Supplementary Information (SI) for Energy & Environmental Science. This journal is © The Royal Society of Chemistry 2024

## High triboelectrification and charge collection efficiency of direct current

triboelectric nanogenerator achieved by tri-synergistic enhancement strategy

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**Supplementary Figure 2:** The Output characteristics of dielectric materials with negative charge.

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**Supplementary Figure 4:** The Output characteristics of 1-unit TDD-TENG slider with different stator.

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**Figure S1.** Photographs of corona discharge of TDD-TENGs. a) Corona discharge photo of 1-unit TDD-TENG without PTFE powder treatment. b) Corona discharge photo of 1-unit TDD-TENG with PTFE powder treatment. c) Corona discharge photo of 4-unit TDD-TENG without PTFE powder treatment. d) Corona discharge photo of 4-unit TDD-TENG with PTFE powder treatment.



**Figure S2.** Output charge of dielectric materials with negative charge friction with PU.



Figure S3. Output charge of dielectric materials with positive charge friction with PU.



Figure S4. Output charge of 1-unit TDD-TENG slider with different stator.



**Figure S5.** Output charge of a) single PTFE slider and b) 1-unit TDD-TENG with different thickness of PTFE.



**Figure S6.** Output of 1-unit slider of TDD-TENG at different motion speed. a) Output charge and b) Output current of 1-unit TDD-TENG at different motion speed.



**Figure S7.** Scanning electron microscopy (SEM) images of PU foam with different thickness.



Figure S8. EDS mapping of PU foam after friction with PTFE.



**Figure S9.** Output charge of 1-unit slider with same material. a) Output charge of 1unit slider of PA b) Output charge of 1-unit slider of PTFE.



**Figure S10.** Output charge comparation of 1-unit slider of PTFE/X with and without PTFE pre-friction. a) Output charge of 1-unit slider of PTFE/X without PTFE pretreatment b) Output charge of 1-unit slider of PTFE/X with PTFE pretreatment



**Figure S11.** Output charge comparation of 1-unit slider of X/PA with and without PTFE pre-friction. a) Output charge of 1-unit slider of X/PA without PTFE pretreatment b) Output charge of 1-unit slider of X/PA with PTFE pretreatment



**Figure S12.** Output charge comparation of 1-unit slider of PTFE/X and X/PA with and without PTFE pretreatment. a) Output charge of 1-unit slider of PTFE/X with/without PTFE pretreatment b) Output charge of 1-unit slider of X/PA with PTFE pretreatment



Figure S13. Scanning electron microscopy (SEM) images of PU foam with spontaneously introduced PTFE and externally added PTFE power with size of  $10 \mu m$ .



**Figure S14.** Photos of PU with spontaneously introduced powder and externally added powder.



**Figure S15.** Output charge of single a) PTFE and b) PA slider rubbed with PU coated excessive PTFE power.



**Figure S16.** Output charge comparation of 1-unit slider of PTFE/PA with externally addation of different mass of PTFE power.



Figure S17. Morphology of PTFE film after 30 minutes of friction with thickness of  $20 \ \mu m$ .



Figure S18. Morphology of PA film after 30 minutes friction with thickness of 20  $\mu$ m.



Figure S19. TDD-TENG sliders with different units of PTFE/PA.



**Figure S20.** Output a)charge and b)current of TDD-TENG with different units of PTFE/PA without preatment of PTFE.



**Figure S21.** The output charge of 4-unit TDD-TENG at different motion speed with pretreatment of PTFE.



**Figure S22.** The output charge of traditional binary DC-TENG with blank areas in different units with/without pretreatment of PTFE.



**Figure S23.** The output a) current and b) voltage of sliding TDD-TENG with different units at load resisitances from  $0.01M\Omega$  to  $1G\Omega$  with pretreatment of PTFE at motion speed of  $0.72 \text{ m s}^{-1}$ .



Figure S24. The output power of sliding TDD-TENG with different units at load resisitances from 0.01 M $\Omega$  to 1 G $\Omega$  with pretreatment of PTFE at motion speed of 0.72 m s<sup>-1</sup>.



**Figure S25.** The leakage current of PU foam at different voltages (200-3000 V) with thickness of 2 mm, 3 mm, 4 mm, 5 mm.



Figure S26. The mechanism of DC-output on bottom electrode.



**Figure S27.** The discharge images of bottom electrodes. a) The photograph of the back of stator with a pair of bottom electrodes in light. b) The photograph of discharge on the back of stator in dark.



**Figure S28.** The output charge on unilateral and bilateral bottom electrode with at different unit on slider.



Figure S29. Photograph of rotary TDD-TENG with different unit.



**Figure S30.** The output charge of rotary TDD-TENG with different unit at speed of 5 rpm.



**Figure S31.** The comparation of output charge and current of different units' rotary TDD-TENG with/without pretreatment of PTFE.



**Figure S32.** Crest factor comparation of different units' rotary TDD-TENG with/without PTFE treatment.



**Figure S33.** DC output on bottom electrodes of 3-unit TDD-TENG at rotary speed of 5 rpm.

**Supplementary Note S1.** The calculation process of average power density of TDD-TENG.

The average power density is an important indicator for evaluating the output performance of TENG. The calculation method for the average power density in this work is as follows: Firstly, calculate the RMS current under the optimal matching impedance using Origin Lab software, with the following formula

$$I_{RMS} = \sqrt{\frac{1}{n} \sum_{t=1}^{n} I_t^2}$$

Among them, in sliding mode, n is the time taken to slide a distance of 120 mm, in rotary mode, n is 4s,  $I_t$  is the current value corresponding to each time point. Secondly, the average power density is calculated by

$$P_{er} = \frac{I_{RMS}^2 R}{S_f}$$

R is the resistance at the best matching impedance point,  $S_f$  is the total squre of PA and PTFE film on the slider.

**Supplementary Note S2.** The calculation process of maximum charge density of TDD-TENG.

The maximum charge density of rotatary TENG with different units were calculated as follow, by calculating the maximum transfer charge and friction area of PA and PTFE films on rotator for rotating TDD-TENGs with different units at a speed of 5 rpm, the maximum charge density can be obtained in following formula:

$$MCD = \frac{Q}{S_f}$$

Q is the maximum transfer charge at the speed of 5 rpm, S<sub>f</sub> is the total square of PA and PTFE at the speed of 5 rpm. And the corresponding data are shown in Table S2.

**Supplementary Table S1.** Relevant data for calculating *I<sub>RMS</sub>* at the optimal matching impedance of sliding TDD-TENG.

Unit	$\sum_{t=1}^{n} I_t^2$	$t_1$	tn	$\frac{1}{n} \sum_{t=1}^{n} I_t^2$	Irms (A)
4-unit	9.93E-11	22.4	23.001	1.65E-10	1.2857E-05

**Supplementary Table S2.** Relevant data for calculating the average power density of at the optimal matching impedance sliding TDD-TENG.

	-					
						Average
Unit	I <sub>RMS</sub>	R	Р	$S_{f}$	power	
	(A)	$(M\Omega)$	(W)	(m <sup>2</sup> )	density	
						(W m <sup>-2</sup> )
					0.0036	
					(0.054-	
4-unit	1.2857E-05 400 MG	400 ΜΩ	0.066121198	0.002×9)×	18.36699945	
					0.1	

Unit	Δt (s)	$\sum_{t=1}^{n} l_t^2$ (A)	I <i>rms</i> (A)	I <sub>peak</sub> (A)	CF
3	4.000	8.59E-11	4.63E-06	6.96E-06	1.502
3 (PTFE)	3.999	7.36E-10	1.36E-05	1.47E-05	1.084
6	3.999	2.50E-10	7.91E-06	1.04E-05	1.315
6 (PTFE)	3.999	1.23E-09	1.752E-05	1.90E-05	1.084
12	3.999	5.38E-09	3.668E-05	3.93E-05	1.071
12 (PTFE)	3.999	7.72E-09	4.39E-05	4.60E-05	1.047
18	3.999	5.94E-09	3.85E-05	4.23E-05	1.097
18 (PTFE)	3.999	1.21E-08	5.50E-05	5.77E-05	1.048
24	3.999	4.57E-09	3.38E-05	4.04 E-05	1.195
24 (PTFE)	3.998	1.41E-08	5.94E-05	6.16 E-05	1.036
30	3.999	7.36E-09	4.29E-05	5.20E-05	1.212
30 (PTFE)	3.998	2.65E-08	8.14E-05	8.37E-05	1.027
36	3.999	1.32E-8	5.76E-05	6.31E-05	1.096
36 (PTFE)	3.998	2.16E-08	7.36E-05	8.06E-05	1.095

**Supplementary Table S3.** *I*<sub>*RMS*</sub> and Crest Factor of TDD-TENG with different output units.

Unit	t (reach 20 μC at 5 rpm) (s)	Transfer Charge (μC s <sup>-2</sup> )	S <sub>total</sub> (mm <sup>2</sup> )	$S_f$ (at 5rpm) (mm <sup>2</sup> s <sup>-2</sup> )	Maximum charge density (mC m <sup>-2</sup> )
3	26.585	0.752	18540	1545	0.487
3 (PTFE)	25.834	0.774	18540	1545	0.501
6	17.75	1.127	18240	1520	0.741
6 (PTFE)	16.127	1.240	18240	1520	0.816
12	7.22	2.770	17640	1470	1.884
12 (PTFE)	4.094	4.885	17640	1470	3.323
18	4.96	4.032	17040	1420	2.839
18 (PTFE)	3.317	6.030	17040	1420	4.246
24	2.974	6.725	16440	1370	4.909
24 (PTFE)	2.341	8.543	16440	1370	6.236
30	2.122	9.425	15840	1320	7.140
30 (PTFE)	2.074	9.643	15840	1320	7.305
36	2.329	8.587	15240	1270	6.761
36 (PTFE)	2.167	9.229	15240	1270	7.267

**Supplementary Table S4.** Relevant data for calculating maximum charge density of TDD-TENG.

**Supplementary Table S5.** Relevant data for calculating average power density of rotary TDD-TENG.

Unit	I <sub>RMS</sub> (A)	<i>R</i> (ΜΩ)	Р (W)	$S_f$ (m <sup>2</sup> )	Average power density (W m <sup>-2</sup> )
30-unit (30 rpm)	1.89 E-05	200 ΜΩ	7.14E-02	0.66	1.08E+01

Ref	Article	Tribo-materials	AC/DC	Average power density (W m-2)
1	Zhang et.al (2014) Adv. Energy Mater	Al/PVC	DC	0.025
2	Zhang et.al (2024) Energy Environ. Sci	NC/PU	DC	0.071
3	Shan et.al (2021) Energy Environ. Sci	PA/Kapton	DC	0.095
4	Chen et.al (2022) Adv. Energy Mater	PTFE yarn	DC	0.18
5	Zhao et.al (2021) Nat. Commun	Cu/PVC	DC	0.2
6	Lei et.al (2020) Energy Environ. Sci	PA/Kapton/Cloth	DC	0.24
7	Zhang et.al (2023) Nat. Commun	Cu/PVC	DC	0.24
8	Liu et.al (2019) Sci. Adv	Cu/PTFE	DC	0.35
9	Zeng et.al (2022) Adv. Mater	PU/PTFE	DC	$0.398 \ W \ m^{-2}  Hz^{-1}$
10	He et.al (2022) ACS Nano	PTFE/PA	DC	0.89
11	Qiao et.al (2021) Nona Energy	Cu/FEP	DC	0.96
12	Chen et.al (2021) Energy Environ. Sci	Rabbit fur/PTFE	DC	1.11
13	Li et.al (2022) Energy Environ. Sci	PVC/Cu	DC	1.98
14	Luo et.al (2018) Adv. Energy Mater	Al/FEP	DC	2.03
15	Nat. Commun	ETFE/PI	DC	2.32
16	Adv. Energy Mater	PTFE/PA	DC	3
17	Fu et.al (2022) Nano Micro Letters	FEP/Al/PA	DC	3.1
18	Du et.al (2022) Adv. Funct. Mater.	FEP/PA	DC	4.2
19	Liu et.al (2022) Small	Cu/PTFE	DC	4.84

**Supplementary Table S6.** Comparison of charge density and average power density of DC-TENG.

20	Ryu et.al (2018)	PTFF/MC Nylon	DC	49	
20	Energy Environ. Sci		Бе	1.9	
21	Meng et.al (2022)	A 1/DD	DC	5 4	
	Energy Environ. Sci	Al/PPy		5.4	
22	He et.al (2022)		DC	$5.74 \text{ W m}^{-2} \text{ Hz}^{-1}$	
ZZ.	Adv. Energy Mater	ΓΓΕ/ΓΑ	DC		
23	Li et.al (2022)		DC	( <b>15</b> W? II]	
	Energy Environ. Sci	FIFE/FA/FEI	DC	0.15 W III 11Z	
24	Sun et.al (2022)	DTEE/DMMA/Dubhan	DC	7.9	
24	Energy Environ. Sci	PIFE/PININIA/Kubber			
25	Shan et.al (2022)	DTEE/Clath	DC	0.8	
23	Nano Energy	PTFE/Cloui	DC	9.8	
26	Li et.al (2024)		DC	0.00	
26	Energy Environ. Sci	PIFE/PU	DC	9.98	
27	Shan et.al (2023)		DC	12.4	
	Adv. Funct. Mater.	PIFE/PI/PU	DC		
	This work	PTFE/PA/PU	DC	18.37 (Sliding)	

## Reference

- C. Zhang, T. Zhou, W. Tang, C. Han, L. Zhang and Z. L. Wang, *Advanced Energy Materials*, 2014, 4.
- 2. X. Zhang, D. Ren, H. Wu, J. Wang, X. Li, H. Yang, Q. Li, Q. Yang, J. Zhu and Y. Xi, *Energy & Environmental Science*, 2024, **17**, 4175-4186.
- C. Shan, W. Liu, Z. Wang, X. Pu, W. He, Q. Tang, S. Fu, G. Li, L. Long, H. Guo, J. Sun, A. Liu and C. Hu, *Energy & Environmental Science*, 2021, 14, 5395-5405.
- 4. R. Cheng, C. Ning, P. Chen, F. Sheng, C. Wei, Y. Zhang, X. Peng, K. Dong and Z. L. Wang, *Advanced Energy Materials*, 2022, **12**.
- Z. Zhao, L. Zhou, S. Li, D. Liu, Y. Li, Y. Gao, Y. Liu, Y. Dai, J. Wang and Z. L. Wang, *Nat Commun*, 2021, **12**, 4686.
- R. Lei, Y. Shi, Y. Ding, J. Nie, S. Li, F. Wang, H. Zhai, X. Chen and Z. L. Wang, *Energy & Environmental Science*, 2020, 13, 2178-2190.
- J. Zhang, Y. Gao, D. Liu, J. S. Zhao and J. Wang, *Nat Commun*, 2023, 14, 3218.

- X. Y. Di Liu, Hengyu Guo, Linglin Zhou, Xinyuan Li, Chunlei Zhang, Jie Wang, Zhong Lin Wang, *Sci. Adv*, 2019, 5, eaav6437.
- Q. Zeng, A. Chen, X. Zhang, Y. Luo, L. Tan and X. Wang, *Adv Mater*, 2023, 35, e2208139.
- L. He, C. Zhang, B. Zhang, O. Yang, W. Yuan, L. Zhou, Z. Zhao, Z. Wu, J. Wang and Z. L. Wang, *ACS Nano*, 2022, 16, 6244-6254.
- G. Qiao, J. Wang, X. Yu, R. Jia, T. Cheng and Z. L. Wang, *Nano Energy*, 2021, **79**.
- P. Chen, J. An, R. Cheng, S. Shu, A. Berbille, T. Jiang and Z. L. Wang, Energy & Environmental Science, 2021, 14, 4523-4532.
- X. Li, C. Zhang, Y. Gao, Z. Zhao, Y. Hu, O. Yang, L. Liu, L. Zhou, J. Wang and Z. L. Wang, *Energy & Environmental Science*, 2022, 15, 1334-1345.
- J. Luo, L. Xu, W. Tang, T. Jiang, F. R. Fan, Y. Pang, L. Chen, Y. Zhang and Z. L. Wang, *Advanced Energy Materials*, 2018, 8.
- Y. Gao, L. He, D. Liu, J. Zhang, L. Zhou, Z. L. Wang and J. Wang, *Nat Commun*, 2024, 15, 4167.
- C. Shan, W. He, H. Wu, S. Fu, Q. Tang, Z. Wang, Y. Du, J. Wang, H. Guo and C. Hu, *Advanced Energy Materials*, 2022, 12.
- S. Fu, W. He, H. Wu, C. Shan, Y. Du, G. Li, P. Wang, H. Guo, J. Chen and C. Hu, *Nanomicro Lett*, 2022, 14, 155.
- Y. Du, S. Fu, C. Shan, H. Wu, W. He, J. Wang, H. Guo, G. Li, Z. Wang and C. Hu, *Advanced Functional Materials*, 2022, 32.
- L. Liu, Z. Zhao, Y. Li, X. Li, D. Liu, S. Li, Y. Gao, L. Zhou, J. Wang and Z.
   L. Wang, *Small*, 2022, 18, e2201402.
- 20. H. Ryu, J. H. Lee, U. Khan, S. S. Kwak, R. Hinchet and S.-W. Kim, *Energy & Environmental Science*, 2018, **11**, 2057-2063.
- J. Meng, C. Pan, L. Li, Z. H. Guo, F. Xu, L. Jia, Z. L. Wang and X. Pu, Energy & Environmental Science, 2022, 15, 5159-5167.
- W. He, C. Shan, H. Wu, S. Fu, Q. Li, G. Li, X. Zhang, Y. Du, J. Wang, X. Wang and C. Hu, *Advanced Energy Materials*, 2022, 12.

- 23. Q. Li, S. Fu, X. Li, H. Chen, W. He, Q. Yang, X. Zhang, H. Yang, D. Ren and Y. Xi, *Energy & Environmental Science*, 2023, **16**, 3514-3525.
- D.-J. Sun, W.-Z. Song, C.-L. Li, T. Chen, D.-S. Zhang, J. Zhang, S. Ramakrishna and Y.-Z. Long, *Nano Energy*, 2022, 101.
- 25. C. Shan, W. He, H. Wu, S. Fu, G. Li, Y. Du, J. Wang, Q. Mu, H. Guo, B. Liu and C. Hu, *Nano Energy*, 2022, **104**.
- K. Li, C. Shan, S. Fu, H. Wu, W. He, J. Wang, G. Li, Q. Mu, S. Du, Q. Zhao,C. Hu and H. Guo, *Energy & Environmental Science*, 2024, 17, 580-590.
- C. Shan, K. Li, H. Wu, S. Fu, A. Liu, J. Wang, X. Zhang, B. Liu, X. Wang and
   C. Hu, *Advanced Functional Materials*, 2023, 34.