## A FLEXIBLE AND WEARABLE GENERATOR WITH FLUOROCARBON PLASMA INDUCED WRINKLE STRUCTURE

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#### ABSTRACT

This work presents a flexible and wearable energy harvester for harvesting muscle motion energy. A novel single-step fluorocarbon plasma induced wrinkle structure is employed as the friction layer to increase its performance by enlarging the contact area and introducing material modification at the same time. Under bending or pressing, this device could produce an alternating current. Additionally, by adjusting the spin-coating speed, this wrinkle morphology and the thickness of PDMS (polydimethylsiloxane) film could be controlled. Therefore, through the optimizing of this parameter, 225 V peak voltage and 375 µA maximum current is achieved under finger typing. Moreover, this device is successfully mounted on an adult's arm to scavenge the mechanical energy during his motion. Due to its well flexibility, simple manufacturing process, and high output performance, the generator has much potential for powering up wearable electronics or e-skin.

#### **INTRODUCTION**

Flexible electronic devices, such as flexible electronic screen, organic light emitting diode (OLED), and flexible tactile sensor, have been thought to be the next-generation electronic devices. One of the most important technical issue blocking their development is to provide flexible batteries for these devices. Among all the optional solutions, flexible energy harvester, scavenging mechanical energy from the environment and human body, is a promising alternative technology [1-2]. To date, the adopted mechanisms mainly include effects of piezoelectric that using nanowires, nanofibers, and bulk material (e.g. PZT) [3-5], and triboelectric that employing the contact-electrification effect and electrostatic induction between two different flexible materials [6-7].

However, fabrication process for these piezoelectric nanomaterials is quite difficult and expensive. The high electric polarization voltage for piezoelectric materials would also bring more potential danger and challenge. Meanwhile, the output performance of these devices are still too low to supply power for some low-consumption device. The fabrication process for triboelectric nanogenerators (TENGs) is simpler, and it could provide more considerable performance and higher power density in a relative low price [8-9]. Besides, the most adopted methods to enhance the performance of vertical-contact-mode TENGs are the utilization of micro/ nano structures and material modifications, both of which lead to a higher surface charge density [10-12]. But there is no work that combines these two methods together to further increase the surface charge density yet. Simply adding those up would also make it difficult and expensive for the fabrication process.

Thereby, aiming at obtaining a high performance flexible generator, we introduce a novel method based on fluorocarbon plasma treatment to combine these two methods together in a single step, obtaining a high performance flexible TENG. By treating the uncured PDMS, wrinkle structure is observed, and fluorocarbon copolymer is deposited at the same time. In addition, this winkle morphology and the thickness of PDMS film can be simultaneously controlled and adjusted by altering the spincoating speed of its mixture, which leads to further dramatically enhancement of the performance of the generator. Meanwhile, we investigated and characterized the morphology change systematically and quantitatively at the aid of laser scanning microscope (LSM). Facilitated by this novel method and its flexibility, this TENG could be employed to harvest the mechanical energy during the body motion, even on one's arm.

#### STRUCTURAL DESIGN

The 3D view of this wrinkle structure based TENG and its serial connection are diagramed in Figure 1. We use a thin polyethylene terephthalate (PET) film as the substrate and a thin indium tin oxide (ITO) as the electrode, which make the total structure flexible. Wrinkle structured PDMS is fabricated on the ITO film of bottom layer, while an archshaped structure is adopted for this TENG. This arch-shaped structure can be pressed to contact with each other under outer force, while which would separate rapidly under the elastic energy of this structure after the leaving of this force. Besides, a novel serial TENGs design is proposed by the connection of these arch-shape TENGs, which could provide better accommodation to curved surface (e.g. a person's arms).



Figure 1: Schematic diagram of the wrinkle TENG and its serial connection.

### **FABRICATION PROCESS**

The fabrication process for the serial TENGs is shown in Figure 2, schematically. It begins with an ITO-coated PET film, the thickness of the PET and ITO layer are about 125 µm and 180 nm separately (Figure 2a). After the PDMS elastomer and cross-linker (Sylgard 184, Tow Corning) were thoroughly mixed in a 10:1 ratio (w/w) and degassed, it was spin-coated onto the ITO layer (Figure 2b). Before the PDMS cured, a C<sub>4</sub>F<sub>8</sub> plasma treatment process was carried out in an inductively coupled plasma etching (ICP) machine as shown in Figure 2c. During this process, the fluorocarbon copolymer was deposited on the surface of uncured PDMS, and the wrinkle structure would be formed spontaneously at the same time (Figure 2d). Then, after the PDMS was cured at 90°C for 30 minutes, the processed PDMS film was assembled with another PET/ITO film to form an arch-shape TENG by using polyamide (PI) tape to bond two ends of the two films (Figure 2e). Finally, three of arch-shape TENGs were connected by PI tape to form the serial TENGs (Figure 2f).



Figure 2: Fabrication process of the wrinkle structure and the serial TENGs.



Figure 3: (a) Photographs of the device, (b-c) its deformation under press and bending, the conformal contact with a curved surface.

The photo of fabricated wrinkle TENG is shown in Figure 3a, which has a size of 40 mm  $\times$  50 mm, and the maximum gap size of which is fixed at 3 mm. As exhibited

in Figure 3b-c, two layers of this device would be brought into periodic contact and separate under finger's press or bending. Additionally, the serial TENGs could contact conformally with a curved surface.

# CHARACTERIZATION OF WRINKLE STRUCTURE

The wrinkle morphologies under different spin coating speed is first studied by scanning electron microscope (SEM) as shown in Figure 4. Apparently, disordered wrinkle structure was observed in the PDMS surface. Comparison of parameters proves that the size of typical wrinkle structure became smaller as the increase of spin-coating speed, and the thickness of PDMS film shown the same trend. Therefore, the wrinkle scale is related to the thickness of the PDMS film.



Figure 4: SEM photos of the wrinkle structure under different spin coating speeds with a scale bar of 50  $\mu$ m.



Figure 5: 3D views of the wrinkle structure and the morphology as a function of the spin coating speeds.

Furthermore, CLM is employed to obtain the quantitatively correlation between the spin-coating speed and the wrinkle structure. The 3D views of the obtained wrinkle structures under the spin-coating speed of 1000 rpm and 5000 rpm are shown in Figure 5a-b, respectively. Compared with 5000 rpm, the wrinkle structure under 1000 rpm had larger scale structure and its aspect-ratio was lower. Figure 5c plots

the topography profiles of wrinkle structures under 1000 rpm, 5000 rpm and 8000 rpm, respectively. Similarly, as the increase of the spin-coating speed, more waves with smaller amplitudes were observed. Besides, the average heights and lengths for these profiles are shown in Figure 5d. Specifically, the average height for this structure decreased from 4  $\mu$ m to 3.6  $\mu$ m while the average length reduced from 11.5  $\mu$ m to 6.7  $\mu$ m, which is to say that both the height and length of wrinkle structure decreased with the spin coating speed, and correspondingly, the aspect ratio increased from 0.35 to 0.54. Moreover, it can be concluded that there is a dramatically increase in the surface area of PDMS film as the spin-coating speed increases.

#### ELECTARICAL MEASUREMENT

The output voltage was measured using a digital oscilloscope (Agilent DSO-X 2014A) via a 100 M $\Omega$  probe (HP9258), and the current was amplified through a SR570 low noise current amplifier from Stanford Research systems. The upper surface of the generator was fixed onto a certain place. Then a sinusoidal signal with an amplitude of 1.5 V was generated from the signal source module of the oscilloscope and amplified by an amplifier (SINOCERA YE5871A) to power the modal shaker (JZK-10) to provide a periodic and stable external force to the device. Besides, the operational distance between the vibrational system and the device was controlled to be 5 mm to ensure a comparable movement for different TENGs.



Figure 6: The time trace output voltage and current of a TENG and its enlarged view in a single cycle.

Figure 6 shows the obtained output voltage of a TENG using 1000 rpm-spin-coated PDMS as the contact material. It had a maximum value of 170 V, while the Root-Mean-Square (RMS) value was 48 V. And the observed maximum current for this device was 184  $\mu$ A as plotted in Figure 6b.

Figure 7a gives the correlation between the electrical performance of the generator and the spin-coating speed of PDMS film. Obviously, as the speed accelerates, the output voltage decreased firstly, and a minimum value of 130 V was

achieved at the speed of 6000 rpm, decreased by 24 %. Oppositely, during this process, the current increased from 184 µA to 276 µA, increased by 50 %. Afterwards, both the voltage and current shown an increasing trend with the spincoating speed ranging from 6000 to 7000 rpm. And the voltage increased from 130 V to 190 V, increased by 46.2 %. At the same time, the current increased from 276 µA to 375 µA by 35.5 %. Finally, both the voltage and current decreased with the spin-coating speed. To further investigate this relationship, these generators were used to charge a 1 µF capacitor. Figure 7b plots the stored charges by these devices in a single-operation cycle and the corresponding surface charge density generated by contact-electrification effect. Similarly, both these two parameters increased firstly with the spin-coating speed, and a maximum value of 641 nC charge with a surface charge density of 196  $\mu$ C/m<sup>2</sup> were observed at the speed of 7000 rpm. Then, both of them reduced with this speed.



Figure 7: (a) The peak output voltage and current as a function of spin coating speeds. (b) The stored charges and corresponding surface charge density under different spin coating speeds.

This phenomenon could be divided into three stages to analyze. As developed by previous theory of contact-mode TENG, the output voltage is determined by [13]:

$$V = -\frac{Q}{S\varepsilon_0} \left( \frac{d}{\varepsilon_1} + x(t) \right) + \frac{\sigma x(t)}{\varepsilon_0}$$
(1)

Where -Q is the transferred charges between two electrodes, S and  $\sigma$  represent the contact area of material and the accumulated charges on its surface, respectively, while  $\varepsilon_0$ and  $\varepsilon_1$  are the permittivity of vacuum and the relative permittivity of PDMS, and x(t) is the separation distance between two plates. And from this equation, we can conclude that the output voltage decreases with the reducing of thickness of PDMS film and increases with the surface charge density. Meanwhile, the transferred charges in short-circuit state is defined by:

$$Q_{SC} = \frac{S\sigma x(t)}{d_{\ell_{\varepsilon_1} + x(t)}}$$
(2)

Meanwhile, the short-circuit current  $I_{SC}$  is the differential of  $Q_{SC}$ . Therefore,  $I_{SC}$  is proportional to  $\sigma$ , while it increases with d monotonously. Additionally, the increase of spin-coating speed leads to the reduce of the thickness and larger effective contact area of PDMS film at the same time. And the surface charge density is greatly influenced and enhanced by the increase of effective contact area. Consequently, the output voltage decreased with the spin-coating speed firstly, and then increased with it at the

balanced effects of reducing thickness of PDMS film and increasing of surface charge density. Meanwhile, the  $I_{SC}$  increased with this speed under the cooperating of these two effects.

#### DEMONSTRATION

As a demonstration, the serial TENGs was put around an adult's arm to harvest its muscle motion energy (Figure 8). During its contraction and expand process, the generators could produce peak voltage of 10 V at a relative low frequency (1.1 Hz) and 20 V at a higher frequency (2.5 Hz).



Figure 8: The serial TENGs mounted on an adult's arm and the output voltage under different motion frequencies.

#### CONCLUSIONS

In summary, a novel wrinkle structure formation process induced by single-step C<sub>4</sub>F<sub>8</sub> plasma treatment is proposed to fabricate flexible TENG in this work. It could deposit fluorocarbon polymer on the PDMS film and enlarge the effective surface area at the same time. Additionally, this wrinkle structure could be controlled by varying the spincoating speed of PDMS film, resulting in smaller scale structure and larger aspect-ratio as the increase of this speed. And the performance of TENG is optimized at a speed of 7000 rpm. A 225 V peak voltage and 375 µA maximum current is achieved under finger typing, and a maximum surface charge density of 196  $\mu$ C/m<sup>2</sup> was observed. Facilitated by its flexibility and high-output ability, this TENG was successfully put on an adult's arm to scavenge its mechanical energy into electricity. This work provides an optimizing method for enhancing the performance of TENG.

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