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Review

Soft triboelectric nanogenerators for mechanical energy scavenging and self-powered sensors

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ABSTRACT

Efficient scavenging and converting energy from surrounding environment to achieve the self-powered operation of electronics has attracted tremendous interest to meet the energy needs of the rapidly growing Internet of Things and portable devices. As a promising technology, the triboelectric nanogenerator (TENG) possesses the capability of efficient utilization of ambient mechanical energy for energy supply and self-powered sensing. By integrating the soft materials into TENG, the soft TENG can be triggered by a quite weak external mechanical stimulation and realize a better conversion of micro mechanical energy. Meanwhile, the deformable soft TENGs further expand the application scenarios of TENGs. Here, the advances of soft TENGs by focusing on the fundamentals, candidate materials, promising structures, applications in energy scavenging and self-powered sensors are reviewed. In particular, the wearable and implantable soft TENGs, through which the related self-powered sensing or biomechanical energy harvesting can be realized in *vivo* and in *vitro* respectively, are summarized. Finally, the unique advantages and possible focuses for scientists in future of soft TENGs are emphasized.

1. Introduction

With the recent continuous advancement of Internet of Things and portable electronics, realizing self-powered operation of the intelligent devices is a desirable alternative, which can avoid additional economic burden and environmental problems caused by regular replacement of power sources [1–3]. Efficient energy harvesting from living environment can be an ideal solution to substitute for conventional power technologies [4–14]. In particular, the mechanical energy as the most widely distributed clean energy ubiquitously existing in our daily life. Since 2012, the triboelectric nanogenerator (TENG) has been extensively studied as an emerging technology in the field of energy scavenging, which can realize the high-efficiency conversion of mechanical energy to electrical energy [15–21]. Using TENG, diversified forms of mechanical energy existed in ambient environment including wind [22–33], vibration [34–37], human body motion [38–45], raindrop [46–48], water wave [49–52] and rotating motion [53,54], can be harvested efficiently. Meanwhile, the TENG-based self-powered sensors with different functions have been proposed and widely applied to environmental sensing [55–59] and human motion monitoring [60–63].

Moreover, the utilization of soft materials will further improve the response of TENGs to micro mechanical stimulation [64–67], and expand the application scenarios of TENGs, such as wearable and stretchable electronics [68–72], implantable self-powered sensing and energy scavenging [73–79], and so forth. In general, compared with the case in the rigid material based TENGs, the soft materials based soft TENGs can realize the more efficient energy conversion through an ideal contact between the soft friction materials, and possess a better prospect in flexible sensing and energy supply of portable electronics. Over the years, the soft TENGs have attracted tremendous efforts and greatly advanced, especially in mechanical energy scavenging and self-powered sensors, as shown in Fig. 1.

Herein, the recent progress of soft TENGs are comprehensively summarized, meanwhile, the applicability of soft TENGs in various

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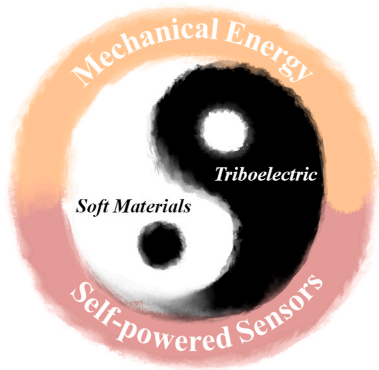

Fig. 1. By integrating the soft materials into triboelectric nanogenerator (TENG), the soft TENG has attracted tremendous efforts and been greatly advanced, especially in mechanical energy scavenging and self-powered sensors.

application scenarios is emphatically introduced. This review begins with the working principles and invention of soft TENGs. Thereafter, the promising soft materials, various structures with unique advantages and applications of soft TENGs in energy scavenging and self-powered sensors are systematically elaborated. Moreover, the soft TENG based hybridized nanogenerators and self-charging systems are also reviewed in detail. Particularly, it has been indicated in this paper, the wearable and implantable soft TENGs, which can be a significant part in both *vitro* and *vivo*, maximize their potential of applications. At last, the unique advantages of soft TENGs are concluded, and the possible challenges and promising research directions in the future are pointed out.

2. Soft triboelectric nanogenerator

2.1. Fundamentals and invention of soft TENGs

Contact electrification, also known as triboelectrification, was often ignored or even regarded as hazardous until the concept of TENG was proposed in 2012 [15]. In typical TENGs utilize the integration of electrostatic induction and triboelectric effect, an alternating current (AC) output could be generated by realizing the periodic change of distance or contact area between two friction layers with opposite electrostatic charges on the surface after contact electrification [80–84]. According to different structures and manners of operating, the working principle of TENGs are divided into four main categories [85,86], including lateral sliding mode, vertical contact-separation mode,

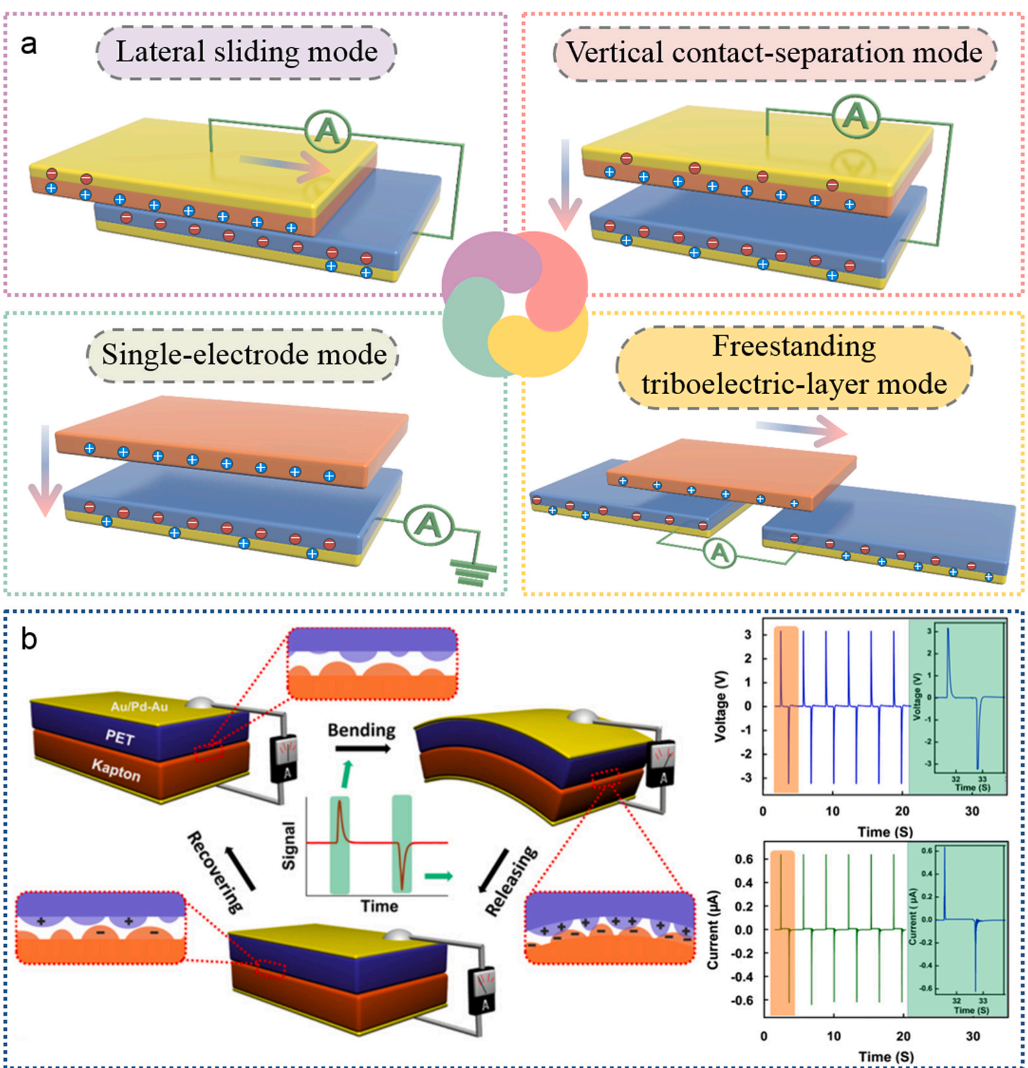

Fig. 2. Fundamentals and invention of soft TENGs. (**a**) The operation mechanism of four fundamental modes of TENGs, including lateral sliding mode, vertical contact-separation mode, single-electrode mode, freestanding triboelectric-layer mode. (**b**) Schematic diagram of the first soft TENGs.
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single-electrode mode and freestanding triboelectric-layer mode, as illustrated in Fig. 2a. As for the soft TENG, it can be also categorized into these four fundamental modes. In 2012, the first soft TENG, which is operated in the vertical contact-separation mode, was introduced by Fan et al. [15]. The Fig. 2b exhibits the structural diagram and working mechanism of the soft TENG. Soft polyethylene terephthalate (PET) and Kapton sheets are stacked together as a sandwiched structure, and their back sides are coated by a layer of Au-Pd alloy film as electrodes. Under the external force, the device will be bended, and two sheets will contact each other, and then the electrons transfer from PET surface to the Kapton surface due to their difference in triboelectric polarity. After the external force is removed, the positively charged and the negatively charged surfaces with equal charge are separated from each other, lead to a potential difference between the bottom and the top electrodes, thus, the electrons will flow through external circuit to balance the potential difference. As the two friction surfaces approach again, the potential difference between the electrodes will be weakened and then the electrons will be driven flow backward through the external circuit. Therefore, an AC output can be generated under the alternating potential caused by the periodic contact and separation between two thin sheets. An output current of 0.6 μA and output voltage of 3.3 V can be obtained during the process of bending and releasing of the soft TENG.

2.2. Material selection

The contact electrification effect almost exists in any friction process between any two different materials or even the same kind of materials [87]. Furthermore, many deformable (e.g., stretchable, folding and

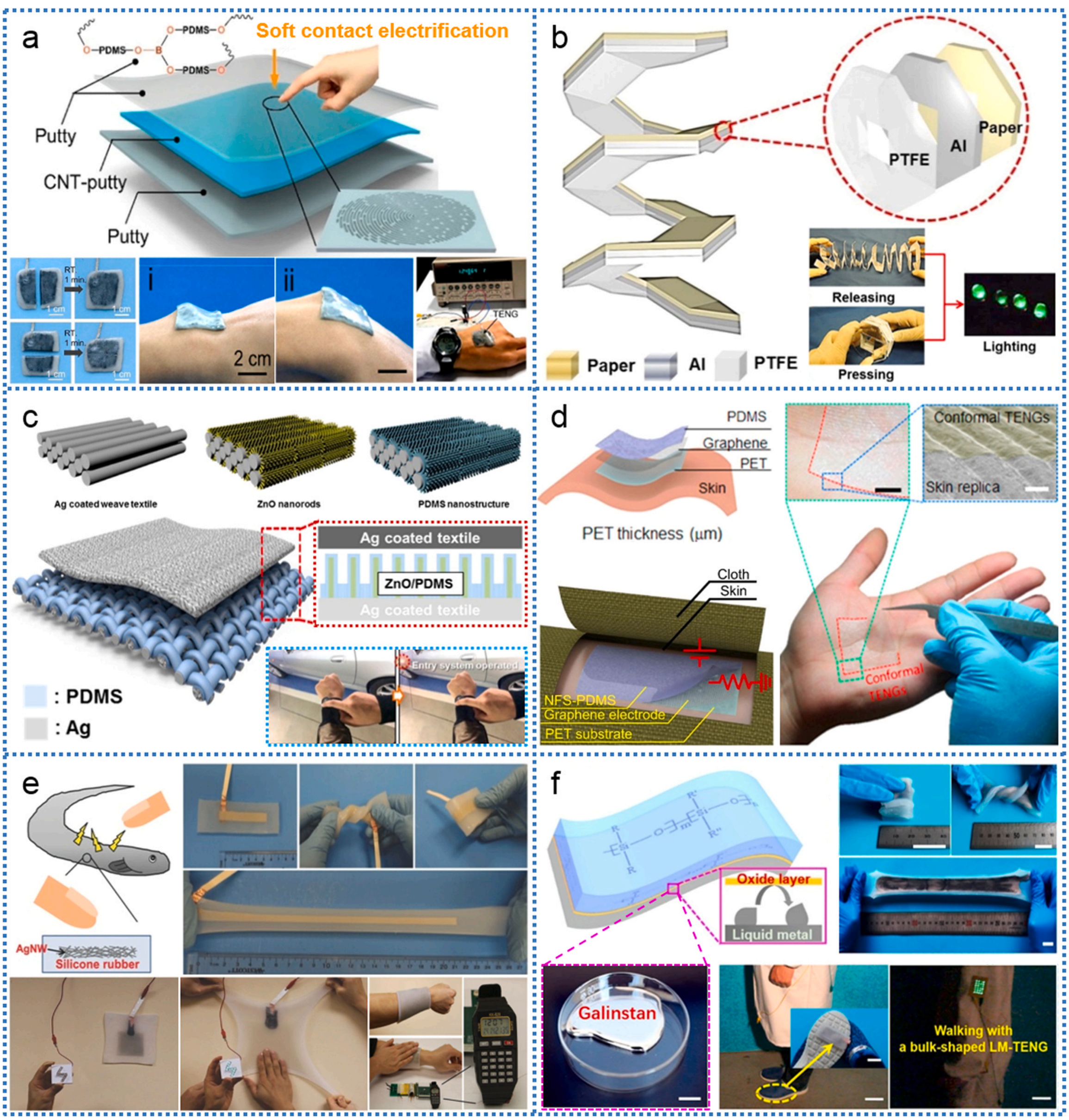

Fig. 3. Promising candidate materials of soft TENGs. (**a**) Schematic of viscoelastic-polymer-based soft TENG rely on soft contact electrification. (**b**) Schematic of paper-based soft TENG for self-powered pressure sensing. (**c**) Schematic of textile-based wearable soft TENG for realizing the remote control. (**d**) Schematic of graphene-based conformal soft TENG that can be attached on the human skin. (**e**) Schematic of silicone-rubber-based with high stretchability. (**f**) Schematic of liquid-metal-based soft TENG for harvesting the mechanical energy of human walking.

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flexible) soft materials with their own advantages, such as some soft polymers [88–91], papers [64,92], hydrogel [93,94], sponge [95,96], fabrics [97,98] and so forth, can be selected to fabricate various soft TENG. However, considering some specific requirements for soft TENGs, it is necessary to select appropriate soft materials to meet the needs of different application scenarios.

One of the advantages of using soft materials is that an ideal contact between materials can be realized, thus, lead to a more effective contact electrification process. A single-electrode mode soft TENG based on the soft contact electrification at the friction interface was reported by Chen et al. [99]. Fig. 3a shows the configuration of the soft TENG, which consists of viscoelastic polymer named putty as the triboelectric layer, while the putty matrix dispersed by carbon nanotubes (CNT-putty) as the electrode sandwiched by the triboelectric layer. Under the ideal contact at interface, contact area between the triboelectric layer and other objects is increased, thus, the larger output can be obtained. At room temperature, the soft TENG exhibit excellent self-healing ability, and the output voltage in original state (before cutting) is basically the same as that after cutting and self-healing. Moreover, because of the ultra-soft property of viscoelastic polymer, the soft TENG is shape-adaptive and can be adhered to various surfaces. Such a device can be well adapted to the surface of a moving wrist and can be applied as a soft power supply for driving an electronic watch by tapping the surface of putty film, the developed soft TENG has a great potential to be applied in soft electronics and robotics.

In addition, the paper is a collapsible, lightweight and low-cost material with outstanding ability to lose electrons, thus, it is a promising candidate choice for positive triboelectric materials [100]. As illustrated in Fig. 3b, a paper-based stacked soft TENG, which is operated in single-electrode mode and composed of paper (as the positive triboelectric material and the substrate), polytetrafluoroethylene film (PTFE, as the negative triboelectric material) and Al electrode, was introduced by Yang et al. [101]. With the application and release of external force, the paper and PTFE contact and separate periodically, providing an alternating electric output. The soft TENG, which is capable of harvesting the micro mechanical energy in stretching and twisting and realizing a peak power density of 0.14 W/m^2, can be used as a portable power source to drive small electronics. Moreover, since the electrical output of the fabricated soft TENG will increase with the increased external force applied, the self-powered pressure sensing can be also demonstrated.

It is well known that the textile, which is flexible and skin-friendly, is one of the most excellent wearable materials, has been widely utilized in wearable electronics [102]. In 2015, Seung et al. developed a soft TENG operated in vertical contact-separation mode, which consists of a layer of Ag-coated conductive textile and a layer of Polydimethylsiloxane (PDMS) triboelectric layer with the conductive textile as its back electrode [97]. As exhibited in Fig. 3c, the ZnO nanorod arrays film was grown on conductive fabric as the template for PDMS, and hence the nanoscale pattern was implemented onto the surface of triboelectric layer to improve the output performance of the textile based soft TENG. Furthermore, the total power output of the soft TENG can be further promoted through a multilayer-stacked design, and a remote control can be driven by the fabricated wearable soft TENG to control the vehicle entry system.

Additionally, the soft materials with ultrathin thickness can achieve perfect conformal contact on the skin, resulting from the enough static adhesion force can be formed at the interface [103]. Chu et al. proposed a conformal soft TENG, which can be attached on the human skin for harvesting the mechanical energy of human motions [104]. As shown in Fig. 3d, the single-electrode mode soft TENG is composed of a PDMS film with a thickness of 1.5 μm as the triboelectric layer, a PET substrate with a thickness of 0.9 μm and a bilayer graphene electrode. The electrical power can be generated by the repeated contact between PDMS and clothing or the human body and depends on the effective contact area. Thus, this wearable device can be utilized as a self-powered tactile sensor for detecting the number of touching fingers.

In addition, the silicone rubber with superior stretchability is widely considered as a competitive candidate material for stretchable electronics [105–111]. Lai et al. demonstrated a super-stretchable soft TENG in single-electrode mode, which has a silver nanowires (AgNWs) film with high conductivity and stretchability as the conductive layer and the silicone rubber as both the encapsulation and the triboelectric layer [112]. As displayed in Fig. 3e, the AgNWs film is sealed by the silicone rubber and the whole device shows a good stretchability. Due to the excellent mechanical reliability of the soft TENG, the device is capable of operating under intense stretching and can be used as a sustainable and portable power supply to drive wearable electronic devices. In addition to AgNWs, there are also some liquid conductive materials which can be used as flexible electrodes of soft TENG. Following the similar structure exhibited in Fig. 3e, Yang et al. used Galinstan, a kind of liquid metal, as the electrode of soft TENG, which can maintain a continuous conductivity under the large tensile strain of 300% [113]. As illustrated in Fig. 3f, the soft TENG is capable of operating stably under various deforms, and the short circuit current, open circuit voltage, and average power density can reach up to 15.6 μA, 354.5 V, and 8.43 mW/m^2, respectively. Moreover, this soft TENG can be attached to the sole of the shoe and drive several LEDs by converting the mechanical energy of human walking.

Although biomaterials have not been widely used in the field of glycosylation up to now, but due to their good biocompatibility and biodegradability, it has been reported that recombinant spider silk proteins have been used to construct soft TENG. This TENG was based on recombinant spider silk proteins (MaSp1). The conversion efficiency of mechanical energy into electrical energy reached 47.3%. Moreover, the high mechanical properties of spidroins allow a long operation of the device which can sustain at least 35,000 cycles.

2.3. Various structures of soft TENGs

Various soft TENGs with different structures and working modes have their own unique advantages. According to different application scenarios, the harvesting of various mechanical energy and the self-powered sensing of various environmental stimuli can be realized via the appropriate structural design. The vertical contact-separation mode soft TENGs, of which the output is proportional to the variation of the clearance between the two layer of friction materials, can be used as the self-powered pressure sensor. Ha et al. demonstrated a soft TENG operated in vertical contact-separation mode with the interlocked microridge structure [114]. As shown in Fig. 4a, the microridge structure on the surfaces of P(VDF-TrFE) [poly(vinylidenefluoride-co-trifluoroethylene)] film and PDMS film ensure the necessary working distance between the triboelectric layers instead of needing an additional spacer. Furthermore, due to that the PDMS film and P(VDF-TrFE) film are interlocked, the soft TENG is very sensitive to compressive and bending strains. As an outstanding multifunction self-powered sensor, it can sense the bending motion of fingers and the tiny pulse of the artery, and its bending and pressure sensitivity can reach up to 0.1 V/° and 0.55 V/kPa, respectively.

For TENG with single-electrode structure, because only one electrode is needed, there is no need to connect the two parts of friction materials, so the structure can be greatly simplified and can adapt to more forms of deformation. Chen et al. proposed a single-electrode mode soft TENG with three-dimensional configuration by 3D printing technique [115]. Fig. 4b depicts the configuration of the soft TENG, which is consisted of composite resin parts (part I and part II) as the triboelectric materials and the ionic hydrogel is selected as the electrode. Meanwhile, the whole device is ultra-flexible, a compressive deformation will occur under the external stress, and the surfaces two triboelectric layers will be contacted and charged. Then, with the release of external force due to elastic recovery of the whole structure, the two charged surfaces will separate and produce an electrical signal. In addition, the fabricated soft

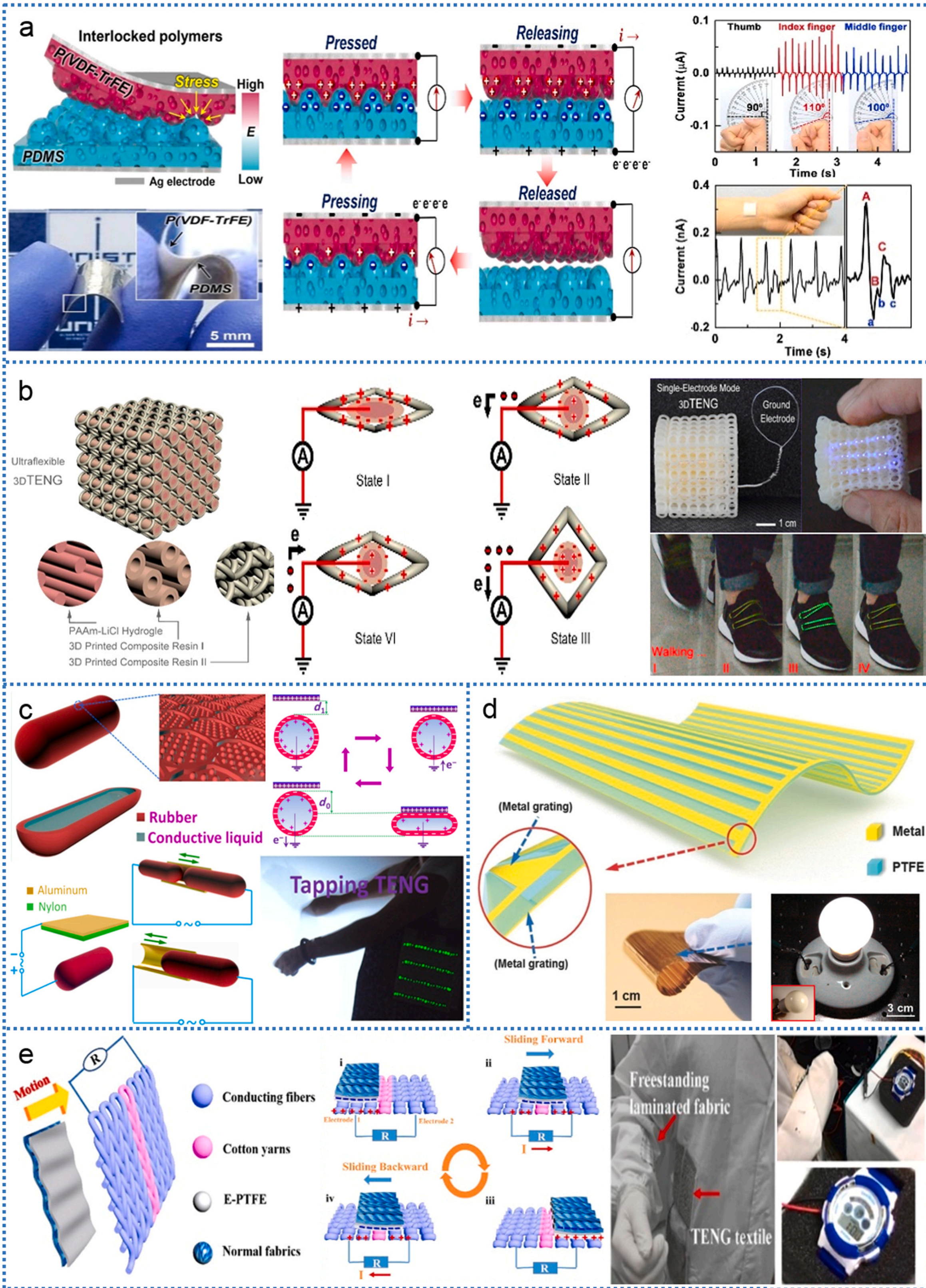

Fig. 4. Various structures of soft TENGs. (**a**) Schematic of vertical contact-separation mode soft TENG with the interlocked micro-ridge structure. (**b**) The configuration of a single-electrode mode soft TENG with three-dimensional structure. (**c**) The single-electrode mode soft TENG with package structure. (**d**) The micro-grating based soft TENG in lateral sliding mode. (**e**) The wearable textile-based soft TENG in freestanding triboelectric-layer mode.
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TENG can be integrated into the insole as a portable energy source for harvesting the biomechanical energy, and it will not affect the walking and compromise comfort because of its superior softness. In addition, Yi et al. developed a soft TENG operated in single-electrode mode with package structure by employing NaCl solution as the electrode and rubber with nanorods nanostructures on the surface as both the electrification layer and sealing layer, as illustrated in Fig. 4c [116]. By utilizing the liquid electrode, the soft TENG has unlimited deformation ability in theory. Based on the above fabricated unit, soft TENG can be further applied to three other working modes to achieve different functions. For example, in a single-electrode structure, it can be designed to be worn on the wrist like bracelet, which is capable of converting the energy of tapping motion into electrical energy and drive 80 LEDs to work.

Furthermore, Zhu et al. reported a micro-grating based soft TENG in lateral sliding mode, which can scavenge the mechanical energy during sliding motion between surfaces, and the energy conversion efficiency can reach up to 50% [117]. As depicted in Fig. 4d, the soft TENG consisting of a PTFE film with metal micro-gratings on either side. Thanks to the micro-grating design, when two layers of PTFE films with the above structure are rubbed, the alternating charge transfer with high frequency can be realized, thus, an enhanced output current can be obtained. Under a relative sliding velocity of 1 m/s, a globe light can be powered directly. Moreover, the soft TENG, which is shape-adaptive, can be affixed onto curved surfaces, and diversified forms of motions can be utilized such as relative rolling between the two cylindrical tubes.

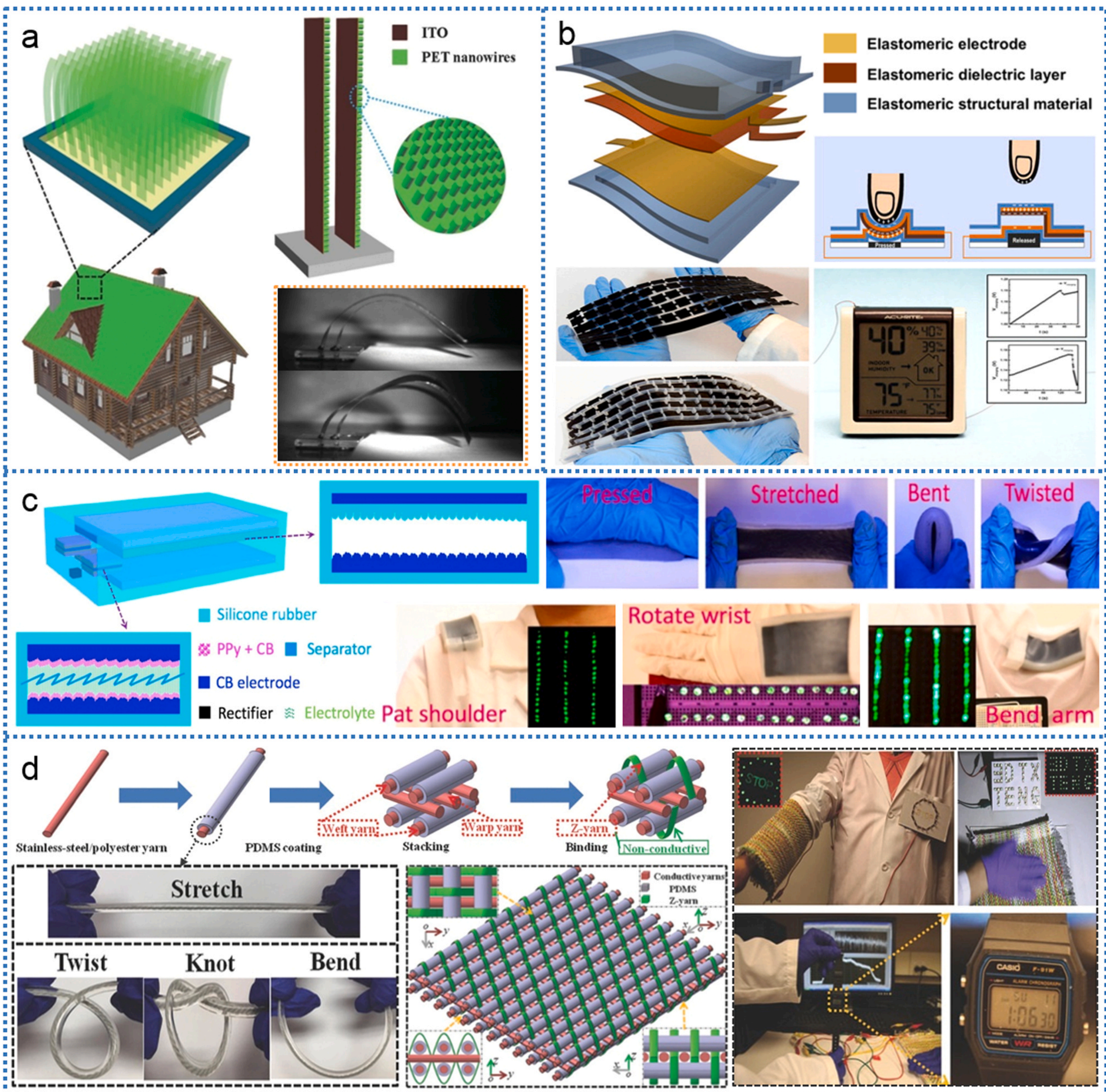

Fig. 5. Soft TENGs for scavenging mechanical energy. (a) The lawn structured soft TENG for scavenging wind energy. (b) The elastomer-based soft TENG for harvesting the tapping energy during typing. (c) The stretchable soft TENG for scavenging energy from various forms of deformation. (d) The textile-based soft TENG for converting mechanical energy from human motion.

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Additionally, for the structure of freestanding triboelectric-layer mode, in which the independent friction layer can move freely and does not need to connect with the other part of the TENG through wires, so it can scavenge multiple forms of mechanical energy more flexibly and conveniently. Huang et al. demonstrated a wearable soft TENG based on textile in freestanding triboelectric-layer mode, which can scavenge and convert the mechanical energy of the human motion [118]. Fig. 4e shows the structure diagram of the soft TENG, which is composed of two separate conductive fabrics (as the electrode and the triboelectric material) separated by cotton yarns and an expanded PTFE film (as the freestanding triboelectric material) attached to common fabric (as the substrate). After contact electrification, when the freestanding layer reciprocates on the surfaces of the two conductive fabrics, even without direct physical contact, the electrons will be driven and transfer alternately between the two electrodes. The freestanding layer and the separate electrode can be respectively fixed on the wrist and waist side of the clothes to scavenge the mechanical energy of various human motions (beating arm, walking and running, for instance). During swinging the arm, the soft TENG will operate and a capacitor can be charged, meanwhile, the stored energy can drive the watch sustainably.

3. Applications

3.1. Soft TENGs for scavenging mechanical energy

As a novel energy technology, soft TENGs possess the same advantages as traditional TENGs, such as the stable electrical output, variable structure, low cost and high energy conversion efficiency. In particular, due to the base material and overall structure are soft, the soft TENG can respond to a quite weak environmental stimulation, and can adapt to curved surfaces [63,99] or even moving surfaces to realize the scavenging of mechanical energy of various actions, such as stretching, torsion, bending and extrusion [115,119].

Wind energy is one of the most accessible and sustainable energy in nature, however, the traditional wind power equipment is difficult to be applied in our daily life, because of its large size and dangerousness [13]. Zhang et al. developed a soft TENG with lawn structured for scavenging wind energy, which can be installed on the rooftop [120]. As exhibited in Fig. 5a, the proposed soft TENG consists of a vertical array of free-standing polymer strip, which is made by a PET strip with indium tin oxide (ITO) electrode deposited on the back. Each polymer strip can swing freely in the wind, thus, when the wind passes, a contact-separation among strips will be realized and an AC output can be delivered. Due to that the strip is very flexible, the soft TENG can operate in the wind from arbitrary direction and scavenge the wind energy at the same time. Moreover, owing to surface modification of PET film, the open-circuit voltage and the short-circuit current of the soft TENG can reach up to 98 V and 16.3 μA, respectively. This system provides an ingenious way to harvest wind energy and can be a sustainable power supply for household electronic devices.

With the development of computer technology, in the work and our daily life, the human-computer interaction is indispensable, which is mainly realized by tapping the keyboard. In order to harvest the biomechanical energy used for typewriting, Li et al. successfully designed a soft TENG based on flexible elastomer materials, which can scavenge the tapping energy during typing [121]. As shown in Fig. 5b, in the soft TENG based on the coordinating of single-electrode mode and contact-separation mode, the structural layer will be charged after contact with the skin and the dielectric layer will contact the bottom electrode under a small pressure of fingers. When the finger is lifted, the skin will separate from the structural layer, the dielectric layer is separated from the bottom electrode, and then the current signal can be produced under the difference of potential between the bottom and top electrodes. Particularly, the dual-mode soft TENG can be attached to the keyboard as a keyboard membrane to effectively harvest the micro mechanical energy during the finger tapping. In a normal typing speed, by tapping one hour on the keyboard, a capacitor could be charged to 1.15 V and the stored electric energy can directly drive the thermometer and hygrometer.

As mentioned above, the soft TENG made of stretchable materials can withstand and scavenge energy from various forms of deformation. Yi et al. reported a soft TENG that can be subject to various deformations, meanwhile, these deformations can be converted into electricity [119]. As exhibited in Fig. 5c, the soft TENG consists of two triboelectric parts separated by air, including silicone rubber with electrode on backside and a compound electrode made of carbon black (CB) and silicone rubber. Due to the outstanding flexibility of the soft TENG, it can operate under various deformations (pressing, bending, stretching, and twisting, for example) without any physical damage. When the fabricated TENG is pressed, bent, stretched or twisted, the triboelectric layers will contact and separate repeatedly, and resulting in an AC electrical output. Furthermore, it can be attached to the curved and moving surfaces of the human body for scavenging the mechanical energy of tapping shoulder, wrist rotation the and arm bending. In addition, two series stretchable supercapacitors for storing electrical energy harvested by TENG can be encapsulated in silicone rubber together with the soft TENG to form a self-charging energy system, which can be used as a reliable power supply of wearable electronics.

Since the human body is an ever-present source of mechanical energy, and clothing is requisite for everyone, therefore, textile-based soft TENG is a promising strategy, which can harvest the neglectful energy of human motion at any time. Dong et al. developed a soft TENG with 3D orthogonal woven structure by using stainless steel/polyester conductive yarn as the electrode, PDMS coating as the triboelectric layer and non-conductive Z-yarn woven in the thickness direction for fixing the whole structure [122]. Here, the conductive yarn covered by PDMS triboelectric layer acts as an energy-harvesting unit that can withstand a variety of mechanical deformation, as depicted in Fig. 5d. In virtue of the adoption of 3D braiding scheme with two layers of energy collection unit, which is different from the simple cross strategy in the traditional 2D plane braiding model, the manufactured TENG can achieve higher output power, and the power density can reach up to 263 mW/m^2 can be obtained. Furthermore, the soft TENG could be applied as a wearable textile to scavenge human motion energy and driving portable electronics.

3.2. Soft TENGs hybridize with other types of generators

During past years, tremendous research efforts aim to boost the energy harvesting efficiency of nanogenerators have been dedicated, and great progress has been achieved, in which the hybridized nanogenerator is one of the most optimum choice [123–129]. Compared with the nanogenerators based on a single effect, by integrating multiple effects in one device, the hybridized nanogenerators is desirable to realize a larger electric power [128,129]. As for hybridized nanogenerator, the way to achieve the more efficient harvesting of mechanical energy is to convert the energy to the maximum extent by combining multiple correlation effects [2]. As we know, the mechanical energy can be harvested based on triboelectric effect, magnetoelectric effect and piezoelectric effect. Moreover, it has been proved that the soft TENG is an outstanding platform to hybridize with other types of generators for scavenging mechanical energy.

By using the coupling of electromagnetic effect and contact electrification effect, an enhanced electrical output can be generated under a same mechanical stimulation. Zhang et al. introduced a mechanically flexible hybridized triboelectric-electromagnetic nanogenerator for harvesting the mechanical energy in stretching [130]. Fig. 6a shows the schematic diagram of the soft hybridized nanogenerator, including the TENG part that consists of triboelectric layers (FEP film and PDMS film) and electrodes (conductive glass fabric and ITO), and the electromagnetic generator (EMG) part consisting of a Cu coil and a NdFeB magnet. With the application and release of the tensile force, the upper and lower

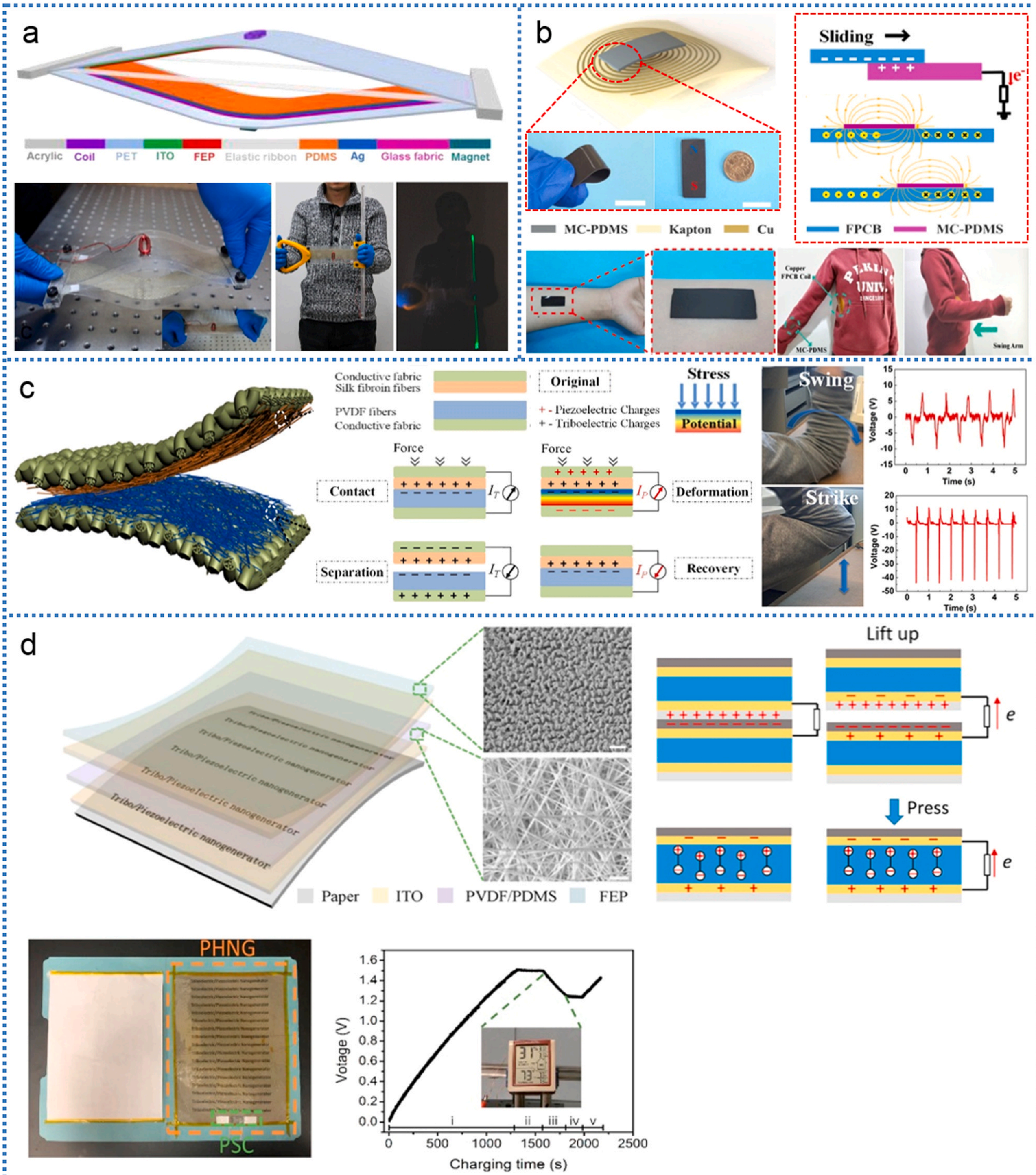

Fig. 6. Soft TENG hybridized with other types of generators. (a) The stretchable hybridized electromagnetic-triboelectric nanogenerator for scavenging the mechanical energy in stretching. (b) The soft hybridized electromagnetic-triboelectric nanogenerator for harvesting mechanical energy in sliding motion. (c) The wearable fiber based hybridized piezoelectric-enhanced triboelectric nanogenerator. (d) The paper-based transparent piezoelectric-triboelectric hybridized nanogenerator.

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layers of hybridized nanogenerator will contact and separate continuously, meanwhile, the TENG part and the EMG part will convert the applied mechanical energy into electrical output simultaneously based on their corresponding working principles, therefore, in this case, the mechanical energy can be harvested more efficiently. Furthermore, thanks to the flexibility of the whole device, the hybridized nanogenerator can be integrated into a bus handgrip to scavenge the biomechanical energy it's pulled by someone. Recently, by combining of soft TENG and EMG, Wan et al. reported soft hybridized nanogenerator, which can harvest mechanical energy in sliding motion [131]. As exhibited in Fig. 6b, the hybridized nanogenerator consists of two parts: a magnetic and conductive MC-PDMS film made of PDMS, NdFeB particles and multi-walled carbon nanotubes, and a Kapton film with copper coil inside. When the Kapton film and MC-PDMS film slide relatively, the TENG part operated in single-electrode mode, meanwhile, and the current can be generated in the copper coil owing to the change of magnetic field. Due to the different output characteristics, the output voltage of EMG part is limited but the output current is larger, while the output voltage of TENG part is large but the output current is limited, these two components can complement each other to achieve a faster charging speed than that of the single component. Moreover, the soft MC-PDMS membrane can be fixed on clothes or attached to human skin as a wearable energy device to harvest the sliding mechanical energy of human motion.

By combining the triboelectric effect and piezoelectric effect, Guo et al. developed a hybridized nanogenerator based on textile, in which the enhanced output resulting from the positive interaction of the two effects can be obtained [132]. Fig. 6c exhibits the structure and working mechanism of the hybridized nanogenerator, by selecting polyvinylidene fluoride (PVDF) fibers with piezoelectric effect after polarization treatment as triboelectric material, the hybridized nanogenerator incorporates a vertical contact-separation mode soft TENG and a typical piezoelectric nanogenerator. Particularly, by choosing an appropriate polarization direction for PVDF, the current outputs generated by triboelectric effect and piezoelectric effect will have the same direction, thus, the cooperative work can be realized. Because of the outstanding flexibility of the overall structure, the soft hybridized nanogenerator can be perfectly combined with ordinary clothes. For example, it can be attached to human elbows to scavenge the mechanical energy of body movement and identify different movements. Based on the similar principle, He et al. demonstrated a transparent soft piezoelectric-triboelectric hybridized nanogenerator [92]. As depicted in Fig. 6d, an energy harvesting unit is based on the paper substrate and has different functional layers, including a negative triboelectric layer (FEP film), positive triboelectric layer (paper) and a piezoelectric layer (PVDF-PDMS nanocomposite film) sandwiched by the top and bottom ITO electrodes. The soft TENG part is operated in contact separation mode, when the papers were flipped, one of the fabricated nanocomposite paper contacts with another paper, the paper will be positively charged while the surface of FEP will be negatively charged. Meanwhile, the piezoelectric part can be triggered when the paper squeezes each other under the external force. In the synergic working process, maximum power density can reach up to 286.5 mW/m^2. Additionally, a paper-based self-charging system was realized by

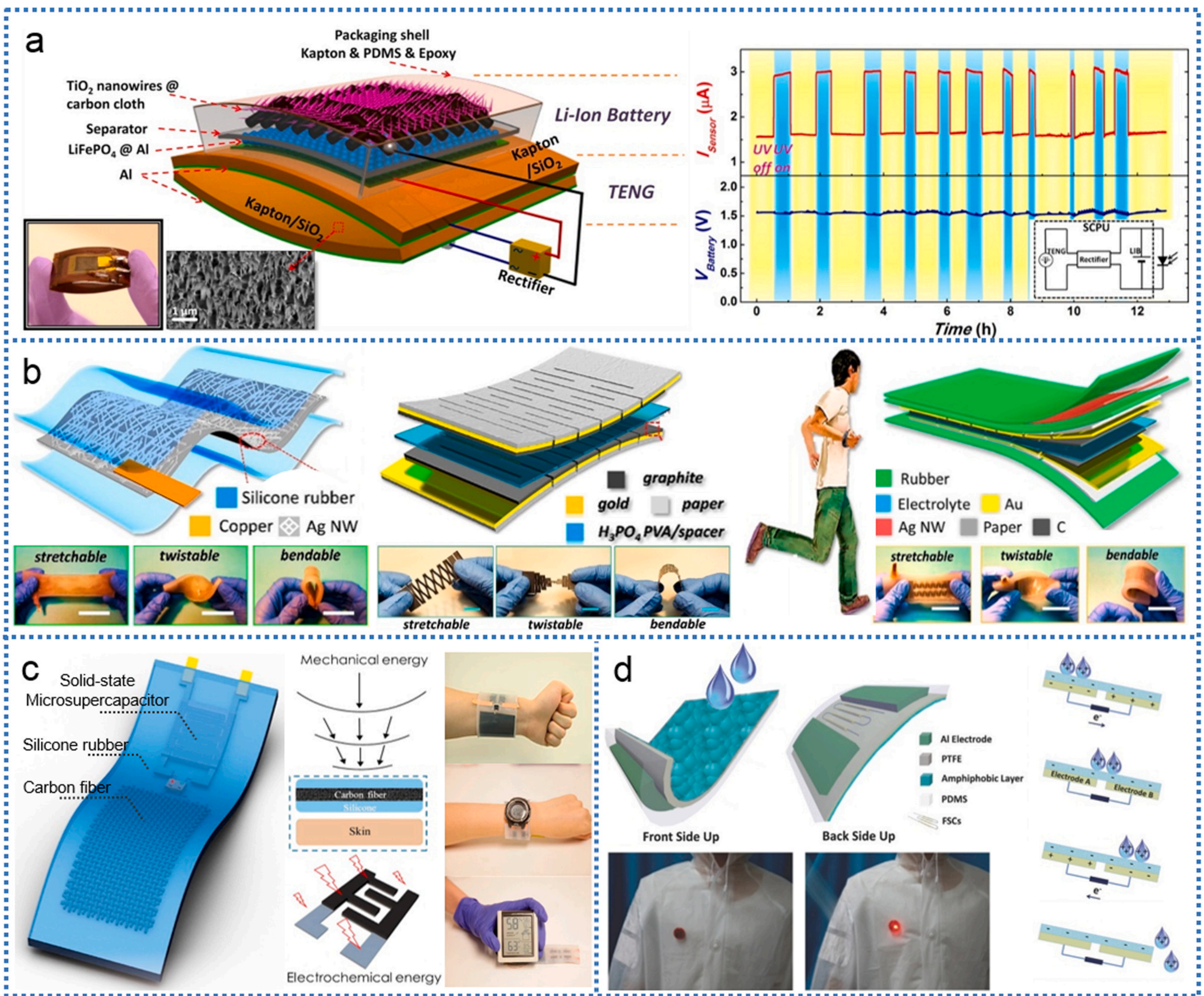

Fig. 7. Soft TENGs for self-charging systems. (a) The self-charging system based on arch shaped soft TENG and flexible lithium-ion battery. (b) The flexible self-charging system based on soft TENG and graphite-paper-based supercapacitor. (c) The wearable self-charging power unit with integral package structure. (d) The self-charging power system based on the soft hydraulic TENG and fiber supercapacitor for scavenging water energy.
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combining paper-based hybridized nanogenerator and paper-based supercapacitors, which can be used as a sustainable power supply for temperature-humidity sensor. Due to its lightweight, flexible and transparent structure, the reported soft device can harvest mechanical energy without affecting the reading, which has a good prospect in practical application.

3.3. Soft TENGs for self-charging power systems

As an outstanding sustainable energy source for portable electronic devices, the self-charging power systems based on soft TENG and various soft energy storage components with excellent flexibility have attracted great scientific interest [133]. While the environmental mechanical energy is harvesting by the soft TENG, the generated electrical energy can be converted into chemical energy simultaneously, which can be stored in the soft batteries or soft supercapacitors.

In 2013, based on the integration of arch shaped soft TENG and flexible lithium-ion battery, the flexible self-charging power system was introduced by Wang et al. for the first time [134]. As illustrated in Fig. 7a, the vaulted soft TENG is operated in vertical contact-separation mode, which is composed of two Kapton films with Al electrodes on the back side. Meanwhile, the flexible lithium-ion battery with the top surface of the TENG as substrate is composed of the composite cathode made of $LiFePO_4$, active carbon and binder, the TiO_2 nanowires grown on the soft carbon cloth as the anode, and the polyethylene (PE) film for separating. As the external vibration is applied to the soft TENG, the upper and lower parts of the arched TENG are continuously contacted and separated to realize an AC output, and then the generated electric energy can be stored in the top soft battery after rectification. In particular, the self-charging power system can simultaneously scavenge energy and provide external power. In this case, electrochemical reactions related to charging and discharging occur concurrently on the electrodes of the battery. When the soft self-charging power system is working under a sustaining deformation, the produced direct current (DC) output can maintain a relatively stable value within 12 h and continuously power the UV sensor.

Furthermore, the flexible supercapacitor is another ideal energy storage device which can be integrated with soft TENG to demonstrate the self-charging energy systems. Guo et al. fabricated a flexible self-charging package by a graphite-paper-based supercapacitor with capacitance of 1 mF/cm^2 and a single-electrode mode soft TENG [135]. The energy harvesting unit is made of the AgNWs film electrode embed in silicone triboelectric layer, while the energy storage unit is based on a kirigami structure, as described in Fig. 7b. Both two functional units can withstand complex mechanical deformation due to their proper material selection and structural design, thus, the entire self-charging power system that built by encapsulating the two functional units is stretchable, twistable and bendable. The proposed power package can scavenge the energy generated in human movement under various mechanical deformation, moreover, it can be fixed on the human body continuously supply power for the wearable electronics. In addition, by using the MXene-based supercapacitor with a larger capacitance reach up to 23 mF/cm^2 as the energy storage unit, Jiang et al. developed another self-charging power system with integral package structure [136]. As depicted in Fig. 7c, the soft TENG part consists of the carbon fiber electrode and the silicone triboelectric layer, meanwhile, the solid-state supercapacitor with interdigital structure fabricated by choosing $Ti_3C_2O_2$ as the active material and polyvinyl alcohol (PVA)/H_3PO_4 gel as the electrolyte. Subsequently, these two components are connected by a rectifier and sealed together by silicone to form the wearable self-charging system. The flexible system, which can be worn on the wrist, will contact the skin repeatedly while moving, causing the soft TENG module to work and charge the supercapacitor module. The developed power system can successfully drive the thermometer and electronic meter to work and is a reliable energy source for wearable devices.

In addition to biomechanical energy, some micro mechanical energy in surrounding environment, such as the energy of raindrop falling, can also be harvested and stored by the self-charging energy system based on soft TENG. By integrating the soft hydraulic TENG and fiber supercapacitor, Zhang et al. developed a self-charging power system to scavenge water energy to continuously drive electronics [48]. As illustrated in Fig. 7d, the whole system is based on soft TENG, which can harvest the mechanical energy of water droplets. Meanwhile, the soft TENG is encapsulated by PDMS together with the fiber supercapacitors on the back side, thus, the water penetration can be avoided. When the raindrop flow on the surface of the hydrophobic layer, a unbalanced potential between the back electrodes can be caused, which will drive the electrons to transfer back and forth through the external circuit, and in the meantime the fiber supercapacitors on the back can be charged after rectification. Furthermore, the system can be further designed as a raincoat, which can convert the mechanical energy of water like raindrops to light the LED sustainably.

3.4. Soft TENGs for self-powered sensors

As mentioned above, electrical output can be generated by TENG under external mechanical stimulation, and consequently the scavenging of mechanical energy is realized. On the other hand, the electrical signal induced by external stimulation contain the information of these stimuli such as magnitude, frequency and so forth. Therefore, the electrical signal generated by TENG can be directly used to monitor the changes of ambient environment and sense the external mechanical stimulation, without any other power supply, and finally realize the self-powered sensing [137]. In particular, the self-powered sensors based on soft TENGs has higher sensitivity and wider application scenarios, compared with other TENG based self-powered sensor systems using rigid materials.

In 2013, Yang et al. reported a soft TENG based self-powered tactile sensor operated in single-electrode mode [63]. As shown in Fig. 8a, in a typical working process, when the finger contacts and separates from the PDMS (triboelectric layer), a pulse electrical signal will be generated, through which the occurrence of touch can be reflected. Moreover, with the enhancement of applied pressure, the contact area between skin and triboelectric layer will increase, so more electrostatic charges will be generated on the PDMS film surface after contact electrification, resulting in an increased output voltage. For the fabricated self-powered pressure sensor, the pressure detection sensitivity of each sensor unit in the displayed 4×4 tactile sensor arrays can reach up to 0.29 V/kPa. Because of the flexibility of the whole system, it can be attached to a curved surface, such as the surface of a tube, to sense the distribution of pressure applied by the hand.

However, due to the difference in triboelectric polarity of different materials, when the objects of different materials contact the surface of pressure sensor based on TENG operated in single-electrode mode, even under the same pressure, the amplitude and polarity of triboelectric charges on the sensor surface can be quite different, which will lead to a different electrical output, and ultimately affect the accuracy of the pressure sensor [138]. To solve this problem, Ren et al. adopted the design of coating a charge shielding layer on the top surface of the TENG operated in single-electrode mode based on the contact and separation between the inner materials, which can avoid the influence of the top electrostatic charge on the output performance of TENG [138]. As illustrated in Fig. 8b, the self-powered pressure sensor based on the soft TENG consists of a PDMS film as triboelectric layer and PDMS-CB electrode made of carbon black and PDMS, which are separated by the spacer. The shield film cover on top is highly conductive and grounding through external circuit, thus, electrostatic charge generated by the triboelectrification process between the external object and the top surface of the sensor can be shielded. Therefore, under the same pressure, there is almost no difference in the voltage output generated when the sensor is pressed by objects with different materials. Moreover, an

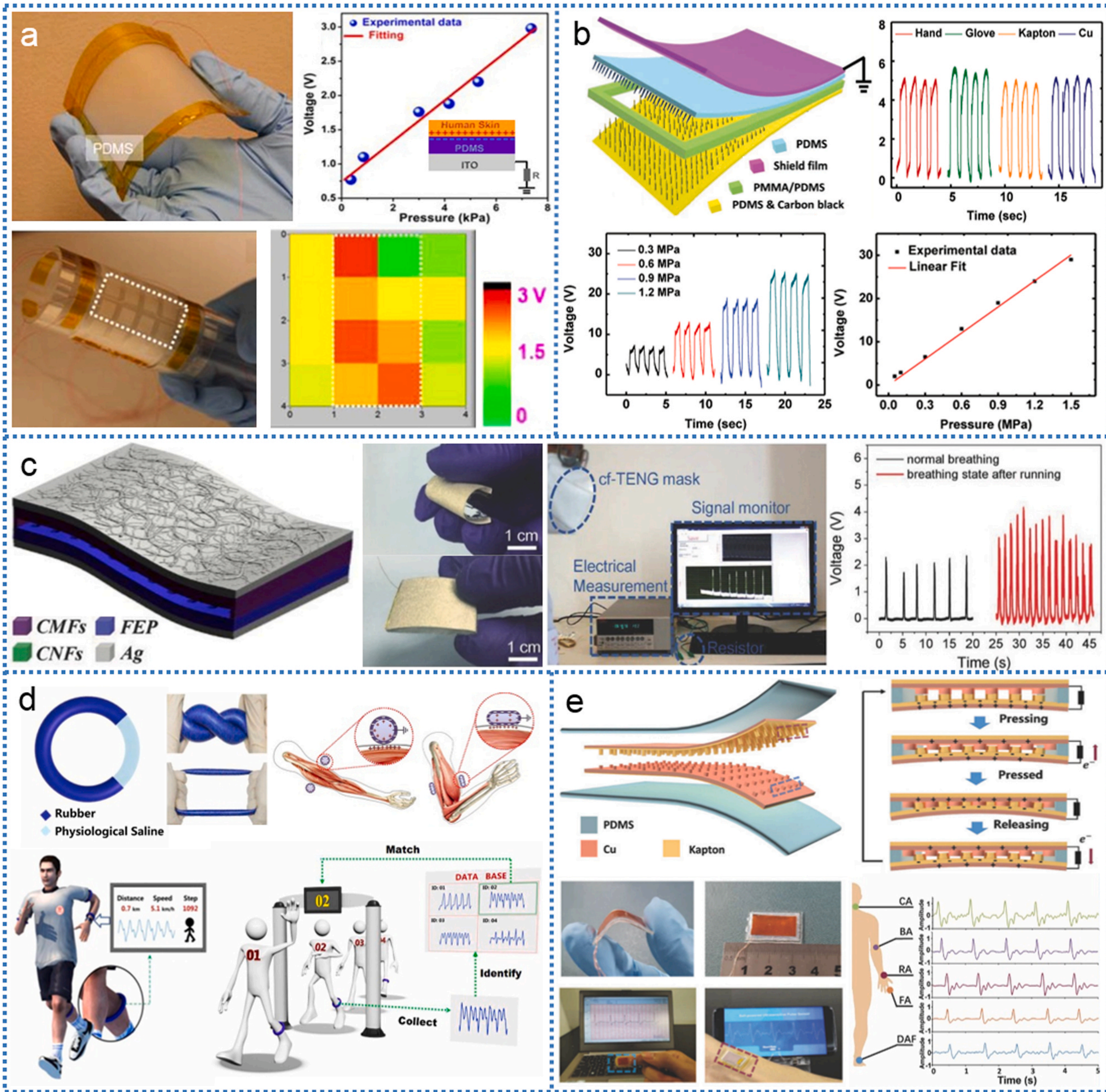

Fig. 8. Soft TENGs for self-powered sensors. (a) The flexible self-powered tactile and pressure sensor. (b) The self-powered pressure sensor with a charge shielding layer. (c) The antibacterial wearable sensor implanted into the mask for realizing self-powered breathing Monitoring [139]. (d) The wearable self-powered sensor for achieving the recognition of the identity. (e) The self-powered pulse sensor with ultra-high sensitivity for monitoring the heart rhythm.
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enhanced sensitivity of 51 kPa/V can be obtained by printing burr arrays on the inner surfaces of triboelectric layer and electrode.

In addition, the soft TENGs with outstanding flexibility and shape adaptability can be designed as various wearable self-powered sensors. He et al. reported an antibacterial soft TENG based on cellulose fibers for realizing self-powered breathing Monitoring [139]. Fig. 8c depicts the schematic diagram of the contact-separation mode TENG, the 2D cellulose microfibers/nanofibers composite paper and FEP film as the triboelectric material with micropores structure for removing $PM_{2.5}$ and allowing the air through smoothly, meanwhile, the Ag nanofibers electrode were deposited on their backside. Soft TENG with good flexibility can be implanted into the mask without reducing the comfort, and can be driven by the respiratory airflow and realize the monitoring of breathing. As a wearable self-powered sensor, the respiratory rate and respiratory intensity of the wearer before and after running can be well monitored at the same time. Han et al. proposed a stretchable TENG band for self-powered identity recognition by distinguishing the change of each person's muscle morphology during walking [140]. As illustrated in Fig. 8d, in the fabricated soft TENG, the physiological saline as liquid electrode sealed by a rubber tube as the electrification layer. The stretchable device can be worn on the arm or leg, and the area of the charged interface between the rubber tube and the skin will increase or decrease as the muscle contracts or relaxes, resulting in the alternating potential difference between the ground and the electrode, thus generating an AC signal. Due to the different muscle change degree of each person during a certain movement and the original muscle morphology, a unique electrical signal will be obtained even when they complete a same action. Therefore, according to the electric signals with specific waveform generated by each person in the process of walking, the recognition of their identity can be realized.

Additionally, thanks to the soft structure, a small-scale deformation can be sensed by soft TENG sensitively. Ouyang et al. reported a self-powered pulse sensor, which is composed of Kapton film with a Cu electrode deposited on back side (as the electrification layer) and Cu film (as another electrification layer and electrode), and the whole device was sealed by PDMS [141]. As exhibited in Fig. 8e, the surfaces of the two friction layers possess soft nanostructures, which lead to an enhancement in the contact area at the interface under an external force and enhance the electron transfer process in the triboelectrification process. Particularly, the continuous contact between the triboelectric layers achieves a high signal-to-noise ratio, so an ultra-high sensitivity of the sensor can be obtained. The self-powered sensor can be triggered by the pulse with any amplitude and can be fixed on various positions of the human body due to its good flexibility. Moreover, the device can monitor the heart rhythm accurately in real time, and has great potential for health diagnosis in the field of clinical medicine.

3.5. Wearable soft TENGs

By virtue of its superior flexible structure, the soft TENG has been widely used as a wearable electronics for scavenging mechanical energy and self-powered sensors. The soft TENGs can be integrated with other daily wear products such as shoes [115,142], socks [98,143], clothes [118,121,132] and so forth, to realize the wearable functions. In 2013 Zhu et al. developed a power generation insole based on contact-separation mode soft TENG [144]. As illustrated in Fig. 9a, the soft with multilayer folding structure, which consisting of the Kapton substrate, the PTFE film with Cu electrode on the back side as electrification layer and the Al foil as another electrification layer, was packed inside the insole. During walking, the foots will periodically apply and release pressure on the constructed power generation insole, resulting in periodic contact and separation between the friction layers in the soft TENG, and finally realize an AC output. Moreover, as a wearable energy harvesting device, it can be used as a sustainable energy supply for LED, mobile phone and other portable electronic devices. Clothing is one of the excellent wearable platforms that can be integrated with soft TENG. Jung et al. introduced a wearable soft TENG based on fabric that can be woven onto the armpit area of the clothes [145]. As shown in Fig. 9b, based on carbon fabric, the soft TENG consists of two parts, one of which is composed of alternating polyurethane (Pu) and polyimide (PI), and another part is made up of PDMS and Al with a similar design. When swinging the arm at 1.5 Hz, an electrical output with 0.18 $\mu W/cm^2$ power density can be produced by fabric-based soft TENG and stored in the super capacitor after rectification. Furthermore, because the faster the friction speed the faster the charge accumulation, so the human movement can be monitored through the speed of charge accumulation, such as walking, running and sprinting.

The soft TENGs that are shape-adaptable and stretchable can be attached directly to the human skin. A shape-adaptable soft TENG proposed Yang et al. [146], consisting of Kapton film with wavy structure as triboelectric layer and sandwiched between two stretchable serpentine Cu electrodes deposited on PDMS, as exhibited in Fig. 9c. The Kapton film will be in full contact with Cu electrodes and electrified under the vertical compression or transverse tension of external force, and the two will separate after releasing the external force. Under the periodic force, Cu and Kapton film contact and separate periodically, and an alternating current signal can be obtained. The good stretchability of the device arising from the waveform structured triboelectric layer and the serpentine design of the electrode. Meanwhile, the PDMS substrate is shape-adaptive and highly stretchable. Thus, the flexible soft TENG could be attached to the moving and curved human skin as a self-powered sensor, by which the movement of elbow, knee and muscle, even the tiny movements such as swallowing can be well monitored. Recently, Anaya et al. realized a wearable wireless sensing based on the near-field electrostatic induction [147]. In the process of near-field electrostatic induction, when the two charged surfaces move relative to each other after contact electrification a variable electric field $\vec{E}$ in space can be caused, and will further lead to a changing voltage between the grounded conductor in the near field and the ground. As described in Fig. 9e, the soft TENG can be attached to the Orbicularis Oculi muscle, when the eye is closed, the muscle contract, causing the device to stretch and the two triboelectric layers (PEDOT: PSS film and silicone film) contact, while when the eye is open, the muscle will relax and the two triboelectric layers separate from each other. Therefore, during blinking, an alternating electrical signal will be obtained between the ground and the metal electrode placed on the glasses leg, and the monitoring of eye movement is realized. Moreover, based on the same concept, a PDMS film can be adhered to the eyelid to realize wireless sensing of eye movement based on the triboelectrification process between skin and PDMS film during blinking. Different from the traditional contact-separation working mode in TENG, the reported soft TENG does not need any connection between the triboelectric material and the electrode, which widens the design options of wearable TENG.

3.6. Implantable soft TENGs

Different from the *in vitro* application scenarios of soft TENGs as described above, there are some stringent conditions that need to be met for the purpose of realizing the application of soft TENGs *in vivo* [75,76,148–150]. Firstly, it is necessary to ensure that the implanted devices can not cause any damage to human organs, thus, the enough soft structure and appropriate size for the implantable TENGs is indispensable. Secondly, in order to ensure the biocompatibility and sealing of the implantable TENGs, the whole device needs to be strictly encapsulated by the biocompatible materials such as PDMS. Finally, due to that the amplitude of mechanical stimulation in *vivo* is much smaller than that *in vitro*, it is necessary to be sensitive enough for implantable TENG to be triggered by the tiny motion.

The first implantable soft TENG which is capable of scavenging mechanical energy in a living animal was introduced by Zheng et al. [76]. As illustrated in Fig. 10a, the PDMS thin film with pyramid arrays on the surface and Al film with patterned nanostructure acted as the triboelectric layers, of which the surface modification enables the high sensitivity of the device. Meanwhile, the Kapton film and the Au film deposited on the back of Kapton film are selected as the substrate and the other electrode of the soft TENG, respectively. The fabricated device is encapsulated by a layer of PDMS for realizing the biocompatibility and leak-proofness, and can be implanted under the skin of rat chest. In the process of breathing, the thoracic of the rat will expand and contract periodically, which makes the triboelectric layer contact and separate periodically, and then resulting in AC signal. Moreover, under the stimulation of small amplitude mechanical movement, the output current and voltage of this device can reach 0.14 μA and 3.73 V respectively, which can drive the pacemaker to work and regulate the heart rhythm of the rat.

Liu et al. demonstrated a self-powered pressure sensor based on soft TENG in contact-separation mode, which can be implanted into the ventricle and atrium of a pig [148]. As exhibited in Fig. 10b, the soft TENG is fabricated by employing the surface modified PTFE (nano-PTFE) film with Au electrode coated on the back as the electrification material and Al as the other electrode and electrification layer. The two electrification layers separated by the spacer made by ethylene-vinyl acetate (EVA), which will contact each other and charged under the action of external pressure, and then separate again after the pressure is released, therefore, a periodic electrical signal can be generated by the implantable TENG as the heart beats periodically. The larger the external pressure, the larger the effective contact area between the triboelectric materials, so an enhanced electrical signal can be obtained. As a self-powered pressure sensor, the device shows an excellent

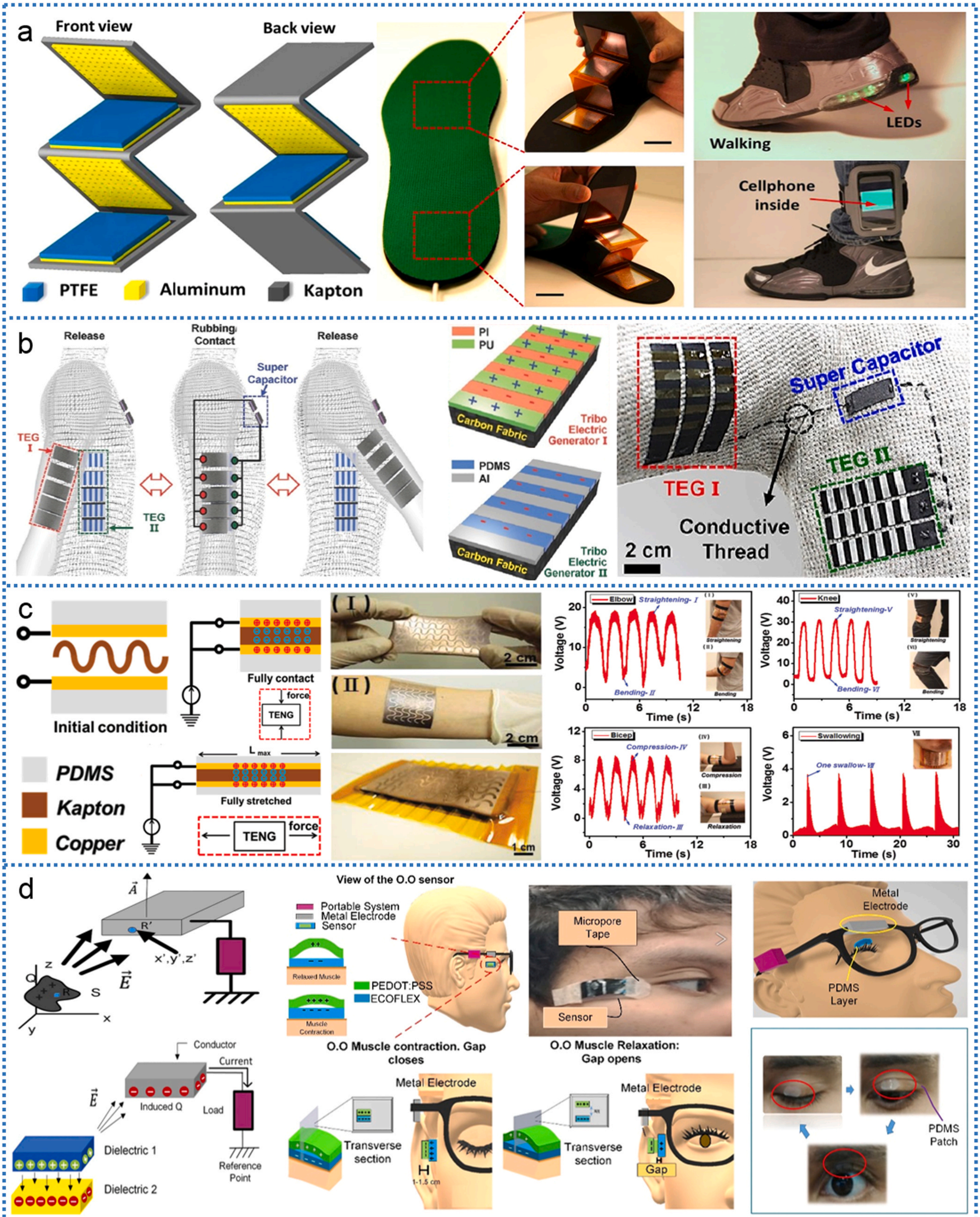

Fig. 9. Wearable soft TENGs. (a) The soft TENG with multilayer folding structure as power generation insole. (b) The fabric-based wearable soft TENG for scavenging mechanical energy during human movement. (c) The shape-adaptable soft TENG attached to human skin for human motion monitoring. (d) The soft TENG attached to the Orbicularis Oculi muscle and eyelid based on near-field electrostatic induction.

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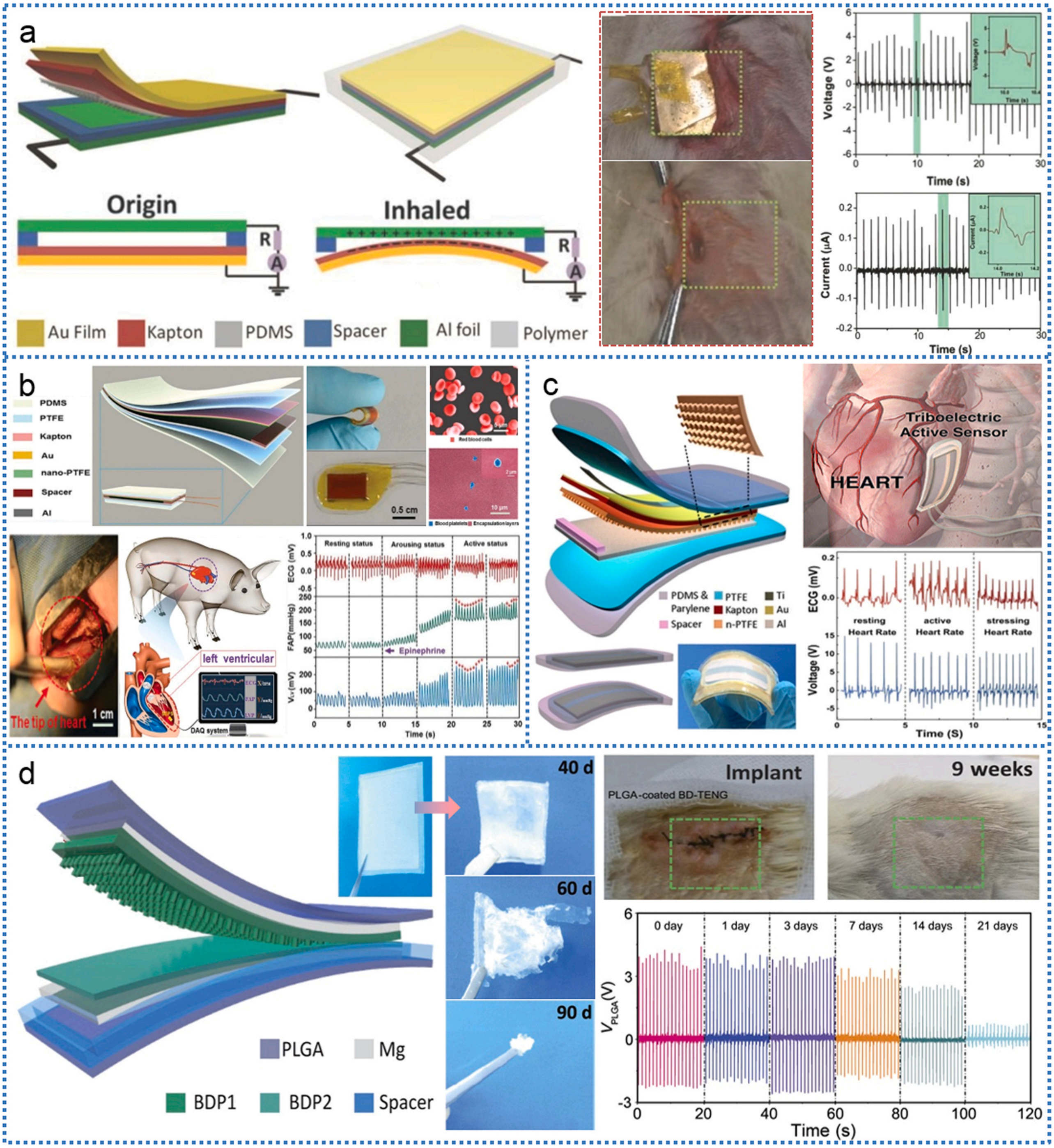

Fig. 10. Implantable soft TENGs. (a) The implantable soft TENG for scavenging biomechanical energy in *vivo* during the process of breathing. (b) The implantable soft TENG to realize self-powered monitoring of endocardial pressure. (c) The implantable soft TENG to realize continuous monitoring of heart rhythm and heart rate for real-time diagnosis. (d) The implantable soft TENG based on degradable materials for harvesting biomechanical energy.
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sensitivity which can reach up to 1.195 mV/mmHg and an outstanding linearity. Moreover, the encapsulation layer of the device including a layer of PTFE film (the first layer) and a layer of PDMS film (the second layer), which is durable and can be well compatible with blood. By virtue of the flexibility and biocompatibility, the soft TENG can be implanted into the heart to realize self-powered monitoring of endocardial pressure (EP) without affecting the normal beating of the heart and even detect arrhythmia effectively. The proposed implantable soft TENG has a great application prospect in implantable medical monitoring.

In addition, implantable soft TENG can realize continuous monitoring of heart rhythm and heart rate for real-time diagnosis. Ma et al. proposed an implantable soft TENG consists of triboelectric layers (nanostructured PTFE film and Al film), substrate (Kapton film), electrodes (Au film coated on the back of substrate and Al film), encapsulation layers (PTFE film, PDMS film and Parylene coating layer), elastic Ti strip and spacer [149]. As illustrated in Fig. 10c, the soft device can be attached to the pericardium. The contact-separation mode soft TENG will be driven by the beating of the heart, thus, the generated electrical signal contains abundant physiological information of the heart. By recording and comparing the working signals of electrocardiogram (ECG) and implantable soft TENG in different states of pig, indicating that the heart rhythm and the heart rate can be monitored accurately by the self-powered sensor. Furthermore, when the soft TENG is implanted into the right and left lateral wall of the heart adjacent to the lung, the peak output of the soft TENG will fluctuate periodically with the periodic breathing, indicating that the implantable soft TENG can be also applied for self-powered respiration monitoring.

In particular, the implantable soft TENG based on degradable materials can be directly degraded and absorbed in *vivo* at the end of its operation cycle. Zheng et al. developed a biodegradable and implantable soft TENG for scavenging biomechanical energy in *vivo* [150]. As illustrated in Fig. 10d, the poly(caprolactone) (PCL) film and the poly (L-lactide-co-glycolide)(PLGA) film with nanorod arrays etched on the surface were selected as the triboelectric layers, meanwhile, the ultrathin Mg film coated on the triboelectric layer as the electrode. When the soft TENG with PLGA as the encapsulation layer was immersed in phosphate buffered saline, it would be degraded completely within 90 days. The soft TENG coated by PLGA shows excellent biocompatibility. Nine weeks after implantation into the rat, the wound healed well without obvious inflammatory reaction. As a biomechanical energy harvester, the fabricated soft TENG can produced an output voltage of 4 V and work sustainably for 3 weeks. After implanted, the soft TENG will be degraded in the body spontaneously instead of removing it surgically, so it can be a promising transient implanted medical device.

4. Summary and perspectives

In summary, the application of soft materials further promotes the development of the TENG, of which the practical perspectives have been further improved. Since the first soft TENG was proposed in 2012, tremendous scientific efforts have been devoted to this field and rapid progress has been attained. In this review, the development and advances of soft TENGs including the fundamentals and invention, promising candidate materials, various structures with their own advantages and applications in energy scavenging and self-powered sensors are systematically summarized, with emphasis on their applicability in various practical application scenarios.

As a newly promising energy technology, the soft TENG possess various unique advantages: 1) the soft TENGs can be triggered by a tiny external mechanical stimulation and realize a more effective energy harvesting of micro mechanical energy. On the other hand, a higher sensitivity of the self-powered sensors based on soft TENGs can be realized. 2) The soft TENGs can be deformable (e.g., stretchable, folding and twistable), meanwhile, the mechanical energy in these deformations can be scavenged. In addition, the soft TENGs can be attached to the curved surface and moving surface to realize the harvesting of mechanical energy and self-powered sensing. 3) Thanks to its flexible structure, the soft TENGs can be used as wearable and implantable electronics for realizing human motion sensing and implantable medical monitoring or scavenging biomechanical energy to provide sustainable energy for portable electronic devices.

In future, the following research aspects is desired to be focused. 1) The output performance of soft TENG need to be further improved for driving high-power portable electronic devices. 2) The soft multi-effect coupling nanogenerators can be constructed based on the soft TENGs to achieve higher energy conversion efficiency. 3) Recently, researchers are working on direct current triboelectric nanogenerator (DC-TENG), which can directly provide DC output without any rectification methods [151–153]. As we all know, compared with AC output, the direct current (DC) output is more suitable for driving electronic devices and energy storage. Therefore, the soft DC-TENGs will be one of the focuses of future research. 4) Exploring more superior soft materials and new structures of soft TENGs to achieve more functions and further expand their application scenarios. With the progress of soft TENG in recent years and the more and more scientific research efforts in the future, the greater breakthroughs and more promising practical prospects in this field can be expected.

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.

Acknowledgments

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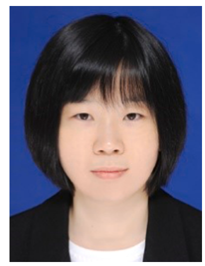

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