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

Realizing High Comprehensive Energy Storage Performance in Lead-Free Bulk Ceramics via Designing Unmatched Temperature Range

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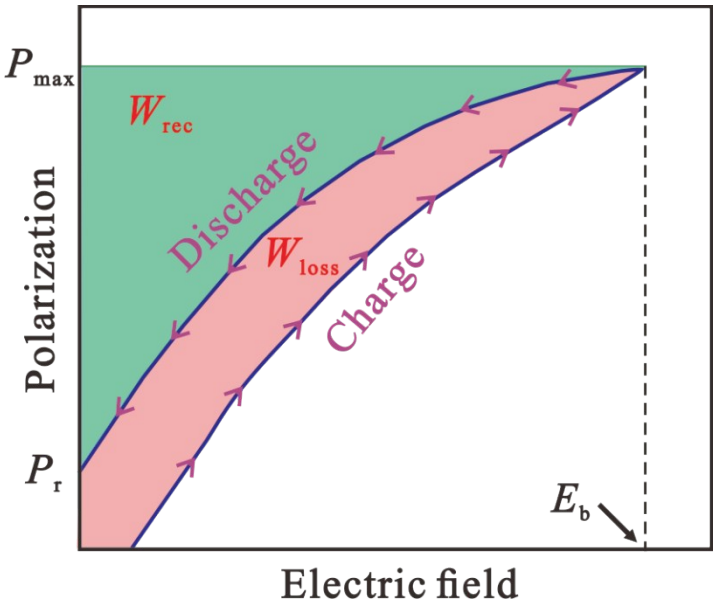

Fig. S1. Schematic diagram of calculating the energy storage properties of dielectric ceramics.

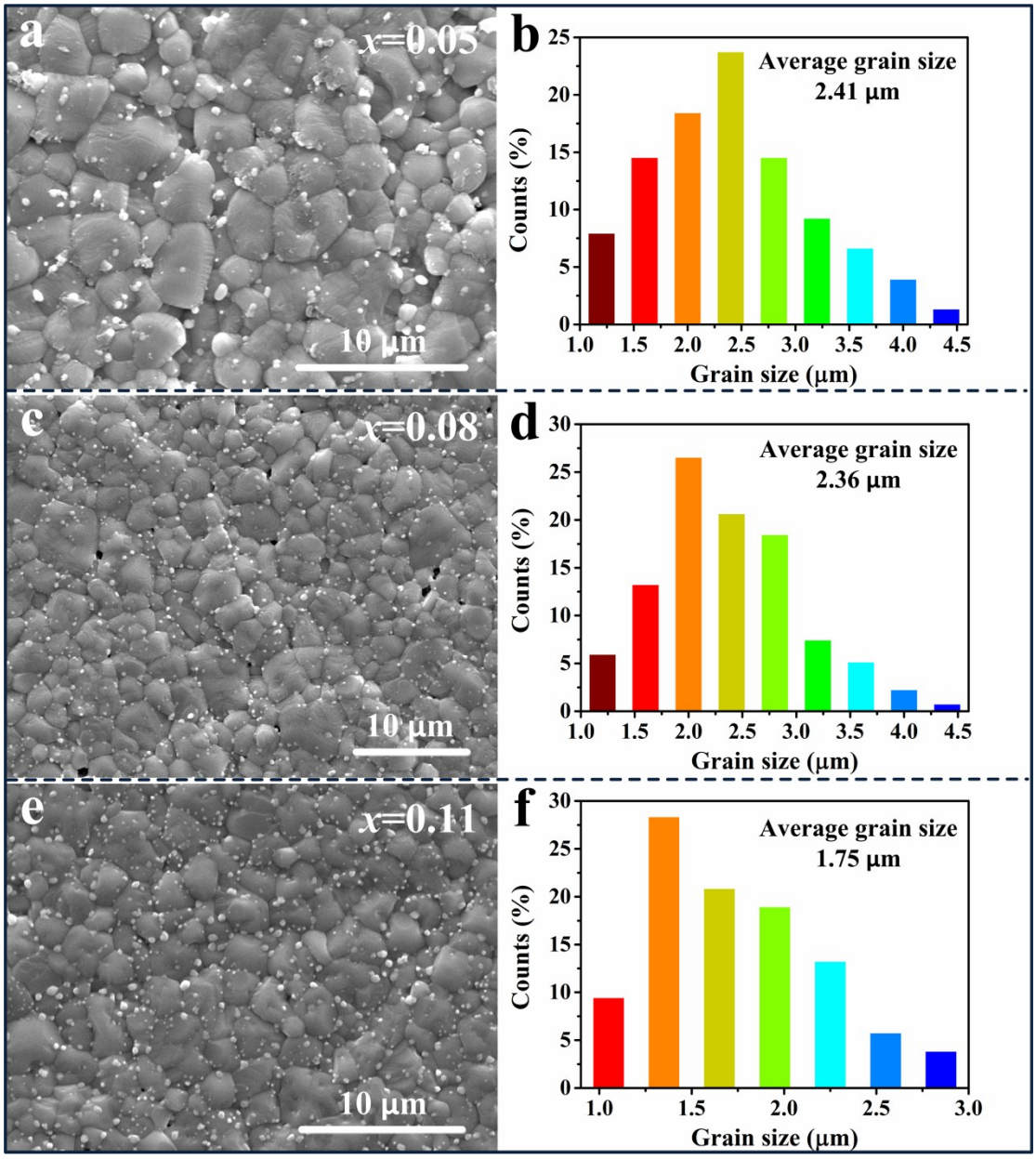

Fig. S2. SEM images of the thermally etched surfaces of (1-*x*)NN-*x*BNH ceramics with (a) *x*=0.05, (c) *x*=0.0.8 and (e) *x*=0.11. Grain size distribution and average grain size of (1-*x*)NN-*x*BNH ceramics with (b) *x*=0.05, (d) *x*=0.0.8 and (f) *x*=0.11.

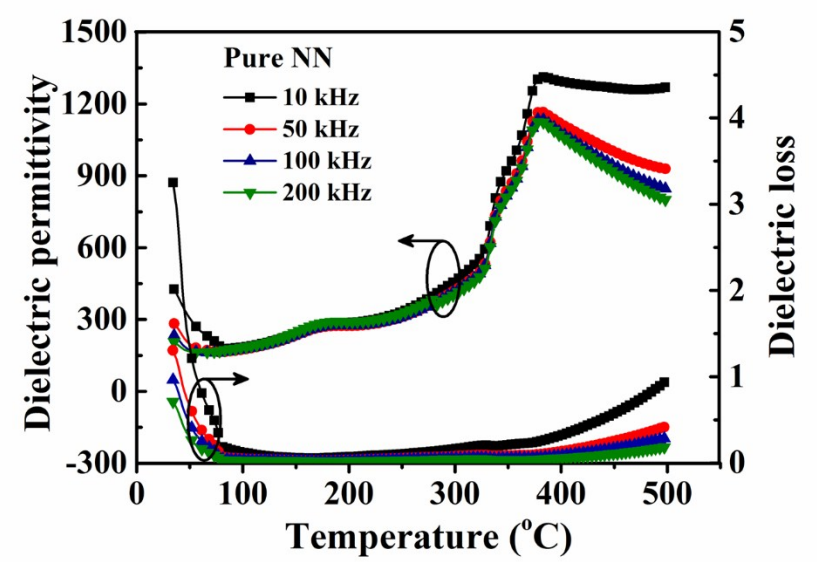

Fig. S3. Temperature dependence of dielectric permittivity and loss of pure NN ceramics from 20 ºC to 500 ºC.

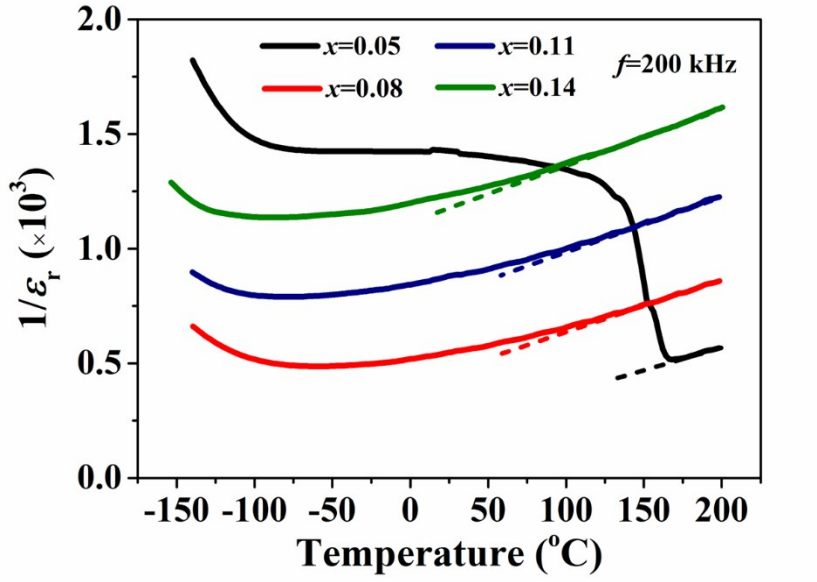

Fig. S4. Temperature dependence of the reciprocal of dielectric permittivity to determine the T_B values of (1-*x*)NN-*x*BNH ceramics, from which the dielectric responses start to follow Curie-Weiss law.

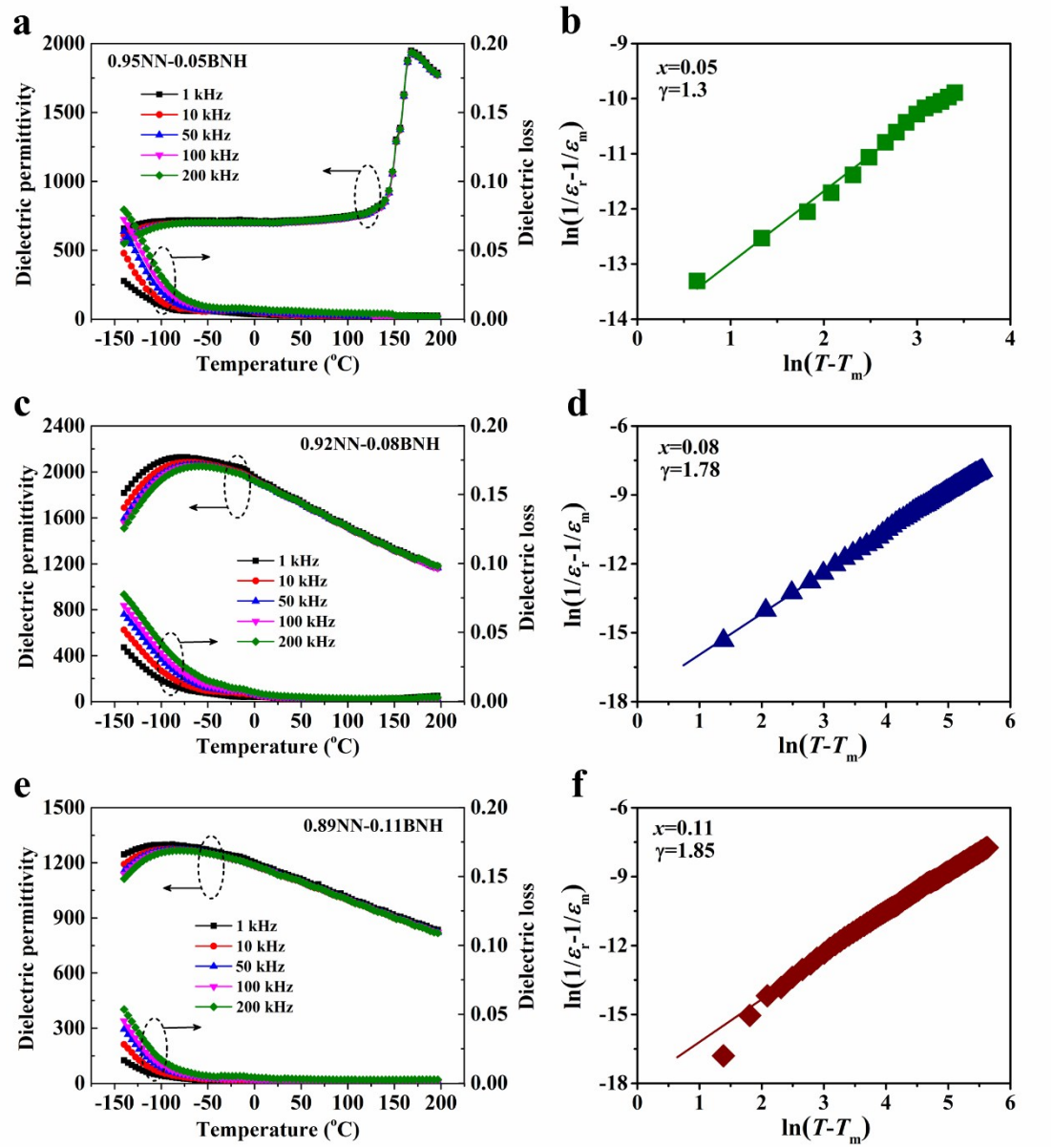

Fig. S5. Temperature dependence of dielectric permittivity and loss of (1-*x*)NN-*x*BNH ceramics from -150 ºC to 200 ºC with (a) *x*=0.05, (c) *x*=0.08 and (e) *x*=0.11. $\ln(T-T_m)$ as a function of $\ln(1/\varepsilon_r-1/\varepsilon_m)$ for (1-*x*)NN-*x*BNH ceramics at 200 kHz with (b) *x*=0.05, (d) *x*=0.08 and (f) *x*=0.11.

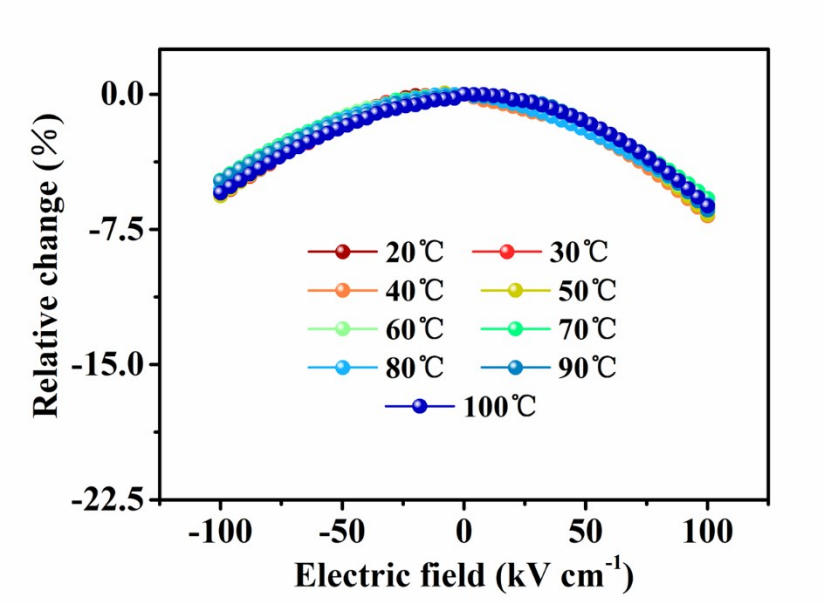

Fig. S6. Relative change of dielectric permittivity as a function of bias electric field for 0.86NN-0.14BNH ceramics from 20 ℃ to 100 ℃.

Table S1. The energy storage performance of lead-free bulk ceramics.

Materials	W_{rec} (J cm^{-3})	η (%)	Ref.
$0.95(0.8Bi_{0.5}Na_{0.5}TiO_3\text{-}0.2SrTiO_3)\text{-}0.05NaNbO_3$	0.73	55	1
$0.852Na_{1/2}Bi_{1/2}TiO_3\text{-}0.028BaTiO_3\text{-}0.12K_{1/2}Bi_{1/2}TiO_3$	0.83	87	2
$0.7[0.94(Na_{1/2}Bi_{1/2}TiO_3)\text{-}0.06(BaTiO_3)]\text{-}0.3SrTiO_3$+11mol% MnO	1.06	68	3
$0.62Bi_{0.5}Na_{0.5}TiO_3\text{-}0.06BaTiO_3\text{-}0.32(Sr_{0.7}Bi_{0.2})TiO_3$+MnO	1.07	92	4
$0.8(0.775Na_{0.5}Bi_{0.5}TiO_3\text{-}0.225BaSnO_3)\text{-}0.2BaZrO_3$	2.08	88.8	5
$\{Bi_{0.5}[(Na_{0.8}K_{0.2})_{0.90}Li_{0.10}]_{0.5}\}_{0.96}Sr_{0.04}(Ti_{0.975}Ta_{0.025})O_3$	2.42	64.3	6
$(Na_{0.5}Bi_{0.5})_{0.8}Ba_{0.2}Ti_{0.8}Sn_{0.2}O_3$	2.347	71	7
$0.85(0.65Bi_{0.5}Na_{0.5}TiO_3\text{-}0.35Bi_{0.1}Sr_{0.85}TiO_3)\text{-}0.15KNbO_3$	1.32	82.5	8
$0.9Bi_{0.48}La_{0.02}Na_{0.48}Li_{0.02}Ti_{0.98}Zr_{0.02}O_3\text{-}0.1Na_{0.73}Bi_{0.09}NbO_3$	2.04	55	9
$0.9(0.92Bi_{0.5}Na_{0.5}TiO_3\text{-}0.08BaTiO_3)\text{-}0.1NaNbO_3$	0.71	60	10
$0.8(0.92Bi_{0.5}Na_{0.5}TiO_3\text{-}0.08BaTiO_3)\text{-}0.2Na_{0.73}Bi_{0.09}NbO_3$	1.36	73.9	11
$0.475Bi_{0.5}Na_{0.5}TiO_3\text{-}0.525Ba_{0.85}Ca_{0.15}Ti_{0.9}Zr_{0.1}O_3$+5wt% MgO	1.04	70	12
$0.7[0.94Bi_{0.5}Na_{0.5}TiO_3\text{-}0.06BaTiO_3]\text{-}0.3SrTiO_3$	0.98	82	13
$0.65Bi_{0.5}Na_{0.5}TiO_3\text{-}0.3Sr_{0.7}Bi_{0.2}TiO_3\text{-}0.05LaTi_{0.5}Mg_{0.5}O_3$	1.32	75	14
$0.9(Na_{1/2}Bi_{1/2})_{0.92}Ba_{0.08}TiO_3\text{-}0.1Bi(Mg_{1/2}Ti_{1/2})O_3$	2	88	15
$0.9(Bi_{0.5}Na_{0.5})TiO_3\text{-}0.1KNbO_3$	1.17	82	16
$0.84(Bi_{0.5}Na_{0.5})TiO_3\text{-}0.16(K_{0.5}Na_{0.5})NbO_3$	1.2	71	17
$0.55Bi_{0.5}Na_{0.5}TiO_3\text{-}0.45Ba_{0.85}Ca_{0.15}Ti_{0.85}Zr_{0.1}Sn_{0.05}O_3$	1.21	72.8	18
$0.7(Na_{0.5}Bi_{0.5})TiO_3\text{-}0.3SrTiO_3$	0.65	73	19
$0.75Na_{0.5}Bi_{0.5}TiO_3\text{-}0.25BaSnO_3$	1.91	86	20
$0.4(Na_{0.5}Bi_{0.5})TiO_3\text{-}0.225BaTiO_3\text{-}0.375BiFeO_3$	1.4	90	21
$0.95(Na_{0.5}Bi_{0.5})TiO_3\text{-}0.05BaTiO_3$+3mol% $BaO\text{-}B_2O_3\text{-}SiO_2$	0.68	71	22
$0.96(0.8Bi_{0.5}Na_{0.5}TiO_3\text{-}0.2Bi_{0.5}K_{0.5}TiO_3)\text{-}0.04BaZrO_3$	0.73	75	23
$0.82[0.94Bi_{0.5}Na_{0.5}TiO_3\text{-}0.06BaTiO_3]\text{-}0.18K_{0.5}Na_{0.5}NbO_3$+0.4mol% ZnO	1.03	72.7	24
$0.91(0.84Na_{0.5}Bi_{0.5}TiO_3\text{-}0.16K_{0.5}Bi_{0.5}TiO_3)\text{-}0.09K_{0.5}Na_{0.5}NbO_3$	1.51	65	25
$0.84[0.934Na_{0.5}Bi_{0.5}TiO_3\text{-}0.07K_{0.5}Na_{0.5}NbO_3]\text{-}0.16SrTiO_3$	0.59	64	26
$0.85Ba_{0.04}Bi_{0.48}Na_{0.48}TiO_3\text{-}0.15SrZrO_3$	1.32	56	27
$0.475Bi_{0.5}Na_{0.5}TiO_3\text{-}0.525Ba_{0.85}Ca_{0.15}Ti_{0.9}Zr_{0.1}O_3$	1.06	82.37	28
$0.9(Bi_{0.5}Na_{0.5}TiO_3\text{-}BaTiO_3)\text{-}0.1NaTaO_3$	1.2	74.8	29
$Bi_{0.487}Na_{0.427}K_{0.06}Ba_{0.026}TiO_3$+2.2wt% CeO_2	0.94	54	30
$(Na_{0.5}Bi_{0.5})_{0.4}Sr_{0.6})Ti_{0.99}Sn_{0.01}O_3$	2.35	64	31
$0.94(0.65Bi_{0.5}Na_{0.5}TiO_3\text{-}0.35Bi_{0.1}Sr_{0.85}TiO_3)\text{-}0.06(K_{0.5}Na_{0.5}NbO_3)$	2.65	84.6	32
$0.94Bi_{0.5}K_{0.5}TiO_3\text{-}0.06La(Mg_{0.5}Ti_{0.5})O_3$	2.08	68	33
$0.96(0.65Bi_{0.5}Na_{0.5}TiO_3\text{-}0.35Sr_{0.85}Bi_{0.1}TiO_3)\text{-}0.04NaNbO_3$	3.08	81.4	34
$(SrTiO_3+Li_2CO_3)/(0.94Bi_{0.54}Na_{0.46}TiO_3\text{-}0.06BaTiO_3)$	2.41	68	35
$0.5SrTiO_3\text{-}0.5(0.95Bi_{0.5}Na_{0.5}TiO_3\text{-}0.05BaAl_{0.5}Nb_{0.5}O_3)$	1.89	77	36
$0.8SrTiO_3\text{-}0.2(0.93Bi_{0.5}Na_{0.5}TiO_3\text{-}0.07Ba_{0.94}La_{0.04}Zr_{0.02}Ti_{0.98}O_3)$	2.83	85	37
$0.9SrTiO_3\text{-}0.1(Bi_{0.48}La_{0.02}Na_{0.48}Li_{0.02}Ti_{0.98}Zr_{0.02}O_3)$	2.59	85	38
$Sr_{0.98}Ca_{0.02}TiO_3$	1.98	72.3	39
$(Sr_{0.98}Ca_{0.02})(Ti_{0.98}Zr_{0.02})O_3$	2.77	77.7	40
$SrSn_{0.05}Ti_{0.95}O_3$	1.1	86.02	41

Materials	W_{rec} (J cm^{-3})	η (%)	Ref.
$Sr_{0.3}(Bi_{0.7}Na_{0.94}Li_{0.03})_{0.5}TiO_3$	1.70	87.2	42
$0.45SrTiO_3$-$0.2Na_{0.5}Bi_{0.5}TiO_3$-$0.35BaTiO_3$	1.78	77.06	43
95wt%$Ba_{0.4}Sr_{0.6}TiO_3$-5wt% MgO	1.5	88.5	44
$Ba_{0.5}Sr_{0.5}TiO_3$+1.0wt% SiO_2	2.5	80	45
$Ba_{0.4}Sr_{0.6}TiO_3$+9wt% (Bi_2O_3-B_2O_3-SiO_2)	1.98	90.57	46
$Sr_{0.98}Ca_{0.02}TiO_3$	1.95	72.3	47
$Ba_{0.3}Sr_{0.7}TiO_3$	1.13	86.8	48
($SrTiO_3$+0.5wt%Li_2CO_3)/($0.93Bi_{0.5}Na_{0.5}TiO_3$-$0.07Ba_{0.94}La_{0.04}Zr_{0.02}Ti_{0.98}O_3$)	2.72	74	49
$0.9CaTiO_3$-$0.1BiScO_3$	1.55	90.4	50
$0.96NaNbO_3$-$0.04CaZrO_3$	0.55	63	51
$0.8NaNbO_3$-$0.2SrTiO_3$	3.02	80.7	52
$NaNb_{0.4}Ta_{0.6}O_3$	0.9	87	53
$0.9NaNbO_3$-$0.1Bi(Mg_{2/3}Nb_{1/3})O_3$	2.8	82	54
$0.77NaNbO_3$-$0.23BaTiO_3$+1.5mol% Bi_2O_3	1.5	68.2	55
$0.61BiFeO_3$-$0.33BaTiO_3$-$0.06Ba(Mg_{1/3}Nb_{2/3})O_3$	1.56	75	56
$0.66BiFeO_3$-$0.33BaTiO_3$-$0.06La(Mg_{1/2}Ti_{1/2})O_3$	1.66	82	57
$0.60BiFeO_3$-$0.34BaTiO_3$-$0.06Sr(Al_{0.5}Nb_{0.5})O_3$	1.75	65	58
$0.75(Bi_{0.85}Nd_{0.15})FeO_3$-$0.25BaTiO_3$	1.82	44.4	59
$0.60BiFeO_3$-$0.34BaTiO_3$-$0.06Ba(Zn_{1/3}Ta_{2/3})O_3$	2.56	72	60
$0.65BiFeO_3$-$0.3BaTiO_3$-$0.05Bi(Zn_{2/3}Nb_{1/3})O_3$+0.1wt% Mn_2O_3	2.06	53	61
$0.60BiFeO_3$-$0.34BaTiO_3$-$0.06Sr(Al_{0.5}Nb_{0.5})O_3$	1.75	81	62
$0.62BiFeO_3$-$0.3BaTiO_3$-$0.08Nd(Zr_{0.5}Zn_{0.5})O_3$	2.45	72	63
$0.75(Bi_{0.85}Nd_{0.15})FeO_3$-$0.25BaTiO_3$+0.1 wt% MnO_2	1.82	41.3	64
$0.61BiFeO_3$-$0.33(Ba_{0.8}Sr_{0.2})TiO_3$-$0.06La(Mg_{2/3}Nb_{1/3})O_3$+$MnO_2$+$BaCu(B_2O_5)$	3.38	59	65
$Bi_{0.83}Sm_{0.17}Fe_{0.95}Sc_{0.05}O_3$	2.21	76	66
$0.85(K_{0.5}Na_{0.5})NbO_3$-$0.15SrTiO_3$	4.03	52	67
$0.9(K_{0.5}Na_{0.5})NbO_3$-$0.1Bi(Mg_{2/3}Nb_{1/3})O_3$	4.08	62.7	68
$0.8(K_{0.5}Na_{0.5})NbO_3$-$0.2Sr(Sc_{0.5}Nb_{0.5})O_3$	2.02	81.4	69
$0.92K_{0.5}Na_{0.5}NbO_3$-$0.06Sr(Zn_{1/3}Nb_{2/3})O_3$	1.5	50	70
$0.9(K_{0.5}Na_{0.5})NbO_3$-$0.1Bi(Mg_{2/3}Nb_{1/3})O_3$+1.0mol% CuO	4.02	57	71
$0.8(K_{0.5}Na_{0.5})NbO_3$-$0.2Sr(Sc_{0.5}Nb_{0.5})O_3$+0.5mol% ZnO	2.6	73.2	72
$0.9K_{0.5}Na_{0.5}NbO_3$-$0.1BiFeO_3$	2	60.5	73
$(Ag_{0.97}Bi_{0.01})NbO_3$	3.0	55	74
$Ag(Nb_{0.85}Ta_{0.15})O_3$	4.2	69	75
$AgNbO_3$+0.1wt% MnO_2	2.3	56	76
$AgNbO_3$+0.1wt% WO_3	3.3	50	77
$Ag(Nb_{0.8}Ta_{0.2})O_3$	3.7	57	78
$(Ag_{0.92}Ca_{0.04})NbO_3$	3.55	56.3	79
$(Ag_{0.94}La_{0.02})NbO_3$	4.4	70	80
$(Ag_{0.88}Gd_{0.04})O_3$	4.5	64	81
$(Ag_{0.96}Ba_{0.02})NbO_3$	2.3	46	82
$AgNbO_3$	2.1	40	83
$0.91BaTiO_3$-$0.09BiYbO_3$	0.71	82.6	84

Materials	W_{rec} (J cm^{-3})	η (%)	Ref.
$0.9BaTiO_3$-$0.1Bi(Mg_{2/3}Nb_{1/3})O_3$	1.13	96	85
$0.88BaTiO_3$-$0.12Bi(Mg_{1/2}Ti_{1/2})O_3$	1.81	88	86
$0.9BaTiO_3$-$0.1BiInO_3$	0.753	89.4	87
$0.9BaTiO_3$-$0.1BiNbO_4$	0.797	92.5	88
$0.86BaTiO_3$-$0.14Bi(Zn_{1/2}Ti_{1/2})O_3$	0.81	94	89
$0.85BaTiO_3$-$0.15Bi(Zn_{2/3}Nb_{1/3})O_3$	0.79	93.5	90
$0.85BaTiO_3$-$0.15Bi(Zn_{1/2}Sn_{1/2})O_3$	2.41	91.6	91
$0.88BaTiO_3$-$0.12Bi(Ni_{2/3}Nb_{1/3})O_3$	2.09	95.9	92
$0.9BaTiO_3$-$0.1(Bi_{0.9}Na_{0.1})(In_{0.8}Zr_{0.2})O_3$	1.33	88	93
$0.88BaTiO_3$-$0.12Bi(Li_{0.5}Nb_{0.5})O_3$	2.032	88	94
$0.85BaTiO_3$-$0.15Bi(Mg_{0.5}Zr_{0.5})O_3$	2.9	86.8	95
$0.92(0.92BaTiO_3$-$0.08K_{0.5}Bi_{0.5}TiO_3)$-$0.08NaNbO_3$	1.96	67.4	96
$0.93BaTiO_3$-$0.07YNbO_4$	0.614	87	97
$0.9BaTiO_3$-$0.1Bi(Mg_{2/3}Nb_{1/3})O_3$+0.3wt% $MnCO_3$	1.7	90	98
$0.92(0.65BaTiO_3$-$0.35Bi_{0.5}Na_{0.5}TiO_3)$-$0.08Na_{0.73}Bi_{0.09}NbO_3$	1.70	82	99
$0.85Ba(Zr_{0.2}Ti_{0.8})O_3$-$0.15(Ba_{0.7}Ca_{0.3})TiO_3$	0.94	72	100
$Ba_{0.95}Ca_{0.05}Zr_{0.3}Ti_{0.7}O_3$	0.59	72.8	101
$Ba_{0.9995}La_{0.0005}TiO_3$+15mol% (65PbO-20$B_2O_3-15SiO_2$)	0.564	54.2	102
$0.88BaTiO_3$-$0.12Bi(Mg_{1/2}Ti_{1/2})O_3$+4mol% (25%$SiO_2$-75%$B_2O_3$)	1.971	94.5	103
$0.92(0.65BaTiO_3$-$0.35Bi_{0.5}Na_{0.5}TiO_3)$-$0.08SrY_{0.5}Nb_{0.5}O_3$	1.36	74.3	104
$0.87BaTiO_3$-$0.13Bi(Zn_{2/3}(Nb_{0.85}Ta_{0.15})_{1/3})O_3$	1.44	92.5	105

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