Supporting Information

A Ternary Electrification Layered Architecture for High Performance Triboelectric Nanogenerators

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Supporting Figures:

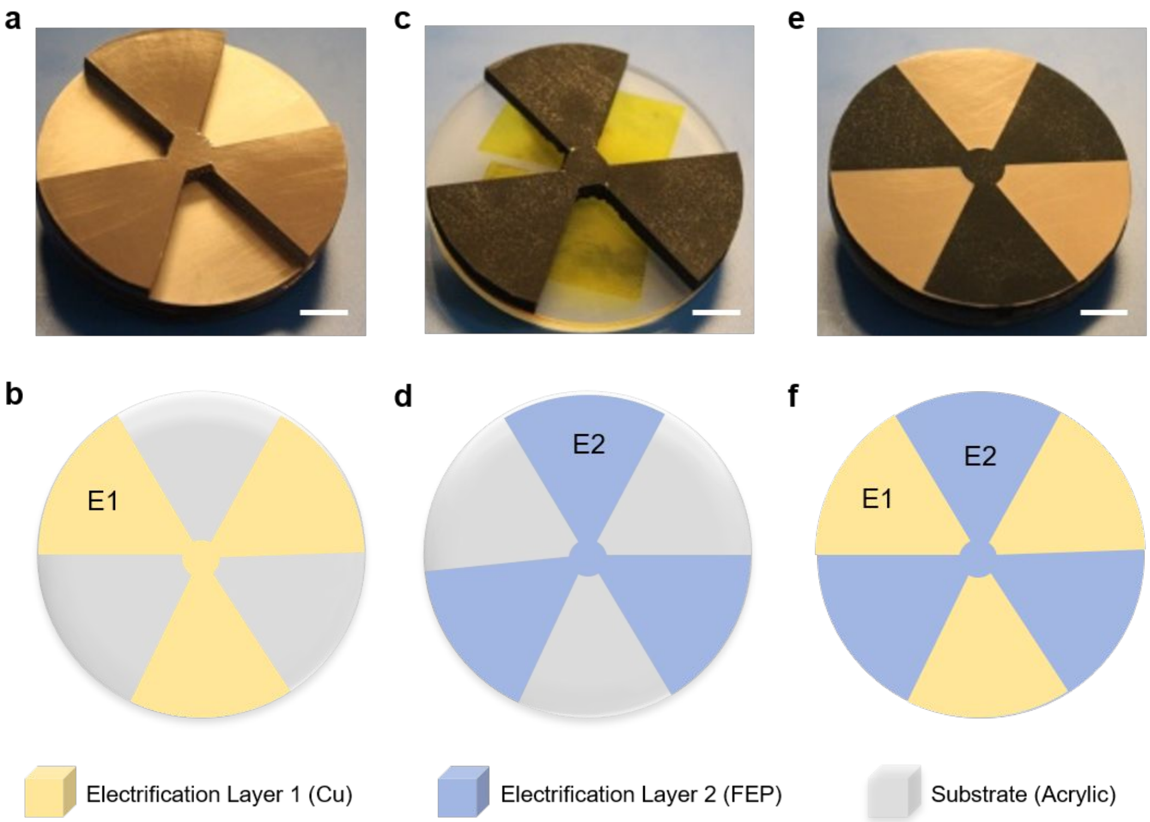

Figure S1. Schematic illustration of rotator with (a-b) Cu as electrification layer (EL1), (c-d) FEP as electrification layer, and (e-f) Cu/FEP as electrification layer (EL1/EL2). (The scale bars are 2 cm)

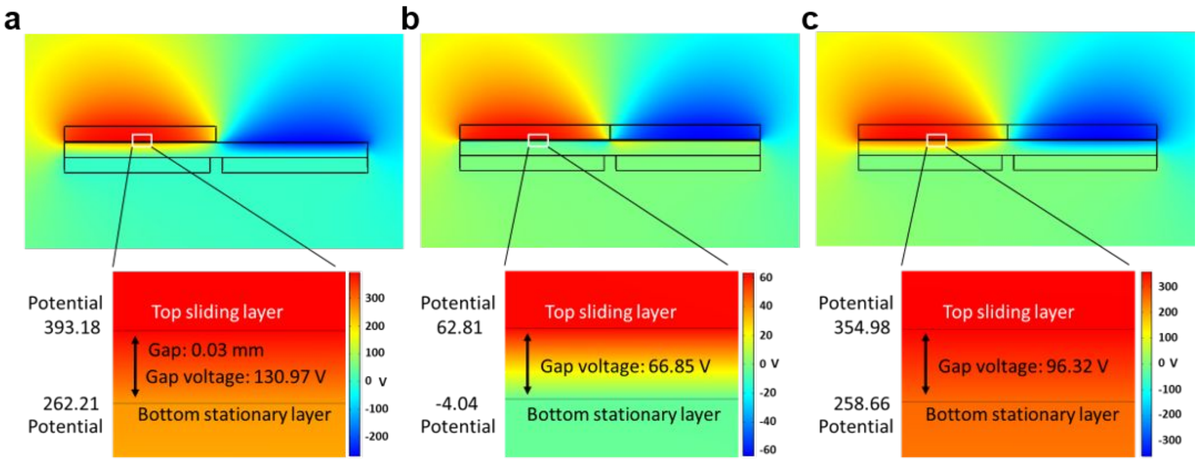

Figure S2. The simulation results of potential distribution between the two electrodes when the air gap is 0.03 mm. (a) EL1@EL3 based BEL-TENG, (b) The initial state of EL1/EL2@EL3 based TEL-TENG and (c) the saturated state of EL1/EL2@EL3 based TEL-TENG.

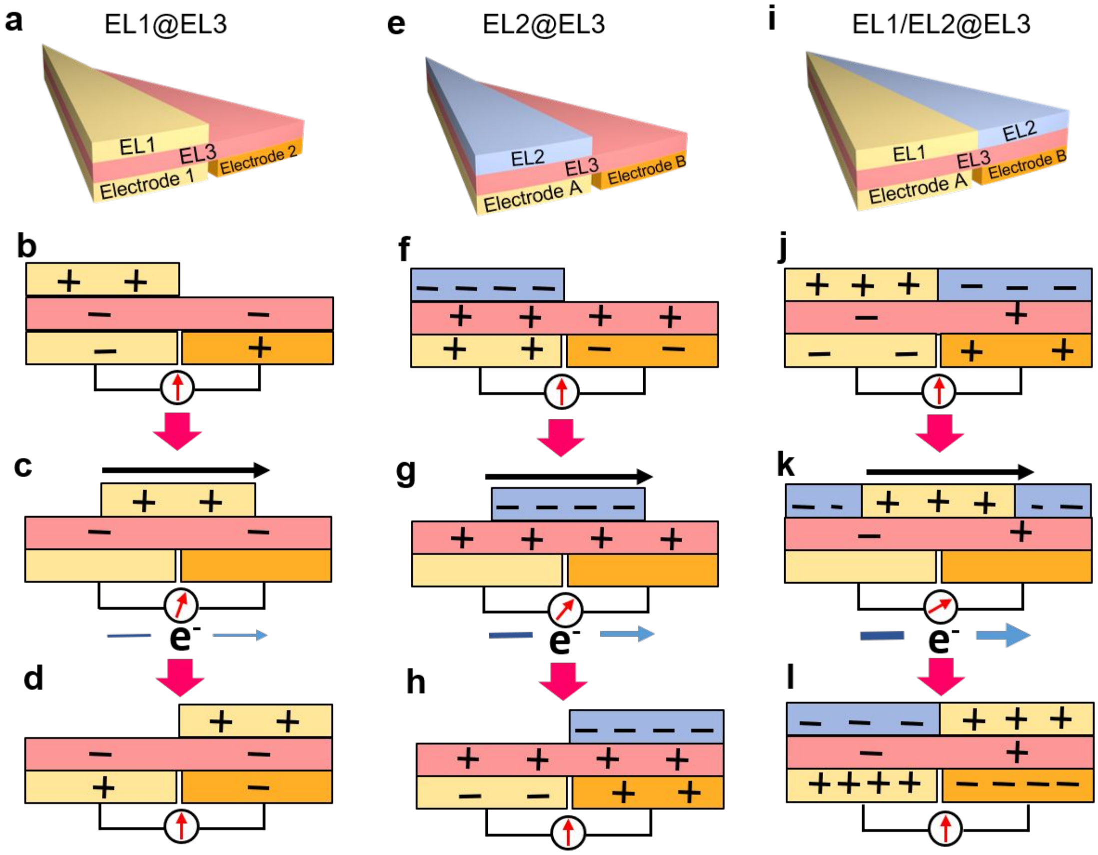

Figure S3. The operating principle of (a-d) EL1@EL3 based BEL-TENG, (e-h) EL2@EL3 based BEL-TENG and (i-l) EL1/EL2@EL3 based TEL-TENG.

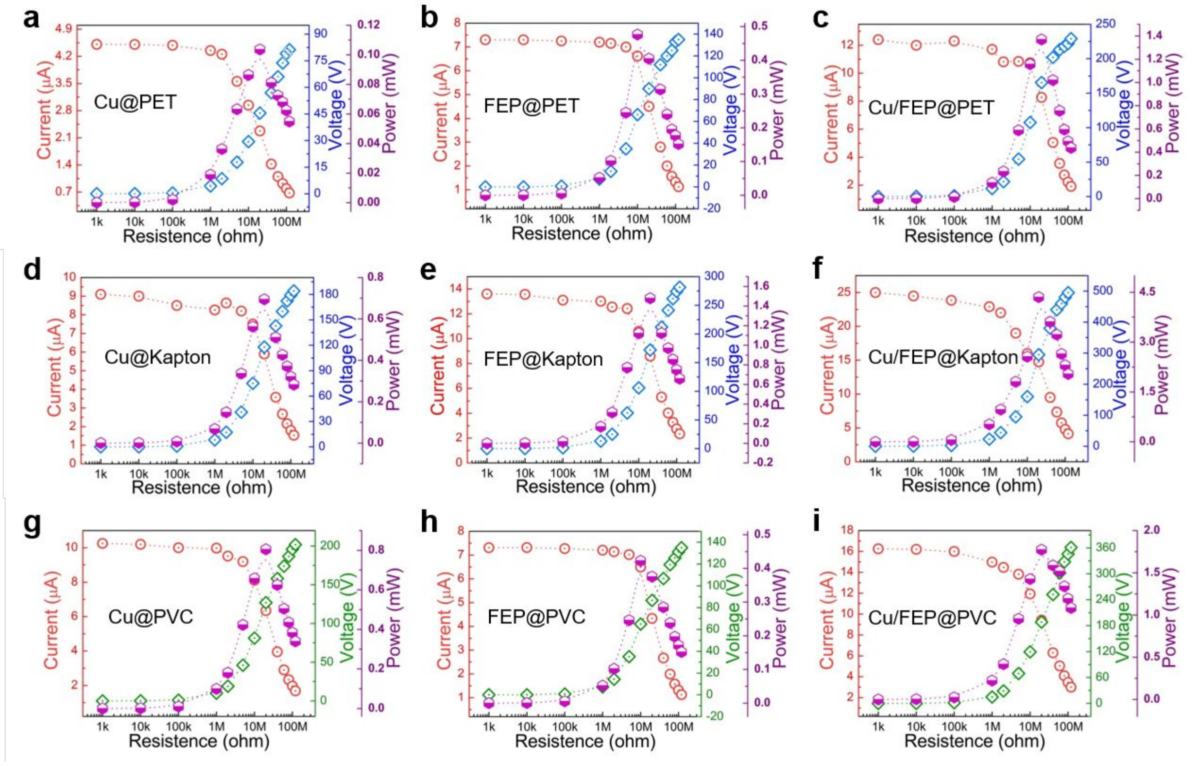

Figure S4. The output performance of BEL-TENG and the TEL-TENG with different intermediate electrification materials under variable external load resistances. (a-c) The performance of output power for the BEL-TENG and the TEL-TENG with PET as the intermediate material. (d-f) The performance of output power for the BEL-TENG and the TEL-TENG with Kapton as the intermediate material. (g-i) The performance of output power for the BEL-TENG and the TEL-TENG with PVC as the intermediate material. The unit center angle of 60° was used and the rotating speed of 600 rpm was fixed.

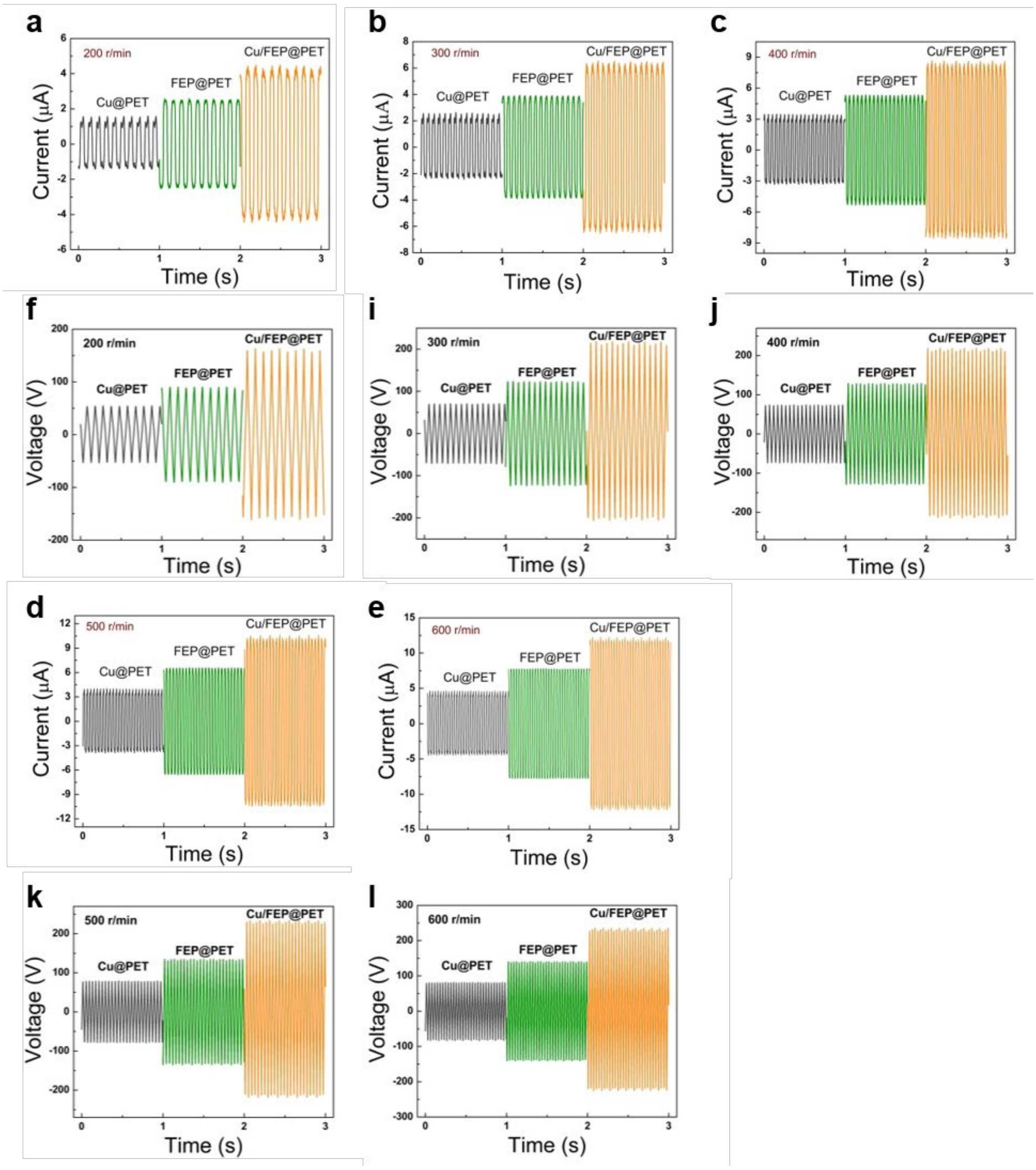

Figure S5. The electric output performance of short-circuit current (a-e) and voltage (f-l) for the BEL-TENG and the TEL-TENG with PET as the intermediate material. All of electric signals were measured at an external load resistance of 120 MΩ with a rotating speed of 600 rpm.

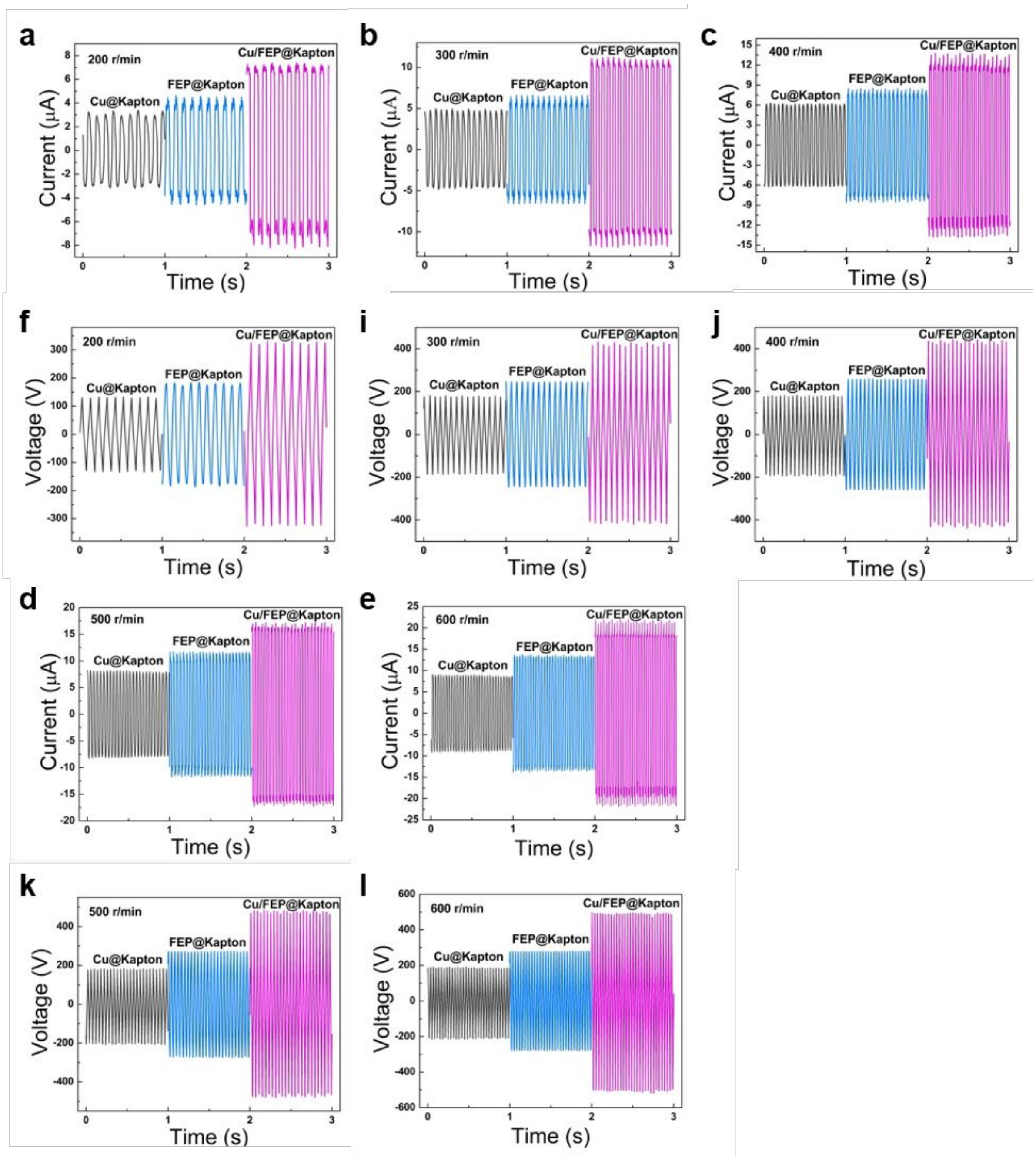

Figure S6. The electric output performance of short-circuit current (a-e) and voltage (f-l) for the BEL-TENG and the TEL-TENG with Kapton as the intermediate material. All of electric signals were measured at an external load resistance of 120 MΩ with a rotating speed of 600 rpm.

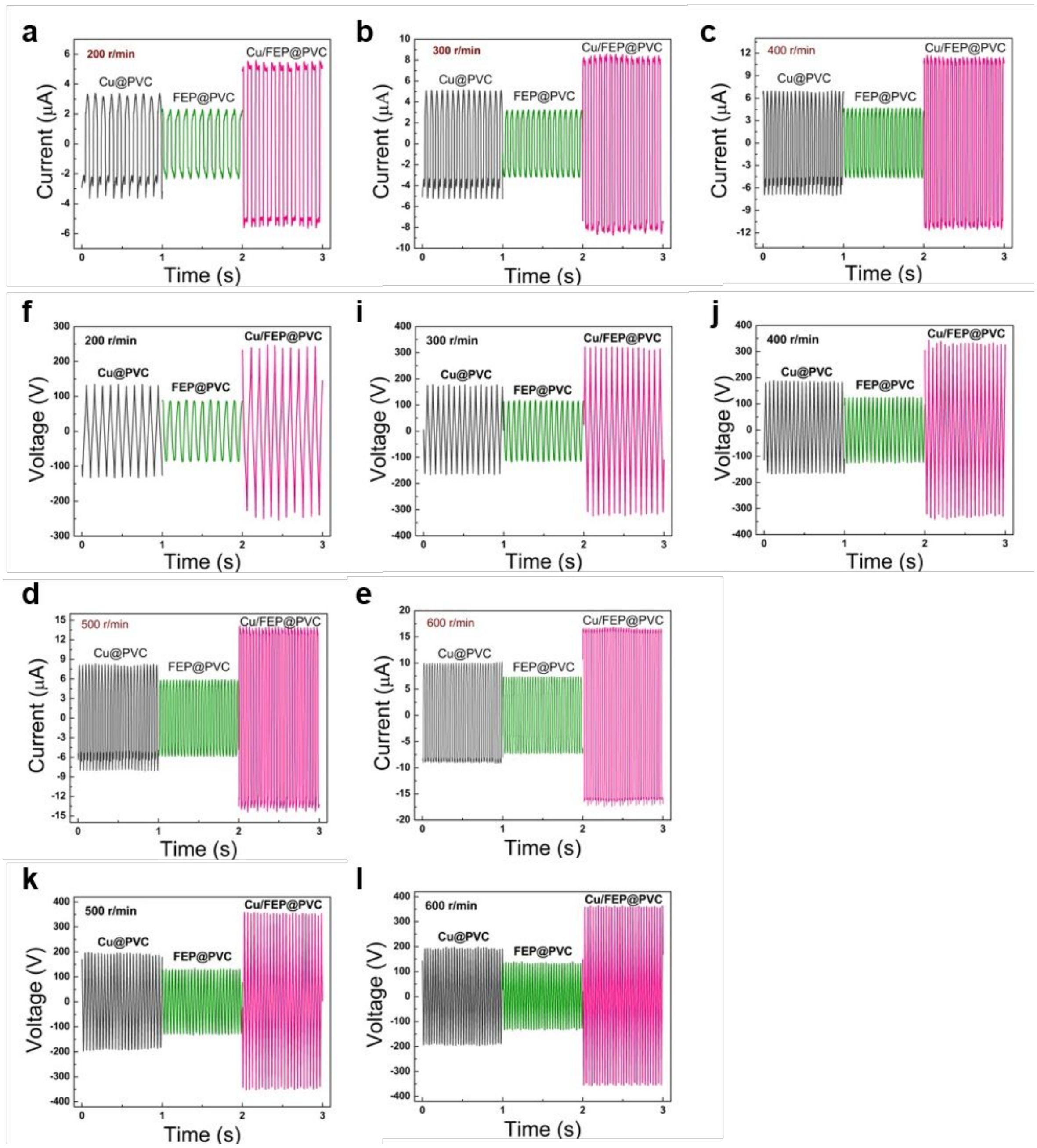

Figure S7. The electric output performance of short-circuit current (a-e) and voltage (f-l) for the BEL-TENG and the TEL-TENG with PVC as the intermediate material. All of electric signals were measured at an external load resistance of 120 MΩ with a rotating speed of 600 rpm.

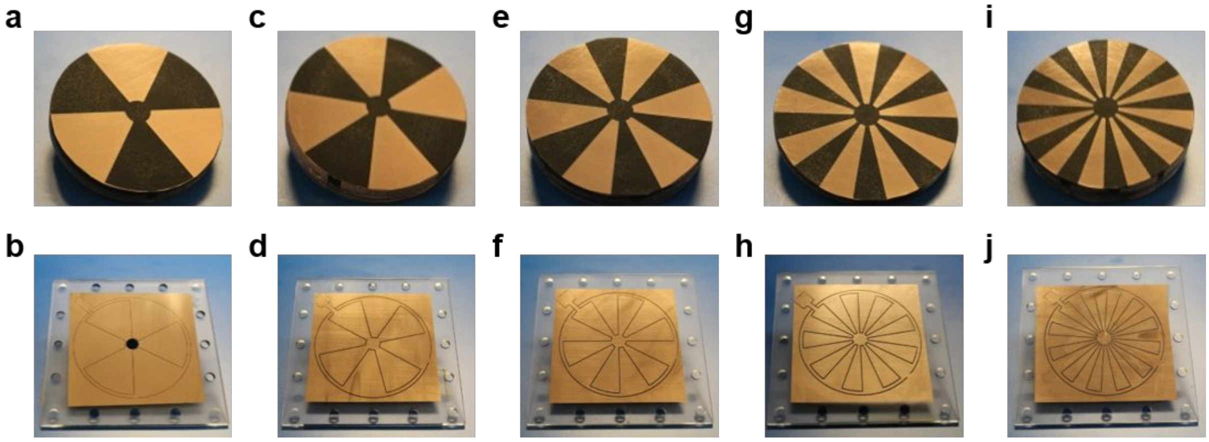

Figure S8. Corresponding photographs of stator and rotator with various unit center angles of (a-b) 60°, (c-d) 45°, (e-f) 30°, (g-h) 20°, and (i-j) 15°.

Supporting Note 1:

We assume the gap between electrodes are ignored. The rotation disc was separated into 2N parts (N is the number of blocks of each material). Then the signals should be periodically with period of $2\pi/N$. After contacting with interlayer, the rotation materials take charge density of $2\sigma_1$ and $-2\sigma_2$ ($2\sigma_1$ and $2\sigma_2$ are the charge density of each piece of the triboelectric material on the rotator, both σ_1, $\sigma_2 \geq 0$), respectively. Therefore, the interlayer will totally take the charge density of $2(\sigma_2-\sigma_1) = (-(2\sigma_1-2\sigma_2))$ to balance the total charge. Then the corresponding interlayer will take charge density of $(\sigma_2-\sigma_1)$, respectively. The electric potential distributions are depicted in Figure 4. Firstly, we can set MACRS (Minimum achievable reference charge state)[1] as the state in Figure 4j. So at that time, the rotation angle $\alpha = 0$, $V_{OC}(0) = 0$, and $Q_{SC}(0) = 0$. For the two other extreme status in Figure 4i and k, it has been reported the capacitance between the two electrodes can be considered as identical,[1-3] and we can assume it as C_p. When the radius of the rotator is R, therefore, the open-circuit voltages can be calculated by the short-circuit charge transfers over C_p:

The state of Figure 4i: $\alpha = -\frac{\pi}{2N}$ or $\frac{3\pi}{2N}$, $Q_{SC} = -(\sigma_1+\sigma_2)\frac{\pi R^2}{2}$, $V_{OC} = -(\sigma_1+\sigma_2)\frac{\pi R^2}{2C_p}$

The state of Figure 4k: $\alpha = -\frac{3\pi}{2N}$ or $\frac{\pi}{2N}$, $Q_{SC} = (\sigma_1+\sigma_2)\frac{\pi R^2}{2}$, $V_{OC} = (\sigma_1+\sigma_2)\frac{\pi R^2}{2C_p}$

These V_{OC} are the maximum and minimum values.

For an arbitrary α , the short-circuit charge transfer ($\alpha_0 = \pi/N$):

$$Q_{SC} = \begin{cases} \alpha(\sigma_1+\sigma_2)NR^2, -\frac{\pi}{2N} < \alpha < \frac{\pi}{2N} \\ (\alpha_0-\alpha)(\sigma_1+\sigma_2)NR^2, \frac{\pi}{2N} < \alpha < \frac{3\pi}{2N} \end{cases}$$

Therefore, the short-circuit current:

$$I_{SC} = \frac{\mathrm{d}Q_{SC}}{\mathrm{d}t} = \begin{cases} \frac{\mathrm{d}\alpha}{\mathrm{d}t}(\sigma_1+\sigma_2)NR^2, -\frac{\pi}{2N} < \alpha < \frac{\pi}{2N} \\ -\frac{\mathrm{d}\alpha}{\mathrm{d}t}(\sigma_1+\sigma_2)NR^2, \frac{\pi}{2N} < \alpha < \frac{3\pi}{2N} \end{cases}$$

These I_{SC} are the maximum and minimum values.

For the TENG with only one material, we can get the corresponding V_{OC} and I_{SC} by simply setting the charge density of the other material as zero. Therefore, due to the proportional relations between V_{OC} or I_{SC} and ($\sigma_1 + \sigma_2$), there will be:

V_{OC} (two materials)$_{max}$ = V_{OC} (material 1)$_{max}$ + V_{OC} (material 2)$_{max}$

V_{OC} (two materials)$_{min}$ = V_{OC} (material 1)$_{min}$ + V_{OC} (material 2)$_{min}$

I_{SC} (two materials)$_{max}$ = I_{SC} (material 1)$_{max}$ + I_{SC} (material 2)$_{max}$

I_{SC} (two materials)$_{min}$ = I_{SC} (material 1)$_{min}$ + I_{SC} (material 2)$_{min}$

Supporting References:

(1) Niu, S.; Liu, Y.; Chen, X.; Wang,S.; Zhou,Y. S.; Lin, L.; Xie, Y.; Wang, Z. L. Theory of Freestanding Triboelectric-Layer-Based Nanogenerators. *Nano Energy* **2015**, *12*, 760-774.

(2) Jiang, T.; Chen, X.; Han, C. B.; Tang, W.; Wang, Z. L. Theoretical Study of Rotary Freestanding Triboelectric Nanogenerators. *Adv. Funct. Mater.* **2015**, *25*, 2928-2938.

(3) Wang, S.; Xie, Y.; Niu, S.; Lin, L.; Wang, Z. L. Freestanding Triboelectric-Layer-Based Nanogenerators for Harvesting Energy from a Moving Object or Human Motion in Contact and Non-Contact Modes. *Adv. Mater.* **2014**, *26*, 2818-2824.

Supporting Table 1

Triboelectric table (tests were performed by Bill Lee (Ph.D., physics). ©2009 by AlphaLab, Inc. (TriField.com), which also manufactured the test equipment used. Column 1: Insulator name. Column 2: Charge affinity in nC/J. Column 3: Notes.

Polyurethane foam	+60	
Sorbothane	+58	Slightly conductive. (120 G ohm cm).
Box sealing tape (BOPP)	+55	More negative if sanded down to the BOPP film.
Hair, oily skin	+45	Skin is conductive. Cannot be charged by metal rubbing.
Solid polyurethane, filled	+40	Slightly conductive. (8 T ohm cm).
Magnesium fluoride (MgF_2)	+35	Anti-reflective optical coating.
Nylon, dry skin	+30	Skin is conductive. Cannot be charged by metal rubbing.
Machine oil	+29	
Nylatron (nylon filled with MoS_2)	+28	
Glass (soda)	+25	Slightly conductive. (Depends on humidity).
Paper (uncoated copy)	+10	Slightly conductive.
Wood (pine)	+7	
GE brand Silicone II (hardens in air)	+6	More positive than the other silicone chemistry.
Cotton	+5	Slightly conductive. (Depends on humidity).
Nitrile rubber	+3	
Wool	0	
Polycarbonate	-5	
ABS	-5	
Acrylic (polymethyl methacrylate)	-10	
Epoxy (circuit board)	-32	
Styrene-butadiene rubber (SBR, Buna S)	-35	Sometimes inaccurately called "neoprene" (see below).
Solvent-based spray paints	-38	May vary.
PET (mylar) cloth	-40	
PET (mylar) solid	-40	
EVA rubber for gaskets, filled	-55	Filled rubber will usually conduct.
Gum rubber	-60	Barely conductive. (500 T ohm cm).
Hot melt glue	-62	
Polystyrene	-70	
Polyimide	-70	
Silicones (air harden & thermoset, but *not* GE)	-72	
Vinyl: flexible (clear tubing)	-75	
Carton-sealing tape (BOPP)	-85	Raw surface is very +, but close to PP when sanded.
Olefins (alkenes): LDPE, HDPE, PP	-90	UHMWPE is below.
Cellulose nitrate	-93	
Office tape backing	-95	
UHMWPE	-95	
Neoprene (polychloroprene, *not* SBR)	-98	Slightly conductive if filled (1.5 T ohm cm).
PVC (rigid vinyl)	-100	
Latex (natural) rubber	-105	
Viton, filled	-117	Slightly conductive. (40 T ohm cm).
Epichlorohydrin rubber, filled	-118	Slightly conductive. (250 G ohm cm).
Santoprene rubber	-120	
Hypalon rubber, filled	-130	Slightly conductive. (30 T ohm cm).

Butyl rubber, filled	-135	Conductive. (900 M ohm cm). Test was done fast.
EDPM rubber, filled	-140	Slightly conductive. (40 T ohm cm).
PTFE (Teflon)	-190	Surface is fluorine atoms-- very electronegative.