Supplementary Information

Strain-Induced Electrification-Based Flexible Nanogenerator for Efficient Harvesting from Ultralow-Frequency Vibration Energy at 0.5-0.01 Hz

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Methods

Fabrication of the SIE-NG

High-purity Au pellets (iTASCO, 99% purity) were purchased for thermal-evaporation deposition. The deposition was performed using a tungsten boat in a high-vacuum evaporation system (Jvac, JVEVA-F30k2p). Substrates were prepared from commercially used 100-μm-thick PET, PI, and UTG. For roughness testing, PET substrates underwent additional polishing using three different grades of sandpaper (**Supplementary Fig. 3**). The substrates were carefully cleaned in a sonication bath each with ethanol acetone, and deionized water several times. Cleaned substrates were attached to a rotating substrate holder in the chamber. Prior to the deposition, the pressure in the chamber was reduced to 1×10^{-6} Torr using a pump to prevent oxidation. The cooling system was operated continuously to maintain the internal temperature of the chamber. The boat temperature was increased by applying a voltage and current, and the Au pellets were melted under a current and voltage of 70 A and 0.6 V, respectively. Au was deposited for 10 min after melting commenced at a pressure of 5×10^{-6} Torr, and 40-nm-thick samples with an area of 70×70 mm^2 were fabricated. The SIE-NG was positioned on the watch strap using a liquid bandage (Mediform Liquid, Mundipharma Korea Ltd.) and then subjected to continuous pressure for a few seconds to achieve sufficient adhesion. The bonding was sufficiently strong to accommodate the effects of the bending forces.

Measurement of surface roughness

The RMS surface roughness of the substrates was investigated via a stylus profilometer (Bruker Dektak XT); range, stylus type, stylus force, scan length, duration was approximately 65.5 μm, radius 2 μm, 3 mg, 5000 μm and 300 sec, respectively. To ensure that every surface area had been polished, the surface profiles of the polished specimens were examined using optical microscopy (Eclipse LV100ND, Nikon Inc., Japan). The surface roughness of the specimens were assessed after uniform profiles were produced.

Measurement of electrical outputs

The fabricated SIE-NG is based on the single-electrode mode, which consists of an Au layer that plays dual roles as an electrification layer and an electrode connected to the ground across an external resistor. The open-circuit voltage and short-circuit current of the devices were measured using an electrometer (Model 6514, Keithley Instruments Inc., USA) and an oscilloscope (Model MDO3000 Tektronix Inc., USA).

UPS measurement and calculation of work function

The work function of the SIE-NG was investigated using UPS (ESCALAB 250Xi, Thermo Scientific). Specifically, the UPS spectra were obtained under He I (21.2 eV) ultraviolet light, and the electron cutoff was measured based on a pass energy of 5 eV and a sample bias of -5 V. Using the UPS data, the work function (Φ) was calculated as follows[1]:

$$\Phi = h\nu - (E_{cutoff} - E_f), \qquad \text{(eq. 1)}$$

where $h\nu$, E_{cutoff}, and E_f represent the UPS spectra-derived excitation photon energy, energy cutoff, and Fermi-level edge, respectively. The Au layers on PET conformally adhered to curved poly(lactic acid) substrates used as bending holders with curvature radii of 49.1, 34.6, and 24.5 mm; these induced corresponding uniaxial bending strains of 0.11%, 0.15%, and 0.18%, respectively, on the layers.

Calculation of tensile strain induced by bending system

A motorized bending stage (Jaeil Optical System) and a computer-aided step motor controller (Ecopia, SMC-100) were used to supply a tensile strain with a specified frequency to the SIE-NG. The tensile strain induced by bending was calculated using the substrate length and the change in length, as follows[2]:

$$\varepsilon = \frac{\pi h \sqrt{\frac{dL}{L} - \frac{\pi^2 h^2}{12L^2}}}{L}, \qquad \text{(eq. 2)}$$

where L, dL, and h are the initial length of the Au/PET layer, the change in L, and the thickness of the Au/PET layer, respectively. **Equation 2** assumes that when a film with a flat surface is bent, the curvature can be described by a sinusoidal curve. The bending stage was equipped with two clamps for securing the thin sheets. One clamp was fixed, whereas the other was connected to a computer and used to bend the thin film via the step motor. The system enabled bending even under an extremely small displacement of only approximately 2 μm during the bending stage. We controlled the maximum bending frequency to 0.5 Hz by setting the travel distance and speed of the moving clamp. All experiments, except for the humidity-stability test, were conducted at room temperature at a relative humidity of ~40%. Optical images and videos were captured using a mobile phone camera.

(a)

(b)

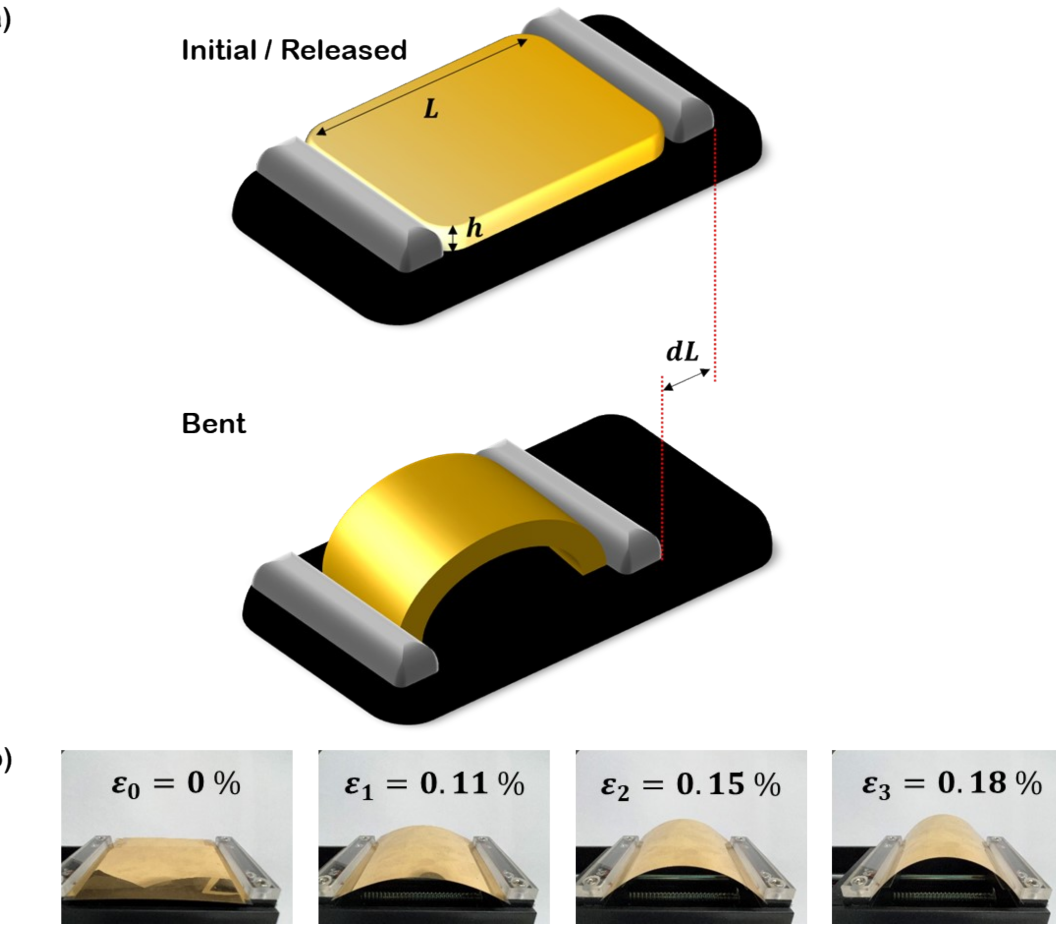

Fig. S1. (a) Schematic of the bending system configuration (b) Snapshot of bending tests for SIE-NG, inducing tensile strain of 0 to 0.18%.

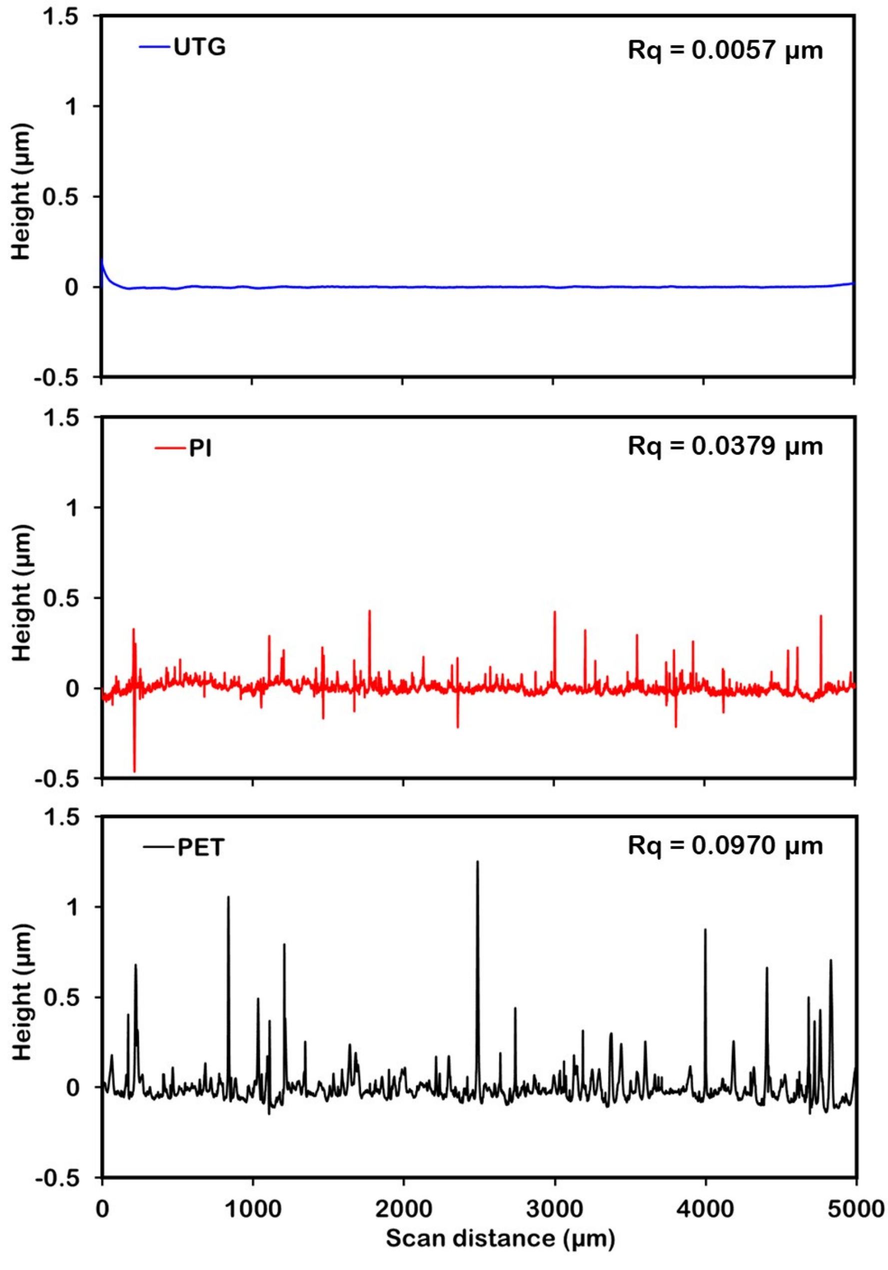

Fig. S2. Surface roughness profiles of PET, PI, and UTG substrates.

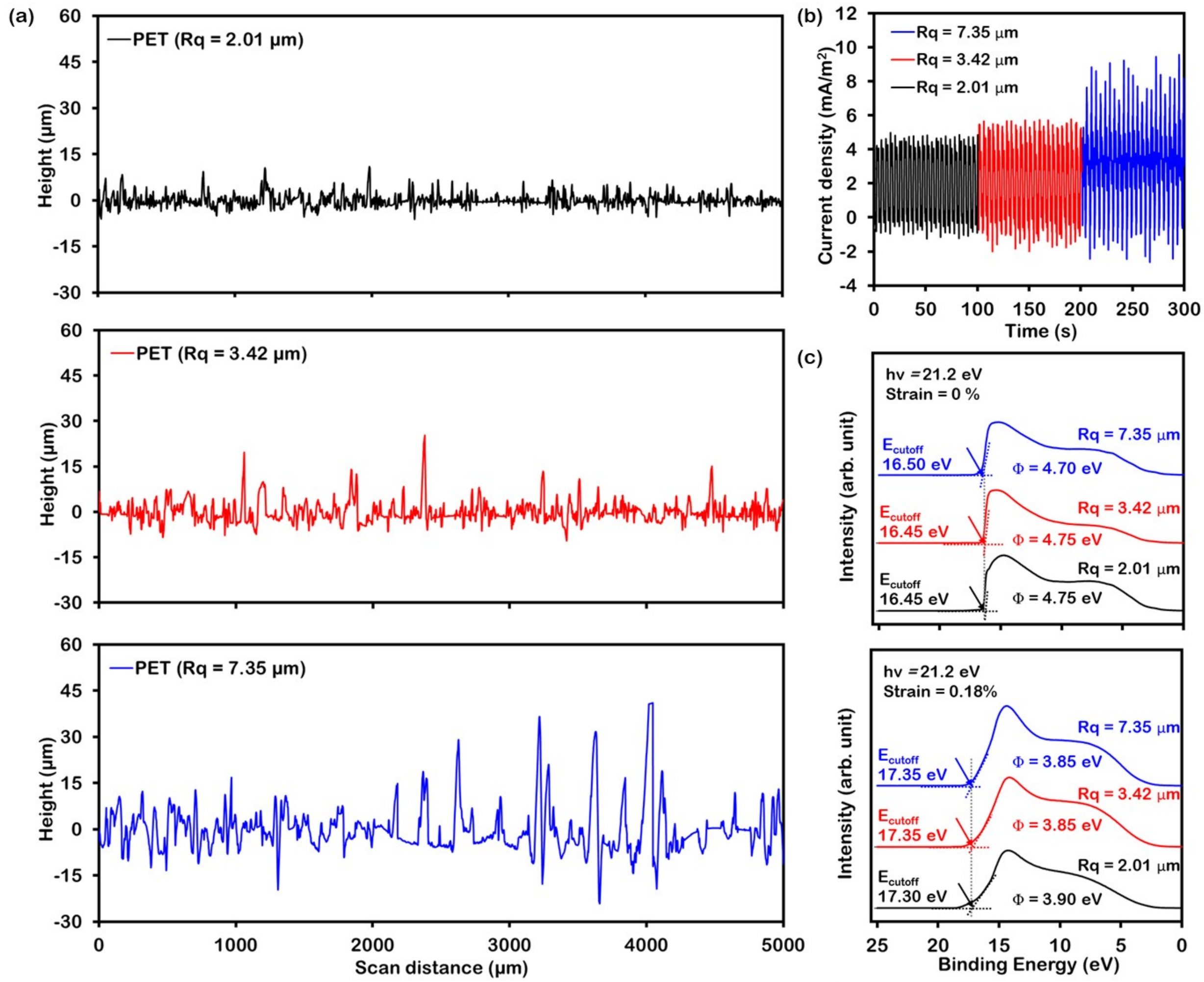

Fig. S3. (a) Surface profiles of polished PET substrates. The substrates were treated with three different grades of sandpaper (P80, P320, and P500). (b) Current densities from Au layer on the polished PET substrates at a frequency of 0.25 Hz. (c) UPS spectra from Au layer on the polished PET substrates under strains of 0% and 0.18%.

Table S1. Summary of the detailed output performances of Au/PET-based SIE-NG at a frequency of 0.25 Hz. The three trials' values of output peak voltage and peak current densities and the derived average and standard deviation.

Strain (%)	V_{peak} (V)			V_{Avg}	V_{Std}	J_{peak} (mA/m^2)			J_{Avg}	J_{Std}
	#1	#2	#3			#1	#2	#3		
0.11	0.50	0.52	0.49	**0.50**	0.01	0.69	0.64	0.59	**0.64**	0.04
0.15	0.77	0.93	0.79	**0.83**	0.07	1.26	1.11	1.27	**1.21**	0.07
0.18	2.31	2.53	2.22	**2.35**	0.13	2.49	2.57	2.66	**2.57**	0.07
0.21	4.32	4.21	4.44	**4.32**	0.09	4.02	3.78	3.86	**3.89**	0.10

Table S2. Summary of the detailed output performances of Au/PI-based SIE-NG at a frequency of 0.25 Hz. The three trials' values of output peak voltage and peak current densities and the derived average and standard deviation.

Strain (%)	V_{peak} (V)			V_{Avg}	V_{Std}	J_{peak} (mA/m^2)			J_{Avg}	J_{Std}
	#1	#2	#3			#1	#2	#3		
0.11	0.31	0.28	0.30	**0.30**	0.01	0.22	0.25	0.27	**0.24**	0.02
0.15	0.40	0.37	0.40	**0.39**	0.01	0.35	0.34	0.43	**0.37**	0.04
0.18	1.42	1.38	1.38	**1.40**	0.02	0.61	0.82	0.80	**0.74**	0.09
0.21	1.99	2.45	2.50	**2.31**	0.23	1.17	1.33	1.27	**1.26**	0.07

Table S3. Summary of the detailed output performances of Au/UTG-based SIE-NG at a frequency of 0.25 Hz. The three trials' values of output peak voltage and peak current densities and the derived average and standard deviation.

Strain (%)	V_{peak} (V)			V_{Avg}	V_{Std}	J_{peak} (mA/m^2)			J_{Avg}	J_{Std}
	#1	#2	#3			#1	#2	#3		
0.11	0.170	0.090	0.130	**0.130**	0.033	0.018	0.024	0.023	**0.022**	0.003
0.15	0.280	0.220	0.250	**0.250**	0.024	0.045	0.039	0.041	**0.042**	0.003
0.18	0.800	0.670	0.712	**0.727**	0.054	0.104	0.067	0.078	**0.083**	0.015
0.21	1.120	1.000	1.030	**1.050**	0.051	0.157	0.143	0.135	**0.145**	0.009

Table S4. Summary of the detailed output performances of Au/PET-, Au/PI-, and Au/UTG-based SIE-NGs at a frequency of 0.25 Hz. The three trials' values of power density and the derived average and standard deviation.

Strain (%)	Device	P_{peak} (mW/m^2)			P_{Avg}	P_{Std}
		#1	#2	#3		
0.11	Au-PET	0.341	0.332	0.285	0.319	0.025
	Au/PI	0.067	0.069	0.080	0.072	0.005
	Au/UTG	0.003	0.002	0.003	0.003	0.0004
0.15	Au-PET	0.970	1.034	1.000	1.001	0.026
	Au/PI	0.139	0.125	0.174	0.146	0.02
	Au/UTG	0.013	0.009	0.01	0.01	0.002
0.18	Au-PET	5.761	6.501	5.895	6.052	0.322
	Au/PI	0.874	1.13	1.107	1.037	0.116
	Au/UTG	0.083	0.045	0.055	0.061	0.016
0.21	Au-PET	17.38	15.93	17.15	16.82	0.636
	Au/PI	2.320	3.271	3.174	2.92	0.427
	Au/UTG	0.176	0.143	0.139	0.153	0.016

Table S5. Summary of the detailed output performances of current density, output voltage, and power density of Au/PET-based SIE-NG under different external loads.

Ohm(Ω)	J_{peak} (mA/m^2)	V_{peak} (V)	P_{peak} (mW/m^2)
1	2.45	2.98	7.31
1.5	2.32	3.25	7.53
2.5	2.21	3.4	7.51
3.3	1.98	3.48	6.90
5	1.75	3.66	6.39
10	1.42	4.04	5.75
20	1.15	4.28	4.91
30	0.83	4.5	3.72
50	0.57	4.61	2.63
100	0.47	4.7	2.21
150	0.40	4.72	1.91

Table S6. Summary of the detailed output performances of Au/PET-based SIE-NG with a tensile strain of 0.21% at different frequencies under external load of 1.5 Ω. The three trials' output peak voltage and peak current densities values and the derived average, standard deviation, and power densities.

Frequency (Hz)	V_{peak} (V)			V_{Avg}	V_{Std}	J_{peak} (mA/m^2)			J_{Avg}	J_{Std}	P_{peak} (mW/m^2)	
	#1	#2	#3			#1	#2	#3			P_{Avg}	P_{Std}
0.01	1.57	1.55	1.72	1.61	0.08	2.35	2.24	2.31	**2.30**	0.04	**3.71**	0.20
0.05	2.01	1.86	1.98	1.95	0.06	3.10	3.06	3.26	**3.14**	0.09	**6.13**	0.32
0.1	2.56	2.01	2.11	2.23	0.24	3.83	3.90	3.97	**3.90**	0.06	**8.67**	0.83
0.25	3.07	2.63	2.92	2.87	0.18	4.06	4.35	4.31	**4.24**	0.13	**12.16**	0.52
0.5	3.35	2.82	3.24	3.14	0.23	4.71	4.85	4.80	**4.80**	0.06	**15.00**	0.94

Table S7. Comparison of output current density of the TENGs and the SIE-NG around 0.5 Hz.

Material	**Size**	**Frequency (Hz)**	**Output current density around 0.5Hz (mA/m^2)**	**Ref.**
PVA, glycerol,and acrylic resin / Kapton	2 x 2 cm^2	0.5 ~ 2.5	0.01	[14] *
PTFE balls / Kapton Nylon	Diameter 94 mm Height 5 mm	0.6 ~ 2.4	0.005	[17] *
Fluorinated ethylene propylene / Copper	Shell diameter 97 mm	0.1 ~ 0.5	2.78	[25] *
Nylon fiber / Polytetrafluoroethylene	Diameter 1.22 mm	0.3 ~ 1.5	0.164	[39]
Rubber sole / Acrylics	300 cm^2	0.25 ~ 2.0	0.0067	[40] *
Nylon / PVC	25 cm^2	0.5 ~ 2.5	0.02	[41] *
PTFE / Aluminum, Copper	351 cm^2	0.5 ~ 2.25	0.02	[42] *
Skin / HCOENPs/BP/PET	7 x 7 cm^2	1.0 ~ 6.0	4.10	[43] **
Au / PET, PI, UTG	7 x 7 cm^2	**0.01 ~ 0.5**	**4.80**	This work

*Note: The current densities were calculated from the size of friction layers in the TENGs.

**Note: The output current density at 1 Hz is tabulated because it is the lowest frequency at which the performance evaluation was conducted.

Table S8. Summary of the detailed current density during cyclic bending for up to 1,000,000 cycles at a frequency of 0.25 Hz with a tensile strain of 0.21%.

Working cycles	J_{peak} (mA/m^2)
100,000	1.96
200,000	2.18
300,000	2.07
400,000	1.92
500,000	2.09
600,000	1.91
700,000	2.11
800,000	1.95
900,000	2.12
1,000,000	2.16

Table S9. Summary of the detailed capacitor charging using 0.33. 1.00, and 2.40 μF capacitors at a frequency of 0.25 Hz with a tensile strain of 0.21%.

Timesteps	Capacitors (μF)	V_{peak} (V)
0 s	0.33	0.04
	1.00	0.02
	2.40	0.01
10 s	0.33	2.89
	1.00	1.55
	2.40	0.33
20s	0.33	4.26
	1.00	2.04
	2.40	0.57
30s	0.33	4.66
	1.00	2.44
	2.40	0.81
40s	0.33	4.72
	1.00	2.92
	2.40	0.89

Reference

1. A. Kiejna and K. F. Wojciechowski, *Metal surface electron physics*, Elsevier, 1996.
2. S. I. Park, J. H. Ahn, X. Feng, S. Wang, Y. Huang and J. A. Rogers, *Advanced Functional Materials*, 2008, **18**, 2673-2684.