High capacitive performances obtained in sandwich structured $Bi_{0.5}Na_{0.5}TiO_3$-based dielectric ceramics

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Experimental section

Material preparation: Oxides and carbonates of Bi_2O_3, TiO_2, $CaCO_3$, Na_2CO_3, and $SrCO_3$ (with purities of ≥99.9%, ≥99.5%, ≥99.99%, ≥99.8%, and ≥99.0% respectively) purchased from Sinopharm Chemical Reagent Co., Ltd. (China) were employed as raw materials. Based on the chemical formulas of BNST and BNCT, the aforementioned starting materials were carefully weighed and ball-milled for 24 hours using anhydrous ethanol and zirconia balls as the milling medium. The milled mixtures were dried at 100°C for 8 hours and then calcined in air at 850°C for 4 hours. After that, it was ball-milled again for 12 hours and dried again at 100°C for 8 hours to obtain BNST and BNCT powders. Subsequently, slip casting slurries were prepared using anhydrous ethanol, triolein, acetone, butyl phthalate, polyethylene glycol, polyvinyl butyral, and BNST and BNCT powders. The obtained ceramic slurries were processed through a tape casting process to prepare the BNST and BNCT films, which were then cut into 12mm×12mm squares. After that, four types of ceramics, including the BNST ceramic, the sandwich structured ceramic with BNCT as the intermediate layer and BNST as the outer layer (denoted as SCS), and the reverse sandwich structured ceramic with BNCT as the outer layer and BNST as the intermediate layer (denoted as CSC), as well as the BNCT ceramic, were prepared through lamination and pressing processes. The obtained ceramic green bodies were sintered at 600°C for 12 hours for glue dispensing treatment, then the after gluing ceramic green bodies were heated to 1200°C at a rate of 4°C/min, held for 12 hours, and finally cooled to room temperature with the furnace.

Characterization: The phase structures of the BNST and BNCT ceramics were characterized by an XRD diffractometer (MiniFlex600, Rigaku). Dielectric properties

were measured at different frequencies and within a temperature range of 25-200°C during the heating process using an impedance analyzer (Keysight E4990A). The *P-E* hysteresis loops were detected using a ferroelectric analyzer (PolyK Technologies, LLC State College, PA, USA), and the charge-discharge performance was evaluated using a charge-discharge platform (CFD-003, Gogo Instruments Technology, China). The surface microstructure of the sintered ceramics was characterized using a field emission scanning electron microscope (FE-SEM, S-4200, Hitachi, Tokyo, Japan), and elemental scanning was performed using an energy-dispersive spectrometer (EDS).

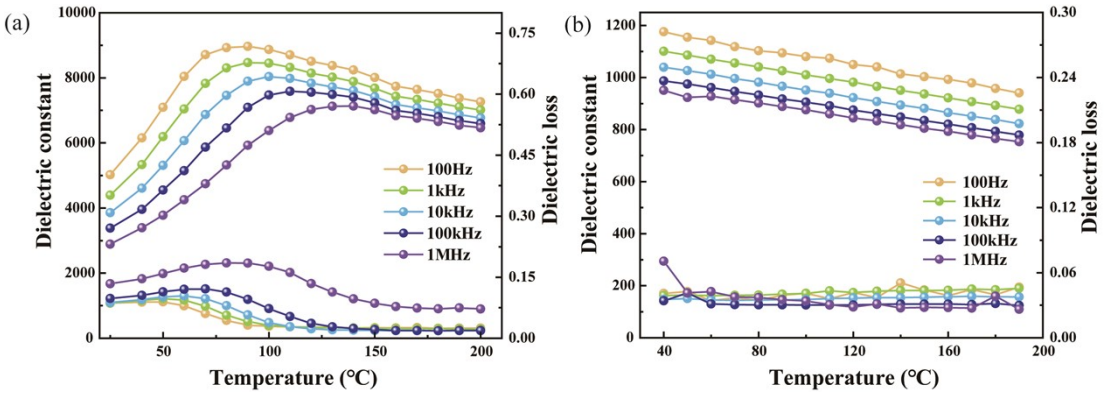

Fig. S1. Dielectric constant and dielectric loss as a function of temperature from 20℃ to 200℃ for (a) BNST, (b) BNCT.

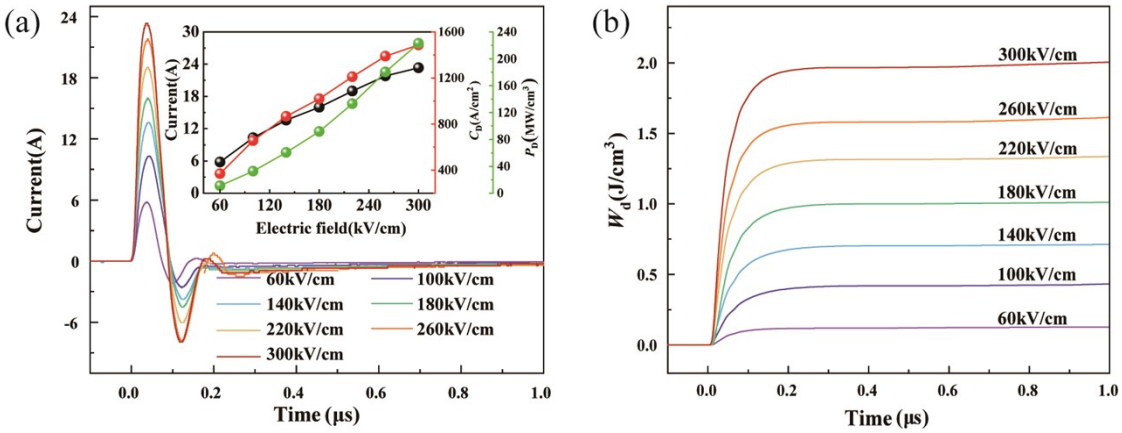

Fig. S2. (a) Underdamped discharge curves of SCS, the insets display the corresponding maximum current, current density and power density as a function of electric field; (b) Discharging energy density versus time for SCS under different electric fields.

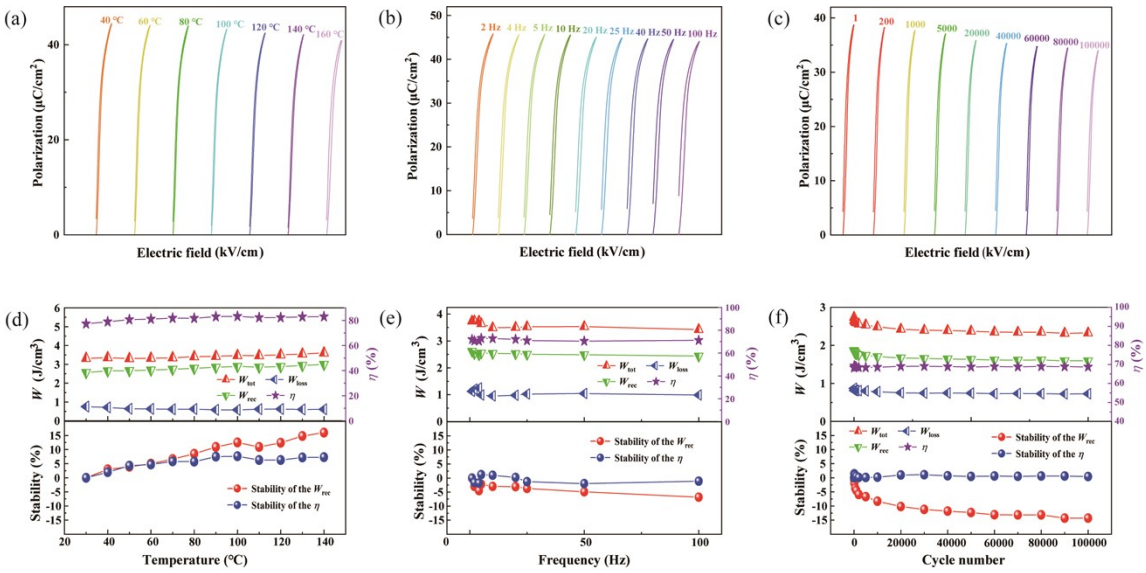

Fig. S3. (a)-(c) Unipolar *P-E* loops within the temperature range of 30-160 ℃, frequency range of 1-100 Hz and cycle numbers of 1-10^5 under 300 kV/cm for SCS; (d)-(e) Energy storage parameters (W_{tot}, W_{rec}, W_{loss} and η) along with the stability of W_{rec} and η at various frequencies, cycle numbers, and temperatures for SCS.

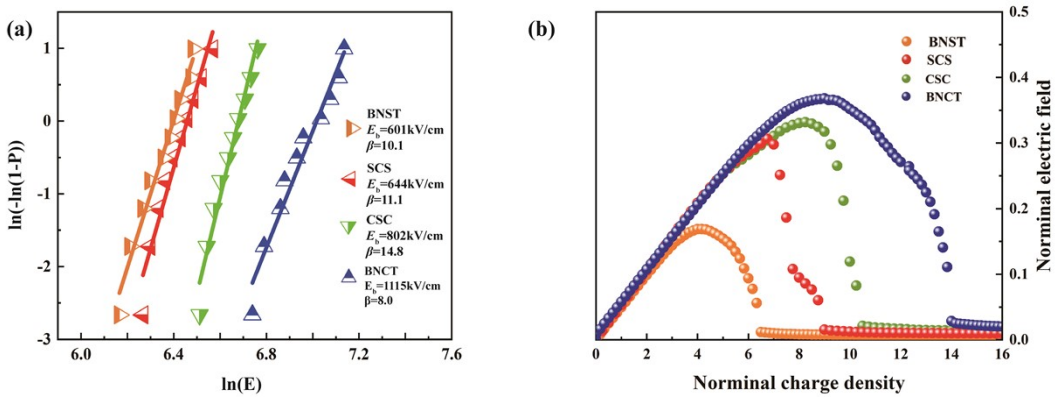

Fig. S4. (a) Weibull statistical failure analysis for the BNST, SCS, CSC and BNCT ceramics; (b) Nominal charge density versus nominal electric field curves for BNST, SCS, CSC and BNCT.