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High energy storage efficiency and excellent recoverable energy storage density realized in $0.65Bi_{0.5}Na_{0.5}TiO_3$-$0.35BaTiO_3$-$SrZr_{0.5}Ti_{0.5}O_3$ ceramics

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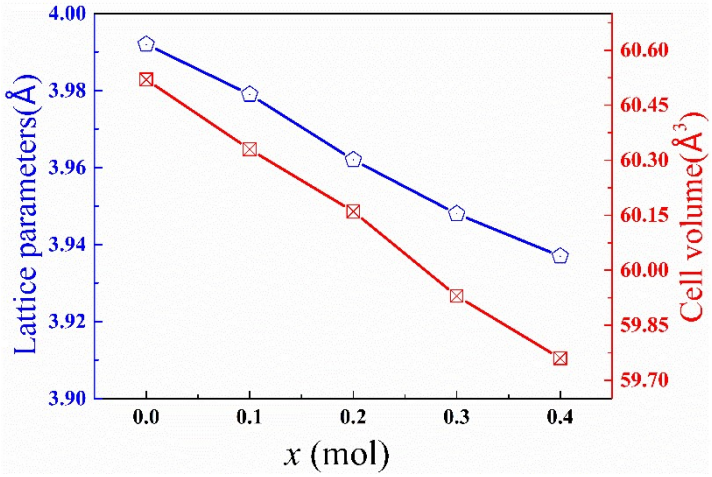

Fig. S1 the corresponding lattice parameters and unit cell volume of the (1-*x*)BBNT-*x*STZ ceramics as a function of *x*.

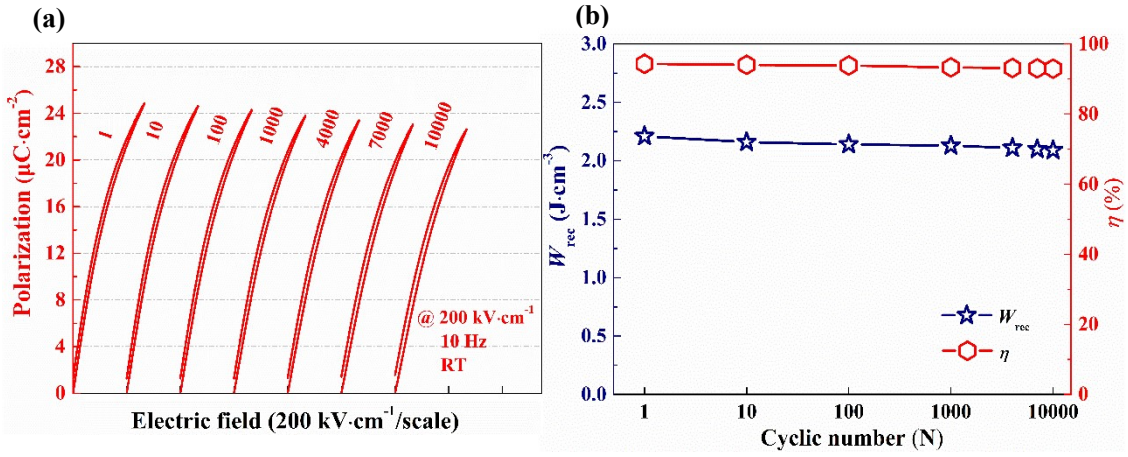

Fig. S2 the unipolar P-E loops (a)and energy storage properties(b) dependence on the fatigue cycles of the 0.7BBNT-0.3STZ ceramic.

Simulation of Dielectric Breakdown Process on Microscale: The development characteristics of breakdown paths in the process of breakdown can be simulated by the finite element method using the breakdown model, as illustrated by the following formula:

$$p(i,j \to i'j') = A\frac{\left(\phi_{i',j'}\right)^m}{\sum\left(\phi_{i',j'}\right)^m} + B\frac{\left(\phi_{i',j'}\right)}{\phi_0} + C$$

where p is the grown probability of electrical tree paths, (i, j) and (i′, j′) represent the coordinates of discrete lattice system in finite element model, φ_0 is the threshold electric potential evaluating the minimum voltage driving electrical tree development, m denotes the fractal dimension whose value is 3 in this work. The first term of the right polynomial manifests the growth direction of electrical trees, the second term reflects the growth difficulty degree of electrical trees, and the last one is related to the dielectric properties of materials. The values of A, B, and C are the weighting coefficients of each term, respectively. In this work, the values of φ_0, A, B, and C for grain boundary are 0.03, 1.0, 8×10^{-16}, and 0.72, respectively, whereas the corresponding values for grain are 0.00054, 1.0, 2.7×10^{-19}, and 0.53, respectively.

The FORC distribution is derived from a series of FORC loops and the Preisach density ρ (α, β) is calculated by the following equation:

$$\rho(\alpha,\beta)=\frac{1}{2}\frac{\partial P^2(\alpha,\beta)}{\partial\alpha\partial\beta}$$

Where ρ (α, β) is the polarization of the FORC loop, α is the reversal electric field, and β is the actual electric field. The Preisach density represents the density distribution of the ideal 'hysteron' and provides the information of the local switching behavior. In this work, we set E_{max}=60 kV·cm^{-1}, Δα=Δβ=4 kV·cm^{-1} and 60 FORC loops were measured.

According to the structural model proposed by Randall et al., the dielectric constant of the grain and grain boundary can be calculated with the macroscopic dielectric constant of the ceramics by the following formula:

$$\frac{1}{\varepsilon'} = \frac{1}{\varepsilon_g'} + \frac{1}{k\varepsilon_{gb}'}$$

Where ε', ε_g' and ε_{gb}' represent the dielectric constant of the ceramics, grain and grain boundary, respectively. k is the ratio of grain size and grain boundary thickness. Based on the above formula and the measured macroscopic dielectric constant and average grain size of BBNT and 0.7BBNT-0.3STZ ceramics, the calculated dielectric constant of the ceramic grain is 2825 for 0.7BBNT-0.3STZ ceramic. The dielectric constant of the grain boundary is estimated to be about 1/10 of the grain.