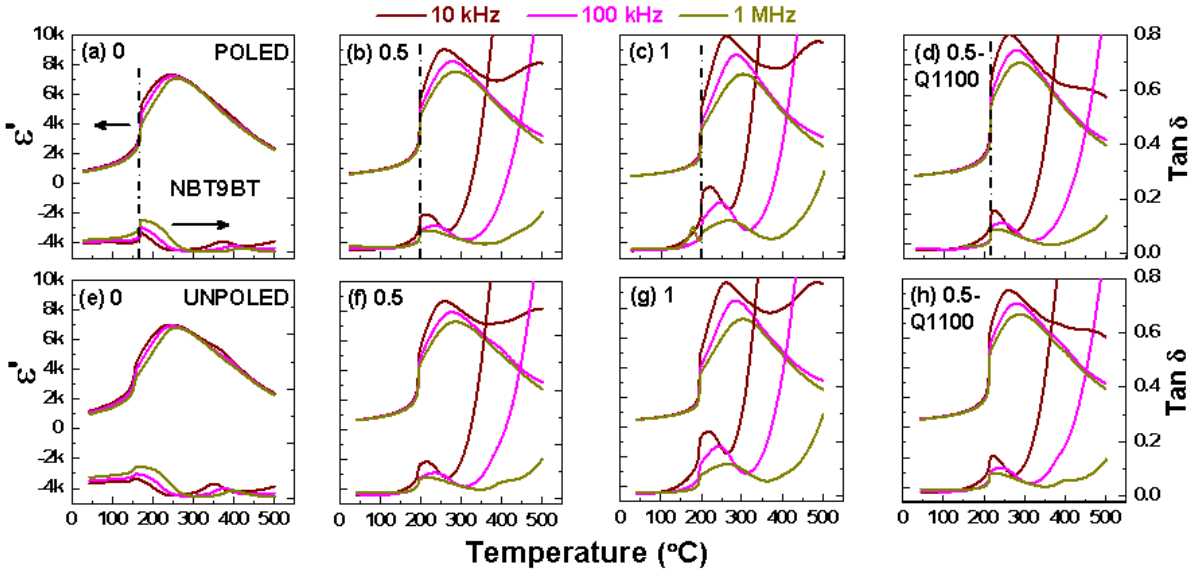

Fig. S4 Temperature- and frequency- dependent permittivity for doped and 0.5-Q1100 NBT9BT samples in the poled (a-d) and unpoled (e-h) state

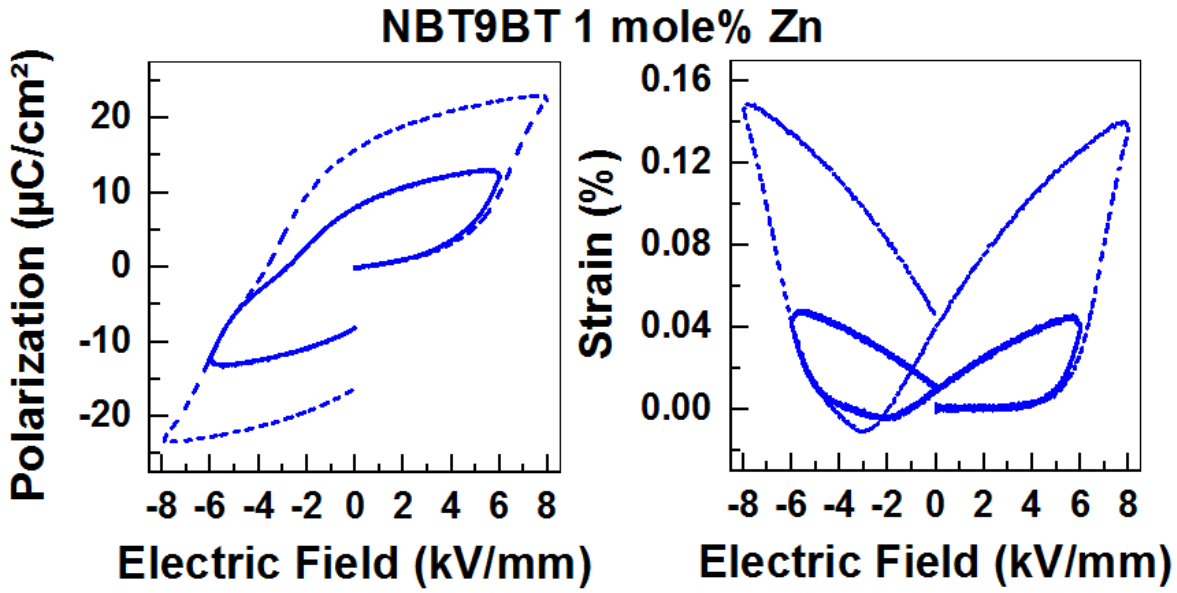

Fig. S5 Polarization- and strain-field hysteresis of 1 mole % Zn^{2+}-doped NBT9BT. The loops are not saturated at 6 kV/mm. The maximum applied field of 8 kV/mm is close to the breakdown strength of the sample.

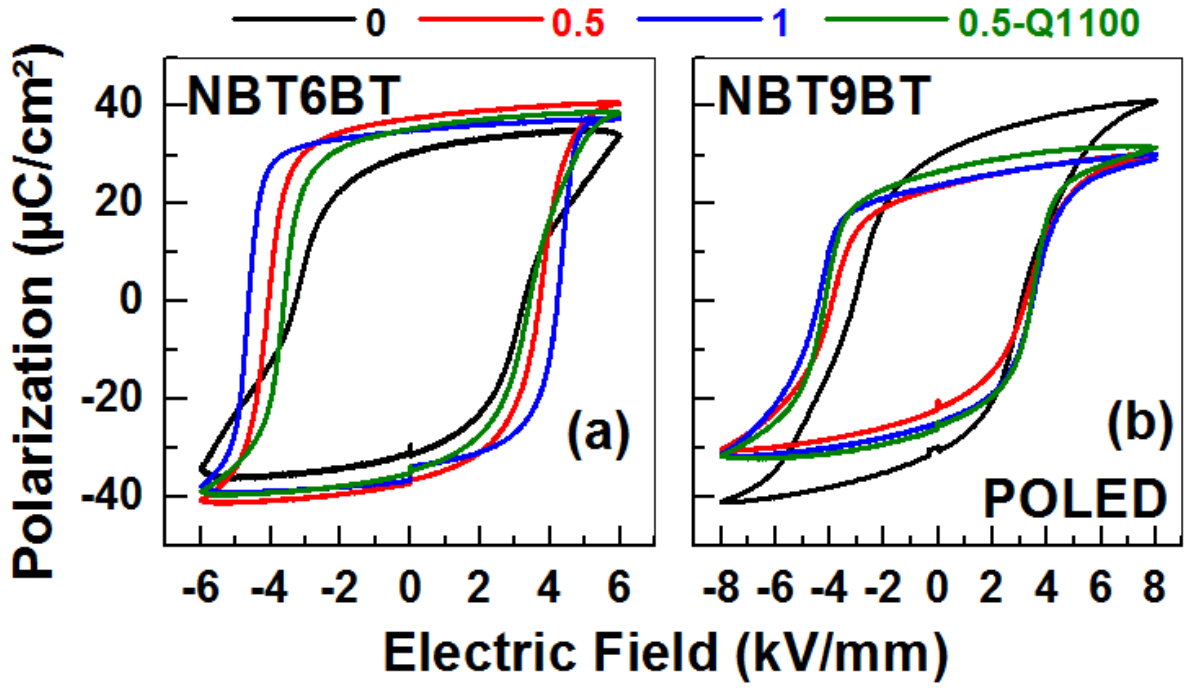

Fig. S6 Polarization-field hysteresis obtained in the poled state.

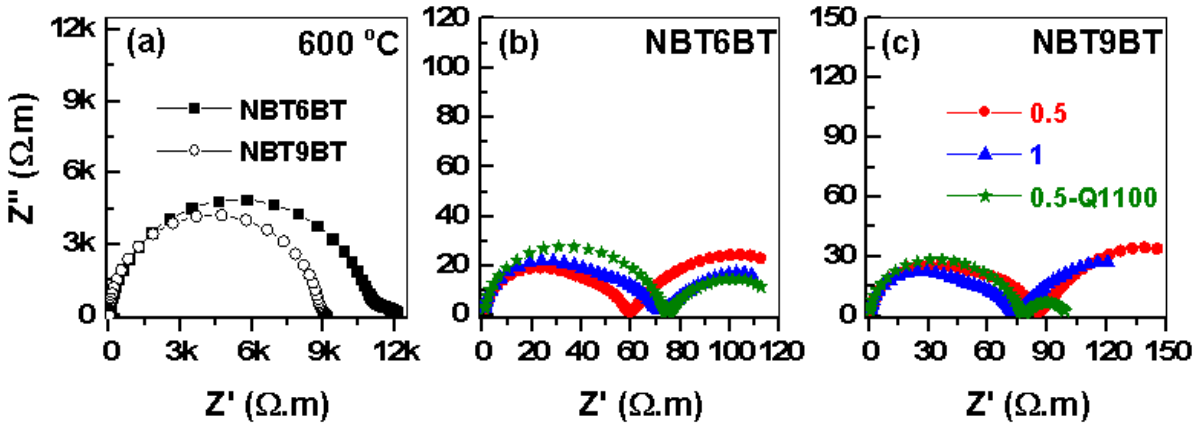

Fig. S7 Nyquist plots of impedance normalized to sample dimensions at 600 °C for (a) undoped, (b-c) doped and 0.5-Q1100 samples

Table S1 Activation energies (E_a) evaluated from the slope of linear fits of the data in Figure 6

Sample	NBT6BT		NBT9BT	
	High E_a (eV)	Low E_a (eV)	High E_a (eV)	Low E_a (eV)
0	1.25	0.72	1.23	1.09
0.5 mole % Zn	0.53	0.79	0.59	0.80
1 mole % Zn	0.61	0.79	0.57	0.82
0.5 mole % Zn – Q1100	0.58	0.85	0.57	0.83

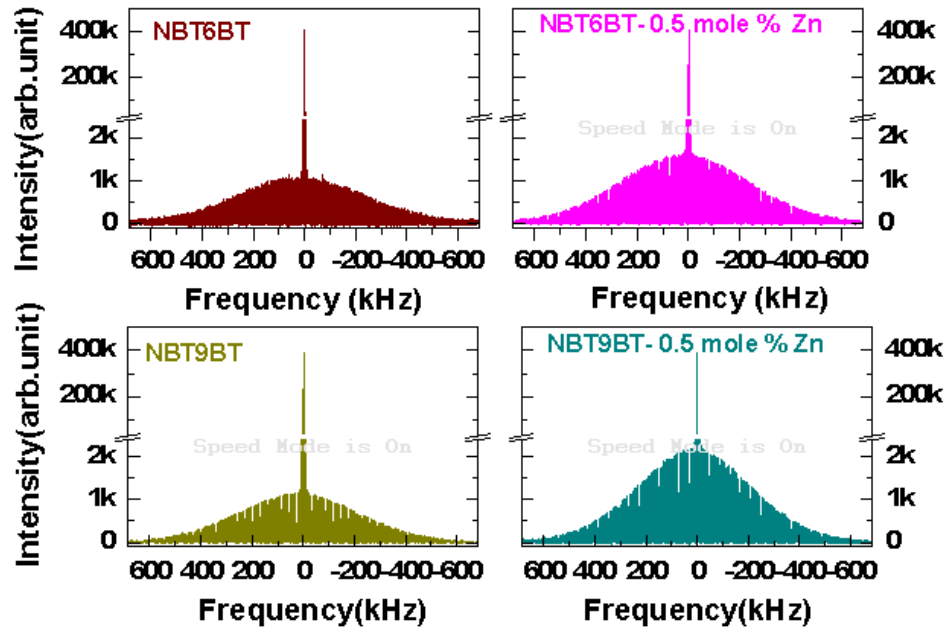

Fig. S8 ^{23}Na MAS NMR spectra of NBT100*y*BT and 0.5 mole % Zn^{2+}-doped NBT100*y*BT samples in the unpoled state normalized to the intensity of the centerband.

Table S2 Refined structural parameters obtained from high resolution x-ray diffraction data using a monochromatized Cu-$K\alpha_1$ radiation (λ = 1.540598 Å, Ge[111]-monochromator) and cubic fraction calculated from the resonance ^{23}Na NMR spectra.

Sample		**NBT6BT**		**NBT6BT-0.5 mole %Zn**	
		Pm$\bar{3}$m	***R3m***	***P4mm***	***R3m***
a, Å		3.8995 (1)	5.4981 (4)	3.8874 (2)	5.5152 (2)
c, Å		3.8995 (1)	6.7936 (7)	3.942 (4)	6.7649 (5)
V, Å^3		59.297 (3)	177.85 (3)	59.570 (4)	178.28 (2)
Unit cell distortion		-	0.620	1.4 %	0.519
Na/Bi/Ba	**x**	0	0	0	0
	y	0	0	0	0
	z	0	0	0	0
Ti/Zn	**x**	0.5	0	0.5	0
	y	0.5	0	0.5	0
	z	0.5	0.510 (5)	0.469 (5)	0.524 (1)
O1	**x**	0.5	0.492 (4)	0.5	0.506 (1)
	y	0.5	0.507 (4)	0.5	0.494 (1)
	z	0	0.084 (4)	-0.11 (2)	0.058 (2)
O2	**x**	-	-	0.5	-
	y			0	
	z			0.629 (5)	
R_p		6.94		6.74	
R_{wp}		9.21		9.06	
R_{exp}		4.62		4.07	
Phase fraction, XRD (%)		68 (3)	32 (3)	33 (4)	67 (4)
Cubic phase fraction, ^{23}Na NMR (%)		27±2		17±2	

Sample		NBT9BT		NBT9BT-0.5 mole %Zn	
		P4mm	***R3m***	***P4mm***	***R3m***
a, Å		3.8927 (1)	5.5269 (3)	3.88960 (7)	5.5283 (2)
c, Å		3.9474 (2)	6.784 (1)	3.9566 (1)	6.787 (1)
V, Å^3		59.816 (4)	179.45 (3)	59.859 (2)	179.64 (3)
Unit cell distortion		1.4 %	0.155	1.7 %	0.105
Na/Bi/Ba	**x**	0	0	0	0
	y	0	0	0	0
	z	0	0	0	0
Ti/Zn	**x**	0.5	0	0.5	0
	y	0.5	0	0.5	0
	z	0.482 (4)	0.519 (2)	0.489 (3)	0.518 (2)
O1	**x**	0.5	0.510 (1)	0.5	0.515 (2)
	y	0.5	0.490 (1)	0.5	0.485 (2)
	z	-0.079 (4)	0.101 (3)	-0.097 (2)	0.093 (3)
O2	**x**	0.5		0.5	
	y	0	-	0	-
	z	0.543 (3)		0.549 (2)	
R_p		5.69		5.58	
R_{wp}		7.46		7.33	
R_{exp}		4.87		3.69	
Phase fraction, XRD (%)		66 (3)	34 (4)	81 (5)	19 (2)
Cubic phase fraction, ^{23}Na NMR (%)		23±2		10±2	