Supporting information for

Two-step sintering and $Sr(Fe_{0.5}Nb_{0.5})O_3$ addition enhancing electrical properties of KNN-based lead-free piezoelectric ceramics

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Table S1. Cell volume and density of 0.95KNNbS-0.05SrFNb ceramics sintered by CS and TSS methods at different conditions.

Sintering conditions (℃)	Theoretical volume ($Å^3$)	Bulk density (g/cm^3)	Theoretical density (g/cm^3)	Relative density (%)
1160	63.81	4.19	4.55	92.08
1165	63.47	4.22	4.58	92.13
1170	63.66	4.18	4.56	91.66
1200-1125 ℃ 6 h	63.87	4.29	4.55	94.28
1200-1125 ℃ 4 h	63.67	4.31	4.56	94.51
1190-1125 ℃ 4 h	63.66	4.28	4.56	93.85

Table S2 Cell volume and density of (1-x)KNNbS-xSrFNb ceramics sintered by TSS.

Composition (mol%)	Theoretical volume ($Å^3$)	Bulk density (g/cm^3)	Theoretical density (g/cm^3)	Relative density (%)
x = 0.01	63.00	4.24	4.57	92.77
x = 0.03	63.23	4.15	4.57	90.81
x = 0.05	63.67	4.31	4.56	94.51
x = 0.07	63.00	3.98	4.63	85.96

Table S3 P_r, E_c, d_{33}*, K_p, Q_m and d_{33} of 0.95KNNbS-0.05SrFNb ceramics sintered by CS and TSS methods at different conditions.

Sintering conditions (℃)	P_r (μC/cm^2)	E_c (kV/cm)	d_{33}* (pm/V)	K_p	Q_m	d_{33} (pC/N)
1160	3.75	8.13	127	0.105	97.5	16±0.8
1165	5.60	11.80	137	0.095±0.008	154.1±14	20±2.5
1170	5.11	8.32	156	0.100	119.3	20±0.5
1200-1125 ℃ 6 h	4.85	7.08	207	0.098	162.4	25±1.5
1200-1125 ℃ 4 h	6.46	7.57	129	0.128±0.003	120.5±7	42±4.5
1190-1125 ℃ 4 h	6.01	7.19	156	0.101	188.3	40±1.5

Table S4 P_r, E_c, d_{33}*, K_p, Q_m and d_{33} of (1-x)KNNbS-xSrFNb ceramics sintered by TSS.

Samples	P_r (μC/cm^2)	E_c (kV/cm)	d_{33}* (pm/V)	K_p	Q_m	d_{33} (pC/N)
x=0.01	12.45	8.85	206	0.251±0.007	329.7±17	94±5.0
x=0.03	22.72	9.47	194	0.285±0.01	143.4±37	133±2.0
x=0.05	6.46	7.57	129	0.128±0.003	120.5±7	42±4.5
x=0.07	13.14	7.34	44			

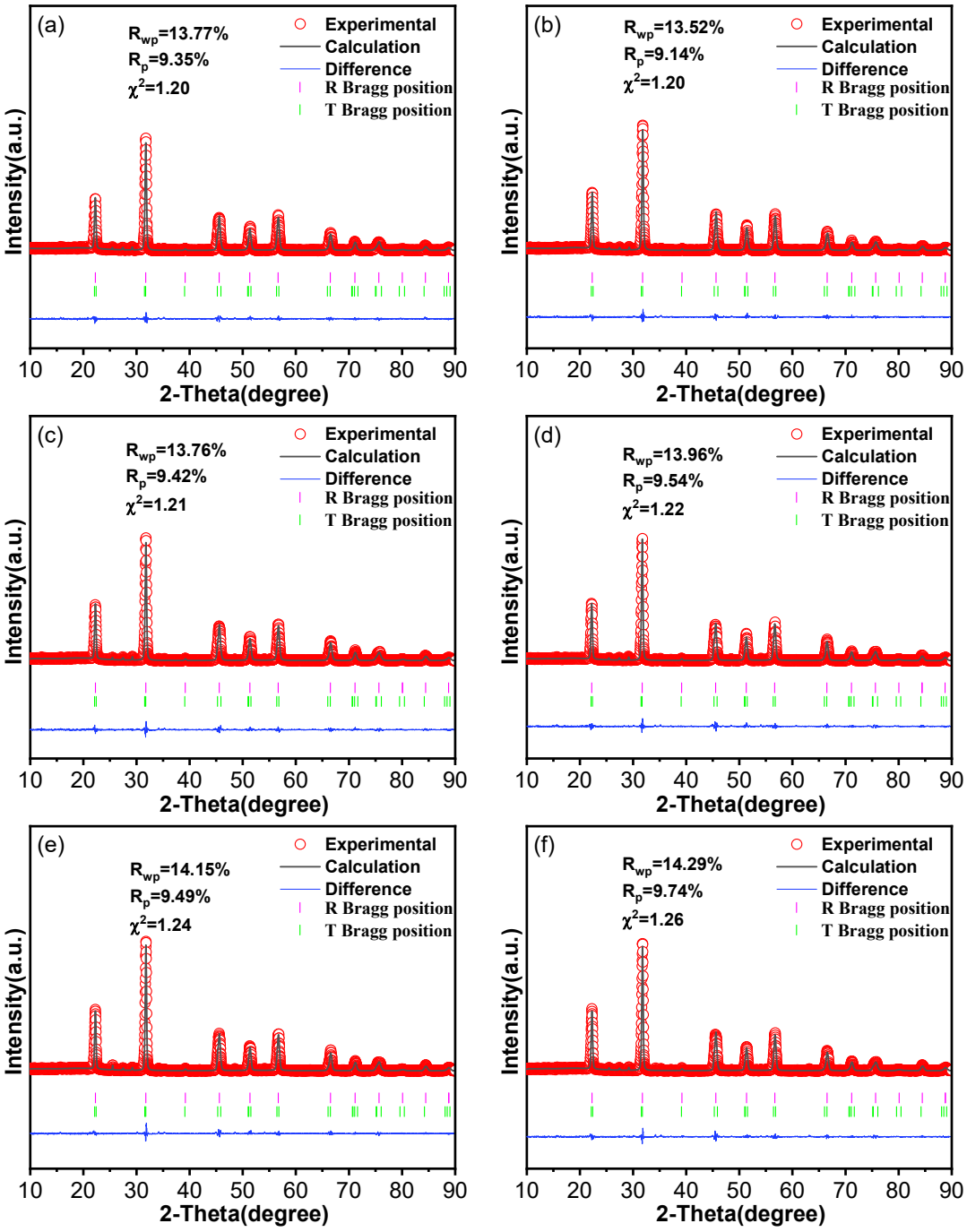


Figure S1. XRD Rietveld refinement results of 0.95KNNbS-0.05SrFNb ceramics sintered by CS and TSS methods at different conditions.

(a) 1160 ℃; (b) 1165 ℃; (c) 1170 ℃; (d) 1200-1125 ℃ 6 h;

(e) 1200-1125 ℃ 4 h; (f) 1190-1125 ℃ 4 h.

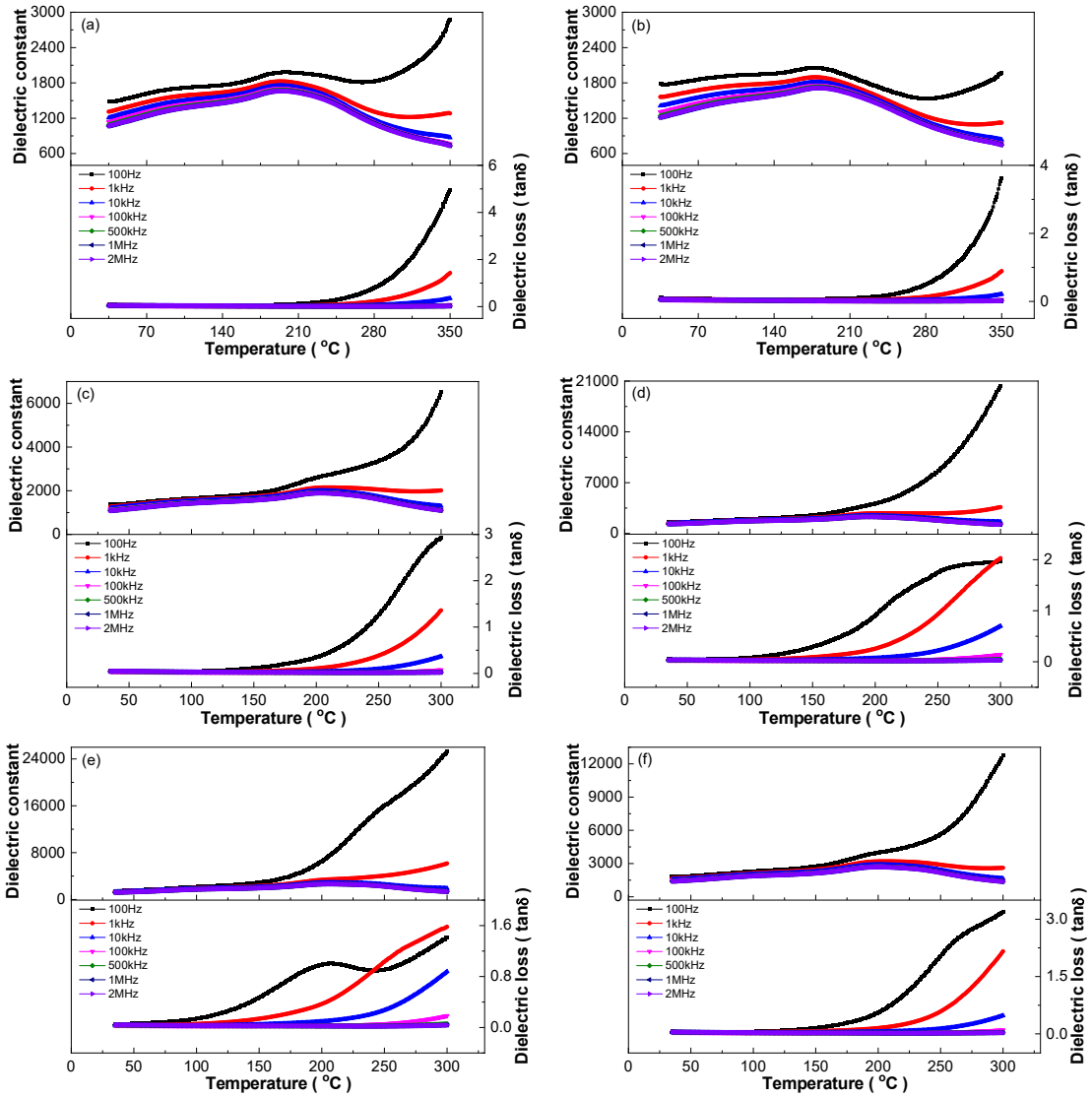


Figure S2. Effects of frequency on dielectric properties of 0.95KNNbS-0.05SrFNb ceramics sintered by CS and TSS methods at different conditions.

(a) 1160 °C; (b) 1165 °C; (c) 1170 °C; (d) 1200-1125 °C 6 h;

(e) 1200-1125 °C 4 h; (f) 1190-1125 °C 4 h.

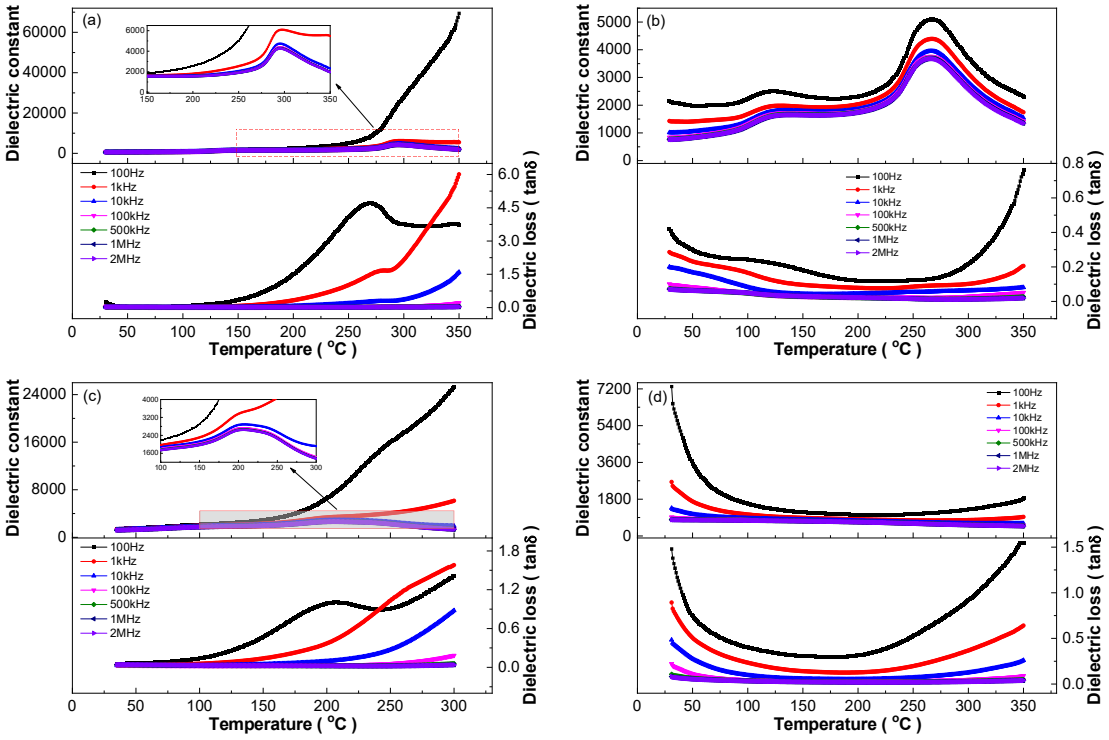
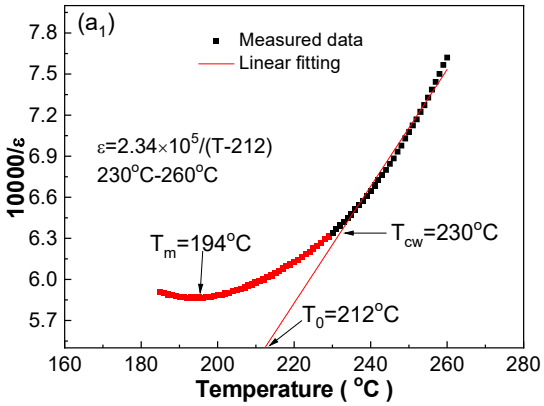
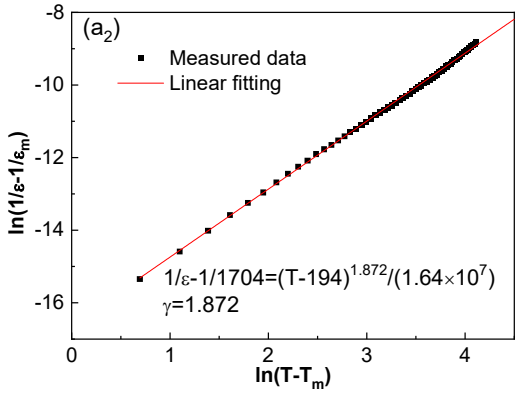
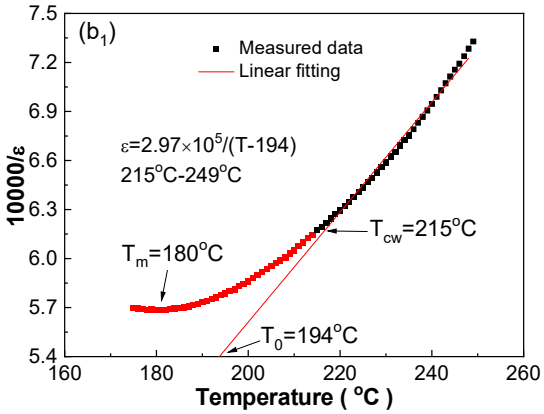


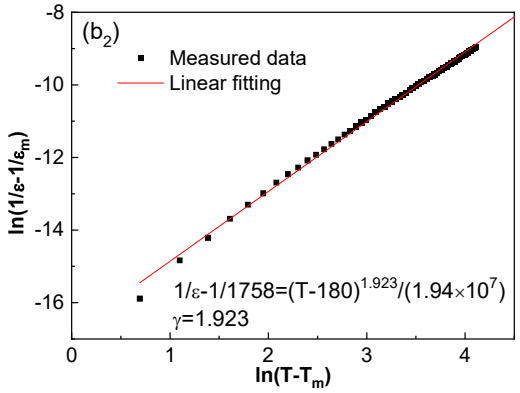
Figure S3. Effects of frequency on dielectric properties of (1-x)KNNbS-xSrFNb ceramics sintered by TSS.

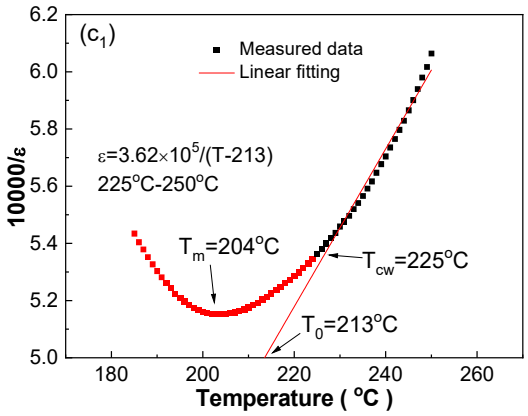
(a) x = 0.01; (b) x = 0.03; (c) x = 0.05; (d) x = 0.07

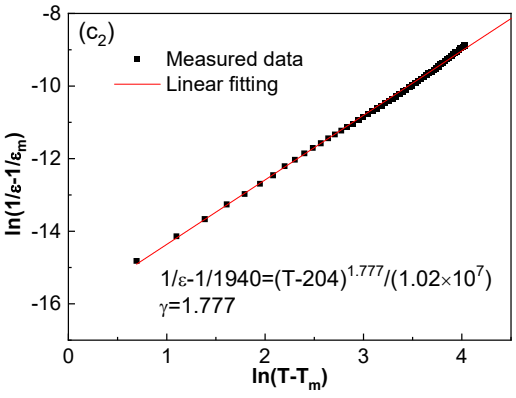

(a1)
Measured data
Linear fitting
ε=2.34×10^5/(T-212)
230°C-260°C
Tm=194°C
Tcw=230°C
T0=212°C
10000/ε
Temperature (°C)


(a2)
Measured data
Linear fitting
1/ε-1/1704=(T-194)^1.872/(1.64×10^7)
γ=1.872
ln(1/ε-1/εm)
ln(T-Tm)


(b1)
Measured data
Linear fitting
ε=2.97×10^5/(T-194)
215°C-249°C
Tm=180°C
Tcw=215°C
T0=194°C
10000/ε
Temperature (°C)


(b2)
Measured data
Linear fitting
1/ε-1/1758=(T-180)^1.923/(1.94×10^7)
γ=1.923
ln(1/ε-1/εm)
ln(T-Tm)


(c1)
Measured data
Linear fitting
ε=3.62×10^5/(T-213)
225°C-250°C
Tm=204°C
Tcw=225°C
T0=213°C
10000/ε
Temperature (°C)


(c2)
Measured data
Linear fitting
1/ε-1/1940=(T-204)^1.777/(1.02×10^7)
γ=1.777
ln(1/ε-1/εm)
ln(T-Tm)

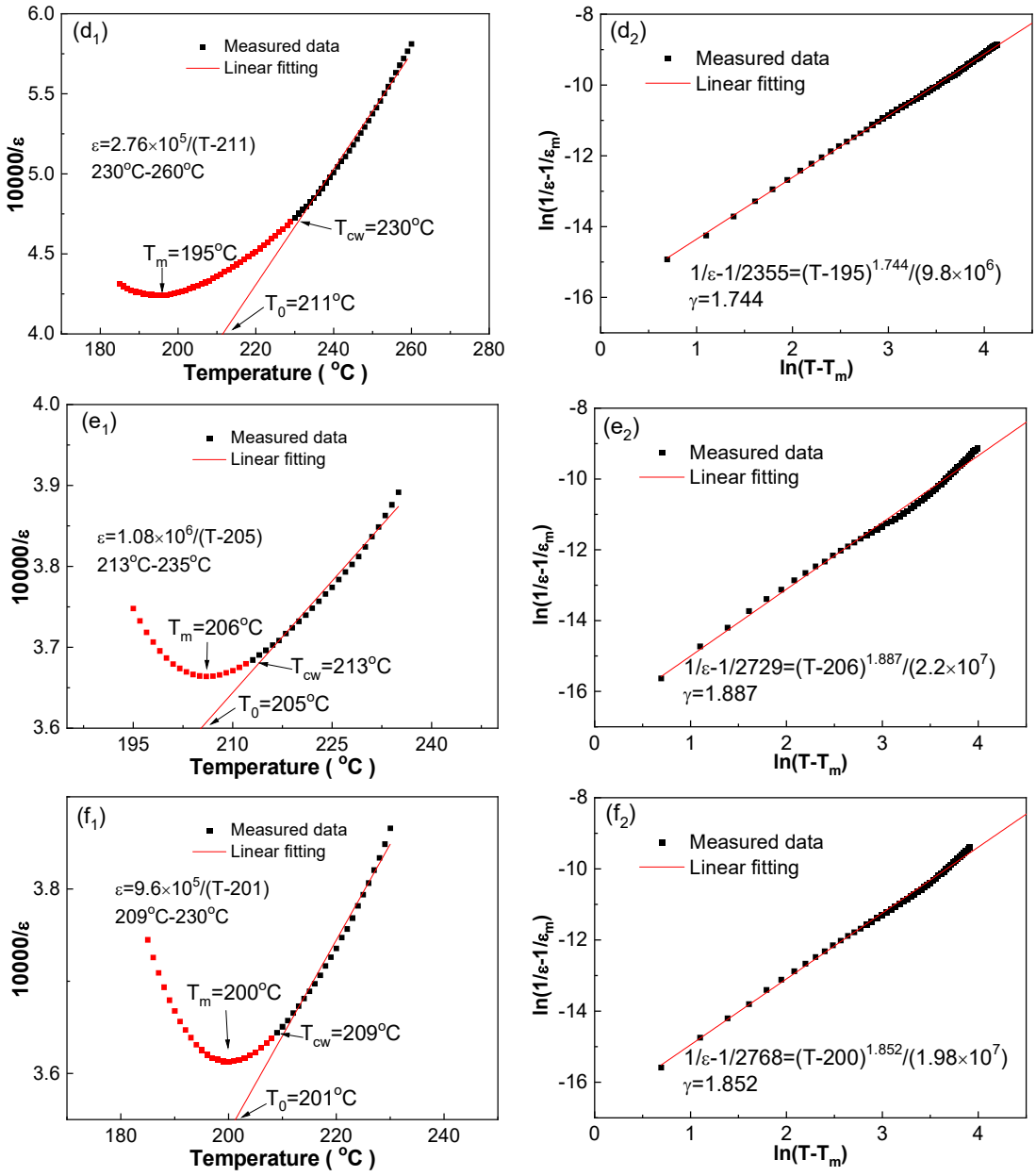


Figure S4. Linear fitting of the dielectric behavior above the T_C temperature of the 0.95KNNbS-0.05SrFNb ceramics sintered by CS and TSS using 100 kHz data according to (a_1)-(f_1) the Curie-Weiss law and (a_2)-(f_2) the exponential law.

(a) 1160 ℃; (b) 1165 ℃; (c) 1170 ℃; (d) 1200-1125 ℃ 6 h;

(e) 1200-1125 ℃ 4 h; (f) 1190-1125 ℃ 4 h.

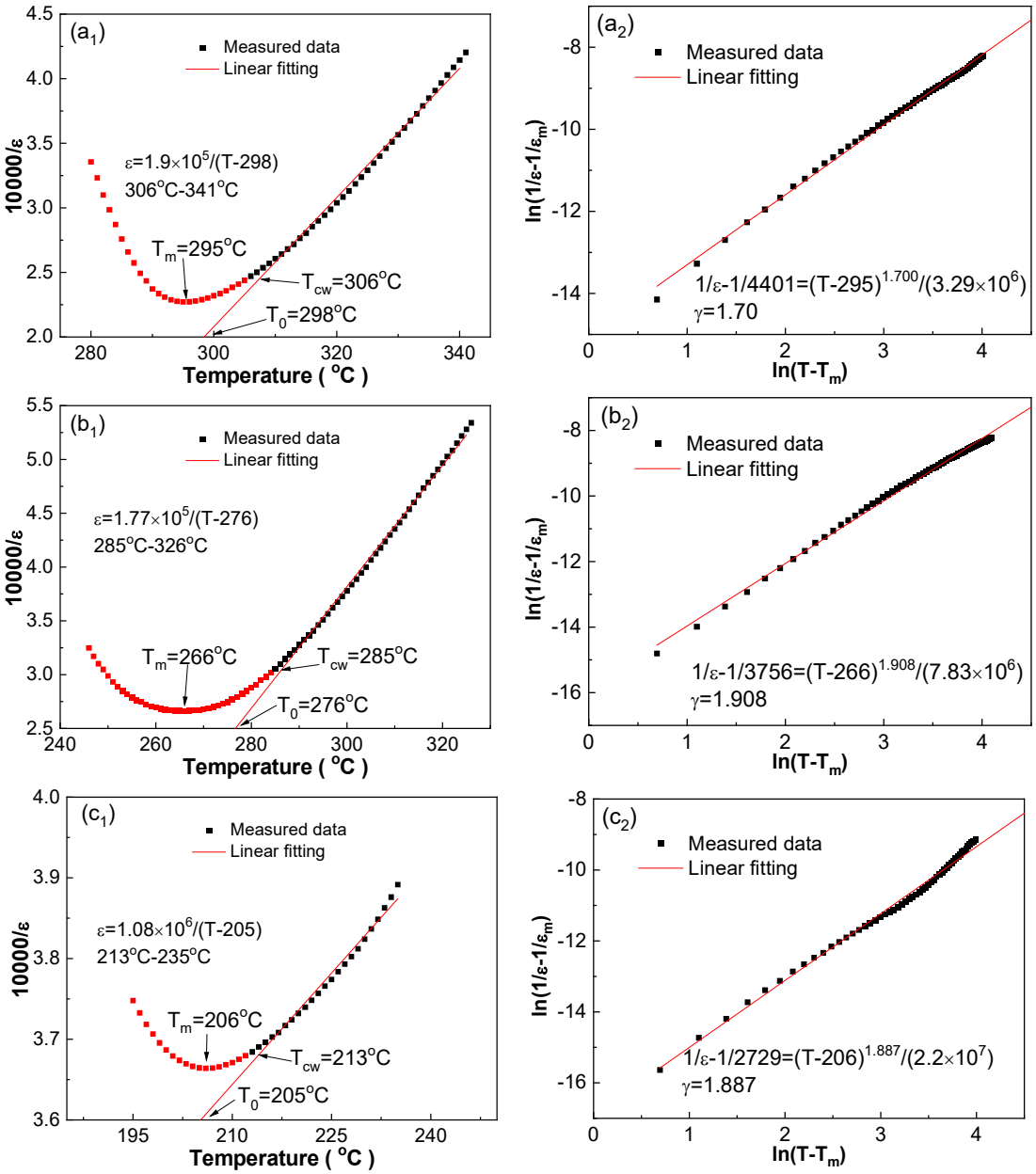


Figure S5. Linear fitting of the dielectric behavior above the T_C temperature of the (1-x)KNNbS-xSrFNb ceramics sintered by TSS using 100 kHz data according to (a_1)-(c_1) the Curie-Weiss law and (a_2)-(c_2) the exponential law.

(a) x = 0.01; (b) x = 0.03; (c) x = 0.05.

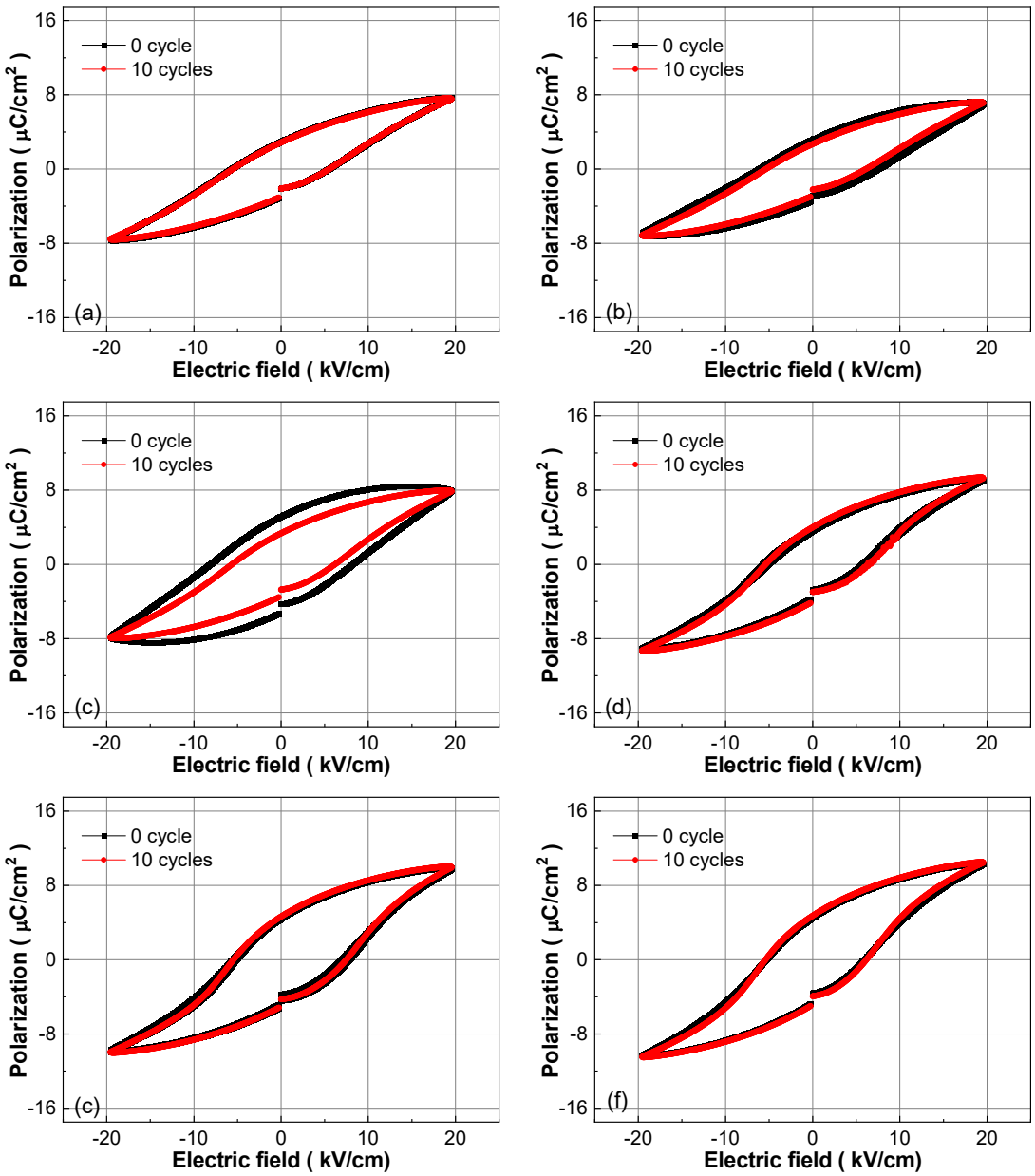


Figure S6. Fatigue test of 0.95KNNbS-0.05SrFNb ceramics sintered by CS and TSS after 10 cycles under 20 kV/cm.

(a) 1160 ℃; (b) 1165 ℃; (c) 1170 ℃; (d) 1200-1125 ℃ 6 h;

(e) 1200-1125 ℃ 4 h; (f) 1190-1125 ℃ 4 h.

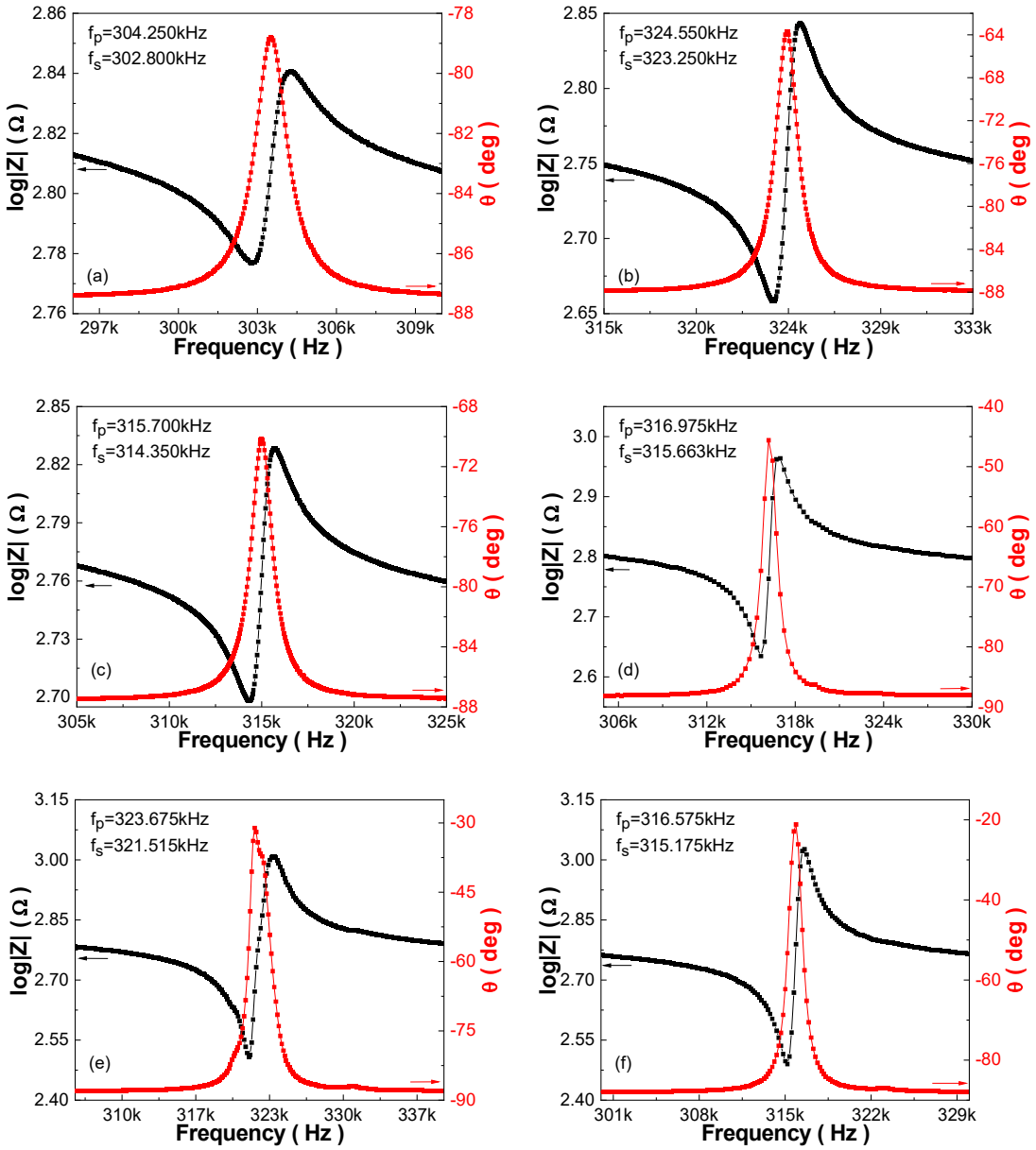


Figure S7. Impedance and phase angle of 0.95KNNbS-0.05SrFNb ceramics sintered by CS and TSS.

(a) 1160 ℃; (b) 1165 ℃; (c) 1170 ℃; (d) 1200-1125 ℃ 6 h;

(e) 1200-1125 ℃ 4 h; (f) 1190-1125 ℃ 4 h.