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Biopolymer and Biomimetic Techniques for Triboelectric Nanogenerators (TENGs)

Zhaoqi Liu, Xiangyu Chen, and Zhong Lin Wang**

Triboelectric nanogenerators (TENGs) play a crucial role in attaining sustainable energy for various wearable devices. Polymer materials are essential components of TENGs. Biopolymers are suitable materials for TENGs because of their degradability, natural sourcing, and cost-effectiveness. Herein, the latest progress in commonly used biopolymers and well-designed biomimetic techniques for TENG is summarized. The applications of natural rubber, polysaccharides, protein-based biopolymers, and other common synthetic biopolymers in TENG technology are summarized in detail. Each biopolymer is discussed based on its electrification capability, polarity variations, and specific functionalities as active and functional layers of TENGs. Important biomimetic strategies and related applications of specific biopolymers are also summarized to guide the structural and functional design of TENG. In the future, the study of triboelectric biopolymers may focus on exploring alternative candidates, enhancing charge density, and expanding functionality. Various possible applications of biopolymer-based TENGs are proposed in this review. By applying biopolymers and related biomimetic methods to TENG devices, the applications of TENG in the fields of healthcare, environmental monitoring, and wearable/implantable electronics can be further promoted.

1. Introduction

Triboelectric nanogenerators (TENGs) have played a notable role in the field of distributed energy networks;[1,2] TENGs can harvest energy from irregular mechanical motions[3–5] and thereby supply power to various wearable and portable informational devices.[6–8] With the rapid development of smart living,[9–11] various wearable.[12–16] and implantable devices.[17–22] have undergone substantial advancement, and TENGs have also received global attention as a related energy supply technology. One of the most notable advantage of TENGs lies in their ultrawide range of material selections,[23–26] as the contact/triboelectrification phenomenon can occur between all types of materials.[27–29] Through material synthesis and modification, TENGs enable not only a high energy output but can also realize various special functionalities.[30–32] Biopolymers have the advantages of renewability, biocompatibility, and outstanding functional attributes, establishing their indispensable roles in diverse TENG applications.[33–35] Their renewability enhances cost efficiency and permits recycling and reusability in line with the concept of environmental sustainability.[36–38] Biopolymers possess excellent biocompatibility,[39,40] making them particularly suitable for biomedical applications that require contact with the human body, including various wearable electronic devices.[41–43] and sensors.[44–46] based on TENG. Exhibiting a range of unique functionalities, including self-healing,[47,48] moisture absorption,[49,50] elasticity,[51,52] antibacterial properties,[53] and abundant surface groups,[54] biopolymers serve as multifunctional promoters for TENG advancement, and self-powered bioelectronic devices have also become a promising research direction in the field of TENGs.

Furthermore, biomimetic techniques have emerged as powerful approaches for designing and fabricating materials and devices. By mimicking the structures, properties, and functionalities of nature, researchers have been able to create advanced materials with outstanding functions and enhanced performance for TENG applications.[55,56] Existing biomimetic strategies for TENG include structural,[57,58] surface texture,[59] and mechanical principle biomimics.[60] Through diversified biomimetic strategies, TENG can enhance the energy output efficiency and achieve multifunctionality and both the stability and sustainability of TENG systems can be improved accordingly. Neural signals within living organisms are transmitted through electrochemical signals. Some bio-inspired TENG devices have successfully mimicked biological functions, such as simulating the transmission of sensory perceptions, including touch,[61,62] hear,[63,64] smell,[65] and even synapses,[66] achieving a biomimetic system based on triboelectricity. However, challenges that require further research

Z. Liu, X. Chen, Z. L. Wang
Beijing Institute of Nanoenergy and Nanosystems
Chinese Academy of Sciences
Beijing 100083, China
E-mail: chenxiangyu@binn.cas.cn; zhong.wang@mse.gatech.edu
Z. Liu, X. Chen, Z. L. Wang
School of Nanoscience and Engineering
University of Chinese Academy of Sciences
Beijing 100049, China
Z. L. Wang
Georgia Institute of Technology
Atlanta, GA 30332-0245, USA

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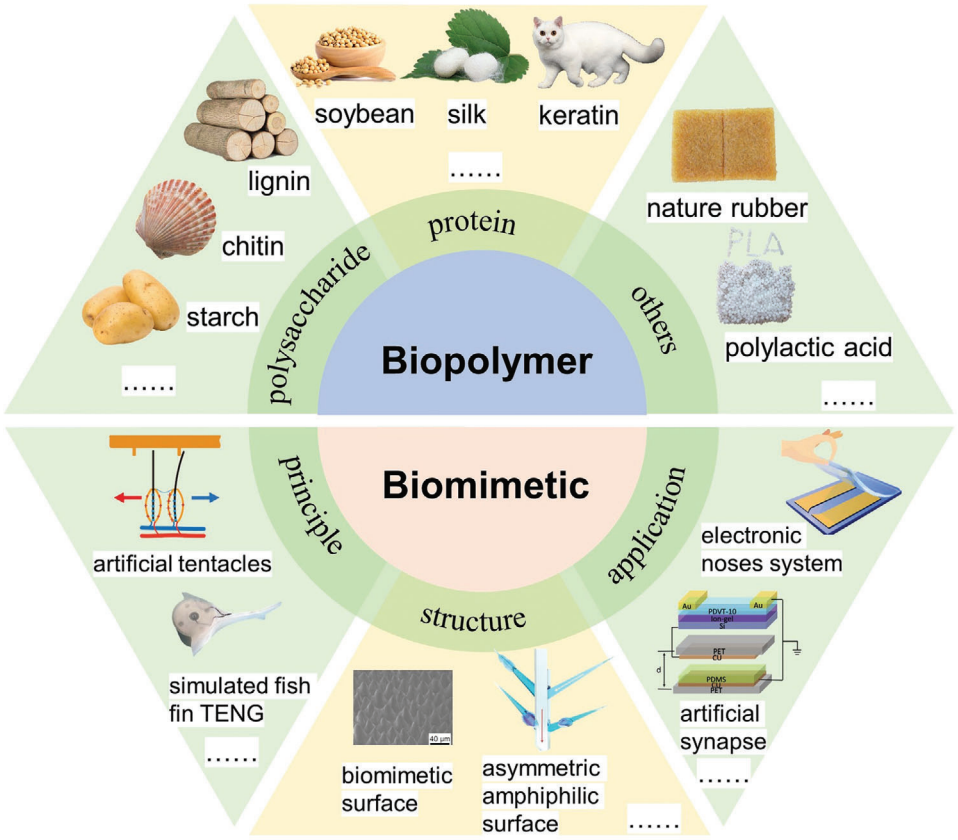

Figure 1. Biopolymer and Biomimetic techniques for TENG related materials. Biomimetic principles: artificial tentacles. (Reproduced with permission).[61] Copyright 2021, Wiley, VCH. simulated fish fin TENG. (Reproduced with permission).[137] Copyright 2022, American Chemical Society. Biomimetic structure: biomimetic surface. (Reproduced with permission).[135] Copyright 2020, Wiley, VCH. asymmetric amphiphilic surfaces. (Reproduced under the terms of the Creative Commons Attribution 4.0 International License).[136] Copyright 2022, the authors. Biomimetic applications: Artificial synapses. (Reproduced with permission).[66] Copyright 2019. Elsevier Electronic nose system. (Reproduced with permission).[139] Copyright 2024, Elsevier.

and resolution remain, such as the complexity of the biomimetic design and difficulties in material selection and fabrication.

This article reviews the biopolymers used in TENGs and self-powered systems in recent years, including polysaccharides, proteins, synthetic biopolymers, and natural macromolecules (**Figure 1**). Each biopolymer was introduced from the perspective of active and functional layers, considering both electrification capability and special functionality as a sensory device. For the active layer, we mainly discuss the triboelectric charge density of materials and their polarity variations as electron donors or acceptors, whereas for the functional layer, we analyze the unique properties and functions of biopolymers, including packaging, degradation, and absorption. Moreover, by examining studies on the application of biomimetic technology to TENG, this article discusses the advantages of biomimetic technology in material and TENG structural design. By summarizing previous TENG devices based on biopolymers and biomimetic designs, the preparation ideas, application prospects, and development directions of TENG-based bioelectronics are proposed. Finally, this article explores further research challenges and development prospects of biopolymers for TENG applications. By developing new functionalities and simple acquisition methods for biopolymers, the performance of bio-related TENG can, not only be enhanced, but various new branches in the field of bioelectronics also be opened up, greatly promoting the future development of green energy and sustainable technologies.

2. Background of TENG

Since its invention by Wang in 2012, TENG has emerged as a notable solution for addressing energy-related challenges.[67] It operates based on the coupling principle of triboelectrification and the electrostatic induction effect.[68] Typically, a triboelectric active layer consists of polymers with different electron gain or loss abilities. When the donor and accepted polymer layers come into contact, the triboelectric effect induces equal and opposite charges on their surfaces, respectively. Owing to the electrostatic induction effect, charges were induced on the electrodes, generating a current in the external circuit to power the load.[69] Generally, triboelectric layer materials exhibit low electrical conductivity or are typically insulators, enabling them to retain charge easily, whereas the electrode materials are composed of highly conductive conductors; consequently, charges induced by electrostatic induction can be readily output to the load. Owing to the wide

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variety and strong electrification capabilities of polymers as well as their excellent insulating properties, they are commonly employed as active layers in TENG systems.

Furthermore, in previous studies, contact-separation mode of TENG has been considered as a parallel-plate capacitor model.[70] and through this simplified model, the influencing factors for evaluating TENG performance are derived to be material and structural factors. Therefore, optimizing the structural design of TENG and breaking through the triboelectric charge density of the triboelectric active layer have become the major methods for improving the TENG output.

Polymers are commonly used as active layers in TENGs because of their excellent dielectric properties and insulating nature. Understanding the charging mechanisms and output enhancement methods for polymers has become a focal point of research. Studies have indicated that the triboelectric output of TENG based on polymers is influenced by factors, such as electron affinity,[71,72] work function,[73] defect states and charge traps,[74] and surface air breakdown.[75,76] This has led to a series of performance enhancement methods, including surface modification, introduction of inorganic fillers, optimization of processing and synthesis methods, and incorporation of new functional groups.

With respect to the structural aspects, enhancing the frequency, increasing the amplitude (to reach a threshold), achieving conformal contact, and achieving complete interlayer separation are pivotal operational parameters for augmenting the TENG output power and designing its impedance. However, increasing the triboelectric charge density and reducing the dielectric constant can boost the output power. Biomimetic technology provides numerous innovative approaches for optimizing TENG structures to achieve high output levels. Additionally, the structural design of TENGs requires careful consideration of factors, such as mechanical durability, material selection, dimensions, weight, environmental adaptability, and manufacturing costs to ensure practical production feasibility.

3. TENG Based on Biopolymers as Triboelectric Materials

Biopolymers are macromolecules derived from living organisms with a molecular structure composed of repeating units of biological origin. Because of their natural origin, biopolymers possess characteristics, such as biodegradability, recyclability, biocompatibility, and excellent functional properties. The advantage of biopolymers used in TENG lies in their biodegradability and recyclability, making TENG more in line with the concept of sustainable development. Moreover, the compatibility of biopolymers with biological systems makes them suitable for wearable devices, aligning well with TENG applications involving human interactions. In flexible electronic devices, biopolymers primarily serve as structural elements owing to their biocompatibility and degradability in humans. The gradual degradation and absorption characteristics of these biopolymers ensure long-term functionality, while avoiding the need for device removal or extraction procedures when used as implantable devices. More importantly, biopolymers have many special properties suitable for TENG, which can improve their output or provide special functions. Biopolymer surfaces contain several active groups, including hydroxyl (- OH), carboxyl (-COOH), amino (-NH2), and aldehyde (-CHO) groups. The ability of the functional groups to gain or lose electrons is closely related to their frictional electrical properties. The rich functional groups can not only help improve the triboelectric output and explore the triboelectric mechanism, but also facilitate the formation of composite materials with special functions for TENG. Additionally, natural rubber (NR) exhibits exceptional extensibility and tear resistance unmatched by synthetic rubber. Gelatin and sodium alginate possess excellent gel-forming characteristics and can form robust gel systems. Chitosan has broad-spectrum antimicrobial properties and exerts notable inhibitory effects on various microorganisms including bacteria and fungi. When integrated into TENG applications, these distinctive attributes confer enhanced functionality to the device.

3.1. Natural Macromolecules

NR is a natural macromolecular compound primarily composed of cis-1,4-polyisoprene with a rubber hydrocarbon content of over 90%. They also contain small amounts of proteins, fatty acids, sugars, and ash. NR is obtained from the latex of rubber trees and exhibits remarkable elasticity and tear resistance, making it suitable for various applications in automotive, manufacturing, and healthcare industries. The excellent properties of NR are attributed to its strain-induced crystallization, which imparts self-reinforcement and contributes to its superior performance compared to synthetic rubbers.[77,78]

When NR is employed as the supporting component of a TENG, its exceptional extensibility and high elasticity are primarily utilized. The NR is integrated within the design of an elastic TENG by fixing it between two copper electrodes, as shown in **Figure 2a**.[79] This innovative structure based on elastic NR enables the output signals of the TENG to exhibit excellent angle detection performance without being restricted by the direction of the stimuli. Utilizing the elasticity of NR, it can also be made into a cavity TENG to achieve self-powered NH_3 sensing.[80] (Figure 2b). NR, which is used as a supporting component of ultra-stretchable TENGs presents unique advantages, exhibits not only exceptional elasticity and toughness unmatched by synthetic rubbers, but also superior biodegradability, thereby posing less environmental harm. However, when influenced by light, heat, and atmospheric oxygen, NR is prone to aging, which leads to a shortened lifespan. Additionally, its dependence on natural sources, such as rubber trees has limitations owing to seasonal and geographical variations. Furthermore, certain individuals may experience allergic reactions to proteins present in NR, making them less common in wearable devices.

In TENG, when employed as the active layer, pure NR assumes the middle position in the triboelectric series; thus, doping materials containing positive or negative functional groups can modify NR into electron acceptors or electron donors. For example, the incorporation of cellulose nanocrystals (CNC) containing high concentrations of ester groups into epoxy natural rubber (ENR) to form a composite material enhances the electron transfer ability of ENR, thereby producing a higher TENG output. Figure 2c illustrates the results of the CNC-ENR composite molecular orbital analysis,[81] which reveals that the highest occupied molecular orbital (HOMO) responsible for electron release

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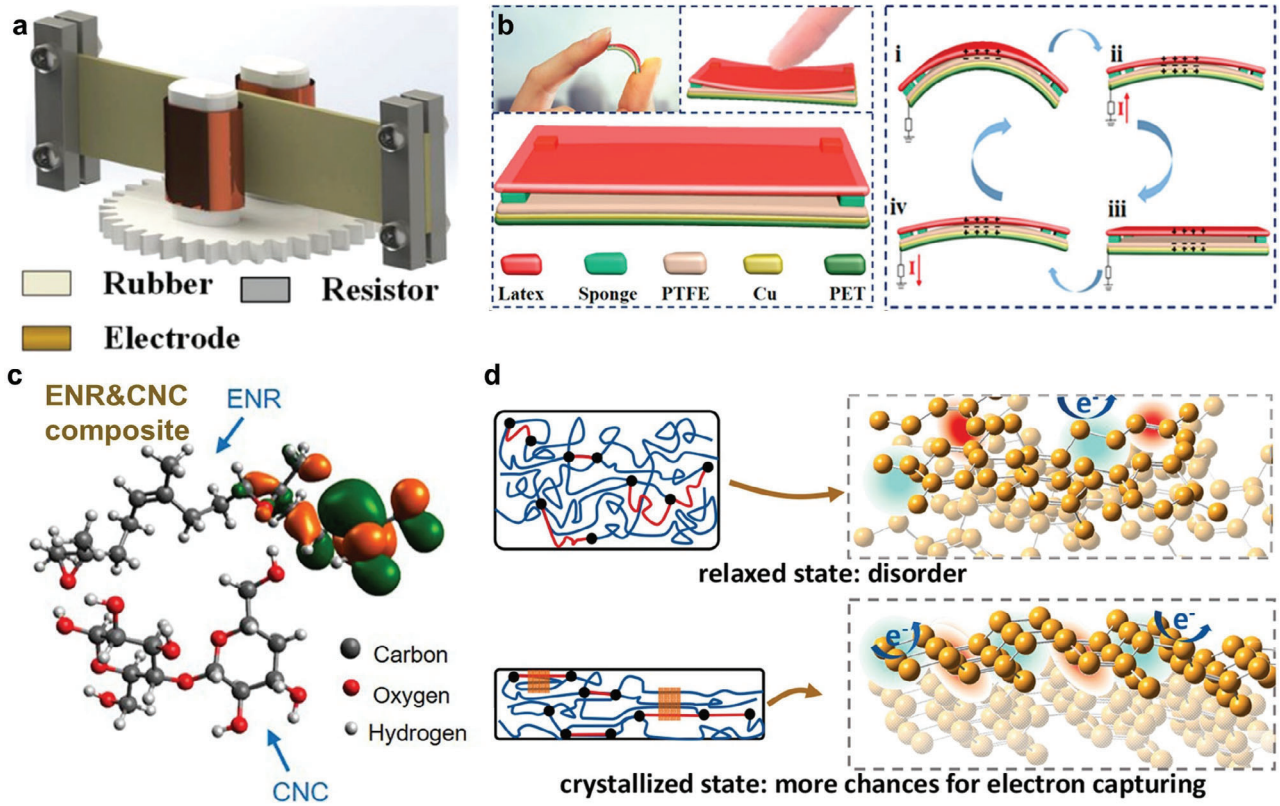

Figure 2. TENG materials based on nature rubbers. a) Schematic of the E-TENG. A flexible and elastic rubber was placed in the middle of the two copper electrodes as one of the triboelectric layers. (Reproduced with permission).[79] Copyright 2019. Wiley, VCH. b) Schematic and working mechanism of the latex-based flexible TENG. (Reproduced with permission).[80] Copyright 2021, American Chemical Society. c) HOMO plots of ENR and ENR/CNC generated using Avogodro-1.2.0 program. (Reproduced with permission).[81] Copyright 2024, Elsevier. d) Schematics illustrating the corresponding NR structures before and after stretching. (Reproduced with permission).[83]Copyright 2022, American Chemical Society.

was situated at the ENR fragment in both ENR/CNC biocomposites, confirming the role of CNC-ENR as an electron donor during contact electrification. Consequently, the output voltage of TENG was increased by 1.75 times, achieving outputs of 80.3 V and 7.4 μA, and a power density of 1.32 $W{\cdot}m^{-2}$ at 9 MΩ. Furthermore, the incorporation of silver nanoparticles (AgNPs) enhances the dielectric constant of NR.[82] The interface polarization effect between AgNPs and the NR matrix contributes to boosting the power density of the TENG device to 0.83 $W{\cdot}m^{-2}$, a six-fold increase compared to previous levels. Moreover, the use of NR in stretchable TENG has distinctive effects, as the crystallization induced by strain and temperature can cause a shift in the triboelectric series and even reverse the triboelectric polarity of elastic materials.[83] (as shown in Figure 2d). This effect is attributed to the notable rearrangement of the surface electron cloud density that occurs during crystallization of the molecular chain. At the initial state, the saturated charge density between NR and a PA film is around 19 $\mu C{\cdot}m^{-2}$. With an applied strain, the charge density of NR (400%) increases to 23.5 $\mu C{\cdot}m^{-2}$, which provides a mechanism distinct from the general understanding of elastic triboelectric materials.

Owing to its toughness, NR can also function as a reinforcing filler, enhancing the mechanical properties of other materials and yielding high-performance composites. For instance, NR-toughened carbon nanotube buckypapers exhibit multifunctionality in electromagnetic interference shielding, thermal conductivity, Joule heating, and TENG.[84] High-performance multifunctional NR-toughened carbon nanotube paper was successfully prepared by the simple vacuum filtration of a carbon nanotube/NR mixed dispersion. The interlinking of NR particles within the entangled network of carbon nanotubes significantly enhanced the tensile strength and fracture strain of the composite buckypaper, while exhibiting a negligible reduction in electrical conductivity.

3.2. Polysaccharide

Polysaccharides have important potential applications in TENG. These naturally sourced biopolymers typically exhibit good friction properties and charge transfer capabilities owing to their rich functional groups, such as hydroxyl and carboxyl groups. They can serve as active or support layers and utilize their excellent compatibility with other materials to provide high-performance triboelectric materials for TENG.

3.2.1. Lignin and Cellulose

Lignin and cellulose are crucial components of plant cell walls. Cellulose is a polysaccharide composed of glucose units linked by

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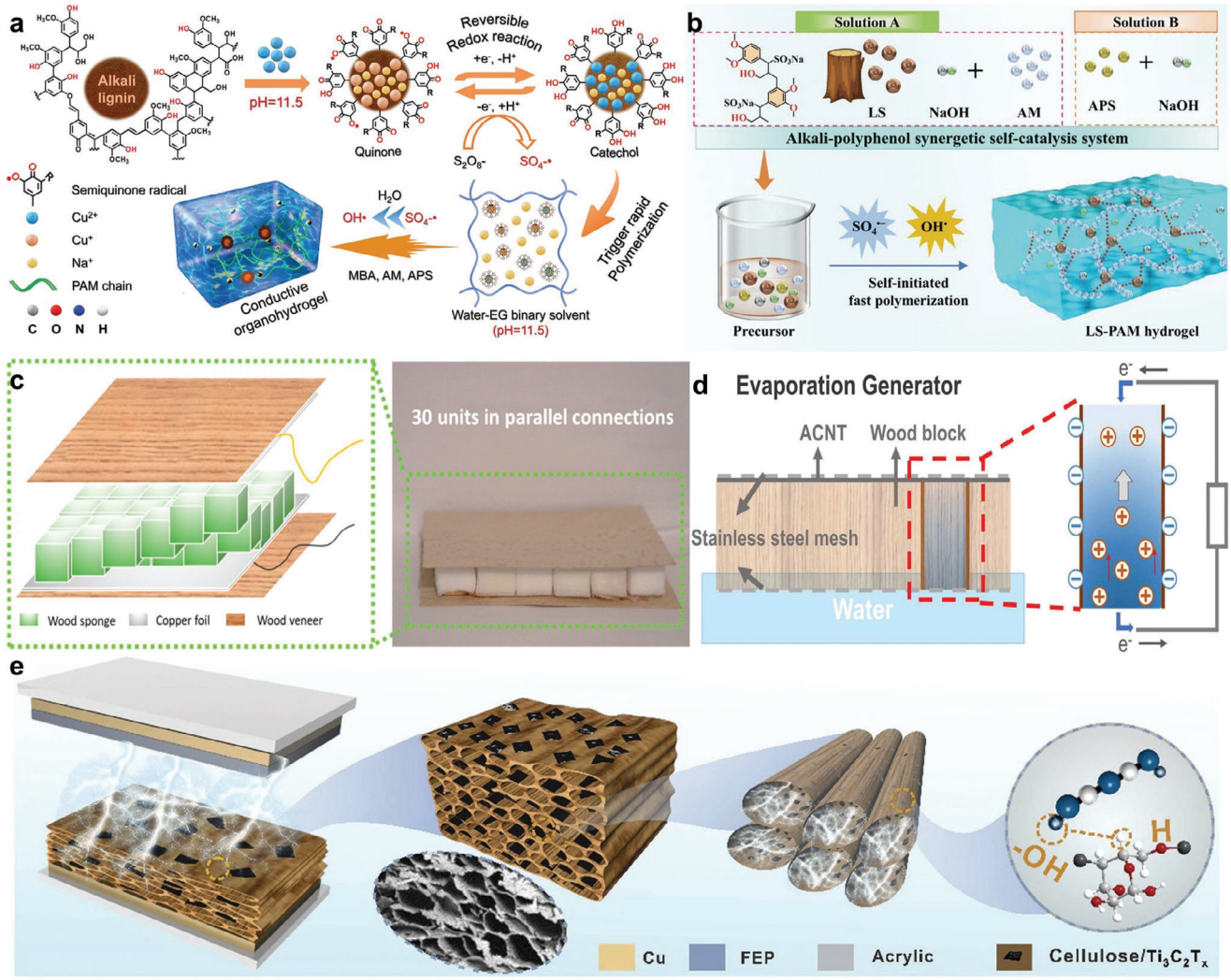

Figure 3. TENG materials and hydrogel based on lignin and cellulose. a) Schematic of the AL-Cu^{2+}-APS dual-catalytic system activating the formation of semiquinone and hydroxyl radicals and triggering the fast polymerization of AM monomers in an alkaline water–EG binary solvent at room temperature. (Reproduced with permission).[89] Copyright 2022, Wiley, VCH. b) Self-initiated polymerization occurs in the precursor, which includes solutions A and B. Reproduced with permission.[90] Copyright 2023, Wiley, VCH. c) Schematic of upscaling by assembling multiple wood sponges. (Reproduced with permission).[97] Copyright 2020, American Chemical Society. d) Schematic of working principle of evaporation generator. (Reproduced with permission).[98] Copyright 2023, Elsevier. e) Schematic of the structure of cellulose/$Ti_3C_2T_x$ composite-based TENG. (Reproduced with permission).[99] Copyright 2023, Elsevier.

β−1,4-glycosidic bonds, and lignin is a complex phenolic polymer that fills the spaces between cellulose, hemicellulose, and pectin in the cell wall, imparting rigidity and hydrophobicity. As both are components of plant cell walls and can be extracted from wood, we introduce them together.[85–88] Lignin plays a reinforcing role in TENG because of its high flexibility and conductivity. Owing to its natural source and exceptional properties, lignin remains a focal point in sustainable energy and renewable material research. Moreover, Lignin contains abundant aromatic structures, aliphatic and aromatic hydroxyl groups, and quinones, which make it suitable for catalytic applications. In a novel lignin-based macromolecular self-catalytic system (AL-Cu^{2+}), the abundant methoxy and phenolic hydroxyl groups of alkali lignin allow it to serve as a polyphenol that participates in a self-catalytic system with metal ions. This system initiated the rapid polymerization of acrylamide monomers in an alkaline water-ethylene glycol (EG) binary solvent, resulting in the synthesis of an organic hydrogel under ambient conditions.[89] (**Figure 3**a). Furthermore, by leveraging the polyphenolic groups within the lignin, an alkali-polyphenol synergistic self-catalysis system was established, as shown in Figure 3b. The alkali-polyphenol synergistic self-catalysis system refers to a chemical process in which alkalis and polyphenols (compounds found in various natural sources, such as plants) work together to accelerate chemical reactions without being consumed in the process. This synergistic effect enhances catalytic activity, leading to more efficient and rapid reactions. The addition of lignin to a hydrogel system can catalyze the rapid gelation of self-healing and self-adhering lignin-based conductive hydrogels, which can be regarded as hydrogel electrodes for flexible TENGs.[90]

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Furthermore, multiple hydrogen bonds between lignin biopolymers and polyacrylamide chains impart excellent adhesion and self-healing properties to lignin-based hydrogels. Lignin is an important biopolymer for constructing hydrogel catalytic systems because of its rich active groups and lignin-based hydrogels can be widely used as electrodes or active layers in TENG owing to their excellent conductivity and sustainable sources of renewable biomass. In addition to catalytic applications, the abundant -OH and -COOH functional groups on the surface of lignin not only provide intrinsic electron-donating capabilities but also offer advantages for surface chemical modification.[91] Cellulose, an insulating biopolymer, is primarily used in TENG to provide supporting structures and equipment stability. For example, it is utilized in the production of nanocellulosic materials and has the potential for energy conversion applications, particularly within the domains of flexible electronic devices and mechanical energy transducers.[92] Yao.[93] et al. used cellulose nanofibrils (CNF) as raw materials and employed a chemical reaction method to attach nitro and methyl groups to the cellulose molecule, altering the frictional polarity of the CNF and consequently significantly enhancing the frictional electricity output. Specifically, the synthesized nitro-CNF exhibited a negative surface charge density of 85.8 $\mu C \cdot m^{-2,}$ whereas methyl-CNF showcased a positive surface charge density of 62.5 $\mu C \cdot m^{-2}$, which demonstrates the potential of utilizing natural, biodegradable, and cost-effective cellulose to create novel composite polymers suitable for TENG applications. Comprising nanoscale cellulose particles, CNC exhibit highly crystalline structures characterized by high surface area and excellent rigidity. Peng.[94] et al. embedded CNC into PDMS thin films and contacted an Al electrode to devise a bio-renewable cellulose-based TENG with a stable power output performance. During contact cycling, the study observed that positive charges generated on the CNC surface facilitated electron transfer, inducing additional charges while enhancing the dielectric constant of the composite material, demonstrating the facilitative role of CNC within the TENG active layer in the electron donor capacity.

In addition, wood is often used as a structural support for TENG because of its loose and porous structure.[95,96] For instance, wood can be processed to create a sponge and studies have demonstrated that wooden sponges are capable of generating electricity without requiring electrical polarization, which is attributable to the inherent piezoelectric properties of crystalline cellulose. The piezoelectric effect in wood is due to the mechanical deformation of crystalline cellulose, the unit cell of which closely resembles monoclinic symmetry class 2. As shown in Figure 3c, two copper foils were attached to the radial part of the wooden sponge and then covered with two wooden veneers to form a small nanogenerator that could light up an LED by tapping.[97] When employed as a structural component in TENG systems, natural wood exhibits abundant porous microchannels and shows substantial potential for ion conveyance. Consequently, Figure 3d shows liquid-solid power generation, which efficiently captures electrical energy from the interaction between the water flow and microporous channels.[98] Wood floats effortlessly in water because of its relatively low density, making it a promising material for harnessing blue energy resources. Natural wood or bamboo can be processed into multilayered porous cellulose templates as supporting materials after the removal of a portion of lignin and hemicellulose, as shown in Figure 3e.[99] The resulting cellulose scaffold exhibited a well-organized one-dimensional layered structure and owing to its numerous polar functional groups (-OH), provides abundant active sites for subsequent reactions, such as facilitating the attachment of MXene. The unique advantage of wood as a supporting material is demonstrated by its hierarchical pore structure, which facilitates the diffusion of water molecules, thereby enhancing their interaction with $Ti_3C_2T_x$. The high hydrophilicity and distinctive porous structure of MXene attached to the cellulose template offer promising applications as a wet-sensitive triboelectric material.

3.2.2. Chitosan

Chitosan is a polysaccharide biopolymer typically derived from the exoskeletons of crustaceans. It is known for its biocompatibility, biodegradability, and antimicrobial properties, making it widely applicable in the fields of medicine, the food industry, and environmental applications.[39,45,100,101] Chitosan, which is abundant in $-NH_2$ and -OH functional groups,[102] is commonly utilized as an electron donor in the active layer of TENG. However, Fang et al. indicated that the triboelectric polarity of chitosan could be effectively regulated by adjusting its crystallinity, thereby expanding its potential applications.[103] as shown in **Figure 4a**. This is attributed to the presence of a certain amount of crystalline structure in the high-crystallinity chitosan membrane, resulting in an ordered arrangement of molecular chains. The crystalline structure increases the exposure of the carbon skeleton structures and expands the effective contact positions with the opposite material, while simultaneously reducing the opportunity for -OH and $-NH_2$ group interactions. Owing to the weak electron-donating characteristics exhibited by the carbon skeleton structure (C-C) and methylene (-CH2-) of the chitosan chains, those ultimately transform from electron-donating to electron-accepting upon contact with the polyamide. Consequently, the charge density of the optimized performance reached 262 $\mu C \cdot m^{-2}$, 2.45 times that of the pure chitosan film. In addition, Figure 4b shows that Sun et al. proposed improving the dielectric constant and triboelectric properties of chitosan blends by optimizing the ion and interface polarizations.[104] By incorporating ions and molecules into chitosan to form a blend, the migration rate of the molecular polar groups can be enhanced. For example, adding polyvinyl alcohol (PVA) to chitosan can increase the interface polarization by creating more interfaces between PVA and chitosan, or by adding metal chlorides, such as LiCl or NaCl, where the metal cations form coordination bonds with the -OH groups in chitosan, disrupting the hydrogen bonding content in the blend and thereby enhancing the migration rate of polar groups. Ultimately, cooperation between interface polarization and ion polarization increases the dielectric constant of the blend. The output voltage enhanced film can reach three times (18 V) of the initial output voltage, up to 25 times (200 V) of the initial value after the corona charge injection. Considering the excellent mechanical properties and transparency of chitosan, Zheng.[105] et al. developed an environmentally friendly, flexible, and transparent chitosan@starch composite film via drop casting. This film demonstrated a high transmittance of over 88% in the visible light range. Additionally, the optimized chitosan@starch-FEP

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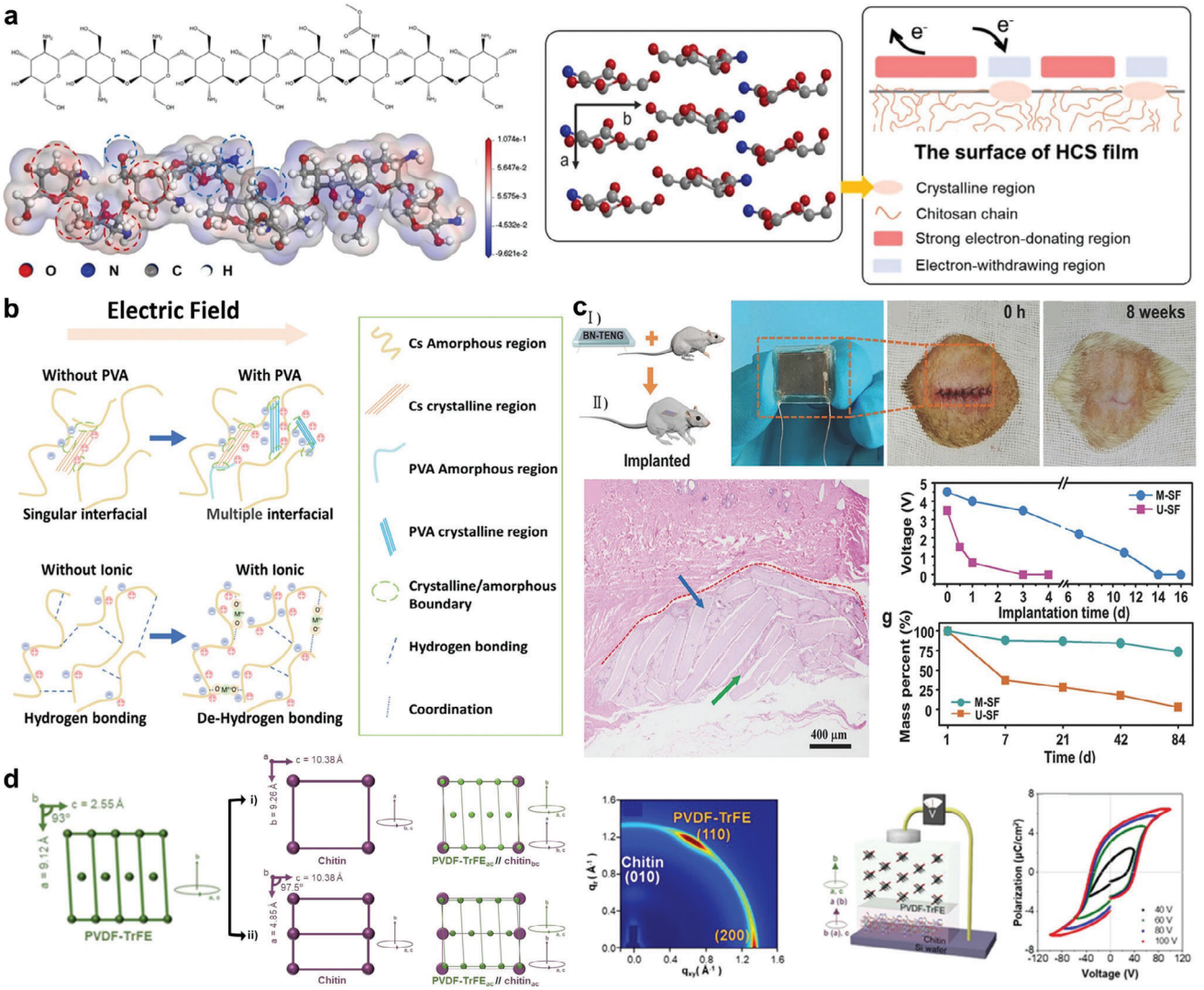

Figure 4. Chitosan as active layer and support layer in TENG. a) Electrostatic potential maps and molecular chain arrangements of the chitosan molecules. (Reproduced with permission).[103] Copyright 2023, Elsevier. b) Schematic of the effects of ions, molecules, and pH on the molecular structure of CS. (Reproduced with permission).[104] Copyright 2021, Wiley, VCH. c) Degradation and triboelectric properties of completely biodegradable TENG fabricated from chitosan. (Reproduced with permission).[106] Copyright 2018, Wiley-VCH. d) Schematic of the proposed epitaxial relationship of the PVDF-TrFE/chitin contact planes. (Reproduced with permission).[107] Copyright 2020, American Chemical Society.

based TENG exhibits remarkable performance, with an open-circuit voltage of approximately 1080 V, short-circuit current of around 16.9 mA·m^{-2}, and a maximum power density of about 5.07 W·m^{-2}.

Chitosan, which possesses special features, such as antibacterial properties, can be employed as a supporting layer in TENG to create a completely biodegradable device. Jiang.[106] et al. used chitosan to produce a completely biodegradable natural material-based TENG. Research has indicated that the manufactured chitosan TENG can be completely degraded and absorbed by rats. Given its impressive in vitro and in vivo electrical outputs, good biocompatibility, adjustable biodegradability, and bioabsorbability, chitosan has great potential for future use as a flexible biodegradable TENG power source (Figure 4c). As shown in Figure 4d, Eom.[107] et al. investigated the epitaxial growth of PVDF-TrFE on a chitin film to develop materials for TENGs. The crystalline structure of chitosan can induce the growth of special crystalline structures of PVDF-TrFE on the surface. The PVDF-TrFE film with special orientation exhibits ferroelectric effects, which is advantageous for accumulating surface charges in TENG applications. In contrast to traditional synthetic flexible polymer substrates, such as polydimethylsiloxane, polyimide, and polyetherimide, chitosan leaves non-toxic residues upon decomposition. The major advantages of chitosan as an epitaxial layer include its natural abundance, ecological friendliness, and robust mechanical properties. Tian.[108] et al. developed a yarn-based TENG by incorporating chitosan fibers during spinning, resulting in the creation of a highly flexible fabric TENG. Operating in the single-electrode mode, the fabricated TENG, driven by a 100 N, 5 × 5 cm^2, and 3 Hz mechanical stimulus,

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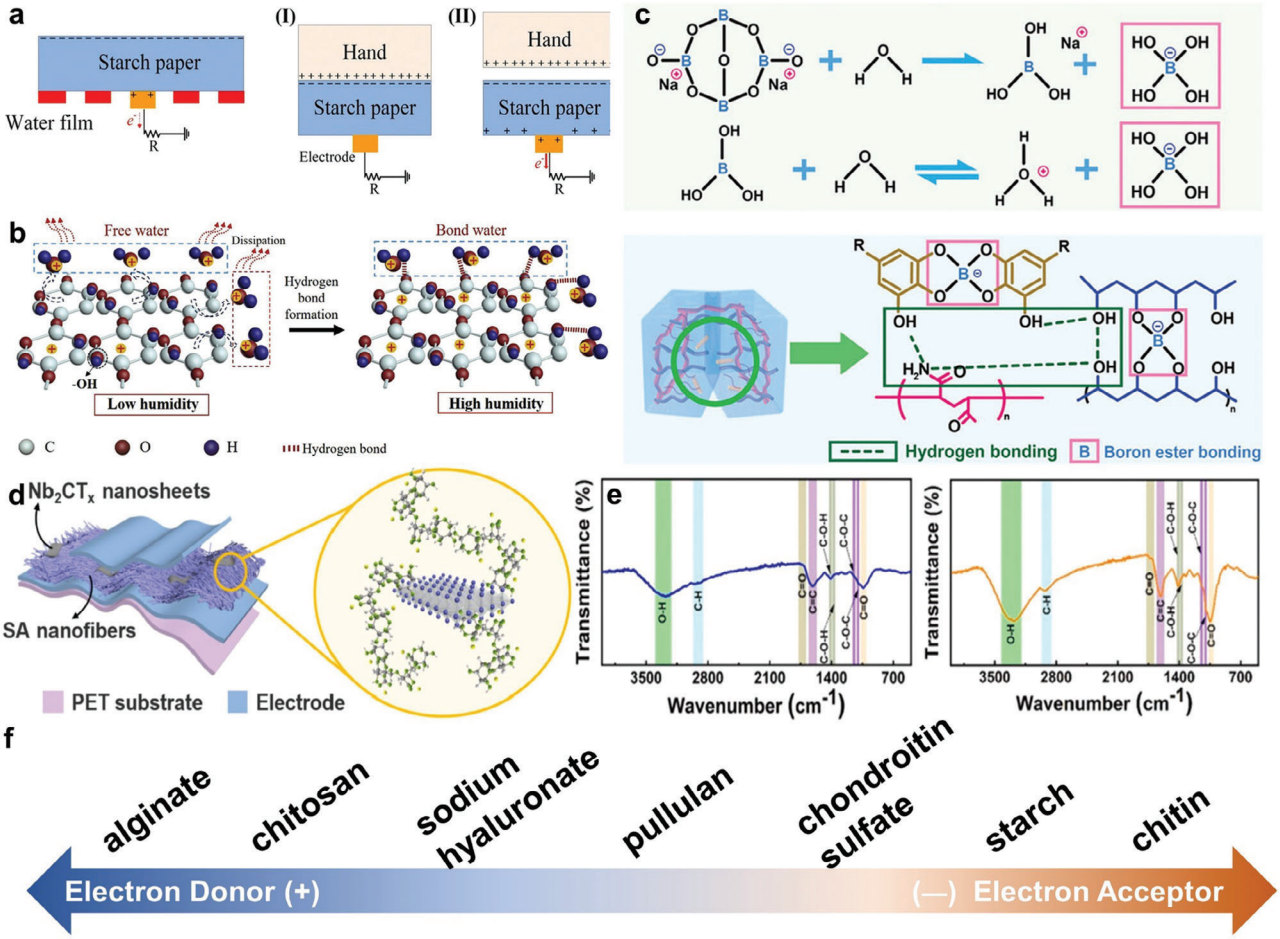

Figure 5. TENG materials based on starch and sodium alginate. a) Working principle of starch-based sweat sensor. (Reproduced under the terms of the Creative Commons Attribution 4.0 International License).[112] Copyright 2018, the authors. b) Mechanism of water molecule immobilization on the biofilm surface and participation in triboelectric charging. (Reproduced with permission).[50] Copyright 2020; Elsevier. c) Self-healing mechanism with the combined effects of dynamic boron ester and hydrogen bonding. (Reproduced with permission).[118] Copyright 2023, Wiley, VCH. d) Schematic of the moist-electric device.[119] Copyright 2022, Elsevier e) FT-IR spectral analysis of spray-coated seagrass films, where different colors highlight their corresponding characteristic peaks. (Reproduced with permission).[120] Copyright 2021, Elsevier. f) Triboelectric series of commonly used polysaccharide biopolymers.

achieved electrical output of 31.3 V, 1.8 μA, and 15.8 mW·m^{-2}. Furthermore, it exhibited satisfactory comfort and outstanding antibacterial properties,[109,110] benefitting to its excellent broad-spectrum antibacterial characteristics. Menge et al. designed a TENG based on chitosan/alginate/silver nanocomposite multilayers, which exhibited antibacterial activity against Escherichia coli strains. Consequently, it is a promising TENG material that is suitable for wearable and implantable electronic devices.

3.2.3. Starch

Starch has garnered attention in the field of TENG owing to its flexibility, renewability, and dielectric properties.[111] Additionally, starch exhibits hygroscopicity and water molecules enter the molecular structure of starch when in contact and interact with the hydrogen bonds within the starch molecules. This process, known as hydration, is the fundamental principle underlying the water absorption of starch. Consequently, starch has extensive utility in humidity TENG and sensor applications.

Zhu.[112] assembled a TENG using a starch film, capitalizing on the hygroscopic properties of starch. They harnessed the coupling effect between the human skin and starch films to develop a self-powered sensor for detecting human sweat. The introduction of water vapor led to the creation of conduits for water within the starch paper surface and interior, thereby reducing the electronic resistance of the starch paper, as the working mechanism is shown in **Figure** 5a. This effect became notably pronounced when a thin layer of water formed around the electrode surfaces, enabling the sensor to sensitively perceive humidity changes and detect sweat.

Figure 5b shows that Wang et al. developed a novel starch-based biodegradable TENG.[50] In high-humidity environments, hydroxyl-rich starch molecules spontaneously form hydrogen bonds with water molecules, thereby anchoring the water molecules to the surface. Consequently, water molecules

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participate in triboelectric charging as more positively charged materials, resulting in a higher TENG output. Unlike traditional polymer-based TENGs, the production yield of starch-film-based TENG increases with increasing environmental humidity. The output current and voltage escalated from 6.2 μA to 110 V and 16.6 μA to 330 V, respectively, as the ambient humidity increased from 15% to 95%. At 95% humidity, it outperformed the traditional nylon-11-based TENGs by approximately 12-fold. This innovation greatly expands the application scope of energy harvesting and self-powered sensors, particularly in high-humidity conditions, such as overcasting, foggy weather, or exposure to water and sweat.

3.2.4. Sodium Alginate

Sodium alginate is a natural polysaccharide extracted from seaweeds and is rich in -OH and -COOH functional groups. It is commonly used in the food industry as a thickening, stabilizing, and gelling agent. Additionally, it has applications in pharmaceuticals, textiles, and various biomedical fields owing to its biocompatibility and non-toxic nature. The -OH and -COOH functional groups in the sodium alginate molecular structure can form hydrogen bonds with water molecules, thereby creating a three-dimensional network structure that gives it excellent gel-forming properties for use in the preparation of hydrogels. Hydrogels are primarily used in TENG to fabricate flexible and stretchable electronic devices. TENG based on hydrogels can easily be affixed to specific areas of the body and their flexibility makes them suitable for capturing energy in conjunction with human motion.[113,114] In most cases, incorporating conductive polymers, metal nanoparticles, or carbon-based nanomaterials into hydrogels enhances their electrical conductivity, making them suitable as electrodes in TENG.[115–117] When serving as the active layer in TENG, hydrogels must exhibit good insulating properties and require relatively distant materials along the triboelectric series to achieve high surface charge density during contact electrification. As shown in Figure 5c, Li et al. employed polyvinyl alcohol-polyacrylamide double-network hydrogels.[118] in the presence of sodium alginate and TA-modified cellulose nanocrystals for in situ polymerization to construct self-healing pressure-sensitive sensors and TENG. These are intended for all-weather, self-powered, and intelligent monitoring applications. Owing to the hydrogen and borate ester bonds, the resulting strain sensor demonstrated rapid self-healing within 1 min and restored its sensing capability with a self-healing efficiency of 97.4%. Zhao et al. used sodium alginate and Nb_2CT_x composites to create a humidity generator.[119] (Figure 5d). As the generator is stimulated by humidity, water molecules are first adsorbed on the surface of sodium alginate through hydrogen bonds and then due to the Grotthus chain reaction ($H_2O+H_3O^+ \leftrightarrow H_3O^+ + H_2O$), physical adsorption occurs to form water membrane and ionize the charge carrier. During the adsorption of several water molecules, cations dissociate from the Nb_2CT_x/SA complex. Driven by the ion concentration difference and flow potential, the positive charges spontaneously migrate along the nanochannel from the permeable side to the dry side, resulting in a potential difference and diffusion current between the two electrodes. In electric humidity generators, sodium alginate plays a special role in the preparation of hydrogels. On the other hand, utilizing the high electron donating ability of oxygen rich -OH and -C = O (Figure 5e), Saqib.[120] et al. found that TENG made from polyethylene terephthalate (PET) film sprayed with seaweed has excellent output performance, with an open circuit voltage of 288 V, a short circuit current of 40 μA, and an instantaneous power of 1690 μW (power density of 70.42 μW cm$-^2$). Figure 5f illustrates the triboelectric series of common pure polysaccharide biopolymers, showing their different functions as electron donors and electron acceptors owing to the differences in their primary functional groups. The strength of the electron donating ability of the side groups followed the order carboxylate group > amino group > hydroxyl group > N-acetyl amide group. Their positions in the triboelectric series generally determine whether they act as electron donors or acceptors.[121] However, because of their rich surface functional groups, they can be modified to extend their positions toward both ends of the triboelectric series, thus enhancing their electron-donating or electron-accepting abilities and expanding the output of TENGs.

3.3. Protein

Proteins are biopolymers, including but not limited to silk fibroin,[122] keratin, collagen,[123] and gelatin.[124,125] Among the protein-based biopolymers, natural silk fibers are widely used in the manufacture of flexible devices owing to their excellent mechanical properties. Silk fiber, a composite of silk fibroin and collagen, is a natural amphiphilic block copolymer that contains several -COOH, -NH2, and -OH functional groups, which endow it with strong electron-donating capabilities when used as the active layer of TENG. These functional groups are hydrophilic and can adsorb water molecules to maintain dry surfaces. Taking advantage of this feature, **Figure 6**a shows a high-output silk-based TENG with durability and moisture resistance.[49] developed by He et al. Utilizing the moisture absorption of these functional groups of the silk fiber, the output of the TENG was stable after 1.5 million cycles, even at a relative humidity of up to 90%. In addition, TENG made of fluffy mulberry silk fibers have a certain degree of elasticity and flexibility, thus avoiding poor contact under small external vibrations. Therefore, even under low pressure, the TENG can avoid the output attenuation caused by small contact areas. Moreover, utilizing recombinant spider silk proteins (RSSP) through genetic engineering, a biologically functional TENG with well-defined mechanical and chemical properties and a high-power output is proposed in Figure 6b. In short, E. coli cells carrying silk expression plasmids were treated with isopropyl-β-D-thiopyranogalactoside to induce silk expression, resulting in RSSP.[126] By constructing RSSPs with different repetitive units through genetic engineering, the output voltage of the 32mer (32 repeating units in one protein molecule) RSSP was enhanced by two times compared with 4mer RSSP, which shows an enhanced triboelectric effect via genetically engineered RSSP. Through genetic optimization of the triboelectric output, the obtained RSSP is positively positioned in the triboelectric series and exhibits high performance when in contact with other materials. Figure 6c shows the structure of collagen in chicken skin,[127] where the triple-helix peptide chain has the basic backbone of the amino acid glycine-X-Y, where X can be

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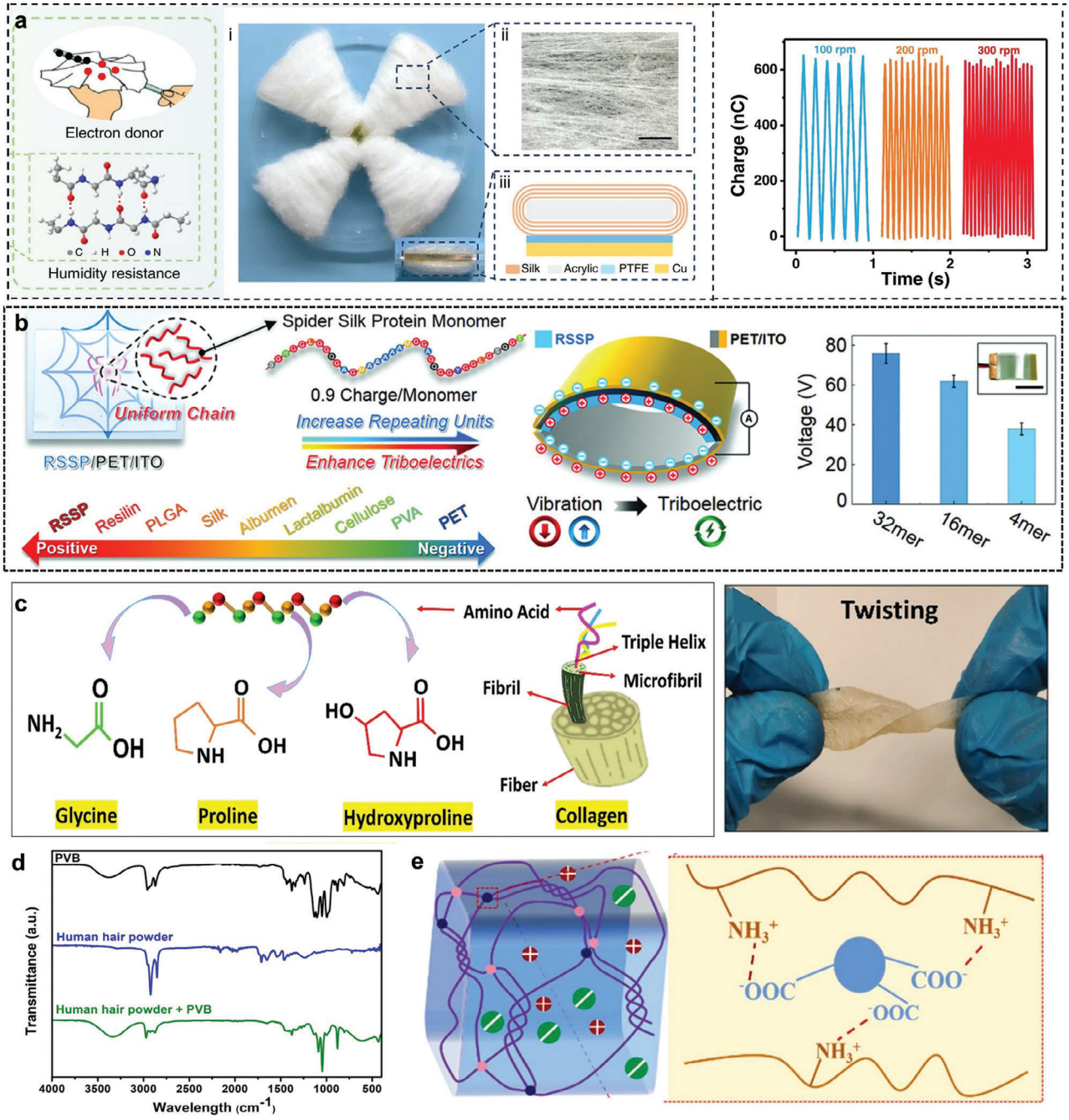

Figure 6. Production and applications of protein based TENG a) Photograph of the rotor made of mulberry silk in MS-TENG, and a diagram of silk and its protein molecular structure. (Reproduced with permission).[49] Copyright 2023, Elsevier. b) Postulated mechanism of genetic modulation of RSSP and working principle of TENG based on RSSP. (Reproduced with permission).[126] Copyright 2018, Wiley-VCH. c) Structure of the collogen and amino acids from chicken skin and flexible TENG. (Reproduced under the terms of the Creative Commons Attribution 4.0 International License).[127] Copyright 2023, the authors. d) FTIR spectra of the pristine PVB film (black), human hair particles (blue), and H-PVB film (green). (Reproduced with permission).[128] Copyright 2022, American Chemical Society. e) Schematic of GNOH synthesized via simple immersion strategy. (Reproduced with permission).[129] Copyright 2022, Elsevier.

proline or lysine and Y is a hydroxylated form. This structure endows the collagen with stable, hydrophilic, and charged properties. These amino acid functional groups contain numerous electron-donating groups, such as -NH_2 and -OH groups, providing an electron-donating ability for collagen in triboelectricity. The 3 × 3 cm^2 sections of skin were utilized in creating the chicken skin TENG (CS-TENG) device, generating an open-circuit voltage of 123 V, a short-circuit current of 20 μA, and a power density of 0.2 $mW \cdot cm^{-2}$ at 20 MΩ. This biocompatible CS-TENG demonstrated exceptionally durable and stable endurance,

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sustained over 52000 cycles. Owing to the inherent structural strength and elasticity of collagen, the CS-TENG exhibited excellent resilience, enabling torsional deformation without damage or performance loss.

Keratin is a fibrous structural protein abundantly found in vertebrate hair, nails, feathers, and horns. Its notable features include a high sulfur content owing to the presence of cysteine disulfide bonds, imparting significant structural stability. Additionally, keratin contains amino acid residues such as glycine, alanine, and serine, which contribute to its mechanical strength and resilience. Keratin is rich in various functional groups, including -NH_2, -COOH, and -SH. These groups are all electron-donating functional groups and when used as the active layer in TENG, they can be positioned towards the positive end of the triboelectric series. Owing to the influence of keratin, human hair exhibits stronger electron-donating ability than commonly used positive materials for TENG, such as nylon-66, indicating its potential for application as a positive triboelectric material in TENG. As depicted in Figure 6d, hair powder has been incorporated into polyvinyl butyral (PVB), which leads to a substantial increase in its electron-donating capability.[128] It was confirmed that at 5 wt% human hair, the maximum output performance was achieved, with VOC and ISC values increasing 1.9 times and 3.1 times, respectively, compared with those of the original TENG. Gelatin is a mixture of peptides and proteins produced by partial hydrolysis of collagen extracted from the skin, bones, and connective tissues of animals. Gelatin is rich in several functional groups, including -CONH, -NH_2, -COOH, and -OH. These functional groups contribute to their unique properties such as their ability to form gels. Figure 6e shows the development of a highly conductive gelatin/NaCl organohydrogel (GNOH) has been achieved by preparing a hydrogel using a mixture of gelatin and NaCl solution followed by immersion in a glycerol/water binary solvent.[129] During the immersion process, water molecules within the gelatin pre-hydrogel were partially exchanged with the surrounding glycerol molecules, leading to the formation of abundant hydrogen bonds, thereby facilitating the transition from a hydrogel to an organic hydrogel. This transformation endowed the organic hydrogel with outstanding anti-freezing and anti-drying properties. Furthermore, the hydrogen bonding interaction between gelatin and glycerol suppressed phase separation, effectively promoting the formation of an organic hydrogel. The distinctive functional attributes of gelatin play a crucial role in the fabrication of hydrogels and their application in flexible electronic devices, such as TENG.

3.4. Synthetic Biopolymers

Synthetic biopolymers are polymers with bio-based characteristics that are artificially manufactured, typically derived from natural resources, and synthesized through chemical processes or biotechnological methods, including the use of polyhydroxyalkanoates (PHA), polylactic acid (PLA), poly[(R)−3-hydroxybutyric acid] (PHB) and polybutylene succinate (PBS). These materials are biodegradable and biocompatible and have wide applications in medical devices, biomedical materials, packaging, and textiles. In the field of TENG, biopolymers can serve as triboelectric active layers or support materials.[18] Owing to their biocompatibility and biodegradability, these biopolymers have potential advantages for TENG applications in wearable devices and medical health monitoring. PHB can be added as a filler and nucleating agent to PLA, and the PHB particles act as crystallization templates,[130] guiding and enhancing the crystallization of PLA. This enhancement leads to an improved overall crystalline performance in the resulting polymer and the synthesized polymer is completely biodegradable. PLA is easily processed and serves as a biodegradable thermoplastic polymer with excellent formability and versatility in 3D printing.[131,132] Consequently, it can be readily fabricated into various specialized shapes for use as structural layers in TENG. This confers an advantage to PLA in TENG applications owing to its flexibility and adaptability, catering to the diverse shapes and design requirements of TENG devices. Furthermore, additional synthetic bio-based polymers, such as a fully bio-based elastomer fabricated using a dual dynamic cross-linking network employing multiple hydrogen bonds and β-hydroxy esters.[133] are designed for use in self-powered triboelectric nanogenerators operating under low-temperature conditions.

4. TENG Based on Biomimetic Techniques

Biomimetic techniques are interdisciplinary approaches that draw inspiration from biological systems, processes, and structures, and leverage these insights to develop innovative engineering solutions. This field involves the simulation of biological mechanisms, often resulting in the development of novel materials, structures, and designs. Biomimetic techniques enhance the efficiency, sustainability, and adaptability of human systems by closely mimicking natural phenomena. TENG based on biomimetic technology are expected to achieve a higher output, optimized structure, and higher efficiency by simulating the surface morphology, design principles, and biological structures in nature.

4.1. Surface Patterning and Texturing Replicating Natural Structures

Surface structuring or texturing biomimetic techniques represent straightforward surface modification approaches that enable the emulation of natural surface morphologies to create specialized microstructured surface layers. A prime example of surface texture biomimetics involves the emulation of the superhydrophobic surface microstructure found on lotus leaves. The surface morphology of a TENG is intricately linked to its output and previous studies have demonstrated that complex surface morphologies (such as nanoforest structures) can increase the effective contact area of a TENG, thereby enhancing its performance. Moreover, in solid-liquid contact electrification, the surface morphology significantly influences the hydrophobic properties, exerting a profound impact on the TENG performance. **Figure 7a** shows the laser shock imprinting technique.[134] developed by Jin et al. Through a reverse molding process, a hierarchical microstructure from natural templates (such as bamboo leaves) is simulated, transferred, and then integrated into the active-layer film in TENG devices. In solid-liquid TENG setups,

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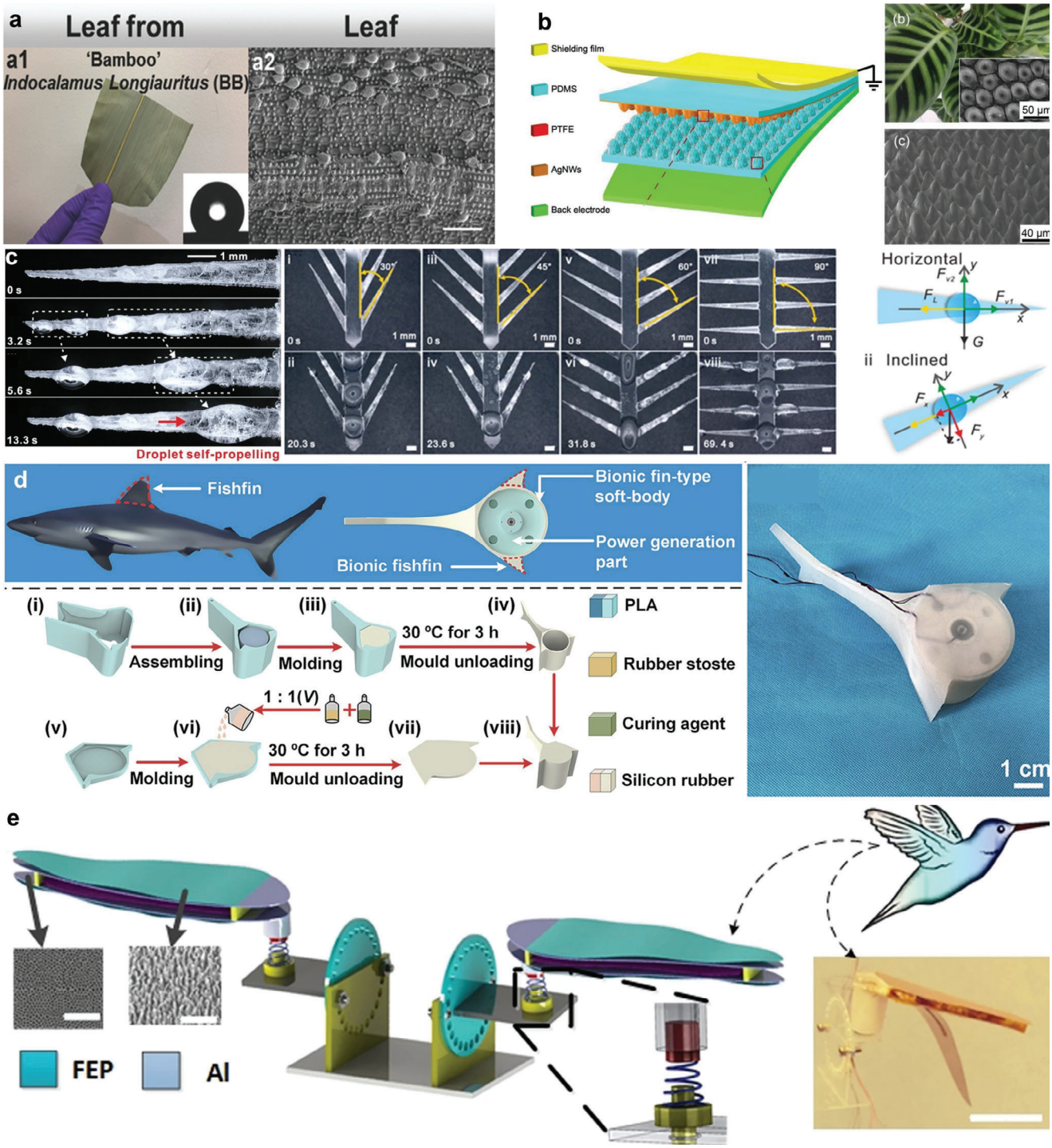

Figure 7. Surface morphology biomimetic and structural biomimetic in TENG materials. a) Leaves from Bamboo and SEM images of the original leaves. (Reproduced with permission).[134] Copyright 2018, Wiley-VCH. b) Structural illustration of TENG e-skin sensor and micromorphology of biomimetic microstructures. (Reproduced with permission).[135] Copyright 2020, Wiley, VCH. c) Fog harvesting on the ACEC@FEP spine and in an array at different angles between the spine and hydrophobic channels (30°, 45°, 60°, and 90°). (Reproduced under the terms of the Creative Commons Attribution 4.0 International License).[136] Copyright 2022, the authors. d) Fabrication schematic of bionic fin-type soft body. (Reproduced with permission).[137] Copyright 2022, American Chemical Society. e) 3D modeling of hummingbird-wing-stimulated TENG wind harvester. (Reproduced under the terms of the Creative Commons Attribution 4.0 International License).[138] Copyright 2017, the authors.

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this film with a bio-inspired microstructure surface expands the TENG output. Moreover, this method is compatible with both optical and soft lithography techniques. Figure 7b shows the surface biomimetic simulation of leaves.[135] as a triboelectric layer material for the TENG. This surface-texture biomimetic approach aims to enhance the overall triboelectric output by increasing the microscale and high-density nanostructures on the surface of the active layer.

4.2. Biomimetic Structural Design of TENG

The biomimetic structural design of a TENG refers to the emulation and incorporation of natural structural features into its development. This involves drawing inspiration from biological structures to create innovative structures. By replicating and adapting these natural structures, researchers aim to improve the efficiency, robustness, and adaptability of TENG or design new working modes for TENG, paving the way for more sustainable and versatile energy harvesting and sensing technologies.

Figure 7c shows a self-driving triboelectric adsorption asymmetric amphiphilic surface inspired by cactus spines and the Coleoptera beetle to obtain clean water from the atmosphere.[136] Laser-carved fluorinated ethylene propylene (FEP) was used as an artificial spike, and an asymmetric amphiphilic surface was constructed using the surface chemical method of nucleophilic substitution. The droplet TENG converts the mechanical energy generated by a droplet falling into electrical energy through the volume effect, thereby achieving excellent output performance and further enhancing electrostatic adsorption through external charges. The water collection efficiency reaches 93.18 $kg{\cdot}m^2{\cdot}h$. This biomimetic strategy provides ideas for the design of water-collection systems and solid-liquid TENGs. Furthermore, by mimicking the limb structures and behavioral patterns of animals, it is possible to develop TENGs with unique structures and functionalities. Animals offer abundant unique limb structures and behavioral patterns, such as the wings of birds, legs, and insects. These naturally evolving structures and mechanisms exhibit excellent performance and adaptability, making them valuable sources of inspiration. Incorporating these biological principles into the design of TENG may lead to more efficient, flexible, and functional energy conversion and collection systems. For example, as shown in Figure 7d, a flexible biomimetic fin structure triboelectric–electromagnetic generator (SF-TEG) with a swing-rotation mechanism was proposed.[137] It consisted of a biomimetic fin-shaped soft-body TENG and an electromagnetic generator (EMG). The lift and drag coefficients of the biomimetic fin-shaped body are enhanced by 9.6 and 2.6 times, respectively, compared to a circular bluff body. Under the driving force of the vortex effect, the soft body vibrates, causing the TENG to collect mechanical energy. The swing-rotation mechanism enables the collection of hydrokinetic energy from water flow. The hummingbird-inspired TENG emulates the structure and flutter mechanics exhibited by hummingbird wings.[138] (Figure 7e). This shape-adaptive, lightweight TENG is specifically designed to harness wind energy on a small scale. Using this method, the flutter motion confined between the two surfaces induces contact electrification during oscillation. The hummingbird TENG (H-TENG) device, weighing only 10 g, was one of the lightest recorded TENGs then. By incorporating a network of six TENG units, the hybrid design achieves a peak electrical output of 1.5 $W{\cdot}m^{-2}$ at a wind speed of 7.5 $m{\cdot}s^{-1}$.

4.3. Biomimetic Principle for Application in TENG

The biomimetic principle is the most difficult and complex mold of biomimetic techniques, and involves the study of the operational principles and processes of animal or plant systems to fabricate an artificial system that imitates the functionalities of living organisms. By emulating the underlying mechanisms and processes observed in biological organisms, biomimetic systems can be developed to replicate and achieve the desired functions in their natural counterparts.

Inspired by the human olfactory system, e-noses emulate the olfactory capabilities by utilizing an array of gas sensors to detect and recognize various gases (**Figure 8a**), mimicking the roles of cells and nerves in the human olfactory system.[139] The working principle of an e-nose involves detecting and identifying the presence of different gases using a gas sensor array, transmitting the collected data to a receiver, and providing real-time information about the gas composition and concentration in each environment. The TENG enables a self-powered supply system by generating a high displacement current, thereby facilitating the real-time emission and transmission of electromagnetic wireless signals for gas sensing. This enables the realization of a unique self-powered, wireless, multidimensional e-nose system. This self-powered wireless e-nose system comprises two main components: a TENG for harvesting external mechanical energy and a tip-to-tip metal structure exposed to the test gas, which accumulates the charges generated by the TENG to initiate gas discharge. This artificial olfactory system emulates the functionality of the human nose and employs deep learning techniques to accurately identify 60 different gas environments, achieving a precision of 93.8%. Tactiles are one of the most important sensing abilities of humans and are also used in intelligent robots. Inspiration from animal vibrissae has led to the creation of artificial tactile systems that primarily rely on a tactile system comprising mechanoreceptors, neural transmission, and the brain to determine direction. Owing to the leverage effect, vibrissae can amplify weak touch signals, thereby providing animals with sensitive tactile perception. Figure 8b shows a biomimetic whisker mechanoreceptor.[61] consisting of a 0.2 mm thick FEP strip similar to an animal vibrissa and a biomimetic follicle covered with two metal electrodes, similar to the sensory nerves of an animal follicle, which can sense the deformation of the biomimetic vibrissa. When the whisker is deflected in different directions, the electrode senses different types of charges and generates triboelectric signals. The detected signals can also be transmitted to the central brain through WIFI, which can uniformly schedule robot clusters. By analyzing the collected data, coordination and cooperation between robots can be achieved, thereby realizing the biomimetic principles of the system. Chen et al. simulated the neural workings of human touch and constructed a virtual tactile system.[140] When the system interacts with objects, artificial sensors convert mechanical stimuli into electrical potential signals and encode them into spike sequences to represent tactile information. Subsequently, the spiking neural network

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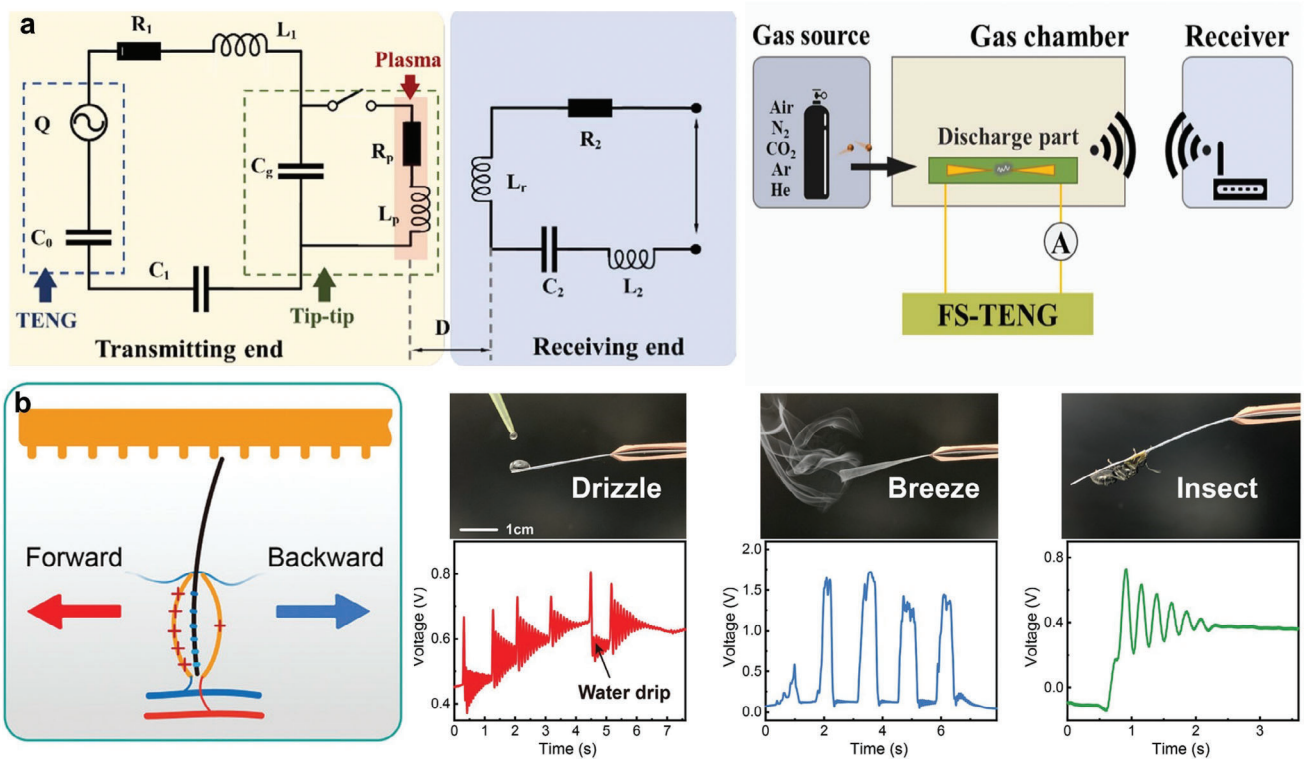

Figure 8. Biomimetic construction of virtual sensation based on biological principles. a) Equivalent electric circuit for a wireless transmission system. (Reproduced with permission).[139] Copyright 2023, Elsevier. b) Schematic of forward and backward stimulation applications. (Reproduced with permission).[61] Copyright 2021, Wiley, VCH.

processed the spike sequences for object recognition and feature extraction. Based on the principle of triboelectricity, Shi et al. developed a virtual tactile device using a TENG and a suspended electrode array,[141] achieving a self-powered, skin-integrated, safe, and painless virtual tactile system. The static electricity triggered by the TENG can induce significant electrotactile (ET) stimulation, while the controlled distance between the ET electrodes and human skin can regulate the inductive discharge current. The advantage of TENG lies in its ability to trigger non-contact static electricity discharge as a stimulus due to its high voltage and its limited triboelectric charge can maintain the inductive current on the skin below 25 μA, thereby generating notable and painless tactile sensations.

5. Summary and Prospects for Biopolymers in TENG

5.1. Summary

In this review, we have discussed the research progresses of biopolymers and biomimetic technique in the field of TENG. Biopolymers, being degradable and derived from natural raw materials, offer low costs after simple purification or processing, aligning with the "low-carbon" environmental protection concept. Furthermore, they possess unique functionalities such as strength, moisture absorption, and stretchability. **Table 1** summarizes the special properties, functional groups, cost, and durable time for triboelectric biopolymers discussed in this paper. By exploring the diverse properties and functionalities of various biopolymers and incorporating them into TENGs, we are able to significantly enhance the performance and stability of TENG. Meanwhile, special surface functional groups make biopolymers suitable for modification and enhance their electron donating or accepting abilities in TENG. We have listed many representative materials and analyzed their unique performance and functional characteristics, while some classic application examples corresponding to each material type are all exhibited.

On the other hand, we also discussed and summarized the application of biomimetic technology in TENG. The application of various biopolymers reflects a kind of application of biomimetic technology, while the implementation of many biomimetic structures and functions also requires the assistance of biopolymers. Therefore, the applications of biomaterials and biomimetic technology complement each other. Existing biomimetic strategies for TENG include structural, surface texture, and mechanical principle biomimics. Surface texture biomimetic refers to designing and fabricating artificial surfaces with similar properties by mimicking the micro or nanoscale structures found on biological surfaces. Structural biomimetic involves emulating natural structures of animals or plants in order to obtain more efficient artificial structures, and then achieving specific functionalities or enhancing TENG output. Mechanical principle biomimetic entails mimicking the working systems of animals or plants to achieve functional replication at a fundamental level. Some bio-inspired TENG devices have successfully mimicked biological functions, such as simulating the transmission of sensory perceptions like

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Table 1. Special properties of biopolymers in TENG applications.

Type	Biopolymer	Special properties	Surface functional groups	Triboelectric performance (in triboelectric series)	Durability	Cost	Degradation time/Method
Natural macromolecules	nature rubber	ultra stretchable, strain induced crystallization	C = C, -CO = NH-	Slight negative	High durability	High	>24 months / Enzymatic
Polysaccharide	lignin	catalyze	-OH (phenolic hydroxyl), -COOH, $-OCH_3$	Quite positive	Moderate	Low	Several months / Microbial degradations
	cellulose	water solubility, high strength and toughness	-OH	Very positive	High	Low	Several weeks / Enzymatic
	chitosan	antibacterial, biocompatible, implantable	-OH, $-NH_2$, $-NHCOCH_3$	Quite positive	Moderate	Moderate	>20 weeks / Enzymatic
	starch	moisture absorption, humidity sensitive	-OH	Quite positive	Low	Low	1 h / Enzymatic
	sodium alginate	gelation	$-COO^-Na^+$, -OH	Quite positive	Low	Low	About 80 days / Hydrolytic disintegration
Protein	silk protein	fluffy and moisture absorption	-COOH, -OH, some nitrogen-containing groups	Quite positive	Moderate	High	6 weeks / Proteolysis
	collagen	elasticity	-OH, -COOH, $-NH_2$	Quite positive	High	Moderate	12 h / Enzymatic
	keratin	strong electron donor capability	-COOH, -OH, some nitrogen-containing groups	Very positive	High	High	About 1 week / Enzymatic
Synthetic biopolymers	PLA	biodegradable, used in 3D printing	-COOH, -OH	Slight negative	Moderate	Low	>24 months / Hydrolytic
	PHB	biodegradable	-COOH, -OH	Quite negative	High	Moderate	>24 months / Bacterial fermentation

touch and smell, achieving a biomimetic system based on triboelectricity.

Despite the notable progresses in the field, the extensive utilization of biopolymers in TENG applications is still confronted with several challenges and limitations. A key issue is the relatively weak triboelectric performance of biopolymers, which hampers the high energy output potential of bio-based TENGs. Additionally, the biodegradability of biopolymers, although environmentally advantageous, can lead to reduced durability of TENG devices. The mechanical properties and stability of bio-based TENGs also require careful evaluation to guarantee their dependability and efficacy in real application scenarios. The development of biopolymers with enhanced functionalities and their strategic incorporation with TENGs to capitalize on their benefits, as well as the application of biomimicry to refine the structural design of TENGs, represent significant challenges for the future. The intricate, often hierarchical structures inherent in natural systems pose a considerable challenge to accurate replication using contemporary fabrication technologies, and the seamless amalgamation of these biomimetic principles with current technological frameworks remains an unresolved issue. The large-scale production and broad application of biopolymers endowed with high charge density, precise charge control, and multifunctionality are indispensable for boosting TENG performance in forthcoming developments. Moreover, the exploration of novel properties of existing biopolymers, especially their electrical attributes,[142,143] can further enhance their integration with TENG applications, thereby expanding the scope and impact of TENG technology in energy harvesting and sensing devices.

5.2. Prospect for Biopolymers and Biommic Techniques in TENG

Figure 9 shows the possible study approaches of biopolymers with TENG and the details are summarized as follow:

5.2.1. Exploring Other Candidate Biopolymers for TENG

The exploration of alternative biopolymers for TENG not only expands the application scope of biopolymers but also introduces a broader range of multifunctional sustainable materials to TENG. In addition to existing biopolymers, some noteworthy potential biopolymers include DNA,[144] polydopamine,[123] glucomannan from konjac.[145] as well as. microbial metabolites.[146,147] such as antibiotics, enzymes, and amino acids. By delving into these and other biopolymers, researchers can broaden the spectrum of sustainable materials available for TENG.

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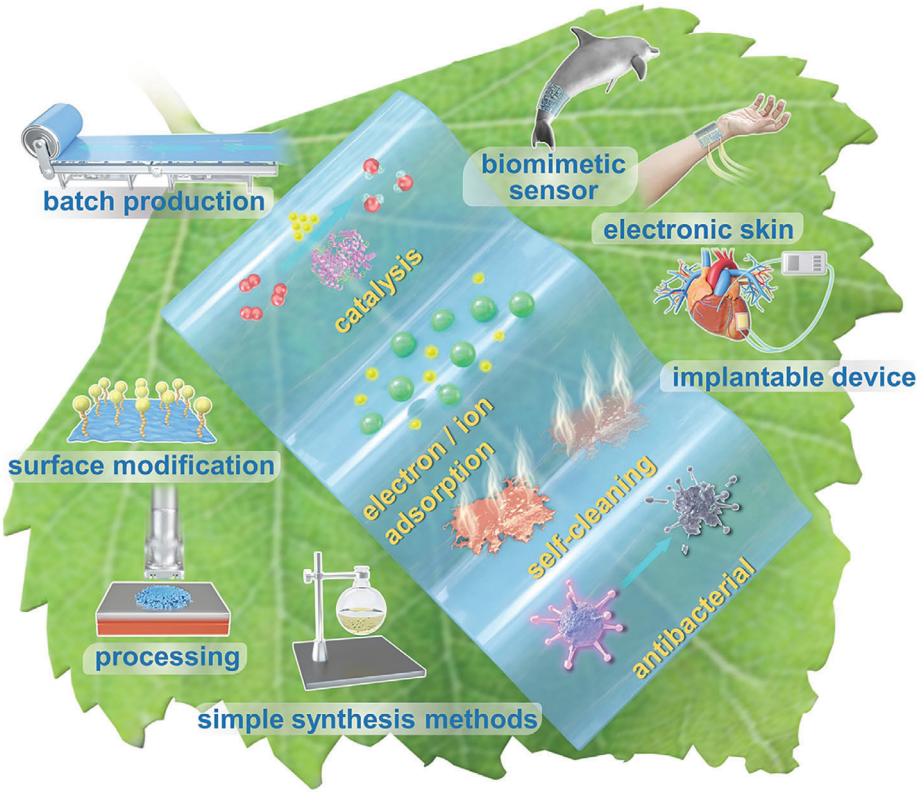

Figure 9. Prospect of biopolymers and biomimetic techniques in TENG technologies.

5.2.2. Boosting the Triboelectric Charge Density of Biopolymers

Improving the triboelectric charge density of polymers involves various methods have ever been studied including surface modification, treatment, synthesis, and processing, which can also be applied to biopolymers for TENG. Surface modification.[148,149] can bring in new functional groups or chemical substances onto the biopolymer surface to alter its surface properties, thereby enhancing the triboelectric charging. Possible surface modification methods include physical or chemical processes, such as plasma treatment or ion radiation,[150] to improve triboelectric performance. Surface grafting.[151] on polymers can also be employed for surface modification, thereby obtaining materials with tunable triboelectric series. Doping with fillers is a simple but effective way to improve the triboelectric performance, which can also be used on biopolymers. Some fillers have excellent triboelectric properties or conductivity,[152] and composite materials formed by doping can be used as electrodes or triboelectric active layers due to these properties. It is also possible to introduce new interfaces in the bulk polymers after doping, resulting in more traps in the composite material, which can better store charges.[153] In addition, biopolymers can also be used as fillers to enhance the performance of other materials.[48] Processing methods also play a crucial role in enhancing the triboelectric charge density of biopolymers. By optimizing processing techniques such as extrusion, compression molding, or injection molding, the molecule structure of bulk biopolymers can be controlled to effectively enhance their triboelectric charging efficiency. Furthermore, specific processing conditions and parameters such as temperature, pressure, and velocity can be employed to improve the triboelectric properties of biopolymer polymers,[154] thereby increasing the triboelectric charge density. Additionally, novel simple synthesis methods involve designing and synthesizing biopolymers with specific structures and properties, such as copolymerization and functionalization.[155,156] The integrated application of these methods can effectively enhance the performance of biopolymers in terms of triboelectric charge density, thereby driving further advancements in their applications within TENG devices. The ultimate goal is to achieve batch production of high-performance triboelectric biopolymers, leading to an increased efficiency in harvesting renewable energy. High-performance triboelectric polymers can provide higher output with smaller size of device, indicating better signal-to-noise ratio for signal transmission.

5.2.3. Developing Special Functions for Biopolymer-Based TENG

Developing biopolymers with special properties specialized for TENG applications holds significant promise for enhancing the comprehensive performance of TENG devices. At the same time, the biomimetic methods based on specific biopolymers are also good approaches for promote the applications of TENG. Some research prospects that can be engineered of biopolymers for TENG applications include:

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- Antibacterial properties: Incorporating antibacterial agents or designing biomaterial surfaces with antibacterial properties can prevent microbial growth on TENG devices. This is particularly important for applications where TENG devices come into contact with organisms, as biomedical sensors or wearable electronics. For example, antimicrobial peptides.[157] are a type of alkaline polypeptide substances with antibacterial activity that are induced in the body of insects, which have strong alkalinity, thermal stability, and broad-spectrum antibacterial properties.
- Self-Cleaning Surfaces: Biopolymers with self-cleaning properties can repel dirt, dust, and other contaminants, thereby maintaining optimal performance over time.[158] Self-cleaning surfaces can reduce maintenance requirements and ensure long-term stability and reliability of TENG devices, especially in outdoor or harsh environments. For instance, enzymes are a type of biocatalyst, which are expected to be applied in self-cleaning TENG.
- Ion or electron adsorption capacity: Researches have shown that polymers that can adsorb ions or electrons from the surrounding environment are beneficial for enhancing the triboelectric properties of TENG active layers.[159,160] It is expected to improve the efficiency of triboelectric charging and energy generation by designing biopolymers with strong electron or ion adsorption capabilities to regulate surface charge density.

5.2.4. Application Areas of Biopolymer-Based TENG

- High voltage TENG: In high-voltage TENG applications, biopolymers can be explored as dielectric materials. For instance, soft rabbit fur and fluffy silkworm silk have been utilized in high-voltage TENG applications. Soft rabbit fur.[161] exhibits excellent triboelectric properties, reducing triboelectric resistance and extending device lifespan, and fluffy silk possesses moisture absorption capabilities, allowing TENG to perform well even in high-humidity conditions.
- contact-electro-catalysis (CEC): CEC is a new catalytic mode based on contact electrification, and biopolymers may show potential applications in CEC.[162–164] By utilizing the special surface properties and functional groups of biopolymers, they can serve as effective carriers or supporting materials, or biopolymers themselves may also participate in contact-electro-catalysis, which is expected to bring new breakthroughs and applications in the field of CEC.
- Virtual sensory sensing system: virtual sensing relies on the ability to accurately detect and interpret physical parameters using smart materials and devices, which can draw inspiration from biomimetic techniques. Biopolymers, with their sensory and responsive properties, can be integrated into virtual sensing platforms to develop robust and versatile sensing systems for applications such as health monitoring, environmental sensing, and human-machine interfaces. Biopolymers have natural biocompatibility advantages over other materials, and can be used in applications such as electronic skin, implantable devices, drug delivery systems, etc., reducing adverse reactions and rejection to the human body.
- Fully bio-based energy devices: Considering future development trends, the integration of biological materials in energy devices represents a promising frontier. Currently, chitosan, starch,[165] and gelatin.[166] have been utilized in the fabrication of bio-based supercapacitors, while battery electrolytes and separators.[167] have also seen the realization of biopolymer production. Incorporating bio-based TENG with these materials holds the potential for achieving entirely bio-based energy devices.

In the future, the development of biomimetic technology requires interdisciplinary cooperation. The development direction of biomimetic technology in TENG includes the development of biomimetic sensing devices, self-healing materials, and intelligent materials and structures for innovative medical devices, wearable technology, and environmental monitoring. For instance, drawing inspiration from animal sensory organs such as the human eye or ear, researchers can design TENG-based sensors that replicate the efficiency and specificity found in natural sensory systems. These biomimetic sensors could find applications in health monitoring, environmental sensing, and industrial diagnostics, offering improved accuracy and functionality. These efforts will bring TENG more intelligent, reliable, and multifunctional solutions, promoting innovative development in the industry.

By incorporating these special properties of biopolymers and biomimetic techniques into TENG devices, researchers can develop new opportunities for applications in diverse fields, including healthcare, environmental monitoring, wearable electronics, and beyond. Moreover, ongoing advancements in biopolymer synthesis, surface engineering, and fabrication techniques are expected to further expand the capabilities and versatility of bio-related TENG devices in the future.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

biomimetic techniques, biopolymers, bio-related devices, self-powered system, triboelectric nanogenerator

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Zhaoqi Liu received her B.E. degree in Chemical Engineering and Technology from Dalian University of Technology in 2020. Now, she is a Ph.D. candidate of Prof. Xiangyu Chen in Beijing Institute of Nanoenergy and Nanosystems, Chinese Academy of Sciences. Her current research interests include high performance triboelectric materials and triboelectric nanogenerators.

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Xiangyu Chen received his B.S. degree in Electrical Engineering from Tsinghua University in 2007 and his Ph.D. in Electronics Physics from Tokyo Institute of Technology in 2013. Now he is a professor in Beijing Institute of Nanoenergy and Nanosystems, Chinese Academic of Sciences. His main research interests have been focused on the field of functional dielectric materials, self-powered nano energy system and the nonlinear optical laser system for characterizing the electrical properties of the devices.

Zhong Lin Wang received his Ph.D. from Arizona State University in physics. He is now the director of Beijing Institute of Nanoenergy and Nanosystems. Prof. Wang has made original and innovative contributions to the synthesis, discovery, characterization and understanding of fundamental physical properties of oxide nanobelts and nanowires, as well as applications of nanowires in energy sciences, electronics, optoelectronics and biological science. His discovery and breakthroughs in developing nanogenerators established the principle and technological road map for harvesting mechanical energy from environment and biological systems for powering personal electronics. His research on self-powered nanosystems has inspired the worldwide effort in academia and industry for studying energy for micro-nano-systems, which is now a distinct disciplinary in energy research and future sensor networks. He coined and pioneered the field of piezotronics and piezophototronics by introducing piezoelectric potential gated charge transport process in fabricating new electronic and optoelectronic devices. Details can be found at: www.nanoscience.gatech.edu.

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