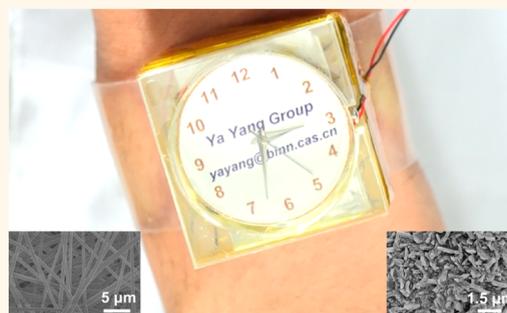


Hybridized Electromagnetic–Trieboelectric Nanogenerator for a Self-Powered Electronic Watch

Ting Quan,[†] Xue Wang,[†] Zhong Lin Wang,^{*,†,‡} and Ya Yang^{*,†}

[†]Beijing Institute of Nanoenergy and Nanosystems, Chinese Academy of Sciences, Beijing 100083, China and [‡]School of Materials Science and Engineering, Georgia Institute of Technology, Atlanta, Georgia 30332-0245, United States

ABSTRACT We report a hybridized nanogenerator including a triboelectric nanogenerator (TENG) and six electromagnetic generators (EMGs) that can effectively scavenge biomechanical energy for sustainably powering an electronic watch. Triggered by the natural motions of the wearer's wrist, a magnetic ball at the center in an acrylic box with coils on each side will collide with the walls, resulting in outputs from both the EMGs and the TENG. By using the hybridized nanogenerator to harvest the biomechanical energy, the electronic watch can be continuously powered under different motion types of the wearer's wrist, where the best approach is to charge a 100 μ F capacitor in 39 s to maintain the continuous operation of the watch for 456 s. To increase the working time of the watch further, a homemade Li-ion battery has been utilized as the energy storage unit for realizing the continuous working of the watch for about 218 min by using the hybridized nanogenerator to charge the battery within 32 min. This work will provide the opportunities for developing a nanogenerator-based built-in power source for self-powered wearable electronics such as an electronic watch.



KEYWORDS: hybridized · electromagnetic · triboelectric · self-powered · electronic watch

As wearable electronics are rapidly increasing, more recently, efforts have been focused on developing built-in energy harvesters for potential applications in extending the operation time of Li-ion batteries and ultimately realizing the self-charging capability of wearable electronics without external power sources.^{1–4} The scavenging of waste biomechanical energy from human locomotion is an ideal solution for sustainably powering these wearable electronics. Previous existing approaches have been demonstrated to convert biomechanical energy into electricity by utilizing different mechanisms such as electromagnetic,^{5,6} piezoelectric,^{7–11} or triboelectric effects.^{12–15} However, the output power of reported types of small-size energy harvesters alone still remains at quite a low level, where the produced electric energy is not enough to sustainably power some wearable electronics with larger power demands, especially under the operation conditions of small biomechanical forces

induced by body movements. Moreover, the energy harvester needs to work under a low frequency to effectively scavenge biomechanical energy from the natural motions of the human body.

Here, we have developed a hybridized electromagnetic–triboelectric nanogenerator that can deliver an output performance much higher than that of the individual energy-harvesting unit due to the cooperative operation of electromagnetic and triboelectric mechanisms under the same mechanical motions.^{16–19} We demonstrated a hybridized electromagnetic–triboelectric nanogenerator to convert the biomechanical energy from the natural motions of the wearer's wrist into electricity for sustainably powering an electronic watch. The transparent polyvinylbutyral (PVB) nanowire–polydimethylsiloxane (PDMS) composite film was used as both the watch belt and triboelectric nanogenerator materials, where the elastic modulus of the composite film has been enhanced to 6.6 MPa as compared with

* Address correspondence to
yayang@binn.cas.cn,
zlwang@gatech.edu.

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that of pure PDMS film (4.4 MPa). The electromagnetic generator consists of a magnetic ball at the center in an acrylic box with coils on each side. The fabricated hybridized nanogenerator has the total dimensions of $3.6\text{ cm} \times 3.6\text{ cm} \times 3\text{ cm}$ and the light weight of 46.9 g, which can be integrated in a commercial electronic watch to realize its self-powered function. The self-powered electronic watch can continuously work for 456 s by using a capacitor of $100\ \mu\text{F}$ charged by the hybridized nanogenerator in 39 s. Moreover, a homemade Li-ion battery was utilized to increase the continuous operation time of the electronic watch, where the working time can be up to 218 min after charging the Li-ion battery in 32 min by using a hybridized nanogenerator to scavenge the biomechanical energy from the natural motions of the wearer's wrist.

RESULTS AND DISCUSSION

Figure 1a illustrates the schematic diagram of the fabricated hybridized electromagnetic–triboelectric nanogenerator, where the electromagnetic generator (EMG) consists of a magnetic ball in an acrylic box and

six coils on each side of the acrylic box. The triboelectric nanogenerator (TENG) at the bottom of the acrylic box is composed of a nylon film on a Cu electrode and a PVB nanowire/PDMS composite film on a Cu electrode, where the transparent composite film acts as both the watch belt and triboelectric nanogenerator materials. The collision behavior between the magnetic ball and each wall of the acrylic box can induce the work of both the EMGs and the TENG. Figure 1b,c displays the photographs of the fabricated hybridized nanogenerator, which has the small external dimensions of $3.6\text{ cm} \times 3.6\text{ cm} \times 3\text{ cm}$ and a light weight of 46.9 g. Figure 1d shows a scanning electron microscopy (SEM) image of the fabricated PVB nanowires that were synthesized *via* an electrospinning method, indicating that the diameters of the PVB nanowires are $200 \pm 50\text{ nm}$ approximately. A surface SEM image of the PVB nanowire/PDMS composite film in Figure 1e shows that some PVB nanowires can still be observed on the surface of the composite film. Figure 1f illustrates an elastic modulus mapping figure of the PVB nanowire/PDMS composite film by using atomic force microscopy (AFM), clearly showing the distribution of the

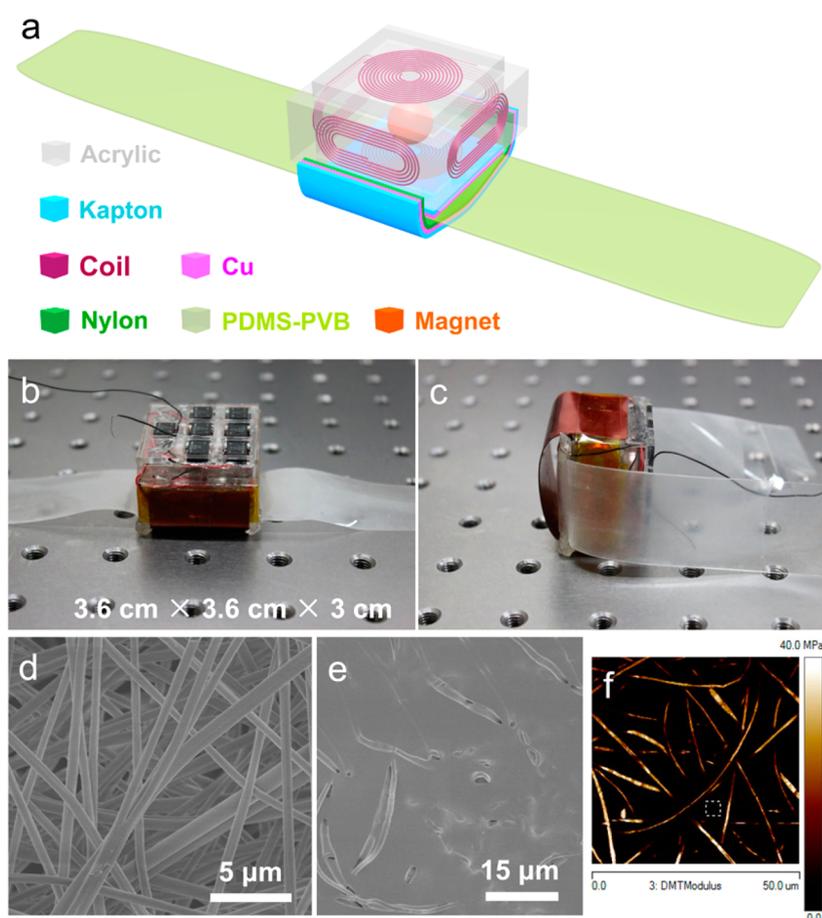


Figure 1. (a) Schematic diagram of the hybridized nanogenerator. (b,c) Photographs of the fabricated hybridized nanogenerator. (d,e) SEM images of the PVB nanowires (d) and the surface of the PVB nanowire/PDMS composite film (e). (f) Measured elastic modulus mapping figure of the PVB nanowire/PDMS composite film. The elastic modulus in the area of the white dashed lines is about 4.4 MPa.

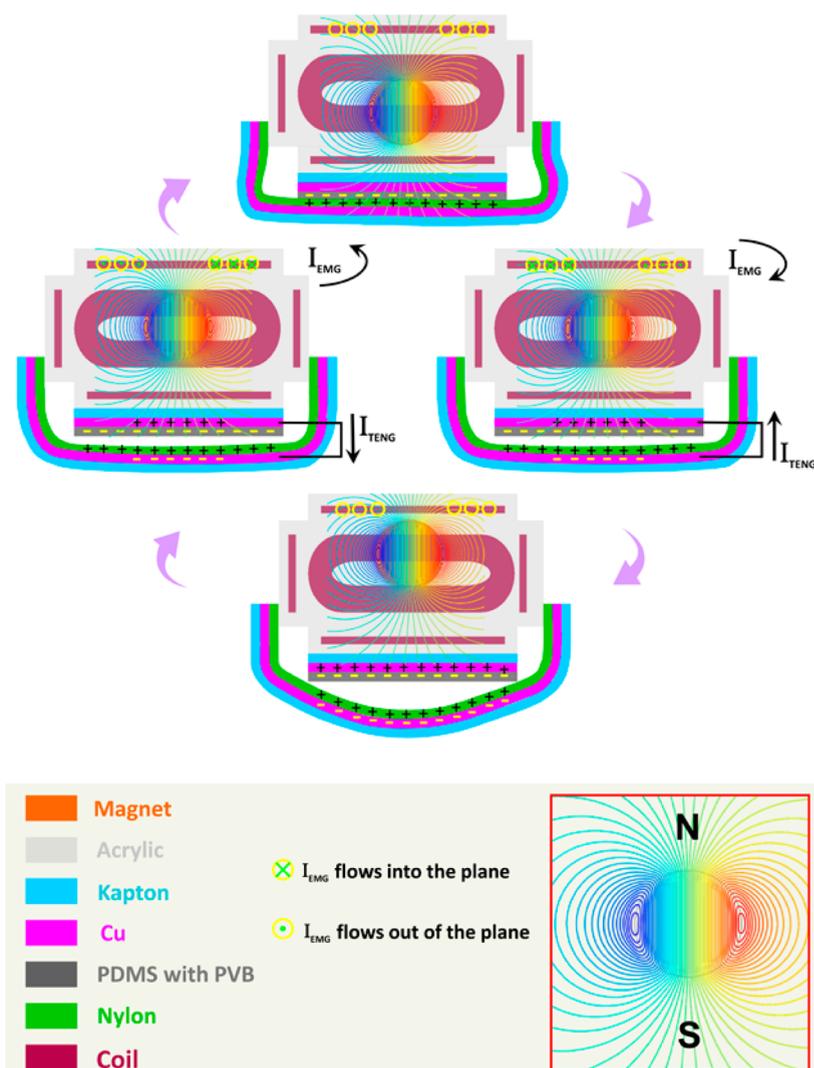


Figure 2. Working principle of the hybridized electromagnetic–triboelectric nanogenerator. When the magnetic ball was moved up and down in the acrylic box, both the triboelectric nanogenerator and electromagnetic generator can deliver the output voltage/current signals.

elastic modulus, where the bright lines indicate an elastic modulus much larger than that of the dark area in the dashed lines. The measured average elastic modulus of the composite film is about 6.6 MPa, which is obviously larger than that of the pure PDMS film with a corresponding value of about 4.4 MPa (Figure S1). The enhancement of the elastic modulus is beneficial to the robustness and mechanical durability of the device.

Figure 2 displays the electricity generation process of one top EMG and the TENG when the magnetic ball moves in one cycle along the vertical direction. At the initial state, under a compressive force applied on the hybridized nanogenerator, the PVB nanowire/PDMS composite film and the nylon film are brought into full contact with each other, resulting in the negative and positive triboelectric charges appearing on the surfaces of the composite film and the nylon film due to the different triboelectric polarities, respectively. The

nature of the insulator can ensure that the produced triboelectric charges exist for a long time.²⁰ Moreover, the magnetic field induced by the magnetic ball goes through the top coil. There is an observed current/voltage signal for both the TENG and the EMG. When the applied force is released and the magnetic ball is moved up, the electrons can be driven to flow between two Cu electrodes due to the electrostatic induction, resulting in the output current/voltage signals for the TENG. The magnetic flux through the top coil can be increased, inducing the electrons to flow in the top coil due to electromagnetic mechanism, resulting in the observed current/voltage signals for the EMG. When the magnetic ball is moved back from top to bottom and the distance between the PVB nanowire/PDMS composite film and the nylon film is decreased, the opposite output voltage/current signals can be observed. Thus, both the TENG and the EMG can deliver AC output signals in one operation cycle. Although the

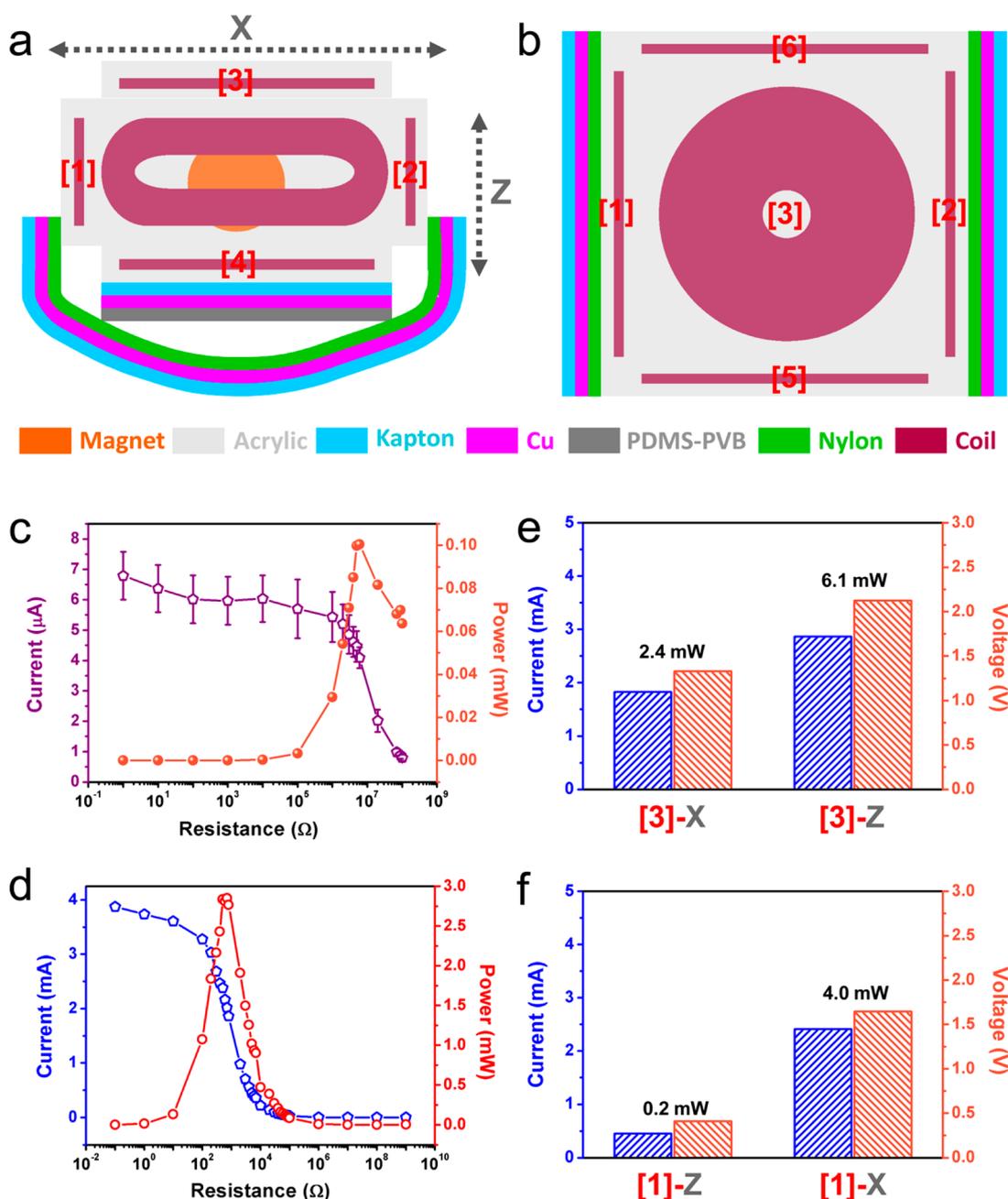


Figure 3. (a,b) Schematic diagrams of the fabricated hybridized nanogenerator with two different view angles. (c,d) Output current and power of the TENG (c) and the EMG (d). (e,f) Output current, voltage, and power of the EMGs under the different movement directions.

working process of only one EMG has been described in Figure 2, the movement of the magnetic ball in the acrylic box can induce the simultaneous working of all six EMGs due to the changes of magnetic flux through these coils.

As depicted in Figure 3a,b, the schematic diagrams of the cross section and top view of the hybridized nanogenerator clearly indicate the relative positions of the TENG and six EMGs. Under a vibration frequency of 22.5 Hz along the direction Z in Figure 3a, the TENG delivers the largest output power of 0.1 mW under a loading resistance of $6 \times 10^6 \Omega$ (Figure 3c).

As presented in Figure 3d, the EMG-3 can produce the largest output power of about 2.8 mW under a loading resistance of 700Ω along the direction X in Figure 3a, while the largest output power of EMG-3 can reach 6.1 mW along the direction Y in Figure 3a, as displayed in Figure 3e. As illustrated in Figure 3f, the largest output powers of EMG-1 are about 0.2 and 4 mW along the directions Z and X, respectively. All of the six EMGs can simultaneously work under the same movements of the magnetic ball in the acrylic box.

Figure 4a presents a photograph of the fabricated electronic watch, which can be integrated with the

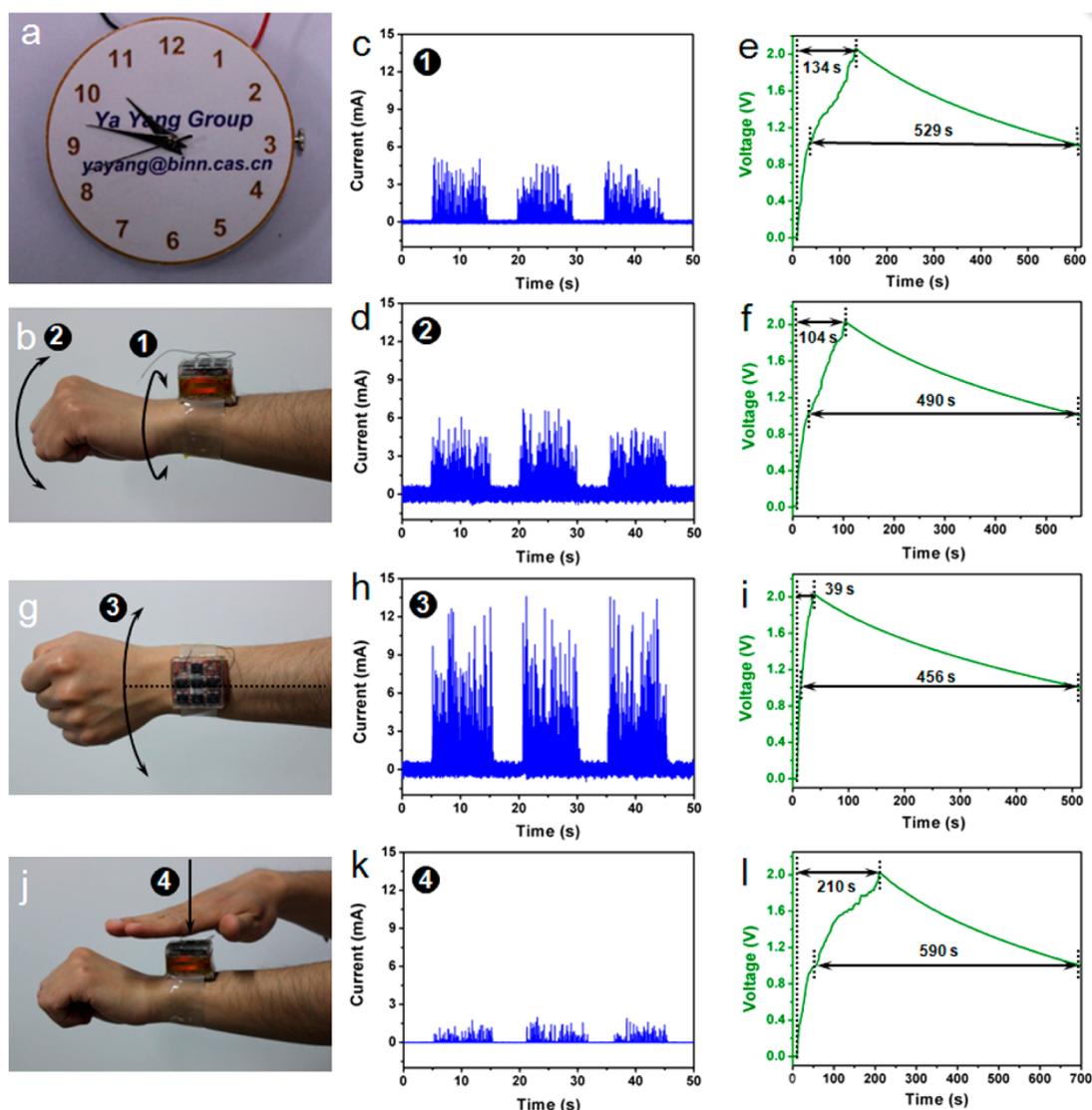


Figure 4. (a) Photograph of the fabricated electronic watch. (b–f) Photograph of the two different motion types (b), corresponding output current signals (c,d), and corresponding charging–discharging curves of a 100 μ F capacitor (e,f). (g–i) Photograph of another motion type (g), corresponding output current signals (h), and corresponding charging–discharging curve of a 100 μ F capacitor (i). (j–l) Photograph of another motion type (j), corresponding output current signals (k), and corresponding charging–discharging curve of a 100 μ F capacitor (l).

hybridized nanogenerator in Figure 1, where the TENG and EMGs were connected in parallel after using the rectification circuits. Under a constant voltage of 1.1 V, Figure S2a displays that the pulse current through the electronic watch is about 0.35 mA. The smallest working voltage of the electronic watch is about 1.06 V. The corresponding pulse power of the device is about 0.4 mW, which can be increased with increased applied voltage (Figure S2b). To evaluate the possibility of our hybridized nanogenerator to sustainably power the electronic watch, we measured the output current of the hybridized nanogenerator under the different motions of the wearer's wrist and the charging–discharging process of a 100 μ F capacitor that was directly connected to the electronic watch. As displayed in Figure 4b, the different motion methods of the wearer's wrist

can induce the different output current signals of the hybridized nanogenerator, as shown in Figure 4c,d. For the motion method 1, Figure 4e shows that the capacitor can be charged from 0 to 2 V in 134 s, where the electronic watch can work when the voltage of the capacitor is greater than 1.06 V, resulting in the continuous working time of the electronic watch for about 529 s. As presented in Figure 4f, the self-charging time of 104 s can induce the continuous working time of the watch for about 490 s. Figure 4g illustrates a photograph of another motion method of the wearer's wrist, which can result in the largest output current of up to 12 mA, as illustrated in Figure 4h. A self-charging time of only 39 s can sustain the continuous operation of the electronic watch for 456 s (Figure 4i). The produced smallest output current is less than 1 mA

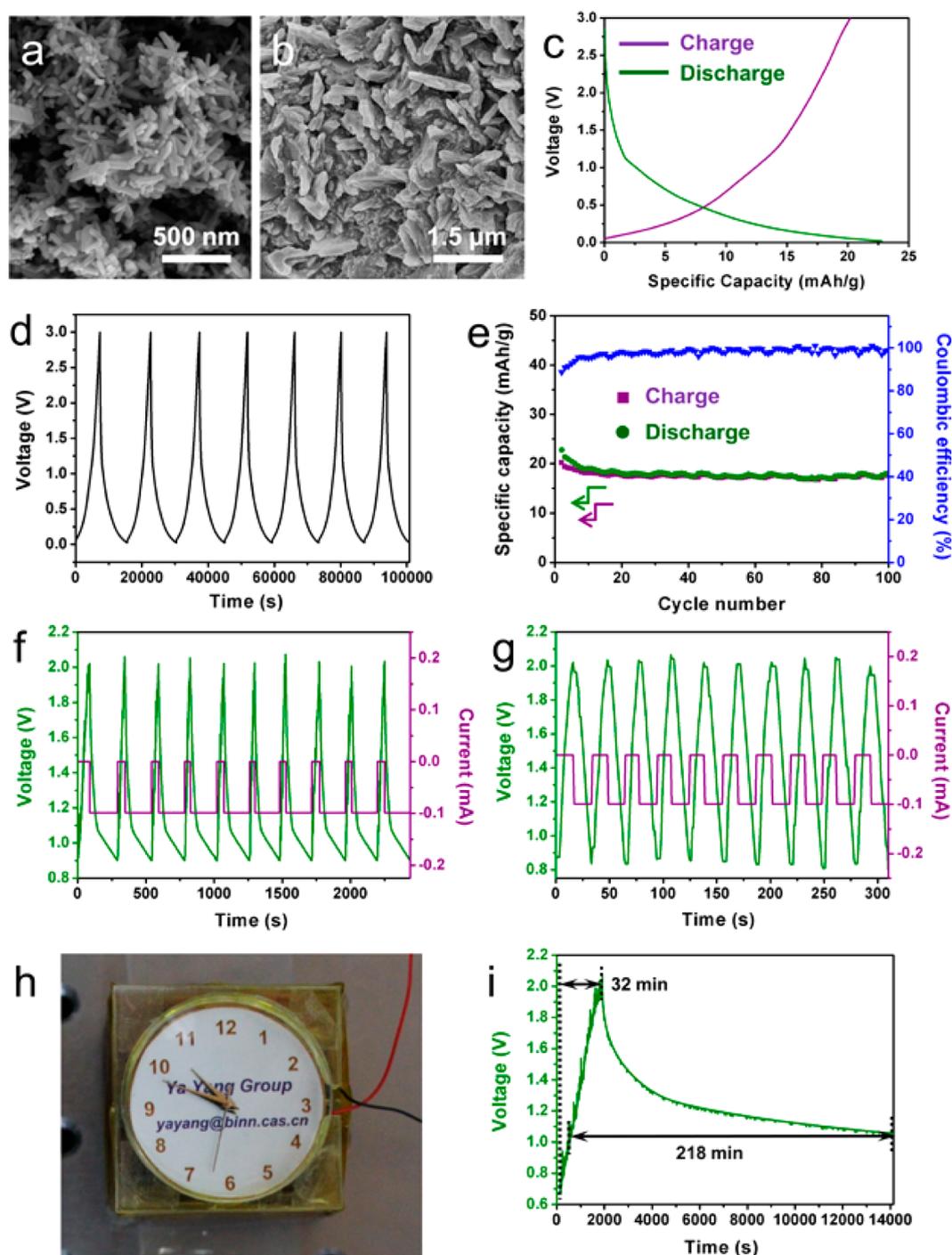


Figure 5. (a,b) SEM images of TiO_2 nanowires coated with carbon (a) and $\text{Li}_4\text{Ti}_5\text{O}_{12}$ nanomaterials coated with carbon (b). (c) Charge and discharge curves of the Li-ion battery. (d,e) Stability test of the Li-ion battery. (f,g) Charge and subsequent constant discharge curves of the Li-ion battery (f) and the $100\ \mu\text{F}$ capacitor (g) that were charged by using the hybridized nanogenerator. (h) Photograph of the fabricated self-powered electronic watch. (i) charge and discharge curves of the Li-ion battery that is connected to the electronic watch.

(Figure 4k) by compressing the hybridized nanogenerator as shown in Figure 4j. For this motion method, the electronic watch can also be continuously powered for 590 s after the capacitor was charged by the hybridized nanogenerator within 210 s, as presented in Figure 4l. These results clearly indicate that the hybridized nanogenerator can effectively convert the

mechanical energy from natural motions of the wearer's wrist into electricity for sustainably powering the electronic watch.

Although the self-powered electronic watch has been realized in Figure 4, the continuous operation time of the electronic watch is less than 400 s after the voltage of the capacitor is charged to 2 V. To increase

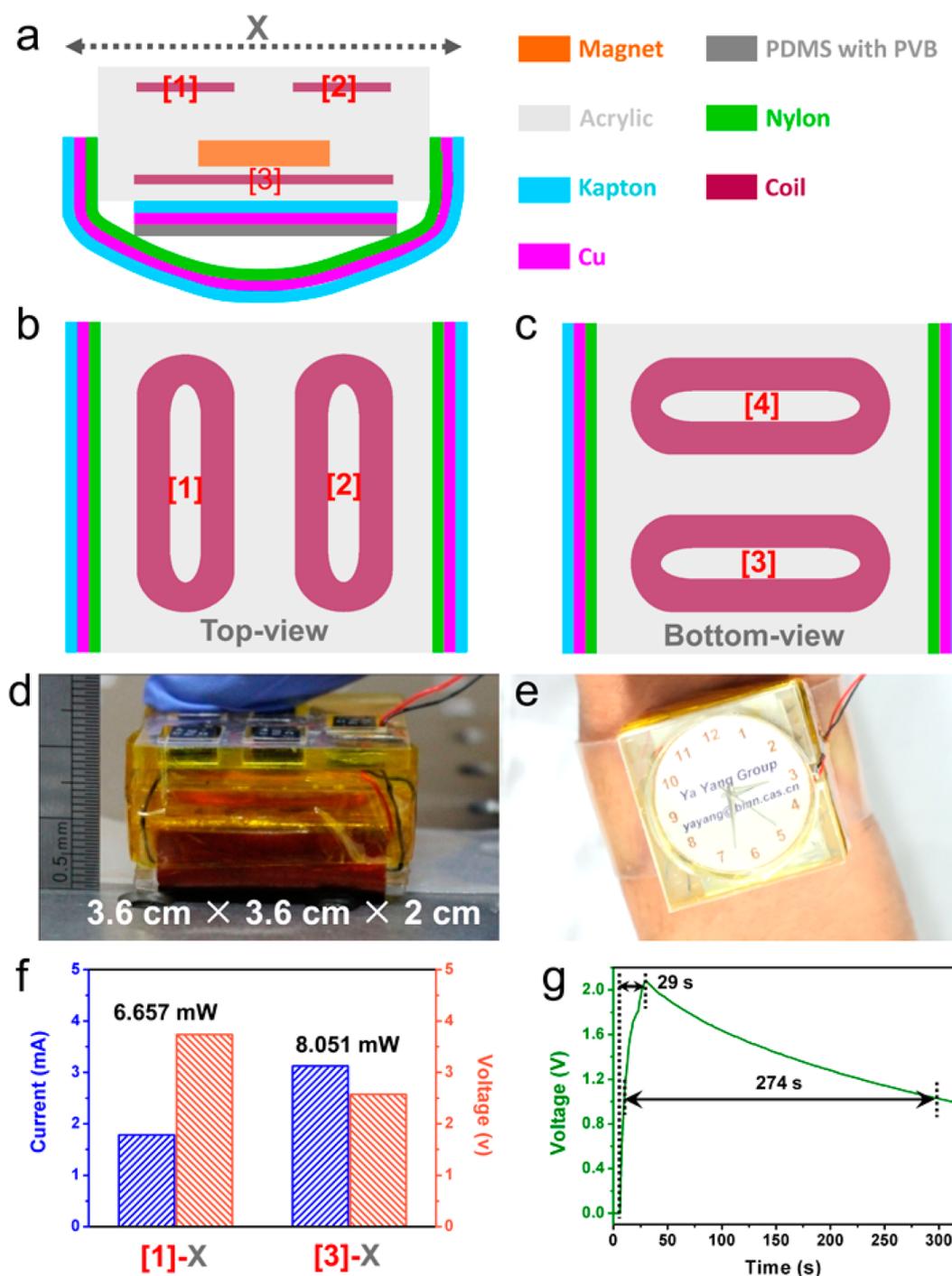


Figure 6. (a–c) Schematic diagrams of the hybridized nanogenerator with three different view angles. (d) Photograph of the fabricated hybridized nanogenerator. (e) Photograph of the self-powered electronic watch. (f) Measured output current, voltage, and power of the hybridized nanogenerator. (g) Charging–discharging curve of a 100 μ F capacitor that is connected to the electronic watch.

the continuous working time of the electronic watch under the low voltages of less than 2 V further, we fabricated a Li-ion battery that was utilized to replace the capacitor in Figure 4. The fabricated Li-ion battery consists of $\text{Li}_4\text{Ti}_5\text{O}_{12}$ nanomaterials coated by carbon as the anode, the foam nickel as the buffer layer, and a Li film as the anode. Figure 5a shows a SEM image of the grown TiO_2 nanowires coated by carbon, indicating

that the diameters of the nanowires are less than 100 nm. As presented in Figure 5b, the SEM image of the obtained $\text{Li}_4\text{Ti}_5\text{O}_{12}$ nanomaterials coated by carbon exhibits a size larger than that of the TiO_2 nanowires. Figure 5c illustrates the specific charge–discharge capacity curves of the fabricated Li-ion battery for the first time, where the specific charge and discharge capacities are about 20.2 and 22.8 mAh/g,

respectively, where the corresponding Coulombic efficiency is about 88.6%. As displayed in Figure 5d,e, the fabricated Li-ion battery exhibits stable charge–discharge characteristics, where the specific charge capacity is about 17.8 mAh/g after 100 charge–discharge cycles. As shown in Figure 5f, the fabricated Li-ion battery was charged and discharged by the hybridized nanogenerator attached to vibration equipment under a vibration frequency of 22.5 Hz, where the constant discharging current is 0.1 mA. The voltage of the Li-ion battery was increased from 0.9 to 2 V after it was charged by using the hybridized nanogenerator in 86 s. As compared with the 100 μ F capacitor used in Figure 5g, the fabricated Li-ion battery has a much greater charging and discharging time by using the hybridized nanogenerator.

Figure 5h illustrates the self-powered electronic watch that consists of a hybridized nanogenerator as the power source, an electronic watch as the function unit, and a fabricated Li-ion battery as the energy storage unit. By harvesting the biomechanical energy from natural motions of the wearer's wrist, the produced electric energy can be utilized to charge the Li-ion battery to sustainably power the electronic watch. As displayed in Figure 5i, the fabricated Li-ion battery can be charged by the hybridized nanogenerator from 0.66 to 2 V in 32 min, which can be utilized to sustain the continuous operation of the electronic watch for 218 min. As compared to the working time (<400 s under a charged voltage of 2 V by using a 100 μ F capacitor) of the electronic watch in Figure 4, the continuous working time of the electronic watch using the Li-ion battery has been enhanced greater than 30 times. Moreover, we can easily calculate that with a charging time of only 3.5 h using the hybridized nanogenerator to convert natural motions of the wearer's wrist into electricity, a continuous working time for the electronic watch in 24 h can be realized.

To decrease the dimensions of the hybridized nanogenerator further, we also investigated the possibility of using a small magnetic sheet to replace the magnetic ball in Figure 1. Figure 6a–c illustrates the schematic diagrams of the hybridized nanogenerator with the different view angles, clearly showing the relative positions of the TENG and EMGs. Figure 6d displays a photograph of the fabricated hybridized nanogenerator with the corresponding size of 3.6 cm \times 3.6 cm \times 2 cm, which is smaller than that of the hybridized

nanogenerator in Figure 1. As shown in Figure 6e, the self-powered electronic watch can be achieved by using the hybridized nanogenerator to scavenge the biomechanical energy from the natural motions of the wearer's wrist, which can also be seen in the Supporting Information movie. Figure 6f presents the output powers of the two EMGs in the hybridized nanogenerator, where the EMG-3 can deliver the largest output power of about 8 mW when the magnetic sheet was moved in the acrylic box along the direction *X* in Figure 6a. Moreover, the charge and discharge data in Figure 6g show that a continuous operation time of 274 s can be achieved by using the hybridized nanogenerator to charge a 100 μ F capacitor in 29 s. In this study, although the magnetic sheet has been used to replace the magnetic ball, we found that the motions of the magnetic sheet are much more difficult than the magnetic ball. Thus, other effective methods to largely decrease the total dimensions of the hybridized nanogenerator will be the focus of future research. Although the EMGs have a contribution for the produced electric energy that is larger than that of TENG, the TENG has some advantages, such as low weight, small volume, and flexible properties, which are important for wearable electronics.

CONCLUSIONS

In summary, a self-powered electronic watch has been realized by using a hybridized electromagnetic–triboelectric nanogenerator for scavenging biomechanical energy from natural motions of the wearer's wrist to sustainably power it. The working mechanism of the hybridized nanogenerator is to utilize the collision between a magnetic ball and the coils to induce the simultaneous working of TENG and EMGs. The produced electric energy by the hybridized nanogenerator can be stored in a 100 μ F capacitor to provide continuous operation of the electronic watch for 456 s after the capacitor is charged for 39 s. To increase the continuous working time of the electronic watch, a homemade Li-ion battery instead of the 100 μ F capacitor was utilized as the storage unit in the self-powered system, where a continuous working time of 218 min has been realized by using the hybridized nanogenerator to scavenge biomechanical energy from natural motions of the wearer's wrist to charge the Li-ion battery for 32 min. The fabricated hybridized electromagnetic–triboelectric nanogenerator has potential applications for biomechanical energy harvesting and self-powered personal electronics.

EXPERIMENTAL SECTION

Fabrication of the Hybridized Nanogenerator. The fabricated hybridized nanogenerator includes six EMGs and one TENG. For the EMGs, a magnetic ball was put into an acrylic box with each

coil on each side. The movement of the magnetic ball in the acrylic box can result in the change of magnetic flux through each coil, inducing the simultaneous working of six EMGs. The TENG consists of a nylon film with a thickness of 50 μ m as the triboelectric material, a PVB nanowire/PDMS composite film

as another triboelectric material, and two Cu electrodes. The change of distance between the nylon film and the composite film can result in the working of the TENG. The fabrication of the composite film is as follows: First, the PVB nanowires were fabricated by using an electrospinning method, where the PVB powders and the ethanol solution in a ratio of 3 g/50 mL were mixed for the fabrication of PVB nanowires. After the PVB nanowires were grown on the conductive substrate, a PDMS solution was dropped on the surface of the PVB nanowires, where the PDMS elastomer and the cross-linker were mixed in a ratio of 10:1 (w/w). After the PDMS solution was uniformly coated on the surface of PVB nanowires, the composite film was then dried at 80 °C for 3 h in an oven. The obtained PVB nanowire/PDMS composite film was utilized as the triboelectric material in the TENG device. The TENG and six EMGs were connected in parallel after using seven rectification circuits. The hybridized nanogenerator has the total mass of about 46.9 g and external dimensions of 3.6 cm × 3.6 cm × 3 cm.

Fabrication of the Li-Ion Battery. The fabricated Li-ion battery is based on the $\text{Li}_4\text{Ti}_5\text{O}_{12}$ nanomaterials coated with carbon as the anode with the raw materials of TiCl_4 , PVA (polyvinylalcohol), and LiCO_3 . The detailed fabrication process is as follows: First, TiO_4 was prepared by a hydrothermal method. Then, 38.32 g of pure TiO_4 liquid was mixed with some pure water in a clean container, which was placed in an ice–water bath with the temperature less than 5 °C. The solution was stirred thoroughly until there was no white sediment or white smoke, where the concentration of TiCl_4 was 0.5 M. The obtained precursor solution was poured into a Teflon-lined stainless steel autoclave and heated at 160 °C for 24 h. After being cooled, the product was centrifuged and the mixed solution was washed with ethyl alcohol several times to remove the impurity and then dried in a vacuum-drying oven at 80 °C for 24 h. After that, the obtained precursor TiO_2 was mixed evenly with PVA (10 wt %) and then put in the tube furnace under the protection of N_2 at 700 °C for 1 h to produce the TiO_2 nanowires coated with carbon. Then, the powder was mixed with Li_2CO_3 and put in the tube furnace under the protection of N_2 at 800 °C for 10 h to generate the final products. The molar ratio of TiO_2 and Li_2CO_3 was 4:5. The working electrode of the Li-ion battery was fabricated by mixing the carbon-coated Li_2CO_3 (8 wt %) with super-C (1 wt %) and adhesive (1 wt %). Then, the slurry coated on the Cu foil was dried in the vacuum-drying oven at 120 °C for 12 h and then was cut into disks with a radius of 10 mm. After that, the battery was assembled in the argon-filled glovebox, using a CR2025 button-type battery as the test model, lithium foil as the counter electrode, CR2300 porous polypropylene membrane as the diaphragm, the mixture of 1 mol/L LiPF_6 and EC/DMC/EMC (volume ratio was 1:1:1) (EC is ethylene carbonate; DMC is dimethyl carbonate; EMC is ethyl methyl carbonate) as the electrolyte, and foam nickel as the buffer layer.

Measurement of the Hybridized Nanogenerator. The output current signals of the TENG under the different loading resistances were measured by using a low-noise current preamplifier (Stanford Research SR570). The output voltage and current signals of the EMGs under the different loading resistances were performed by using a programmable electrometer (Keithley model 6514). The elastic modulus mapping figure of the PVB nanowire/PDMS composite film was performed by using an AFM (Dimension Icon). An electrodynamic shaker was used to produce the periodic vibrations with the frequencies ranging from 2 to 100 Hz.

Conflict of Interest: The authors declare no competing financial interest.

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Supporting Information Available: The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acsnano.5b05598.

Movie file displays the continuous working of the self-powered electronic watch by using the hybridized nanogenerator to scavenge the biomechanical energy from natural motions of the wearer's wrist (AVI)

Additional figures of the elastic modulus mapping figure of the pure PDMS film, the measured current through the electronic watch under a constant voltage of 1.1 V, the measured current signals, and the corresponding powers of the electronic watch under the different applied voltages (PDF)

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