

Supplementary Information

Global-optimized energy storage performance in multilayer ferroelectric ceramic capacitors

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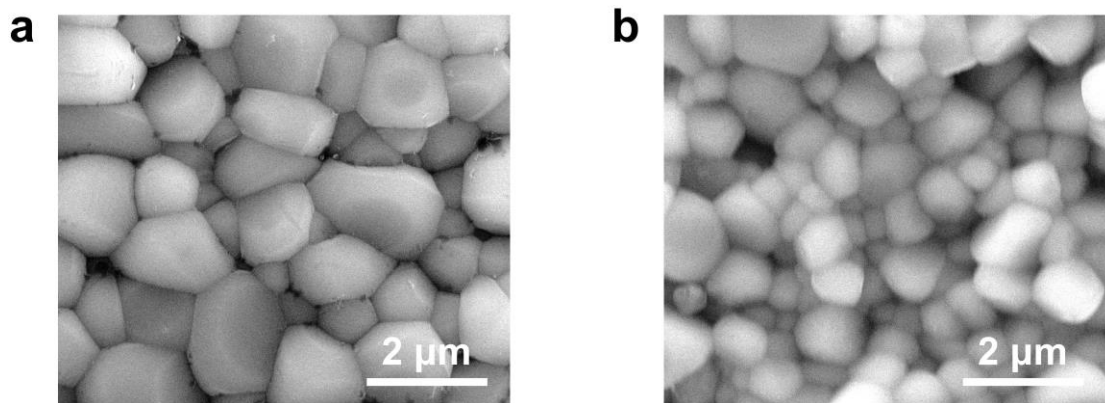
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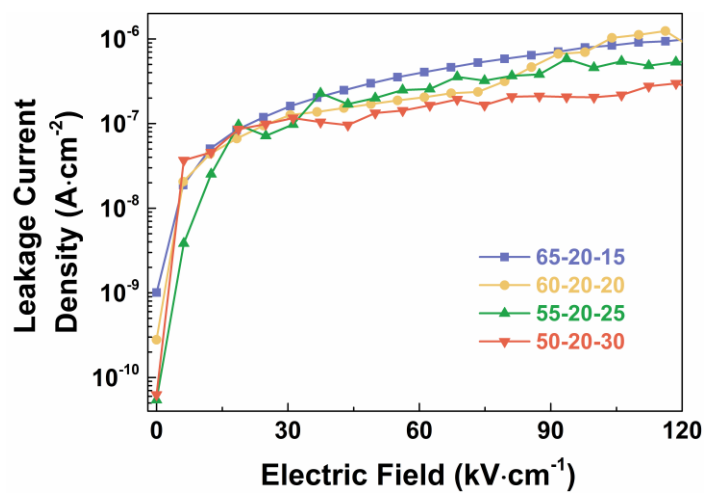
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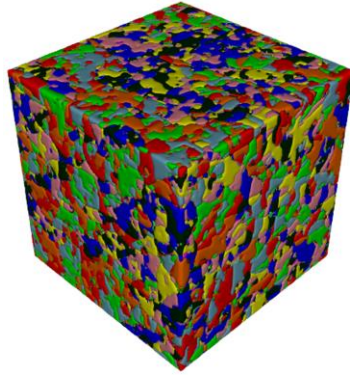
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Supplementary Fig. 1 SEM images of (1-x)BNT-0.20NN-xST ceramics. a, 60-20-20. b, 50-20-30.

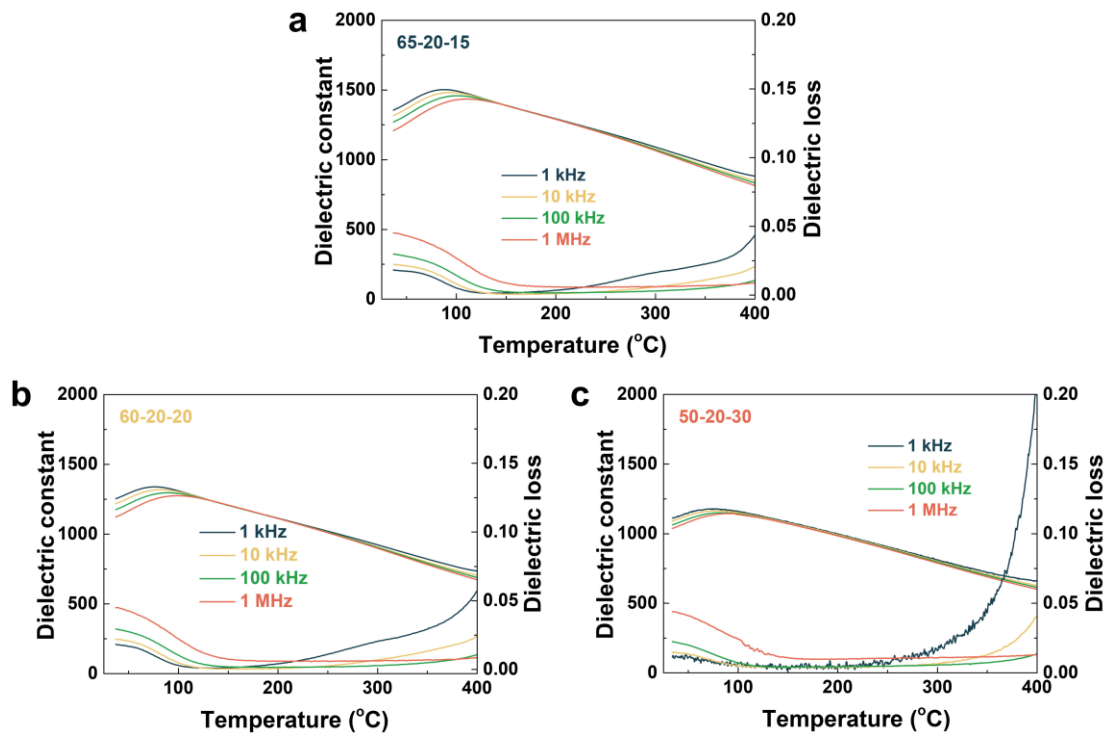


Supplementary Fig. 2 Leakage current densities as a function of electric field of (1-x)BNT-0.20NN-xST ceramics.

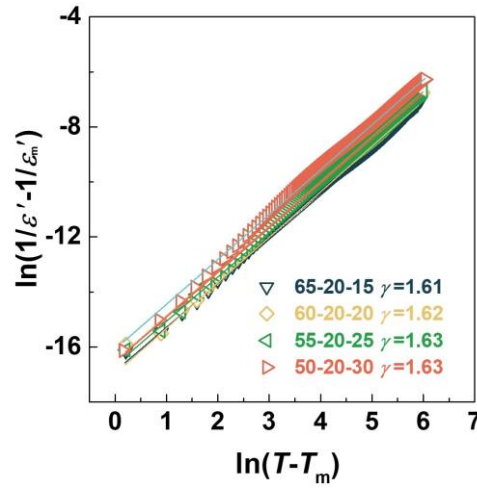


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|------------|------------|------------|------------|
| R1(+,+,+) | R1(-,-,-) | R2(+,-,+) | R2(-,+,-) |
| O1(+,+,+) | O1(-,-,0) | O2(+,-,0) | O2(-,+,0) |
| O5+(0,+,+) | O5-(0,-,-) | O6+(0,+,-) | O6-(0,-,+) |
| R3+(-,-,+) | R3-(+,+,-) | R4+(-,-,+) | R4-(+,-,-) |
| O3+(-,0,+) | O3-(+,0,-) | O4+(-,0,+) | O4-(+,0,-) |
| T1+(-,0,0) | T1-(+,0,0) | T3+(0,0,+) | T3-(0,0,-) |

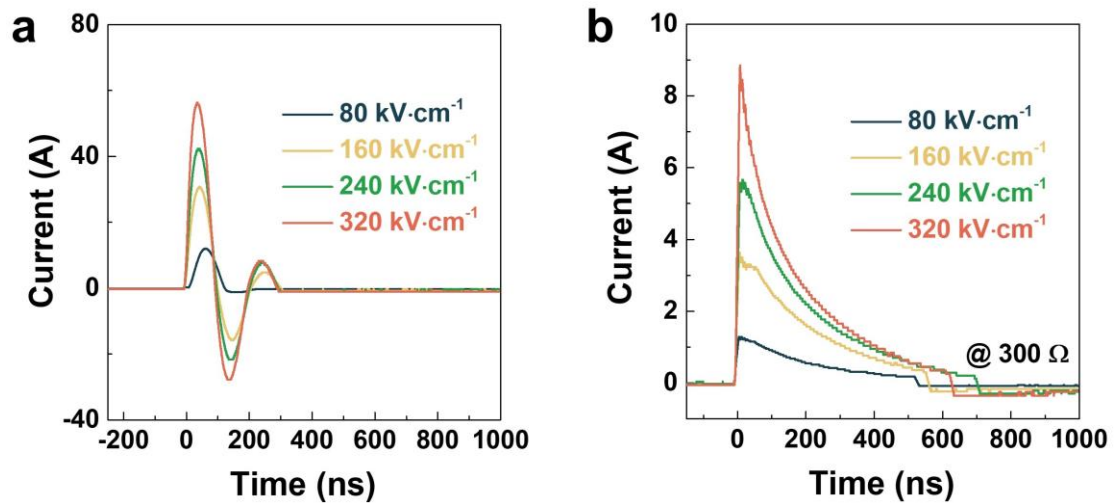
Supplementary Fig. 3 Phase-field simulated three-dimensional domain structures of 55-20-25 ceramics. The different colors represent different polarization directions of the domains. For clarity, the cubic (C) matrix is not depicted.



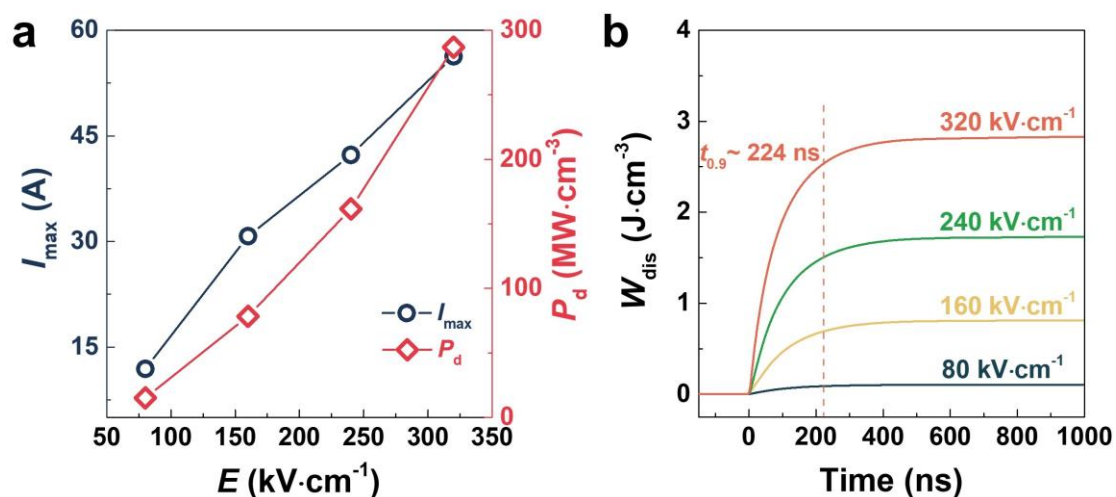
Supplementary Fig. 4 Temperature-dependent dielectric constant and dielectric loss of (1-x)BNT-0.20NN-xST ceramics. a, 65-20-15. b, 60-20-20. c, 50-20-30.



Supplementary Fig. 5 Characterization of the diffuseness factor γ for (1-x)BNT-0.20NN-xST ceramics. The relaxor diffuseness factor γ can be calculated from the modified Curie-Weiss law ($\frac{1}{\epsilon'} - \frac{1}{\epsilon_m'} = \frac{(T-T_m)^\gamma}{C}$), where ϵ' is the dielectric constant at the temperature T , T_m is the temperature at which ϵ' reaches the maximum value (ϵ_m'), and C is a constant. The γ value is used to reflect the relaxor feature, which varies from 1 (corresponding to normal ferroelectrics) to 2 (corresponding to ideal relaxor ferroelectrics).

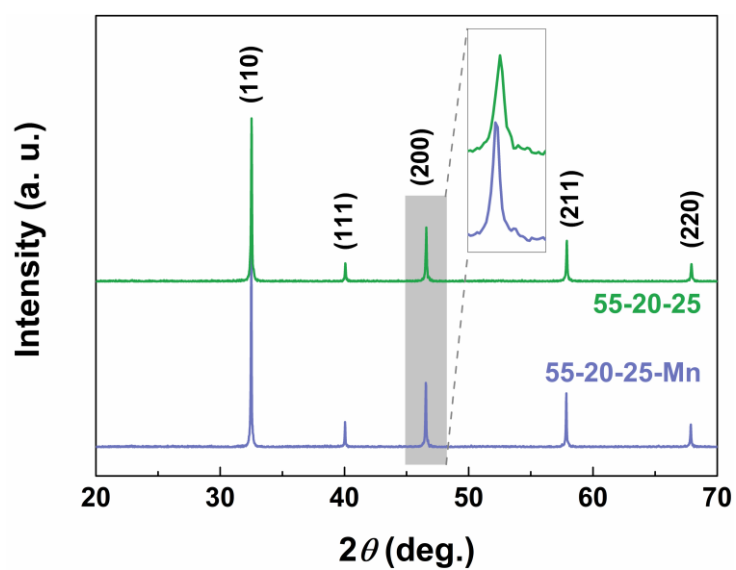


Supplementary Fig. 6 Charge-discharge performances of 55-20-25 ceramics. **a**, Underdamped discharge waveforms, and **b**, overdamped discharge waveforms at different electric fields.



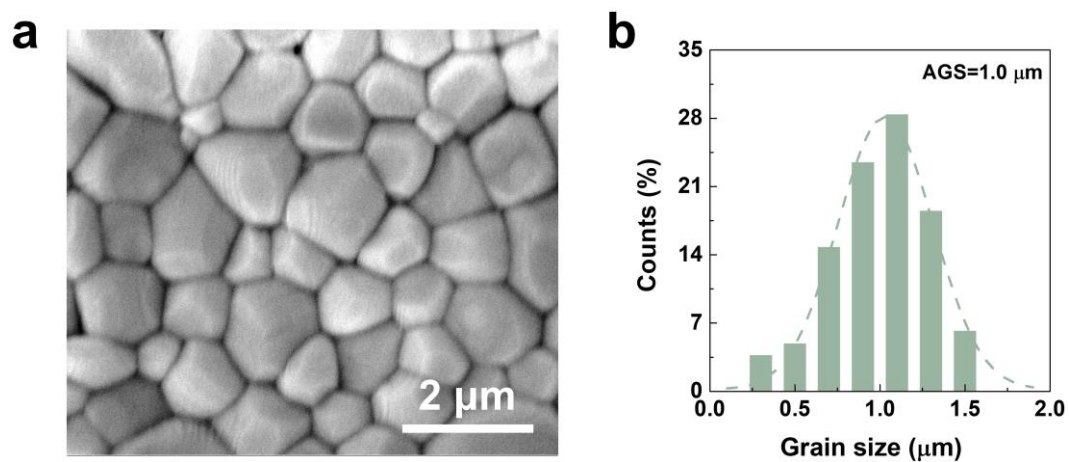
Supplementary Fig. 7 The calculated discharge behavior parameters of 55-20-25

ceramics. **g**, $I_{\max}$, and P_d values, and **h**, W_{dis} as a function of time.

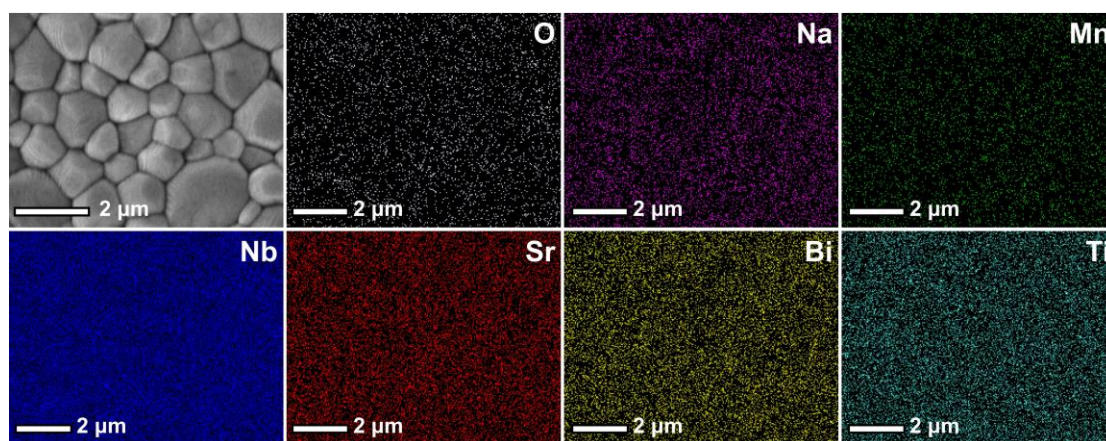


Supplementary Fig. 8 Effect of adding MnO_2 on the phase structure of ceramics.

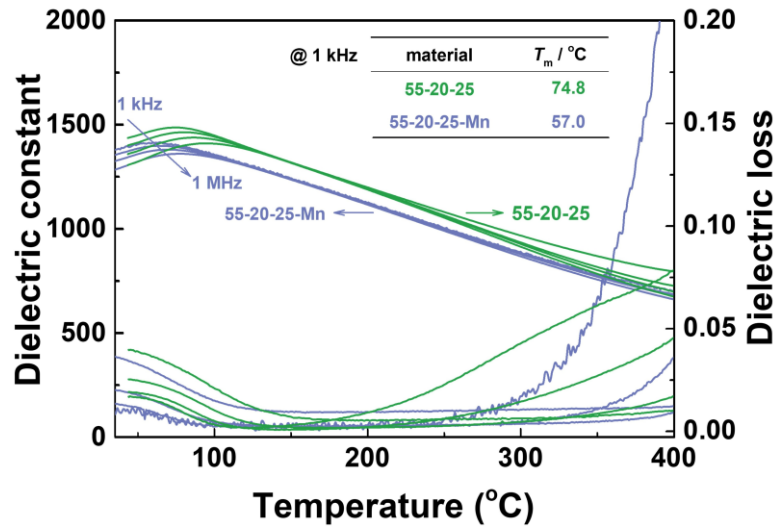
XRD patterns of 55-20-25 and 55-20-25-Mn ceramics.



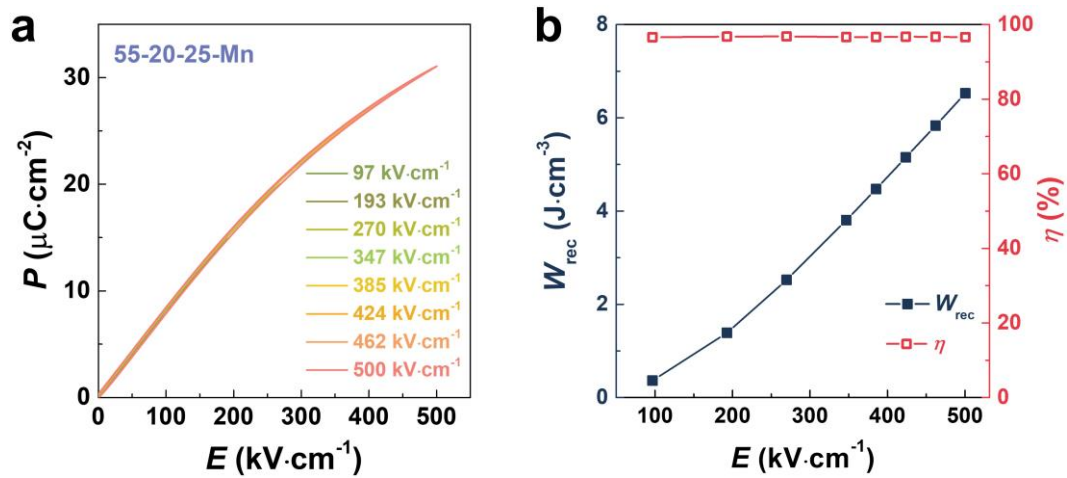
Supplementary Fig. 9 Microstructures of 55-20-25-Mn ceramics. **a**, SEM image of the natural surfaces and **b**, grain size distribution of 55-20-25-Mn ceramics.



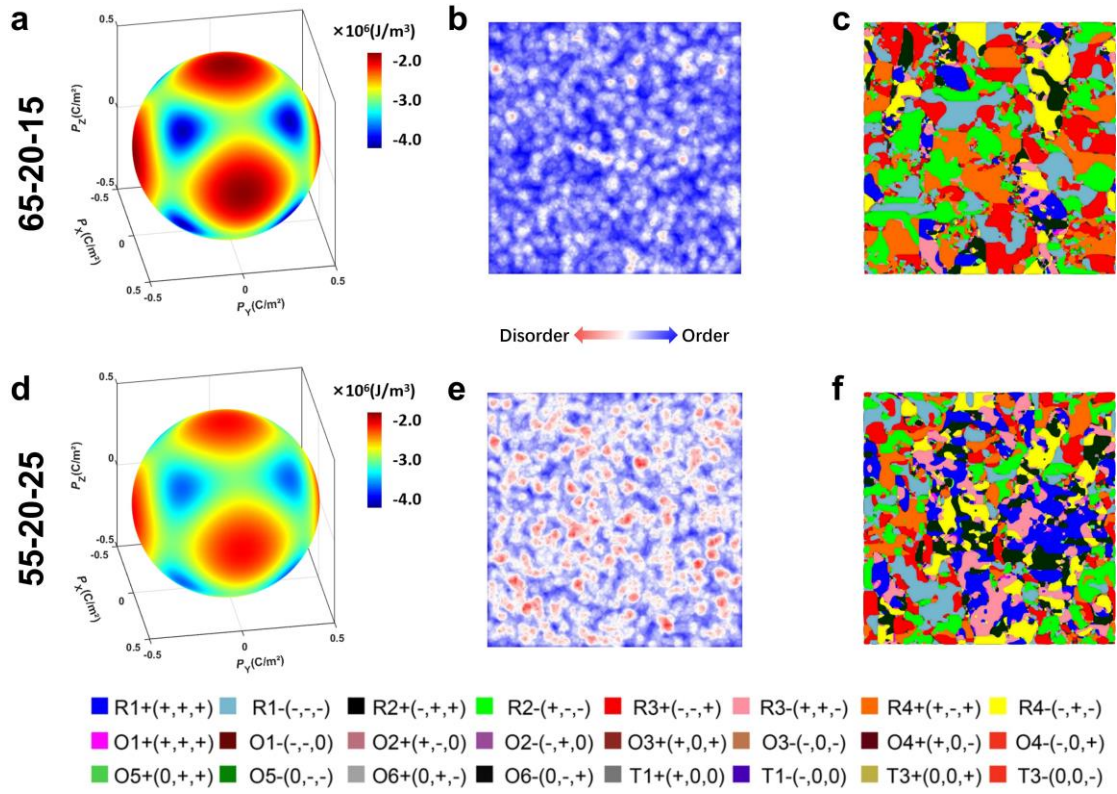
Supplementary Fig. 10 Energy-dispersive X-ray (EDX) spectrometry element-mapping images of 55-20-25-Mn ceramics. All elements are evenly distributed over the observed range.



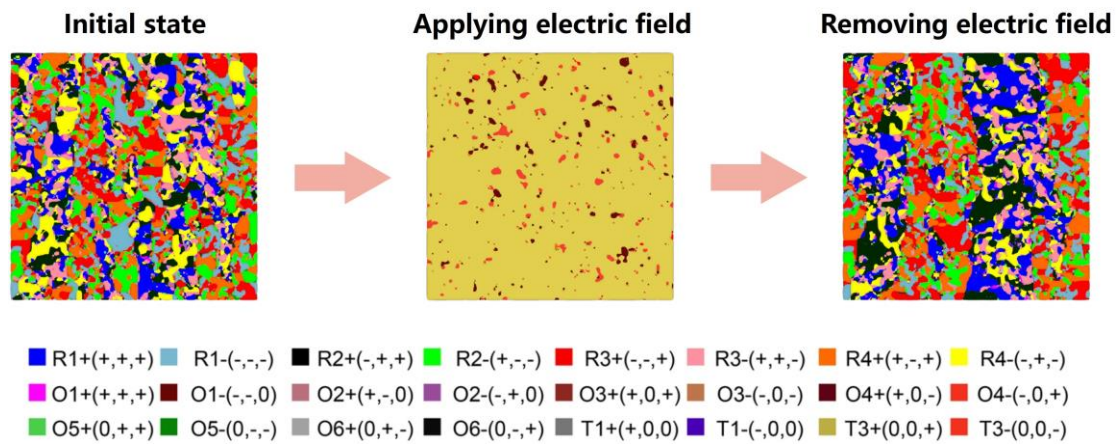
Supplementary Fig. 11 Temperature-dependent dielectric constant and dielectric loss of 55-20-25 and 55-20-25-Mn ceramics. The insert table presents the T_m value of the samples at 1 kHz.



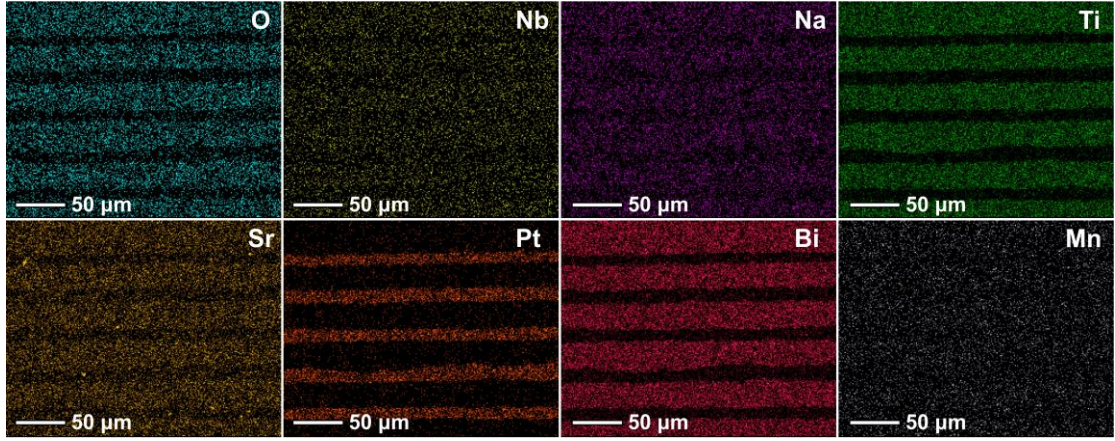
Supplementary Fig. 12 Energy storage properties of 55-20-25-Mn ceramics. **a**, P - E loops, **b**, W_{rec} and η values at various electric fields.



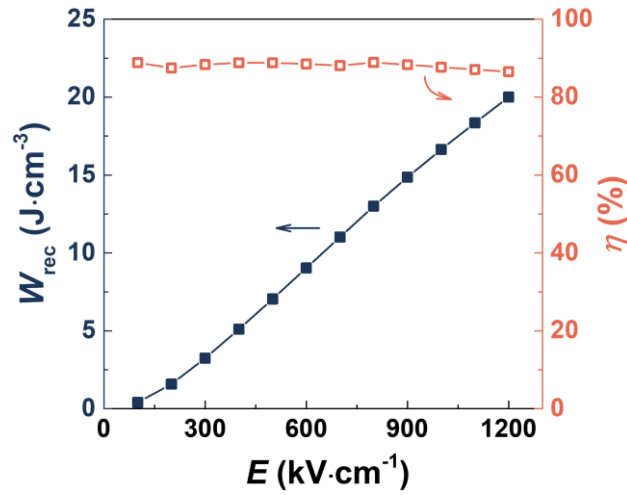
Supplementary Fig. 13 Evolution of polar structures simulated by the phase-field model. **a, d**, The Landau energy profiles, **b, e**, random field distribution, and **c, f**, simulated domain structures of representative compositions of 62-20-15 and 55-20-25. The effect of polar disorder is characterized by the local random field variance. A larger random field corresponds to higher degree of polar disorder.



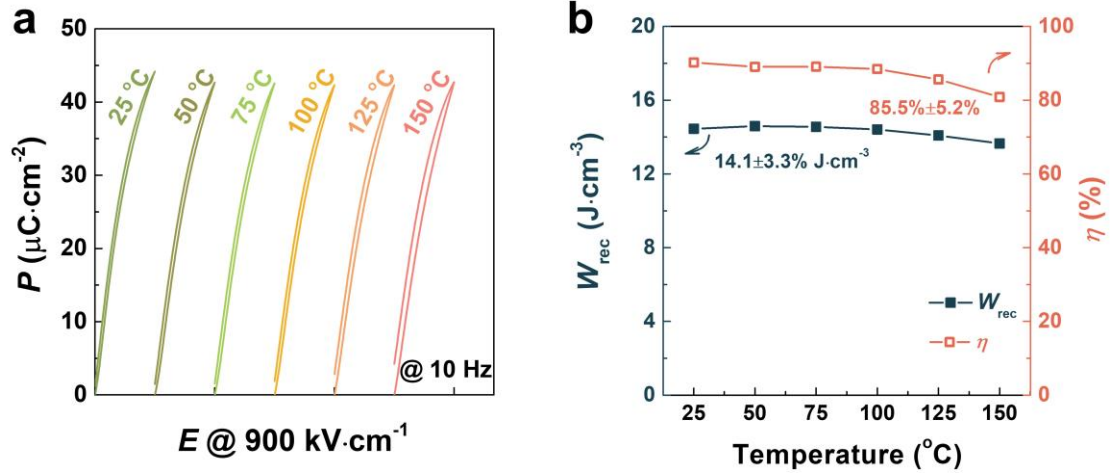
Supplementary Fig. 14 Evolution of polar structure under applied electric field.



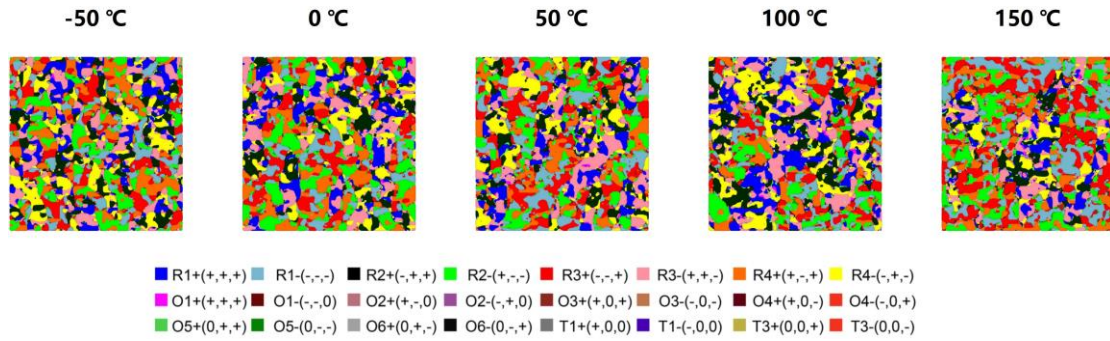
Supplementary Fig. 15 SEM image of the cross-section area with corresponding element distribution. The MLCCs shows a good quality interface between the dielectric layer (Nb, Na, Ti, Bi, Sr, Mn, O elements) and the electrode layer (Pt elements).



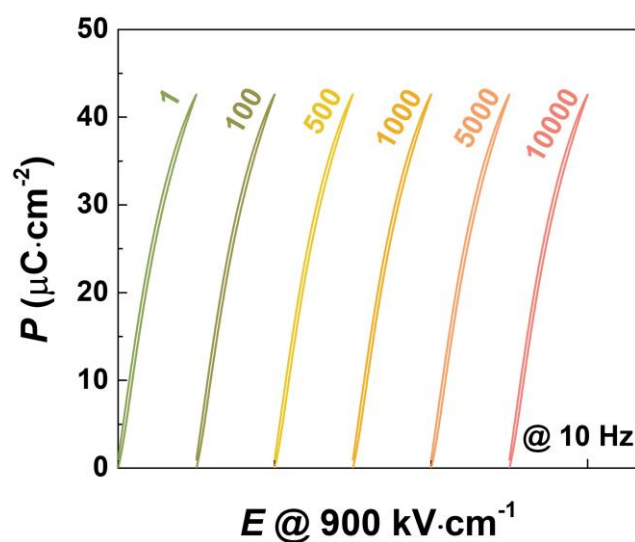
Supplementary Fig. 16 Energy storage properties of 55-20-25-Mn MLCCs. W_{rec} and η values as a function of electric field up to E_b .



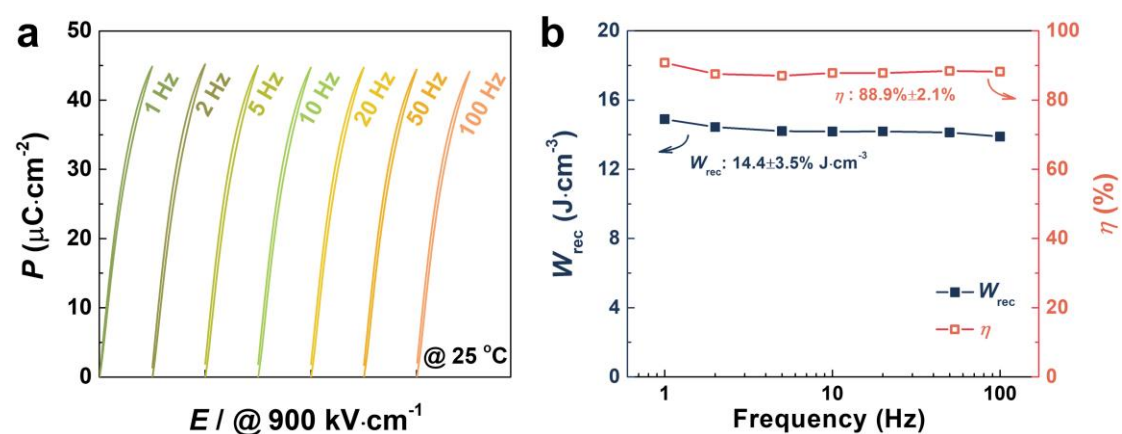
Supplementary Fig. 17 Energy storage properties-dependent temperature of 55-20-25-Mn MLCCs. **a**, P - E loops, **b**, W_{rec} and η at elevated temperatures and 900 $\text{kV}\cdot\text{cm}^{-1}$.



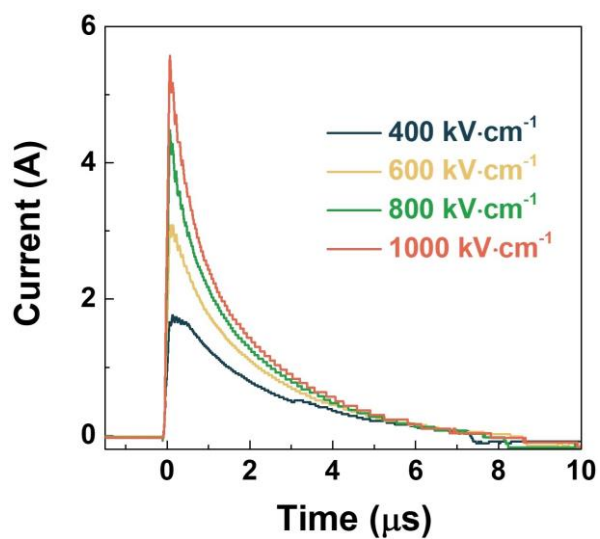
Supplementary Fig. 18 Evolution of polar structures simulated by the phase-field model at different temperatures. Simulated domain structures of 55-20-25-Mn over a broad temperature range from 25 to 150 $^{\circ}\text{C}$.



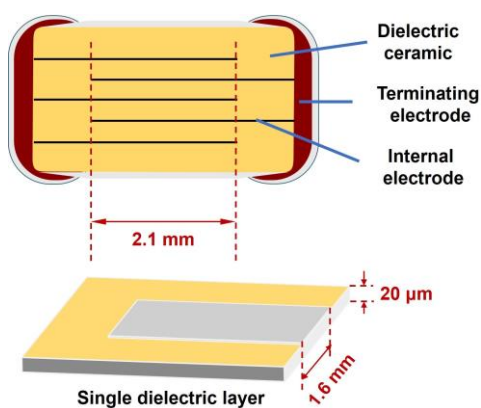
Supplementary Fig. 19 P - E loops after different cycling numbers under 900 $\text{kV}\cdot\text{cm}^{-1}$ for the 55-20-25-Mn MLCC.



Supplementary Fig. 20 Energy storage properties-dependent frequency of 55-20-25-Mn MLCCs. **a**, P - E loops, **b**, W_{rec} and η at various frequencies and 900 $\text{kV}\cdot\text{cm}^{-1}$.



Supplementary Fig. 21 Overdamped discharging waveforms at various electric fields for the 55-20-25-Mn MLCC.



Supplementary Fig. 22 Structure diagram of MLCC.

Supplementary Table 1 T_m of (1- x)BNT-0.20NN- x ST ceramics at 1 kHz.

x / mol	0.15	0.20	0.25	0.30
T_m / °C	87.9	76.2	74.8	69.4