Cellulose for Sustainable Triboelectric Nanogenerators

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Triboelectric nanogenerators (TENGs) are an emerging technology capable of converting the ambient mechanical energy into electricity, promising for complete utilization of the distributed energy to improve the quality of human life. However, realizing material innovation for good trade-off between the high triboelectric output and environment friendliness remains huge challenge, but it is of great importance for sustainable development of TENGs. Cellulose is a typical natural polymer that promises to address the challenge. Herein, the sustainable TENGs that originate from cellulose and potentially back to the natural world are focused on, realizing on-spot TENGs. First, the sources, forms, morphologies, structures, and modifications of cellulose are systematically summarized, indicating the commercial and noncommercial cellulose materials’ potential for TENGs’ application. Second, the dominating additive properties of cellulose such as hydrophobicity, self-cleaning, corrosion resistance, optical transparency, electrical conductivity, and degradability are introduced, which can effectively steer the development of TENGs. Thereafter, the TENGs with tailorable functions enabled by less-refined cellulose and various physical/chemical modified cellulose are reviewed, as well as plants-supported on-spot TENGs with environment adaptivity and transient property, embracing a sustainable future of versatile cellulose to adapt the distributed network of multiscenario TENGs for energy management and information communication.

1. Introduction

The rapid development of modern society accompanied with increasing demand of energy, especially the omnipresent electronics, requires a mass of electricity as power. Nowadays, rechargeable bulky batteries as the dominating power source are applied for triggering most electronics, however, because of the limited lifetime, ultimately, the batteries would be invalid and be treated as electronic waste (e-waste), leading to serious environmental contamination. In terms of sustainable development of our society, the issue should be solved step by step. Therefore, new strategies are sought for weakening the dependence of the traditional power sources, even pursuing the ultimate replacements. Triboelectric nanogenerator (TENG) is an emerging technology capable of converting the ambient mechanical energy into electricity by coupling electrostatic induction and triboelectrification. A typical TENG comprises two electrodes and at least one insulating material as the triboelectric active layer. The device can be easily triggered by external mechanical force to generate electrons transfer between two triboelectric layers, producing electricity output in the external circuit. The simple operation enables TENGs to work without limitations of time and space; in theory, any scenes that have mechanical stimuli can trigger the TENGs to generate electricity by triboelectric effect. Therefore, TENG has been recognized as a promising means for harvesting the distributed energy, which can be utilized on spot or on demand to power the electronics as well as realize self-powered sensing.

Another attractive advantage of the TENG is the simple configuration; any two distinct and even the same materials can serve as triboelectric active materials, which also largely determine the electricity output due to their electronegativity difference; the higher electronegativity can easily build a larger electric potential difference with the coupled material, generating higher electricity output. To obtain high power density, currently, most TENGs are fabricated using commercial polymers films as triboelectric active layers, such as polydimethylsiloxane (PDMS), polyimide, and various fluorine-contained polymers including polytetrafluoroethylene (PTFE), fluorinated ethylene propylene (FEP), perfluoroalkoxylalkane (PFA), polyvinylidine fluoride (PVDF), etc. Although those polymers possess...
high electron affinity and can easily produce superior electricity, their chemical stability delivers poor degradability for the devices. Predictably, if a large number of TENGs penetrate into every corner of our life in the future, the concomitant e-waste would be big trouble for us. Thus, the sustainability of TENGs is of great importance for this kind of new energy strategy. Tremendous efforts have been made to exploit various natural materials as triboelectric active materials for biocompatible and degradable TENGs, such as cellulose, gelatin, chitosan, alginate, and protein, promising to address the low degradability of TENGs. However, most of the natural materials are located closely to the zero point in the triboelectric series, and it is hard to build high electric potential difference with most of other materials, limiting their extensive application in TENGs.

Cellulose is one of the typical natural polymers, with the most abundant resource in nature; it is of great significance to exploit its application potential in the field of TENG, building a sustainable development route of TENGs that can come from nature and be able to go back to nature. In this review, we systematically summarize the commercial and noncommercial cellulose materials in terms of dimension, morphology, and chemical structure, bridging the gap with their properties and application potential in TENGs. Thereafter, we introduced the recent progress of cellulose-based TENGs; first is the strategies for triboelectric output improvement, including less-refined cellulose, physically modified cellulose, and chemically modified cellulose. Second, functions of cellulose in the TENGs were introduced, sorted as environment adaptive cellulose, degradable/transient cellulose, as well as the origin of cellulose such as wood, cotton, and plants. It aims to build a cycle for cellulose materials with the core application of TENGs, including the direct application of cellulose for low-cost TENGs, modification utilization of cellulose for high-performance TENGs, and pristine cellulose for degradable TENGs; furthermore, it targets in situ TENGs that are directly supported by plants such as leaves and wood, achieving on-spot devices on the origin of cellulose. In brief, taking cellulose as a model, this review provides insights and prospects for constructing sustainable TENGs and hopefully could inspire the community to settle the balance issue between triboelectric performance, stability, degradability, and transients in the near future, achieving the on-site assembly, real-time communication, and wireless transmission of TENGs in sustainable way.

2. Triboelectric Nanogenerators

2.1. Mechanism and Advantages

TENG is an emerging technology with a simple device structure, configured by two electrodes and at least one insulating material as the triboelectric active layer, and insulating spaces are needed for periodically separating two triboelectric materials. TENGs are usually operated in four kinds of modes. The device works based on the mechanism refined from the ancient electrification phenomenon, when two dielectric materials contact each other upon being excited by external mechanical force; electrons transfer from the negative relative material to the positive material, and the bound charges on the surface cause equal opposite charges in the back electrodes due to electrostatic induction. Accordingly, there will be electric potential difference between the two electrodes, causing electron flow in the external circuit, generating electricity as triboelectric output. Round-trip flow of electrons occurs between contiguous two contacts, so called “one cycle,” and continuous electricity can be generated when periodical contact and separation are applied on the TENG.

Because of the simple device structure and unlimited material option of the TENGs, low-cost superiority and application feasibility in almost any scene are inherent. The working mode of periodic contact separation enables TENGs highly suitable for harvesting the distributed energy around us, converting the ubiquitous tiny mechanical vibration/friction, rotary motion, oscillating motion, and expanding/contracting motion into electricity, serving as both power sources and self-powered sensors. Moreover, structure features and operation mode determine that TENGs are flexible and deformable, highly suitable for sensing, wearables, soft electronics, and intelligent interface applications that possess distributed requirements. To meet the large-scale application, TENGs with higher comprehensive performance are desired.

Figure 1. A cyclic schematic of the sustainable TENGs from plants, to cellulose, and to plants. TENGs can be fabricated by cellulose extracted from the plants; cellulose with diversity in tailorable size, morphology, structure, and property can well meet the application requirements of TENGs in processing strategy, output performance, sensitivity, durability, degradability, and transients. Ultimately, cellulose inspired researchers such that on-spot TENGs could be constructed on plants, such as cotton, wood, and leaf, which render TENGs back to the origin of cellulose and promise attractive application potentials in situ energy management and information communication.
2.2. Challenges of TENGs in Terms of Sustainable Development

As admitted in the TENG community, the main challenges in this field remain in the following directions: 1) The fundamental mechanism of contact electrification. 2) Improving the output power density of TENG. 3) Addressing the conflict between the durability and degradability of the TENG. 4) Encapsulation and immobilization of the TENG to adapt to harsh environments. 5) Effective energy storage for the TENG. 6) Sensing accuracy and the big data network of TENGs. These challenges require consistent efforts from all community members. Considering the sustainability in terms of energy and environment, solutions to address the challenges of performance durability, degradability, transients, and environmental adaptivity of TENGs are of pressing need (Figure 2b). Most of the commercial silicone-/fluorine-containing polymers provide high output and good stability for the TENG but do not degrade. Natural polymers deliver fast degradability but have lower power density. Both materials are short of self-adaptivity in the application environment. Therefore, a series of multifunctional materials to satisfy the diverse property requirements of TENGs are exploited, and supporting the sustainable development of TENGs is of great importance.

3. Cellulose

Cellulose is the most abundant natural renewable polymer in the world, with output of $10^{11} - 10^{12}$ tons per year. It is mainly a macromolecular polysaccharide composed of D-glucose with β-1,4 glycosidic bonds. The adjacent glucose rings in the crystal area are inverted, and hydroxyl and hydrogen atoms in the adjacent glucose ring are distributed on both sides of its plane. Cellulose does not exist as a monomer, but through the interaction of van der Waals forces and hydrogen bonds, cellulose molecular chains are linked together to form the basic units of cellulose fiber (CF), which is called nanoscale fibrils. The abundant hydroxyl groups offer interaction between chains and hydrogen bonds, enabling tight connection between the nanoscale fibrils to generate microfibrils. Cellulose is the main component of plant cell walls, which widely exists in various plants, and is almost inexhaustible in nature. The microfibrils with the high alignment of molecule structure and high content of crystal phase render cellulose a water-insoluble substance with high toughness to support the growth of plants. The superior mechanical properties and degradability make cellulose an important material for the sustainable development of the industry. For the last thousands of years, cellulose has been used as plant fiber and wood for clothing, building materials, and energy materials, while cellulose has been used as a chemical material for about 150 years.

Owing to the physical processability and chemical structure with abundant active groups of hydroxyl, cellulose can be processed into different forms with nano-/micromension and modified into various functional materials with different active functions for wide applications in our life, which include commercial cellulose materials, commercial-modified cellulose materials, and noncommercial cellulose. Wide applications of cellulose materials have been demonstrated in constructing "green electronics", due to the enriching properties enabled by size as well as active groups. TENGs highly rely on the insulating active layer, flexible substrate, electrode, and spacers to construct devices with advanced structures and triboelectric performance for the generation of high electricity. Cellulose materials deliver great application potential in TENG development, offering new opportunities to balance the output performance, mechanical durability, and degradability of TENGs, promising sustainable development in the future to tackle the huge challenges of TENGs.
3.1. Commercial Cellulose with Dimension Variety

Cellulose can be extracted from various plants such as wood, cortex, cotton, flax, etc., as well as generated by specific bacteria such as acetobacter, agrobacterium, rhizobium, and sarcina. The product size of cellulose ranges from micro- to nanoscale according to different processing methods, including swelling, oxidation, and hydrolyzation that enable the size change of cellulose materials. Due to the mature processing methods, various cellulose products have been developed for commercialization applications, such as microcrystalline cellulose (MCC), cellulose nanofibrils (CNFs), cellulose nanocrystalline (CNC), bacterial cellulose (BC), etc., which possess diverse properties as a result of the dimension difference.

MCC can be extracted from a wide range of plants (wood: hardwood, cork, and cotton staple) and bacteria (non-wood) and it appears as white, odorless, crystalline powder with dimension of around 20–80 μm (see Figure 3a). It is a kind of hydrolyzed cellulose with leveling off degree of polymerization, a partially depolymerized non-fibrous form of cellulose. Therefore, the degree of polymerization of MCC is lower than that of the starting material, and its crystallinity relies largely on that of the raw material and the processing technology. Besides cultivation and purification of bacteria to obtain MCC, the commonly used processing methods of MCC from wood or plants include the mechanical method, acid hydrolysis and enzymatic hydrolysis, or both, or a set of two or more strategies, aiming to remove lignin, hemicellulose, and other impurities. Acid extraction is the most commonly utilized process for the commercial production of MCC due to its low production cost and high efficiency. Due to nontoxicity and biocompatibility, MCCs can serve as fillers, disintegrants, dispersants, and binders, widely being applied in cosmetics, food, pharmaceuticals, and other fields.

In addition, cellulose has been widely processed into nanocellulose to enrich their functions and applications. There exist three types of nanocelluloses including CNC, CNF, and BC that have been commercialized. They possess different crystallinity and surface chemical properties because of the difference of starting materials and processing methods. CNC was obtained by acid hydrolysis or the thermal control method to remove the amorphous regions of cellulose, and the size and crystallinity of CNC depend on the raw materials. Usually, CNC has a wide needle-like and rod-like appearance with size of about 100 nm (see Figure 3b). CNC is a tasteless product with a lot of advantages in properties, such as low expansibility, good mechanical properties, biocompatibility, biodegradability, and optical transparency, being promising as a sustainable replacement for traditional nanomaterials to deliver good economic benefits.

CNF is a new form of cellulose produced by the mechanical treatment of cellulose (see Figure 3c). It appears as an ultrafine nanofibril with a diameter of only a few nanometers and a length of only a few micrometers. Compared with MCC and CNC, CNFs appears as nanofibrils containing both crystalline and amorphous regions and lower degree of crystallinity, resulting in a more flexible cellulose-derived product. Common applications such as a thin membrane and the object matrix of CNC have been widely used for electronic devices that are flexible, transparent, and...
degradable.\textsuperscript{[51]} Accordingly, TEMPO-oxidation was used to treat the CNF to propose an emerging cellulose product of TEMPO-oxidized cellulose nanofibers (tCNF), which possess much higher optical transparency and flexibility, exhibiting great potential in next-generation optoelectronic devices.\textsuperscript{[52]}

Besides the direct extraction and purification on wood-type materials, BC (see Figure 3d) is another pure cellulose with high crystallinity produced by several kinds of bacteria, such as agrobacterium, rhizobia, and gluconobacter. At the microlevel, BC has a complex three dimension (3D) porous network structure, possessing high crystallinity, polymerization degree, high water content (\(\approx 99\%\)), and good plasticity.\textsuperscript{[53]} By virtue of different cultivation methods, BC can be produced into different shapes such as particles, thin fibers, thin films, and hydrogels.\textsuperscript{[54]} With diversity in appearance and processibility, BC has been widely applied in flexible optical and electrical devices for energy and environmental applications.

### 3.2. Commercial Cellulose with Modified Structures

Cellulose contains abundant hydroxyl groups and polar groups that are active for being derived into different active groups by chemical modification, achieving various derived cellulose materials with varying properties. Based on the targeted active groups of hydroxyl, the commercial chemical modification strategies available on cellulose usually involve esterification and etherification to introduce new groups for hydroxyl replacement. The mature commercial derived-cellulose materials include methyl cellulose (MC), ethyl cellulose (EC), carboxymethyl cellulose (CMC), hydroxypropyl cellulose (HPC), hydroxypropyl methyl cellulose (HPMC), cellulose acetate (CA), etc. The commonly used products include CMC, MC, EC and CA, of which, CMC, MC and EC are typical etherified cellulose.

MC is a nonionic cellulose ether, modified by introducing methyl into cellulose. It is a water-soluble polymer and can serve as a thickener for adhesives (Figure 3e).\textsuperscript{[55]} EC is a long-chain polymer of \(\beta\)-glucose units linked by acetal and appears as white or pale brown powder at room temperature (Figure 3f).\textsuperscript{[56]} It is commonly used in the preparation of coating materials, tablet adhesives, microcapsules and microspheres, matrix-controlled release tablets, and flexible transparent films.\textsuperscript{[57,58]} CMC is produced through carboxymethylation on the cellulose; it is an anionic water-soluble biopolymer with hydrophilicity, biological adhesion, and pH sensitivity (Figure 3g).\textsuperscript{[59]} It is one of the most stable modified cellulose materials, showing resistance to water, dilute alkali, and concentrated alkali. CA (Figure 3h) is a cellulose-derived polymer compound obtained by esterifying the hydroxyl groups with acetic acid. CA usually serves as plasticizers for injection production and makes cigarette filters significantly reduce the nicotine content.

### 3.3. Noncommercial-Modified Cellulose

Besides the commercialized modified cellulose with mature processing methods, abundant hydroxyl groups of cellulose allow more modifications to graft various active groups to obtain a variety of derived cellulose materials, such as esterified cellulose, etherified cellulose, sulfonated cellulose, nitrocellulose (NC), etc., which mostly concentrate on the new materials’ development for further industrialization and commercialization.

Cellulose acetate butyrate (CAB, Figure 4a) is a typical mixed cellulose ester. It is an important cellulose derivative with higher melting point, better transparency, and solubility, being suitable for fabrication of a plastic sheet/film with high transparency and good weather resistance.\textsuperscript{[60]} Sulfonated cellulose (SC, Figure 4b) is also a kind of ester cellulose, through the introduction of glycoside modification in cellulose –C–S– to form strong adsorption of cellulose xanthate ester.\textsuperscript{[61]} NC (Figure 4c), obtained by electrophilic attack by nitrate ion to eliminate hydrogen proton, is known as cellulose nitrate ester, appearing as white or pale yellow cotton. According to various nitrogen contents, NC can be applied to fabricate films, clothes, paint, and explosives for extensive application in the industry.\textsuperscript{[62]} Cyanethyl cellulose (CEC) is a typical cellulose ether synthesized from cotton first

Figure 4. SEM and chemical structure of the typical noncommercial-modified cellulose. a) CAB, Reproduced with permission.\textsuperscript{[224]} Copyright 2017, Elsevier. b) SC, Reproduced with permission.\textsuperscript{[61]} Copyright 2021, Springer Nature. c) NC, Reproduced with permission.\textsuperscript{[225]} Copyright 2019, Elsevier. d) CEC, Reproduced with permission.\textsuperscript{[226]} Copyright 2003, Springer Nature.
(Figure 4d). It possesses a variety of merits such as antibacterial property, acid resistance, regulable waterproofness, and dielectric constant and has been widely used in functional textiles, luminescent devices, and displays.

Meanwhile, some emerging cellulose-modified materials were proposed by Xiong et al. They focus on the simultaneous graft of diverse active groups on cellulose, enriching the material function for molecule/ion trapping and releasing. For instance, Xiong et al. developed amphiprotic cellulose (APC) with anionic (2-acrylamide-2-methyl propane sulfonic acid, AMPS) and cationic groups (3-chloro-2-hydroxypropyl trimethyl ammonium chloride, CTA) on the main chain; they demonstrated that the modification route to graft CTA first followed by grafting AMPS can well introduce both groups successfully; the resulting APC shows superiority in removing harmful contaminants with charge diversity from industrial wastewater, such as various dyes and metal ions.[63,64] Using a similar strategy, a poly(amidoxime-hydroxamic acid) cellulose derivative (pAHA-cellulose) was developed by Jiao et al. via free radical polymerization followed by an oximation process; pAHA-cellulose delivers excellent capability for removal of different metal ions (Cu$^{2+}$, Zn$^{2+}$, Pb$^{2+}$, Cr$^{3+}$) from aqueous solutions.[65] These medication strategies to graft diversified groups on cellulose provide inspiration to enrich the triboelectric properties of cellulose-derived materials. Due to the complicated modification process of the abovementioned cellulose, their commercialization for large-scale application is still not high enough.

Diversity of cellulose materials in morphology and structure provides important inspirations for researchers, especially the multifunction requirement of flexible equipment and electrical devices, enriching the efforts to explore various strategies to realize different functional cellulose and cellulose derivatives and being promising to meet the application requirement of emerging flexible devices and optoelectronic devices, which are toward a sustainable future to reduce the e-waste and improve human life.[66–68]

4. Emerging Cellulose with Customized Morphologies and Properties

4.1. Emerging Cellulose Materials with Customized Morphologies

The active groups of cellulose allow the generation of abundant cellulose derivatives, which can be designed into a variety of cellulose-based materials with different sizes and morphologies, including materials with shapes from 0D) to 3D. They are created as emerging cellulose materials to meet and adapt different application requirements, also exhibiting great potential of cellulose in the sustainable development of functional materials.[69–71]

Spherical cellulose with 0D morphology has been widely developed by different methods.[72–79] For instance, Hu et al. observed that certain strains of the bacterium *Glucanacetobacter xylinus* can produce sphere-like cellulose particles (SCP) under orbital shaking cultivation (Figure 5a). The diameter, amount, and formation process of SCP were clearly revealed by controlling the initial glucose concentration and culture temperature.[74] They demonstrated rapid attachment and extension of human osteoblasts grown on SCP, verifying good biocompatibility and application potential of SCP for biomedicine.

Cellulose nanocrystals with one dimension (1D) morphology show advantages in high mechanical performance for composite reinforcement. Xiong et al. demonstrated a combination of 1D cellulose nanocrystals (Figure 5b) and two dimension (2D) graphene oxide (GO) sheets, which delivers synergetic properties’ improvement in high mechanical strength and excellent toughness, promising extensive applications such as ballistic protection, electromagnetic interference shielding, and wearable electronics.[70]

1D cellulose material was also developed to integrate new functional components to enrich the applications.[76–80] For instance, Shang et al. demonstrated superhydrophobic (water contact angle of 161°) and superoleophobic (oil contact angle of 3°) CA nanofiber membranes for robust oil–water separation properties; CA nanofibers were modified by a fluorinated polybenzoxazine (F-PBZ) functional layer doped with silica (Figure 5c).[77] The membrane presents efficient separation for oil–water mixtures and excellent stability to adapt a wide range of pH conditions, delivering good candidates for oil-polluted water treatments and oil spill cleanup.

Furthermore, cellulose membranes (2D) are widely used to replace traditional rigid components for flexible electronics development.[81–85] Xie et al. applied a BC membrane as a template to synthesize a hybrid electrolyte membrane with a hybrid Li$_3$La$_2$Zr$_2$O$_12$ (LLZO)-interconnected network and poly(ethylene oxide) (PEO) (Figure 5d).[82] The porous microstructure and water wettability enable BC to adsorb the precursors to produce porous LLZO nanofiber membranes after calcination. It inspires an inexpensive but efficient strategy for hybrid electrolyte membrane fabrication for electrochemical energy storage systems.[83]

In addition to 0D–2D morphology, cellulose materials were widely developed into 3D aerogel and hydrogel, enriching its applications in the fields such as oil–water separation, thermal management, and electromagnetic shielding.[86–91] Song et al. reported a top-down approach to fabricate an anisotropic wood aerogel (Figure 5e) directly from natural wood by a simple chemical treatment. The layer-structured wood aerogel with anisotropic structural properties shows good mechanical compressibility and fragility resistance, demonstrated by a high reversible compression of 60% and stress retention of ≈90% after 10 000 compression cycles.[87] By incorporating with carbon nanotubes (CNTs), the wood aerogel exhibits an anisotropic thermal conductivity with an anisotropy factor of ≈4.3. In comparison, cellulose-based hydrogel with capability to contain more objects presents good potential for e-skin and bioelectronic applications.[92–96] Ding et al. developed a CNF–polypyrrole (PPy) hybrid hydrogel with polyvinyl alcohol (PVA)–borax (PB) matrix.[93] The CNF–PPy/PB hybrid hydrogel shows low density (≈1.2 g cm$^{-3}$) with high water content (≈94%), demonstrating attractive properties such as excellent biocompatibility, plasticity, pH sensitivity, and rapid self-healing ability without additional external stimuli (Figure 5f). Therefore, the morphology of cellulose-based materials can be customized to meet various application requirements.
4.2. Emerging Cellulose Materials with Tailorable Properties

The morphology diversity of cellulose materials exhibits the versatile processabilities of cellulose; it is conducive in incorporating and endowing a variety of properties on cellulose materials, such as hydrophobicity, self-cleaning, corrosion resistance, optical transparency, electrical conductivity, and degradability. The derived multifunctional cellulose materials are of great importance to extend the application of cellulose in different fields.

The intrinsic hydrophilicity of cellulose largely limits its applications in oil absorption, electronic devices, and intelligent textiles. Tremendous efforts have been applied to develop various hydrophobic cellulose materials to allow their application in other fields. Typically, Xiong et al. developed hydrophobic cellulose oleyl ester nanoparticles (HCOENPs) for decorating the daily textiles, enabling functional textiles with superhydrophobicity, self-cleaning, and anti-fouling properties, which were further demonstrated as wearable all-fabric-based TENGs for both water electrostatic and mechanical energy harvesting (Figure 6a).[89] Cellulose-derived nanocoating can well maintain the intrinsic breathability of textiles, achieving washable and environment-adaptive textile TENGs capable of screening the negative effect of moisture or water stimuli.

Self-cleaning capability can also be realized by cellulose-derived materials with functional coating or component mixing. Bedford et al. developed a king of core-shell CA fibers with TiO₂ shell by coaxial electrospinning (Figure 6b),[102] which shows superior capability to degrade the dye stains than that of the fibers only decorated with TiO₂ on the surface. In addition, corrosion resistance can be realized using some of the cellulose with ultrahigh crystallinity, such as CNC, which is obtained by sulfuric acid hydrolysis in most cases; thus, it delivers certain acid corrosion resistance. Better performance are realized on a few more cellulose derivatives.[106–110] Typically, Luo et al. demonstrated a conductive CNT–CNF film as current collector for electrochemical application (Figure 6c).[107] Compared with commercial
carbon-based current collectors, the CNT–CNF film exhibits superior long-term electrochemical stability and corrosion resistance in a 5.0 mol L\(^{-1}\) sulfuric acid electrolyte, suggesting the advantages of CNFs in corrosion resistance applications.

In addition to the traditional functions, some of the cellulose materials exhibit merits for flexible electronics application. Optical transparency is one of the important properties for next-generation optoelectronic devices.\(^{111}–115\) Xiong et al. developed an EC-based foldable transparent conductor embedded with an innovative silver nanowire-bundle micromesh (AgBM), allowing less amount of AgNWs to achieve explicit conductive paths and high transparency to balance the optical–electrical performance (Figure 6d).\(^{112}\) They presented a representative EC conductor with low sheet resistance of 25 \(\Omega\), ultrahigh transmittance of 97%, and low haze of 2.6%, which demonstrates extreme deformability (internal bending radius of 5 \(\mu m\)) and waterproofing properties enriched by protection with a hydrophobic cellulose-derivative coating, opening up new possibilities for low-cost and scalable transparent conductors to replace indium tin oxide (ITO) for future flexible electronics.

To accommodate the application requirements of electronic devices, efforts are also dedicated to realize conductivity in cellulose materials.\(^{116}–121\) For instance, Liu et al. reported a nanostructured-reduced GO (rGO)/cellulose fiber (CF) composite paper by combining “dipping and drying” with a hydrothermal process (Figure 6e).\(^{117}\) rGO sheets were uniformly coated on the CF network and assembled into a paper with microscale porous conductive networks, which show high conductivity for broad applications in flexible electronics, including energy-storage electrodes, antistatic packages, electromagnetic shielding, and sensors. Furthermore, Wang et al., by controlling the continuous reaction process and isolating oxygen, directly extracted intrinsically conductive CNF from biomass, where the confined ranges of molecular chains of CNFs were converted to highly graphitized carbon at 90°C and atmospheric pressure, while large-scale twisted graphene films were synthesized bottom up from CNFr-graphene (CNFene) suspensions.\(^{118}\) The conductivity of the best CNFene can be as high as 1.099 S cm\(^{-1}\), and the generality of this synthetic route has been verified from multiple biomass cellulose sources. These findings break through the conventional notion that nanocellulose cannot conduct electricity by itself and are expected to extend the application potential of pure nanocellulose to electronic devices, energy storage, catalysis, and sensing.
For electrical or nonelectrical applications, another important merit of cellulose is its biodegradability, promising to replace the majority of traditional nondegradable materials. For example, Xia et al. reported a facile in situ lignin regeneration strategy to synthesize a high-performance biodegradable plastic from lignocellulosic resources. In this process, the porous matrix of natural wood is deconstructed to form a homogeneous cellulose–lignin slurry that features nanoscale entanglement and hydrogen bonding between the regenerated lignin and cellulose micro-/nanofibrils. The resulting lignocellulosic bioplastic shows high mechanical strength, excellent water stability, ultraviolet light resistance, and improved thermal stability. Furthermore, bioplastic can be easily recycled or safely biodegraded in the natural environment. This in situ lignin regeneration strategy involving only green and recyclable chemicals provides a promising route to produce strong, biodegradable, and sustainable lignocellulosic bioplastic as a promising alternative to petrochemical plastics. Furthermore, Wang et al. designed all-natural biodegradable straws (Figure 6f) by hybridizing CNFs and microfibers in a binder-free manner. Straw-like CNFs were fabricated by rolling up the wet hybrid film and sealed with internal hydrogen bonding formed among the CFs after drying. The fabrication strategy with low-cost raw materials and binder-free hybrid design can potentially be a suitable solution to solve the environmental challenges brought by the enormous usage of plastics straws.

In summary, diversity in structure, dimension, morphology, and properties endows cellulose with versatile processabilities, capable of producing a variety of cellulose derivatives that have diverse performances and functions. It demonstrates the great application potential of cellulose from scientific research to industry as well as commercialization. Especially, the fast-growing requirement of electronic devices around human beings, accompanied with the generation of a lot of e-waste, has been a huge concern in modern society. Cellulose exhibits excellent potential in this field, promising to build a sustainable future for transient and recyclable electronics.

5. Cellulose for High-Output-Performance TENGs

The development of TENGs aims to harvest the distributed energy from the environment; therefore, it is foreseeable that a mass of TENG devices will be required in the future to construct a distributed network for harvesting the ubiquitous energy around human beings. Accordingly, a lot of e-waste from TENGs would be generated to cause severe environmental concerns. Cellulose exhibits superiorities in friendliness, skin affinity, biocompatibility, as well as degradability and recyclability, exhibiting attractive potential for the sustainable development of TENGs, especially replacing the traditional nondegradable triboelectric active layers that are crucial for TENGs’ electrical output performance. However, pure cellulose shows neutral characteristics compared with other materials in the triboelectric series, as triboelectric active materials. Its low electronegativity limits the electrical output of TENGs to meet the application requirements. Meanwhile, a variety of functions are also required for cellulose materials in serving as other components of TENGs, such as substrate, current collector, spacer, and encapsulation layer.

Fortunately, cellulose offers diversified possibilities for modification in structure, size, and properties. Tremendous efforts have been dedicated on cellulose modification to improving the output performance of TENGs; according to different processing methods of cellulose, it can be divided into three categories: less refined, physical, and chemical treatments to attain cellulose-based TENGs with high output.

5.1. Less-Refined Cellulose-Based TENG

5.1.1. Origin Forms of Cellulose-Based TENGs

Due to the simple working mechanism of TENGs, cellulose is an intrinsic insulating material with slight positive electron affinity that is competent for the triboelectric active layer. Therefore, reasonably selected cellulose in the original form, such as flowers, leaves, and wood, can be used directly to manufacture TENGs. Singh et al. reported a simple TENG, utilizing natural seeds (mustard, basil, flax) and electrospinning PVDF fibers as triboelectric materials (Figure 7a). Comparing the morphology as well as output performance of the three seeds, they found that the TENG properties of different types of seeds generally depended on the cellulose content, dielectric constant, and surface morphological features. Taking these factors into consideration, the flax seed-based TENG rendered an electrical output with an average open-circuit voltage ($V_{oc}$) of 126 V and maximum power density 324 mW m$^{-2}$, under an impact force of 40 N at 25 Hz, operating in the contact-separate mode. Xia et al. creatively used tea leaves (after drinking) and waste aluminum plastic bags to construct TENGs, which offered an economical method of fabricating TENGs with entire waste materials (Figure 7b). Aiming to enlarge the effective contact area, tea leaves were made into tea film and tea powder, being able to obviously increase the TENGs’ output. The output power density of the tea powder-based TENG reached 488.88 μW cm$^{-2}$. Chen et al. designed a rose petal-based TENG, which first proved that the continuous process of contact and separation of water from the surfaces of petals could generate electrical charge. The triboelectrification effect on the petal epidermis is mainly derived from a periodic array of micropapillae with cuticular folds in the nanometer scale and its great hydrophobicity. Working in the single-electrode mode, the natural petal-TENG was able to attain a maximum $V_{oc}$ of 30.6 V and short-circuit current (I$sc$) of 0.78 μA from a fresh rose petal (3 cm × 3 cm).

What is more, Hao et al. compared the output performance of six natural woods, of which New Zealand pine was chosen for TENG fabrication because of its highest $V_{oc}$ and I$sc$.[131] The maximum output power density acquired the value of 158.2 mW m$^{-2}$ at a loaded resistance of 50 MΩ. The harvested energy was saved in the capacitor to render a new assumption for the application of TENGs in smart homes and electronics. In view of unsatisfactory mechanical properties and the output performance of natural wood, Luo et al. proposed a TENG with modified wood-based film, which was prepared in a simple two-step method, involving a boiling process in the aqueous mixture of NaOH and Na$_2$SO$_3$, followed by hot pressing. Through chemical treatment, not just the structure changed from open-lattice cell lumina to crumpled ones with shrunken diameters and irregular shapes, but also the flexibility and wear resistance were improved. As far as the
wide application of wood materials in the sports industry is concerned, taking table tennis as an example, this equipment is suitable for smart table tennis table innovation for table tennis training and auxiliary systems (Figure 7c). It could provide important functions such as sports big data analysis of point distribution statistics and controversial marginal ball judgments. Apart from maintaining the original characteristics of wood, this flexible, durable, and intelligent TENG is of great significance for the sustainable development of sports equipment. Therefore, in the future, with a series of advantages such as being self-powered, maintenance-free, and cost-effective TENGs will have broader application prospects in the sports industry. Therefore, it can provide an important basis for sports big data analysis, such as the collected sports data can be analyzed to enhance sports performance and develop scientific sports plans and competition strategies, etc.\[133\]

5.1.2. Cellulose Paper-Based TENGs

Being part of the most influential inventions of mankind, paper is one of the most widely used cellulose products in our daily life, which is usually made of plant fiber as raw material. Through different process ratios and raw material selections, a variety of products with various thicknesses and surface roughnesses can be manufactured. A great number of approaches have been explored to endow electronic functions on paper as alternatives for designing electronic products;\[134\] paper-based TENG is one of the typical demonstrations. From the perspective of TENG development, cellulose paper as a component has the advantages such as low cost, environmental protection, diversity for modification, and good physical properties;\[135\] Researchers developed paper-based TENGs with better performance using different types of cellulose papers, which served as different components for TENGs as well as optimized the working mode of TENGs.

Zhong et al. creatively reported a paper-based nanogenerator, which branched out a new field of research, not only in paper applications, but in the progress of new sources of energy for a range of applications in sensing and actuating as well.\[136\] In that study, a piece of commercial printing paper was applied as substrate material. By sticking the TENG with a movable object, such as the page of a book, mechanical energy could be converted into electricity when operating turning a book page. Similarly, Mao et al. also selected a piece of commercial printing paper to design a TENG.\[137\] In contrast, paper without modification was used as a triboelectric layer material. The maximum power density was about 53 W m$^{-2}$ at a load resistance of 8 MΩ.

Figure 7. Less-refined cellulose-based TENGs. a) The mustard seeds-based TENGs. Reproduced with permission.\[128\] Copyright 2019, American Chemical Society. b) TENG based on waste tea leaves and tea leaves powder. Reproduced with permission.\[129\] Copyright 2019, Elsevier. c) Modified wood-based TENGs for smart table tennis table. Reproduced under the terms of the CC-BY Creative Commons Attribution 4.0 International License (https://creativecommons.org/licenses/by/4.0/).\[132\] Copyright 2019, The authors, published by Springer Nature. d) Schematic structure of the PSC and the SEM image of the a-CNF/polymer electrode. Reproduced with permission.\[138\] Copyright 2017, Elsevier. e) The structural diagram of ionogel infiltrated paper and the all-paper-based flexible self-powered sensor. Reproduced with permission.\[139\] Copyright 2019, Elsevier. f) The SEM image of PVDF and CNT-coated cotton thread with high magnification and the digital photograph of FBGs woven into fabric. Reproduced with permission.\[141\] Copyright 2014, American Chemical Society. g) Schematic diagram of the difference in water diffusion between the CF-based film and regenerated cellulose film. Reproduced with permission.\[150\] Copyright 2020, Wiley-VCH GmbH.
Moreover, He et al. proposed an ultrathin paper-based self-powered system, which is composed of a paper-based triboelectric/piezoelectric hybrid nanogenerator (PHNG) and a paper-based supercapacitor (PSC) (Figure 7 d). Via the bottom-up method, a common paper was adopted as the substrate to fabricate PHNG. Then, to achieve good and firm adhesion, a diluted H₃PO₄/PVA gel and a-CNFs to make a PSC composite electrode is chosen. Collecting and converting energy and integrating TENG with PENG together can continuously charge the supercapacitor.

In addition, with a flat wood substrate, Gu et al. applied four commercial paper products (weighing paper, printing paper, tissue paper and crafting paper) to pair with NC paper to fabricate TENGs. Among them, the electrical output performance of weighing paper and printing paper was better. To further optimize the electrification ability of weighing paper, the mechanical exfoliation method was used to make the surface microstructure. On this basis, a cellulose-based energy collection floor was developed, exhibiting output performance with a maximum output voltage and current value of 360 V and 250 μA, respectively. Wu et al. constructed a card-type TENG, which was able to be triggered simply by shaking. The multilayer TENG also was manufactured to increase the effective contact area and greatly enhance the output. What is more, not just the materials and methods used in the processing reduce the cost and environmental pollution as much as possible, but it has the strengths of small size, being lightweight, and is easy to carry and use as well. For the first time, Liu et al. showed an all-paper-based flexible self-power sensor with a flexible electrode of a conductive ionogel infiltrated paper (Figure 7e). In the range of 0.45 N–6.5 N of 1 Hz impulsive force, the TENG-based sensor presented good linearity. Therefore, the ionogel-infiltrated paper had great potential to be applied in wearable and disposable electronics.

5.1.3. CF/fabric-Based TENG

In the traditional textile field, cellulose usually exists in the forms of yarn or fabric. In addition, due to the good skin affinity and softness of cotton fibers or fabrics, they are suitable for being processed into wearable textiles. For instance, Zhong et al. presented a metal-free fiber-based generator (FBG), with an average output power density of 0.1 μW cm⁻² (Figure 7f). It consisted of two entangled modified-cotton threads: one was coated by CNT and the other one was coated by both PVDF and CNT. The fiber generator could operate singly or multiple woven fabrics in parallel. Weaving the made yarn into the fabric also could form a smart textile. Chen et al. prepared a self-charging power textile based on TENG and supercapacitor with cotton thread via the traditional weaving craft. By means of reasonably arranging the density of warp and weft during the weaving process, the TENGs based on different working modes were able to be attained. Through integrating it into clothing, the mechanical energy generated during human movement could be converted into electrical energy and stored. Mule et al. introduced an interlaced mesh cotton fabric as a support frame to deposit conductive polymer PPY by the in situ chemical polymerization process to replace the traditional metal electrode. In the single-electrode mode, the device responded significantly when lightly touched by a human hand, and the corresponding $I_{sc}$ and $V_{oc}$ values were 180 V and 5.3 μA, respectively.

In addition, cellulose can be dissolved in some solvents so that cellulose can be regenerated through a phase conversion process by immersing in a water bath to be applied in fabricating TENG. For example, Zhang et al. selected regenerated cellulose and commercial cellophane as triboelectric materials and graphite foils as electrodes to assemble TENGs (Figure 7g). Generally speaking, the combination of water molecules and cellulose macromolecules would cause the triboelectric effect of fibrillated cellulose and paper to be relatively weak. As based on fibers, commonly used microfibrillated cellulose (MFC) or CNF films belong to porous materials, which means that there exist enough spaces between the fibers that allow water to enter and bind to the fibers. However, in this study, the dense film inhibited the diffusion and bonding of water into the film, so it showed high triboelectric effect. It would significantly contribute to the development of regenerated cellulose-based TENGs.

5.2. Physically Modified Cellulose-Based TENGs

According to whether extra materials are added in the modification process of cellulose, physical modification can be classified into two types: the nonadditive modification and the additive one. Nonadditive modifications generally refer to the methods for fabrication of micro-/nanostructures on the surface, aiming to improve effective contact area or roughness. While the additive modification mainly enhances the TENG performance by introducing additional nanomaterials into the contact layer to tune the dielectric performance, increases the surface charge density, and prolongs the decay time of the charge to reduce the charge loss, it enables higher triboelectric output.

5.2.1. Nonadditive Modifications

Nonadditive physical modification can greatly enhance the output performance by means of increasing the specific surface area and effective contact area of the triboelectric active material, including wet ball milling, freeze-drying method, mechanical exfoliation, template method, printing method, etc. Relying on template method, He et al. described a new approach to introduce CNFs into the big pores of the cellulose microfiber (CMF) skeleton to generate a nanostructured CMF/CNF paper; as a template it was used to load silver nanofibers to construct a TENG, which relied on hierarchical nanostructure and involuntary motions to trigger electricity generation. The peak power density reached 7.68 μW cm⁻², at the external load resistance of 20 MΩ (Figure 8a). Furthermore, inspired by the unique network structure of natural leaf venation, Sun et al. proposed a novel hybrid nanogenerator based on high-performance electrodes by the template method. The leaf veins were acquired by soaking in potassium hydroxide solution and rinsing to obtain intact leaf veins so that they could be molded to add AgNWs for preparation of transparent electrodes. The flexible triboelectric–piezoelectric–thermoelastic hybrid generator made by these electrodes shows the maximum $V_{oc}$ of the mechanical part up to 55 V (Figure 8b). Via versatile electrospinning method, Yan et al. utilized carboxymethyl cellulose sodium (CMC-Na) to
manufacture nanoﬁber as triboelectric layer. The ﬁlm had the features of large speciﬁc surface area and high porosity, and the output power of this TENG with an effective area of 3 cm \times 3 cm exhibited 120 mW m^{-2}. In addition, CMC-Na belonging to a thickening agent is usually applied to the food industry, which is nontoxic to the body, has no pollution to the environment, and can be easily dissolved in water (Figure 8c). Similarly, nanofibers can also be attained by other methods. For example, the solution of soy protein and lignin was blown into the nanofibers membrane by An et al., with a fast and convenient solution blowing method. Compared with the nanofiber ﬁlm prepared by the electrospinning method, this fluffy and porous ﬁlm was conducive in rendering higher voltage through triboelectricity.

Zhang et al. developed a cellulose II aerogel by a combination of the dissolution-regeneration method and freeze-drying method and adopted lithium bromide trihydrate as a solvent. Compared with the nanofiber ﬁlm prepared by the electrospinning method, this fluffy and porous ﬁlm was conducive in rendering higher voltage through triboelectricity. Zhang et al. developed a cellulose II aerogel by a combination of the dissolution-regeneration method and freeze-drying method and adopted lithium bromide trihydrate as a solvent. Due to the special 3D nanostructure, which was different from the 2D structure of the ﬁlm, the induced charges were able to not merely exist on the contact surface but also distribute on the surface of the structural network. Qian et al. combined printing method with the freeze-drying method to design an all-printed TENG (AP-TENG) based on a hierarchical micro-/nano-3D structure, which greatly boosted the effective contact area as well as electronic transmission. At an external resistance of 10 MΩ, a maximum power density of 29 mW m^{-2} can be achieved. In addition, the all-printed structure not only played a vital role in enhancing electrical performance, but also was helpful to improve its roughness and durability. Thus, the TENG was undoubtedly suitable for use as a stable and reliable energy source for various electronic equipment. To increase material surface roughness, Wang et al. utilized lithography technology and inductively coupled plasma (ICP) etching, respectively, to process 317 L stainless steel (317 L SS) and EC (Figure 8f). By exploring the optimal etching power density, the optimal process conditions were successfully attained, acquiring the maximum power output of the TENG, with the I_{sc} and the V_{oc} of the TENG reaching 50 μA and 245 V, separately.

5.2.2. Additive Modifications

As far as additive physical modification is concerned, on the one hand, materials with micro- and nanostructures such as nanowires or nanorods can be introduced into changing the physical morphology of the materials to increase the surface roughness. On the other hand, nanoparticles with high dielectric constant can be added to frictional materials as charge trapping sites to transform single materials into functional composites to increase the retention of frictional charge.

Shi et al. added BaTiO_{3} nanoparticles into cellulose solution to create a composite cellulose paper, via the freeze-drying and compressing process. After dielectric modulation, the TENG generated an output voltage of 88 V and current of 8.3 μA, with a peak power of 141 μW.
paper that consisted of tCNF matrix embedded with a semiconductor 2D materials of phosphorene (BP) nanosheets, first evidencing that the BP could enhance the triboelectric output of the hybrid paper (Figure 9a).[170] Zhang et al. doped silver nanoparticles into the polyethyleneimine (PEI)-modified nanocellulose film and studied the effect of the particle coating on the output performance.[172] Due to the introduction of nanoparticles, under a certain pressure, the film would slightly deform to fill the space formed by the nanoparticles, which undoubtedly increases the effective contact area. Making a contrast with the film without doping, the film could indeed generate more surface charges to enhance the triboelectric output.

In addition to the above methods of modifying cellulose, cellulose or its derivatives also can be incorporated into the preparation of TENGs as an additive to improve output performance.[174,175] As a proof, with the gel-casting technique, Chandrasekhar et al. explored the TENG formed by adding different weight percentages of MCC powder to the PDMS solution for preparing composite films (Figure 9b).[176] The peak output voltage and current for the pure PDMS-based TENG were ≈9 V and 0.8 μA, respectively, while the 5 wt% cellulose/PDMS-based TENG exhibited electrical outputs of about 28 V and 2.8 μA, which successfully proved that the presence of MCC indeed increased the charge density during contact and separation. Medeiros et al. showed a low-cost and facile method of the sequential spray deposition of alkylated organosilanes, EC, conductive nickel nanoparticles, and PVDF to develop a flexible electronic device based on TENG (Figure 9c).[177] After treatment, the device delivers an advantage in human—machine interfaces, such as a flexible keypads and wireless music players. Collecting energy from the contact with user interaction, the power density could be up to 300 μW cm⁻².

5.3. Chemically Modified Cellulose TENG

Chemical modification is one of the most commonly used methods in cellulose modification. Generally, the treatment methods mainly include esterification, etherification, crosslinking, and surface polymerization.[178] Chemical modification directly regulates the charge donation or attraction capacity of the material surface. Different techniques can introduce the ability to gain or lose electrons, resulting in the enhancement of tribological electrical properties of the material. Furthermore, chemical bonding greatly strengthens the friction durability of the material.[179] Next, the chemically modified cellulose-based TENG is systematically introduced, which could well inspire the innovative modification of cellulose.

5.3.1. Derived Cellulose-Based TENG

As summarized earlier, cellulose derivatives possess diversity in structures and properties that depend on different modification

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**Figure 9.** Additive physically and chemically modified cellulose-based TENGs. a) tCNF/BP hybrid paper for TENG performance enhancement. Reproduced with permission.[170] Copyright 2017, Wiley-VCH GmbH. b) Cellulose/PDMS-based TENG templated by sand paper. Reproduced with permission.[176] Copyright 2017, The Royal Society of Chemistry. c) EC-based TENG touch paper by spray-coating technique. Reproduced with permission.[177] Copyright 2020, Elsevier. d) EC/graphene foam-TENG embedded with PS microspheres. Reproduced with permission.[180] Copyright 2018, Elsevier. e) TENG based on electrosprun CA nanofibers. Reproduced with permission.[182] Copyright 2018, Elsevier. f) TENG based on porous NC membrane. Reproduced with permission.[134] Copyright 2019, Elsevier. g) TENG based on BC/ZnO hydrogel treated with aminosilane. Reproduced with permission.[185] Copyright 2020, American Chemical Society. h) Nanocellulose fiber grafted with allicin for TENG. Reproduced with permission.[186] Copyright 2020, Elsevier. i) PEI-crosslinked CA for TENG. Reproduced with permission.[187] Copyright 2020, Elsevier.
and processing methods. Taking advantage of their characteristics, a series of cellulose derivative-based TENGs have been systematically studied and applied. Below, we will separately describe some typical applications of cellulose ethers and cellulose esters in TENGs.

Zhao et al. manufactured a TENG-based self-adaptive motion sensor, which is composed of the composite consisting of graphene foam (GF) coated with EC and polystyrene microspheres (PSMs) dispersed in the hole of the foam (Figure 9d)[180]. Upon contacting, electrons were injected from EC into PSMs to generate surface triboelectric charge. When separating, the negative charge on the EC in the hole induced a negative charge on the upper electrode, and the negative charge on the PSMs was lower. Positive charges were induced on the electrodes, so the electrons in the lower electrode moved to the upper electrode through the external circuit, reached the electrostatic balance, and generated a positive current signal. Šutka et al. prepared a highly porous EC as the triboelectric layer material for TENG devices by means of a simple immersion precipitation approach.[181] When the load resistance was 10^8 Ω, the paired contact between EC and tin-doped In2O3 and PDMS and poly-methyl methacrylate, respectively, produced peak voltages of ≈5, 4.5, and 6 V. In addition, these devices offered stable and continuous outputs, even with 1000 cycles without degradation.

Li et al. utilized CA for fabricating a multilayered fiber-based TENG in contact-separation mode, by the electrospinning method (Figure 9e).[182] CA had the advantages of great biodegradability, chemical stability, ease of processing, and much more importantly, excellent positive triboelectricity. The contact angle of CA was 135°, which was beneficial to reducing the dissipation of triboelectric charges and improving the ability to adapt to the influence of humidity in practical applications.[183] Chen et al. introduced nitro groups into the cellulose film through a chemical reaction and changed the triboelectric polarity of the cellulose from positivity to negativity and then obtained TENG with NC membrane (Figure 9f).[185] The usage of NC to replace commonly used polymers is undoubtedly conducive to environmental friendliness. The NC-based TENG showed the highest voltage and \( I_{sc} \) values of 103.2 V and 19.4 \( \mu \)A, respectively, which were higher than the PTFE-based TENG, attributed to the highly porous structure of the NC film.

5.3.2. Targeted Functional Cellulose-Based TENG

As the natural cellulose is rich in hydroxyl groups and belongs to hydrophilic materials, the ability to gain or lose electrons of cellulose can be efficaciously enhanced, by grafting functional groups with strong friction polarity on the surface of cellulose, such as carboxylation, esterification, sulfonation, silanization, and other chemical modification methods.[184] The existence of these functional groups renders cellulose to acquire more properties other than its own, such as hydrophobicity, friction polarity, and so on. Jakmuangpak et al. retained the 3D nanostructure of BC and incorporated ZnO nanoparticles into the BC film and then treated it with aminosilane functionalization to synthesize a composite nanofilm (Figure 9g).[187] The modified film improved the hydrophilicity and the adhesion between it and the substrate. The TENG exhibited the \( V_{oc} \) and \( I_{sc} \) of 5 V and 7 \( \mu \)A, which are similar to the performance of typical polymer pairs-based TENGs. By means of an novel strategy, Roy et al. used fresh garlic juice with a great amount of the natural sulfur-rich compound “allicin” to graft CNFs (Figure 9h).[186] As sulfur has a higher nucleophilic property than oxygen atoms, through the thiolation of allicin, the peak output current value rose from 0.8 to 5.13 \( \mu \)A, which is about 6.5 times higher than pure cellulose-based TENG (1.23 V, 0.80 \( \mu \)A). Bai et al. adopted the method of regulating the ratio between CA and water-soluble PEI to form abundant termite nest-like porous structure composite films as the triboelectric materials (Figure 9i).[187] By exploring the influence of CA/PEI mass ratio and thickness on electrical properties, the CP performance was gradually optimized. At the CA/PEI mass ratio of 3:1, the TENG rendered the best performance with the peak \( V_{oc} \) of 454 V.

In addition, although natural cotton fibers are not conductive, a simple pyrolysis process can convert them into highly conductive carbon ones and still maintain the great integrity and flexibility.[188] Making a comparison with conventional carbon precursor materials, it is of low cost and environmental friendliness.[189] As a proof, Long et al. carbonized the mixture of glucose and dicyandiamide into multilayered nanosheets, subsequently dispersed them in CNF suspension, and then converted into carbon aerogels, including directional freeze-casting, freeze-drying, and carbonization treatment.[190] Because of the large specific surface area and conductivity, the carbon aerogel was used to fabricate a TENG as triboelectric layer. \( I_{sc} \) and \( V_{oc} \) of the device were able to reach 3 \( \mu \)A and 38 V.

As summarized above, a variety of strategies have been explored to decorate and modify cellulose, enriching the function of cellulose to serve as different components of TENGs, such as triboelectric active material, substrate, and electrode. The efforts demonstrated great application potential of cellulose in triboelectric performance improvement, which is one of the most important properties for electrical devices. The available examples well evidenced the feasibility, more importantly, inspiring the community to consider the prospect of cellulose and explore more innovative methods to address the challenge of output performance for cellulose-based TENG.

6. Cellulose for Environment Adaptive TENGs

Some changes in the working environment, such as air pressure, temperature, humidity, and corrosion, often affect the triboelectric output performance of TENGs.[191,192] In consideration of these aspects, scientists have gradually dedicated to improving the adaptive capabilities of TENGs in various harsh environments. It not only maintains their stable electrical output, but is also committed to taking advantage of its environmental response characteristics to achieve breakthroughs in the advancement and application of TENGs. Cellulose and its derivatives exhibit versatile functions in this field.

Wind is one of the most common forms of mechanical energy. In terms of converting wind energy, researchers have made different attempts, for instance, Luo et al. designed a self-powered mobile disinfection and infection control system based on wind-driven TENGs, to reduce the risk of disease transmission.[193] Ma et al. developed a degradable single-electrode mode TENG with...
the use of wheat straw strips coupled with commercial polymer films. The maximum output power density of WS-TENG is 404 mW m$^{-2}$, which can light up over 60 light-emitting diodes (LEDs). By designing into different shapes, the TENG can act as windmill and bionic lawn to detect the wind speed and wind direction (Figure 10a).\textsuperscript{194} It is a typical example of cellulose-based TENG to utilize wind energy for self-powered sensing.

Nie et al. fabricated a silica/elastomer-coated cellulose paper with further treatment by triethoxy-1H,1H,2H,2H-tridecafluoro-n-octylsilane (PFOTES), as the external layer, which was coupled with the PTFE film to construct a dual-mode TENG for energy harvesting from water drop (Figure 10b).\textsuperscript{195} A similar device based on PFOTES-modified CNF paper was developed to resist humidity, whose $I_{sc}$ achieved 9.3 $\mu$A, about twice of the pure CNF-based TENG.\textsuperscript{50} At 70% relative humidity, output performance still maintained about 70% of the maximum values and good stability. Undoubtedly, the hydrophobic treatment could effectively improve the screening capability for moisture, enabling a reliable TENG even in high humidity conditions.

A more severe environment such as a cold condition below subzero temperature is also a possible scenario that the TENGs would encounter, which would severely reduce its performance. An antifreezing hydrogel was synthesized by Bao et al. via the one-step radical polymerization of acrylamide monomer in hydroxyethyl cellulose (HEC) aqueous solution; with the assistance of lithium chloride (LiCl), the hydrogel can sustain $-69 \, ^\circ C$ without freezing (Figure 10c).\textsuperscript{196} The HEC component not only serves as the physical crosslinking point to improve mechanical properties but also improves water retention of the hydrogel through hydrogen bonds. Accordingly, the freezing point of the hydrogel could be adjusted by regulating the amount of LiCl. A single-electrode TENG based on the hydrogel was demonstrated for collecting biomechanical energy at temperatures below 0, with 626 mW m$^{-2}$ instantaneous peak power density at 2.5 Hz.

In addition to adapting the surrounding environment of TENG, the higher target is to adapt different contact objects, providing self-powered identification and sensing capabilities. Human skin and water are the most common contacted objects by TENGs. However, as is known to all, the common decorative material of fluorine not just pollutes rivers and endangers aquatic life; it can even adversely threaten human life.\textsuperscript{197–200} To avoid these issues, Xiong et al. synthesized a cellulose-based hydrophobic coating of HCOENP to construct a skin-triggered breathable sandwiched all-textile TENG (Figure 10d).\textsuperscript{201} With the coating of BP and HCOENP on PET fabric in sequence to obtain the HBP fabric to serve as the triboelectric active layer of TENG, the device can be triggered by human skin contact to produce relatively high output performance ($\approx 250$–$880 \, V, \approx 0.48$–$1.1 \, \mu A \, cm^{-2}$). More interesting is that the fabric not merely maintained good output performance after various extreme conditions, such as deformation and washing, but also collected a stable electrical
output when applied to different body parts. Moreover, this textile TENG can sensitively perceive the voluntary and involuntary contacts with the human body, delivering excellent potential for practical applications. This research supplies a facile route for the development of novel and multifunctional wearable electronics.

In addition, water condition is also a common scenario for TENG applications. Wang et al. creatively proposed a TENG for collecting blue energy—water energy in 2013. In that work, the concept of water—solid contact electrification was used for energy harvesting for the first time. Considering comfortable wearable application, Xiong et al. reported for the first time a wearable all-fabric-based TENG decorated by cellulose-derived HCOENP coating for water energy collection (Figure 10e). A series of TENGs were designed based on cotton, linen, silk, PET, and other fabrics by dip coating, all of which has been proven to be able to efficaciously harvest water energy. Typically, triggering by tap water under a load resistance of 100 MΩ, the PET fabric-based TENG can generate an output power density of 0.30 W m⁻². This strategy is universal for various types of fabrics but can retain the original advantages of the fabric as well, including flexibility, breathability, comfort, etc., contributing to the important advancement of breathable wearable self-power systems for potential outdoor applications.

Water energy harvesting is a single function for traditional TENGs. To monitor more information in the water resource, such as irrigation water, waste water, and seawater, Xiong et al. further developed a TENG with shape memory hierarchical architectures, which was creatively triggered by hot water to recover to their original morphology; it not only prolongs the lifetime of device, but also builds a linear relationship between the time of device, but also builds a linear relationship between the water-soluble ingredients, so that it could easily dissolve in water and acted as an electrode by adding AgNWs. Apart from being light weight as well as having outstanding foldability, the device can light up seven commercial LEDs at the contact-separation mode. AgNW/CNF paper could be dispersed in deionized water by sonication in 55.75 °C for 30 min (Figure 11a). Besides the external stimuli for accelerated degradation, dissolvable TENG is also popular in the community. Khandelwal et al. manufactured a biocompatible and biodegradable TENG that was made of a laver, an edible silver leaf, and an edible rice sheet. Operating in contacting separating with a fluorinated ethylene propylene, the device exhibits the maximum outputs of 23 V and 315 nA. The device components were soaked in phosphate-buffered saline (PBS) solution to simulate the degradation in gastric juice, and the obvious degradation process of the materials can be clearly observed (Figure 11b). Jo et al. deposited various concentrations of CNTs or silver nanowires on sodium lauryl glutamate (SLG) paper to form electrodes. The resultant SLG conductive paper was utilized as both substrate and dielectric layer (Figure 11c). Interestingly, in addition to the advantages of paper, it also contained both oil-soluble and water-soluble ingredients, so that it could easily dissolve in water and oil. In addition, soil is one of the most extensive matrices for object degradation, which provides a comprehensive decomposition through the erosion of water/moisture and bacteria. Gao et al. constructed an arch-structured transparent nanogenerator assembled by the pure BC film and BC–CNT–PPy composite film. They proved that cellulolytic enzyme can be completely degraded within 8 h, and the remaining CNT can be recycled for potential regeneration (Figure 11e). Moreover, transient TENGs with controllable degradability for bioelectronic application are of great importance. Jiang et al. reported a totally bioabsorbable natural material-based TENGs for energy harvesting and interactive applications, which were assembled by the pure BC film and BC–CNT–PPy composite film. They proved that cellulolytic enzyme can be completely degraded within 8 h, and the remaining CNT can be recycled for potential regeneration (Figure 11e). The operating time of TENGs can be adjusted from a few days to several weeks, so after completing its function, it can be completely degraded and absorbed in the body, successfully avoiding secondary operations and other side effects. Being a transient electronic product, this product with good output performance and biocompatibility renders a novel method for implantable medical products and has great development prospects.
Therefore, on the one hand, modified cellulose materials can enrich the functions of TENGs; on the other hand, the intrinsic advantages enable cellulose superior in the development of degradable, recyclable, renewable, and transient TENGs. Ecofriendliness and biocompatibility are the seductive directions for future development of electrical devices, aiming to reduce e-waste disposal for better environment protection. The diverse degradation routes of cellulose-based TENGs allow their application as distributed networks and not be limited by application scenarios. Moreover, the degradation scene can transform from in vitro to in vivo, especially the on-spot and on-demand application in vivo that would need controllable degradability of TENGs. Typically, as an ideal application, the device can well disappear after completing its function in vivo, without any side effects. Undoubtedly, this will be an attractive research direction for TENGs, and hopefully cellulose can play an important role.

8. Cellulose-Inspired on-Spot TENGs Based on Plants

Cellulose inspired more research of TENGs returning to its origin, such as devices based on or supported by different parts of plants. Meanwhile, on-spot electrical devices have attracted increasing attention due to their ability to self-operate without need of external intervention. The distributed working scenarios enable TENGs to be highly suitable to construct on-spot device especially integrating them in the environment around, such as ubiquitous plants that are mainly composed of cellulose. It promises to realize plant robotics that can harvest energy from the environment, achieving power sources and self-powered sensors that can detect the information for plant and environment, transmitting and communicating the information via the wireless process. In this section, we will introduce the emerging plant-inspired and plant-supported TENGs, which proposes a concept that TENGs finally can return to the forms of origin of cellulose, embracing nature.

Li et al. developed a self-charging device that was composed of a TENG and microsupercapacitor (Figure 12a).[212] The TENG was constructed by a graphene electrode and a superhydrophobic PDMS layer that was replicated with the bionic hierarchical structure of the lotus leaf. The hybrid device can attach on the leaves to harvest electricity from the vibration of the adjacent leaves and contact separation from the raindrops. They demonstrated a maximum output voltage of 170 V at a frequency of 25 Hz. In practical application, it is hard to achieve high mechanical contact frequency at the plant-based scenario. Moreover, the PDMS-based device with low breathability does not allow long-term on-plant usage. The potential dangers to plant respiration and growth ran counter to the original purposes.

Therefore, Lan and Xiong et al. described a waterproof and breathable fiber-based TENG (WB-TENG) to establish an intelligent interface between plants and the environment for real-time monitoring of plant health and harvesting natural energy (Figure 12b).[213] By means of the electrospinning and electro-spraying method, a composite layer-structured PVDF fiber film with adjustable components and morphology was produced; the hydrophobic functional components of fluorinated CNT could be
combined between the nanofibers, but it also boosted the roughness. In addition to good environmental adaptability of the TENG, the key was that it can self-attach on the plant and not affect the health of the plant; stable output performance was proved when the WB-TENG adhered to the plant for 55 days.

In addition to the attachable TENG, TENG-based artificial leaves were further developed for integrating with the tree. Feng et al. displayed a live flower and artificial leaves-based TENG tree, whose $V_{oc}$ and $I_{sc}$ can acquire maximum values of 0.71 $\mu$A and 6.2 V, respectively (Figure 12c).\textsuperscript{[214]} In that study, different varieties of fresh leaves were applied to fabricate TENGs, with electrical output performance of 15 $\mu$A and 430 V. Subsequently, higher performances were achieved on the TENGs based on the dried leaf (25 $\mu$A and 560 V), leaf powder (26 $\mu$A and 660 V), and poly-L-lysine-modified leaf powder (60 $\mu$A and 1000 V). Constructing the wind-driven TENG tree to simulate the energy harvesting process of real trees has broad prospects for resource utilization and environmental friendliness.

Furthermore, Jie et al. directly choose natural leaves to couple with poly(methyl methacrylate) (PMMA) to assemble a leaf-TENG, utilizing the natural leaves as triboelectric active materials and electrodes (Figure 12d).\textsuperscript{[215]} Operating in single-electrode mode, the peak values of the open-circuit voltage and short-circuit current reach $\approx$230 V and 9.5 $\mu$A, respectively. Through integrating TENGs into the tree-shaped energy harvester, it is beneficial to realize large-area environmental mechanical energy collecting. Furthermore, the electrical energy collected can be stored or directly power smart devices.

Therefore, cellulose not only can serve for TENGs as processed or modified forms after extraction from the plants; its origin of plants (wood, leaves) directly supporting TENGs inspired a more promising direction of on-spot self-powered devices, which highly match with the distributed application forms of TENGs, capable of harvesting energy from the environment (rain, wind) and achieving in situ self-powered sensing for wireless information communicating. It demonstrates a cycle utilization concept of cellulose material for TENG construction, indicating the versatile application of cellulose for TENG community to build a sustainable future.

9. Challenges and Prospects in the Development of Cellulose-Based TENG

The current advances have demonstrated a charming potential of cellulose for versatile applications in TENG. However, the already proven cases of cellulose for TENGs still have not fully revealed their practicability in terms of triboelectric performance enhancement, diverse environment adaptivity, controlled degradability, and multiscenario on-spot application. The main challenge is how to build one-to-one correspondence between property requirement of the TENG components and the matchable performance of cellulose or its derivatives (Figure 13). Typically, 1) the triboelectric active layer usually requires high dielectric performance and high electron affinity, rendering induction of high surface charge density as well as prolonged

Figure 12. Cellulose-inspired on-spot TENGs based on plants. a) Lotus leaf-inspired PDMS-based hybrid device of MTENG and MSC. Reproduced with permission.\textsuperscript{[212]} Copyright 2020, Elsevier. b) A breathable fiber-based TENG self-attached on plant for environmental energy harvesting. Reproduced with permission.\textsuperscript{[213]} Copyright 2021, American Chemical Society. c) TENG designed as artificial leaf integrated with the tree.\textsuperscript{[214]} d) Various leaves coupled with PMMA film for on-spot TENGs. Reproduction with permission.\textsuperscript{[215]} Copyright 2018, Wiley-VCH GmbH.
Figure 13. Challenges and prospects in the sustainable development of cellulose-based TENGs.

decay time of the charge. Accordingly, functional groups with high triboelectric polarity are recommended to be grafted on cellulose. 2) Environment adaptivity of TENG includes properties such as mechanical deformability, durability, waterproofness, self-healing, corrosion resistance, optical transparency, and environment friendliness; cellulose-derived materials could be designed as flexible substrates, smart coatings, and adhesives to approach these targets. 3) Controllable degradability is important for future applications of TENGs in vitro and in vivo. Cellulose-based materials are suggested to be modified with a good balance between their mechanical stability and biodegradability. 4) Multiscenario on-spot applications of TENGs require a combination of the properties aforementioned. Chemical, mechanical, optical, and electrical properties of cellulose-based materials should be comprehensively considered during modification and processing.

The challenges are accurate modification strategies to tailor the properties of cellulose-based materials. To this end, the first difficulty remained on the intrinsic rigid molecule chains of cellulose, which have relatively poor solubility in most media. To cope with the versatile function requirements of the components of TENGs, a promising strategy is integrating multiple processing methods for cellulose, including regulation on the dimension, morphology, shape, structure, and properties. Hopefully, cellulose-derived materials could well serve different roles in TENGs, capable of first ensuring the considerable electrical output and then enriching the environmental adaptivity of the device, such as the on-plant TENGs that could be a new kind of on-spot soft robotics, achieving a distributed intelligent network for energy management, sensing, and communication. In addition, controllable degradability and recyclability are of great importance for the future development of distributed TENGs, which are not only important for disposing the devices to reduce the e-waste, but also attractive for biomedical applications, especially in vivo utilization of TENGs to wirelessly monitor physiological information and human health. As a proof of concept, we believe that the versatile derivatives of cellulose and other natural materials with tailorable properties could well build a sustainable future for TENG development, promising an ecofriendly and sustainable distributed network for energy harvesting, information communication, and Internet of Things.

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Conflict of Interest

The authors declare no conflict of interest.

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cellulose, environment adaptivity, sustainability, transient electronics, triboelectric nanogenerators

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