

Supporting Information for

Superior energy storage performance and transparency

in $(\text{K}_{0.5}\text{Na}_{0.5})(\text{Nb}_{0.97}\text{Ta}_{0.03})\text{O}_3$ -based ceramics

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1. Experiments

1.1 Preparation of materials

$(1-3x)(\text{K}_{0.5}\text{Na}_{0.5})(\text{Nb}_{0.97}\text{Ta}_{0.03})\text{O}_3-x\text{LiNbO}_3-2x\text{SrCO}_3$ ceramic ($x = 0.04, 0.06, 0.08, 0.10, 0.12, 0.14$) materials were fabricated by traditional solid-phase way. The raw stuff used in this experiment are Na_2CO_3 (99.5%), K_2CO_3 (99%), Nb_2O_5 (99.5%), Ta_2O_5 (99.99%), Li_2O_3 (99.99%) and SrCO_3 (99.95%). The raw materials are dried in an 90 °C oven for more than 4 h and then weighed. The slurry is ball-milled in a polyethylene bottle using ethanol as a medium for 16 h and then desiccated at 90 °C. After that, the powder is pressed into thin circular plate with a diameter of 30 mm at 8 MPa in a mold using a tablet press. All ceramic fragments are calcined at 950~980°C (980 °C for KNNT-0.04SLN and KNNT-0.06SLN, 970 °C for KNNT-0.08SLN, 960 °C for KNNT-0.10SLN and KNNT-0.12SLN, 950 °C for KNNT-0.14SLN) for 3 h and then ball-milled for another 16 h. Then, the powder is pressed into discs with a diameter of 10 mm at 200 MPa. The pellets are sintered at 1130~1180 °C for 3 h. The samples were ground and polished before testing the hysteresis loops. The thickness and electrode area of samples are 0.07 mm and 1 mm, respectively.

1.2 Characterization

X-ray diffraction (XRD, D8 Advance, Bruker, Germany) is applied to check the structure of the sample. The topography of the samples was investigated by scanning electron microscopy (SEM, Zeiss Ltd., Germany) and transmission electron microscopy (TEM). The elemental composition and distribution is carried out by energy dispersive spectroscopy (EDS). A ferroelectric test system (TF-3000, Aix ACCT, Germany) was applied to test the polarized electric field (P - E) hysteresis loop and the energy storage density was acquired from the P - E loop. The optical transmission spectra were determined by ultraviolet-visible near-infrared spectroscopy (UV-2600, Shimadzu, Japan). The performance on charge/discharge was evaluated by the CFD-003 plus test system (Tongguo Technology Co., Ltd.). The dielectric-temperature spectrum was measured by the high and low temperature dielectric test system (HCT1821, Tongguo Technology Co., Ltd.). The local piezoelectric properties were conducted using a commercial atomic force microscopy

(Cypher ES, OXFORD Instruments). The manufacturer of the probe (Silicon probe, Ptlr reflex/tip coated) is NanoWorld, with the spring k of 2.8 N/m and frequency of 75 kHz. The hardness was measured by HVS-1000A digital microscope Vickers hardness tester, and the load of 4.9 N was applied for 10 s.

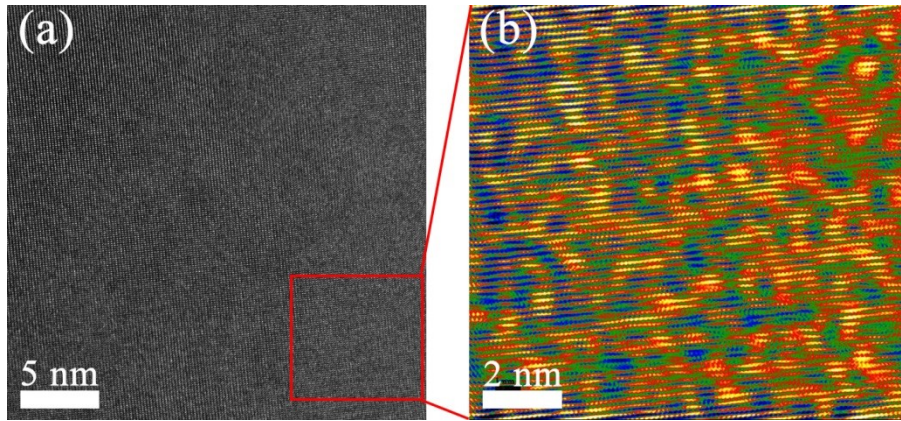


Figure S1. (a) High-resolution (HR) TEM image of the KNNT-0.10SLN ceramic, (b) the inverse fast Fourier transformation (FFT) pattern for KNNT-0.10SLN ceramic.

The yellow areas represent the local polar regions. [Figure S1](#)(b) shows that there are polar nano-regions (PNRs) in the sample.

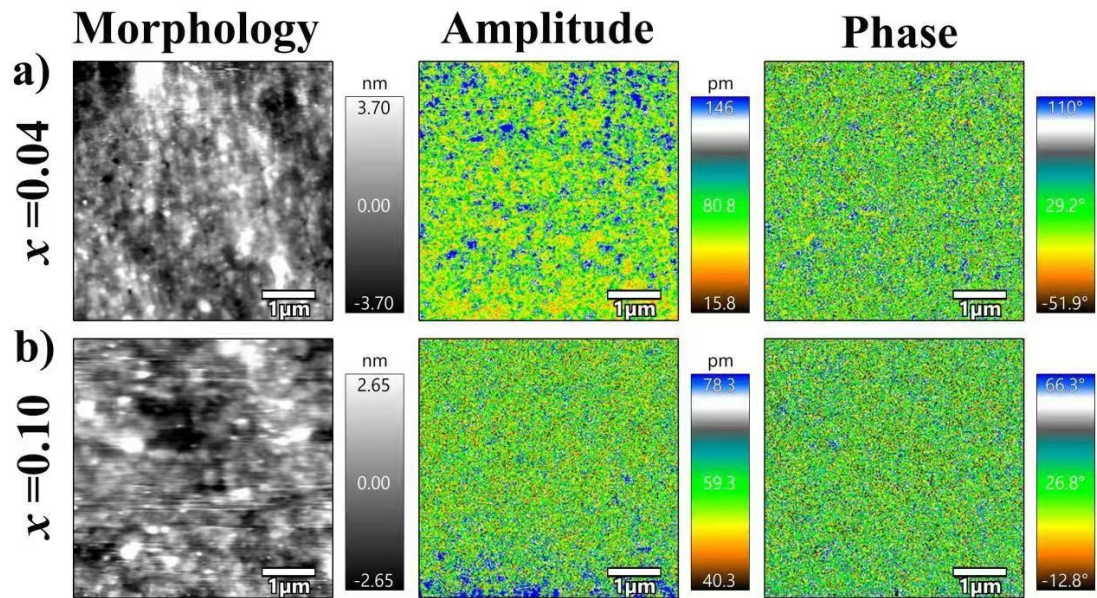


Figure S2. Out-of-plane PFM images for the (a) $x = 0.04$ and (b) $x = 0.10$ ceramics.

Amplitude curve and phase curve testing using ‘DARTSSPFM’ out-of-plane mode. The point of interest is selected from the available topography for single point scanning. Adjust the maximum DC voltage, the duration of the voltage, the phase value of the waveform, and the duration of a single pulse. Then, tune the peak and find the resonance peak before performing ‘Single Force’. Continuously increasing the DC voltage can help the domain flip.

As can be seen, there are no macroscopic ferroelectric domains in both samples, only nano-domains exist.

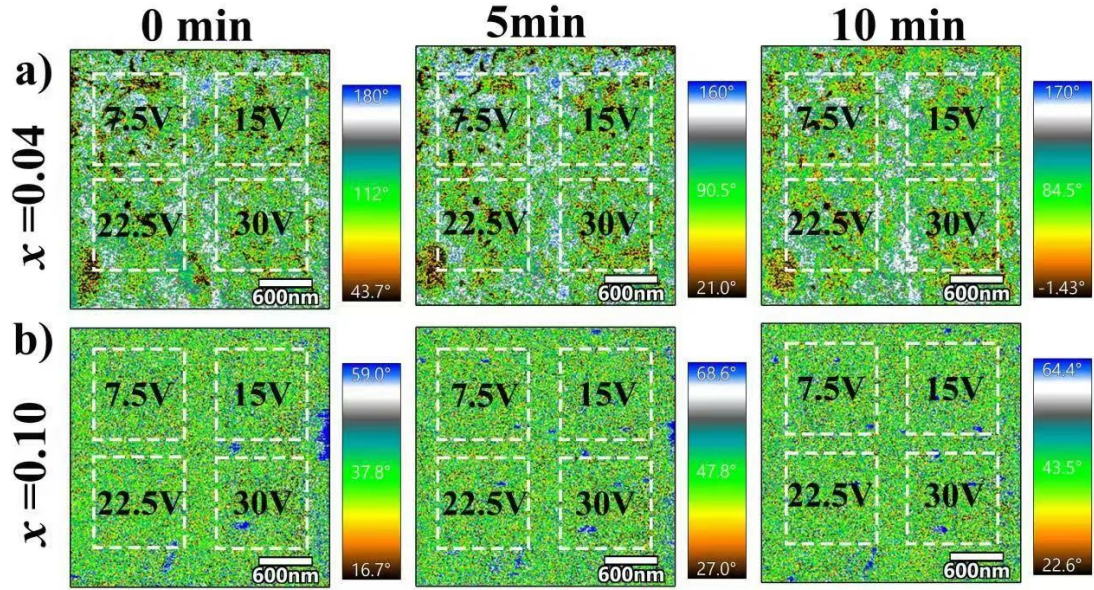


Figure S3. PFM phase images after poling and relaxation durations for the $x = 0.04$ and 010 ceramics.

Design a grayscale image of the write domain based on the flipping voltage. Select 3 μm for scan size, -30V for min bias, and 30V for max bias. Apply a negative DC voltage of -30V to a 3 $\mu\text{m} \times 3 \mu\text{m}$ square area, and apply positive DC voltages of 7.5V, 15V, 22.5V, and 30V to the four 1 $\mu\text{m} \times 1 \mu\text{m}$ square areas. Then, tune the peak and perform ‘Do Scan’ after finding the resonance peak. After applying the polarization voltage, perform tuning peak scanning at intervals of 0, 5, 10, and 30 minutes to observe the relaxation behavior of the sample.

From the local polarization experiment, it is seen that no domain inversion is observed in the two samples even under the electric field of +30 V, indicating the existence of highly dynamic PNRs.

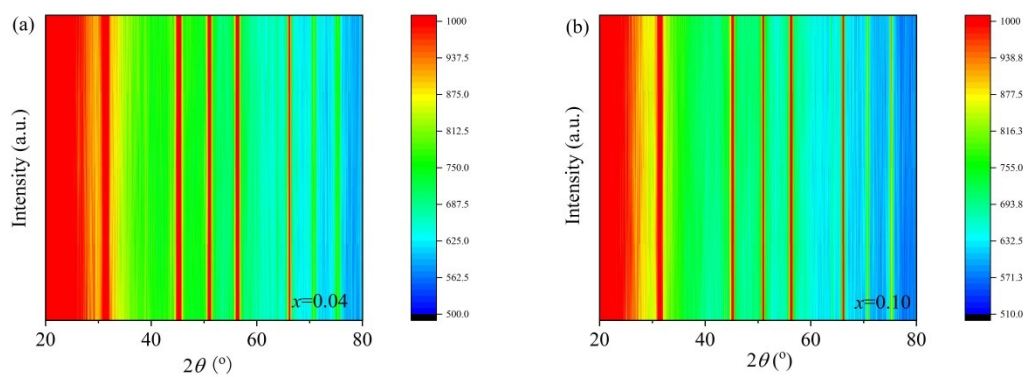


Figure S4. The in-situ XRD patterns for the (a) $x=0.04$ and (b) $x=0.10$ ceramics at 40~140 °C.

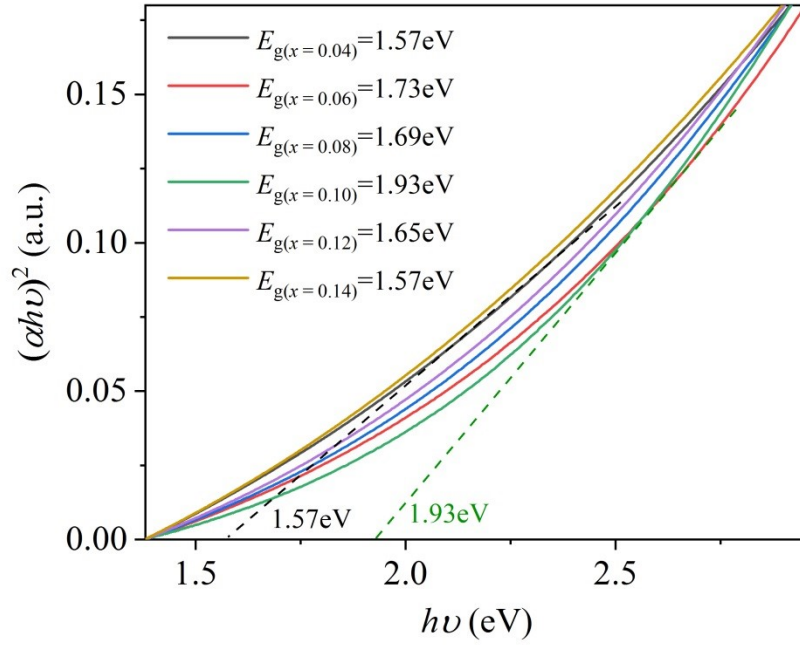


Figure S5. The $(\alpha h\nu)^2$ as functions of $h\nu$ for the KNNT- x SLN ceramics.

It can be seen that the $x = 0.10$ sample has the largest band gap (E_g), which has a positive effect on the sample's high E_b .