Doctoral Dissertation (Shinshu University)

Study on conductive hydrogels in flexible and wearable triboelectric devices towards energy-harvesting and sensing applications

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Abstract

Flexible triboelectric nanogenerators (TENGs) based on the combination of contact-electrification and electrostatic induction have attracted increasing interest since their advent in 2012. In comparison with other flexible electrodes, hydrogels that combine the integrated attributes of transparency, stretchability, tunable conductivity, self-adhesion, self-healing, biocompatibility, ease of processing, and the ability to customize different shapes provide great potential as current collectors in TENGs for wearable applications. However, the development of hydrogel-based TENGs (H-TENGs) is currently an emerging field, but research is lagging far behind other common flexible TENGs. In the work, a series of conductive hydrogels are prepared and then utilized as current collector for TENGs that harvest mechanical energy and act as multi-functional self-powered sensors *via* the triboelectric effect. Detailed research contents are as follows:

(1) A dual-network polyacrylamide (PAAM)/ polyacrylic acid (PAA)/ graphene (GR)/ poly (3,4-ethylene dioxythiophene): poly (styrene sulfonate) (PEDOT: PSS) (MAGP) conductive hydrogel composed of dual-cross-linked PAAM and PAA as well as PEDOT: PSS and GR as conducting component. A wearable strain sensor is fabricated by sandwiching the MAGP hydrogels between two dielectric carbon nanotubes (CNTs)/ Poly (dimethylsiloxane) (PDMS) layers, which can be utilized to monitor delicate and vigorous human motion. In addition, the hydrogel-based sensor can act as a deformable

triboelectric nanogenerator (D-TENG) for harvesting mechanical energy to power small electronics, which provides a potential approach for the development of deformable energy sources and self-powered strain sensors.

(2) A scalable fabrication of core-sheath-structured elastomer triboelectric fibers that combine silicone hollow tubes with gel-electrodes is presented. Gel-electrodes were fabricated *via* a facile freeze–thawing process of blending polyvinyl alcohol (PVA), gelatin, glycerin, poly (3,4-ethylene dioxythiophene): poly (styrene sulfonate) (PEDOT: PSS), and sodium chloride (NaCl). Such fibers can also be knitted into deformable triboelectric nanogenerator textiles with high electrical outputs, which could work as reliable power supplies for small electronics. Moreover, we demonstrate fabric materials recognition, Morse code communication, and human-motion-recognition capabilities, making such triboelectric fiber platform an exciting avenue for multifunctional wearable systems and human–machine interaction.

(3) Based on the novel covalent-like hydrogen bond engineering, we developed a syrup-modified gelatin/Ag-hydrogel, which is entirely derived from natural or safe constituents and highly stretchable, yet fully biodegrades or recyclable. Meanwhile, strong covalent-like hydrogen bonds endow it with several favorable features of adhesion, rapid self-healing and antifreeze. A series of multi-modal application scenarios are further demonstrated to verify the adaptability of this platform that integrates various attributes.

In summary, this work develops a series of conductive hydrogels, respectively, as

well as shows how these hydrogels incorporated into wearable TENG and further demonstrate potential applications in wearable sensors and energy harvesters. we believe above work will be useful to researchers and engineers in this emerging field and spur research and development of this important area.

Chapter 1

General introduction

Chapter 1: General introduction

With the advent of the Internet of Things (IoT), Big Data and Artificial Intelligence (AI) era, the rapid development of next-generation portable and wearable electronics such as haptic sensors, electronic skins and soft robots has challenged the corresponding power devices that they should be quite transparent, flexible and variable. However, traditional power sources such as capacitors and batteries are simply unable to meet the demands of the development of flexible, wearable electronic devices due to their rigid nature which can barely withstand violent deformation. A promising strategy to alleviate this challenge is the triboelectric nanogenerator (TENG) based on the principles of friction-generated electricity and electrostatic induction coupling to harvest ambient mechanical energy and convert it into electrical output, which has the advantages of a wide range of material options, simple device structure, and light weight.

In terms of TENG, stretchable and transparent TENGs are in high demand as wearable devices are highly desired for their stretchability and compliance on curved surfaces, while transparency contributes to potential applications in information visual transmission. Flexible TENG requires that both the triboelectric layer and the electrodes are flexible. Compared to the wide range of flexible triboelectric layer candidates, flexible electrode materials with good performance under deformation such as tension and bending without damage are still limited. To impart stretchability properties to electrodes, researchers have proposed strategies for structural design and material novelty.

Conventional metallic materials are not inherently stretchable and stretchability can be obtained by designing geometrical structures such as wavy structures, island bridge structures and interlocking sheaves. However, the high cost and complexity of the process still yielded limited stretchability. Materials engineering is another promising approach that usually results in electrodes with higher stretchable properties than the structural designs described above. Flexible conductive materials such as liquid metals, conductive elastic polymers embedded with conductive fillers, and conductive hydrogels are promising candidates for stretchable electrodes. Among them, liquid metals with high electrical conductivity and arbitrary stretchability are considered promising candidates. However, the high cost and the risk of harmful liquid metal leakage limit their practical application. Conductive fillers of conductive elastomeric polymers such as conductive silver paste, silver nanowires and MXene nanosheets have limitations such as high cost and complicated preparation process. In particular, the transparency of these devices is limited due to the doping of the conductive filler. In contrast, conductive hydrogels, which combine the electronic properties of conductors with the excellent properties of hydrogels such as high transparency, high stretchability, self-adhesion, biocompatibility, ease of processing, and the ability to customize different shapes, are promising candidates for stretchable electrode materials.

In this chapter, I summarize recent progress and the current status of hydrogels as electrodes in TENG. Particularly, the merits of using hydrogels as electrodes in TENG are discussed. The advanced functions of hydrogel-based TENG (H-TENG) are classified as well as wearable applications of H-TENGs are presented (**Fig. 1-1**). Finally, the

challenges and opportunities for future development of H-TENGs are discussed.

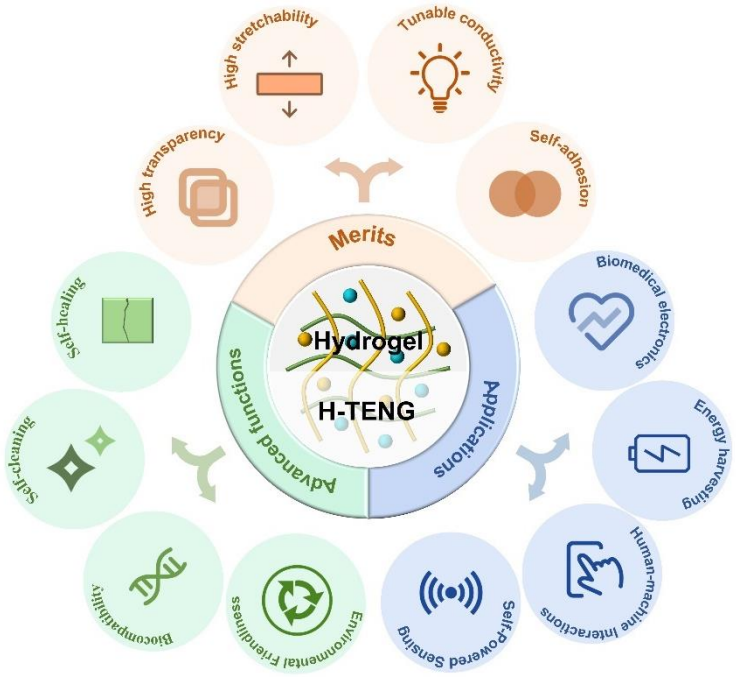

Fig. 1-1. The development of H-TENGs: advanced functions and wearable applications.

1.1 Principle and working mechanism of TENGs

Contact-electrification, also called triboelectrification, is the electricity that occurs when almost any two different materials come into contact with each other. Briefly, as shown in **Fig. 1-2(a)**, the electron clouds of material A and material B, separated by a distance of d, are formed by their outer-shell electrons, which are loosely bonded to the dual potential well formed by their respective atoms. When material A and material B are in contact (**Fig. 1-2(b)**), their electron clouds overlap, allowing electron transfer between the materials. Specifically, atoms with a lower electronegativity (in this case atoms of material A) tend to give electrons to atoms with a higher electronegativity (i.e., atoms of material B). Then, when materials A and B are separated from each other, the transferred electrons will remain in material B due to the energy barrier (**Fig. 1-2(c)**), as long as the

temperature is not too high. On the other hand, at high temperatures, when materials A and B separate, the electrons are more inclined to jump out of the potential well due to the large energy fluctuations of these particles. These jumping electrons either return to the material from which they originated or are emitted into the air (**Fig. 1-2(d)**)[1]. After separating the two materials at a not too high temperature, the transferred charge creates an electric field between them. By utilizing electrostatic induction, the variation in electric field strength generates a flow of electrons when two materials are separated, thereby generating electricity. Therefore, by utilizing contact initiation and electrostatic induction, it is possible to construct a triboelectric device (i.e., TENG).

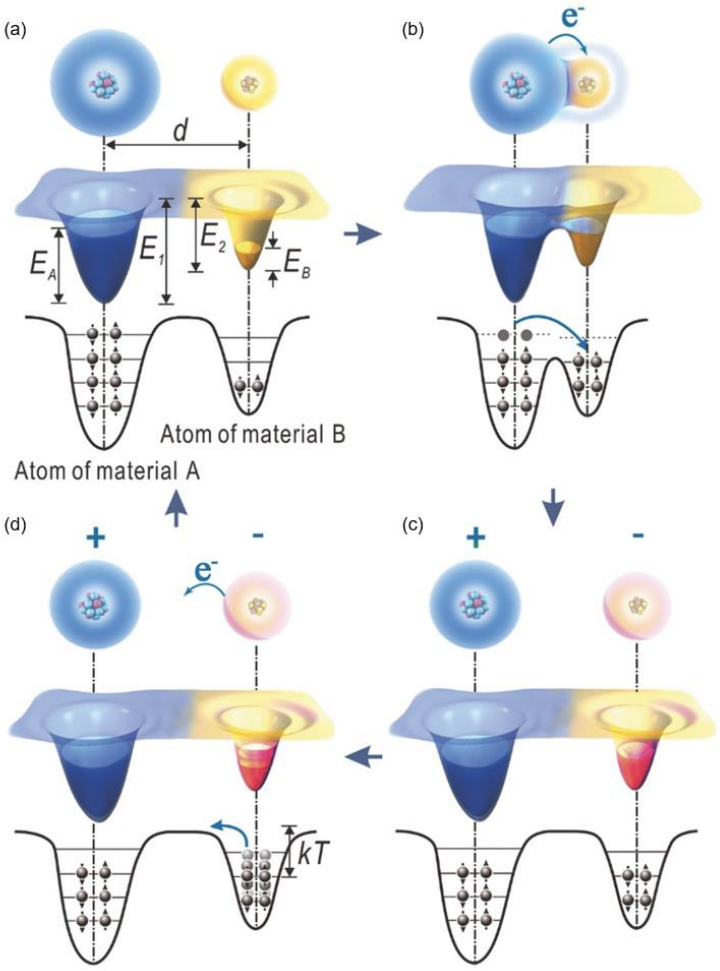

Fig. 1-2. Schematic of an electron cloud-potential well models of two atoms belonging to two materials A and B, respectively. When they are: (a) before contact, (b) in contact, and (c) after contact, show that electrons are forced to have electrons that can overlap after transferring from one atom to another. (d) Once kT approaches the barrier height, the charge is released from the atom at an elevated temperature T. d, distance between two nuclei; EA and EB, occupied energy levels of electrons; E_1 and E_2, potential energies for electrons to escape; k, Boltzmann constant; T, temperature. Reproduced with permission[1]. Copyright 2018, Wiley VCH.

Based on the circuit connection methods and modes of operation, TENGs can be classified into four different working modes (**Fig. 1-3**), i.e., vertical contact–separation (CS) mode, single-electrode (SE) mode, freestanding triboelectric-layer (FT) mode, and lateral-sliding (LS) mode. The vertical CS mode is the most common of the four modes of operation due to its simple structure and method of working, which relies on the relative motion between the two triboelectric layers perpendicular to the interface (**Fig. 1-3(a)**). The SE mode is based on the reference electrode and is versatile in that it can harvest energy from freely moving objects without connecting an electric conductor (**Fig. 1-3(b)**), which has better adaptability in design since no additional electrode layer is required. The FT mode typically generate electrical output through a sliding dielectric layer and the connecting conductive layer located beneath it (**Fig. 1-3(c)**). The LS mode depends on a relative displacement parallel to the direction of the interface (**Fig. 1-3(d)**), producing a relatively stable output that meets the continuous power supply requirements of a wider range of applications. Among them, the single-electrode mode has a simpler structure and is easy to integrate and carry. Therefore, it is widely used in the flexible and

wearable applications.

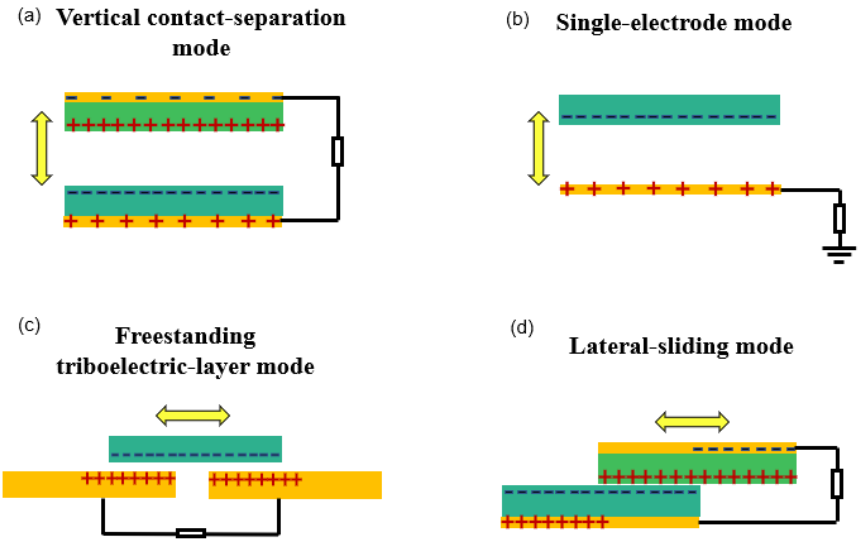

Fig. 1-3. Schematic illustrations of four basic operation modes of triboelectric nanogenerators (TENGs). (a) vertical contact–separation mode. (b) Single-electrode mode. (c) Freestanding triboelectric-layer mode. (d) Lateral-sliding mode.

1.2 Merits of conductive hydrogels as electrodes in TENGs

This section will focus on the advantages of hydrogels as electrodes for TENGs and will also discuss the reasons why they provide additional advantages for TENGs.

1.2.1 High transparency

High optical transparency of hydrogels is prerequisites for wearable devices that demand visualization such as next-generation displays and touch panels, etc. High transparency is also significant for visual transmission of aesthetics in wearable applications of TENGs that can yield a perfect visual fit with the human body. Therefore, many reports have highlighted the high transparency of H-TENGs benefit from the flexible and transparent hydrogel, such as gelatin/sodium chloride (NaCl) organogel (**Fig.**

1-4(a)) prepared by Wu *et al*. [2] and PVA/phytic acid (PA) hydrogel developed by Yang *et al*.[3] With this in mind, transparency can allow for seamless integration with other devices that can take advantage of the H-TENG's energy harvesting and optical information collection. For instance, Guo *et al*. [4] have prepared transparent H-TENG for Touch Panel (**Fig. 1-4(b)**). Liu *et al*. [5] have prepared integrated H-TENG for energy harvesting and touch sensing without optical information loss (**Fig. 1-4(c)**). The development of this feature of transparency may facilitate the integration of the H-TENG energy harvester into larger everyday devices.

1.2.2 High stretchability

Stretchability is an important property of hydrogels that is essential for the fabrication of wearable sensing and monitoring. Some hydrogels can be strained over 2000%. For example, a polyacrylamide (PAM)/carbon quantum dot/lithium magnesium silicate hydrogel was prepared through a chemically cross-linked network, which could achieve an elongation at break of 2480%.[6] In wearable sensing and monitoring applications, high flexibility and stretchability are key factors when choosing materials. In particular, flexible TENGs are often used as strain sensors to monitor human motion. [7-9] Compared with bare hydrogel sensors, H-TENG sensors not only can charge the electronics but also protect the internal hydrogel from contamination/damage and also impede water loss from the hydrogel and are further used in wearable applications. For example, self-polymerized dopamine-Fe-polyacrylic acid hydrogel[10] exhibits outstanding stretchability and toughness, which is able to withstand high levels of extension (up to 2000%) without fracture (**Fig. 1-4(d)**). The gelatin/NaCl organogel

(GNOH) prepared by Wu *et al*.[2] has good flexibility and mechanical strength and can be stretched to three times its initial length even with a knot tied in the fascia (**Fig. 1-4 (e)**). The zwitterionic network hydrogel developed by Guo *et al*. [4] can be stretched to 11500% of its initial length and is considered to be "Ultra-stretchability" (**Fig. 1-4(f)**). Therefore, the H-TENGs could not only be stretched, but also be rolled, folded and twisted without mechanical failure, and maintain a stable and good output.

1.2.3 Tunable conductivity

One of the unique merits of hydrogels over other flexible electrodes is the adjustable conductivity. Typically, conductive hydrogels consist of conductive materials and a network of cross-linked polymers. The polymer network provides the framework, while the conductive material gives the hydrogel its conductivity. The introduction of conductive additives is convenient and allows hydrogel precursors to be tailored for conductivity. To date, a variety of conductive materials including electronically and ionically conductive materials have been used to manufacture conductive hydrogels for wearable devices. Widely used electronically conductive materials include conductive polymers[11-13], carbon-based materials[14-16], MXenes[17-19] and metallic materials[20-22], while ionic materials include polyelectrolytes[23] and electrolytes[24]. Hence, both electronically and ionically conductive materials can be optimized by optimizing the loading and designing a suitable hydrogel network structure to impart the appropriate conductivity to the conductive hydrogels. For example, the addition of 1.3 wt.% poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT: PSS) to γ-polyglutamic acid/ (3-glycidyloxypropyl) trimethoxysilane (γ-PGA/PEDOT:PSS)

hydrogel changed the appearance from transparent to black (**Fig. 1-4(g)**), while the conductivity increased from "1.3 S/m" to 12.5 S/m.[25] The conductivity of dopamine-Fe-polyacrylic acid hydrogel increased with increasing Fe^{3+} content and reached the highest value of 38 S/m (**Fig. 1-4(h)**). [10] Furthermore, even after the hydrogel has been synthesized, the conductivity can still be increased *via* a simple soaking strategy. In the circumstances, the hydrogel is immersed in a salt solution to allow the transfer of ions into the hydrogel. For instance, Wang *et al*.[26] immersed the polyvinyl alcohol (PVA)/hydroxyethyl cellulose hydrogel in a sodium chloride solution; the resulting multi-crosslinked hydrogel possessed a good conductivity of 5.77 S/m at 25 ºC and were used as flexible sensor for human movements monitoring and handwriting recognition.

1.2.4 Adhesion

A neglected point of laminated structured H-TENGs is the triboelectric layer–electrode interfacial integrity, where mechanical deformation and effects of interface slippage and delamination lead to H-TENGs performance deterioration or even dysfunction in practical applications. A common approach is to build all-in-one configurations that consistently rely on complex instrumentation or redundant manufacturing processes. In contrast, self-adhesive hydrogel electrodes engineered with the required compliance and flexibility can form reliable, interfacing contacts with the curvilinear surface of triboelectric layer, when the H-TENGs are subjected to arbitrary deformation, which is crucial for H-TENGs yet challenge. Therefore, much H-TENGs on adhesive hydrogels such as doped with alkali lignin macromolecule[27], lignosulfonate sodium[28] and PEDOT: PSS[29] have been developed (**Fig. 1-4(i-k)**).

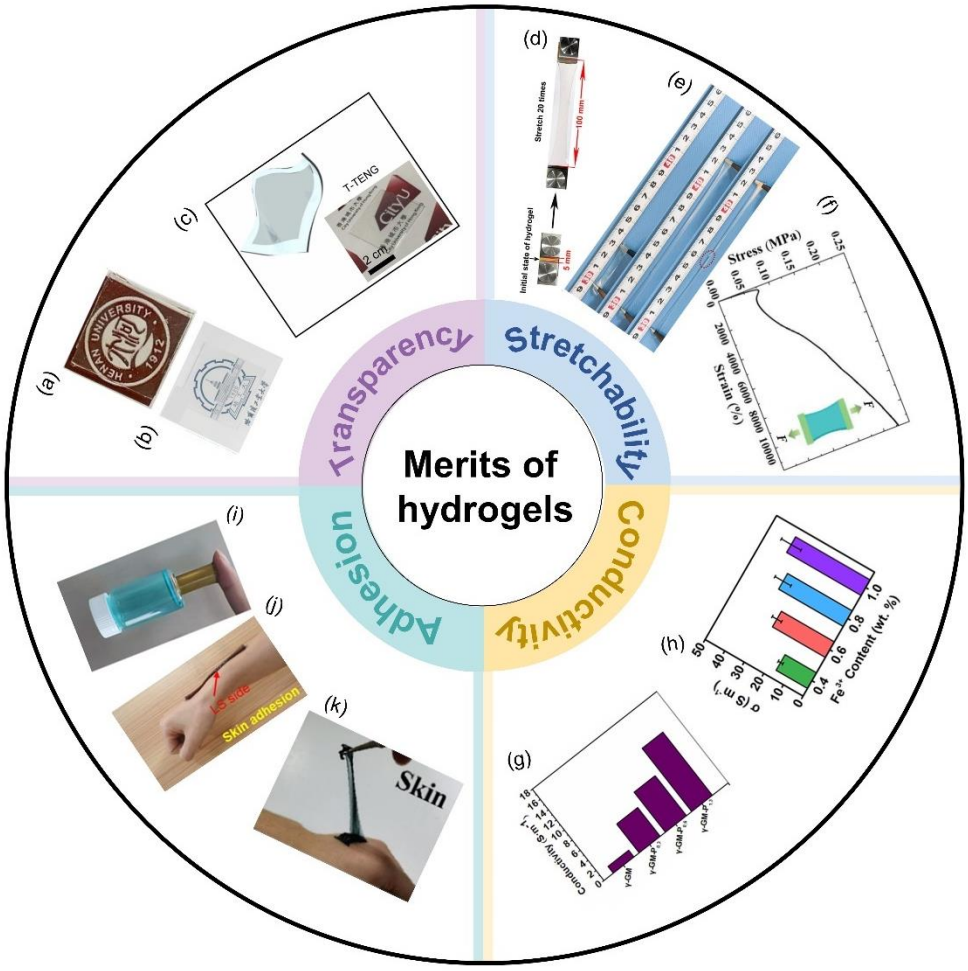

Fig. 1-4. (a) Transparency of the GNOH. Reproduced with permission.[2] Copyright 2022, Elsevier. (b) Transparency of H-TENG. Reproduced with permission.[4] 2022 Wiley-VCH. (c) Schematic illustration and optical image of the H-TENG. Reproduced with permission.[30] Copyright 2022, Elsevier. (d) Digital images of the hydrogel tensile test (0.6 wt.% Fe^{3+}). Reproduced with permission.[10] Copyright 2019, American Chemical Society. (e) The excellent mechanical properties under stretching/knotted stretching. Reproduced with permission.[2] Copyright 2022, Elsevier. (f) Stress–strain curve of the hydrogel. Reproduced with permission.[4] Copyright 2022 Wiley-VCH. (g) Conductivity of γ-PGA/PEDOT: PSS hydrogels. Reproduced with permission. [25] Copyright 2022, Elsevier. (h) Dopamine-Fe-polyacrylic acid hydrogel conductivity as a function of Fe^{3+} contents. Reproduced with permission.[10] Copyright 2019, American Chemical Society. (i-k) Photographs of

the hydrogel adhering on the surface of various substrates. Reproduced with permission. [27-29] Copyright 2022, Wiley-VCH. Copyright 2021, Elsevier. Copyright 2022, Royal Society of Chemistry.

1.3 Advanced functions of hydrogel-based TENGs

The unique properties of hydrogels include self-healing, self-cleaning, biocompatibility, recyclability and degradability endowing H-TENG with some advanced functions.

1.3.1 Self-Healing

Self-healing is a unique property of conductive hydrogels, a concept originated from the function of biological skin and bone.[31] The self-healing behavior of hydrogels is through reversible chemical bonding (e.g., acyl hydrazone, Schiff base, borate ester, and disulfide bonds) and noncovalent interactions (e.g., hydrogen bonds, ionic interactions, metal coordination, host–guest interactions, and hydrophobic interactions). Applying this property to TENG equipment can restore the original performance after damage, thus achieving significant durability and extending the life of the equipment, and reducing maintenance costs. For instance, Liao *et al*. [22] have prepared a PVA-stabilized LMPs (PVA-LMPs) hydrogels with good self-healing ability. The self-healing behavior can lead to the reconstruction of conductive pathways in the PVA-LMPs hydrogel network. Upon contact, some PVA-LMPs could be reconstituted between the broken surfaces of PVA-LMPs hydrogels, and ion diffusion channels are constructed through dynamic cross-linking bonds between PVA and borate ions, so that both electrical and ionic conduction pathways can be restored. (As illustrated in **Fig. 1-5(a)**). Ge *et al*. [32] have reported a

human muscle-inspired self-healing PAA-PANI binary networked-hydrogel (PPBN-hydrogel) (detailed self-healing mechanism for PPBN-hydrogel is shown in **Fig. 1-5 (b)**). Chemical bonds are separated in the interface of the break, and reversible bonding could heal the separated surfaces through metal coordination, "dynamic zipper", and hydrogen bonding. The rejuvenation of the polymer matrix helps to rebuild the interface of the break interface. Gao *et al*.[33] have reported a self-healing polyzwitterion-clay nanocomposite hydrogel *via* noncovalent bonding (as illustrated in **Fig. 1-5(c)**). When two freshly cut hydrogels are placed together, the dangling chains of surfaces are first touched to form the electrostatic attraction between the zwitterion groups, which causes a quick interfacial adhesion and reconstruction of the ion channel. The polymer chains then further diffuse until bridging of adjacent nanoclays occurs at the interface, leading to an increase in healing efficiency. Interestingly, Huang *et al*. [34] have reported a self-healable PAM/clay H-TENGs with good recovery in a broad temperature range. As shown in **Fig. 1-5(d)**, the obtained organogel heals rapidly from -30 to 80 ℃, maintaining the same resistance. The organogel has ultrafast electrical self-healability over a broad range of temperatures, which facilitates applications in electronic devices.

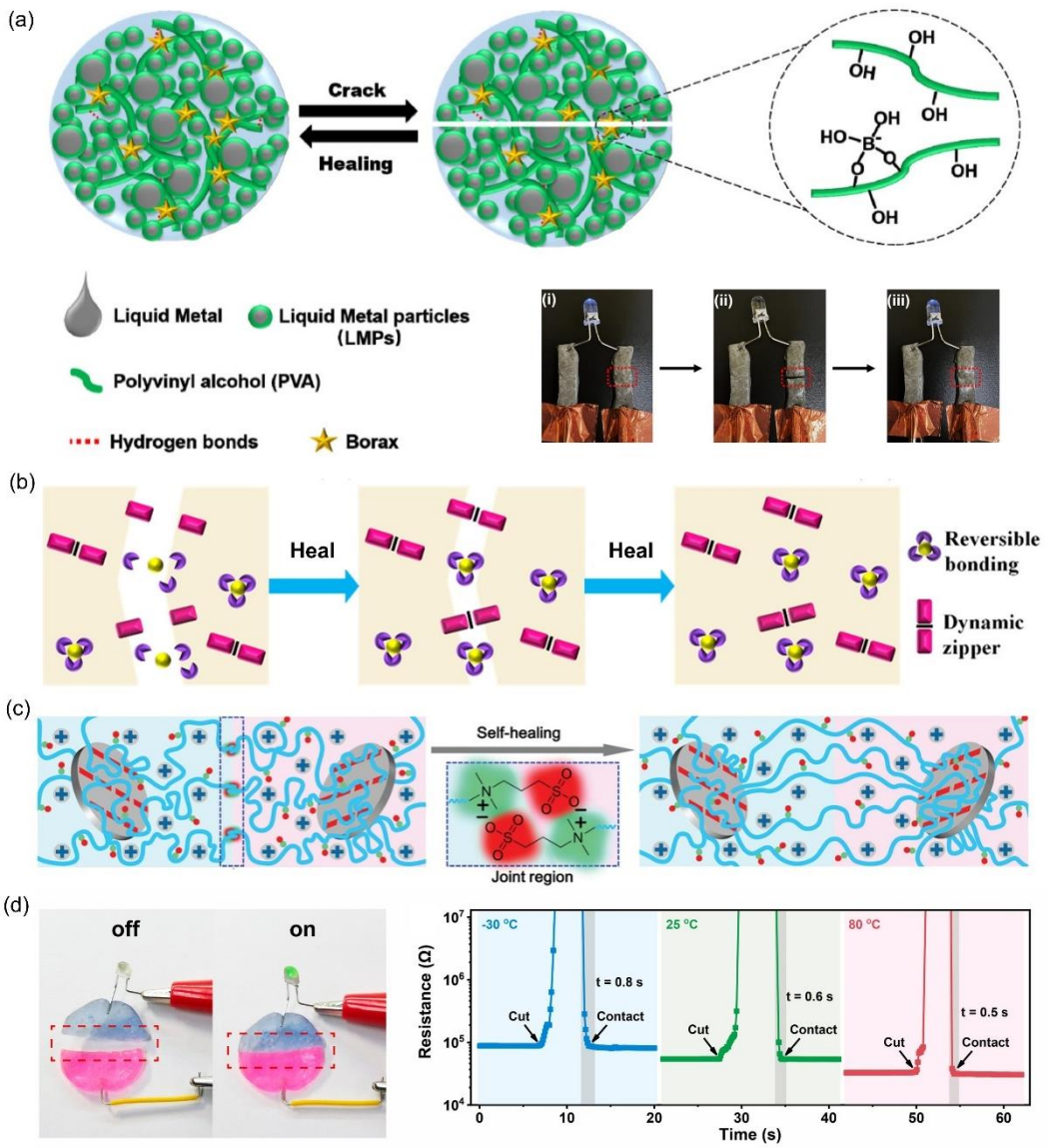

Fig. 1-5. (a) Schematic illustration of PVA-LMPs hydrogel could be reversibly self-healed. Reproduced with permission. [22] Copyright 2019, American Chemical Society. (b) Schematic illustration of the self-healing mechanism for PPBN-hydrogel. Reproduced with permission. [32] Copyright 2019, American Chemical Society. (c) Schematic illustration of cut hydrogels healed by electrostatic attraction between polyzwitterions and rebuilding polyzwitterion–clay adsorption. Reproduced with permission. [33] Copyright 2020, Wiley-VCH. (d) Photographs of damaged and healed PAM-Clay organohydrogel, and resistance variation during the process of cut and contact. Reproduced with permission. [34] Copyright 2021, Elsevier.

1.3.2 Self-Cleaning

Triboelectric layers and hydrogels are susceptible to contamination during daily use. Because of the reduced surface charge density, contaminated TENGs show a lower output. Hence, it is crucial to combine TENGs with self-cleaning capabilities. So far, there has only been one research on the self-cleaning property of H-TENG. Lee *et al*. [35] have prepared a self-cleaning, transparent and attachable ion communicator (STAIC) through chemical functionalization of the PDMS surface. As shown in **Fig. 1-6(a)**, the STAIC with the surface treatment showed good self-healing property compared to STAIC without the surface treatment. Self-cleaning properties were achieved by surface treatment, resulting in recovery of transmittance (40.6% increase) and electrical output voltage (19.4% increase) compared to the STAICs without the surface treatment after simulated dust coverage (**Fig. 1-6(b-c)**).

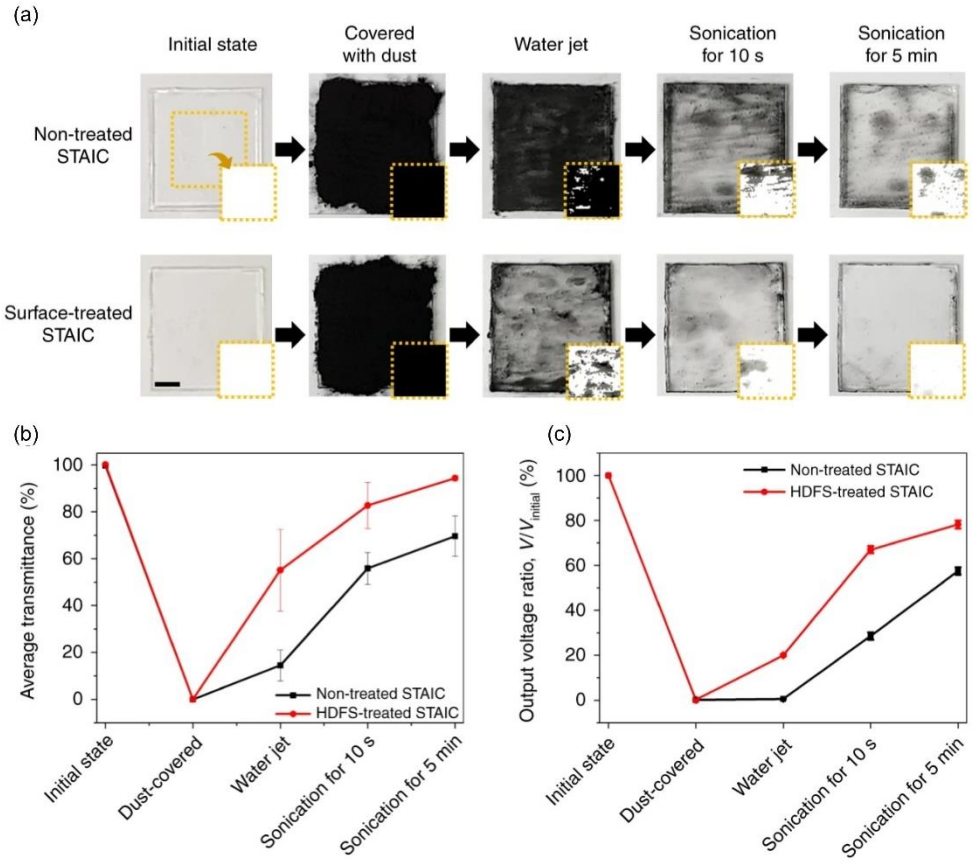

Fig. 1-6. (a) Self-cleanability of HDFS-treated STAICs was examined by cleaning surfaces contaminated by activated charcoal powders. (b) The average transmittance at 550 nm was measured according to the flow. Ten points on STAICs were randomly probed to calculate the average transmittance. (c) Output voltages of STAICs were studied according to the cleaning processes. Reproduced with permission. [35] Copyright 2018, Springer Nature.

1.3.3 Biocompatibility

Wearable or implantable devices come into direct contact with the human body and are not expected to cause harm to human health. Wearable or implantable devices with biocompatibility endows them with safety and long-term usability in on-body applications. Therefore, biocompatibility is critical concern for TENGs in on-body

applications. Because of their unique hydration environment and adjustable physicochemical properties, conductive hydrogels have been widely used as good candidates for biocompatible materials for a variety of biological devices such as biosensors, tissue engineering, artificial skin, drug delivery and cell culture. For instance, As shown in **Fig. 1-7(a)**, the flexible edible triboelectric hydrogel sensor fabricated by Guo *et al*. [36] includes three layers of gelatin, agar hydrogel, and seaweed for full range of infant movement monitoring. Since all materials of the flexible biocompatible sensor are edible, the risk of accidental infant ingestion can be fundamentally eliminated. Chen *et al*.[37] prepared an implantable, programmable and battery-free neurostimulators based on H-TENG. **Fig. 1-7(b)** presents live/dead fluorescence staining showing that cells on the H-Teng device and on the PAM/graphene hydrogel exhibit similar density and morphology to the reference cells. Electroceuticals based on electrical stimulation of the vagus nerves show great promise for therapeutic applications in a wide range of diseases and clinical conditions. Gogurla *et al*. [38] prepared an artificial energy-generating skin (EG-skin) by using silk hydrogel. It provides a method for implementing a soft, stretchable and biocompatible energy harvester in a bioelectronic device (**Fig. 1-7 (c)**). Du *et al*. [39] prepared a single-electrode H-TENG E-skin patch through utilizing conductive and photothermal hydrogels for sensing motion and promoting wound healing. The immunofluorescence staining images in **Fig. 1-7(d)** confirmed that the in vitro biocompatibility of H-TENG E-skin patch, indicating that this is biologically safe.

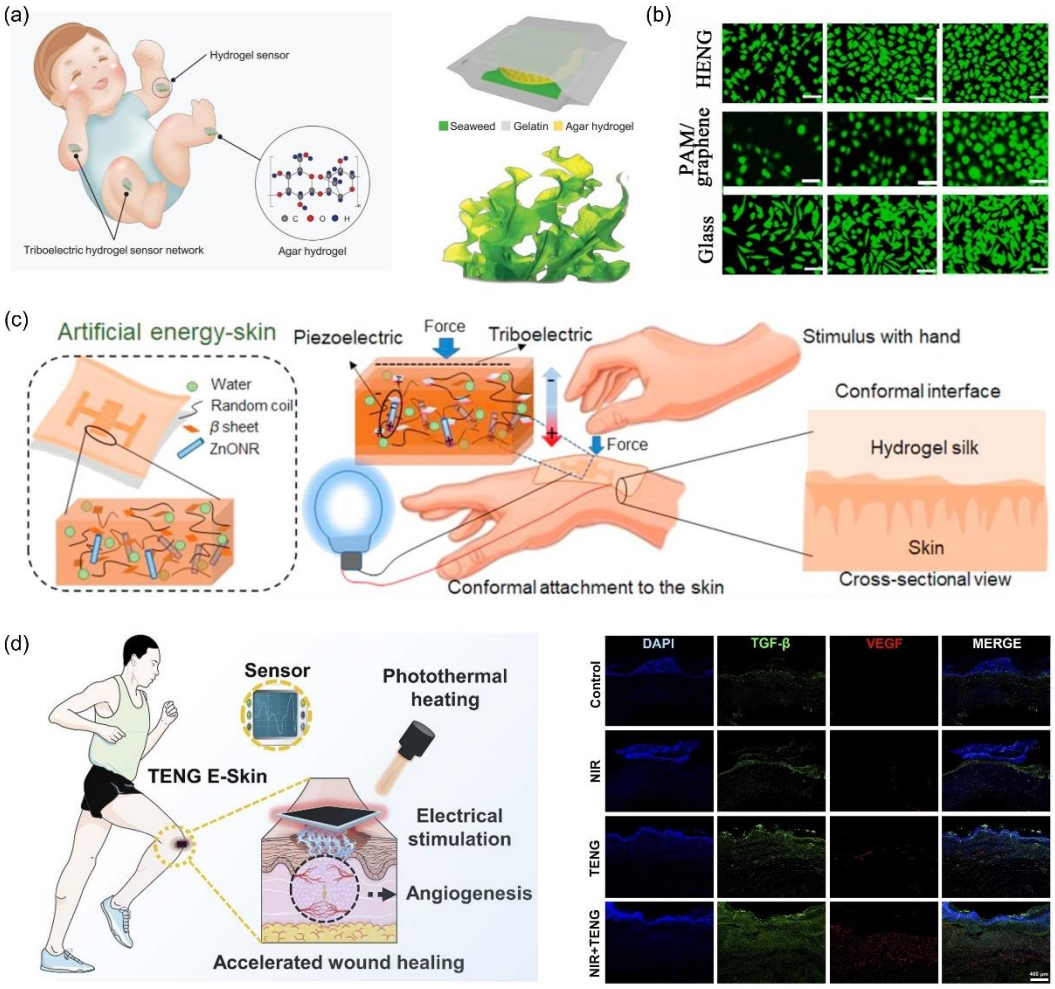

Fig. 1-7. (a) Schematic diagram of a flexible edible triboelectric hydrogel sensor for body area sensor network system for infant movement monitoring. Reproduced with permission.[36] Copyright 2022, Wiley-VCH. (b) Live/dead cellular staining of the mouse fibroblast (L929) seeded on H-ENG, PAM/graphene hydrogel and glass plates as control. (Scale bar = 100 μm). Reproduced with permission.[37] Copyright 2021, Elsevier. (c) Schematic illustration of working principle of artificial EG-skin using silk hydrogel. Reproduced with permission. [38] Copyright 2020, Elsevier. (d) Functions of the wearable H-TENG E-skin patches in the wound and immunofluorescence images of stained nucleus that were cultured in wound tissues among different groups. Reproduced with permission.[39] Copyright 2022, Elsevier.

1.3.4 Environmental friendliness

E-waste already has a negative impact on the environment and human health. Environmental friendliness is therefore a parameter correlated with the influence of recent/future materials that we exploit and implement into next generation technology. The development of TENG based on degradable materials strongly facilitates the next generation of green energy techniques that will effectively prevent the pollution and harms resulting from metals and virtually non-degradable plastic materials. We therefore need to develop environmentally friendly electronics, which depends on emerging recyclable and biodegradable polymer technologies. The typical degradable materials for TENG are animal-based degradable materials (e. g., chitosan/chitin, silk, egg white and derivatives, peptides, and gelatin), plant-based degradable materials (e. g., paper, cellulose, leaves, rice paper (starch), wood, and alginate.) and artificial degradable materials (e. g., PVA, PHB/V, PLA and PCL). For example, Kim *et al*. [40]developed a simple method for preparing a hyaluronic acid (HA) hydrogel membrane and fabricating biocompatible, biodegradable, and high-performance TENGs based on a HA hydrogel film (HA-TENGs). The HA-TENG may be a proving of concept for future implantable and biodegradable devices (**Fig. 1-8(a)**). Xu *et al*. [41] presented a novel H-TENG with complete flexibility and environmental friendliness by utilizing physically linked PVA hydrogel as substrate materials. The utilization of PVA hydrogel with biodegradability shows the recyclability, which affords the H-TENG with merits of environmental friendliness and low cost (**Fig. 1-8(b)**). Zhang *et al*. [42] assembled a biodegradable TENG *via* using chitin hydrogel films, which provides new insights for the utilization of

marine wastes (**Fig. 1-8(c)**). Ghosha *et al*. [43] demonstrated an entirely biodegradable ferroelectric nanogenerator-driven skin sensor based on eatable porcine skin gelatin, demonstrating potential for wearable electronics with zero waste fingerprint (**Fig. 1-8(d)**).

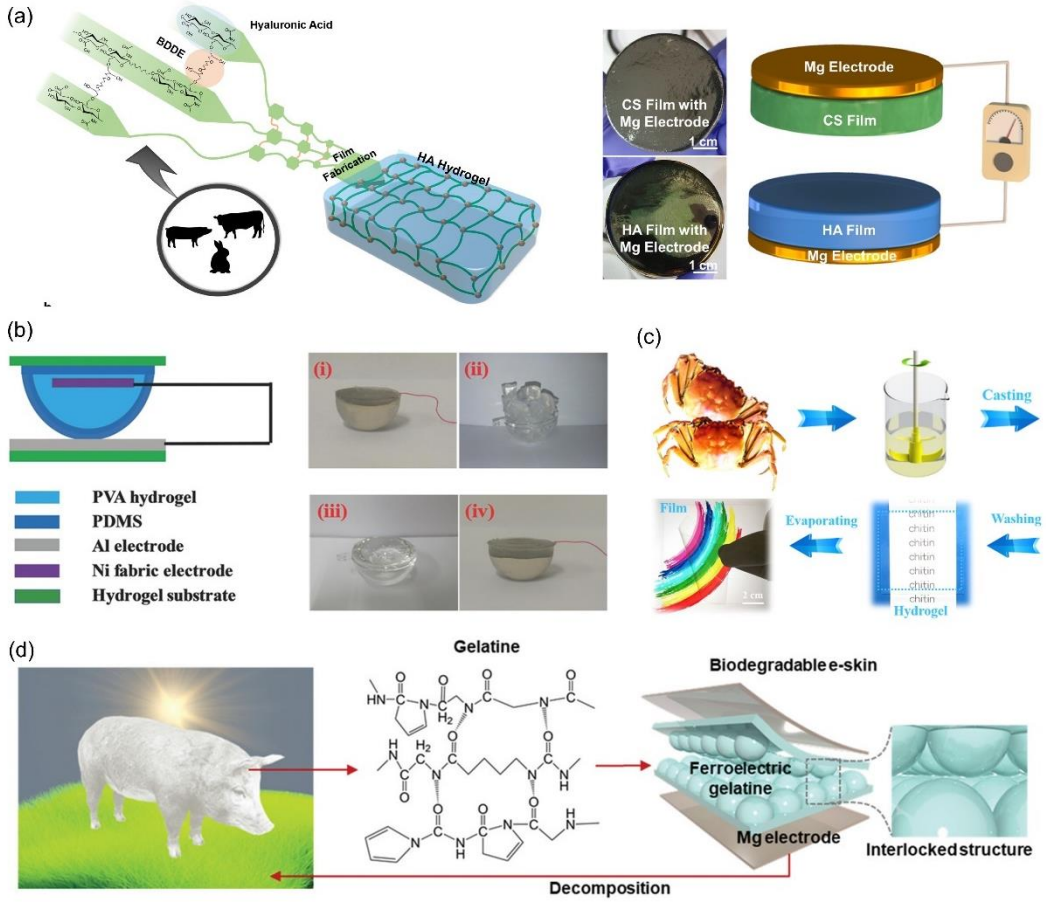

Fig. 1-8. (a) Schematic illustration and working principle of biocompatible and biodegradable TENGs based on HA hydrogel films. Reproduced with permission. [40] Copyright 2021, Elsevier. (b) Schematic of the PVA H-TENG and photograph of the recycling process of PVA hydrogel. Reproduced with permission. [41] Copyright 2016, Wiley-VCH. (c) Preparation process of chitin hydrogel film. Reproduced with permission. [42] Copyright 2022, Elsevier. (d) Schematic diagram showing the concept of gelatin-based transient "green" TENG from biosynthesis to decomposition. Reproduced with permission. [43] Copyright 2021, Wiley-VCH.

1.4 Applications of hydrogel-based TENG devices

Similar to other flexible TENGs, H-TENGs are widely applied for self-Powered monitoring/sensing and energy harvesting. In addition, they can also be used in other wearable applications such as human-machine interactions and bio-medical electronics.

1.4.1 Self-Powered sensing/monitoring

H-TENGs have been used in a wide range of different types of sensors for wearable applications such as biomechanical motions monitoring, temperature sensing, pressure sensing, self-powered sweat sensing, and smart farming.

For instance, Kim *et al*. [44] have prepared a bio-hydrogel based on all ocean biomaterials and integrated biocompatible catechol-chitosan-diatom hydrogel (CCDHG) electrode for assembly of CCDHG-TENG and M-shaped film and to develop diagnostics for Parkinson's disease (**Fig. 1-9(a)**). As shown in **Fig. 1-9(b),** Ghosh *et al*. [43] demonstrated an entirely biodegradable and ferroelectric gelatin e-skin nanogenerator with the capability of sensing temperature and pressure. Sheng *et al*. [45] have prepared sodium alginate/zinc sulfate/poly acrylic-acrylamide (SA-Zn) H-TENGs as a training band sensor to perform real-time motion monitoring. As shown in **Fig. 1-9(c)**, the self-powered intelligent belt system composed of signal collection, processing system and a real-time visualization. This device offers a wide range of promising possibilities for the development and applications of self-powered health and exercise monitoring platforms. Qin *et al*. [46] have fabricated an entirely flexible self-powered sweat sensor on the basis of the 2,2,6,6-tetramethylpiperidine-1-oxyl radical (TEMPO)-oxidized CNFs/

polyaniline-PVA/ borax hydrogel (CPPH). Wireless transmission of sensor output to the mobile phone application offers new possibilities for convenient real-time health monitoring (**Fig. 1-9(d)**). Hsu *et al*.[47] has successfully developed a smart farming system consisting of H-TENG and hydrogel supercapacitors based on versatile polyacrylic acid/ reduced graphene oxide/ polyaniline (PRP) hydrogel. PRP-TENG can harvest clean energy, for example, acoustic energy, wind energy and rainfall, and convert it to electricity and stored it in the hydrogel supercapacitor to provide power for plant growth boosting, indicating its potential for green agriculture (**Fig. 1-9(e)**).

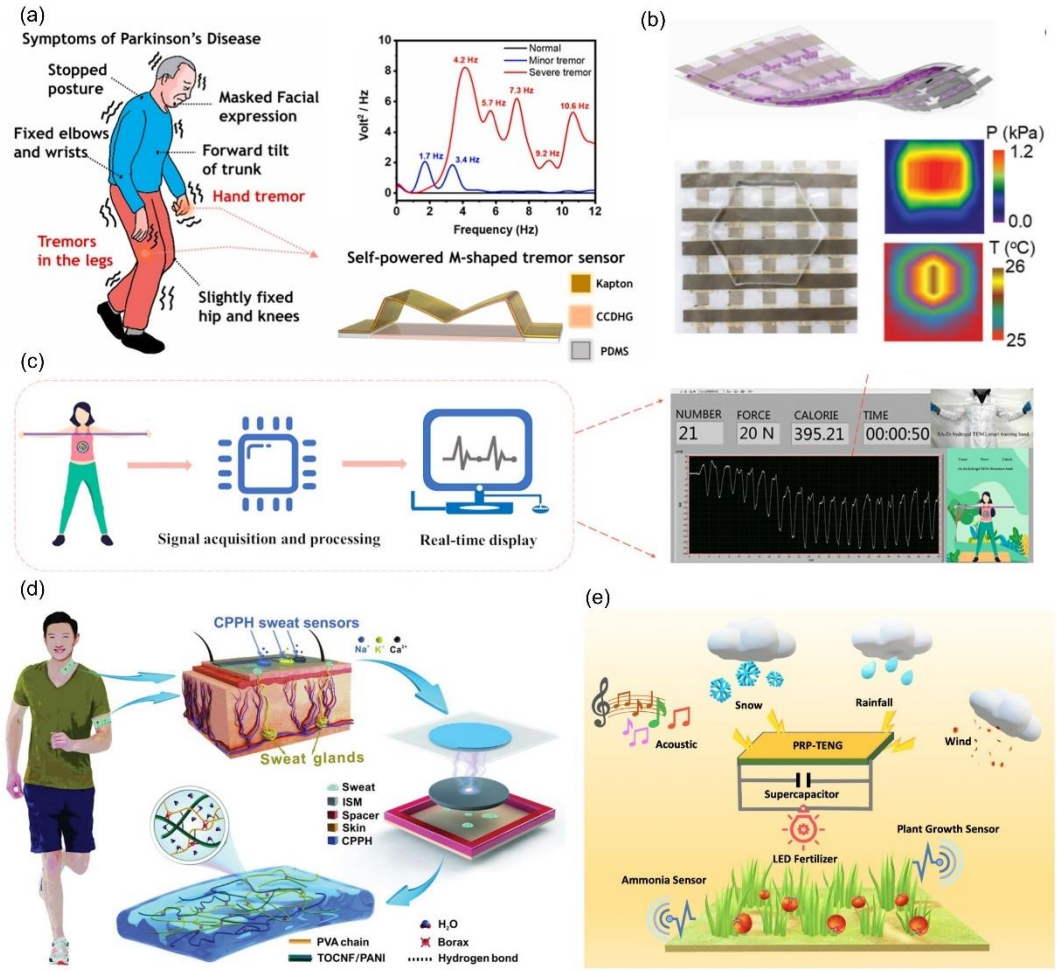

Fig. 1-9. (a) Typical symptoms of Parkinson's disease, schematic image of tremor sensor and the power spectral density of the voltage signal from tremor sensor with different motions. Reproduced

with permission. [44] Copyright 2021, Elsevier. (b) Schematic diagram of the sensor array matrix and Photograph of the sensor network with object placed on it to plot the spatial distribution of pressure (P) and temperature (T) upon contact. Reproduced with permission. [43] Copyright 2021, Wiley-VCH. (c) Demonstration in the application of the SA-Zn-TENG intelligent training band. Reproduced with permission. [45] Copyright 2021, American Chemical Society. (d) Schematic of self-powered sweat sensing based on CPPH-TENG. Reproduced with permission. [46] Copyright 2022, Wiley-VCH. (e) Schematic of a smart farming system based on PRP-TENG. Reproduced with permission. [47] Copyright 2021, Elsevier.

1.4.2 Energy harvesting

Converting biomechanical energy from physical motions into electrical energy is one of the most versatile and critical applications of TENGs. The electricity derived from H-TENGs harvesters holds promising applications for powering and charging wearable electronic products.

Fig. 1-10(a) schematically shows a typical versatile circuit with rectifier and capacitor. On the one hand, the circuit can be linked directly to the LED in order to light it up immediately. On the other hand, the circuit can be linked to a capacitor to store the electrical energy created from the TENG and then for powering[27]. Under tapping with one hand at a frequency of 2 Hz, the cellulose-derived ionic H-TENG proposed by Lu *et al*. [48] can charge a capacitor with an increased capacitance from 1 μF to 10 μF (**Fig. 1-10(b)**) and drive the watch with the assistance of the capacitor (**Fig. 1-10(c)**). Much literatures on H-TENG have reported that energy of the tapping movement is adequate to

illuminate LEDs linked in series, as demonstrated in **Fig. 1-10(d-e)**. [49, 50] Sun *et al*. [27] proposed an organogel TENG (O-TENG), which has been utilized to charge a 2.2 μF capacitor and can further use the stored electricity to power an electronic watch (**Fig. 1-10(f)**). Li *et al*. [51] fabricated a transparent solid polymer electrolyte (SPE) TENG (SPE-TENG) based on phosphoric acid/polyvinyl alcohol hydrogel. The SPE-TENG can generate electricity with gentle finger tapping and stored it in a 22 μF capacitor capable of turning on a commercial hygrothermograph, as shown in **Fig. 1-10(g)**. The electricity generated by poly(lipoic acid)-based organohydrogel TENG proposed by Khan *et al*.[52] could be stored in a capacitor to power up calculator devices (**Fig. 1-10(h)**). The Ag@rGO/PVA-PAAm organohydrogel-based TENG (Ag@rGO/ PVA-PAAm-TENG) proposed by Zhang *et al*. [53] could harvest biomechanical energy and then power electronic watch in snowing weather, verifying its potential at subzero temperature(**Fig. 1-10(i)**).

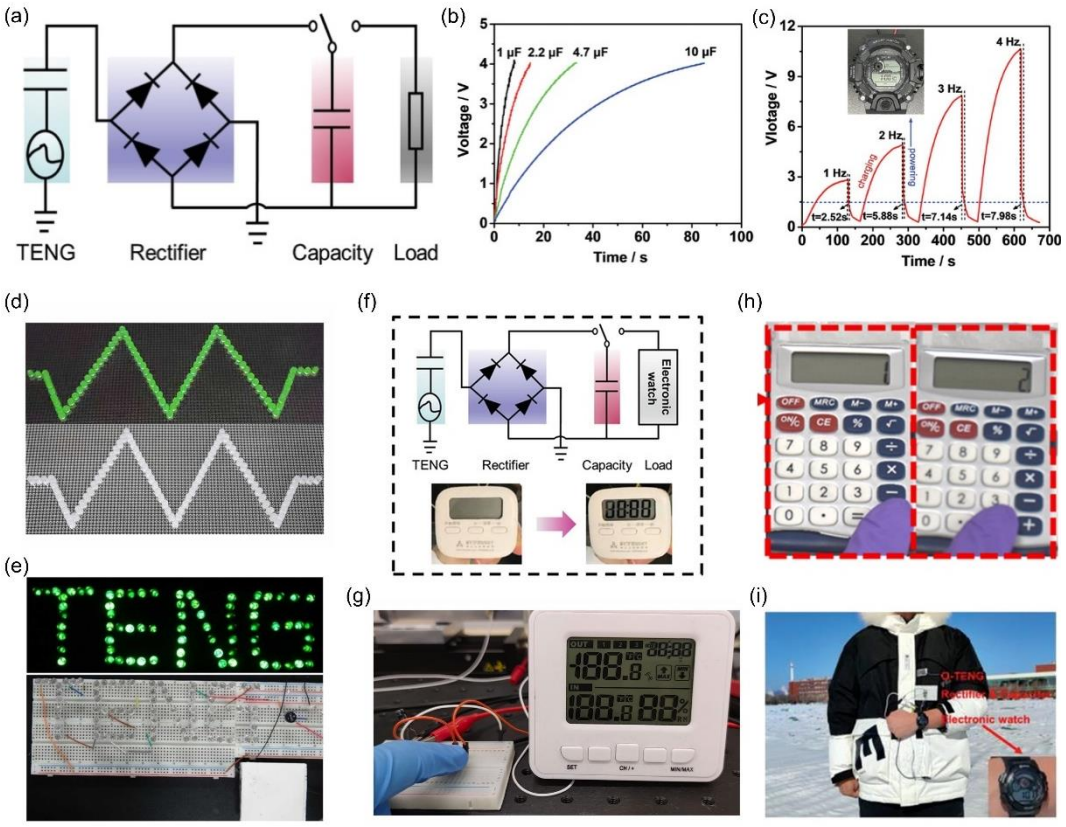

Fig. 1-10. (a) Equivalent circuit diagram of the self-charging power system based on H-TENG. Reproduced with permission.[27] Copyright 2022, Wiley-VCH. (b) The charging performance of the cellulose-derived ionic hydrogel-based H-TENG for the different capacitors. Reproduced with permission.[48] Copyright 2022, Wiley-VCH. (c) The charging/discharging performance of cellulose-derived ionic H-TENG for the 4.7 μF capacitor under different frequencies. Reproduced with permission.[48] Copyright 2022, Wiley-VCH. (d-e) Photograph shows the H-TENG applied to power LEDs. Reproduced with permission.[49] Copyright 2021, Elsevier. Reproduced with permission. [50] Copyright 2022, Elsevier. (f) Photographs of the O-TENG powering an electronic watch. Reproduced with permission.[27] Copyright 2022, Wiley-VCH. (g) A commercial temperature/humidity meter powered by the SPE-TENG. Reproduced with permission.[51] Copyright 2021, Elsevier. (h) Photographs of the harvested energy to drive a calculator. Reproduced with permission. [52] Copyright 2021, Elsevier. (i) Demonstration of the Ag@rGO/PVA-PAAm-TENG for harvesting biomechanical

energy at subzero temperatures. Reproduced with permission.[53] Copyright 2022, Wiley-VCH.

1.4.3 Human-Machine interactions

A user-interactive interface offers a way to communicate between the human beings and the machine.[54] User-interactive interfaces is capable of sensing external stimuli and providing interactive response to the users.[55] Flexible and wearable human-machine interfaces (HMIs) such as artificial skins, use-interactive interfaces, and wearable medical equipment are being developed rapidly in recent years. The integration of H-TENGs into HMI systems has attracted research interest as hydrogels show the advantages of excellent transparency, biocompatibility, flexibility, stretchability and self-healing properties, while TENGs are a unique power source.

Guo *et al*.[56] have proposed a flexible transparent artificial electroreceptor consisting of the starch-based polyacrylamide/LiCl organogel, PDMS elastomers and elastomeric electret. A touchless control pad managed by the electroreceptor can be applied to operate the popular electronic game Super Mario. During a coronavirus disease (COVID-19) pandemic, touch-based HMI is a high risk and potential vector for a variety of bacteria and viruses. Therefore, HMIs that beyond the contact-mode paradigm and customized terminal designs have great potential of interaction in the future **(Fig. 1-11 (a))**. Yang *et al*. [3] reported a PVA/PA H-TENG that can be applied to medical nursing HMI system fixed on patient's fingers. The demand for distress calls such as the nursing message of "hungry", "help", and "thirsty" can be transmitted with just a gentle bend finger in the diagnosis (**Fig. 1-11(b)**). Another example about HMI systems can be traced

in the Wu *et al*.'s study[57], where an ion-conducting organogel-based multifunctional HMI (MHMI) was presented. The one-dimensional (1D) MHMIs can be utilized as a linear controller, and the two-dimensional (2D) MHMI can operate with a simple touch to operate the motion of external devices in a 2D coordinate system (**Fig. 1-11(c)**). Tao *et al*. [58] fabricated a self-powered tactile hydrogel sensor (THS) using a micro-pyramid-patterned PDMS membrane acting as electrification layer and a carrageenan/acrylamide/lithium bromide hydrogel as the electrode. THS with a good self-power sensing capability is utilized as a switching button to operate the robot hand through the intuitionistic signal processing method. As shown in the **Fig. 1-11(d)**, the robotic hand could be actuated to grasp the mango and the coil under the control of the THS, respectively.

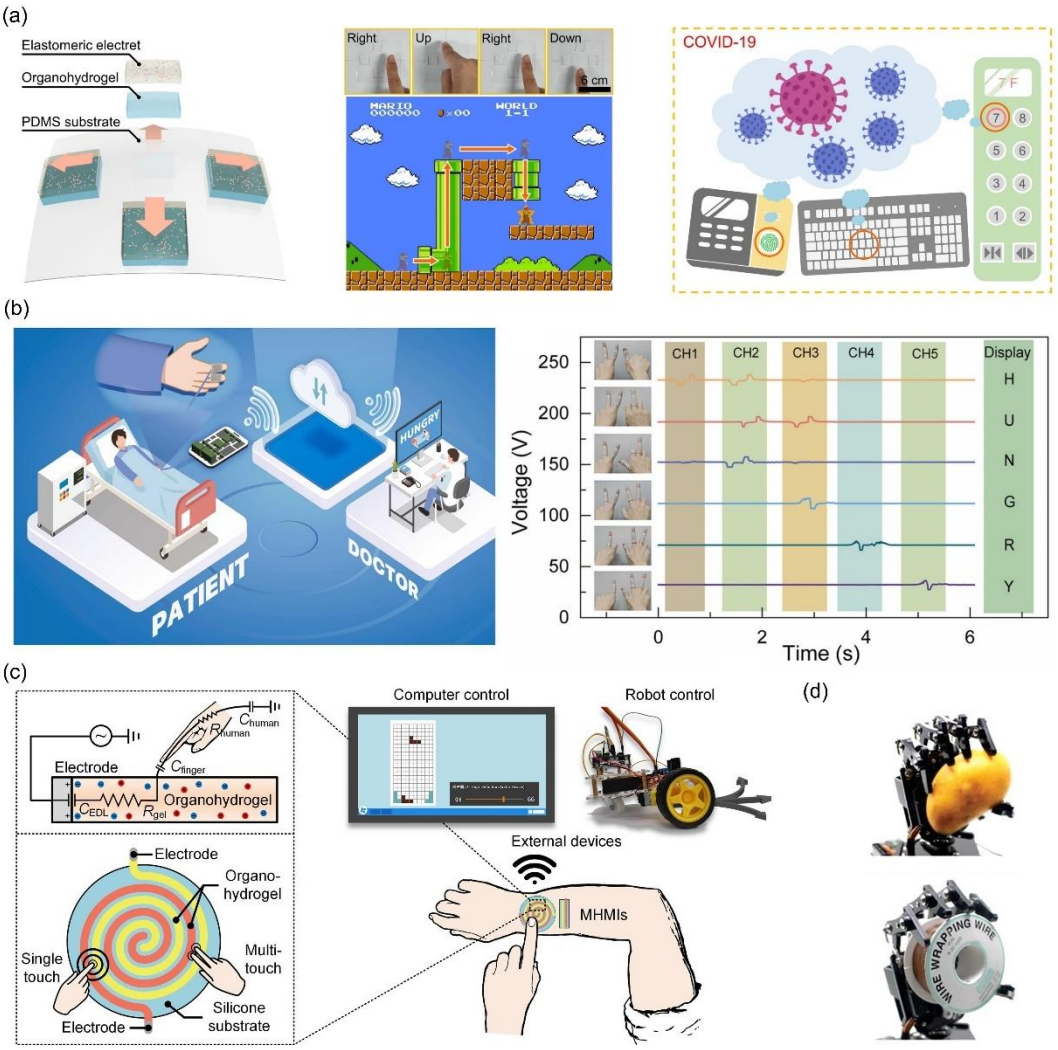

Fig. 1-11. (a) The schematic diagram of touchless control pad and demonstrations of playing Super Mario using touchless control pad and envisioned application of the touchless control pad to prevent the risk of viruses during the COVID-19 pandemic. Reproduced with permission.[56] Copyright 2022, Science. (b) Schematic of a medical nursing system for delivering the patient's help message such as "hungry". Reproduced with permission.[3] Copyright 2022, Elsevier. (c) Schematic diagram of organogel-based MHMI and touch detection principle of MHMI and various applications of MHMI. Reproduced with permission.[57] Copyright 2022, Elsevier. (d) The photograph of grabbing a mango and a coil by robot hand under the control of THS. Reproduced with permission.[58] Copyright 2022, Wiley-VCH.

1.4.4 Biomedical/Implantable electronics

Flexible TENGs exhibit great potential for power supply in biomedical devices, promotion of cell proliferation and differentiation, drug delivery, and direct electrical stimulation. Owing to the unique merits of biocompatibility and biodegradability, hydrogels are commonly utilized in implantable and biomedical applications. Accordingly, some researchers have also regarded H-TENGs as wearable and implantable biomedical application.

As shown in **Fig. 1-12(a)**, Sheng *et al*.[59] developed a TENG (OFS-TENG)-based organogel/silicone fiber-helical sensor with stability, stretchability and biocompatibility, which can be applied for implantable real-time ligament strain monitoring. Jeong *et al*. [60] proposed an interwoven design of ion-containing gel fibers wrapped with elastomers, thus enabling wearable iTENG patch to accelerate wound healing (**Fig. 1-12(b)**). The H-TENG with photothermal properties prepared by Yang *et al*.[61] was composed of a PVA/sodium borate/carbon nanotubes (CNTs) modified *via* poly(dopamine) (PDA) (PDA-CNTs) hydrogel sandwiched between two self-healing silicone elastomeric membranes. The PDA-CNT in the hydrogel offers effective photothermal therapy for the H-TENG, which would help the recovery of human injury under near-infrared (NIR) laser irradiation. Photothermal therapy is wound radiation with a NIR laser to raise the wound temperature to facilitate microcirculatory blood flow for wound pain relief. Temperature of PDA-CNTs/PVA hydrogels and TENG rose significantly above 60 ^{o}C under NIR radiation, while the temperature of the pure PVA hydrogels varied from 20 to 29.3 ^{o}C (**Fig.**

1-12(c)). Nguyen *et al*. [62] developed an engineering electroactive dressing comprising a layer of polydopamine-crosslinked carboxymethyl chitosan conductive hydrogel and interdigitated array (IDA) electrodes. The electroactive dressing designed to promote wound healing. The monitoring system integrated with IDA electrode and a self-powered TENG that detects resistance or output current on the injured tissue for non-invasive real-time monitoring of the entire healing process (**Fig. 1-12(d)**).

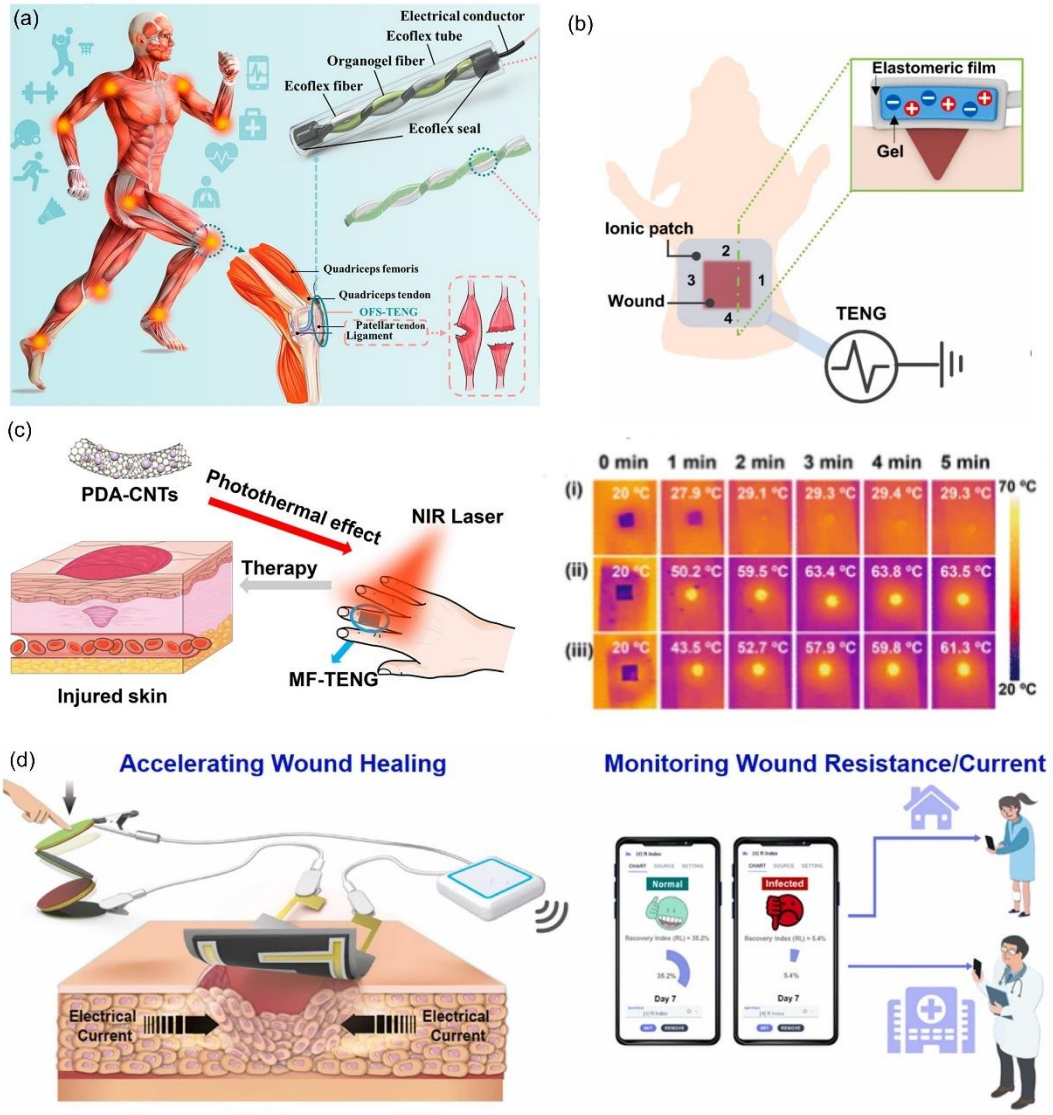

Fig. 1-12. (a) Application and architecture of the OFS-TENG. Reproduced with permission.[59] Copyright 2022, American Chemical Society. (b) Schematic of the iTENG patch applied to the wound

site. Reproduced with permission.[60] Copyright 2021, Elsevier. (c) Simulation of the photothermal treatment of H-TENG. Reproduced with permission.[61] Copyright 2021, American Chemical Society. (d) The wound monitoring system integrated with smart electroactive dressings and a self-powered TENG that simultaneously facilitate wound healing and non-invasively monitor the progress of healing. Reproduced with permission.[62] Copyright 2022, Elsevier.

1.5 Purpose and significances of research

The rising popularity of next-generation of portable and wearable electronics is stimulating a rise in demand for power supplies. Nevertheless, the rigid/complex configuration and large size of most traditional power supplies no longer meet the critical requirements of wearable electronics. Consequently, flexible TENGs have attracted much interest due to its simple structure and manufacturing, light weight, and low cost. Apart from triboelectric materials, electrodes are also important for integration of flexible TENG in wearable applications. Typically, there are four types of electrodes in flexible TENGs: i) metal plates; ii) carbon sheets; iii) conductive polymer films; and iv) hydrogel films. Despite the high electrical conductivity, metal plates are usually not suitable for flexible and wearable due to limited flexibility and stretchability. Carbon sheets have poor electrical conductivity despite their lower cost. Similar drawbacks plague conductive polymer films, which show poor conductivity despite better stretchability. Hydrogels that combine the integrated attributes of high transparency, high stretchability, biocompatibility, and self-healing abilities, and have thus emerged as popular materials from which to fabricate wearable TENGs. Therefore, this research aimed to develop

conductive hydrogels, which was then incorporated into flexible TENG suitable for wearable application.

1.6 Outline of dissertation

In this study, two strategies were developed to assemble conductive hydrogel into flexible TENG for energy harvesting and self-powered sensing.

In chapter 2, a dual-network polyacrylamide (PAAM)/ polyacrylic acid (PAA)/ graphene (GR)/ poly (3,4-ethylene dioxythiophene): poly (styrene sulfonate) (PEDOT: PSS) (MAGP) conductive hydrogel composed of dual-cross-linked PAAM and PAA as well as PEDOT: PSS and GR as conducting component. A wearable strain sensor is fabricated by sandwiching the MAGP hydrogels between two dielectric carbon nanotubes (CNTs)/ Poly (dimethylsiloxane) (PDMS) layers, which can be utilized to monitor delicate and vigorous human motion. In addition, the hydrogel-based sensor can act as a deformable triboelectric nanogenerator (D-TENG) for harvesting mechanical energy to power small electronics.

In chapter 3, a scalable fabrication of core-sheath-structured elastomer triboelectric fibers that combine silicone hollow tubes with gel-electrodes is presented. Gel-electrodes were fabricated *via* a facile freeze–thawing process of blending polyvinyl alcohol (PVA), gelatin, glycerin, poly (3,4-ethylene dioxythiophene): poly (styrene sulfonate) (PEDOT: PSS), and sodium chloride (NaCl). Such fibers can also be knitted into deformable triboelectric nanogenerator textiles with high electrical outputs, which could work as reliable power supplies for small electronics. Moreover, we demonstrate fabric materials

recognition, Morse code communication, and human-motion-recognition capabilities.

In chapter 4, based on the novel covalent-like hydrogen bond engineering, we developed a syrup-modified gelatin/Ag-hydrogel, which is entirely derived from natural or safe constituents and highly stretchable, yet fully biodegrades or recyclable. Meanwhile, strong covalent-like hydrogen bonds endow it with several favorable features of adhesion, rapid self-healing and antifreeze. A series of multi-modal application scenarios are further demonstrated to verify the adaptability of this platform that integrates various attributes.

In chapter 5, the conclusions were presented.

Chapter 2

Stretchable, adhesive, self-healable, and conductive hydrogel-based deformable triboelectric nanogenerator for energy harvesting and human motion sensing

Chapter 2: Stretchable, adhesive, self-healable, and conductive hydrogel-based deformable triboelectric nanogenerator for energy harvesting and human motion sensing

2.1 Abstract

Hydrogels that combine the integrated attributes of being adhesive, self-healable, deformable, and conductive show great promise for next-generation soft robotic/energy/electronic applications. Herein, we reported a dual-network polyacrylamide (PAAM)/ polyacrylic acid (PAA)/ graphene (GR)/ poly (3,4-ethylene dioxythiophene): poly (styrene sulfonate) (PEDOT: PSS) (MAGP) conductive hydrogel composed of dual-cross-linked PAAM and PAA as well as PEDOT: PSS and GR as conducting component that combines these features. A wearable strain sensor is fabricated by sandwiching the MAGP hydrogels between two dielectric carbon nanotubes (CNTs)/ Poly (dimethylsiloxane) (PDMS) layers, which can be utilized to monitor delicate and vigorous human motion. In addition, the hydrogel-based sensor can act as a deformable triboelectric nanogenerator (D-TENG) for harvesting mechanical energy. The D-TENG demonstrates peak output voltage and current of 141 V and 0.8 μA, respectively. The D-TENG could easily light 52 yellow LEDs simultaneously and demonstrated the capability to power small electronics, such as a hygrometer thermometer.

Chapter 3

Deformable textile-structured tribo-electric nanogenerator knitted with multifunctional sensing fibers for biomechanical energy harvesting

Chapter 3: Deformable textile-structured triboelectric nanogenerator knitted with multifunctional sensing fibers for biomechanical energy harvesting

3.1 Abstract

Fibers and textiles that harvest mechanical energy via the triboelectric effect are promising candidates as power supplies for wearable electronics. However, triboelectric fibers and textiles are often hindered by problems such as complex fabrication processes, limited length, performances below the state-of-the-art of 2D planar configurations, etc. Here, we demonstrate a scalable fabrication of core-sheath-structured elastomer triboelectric fibers that combine silicone hollow tubes with gel-electrodes. Gel-electrodes were fabricated via a facile freeze–thawing process of blending polyvinyl alcohol (PVA), gelatin, glycerin, poly (3,4-ethylene dioxythiophene): poly (styrene sulfonate) (PEDOT: PSS), and sodium chloride (NaCl). Such fibers can also be knitted into deformable triboelectric nanogenerator textiles with high electrical outputs up to 106 V and 0.8 μA, which could work as reliable power supplies for small electronics. Moreover, we demonstrate fabric materials recognition, Morse code communication, and human-motion-recognition capabilities, making such triboelectric fiber platform an exciting avenue for multifunctional wearable systems and human–machine interaction.

Chapter 4

Fully biofriendly, biodegradable and recyclable hydrogels based on covalent-like hydrogen bond engineering towards multimodal transient electronics

Chapter 4: Fully biofriendly, biodegradable and recyclable hydrogels based on covalent-like hydrogen bond engineering towards multimodal transient electronics

4.1 Abstract

Biodegradable or recyclable transient electronics originated from natural polymers are considered as significant solution for electronic waste (e-waste) management, sustainable development, and emerging implantable devices. Yet, they are mostly struggling from mismatched mechanical features with human tissue and inferior environmental adaptability. In this work, based on the novel covalent-like hydrogen bond engineering, we developed a syrup-modified gelatin/Ag-hydrogel, which is entirely derived from natural or safe constituents and highly stretchable, yet fully biodegrades or recyclable. Meanwhile, strong covalent-like hydrogen bonds endow it with several favorable features of adhesion, rapid self-healing and antifreeze. A series of multi-modal application scenarios are further demonstrated to verify the adaptability of this platform that integrates various attributes. Overall, this work provides the feasibility of modified natural polymers to replace synthetic polymers, and provides a general toolbox for the construction of functional units for “green electronics” and the expansion of application models.

Chapter 5

Overall conclusions

Chapter 5: Overall conclusions

5.1 Overall conclusions

TENG represents is an emerging technology to realize self-powered sensing and energy harvesting. Portable, flexible, wearable, and self-powered electronic devices based on TENGs are much desired, whereas the complex preparation processes and high cost of conventional flexible electrodes prevented their practical applications. Hydrogels that combine the integrated attributes of transparency, stretchability, tunable conductivity, self-adhesion, self-healing, biocompatibility, ease of processing, and the ability to customize different shapes, and have thus emerged as popular materials from which to fabricate wearable TENGs. In this work, a series of conductive hydrogels are prepared and then utilized as electrode for TENGs that harvest mechanical energy and act as multi-functional self-powered sensors *via* the triboelectric effect. Based on the above research, the main conclusions are as follows:

In chapter 2, we have prepared a hydrogel that combines integrated properties with stretchability, adhesivity, self-healing ability, and conductivity. The hydrogel could be stretched to up to approximately 500% before breaking. It also showed strong adhesion to various materials including glass, iron, wood, and ceramics. Besides, it exhibited a good self-healing ability without the aid of any external energy or chemicals when mechanically damaged. The performances of the hydrogel-based wearable strain sensor and D-TENG are demonstrated. The strain sensor presents good long-term stability and

durability at least 1000 cycles, and it can be utilized as a body motion sensor to monitor delicate and vigorous human motion. Furthermore, the D-TENG with area of 42 cm^2 can generate an open-circuit voltage of 141 V and a short-circuit current of 0.8 μA and therefore directly power 52 LEDs. In addition, the D-TENG can charge diverse capacitors, and demonstrate the capability to power small electronics, such as a hygrometer thermometer. This D-TENG with merits including intrinsic stretchability, energy-extraction, and actively-sensing abilities, can meet wide application needs ranging from deformable/portable electronics to smart interfaces. This study provides the feasibility of developing stretchable power generation devices as well as sensors based on hydrogels.

In chapter 3, we demonstrated a facile and scalable manufacturing approach of stretchable gel-electrode-based triboelectric fiber (GETF) and gel-electrode-based triboelectric textile (GETT). The GETF with a core-sheath structure consists of silicone hollow tubes and gel-electrodes; further, the gel-electrode was synthesized *via* a simple freeze–thawing technique of blending polyvinyl alcohol, gelatin, glycerin, PEDOT: PSS, and NaCl. The produced GETFs exhibit good stretchability and flexibility. Additionally, the gel-electrode avoided problems, including cracking of metallic wire-based triboelectric fiber and leakage of liquid conductor-based triboelectric fiber. Particularly, the fabrication process we developed is scalable and enables unlimited-length fibers to be yielded, leading to possibilities for large-area and flexible fiber and textile-based electronics. Integrated *via* textile-based techniques, a 4.5 cm × 4.5 cm textile could be fabricated that demonstrates high electrical outputs up to 106 V and 0.8 μA, which could work as reliable power supplies for small electronics. Our demonstrated fabric material

recognition, Morse code communication, and human-motion-recognition capabilities are being developed for wearable systems and smart textiles. Overall, this work provides a new path for energy harvesting and functional fiber processing and provides opportunities for the advancement of other functional fiber and textile-based electronics.

In chapter 4, aiming to solve the e-waste problem, a syrup-modified gelatin hydrogel is proposed via the covalent-like hydrogen bond engineering strategy, which combines a series of interesting features of improved mechanical behaviors (strength ~ 54.2 kPa; stretchability ~ 329%), strong interfacial adhesion, self-healing (strain healing efficiency up to 88.4%), freeze resistance (freeze-resistance at -28.9 °C), reusability and degradability. Based on this green-toolbox, a new set of e-skin platforms towards multimodal capabilities of flexible strain sensors (GF up to 3.42) and TENG is further created. In several proof-of-concept demonstrations, we display its possible applications range from fully-ranged human motion monitoring, biomechanical energy harvesting (V_{oc} from the GAPS-TENG treated with a hand tapping reach up to 38 V) or self-powered information transmission. To sum up, we believe the results will provide some beneficial inspiration for improving the mechanical performance of non-covalent crosslinking hydrogel, shows a wide potential of fully natural polymer-based electrodes.

In summary, this work develops a series of conductive hydrogels, respectively, as well as shows how these hydrogels incorporated into wearable TENG and further demonstrate potential applications in wearable sensors and energy harvesters. we believe above work will be useful to researchers and engineers in this emerging field and spur research and development of this important area.

5.2 Perspectives

The development of hydrogels as current collectors for TENGs has been rapid recently, with good progress made in terms of transparency, conductivity, stretchability, self-adhesion, self-healing, biocompatibility and environmental friendliness. However, the development of commercial hydrogels and H-TENGs still faces challenges such as performance and environmental stability parameters that are important to achieve commercial applications of H-TENGs. Opportunities and challenges coexist. The simple fabrication of highly conductive and durable hydrogels will provide an excellent alternative to H-TENGs for future wearable and biomedical applications.

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ACCOMPLISHMENTS

Journal Publications

[1] **Li Dong**, Mingxu Wang, Jiajia Wu, Chunhong Zhu, Jian Shi, Hideaki Morikawa. Stretchable, Adhesive, Self-Healable, and Conductive Hydrogel-Based Deformable Triboelectric Nanogenerator for Energy Harvesting and Human Motion Sensing. ACS Applied Materials & Interfaces. 2022, 14, 7, 9126-9137.

[2] **Li Dong**, Mingxu Wang, Jiajia Wu, Chunhong Zhu, Jian Shi, Hideaki Morikawa. Deformable Textile-Structured Triboelectric Nanogenerator Knitted with Multifunctional Sensing Fibers for Biomechanical Energy Harvesting. Advanced Fiber Materials. 2022, 4, 6, 1-14.

[3] **Li Dong**, Mingxu Wang, Jiajia Wu, Chenyang Zhang, Jian Shi, Keimei Oh, Lirong Yao, Chunhong Zhu, Hideaki Morikawa. Fully biofriendly, biodegradable and recyclable hydrogels based on covalent-like hydrogen bond engineering towards multimodal transient electronics. Chemical Engineering Journal. 2023, 458, 141276.

Conference

[1] **Li Dong**, Chunhong Zhu, Hideaki Morikawa. Stretchable, Adhesive, Self-Healable, and Conductive Hydrogel-Based Deformable Triboelectric Nanogenerator for Energy Harvesting and Human Motion Sensing. (Oral presentation). 2022 Textile Society Annual Meeting. (June 8-10, 2022, Tokyo, Japan).

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