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Non-Polar Nanocluster Confinement Engineering Realizes High Capacitive Energy Storage in Pb-Free High-Entropy Relaxors

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Abstract:

Dielectric ceramic capacitors with ultrahigh power density have become indispensable in modern power electronics, yet the persistent challenge of achieving superior energy density with high energy efficiency remains a critical bottleneck for practical applications. Herein, we propose an effective non-polar nanocluster confinement strategy through phase-field simulation-guided design of high-entropy $(\text{Bi}_{0.2}\text{Na}_{0.2}\text{Ba}_{0.2}\text{Sr}_{0.2}\text{Ca}_{0.2})(\text{Ti}_{1-x}\text{Sn}_x)\text{O}_3$ lead-free relaxor ferroelectrics. The incorporation of Sn^{4+} ions with low ionic polarizability leads to the formation of localized non-ferroelectric perovskite units, which constitute robust non-polar nanoclusters, being further stabilized and rendered immobile against electric fields by the substantial local random fields inherent to the high-entropy configuration. Consequently, these engineered non-polar nanoclusters serve as effective pinning centers to impede the merging and growth of polar nanodomains under electric fields, thereby reconciling the inherent conflict between polarization enhancement and hysteresis reduction. The optimized composition ($x = 0.06$) exhibits a high recoverable energy density of $\sim 18.5 \text{ J}\cdot\text{cm}^{-3}$ together with an ultrahigh energy efficiency of $\sim 92.4\%$ in multilayer ceramic capacitors, representing a competitive combination among lead-free counterparts. This approach not only establishes a viable paradigm for next-generation energy storage dielectrics but also provides fundamental insights for designing functional materials with tailored electrical properties.

Keywords: Capacitive energy storage; Relaxor ferroelectrics; High entropy; Non-polar nanocluster; Ex-/in-situ multiscale structure evolution.

Introduction

High-performance dielectric energy-storage capacitors are critical components for next-generation high/pulsed power electronic systems, driven by escalating demands for advanced energy conversion and management technologies¹⁻³. Distinguished by their unique electrostatic energy-storage mechanism, dielectric capacitors offer unparalleled advantages, including the highest power density ($\sim 10^7$ - 10^8 W/kg) among energy-storage devices, ultra-fast charge/discharge rates on nanosecond timescales, and exceptional operational longevity exceeding 10^5 cycles⁴⁻⁸. These characteristics make them indispensable in mission-critical applications such as medical defibrillators, electromagnetic armor systems, and hybrid electric vehicle powertrains.

Multilayer ceramic capacitors (MLCCs) have attracted considerable attention as promising candidates for next-generation energy storage⁸⁻¹⁶. In contrast to bulk ceramic capacitors, their layered architecture enhances dielectric breakdown strength (E_B) through the thinning of individual ceramic layers and improves volumetric efficiency via compact stacking of alternating dielectric and electrode layers⁸⁻¹⁶. While thin-film capacitors can attain higher E_B and thus larger energy storage density (W_{rec}) owing to their sub-micron thickness, their limited active volume substantially restricts the total storable energy, making them less suitable for macro-scale applications compared to MLCCs^{3,14}. Nevertheless, the energy-storage performance of MLCCs remains fundamentally limited by the intrinsic properties of their ceramic dielectrics, particularly the challenge of achieving both high W_{rec} and high energy efficiency (η) simultaneously.

The energy storage performance of dielectric materials is governed by their polarization response under electric fields, where achieving high maximum polarization (P_{max}) with minimal hysteresis is essential for obtaining high W_{rec} and η ⁴⁻⁸. Recent advances have explored various strategies to optimize polarization response, including nanodomain engineering^{8,11,16}, superparaelectric design^{17,18}, defect engineering¹⁹, multiphase construction^{3,13}, lattice strain modulation¹⁴, relaxor antiferroelectric (AFE) strategy^{20,21} and composite ceramic design^{9,22}, etc. Especially, high-entropy ferroelectric (FE) materials featuring multi-elemental occupation of

equivalent lattice sites have recently demonstrated exceptional potential for energy storage because that the atomic-scale compositional heterogeneity induces substantial local structural disorder and generates large random electric/stress fields^{3,12,23-26}. These features are highly desirable for enhancing energy storage performance. The above advancements have also catalyzed extensive exploration of various lead-free FE and AFE ceramic systems, including BaTiO₃ (BT)^{3,11,13,18,23,26}, Bi_{0.5}Na_{0.5}TiO₃ (BNT)^{9,10,14}, K_{0.5}Na_{0.5}NbO₃ (KNN)^{27,28}, BiFeO₃ (BF)^{16,25}, NaNbO₃ (NN)^{20,24}, and AgNbO₃ (AN)-based materials^{19,21}. Despite these advances, the intrinsic trade-off between W_{rec} and η remains a critical bottleneck in both relaxor FEs and AFEs. Excessive polarization hysteresis and consequent η degradation under high electric fields continue to impede progress, primarily due to the electric-field-induced growth of nanodomains. Therefore, the synergistic achievement of superior W_{rec} ($>15 \text{ J cm}^{-3}$) and ultrahigh η ($>90\%$) presents a formidable challenge in lead-free ceramics and MLCCs (Table S1, Supplementary Information)^{4,7,9-16,18-30}.

Herein, we propose a novel strategy of non-polar nanocluster confinement to inhibit the electric-field-induced growth of polar nanodomains in high-entropy relaxor FEs, thereby enabling concurrent enhancement of polarization and reduction of hysteresis, as schematically illustrated in Fig. 1. We selected a representative high-entropy relaxor composition, (Bi_{0.2}Na_{0.2}Ba_{0.2}Sr_{0.2}Ca_{0.2})TiO₃, which exhibits significant A-site cationic disorder, as the matrix³¹⁻³³. Sn⁴⁺, which possesses extremely low ionic polarizability compared to other commonly used B-site cations, was introduced at the B-site^{34,35}. Given that SrSnO₃, CaSnO₃, and BaSnO₃ are all non-FE perovskites^{36,37}, the Sn-rich regions that form at the atomic scale are expected to behave as non-polar clusters, exhibiting limited responsiveness to electric fields and thus not contributing to polarization growth. Furthermore, the incorporation of Sn⁴⁺ coupled with different A-site cations introduces varied lattice distortions, which enhances local random fields. This effect not only reduces the size of the polar domains but also further confines the Sn-rich non-polar nanoclusters. Under increasing electric fields, these confined non-polar regions effectively impede the merging and growth of adjacent polar nanodomains, as depicted in Fig. 1a. This design concept is

corroborated by our phase-field simulations of the domain structure evolution in $(\text{Bi}_{0.2}\text{Na}_{0.2}\text{Ba}_{0.2}\text{Sr}_{0.2}\text{Ca}_{0.2})(\text{Ti}_{1-x}\text{Sn}_x)\text{O}_3$ (BNBSCT_{1-x}S_x) ceramics (Fig. 1b). The simulated P - E loops demonstrate an improved polarization-field response along with significantly reduced hysteresis (Fig. 1c). Experimentally, we realized a desirable polarization-field response characterized by an extremely low hysteresis and a high $P_{\max}$ in BNBSCT_{1-x}S_x bulk ceramics with $x = 0.06$. This yields a superior W_{rec} of $\sim 18.5 \text{ J cm}^{-3}$ and an ultrahigh η of $\sim 92.4\%$ under $\sim 108 \text{ kV mm}^{-1}$, representing a notable advancement in the comprehensive energy-storage properties of lead-free MLCCs.

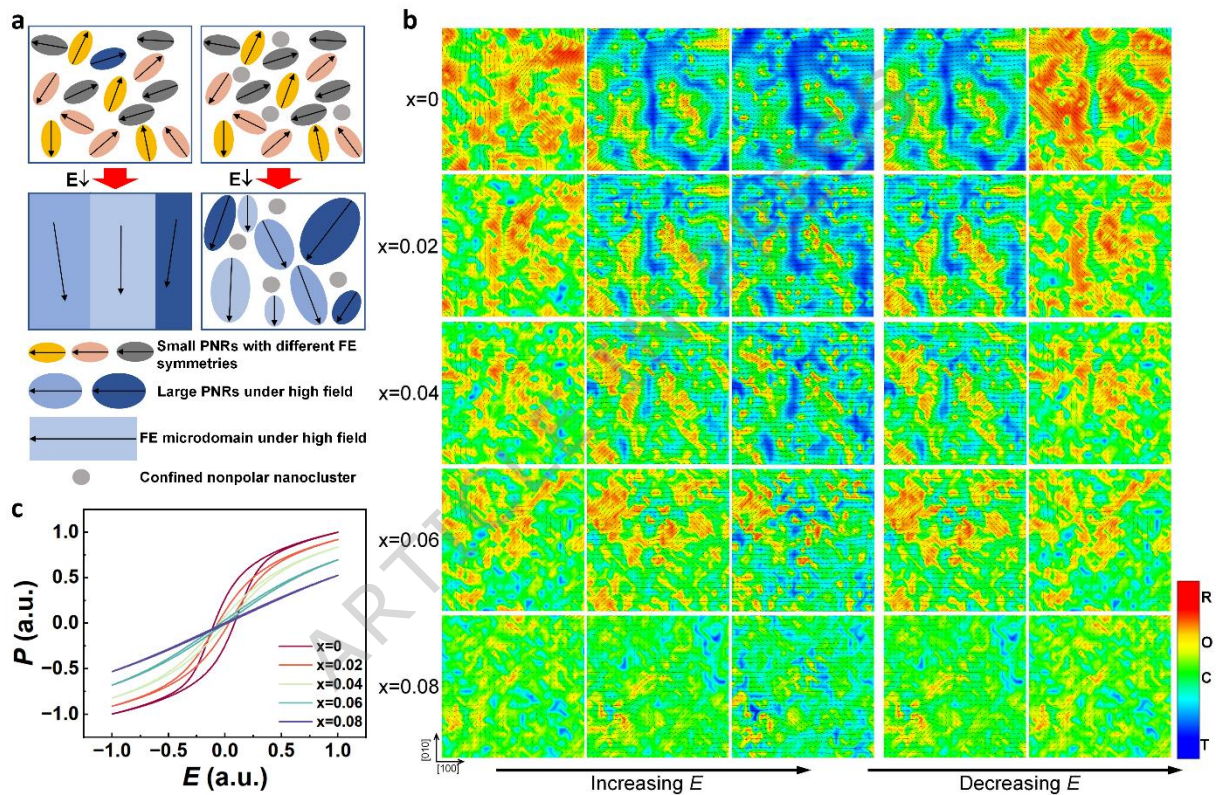


Fig. 1: Strategy for non-polar nanocluster confinement engineering in energy-storage relaxor FEs. a Schematic ground state and high-field state of relaxor FEs with/without confined non-polar nanoclusters. b Phase-field simulation of domain structure evolution during loading/unloading electric fields of BNBSCT_{1-x}S_x compositions. c Simulation results of P - E loops of BNBSCT_{1-x}S_x compositions (R: rhombohedral; O: orthorhombic; T: tetragonal; C: cubic).

Results and Discussion

Guided by the phase-field simulation predictions, we designed and synthesized a series of

BNBSCT_{1-x}S_x high-entropy ceramics. X-ray diffraction patterns (Fig. S1) confirms the formation of single-phase perovskite structures across all compositions. Microstructural analysis reveals dense ceramic packing with well-defined grain boundaries (Fig. S2), where the refined grain morphology significantly enhances $E_B^{7,29}$. To experimentally validate the phase-field predictions regarding domain confinement, we investigated the electric-field-induced domain evolution using in-situ piezoresponse force microscopy (PFM), as shown in Fig. 2a. At low voltage (10 V), featureless amplitude images indicative of polar nanoregions are observed across all compositions, confirming their relaxor FE nature^{38,39}. This behavior is further evidenced by temperature-dependent dielectric spectroscopy (Fig. S3), revealing typical relaxor characteristics^{40,41}. These characteristics likely originate from strong A-site ion disorder, which induces large local random electric/stress fields^{40,41}. Under higher voltages (20-30 V), the PFM amplitude responses diverge significantly. The $x = 0$ sample develops micron-scale FE domains exhibiting strong piezoelectric activity, indicating an electric-field-induced transition from short-range to long-range polar order. In contrast, Sn-doped specimens maintain significantly finer domain configurations, demonstrating that this transition is progressively inhibited with increasing Sn content, which is consistent with our phase-field simulation results. This suppressed domain evolution is directly reflected in the field-dependent polarization responses. As displayed in Fig. 2b and S4, increasing Sn content induces remarkable slimming of P - E loops, coupled with broadening and suppression of current density peaks in current density-field (J - E) curves. It is noted that the current density does not return to zero when the electric field is withdrawn. This is a common feature observed in dielectric and FE materials, which can be understood from the fundamental relationship $J \propto dP/dt = (dP/dE) \cdot (dE/dt)^{42}$. Under a triangular voltage waveform, dE/dt value is constant, and dP/dE is proportional to the differential dielectric constant of the material^{42,43}. Thus, a finite background current is a natural manifestation of the material's intrinsic dielectric polarizability rather than an artifact or a significant loss mechanism^{42,43}. This physical interpretation confirms that the observed non-zero current does not contradict the low-energy-loss characteristics of our ceramics. The

optimized $x = 0.06$ composition achieves significantly improved energy storage performance in bulk ceramic form with $W_{rec} = 8.1 \text{ J cm}^{-3}$ and $\eta = 95.0\%$, representing 40% and 22% improvements respectively compared to the undoped counterpart. While W_{rec} decreases slightly with further Sn addition, η remains around 95.0%, likely due to a gradual reduction in P_{max} associated with increased non-polar phase content¹⁷.

Comparative analysis of field-dependent polarization behavior for two representative compositions (Fig. 2d-g) reveals fundamentally different energy loss mechanisms. The undoped ceramic ($x = 0$) exhibits pronounced polarization hysteresis under increasing fields, manifested by the rapidly enlarging difference between the total energy storage density W_{total} and W_{rec} and a dramatic η decrease from 96% to 78% as the field intensifies from 5 kV mm^{-1} to 40 kV mm^{-1} . In striking contrast, the $x = 0.06$ composition maintains significantly suppressed polarization hysteresis and ultrahigh efficiency ($\eta \geq 95\%$) even at 55 kV mm^{-1} , demonstrating superior field stability. Analysis of the dynamic scaling behavior further supports this conclusion: the $x = 0.06$ ceramic exhibits a relatively small exponent α value in its two linear stages compared to the $x = 0$ ceramic^{12,28}. Moreover, the turning point between these stages occurs at higher electric fields for $x = 0.06$. These results confirm that Sn^{4+} introduction effectively suppresses the electric-field-driven growth process from polar nanodomains to micrometer-scale FE domains, resulting in minimal polarization hysteresis even under high electric fields^{12,28}.

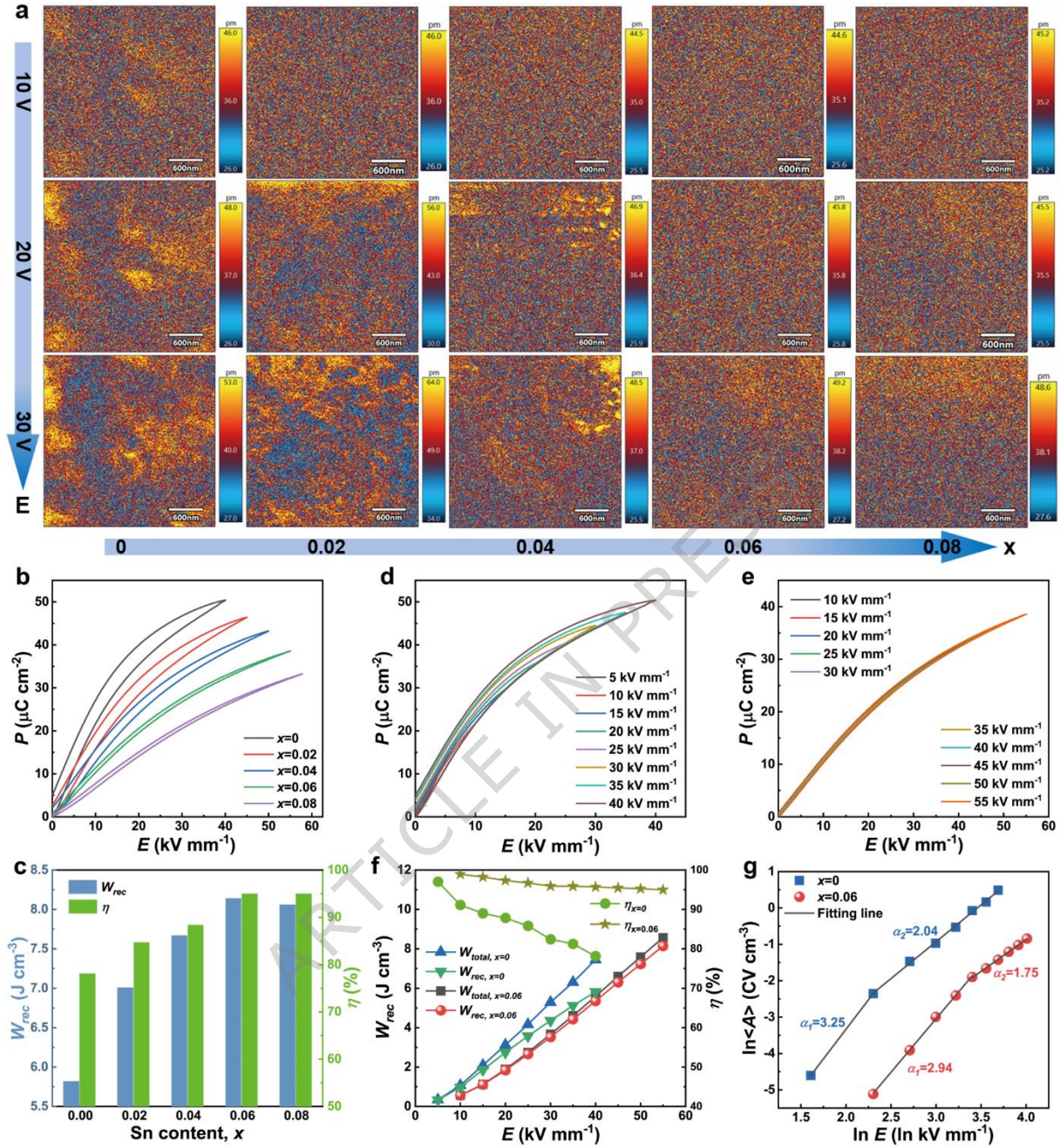


Fig. 2: Polarization-field response characteristics and energy-storage performances of BNBSCT_{1-x}S_x bulk ceramics. **a** In-situ PFM results of the BNBSCT_{1-x}S_x ceramics measured under different voltages. **b** The P - E loops of BNBSCT_{1-x}S_x ceramics measured under the maximum testable electric fields and 10 Hz, and **c** corresponding W_{rec} and η values. P - E loops of the **d** $x = 0$ and **e** $x = 0.06$ ceramics measured at different electric fields and 10 Hz. **f** Energy-storage properties and **g** curves of $\ln\langle A \rangle$ as a function of $\ln E$ for the $x = 0$ and $x = 0.06$ ceramics.

The mechanism behind the suppressed field-induced domain evolution was elucidated through multiscale structural characterization. Atomic-resolution energy-dispersive X-ray spectroscopy (EDS) mapping (Fig. 3a) reveals distinct elemental distribution patterns. Primary elements in the lattice, namely A-site cations (Ba, Bi, Ca, Sr) and B-site Ti, exhibit well-defined periodic distributions consistent with the atomic lattice, indicating a disordered coexistence of multiple elements occupying equivalent lattice sites. In contrast, Na and Sn display a random arrangement with significant intensity fluctuations. The weak signal from Na is related to its low atomic weight, whereas the sparse distribution of Sn is attributed to its low concentration. Although Sn demonstrates macroscopically uniform distribution similar to other elements (Fig. S5), atomic-scale heterogeneity is evident, suggesting local compositional variation and structural non-uniformity. This observation is further supported by the atomic fraction variation of A-site and B-site elements (Fig. S6 and Fig. 3a, respectively), where Sn exhibits relatively disordered changes compared to other elements.

Transmission electron microscopy (TEM) analysis of the $x = 0.06$ ceramic (Fig. 3b) shows no discernible FE domain contrast, indicating the absence of long-range FE order. High-resolution TEM images and corresponding selected-area electron diffraction (SAED) patterns acquired along the $[100]_c$ and $[110]_c$ zone axes (Fig. 3c) further confirm the lack of large-scale FE or ferroelastic domains across multiple length scales, supporting the effective disruption of long-range ferroic ordering. To further probe the local polarization configurations, we acquired high-angle annular dark-field (HAADF) images with Z-contrast along the $[001]_c$ and $[110]_c$ directions (Fig. 3d and 3e, respectively). Local polarization vectors were mapped by quantifying the displacements of B-site cations relative to their four nearest A-site neighbors, with arrow color and length indicating the direction and magnitude of polarization, respectively. Along the $[100]_c$ zone axis (Fig. 3d), the T phase exhibits polarization oriented along $[001]$, while both R and O phases show alignments along $[110]$. Along the $[110]_c$ zone axis (Fig. 3e), the R and O phases are distinguishable by their polarization directions: $[111]$ for R and $[110]$ for O.^{2,15,43} Thus, our analysis reveals localized

regions with distinct polar symmetries (R, O, T). The coexistence of PNRs with multiple FE symmetries reduces polarization anisotropy, implying a low energy barrier for polarization reorientation. This is in favor of realizing a high P_{max} . Critically, we identified regions exhibiting near-zero polarization magnitude, which we attribute to non-polar clusters. These are likely caused by local compositional fluctuations, as supported by intensity variations in the fitted B-site and A-site atomic columns (Fig. 3f-g and Fig. S6). Notably, some non-polar nanoclusters (yellow dashed circles in Fig. 3d-e) exhibit enhanced B-site intensity, suggesting Sn-rich regions. The extremely low ionic polarizability of Sn^{4+} renders these Sn-rich clusters less responsive to electric fields. Furthermore, local unit cells containing Sn^{4+} coupled with different A-site cations (e.g., CaSnO_3 , BaSnO_3 , SrSnO_3) are paraelectric with diverse crystal structures. These unit cells not only resist polarization under external fields but also introduce strong random electric and stress fields due to their distinct structural distortions, further suppressing polarization response. Consequently, these confined non-polar nanoclusters effectively inhibit the merging and growth of adjacent PNRs under electric fields, accounting for the extremely low polarization hysteresis observed in our electrical measurements.

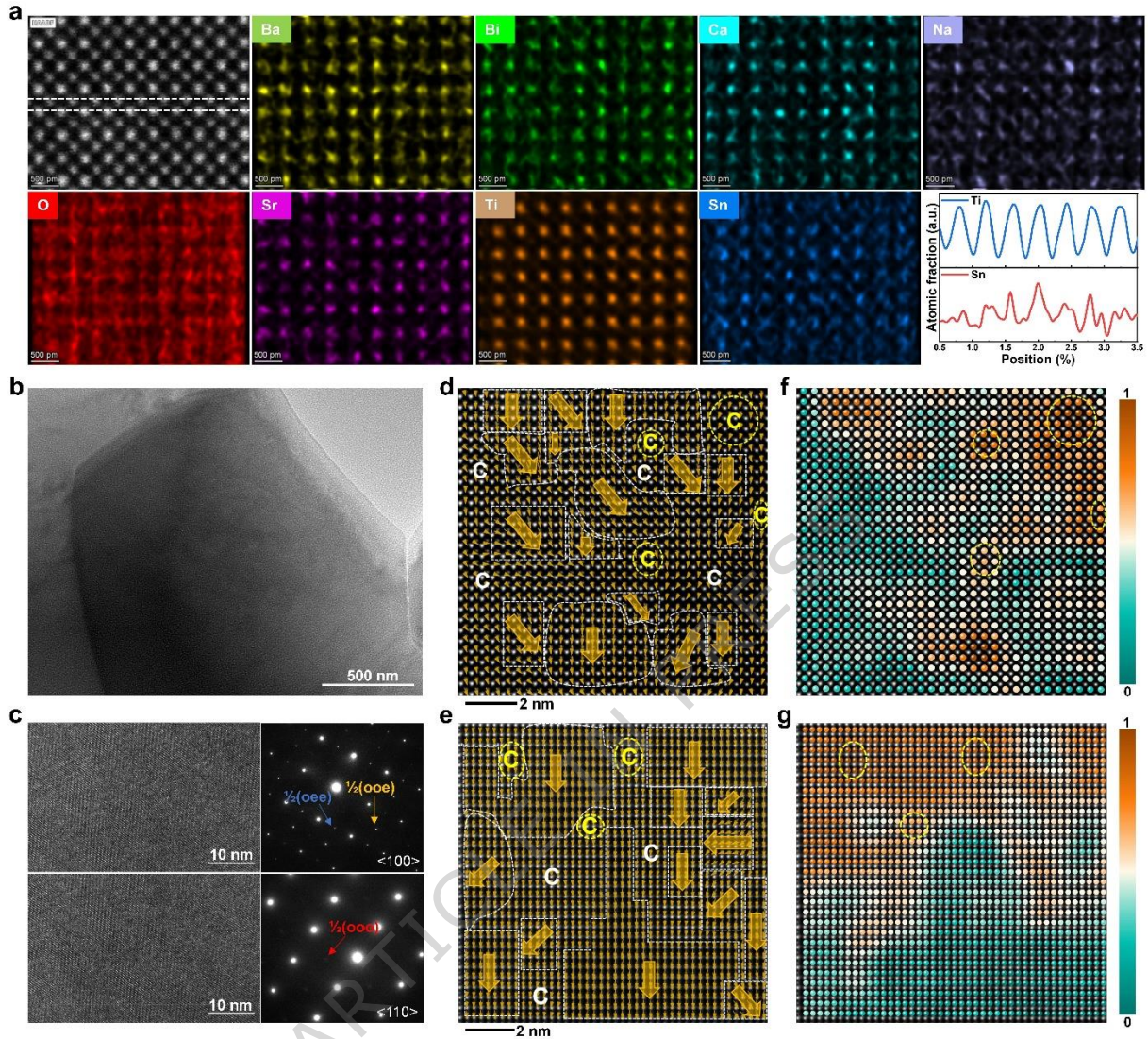


Fig. 3: Hierarchical Domain structures of the $x = 0.06$ ceramic. **a** Atomic EDS mapping of element distributions and atomic fraction fluctuation of Ti and Sn at B site. **b** Bright-field TEM image. **c** HR-TEM and corresponding SEAD images along $[100]_c$ and $[110]_c$ directions. Atomic-resolution HAADF images with cation displacement vectors along **d** $[100]_c$ and **e** $[110]_c$ directions. The intensity distribution of fitted B-site atomic columns **f** $[100]_c$ and **g** $[110]_c$.

MLCCs fabricated with the optimized $x = 0.06$ composition exhibit exceptional structural and functional integrity. Cross-sectional SEM analysis (Fig. 4a) confirms a well-laminated architecture comprising five highly dense dielectric layers (average thickness $\approx 16 \mu\text{m}$) interfaced with continuous metallic electrodes. EDS mapping demonstrates negligible elemental interdiffusion at

electrode-dielectric interfaces, preserving interfacial stability. The thickness of different dielectric layers is precisely controlled within a narrow range of $\pm 0.8 \mu\text{m}$, ensuring uniform field distribution. Field-dependent polarization responses (Fig. 4b-c) highlight the material's superior energy storage dynamics. The slim P - E loops exhibit strongly field-amplified P_{max} ($49.8 \mu\text{C cm}^{-2}$ at 108 kV mm^{-1}) with ultralow remnant polarization ($P_r < 0.8 \mu\text{C cm}^{-2}$), corresponding to hysteresis losses below 8% of total energy input. This behavior directly manifests the effectiveness of the non-polar nanocluster confinement mechanism in suppressing long-range FE ordering under high electric fields while enabling rapid polarization reversal, which is also consistent with the phase-field simulation results. Consequently, a large W_{rec} of 18.5 J cm^{-3} and a high η of 92.4% were obtained under 108 kV mm^{-1} (Fig. 4d). As summarized in Fig. 4e and Tabs S1 (Supplementary Information), this performance surpasses most recently reported bulk ceramics and MLCCs, demonstrating the exceptional efficacy of our design strategy^{3,4-7,9-16,18-30,44-50}.

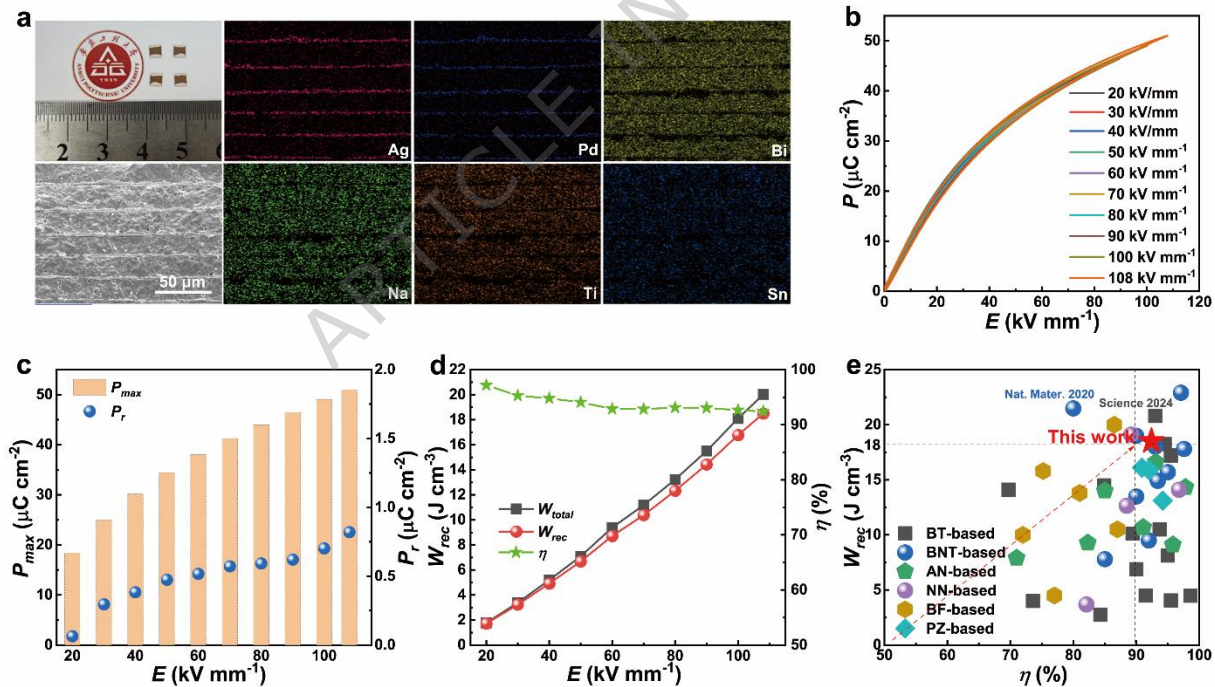


Fig. 4: Energy-storage performances of the $x = 0.06$ MLCCs. **a** The optical photo, cross-section image and EDS images of representative elemental distributions of the MLCCs. **b** P - E loops and **c** P_{max} and P_r values of the MLCCs measured at various electric fields and 10 Hz. **d** Field-dependent W_{rec} and η values of the MLCCs. **e** A comparison of energy-storage performances between the $x = 0.06$ MLCC and other recently-reported MLCCs^{3,4-7,9-16,18-30,44-50}.

The temperature stability of energy-storage performance is crucial for practical applications^{7,51,52}. Temperature-dependent in-situ Raman spectra (Fig. 5a) reveal minimal changes in peak shape and intensity between 25 °C and 175 °C, indicating temperature-insensitive local structural features^{49,52}. The distinctive broadening and smoothing of Raman peaks at 200 °C correlate with reduced unit cell polarity. This remarkable thermal stability is attributed to the complex lattice stress conditions induced by the heterogeneous nanodomains within the material^{52,53}. The variation of capacitance ($\Delta C/C$) at 0.1-10 kHz is less than 15% over a wide temperature between 0 °C and 250 °C (Fig. 5b), further confirming the temperature-insensitive structural features⁴⁹. Furthermore, the material exhibits suppressed defect mobility at high temperatures. This is evidenced by effectively suppressed high-temperature dielectric loss ($\tan \delta$; Fig. 5b), indicating inhibited defect mobility that significantly reduces high-temperature energy losses^{7,51}. Consequently, temperature-dependent P - E loops measured over a broad temperature range (20-170 °C) demonstrate exceptional thermal stability, maintaining low hysteresis and high P_{max} (Fig. 5c). This results in minimal variations in energy storage parameters ($\Delta W_{rec} < 2\%$, $\Delta \eta < 2\%$; Fig. 5e). Moreover, across a wide frequency range (1-100 Hz), the MLCCs exhibit outstanding frequency stability (Fig. 5d), characterized by $W_{rec} = 8.9 \pm 0.1 \text{ J cm}^{-3}$ and $\eta = 93.4\%$ with variations less than 2% (Fig. 5f).

The charge/discharge performance further confirms the application potential of our MLCCs. Regular underdamped discharge waveforms are observed under electric fields from 20 to 60 kV mm⁻¹ (Fig. 5g). The inset of Fig. 5g shows that the power density (P_D) increases with electric field, reaching an ultrahigh value of $\approx 169.9 \text{ MW cm}^{-3}$ at 60 kV mm⁻¹. For overdamped discharge processes, the $x = 0.06$ MLCC exhibits stable oscillating waveforms from 20 to 60 kV mm⁻¹ with a fixed load resistance (Fig. S8). As shown in Fig. 5h, an ultrahigh discharge energy density (W_D) of $\approx 7.4 \text{ J cm}^{-3}$ is achieved at 60 kV mm⁻¹, with 90% of this energy released within $\approx 52.0 \text{ ns}$. This rapid discharge speed is attributed to the fast polarization-field response, as evidenced by the

frequency-insensitive P - E loops in Fig. 5d. These comprehensive results demonstrate that the $x = 0.06$ MLCCs exhibit outstanding overall properties with excellent stability, showing promising application prospects for advanced pulse-power capacitors.

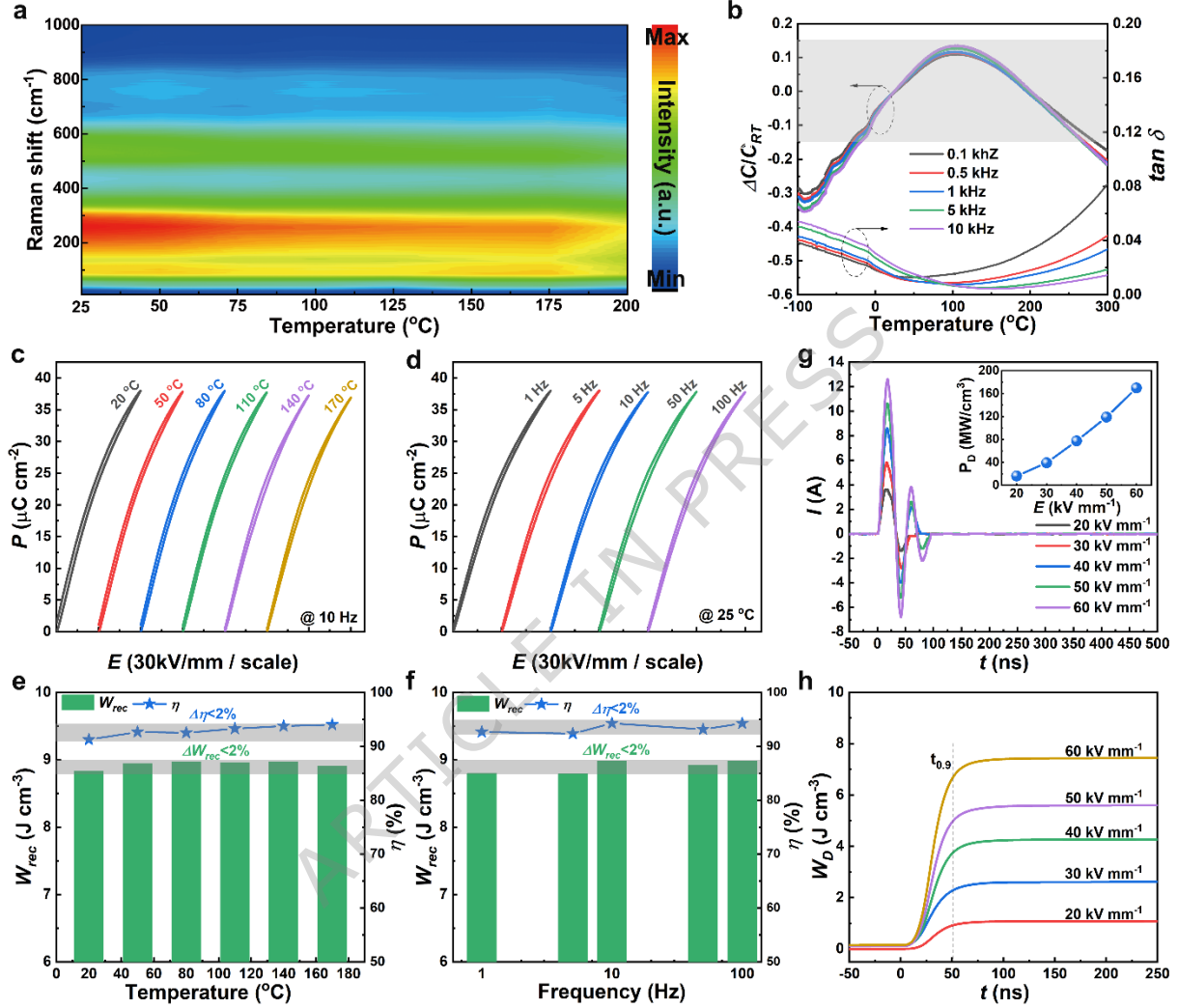


Fig. 5: Stability and charge/discharge performances of the $x = 0.06$ MLCCs. Temperature dependent **a** Raman spectra and **b** $\tan \delta$. **c** P - E loops under different temperatures at 60 kV mm^{-1} and 10 Hz . **d** Room-temperature P - E loops under different frequencies at 60 kV mm^{-1} . **e** W_{rec} and η as a function of temperature at 60 kV mm^{-1} . **f** W_{rec} and η as a function of frequency at 60 kV mm^{-1} . **g** Overdamped discharge waveforms and field-dependent P_D values. **h** W_D as a function of electric fields.

In summary, we have demonstrated an effective strategy of non-polar nanocluster confinement engineering to mitigate the long-standing trade-off between W_{rec} and η in dielectric ceramics via a rationally designed high-entropy BNBSCT_{1-x}S_x system. By leveraging phase-field simulations and analyzing multiscale structure evolution, we found that the Sn-riched non-polar nanoclusters within a high-entropy relaxor FE matrix are hardly responsive to external electric fields owing to the weakly FE-active Sn⁴⁺ ions and large random fields. These confined nanostructures act as non-polar pinning centers to inhibit the polar nanodomain merge during high electric field loading, achieving a collaborative optimization of polarization and hysteresis. The optimized composition yields an excellent energy-storage performance in MLCCs, achieving a W_{rec} of $\sim 18.5 \text{ J}\cdot\text{cm}^{-3}$ together with an ultrahigh η of $\sim 92.4\%$. This represents a competitive combination of properties among state-of-the-art lead-free counterparts. Our work not only provides a viable design pathway toward high-efficiency, high-energy-density dielectrics for next-generation power electronics but also offers broader insights into the control of nanoscale polarization behavior in functional materials through local compositional engineering.

Methods

Fabrication of bulk ceramics and MLCCs

The BNBSCT_{1-x}S_x perovskite powders were synthesized via a conventional solid-state reaction route employing high-purity precursors (> 99%), including BaCO₃, CaCO₃, SrCO₃, Na₂CO₃, Bi₂O₃, TiO₂ and SnO₂. The powders were weighed, thoroughly mixed, and then ball-milled in ethanol using zirconia grinding media for 6 hours. The dried mixture was subsequently calcined at 900 °C for 8 hours in covered alumina crucibles. For bulk ceramic samples, the calcined powders were compacted into pellets and sintered at 1180-1220 °C for 2 hours within sealed crucibles. For MLCC fabrication, the calcined powder with composition $x = 0.06$ was dispersed in a solvent along with a dispersant and ball-milled for 12 hours. Binders and plasticizers were then introduced, followed by additional milling for another 12 hours. The resulting slurry was filtered, de-aired,

and tape-cast to form green sheets. Ag/Pd electrode paste was screen-printed onto these sheets, which were then stacked, laminated (50 °C, 5 minutes), and subjected to warm isostatic pressing (50 MPa, 55 °C) to improve green density. Binder removal was conducted via thermal debinding at 600 °C for 2 hours with a heating rate of 0.5 °C/min. The MLCCs were finally sintered at 1210 °C for 2 hours.

Structure Characterizations

The XRD data was collected on an X-ray diffractometer (D/Max-RB, Rigaku, Japan). The grain morphology of the ceramics was taken by a field-emission scanning electron microscope (FE-SEM, SU8020, JEOL, Japan). The domain morphology evolution with bias voltage was performed by using a PFM (AsylumResearch MFP-3D Origin+, Oxford Instruments, UK). Raman spectra were collected on well-polished samples through a Raman spectrometer (LabRam HR Evolution, HORIBA JOBIN YVON, Longjumeau Cedex, France). The atomic-scale STEM HAADF images were performed on a probe-corrected Hitachi HF5000 at 200 kV, while the domain morphology observations and SEAD measurements were carried out on a JEOL JEM-F200 microscope at 200 kV. Specimens for STEM and TEM measurements were prepared by a conventional approach combining mechanical thinning and finally Ar⁺ ion-milling in a Gatan PIPS II.

Electric Property Measurements

Temperature/frequency-dependent dielectric properties were measured by an LCR meter (E4980A, Agilent Technologies, Inc., USA) connected with a high/low-temperature probing stage (HCT1821, Tongguo technology, China). The *P-E* loops were obtained through an FE measurement system (TF Analyzer 2000E, aixACCT Systems GmbH, Germany). The energy release properties were studied via a charge-discharge platform (CFD-003, Tongguo technology, China).

Grain size distribution

Grain size distribution was analyzed using Nano Measurer software. To ensure statistical representativeness, the average grain size was determined from a sample of 200 grains, with a relative deviation of less than 5%.

Phase-field simulations

The details of phase-field simulation about domain structure and polarization configuration are elaborated in the Supplementary Information.

Acknowledgements

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Author contributions

This work was conceived and designed by A.X. and R.Z. Bulk ceramics and MLCCs were fabricated by A.X., Z.L., X.W., A.T., and Y.Z., who also performed the measurements of energy storage, charge-discharge, and dielectric properties. XRD and PFM characterizations were conducted by A.X., Y.Z., X.G., and X.X. TEM and STEM characterizations were carried out by H.T. and N.L. Raman spectra were collected by X.J. and X.E. Phase-field simulations were

performed by L.H. Funding and laboratory equipment support were provided by A.X., T.H., and R.Z. The manuscript was drafted by A.X. and revised by R.Z. All authors contributed to data analysis and discussions.

Competing interests

The authors declare no competing interests.

Data availability

All data supporting this study and its findings are available within the article and its Supplementary Information. Any data deemed relevant are available from the corresponding author upon request.

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Editor's summary:

The authors reconcile the conflict between polarization and hysteresis by confining polarization response of non-polar nanoclusters in high-entropy relaxors, thus improving the energy storage properties of multilayer ceramic capacitors.

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