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Advances in Green Triboelectric Nanogenerators

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Triboelectric nanogenerator (TENG), an emerging energy conversion technology, offers innovative pathways for energy harvesting and self-powered sensing. To achieve superior performance, researchers commonly employ substantial quantities of original or treated polymers, resulting in high energy and precise sensing. Nevertheless, the sustainable development of TENGs faces significant challenges related to environmental compatibility, pollution hazards, and high production and disposal costs. To address this issue, numerous green materials for diverse TENGs are introduced and advanced. These materials may encompass natural resources, household waste, and recyclable materials, among others. Consequently, a review of the progress in TENGs based on green materials, which can be called green TENGs, becomes imperative to advance its sustainable development. To this end, this work comprehensively elucidates the development of green TENGs from the perspective of materials processing and treatment degree for the first time. Various green TENGs, including food waste, discarded daily-use items, plant organs, biodegradable industrial products, and natural cellulose, are meticulously categorized. This review not only systematically synthesizes the latest research advancements in green TENGs, but also offers insight into their processing methodologies, working characteristics, and potential application scenarios. Finally, it envisions the challenges, proposed solutions, and future research directions for the development of green TENGs.

1. Introduction

The popularization and development of the Internet of Things (IoT) requires a proper way to keep vast distributed wireless sensor nodes working properly.[1] The adoption of self-powered systems has become the most promising and sustainable solution.[2] Among different energy harvesting and self-powered sensing technologies, Triboelectric nanogenerator (TENG) is one of the most effective means, which has been widely concerned and was first invented by Prof. Wang in 2012.[2a] It is characterized by high energy conversion efficiency, simple fabrication, low cost, and high power density, which makes it easier to build self-powered sensors/systems. Through the coupling of two effects, triboelectrification and electrostatic induction,[3] TENG converts mechanical energy acting between two contact surfaces made of different materials into electrical energy. It has been widely developed for mechanical energy harvesters, including ocean wave energy,[4] wind energy,[5] vibration energy,[6] biomechanical energy, etc.[7] In addition, as a self-powered sensor, TENG has a very high sensitivity and is promising for many application scenarios.[8] For example, it can be used for sensing temperature and humidity in natural environments,[9] navigation monitoring in smart transportation,[10] monitoring of equipment operation status in industrial production,[11] and monitoring of human micro-activities as well as health status.[12] Generally, triboelectric layers of TENGs mainly consist of fluorinated ethylene propylene (FEP), Polyvinylidene Fluoride (FVDF), polytetrafluoroethylene (PTFE), polydimethylsiloxane (PDMS), polyamide fibers (PA), copper, and other polymers.[13] Most of the TENGs made from these materials show good output performance. However, since most of these materials are non-biodegradable or very expensive, they challenge the sustainable development of TENGs and environmental protection. Therefore, using environment-friendly and green materials to design and manufacture TENG devices is essential.

The above issues have received extensive attention from scholars, and the number of articles on green TENGs, which is the use of waste from production and life, natural plants, or polymer degradable materials as the triboelectric layer of TENGs, has increased significantly in recent years. For example, tea leaves,[14] milk cartons,[15] lotus leaves,[16] hydrogel,[17] mulberry silk and other materials are utilized to fabricate TENG.[18] To facilitate the development of green TENGs, some wonderful reviews are presented. Herein, TENGs made from recyclable materials are summarized in a review by Wang et al.[19] Slabov et al.[20] reviewed advances in the field of TENG using plants from natural materials, plant-processed products, and naturally degradable materials. Relying on the categorization idea of refined, hybridized,

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Figure 1. Green TENG based on food waste, discarded daily-use items, plant organs, biodegradable industrial products, and natural cellulose. Reproduced with permission.[25] Copyright 2020, Elsevier. Reproduced with permission.[26] Copyright 2020, Elsevier. Reproduced with permission.[27] Copyright 2018, Elsevier. Reproduced with permission.[28] Copyright 2022, Elsevier. Reproduced with permission.[29] Copyright 2018, Elsevier. Reproduced with permission.[30] Copyright 2020, American Chemical Society. Reproduced with permission.[12c] Copyright 2023, Elsevier. Reproduced with permission.[31] Copyright 2022, Elsevier. Reproduced with permission.[32] Copyright 2018, Elsevier. Reproduced with permission.[33] Copyright 2020, Elsevier.

post-treated, or untreated, Song et al.[21] discussed recent advances in natural biomaterials for ecological TENG and their energy harvesting properties. In particular, TENG based on degradable materials and their applications were summarized by Chao et al.[22] They classified degradable materials into animal-based degradable materials, plant-based degradable materials, and artificial degradable materials, and emphasized that TENG based on degradable materials have healthcare effects in vivo, which promotes the development of degradable TENG in different fields. Moreover, recent developments in wearable sensors based on the bio-derived material TENGs are discussed by Yang et al.[23] Although these efforts have played a crucial role in the development of green TENGs, existing studies have often focused on highly industrialized materials, lacked effective attention to TENGs based on food wastes and industrial wastes, or studied only single green materials and lacked attention to green composites, which makes a comprehensive review covering different green TENGs is still in its infancy.[19,24] Therefore, there is an urgent need for a systematic review of comprehensive green TENGs to promote the green development of TENGs.

In order to solve the above problems and promote the combination of direct or indirect environmental protection. We have collected articles based on TENGs made from environmentally friendly materials or recycled wastes and comprehensively and systematically reviewed the research progress of green TENGs in recent years from the perspectives of different materials and different processing levels. As shown in **Figure 1**, according to the recycling difficulty, finishing degree, recycling urgency, and cost-effectiveness of the relevant materials, we divided them into food waste, discarded daily-use items, plant organs, biodegradable industrial products, and natural cellulose. In addition, the processing methods and techniques, working properties, and applications for green-type TENGs are elaborated. Finally, the challenges, solutions, and subsequent research directions for TENG based on green materials are envisioned.

2. Working Mechanisms and Working Modes of TENG

As shown in Equations (1–4), the operation mechanism of TENG originates from the Maxwell displacement current.

$$\nabla \cdot \boldsymbol{D} = \rho_f \tag{1}$$

$$\nabla \cdot \boldsymbol{B} = 0 \tag{2}$$

$$\nabla \times \boldsymbol{E} = -\frac{\partial \boldsymbol{B}}{\partial t} \tag{3}$$

$$\nabla \times \boldsymbol{H} = \boldsymbol{J}_f + \frac{\partial \boldsymbol{D}}{\partial t} \tag{4}$$

Where Equation (1) is Gauss's law, ∇ is the vector differential operator, $\boldsymbol{D}$ is the potential shift vector, and ρ_f is the free charge density. Equation (2) is Gauss's law of magnetism, $\boldsymbol{B}$ is the magnetic field, which denotes that the dispersion of the flux density is 0. Equation (3) is Faraday's law, which denotes the rate of change of the flux density in time at the point for which the spin of the strength of the electric field is negative. Equation (4) is Maxwell–Ampere's law, $\boldsymbol{H}$ is the magnetizing field, and $\boldsymbol{J}_f$ is the free current density, which indicates that the spin of the magnetizing field strength is the sum of the free current density and the displacement current density $\frac{\partial \boldsymbol{D}}{\partial t}$ at that point.[34] After that, as shown in **Figure 2a**, Wang further developed and extended Maxwell's

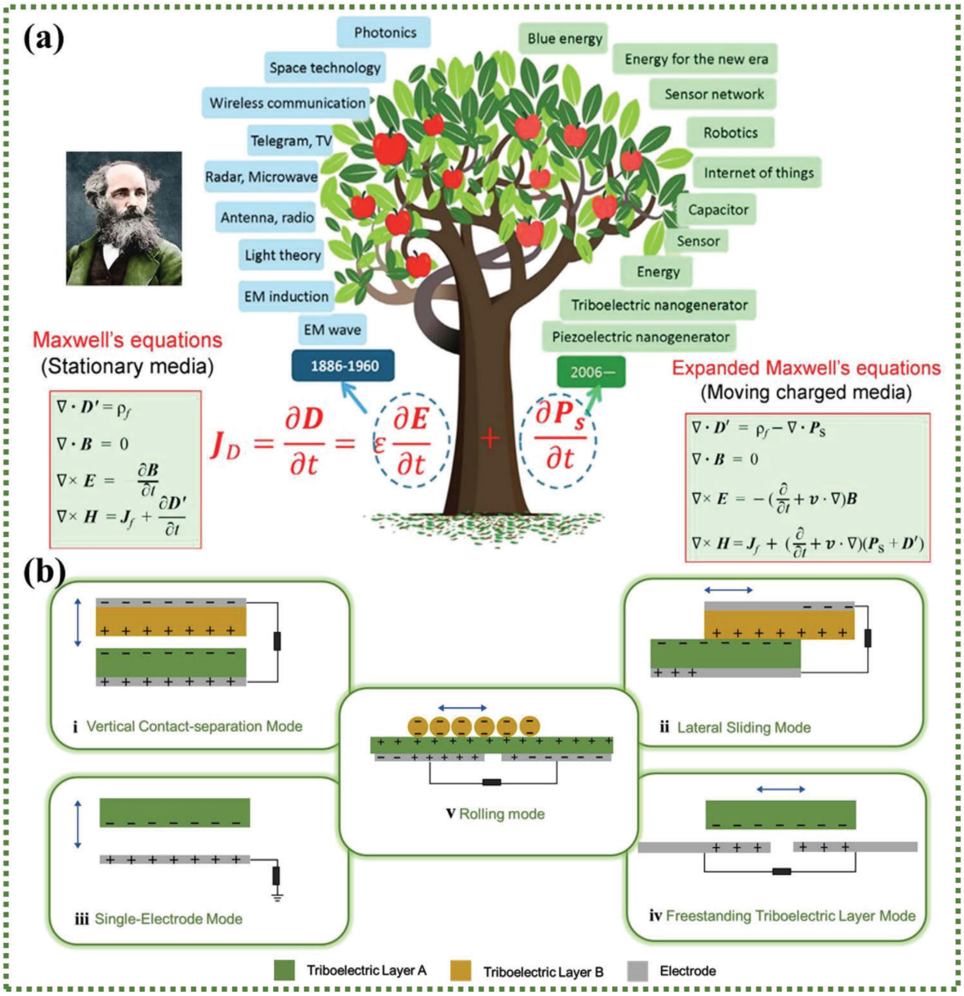

Figure 2. a) Theoretical foundation for TENG based on Maxwell's displacement current. Reproduced with permission.[34] Copyright 2018, Springer Nature. b) Four fundamental modes of TENG. **i)** Contact-separation mode. **ii)** Lateral-sliding mode. **iii)** Freestanding mode. **iv)** Single-electrode mode. **v)** Rolling mode.

system of equations, and based on the extended system of equations, he developed the basic theory of TENG.[35]

The TENG not only performs energy harvesting but also acts as a self-actuating sensor that converts mechanical energy into electrical signals.[36] As shown in Figure 2b,[37] the TENG can be categorized into four basic forms of motion based on the electrode structure of the TENG and the difference of electrostatic induction generated between the TENG layers: **i)** vertical contact separation,[38] **ii)** lateral sliding,[39] **iii)** single electrode,[40] **iv)** freestanding layer mode, and **v)** rolling mode.[41] These five operating modes can be universally adapted to energy harvesting and self-powered sensing in daily production life.

3. TENG Based on Food Waste

Food waste is the waste of food scraps, leftover food and beverage ingredients, peels, and other organic substances generated in our daily lives. However, the problem of food waste is becoming more and more serious. A United Nations report shows that globally, more than 100 million tons of food waste from homes, restaurants, and stores are generated each year.[42] The advent and quick advancement of TENG technology in recent years has inspired researchers to concentrate on TENG based on food waste as an environment-friendly class of materials, offering a fresh approach to this issue.

3.1. TENG Based on Food Material

Edible foodstuffs were used directly as the triboelectric layer of the TENG. Jiao et al. created sandwich-structured TENG using cabbage leaves/wheat bread as shown in **Figure 3**a, where the microstructure of the cabbage leaves is shown in Figure 3b. Figure 3c shows the 3D structure of the sandwich-structured TENG, through which a commercial LED can be driven and a commercial alarm can be activated as shown in Figure 3d.[26] Similarly, Khandelwal et al. fabricated a single electrode lightweight TENG (E-TENG) using natural silver leaf as a triboelectric layer. It has a current of 315 nA and a maximum output voltage of 23 V.[44] It can be used to power hygrometers,

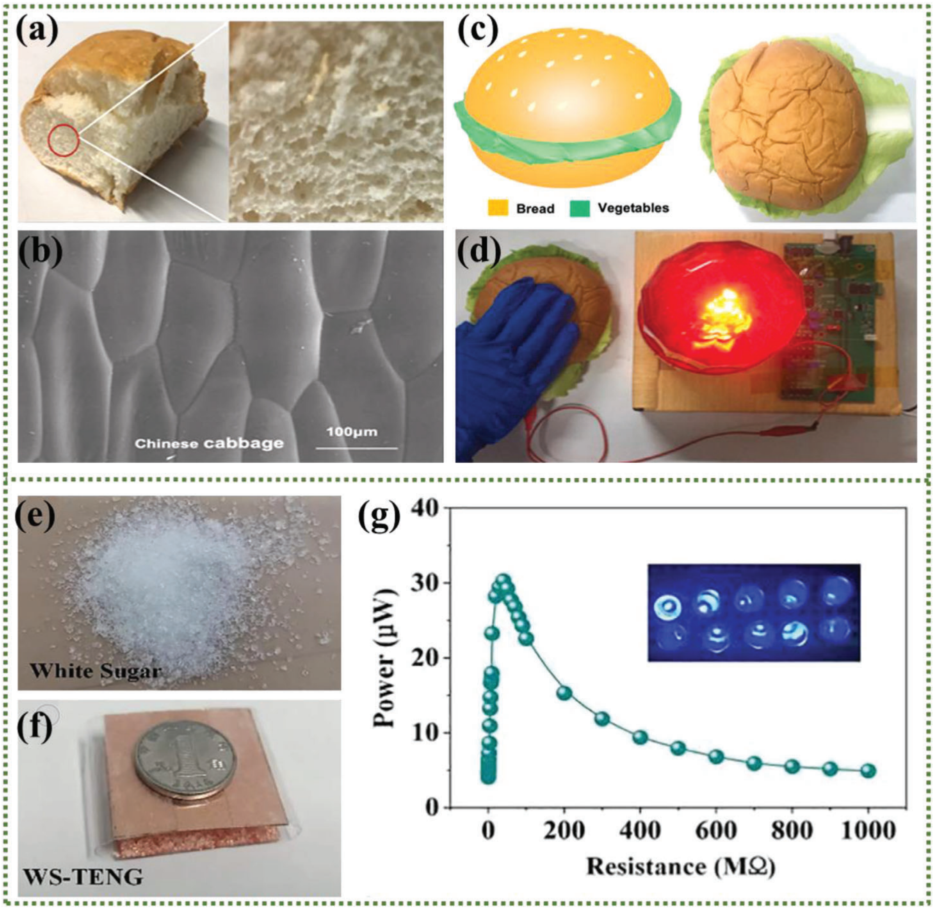

Figure 3. Edible material-based TENG. a) Structural diagram of sandwich bread and its structural enlargement. b) Microstructure of cabbage. c) 3D schematic and actual picture of sandwich TENG. d) Picture of sandwich TENG used to collect mechanical energy. Reproduced with permission.[26] Copyright 2020, Elsevier. e) White sugar material schematic. f) Schematic diagram of TENG structure based on white sugar particles. g) Picture demonstrating the performance of WS-TENG as an energy harvesting device. Reproduced with permission.[43] Copyright 2020, Elsevier.

watches, LEDs, and so on. In addition to the above materials, some scholars have utilized discarded onion skins (OT) as a dielectric layer of TENG for harvesting the energy generated by humans during activities such as walking, running, and jumping.[45] On this basis, Zhang et al. compared the heterogeneous friction-triggered effect (HTE) of leeks, green onions, and onions and utilized their HTE to fabricate a single-electrode TENG, which outputs power densities as high as 35 W m^{-2}, which enables energy harvesting, gas sensing, and humidity sensing.[46]

Moreover, industrial deep-processed foodstuffs have also been utilized to manufacture TENG, which has superior output characteristics due to its regular shape. As shown in Figure 3e, Liu et al. fabricated a white sugar novel triboelectric nanogenerator (WS-TENG), and its structure is shown in Figure 3f. Figure 3g demonstrates the ability of the WS-TENG to complete mechanical energy harvesting to light a commercial electric light bulb.[43] Meanwhile, since white sugar is sensitive to humidity, the WS-TENG can be used as a self-powered humidity sensor. In addition, Xie et al. utilized instant noodle powder to create vertically separated TENGs that can capture mechanical energy under different operating conditions and detect human touch states.[47]

In this subsection, we illustrate the edible materials by dividing them into two efforts according to the degree of refinement of the material and the stability of the output properties from low to high. First, materials such as vegetable leaves, bread, and onion skins are directly utilized by the researchers. This type of TENG production shortens the production cycle and the production process tends to be relatively simple, which facilitates the production of large quantities quickly. However, the relatively low degree of standardization of these more primitive materials results in poor durability and smoothness of the device's energy output. Therefore, as research progresses, researchers begin to make TENGs from industrial foods (sugar and flour) or by refining (dissolving, filtering, grinding) collected edible ingredients, which results in TENGs that are highly similar in physical properties, such as size and shape, and thus make significant advances in performance output.

3.2. TENG Based on Food Scraps

Another key factor in the generation of food waste is food scraps generated during food processing or consumption, such as fish

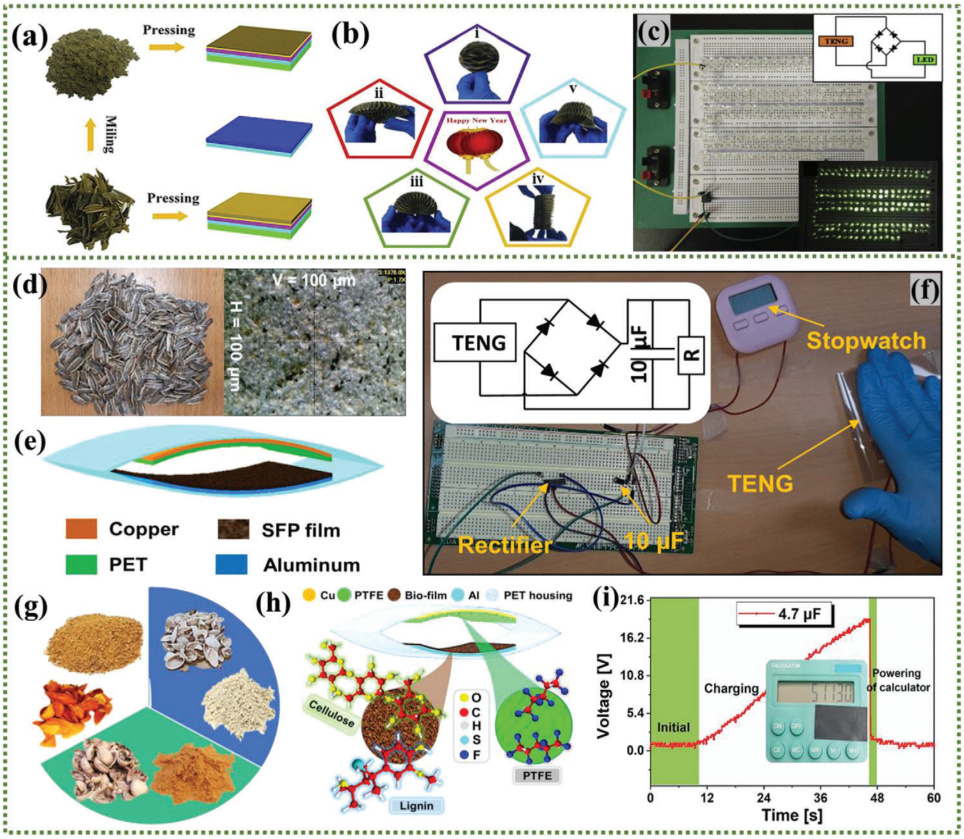

Figure 4. TENG based on food scraps: a) Processing Flow of Waste Tea Leaves. b) Schematic diagram of different structures of TAP-TENG. c) Performance demonstration picture of TAP-TENG as an energy harvesting device. Reproduced with permission.[14] Copyright 2019, Elsevier. d) Acquisition of material and processing process of sunflower seed shell-based TENG (SFP-TENG). e) 3D structure and physical picture of SFP-TENG. f) SFP-TENG as an energy harvesting device to collect vibration energy and power a sports watch. Reproduced with permission.[49] Copyright 2021, Elsevier. g) Photographs of the various WFSs and their powders. h) Structure based on WFS-TENG. i) Charge-discharge curves of TENG devices based on the PTFE layer in the Pi-WFS system. Reproduced with permission.[50] Copyright 2021, Elsevier.

scales, egg shells, nut shells, and coffee grounds,[48] which are food scraps or inedible wastes that are usually discarded as garbage and cause environmental pollution. Some scholars have combined these materials with TENG to create green-type TENG.

As research progressed, many food residues began to be used as triboelectric materials for TENG. Xia et al. achieved an energy output of 42.8 μA, 792 V, and 488.88 μW cm^{-2} using an aluminum-plastic bag/tea leaf fabricated TENG (TAP-TENG).[14] As shown in **Figure** 4a, the TAP-TENG film is fabricated by collecting discarded tea leaves after drinking, grinding them into tea powder with a blender, and then spreading the powder on double-sided adhesive tape for compaction. Figure 4b(i–v) shows a honeycomb structure based on a conventional lantern shape, where energy output is realized by stretching, compression, and bending. The honeycomb lantern structure of TAP-TENG can be used to harvest mechanical energy such as tapping, pressing, and stroking, and Figure 4c demonstrates that it can be used to power small electronic devices such as electronic thermometers, electronic watches, and so on through the power management circuit. As a typical drink in daily life, coffee produces much residue after drinking. The TENG is made by embedding the waste coffee residue as a raw material into a silicone rubber elastomer, which can collect the energy generated by the human body's movement. At the same time, with its ultra-high sensitivity, it is used to sense physiological signals from the human body and develop an intelligent tactile epidermal controller.[48a]

In addition, the researchers utilized inedible parts produced in daily life such as fruit shells or peels to create TENG. For example, using powder ground from discarded peanut shells and making a film to design an environmentally friendly TENG for wind energy collection or as a wind speed sensor.[48d] Shaukat et al. made a TENG using sunflower seed husk and obtained a performance output of 28.5 μA, 488 V, and 1200 μW.[49] As shown in Figure 4d, they collected sunflower husk and cleaned, dusted, dried, and blended it into powder. The powder was then combined with an aluminum film to make a TENG based on biowaste sunflower husk powder (SFP-TENG). Figure 4e illustrates the 3D structure of the SFP-TENG with vertical separation. As shown in Figure 4f, the SFP-TENG can derive mechanical energy from the daily movement of the human body to power microelectronic devices. As shown in Figure 4g, Saqib et al. selected wood fiber mass waste fruit shells (WFS) of almonds (A), walnuts (W), and pistachios (Pi) as anode materials for TENG.[50] They used three different WFS as anode materials with PET and PTFE to make the WFS-TENG as shown in Figure 4h. Finally, the Pi-WFS-based TENG was analyzed and compared to achieve a better

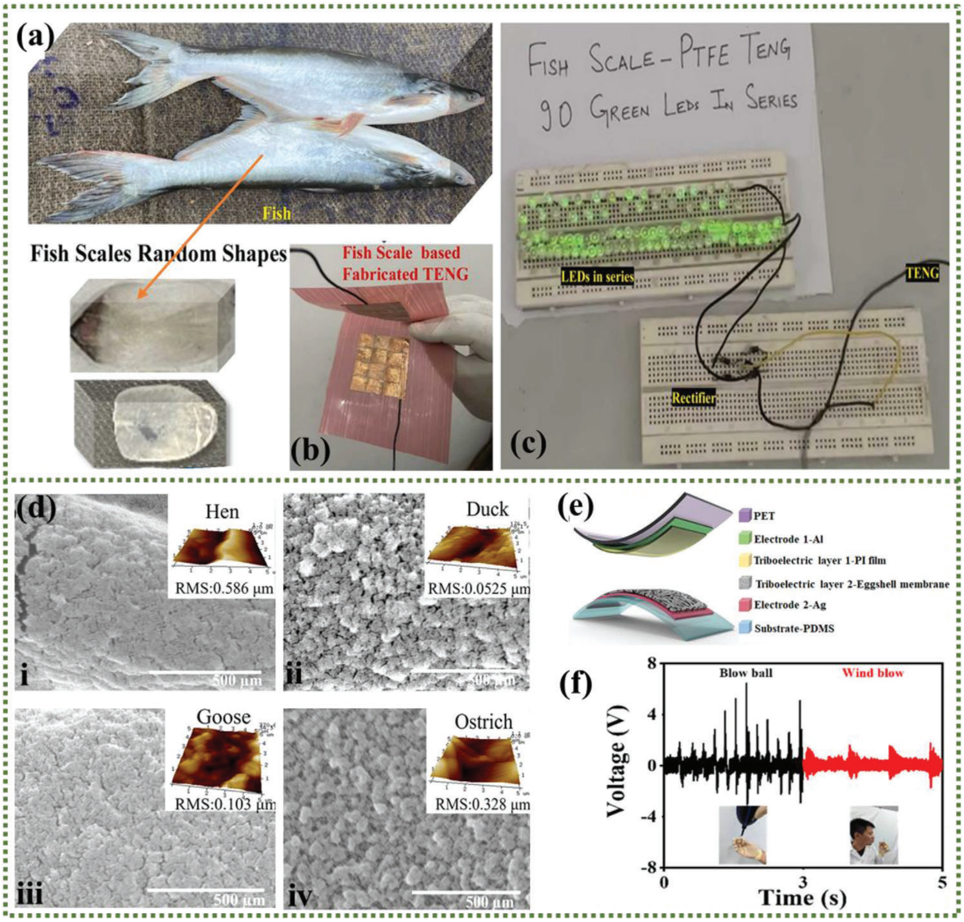

Figure 5. TENG based on animal-based food scraps. a) Selection of fish scale material. b) Physical photo of fish scale TENG. c) Picture of fish scale TENG as energy harvesting device lighting LED bulb. Reproduced with permission.[48b] Copyright 2022, Elsevier. d) SEM images of various types of ESM: (i) hen, (ii) duck, (iii) goose, and (iv) ostrich. e) 3D schematic of the EM-TENG device. f) EM-TENG as a smart sensor for detecting body movement. Reproduced with permission.[55] Copyright 2022, American Chemical Society.

output performance of 700 V, 95 mA, and 416.14 mW cm^{-2}, and can power the small electronic device in Figure 4i. Similar to the above, Kim et al. creatively made mandarin peel powder (MPP) from citrus peels and fabricated TENG (MMP-TENG) based on the film made from MPP. It achieves an energy output of 156 V, 2 μA, 5.3 μW cm^{-2}, and harvests energy to light the LED.[51]

In addition to the above forms of food waste, many food processing processes leave behind food residues,[52] such as fish bladders and eggshells,[48c] and these materials have attracted a lot of attention from scholars, and TENGs made from these materials have shown notable performance. As shown in **Figure 5**a, Singh et al. collected fish scales from fish market garbage, washed and dried the collected fish scales with distilled water and ethanol, and then cut different sizes of fish scales into a uniform size of 1 cm × 1 cm. As shown in Figure 5b, they made a fish scale TENG by pasting 12 treated fish scales with an area of 1 cm^2 onto the adhesive surface of the electrodes and achieved an energy output of 1.7 μA on a 1 MΩ resistor. Figure 5c shows the illumination of 90 commercial green LEDs using the fabricated fish scale-PTF TENG.[48b] Similarly, a TENG based on a fish bladder membrane was designed, and the output current density and charge density were close to 4.56 mA m^{-2} and 25 μC m^{-2}, respectively.[52]

In addition, Zhang et al. completed the purification by washing, drying, and dissolving the raw powder of crab chitin. The fabricated TENG achieved a power output of 182.4 V, 4.8 μA, and 1.25 W m^{-2}.[53]

Eggs are one of the most common foods in our daily lives, improving our quality of life while producing an unavoidable waste, eggshells. egg shell membrane (ESM) has been utilized as a triboelectric material to create TENGs with excellent power generation performance and potential as sensors.[48c,54] Lin et al. further investigated the TENG based on the eggshell membrane, as shown in Figure 5d(i–iv). After comparing the electrical properties of different eggshell membranes, they found that ostrich ESMs were superior to other ESMs due to their rich functional groups, high roughness, high surface charge, and high dielectric constant. Figure 5e shows a TENG (EM-TENG) made of ostrich eggshell membrane, achieving a power output of 300 V and 0.18 mW. As shown in Figure 5f, they also used the EM-TENG to detect air blowing out of the mouth to collect wind energy or to fix it on one leg to detect leg movement.[55]

In this subsection, we focus on TENG based on food scraps. We divide the chapter into two main parts: plant-based food scraps and "meat, egg, and aquatic" food scraps. First, we divide plant-based food scraps into two parts: beverage-based and peel-based. Since beverage-type waste (waste tea leaves, coffee grounds) contains more water, the researchers dry and ground it to improve the output performance of the TENG. For peel food waste, such as orange peels, nut shells, and other materials, the researchers focus on powdering them and making a thin film to further enhance their output capability. Second, for the category of food waste about "meat, egg and aquatic", we mainly mention aquatic waste and eggshells. Aquatic wastes are varied, and scholars have assembled fish scales or used fish floats to create TENGs. However, the standardization of these materials is relatively low, resulting in poorer smoothness of energy output. Compared to aquatic waste, researchers have studied eggshell membranes more deeply, and they have analyzed the power generation characteristics of different types of eggshell membranes so that TENGs made from these materials can achieve a smoother output, which is also a promising material for the development of this subsection.

This subsection has been categorized into edible food, food residues, and food trimmings to provide a systematic and progressive review of previous research results. These seemingly useless wastes have been given new life by TENG researchers to turn these materials into treasures, and the green-type TENGs made of these materials have shown remarkable energy output and stable and sensitive self-powered sensing performance, which provide a new direction for subsequent large-scale applications and environmental protection.

4. TENG Based on Discarded Daily-Use Items

A large amount of industrial waste is generated in our daily lives, including recyclable, non-recyclable, hazardous ones, etc. These industrial wastes are found in almost all corners of our daily life and seriously jeopardize the aesthetics and environmental quality. In this section, TENGs based on industrial wastes such as PET plastic bottles,[56] old clothes,[57] and cigarette butts are systematically reviewed.[28] In addition, the output performance of TENGs based on recycled and reused materials and application scenarios as self-powered sensors are discussed in detail.

4.1. TENG Based on Recyclable Waste Materials

For example, plastic bottles, milk cartons, and other plastic products are recyclable waste from daily life. However, they cannot be recycled well due to their large quantity. With the rapid development of TENG technology, they are used as triboelectric materials for TENG thus adding a new way to reuse this type of waste.

Milk cartons or other packaging boxes are collected and used as material for making TENGs, such as the aluminum layer inside the carton is extracted and used as a TENG electrode. Costa et al. first used recyclable milk carton aluminum paper (ARP) as the substrate and the bottom electrode of the TENG to make a TENG, which outputs a power density of up to 1.6 μW cm^{-2} when operated at 50 Hz vibration frequency.[15] Li et al. further simplified the fabrication process by directly utilizing the laminated structure of milk carton packages to prepare TENGs.[58] The initial charge density of TENGs based on used milk cartons is 0.035 mC m^{-2}, which can be increased to 1.00 mC m^{-2} by combining with charge excitation circuits. Similarly, an output voltage of 4 V and an instantaneous power density of 11.8 nW cm^{-2} were generated by hand-exciting a TENG made from discarded food packaging aluminum foil and laboratory film.[59] Based on the above studies, Zhou et al. processed a milk carton to turn it into a fully enclosed arc nanogenerator (AS-TENG), which is illustrated in **Figure 6**a. As shown in Figure 6b, they developed a self-powered wireless environmental monitoring system using AS-TENG as a power source for real-time monitoring of water quality (pH) in the natural environment.[27] Then, a self-powered early warning system for landslide hazards is successfully achieved as depicted in Figure 6c. Furthermore, Moreira et al. and Sankar et al. fabricated TENGs by collecting aluminum material from used cartons.[60] The fabricated TENGs showed good stability and output performance.[61]

Besides aluminum boxes, plastic bottles, which are another main industrial waste product in life, have also been employed to fabricate TENGs based on recycled materials. As shown in Figure 6d, Roy et al. made a highly elastic, spongy PET electrostatically spun aerogel (ppp aerogel) by washing recycled PET bottle flakes by mixing and stirring trifluoroacetic acid and methylene chloride, filtering, and heating.[56] Figure 6e shows the schematic structure and power generation method and achieves an electrical output of 67.7 V, 9.4 μA. Figure 6f demonstrates its excellent output performance, which can instantly light up nearly 36 colors of LEDs by pressing it by hand. Similarly, Sukumaran et al. developed a system that can be used for vehicle safety alerts and tire motion signal acquisition using TENG made from waste plastic bottles.[62]

We categorize recyclable waste into carton waste and plastic bottle waste by material according to the recycling material. For carton waste such as milk cartons, researchers mainly extract aluminum film from it to be used as TENG-positive electrodes, thus indirectly realizing the recycling of waste metals. For plastic products, the processing process is relatively complex compared to cartons, usually after crushing, melting, and other processes to remove impurities or make them into powder. Although this process is complicated, it further improves the output and sensing characteristics of TENG, which is conducive to subsequent promotion and development.

4.2. TENG Based on Unrecyclable Industrial Wastes

The above is mainly about the garbage that is easy to recycle or easy to handle, but some of the garbage in our life, such as some plastic film, foam, etc., cannot be recycled, or the cost of recycling is high. The ways and means to deal with these wastes have become a serious problem. To address these issues, Jalili et al. used waste polymers such as polyurethane foam (PU), polypropylene (PP), and other waste polymers to manufacture environment - friendly, simple, and low-cost TENGs without any physical or chemical functionalization. In addition, an MF-based TENG with an area of 3 × 3 cm^2 was found to achieve an electrical output of 1830 V, 96.68 mW cm^{-2}, and illuminate 100 commercial blue LEDs in a comparative experiment.[63] As shown in **Figure 7**a, Nawaz et al. created a Waste Polystyrene TENG (WPS-TENG) using polystyrene (PS) extracted from packaging waste. Figure 7b

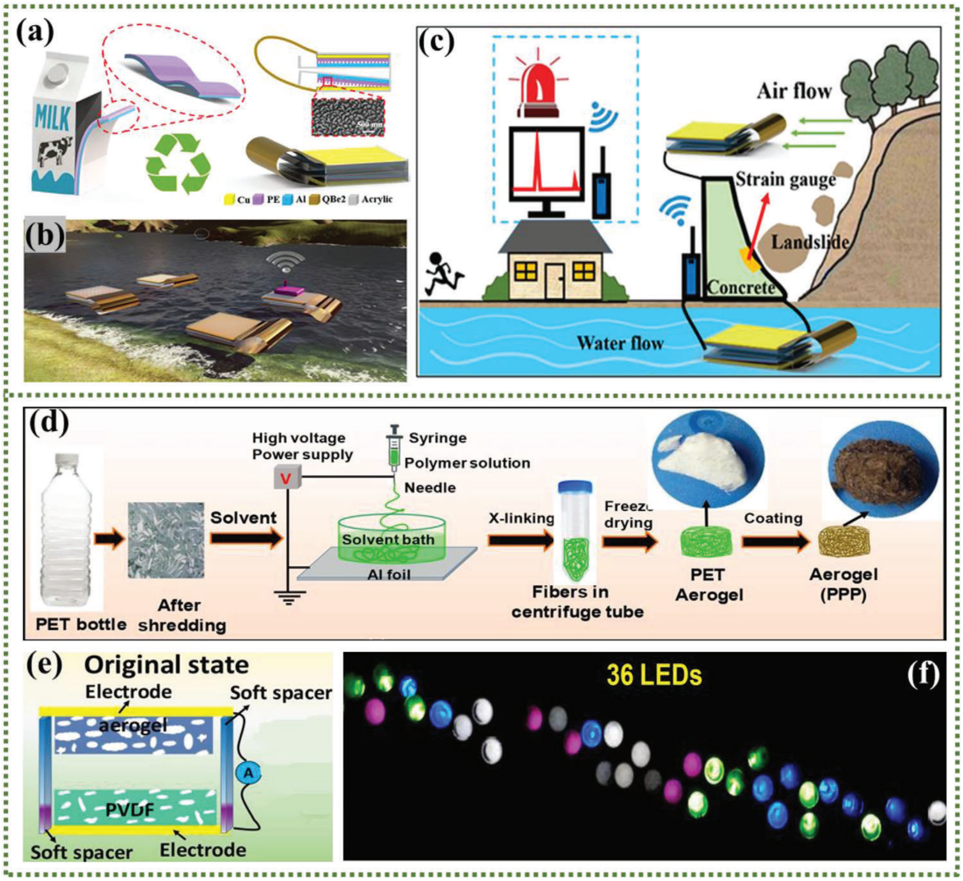

Figure 6. TENG based on recyclable waste materials. a) Schematic diagram of the material source fabrication method and 3D structure of AS-TENG. b) Application scenario of AS-TENG as a self-powered sensor. c) Schematic diagram of the application scenario of AS-TENG as a sensor for landslide warning. Reproduced with permission.[27] Copyright 2018, Elsevier. d) Flow chart of 3D PPP aerogel made from PET waste bottles. e) Schematic diagram of the 2D structure of TENG based on PET material. f) Photograph of 36 LEDs of different colors lit by manual pressing. Reproduced with permission.[56] Copyright 2018, Elsevier.

shows that the WPS-TENG can convert mechanical energy from human tapping into electrical energy to power 120 LEDs. In addition, Figure 7c shows that it successfully implements a self-powered sensor to realize the detection of vehicle speed under different loads.[10] Meanwhile, the TENG, which was prepared using PVC cling film and nylon as triboelectric material, was able to achieve 31 V, 3.1 A, and 11.9 mW m^{-2} power output, and was able to illuminate 28 commercial LEDs.[64] Han et al. went a step further by utilizing a novel TENG (WPB-TENG) made from waste plastic bags (WPB) and PTFE film that could drive 40 commercial LEDs as well as power flexible vertically interconnected paper-and-pencil strain sensors.[65]

Industrial waste products can be further processed into powder to further improve TENG's output performance. A WRP-based TENG was designed by making a powder of waste rubber (WRP) with fluorine on the surface as a negative electrode and an aluminum film as a positive electrode. The maximum output voltage and charge density are 265 V and 75 mA m^{-2}, respectively, and can directly drive up to 100 commercial LEDs.[67] As shown in Figure 7d, Nawaz et al. used discarded pencil lead as TENG material.[66] Figure 7e shows the fabrication process of the flexible composite film: A flexible composite film is made by converting graphite into an excellent powder and incorporating this material into a PDMS matrix. Figure 7f shows the 3D structure of this TENG. Figure 7g shows that circuit optimization can light up 22 LEDs.

In this part, non-recyclable materials are reviewed according to the depth of finishing. First, for some materials such as plastic films and foams, researchers can make TENG without much processing, which shortens the production cycle and has a lower processing depth to be favorable for the production of large quantities quickly. However, there are also problems with the durability of the device and the smoothness of the energy output. Some scholars have gone a step further and processed industrial waste into powder form to produce TENGs with better power output and sensing performance.

4.3. TENG Based on Hazardous Industrial Wastes

Hazardous wastes such as batteries and cigarette butts are also generated in daily life. These wastes seriously damage the natural environment and cannot be recycled, but can only be incinerated or buried. TENG researchers have provided a new

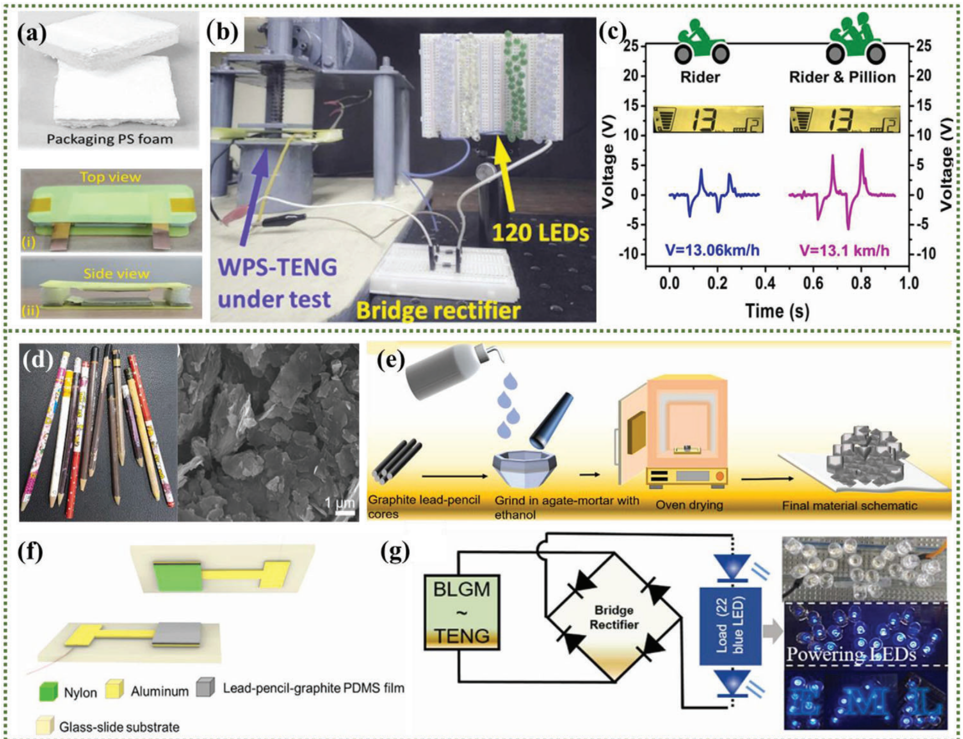

Figure 7. TENG based on unrecyclable industrial wastes. a) Material source of WPS-TENG and schematic of the structure of WPS-TENG. b) WPS-TENG powers 120 LEDs connected in series. c) Detection of vehicle speed under different operating conditions. Reproduced with permission.[10] Copyright 2022, Elsevier. d) Digital image of grades based on discarded graphite pencils. e) Schematic diagram of the material synthesis process. f) 3D schematic diagram of WRP-TENG based on discarded graphite-pencil. g) As an energy harvesting device to drive 22 commercial LEDs. Reproduced with permission.[66] Copyright 2022, Elsevier.

way to deal with hazardous wastes by collecting and reusing them.

Rani et al. were the first to utilize discarded cigarette filters (CF) to fabricate TENGs.[28] The fabricated CF-TENG achieved an output performance of 42.8 V, 0.86 μA, and 63.2 mW m^{-2}, and was capable of powering 44 LEDs and an LCD timer. Bukhari et al. fabricated graphite and plastic-based TENG (GP-TENG) by collecting waste plastics and electronic wastes and achieved 83.88 V, 101 μA, and 26.54 μW cm^{-2} electrical performance.[62] It could power a digital calculator and 19 blue-emitting LEDs. Alternatively, the TENG can be prepared from waste electronic components to provide power for load systems.[68] In addition to this, Natarajan et al. recycled used LIB graphite anodes and metal aluminum shells to make a TENG, which can charge a commercial capacitor with a maximum voltage of 1.4 V in 100 s by applying a force of 0.2 m s^{-2}, thus providing a sustainable power source for various applications.[69] As shown in **Figure** 8a(i–viii), Chougale et al. innovatively converted expired drugs into novel friction electric materials.[70] These expired drugs were dissolved, stirred, and uniformly sprayed with a nebulizer to form a thin film, which showed a strong positive potential due to the high electron-donating ability of the hydroxyl and carbonyl functional groups. Figure 8b illustrates the TENG device diagram. As shown in Figure 8c, the drug-based TENG can harvest energy to power a watch.

In addition to the above mentioned TENG made from hazardous industrial waste materials for energy harvesting can also be further used as a self-powered sensing device. As shown in Figure 8d, Sahu et al. fabricated a single-electrode TENG. Figure 8e shows a four-finger woven TENG based on worn textiles such as jute, cotton, wool, and waste materials such as cigarette fibers from garbage cans.[57,71] They introduced digital signal processing techniques to realize practical self-powered applications in innovative sports, such as the self-powered pulse force recognition and fringe ball judgment system demonstrated in Figure 8f. They also collected waste plastics from the lab to create a waste-based, vertically separated TENG. It is mounted in different locations in the laboratory to collect mechanical energy from various locations, and it can also be used as a self-powered tracking device to determine the location of a human body in an emergency.

Hazardous wastes generated in daily life such as batteries, cigarette butts, expired medicines, etc., which seriously pollute the environment and jeopardize human life and health, have become an urgent problem to be solved. TENGs based on them are discussed in aspects of energy harvesters and sensors according to different application methods. Many scholars have focused on the pre-processing of waste and energy harvesting. It is worth noting that some scholars have further utilized these hazardous wastes to make self-powered sensors and realized the monitoring of fringe balls in sports activities. This is favorable to the multidimensional development of TENG based on hazardous waste.

This subsection provides an overview of three main areas: recyclable industrial wastes, unrecyclable industrial wastes, and

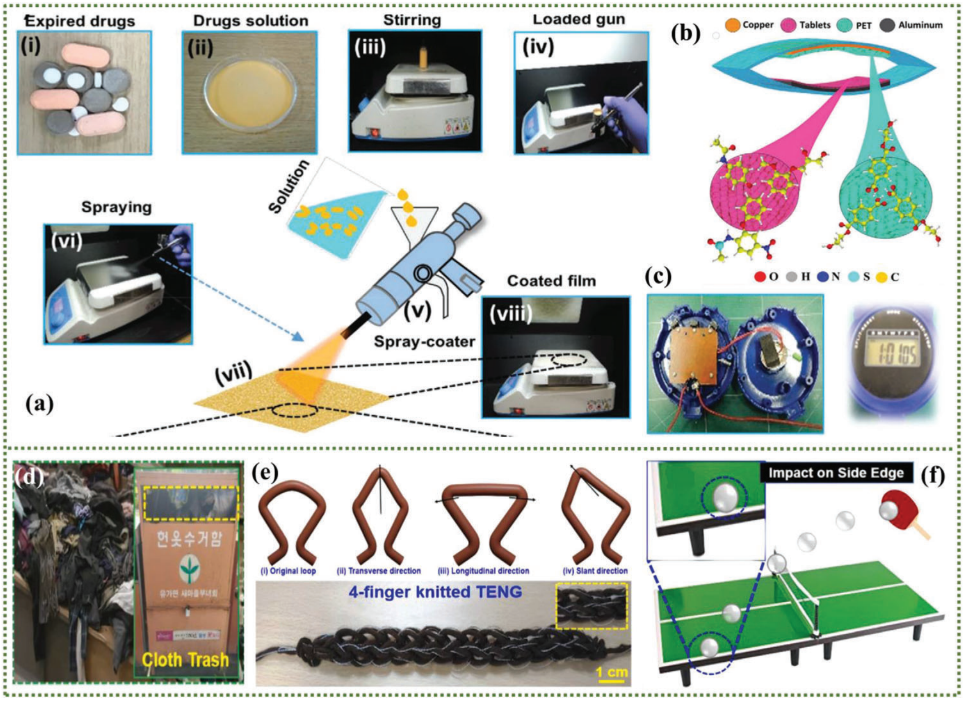

Figure 8. TENG based on hazardous industrial wastes. a) Complete fabrication process of the expired pharmaceuticals based TENG. b) The schematic diagram of the fabricated expired pharmaceuticals-based TENG. c) Implementation diagram of the TENG connected to a stopwatch to realize the power supply to a low-power stopwatch. Reproduced with permission.[70] Copyright 2021, Wiley-VCH. d) Material source of F-TENG. e) Deformation of the knitted textile and digital image of the 4-finger knitted TENG. f) Self-powered edge ball detection system. Reproduced with permission.[57] Copyright 2022, Elsevier.

hazardous wastes. Recyclable wastes are relatively easy for scholars to utilize directly or with minimal treatment to achieve a good performance output, but unrecyclable or costly to recycle industrial wastes due to irregular shapes are usually ground into granules and then made into films to make TENG. while the treatment of those hazardous wastes is more important grinding, dissolving, and other deeper treatments are usually done, but it is much more difficult than the treatment with incineration or burial, this technology not only provides a reasonable treatment of electronic waste, but also realizes the resource utilization of waste electronic components, which provides a new idea for electronic waste treatment.

5. TENG Based on Plant Organs

There are many existing natural materials in nature, the most common of which are the trees, flowers, and other plants that exist all around us all the time. From the direction of environmental protection and low cost, some research scholars use these natural materials to make environment-friendly TENG to achieve various applications such as TENG energy collection and self-powered sensors without harming the environment. Corresponding research has been conducted by previous researchers in this area, and a review on the development of biomaterial wearable pressure sensors written by Hong Pan et al. shows that many researchers have utilized plant organs to fabricate TENGs for wearable sensors that are more environmentally friendly.[24d]

5.1. TENG Based on Leaves

The leaves of some plants are used as TENG's power generation material for energy harvesting. Choi et al. revealed for the first time the existence of discrete liquid-solid contact electrochemistry and the resulting net charge on the surface of natural lotus leaves.[16] As shown in **Figure 9a**, they directly used the surface of a natural lotus leaf to create the first natural lotus leaf-based triboelectric nanogenerator (LL-TENG). As shown in Figure 9b, the LL-TENG has a relatively high energy output performance and can light up 30 LEDs. Figure 9c demonstrates its sustainable power output performance for a month in an external dusty environment with the help of the prominent "Lotus Effect". It is also possible to use the cuticle and internal conductive tissues of leaves as triboelectric materials and electrodes to fabricate TENGs and derive energy from water droplets.[30] Meanwhile, Kim et al. collected leaves to make a fan-shaped TENG and found that its performance output was positively correlated with the number of leaves and vibration frequency.[74] A 3-leaf structure was finalized, which can power 20 LEDs by manual shaking. Saqib et al. fabricated the first natural seagrass-based TENG using a simple spraying technique and compared the friction characteristics of two seagrasses (Zostera marina and Phyllospadix japonicus). The sprayable Phyllospadix japonicus seagrass thin can quickly charge various commercially available capacitors and power ≈150 LEDs and other microelectronic devices.[75]

Research on leaf-based TENG is not limited to energy harvesting it can also be transformed into a self-powered sensor.

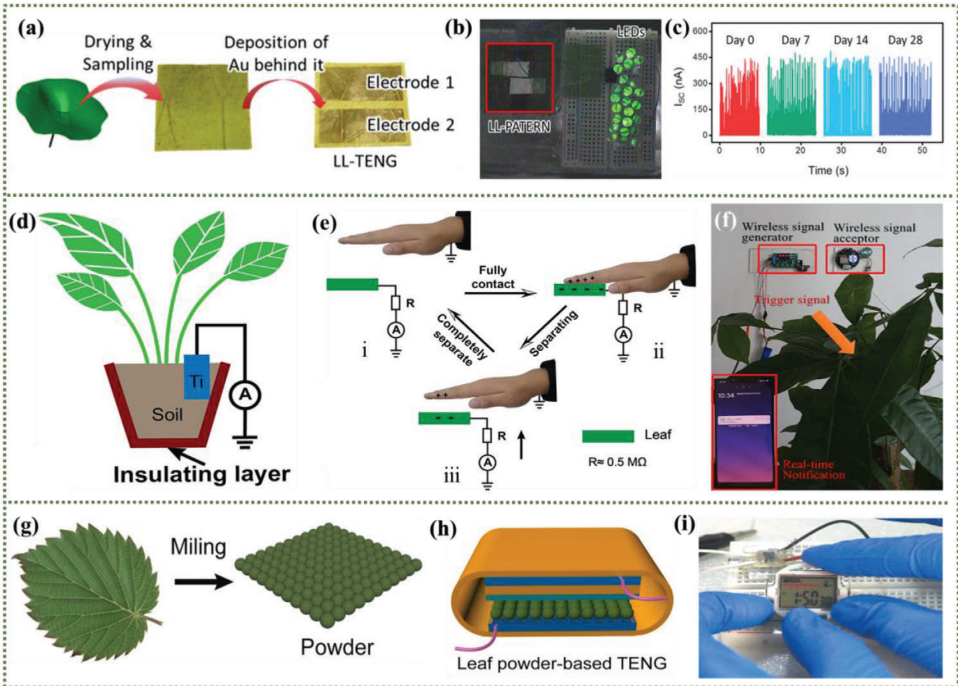

Figure 9. TENG based on plant leaves. a) Manufacturing process of LL-TENG: Selected lotus leaves are dried and gold is deposited on the back of the leaves. b) Power Performance Output Performance of LL-TENG. c) Plot of power output performance of LL-PATERN versus time. Reproduced with permission.[16] Copyright 2017, Elsevier. d) Schematic images of a green flower-based triboelectric generator. e) Schematic of charge transfer in a purely natural leaf. f) Schematic diagram of a wireless contact sensor based on a natural leaf made of TENG. Reproduced with permission.[72] Copyright 2021, Wiley-VCH. g) Process for making powder from leaves. h) 3D structure of a leaf powder-based TENG. i) Spreadsheets powered by leaf powder-based TENG. Reproduced with permission.[73] Copyright 2018, Elsevier.

Luo et al. used plant leaves and composite films to make a live plant leaf-based triboelectric nanogenerator (LPL-TENG), which can be applied in different scenarios such as detecting plant humidity and detecting wind size to promote smart agriculture.[76] Feng et al. designed an all-green TENG based on natural living plants that utilize stems and soil to transfer electrons.[72] Figure 9d shows a single electrode three-phase generator (GFTEG) based on a green blade where the blade, soil, and titanium sheet act as conductive layers. As shown in Figure 9e(i), when the hand and the leaf are completely separated neither surface is charged. As the hand slowly approaches the leaf to reach full contact between the hand and the leaf as shown in Figure 9e(ii), the hand is positively charged the leaf surface is negatively charged. Immediately afterward, as shown in Figure 9e(iii), when the hand and the leaf are separated, the charge of the hand flows rapidly to the ground in an uncharged state because the person is in contact with the ground. The device can also be designed as a contact alarm sensor as shown in Figure 9f. When a hand touches a leaf, the electrical signal is amplified and powers a wireless signal transmitter, which triggers an alarm signal.

The shape of the various leaves was not favorable to the output performance of TENG, thus it was further powdered to improve its output performance. As shown in Figure 9g, Feng et al. ground plant left into powder and made a vertically separated Leaf-based TENG shown in Figure 9h. For the vertically separated TENG, after surface modification, it can easily power the commercial meter in Figure 9i.[73] Babu et al. used extracted Rumex leaf powder and PET as a triboelectric layer. The TENG produces 3.86 V and 3.78 μA electrical output when tapped by hand.[77]

TENGs made based on leaves are divided into three parts on the basis of the depth of material processing and different application areas. The first is the direct utilization of plant leaves for energy harvesting, e.g., studying basic principles such as the lotus leaf effect. This part does not carry out deep processing of leaves, but mainly studies the power generation principle of TENGs based on leaves, providing a theoretical basis for subsequent work. Some scholars directly use leaves to realize self-powered sensing such as using leaves to detect plant humidity, the level of the wind, and so on. However, due to the inconsistency of leaf size and surface details, the energy output and sensing performance of the device need to be improved. On the basis of the above, scholars deep-processed the leaves and crushed them to further improve the output performance of the device, which will help the subsequent development of leaf-based TENGs.

5.2. TENG Based on Flowers and Fruits

In addition to using plant leaves, plant flowers or fruits can also be used as triboelectric materials. Chen et al. creatively constructed TENG using rose and methyl methacrylate.[29] As shown in **Figure 10a**, they selected red roses from China and studied their microstructure by ESEM. Figure 10b shows the operating mode of TENGs using contact separation. Figure 10c

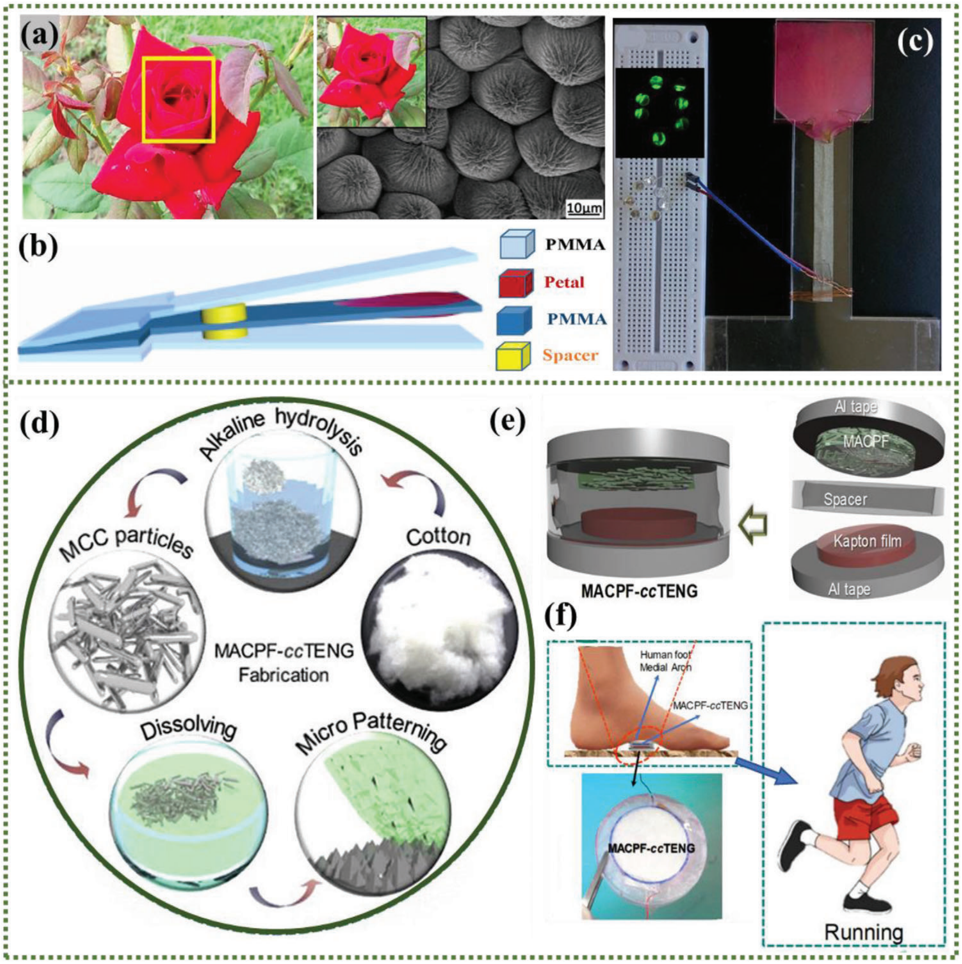

Figure 10. TENG based on flowers and fruits. a) Selection of Chinese red rose petals and their ESEM images. b) Schematic illustration of the petals electricity. c) Chinese red roses as a three-point material to light up the LED Lighted photo. d) CPF-pTENG raw material (cotton) selection and processing flow. Reproduced with permission.[29] Copyright 2018, Elsevier. e) Schematic of the 3D model of the MACPF-based ccTENG device. f) The TENG is placed under the arch of the foot to collect mechanical energy from daily human life. Reproduced with permission.[12a] Copyright 2019, Elsevier.

demonstrates that a TENG made from fresh rose petals (3 cm × 3 cm) can harvest mechanical energy and light up several LEDs. Similarly, Patil et al. crushed Delonix regia flowers (DRFs) as upper electrodes. Combining them with a PTFE film to create a DRF/PTFE-structured TENG can light up 210 light-emitting diodes and run low-power electrons for a few seconds.[78]

Utilizing plant fruits to make TENG for energy harvesting and self-powered sensing. As shown in Figure 10d, Graham et al. used waste cotton to make microcrystalline cellulose (MCC) particles, which were further dissolved into polyvinyl alcohol (PVA) to create novel triboelectric materials.[12a] Figure 10e shows the MACPF-ccTENG fabrication process: waste cotton is combined with a ring-shaped PDMS spacer after a series of treatment processes such as washing, refining, crystallization, and drying, and sealed with cellophane tape. In addition to this, a coin cell type TENG based on MACPF as shown in Figure 10f was developed and placed on the inside of the human foot arch to harvest mechanical energy from daily life to achieve a performance output of ≈600 V, 50 μA, and 84.5 W m^{-2}. Corn, as a common crop was used to develop a self-powered multi-channel wireless agricultural sensing system based on a corn husk composite membrane, pulsed TENG (CH-Pulsed-TENG).[79] The system can simultaneously collect and transmit four different signals of farmland temperature, humidity, light intensity, and soil moisture. Similarly, Zhu et al. utilized disposable eco-friendly starch paper to fabricate TENG for human sweat sensing.[80] Since its output performance varies with the absorbed moisture content it can be used for human sweat sensing.

The plant organs are talked about in the items of flowers and fruits. Scholars' research on the use of flowers as tribo-materials is still in its infancy. The flowers in this area are not highly processed, and the focus is on the physical properties of the thin film-like structure of the flowers as well as the power generation properties, thus providing a basis for the follow-up research. For fruits, scholars mainly centralize further purification or combining with polymer materials to achieve better performance, which realizes a multi-channel wireless agricultural sensing system and mechanical energy harvester as a self-powered electricity source. The method of combining the fruits of plants with existing

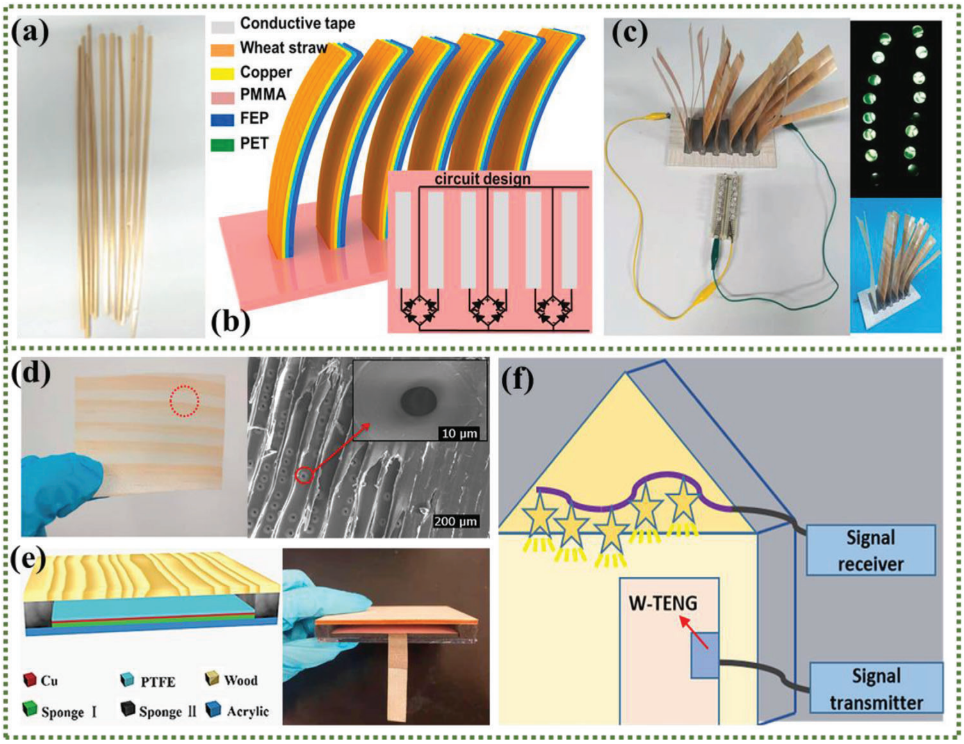

Figure 11. TENG based on plant stalks. a) Material selection of WS-TENG. b) WS-TENG-based Lawn Structure Diagram and Circuit Management. c) LEDs light up with WS-TENG lawn. Reproduced with permission.[81] Copyright 2021, Elsevier. d) Scanning electron microscope images of the surface texture and micromorphology of New Zealand pine. e) Schematic diagrams of W-TENG and actual photographs. f) Schematic diagram of the operation of W-TENG as a doorbell sensor. Reproduced with permission.[84] Copyright 2020, Elsevier.

polymer materials has been found to be beneficial to the performance of the device and is a point of interest for subsequent development.

5.3. TENG Based on Plant Stalks

In addition to the leaves, flowers, and fruits of the plants mentioned above, the stems of the plants can also be used to make TENG. As shown in **Figure 11a**, Ma et al. fabricated friction nanogenerators for converting low-frequency, small-amplitude mechanical energy into electrical energy using straw in a single-electrode mode.[81] The WS-TENG was also fabricated in other shapes, as shown in Figure 11b it was made in the shape of a lawn to simulate the oscillation of grass in the wind and achieved a power output of 404 mW m^{-2}. As shown in Figure 11c, the lawn-shaped WS-TENG is used for wind energy harvesting and lighting multiple LEDs. Bang et al. achieved 90.1 V, 114.4 nA cm^{-2}, and 54.53 mW energy output using a TENG made of natural wood at 8.2 N with a 4.7 × 10^{6}Ω load.[82] Paper as a straw-processed product was used as a support element to propose a stacked TENG, which can directly light up 101 LEDs.[83]

Plant straw-based TENGs can also be used as self-powered sensors. For example, in Figure 11d, Hao et al. fabricated a wood TENG (W-TENG) by studying the structure of New Zealand pine trees. Figure 11e shows its 3D structural diagram and actual structure as well as the vertical-separation mode of operation.[84] As shown in Figure 11f), W-TENG can be used as a doorbell alarm, which will trigger the alarm and light up the light bulb when someone passes by. Similarly, Cai et al. utilized a TENG made from the porous structure of wood and the ammonia-sensitive properties of carbon nanotubes to achieve real-time monitoring of the quality of perishable foodstuffs, which can be used for real-time wireless food quality detection in cold chain supply.[85] Alluri et al. fabricated TENG using natural bio-organic material extracted from a tropical desert plant (Aloe vera). They were used as a self-powered finger-monitoring sensor to monitor the bending angle of human fingers.[86] Park et al. fabricated the mechanically durable and sustainable wood TENG (wood-TENG) and used it as a kinetic energy harvester for humans.[87]

For energy harvesting, scientists rely on the excellent flexibility, support, and plasticity of plant stalks to harvest energy from the environment. Some scholars also utilize its 3D porous structure sensitive to certain substances to achieve real-time monitoring of the concentration of specific substances. Plant stalks are more widely available in nature, and their excellent flexibility and high processability facilitate the comprehensive development of TENGs based on plant stalks in all aspects and multiple fields.

This section synthesizes previous research results mainly from the organs of plants. Plant materials are widely available in nature and can be easily obtained and processed. With the leaves, flowers, fruits, and stalks of plants, TENG realizes the collection of wind and water energy in nature and self-powered sensing in the fields of smart agriculture and smart wearable, which is truly taken from nature and used in nature. These studies show that plant organ-based TENGs have a broad potential application in environmental energy harvesting and self-powered sensors.

6. TENG Based on Biodegradable Industrial Products

The development of sustainable technologies using biodegradable materials from biomass is essential to reduce the impact of non-degradable wastes on the environment. Currently, many scholars combine biodegradable materials with TENG to manufacture green TENG. (mainly categorized into PLA,[88] PBAT,[12c] PBS,[89] gelatin,[90] etc.). After use, biodegradable materials are decomposed into fragments or harmless gases more quickly under the combined effect of the natural environment, thus reducing the bad impact on the environment. In terms of safety, even if a small amount of material is produced or remains during the degradation process of biodegradable materials, it is harmless to the environment and does not affect the survival of human beings and other organisms. In this section, relevant papers will be reviewed from two aspects: solid biodegradable materials and colloidal biodegradable materials.

6.1. TENG Based on Solid Biodegradable Materials

For the study of solid-type biodegradable TENG, some scholars have used methods such as cellulose,[91] metal-type,[92] or enhancement of production technology to prepare TENG. This subsection focuses on the degradation of biodegradable materials from bio-based and petrochemical-based biodegradable materials.[31]

TENG based on petrochemical-based degradable materials: As shown in **Figure 12a**, Liu et al. fabricated fully degradable TENG (FD-TENG) with regenerated cellulose (RC) and PBAT as positive and negative friction layers.[12c] The output performance of the chemically modified RC film can be found to reach 160 V and 10 μA, which are 1.6 and 1.42 times higher than that of the data without chemical modification, as shown in Figure 12b. Figure 12c shows that FD-TENG can be used to charge different electronic devices, monitor human movement, or act as a burglar alarm. poly-ϵ-caprolactone (PCL), a common petrochemical-based biodegradable material, is also used to fabricate TENGs. TENGs made of PCL/ethyl cellulose (EC) composite as the positive electrode can be attached to the human body for energy harvesting and sensing human motion.[93] Similarly, PCL was taken by Parandeh et al. to combine with graphene oxide (GO) and cellulose paper to fabricate TENG and they used electrostatic spinning to fabricate PCL/GO fiber layers consisting of GO nanosheets with different concentrations.[94] After comparison, it was found that PCL/4 wt.% GO cellulose paper yielded the best performance output, driving at least 21 blue LEDs continuously by manual tapping.

TENG based on bio-based degradable materials: As shown in Figure 12d, Wang et al. prepared green-centered green cellulose nanocrystals (CNC)/poly-β-hydroxybutyrate (PHB) nanocomposites using high pressure. Figure 12e shows the schematic 3D structure and working mode of the TENG fabricated based on this material, which was found to exhibit good signal stability after more than 20 000 cycles through experimental studies.[12b] As shown in Figure 12f, it can be fabricated into an arch sensor placed under the human foot to realize accurate real-time monitoring of various human movements. Funayama et al. were the first to utilize laser-induced graphitization of poly (lactic acid) (PLA) material and fabricated a TENG, which can output 1.98 mW m^{-2} energy density at a contact frequency of 1 Hz and an applied pressure of 1 N at a load resistance of 200 MΩ.[88b] Khandelwal et al. went a step further and prepared TENGs using oriented polylactic acid (aPLLA) fibers and chitosan as the active layer.[95] The comparison between aPLLA fiber and random PLLA (rPLLA) fiber revealed that the TENG based on aPLLA fiber exhibited superior performance, yielding an output voltage of 45 V and a power of 9 μA. Xiao et al. went a step further by fabricating ultrasound-driven injectable single-electrode TENGs (I-TENGs) that could alleviate infections and chronic inflammation in humans under their miniaturization, integration, and biocompatibility.[96]

Solid biodegradable materials in this part are classified into two parts: petrochemical-based biodegradable materials and bio-based biodegradable materials. Petrochemical-based biodegradable materials (PBAT, PCL) are mainly polymers made from fossil feedstocks such as petroleum and polymerized by corresponding reactions. Since petroleum-based biodegradable materials usually have excellent soft properties and elasticity, researchers have prepared human wearable devices to realize the monitoring of human activities. Bio-based degradable materials (PLA, PHA) are mainly based on natural substances such as starch and cellulose, which are used to generate plastics in the presence of microorganisms. The easy processing and low cost of bio-based biodegradable materials will enable researchers to further improve the preparation method and hopefully realize large-scale production.

6.2. TENG Based on Gelatin-Based Biodegradable Materials

Since most of the materials described in 6.1 are solids, their slow degradation rate limits the development of green-type TENGs to a certain extent. TENGs based on gelatin-based biodegradable materials have come a long way in recent years.[97] This is mainly due to gelatin-based materials' compact, lightweight, flexible, and stretchable nature. Using them as energy harvesters is expected to revolutionize the market for wearable electronics. They can also be implanted in the body as pacemakers or to eliminate inflammation in the body, among other things, thus providing a new direction for the detection of the body's physiological health, state detection, and disease treatment.

As shown in **Figure 13a**, Kim et al. prepared pure cross-linked HA hydrogel films using solvent evaporation, and TENG (HA-TENG) was fabricated using these HA hydrogel films.[98] Figure 13b shows the structure and power generation mode of the TENG made by combining HA hydrogel films in phase with PTFE films. An electric power output of ≈20 V, ≈0.4 μA, and 5.6 mW m^{-2} was realized. Figure 13c shows its use as an energy harvesting device, which can light multiple LEDs, while Figure 13d shows that the crosslinked HA hydrogel film has a different degradation mechanism than the pure HA hydrogel film thus achieving different degradation rates. Pan et al. fabricated a TENG (BD-TENG) using a gelatin membrane and electrospun

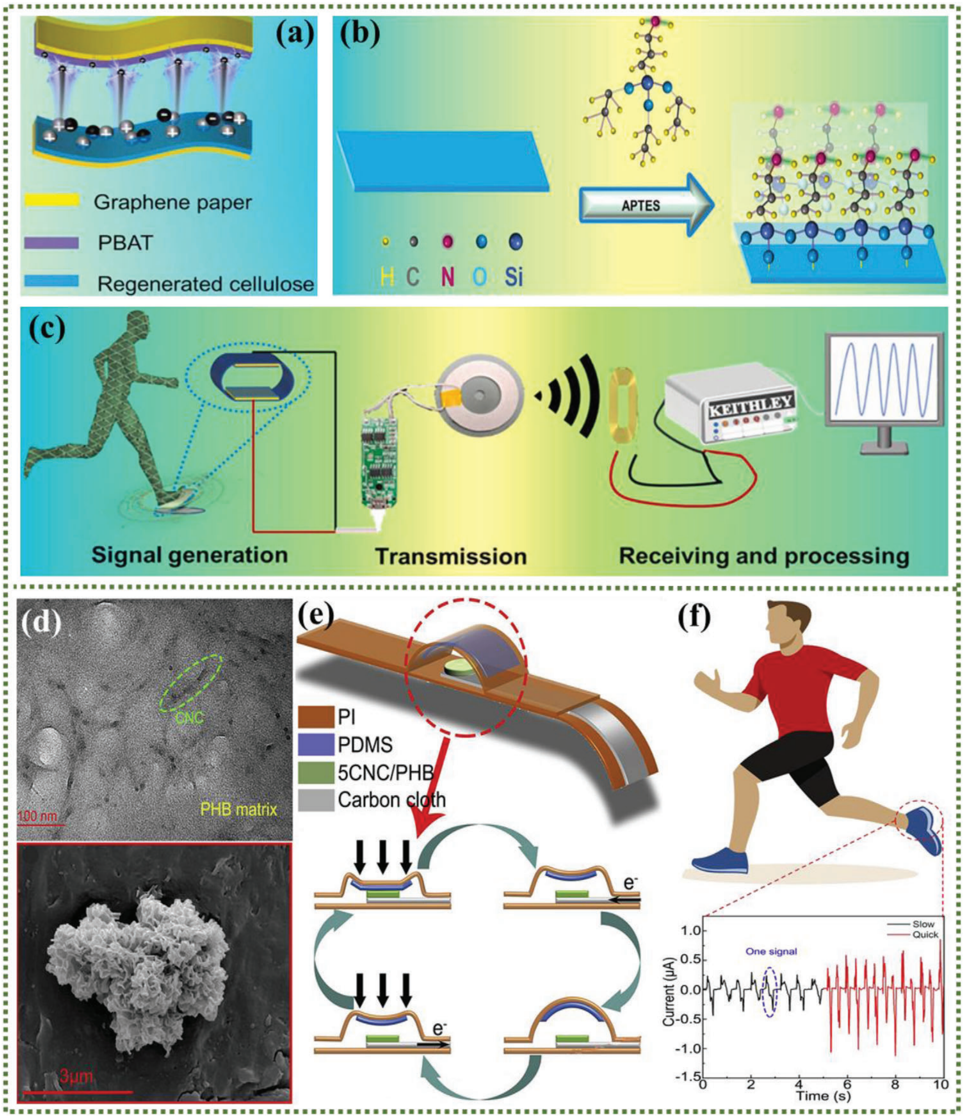

Figure 12. TENG based on solid biodegradable materials. a) Schematic of electron transfer between RC and PBAT materials. b) Schematic diagram of the RC film modified with APTES that contains NH2 groups. c) Schematic diagram of collecting human motion and sensing human motion wirelessly. Reproduced with permission.[12c] Copyright 2023, Elsevier. d) Picture of CNC's morphology in the PHB matrix SEM. e) Schematic of the structure of a sensor used for behavioral monitoring and how it works. f) Biological TENG sensors fixed on human shoes outputting current at different speeds. Reproduced with permission.[12b] Copyright 2022, Elsevier.

PLA nanofiber membrane.[99] It can harvest energy from the wind or motion such as a heartbeat, providing a promising green micro-power source for environmental monitoring and biomedical implants. Similarly, inspired by the surface microstructure of leaves, Shi et al. fabricated a fish gelatin-based TENG that can be attached to the human body to detect motion signals or to collect mechanical energy from the body.[100]

With the properties of gelatin-like materials, it is easier to be used as a wearable or implantable sensor. Li et al. prepared PEGDA/Lap hydrogels by combining polyethylene glycol diacrylate (PEGDA) with soapstone.[17] Figure 13e shows a schematic of its triple network structure. Figs i-iii show the whole process of PEGDA from chemical cross-linking of oligomers to form a network to secondary interaction of high molecular weight PEG chains with Laponite nanoparticles to photocrosslinking between the photoinitiator and the acrylate groups on PEGDA. Figure 13f shows the excellent stretchability of this composite gel material and the schematic structure of a single electrode TENG constructed based on this material. As shown in Figure 13g, BS-TENG can be used as a human health and motion detector to realize real-time monitoring of respiration and various body movements, which plays an important role in the biomedical field. Meanwhile, Ghosh et al. developed a 3D microstructure-based, fully healing but fully biodegradable high-performance ionic bio-gel device by using ionically crosslinked gelatin from biomass resources. It can be used as an electronic skin to monitor low-frequency vital signs and high-frequency sound waves or applied to robotic tactile sensing.[101] Dong et al. developed a

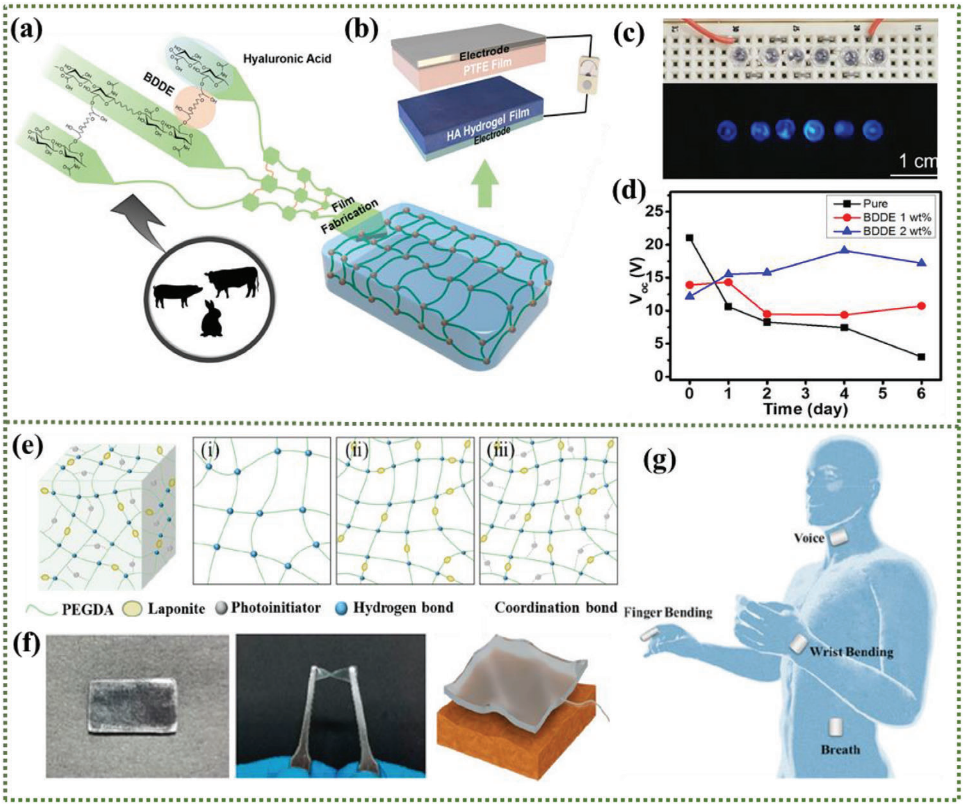

Figure 13. TENG based on gelatin-based biodegradable materials. a) HA Hydrogel film production process: Hyaluronic acid extracted from the mammalian body is made into a hydrogel film. b) Schematic diagram of the HA-TENG structure and the operating modes employed. c) HA-TENG made as an energy harvesting device to light up the LEDs bulb. d) Variation of the power output of HA-TENG with time when exposed to high humidity. Reproduced with permission.[98] Copyright 2020, Elsevier. e) The design principle of the PEGDA/Lap hydrogel. f) BS-TENG good flexibility and stretchability. g) BS-TENG for joint motion monitoring. Reproduced with permission.[17] Copyright 2023, American Chemical Society.

molasses-modified gelatin/ag hydrogel.[102] It can be applied to human motion monitoring, biomechanical energy harvesting, or self-powered information transmission. Similarly, cellulose carbon nanotube aerogel TENG (CCA-TENG) prepared by Wang et al. is characterized by high yield, moisture resistance, simplified structure, and biodegradability, which can be used for environmental energy harvesting and self-powered sensing.[13b]

This subsection divides the degradable materials into two major parts: solid and gelatin, and focuses on the petrochemical-based and bio-based materials in the solid degradable materials: PBAT has better ductility and elongation at break, PCL has good mechanical properties and low upper temperature, but it is expensive, while PLA has reliable biosafety, easy processing and low-cost features that are more widely used. In contrast, the good physical and chemical properties of gelatin-based materials, have low-cost advantages, so that it has a wide range of applications in the pharmaceutical industry, clinical medicine, and clinical treatment, and thus are widely used in wearable, implantable sensors to detect human health.

Gelatin-based materials have received much attention from scholars due to their flexible and stretchable properties, and scholars usually prepare gelatin-based materials by cross-linking through physical or chemical methods. This subsection discusses gelatin-based TENGs energy harvesters and wearable devices according to their different application scenarios. When used as an energy harvester, it may be used to collect the weak mechanical energy of the human body to clean inside the blood vessels of organisms by utilizing its simple structure and small size. With the deepening of research, gelatin-based materials can be more easily used as wearable or implantable sensors to realize the detection of vital signs and health conditions or for robotic tactile sensing by virtue of their fast degradation speed and controllable degradation.

7. TENG Based on Cellulose Materials

This subsection will apply cellulose in environmental protection and low-cost TENGs in two directions: animal fibers (protein fibers) and plant cellulose. There are a lot of review papers related to this direction summarizing the optimization of the structure and properties of cellulose-based TENGs and presenting the status of their applications in areas such as energy harvesting, sensors, and wearable electronics.[24a,103]

7.1. TENG Based on Biofiber Materials

Protein fibers are fibers formed from the hair or secretions of animals, the main types of which are the hair and silk of various

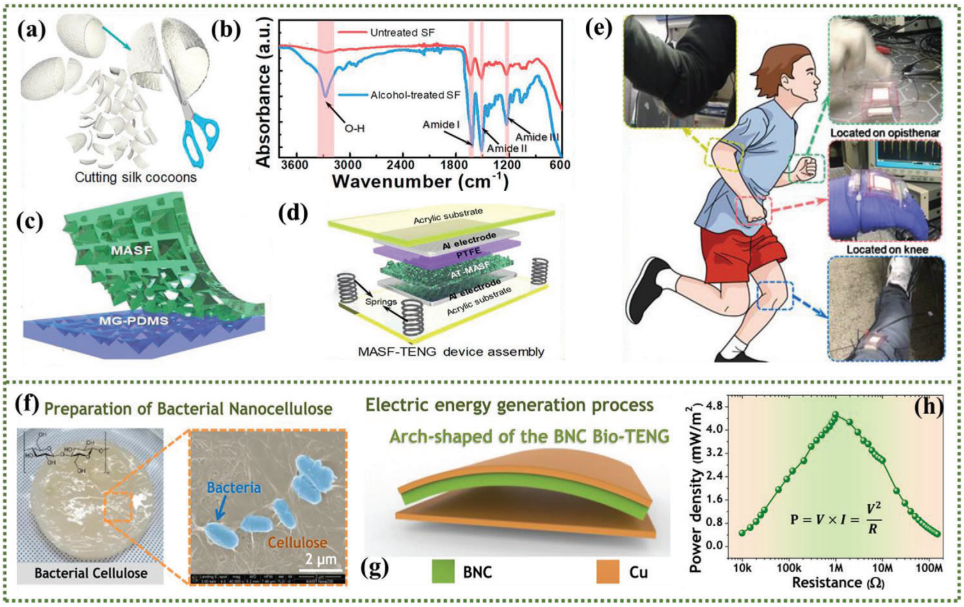

Figure 14. TENG based on biofiber materials. a) Materials used in AT-MASF-TENG. b) Comparison of FTIR spectra of silk before and after alcohol treatment. c) Changes in SF structure before and after alcohol treatment. d) Schematic of the 3D structure of AT-MASF-TENG. e) Schematic for sensing the movement of human hands, arms, knees, and other joints. Reproduced with permission.[104] Copyright 2021, Elsevier. f) Preparation of regenerated bacterial nanocellulose and microscopic schematic. g) 3D schematic of BNC Bio-TENG. h) Output Instantaneous Power Data Plot. Reproduced with permission.[105] Copyright 2017, Elsevier.

animals. It has the advantages of being green and non-polluting, having a wide source of raw materials, and being biodegradable, and can effectively solve the excessive dependence of petroleum-based chemical fibers on petroleum-based chemicals. In addition to this, the microstructured silk protein film has a high surface roughness and can be used as a triboelectric layer of TENG to improve its output performance.

A review on silk fibroin (SF) published by Niu et al. summarizes the working principle of SF-based TENGs and describes their working structures, properties, and application scenarios.[106] It was also found that SF-based TENGs are promising for emerging energy and electronic applications as well as biomedical applications. Dudem et al. extracted crystalline silk particles, SMP, from discarded cocoons of the domestic silkworm, Bombyx mori, through a simple and inexpensive one-step alkalolysis process and used SMP for the first time to enhance the surface charge density of a material such as polystyrene to give a TENG output of ≈280 V, 17.3 μA, 32.5 nC, and 14.4 W m^{-2}.[104] In addition to this, as shown in **Figure 14a**, they further targeted silkworm cocoons using alcohol annealing treatment to enhance the silk film (SF).[33] Figure 14b exhibits the change in the structural properties of silk after subjecting it to alcohol immersion treatment, increasing its β-sheet crystal domains. The silk was dissolved and the purified silk solution was coated on microgrooved polydimethylsiloxane (MG-PDMS) molds, then the microarchitectured SF (MASF) shown in Figure 14c was cast at room temperature and the vertically separated TENG shown in Figure 14d was made by the combination of the MASF, PTFE, and the support device phase. Figure 14e demonstrates the AT-MASF-based TENG for sensing during body-centered activities. Similarly,[18,107] some related scholars have achieved notable energy harvesting by directly utilizing silk secondary processing based on silk[108] or improved processing.[109]

In addition to silk, some scholars have also utilized the hair or fibers of other organisms. Rabbit hair was used as a tribo-material by Han et al. to fabricate a soft contact rotating TENG (SCR-TENG). It can realize the collection of wind energy to light up a light bulb for night indication or integrated detection of soil moisture and ambient temperature and humidity.[110] Wang et al. made a 3D warp-knitted terry fabric TENG (WKTF-TENG) by varying the fabric terry's height and density and utilizing the pile's 3D structure.[111] The results showed that the terry fabric has high output and good abrasion resistance, especially in the transverse sliding mode, and is an excellent triboelectric material that can be used as a self-powered motion monitoring sensor and energy harvesting device.

Special bacterial cellulose (BC) with high purity, polymerization, crystallinity, and hydrophilicity can be used to produce biodegradable TENG. As shown in Figure 14f, Kim et al. processed bacterial cellulose particles into bacterial nanocellulose films for the first time.[105] A TENG was fabricated based on this film as shown in Figure 14g. Figure 14g exhibits its peak power density of ≈4.8 mW m^{-2} at a load resistance of 1 MΩ. Similarly, Pongampai et al. used soft bacterial cellulose/carbon nanotubes (CNTs) instead of metallic aluminum as electrodes. They altered the surface roughness of the device's structural components, which resulted in more than a 1.1-fold increase in the surface charge density and a contact efficiency (η) of 91.67%.[112]

Biological fibers in this subsection are mainly sorted into protein fibers and bacterial fibers. Among them, protein fibers, such as mulberry silk and animal hair, exist in nature, are widely available and easy to process, and are used in large quantities as tribo-materials for TENGs. Protein fibers are relatively mature in

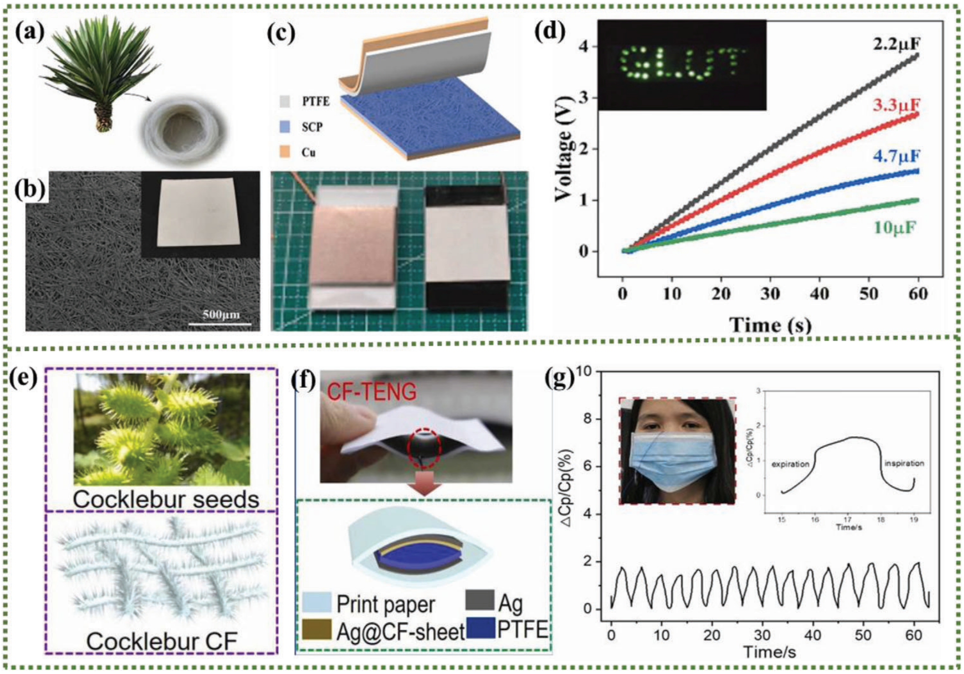

Figure 15. TENG based on lignocellulosic materials. a) The production flow chart of SCP. b) Microstructure of sisal fiber. c) Schematic and physical drawings of the 3D structure of SCP-TENG. d) The charging curve of SCP-TENG to the capacitor and the lit LED lamps. Reproduced with permission.[115] Copyright 2022, Elsevier. e) Schematic based on aleurone fiber. f) Schematic of CF-TENG structure g) Monitoring human motions. response signals of respiration. Reproduced with permission.[117] Copyright 2022, Elsevier.

development and have achieved good performance in energy harvesting and sensing. Bacterial cellulose is a natural nanostructured polymer material mainly produced by bacteria. TENG based on bacterial cellulose is still required to be paid attention to, but its unique microstructure as well as the advantages of high hydrophilicity and high Young's modulus can effectively enhance the output and sensing performance of TENG, and it will have a broader development space in the future.

7.2. TENG Based on Lignocellulosic Materials

Compared to silk, animal, microbial cellulose, and the unmultiprocessed material from plant organs in Section 3. The wide distribution, easier availability, and superior triboelectric properties of the subsequently processed plant cellulose have received much attention from scholars.

Lignocellulose nanofibers (LCNF) in wood were first extracted by Tanguy et al.[113] And the TENG fabricated based on LCNF nanopaper produced 1.6 and 1.2 times higher voltage and current than the TENG using PTFE as triboelectric material. Allicin was first extracted by Roy et al. and was found to enhance the triboelectric properties of cellulose nanofibers (CNFs), resulting in the development of TENGs that achieved 7.9 V and 5.13 μA power output.[114] As shown in **Figure 15a**, Li et al. made sisal cellulose paper (SCP) from sisal cellulose and used it as a friction layer.[115] Their study of the microstructure of sisal cellulose revealed that sisal cellulose has a porous structure as shown in Figure 15b and found that SCP is hydrophilic, with a contact angle of 55.93° with water. They dissolved, dispersed, and filtered sisal to make SCP films and combined it with PTFE films to make the TENG structure shown in Figure 15c. It can be used as a battery to charge a capacitor and can light up to 32 LEDs as shown in Figure 15d.

In terms of self-powered sensors, Zhang et al. used slotted bamboo fibers as triboelectric layers to enhance the TENG output performance. It can also be used as a sensor to detect the bending state of individual joints.[116] As shown in Figure 15e, Lin et al. received the inspiration for the structure of aleurone by refining plant fibers into aleurone-like seeds through high-speed shearing fabricated a TENG (CF-TENG), the 3D structure is shown in Figure 15f. The ear-shaped structure of the fiber design enables the TENG to exhibit excellent output performance.[117] In addition, the CF-TENG exhibits fast response and high flexibility when used as a pressure sensor and can be used as a susceptible sensor to human motion, as shown in Figure 15g. Zhang et al. prepared hydrophilic TENG from natural cellulose for self-powered sensing in high-humidity environments.[118] Yang et al. treated cellulose filter paper (CFP) with PVDF coating and Cu coating as friction layer and electrodes to form a TENG.[119] The CFP-based TENG can be used as a wearable interface for controlling computer programs. Kim et al. developed a chitosan-silica TENG for wearable devices that can be attached to the skin for sensing detection.[120]

Cellulose, found in large quantities in green plants and marine organisms, is one of the most widely distributed and abundant natural polymer materials in nature and is now broadly utilized as part of the tribo-material of TENG. In this subsection, we classify plant fiber-based TENG into two categories based on their application scenarios: energy harvesters and sensors. As energy harvesters, researchers have improved the energy output of

TENGs by subjecting cellulose to processes such as purification. In addition, TENGs made based on plant fibers can also be used for pressure and humidity sensing by virtue of their structural flexibility and sensitivity to temperature and humidity changes.

Natural fibers, as fibers obtained directly from nature's original or artificially cultivated plants and from artificially bred animals, have good chemical and mechanical properties, and natural fibers are highly productive, environmentally friendly, and readily accessible and have been widely used in the field of TENG. This subsection is divided into two parts, biological fibers, and plant cellulose, and provides a systematic review of material acquisition, processing, structural mode of operation of TENG, output properties, and application scenarios as self-powered sensors. Particularly: bacterial fiber (BC) cellulose has become a novel nanomaterial by virtue of its high purity, high degree of polymerization, high crystallinity, high hydrophilicity, high Young's modulus, and many other advantages that have greatly contributed to the prospects of the development of green-type TENGs.

8. Summary and Perspective

In summary, the concept of environmental protection is deeply rooted. The application of green, low-cost, and sustainable energy harvesting devices and sensors is particularly important. **Table 1** summarizes a few representative materials and devices from each section, explaining the origin of the material, the type of material, the use of the material, the power generation performance of the TENG made from the green material, the specific advantages, and the application scenarios as a self-powered sensor.

It is easy to find that these green-type TENGs can not only achieve remarkable energy collection but also be used as self-powered sensors in a wide range of applications such as wireless contact alarms, sensing in smart industry and agriculture, human behavior monitoring, human vital signs monitoring and human health monitoring. Compared with polymeric non-biodegradable materials, green materials have many advantages such as being cost-effective, renewable, biodegradable, and environmentally friendly.[12b,14,16,27,104] Therefore, it has a broad application prospect in the field related to TENGs, but the actual situation of green materials is complicated, they come from a variety of garbage or industrial wastes generated from nature or daily life production, and their quality varies, and although they can be treated by surface physical and chemical modification, etc., the output performance is still low in some cases.[29] In addition, the effect of different treatments of waste materials during collection on the output performance of TENGs is neglected. Second, the effect of different levels of broken, polluted, and obsolete waste on the performance of TENGs also deserves further investigation. The last is that the output is still low compared to mature industrially produced polymeric materials which cannot be produced on a large scale.[26]

Therefore, there are still many challenges and opportunities for future green TENG research as shown in **Figure 16**.

1) Stability/Sensitivity: As summarized in this paper it can be seen that different types of energy harvesting and self-powered sensors based on the green-type TENG have been validated in the laboratory.[10,46,77,93,101] However, the same problem of device stability is faced as in the case of normal TENG (using non-degradable polymer material as a triboelectric layer). First, changes in the natural environment (including humidity, temperature, and pH value) affect the performance output of green-type TENGs. For example, TENGs using food-based materials as triboelectric layers will rapidly deteriorate and deteriorate under high humidity and high temperature, which will severely affect the power output of the TENGs or result in a decrease in sensing sensitivity.[26] Wearable sensors or implantable sensors can be contaminated by human sweat, especially the salts and other substances in human sweat can contaminate the sensor device, thus impairing the stability and sensitivity of the sensing device.[13b,18,79] Second, the physical condition of different green materials varies. For example, factors such as the degree of decay of food waste,[43]
16 the shape and size of plant leaves or fruits,[12a] and the degree of crushing and oxidization of recycling wastes can affect their output performance.[57] Finally, different recycling processing methods also affect the output properties of green materials, for example, dissolved or powdered treatment is relatively more stable than with direct utilization.
2) Percentage of Green Materials: Most of the current studies from the full paper focus on designing, processing, and applying a single green material to the TENG while little research has been done on the optimization of composites derived from green materials.[15,25,26,28,76] For example, Subsections 3–5 of this paper are mainly based on food waste, plant organs, and industrial waste as triboelectric materials for TENG. These relatively single materials such as TENGs made simply by using leaves and plastic bottles compared with the gelatin-based and cellulose-modified materials in Subsections 6 and 7 in terms of both power generation performance and application scenarios as self-powered sensors, which are very insufficient thus hindering the development of green-based TENGs. Second, some articles mentioned in the green TENG to ensure its output performance and stability inevitably use such as PTFE, PVDF, and other non-degradable polymer materials and its combination, only part of the green material manufacturing from the real to achieve the TENG all green material manufacturing there is still a long way to go.
3) Problems Encountered in Large-Scale Production: With the aggravation of global environmental pollution and the enhancement of environmental protection awareness, TENG, as a new emerging technology, has great advantages in low-grade energy harvesting and self-powered sensing, which contributes to the realization of the dual-carbon policy proposed by the state, and the large-scale industrialized production of TENG devices is conducive to the protection of the environment and the harvesting of energy in the natural environment. However, it is difficult to realize the large-scale production of TENG devices, whether it is the traditional TENG made of non-degradable materials or the TENG based on green materials. This is mainly due to the inherent characteristics of natural materials based on green materials such as food waste,[49] plant organs,[73] and cellulose,[117] such as inflexibility, poor abrasion resistance, and structural fragility that do not allow for a long period of time and thus

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Table 1. Summary of the output performance and applications of green TENGs.

Materials Sources	Materials Category	Material Type	Device Performance	Sensing performance	Benefits/Application Scenarios	Refs.
food waste	food material	Leeks, scallions, and onions	35 W m^{-2}	The signal response varies linearly over the 28–56% humidity range.	High-Performance TENG Technology Based on Semi-Cellular Plant Skin As gas sensor and humidity sensor	[46]
		white sugar	6.35 μA and 95.68 V	Realization of long-term sensing in high humidity	Collects mechanical energy. Long-term durability and reliability. Self-powered humidity sensor.	[43]
	food scraps	coffee grounds	50 V, 52 nC, and 2.1 μA	Highly sensitive pressure sensor for pressure detection from 0 to 5.6 kPa	Metal-free and environmentally friendly TENG. Can be used as a supercapacitor Can be used as an electronic skin sensor	[25]
		fish swim bladder	4.56 mA m^{-2} and 25 μC m^{-2}	Sensitivity to humidity and acceleration of ≈446nA s^2 m and 50nA/%RH. Non-contact position sensing performance in the 0–27 mm range	Good biocompatibility and degradability Multifunctional, ultra-flexible, and highly sensitive smart electronic skin	[52]
industrial product waste	recyclable waste	milk carton	600 V and 40 μA	Enables long-term stable wireless sensing	Harvesting of wind and current energy. Enables pH detection in the environment and early monitoring of landslides.	[27]
		PET bottles	67.7 V and 9.4 μA	\	Fluffy, lightweight, and flexible Adsorption of toxic and carcinogenic heavy metals, purification of contaminated wastewater	[56]
	unrecyclable industrial wastes	Waste Polystyrene	4.05 W m^{-2} and ≈90 μC m^{-2}	The error between the sensor and the actual vehicle speed is less than 3%.	Stable operation Real-time speed detection	[10]
		discarded pencil lead	187 V, 28 μA, and 1.2 mW cm^{-2}	BLGM/PDMS films contribute to the sensing properties of the films.	High flexibility High energy output	[66]
	hazardous industrial wastes	expired pharmaceutical	561 V and 53 μA	\	Good stability (12 000 cycles) Provides a new way to dispose of expired drugs	[70]
		worn-out textiles	4.2 V and 2.7 nA	The knitted F-TENG showed an accuracy of 99.64% during practical exercises using the ANN technique.	Easy to knit, versatile construction Harvesting energy from physical activity. Realizes self-powered rim ball judgment system, etc.	[57]
plant organs	leaves	plant leaves.	153.7 V, 2.2 μA and, 253.89 μW	Responds to plant leaf moisture with a sensitivity of ≈ −3.0 V/% RH	TENG based on living plant leaves. Can be used as self-powered smart agricultural sensing.	[76]
		leaves' cuticles	4.6 μA and 35.3 V	Realization of remote transmission of warning signals	All green manufacturing harvests contact, water and wind energy. Wireless contact warning sensor.	[72]

(*Continued*)

Table 1. (Continued).

Materials Sources	Materials Category	Material Type	Device Performance	Sensing performance	Benefits/Application Scenarios	Refs.
	flowers/fruits	rose petal	0.6 V, 0.78 µA, and 27.2 mW m^{-2}	\	Harvesting environmental wind energy, impact energy from rainwater	[29]
		corn husk	3.2 kV	Four different farm signals can be captured and transmitted wirelessly from up to 1.7 km away	Wear-resistant, moisture-proof, non-polluting, and other advantages Realize intelligent agriculture	[79]
	plant stalks	wheat straw	404 mW m^{-2}	All-organic upconverter shows linear dynamic sensitivity toward infrared intensity down to 34 µW cm^{-2}.	Low-cost, small, and portable Versatile construction Collect wind energy and measure wind power	[81]
		natural wood	7 V, 2.4 µA, 16 nC, and 1 nC cm^{-2}	Excellent stability at high humidity (75%) and low temperatures (−18 °C). Exhibits a 0.85 ammonia sensing response at 500 ppm ammonia.	Can be used as an NH3 sensor. Real-time wireless food quality assessment in the cold chain supply chain is realized	[85]
polymer biodegradable materials	solid biodegradable materials	recycled cellulose and PBAT	100 V, 7 µA, and 637 mW m^{-2}	Collecting and transmitting human walking signals at a distance of 3 meters.	Power generation efficiency is 2.22, 3.04, and 5.05 times higher than copper electrodes, respectively. Used as a doorbell or burglar alarm. Completely degraded within three months.	[12c]
		Polyhydroxy butyrate (PHB)	248.8 V cm^3 and 2.5 mA cm^{-3}	Accurately recognizes and detects the activity of different parts of the body based on current signals.	Soft and durable (20 000 cycles) Arch sensors for accurate real-time monitoring of various body movements	[12b]
	gelatin-based biodegradable materials	PEGDA/Lap Nanocomposite Hydrogel	10.4 V and can operate stably for 106 cycles.	Good mechanical sensing properties Enables controlled degradation of sensors	Soft physical characteristics Real-time monitoring of breathing and various body movements is possible. Degradation is controllable and will become the mainstream of diagnosis and treatment.	[17]
		pigskin gelatin	≈325 mW m^{-2}	high-resolution mechano- (≈9 Pa) thermo- (≈0.03 K) transduction functionalities with long-term stability.	Excellent self-healing capability and stable performance output Can be used as multi-functional electronic skin, self-powered human physiological signal monitoring/robotic tactile sensing	[101]
cellulose materials	biofiber materials	Crystallized silk particles	≈280 V, 17.3 µA, 32.5 nC, and 14.4 W m^{-2}	Sensitively detects the movement of the human body's joints by means of different voltages and currents.	Beneficial to the commercialization of TENG, opening up a new avenue for the development of biocompatible TENG on a large scale. As a sensor of human activity	[104]
		Bacterial nanocellulose	≈8.1 µC m^{-2} and ≈4.8 mW m^{-2}	\	Advancing the development of biocompatible TENG for related research.	[105]

(*Continued*)

Table 1. (Continued).

Materials Sources	Materials Category	Material Type	Device Performance	Sensing performance	Benefits/Application Scenarios	Refs.
	lignocellulosic materials	disposable face pack	101.2 V, 2.65 μA, and 20.56 μW cm^{-2}	High sensitivity (0.8/1%) within 40–90% relative humidity (RH).	It can reduce environmental stress and help solve the energy crisis. Can be used as a wearable sensor to monitor the flexion status of joints.	[116]
		cellulose template-based	\	Precise recognition of different degrees of joint flexion through voltage signals	Humidity sensing and respiration monitoring in high humidity environments.	[118]

lead to frequent replacements that are not well suited for use in real-world situations. Second, based on these materials, the secondary processing of the process such as dissolution, crushing, centrifugation, modulation, and other processes are more complex and difficult to achieve industrialized production. Finally, as shown in Sections 3 and 4, the performance outputs of TENGs made from bread,[26] leaves,[77] and other materials with different structural dimensions and detailed textures are not well harmonized.

4) Economics of Material Recycling: Materials such as food scraps and industrial waste are found in complex environments.[46,48c,58,63,66] In addition to themselves, there is a high probability that they include other impurities such as dirt and moisture. In order to ensure the output performance of the TENG and the sensitivity of sensing, it is necessary to sort and clean them. However, this production and living waste from a wide range of sources and complex recycling paths, the relevant enterprises are small, recycling equipment is simple, outdated technology, and the sorting of waste and other materials is not enough mechanization of the required useful waste and impurities are difficult to separate effectively.[121] This results in high costs in the recycling chain and a low utilization rate of recycled resources.

Promote the development of green-type TENGs to overcome the challenges raised above. First, develop new waterproof high-temperature and high-pressure resistant encapsulation processes. Improve its ability to withstand extreme environments to prevent the stability and sensitivity of TENG caused by moisture and heat of the material. Second, for the problem that the use of green materials does not account for a high proportion: increase the optimization research on composite materials derived from recycled materials. Through the development of new materials, such as the formulation of new gelatin materials, the manufacture of composite cellulose materials to enrich the variety of materials to gradually replace the non-degradable polymer materials. Therefore, future research can focus on the study of green composite materials or hybrid materials. In addition, in the case of high recycling costs and poor economy, it is possible to realize low-cost recycling of materials by improving recycling methods or by cooperating with reliable and well-equipped factories. Finally, for the problem of difficult industrialized

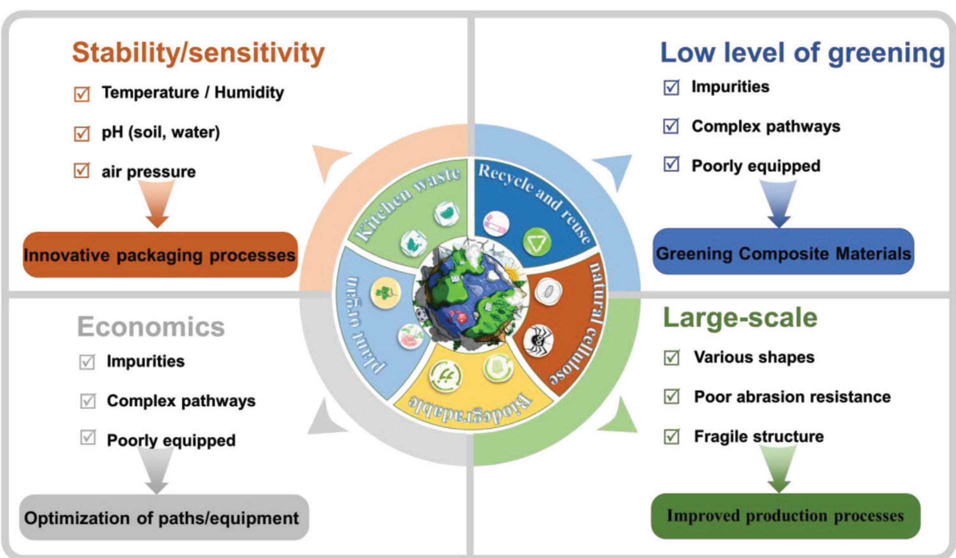

Figure 16. Challenges and prospects for the development of green-type TENG.

production: according to the actual situation of different materials, we should focus on researching and improving the material recovery process. For example, according to the degree of decay of food waste, the shape and size of plant leaves or fruits, and the degree of crushing and oxidation of recycled wastes to implement graded processing, to distinguish the state of different types of green, and to develop simple, safe, low-cost, environmentally friendly recycling methods to deal with the material, and at the same time, to improve the material processing process to simplify the processing of materials thereby realizing the industrialized production of green TENG.

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Conflict of Interest

The authors declare no conflict of interest.

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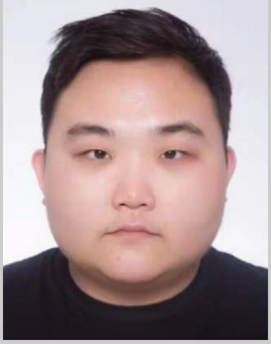

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