

Supporting Information

Improved Energy Storage Performance in Flexible (PbLa)ZrO₃ Thin Films via

Nanocrystalline Engineering

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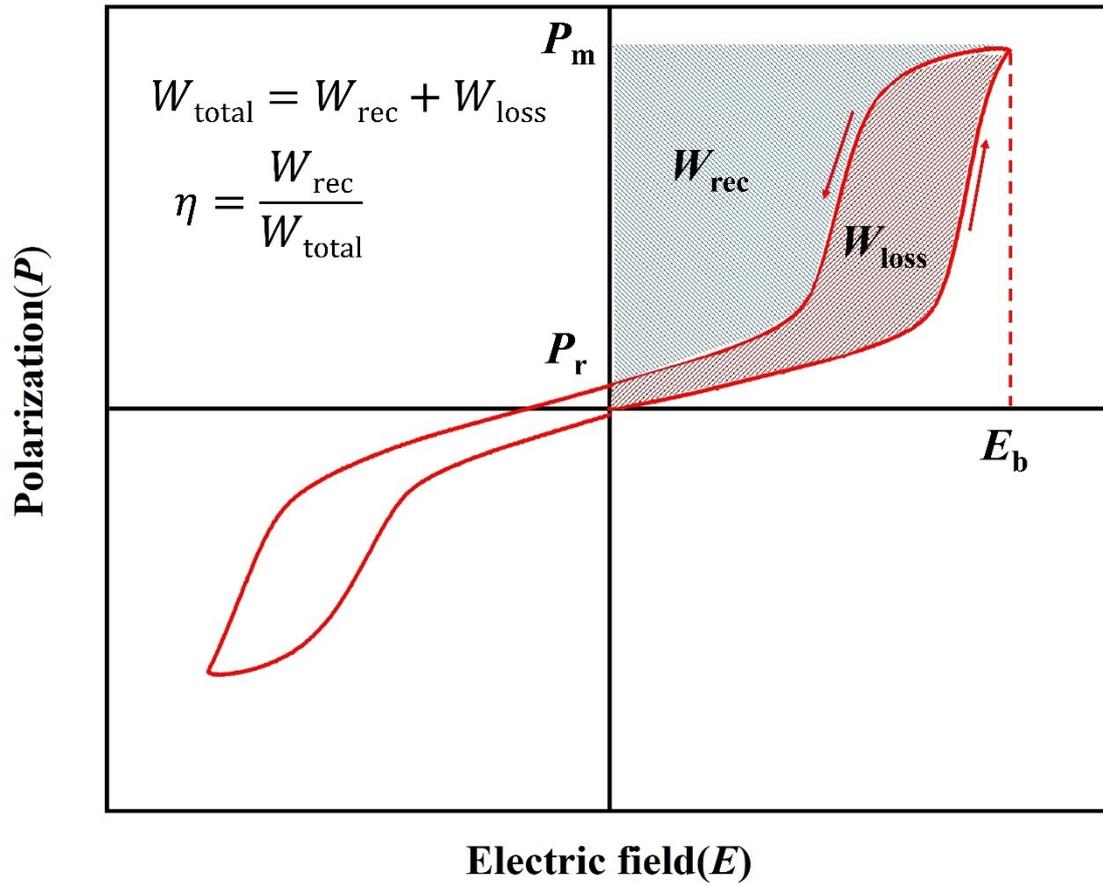


Figure S1. Schematic diagram for the energy storage of antiferroelectric dielectric.

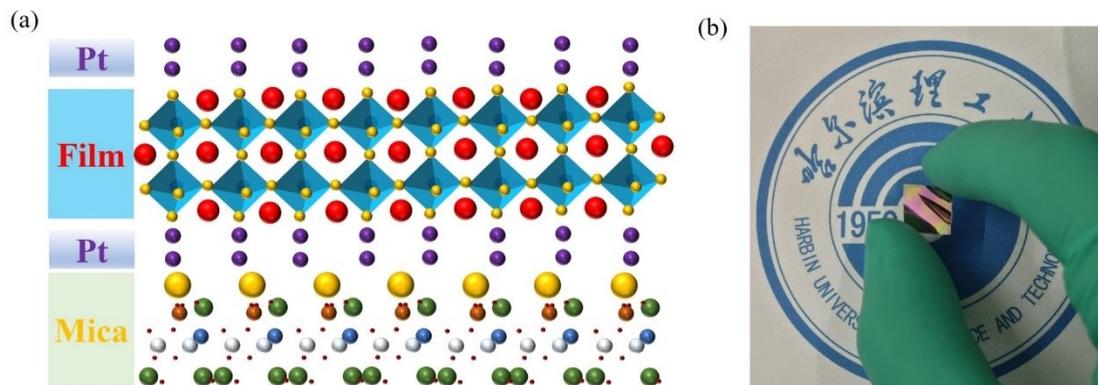


Figure S2. (a) The structure diagram of flexible film, (b) The actual photograph of flexible film.

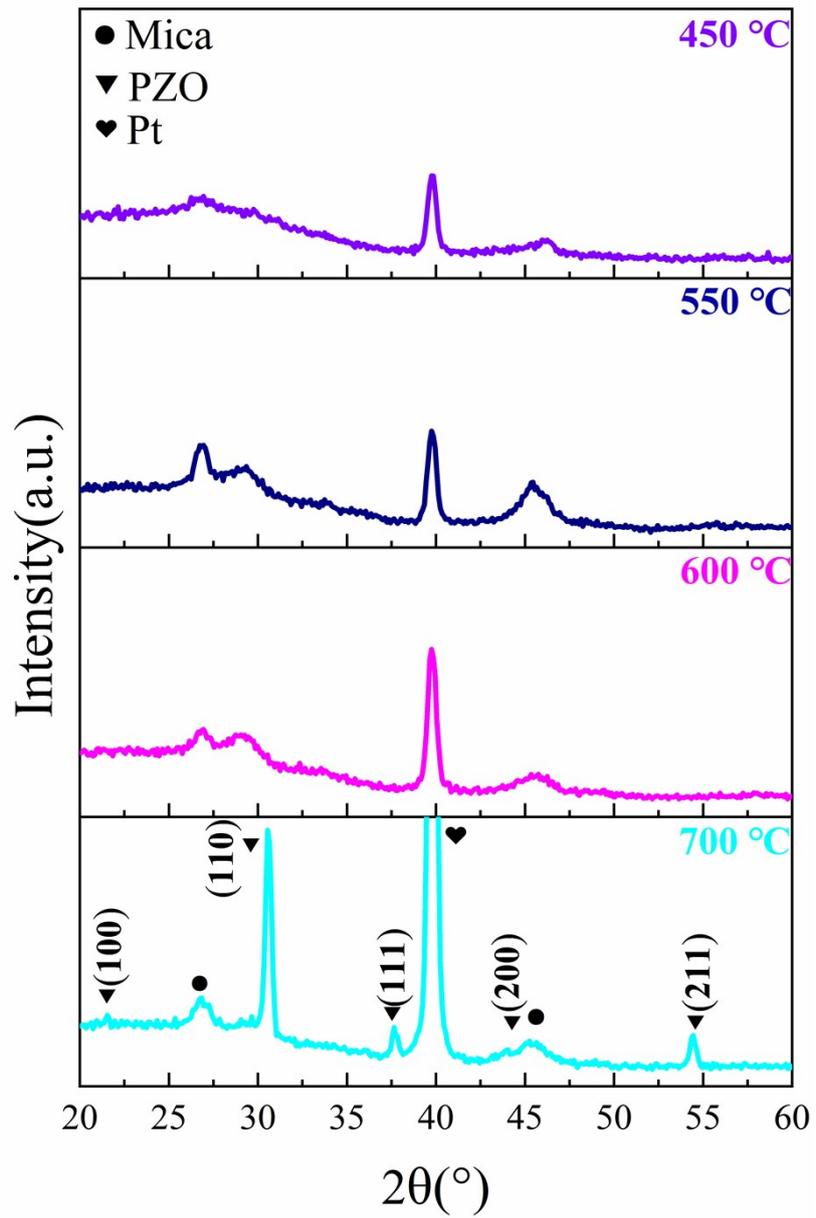


Figure S3. The XRD patterns of flexible films.

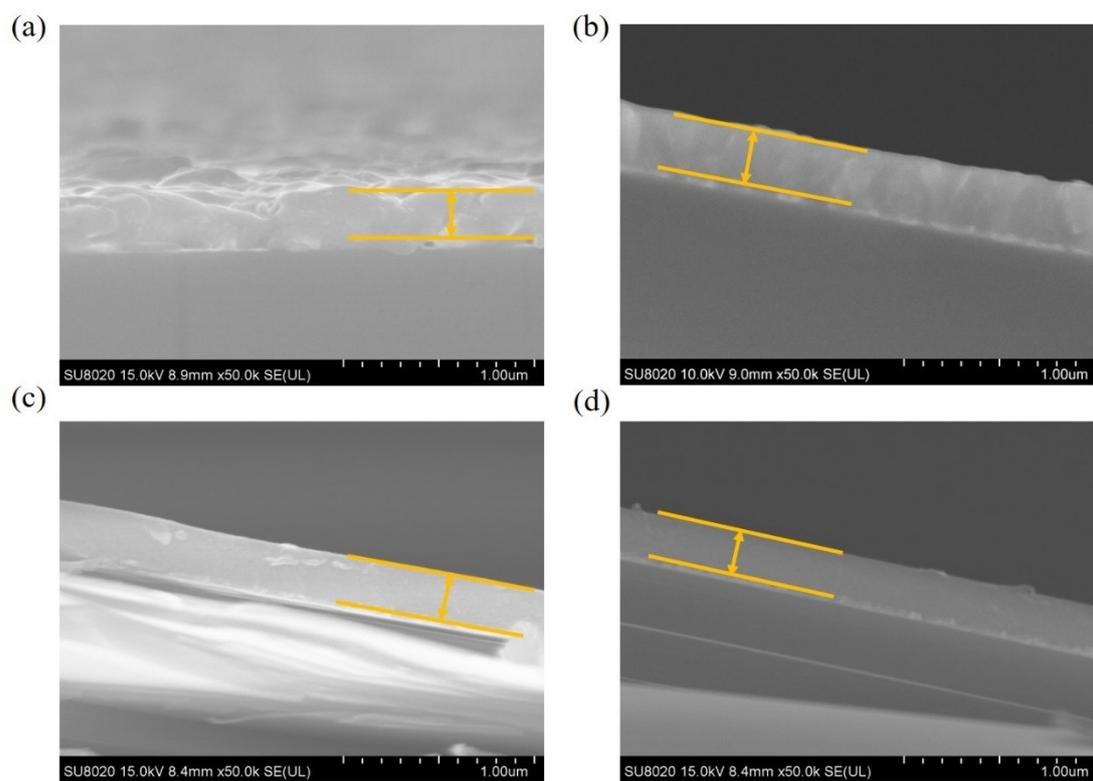


Figure S4. The cross-section SEM micrographs of flexible films at different annealed temperatures. (a) 700 °C, (b) 600 °C, (c) 550 °C, (d) 450 °C

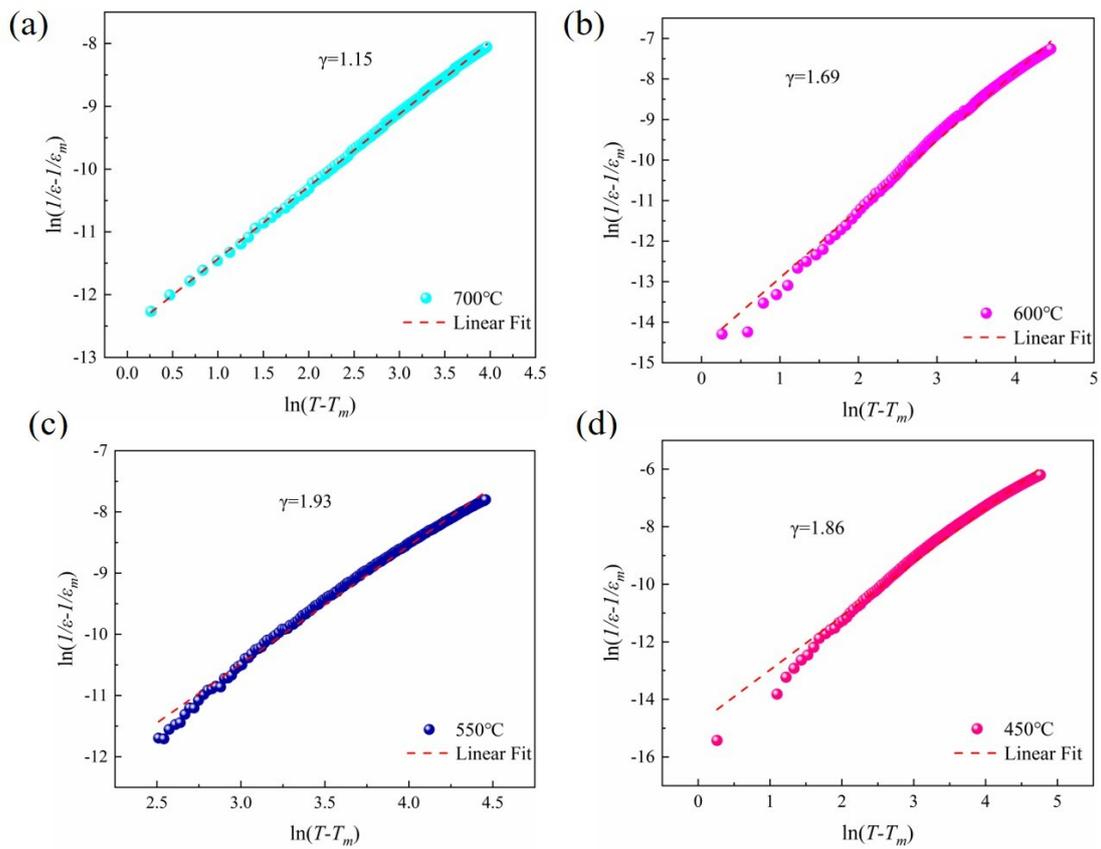


Figure S5. The relationship between $\ln(1/\epsilon - 1/\epsilon_m)$ and $\ln(T - T_m)$ of flexible films at different annealed temperatures. (a) 700 °C, (b) 600 °C, (c) 550 °C, (d) 450 °C.

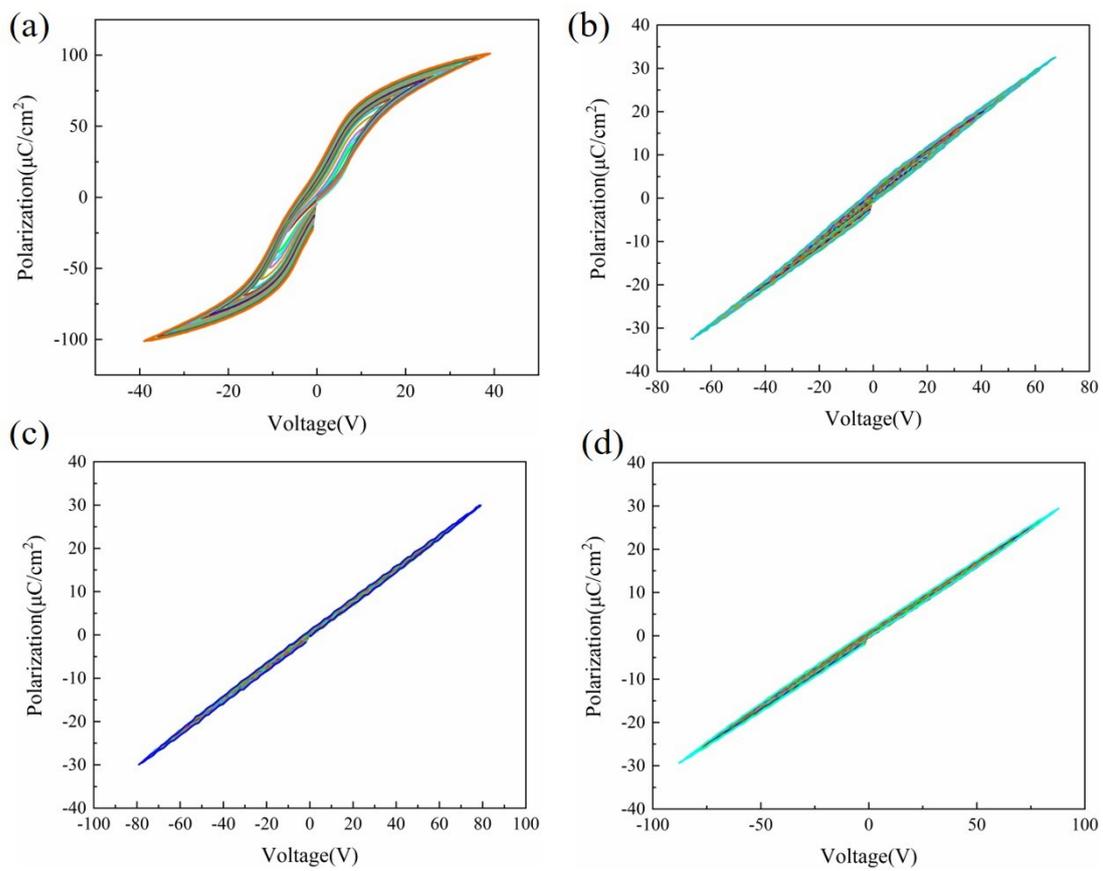


Figure S6. The P - E loops of flexible films at different annealing temperatures. (a)

700 °C, (b) 600 °C, (c) 550 °C, (d) 450 °C.

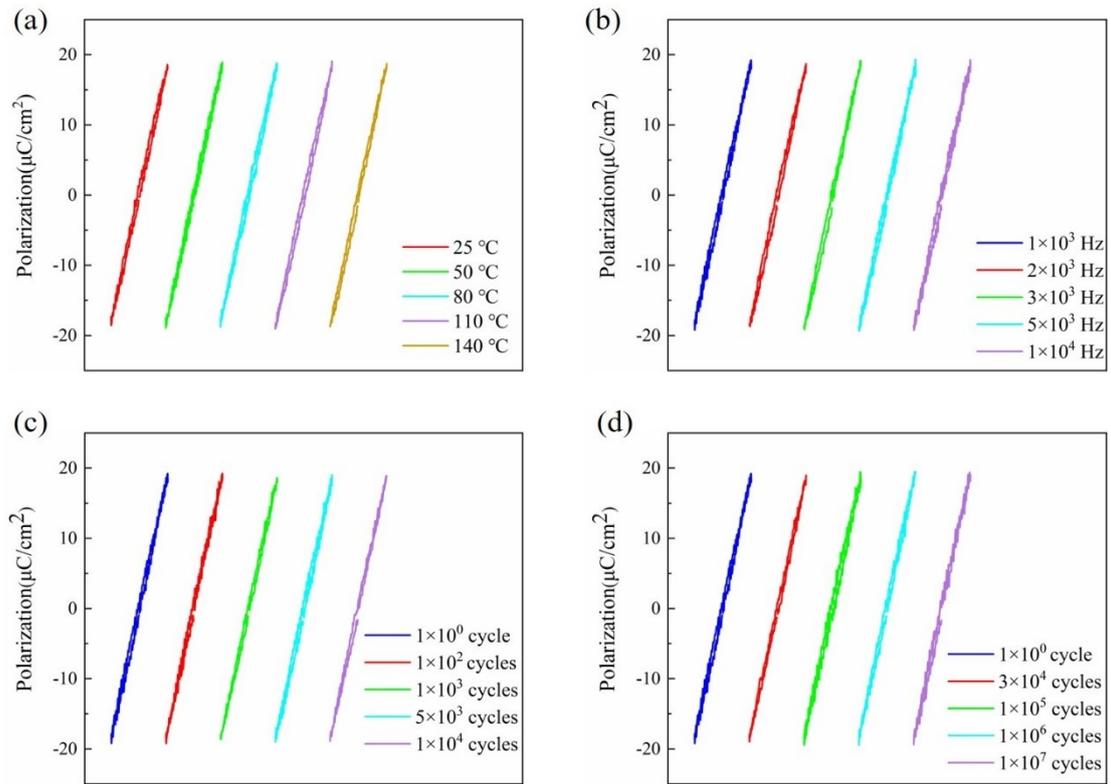


Figure S7. The *P-E* loops of flexible (PbLa)ZrO₃ films annealed at 550 °C under different test conditions. (a) 25-140 °C, (b) 1 k-10 kHz, (c) 10⁴ bending cycles, (d) 10⁷ electric fatigue cycles.

Supplementary Note 1.

The energy storage density (W_{rec}) and charge-discharge efficiency (η) are two key parameters to evaluate the energy storage performance of dielectric films. W_{rec} and η can be expressed by calculating the integral area of the P - E loop. [1]

$$W_{total} = \int_0^{P_m} EdP \quad (1)$$

$$W_{rec} = \int_{P_r}^{P_m} EdP \quad (2)$$

$$\eta = \frac{W_{rec}}{W_{total}} \times 100\% \quad (3)$$

Where P_m and P_r are the maximum polarization and remanent polarization of the dielectrics under an external electric field, respectively.

Supplementary Note 2.

The relaxor dispersion degree can be evaluated using the modified Curie-Weiss equation:[2]

$$1/\varepsilon - 1/\varepsilon_m = (T - T_m)^\gamma / C \quad (4)$$

where γ is the relaxor diffuseness factor, C is Curie constant, and ε_m is the maximum dielectric constant at T_m . $\gamma = 1$ indicates normal ferroelectric and $\gamma = 2$ is corresponds to classical relaxor ferroelectric.

Supplementary Note 3.

The breakdown strength values of the thin films are also analyzed by Weibull distribution: [3]

$$X_i = \ln(E_i) \quad (5)$$

$$Y_i = \ln(-\ln(1 - i/1 + n)) \quad (6)$$

where i is the serial number of the samples, n is the number of samples, E_i is the breakdown strength of each sample, X_i and Y_i are two parameters.

References

- [1] C. Yin, T.D. Zhang, Z.Z. Shi, C.H. Zhang, Y. Feng, Q.G. Chi. High energy storage performance of all-inorganic flexible antiferroelectric–insulator multilayered thin films. *ACS Appl. Mater. Interfaces* 14 (2022) 28997-29006.
- [2] B.B. Yang, Y. Zhang, H. Pan, W.L. Si, Q.H. Zhang, Z.H. Shen, Y. Yu, S. Lan, F.Q. Meng, Y.Q. Liu, H.B. Huang, J.Q. He, L. Gu, S.J. Zhang, L.Q. Chen, J. Zhu, C.W. Nan, Y.H. Lin. High-entropy enhanced capacitive energy storage. *Nat. Mater.* 21 (2022) 1074-1080.
- [3] J. Luo, H. Zhu, T.Y. Zheng, H. Qian, Y.F. Liu, Y.N. Lyu. A slush-like polar structure for high energy storage performance in a $\text{Sr}_{0.7}\text{Bi}_{0.2}\text{TiO}_3$ lead-free relaxor ferroelectric thin film. *J. Mater. Chem. A* 10 (2022) 7357-7365.