Modification of high-performance sodium niobate energy storage ceramics via introducing a high-entropy phase

Shuai Zou [a], Wenjie Fan [a, b], Lida Tong [a, b], Yaohang Gu [a, b, c, *], Xiaoyan Zhang [a, b, c, *], Xiwei Qi [d, *]

[a] School of Materials Science and Engineering, Northeastern University, Shenyang 110819, China
[b] School of Resources and Materials, Northeastern University at Qinhuangdao, Qinhuangdao 066004, China
[c] Key Laboratory of Dielectric and Electrolyte Functional Materials Hebei Province, Qinhuangdao 066004, China
[d] School of Materials Science and Engineering, Shijiazhuang Tiedao University, Shijiazhuang 050043, China

[*] Corresponding author.
E-mail addresses: guyaohang@neuq.edu.cn (Y. Gu), xyaaa2005@163.com (X. Zhang), qixiwei@mail.neu.edu.cn (X. Qi).

Table S1. Refined structural parameters of (1-*x*)NN-*x*BLTDTMH ceramics.

x	Space group	Fraction (%)	Lattice parameters	V (Å^3)	V_{actual} (Å^3)	R_{wp} (%)	R_p (%)	χ^2
0.00	*Pbcm*	98.62	a = 5.50292 Å b = 5.56823 Å, c = 15.51629 Å, α = β = γ = 90^o	475.443	475.558	11.66	7.15	2.15
	Pbma	1.380	a = 5.59161 Å, b =15.6702 Å, c = 5.52135 Å, α = β = γ = 90^o	483.790				
0.05	*P2$_1$ma*	69.96	a = 5.55831 Å b = 7.81044 Å, c = 5.52075 Å, α = β = γ = 90^o	239.671	308.942	7.80	5.47	1.56
	Pbma	30.04	a = 5.46671 Å, b =15.60442 Å, c = 5.51278 Å, α = β = γ = 90^o	470.269				
0.10	*P2$_1$ma*	57.34	a = 5.55578 Å, b =7.8212 Å, c = 5.50745 Å, α = β = γ = 90^o	239.315	341.124	6.28	4.76	1.32
	Pbma	42.66	a = 5.56991 Å, b =15.55362 Å, c = 5.51722 Å, α = β = γ = 90^o	477.969				
0.15	*P2$_1$ma*	100	a = 5.57376 Å, b =7.83869 Å, c = 5.51386 Å, α = β = γ = 90^o	240.906	240.906	6.46	5.09	1.34

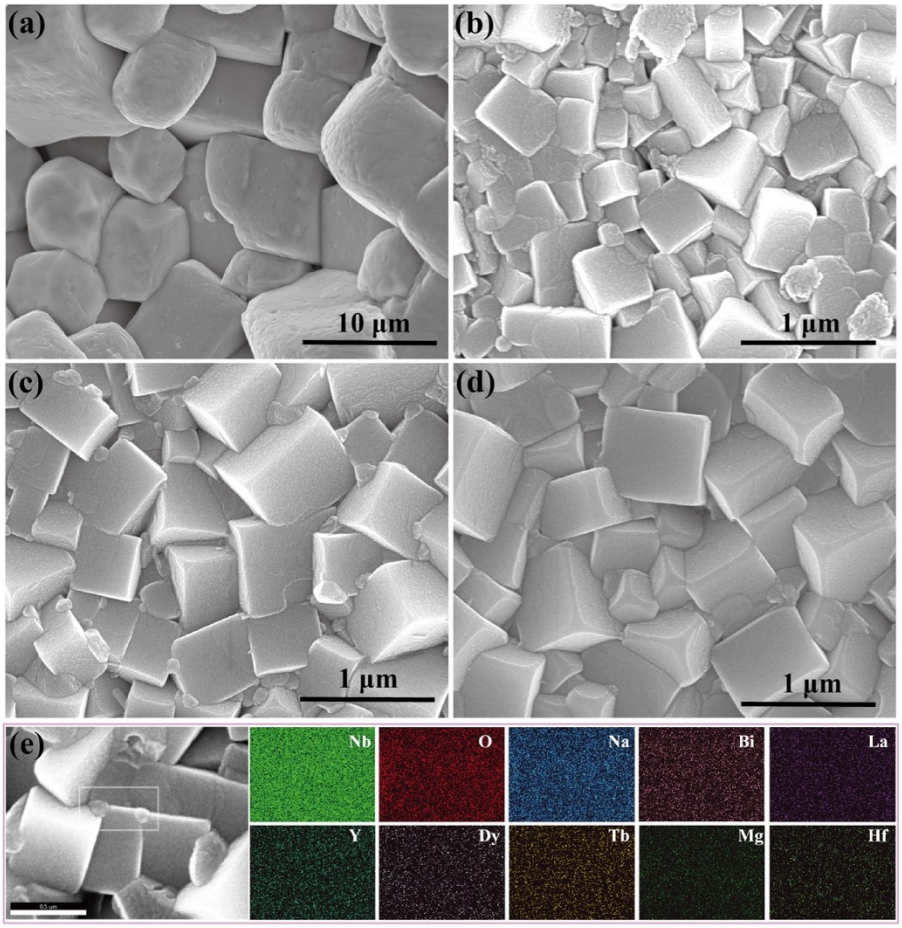

Figure S1. SEM micrographs of (a) $x = 0$; (b) $x = 0.05$; (c) $x = 0.10$; (d) $x = 0.15$; (e) EDS mapping images for $x = 0.10$.

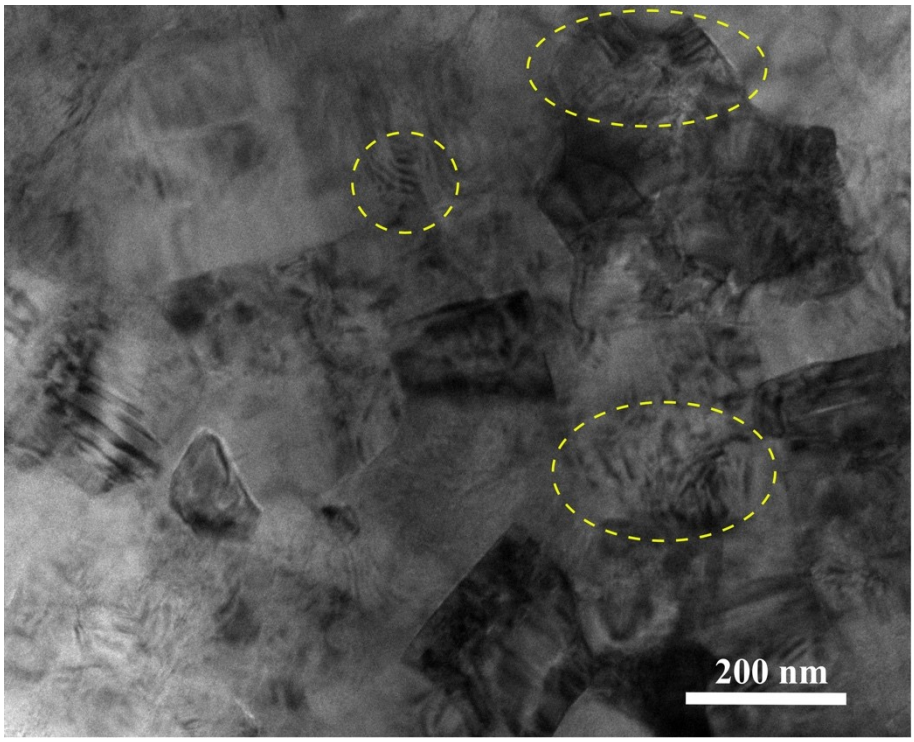

Figure S2. Domain structure of 0.90NN-0.10BLYDTMH.

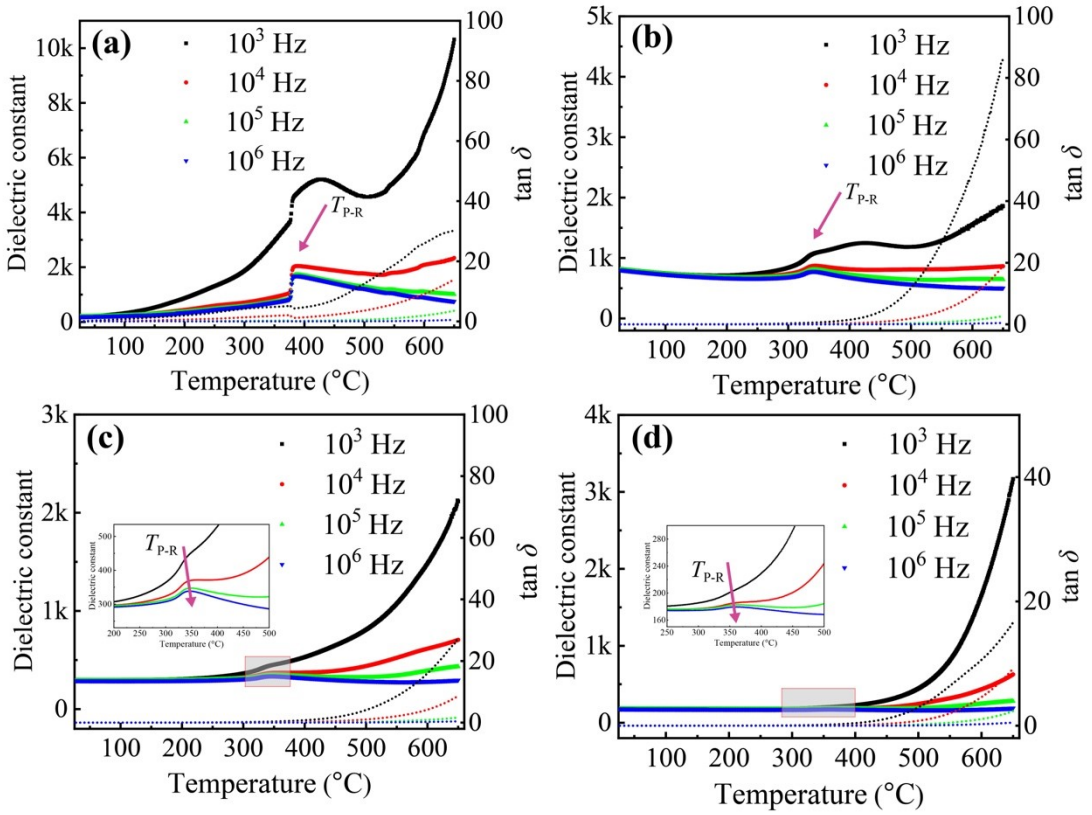

Figure S3. Temperature-dependent dielectric permittivity (ε_r) and dielectric loss (tanδ) at different frequencies (a) x = 0.00; (b) x = 0.05; (c) x = 0.10; (d) x = 0.15.

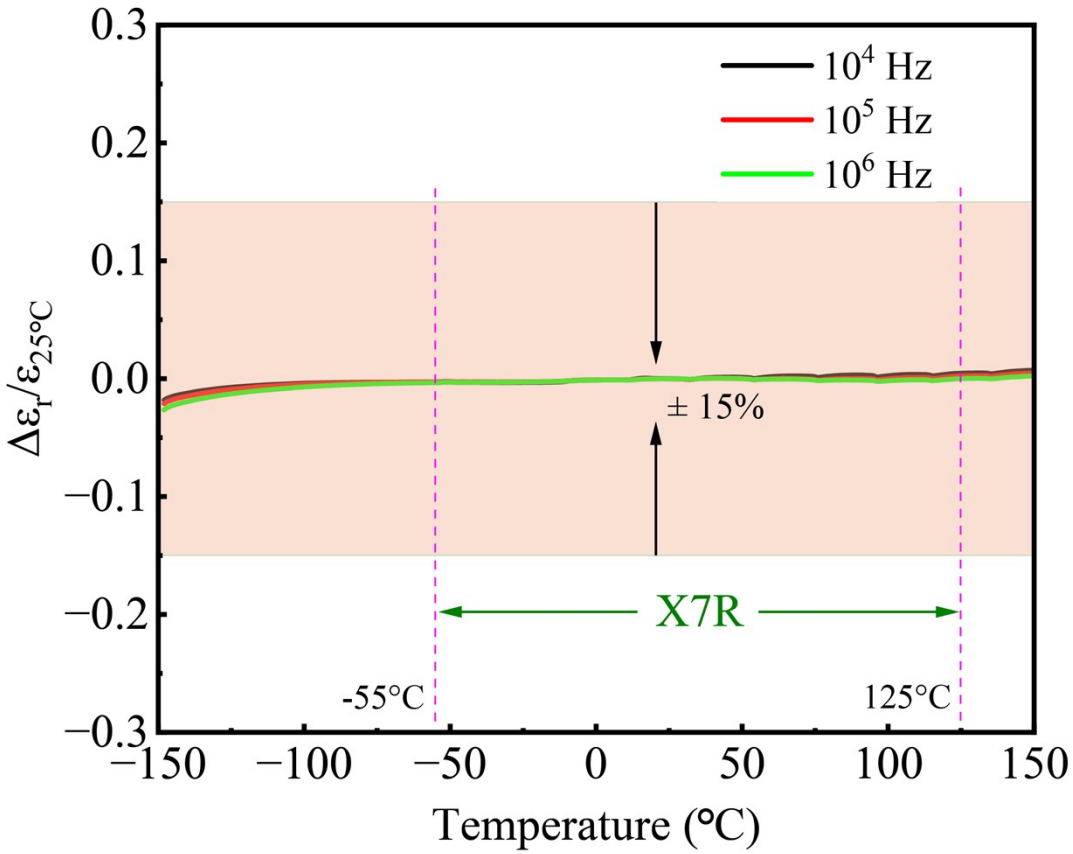

Figure S4. The thermal stability of 0.90NN-0.10BLYDTMH ceramics is measured across the temperature range from -150°C to 150°C at different frequencies.

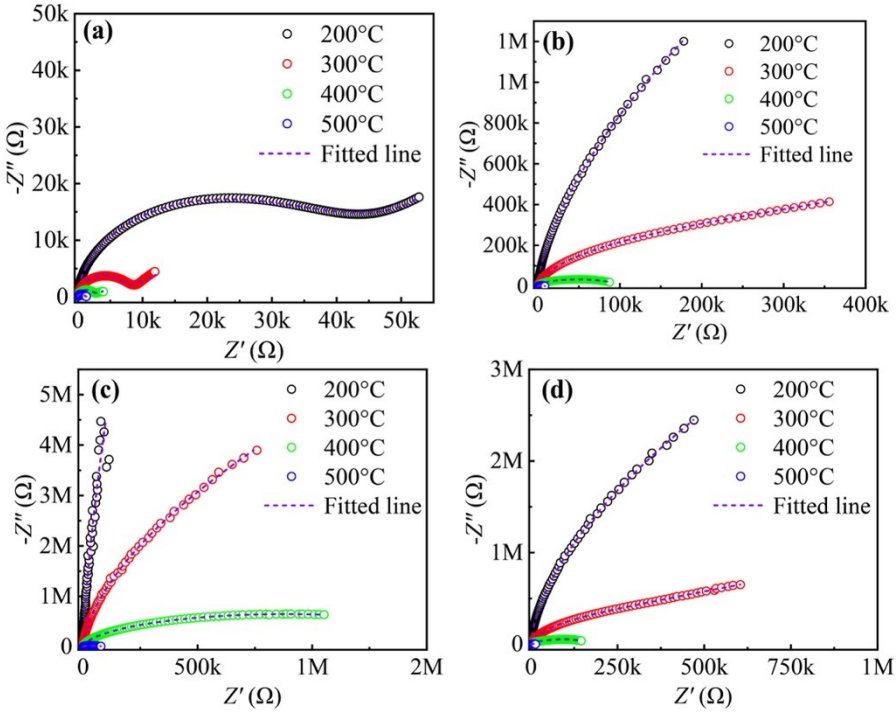

Figure S5. Nyquist plots for all samples of (1-*x*)NN-*x*BLYDTMH at various temperatures: (a) $x = 0.00$; (b) $x = 0.05$; (c) $x = 0.10$ and (d) $x = 0.15$.

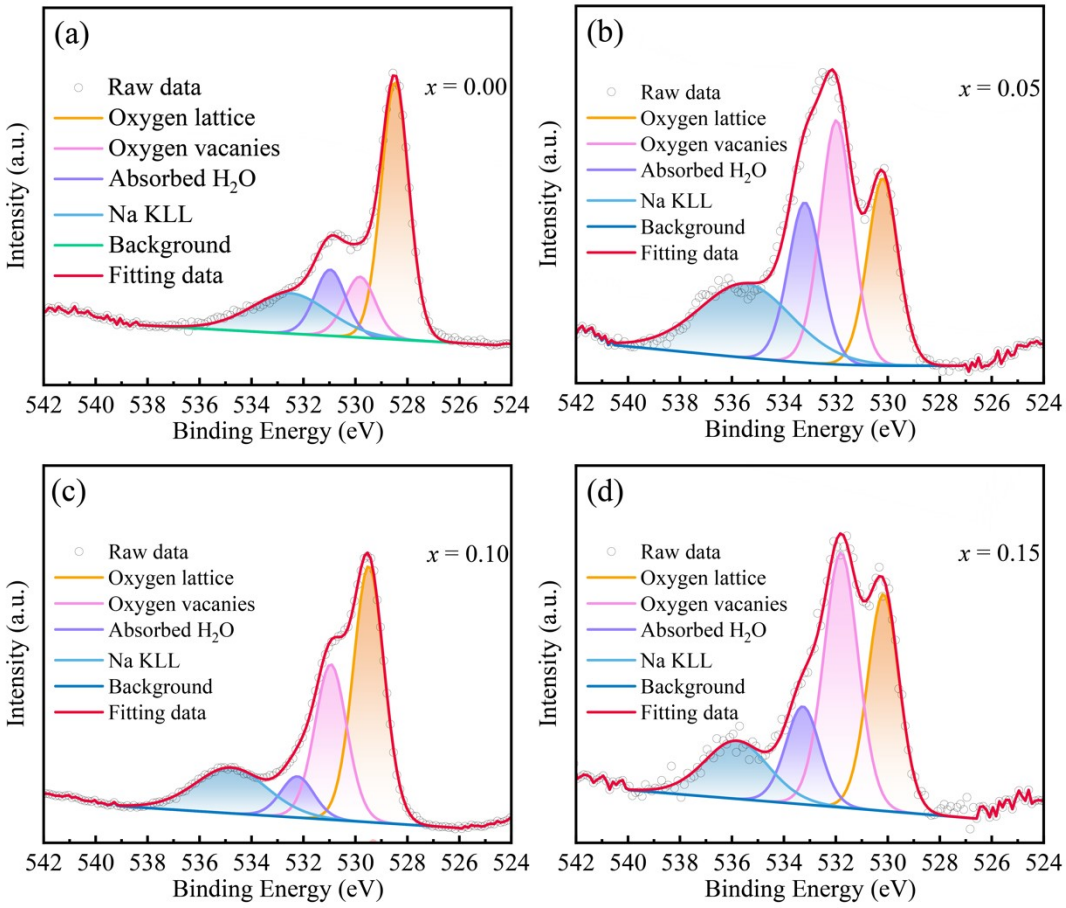

Figure S6. XPS spectra of (1-*x*)NN-*x*BLYDTMH (a) $x = 0.00$; (b) $x = 0.05$; (c) $x = 0.10$; (d) $x = 0.15$.

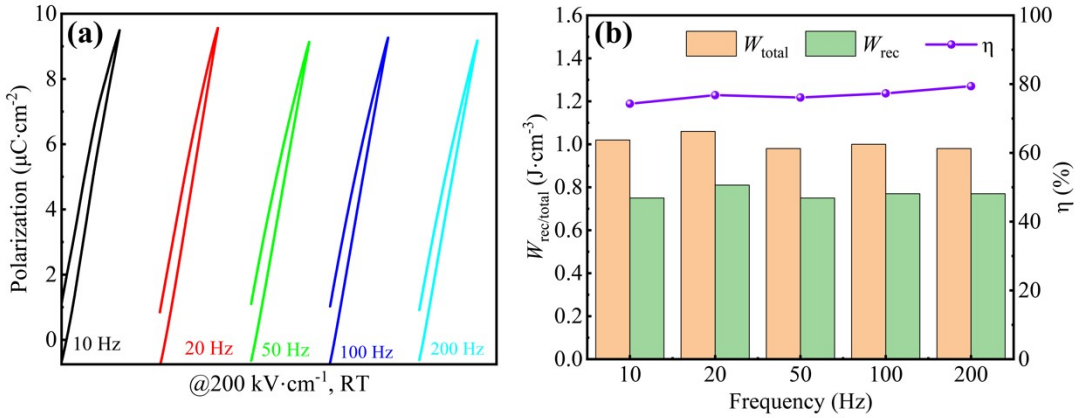

Figure S7. (a) Unipolar *P*–*E* loops at 10-200 Hz. (b) Variations of W_{total}, W_{rec}, and η within the frequency range of 10 to 200 Hz.

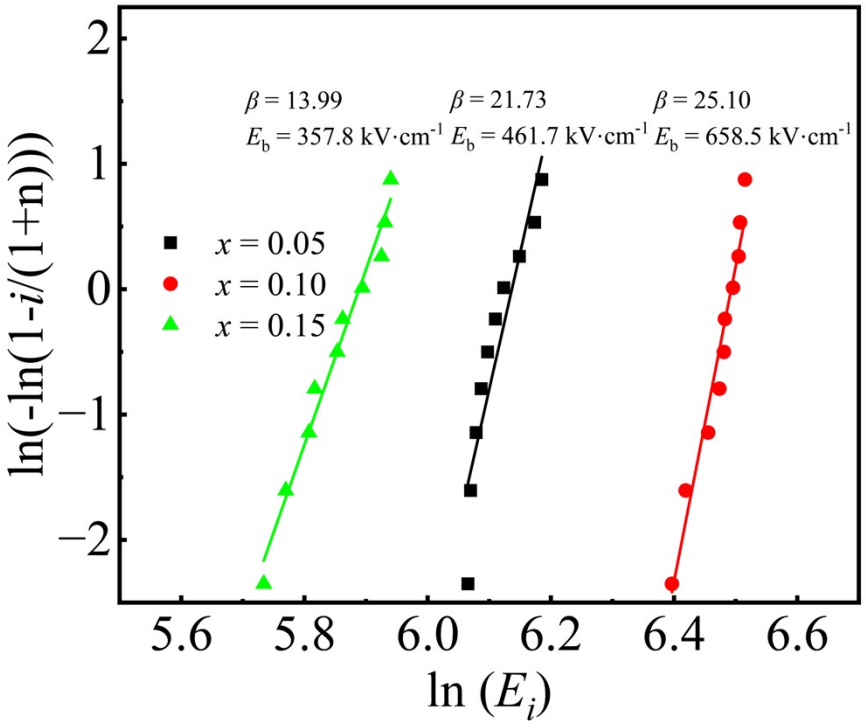

Figure S8. Weibull distribution analyses for evaluating the E_b of (1-x)NN-xNLYDTMH ceramics.

Table S2. Summary of energy-storage properties obtained from recently reported NN-based bulk ceramics.

Composition	P_{max} μC·cm^{-2}	BDS kV·cm^{-1}	W_{rec} J·cm^{-3}	η %	Ref.
$0.74NaNbO_3$-$0.26Sr(Mg_{1/3}Nb_{2/3})O_3$	~25	453	4.28	89.3	[1]
$0.89NaNbO_3$-$0.11(Bi_{0.5}K_{0.5})ZrO_3$	~43	~400	4.4	~41	[2]
$0.85(0.96NaNbO_3$-$0.04Bi(Li_{1/3}Hf_{2/3})O_3)$-$0.15Sr(Sc_{0.5}Nb_{0.5})O_3$	~23	~890	7.05	92.2	[3]
$0.8(0.7AgNbO_3$-$0.3NaNbO_3$-$0.2Bi(Sr_{0.7}Bi_{0.2})O_3$	48.9	478	7.53	74	[4]
$0.825NaNbO3$-$0.1BaTiO_3$-$0.075BiFeO_3$	45	300	~5.0	~82.1	[5]
$0.825NaNbO_3$-$0.175BaZrO_3$	~32	450	~5	85	[6]
$0.85NaNbO3$-$0.15Bi(Mg_{0.2}Hf_{0.2}Ni_{0.2}Zn_{0.2}Ta_{0.2})O_3$	~40	200	5.11	77	[7]
$0.68NaNbO_3$-$0.32(Bi_{0.5}Li_{0.5})(Zr_{0.04}Ti_{0.96})O_3$	~40	530	8.5	90	[8]
$0.90(0.80NaNbO_3$-$0.20Sr_{0.7}Bi_{0.2}TiO_3)$-$0.10BaSnO_3$	~30	531	5.82	86.7	[9]
$0.9NaNbO_3$-$0.1La(Mg_{0.5}Zr_{0.5})O_3$	~22	575	7.0	85.9	[10]
$0.95NaNbO_3$-$0.05La(Mn_{0.5}Ni_{0.5})O_3$	~30	570	6.24	85	[11]
$0.92NaNbO_3$-$0.08Bi_{2/3}HfO_3$	~30	250	4.26	~50	[12]
$0.83NaNbO_3$-$0.12Bi(Mg_{2/3}Nb_{1/3})O_3$-$0.05CaZrO_3$	~23.5	640	5.9	85	[13]
$0.82NaNbO_3$-$0.03NaSbO_3$-$0.15(Na_{0.5}La_{0.5})TiO_3$	~35	470	6.05	80.5	[14]
$0.94(Na_{0.88}Sm_{0.04})NbO_3$-$0.06BiFeO_3$	~28	460	4.0	80	[15]
$0.88NaNbO_3$-$0.12BiFe_{1/3}Zn_{1/3}Ti_{1/3})O_3$	~40	490	6.8	82	[16]
$0.87NaNbO_3$-$0.13Sr(Mg_{1/3}Sb_{2/3})\ O_3$	~27	640	6.17	76.72	[17]
$0.9NaNbO_3$-$0.1Bi(Zn_{0.5}Sn_{0.5})O_3$	~19	350	3.14	83.3	[18]
$0.68NaNbO_3$-$0.32Bi_{0.5}Li_{0.5}TiO_3$	~47.4	480	8.73	80.1	[19]
0.90 $NaNbO_3$-$0.10(Bi_{0.2}La_{0.2}Y_{0.2}Dy_{0.2}Tb_{0.2})(Mg_{0.5}Hf_{0.5})O_3$	32.1	670	6.09	52	This work

References

1. Q. Dong, X. Dong, Y. Pan, X. Wang, J. Wang, P. Nong, M. Xu, D. Zeng, L. Deng, H. Zhang, H. Zhou and X. Chen, *Ceram. Int.*, 2023, **49**, 8081-8087.
2. A. Xie, J. Fu and R. Zuo, *J. Materiomics*, 2022, **8**, 618-626.
3. L. Lv, Z. Pan, J. Hu, Z. Zhou, J. Liu, P. Li, H. Xie and J. Zhai, *Chem. Eng. J.*, 2024, **498**.
4. Z. Hu, Z. Liu, B. Han, H. Peng, K. Dai, Z. Xu, Z. Fu, Z. Hu and G. Wang, *J. Adv. Ceram.*, 2024, **13**, 1282-1290.
5. A. Xie, T. Hu, J. Lei, Y. Zhang, X. Wei, Z. Fu and R. Zuo, *Small*, 2024, **20**, e2309796.
6. J. Zuo, A. Xie, J. Liu, Y. Zhang and R. Zuo, *Ceram. Int.*, 2024, **50**, 6332-6339.
7. R. Zhao, K. Wang, W. Li, Y. Yuan, X. Tang, J. Hu, H. Fan, K. Chen, W. Song, Y. Jiang and X. Guo, *Chem. Eng. J.*, 2024, **495**.
8. A. Xie, J. Lei, Y. Zhang, A. Rahman, X. Jiang, T. Li, X. Xie, L. Liu, C. Zhou, S. Yin, H. Ma, X. Fang and R. Zuo, *J. Mater. Chem. A*, 2024, **12**, 9124-9131.
9. Z. Wang, D. Li, W. Liu, X. Liang, W. Zhao, J. Liu, J. Ren, T. Zhou, D. Xu, W. Liu and D. Zhou, *J. Mater. Chem. A*, 2024, **12**, 19551-19558.
10. J. Huang, M. Xu, Y. Pan, Y. Huang, H. Zhou, X. Li and X. Chen, *J. Mater. Chem. C*, 2024, DOI: 10.1039/d4tc03151b.
11. Q. Dong, Y. Pan, X. Chen, X. Li and H. Zhou, *J. Energy Storage*, 2024, **101**.
12. L. Zhang, Y. Pu, M. Chen, Y. Shi, J. Shang, Y. Yang and D. Zhao, *Chem. Eng. J.*, 2023, **457**.
13. L. Zhang, Z. Chen, G. Luo, F. Ai and N. Luo, *J. Eur. Ceram. Soc.*, 2023, **43**, 6077-6083.
14. A. Xie, J. Chen, J. Zuo, J. Liu, T. Li, X. Jiang and R. Zuo, *ACS Appl. Mater. Interfaces*, 2023, **15**, 22301-22309.
15. X. Meng, Z. Yang, Y. Yuan, B. Tang and S. Zhang, *Chem. Eng. J.*, 2023, **477**.
16. Y. Lu, J. Zhang, K. Zhou and L. He, *J. Mater. Sci.*, 2023, **58**, 11886-11893.
17. Q. Dong, P. Nong, Y. Pan, D. Zeng, M. Xu, H. Zhou, X. Li and X. Chen, *J. Mater. Chem. C*, 2023, **11**, 13120-13128.
18. X. Dong, X. Li, H. Chen, Q. Dong, J. Wang, X. Wang, Y. Pan, X. Chen and H. Zhou, *J. Adv. Ceram.*, 2022, **11**, 729-741.
19. A. Xie, R. Zuo, Z. Qiao, Z. Fu, T. Hu and L. Fei, *Adv. Energy Mater.*, 2021, **11**.