## **Photo-stimulated Triboelectric Generation**

## **Supporting Information**

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## Experimental

**Preparation of Active blend layer.** A mixture of Poly(3-hexylthiophene) (P3HT) (EMindex Co.) and [6,6]-phenyl C61-butyric acid methyl ester ( $PC_{61}BM$ ) (EMindex Co.) was used as the active layer. P3HT and  $PC_{61}BM(1:0.8)$  were dissolved in Dichlorobenzene at a concentration of 1wt. % and the P3HT:PC<sub>61</sub>BM solution, and then stirred overnight at 80°C.

**Preparation of TiO**<sub>x</sub>. Titanium(IV) isopropoxide (Ti[OCH(CH3)2]4, 99.999%, Aldrich) was dissolved in isopropyl alcohol (IPA) at a concentration of 0.2 vol.% and the Titanium isopropoxide solution was heated to 80 °C for 2h under magnetic stirring. The thickness of the TiO<sub>x</sub> layer was controlled by the concentration of Titanium isopropoxide solution (from 0.1 to 0.4 vol.%).

*Device fabrication of P3HT/PCBM photovoltaics cell.* All devices were fabricated on indium tin oxide (ITO) patterned glass substrates, which were cleaned by detergent and sonicated in deionized water, acetone, and isopropyl alcohol for 10 min, respectively. The ITO substrates were exposed to  $O_2$  plasma for 20 min prior to spin-coating with poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS, AI 4083, Clevios) at 5000 rpm for 40 s. Next, the PEDOT:PSS-coated substrates were annealed at 140 °C for 10 min in air, and then taken into a nitrogen-filled glove box. The active blend layer of P3HT:PC<sub>61</sub>BM (EMindex Co.) was spin-coated onto PEDOT:PSS layer at 1100 rpm for 58 s. The Titanium isopropoxide solution was spin-coated onto the active blend layer at 4000 rpm for 40 s and was annealed at 80 °C for 10 min. And then, AI (100nm) was thermally evaporated on to the TiO<sub>x</sub> layer in a vacuum (<10<sup>-6</sup> Torr). The area of the AI electrode defines the active area of the device as 13.0 mm<sup>2</sup>.

*Fabrication of TENG.* The TENG consisted of two parts. On one side, The Al film was attached on a polyimide layer with a double-sided adhesive, used as a top electrode. On the other side, the TiOx/P3HT:PC<sub>61</sub>BM/PEDOT:PSS/ITO cell mentioned above was used as the

bottom layer. The top electrode was connected to the  $TiO_x$ . The TENG has an active area of around  $1 \times 1$  cm<sup>2</sup>. Four springs were installed on the acrylic plate to form a gap between the two layers and to support the layer.

*PV measurements.* Power conversion efficiency was determined from J-V curve measurements (Iviumstat, Ivium Technologies, Eindhoven, The Netherlands) under a 1 sun, AM 1.5G spectrum from a solar simulator (Portable Solar Simulator PEC-L01, Peccell Technologies Inc., Kanagawa, Japan; 1000 Wm<sup>-2</sup>).

*TENGs characterization and Outputs measurements.* AFM/KPFM Observation. To measure the surface potential of the device, Kelvin probe force microscopy (KPFM, Park Systems XE-70) measurements were performed with a frequency modulation (FM) mode, using Au/Wcoated silicon tips (radius < 25nm, force constant 3 N m–1, and resonance frequency of 75 kHz). 1  $\mu$ m × 1  $\mu$ m size KPFM images were scanned at a scanning speed of 0.5 Hz, and set at a point of 10 nm from a sample in atmospheric pressure at room temperature.

A pushing tester (Labworks Inc., model no. ET-126-4) was used to create vertical compressive strain in the nanogenerator. A Tektronix DPO 3052 Digital Phosphor Oscilloscope and a low-noise current preamplifier (model no. SR570, Stanford Research Systems, Inc.) were used for electrical measurements. The charge density from the output signals was measured using a Keithley 6514 system electrometer. The light was completely blocked by using a blackout curtain. This state is described as under dark. Then, AM 1.5G spectrum was applied to the device using a solar simulator. This state is described as under light.



**Figure. S1.** (a) The fabrication process of organic photovoltaic cells with  $Al/TiO_x/P3HT:PC_{61}BM/PEDOT:PSS/ITO/glass.$  (b) The current-density versus voltage (*J–V*) characteristics with and without TiO<sub>x</sub> layer. (c) The energy band diagrams for  $Al/TiO_x/P3HT:PC_{61}BM/PEDOT:PSS/ITO/glass under light illumination.$ 



Figure. S2. The surface potential map of the polyimide layer under dark condition.



**Figure. S3.** The output voltages of the triboelectric nanogenerators consisting of the Al/Polyimide// $TiO_x/ITO/glass$  in (a) and Al/Polyimide// $TiO_2/ITO/glass$  in (b) with the incident white light.



Figure. S4. The topography and surface potential maps of  $TiO_x/ITO/glass$  in (a) and  $TiO_2/ITO/glass$  in (b) with white light illumination.



**Figure. S5.** The core level spectra of O 1*s*, C 1*s*, and Ti 2*p* of the  $TiO_x$  layer before and after the physical contact. (a) under dark condition. (b) light illumination.



**Supplementary Movie 1.** The increase in the output voltage and current by the light illumination. (a) output voltage. (b) output current.



**Supplementary Movie 2.** Current variation under a step-like voltage difference. (a) applied 0 V. (b) applied 20 V. (c) applied - 20 V.