Electronic Supplementary Material (ESI) for Energy & Environmental Science.

Electronic Supplementary information

Inversely polarised ferroelectric polymer contact electrodes for triboelectric-like generators from identical materials

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Experimental details: The morphology of the samples were characterized by using scanning electron microscopy (SEM) recorded by FEI FIB-SEM Helios NanoLab 600, as well as by using atomic force microscope (Smena, NT-MDT) in semi-contact mode. We used the same manufacturer probes (NSG30/Pt, NT-MDT). Samples before SEM measurements were coated with Cu/Pd by using sputtering machine. Crystalline phases of prepared PVDF samples were characterised by attenuated total reflection-Fourier transform infrared spectroscopy (ATR-FTIR) recorded by Bruker Vertex 70 FTIR spectrometer equipped with a Platinum ATR accessory. PVDF films were placed onto the ATR crystal and spectra were collected. 50 scans at a resolution of 4 cm-1 in the range of 4000-400 cm-1 were acquired.

Figure S1. Sample used for piezo-regime tests to measure current and voltage where PVDF film is sandwiched between insulative PET layers. Conductive ITO electrode layers are below PET. Piezo-regime tests are used to demonstrate that the electrostatic induction can be driven by piezoelectric charges.

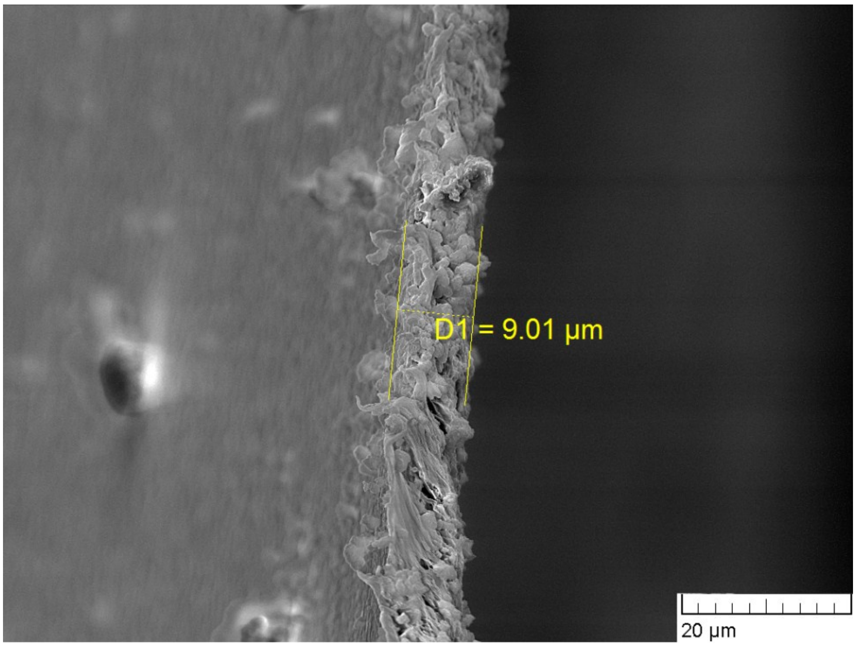

Figure S2. SEM image of cross-section of PVDF film.

Table S1. Output voltage at load resistance $1 \cdot 10^9$ Ω for TENG based on PVDF in combination with polyethyleneterephtalate (PET), polydimethylsiloxane (PDMS), Cu and polymethylmethacrylate (PMMA). The contacting force during the measurements was 10N, frequency – 1Hz and separating distance – 0,5 mm, contacting area 5 cm^2. In table "0" refers to non-polarised PVDF, "-" and "+" to inversely polarised PVDF.

	Peak voltage, V			
	PET	PDMS	Cu	PMMA
PVDF "0"	12	48	8	8
PVDF "+"	100	100	68	75
PVDF "-"	55	53	35	24

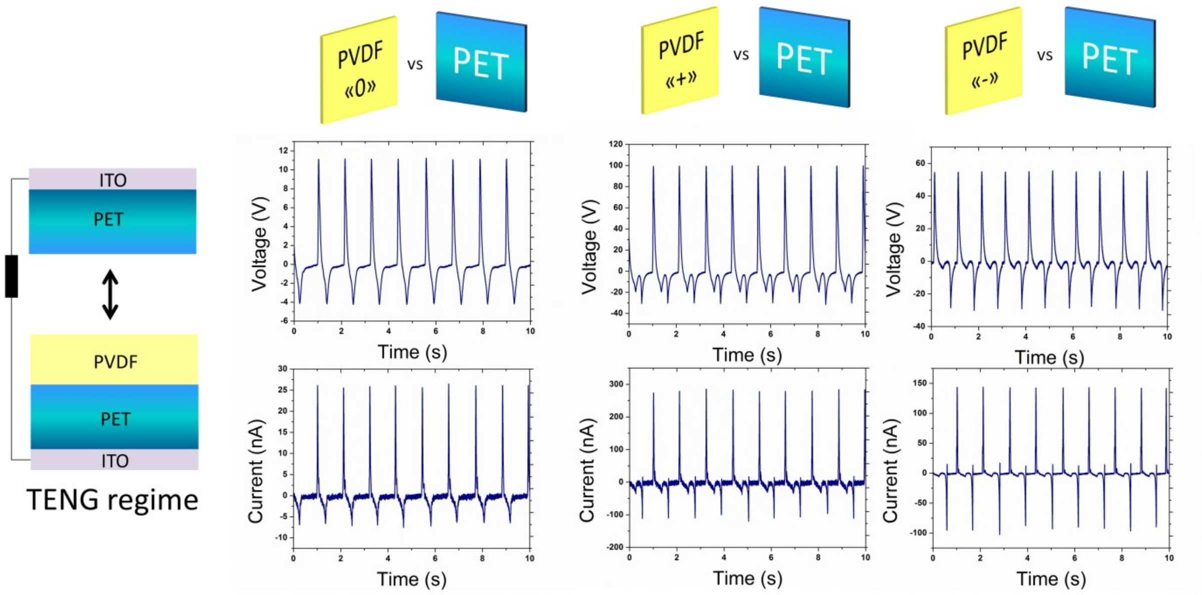

Figure S3. Output voltage (at load resistance $1 \cdot 10^9$ Ω) and short circuit current for TENG based on PVDF in combination with PET: (left) non-polarised PVDF vs PET, (middle) polarised (+) PVDF vs PET and (right) polarised (-) PVDF vs PET. The contacting force during the measurements was 10N, frequency – 1Hz and separating distance – 0.5 mm, contacting area 5 cm^2.

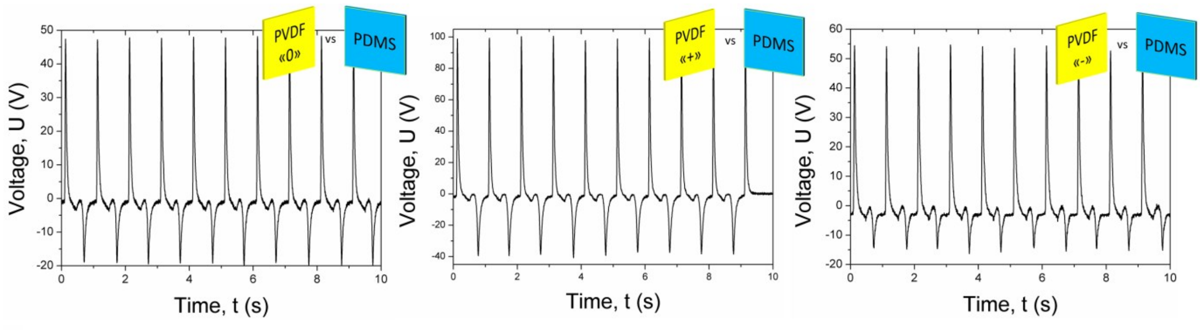

Figure S4. Output voltage at load resistance $1{\cdot}10^9$ Ω for TENG based on different sample combinations: (left) non-polarised PVDF vs PDMS, (middle) polarised (+) PVDF vs PDMS and (right) polarised (-) PVDF vs PDMS. The contacting force during the measurements was 10N, frequency – 1Hz and separating distance – 0,5 mm, contacting area 5 cm^2. PDMS (184, Sylgard) films were obtained by mixing PDMS base with curing agent in a ratio 10:1 and spin coated at 3000 rpm on the same ITO-coated PET. After spin coating, samples were cured at 90°C for 30 min. This procedure was repeated 5 times in order to obtain a PDMS film at optimal thickness of 0.15 µm.

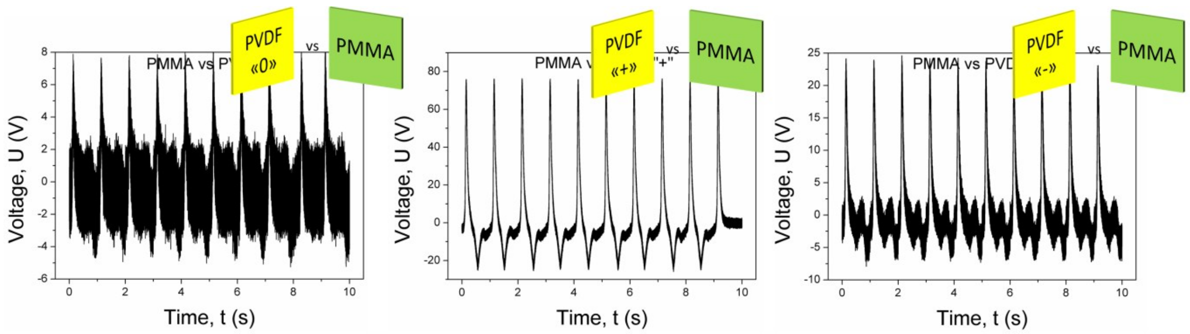

Figure S5. Output voltage at load resistance $1{\cdot}10^9$ Ω for TENG based on different sample combinations: (left) non-polarised PVDF vs PMMA, (middle) polarised (+) PVDF vs PMMA and (right) polarised (-) PVDF vs PMMA. The contacting force during the measurements was 10N, frequency – 1Hz and separating distance – 0,5 mm, contacting area 5 cm^2. PMMA films were spin-coated from 20 wt% PMMA solution in chloroform.

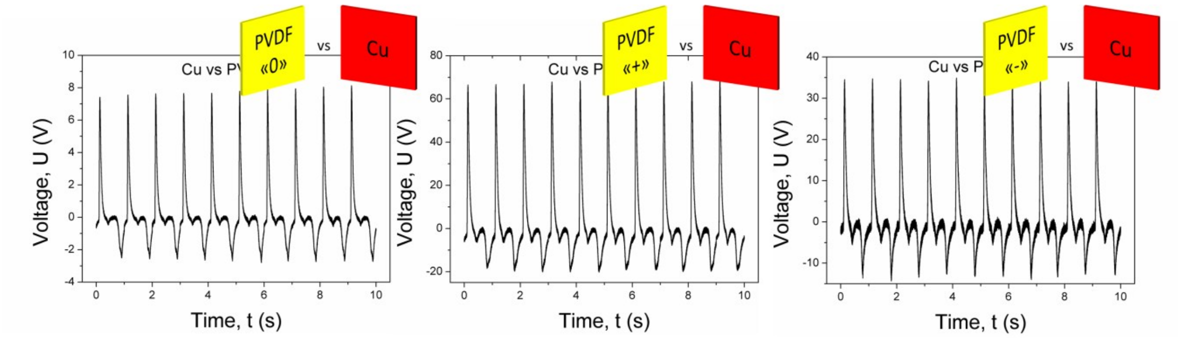

Figure S6. Output voltage at load resistance $1\cdot10^9$ Ω for TENG based on different sample combinations: (left) non-polarised PVDF vs Cu, (middle) polarised (+) PVDF vs Cu and (right) polarised (-) PVDF vs Cu. The contacting force during the measurements was 10N, frequency – 1Hz and separating distance – 0.5 mm, contacting area 5 cm^2.

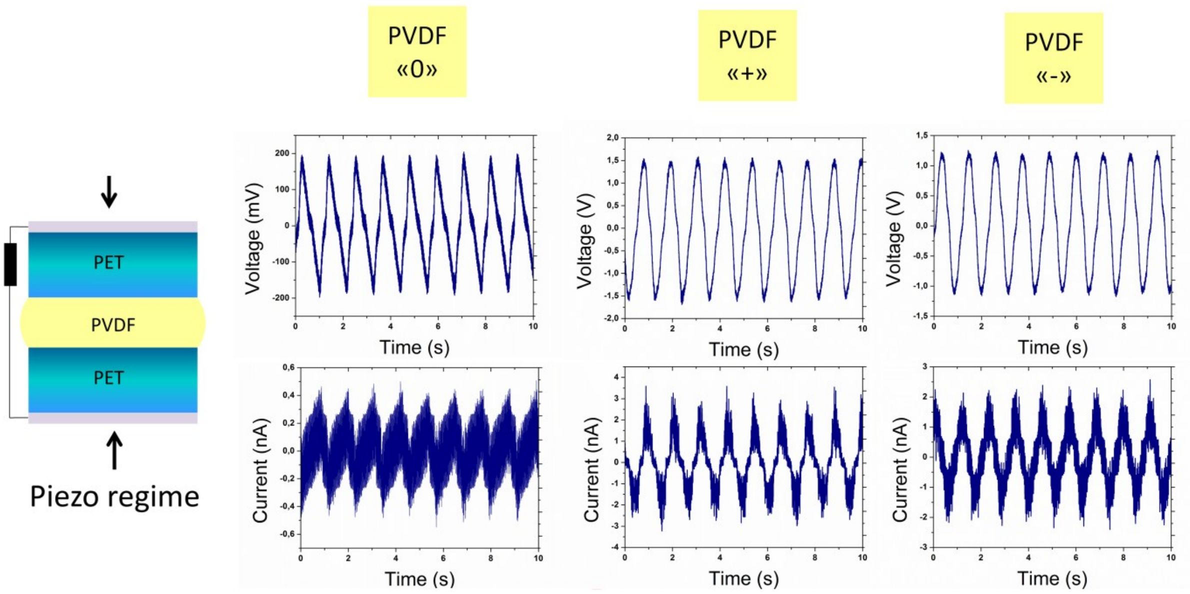

Figure S7. Output voltage (at load resistance $1\cdot10^9$ Ω) and short circuit current for PVDF in piezo regime: (left) non-polarised PVDF, (middle) polarised (+) PVDF and (right) polarised (-) PVDF. The force during the measurements was varied from 1 N to 10 N, frequency – 1 Hz and contacting area 5 cm^2.

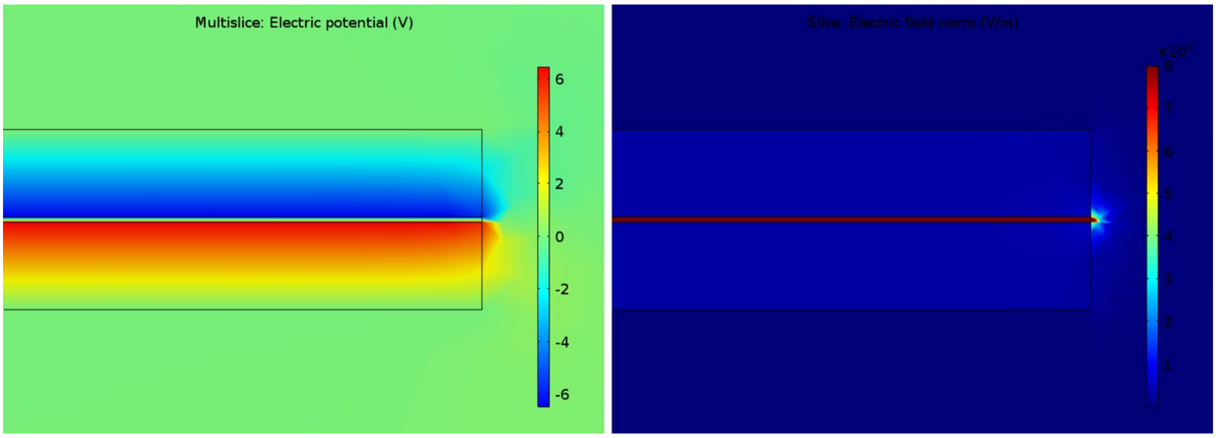

Figure S8. Simulated electrical potential (left) and electric field strength (right) distribution at the cross-section of ITO-PET-PVDF-PET-ITO layered structure, induced by the piezoelectric charge on top and bottom PVDF surface -8.45 nC/cm^2 and 8.45 nC/cm^2 respectively. The simulations were carried out by using COMSOL finite element analysis software.

Table S2. Output peak voltage (at load resistance $1\cdot10^9$ Ω) and short circuit peak current for TENG based on PVDF. Non-polarised and inversely polarised PVDF films were contacted and separated mutually at different combinations. The contacting force during the measurements was 10 N, frequency – 1 Hz and separating distance – 0.5 mm, contacting area 5 cm^2.

Sample combination	Peak voltage, V	Current, nA
0/0	0.2	31
0/-	75	135
0/+	67	126
+/+	6	48
-/-	3	23
-/+	125	180

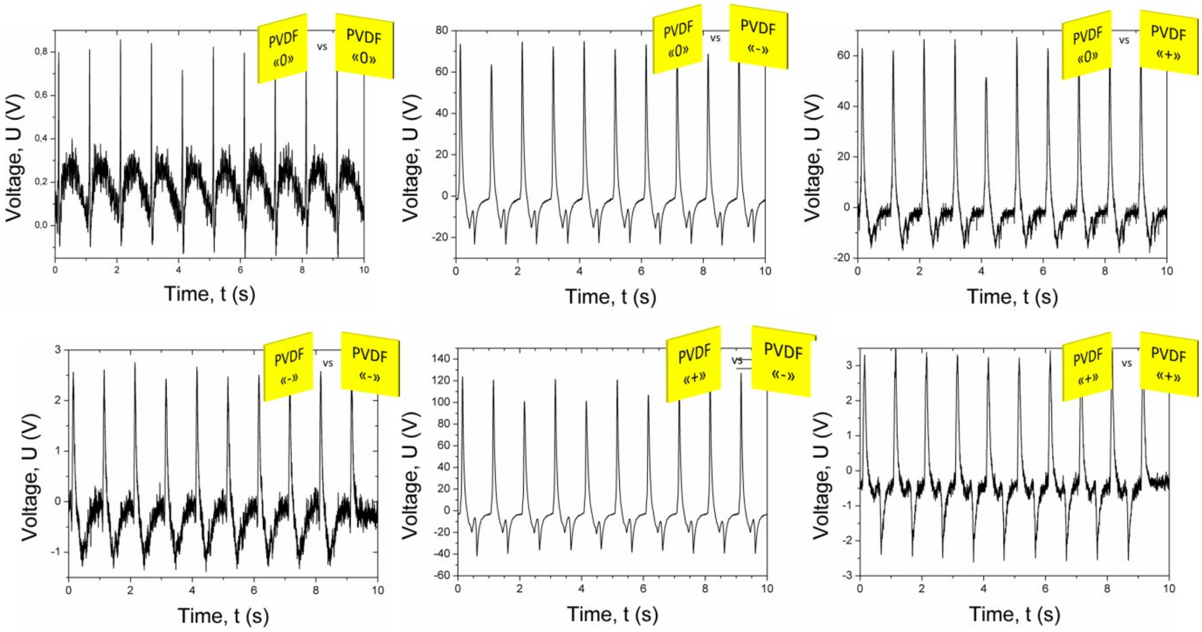

Figure S9. The output voltage for TENG device based on PVDF measured at load resistance $1 \cdot 10^9$ Ω. Non-polarised and inversely polarised PVDF films were contacted and separated mutually at different combinations: (top row, left) non-polarised PVDF vs non-polarised PVDF, (top row, middle) non-polarised PVDF vs polarised (-) PVDF, (top row, right) non-polarised PVDF vs polarised (+) PVDF, (bottom row, left) polarised (-) PVDF vs polarised (-) PVDF, (bottom row, middle) polarised (+) PVDF vs polarised (-) PVDF, (bottom row, right) polarised (+) PVDF vs polarised (+) PVDF.

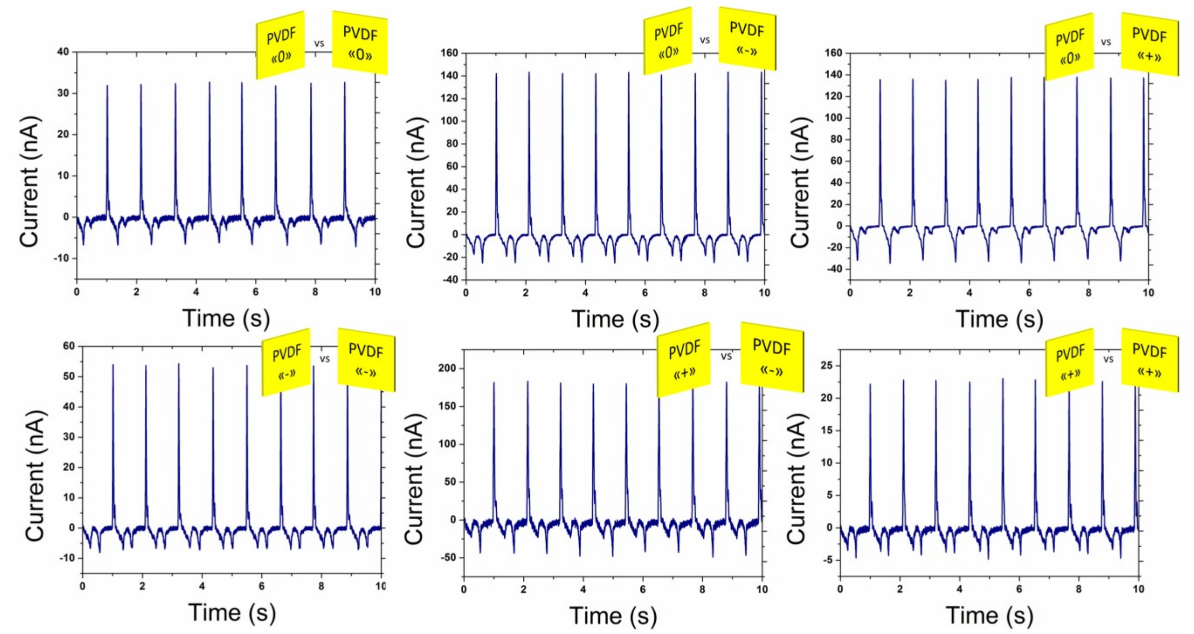

Figure S10. The short circuit current measured for TENG device based on PVDF. Non-polarised and inversely polarised PVDF films were contacted and separated mutually at different combinations: (top row, left) non-polarised PVDF vs non-polarised PVDF, (top row, middle) non-polarised PVDF vs polarised (-) PVDF, (top row, right) non-polarised PVDF vs polarised (+) PVDF, (bottom row, left) polarised (-) PVDF vs polarised (-) PVDF, (bottom row, middle) polarised (+) PVDF vs polarised (-) PVDF, (bottom row, right) polarised (+) PVDF vs polarised (+) PVDF.

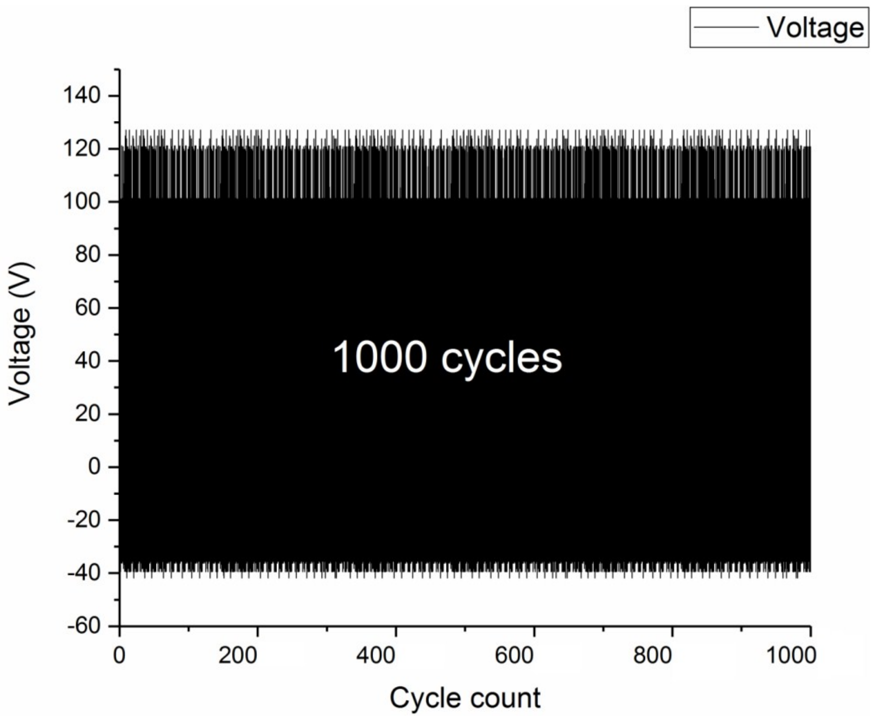

Figure S11. The output voltage 1000 cycle measurement for device based on inversely polarised PVDF. Output voltage was measured at load resistance $1 \cdot 10^9$ Ω. The contacting force during the measurements was 10N, frequency – 1Hz and separating distance – 0.5 mm, contacting area 5 cm^2.

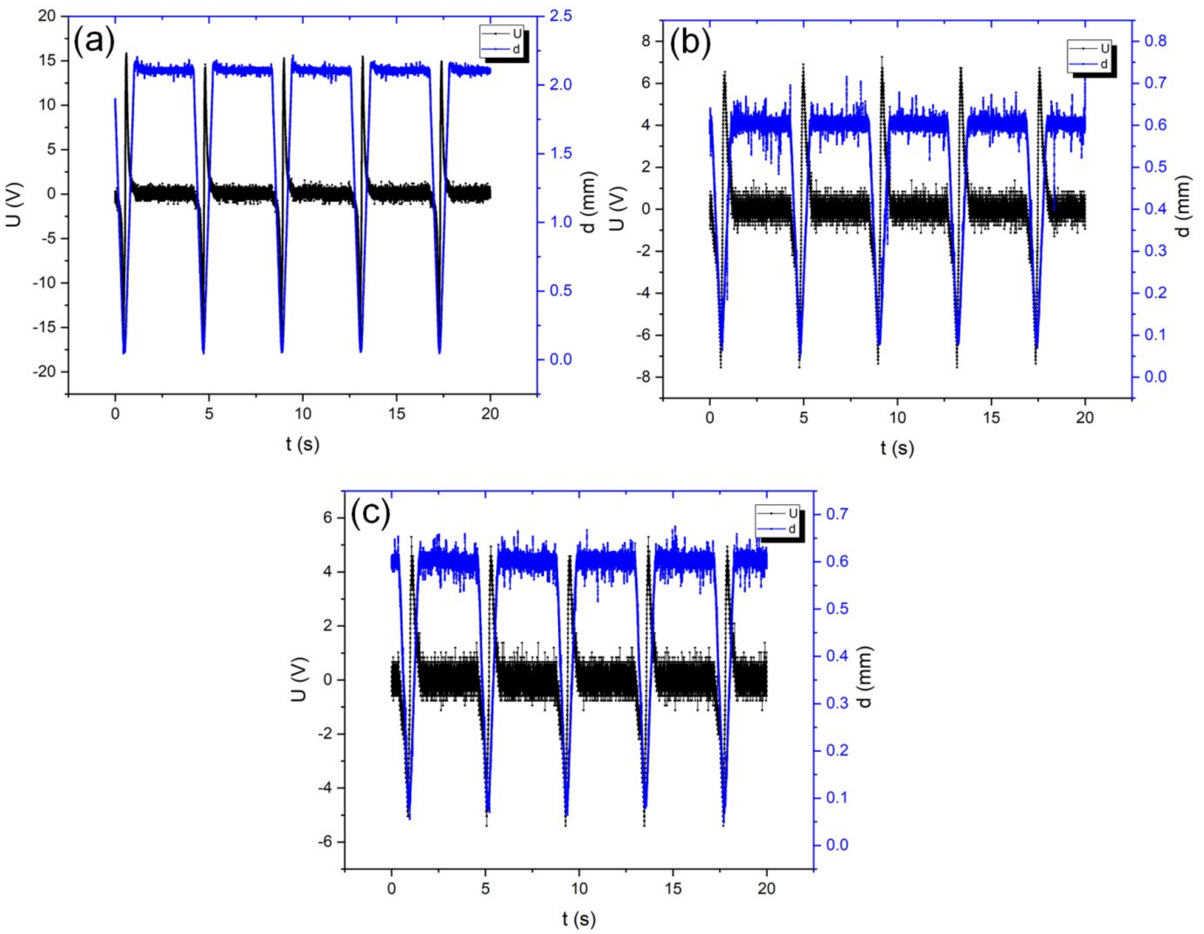

Figure S12. Output voltage (*U, V*) generated by moving TENG electrodes back and forth towards each other without having physical contact. As electrodes a freshly polarised and never contacted PVDF films on conducting electrodes are used. Electrodes are connected by external circuit at load resistance $1 \cdot 10^9$ Ω. In graph (a) polarised PVDF "+" and PVDF "-" are used and separating distance (*d, mm*) during the measurements is varied from 0.05 mm to 2 mm; in graph (b) the same electrode combination is used but separating distance is varied from 0.05 mm to 0.6 mm (smaller voltages are generated due to lower electrical potential when separating gap between electrodes is smaller). Graph (c) shows output voltage measured at non-contact mode in combination PVDF "+" vs ITO, showing that voltage can be generated when electrostatic induction occurs at least on one of two TENG electrodes.

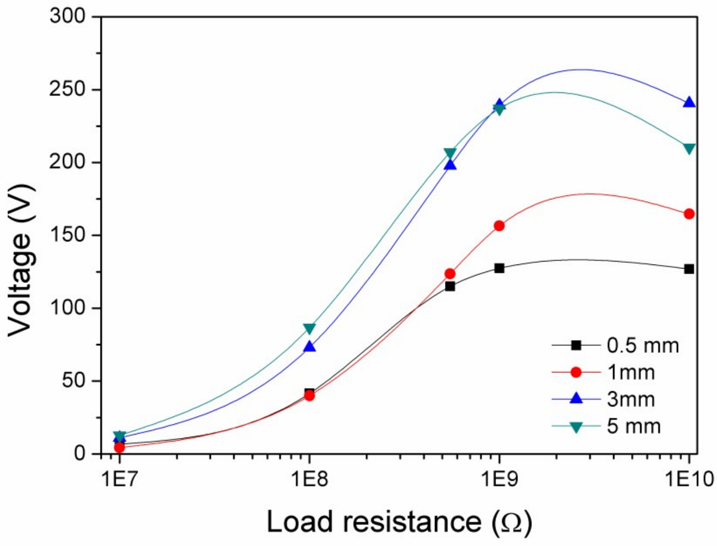

Figure S13. The output voltages at different separating gap between contact electrodes for device based on inversely polarised PVDF. The contacting force during the measurements was 10 N, contacting area 5 cm^2, separating speed 0.5 m/s. The voltage increases by increasing separating gap if the separation speed is maintained the same.

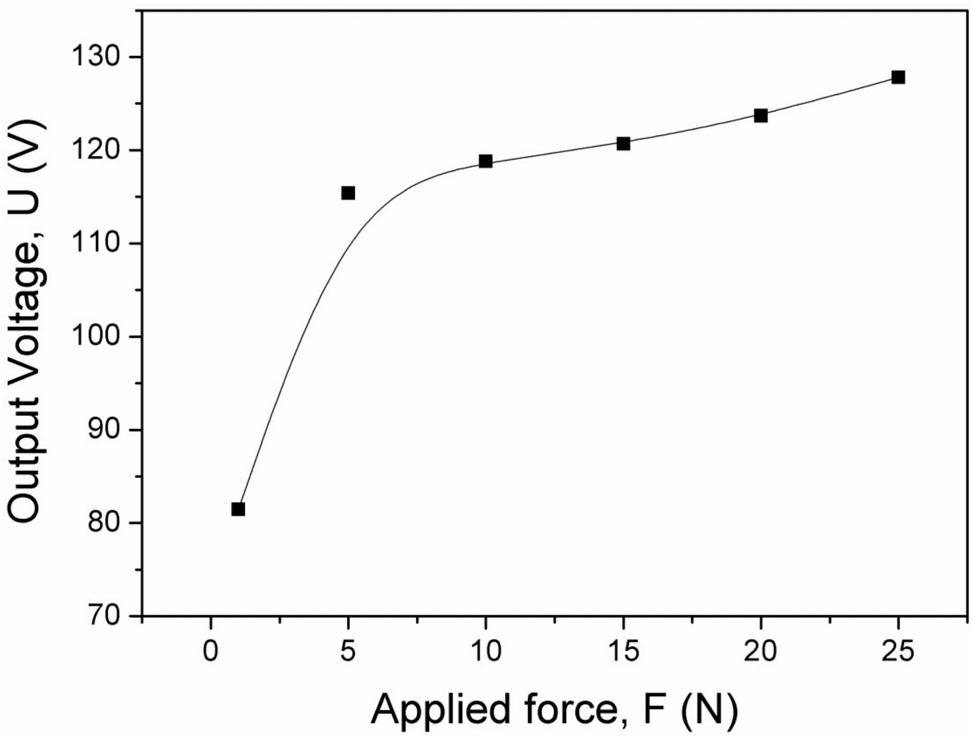

Figure S14. The output voltages at different applied force between contact electrodes for device based on inversely polarised PVDF. Output voltage was measured at load resistance $1 \cdot 10^9$ Ω, frequency – 1 Hz and separating distance – 0.5 mm, contacting area 5 cm^2.

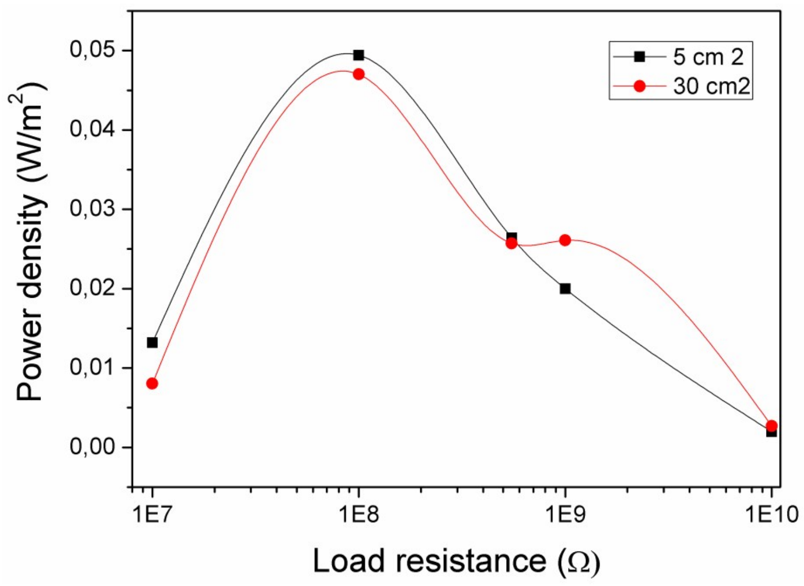

Figure S15. Power density observed from devices based on inversely polarised PVDF with different electrode size. The contacting force during the measurements was 10 N, frequency – 1 Hz and separating distance – 0.5 mm.