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Design of Electrode Materials for Stretchable Triboelectric Nanogenerators

Zhen Wen

Abstract

Triboelectric nanogenerator (TENG), a recently emerging technology that is based on the combination of triboelectric effect and electrostatic induction, has been found to be a promising strategy to harvest large amount of underutilized and low-frequency mechanical energy. One major challenge for TENGs is that the practical application requires flexible, deformable, multifunctional materials to ensure its favorable accommodation to arbitrary surfaces or moving object or harsh environment. Recent research interests mainly focus on the design and fabrication of electrode materials for TENG, making it a perfect candidate for wearable power source. In this chapter, we will introduce a couple of recent achievements regarding highly flexible/deformable TENGs based on stretchable electrodes, including geometrically designed electrode, mixture of conductive materials with elastomeric materials and intrinsically stretchable electrode, etc. In addition, we will address stretchable and self-healing electrodes of flexible TENGs for potential wearable and implantable electronics.

Keywords: triboelectric nanogenerator, electrode design, stretchable electrodes, self-healing, wearable electronics

1. Introduction

Rapid advancement of mobile and portable electronics for health care, environmental sensing and communication is a prominent trend in IoT times [1, 2]. Thus, developing corresponding wearable energy sources to satisfy the wearing demand is of great significance to enhance life quality [3]. There is abundant energy around us in various forms, such as solar energy, thermal energy, mechanical energy etc., which can be converted into electrical energy utilizing energy harvesting techniques. Among them, mechanical energy could be the most extensively distributed one. Nevertheless, mechanical energies like human motion, wind blowing, and ocean waves are almost neglected and wasted in daily life. In 2012, Fan et al. [4] demonstrated a mechanical energy harvester, triboelectric nanogenerator (TENG) based on the coupling effects of triboelectrification and electrostatic induction. Taking advantages of high voltage output, TENG can be used to power commercial digital electronics and can even directly light up LED lights.

In addition to the high electrical output performance, TENG also possesses advantages of light weight, wide-ranging material selection, simple fabrication process, etc. so that it could be regarded as a promising energy source for wearable

electronics [5]. In recent years, tremendous efforts have been put into the development of flexible TENGs which can be attached on human body to harvest human motion energy like walking, patting or twisting [6–8]. Moreover, stretchability of TENG is essential since stretching behaviors happen frequently in human motions such as joint motions. So far, stretchable TENGs show extensive application in many fields, ranging from body mechanical energy harvesting, wearable sensors to monitor human health as well as biocompatible e-skin. The key challenge to fabricate stretchable TENG is to choose stretchable triboelectric materials and electrode materials which are the two main components for TENG [9]. For stretchable triboelectric materials, different kinds of elastomers (PDMS, silicone rubber, VHB, etc.) are the main source due to their softness and stretchability as well as the excellent triboelectric properties.

For stretchable electrode materials, we will introduce three main fabricating strategies in this chapter regarding different properties of conductive materials, including (i) geometrical design of rigid conductive materials, (ii) mixture of conductive materials with elastomers, (iii) intrinsically stretchable and conductive materials. In addition, we will also involve stretchable and self-healing conductive materials. Once the materials are over-stretched, self-healing capability enables the device to last longer lifetime. Meanwhile, the fabrication process of stretchable TENG based on these stretchable electrodes will be discussed as well. Herein, we only involve single-electrode TENG and contact-separation TENG. It should be noted that in some cases of contact and separation mode TENG, stretchable electrode materials can be treated as electrode materials and one of the triboelectric materials at the same time [10, 11].

2. Strategies of stretchable electrodes for TENGs

2.1 Geometrical design of rigid conductive materials

Rigid conductive materials, such as carbon papers, metal foils/wires, certain polymer thin films are all excellent candidates for the electrodes for TENG. But their low strain deformations limit their application in flexible and stretchable TENGs which is a promising developing trend for wearable electronics. In order to overcome this challenge, geometrical designs and engineering have been developed to endow these rigid conductive materials with certain stretchability. Herein, we will introduce several common geometrical engineering strategies.

Wave structural configuration can effectively accommodate large strain to avoid potential fracture of the rigid electrode materials themselves. Such wavy-shaped electrode can be achieved by deposition, coating or transferring the conductive thin film on the pre-stretched elastomeric substrate [14–17]. As **Figure 1a** shows, Wen et al. demonstrated a transparent and stretchable triboelectric nanogenerator based on wrinkled PEDOT:PSS electrode. In this work, poly(dimethylsiloxane) (PDMS) is pre-stretched and fixed on a flat glass panel, which is employed as the elastomeric substrate. Then the conductive material PEDOT:PSS film is blade-coated on this substrate. After releasing the sample from the panel, a wavy-shaped electrode is spontaneously formed (**Figure 1b**). Finally, a stretchable TENG can be obtained by inserting the Al foil as lead wire and encapsulating the whole device with additional PDMS [12]. Such a device is not only applied as body motion harvester but also as the active motion sensor attaching on human skin. Moreover, **Figure 1c** illustrates the fabrication process of a stretchable, transparent and self-healable TENG based on a buckled Ag-PEDOT electrode. The stretchability of this electrode is also achieved *via* a prestrain of substrate (H-PDMS)-transfer of conductive film

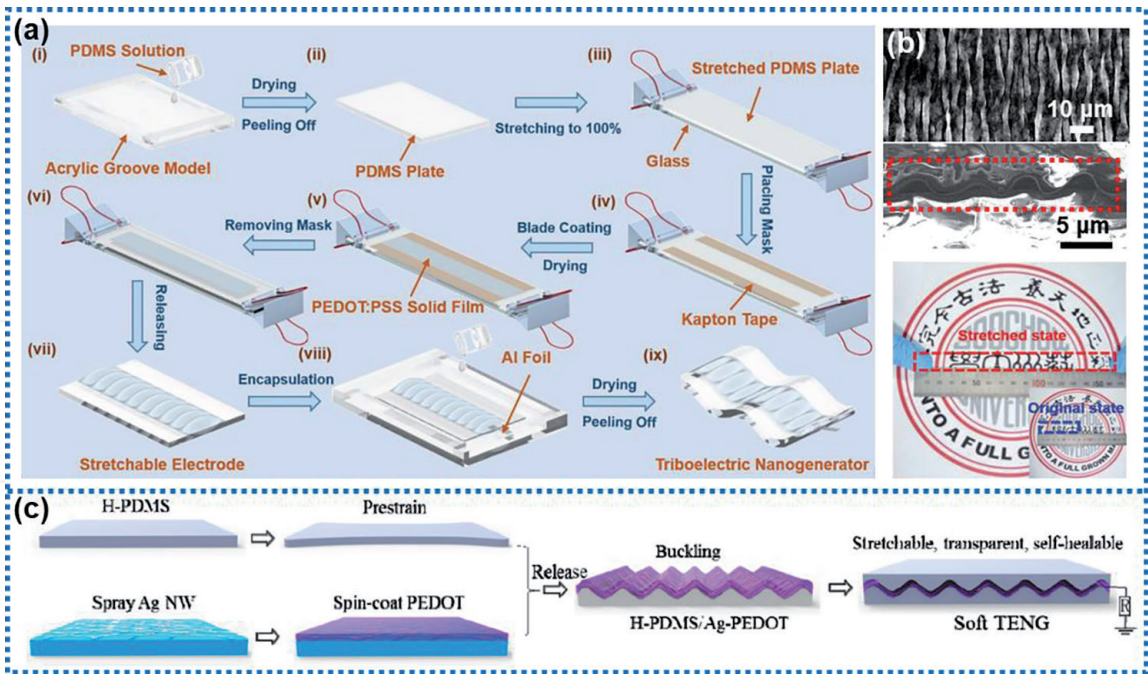


Figure 1. Geometrical design of wave structural configuration for rigid materials. (a) Fabrication process of wavy-structured PEDOT:PSS film based triboelectric nanogenerator. (b) SEM image of a wavy-structured PEDOT:PSS film on an elastic PDMS substrate (up: Front view, down: Side view) and electrode in stretched and original state. Reproduced with permission [12]. Copyright 2018 WILEY-VCH (c) fabrication procedures of the Ag-PEDOT based stretchable electrode and the final soft TENG. Reproduced with permission [13]. Copyright 2018 American Chemical Society.

(Ag-PEDOT)-release of prestrain process. PEDOT here is spin-coated on the Ag NW film at first as an adhesive agent to anchor the networks for successful transfer [13]. Besides, in terms of self-healable ability, we will discuss about it later in this chapter which is also a significant property for stretchable electrodes. In both cases above, the ultimate stretchability of the electrodes is determined by the prestrain level of the elastomeric substrate.

Another strategy to configure stretchable electrode for TENG is to operate geometrical engineering to the existing rigid materials themselves since it is difficult for some conductive rigid materials to adhere to the prestrained substrate. As depicted in **Figure 2a** [18], the wavy structure could be realized easily through paper-folding method. The fabrication process of this stretchable TENG based on sandpaper including following steps: Firstly, the conductive paper is obtained by pencil drawing on the sandpaper. Secondly, the stretchable electrode is achieved by folding the paper into wavy structure. Lastly, the stretchable TENG is fabricated by sealing the carbon paper electrode inside silicone rubber. The ultimate stretchability of this electrode depends on the folding angle of the carbon paper. Take two folds as an example, the length of the electrode after stretching L_{max} can be describe as

$$L_{max} = L_0 / \sin \frac{\theta}{2} \quad (1)$$

Where L_0 is the original length of electrode before stretching, θ is the fold-ing angle (**Figure 2b**). It should be noted that the ultimate stretchability of such stretchable TENG should be determined by a lower one after comparing the stretch-ability of silicone rubber and folded electrode. Here in Zhou's work, the folded carbon paper electrode determines the ultimate stretchability of the entire device. In addition, **Figure 2c** shows that Xie et al. [19] proposed a fiber-shaped stretchable TENG based on the geometric engineering of a steel wire electrode. The steel wire is designed into spiral structure possessing certain stretchability. Subsequently,

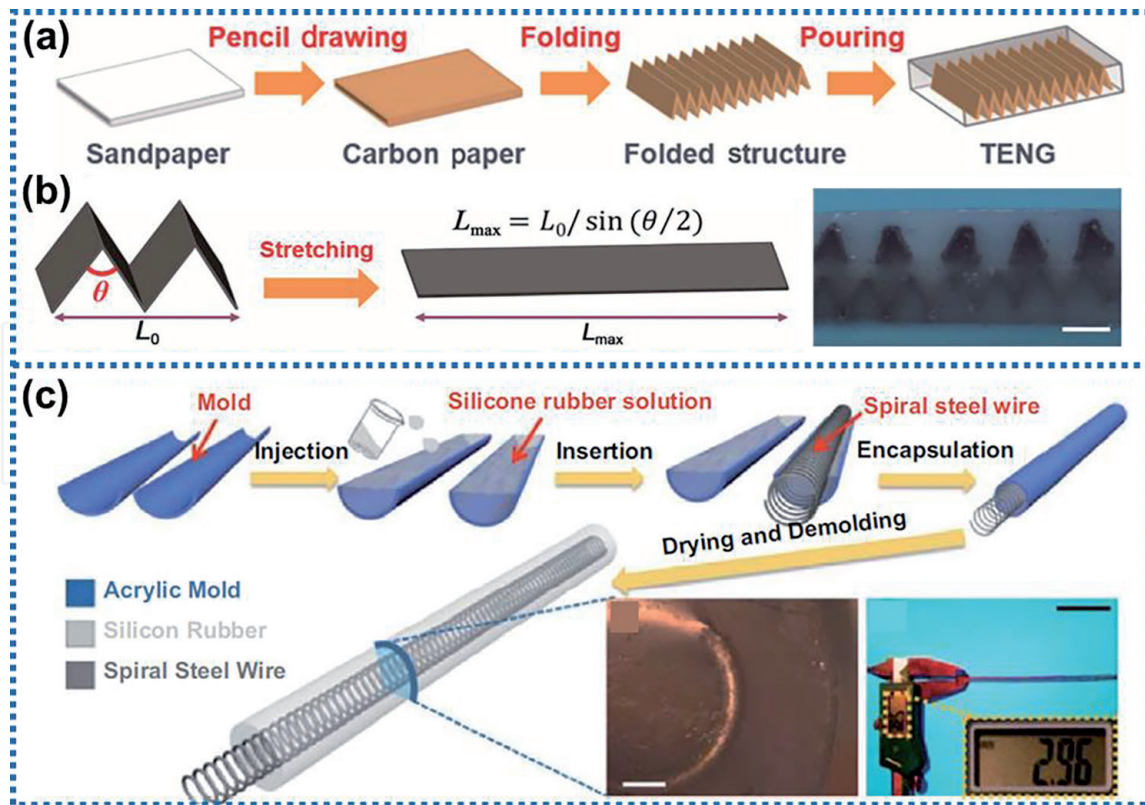


Figure 2. Geometrical engineering of the existing rigid materials for stretchable purpose. (a) Fabrication process of wavy-structured carbon paper electrode and the final TENG. (b) the geometrical relationship between maximum stretchability and folding angle θ . reproduced with permission [18] copyright 2018 springer nature. (c) Fabrication process of the spiral steel wire electrode based TENG. Reproduced with permission [19] copyright 2019 springer.

by inserting such spiral steel wire into silicone rubber, a fiber-shaped stretchable TENG could be obtained. Similar to the previous example, the ultimate strain of this device is also determined by the strain level of the spiral steel wire since the maximum strain level of silicone rubber is much better than these geometrically designed rigid materials. Besides energy harvesting, these fiber-shaped stretchable TENGs are woven into gloves to act as gesture sensor.

The stretchability of rigid materials can also be acquired via kirigami architecture. As illustrated in **Figure 3a**, a silver-coated nylon yarn is employed as the electrode, for which the stretchability is achieved by zigzag arrangement and rhombus interlaced network. Stretchable and soft silicone rubber is selected to seal the yarn electrode network. When tensile strength is applied, the electrode network could be stretched due to gradually straightened zigzag structure and extension of rhombic region (**Figure 3b**). The ultimate stretchability of the TENG is determined by the thickness of the silicone rubber and the basic unit of network electrode including the height h and the length l (**Figure 3c**). With the h and l of 1.24 mm and d of 2.8 mm, the TENG can be stretched to 30% stain, which is eligible for e-skin and pressure sensing application [20]. Origami strategies like **Figure 3d** [21] and **Figure 3e** [22] can also endow the brittle materials with stretchability.

2.2 Mixture of conductive materials and elastomer

In this section, stretchable electrodes for TENG are acquired by directly mixing conductive materials (metal particles, carbon nanotubes, carbon black and so on) with elastomer (PDMS, silicone rubber and so on), taking advantages of the conductive property of conductive materials and the stretchability of elastomers.

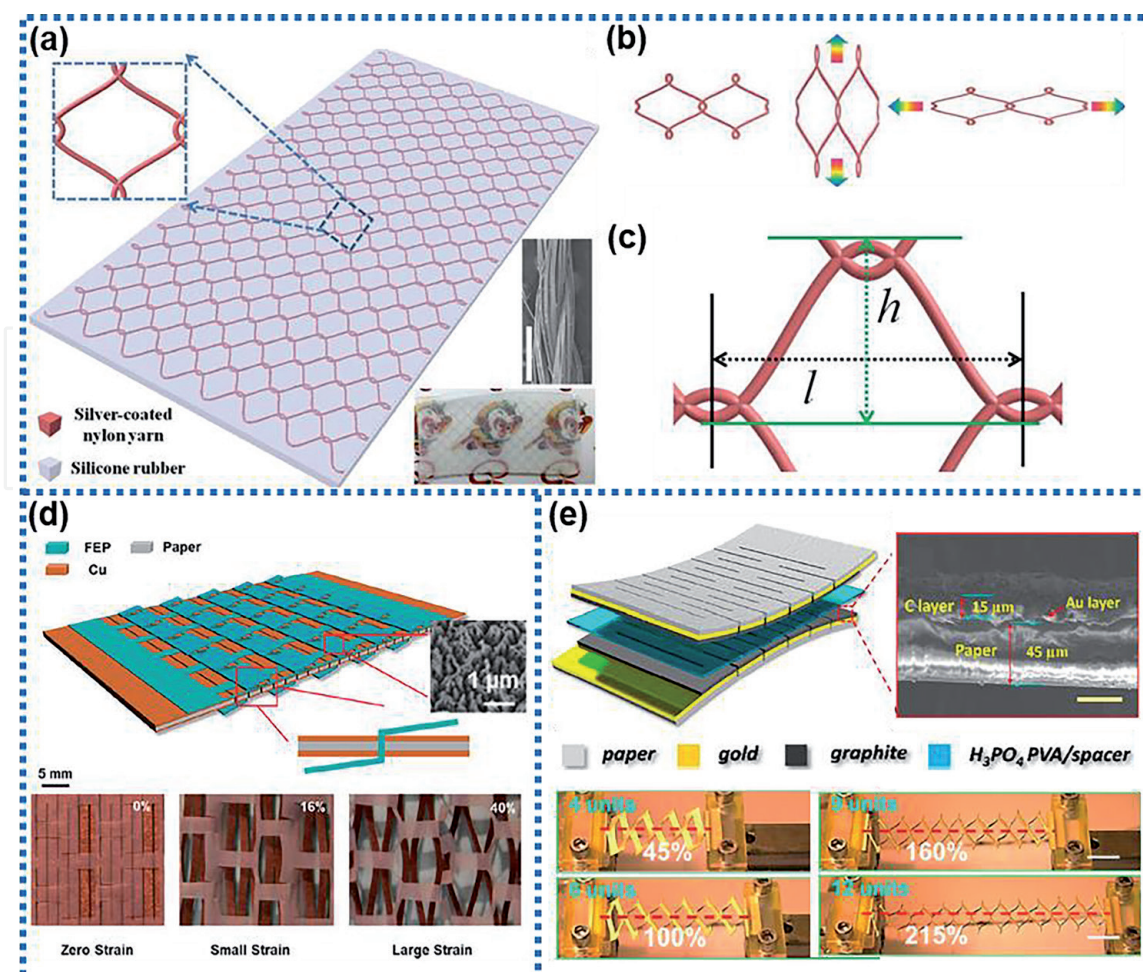


Figure 3. Geometrical engineering of rigid materials by origami architecture. (a) Schematic illustration of the TENG with "chain-link" fence-shaped structure and rhombic unit design. (b) Schematic demonstrating the in-plane tensile behaviors of the repeated rhombic unit in the TENG system. (c) the basic cyclic unit of the yarn conducting network in the SI-TENG system. Reproduced with permission [20] copyright 2018 Wiley-VCH (d) schematic structure of paper-based TENG and the assembled device under different tensile strains. Reproduced with permission [21] copyright 2016 American Chemical Society. (e) Schematic structure of a kirigami based electrode and the related strain property. Reproduced with permission [22] copyright 2016 American Chemical Society.

Different from depositing conductive materials on prestrained elastomer surface, conductive materials in this strategy are embedded into the elastomers, making use of the fluidity of silicone rubber before solidifying.

Figure 4a illustrates a TENG based on a stretchable electrode, combining carbon black with silicone rubber. Herein, carbon black is blended with liquid silicone rubber to form a mixture. Then the mixture is coated over a piece of acrylic plate which has been processed with release agent in advance. After the mixture is solidified, a piece of stretchable conductive carbon black-silicone rubber mixture layer can be peeled off from the plate. **Figure 4b** shows the nano/microstructured surface morphologies of the stretchable electrodes in the original state. Regarding to the proposed stretchable TENG, its triboelectric materials contain one silicone rubber layer and one carbon black-silicone rubber mixture layer. Meanwhile, the stretchable mixture layers act as the electrodes for TENG. An air gap is created between the two layers to realize contact and separation process. With the increasing strain of TENG, electrical output is enhanced resulting from the increasing displacement of two triboelectric layers (**Figure 4c**) [23]. **Figure 4d** demonstrates another super-stretchable TENG via percolating networks effect of silver nanowires and silicone rubber. The fabrication process is as follows: Firstly, Ag NWs suspension is drop-coated on an acrylic plate with a specific shape molded by kapton tapes. Next, the

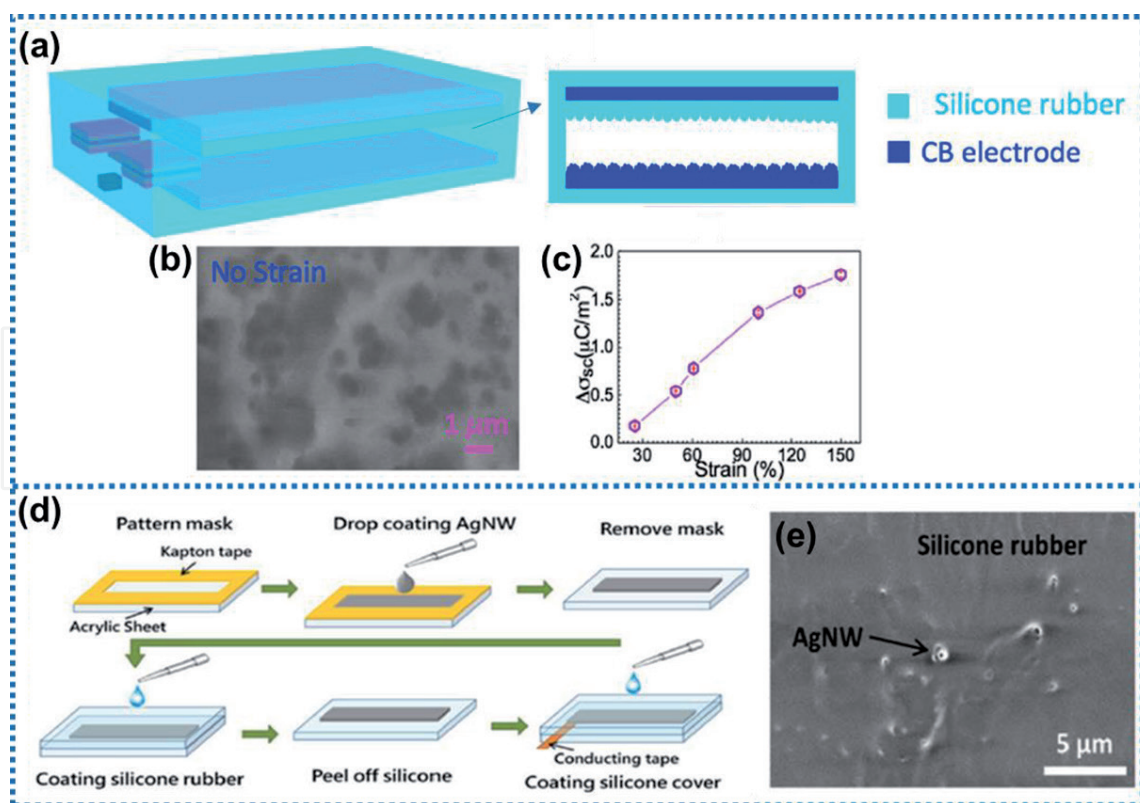


Figure 4.

Mixture of conductive materials and elastomer. (a) Schematic diagram showing the detailed structure of the stretchable TENG. (b) SEM images of the surface morphology of the TENG's electrode in the original state. (c) Relationship between the $\Delta\sigma_{sc}$ and the elongation of the device. Reproduced with permission [23] copyright 2016 American Chemical Society (d) fabrication process of the silver nanowire- silicone rubber electrode based TENG. (e) SEM image of stretchable electrode. Reproduced with permission [24] copyright 2016 Wiley-VCH.

tapes are removed from the sheet and liquid silicone rubber is cast onto the as-fabricated Ag NW network. After solidifying of silicone rubber, Ag NWs-silicone rubber film is peeled off from the acrylic plate. Finally, stretchable TENG is obtained by curing another liquid silicone rubber again over the film with a copper tape as conductive lead for assembling the Ag NW-silicone rubber layer. It is clearly shown in **Figure 4e** that Ag NWs are embedded in the silicone rubber because interspace between Ag NWs allows silicone rubber to percolate into the network structure. The proposed Ag NWs-silicone rubber mixture layer exhibits outstanding stretchability, so as for the fabricated TENG, reaching up to 300% strain [24].

In the work of Lim et al., they fabricated a stretchable TENG based on highly stretchable Au NS-embedded PDMS electrode. As shown in **Figure 5a**, Au NS multilayer film was prepared by transferring monolayer of the Au NSs onto a Teflon coated Si wafer for several times. After each transfer, thermal annealing is conducted to increase the contact between stacked Au NSs. Then, liquid PDMS is poured to the as-prepared Au NS film, percolating into the interspace between the NSs. After curing, the Au NS-embedded PDMS film can be peeled off from the Si wafer. In this stretchable electrode film, Au NSs are densely entangled and partially embedded in the PDMS matrix (**Figure 5b**). Different from directly depositing conductive materials on the surface of elastomer, embedding conductive materials into elastomers presents a more stable and slight resistance change for stretchable electrodes especially under larger strain ($>30\%$) (**Figure 5c–e**). Such TENG is employed to detect movement of fingers [25]. Li et al. proposed electrospinning technique as an effective strategy for the fabrication of conductive materials-elastomer mixed stretchable electrode.

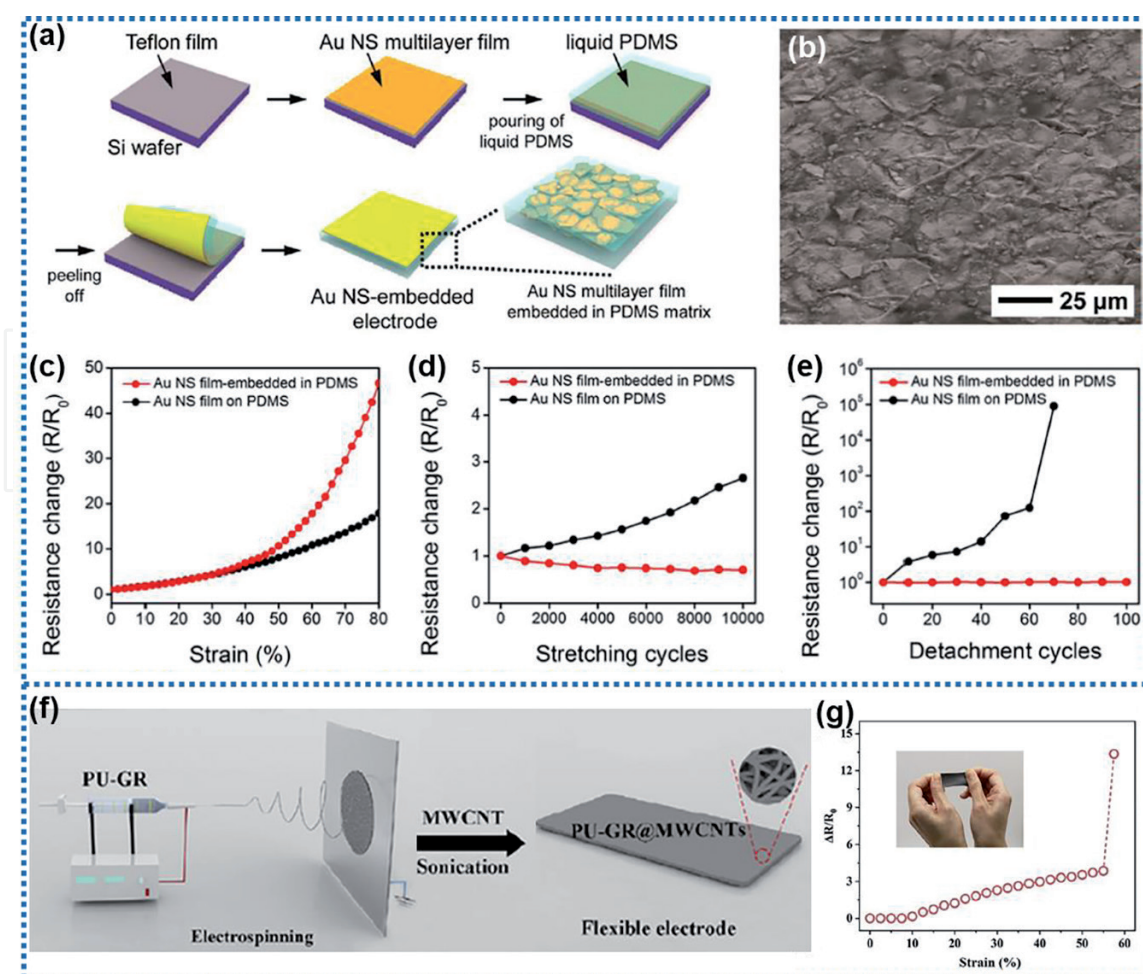


Figure 5. Mixture of conductive materials and elastomer. (a) Fabrication process of the Au NS-embedded electrode. (b) Top-view FE-SEM image of the Au NS-embedded electrode. (c) Change in resistance for Au NS electrodes in response to tensile strain. (d) Variations of R/R_0 for Au NS electrodes during 10,000 cycles of stretching at a 30% strain. (e) Variations of R/R_0 for Au NS electrodes during 100 cycles of the scotch-tape detachment test. Reproduced with permission [25] copyright 2017 Elsevier (f) fabrication process of PU-GR@MWCNTs stretchable electrode. (g) Resistance change of the electrode under different strains. Reproduced with permission [26] copyright 2019 Elsevier.

As **Figure 5f** shows, polyurethane (PU) and graphene (GR) are mixed uniformly as the solution for electrospinning. Then, the prepared PU-GR film is sonicated in the solution of MWCNTs to reduce its internal resistance. Consequently, MWCNTs coated PU-GR (PU-GR@MWCNTs) stretchable electrode is obtained. Such stretchable electrode can withstand a strain of about 15% without a remarkable resistance increase (**Figure 5g**). Researchers also attached this TENG on the leaves to monitor wind speed in the natural environment [26]. In contrast with previously described wavy-structure electrodes whose stretchability mainly depends on the pre-strain level, electrodes based on percolating networks or mixture of conductive materials with elastomers display strong dependence on the capacity of the overlapping of conductive materials in response to the applied strain.

2.3 Intrinsically stretchable conductive materials

Considering the above two strategies, although stretchability can be acquired for electrodes, it is greatly limited by the geometrical engineering technique and conductive pathways of conductive materials when embedded into elastomers.

In this section, we will introduce intrinsically stretchable conductive materials including liquid-state materials with extremely low Young's modulus and prepared

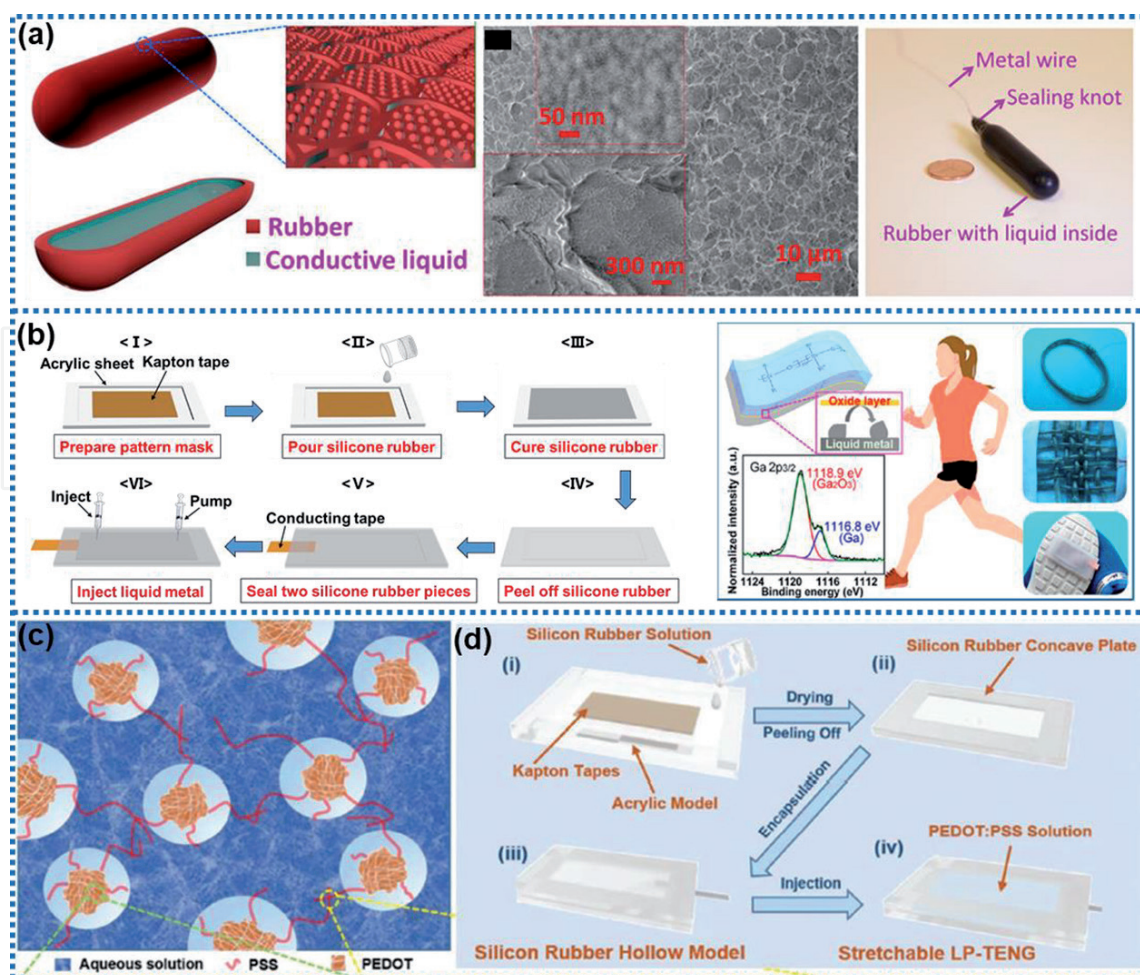


Figure 6.

Conductive liquid-state materials based stretchable electrode. (a) The structure of conductive liquid based TENG unit. Reproduced with permission from the authors [27] (b) fabrication process of liquid metal based TENG. Reproduced with permission [28] copyright 2018 American Chemical Society (c) the PEDOT:PSS particles dispersed in the aqueous solution. (d) Fabrication process of liquid PEDOT:PSS based TENG. Reproduced with permission [29] copyright 2019 the Royal Society of Chemistry.

conductive gels. As shown in **Figure 6a**, a stretchable TENG is composed of conductive liquid inside and a rubber layer outside with etched nanostructures. In the fabrication process, conductive liquid is injected into a hollow cylindrical rubber layer with one terminal open and the other one sealed. After injection, a copper wire is inserted into the liquid and a knot is formed to seal the open terminal. NaCl is used here as the conductive liquid electrode which has certain conductivity and infinite stretchability due to its fluidity at room temperature [27]. Ultimate stretchability only relies on the stretchability of the rubber, which means liquid-state conductive materials can exercise the maximum potential for stretchable electrodes.

However, the performance of TENG is limited due to the low conductivity of ion solution. Thus, in Yang et al.'s work, [28] they demonstrate a super-stretchable and high-performance TENG using liquid metal (Galinstan) electrode which exhibit low Young's modulus, high conductivity at room temperature. Silicone rubber is chosen as the triboelectric layer and encapsulating layer. **Figure 6b** shows the fabrication process of this stretchable bulk-shaped TENG: A silicone rubber model is prepared by encapsulating two concave silicone rubber plates together with a conducting tape inserted to form a cavity in the middle. Single-mode stretchable TENG is obtained after Galinstan is injected into the cavity. The as-prepared stretchable TENG has an ultimate stretchability of 300% depending on the silicone rubber. Meanwhile, its electrical output performance is much higher than the ion-solution based stretchable TENG.

Similar to this work, Shi et al. [29] utilize the liquid-state PEDOT:PSS as electrode for fabricating TENG with silicone rubber as triboelectric layer as well as encapsulating layer. Conductive polymer, PEDOT:PSS are commonly used in organic solar cells, organic light-emitting diodes (OLED) due to its transparency and conductivity while in solid state. As illustrated in **Figure 6c**, PEDOT and PSS particles are dispersed in the aqueous solution and combined by electrostatic attraction. Significant conductivity and stretchability of PEDOT:PSS electrode is attributed to the conjugated bond of PEDOT like other conductive polymers. **Figure 6d** shows similar fabrication process of this polymer liquid electrode based TENG as liquid metal based TENG. Such prepared liquid PEDOT:PSS TENG can also be stretched to 300% at most without any crack.

Moreover, hydrogels, which contains hydrophilic polymer networks swollen with water or ionic aqueous solution, are intrinsically stretchable materials, so that they can be employed as stretchable conductive electrodes for TENG. For example, stretchable electrodes based on polyacrylamide (PAAm)-(lithium chloride) LiCl hydrogel is proposed by Pu et al. for application in the stretchable TENG [30]. PAAm powder is added into LiCl solution, followed by adding N, N'-methylenebisacrylamide, ammonium persulfate, and N, N, N', N'-tetramethylethylenediamine orderly into the solution. Then the solution is transferred into a mold. After treated at 50°C for 2 hours, conductive and stretchable hydrogel is formed. Using elastomer VHB/PDMS as triboelectric materials as well as encapsulating materials, the hydrogel is sealed inside the elastomer to form single-electrode mode TENG (**Figure 7a**). Such VHB-hydrogel based TENG can be stretched to 1000% of original length while the PAAm-LiCl itself can be stretched to 1400% (**Figure 7b**). It means that the conductive hydrogel electrode present enough stretchability for the fabrication of stretchable TENG. The stretchability is only controlled by the encapsulation materials. Zhao et al. [31] utilize PAMPS ionogel as the electrode for stretchable TENG, where the conductivity of the ionogel is

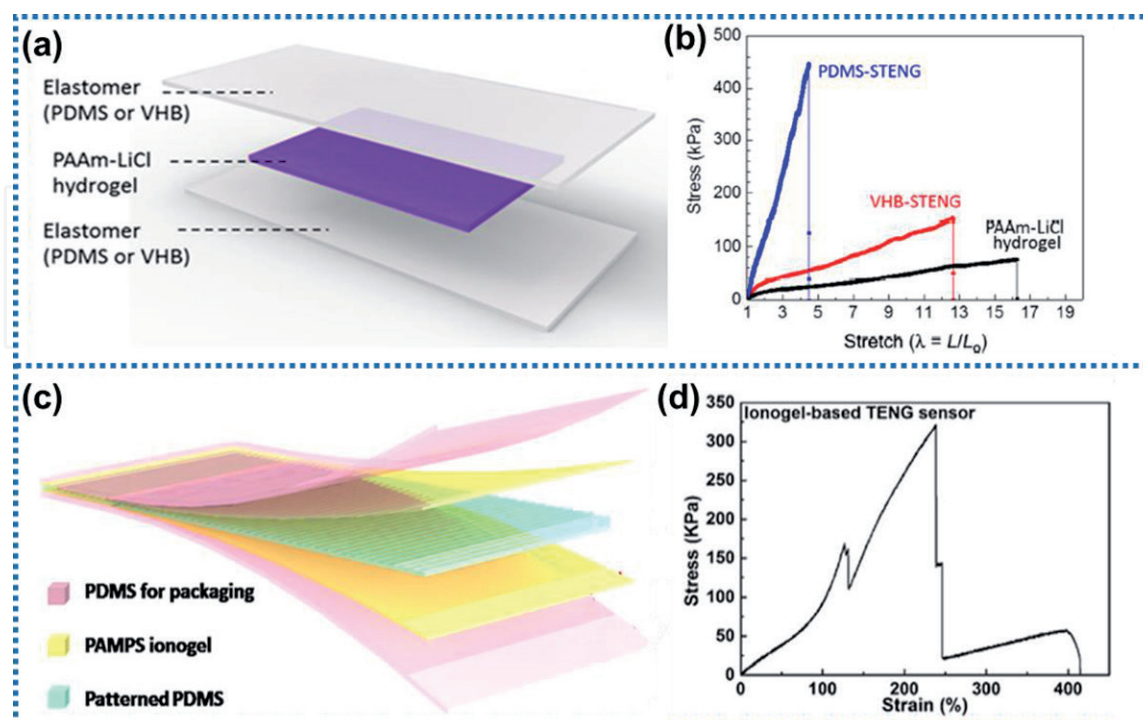


Figure 7. Conductive gels based stretchable electrode. (a) The structure of PAAm-LiCl hydrogel electrode based TENG. (b) Tensile test of the PAAm-LiCl hydrogel, PDMS-STENG and VHB-TENG. Reproduced with permission of the authors [30] (c) the structure of PAMPS ionogel electrode based TENG. (d) Tensile test of the ionogel-based TENG. Reproduced with permission [31] copyright 2019 Elsevier.

attributed to the nonvolatility of ILs. As shown in **Figure 7c**, TENG works in contact and separation mode with patterned PDMS acting as one triboelectric material and ionogel acting as the other triboelectric material. Distance between two triboelectric materials is created due to triangular stripes on the patterned PDMS. The three layers (one patterned PDMS and two ionogel layers) are sealed inside two smooth PDMS films. Such device has an ultimate stretchability of about 125% which is determined by the ionogel film which has lower strain level compared with the patterned triboelectric PDMS and smooth packaging PDMS (**Figure 7d**). Due to the transparency, stretchability and pressure sensitive property of TENG, touching and pressure sensing are demonstrated in both work in **Figure 7**.

2.4 Self-healing conductive materials

When the electrodes are over stretched, fractures would occur to influence its conductivity as well as the output performance of TENG. Hence, self-healing ability is required for electrode materials so that fractured structures can be repaired to maintain performance and elongate the lifetime of devices. The self-healing capability of electrode can be attributed several mechanisms, such as the containing of self-healing agents inside microcapsules, the use of dynamic bonds.

As **Figure 8a** shows, Parida et al. [32] prepared a stretchable and self-healing TENG based on slime ionic electrode. VHB adhesive tape, acting as the substrate and encapsulation material also possesses stretchability and self-healing capability. Although silicone rubber is not intrinsically self-healable, the self-healing process of

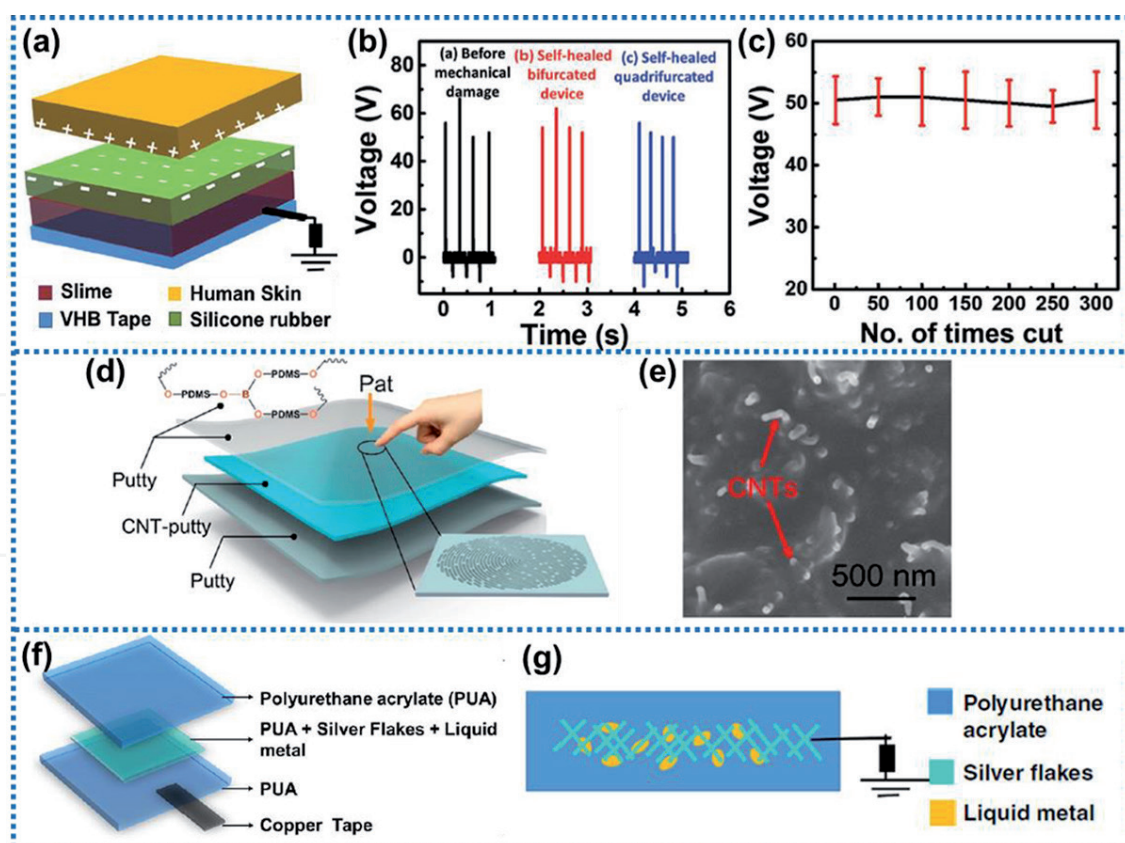


Figure 8.

Self-healing conductive stretchable materials based electrode. (a) The structure of slime electrode based self-healing TENG. (b) Voltage output of the self-healing TENG before damaging and after healing. (c) Stability of the TENG after multiple cut. Reproduced with permission [32] copyright 2017 Wiley-VCH (e) SEM image of CNTs embedded putty. Reproduced with permission [33] copyright 2019 American Chemical Society (f) structure of PUA-silver flake-liquid metal electrode based TENG. (g) Schematic diagram of the stretchable and healable triboelectric nanogenerator indicating silver flakes and liquid metal particles are embedded in the PUA matrix. Reproduced with permission from the authors [34].

slime electrode and VHB tape can bring the damaged silicone rubber film back into contact. Meanwhile, it would not influence triboelectrification with skin or electrostatic induction in electrode because charges over silicone rubber are immovable. Thus, the entire device can be regarded as self-healing TENG. Ultimate stretchability of the device can be as high as 700%. The damaged device can be successfully healed at room temperature without any external stimuli. **Figures 8b** and **c** show that output performance of TENG before damage and after self-healing is comparable to each other, even damaged and recovered for 300 times.

Chen et al. [33] demonstrate a stretchable and self-healing TENG utilizing viscoelastic supramolecular polymer (Putty) as triboelectric materials, mixture of carbon nanotubes and Putty as electrode materials (**Figure 8d**). The percolating method of configuring stretchable electrode is introduced in previous part. **Figure 8e** shows the morphology of the CNT embedded putty. Self-healing capability of CNT-Putty electrode is acquired because of reversible dynamic hydrogen bonds and dative bonds between boron and oxygen in putty, enabling the stretchable TENG to recover from damage at room temperature in 3 min.

Another example of self-healing and stretchable electrode for TENG is fabricated by mixing polyurethane acrylate (PUA) solution, liquid metal particles and silver flakes together. And then the mixture is drop casted and sandwiched between two PUA films. A self-healing and stretchable TENG can be developed after UV cures (**Figure 8f**). This TENG shows the highest stretchability (2500%), compared to previously reported TENGs. Liquid metal here provides effective anchoring between silver flakes in the PUA matrix, ensuring stable electrical performance (**Figure 8g**). Both stretchability and self-healing capability of the device is also attributed to the supramolecular hydrogen-bonding of PUA. It takes about 24 h for this TENG to recover at room temperature [34].

3. Conclusions

Overall, we have introduced three functional strategies in this chapter to fabricate the stretchable electrodes for flexible and wearable TENGs. In the first strategy, we present several methods considering different material properties, including prestrain-coating-release method for conductive materials that can be coated and cured on the surface of stretchable elastomer, structure design and origami architecture method for those existing conductive materials. In the second strategy, conductive and stretchable electrodes are fabricated by embedding conductive materials into the elastomer. In the third strategy, intrinsically stretchable electrode like liquid-state electrode and conductive gel are presented. Besides, we also discussed about the self-healing and stretchable electrodes for TENG application. These stretchable electrodes ensure the great flexibility of TENG so that it can be further applied in wearable electronics.

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Conflict of interest

The authors declare no conflict of interest.

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