Title
A leaf-molded transparent triboelectric nanogenerator for smart multifunctional applications

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Publication Date
2017-02-01

DOI
10.1016/j.nanoen.2016.12.032

Peer reviewed

A leaf-molded transparent triboelectric nanogenerator for smart multifunctional applications

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Highlights

- Natural leaves were introduced as effectively novel mold to fabricate microstructured PDMS in TENG.
- Microstructured PDMS fabricated by leaf was proven to be a promising material for TNEG.
- A relatively fast, environmental friendly and facile galvanic displacement method was used to prepare Ag nanowires.
- Ultra-long silver nanowires were assembled as electrodes in TENG for achieving transparency and flexibility.
- The whole process of TENG fabrication is easy, cost-effective and environmental-friendly.

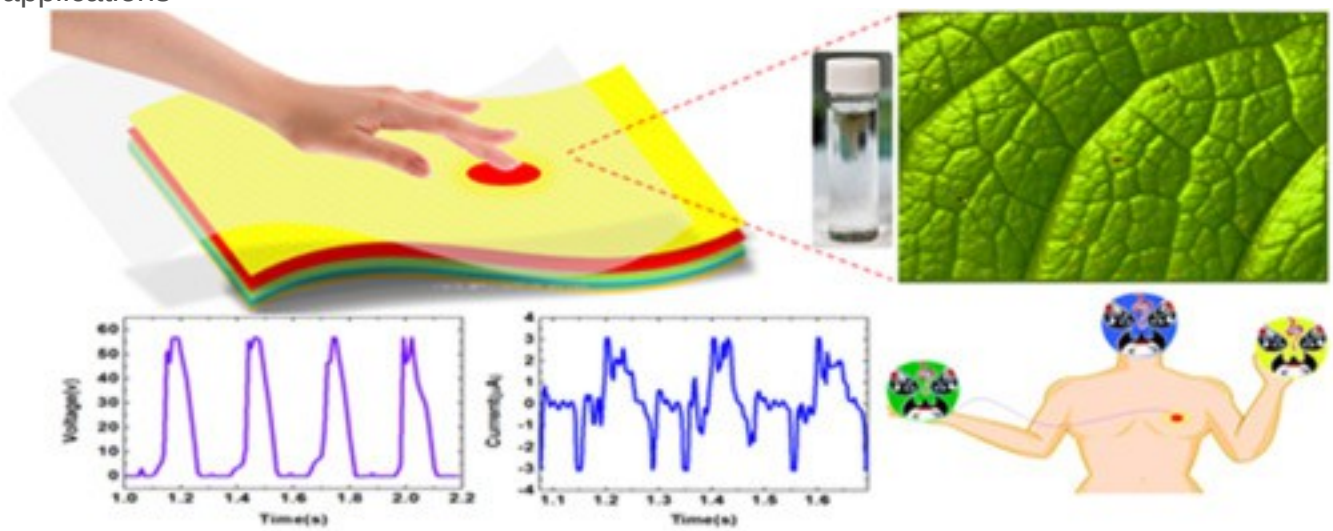
Abstract

Transparent, flexible and highly efficient portable power sources are essential components of the next generation electronics and optoelectronic devices. However, complicated technology, expensive cost, environmental pollution and low-efficient output have limited its development. Herein, based on periodic contact/separation between human skin and the microstructured polydimethylsiloxane (PDMS) film, we demonstrate a transparent flexible triboelectric nanogenerator (TENG) through a relatively simple, low-cost, environmental-friendly and high-efficient method. For the first time, the natural leaves with rich surface textures were introduced to make microstructures on PDMS

films as effective friction surface in the TENG. Furthermore, long silver nanowires (200 μm in length at least) through novel synthesis were assembled as high-performance electrode, resulting in the entirely flexible and transparent TENG. Owing to the unique design, the transparent flexible TENG was eventually obtained with an enhanced output (Voc=56 V; Isc=3.1 μA) and remarkable transparency (88%). Owing to compelling features of the TENG, a self-powered user-interactive wearable system was successfully established by integrating a flexible electrochromic device (ECD). The remarkable color-tunable ability via mechanical control of our system, highly inspired by chameleons, is potentially useful in military camouflage, monitoring human activity visually, as well as replacing performance of face change in Sichuan Opera. Therefore, this research is a substantial advancement toward the construction of transparent nanogenerator and its multifunctional applications in energy conversation, wearable electronics, healthcare, culture experience, and even environmental protection.

Graphical abstract

A leaf-molded transparent triboelectric nanogenerator for smart multifunctional applications



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Keywords

Bio-inspired

Transparent
Triboelectric generator
Silver nanowires
Leaf-molded
Environmental-friendly

1. Introduction

Flexible electronic devices with high transparency, good conductivity as well as remarkable biocompatibility are expected to meet ever developing demands for the next generation of electronic and optoelectronic devices [1], [2], [3], [4]. In particular, when integrated into wearable systems, the independent portable green energy supply is highly desired for realizing self-powered operations of electronic devices [5]. Harvesting energy from the ambient environment like mechanical friction and vibration is widely considered as a promising route due to its abundance and reliability [6]. Recently, triboelectric nanogenerators (TENGs) based on the coupling effect of contact electrification and electrostatic induction [7], [8] have been developed to effectively convert small mechanical energy surrounding human body into electricity. Especially, technological realization of energy harvest from human motions is highly attractive due to the potential applications in wearable multifunctional smart systems [9], [10]. In addition, TENGs with high transparency have aroused more and more attentions for an extended range of applications in wearable electronic devices with displays. Besides transparent substrate, a high-performance transparent electrode, therefore, plays a key role in TENGs for realizing displaying input/output information onboard self-powered wearable electronic devices. Recently, transparent TENGs using ITO or graphene as electrodes have been reported [11], [12], [13]. However, they are hampered by the presence of intrinsic defects resulting in brittleness of ITO as well as technical difficulties. Thus, there is an urgent need for an effective electrode suitable for producing transparent TENGs in wearable electronics with displays.

In addition to characteristic of transparency, output performance of the TENG is of great importance and has been improved drastically through excellent structural design and material optimization in the past decades [14], [15]. Notably, it has been demonstrated that the patterned surface with rich microstructure is an effective approach to enhance the performance of the TENG [16]. However, the conventional patterning process through Si mold is complicated and expensive since it requires multi-steps (spin coating, lithography, etching and so on) [17], [18]. Thus, it is highly desired for a simple,

cost-effective and large-scalable method for patterning process in the fabrication of microstructured surface.

Herein, we present a transparent flexible TENG based on periodic contact/separation between human skin and a patterned PDMS film. As one of the most abundant biomass sources on earth, the leaves with rich surface textures in microscale [19], [20] were introduced as effective mold to make micro-nano structures on the PDMS. Superior to sophisticated conventional processes [17], [18], the patterned method utilized by us is green and low-cost, suitable for mass production. On top of this, silver nanowires were integrated to construct the transparent flexible TENG to serve as flexible electrode. The silver nanowires utilized by us can reach a high aspect ratio up to 4000, which may account for the high transparency of electrodes. Unlike conventional methods using PVP [21], [22], the silver nanowires were synthesized through a relatively fast, environmental friendly and facile galvanic displacement method, therefore suitable for large-scale production [23]. Finally, the TENG fabricated by us has good energy conversion properties (Voc=56 V; Isc=3.1 μA) and remarkable transparency (88%). Owing to the flexibility and transparency characteristics of the TENG, we successfully established a self-powered user-interactive wearable system by integrating a flexible electrochromic device (ECD) capable of reversibly changing optical properties related to externally applied voltage [23], [24]. Through relative response between dynamical friction and the color of smart device, we can monitor human activity visually, potentially useful for real-time health care if the ECD is driven by pulse or heartbeat in the near future. Apart from this, the smart device may be employed in the performance of face change (referring to a popular play Sichuan Opera involving the change of masks in quick succession to show different emotions and feelings of the character) to a certain degree through color changes under successive touching with high entertainment value.

2. Results and discussion

Inspired by gradually optimized leaf surface structures, we replicated leaf morphologies on PDMS for harvesting mechanical energy effectively. A schematic flow chart is shown in Fig. 1. A piece of ramified leaf (10 cm in length), as shown in Fig. 1(a), was used as the mold for fabrication of micro-patterned PDMS in TENG. The rich textures on the leaf surface is mainly caused by the leaf venation embedded in the mesophyll to achieve mechanical stability and other functional properties [25]. The architecture of one branch of the leaf venation with microstructure is further illustrated through a SEM image in the inset of (a). The leaf is then tailored to an appropriate shape as shown

in Fig. 1(b). Subsequently, the fresh leaf was stuck to the bottom of the die in Fig. 1(c), followed by the mixture of PDMS base and cross-linker deposition on the leaf master in Fig. 1(d). Finally, cured at 80–120 °C for 2 h in a vacuum oven, a flexible leaf-mimicking PDMS was obtained as shown Fig. 1(e) and (f), which presents a simple, cost-effective and environment-friendly method for fabrication of mold with rich textures in microscale.

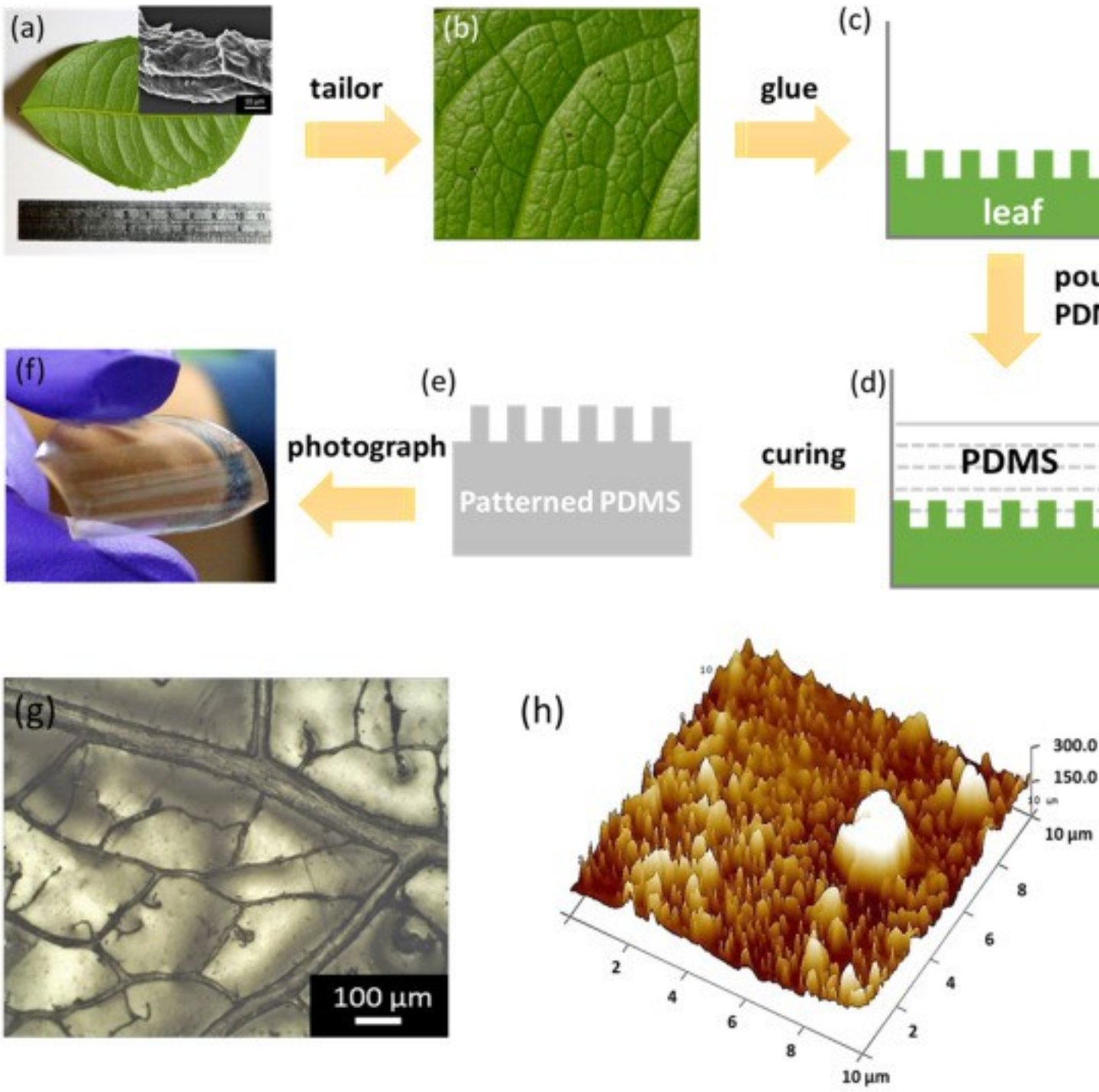

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Fig. 1. (a)-(f). Schematic flow figures of patterned PDMS fabrication process. (a) A fresh leaf was taken as a master mold, inset shows SEM image of one branch of the LV with rich textures. (b) The leaf was cut into appropriate shape. (c) The leaf was stuck in the die. (d) PDMS was deposited on the leaf. (e) The flexible patterned PDMS mold was

obtained. (f) The photograph of flexible microstructured PDMS film. (g) SEM image of patterned PDMS film. (h) AFM image of the patterned PDMS film.

To further characterize the morphology of the PDMS surface, scanning electron microscopy(SEM) and atomic force microscopy(AFM) were employed and the results are shown in Fig. 1(f) and (g) respectively. As illustrated in (f), concave lines (varied from tens to hundreds of micrometers in length along with various widths) interweave with each other to form a naturally arrayed pattern. In addition, the morphology of the PDMS films, including the size, microstructure unit density etc., can be tunable by choosing different types of leaves, as well as different regions of the same leaf. In this work, the leaf mold we chose is the ramified pattern, which is one of the most abundant textures for leaves in nature. To further investigate the condition of leaf-inspired microstructure on PDMS film, AFM has been employed. Clearly evident from the 3D topographic image of AFM in Fig. 1(h), the modified PDMS is greatly roughened in nanoscale, which has average roughness (Ra) of 32.1 nm and root mean square roughness (Rq) of 46.4 nm, respectively. The results indicate that leaf-inspired microstructure on the PDMS film plays an important role in improving effective contact area of the surface, which may account for high output performance of TENG fabricated in later discussion.

In addition to the PDMS surface properties, electro-optical properties of the electrode in TENG is also critically important, especially when combing with smart displays. Nowadays, silver networks stand out among alternatives emerged for replacing traditional ITO as transparent electrodes due to their extraordinary conductivity, flexibility and easy synthesis. As a core component of the transparent TENG, the electrode fabricated by us was composed of long silver nanowires with hundreds of micrometer in length, shown in Fig. 2(a). Deduced from the single silver nanowire marked with red star and a bunch of silver nanowires marked with yellow sphere, the specific length of silver nanowires is as long as 200 μm. As illustrated in Fig. 2(b), the diameter of the long silver nanowires is about 50 nm, which leads to a high aspect ratio over 4000. The high aspect ratio of our silver nanowires is higher than those reported for previous works on silver nanowires, typically in the range of 100–1000, which may account for the high performance of our electrode in later discussion.

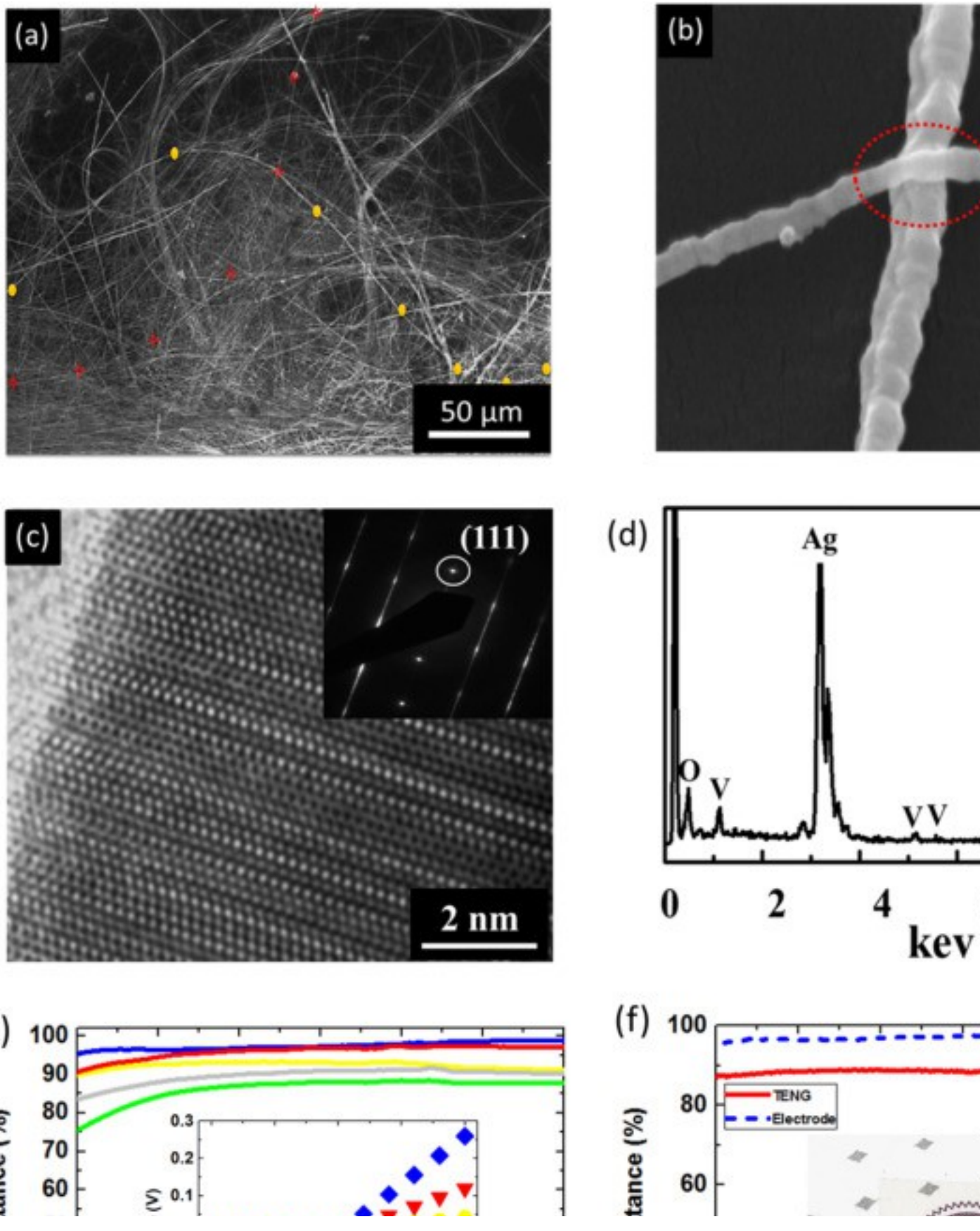
(a)
50 μm
(b)
(c)
(111)
2 nm
(d)
Ag
O
V
VV
0
2
4
kev
(e)
100
90
80
70
60
tance (%)
0.3
0.2
0.1
(V)
(f)
100
80
60
tance (%)
TENG
Electrode

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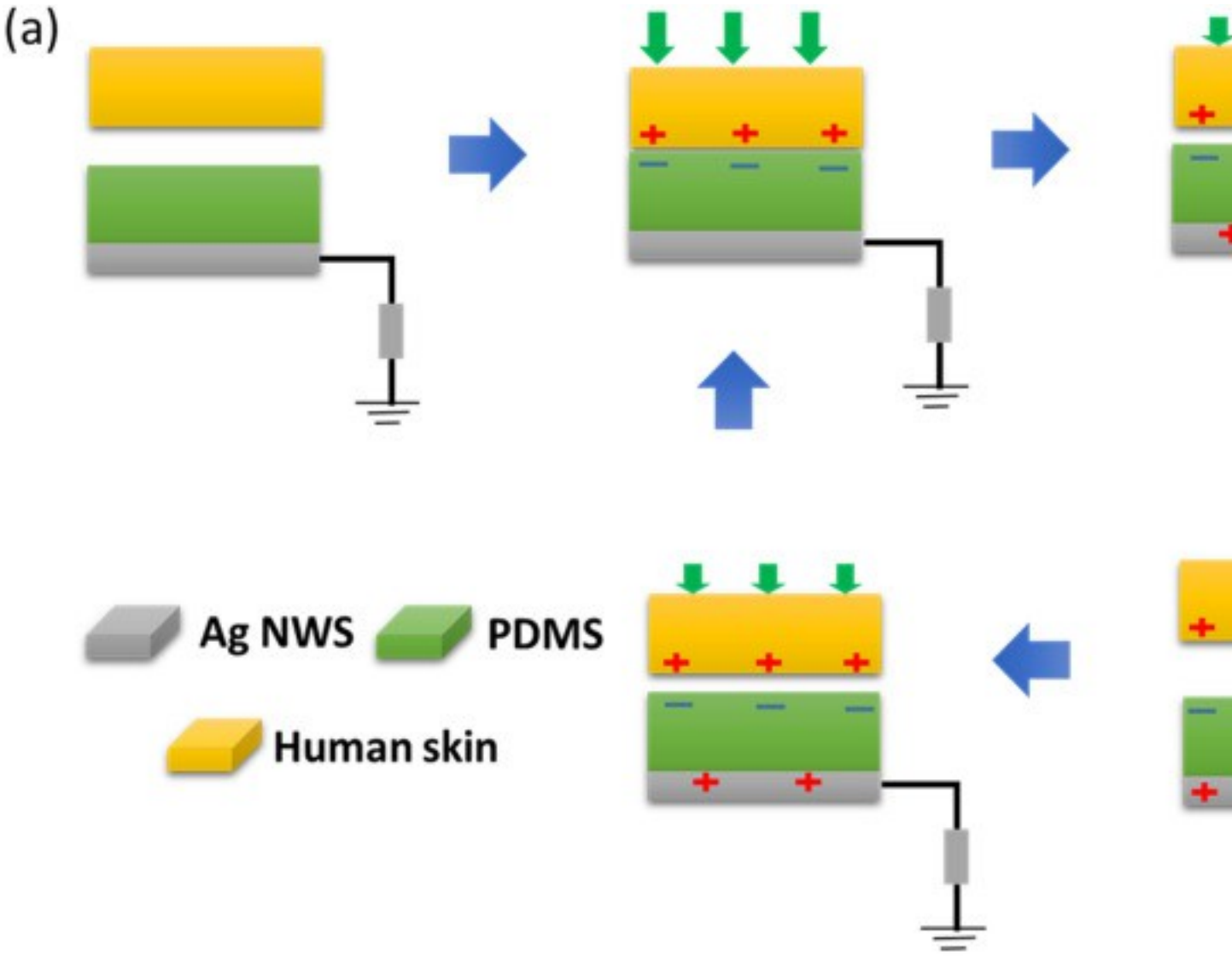
Fig. 2. (a) and (b). SEM images of long silver nanowires. (c) High resolution TEM image of Ag nanowire and the inset electron diffraction pattern. (d) EDS taken from the Ag/VOx core-shell structure. (e) Transmittance and corresponding sheet resistance (inset) of the electrode in the TENG using different density of silver nanowires (blue, red, yellow, gray and green color lines/triangles correspond to 1, 2, 3, 4 and 5 mL Ag NW solution, respectively.). (f) Highest transmittance of electrode and the related transparency of the TENG itself. Inset shows photograph of naked transparent TENG.

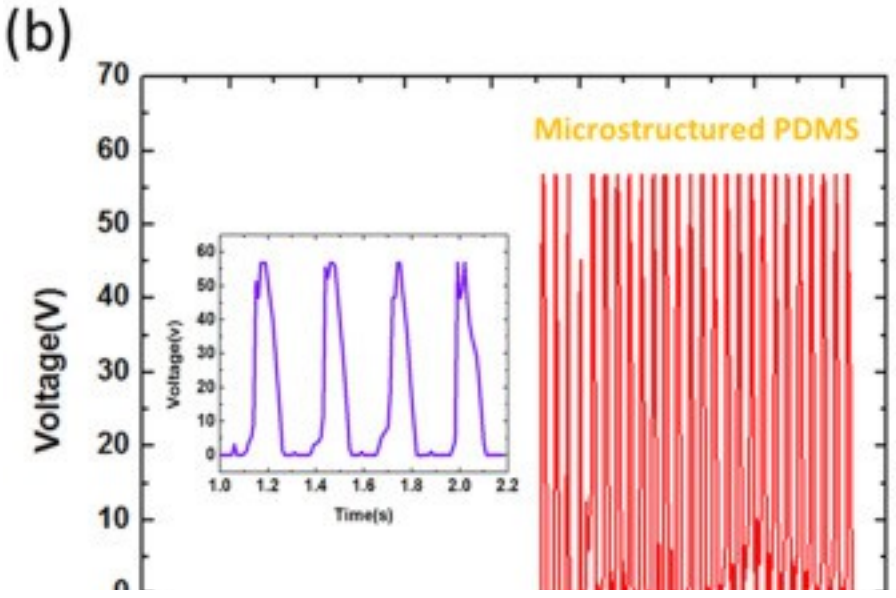
From the HRTEM image in Fig. 2(c) and SAED pattern in the inset, the Ag NWs were found to be crystalline and consist of a high density of nanotwins and stacking faults, with habit planes perpendicular to the axis of the nanowire in the [111] direction. The HRTEM image also shows that the surface of Ag NW is flat and sheathed with a VOx layer (~nm in thickness, in 1.0 mM $AgNO_3$ at room temperature). Fig. 2(d) shows that the sheath is composed of O and V. The Cu peak came from the TEM grid. With low twinning and stacking fault energy in faced-centered cubic Ag, twins and stacking faults are prone to form in the nanowires, particularly, under high growth rate condition. Hence, we have demonstrated a relatively fast, environmental friendly and facile galvanic displacement method to prepare high aspect ratio Ag nanowires on V foil surface, superior to traditional method without adding external capping agent like polymer and surfactant.

The performance of electrode in TENG is further evaluated in Fig. 2(e). Suspensions of silver nanowires in ethanol was diluted down to a concentration of 0.01 mg/mL with deionized water and then sonicated for 30 s. Subsequently, Ag NW conductive films were prepared by spin coating with various volumes of nanowire solution, using 1 mL, 2 mL, 3 mL, 4 mL, and 5 mL solution, separately. Correspondingly, optical transmittance decreases with increasing of Ag NWs density, tuned from 97% to 95%, 93%, 89%, and 86% shown in Fig. 2(e). Simultaneously, the conductivity improves as the density increases, changing from 118.3 Ω/sq to 54 Ω/sq, 19.4 Ω/sq, 6.5 Ω/sq and 2.6 Ω/sq, respectively, as inset shown in Fig. 2(e). To achieve a high transmittance of the TENG, which is mainly determined by the transparency of electrode, we selected 1 mL of silver nanowires solution with concentration of 0.01 mg/mL. As is shown in Fig. 2(f), highly transparent TENG is obtained with optical transmittance of 88%, which may be attributed to the related high transmittance of electrode of about 97%.

According to the triboelectric series, human skin can be used as triboelectric material to harvest biomechanical energy in the touching process by utilizing single-electrode-

based TENG techniques. The working mode of the transparent single-electrode TENG fabricated is schematically depicted in Fig. 3(a). When the skin contacts the PDMS film with leaf-inspired microstructures on its surface fully, charges are generated between them due to the different triboelectric series. Electrons transfer from the triboelectric positive surface (skin) to triboelectric negative surface (PDMS), resulting in the fully balanced paired charges with no electron flow in the external circuit [25], [26], [27]. Once PDMS and skin separated, the negative charges on the surface of the PDMS cannot be compensated by the ones on skin. Subsequently, positive charges on the Ag NWs electrode was induced to reach electrostatic equilibrium, driving free electrons to flow from the Ag NWs electrode to the ground via external load. In addition, output of voltage/current is generated during this electrostatic induction process until the negative charges on PDMS are fully screened. Furthermore, once the skin is reverted to approach the PDMS, the electrons will flow back in the opposite direction until the original state is attained, resulting in reversed output of voltage/current. Based on the mechanism depicted above, the output performance of the TENG was further evaluated during periodical touching between the skin with leaf-inspired PDMS film. As is shown in Fig. 3(b) and (c), the instantaneously generated open-circuit voltage (Voc) and short-circuit current (Isc) reach up to 56 V and 3.1 μA, respectively.


(a)
Ag NWS
PDMS
Human skin


(b)
Voltage(V)
Microstructured PDMS
Voltage(v)
Time(s)

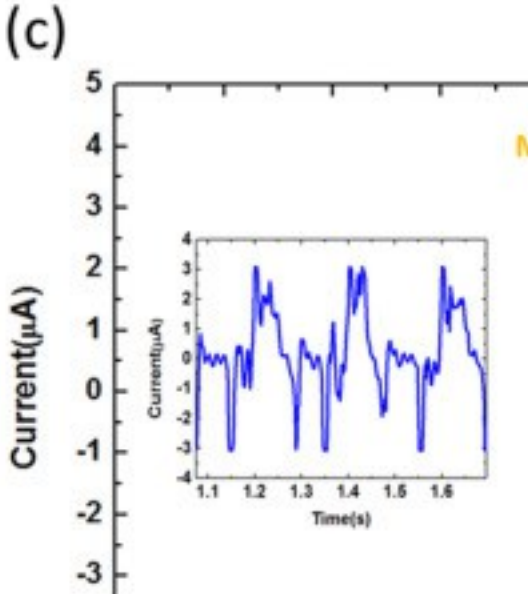

(c)
Current(μA)
Current(μA)
Time(s)

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Fig. 3. (a) Working mode of the transparent single-electrode TENG. (b) Open-circuit voltage (Voc) and (c) Short-circuit current (Isc) of the TENG based on patterned PDMS. Insets show detailed waves of Voc and Isc, respectively.

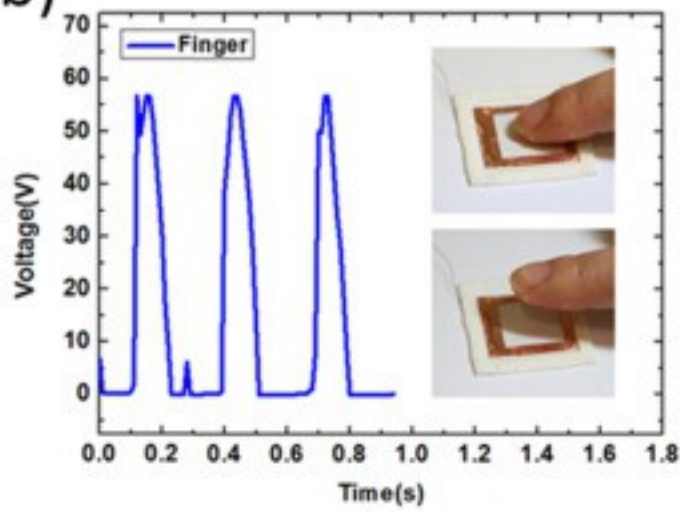
PDMS with leaf-like pattern is preferred to leaf itself due to the difference of triboelectric series between skin and PDMS as well as leaf. In addtion, the leaf is fragile and easy to be destroyed during the successive touching and separating process while the PDMS will stay the intact. The results indicate that the PDMS modified by leaf-inspired microstructure is effective to harvest biomechanical energy from human motions.

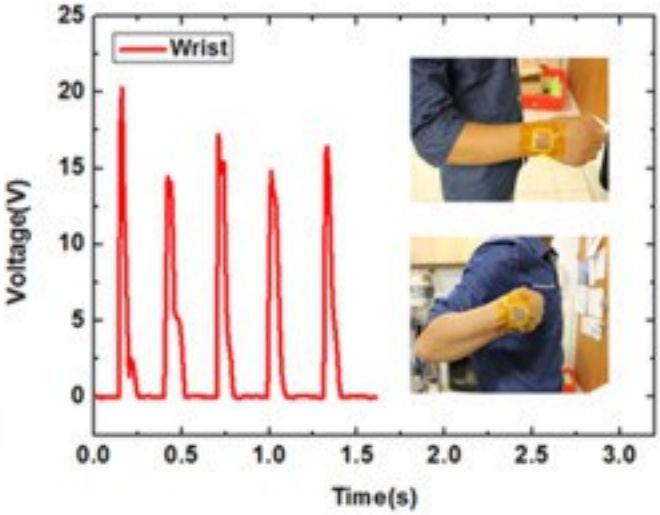
Due to the high output performance and transparent flexible characteristic of the TENG, a self-powered user-interactive smart system is well established. As illustrated in Fig. 4(a), the smart system was realized through integrating a flexible multicolor ECD with a flexible transparent TENG [28], which provides visual response upon successive touching the device. The transparent single-electrode TENG generated an alternating current (ac) output, which was regulated by the full-wave bridge rectifier to convert it into direct-current (dc), flowing through the ECD for charging the Prussian blue (PB) layer and nickel-based Prussian blue analogue (Ni-PBA) layer (both are 3 cm×2 cm in size). The charging cycle will keep taking place for harvesting biomechanical energy and then charging the ECD when the TENG in smart device is subjected to a continuous touching. Notably, the remarkable color-tunable ability via mechanical control of our system is highly inspired by the ability of chameleons. Since it is commonly accepted that the chameleon is able to shift its skin color through controlling the skin pigment cell for purposes in temperature maintenance, camouflage, as well as communication [29]. Besides the potential application in military camouflage, the smart system can be used to replace special effect in the performance of Sichuan Opera to certain degree, as well as monitor human motions visually.

(a)


+
−
PET/Ag NWs
PB
Gel Electrolyte
Ni-PB
PET/Ag NWs

(b)


Finger
Voltage(V)
Time(s)
0
10
20
30
40
50
60
70
0.0
0.2
0.4
0.6
0.8
1.0
1.2
1.4
1.6
1.8


Wrist
Voltage(V)
Time(s)
0
5
10
15
20
25
0.0
0.5
1.0
1.5
2.0
2.5
3.0

(c)

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Fig. 4. (a) Design for construction of smart device. (b) Different ways for driving the smart device. (c) Photographs of the application of smart device in Chinese traditional face changing.

Despite of the simplest way through finger pressing, the flexible TENG can be attached on joint area of the elbow or wrist as a bendable power source depicted in Fig. 4(b) due to its flexibility and searchability characteristics. And as illustrated in Fig. 4(c), the smart device driven by the wearable TENG with different modes can be used to perform Chinese traditional Opera in a new form to certain degree. The first picture shows the pure facial makeup while the remaining 3 pictures show the effect after touching the e-skin through different human motions. When the ECD is not working, its initial color can turn the white region of the makeup to blue while the yellow region changes to green simultaneously. Upon pressing or bending the TENG, ECD can be driven to change color from blue to green and even yellow as well as the white region of facial makeup. In addition, larger dynamical friction and degree of curvature means larger effective contact area of TENG which may lead to larger output driving the colors from blue to yellow (finger touching) while smaller output just enables the display of green (elbow bending) or even maintains blue (wrist bending) within a certain time period. In contrast to the normal form of the art, taking a very skillful person under strenuous training to perform, the remarkable interactions between human body motions and color changes of smart device opens up a possibility for ordinary person to experience traditional face-changing culture in a simple and interesting way in our daily life. Imagine one day, we can enjoy face-change art whenever and wherever possible, in many occasions. Thus, the smart device that combines healthcare with entertainment value may lead to a more healthy and enjoyable life.

3. Conclusions

In conclusion, we have demonstrated a new flexible transparent nanogenerator by combining bio-inspired micro-patterned PDMS film with long silver nanowires network. For the first time, the ramified leaf with rich textures is used as an effective mold to modify PDMS film as friction surface in the TENG. A maximum output up to 56 V and 3.1 μA was obtained, which is almost 60 times as high as that of the previously reported transparent single-electrode TENG based on PDMS and skin [27]. On top of this, leaf is abundant and of low-cost, which is superior to other templates previously proposed in terms of resources and fabrication in large scale. In addition to

transparent polymer materials in TENG, the electrode consists of long silver nanowires showing ultrahigh transparency of ~97%, resulting in an entire transparent flexible nanogenerator. Furthermore, owing to the high transparency of ~88%, the TENG can be integrated with a flexible multicolor ECD to establish a self-powered user-interactive wearable device for the first time. The whole process for TENG fabrication is relatively simple, environmental friendly and facile, which is suitable for mass production. Last but not the least, the smart device is capable of replacing special effect in the performance of Sichuan Opera named face changing through relative interactions between dynamical friction induced by body motions and the color of electronic facial mask. It opens a possibility for people all over the world enjoying fun of Chinese traditional culture in an interesting way. If driven by pulse or heartbeat, the smart device can combine healthcare with entertainment value, leading to a more healthy and enjoyable life.

4. Experimental section

4.1. Fabrication of unmodified PDMS

The PDMS solution was prepared by mixing the Sylgard 184 elastomer with curing agent in 10:1 proportion and stirring for 1 min. Thereafter, the PDMS mixture was kept in vacuum for 30 min to remove air bubbles. After that, the prepared solution was coated onto flexible PET and cured at 60 °C for 2 h. Finally, the PDMS was peeled off from the PET substrate.

4.2. Synthesis of silver nanowires

Silver nitrate ($AgNO_3$) and V foil (99.8%, Alfa Aesar) were employed without further purification. Ag nanowires were prepared on V foil by using 10 mL of Ag salt solution in a 10 mL vial with different concentration. After the synthesis, the V foil was cleaned with D.I. water and blown-dry with N_2 gas.

4.3. Synthesis of PB and Ni-PB NPs

PB and Ni-PB film were synthesized by mixing the transition metal ions provided by FeCl3 and NiCl2, respectively, with ferrocyanide or ferricyanide ions. After stirring mixture of them with an equal molar ratio for 1 h, the precipitates should be centrifuged and washed with DI water. In addition, a filter process is essential to prevent PB and Ni-PBA NPs aggregating in water [30].

4.4. Fabrication of flexible ECDs

The as-prepared water-soluble PB and Ni-PBA were coated on Ag NWs film by spin coatingat 600 rpm for 5 s and then at 1200 rpm for another 20 s. After curing at 100 °C for 10 min, placing an epoxy tape as the spacer to fill in the gel electrolyte(LiClO4:poly(methylmethacrylate):PC:CH3CN=3:7:20:70 by weight) [31]. Finally, the other Ni-PB film was placed on the electrolyte and clipped tightly and sealed to assemble the ECD [32].

Acknowledgments

The research was supported by the Ministry of Science and Technology, Taiwan through Grant nos. MOST 104-2221-E-007-030-MY3 and MOST103-2218-E-011-007-MY3.

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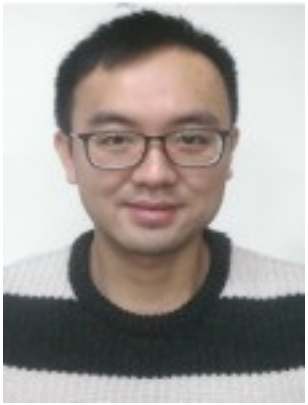

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