Realizing high energy storage performance in $(Na_{0.47}Bi_{0.47})Ba_{0.06}TiO_3$-based ceramics with a slush-like polar state

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1.1 Experimental procedure

Structural and electrical performance characterization

The phase structure of the synthesized ceramics was characterized by X-ray diffraction (XRD; X'Pert PRO, PANalytical, Almelo, The Netherlands). Rietveld refinement of the XRD data was performed using the GSAS software package. Microstructural analysis was conducted using scanning electron microscopy (FE-SEM, Model S4800, Hitachi, Japan). Domain structures were examined by field-emission transmission electron microscopy (FE-TEM, JEM-2100F, JEOL, Tokyo, Japan). Dielectric properties were measured using a precision impedance analyzer (4294A, Agilent Technologies, Palo Alto, CA). Ferroelectric properties were evaluated with a standardized ferroelectric test system (RT66A, Radiant Technologies, Albuquerque, NM, USA). Raman spectra were acquired in the backscattering mode using a micro-Raman spectrometer (XploRA, Horiba Scientific, France) with a 532 nm laser source. Domain morphology and their behavior under electric field application and removal were further investigated by piezoresponse force microscopy (PFM; MFP-3D, Asylum Research, Santa Barbara, CA, USA). Charge-discharge performance was assessed using a charge and discharge test system (PK-CPR1701, PolyK Technologies, PA, USA).

1.2 The dielectric relaxation behavior:

$$\Delta T_{\mathrm{relaxor}} = T_{\mathrm{s}}(10kHz) - T_{\mathrm{s}}(1MHz) \tag{S1}$$

where T_{s}(10 kHz) and T_{s}(1 MHz) are the values of T_{s} at frequencies of 1 kHz and 1 MHz, respectively, as identified on the dielectric temperature spectrum.

1.3 Curie-Weiss model and Vogel-Fulcher theory:

According to the Curie-Weiss model and Vogel-Fulcher theory, the calculation formula is as follows :

$$\varepsilon_{\mathrm{r}} = \frac{C}{T - T_{CW}} \tag{S2}$$

$$f = f_0 \exp[\frac{-E_a}{k_B(T_m - T_f)}] \tag{S3}$$

where C and T_{CW} are the Curie constant and the Curie-Weiss temperature, respectively; f is the test frequency; T_m is the temperature corresponding to the maximum dielectric constant.; f_0 is the characteristic frequency (dynamic transition frequency/ion attempt jump barrier frequency in the double-well model); E_a is the activation energy.

2. Supplementary Figures

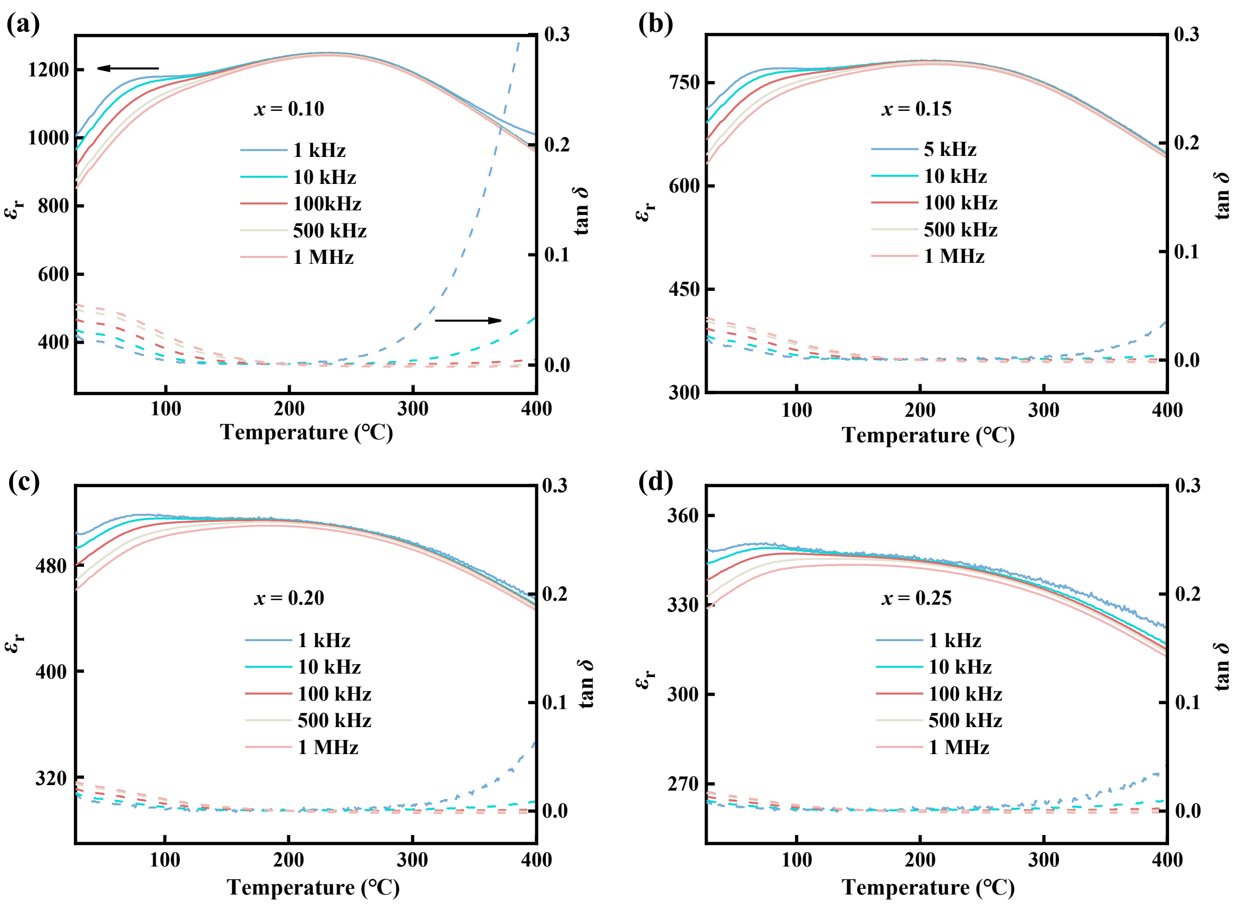

Fig. S1. Temperature-dependent ε_r of xLMN ceramics at various frequencies.

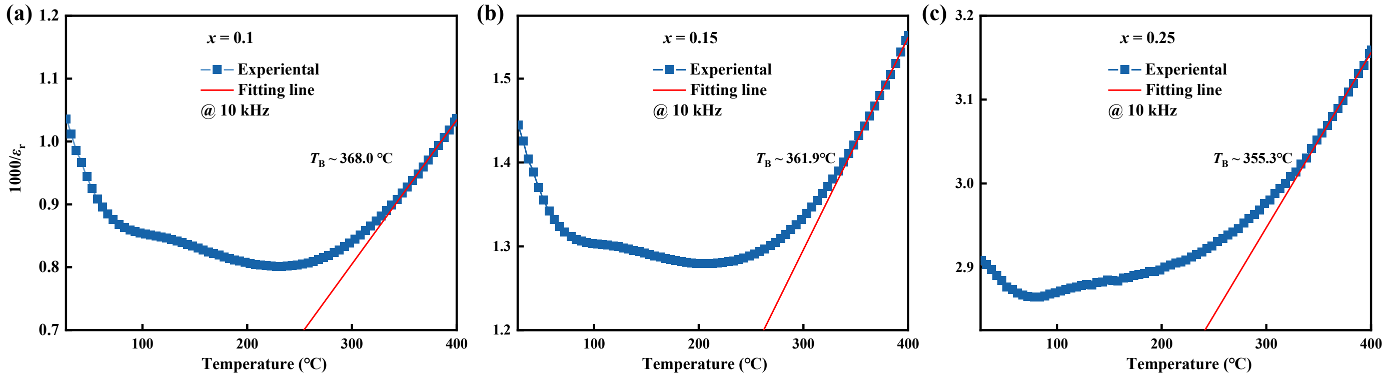

Fig. S2. Temperature-dependent 1000/ε_r of (a) 0.10LMN, (b) 0.15LMN, (c) 0.25LMN ceramics.

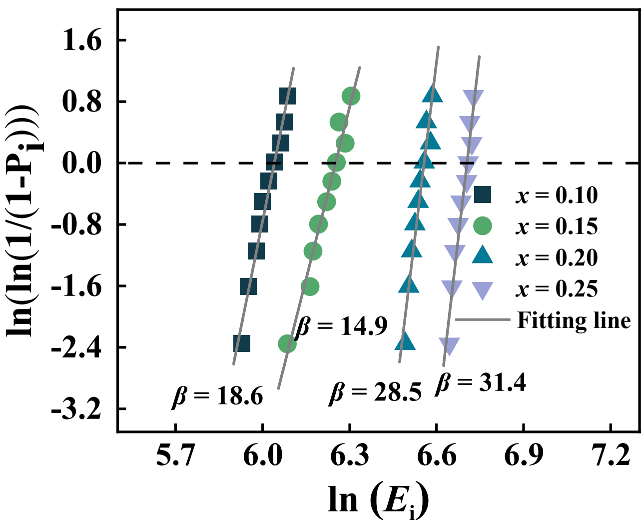

Fig. S3. Weibull distribution function of the breakdown strength for *x*LMN ceramics.

Table S1. Rietveld refinement parameters of the *x*LMN ceramics.

x	Space group	Lattice parameters	*V* (Å^3)	R_{wp} (%)	R_p (%)	χ^2
0.10	*R*3*c* (45.6%)	$a = b = 5.5272(5)$, $c = 13.5401(4)$, $\alpha = \beta = 90^\circ$, $\gamma = 120^\circ$	358.238	3.05	2.29	1.633
	*P*4*bm* (54.4%)	$a = b = 5.5303(3)$, $c = 3.9113(4)$, $\alpha = \beta = \gamma = 90^\circ$	119.627			
0.15	*R*3*c* (35.5%)	$a = b = 5.5316(5)$, $c = 13.5460(7)$, $\alpha = \beta = 90^\circ$, $\gamma = 120^\circ$	358.966	3.46	2.46	2.113
	*P*4*bm* (64.5%)	$a = b = 5.5166(8)$, $c = 3.9031(0)$, $\alpha = \beta = \gamma = 90^\circ$	119.943			
0.20	*R*3*c* (23.6%)	$a = b = 5.5340(5)$, $c = 13.5726(6)$, $\alpha = \beta = 90^\circ$, $\gamma = 120^\circ$	359.983	4.09	2.80	2.814
	*P*4*bm* (76.4%)	$a = b = 5.5429(2)$, $c = 3.9196(7)$, $\alpha = \beta = \gamma = 90^\circ$	120.426			
0.25	*R*3*c* (17.1%)	$a = b = 5.5468(5)$, $c = 13.5869(5)$, $\alpha = \beta = 90^\circ$, $\gamma = 120^\circ$	362.031	4.49	2.88	3.521
	*P*4*bm* (82.9%)	$a = b = 5.5478(4)$, $c = 3.9252(1)$, $\alpha = \beta = \gamma = 90^\circ$	120.812			

Table S2. The V-F fitting parameters of *x*LMN ceramics.

Component	f_0 (Hz)	T_f (℃)	E_a (eV)
x = 0.10	1×10^{11}	29.3	0.026
x = 0.15	5×10^{11}	18.8	0.075
x = 0.20	1×10^{12}	-9.9	0.121
x = 0.25	1×10^{10}	-54.6	0.158