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An ultrathin stretchable triboelectric nanogenerator with coplanar electrode for energy harvesting and gesture sensing

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Stretchable electronics with excellent elastic characteristics are attracting extensive interest in the area of wearable devices and epidermal electronics. Here, we demonstrate an ultrathin stretchable triboelectric nanogenerator (s-TENG) with coplanar electrode for harvesting diverse biomechanical energies and acting as a self-powered gesture sensor. The s-TENG employs electrospun polyurethane nanofibers and conductive nanomaterials as the stretchable electrode. With the coplanar electrode configuration, the device can generate electricity from diverse working situations, such as folding/unfolding of the device and contact/separation with other objects. Facilitated by the increased contact area of nanostructure and paired electrodes design, the s-TENG can generate enhanced instantaneous peak power density of 316.4 μW cm^{-2} when working in the folding/unfolding situation. When in contact with other objects such as cotton cloth and human skin, peak voltages of 330 V and 286 V are obtained, respectively. Thanks to the ultrathin structure of the device, it can be conformally attached on skin and deforms as the body moves. By adjusting the dimensions of the device, the s-TENG can be used to detect human motion in different body parts, showing its great application prospects in sustainable wearable devices, self-powered electronic skins and smart wireless sensor networks.

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Introduction

Flexible and stretchable electronics that are able to deform as desired are drawing massive attention for their potential applications in the field of wearable electronics,[1,2] stretchable sensors,[3,4] epidermal electronics[5] and implantable devices.[6] Correspondingly, a stretchable and sustainable energy source that can integrate with such stretchable electronics is highly desired to provide power for them. However, traditional power supply remains a challenge with rigid structure and limited lifetime, making it unsuitable for integrating with wearable devices. In this regard, it is necessary to develop a new type of power source that can harvest energy from ambient environment. Many efforts have been made in developing flexible and stretchable power generators based on the piezoelectric or pyroelectric effect and so on,[7,8] but their output performance still needs further enhancement for practical applications.

Recently, triboelectric nanogenerators (TENGs) have been successfully developed to harvest mechanical energy and are achieving rapid progress in both academia and industry.[9–11] Based on the conjunction of triboelectrification and electrostatic induction, a TENG has the advantages of low cost, easy fabrication, flexible structure design and very large selections of materials.[12–15] Therefore, they have become promising candidates to be integrated with wearable electronics and as sustainable power sources.[16,17] Nowadays, several efforts have been made to develop stretchable TENGs, such as structure-based TENGs for harvesting body motion energy,[18] rubber-based TENGs as self-powered body motion sensors or self-powered electronic skins,[19–21] fabric-based TENG that can withstand harsh environments,[22] and conductive liquid based TENG as a wearable energy harvester.[23] However, most of the stretchable TENGs presently consist of multiple components with a thickness of over 0.5 centimeters and have a relatively complex structure, which brings inconvenience and uncomfortable experience as wearable devices. Besides, some single-electrode TENGs with a film structure cannot generate high output.[20,24] Thus, it is necessary to develop a simple and thin TENG with high stretchability and high output to meet the demand of sustainable and wearable electronics.

Herein, we developed a stretchable coplanar electrode triboelectric nanogenerator for effective harvesting of diverse biomechanical energies. The device has a simple thin-film structure, enabling it to conformally attach on human skin or cloth and deform as the body moves. To make the whole device stretchable, we adopt an electrospun polyurethane (PU) nanofiber mat as a template to absorb conductive nanomaterials for fabricating the stretchable electrode. After being elongated by 100% of its original length, the electrode retains a resistance of

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lower than 250 Ω cm^{-1}. The rough and porous surface of the soft electrode is also beneficial for enlarging the contact area of the friction layer and thus enhancing surface charge density. In order to make the s-TENG have a high power generation efficiency and extend its operation range, coplanar electrodes are adopted, one of which is coated with micro-patterned PDMS. By virtue of the high output of the device, it can be attached on cloth and skin for fabricating sustainable wearable devices such as self-powered light-up clothes. We demonstrate the device attached on the knee to harvest biomechanical energy when people walk, run and squat. Furthermore, by adjusting the size of the device, it can be can be utilized as a deformable motion sensor for detecting finger gestures. When people bend different fingers, signals can be generated from the s-TENGs attached on the corresponding knuckle, indicating its application potential for fabricating self-powered electronic skins and smart patches.

Experimental details

Fabrication of stretchable electrode

Firstly, a stretchable electrode template was prepared by an electrospinning process. Thermoplastic PU particles were dissolved into a dimethylformamide/tetrahydrofuran (DMF/THF, 2 : 3) mixture with a weight ratio of 13% and stirred with a magnetic spinor for 2 h until the solution became homogenous. The electrospinning process was conducted at a high voltage of 8 kV and the collecting distance was 10 cm. After continuously electrospinning for 30 min, a layer of nanofiber film with thickness of about 40 μm was formed on the collector. To get a stretchable electrode, conductive nanomaterials were added onto the nanofiber film through a drop-drying process. Silver nanowire (AgNW) solution with nanowire length of 10–30 μm was dropped on the PU nanofiber film and then heated on a hot plate to evaporate the solvent. Conductive carbon nanotube (CNT) solution with a concentration of 1 mg ml^{-1} was added onto the nanofiber film with the same method. After repeating this process several times, AgNWs and CNTs were firmly coated on the PU nanofiber surface and formed a conductive network.

Assembling the coplanar electrode nanogenerator

PDMS elastomer and cross-linker (Sylgard 184, Dow Corning) were mixed together at a weight ratio of 10 : 1 and degassed for 10 min to remove bubbles. The micro-patterned PDMS was transferred from an etched silicon wafer by spin coating PDMS solution at a rotation speed of 500 rpm. When the PDMS was partially cured, one piece of electrode with appropriate size was coated on it and then was peeled off with PDMS together after it was cured at 80 °C for 30 minutes. The stretchable substrate was prepared by the same spin coating method and two pieces of electrode with and without micro-patterned PDMS were put on it when the substrate PDMS was partially cured.

Measurement system

The bottom of the stretchable nanogenerator was fixed to a certain surface and the top was flapped by hand to get contact-separation mode output. For folding mode measurement, the nanogenerator was folded along the center line and triggered by a vibrator. The output current of the device was measured by a low-noise current preamplifier (Stanford Research SR570). The voltage was measured by connecting the device in series with a probe that has a resistance of 100 MΩ and all the signals were recorded and displayed through a digital oscilloscope (Agilent DSO-X 2014A).

Results and discussion

The stretchable coplanar induction triboelectric nanogenerator (s-TENG) has a three-layer structure, as sketched in Fig. 1a. Stretchable and biocompatible thin-film PDMS is adopted as the bottom substrate. In the middle layer, two pieces of conductive AgNWs/CNT/PU nanofibers are parallel embedded into flexible PDMS thin film as electrodes. One of the electrodes is coated with micro-patterned PDMS as friction layer while the other one is directly used as another friction layer. As all the materials chosen to fabricate the device are stretchable, the s-TENG can be stretched along arbitrary directions, which could provide conformable accommodation to a stretchable surface like human skin and cloth. Fig. 1b shows a photo of the fabricated device with a size of 1 × 3.5 cm^2. The device has an ultrathin film structure with a thickness of only 230 μm, as shown in Fig. 1c. In fabricating stretchable electronic devices, realizing the stretchability of the electrode is the crucial part. In our design, PU nanofibers are adopted to make up the major component of the electrode for their intrinsic high stretchability over 600%. Fig. 1d–f shows the scanning electron microscopy (SEM) images of the as-spun PU nanofibers before and after coating with AgNWs and CNTs. After the electrospinning process, the PU nanofiber mat forms a porous nonwoven structure, which greatly increases the surface area of the membrane and enables the nanomaterials solution to infiltrate sufficiently. Once the solvent evaporated, the AgNWs are coated along the surface of PU nanofibers, especially at the junctions, forming an interconnected conductive network. After dropping CNTs, PU nanofibers and AgNWs are fully packaged inside them, forming protection for the AgNWs. Besides, some holes are also filled by the CNT clusters and further enhancing the conductivity of the network. In order to generate a friction couple on the coplanar electrode, PDMS with micro-patterned structure (Fig. 1g) is adopted to package one of the electrodes.

Furthermore, the tensile strain capacity of the electrode after each process was investigated through a stress strain gauge with precise length control. Because of the large Young's modulus of CNTs and AgNWs, the stretchability of the electrode decreases a lot after coating with nanomaterials, as exhibited in Fig. 1h. Although the resistance of the electrode increases with elongation (Fig. 1i), this increase is acceptable and will have little negative impact on the TENG's performance, for the inherent impedance of TENG is much larger than this.

Working principle

The working mechanism of the as-fabricated TENG is schematically demonstrated in Fig. 2 with two different working

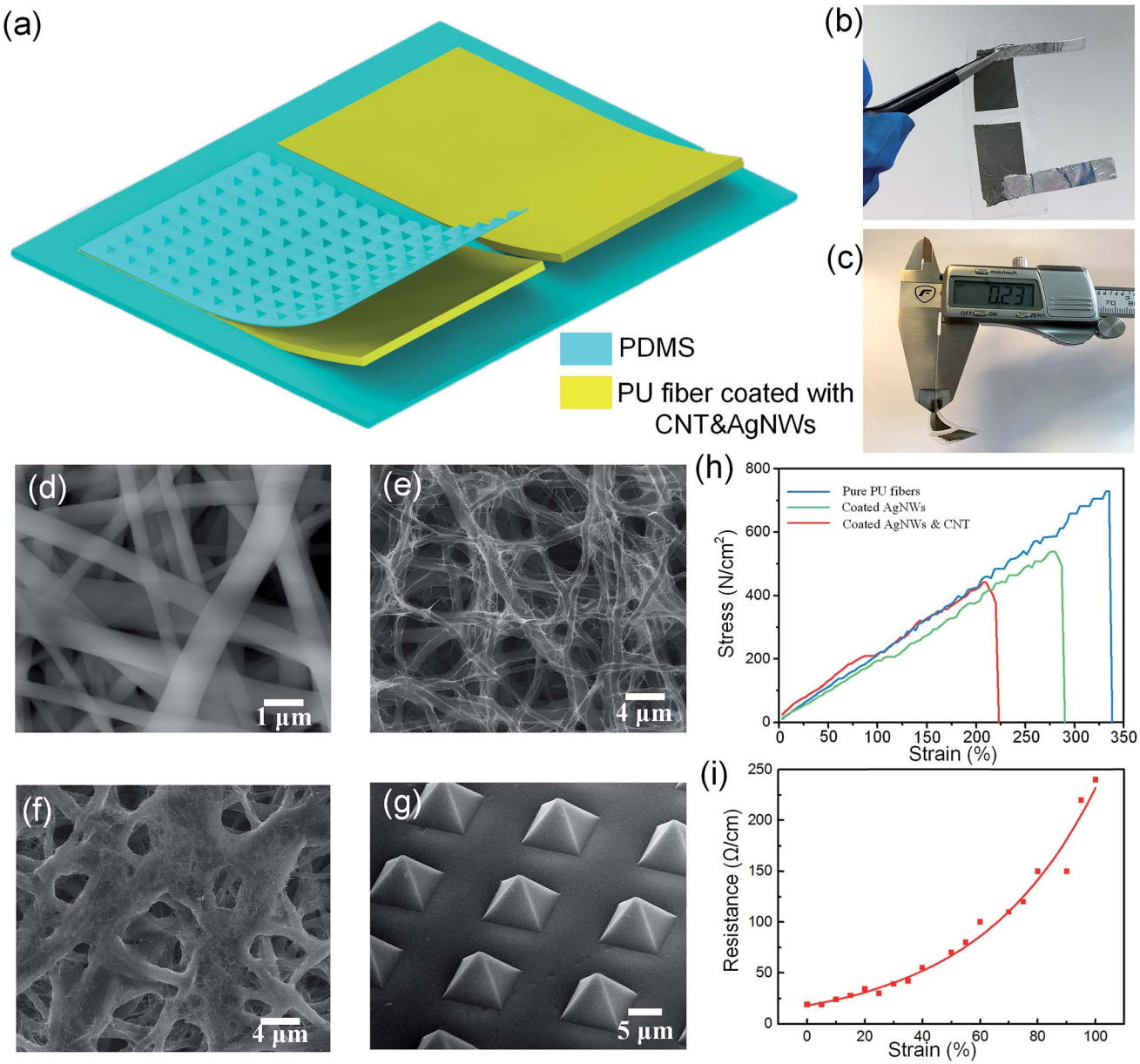

Fig. 1 Structure design of the stretchable TENG. (a) Schematic diagram showing the detailed structure of the s-TENG. (b) Photograph of the as fabricated device. (c) The thickness of the fabricated s-TENG. (d) SEM image of the as-spun PU nanofibers. (e) SEM image of the nanofibers coated with AgNWs. (f) SEM image of the nanofibers coated with CNTs and AgNWs. (g) SEM image of the micro-patterned PDMS. (h) The strain–stress curve of the electrode at different fabrication states. (i) The resistance changes of the electrode with the tensile strain.

situations: contacting with other objects (C-S mode) and folding/unfolding the device (folding mode). Since one of the coplanar electrodes is packaged with PDMS while the other exposes CNTs and AgNWs, there is a large gap of electron affinity in the triboelectric series between them. Therefore, a vast variety of common materials like rayon and plastic can be placed between them and can generate opposite charges on the surface when in contact with the device, as depicted in Fig. 2a(i), where two pairs of friction couple are generated. When the contact material leaves, the negative friction charges on PDMS will remain for a long while and thus the positive charges on the other electrode will transfer to the PDMS electrode by the external circuit because of electrostatic induction (Fig. 2a(ii)). If the contact object totally moves away, the negative triboelectric charges left on the PDMS surface produce asymmetric electric potential between the two electrodes and most of the charges are transferred, as shown in Fig. 2a(iii). Finally, when the object moves back, the transferred charges will be repelled to their original electrode and generate a reverse current in the external circuit.

Besides, the device can be folded and generate opposite triboelectric charges on the friction surfaces by the direct contact between PDMS and nanomaterials, as shown in Fig. 2b. Different from the C-S mode, the CNTs and AgNWs and PDMS

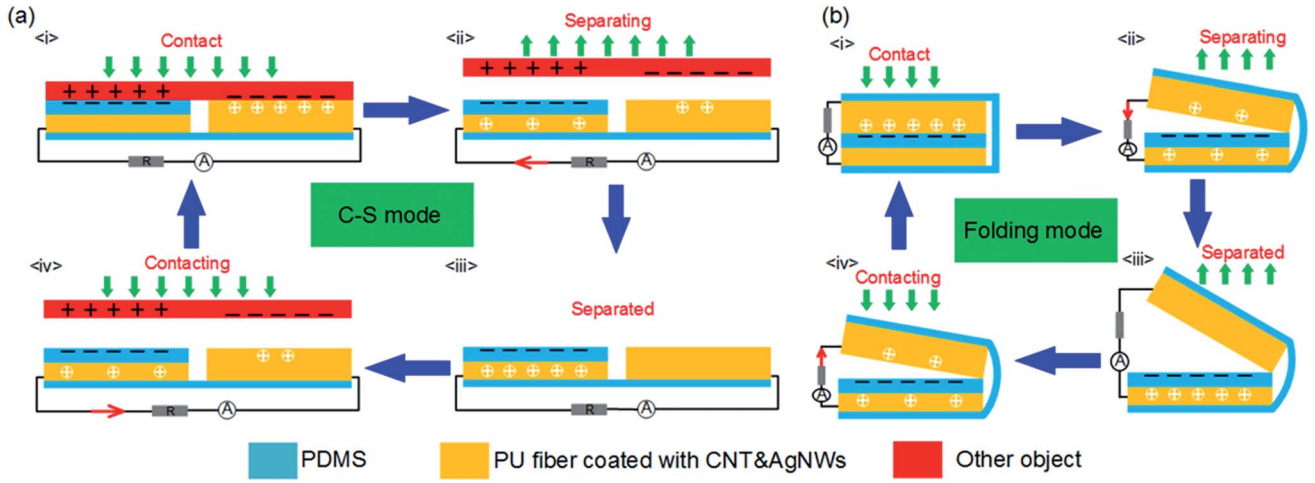

Fig. 2 Working mechanisms of the stretchable TENG. (a) Contacting/separating with other objects. (b) Folding/unfolding the device.

will form a friction couple directly and equal and opposite friction charges will be generated on their surfaces. During the folding/unfolding process, the friction charges generated on the electrode will move forward and back between the two electrodes under the drive of electrostatic induction.

Output characterization

To investigate the electric output performance of the stretchable s-TENG, the device with a size of 3.5 × 3.5 cm^2 is mechanically triggered under folding mode by an electromagnetic vibrator with controlled force at a frequency of 5 Hz. The obtained peak-to-peak output voltage approached 670 V with a contact area of 1.5 × 3.5 cm^2, as shown in Fig. 3a. In addition, the peak short circuit current can reach 29.3 μA, corresponding to a peak current density of 23.9 mA m^{-2} (Fig. 3b). This performance is higher than previously reported stretchable single-electrode TENG (6 mA m^{-2}) and rubber-based TENG (1.35 mA m^{-2}).[20,21] The better results can be attributed to three reasons: (i) the larger difference of the surface charge affinity between PDMS and CNTs and AgNWs; (ii) the increased contact area between micro-structured PDMS and porous non-woven structured nanofibers, resulting in a high surface charge density; and (iii) the coplanar double-electrode TENG having a better charge transfer ability than the single-electrode TENG.[25] The charge output of the device was also measured by two methods. One uses an electrometer with the device triggered by a vibrator, in which the transferred charge in one cycle is about 14.58 nC, as shown in Fig. 3c. Owing to the charge accumulation in the electrometer, the measured curve kept continuously rising overall. The other method involves pressing the device by hand and charging a low-value capacitor after rectifying the output. The transferred charge in one cycle calculated by multiplying the capacitance and voltage is about 27 nC (Fig. 3d). The reason for the difference between the two methods is that the pressure applied by hand is larger than that of the vibrator and therefore more tribo-charges can be generated on the friction layers and therefore more charge can be driven through electrostatic induction. To investigate the actual output power of the s-TENG, we obtained load matching curves using an external load resistor varying from 0.01 MΩ to 200 MΩ, as shown in Fig. 3e. As the resistance increases, the current amplitude decreases while the instantaneous power of the device reaches a maximum value of 3.875 mW at the optimum load resistance of 180 MΩ. This corresponds to an area peak power density of 316.4 μW cm^{-2}. Since the output of the TENG is in the form of pulse which cannot serve as a direct power source for most electronic devices, we employ a capacitor to store the rectified electric output of the device. Fig. 3f shows the charging curve of the s-TENG for a 1 μF capacitor under folding mode. The capacitor can be quickly charged to 10 V in 100 s by the device.

The unique characteristic of the s-TENG in this work is to harvest energy with two electrodes under C-S mode, which leads to better charge transfer ability than the single-electrode TENG and can expand it application area with no need of relative displacement of the device. To verify its electric generation capability, typical output performance of the device including output voltage and short-circuit current was investigated by contacting the device with different common materials, such as skin, butyronitrile and cotton cloth. As can be seen in Fig. 4, the average peak voltages when in contact with butyronitrile, skin and cotton are 210 V, 286 V and 330 V, respectively, while the average peak currents are 8.3 μA, 11.8 μA and 11.2 μA, respectively. In the triboelectric series, the butyronitrile has a strong ability of attracting electrons while the cotton is in the position of easily losing electrons, and the skin is between them. The output voltage is larger when the contact object has a stronger ability of losing electrons. This can be understood from the working mechanism of the device. As mentioned above in Fig. 2a, the movement of electrons between the two coplanar electrodes is mainly the result of the attraction and repulsion of the friction charges on the PDMS, which has a strong electron affinity. Therefore, the easier the loss of electrons for the contact object, the higher is the output of the device. The output currents do not show a regular trend as the voltage does, may be because of the instability of the measurement methods, where the peak current is easily affected by the velocity of movement.

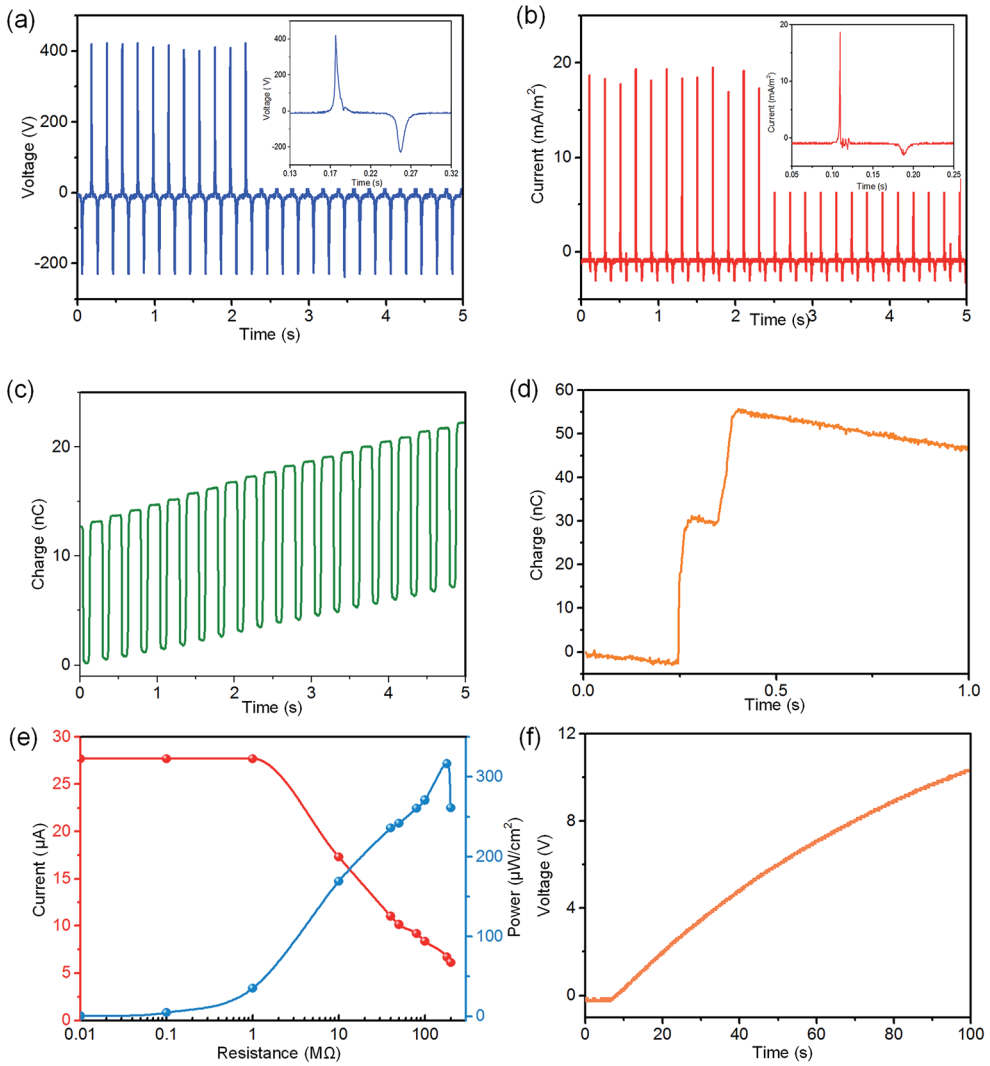

Fig. 3 Output performance of the device at the folding/unfolding state. (a) Output voltage, (b) current and (c) transferred charges of the s-TENG. (d) Transferred charges of the device when pressed by hand in one cycle. (e) Dependence of the output current and peak power on the load resistance. (f) Charging curve of the s-TENG.

Applications

Based on the above measurements and the understanding of the function mechanism, we further propose practical applications of the device as a deformable energy harvester and a self-powered motion sensor. Since the PDMS substrate is biocompatible, the device can be directly put on skin for harvesting diverse body motion energies. As shown in Fig. 5a, the s-TENG is conformally attached on the knee. Owing to the stretchability of the device, it can deform with the movement of the leg and retain its function. When people do various movements like walking, running and deep squatting, the device would make contact and separate from trousers as the leg becomes bent and straight. In these situations, electric outputs can be generated through the C-S working mode. The electricity can be stored in a capacitor for powering low-consumption wearable electronic devices. Besides, the output waveforms of the device are unique under different movements since the contact frequencies and areas vary when the body moves in different ways. As seen in Fig. 5b–d, the amplitude of the output voltage is largest when people run. This is because the contact strength between the device and trousers is greatest under this movement. According to the output differences under various movements, the signal also can be used to judge the movement state of people and may have potential application in preventing falling of older people.

By adjusting the dimensions of the device, the s-TENG could also be utilized to detect human motion in other body parts. To demonstrate a self-powered gesture sensor, we fabricated four s-TENGs with dimension of 1×3.5 cm^2 and attached them on the knuckles of fingers, as shown in Fig. 6a. Once the hand makes a gesture like "OK", "love" and "victory", the fingers would bend and the devices would be folded along the central line. When the finger bends to a certain extent, triboelectric pairs (PDMS and CNTs and AgNWs) on the corresponding device would contact with each other thus generating electric signal through folding mode. Fig. 6b demonstrates the output signals of four

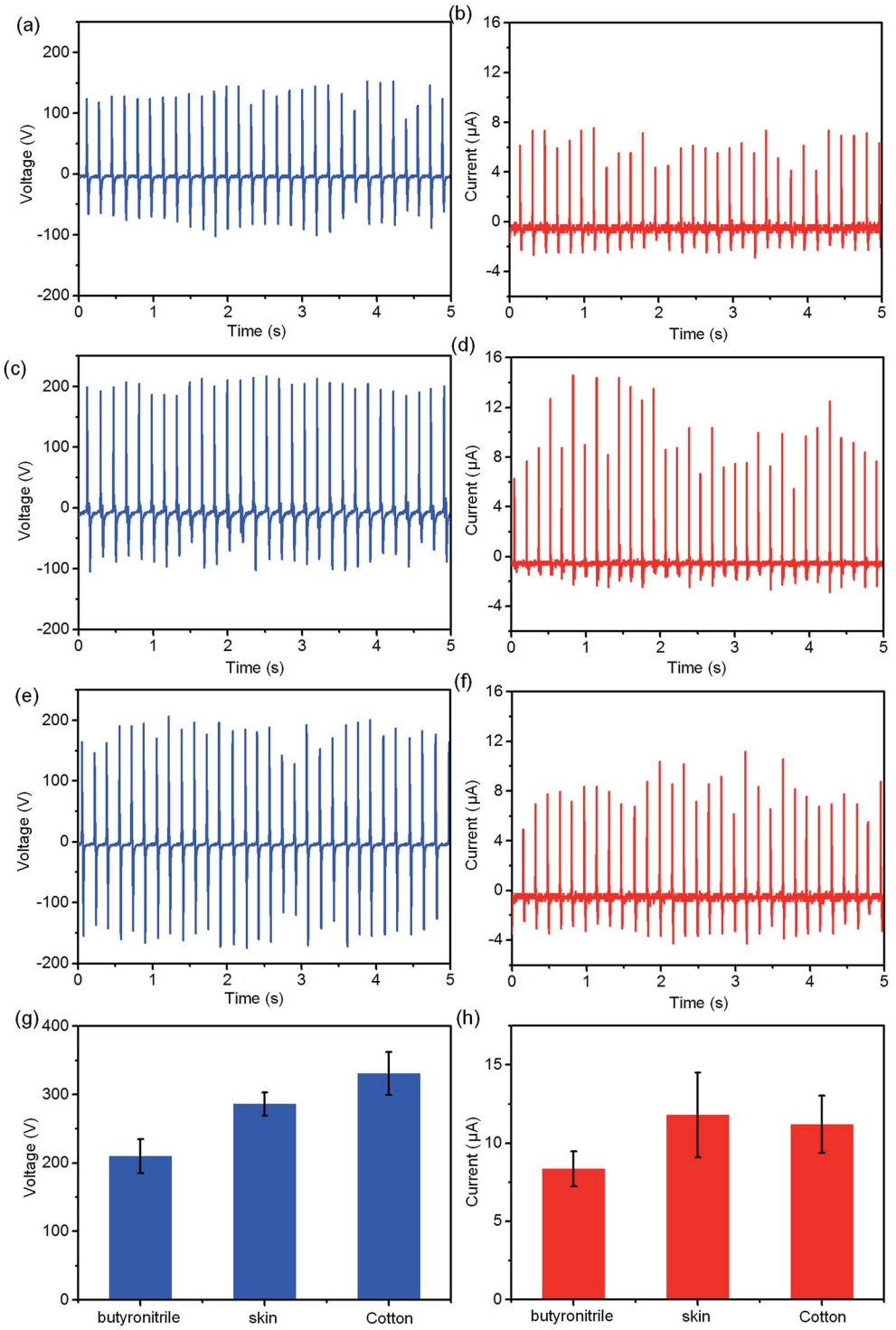

Fig. 4 Output performance of the s-TENG when in contact with other objects. (a) Output voltage and (b) current when in contact with butyronitrile. (c) Output voltage and (d) current when in contact with human skin. (e) Output voltage and (f) current when in contact with cotton cloth. (g) Histogram of the output voltage and (h) current when in contact with the three materials.

devices when the hand makes a fist periodically. The difference of output amplitude between each device is mainly induced by the different contact area on each finger since the device is not folded completely. Similarly, when people make different gestures, the fingers will bend or stay straight in a specific order and the electric signals could be detected in the corresponding electrodes. Fig. 6c demonstrates the electric output of the four TENGs when the hand makes a gesture of "OK" periodically. Under this movement, only the index finger becomes bent while the other three fingers remain straight. Therefore, electric

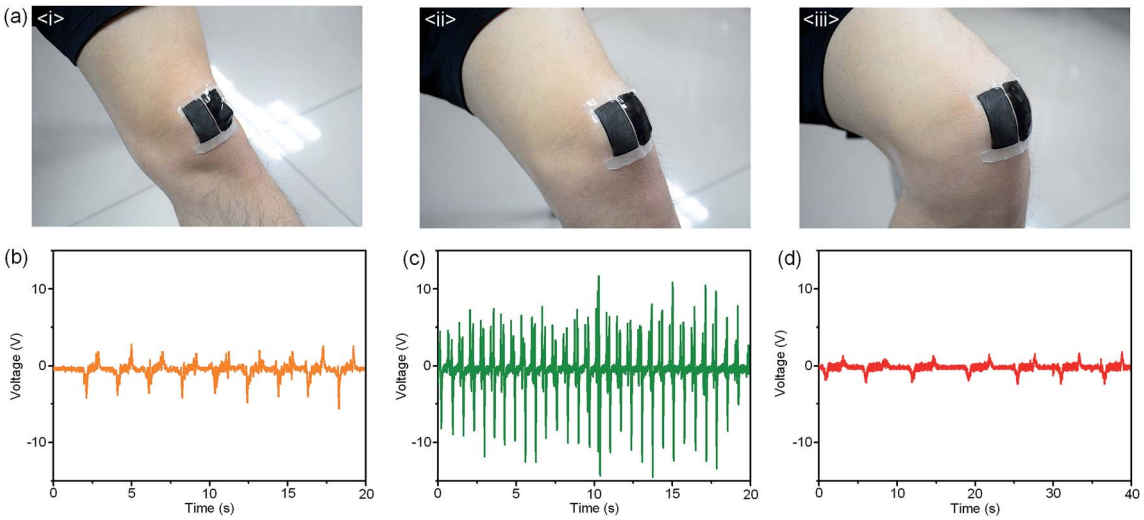

Fig. 5 Application of the s-TENG for energy harvesting on the knee. (a) Optical images of the s-TENG attached on a knee when the leg is (i) straight, (ii) slightly bent and (iii) totally bent. The output of the s-TENG contacting with trousers for (b) walking, (c) running and (d) deep squatting.

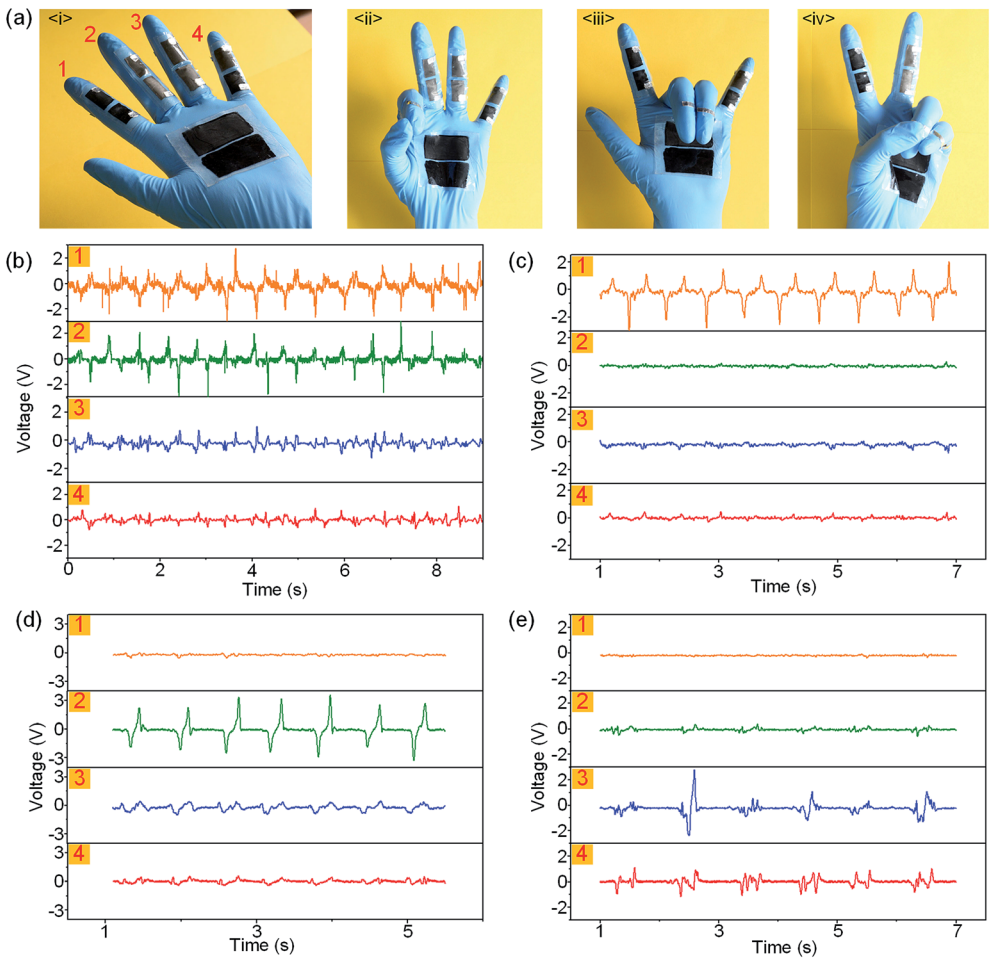

Fig. 6 Demonstration of the s-TENG for gesture sensing. (a) Photographs of the s-TENGs attached on fingers with (i) no gesture, (ii) "OK" gesture, (iii) "love" gesture and (iv) "victory" gesture. Outputs of the four TENGs when the hand makes (b) a fist, (c) the "OK" gesture, (d) "love" gesture and (e) "victory" gesture.

signal can only be generated from the first TENG. In a similar way, when the hand makes the gestures of "love" and "victory", the electric outputs on the "2, 3" and "3, 4" TENGs can be detected, as shown in Fig. 6d and e.

Beyond the application in gesture sensing, the s-TENG may also have potential value in the field of code telegrams, where the telegrapher could send a message by just moving fingers. In this context, the s-TENG as a wearable energy harvester presents great advantages compared with other stretchable TENGs due to its simple film structure, flexible working modes and high output, which might indicate its potential in diverse applications, such as self-powered wearable electronics, smart patches, health care and criminal investigation.

Conclusion

In summary, we have developed a stretchable thin-film structure triboelectric nanogenerator with coplanar electrodes. The s-TENG uses intrinsically stretchable PU nanofibers and conductive nanomaterials as electrode, meaning it retains a high conductivity (<250 Ω cm^{-1}) even after being stretched by 100%. By virtue of the increased contact area of micro-patterned PDMS and nanofiber mat, the device can generate a peak voltage of 670 V and short-circuit current of 29.3 μA in the folding/unfolding working situation, which is higher than those of previously reported stretchable single-electrode TENGs. The coplanar electrodes also enable the device to harvest energy by contacting with other objects such as cotton cloth and human skin, by which a high output voltage of 330 V and 286 V can be derived, respectively. By adjusting the feature size of the s-TENG, we demonstrate it as a deformable energy harvester when attached on skin to harvest biomechanical energy when people walk, run and squat. Besides, the device can be used as a self-powered gesture sensor when attached on a knuckle, which might indicate its potential value in the area of self-powered smart patches, wearable electronics and telegraph communication.

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