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A multi-mode triboelectric nanogenerator for energy harvesting and biomedical monitoring

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ABSTRACT

In the field of exercise physiology, it is of great significance to monitor human body motion status and physiological functions for assessing physical quality and training load. However, the wearable electronics as the mainstream monitoring solution have power consumption and bulk-size issues that limit sustainable operation. Here, we present a multi-mode stretchable and wearable triboelectric nanogenerator (msw-TENG) for biomechanical energy harvesting and physiological functions sensing. The msw-TENG fabricated by a liquid metal and silicone achieves stretchable and highly conductive characteristics at the same time, and realizes conformal contact with skin. The msw-TENG mainly includes contact separation/stretch/press modes, which can be randomly transformed according to the actual applications. The device converts the biomechanical energy of limb movement into electrical energy for directly lighting up commercial LEDs. Additionally, the radial artery pulse signal, joint bending angle and limb stability can be detected in real-time. As a multifunctional biomedical active sensor that is exempt from needing an extra power source, the proposed msw-TENG holds great potentials in the future of exercise monitoring and rehabilitation therapy.

1. Introduction

With the increase of people's health awareness and fitness requirements, wearable electronics have attracted great attention due to their applications in steps, calories, heart rate, breathing and blood pressure monitoring [1–4]. Accurate and comfortable monitoring of physiological signals in real-time is essential for assessing physical quality and training extent [5,6]. Nevertheless, conventional devices have critical demerits such as rigid structures, bulk-size and limited functionality. The emergence of flexible electronic materials is critically needed for promoting changes in this field, which have both flexibility and electronic properties. The devices based on flexible electronic materials should have the characteristics of maintaining the original electronic structure and function in the process of bending, stretching, and torsion. Owing to unremitting efforts of intelligent manufacturing, materials and electronic engineering communities, flexible electronics

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experienced a rapid progress in terms of miniaturization, light weight, stability and environmental friendliness [7–10]. However, the heavy reliance on batteries limits the service life of these electronics and therefore constrains the implementation of real-time or continuous physiological signal monitoring. Therefore, it is critical to realize the self-powered operation of flexible electronics.

Currently, researchers have shown an increased interest in converting mechanical energy into electrical energy via the nanogenerator [11–15], which has advantages of low cost, high energy conversion efficiency, structural diversity and environmental friendliness [16–19]. Furthermore, many research groups have reported flexible nanogenerators with good biocompatibility as wearable/implantable electronics for harvesting biomechanical energy from joint movement [20–22], respiration [23–25], heartbeat [26] and so on. Simultaneously, physiological information about the movement status of the body can also be obtained from the electrical signals generated by nanogenerators [27–30]. This means that the nanogenerator can not only be used as a power source but also directly employed for active sensors.

The emergence of nanogenerators provides the possibility to establish self-powered wearable electric systems [31–33]. The challenge for wearable flexible nanogenerators is the lack of high-performance elastic conductive electrodes in the application scenario of body stretching. Conventional method mostly introduces rigid particles into the elastomer matrix to increase the electrical properties of the material. Because the inherent rigidity of conductive particles does not match the flexibility of the elastic matrix, it is easy to cause the internal stress concentration of the material [34–36]. The result is to increase the stiffness of the entire system, reduce the ductility, and limit the durability of the material in the mechanical response [37]. Moreover, during the stretching process, the conductive particles will be further dispersed, and the conductivity of the electrode will be significantly reduced. There are also research teams that use physiological saline ion solutions as electrodes for fabricating flexible and stretchable nanogenerator[38]. Nevertheless, the relatively low conductivity of ionic solutions limits the electrical performance of the nanogenerator.

In this work, we report a multi-mode stretchable and wearable triboelectric nanogenerator (msw-TENG) based on liquid metal and silicone material. The silicone is employed as an encapsulation and triboelectric layer. The liquid metal EGaIn (Eutectic Gallium-indium alloy) is the stretchable electrode, which has excellent physical and chemical properties including high electrical conductivity, low viscosity, deformability, non-toxic and harmless. Moreover, the construction of the island-bridge structure of the liquid metal based on 3D printing technology further enriches the working modes of the msw-TENG. The output performance of the msw-TENG for harvesting biomechanical energy was measured under contact separation mode. The stretch and press modes of the msw-TENG have demonstrated to accurately monitor the degree of joint bending, pulse signals and limb stability for assessing the body state. As a new energy-harvesting and self-powered sensing device, the developed msw-TENG has versatile applications in wearable electronics for assessing physical quality and training extent.

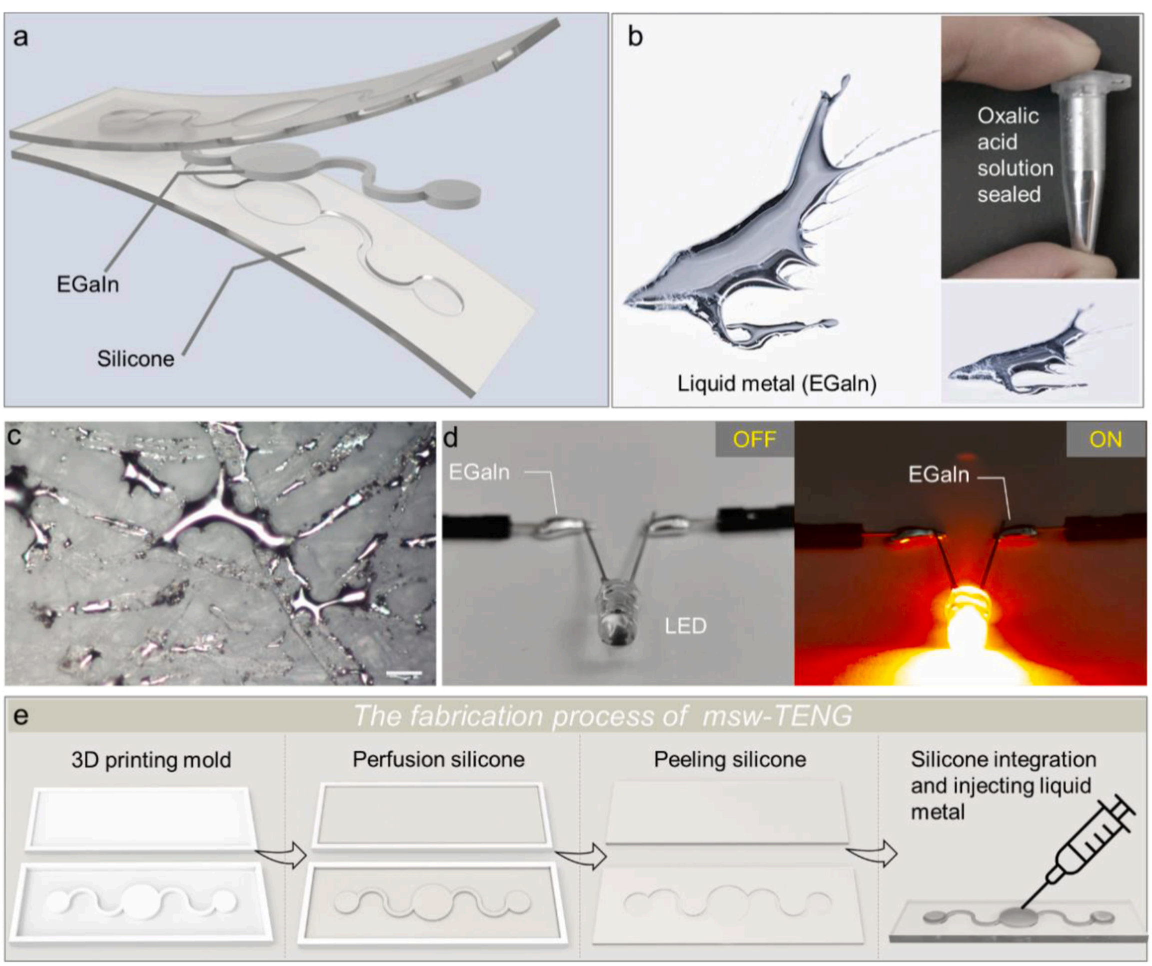

Fig. 1. (a) Schematic diagram of the msw-TENG. (b) Photographs of the liquid metal and sealed by Oxalic acid solution. (c) Microstructure of the liquid metal acquired by metallographic microscope (Scale bar = 20 μm). (d) Photographs showing the conductivity of the liquid metal. (e) The fabrication process of the msw-TENG.

2. Results and discussion

2.1. Structure and preparation of the msw-TENG

The msw-TENG consists of liquid metal (EGaIn) and silicone (Ecoflex 00–10). The liquid metal unit is used as a conductor electrode, which is encapsulated by the silicone unit to form a sealed sandwich structure (Fig. 1a). In addition, to achieve good tensile performance and pressing sensitivity, the liquid metal is distributed in the shape of island bridges based on 3D printing technology. The liquid metal that can flow at room temperature is sealed by oxalic acid solution (Fig. 1b). The fluidity ensures the continuity of the conductive network during the deformation process. The metallographic microscope shows that the liquid metal has a good continuity after any scraping (Fig. 1c). When the liquid metal is

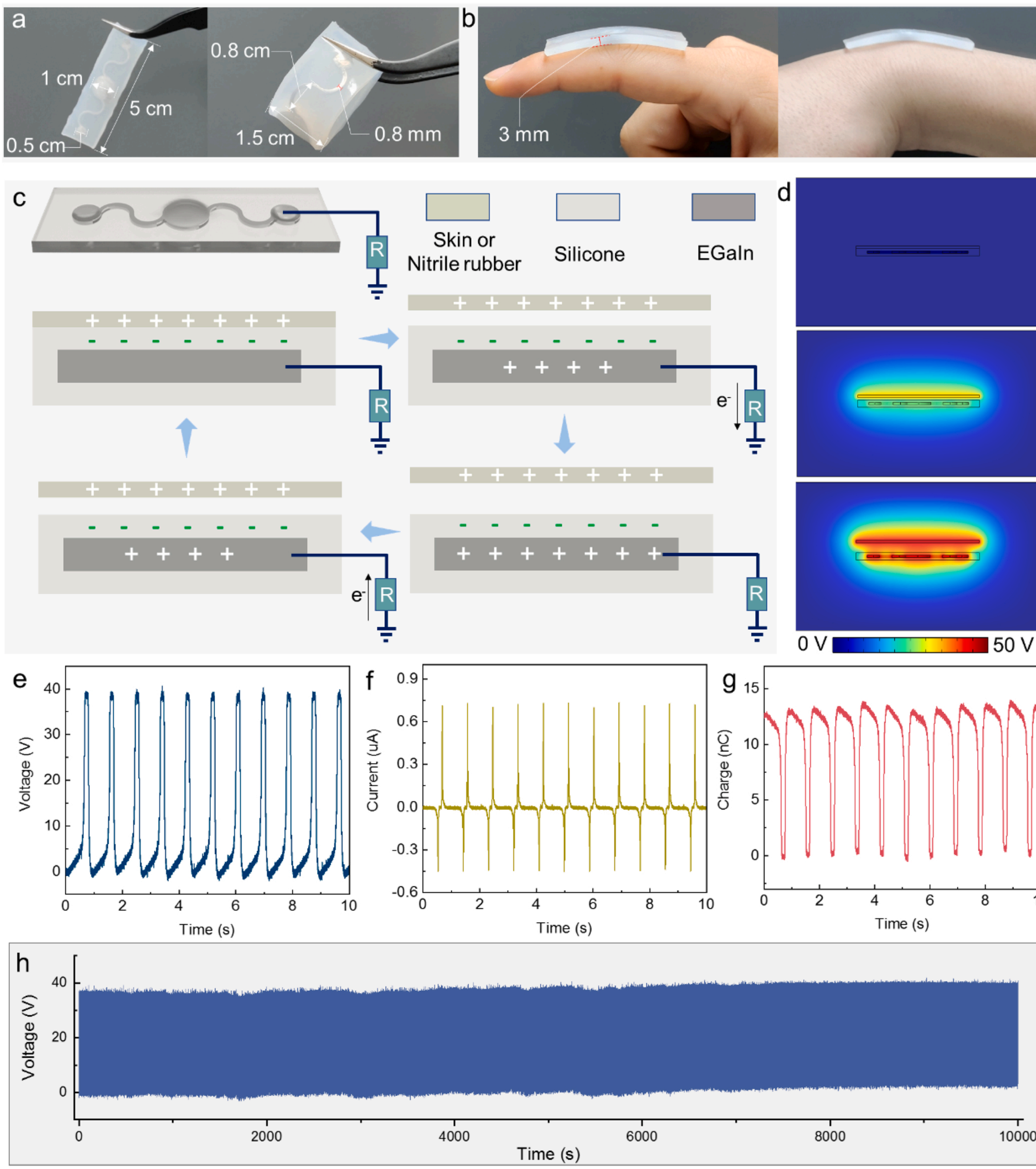

Fig. 2. (a) Photographs of the msw-TENG. (b) The msw-TENG attached to the finger and wrist showing the compliance performance. (c) The energy harvesting mechanism of the msw-TENG. (d) Simulation schematics of the msw-TENG on energy harvesting mode. (e) V_{oc}, (f) I_{sc} and (g) Q_{sc} of the msw-TENG. (h) Durability test of the msw-TENG for ~10,000 working cycles (at 1 Hz).

employed as a conductor for connecting with a red LED, the LED can be lighted by external power supply, showing good electrical conductivity (Fig. 1d). Fig. 1e displays the fabrication process of the msw-TENG. The main steps of processing include 3D printing mold, perfusion silicone, peeling silicone film and injecting liquid metal. See the experimental section for detailed methods.

2.2. Contact separation mode of the msw-TENG for energy harvesting

Mechanical elasticity and flexibility of the final assembled msw-TENG is shows in Fig. 2a and b, with a total dimension of 5 cm × 1.5 cm × 3 mm. The thickness is controlled to be pliable enough to closely fit the surfaces of the skin for showing good compliance performance. In areas containing liquid metal, the diameter of the big circle in the middle is 1 cm, which is twice the diameter of the small circles on both sides. The three circular structures are connected by four semicircles with a diameter of 0.8 cm to form an island-bridge structure. The thickness of the liquid metal area is 1 mm. Fig. 2c exhibits the operating mechanism of the msw-TENG on contact separation mode. In detail, when skin/nitrile rubber is in contact with silicone surface, the same amount of positive and negative charges is generated on the surface of the two materials based on triboelectrification effect. At this point, no electronics flow in the external circuit because of complete balance between the charges. Once the skin/nitrile rubber separates from silicone, electrons flow from the liquid metal to the enameled wire producing positive charges to compensate for the negative charge on the surface of the silicone that results generating current signal. When skin/nitrile rubber approaches silicone, the electrons flowing in the opposite direction (forming a reverse current) leading to the positive charges induced in the enameled wire decrease. Periodic contact and separation

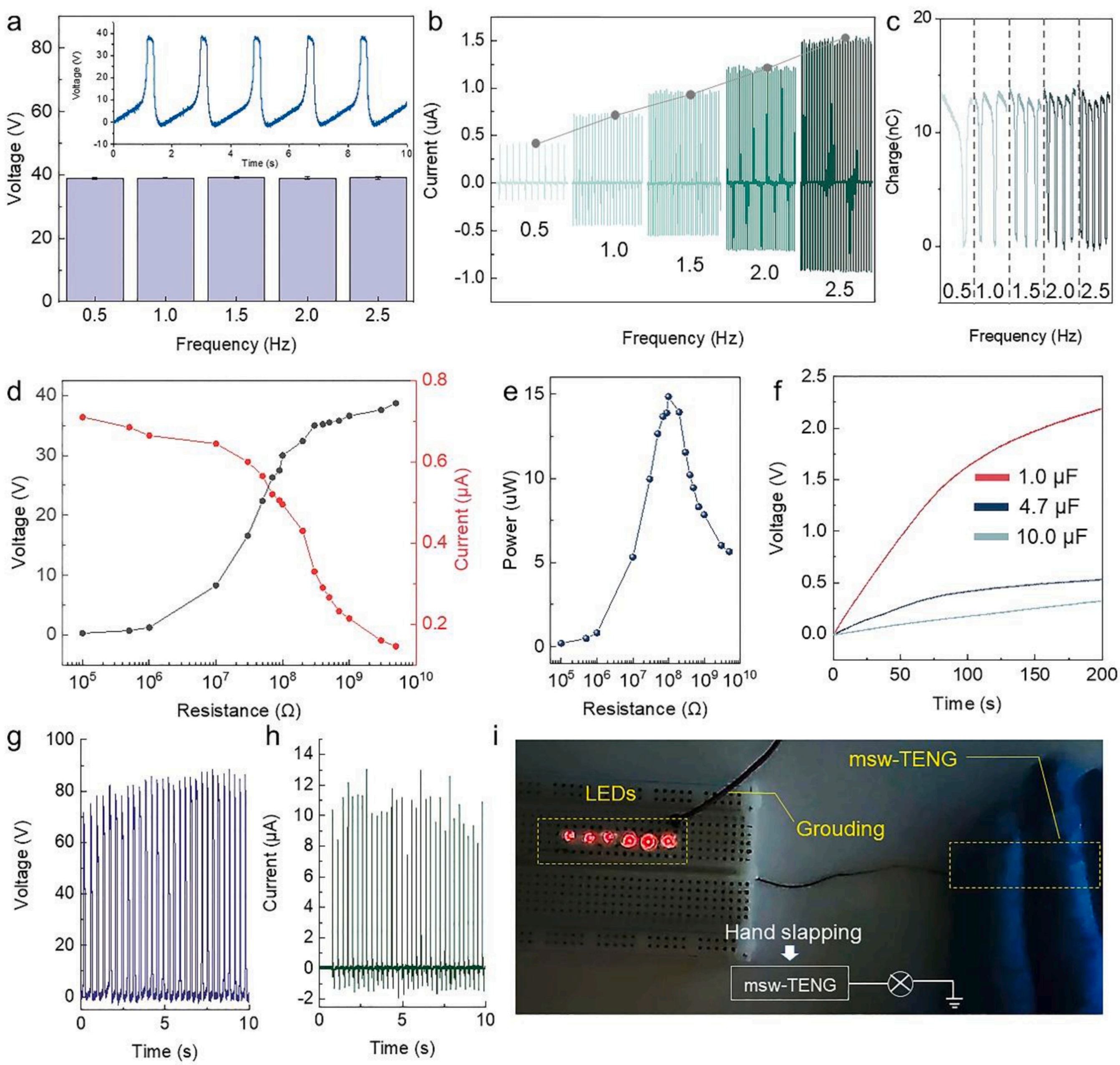

Fig. 3. (a) V_{oc}, (b) I_{sc} and (c) Q_{sc} of the msw-TENG under different working frequency (0.5 Hz, 1.0 Hz, 1.5 Hz, 2.0 Hz and 2.5 Hz). (d) V_{oc}, I_{sc} and (e) Q_{sc} of the msw-TENG when connected with different external load resistance. (f) The charging voltage curves of different capacitors (1.0 μF, 4.7 μF and 10 μF) by the msw-TENG. (g) V_{oc} and I_{sc} of the msw-TENG when wear nitrile rubber gloves to tap the surface of the device. (i) Photograph of the msw-TENG lighting red LEDs.

between the skin/nitrile rubber and the nanogenerator will generate continuous alternating current. The potential distribution of the msw-TENG under the contact separation mode is simulated by COMSOL software (Fig. 2d). Fig. 2e-g show the electrical output performance of the msw-TENG. The Open-circuit voltage (V_{oc}) and short-circuit current (I_{sc}) are about 39.0 V and 0.7 μA, respectively. The short-circuit charge (Q_{sc}) reaches 13 nC. As shown in Fig. 2h and Fig. S1, it demonstrates the V_{oc} stability of the msw-TENG. The V_{oc} does not drop significantly after 10,000 working cycles, which illustrates that the msw-TENG has excellent durability for practical application.

We also measure the electrical output performance of the msw-TENG under different frequency (0.5 Hz, 1.0 Hz, 1.5 Hz, 2.0 Hz and 2.5 Hz). As shown in Fig. 3a and c, it demonstrated that V_{oc} and Q_{sc} remain relatively stable with increasing frequency. In contrast, peak value of the I_{sc} is positively correlated with frequency. When the frequency is 2.5 Hz, the current can reach ~1.5 μA. The electrical output characteristics of the msw-TENG connected with different external load resistance are shown in Fig. 3d and e. The maximum output power is about 15 μW. It also indicates the typical internal resistance of the msw-TENG is ~100 MΩ. To demonstrate the performance of the msw-TENG as energy

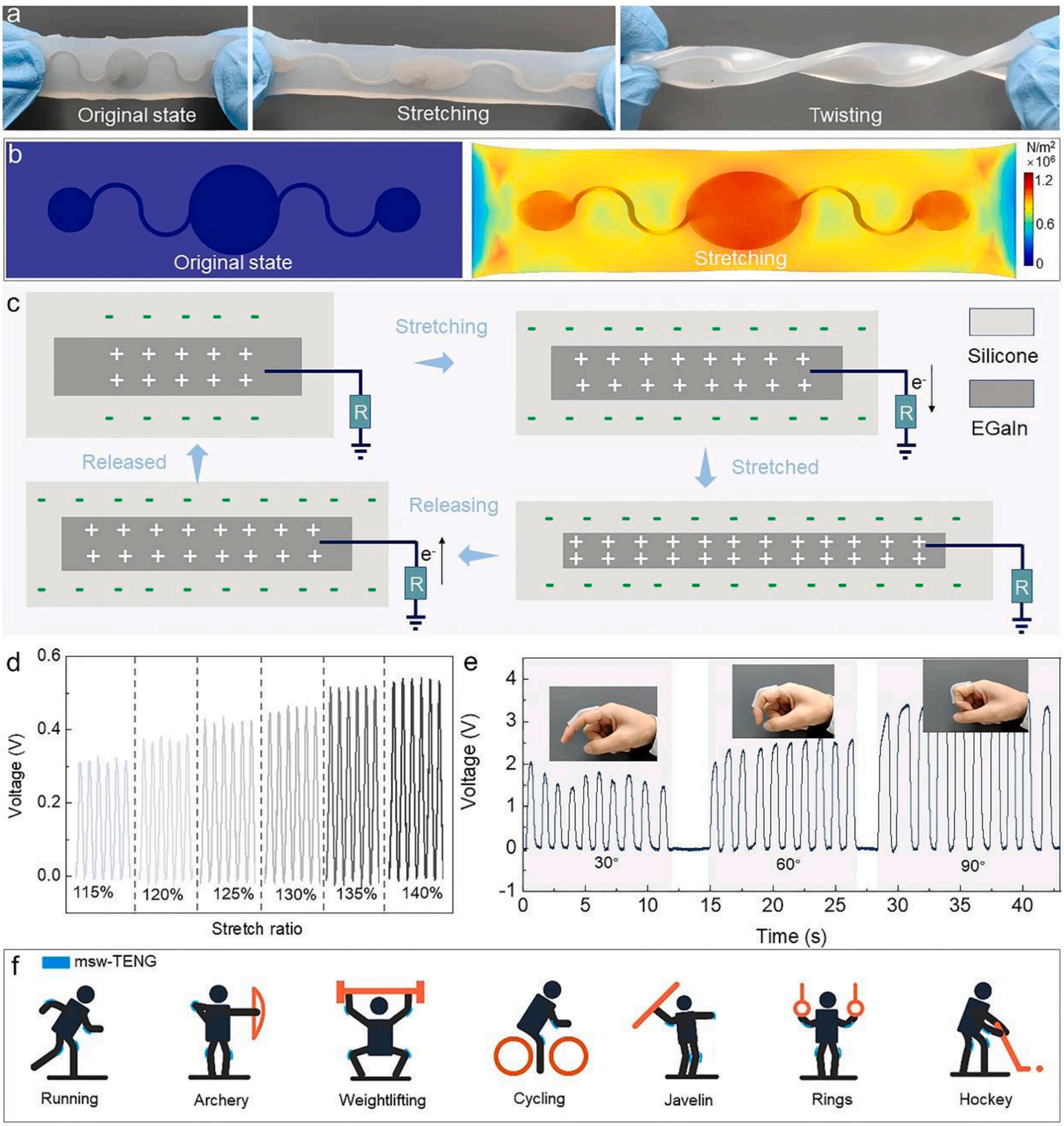

Fig. 4. (a) Original, stretching and twisting states of the msw-TENG. (b) Simulation schematics of the msw-TENG on stretched mode. (c) The working mechanism of the msw-TENG on stretched mode. (d) V_{oc} of the msw-TENG at different strain levels. (e) Output signal of the msw-TENG under motion state of the finger at different bending angles. The insets are the pictures of the msw-TENG attached on the finger. (f) Potential application scenarios for joint monitoring based on the msw-TENG.

harvesting devices, the msw-TENG is employed for charging different capacitor with a rectifier bridge. A 1 μF capacitor can be charged above 2 V within 200 s. At the same time, the maximum value of the charging voltage decreases as the capacitance value increases. When wear nitrile rubber gloves to tap the surface of the device, V_{oc} and I_{sc} of the msw-TENG are ~80 V and 12 μA (Fig. 3g and h), which can directly light up LEDs (Fig. 3i and Video S1). Once the skin of the hand is in contact with the surface of the device, the output of the msw-TENG will decrease due to changes in the triboelectric layer material (Fig. S2).

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2.3. Stretch mode of the msw-TENG for human joint motion detection

Excellently combining high electrical conductivity with high mechanical flexibility is crucial for stretchable materials in practical application including human joint motion. The room temperature flowable liquid metal integrated with flexible stretchable silicone material ensures the continuity of the conductive network during the deformation process. Fig. 4a exhibits flexibility of the msw-TENG under various status of original, stretching and twisting. During the stretching process, the liquid metal maintains good connectivity and outstanding stretchability (~300%) (Fig. S3). The stress distribution of the msw-TENG under stretched mode is simulated by COMSOL software (Fig. 4b). As shown in Fig. S4, the stress-strain curve of the msw-TENG is tested. And interestingly enough, the electrical signal appears along with the stretching of the device because of the coupling effect of frictional electrification and electrostatic induction. The working mechanism of the msw-TENG on stretched mode is shown in Fig. 4c. In the original state, no electronics flow in the external circuit because of complete balance between the charges. When an external tensile force is applied, the liquid metal and silicon are stretched at the same time, and contact area that related to external forces were changed. This means further friction between the two materials. Surface charges transfer then take place at the contact area due to triboelectrification effect. The electrons flow from the liquid metal to the enameled wire producing positive charges to compensate for the negative charge on the surface of the silicone that results generating current signal. Once the external tensile force is released, the electrons flowing in the opposite direction leading to the positive charges induced in the enameled wire decrease. Periodic stretch and release will generate continuous alternating current. Additionally, the V_{oc}, I_{sc} and Q_{sc} increase with the increasing of the stretching length of the msw-TENG (Figs. 4d and S5), which may be attributed to the increase in the effective contact area between liquid metal and silicone. Effective monitoring of the degree of bending of human joints is of great significance for assessing joint flexibility and postoperative rehabilitation. Due to its good flexibility and material biocompatibility, the msw-TENG can be well attached to the joint skin to realize joint movement monitoring. The output signal of V_{oc} for the msw-TENG with different bending angle of the finger is shown in Fig. 4e. With increasing the bending angle of the finger from 30° to 90°, the corresponding V_{oc} of msw-TENG also increases. In the process of using the msw-TENG to monitor the bending degree of the finger, we find that the output value is much greater than the electrical output when simply stretched, which is due to the coupling of stretch mode and contact separation mode. Certainly, through the change of size, the monitoring function of the msw-TENG can also be extended to other application scenarios in the field of sports training (Fig. 4f).

2.4. Pressed mode of the msw-TENG for radial artery pulse and stability detection

The msw-TENG achieves an ultra-sensitive response to small presses due to the flexibility of the silicone material and the ingenious design of the island-bridge structure. When the center of the msw-TENG is pressed by an external force, the silicone is deformed. The deformed silicone squeezes the liquid metal to both sides through narrow channels, and the liquid metal on both sides can easily push up the silicone to make it bulge (Fig. 5a and Video S2). Fig. S6 displays V_{oc} of the msw-TENG changes with pressure in the press mode. The enlarged view of the liquid metal area on both sides before and after applying pressure is shown in Fig. 5b. The above process has caused a significant change in the effective contact area between the liquid metal and the silicone, which builds the basic structure of the triboelectric nanogenerator based on a coupling effect of triboelectrification and electrostatic induction. The working mechanism of the msw-TENG on pressed mode is shown in Fig. 5d. The detailed process of charge transfer is similar to the stretched mode, which means periodic press and release will generate continuous alternating current.

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The msw-TENG as a wearable active sensor for detecting arterial pulse has significance for human medial health information. The msw-TENG can be deformed stably and effectively respond to vascular movement due to the excellent flexibility of the silicone material and the design of the island bridge structure of liquid metal. Fig. 5e exhibits that radial artery pulse signals detected by the msw-TENG under different physiological conditions (at rest, after exercise and after break) in real time. The V_{oc} of the msw-TENG driven by the radial artery pulse has increased from 0.10 V at rest to 0.32 V after exercise. By analyzing the time interval between two adjacent peaks, the heart rate at rest and after exercise is ~72 bmp (beats per minute) and ~120 bmp, respectively. Rest for 15 min after exercise (after the break) and use the msw-TENG again to monitor the radial artery pulse signal, the radial artery pulse generated a V_{oc} of 0.23 V and ~84 bmp. The pulse signal clearly shows the characteristic peaks of peripheral artery waveforms including the pulse pressure (P_1) and late systolic augmentation (P_2), which contain important biomedical and physiological information for diagnosing cardiovascular disease. The radial artery augmentation index (AIx) defined as P_2/P_1 that are strongly related to arterial stiffness. The peak value of radial artery pulse waveform reflects the intensity of the heartbeat. The schematic diagram of radial artery pulse signals detected by the msw-TENG is shown in Fig. 5f. After resting, the pulse wave signal intensity gradually returns to the resting state (Fig. 5g). The recovery speed of heart rate/peak value of pulse wave can indicate the body's acute response and adaptation to external load stimuli, the degree of fatigue and the ability to recover, which is also of great significance in the assessment of athletes' physical fitness. In addition, we can also test the stability of the limbs by touching the surface of the device with the fingers floating, similar to assessing the degree of tremor in Parkinson's patients (Fig. 5h). These signals obtained from the msw-TENG are dominated by tiny biomechanical movements, indicating that the pressed mode of the msw-TENG has a good application in physiological signal detection in biomedical field.

3. Conclusion

In summary, we have demonstrated a novel multifunctional triboelectric nanogenerator for energy harvesting and biomedical monitoring based on liquid metal and silicone materials, which exhibited outstanding stretchability (~300%) and conductivity. As an energy harvester, a power of 15 μW for the msw-TENG was achieved by contact separation mode. As a self-powered sensor, the degree of joint bending could be estimated by the stretch mode, meanwhile, the heart rate and pulse intensity before and after exercise were accurately detected by the press mode. In addition, three different working modes of the device could be randomly transformed according to the actual applications. With robust self-powered characteristics, the proposed msw-TENG exempts the necessity of battery. Furthermore, the good biocompatibility of constituent materials and encapsulation strategy guarantee that contact with the skin does not cause inflammation. This msw-TENG with excellent biomechanical energy harvesting capacity and biomedical

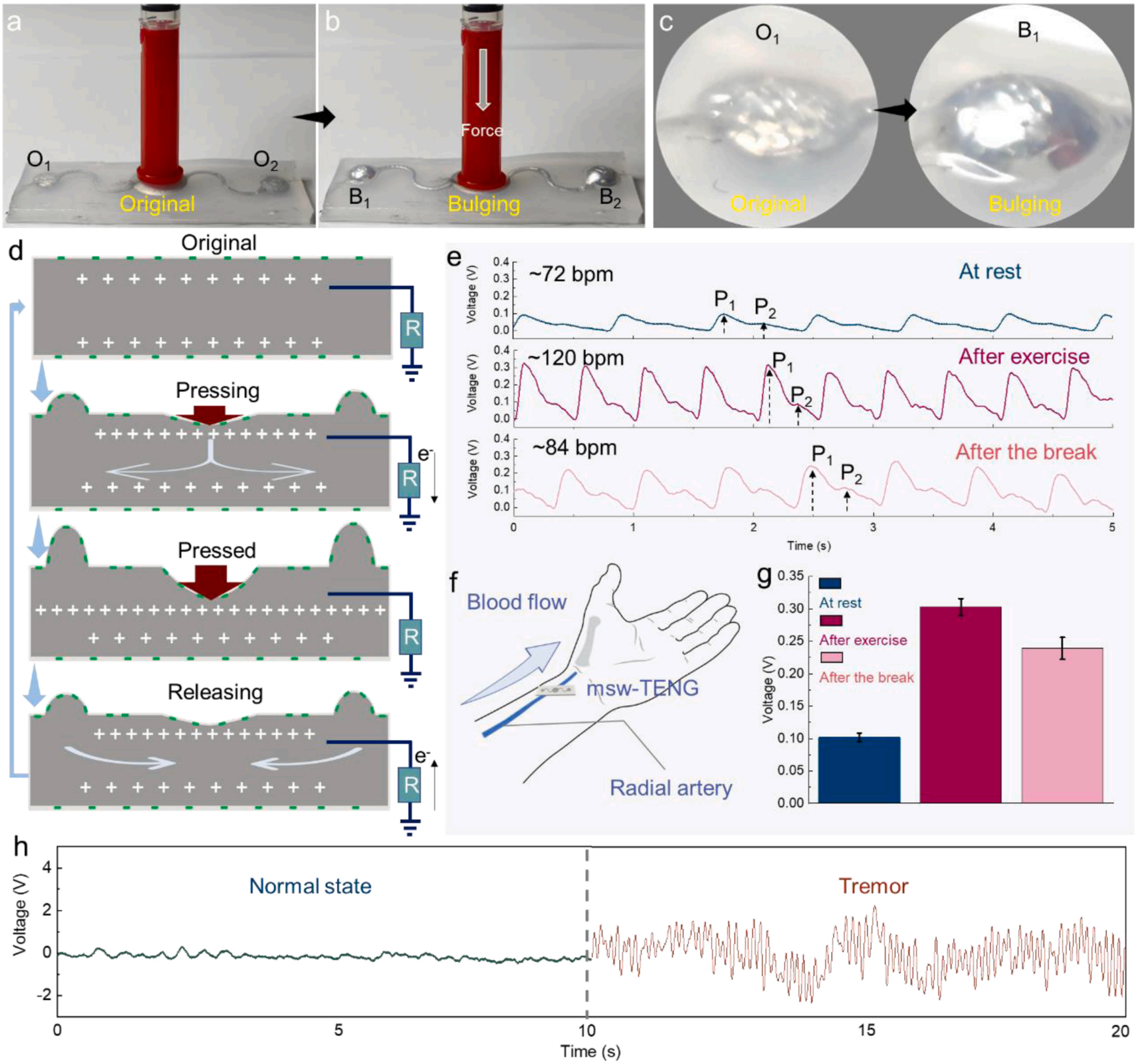

Fig. 5. (a) Photograph of the msw-TENG before pressing at original state. (b) Photograph of the msw-TENG after pressing at bulging state. (c) The enlarged view of the liquid metal area on both sides before and after applying pressure. (d) The working mechanism of the msw-TENG on pressed mode. (e) Output signal of the msw-TENG for monitoring radial artery pulse signals in different motion states, showing different heart rates. (f) Schematic diagram of radial artery pulse signals detected by the msw-TENG. (g) Statistic analysis of peak voltage value of the msw-TENG for monitoring radial artery pulse signals in different motion states. (h) V_{oc} of the msw-TENG in response to the stability of a finger.

monitoring properties has potential applications in wearable electronics, rehabilitation therapy and exercise monitoring.

4. Experimental section

4.1. Fabrication of the msw-TENG

Gallium (99.99% purity) and indium (99.995% purity) were purchased from Shanxi Zhaofeng Gallium Co., Ltd. The EGaIn was prepared by stirring gallium and indium together at 180 °C for 2 h, then sealed and stored at room temperature. Before use, EGaIn usually was washed by sodium hydroxide solution to remove oxides of gallium and indium. We obtained the molds by 3D printing and mixed Ecoflex 00–10 AB glue in 1:1 ratio. Then, the mixture was vacuumed to remove bubbles and injected into the molds. The mixture in the molds was placed in an oven at 30 °C for 2 h to obtain silicone films. Two different structures of silicone film were adhered together by the mixture. Finally, a syringe was employed for injecting liquid metal into the silicone cavity and an enameled wire was attached to the liquid metal for electrical connection.

4.2. Characterization and measurements

A Keithley 6517 electrometer was used for measuring the open-circuit voltage, short-circuit current and short-circuit charge of the msw-TENG, and the data were recorded and collected by an oscilloscope (LeCroy, HDO6104). A linear motor (LinMot E1100) was used to control the msw-TENG working under different frequencies by adjusting the displacement, velocity, acceleration. A metallurgical microscope (Nikon LV100ND) was used to observe the microstructure of liquid metal.

CRediT authorship contribution statement

Y. W., Z. Liu and Z. Li guided the project. Z. Liu, Y. W. and Y. Li conceived idea and designed the experiments. Z. Liu, Y. W. and L. W. fabricated the device. Y. Li, W. R. and Y. Z. accomplished the materials characterization. J. X., Y. G., L. X. and Z. Liu completed COMSOL stimulation and 3D drawing. Z. Liu, Y. W., L. W., X. Q., Y. Liu and Y. Z. performed the electrical characterization. Z. Liu, Y. W., Y. Li., G. X. and Z. Li analyzed the results. Z. Liu, Y. W., Y. Li and Z. Li wrote the manuscript. All authors discussed and reviewed the manuscript.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.nanoen.2021.106715.

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