## Durable Flexible Direct Current Generation through the Tribovoltaic Effect at Contact-Separation Mode

Jia Meng,<sup>a,b</sup> Chongxiang Pan,<sup>a,b</sup> Longwei Li,<sup>b,c</sup> Zi Hao Guo,<sup>b,c</sup> Fan Xu<sup>b,c</sup>, Luyao Jia,<sup>b,c</sup> Zhong Lin Wang<sup>b,c,d,\*</sup> and Xiong Pu<sup>a,b,c,\*</sup>

<sup>a</sup>Center on Nanoenergy Research, School of Physical Science and Technology, Guangxi University, Nanning 530004, P. R. China.

<sup>b</sup>CAS Center for Excellence in Nanoscience, Beijing Key Laboratory of Micro-nano Energy and Sensor, Beijing Institute of Nanoenergy and Nanosystems, Chinese Academy of Sciences, Beijing 101400, P. R. China.

<sup>c</sup>School of Nanoscience and Technology, University of Chinese Academy of Sciences, Beijing 100049, P. R. China.

<sup>d</sup>School of Materials Science and Engineering, Georgia Institute of Technology, Atlanta, Georgia 30332-0245, USA.

\* Corresponding authors.

E-mail: <u>puxiong@binn.cas.cn</u>, <u>zlwang@gatech.edu</u>

Supplementary Note 1: The Optimization experiments of the PPy-coated textile. The PPy-coated textiles prepared have different internal resistances with different dosage ratios of py/FeCl<sub>3</sub>. As the py/FeCl<sub>3</sub> concentration ratio increases, the loading rate of PPy on the fabric gradually increases and tends to be saturated, while the internal resistance of the fabric gradually decreases with the py/FeCl<sub>3</sub> concentration ratio increases (Fig. S4). The physical properties of the fabrics are shown in the Table S1. As shown in Fig. S5, the loading of PPy on the fabric gradually increases with the increase of the py/FeCl<sub>3</sub> concentration ratio. Initially, PPy strives to wrap the surface of the numerous fibers of the fabric. Excess PPy begins to agglomerate at the fiber surface. As the  $py/FeCl_3$  concentration ratio increased from 1/4, the agglomeration of PPy on the surface of the as-prepared fibers became obvious. However, excess PPy agglomerates on the surface, making it easier to peel from the surface. As shown in Fig. S6, with the increase of the  $py/FeCl_3$  concentration ratio, the output of the generator gradually increases to the maximum value and then saturates. Obviously, when the  $py/FeCl_3$  concentration ratio is 1/2, the current output of the generator in the contact-separation mode is better without the agglomeration phenomenon in prepared textile, so the PPy-coated fabric prepared with this ratio is used for the experiment.



Fig. S1 Molecular structure of PPy (a) neutral, (b) polaron and (c) bipolaron.



Fig. S2 SEM images of (a) cotton textile with PPy coatings, (b) a single fiber with PPy coatings, (c) Local magnified single fiber with PPy coatings versus (d) a single cotton fiber without PPy coatings.



Fig. S3 The FTIR spectrum of the PPy and PPy-coated cotton textile.

Fig. S3 shows the FTIR spectra of the PPy-coated cotton textile and PPy. The FTIR spectrum of PPy-coated cotton textile shows the characteristic bands attributable to the ring-stretching mode of pyrrole ring at 1540 cm<sup>-1</sup>, C-C asymmetric stretching vibration at 1458 cm<sup>-1</sup>. The peak at 1294 cm<sup>-1</sup> was assigned to C-N stretching. The peak at 1026 cm<sup>-1</sup> and 960 cm<sup>-1</sup> corresponded to C-H in-plane and out-of-plane deformation vibration.



Fig. S4 The Effect of different py/FeCl<sub>3</sub> concentration ratio on PPy loading and internal resistance of PPy-coated cotton textiles.



Fig. S5 SEM images of the PPy-coated textiles prepared using a series of  $py/FeCl_3$  concentration ratios for (a) 6/1, (b) 4/1, (c) 2/1, (d) 1/1, (e) 1/2, (f) 1/4, (g) 1/6 and (h) 1/8.



Fig. S6 The influence of PPy-coated textiles with different concentration ratio of py/FeCl<sub>3</sub> on the output performance of the generator.



Fig. S7 Schematic diagram of testing force exerted on the flexible DC generator.



Fig. S8 Typical (a) voltage, (b) current and (c) charge output of the generator in approach-separation mode.



Fig. S9  $I_{SC}$  of the generator under contact-separation mode at various separation gap.



Fig. S10 Reverse of the connection to the external measurement. The current reverses its sign when changing the electric circuit connection. Red and purple curves are the corresponding short-circuit current of the device in forward and reverse connection with the electrometer, respectively.



Fig. S11 (a) Relative sliding pressure of the Al film and PPy-coated cotton textile and corresponding V<sub>OC</sub> and I<sub>SC</sub> output in one contact-separation cycle.
(b) Current output trends of the generator in sliding mode under different sliding pressure (v=0.3 m/s). (c) Continuous DC voltage and current outputs of the generator with an Al slider area of 0.7854 cm<sup>2</sup> and a force of ~8 N continuously circularly

sliding on the textile.



Fig. S12  $I_{SC}$  of the generator under contact-separation mode at various operation frequencies.



Fig. S13 Ups spectra of pristine PPy and secondary electron cutoff region (inset photo). Ultraviolet photoelectron spectroscopy (UPS) with an excitation energy of He I (21.2 eV) was employed to characterize the electronic structure of the PPy. The

work function of the material surface can be determined using WF =  $hv - E_{cutoff}$ . Thus, the work functions measured from these spectra for the PPy is 4.19 eV.



Fig. S14 Current-voltage characteristic curves of (a) Al-PPy-Al and Al-PPy-Au and (b) Au-PPy-Au with a contact area of 4 cm<sup>2</sup> under a force of 10 N.



Fig. S15 The V-Q curves of the generator in (a) sliding mode (43 kOhm loading resistance) and (b) contact-separation mode (510 Ohm loading resistance), respectively. The frequency is 6 Hz.



Fig. S16 Voltage profiles of four capacitors charged by five generators connected in series.



Fig. S17 Voltage-time curves of three capacitors charged by three generators connected in series under contact-separation mode.



Fig. S18 The SEM image of the generator in (a) the original state and (b) after being contact-separation 20,000 cycles under the reciprocating mode with a 8 N pressure force and a contact area of 0.785 cm<sup>2</sup>.



Fig. S19 The FTIR spectra of the PPy-coated textile in original state (the red line) and after being contact-separation 20,000 cycles (the purple line) under the reciprocating mode with a 8 N force and a contact area of 0.785 cm<sup>2</sup>.



Fig. S20 The photos of the PPy-coated cotton textile in (a) the original state and (b) after sliding 20,000 cycles under the reciprocating mode with a 8 N force and a slider area of 0.785 cm<sup>2</sup>.

$C_{py}/C_{FeCl3}$	Surface resistance (kΩ/□)	PPy loading (mg/cm²)	Weight gain rate (%)
6/1	49.2986	0.0132	0.1240
4/1	18.8814	0.0186	0.1737
2/1	6.6229	0.2125	2.0755
1/1	4.6430	1.0090	9.9488
1/2	0.9089	1.6181	16.7670
1/4	0.1800	3.3420	31.2523
1/6	0.1400	3.3555	31.4460
1/8	0.0975	3.4320	31.6580

 Table S1 Physical properties of PPy-coated textiles

Principles	Materials (motion mode)	Voltage (V)	Current (mA)	Charge (mC/m <sup>2</sup> )	Current density (A/m <sup>2</sup> )	Power (W/m <sup>2</sup> )	Flexibility	Pressure	Ref.
Tribo-tunneling	carbon aerogel and SiO <sub>2</sub> /Si (slide)	2	15	/	0.2	0.0606	×	/	11
Tribovoltaic effect	Stainless steel and n-Si (slide)	0.02	20	/	0.1	0.00015	×	/	12
Contact electrification	n-type GaN, p-type Si (slide)	130	21.5	23	0.215	2.80	×	/	24
Contact electrification	n-type GaN, p-type Bi <sub>2</sub> Te <sub>3</sub> (slide)	40	340	/	2.56	11.85	×	/	25
Schottky diode	Al, PPy, Au (press)	0.7	290	/	0.624	0.15	×	0.2 MPa	31
polymer-inorganic oxide junction	PPy, SnO <sub>2</sub> , Al, Au (press)	0.25	3.6	/	0.027	0.0018	×	238 MPa	32
Schottky junction	Al, GO-PPy, Au (press)	0.73	175	/	1.319	0.21	×	0.2 MPa	33
Schottky diode	Al, PANI, Au (press)	0.9	45	/	0.339	0.078	×	0.08 MPa	34
Schottky DC	Al, PPy-TiO <sub>2</sub> , Au (press)	0.84	572.7	/	4.31	0.62	×	100 kPa	35
Schottky diode	Al, PPy-coated fabric, Au (press)	3.27	329.69	/	0.8242	0.52	$\checkmark$	270 kPa	36
Tribovoltaic effect	AI, PEDOT-PSS, Ni (slide)	0.7	20	/	0.12	0.0011	$\checkmark$	/	19
Dynamic Schottky	PEDOT-PSS, AI (slide)	0.8	200	/	0.73	0.67	$\checkmark$	/	20
Organic semiconductor heterojunction	PEDOT-PSS, Al alloy (slide)	1	309	/	0.309	0.01167	$\checkmark$	/	21
Tribovoltaic effect	Al, PPy-coated cotton, Au (Contact-separation)	1.06	1256.47	98.72	16.006	5.40036	$\checkmark$	~ 8 N	our work

<b>Table 52</b> A summary of motion mode, output performances and nexiomity of semiconductor-ba	based generators.
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## Abbreviation

PPy, Polypyrrole; PANI, Polyaniline; PEDOT-PSS, Poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate)