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5 **Sustainable and Flexible Microbrush-Faced Triboelectric Generator for**
6 **Portable/Wearable Applications**

7

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1 Triboelectric nanogenerators (TENGs) have been put forward as a state-of-the-art energy
2 scavenging technology for self-powered electronics, but their severe wear and degradation
3 driven by inevitable friction can pose significant durability and sustainability concerns. Here,
4 an array of microfibers is reported that functions as a robust and sustainable TENG in both in-
5 plane sliding and vertical contact-separation modes, with excellent electrical potential as high
6 as 20 V and a high cyclability of 3000. The design flexibility of our microbrush TENG (MB-
7 TENG) on the counter materials facilitates the further improvement of electrical outputs,
8 benefiting numerous applications of human-interactive triboelectrification. Significantly, our
9 MB-TENGs offer sufficient output power for successfully driving a smartwatch as well as an
10 electromyography module. This technology uses a simple and cost-effective manner to provide
11 a robust and reliable monolithic TENG module, which is expected to serve as a promising
12 energy harvesting source for self-powered electronics in the near future.

13

1 Internet of Things (IoT) technology is emerging to take an essential role in smart cities,
2 where advanced electronics exchange information without requiring human intervention.
3 Although IoT holds the promise of enriching the lives of humans, the technology is expected to
4 encounter massive power consumption issues from the billions of advanced electronics
5 connected through the network. Endeavors to meet the energy demands have focused on
6 developing alternative powering technology, which harvests the abandoned energy resources
7 in an independent, sustainable, and maintenance-free manner. In particular, devices for
8 mechanical energy harvesting – including piezoelectric and triboelectric nanogenerators
9 (PENGs^[1-6] and TENGs^[7-14], respectively) – have attracted considerable attention given that
10 mechanical energy is ubiquitous. Indeed, TENGs offer many advantages over conventional
11 energy harvesting technologies, including their light weight, simple fabrication, material
12 selection diversity, and high energy conversion efficiency.^[15-17] However, as TENGs are based
13 on the working mechanism of contact electrification and electrostatic induction, the inevitable
14 friction causes severe abrasion between the layer surfaces in the contact area, thereby leading
15 to a significant degradation in output power over the cycles.^[18-20] The inherent lower durability
16 and sustainability that originate from the working mechanism seriously influence the
17 competitiveness of TENGs compared with other renewable energy harvesting technologies.^[21]
18 Some common working environments in which TENGs will be applied, such as with increased
19 dust, moisture, and temperature, may accelerate the mechanical wear.^[22-28] Therefore, TENGs
20 that exhibit robustness and toughness in the process of repetitive mechanical stimuli are needed
21 urgently. Moreover, the minute control of the operating environments of TENGs has expanded
22 their application range in structural configurations with variable surface shapes by adopting
23 unusual contact-electrification mechanisms and the critical effects on specific materials
24 systems.^[29] Progress in the robustness and reliability of TENGs has been made using various
25 approaches including noncontact mode,^[30,31] rolling structural,^[32,33] liquid-solid contact,^[34,35]
26 self-recovery,^[36,37] and encapsulated TENGs.^[38,39] At the same time, an easy and

1 straightforward route to improve the durability of TENGs is the structural design methodology
2 with suitable materials. Brush-type surfaces have been designed to provide stable and reliable
3 contact compared with the monolithic surface under the same elastic load.^[40] In a recent report,
4 the millimeter-long fur brush has been presented as a lightweight, soft, and low-wear
5 triboelectric material for intriguing TENGs with high elasticity, improved stability, and
6 relatively high density.^[41] Compared with a traditional monolithic design, these brushes offer
7 the following features against the friction: 1) an increased contact area, 2) less contact resistance,
8 3) high resistance to abrasion, 4) less heat generation, and 5) relatively high resilience.
9 Therefore, a tribosurface modified with brushes is a promising candidate for the contact
10 electrification component of durable TENGs. Herein, we report a robust and reliable new-type
11 TENG featuring a surface built with an array of microfibers by using the intrinsic structural
12 advantages of the brush. The array of microfiber bundles (i.e., 6×6, 36 pin-arrays of bundles)
13 implemented into a supporter displays excellent resilience and flexibility, affording a large
14 contact area without any device failure even at large deformation. Our TENG exhibits
15 remarkable capability for mechanical energy harvesting in separate working modes, including
16 in-plane sliding and vertical contact separation. Human being interactive triboelectrification
17 with our TENG reveals outstanding power generation and durability, suggesting that our TENG
18 can serve as a power source for various handheld electronics.

19 The microbrush architecture of our microfiber-based TENG (hereafter referred to as
20 MB-TENG) comprises an array of microfiber bundles implanted into a holder with an electrical
21 lead wire, as schematically illustrated in **Figure 1a**. The microfiber bundles (i.e., nylon) were
22 transplanted from commercially available chopped fiber-bundles to the pin grid array sockets
23 (i.e., holder, 3D printed polylactic acid, PLA), in which the electrical contacts were precoated
24 using Ag paste over the surface in the frame structure, as described in Figure 1b. Each bundle
25 of fibers was tightly plugged and fixed to the electrically conductive sockets through the curing
26 of the Ag paste, while the other side of the bundles was secured as free-standing, allowing

1 multiple degrees of freedom within this geometric configuration (Figure 1b). The fully
2 integrated MB-TENG device module employed approximately 1,980 microfibers in a given
3 area ($20 \times 25 \text{ mm}^2$), as shown in Figure 1c. The surface of an individual microfiber measured
4 by scanning electron microscopy exhibits a rough morphology, facilitating contact
5 electrification by enlarging the effective contact area. As manually counted in Figure 1d, the
6 microfibers in the arrays had a typical average length of ~9-10 mm and an average diameter of
7 ~197.6 μm . Along with the advantageous resilience of the individual microfibers, the presented
8 brush structure possesses excellent mechanical durability in elasticity and flexibility in the
9 operation of the TENG. Thus, the structural novelty enables the MB-TENG to markedly tolerate
10 a wide range of axial deformation (i.e., d_0-d_Δ) by enlarging the contact area against counter
11 materials (Figure 1e); the contact area of a single microfiber bundle with respect to transparent
12 substrates could be visualized under the various axial deformations (Figure S1a, Supporting
13 Information). The representative digital images show an apparent increase in the contact area
14 on both rigid glass and soft polydimethylsiloxane (PDMS) substrates upon an increase in the
15 axial deformation (Figure S1b). Control over the elastic properties of counter materials is a
16 critical factor in engendering a large contact area, with a quantitative analysis revealing a
17 boosted contact area with an increasing deformation up to the highest value of ~17.05 mm^2 for
18 a strain of ~70% (Figure S1c).

19 The typical mechanical tests for the stress-strain response on the 6×6 pin-array of the microfiber
20 bundles compared with the single microfiber bundle are presented in Figure S2. The array
21 structure of the bundled microfibers represents a ~1.5-fold higher maximum compressive stress
22 compared with the single pin-module, and the modulus of resilience calculated from the
23 integration of the stress-strain curve was measured to be $E_{\text{array}} = 14.9 \text{ MN m m}^{-3}$ and $E_{\text{single}} =$
24 9.5 MN m m^{-3} . In particular, the brush structure did not exhibit any permanent deformation
25 even after the stress was removed. These unique structural properties will greatly benefit the
26 materials design for the triboelectrification effects, and are due largely to the remarkable

1 resilience and flexible nature of the brush structure, which also maximizes the contact area by
2 elastically deforming the shape of the individually arranged microfibers.^[42]

3 The triboelectrification of MB-TENG can be operated by repetitive contacts with the
4 counter materials deposited on the Al electrode (Figure 1f), categorized into two different
5 working modes. First, the nylon in the MB-TENG (i.e., the pins of the microfiber bundles)
6 served as a triboelectrically active material that can be rubbed with the counter materials
7 through the in-plane slide mode. Since nylon is one of the most positive materials in the
8 triboelectric series, its surface generally tends to delocalize electrons while in contact with
9 counter materials. Therefore, when the brush bundle comes into contact with the counter
10 material during the sliding process, an equal quantity of positive and negative charges are
11 symmetrically formed on the surfaces of the brush and counter material, respectively. Once the
12 MB-TENG starts to slide along the surface of the counter materials, the mobile brushes stay
13 above the non-charged region of the stationary counter material. The charge unbalance between
14 the surfaces of the brush and counter material then induces the electrical potential difference
15 between two electrodes, leading to the electron flow through the external circuit. As the MB-
16 TENG continues to slide, the brushes are placed on the charged region of the counter material,
17 and thereby the charge balancing drives the reverse electron flow, which causes an alternating
18 current to the external circuit (Figure S3). The representative output current in contact with the
19 polytetrafluoroethylene (PTFE) film shows $0.88 \mu\text{A}$ at the sliding speed of 3 m s^{-1} (Figure 1g).
20 The values of the electrically induced charge were calculated from the integration of two single
21 output current peaks, $Q_{\text{polarization}} = 17.29 \text{ nC}$ and $Q_{\text{depolarization}} = 18.45 \text{ nC}$, indicating that
22 negligible charge loss was observed during sliding. In the second working mode established by
23 the vertical contact-separation process, the compressive and release stimuli drive the periodic
24 electrification between the nylon microbrush and the counter material, resulting in the
25 alternating current (Figure S3). As a representative case, an output current of $0.12 \mu\text{A}$ was
26 obtained in the contact-separation mode at a compressive load of 70 N (Figure 1h). Indeed, we

1 observed similar polarization and depolarization charges, implying that the electrostatic
2 contribution of the measurement apparatus is negligible.

3 The main characteristic features in the sliding mode were thoroughly evaluated for the
4 MB-TENG module using a custom measurement setup (**Figure 2a**), in which the initial contact
5 area was precisely controlled to maintain the discontinuous contact between microfibers by
6 adjusting the distance between the MB-TENG and the counter materials (Figure 2b). The MB-
7 TENG was mounted on a fixed support, and the counter material was moved toward the in-
8 plane direction of the MB-TENG using a programmed linear motor, as presented by a real
9 photograph in Figure 2c. In this measurement configuration, we analyzed the electric outputs
10 using an oscilloscope equipped with a current preamplifier. The output voltages generated from
11 the MB-TENG module could be tuned by varying parameters such as initial contact area, sliding
12 speed, and the number of arrays and counter materials. For example, in the case of PTFE counter
13 materials, the results in Figure 2d show an increased output voltage up to the highest value of
14 ~ 17 V by increasing the contact area (i.e., load-distance) to an enlarged contact area of ~ 240
15 mm^2 at a constant sliding speed of 3 m s^{-1} . As reported previously, a slow sliding speed (0.5 m
16 s^{-1}) resulted in the lowest output potential of ~ 3 V under identical experimental conditions, as
17 an increase in sliding speed gradually boosted output potential (Figure 2e). In this experiment,
18 we found that the electric potential distribution of the MB-TENG was highly dependent on in-
19 plane displacement. The electrification within the physically defined interfaces was computed
20 and visualized as a function of displacement (D) using COMSOL Multiphysics as presented in
21 Figure 2f, in which the output voltage reached a maximum of ~ 4.3 kV for the displacement of
22 25 mm. Although the electrical potentials from the simulation were much greater than the
23 results obtained from the experiments, which may be likely due to the non-ideal open-circuit
24 condition of the measured output voltages,^[43] it is worth noting that an in-plane shift of the
25 microbundle yields the electrical polarization between the MB-TENG and the counter materials.
26 To verify the internal resistance of the MB-TENG, the output voltage and current in contact

1 with the PTFE counter material were investigated using different resistors, as shown in Figure
2 2g, where the maximum power of $7.96 \mu\text{W}$ was collected at a load resistance of $0.3 \text{ G}\Omega$ (Figure
3 S4). The output of a single-bundle MB-TENG module represents $\sim 0.3 \text{ V}$ at the initial contact
4 area of $\sim 240 \text{ mm}^2$ at the sliding speed of 3 m s^{-1} , which was a ~ 55 -fold improved result achieved
5 by increasing the number of microfiber bundles up to 36 pillar arrays of bundles, as shown in
6 Figure 2h. This is probably due to the intercalated interaction of each microbrush bundle acting
7 as a charging pump, and thus the output voltage was a constructively integral state when the
8 polarization and depolarization processes from the microfiber bundles were synchronized in the
9 sliding mode. In addition, the long-term stability was rigorously tested for the MB-TENG under
10 the repetitive sliding mode ($D = 25 \text{ mm}$). Even after three thousand cycles as shown in Figure
11 2i, the voltage levels appeared to be stable compared with its initial stage, demonstrating the
12 outstanding durability and robustness of the MB-TENG module. Moreover, by changing the
13 counter materials, the output current and voltage generated in the sliding mode were tested to
14 confirm the versatility of the MB-TENG, as represented in Figure 2j (see also Figure S5). The
15 resulting levels of the output current and voltage on each sample matched well with the trend
16 of the triboelectric series,^[44,45] and the surface roughness of counter materials was a critical
17 factor. Sufficient electric output voltage levels from contact with a series of counter materials
18 suggest that our MB-TENG can be applied to portable electronics to conveniently generate
19 triboelectricity, facilitating the friction of rough or apparently smooth surfaces.

20 Interestingly, we observed that electrical outputs are almost identical when the counter
21 probe was connected with even ground rather than counter electrodes (Figure S6), provisioning
22 the design flexibility as well as an expanded application of our TENG device. We postulated
23 that a new material design concept featuring a single-electrode mode of the MB-TENG would
24 allow the use of loose fabric counter materials for the limited integration of the electrode, as
25 schematically outlined in Figure 2k. When the MB-TENG was slid on four different fabrics –
26 wool, silk, polyvinyl acetate (PVAc), and cotton fibers – the measured results of the output

1 voltage and current at constant sliding speed increased with the increasing electron affinity of
2 the counter materials in the triboelectric table, reaching 8.0 V and 0.8 μ A for cotton (Figure 2l-
3 m and Figure S7b). Notably, the sidewalls (i.e., ridges and grooves) from the regularly aligned
4 fabrics enhanced the contact area in the sliding mode for effective triboelectrification by
5 interlocking the lateral side of the microfibers in the slightly penetrated configuration of the
6 MB-TENG (the inset in Figure 2k). As a simple demonstration, when the relative humidity was
7 controlled using an ultrasonic humidifier, the electrical output was obviously degraded by
8 increasing the relative humidity from 15% to 55%. It was clear that water layer formation
9 usually results in the depletion of induced charge on the surface of the triboelectric materials
10 (Figure 2n).^[46]

11 The next strategy for triboelectrification was demonstrated by applying the periodic
12 compressive load to the MB-TENG module in a vertical contact-separation mode (**Figure 3a**).
13 A set of spring spacers at the corners were installed between the MB-TENG and the counter
14 materials to retain a gap of ~10 mm when released from the engaged compressive load. The
15 assembled MB-TENG was placed on the digital scale, and the strain of the microfiber bundles
16 was precisely modulated using a programmed linear motor. Similar to the study for a sliding
17 mode, several measurement parameters, including strain and load frequency, altered the
18 electrical outputs by the motions of the compress-release. Figure 3b displays a set of typical
19 output voltage levels in contact separation with the PTFE film linearly increased, with the strain
20 increasing from -10% to -50% and reaching up to 1.2 V at -50% strain as the contact area was
21 enlarged by the external loads. Separately, the output voltages with different frequencies at the
22 applied load of -50 % strain were measured in Figure 3c. The output voltage of 0.3 V was
23 achieved at an initial low frequency of 3 Hz, and the values were increased to 0.4, 0.5, and 1.2
24 V in the frequency ranges of 4.5, 6, and 7.5 Hz, respectively. In the triboelectrification process,
25 this frequency dependence is not surprising, as the response time of the electron flow through
26 the external circuit may not be sufficient under certain conditions.^[7] Thus, at a higher frequency

1 load, the electron flow has a shorter time to be relaxed through the external circuit compared
2 with that at the lower frequency load, and thereby this increases the output voltage and current
3 signals. The electric potential distribution in the repeated contact-separation mode of the MB-
4 TENG in the open-circuit configuration was simulated and visualized according to the contact
5 length (L) using COMSOL, as shown in Figure 3d. As presented, the total surface charges are
6 balanced when two surfaces are in contact with each other. Once two surfaces are then separated,
7 the electric potential can be generated in the vicinity of each surface. Reaching $L = 10$ mm, the
8 open-circuit voltage can be maximized up to ~ 300 V, and the maximum power experimentally
9 generated was 46.72 nW at the load of $100\text{ M}\Omega$ (Figure S8). Within the given conditions (i.e.,
10 7.5 Hz, 5 mm), the trends of the electrical potentials extracted from the COMSOL simulation
11 were in good agreement with the measured values from the experiments (Figure 3e-f). Beyond
12 the friction of the machine-interactive motion, the feasibility of triboelectrification by human
13 motions can expand the viable applications of the MB-TENGs. Thus, we performed the hand-
14 tapping test on the MB-TENG equipped with the same spring-spacer configuration as
15 demonstrated earlier (Figure S9a). The electrical outputs driven by manual tapping motion were
16 found to be much larger than that induced by a programmed linear motor (i.e., 31.2 V in Figure
17 S9b-c). Moreover, the TENG that consisted of a planar nylon film without the surface-relief
18 structure exhibited a relatively low output performance of 7.7 V, as presented in Figure 3g and
19 Figure S10. As previously reported, when the same material has characteristic features other
20 than a flat shape on the friction surface, the contact area increases, which improves the output
21 performance.^[14,47,48] These results imply that our MB-TENG manifests a notable human-
22 interactive energy harvesting device, which represents the maximum output power of $26\text{ }\mu\text{W}$ at
23 $100\text{ M}\Omega$ (Figure S9d-e). More importantly, this trait is particularly advantageous given that the
24 brush has long been a portable common tool used for cleaning, grooming, make-up, painting,
25 etc. In this context, a variety of other counter materials were used to obtain more information,
26 which yielded similar or improved electric output characteristics through a hand-tapping

1 operation (Figure S11). In addition, the triboelectric performance was further enhanced by
2 covering the top surface of the frame structure (i.e., holder, PLA) with a triboelectrically
3 negative material (polyvinyl chloride, PVC film) to collect more electrons from the touching
4 hand, as described in Figure S12a. Surprisingly, as shown in Figure 3h, the output voltage and
5 current in the contact with the PTFE film using the MB-TENG module were remarkably
6 boosted up to 1.6 kV and 200 μ A, respectively, in which the maximum output power was 76.7
7 mW at load resistance of 100 $M\Omega$, as summarized in Figure 3i. This strategy revealed a similar
8 trend by applying other commonly used counter materials such as silicone rubber and polyimide
9 film (Figure 3j and Figure S12b).

10 **Figure 4a** illustrates a practical demonstration of the MB-TENG for extending our
11 scheme to a human-interactive device module.^[49] The typical MB-TENG modules
12 characterized earlier were readily installed at different locations of the human body – including
13 on the hair, clothes, and shoes – to harvest the friction energy driven by arbitrary human motions.
14 In these demonstrations, the electrical output was maximized by selecting and fitting the
15 optimal working mode of the MB-TENG modules as schematically guided in Figure 4b; hair
16 (in-plane sliding), clothes (in-plane sliding), and shoes (vertical contact separation). Such
17 systems provide an efficient platform for portable or wearable energy harvesting where
18 reversible structural flexibility of the TENGs is essential to the operation. Electrical energy
19 collected by the periodic motions either operated the LED arrays or was stored in a lithium-ion
20 battery that powered commercial healthcare devices such as an electromyography (EMG)
21 module or smartwatch, as illustrated in Figure 4c. Figure 4e provides relevant data on the in-
22 plane sliding mode simply by combing long-hair in an axially aligned direction, similar to that
23 measured in the fabric cases (see Figure 2k). Hence, this unusual approach of the MB-TENG
24 successfully acquired an output voltage and current of up to ~150 V and 6 μ A, respectively
25 (Figure 4d), which yielded stable power to light up 100 LEDs (Figure 4e). Another form of the
26 MB-TENG was applied to sportswear (i.e., polyester material) through manual rubbing on the

1 woven fabric surface, providing electrical outputs of 70 V and 1 μ A (Figure 4f). Moreover, the
2 basic MB-TENG module could be integrated into the midsole of shoes and serve as a wearable
3 device for a self-charging power supply through body movement. The measured electric
4 potentials were ~50 V with a different frequency of 0.45 Hz and 1.7 Hz for walking and running,
5 respectively, as shown in Figure 4g. Overall, we successfully demonstrated the harvesting of
6 sufficient electrical energy by utilizing transformable MB-TENG modules at several body
7 locations. To ensure the storage of the instantaneous triboelectrification, commercial capacitors
8 were used, in which the separate levels from each charging capacitive voltage dramatically
9 increased up to ~6 V within 40 s (1 μ F) and ~5 V within 250 s (10 μ F), as summarized in Figure
10 4h; the charged energy was gradually discharged during 200 s due to the inherent characteristics
11 of the capacitor (Figure S13a). As a means to reliably store harvestable energy, the lithium-ion
12 battery (25 mAh) was used and the circuitry was connected with the MB-TENG in the sliding
13 mode, transforming the mechanical movement (i.e., a linear motor for 28 h, Figure S13b) to a
14 power source for small-scale electronic devices. Although the characteristic output degraded
15 by ~2-3 V compared with the initially prepared TENG module (i.e., only degraded 10-15% in
16 performance) after the repeated friction for a long time (i.e., 28 hours), this experiment clearly
17 evidenced the robustness of our MB-TENG device. Relatively, long periodic movement was
18 required at this stage, but the possible wasted mechanical energies could be collected from other
19 possible situations.^[15] Similarly, as a simple test for the power-supply capability of the wearable
20 electronic device, the four 10 F capacitors connected in serial (total capacity: 2.5 F) could be
21 charged to 3.7 V, and then used to drive a smartwatch (Figure S13c-d). Moreover, a
22 representative healthcare monitoring device (i.e., EMG module) was also operated using the
23 lithium-ion battery charged by the MB-TENG, as presented in Figure 4i. Here, the flexible
24 EMG module and epidermal electrodes were attached to the forearm, and the signals of the
25 hand gestures of an open palm and clenched fist were recorded and directly transferred to a
26 smartphone (inset in Figure 4i). Our design approach to the sustainable TENG module to

1 illuminate the energy supply implies potential applications in the future for a variety of research
2 fields such as military, robotics, entertainment, and smart healthcare.^[50,51]

3 In summary, we have reported a simple yet efficient microbrush-based device that
4 exhibits outstanding performance as a robust and reliable TENG, with excellent electrical
5 potential as high as ~20 V and high cyclability of ~3000 in both in-plane sliding and vertical
6 contact-separation working modes. The intuitively succinct design and flexibility of the
7 presented MB-TENG enable the adoption of various forms of counter materials, including
8 polymeric, metallic, and woven fabric materials. Moreover, as the brush has long served as a
9 common handheld tool, the manufactured MB-TENG can be further applied to human-
10 interactive triboelectrification to convert the energy of human motions. Importantly,
11 commercially available small electronic devices such as smartwatches and flexible EMG
12 modules can be successfully powered by harvesting and storing electricity from body motions,
13 allowing the MB-TENG to be used as a universal wearable energy source. We envision that the
14 presented MB-TENGs have viable potential in wearable power sources and can serve not only
15 as a portable energy-harvesting device but also as a transducing medium to interpret signals of
16 human activity recognition and motion-sensing; indeed, a systematic study of the sensory
17 system in the microbrush structure is currently underway.

18

1 **Experimental Section**

2 *Fabrication of the microbrush TENG (MB-TENG).* The basic structure of the MB-TENG
3 comprises an array of nylon microfiber bundles and a 3D printed PLA supporter. To construct
4 the brush structure, the nylon microfiber bundles were extracted from a commercial toothbrush,
5 and were then planted into the PLA supporter (20 mm x 25 mm), in which the array of aperture
6 with a diameter of ~2 mm and interspace of ~1.14 mm was three-dimensionally printed over
7 the surface. The microfiber bundles were fastened to the array of aperture on the supporter with
8 silver paste. The full device employed a 6x6 array of microfiber bundles in a given area.

9 *Characterization of the electric output performance.* A custom-built mechanical test system
10 was constructed with a programmed linear motor (LS Mechapion, APMC-FAL01AM8K) and
11 a sensitive scale. The MB-TENG was mounted on the linear motor to examine the triboelectric
12 performance against the sliding and compressing load. An oscilloscope (Agilent DSO-X-
13 2014A) equipped with a preamplifier (SRS SR-570) was used for low-noise voltage and current
14 measurements while monitoring both the sliding and compressing load to the MB-TENG.

15 *Characterizations.* The morphologies of bundled and individual fibers composing the MB-
16 TENG were observed by a scanning electron microscope (ZEISS SUPRA25, 5-10 kV), optical
17 microscope (Olympus BX51), and digital microscope camera (Hayear, HY-2307). The stress-
18 strain response of fiber bundles was measured using a CT texture analyzer (AMETEK
19 Brookfield).

20 *Numerical simulation via COMSOL.* To compute the triboelectrification of the microbrush, 2D
21 models of an array of a brush with dimensions of 2 mm diameter and 10 mm length were built
22 in COMSOL Multiphysics, matched with the actual dimensions of the MB-TENG. Vertical
23 displacements from 0 mm to 10 mm and horizontal displacements from 0 mm to 25 mm were
24 introduced to the MB-TENG. For potential distribution simulations with an open-circuit
25 condition, the surface charges of 2 nC and 20 nC were set for nylon fibers at the in-plane sliding
26 and vertical contact-separation modes, respectively. The total charge on the PTFE counter

1 material was set to -2 nC and -20 nC at the in-plane sliding and vertical contact-separation
2 modes, respectively.

3
4 **Supporting Information**
5 Supporting Information is available from the Wiley Online Library or from the author.
6

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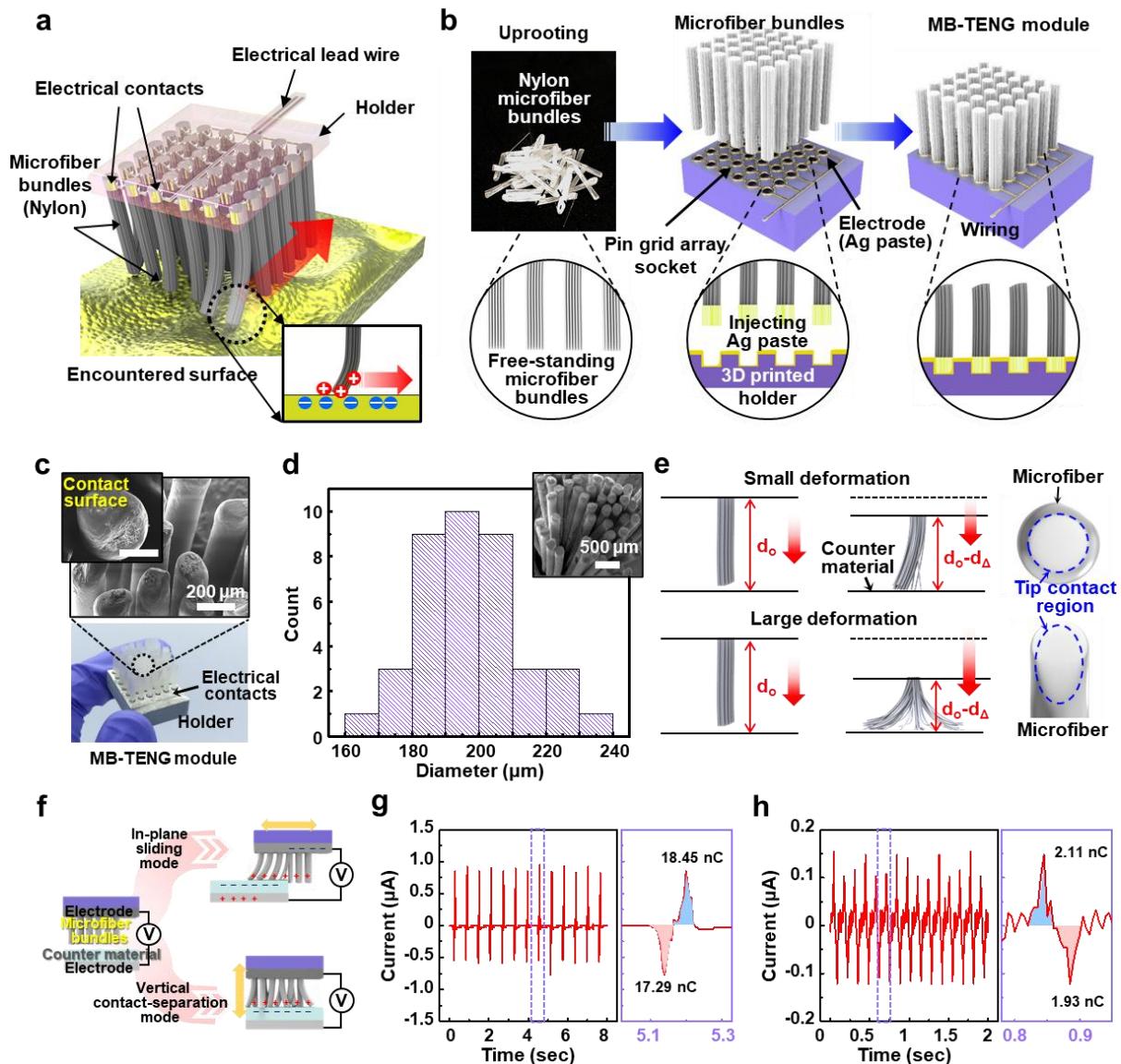
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1
2 **Figure 1 | Concept and design of the MB-TENG-based energy harvesting system. a)**
3 Schematic illustration of the MB-TENG component generating triboelectrification. **b)** The
4 fabrication process to produce the MB-TENG utilizing chopped fiber-bundles and a pin grid
5 array socket connected with electrically conductive Ag paste; corresponding cross-sectional
6 images are presented for each manufacturing process (bottom). **c)** Scanning electron
7 microscopy image of the surface morphology in the microfibers (inset: magnified rough surface,
8 scalebar = 100 μm) as a contact area for triboelectrification, and photograph of the fully
9 integrated MB-TENG module (bottom). **d)** A histogram representing the diameter distribution
10 of the bundled microbrushes in the TENG module. **e)** A deflected single microfiber with tip
11 contact at the axis-dependent deformation; the contact region can be defined as an oval shape
12 by the increased contact area. **f)** The MB-TENG integrated with the counter material to enable
13 two different working modes. Representative output current (left) and transferred charge
14 calculated from a current peak (right), where measured in sliding mode, **g)** and in vertical
15 contact-separation mode, **h)**, respectively.

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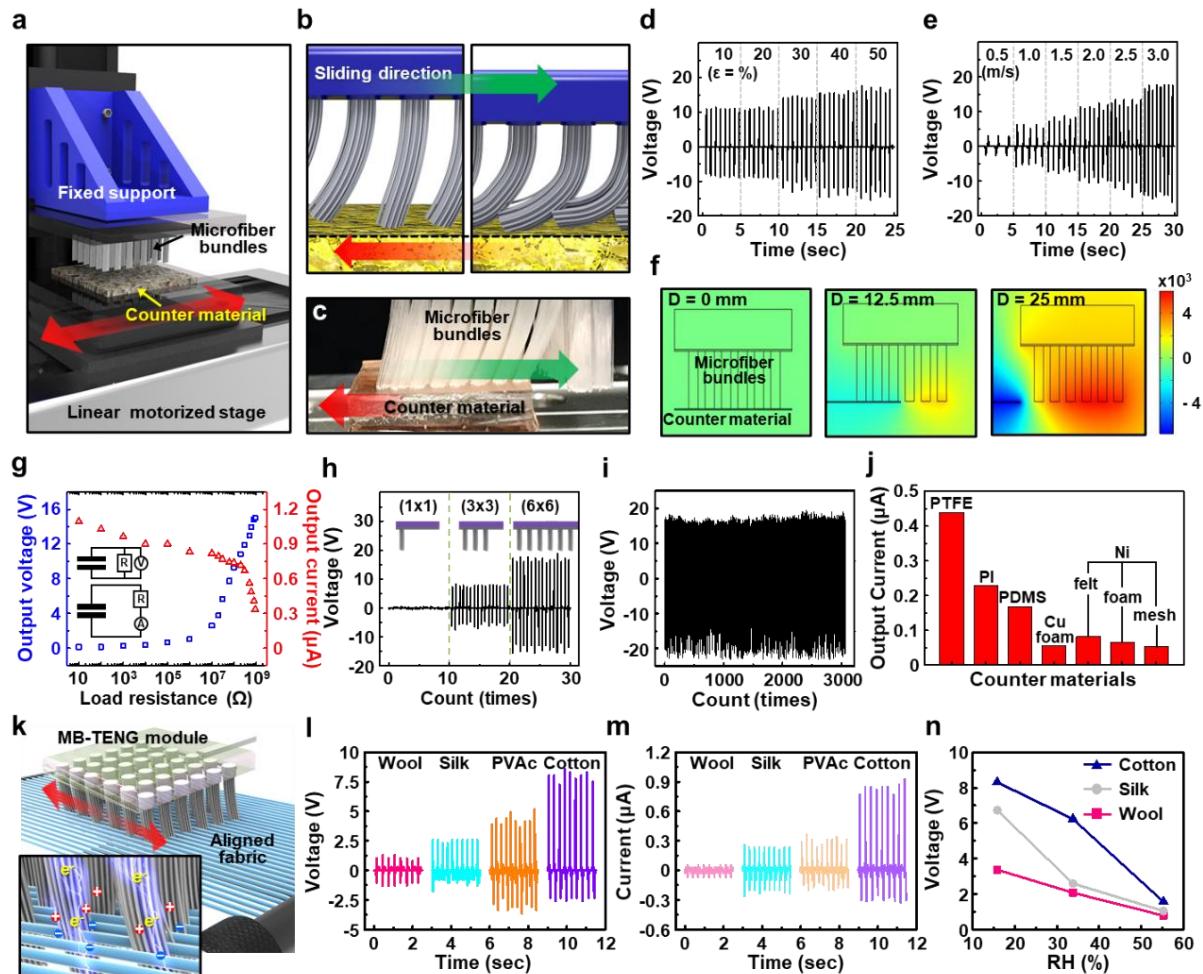
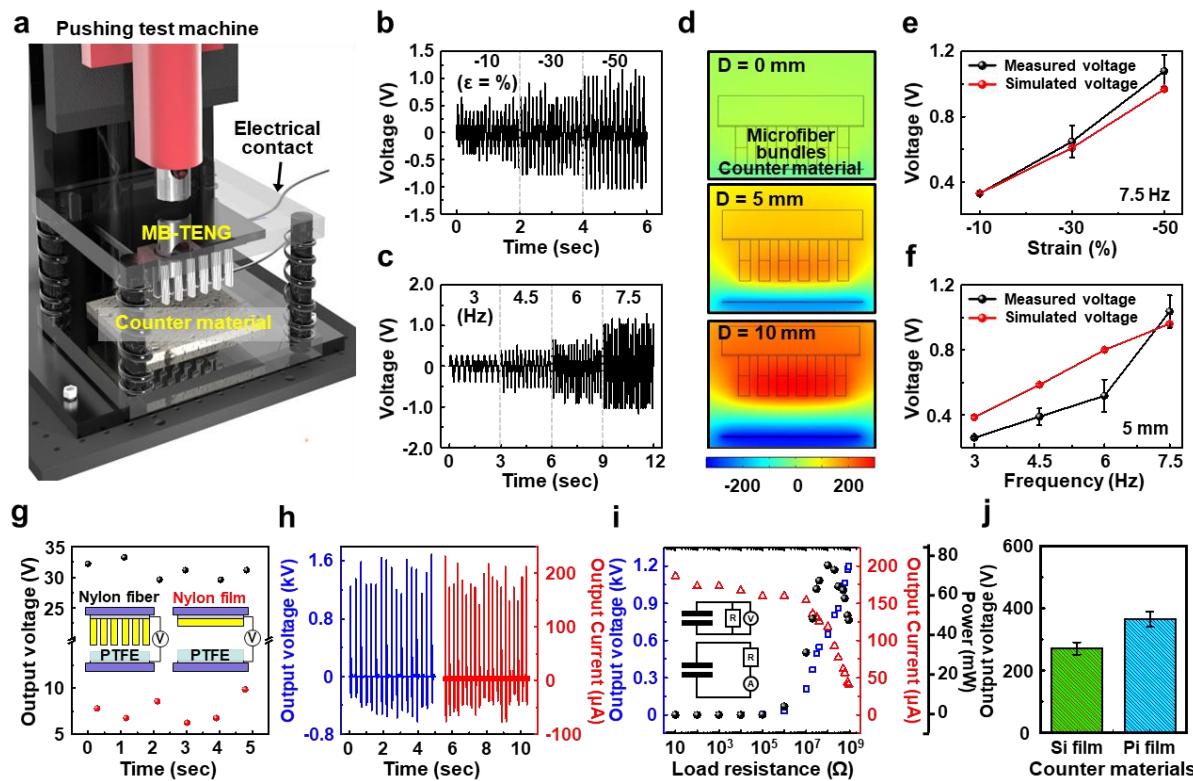


Figure 2 | Characterization of the sliding mode for the MB-TENG. **a)** Schematic illustration of the measurement setup. **b)** Enlarged side view examples of the sliding mode MB-TENG when contacted with a counter material, representing the capability to slide under the tunable vertical distances. **c)** Real digital image of the MB-TENG equipped in the measurement setup. **d-e)** Output voltage of the typical MB-TENG module under different strains from 10% to 50 % at a sliding speed of 3 m s^{-1} and 50% of strain at different sliding speeds. **f)** COMSOL simulation results of the MB-TENG at different separation distances. **g)** Output characteristics depending on the load resistance. **h)** Each representative output voltage performance with the increase in the number of MBs. **i)** Long-term stability of output voltage during 3000 cyclic motions. **j)** Output current of the MB-TENGs matched with various types of counter materials. **k)** Schematic illustration of the sliding MB-TENG mode on regularly aligned fabrics in the single-electrode mode, enhancing the contact area by interlocking the lateral side of the microfibers (inset). **l-m)** Output voltage and current measured from the sliding MB-TENG on various aligned fabrics. **n)** Measured peak voltage values under different relative humidity from 15% to 55%.

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2 **Figure 3 | Characterization of the contact-separation mode for the MB-TENG.** a)
3 Schematic illustration of the measurement setup for the contact-separation mode MB-TENG,
4 consisting of a programmed linear motor and the assembled MB-TENG. b-c) Output voltage
5 of the MB-TENG under different strains at a 7.5 Hz of frequency and under 50% of the strain
6 at different load frequencies. d) COMSOL simulation of the potential distribution in the MB-
7 TENG for the different contact lengths between MBs and the counter material. e-f) Computed
8 and experimental results of the potential differences generated by the MB-TENG under the
9 same conditions as b) and c). g) Comparison of output voltages of the fiber and film structure
10 TENGs (black dots: nylon fiber, red dots: nylon film). h) The output voltage and current of the
11 MB-TENG with PVC film on the top surface of the frame structure. i) The output voltage and
12 current of the MB-TENG measured at different external load resistances varying from 10 Ω to
13 910 MΩ and calculated output power (inset shows a circuit diagram of the measurement
14 system). j) Output voltages of MB-TENGs using other counter materials for silicone and
15 polyimide film.

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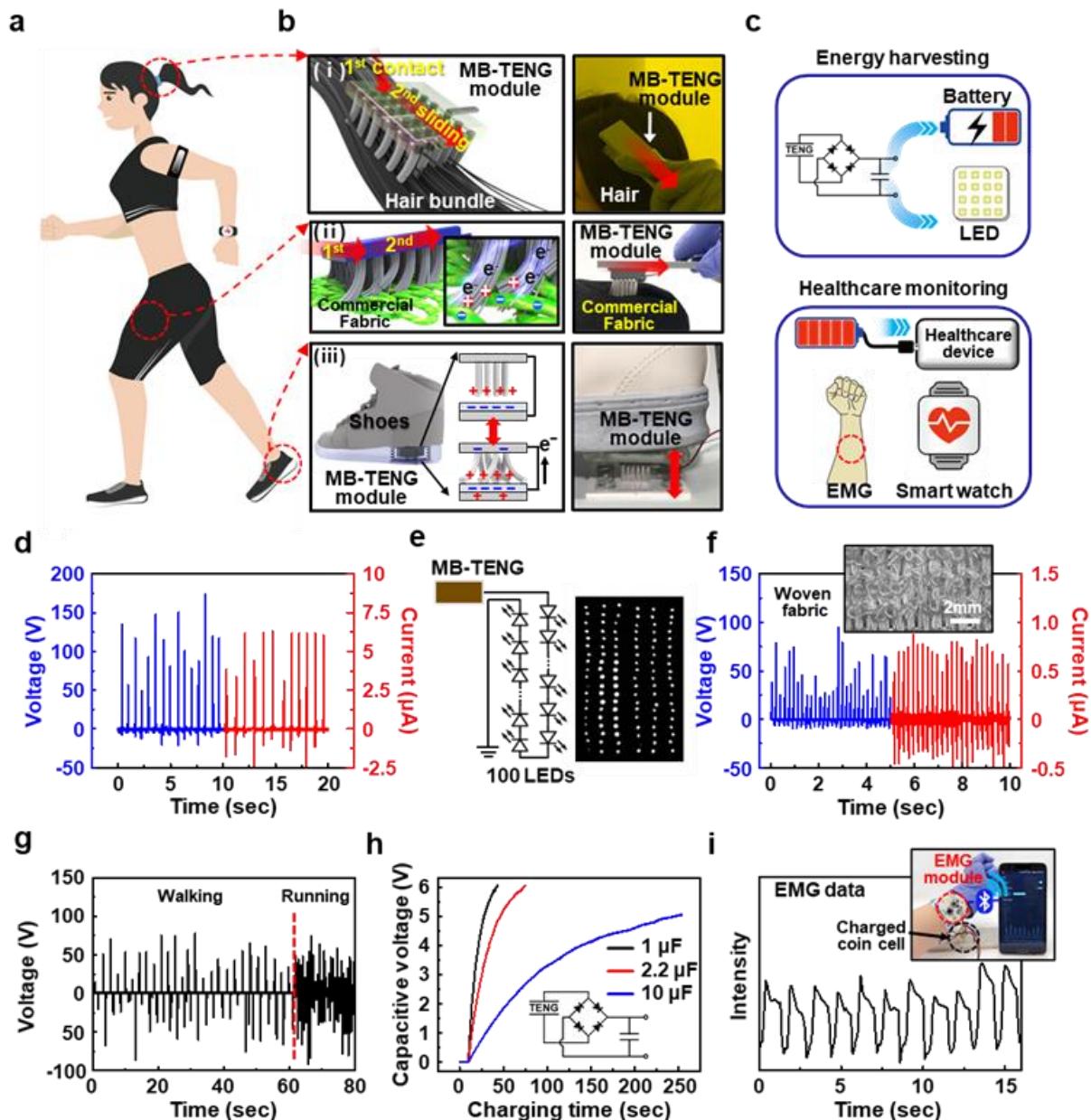


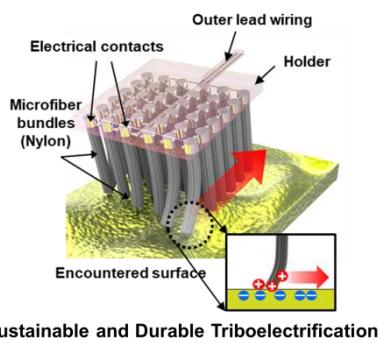
Figure 4 | a) Conceptual illustration of the practical demonstration of the MB-TENGs as a human-interactive device. **b)** Schemes and matched photographs of the MB-TENGs to apply the optimal working modes as (i) combing hair, (ii) sweeping fabrics, and (iii) fixed under a shoe. **c)** Utilization of the harvested energy from the MB-TENGs for lighting up LED arrays, charging a lithium-ion battery, or healthcare monitoring. **d)** Output voltage and current harvested from the case (i). **e)** Circuit diagram for LED arrays connected in a series and actual powering demonstration, lighting up to 100 LEDs. **f)** Output voltage and current measured from the motion of case (ii); the inset shows a photograph of the fabric counter materials. **g)** Output voltage during the process of walking and running with case (iii). **h)** Capacitive charging behavior with hand tapping MB-TENGs; the inset shows a circuit diagram consisting of an MB-TENG and a capacitor. **i)** The measured EMG signals by the continuous hand gestures; the inset shows a photograph of the EMG module and wireless system with a smartphone.

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1 Our microbrush-faced triboelectric nanogenerator featuring high robustness, outstanding
2 sustainability, and excellent durability displays remarkable capability for mechanical energy
3 harvesting in separate working modes. Human being interactive triboelectrification with our
4 device reveals outstanding power generation and durability, suggesting that our device can
5 serve as a power source of various handheld electronics.

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7 Jeonghwa Jeong, Sangheon Jeon, Xiaoting Ma, Young Woo Kwon, Dong-Myeong Shin* and
8 Suck Won Hong*
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10 **Sustainable and Flexible Microbrush-Faced Triboelectric Generator for**
11 **Portable/Wearable Applications**



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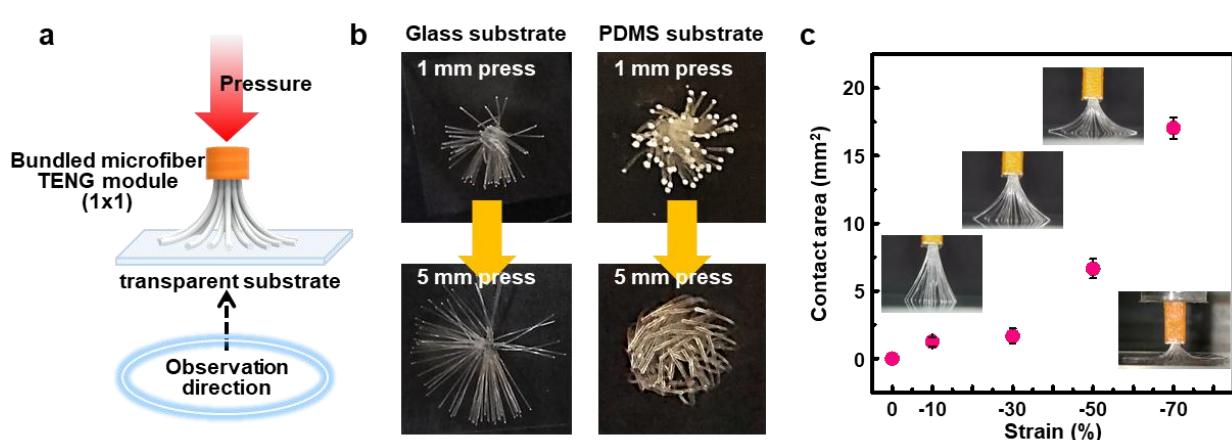
1 Copyright WILEY-VCH Verlag GmbH & Co. KGaA, 69469 Weinheim, Germany, 2018.

2
3 **Supporting Information**
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6 **Sustainable and Flexible Microbrush-Faced Triboelectric Generator for**
7 **Portable/Wearable Applications**
8

9 *Jeonghwa Jeong, Sangheon Jeon, Xiaoting Ma, Young Woo Kwon, Dong-Myeong Shin* and*
10 *Suck Won Hong**

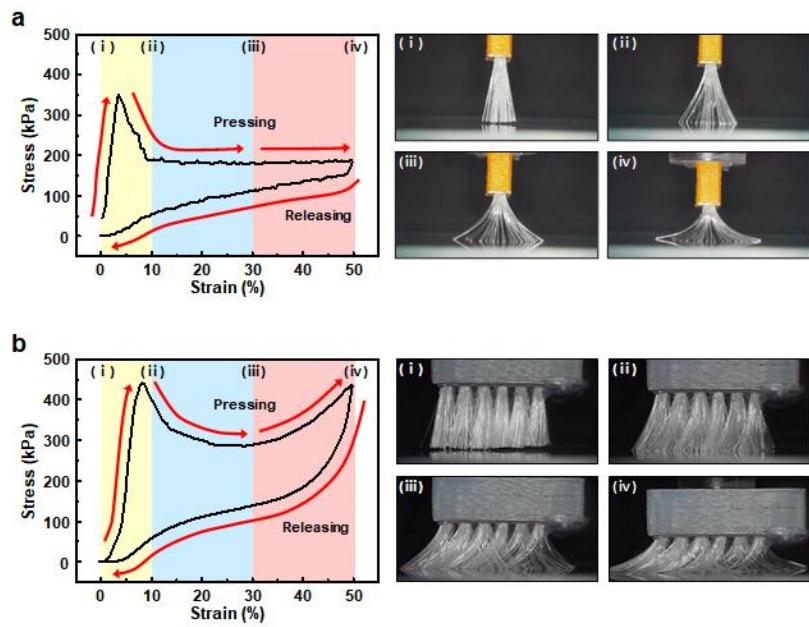
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12 **Supplementary Figures**
13



15 **Figure S1.** Visualization of the estimated contact area for a single microfiber bundle with
16 respect to various axial deformation. a) A schematic drawing on the direct observation for the
17 contact area of a single microfiber bundle, monitored through a transparent substrate (i.e.,
18 smooth glass and tacky PDMS). b) The captured images of the deformed microfibers at 1 mm
19 and 5 mm on each contacted substrate. c) The expanded contact area depending on the fiber-
20 spreading at each axial deformation on a glass substrate.

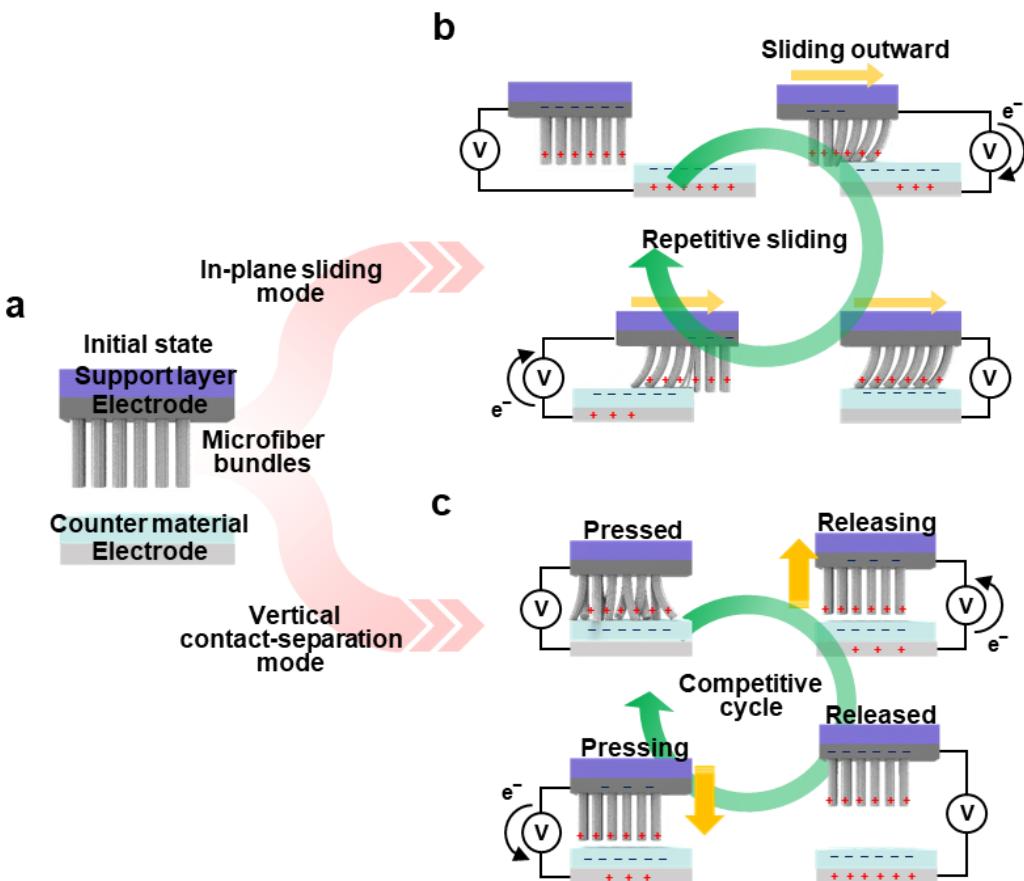
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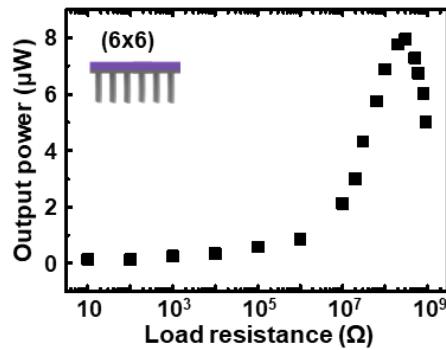
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2 **Figure S2.** Direct comparison of plastic deformation (i.e., press-to-release) for each TENG
 3 module. a-b) The stress-strain graph of a single microfiber bundle and 6×6 pin-array module,
 4 respectively. Increased strain from (i) to (ii) regions in the graph reflects the elastic modulus of
 5 each TENG module based on the microfiber bundles. For the 6×6 pin-array module in b, the
 6 mechanical property at the region (iv) represents the increased stress by the interrupted spread
 7 of the microfiber-pin array.



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2 **Figure S3.** a) The originally designed structure of the MB-TENG. b) The series of schematic
 3 illustrations of the related charge transfer mechanism in the sliding mode of TENG when two
 4 different substances are contacted under the engaged condition. In the process, the periodic
 5 change of the potential difference can be induced on each step between the microbrush and
 6 counter materials through the cyclic frictions. When the two materials are facilitated by sliding
 7 friction, the negative charges from the counter materials generate the electron flows to the
 8 external electrode under a positive charges on the surface of the microbrush. In the following,
 9 the charged state is transformed to the initial state when the two surfaces are separated (i.e.,
 10 released state). Depending on the in-plane sliding distance changes, the triboelectric effect
 11 induced by the potential difference (i.e., opposite triboelectric charges on the surface) can be
 12 increased. c) The series of schematic illustrations of the related charge transfer mechanism in
 13 the contact-separation mode of TENG. Likewise the former case, the potential difference occurs
 14 when the two surfaces are repetitively agitated between the frictional charged surfaces; the
 15 electrons by the triboelectrification can be driven to flow through the external electrode to the
 16 opposite direction. In the two different modes, the shortening of the vertical distance between
 17 the tribologically active material and the enlarging of the frictional area of the microfiber can
 18 lead to a larger electrical potential.

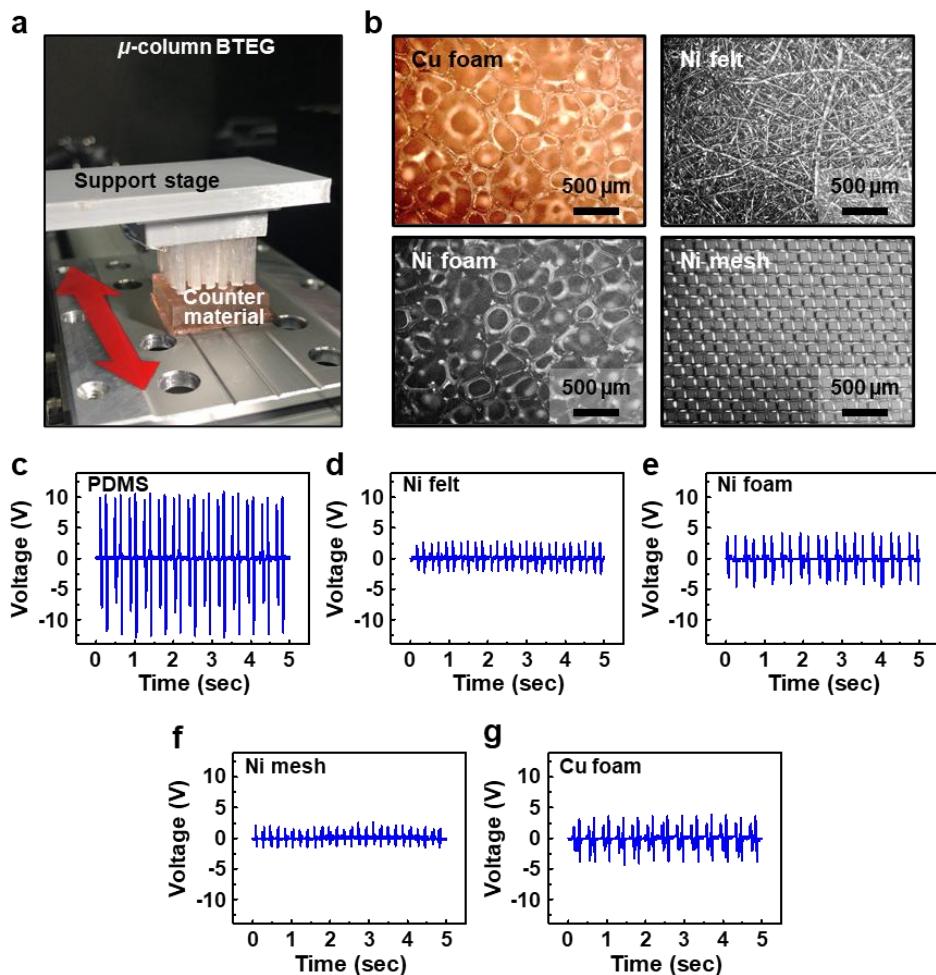


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2 **Figure S4.** Output power according to the load resistance in the sliding mode (6×6 module)
3 with PTFE counter material, representing the maximum power output of 7.96 μW at load
4 resistance of $3 \times 10^8 \Omega$.

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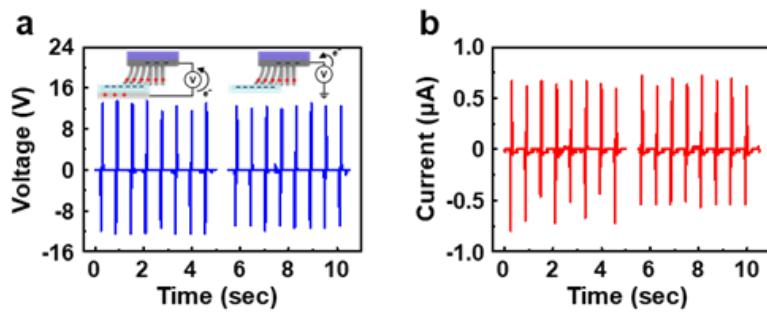
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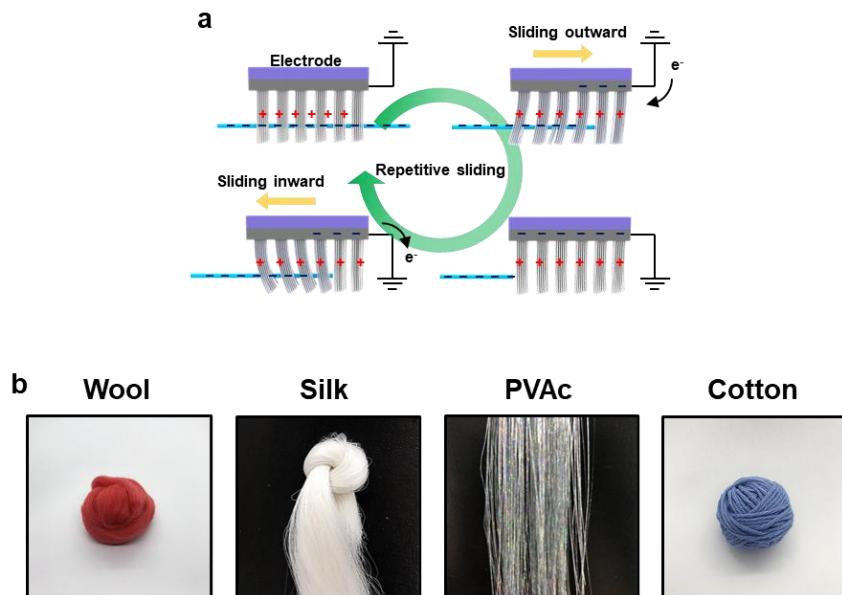
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3 **Figure S5.** a) Actual operating image during the sliding mode test using a linear motor; the
 4 MB-TENG module is fixed to a separate support on the top of the linear motor, and the counter
 5 material is attached to the lower stage of the linear motor to move back and forth. b) Microscopic photographs of counter materials with various shapes (i.e., Cu foam, Ni felt, Ni
 6 foam, and Ni mesh) used in the test. c-g) Representative output voltages from the sliding mode
 7 test with the MB-TENG (i.e., 6x6 module). The levels of the output voltage present in the order
 8 of PDMS, Ni foam, and Cu foam, which is in good agreement with the trend of the triboelectric
 9 series from the nylon microfiber.

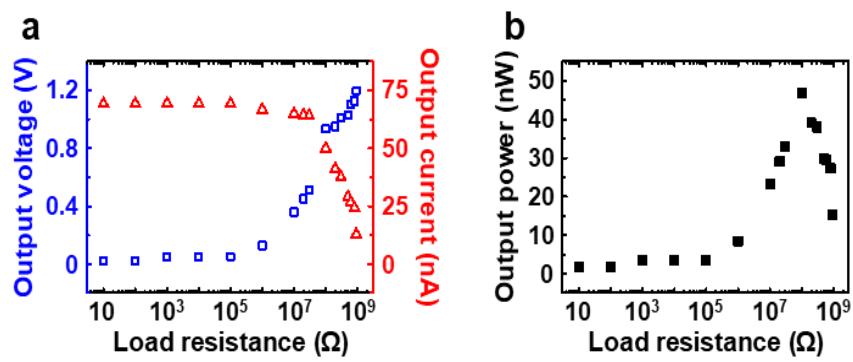
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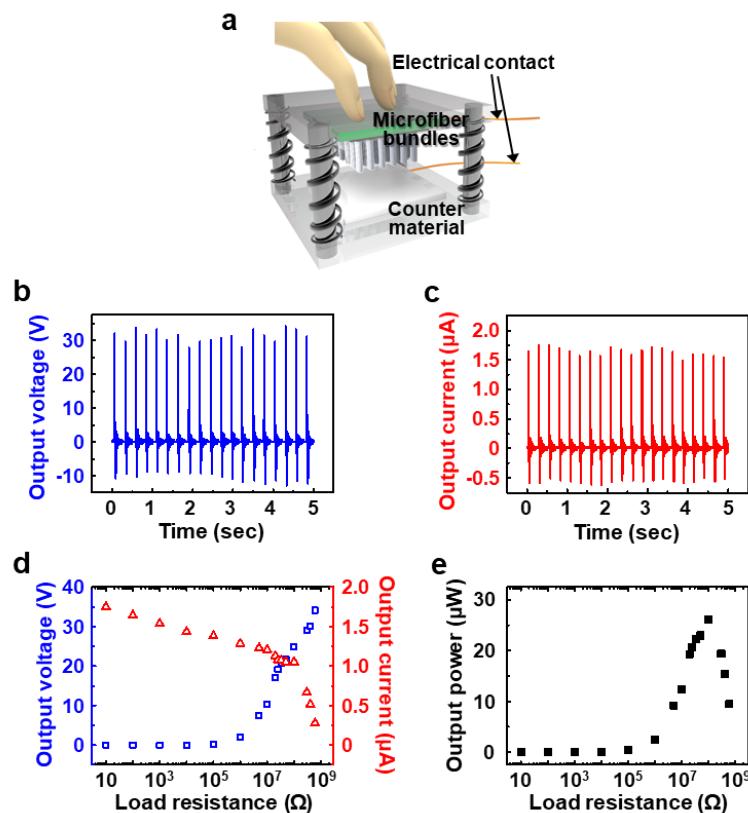
1 **Figure S6.** The representative values of the output voltage and current, separately measured in
 2 a) sliding and b) single-electrode mode using a linear motor.
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5 **Figure S7.** a) The series of schematic illustrations of the related charge transfer mechanism in
 6 the single-electrode mode of the TENG. When the MB-TENG is in complete contact with the
 7 aligned fibers (i.e., counter material), electrons are injected from the nylon microbrushes to the
 8 fibers. As the positively charged nylon microbrushes separate from the negatively charged
 9 aligned fibers, a potential difference can be created between them. This potential difference
 10 induces the electrons to flow from the ground to the electrode until the microbrushes are
 11 completely separated to maintain potential balance. By the nylon microbrushes coming into
 12 contact again with aligned fibers, the positive charge induced in the nylon microbrushes
 13 decreases. In the following, the accumulated electrons can flow from the electrode to the ground
 14 until the electrostatic equilibrium of the two materials is reestablished. b) Photographs of four
 15 different fibers (i.e., wool, silk, PVAc, and cotton) used in the regularly aligned fabrics for the
 16 horizontal sliding mode.
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2 **Figure S8.** a) The output voltage and current, and b) the output power (i.e., 46.72 nW at 108
3 Ω) according to the load resistance in the contact-separation mode of the MB-TENG on the
4 PTFE material using a linear motor.
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2 **Figure S9.** a) A schematic illustration of the hand-tapping contact-separation test on the MB-
 3 TENG equipped with the spring-spacer configuration. b-c) The output voltage and current of
 4 the MB-TENG with PVC counter material. d) The output voltage and current, and e) the output
 5 power of the MB-TENG (i.e., 26.16 μ W at 100 $M\Omega$) measured at different external load
 6 resistances.

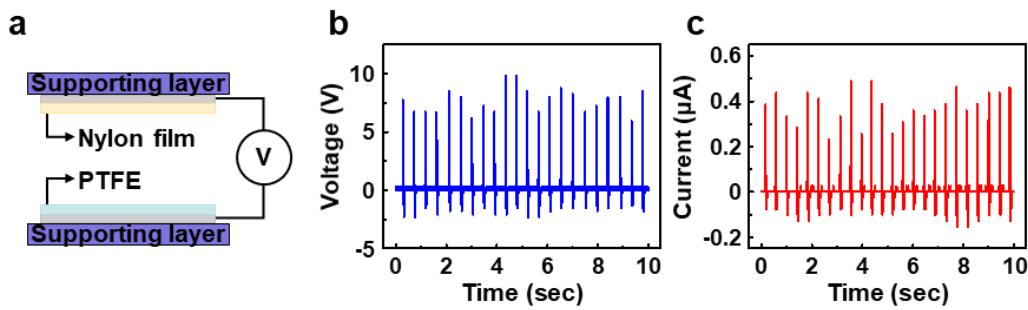
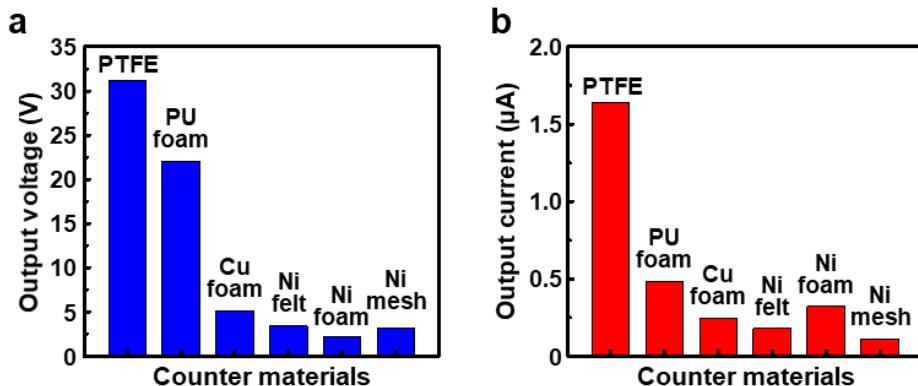


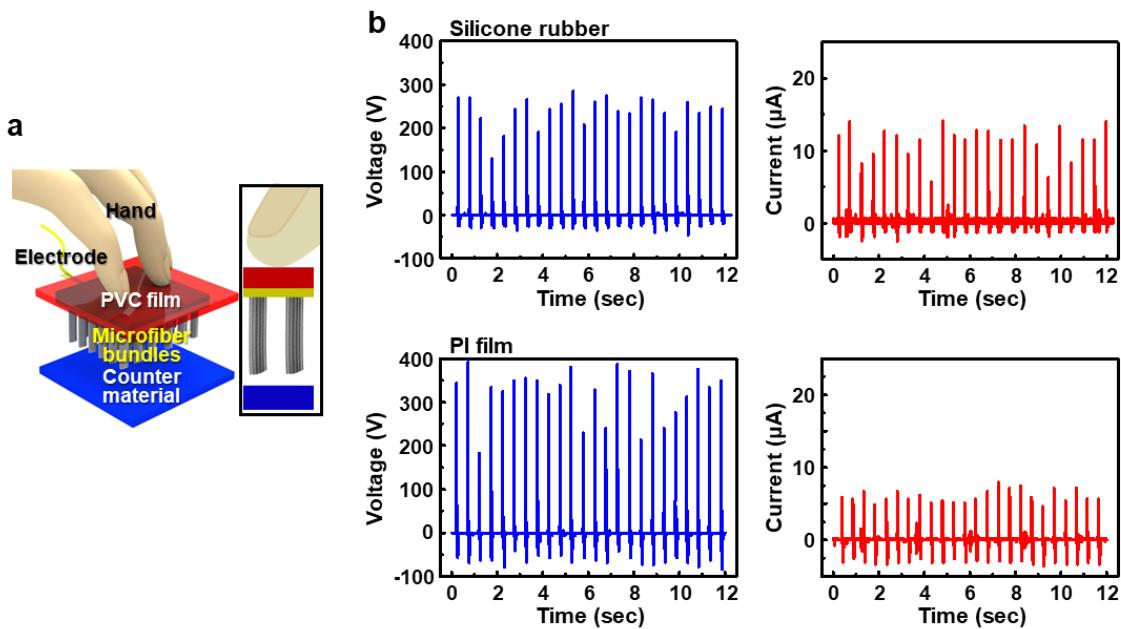
Figure S10. a) A schematic diagram of the hand-tapping test with nylon film. b-c) The output voltage and current in the contact-separation mode using nylon/PTFE films. The actual test area was the same as in the case of the MB-TENG-based module (i.e., $2 \times 2 \text{ cm}^2$). Compared with the microfiber test structure, lower output values were found due to the little effect of the increased contact surface area.

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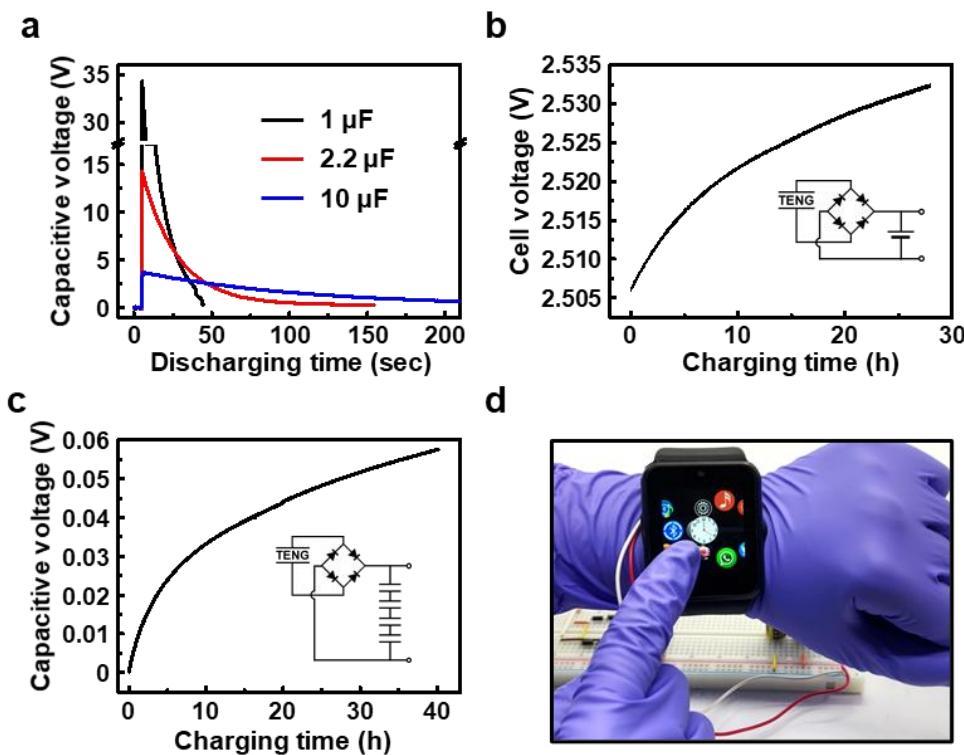


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Figure S11. The contact-separation test based on hand-tapping with a variety of counter materials. a-b) The levels of the output voltage and current; the results generally follow the triboelectric series, but in the structure of metal foam, the difference in output values occurred mainly due to the physical deformation of the counter materials by the repeated external force.



1 **Figure S12.** a) A schematic illustration of the PVC film layer-added MB-TENG by covering
2 the top surface of the frame structure in a hand-tapping operation. b) The levels of the output
3 voltage and current when testing silicone rubber and PI film. The performance of the MB-
4 TENG module can be further enhanced by the additional triboelectrification between the PVC
5 film and the human skin (i.e., hand) besides the frictional charge caused by the contact between
6 the microfiber bundles and the counter material in the contact-separation mode, resulting in
7 much higher levels of the output voltage and current as high as ~210, 350 V and 10, 5 μ A,
8 respectively.



1 **Figure S13.** a) An application of the MB-TENG module (see Figure S12) for the fast
2 charge/discharge process (i.e., 1 min) using 1, 2.2, and 10 μ F capacitors by the hand-tapping
3 operation in the contact-separation mode; for the 1 min charging process, capacitive voltages
4 reached up to \sim 34.34, 14.34, and 3.75 V. b) A demonstration of the charging performance for
5 a lithium-ion battery (coin cell, 3 V, 25 mAh) in sliding mode using a linear motor with a bridge-
6 rectifier circuit; the graph represents \sim 30 mV charging for 30 h. c) A demonstration of the
7 charging performance in sliding mode using a linear motor, in which commercial
8 supercapacitors (i.e., 3.0 V, 10 F) were utilized and serially connected to a bridge-rectifier
9 circuit (inset). d) Real photograph of operating a smartwatch by the charged 2-serial
10 supercapacitors. These demonstrations show that it takes a relatively long time to charge the
11 voltage levels required for a large-scale device operation, but the MB-TENG charged lithium-
12 ion battery and supercapacitors can be applicable to a small-scale device such as a wireless
13 EMG and smartwatch.