Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Supporting Information

Surface texturing and dielectric property tuning toward boosting of triboelectric nanogenerator performance

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Experimental section

Synthesis of $BaTiO_3$ and $CaCu_3Ti_4O_{12}$ particles

The $BaTiO_3$ nanoparticles (NPs) were prepared via hydrothermal reactions. In a typical procedure, 0.05 mol Ti-butoxide was dissolved into 10 ml ethanol and 10 ml deionized water. Then 8 ml ammonium hydroxide was added to the mixture. The solution was heated to 80 °C, followed by adding 25 ml pre-heated (80 °C) barium hydroxide solution (3.75 $mol \cdot L^{-1}$). The well dispersed suspension was transferred into a 100 mL Teflon-lined stainless steel autoclave with a fill factor of ~60 % and was heated at 200 °C for 24 hours. After cooling down to room temperature, the products were separated and washed 3 times with distilled water and ethanol by centrifugation, and then dried in air at 80 ºC overnight.

The $CaCu_3Ti_4O_{12}$ powder was prepared via solid state reaction. Stoichiometric amounts of $CaCO_3$, CuO, and TiO_2 were mixed and calcinated at 970 ºC for 6 hours in air. The obtained material was milled and finally sintered in air at 1050 ºC for 4 hours.

Fabrication of P(VDF-TrFE), PDMS, BTO/PDMS and CCTO/PDMS composite films

A certain amount of P(VDF-TrFE) copolymer powder (70/30 mol%, Piezotech, France) was dissolved in N,N-dimethylformamide (DMF) with stirring for 1 hour at 60 ºC to form a homogeneous solution (15 wt%). The P(VDF-TrFE) solution was spin-coated on ITO-coated PET substrate at 1000 rpm for 30 s, followed by drying at 80 °C on a hot plate to remove the DMF solvent. After that, this layer was annealed at 140 °C for 2 h to enhance the crystallinity of the β phase.

The texture on the surface of the PDMS and PDMS composite films was transferred from the FTO-coated glass (conductive layer). Firstly, the FTO glass was cleaned with deionized water,

ethanol, and acetone successively. Then PDMS elastomer and cross-linker were mixed in the weight ratio of 10:1. Hexane was used as diluent. After a degassing process under vacuum for about half an hour, the mixture was spin-coated on FTO-coated glass at 500 rpm for 30 s and cured at 80 °C for 1 hour. Afterwards, the thin PDMS was peeled off from the glass and then placed on uncured PDMS on the ITO-coated PET film (prepared by spin-coating at 1000 rpm for 30 s), followed by curing at 80 °C for 3 h. The BTO/PDMS and CCTO/PDMS composite films were fabricated in a similar way, except different mass ratios of BTO NPs and CCTO power were, respectively, dispersed into the mixture. Besides, an ultrasonic treatment process was carried out for 1 h to improve the dispersion of the particles.

Material characterizations

The crystalline structures were analyzed using X-ray diffraction (XRD, Bruker D5005, with Cu Kα radiation at $\lambda = 1.541$ Å) and transmission electron microscopy (TEM, Philips FEG CM300). The morphology characteristics were studied using field-emission scanning electron microscopy (FESEM, JEOL FEG JSM 7001F). Fourier transform infrared (FT-IR) spectrum was recorded on a Shimadzu IRPrestige-21 FT-IR spectrophotometer. The dielectric property was investigated by a Solartron analytical 1400 Cell Test System over a frequency range of 0.1 Hz to 10 kHz. By testing the capacitance (C), the dielectric constant (k) can be calculated by: $k = C/(\varepsilon_0 A/d)$, where ε_0 is the vacuum permittivity, A is the sample area, d is the sample thickness. Surface roughness, probe force microscopy (PFM) and Kelvin probe force microscopy measurements were performed using MFP-3D™ Stand Alone Atomic Force Microscope (AFM) with OMCL-AC240TM tips (tip radius, 15 nm; force constant, 2 $N \cdot m^{-1}$; and resonance frequency, 70 kHz). Raman spectrum was

detected using a 532 nm excitation laser with a WITec Raman instrument. Potential distribution was investigated using COMSOL Multiphysics software.

Electrical characterization

A programmed solenoid was used to produce a periodic compressive strain in the TENG with a contact area of 2.5×2.5 cm^2. The gap distance was fixed at 5 mm. A Digital Phosphor Oscilloscope (DPO 7254, Tektronix) and an electrometer (6517B, Keithley Instruments Inc.) were used to measure the electrical output signal.

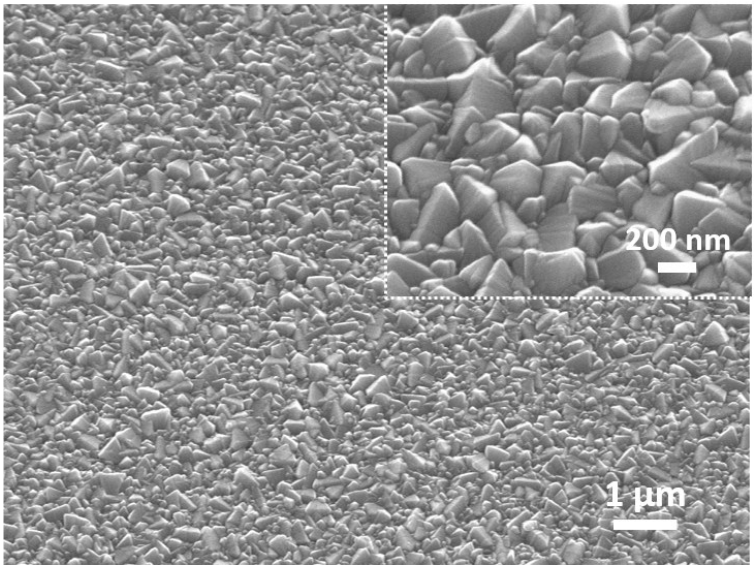

Fig. S1 SEM image of the conductive layer of the FTO-coated glass; inset is an enlarged image.

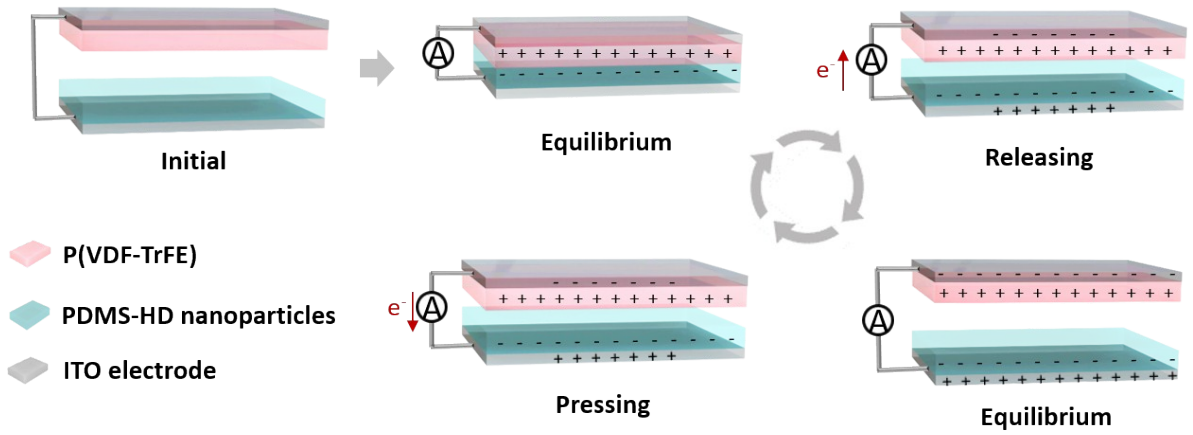

Fig. S2 Schematic illustration of the TENG working mechanism in a full contact and separation cycle.

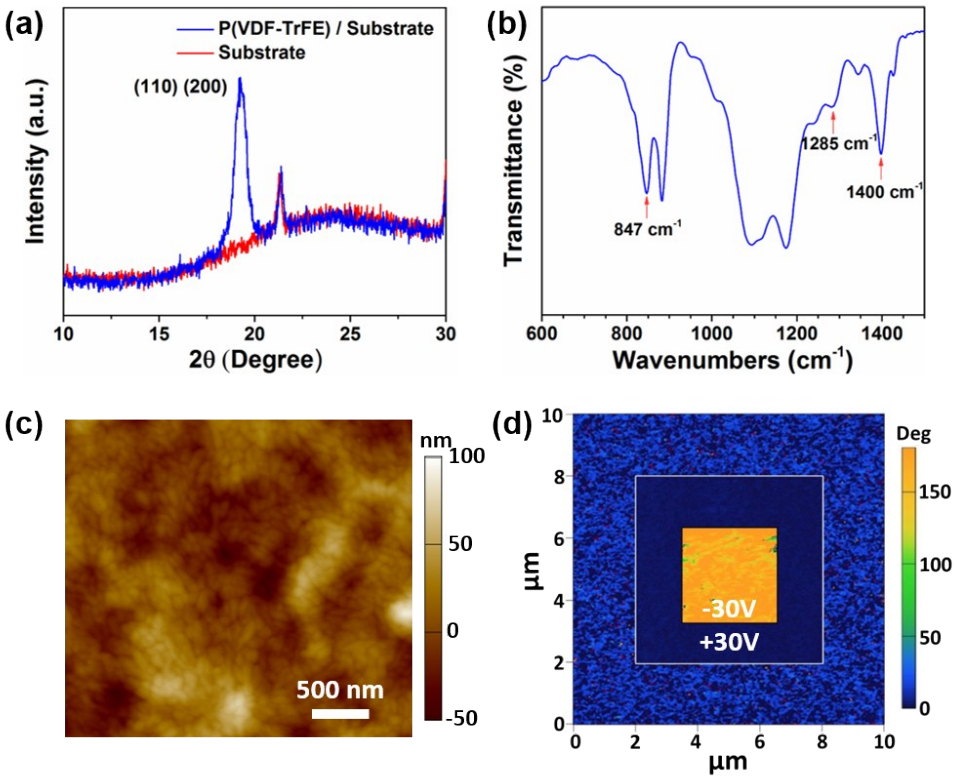

Fig. S3 (a) XRD spectra of P(VDF-TrFE) film and substrate. (b) FTIR spectrum of P(VDF-TrFE) film. (c) AFM images of P(VDF-TrFE) film for the surface topography and roughness. (d) PFM polarization switching of P(VDF-TrFE) film.

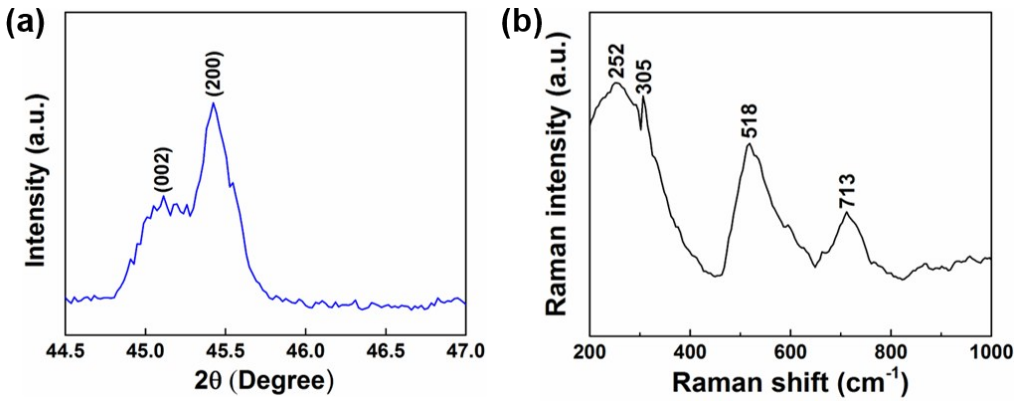

Fig. S4 (a) XRD pattern and (b) Raman spectrum of the synthesized BTO nanoparticles.

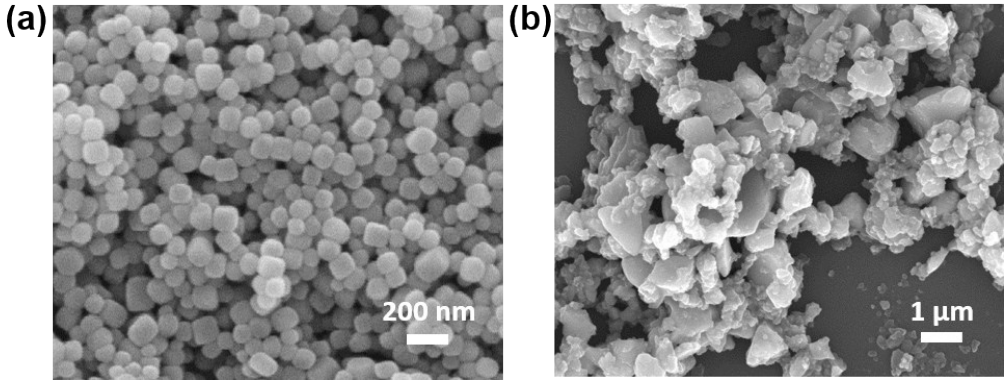

Fig. S5 SEM images of the (a) BTO and (b) CCTO particles.

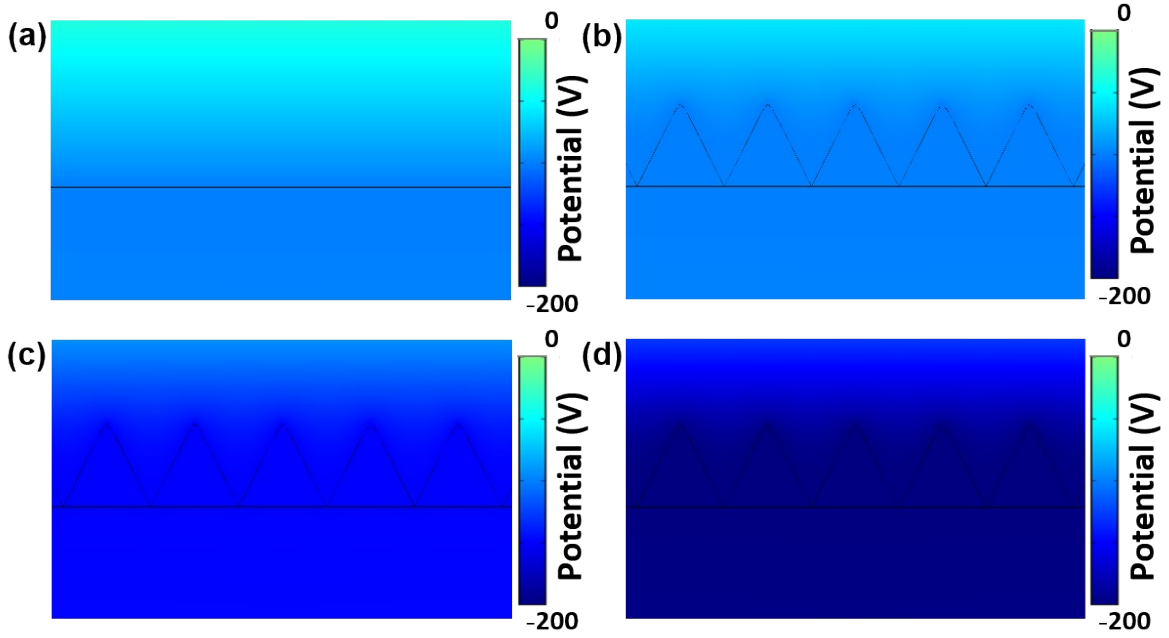

Fig. S6 COMSOL simulation results show the surface potential of the PDMS-based films.

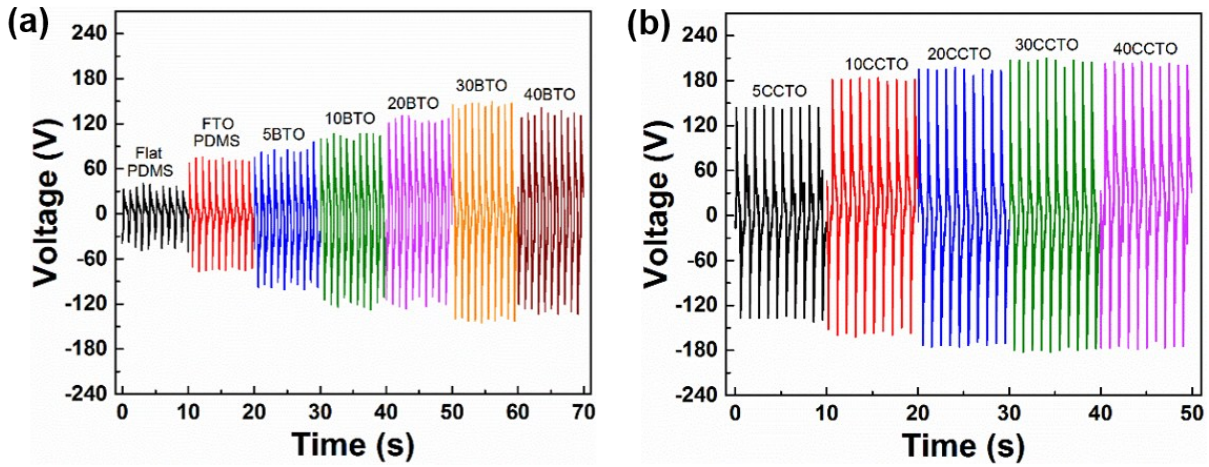

Fig. S7 The output voltage of (a) flat PDMS-, FTO PDMS-, and BTO-based TENGs and (b) CCTO-based TENGs.

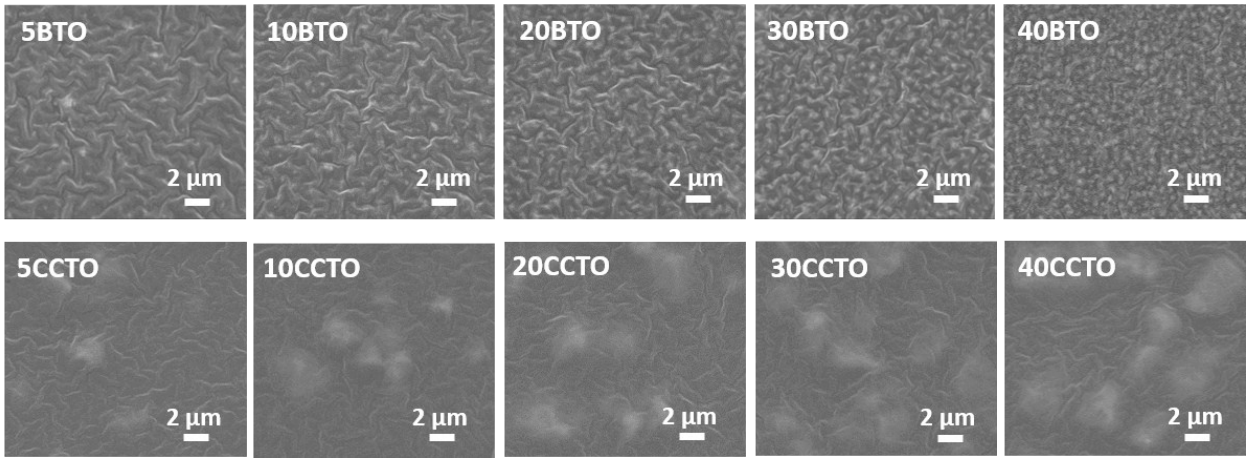

Fig. S8 SEM images of the BTO-PDMS and CCTO-PDMS composite films.

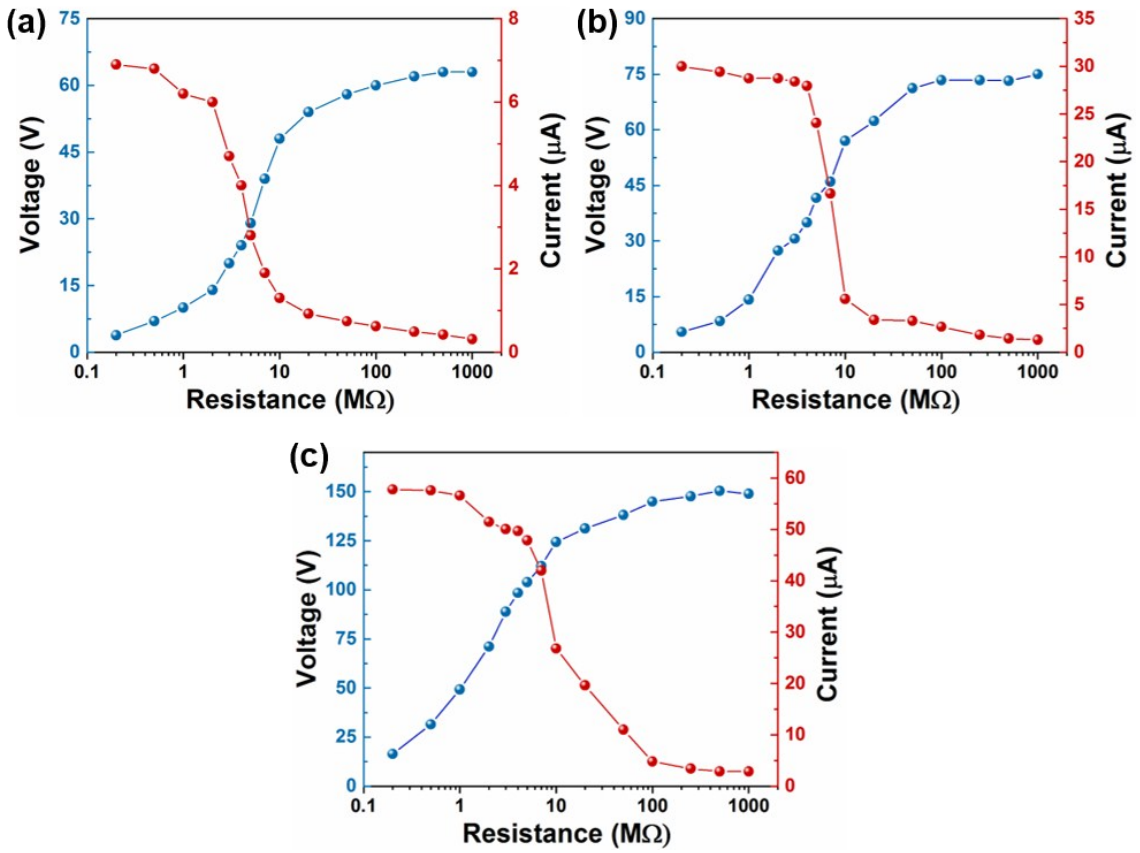

Fig. S9 Output voltage and current of the (a) flat PDMS-based TENG, (B) FTO PDMS-based TENG, and (c) 30BTO-based TENG according to the external load resistance.