Received: 28 September 2020 | Revised: 6 November 2020 | Accepted: 6 December 2020

DOI: 10.1002/eom2.12072

REVIEW

EcoMat FUNCTIONAL MATERIALS FOR GREEN ENERGY AND ENVIRONMENT | WILEY

Triboelectric nanogenerator based on degradable materials

Shengyu Chao[1,2] | **Han Ouyang**[3] | **Dongjie Jiang**[1,2] | **Yubo Fan**[3] | **Zhou Li**[1,2,4]

[1]CAS Center for Excellence in Nanoscience, Beijing Key Laboratory of Micro-Nano Energy and Sensor, Beijing Institute of Nanoenergy and Nanosystems, Chinese Academy of Sciences, Beijing, China

[2]School of Nanoscience and Technology, University of Chinese Academy of Sciences, Beijing, China

[3]Beijing Advanced Innovation Centre for Biomedical Engineering, Key Laboratory for Biomechanics and Mechanobiology of Ministry of Education, School of Biological Science and Medical Engineering, Beihang University, Beijing, China

[4]Center on Nanoenergy Research, School of Physical Science and Technology, Guangxi University, Nanning, China

Correspondence
Zhou Li, CAS Center for Excellence in Nanoscience, Beijing Key Laboratory of Micro-Nano Energy and Sensor, Beijing Institute of Nanoenergy and Nanosystems, Chinese Academy of Sciences, Beijing 100083, China.
Email: zli@binn.cas.cn

Funding information
National Youth Talent Support Program; China Postdoctoral Science Foundation, Grant/Award Number: 2019M660410; Fundamental Research Funds for the Central Universities; National Key R&D Project from Minister of Science and Technology, Grant/Award Numbers: 2016YFA0202703, 2016YFC1102202; National Natural Science Foundation of China, Grant/Award Number: 61875015; National Postdoctoral Program for Innovative Talents, Grant/Award Number: BX20190026; the 111 Project, Grant/Award Number: B13003

Abstract

Green and eco-friendly energy technology are crucial to reduce environmental pollution caused by fossil fuels. Triboelectric nanogenerator (TENG), as an emergency green energy technology, which can get the energy from the surrounding environment and organism. The development of TENG based on degradable materials strongly promote the next-generation green energy technologies that will effectively avoid pollution and hazards caused by metal and hardly degradable plastic materials. In this review, we summarize the TENG based on degradable materials and its applications. The typical degradable materials for TENG are animal-based degradable material, plant-based degradable material, and artificial degradable material. We provide perspectives on the challenges and potential solutions associated with the next-generation degradable TENG. Beyond the material issue, we highlight the full biodegradable devices that show the healthcare function in vivo.

KEYWORDS

bioelectronics, degradable materials, energy harvester, implantable, self-powered, triboelectric nanogenerator

1 | INTRODUCTION

It is an emergency issue to power a huge number of distributed electronic devices. The massive battery usage led to serious environmental concerns.[1] Triboelectric nanogenerator (TENG), as a neotype green energy technology, can get the energy from the surrounding environment and organism.[2-6] It was reported by Pro. Wang and co-workers in 2012,[7] and is supposed to power a huge number of distributed devices. The principle of TENG based on conjunction of the triboelectric effect and electrostatic induction converts.[8] A surface electrostatic charge is generated by the contact between two triboelectric layers and produces an electric field to drive electrons through an external circuit.[9]

Shengyu Chao and Han Ouyang contributed equally to this work.

Exploitation and application of new materials are essential aspects of the research of TENGs. The materials with more functions, including stretchable, self-healable, biocompatible, degradable, even bioabsorbable, and so on have been applied in TENG.[10-13] The development of triboelectric nanogenerator based on degradable materials (DB-TENG) strongly promotes next-generation green energy technologies. The DB-TENG will effectively avoid pollution and harm to the human caused by metal and hardly degradable plastic materials. The triboelectric layer material is the core element of the TENG and crucial for its output. There are many degradable eco-friendly materials such as cellulose, chitin, and silk fibroin show excellent properties in terms of biocompatibility, degradability, and triboelectric effect.[14-16]

Here, we divide the degradable materials into three types by the source, including animal-based degradable material, plant-based degradable material, and artificial degradable material. Animal-based degradable materials are definited in degradable materials obtained from animal sources in this review, such as chitosan, silk fibroin, and gelatin. Similarly, plant-based degradable materials are degradable materials obtained from plant sources, such as leaves, wood and cellulose molecules, alginate, rice paper, and so on. In addition, the cellulose can be classified as plant-based materials by the old Three boundary theory.[17] Artificial degradable materials are degradable materials that have undergone industrial processing, Such as PLA (polylactic acid) and PLGA(poly(lactic-co-glycolic acid), PHB/V (polyhydroxylbutyrate valerate), PCL (polycaprolactone) (Figure 1).

In this review, we have summarized the works which exploiting and applying degradable triboelectric material into TENG. We are concerned about low toxicity materials that can degrade under natural conditions and stable materials which can be used in TENG as triboelectric layers. Because the instability, it is not cover the materials that can be completely chemically decomposed under specific conditions, such as active metals which can degrade in acids.

The concept of the degradation of TENG was first proposed by Qiang Zheng et al.[12] In 2016, they reported the use of artificial degradable materials as the triboelectric layer to make TENG, the first to proposed the concept of "degradable & implantable TENG" and implant fully degradable DB-TENG into the human body for health care. However, attempts to apply degradable materials in TENG began 2 years ago. In 2015, Luca Valentini was the first to use plant-based materials alginate and GO composite membranes on TENG.[18] Two years later, Hyun-Jun Kim et al made animal-based material silk fibroin into a thin film to act as a triboelectric layer by electrospinning, which started an attempt of fine processing on DB-TENG[19]. In 2017, Xiao-Sheng Zhang et al began to use plant-based degradable material paper as a triboelectric layer to make TENG.[21-33] In 2018, Wen Jiang et al reported the use of natural degradable materials to make fully degradable TENG, which put forward new requirements for the bioabsorbability of DB-TENG.[16] In October of the same year, Zhe Li et al applied gold nanorods to DB-TENG, completing the photothermal degradation regulation of implantable TENG.[13] Besides, some studies have reviewed DB-TENG from different prospects.[25-28]From the perspective of materials, Aifang Yu et al[27] introduce the biodegradable/bioabsorbable TENG with degradable materials and discuss the performance optimization of it. Kaushik Paridaa et al[25] have discussed some DB-TENGs with stretchability, self-healability, and bio-compatibility and summarize the TENG with these properties. So far, more degradable materials have been used in TENG to complete functions such as energy harvesting, signal sensing,[29] and medical care.[30]

In this review, we have summarized recent reports of TENG using degradable materials as the triboelectric layer and made milestones. After that, we listed the degradable materials used in TENG, and explained the

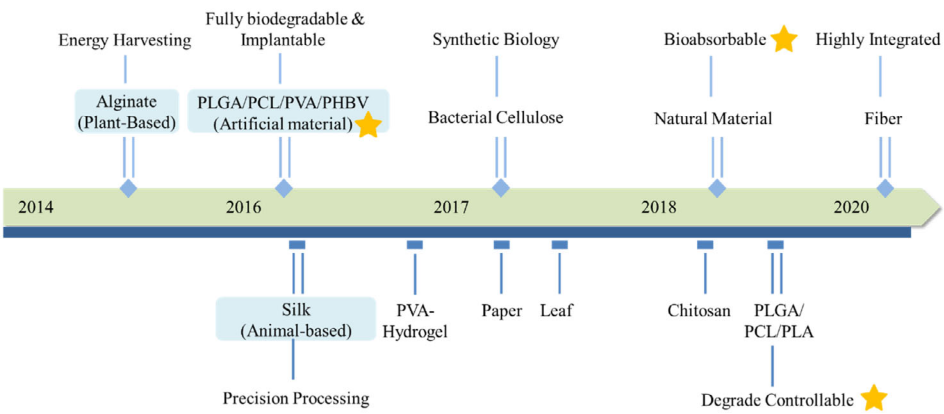

FIGURE 1 Milestones of TENGs which use degradable materials as triboelectric layer. Reference: 2014 to 2016,[18] 2016 to 2017,[12,19,20] 2017 to 2018,[21-23] 2018 to 2019,[13,15,16] 2019 to 2020[24]

characteristics of these materials and their advantages and disadvantages when used in TENG. In addition, in order to introduce the application of DB-TENG, we divide the overall application into three categories and introduce them separately. Finally, we also discussed the challenges, and looked forward to the future research direction of DB-TENG.

2 | TRIBOELECTRIC NANOGENERATOR

The TENG is a kind of energy harvester which can convert mechanical energy to electric energy. In recent years, many significant breakthroughs of theory and application in the area of TENGs, which led it to important energy harvesting techniques. After the contact and separation, two triboelectric layers could carry different kinds charges because of contact electrification effect. And with the movement of materials, the surface electrostatic charge generate a time-varying electric field, which is able to drive the electrons in the external circuit. Considering the effect of different structures, the working model of TENG are further divided into four categories, including vertical contact-separation mode, lateral sliding mode, single electrode mode, and freestanding triboelectric-layer mode.(Figure 2A-D).

2.1 | Vertical contact-separation mode TENG

Vertical contact-separation mode TENG[31] is the most common of the four working modes due to its simple structure design and working method. The structure of the vertical contact-separation mode TENG always includes two triboelectric layers and relevant electrode layers, as shown in Figure 2A. Moreover, some conductive materials can be used as both the triboelectric layer and the electrode layer. Contact electrification is mainly

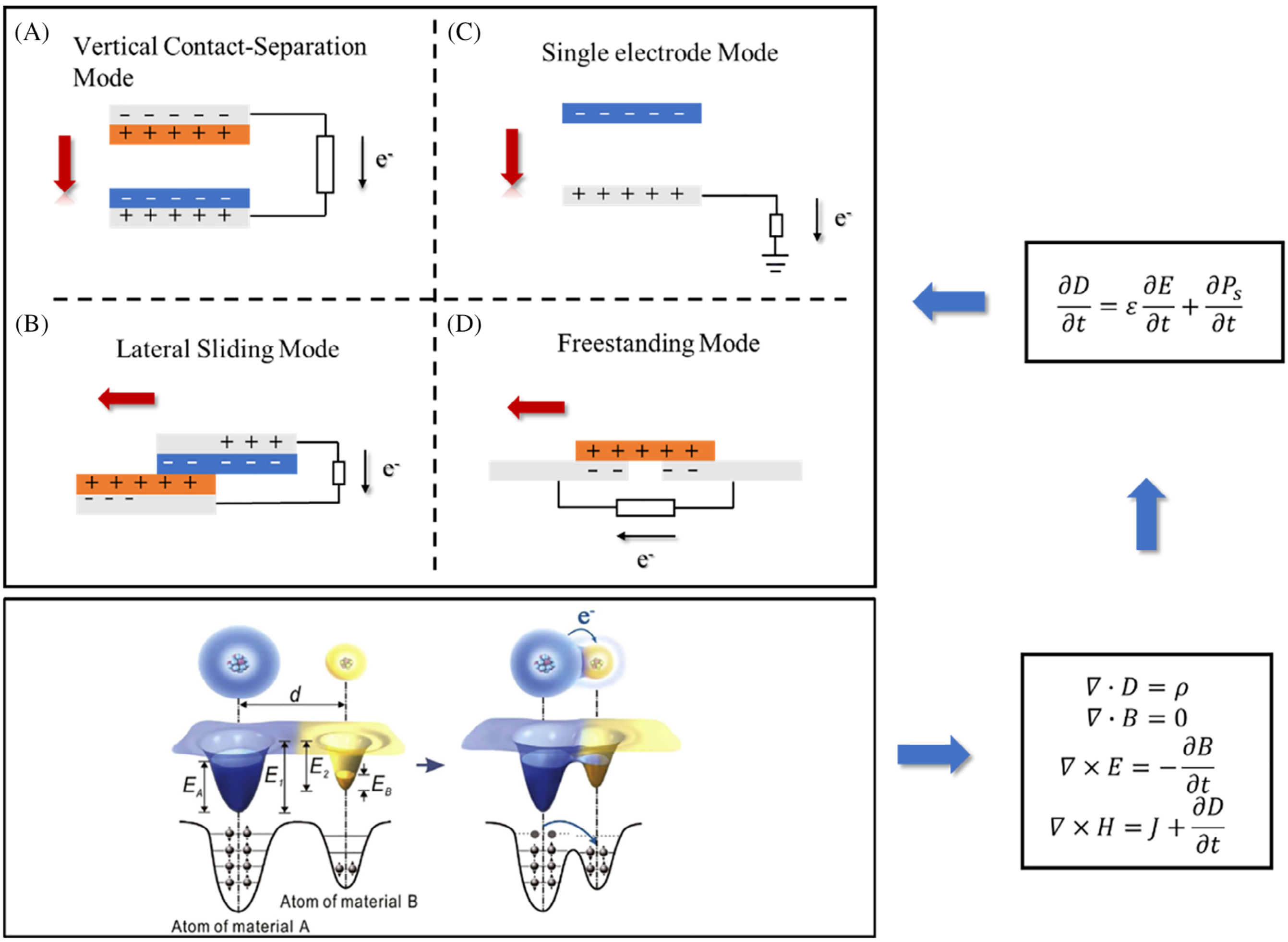

FIGURE 2 (A-D) Schematic diagram of structures and mechanisms of TENG.[31,32,35,36] (e) CE mechanism explained by electron cloud-potential well model at high temperatures[37]

caused by the transfer of surface electrons. Then an electrical potential between two triboelectric layers drove the electrons through external loads due to electrostatic induction.

2.2 | Lateral sliding mode TENG

The working modes of sliding TENG[32] is similar to vertical contact separation, except that the "friction" of the two materials has changed from vertical contact separation to sliding, as shown in Figure 2B. The lateral sliding mode TENG works by friction between two triboelectric layers. Different from the vertical contact-separated mode, the sliding mode produces a relatively stable output, which meets the requirements of continuous power supply for more applications.

2.3 | Single-electrode TENG

The single-electrode mode TENG[33,34] consists of a moving external object and a bottom electrode electrically connected to the reference electrode, which is different from both vertical contact separation working mode and sliding working mode, as shown in Figure 2C. The output of the single electrode mode is only half of the other two modes due to the electrostatic shielding effect.[33] The single-electrode TENG has better adaptability in design since no additional electrode layer is required.

2.4 | Freestanding mode TENG

The freestanding mode TENG is an optimized design for single electrode mode TENG.[35] The triboelectric layer can move and contact with two connected electrodes, which enhance the effect of electrostatic induction. The freestanding TENG also can be divided into a contact separation type and a sliding type according to the way the triboelectric layer contact with each other. Correspondingly, the working principle is similar to the vertical contact separation mode and the lateral sliding mode. Figure 2D shows the freestanding TENG in sliding mode.

3 | MATERIALS FOR TENG

3.1 | TENG based on animal-based degradable materials

The animal-based degradable materials such as silk fibroin,[38,39] gelatin,[40,41] and so on are served as substrates and encapsulation layers and are widely used in transient electronic devices.[42] Recently, the animal-based degradable materials with excellent triboelectric effect have been applied to the triboelectric layer of TENG. Since 2015, The silk fibroin,[19,43] gelatin,[44] and chitin[15,45-47] have begun to emerge in DB-TENG. These TENG using animal-based degradable materials have appeared in broad areas such as energy harvesting, signal sensing, and invasive medical treatment.

There are mainly five animal-based degradable materials, including chitosan/chitin, egg white, silk and derivatives, gelatin, and peptides applied to TENG. Chitin and its derivatives are common in the hard shells of crustaceans (shrimp) and the shells of arthropods (insects). Chitosan can be synthesized from chitin. There are a large number of free hydroxyl and carboxyl groups in the molecular chain, which easily form hydrogen bonds. In terms of electrostatic properties, chitin is a natural polycation polysaccharide in nature. It is easy to lose electrons in the triboelectric effect. Similarly, chitosan also tends to lose electrons as negative charged materials. The chitin and chitosan will also be degraded into small molecules in the environment, and will slowly degrade in the human body, which show good biocompatibility and bioabsorbability.[48] Besides, chitin and chitosan have excellent antibacterial effects and have promising applications in the medical field.[49]

The protein and its derivatives, such as silk, gelatin, egg white, peptides, and so on can serve as a triboelectric layer in TENG. These triboelectric materials usually have a strong ability to transfer electrons due to the carboxyl group. Besides, these proteins can be quickly degraded in environments with microorganisms and proteases, and have a good biocompatibility. Protein is easy to modify and improve by biological and chemical methods to make it more expansible in biomedical applications.

The gelatin is a mixture of protein and peptide which is also one of the most used protein derivative products. Gelatin,[50] is generally derived from the collagen in the connective tissues of animals (such as fish, cattle, etc.) and forms a gel under the incomplete hydrolysis of proteases. In addition, gelatin is expected applied to implantable medical devices due to excellent biocompatibility and biodegradability (Figure 3).

3.2 | TENG based on plant-based degradable materials

In 2013, Z. L. Wang's group began to explore an important plant-based biodegradable material—paper as a component of TENG.[53] But the paper was only used as a spacer, rather than as a triboelectric layer directly applied

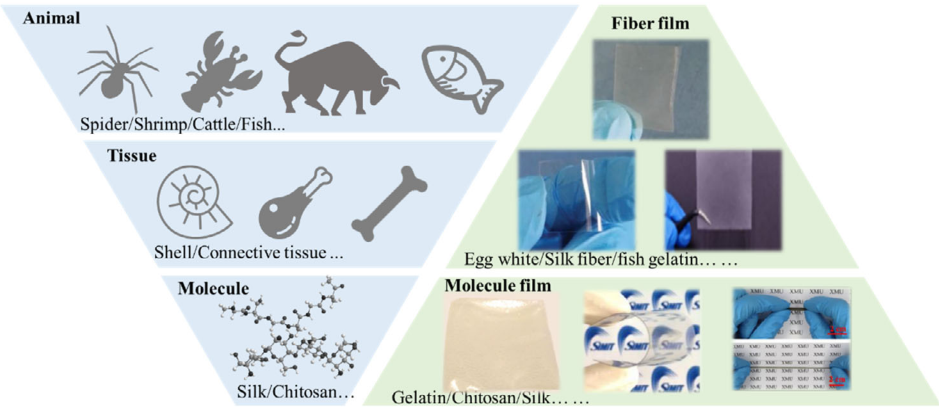

FIGURE 3 Hierarchy chart for animal-based degradable material used in TENG[12,15,16,51,52]

FIGURE 4 Hierarchy chart for plant-based degradable material used in TENG[16,18,60-64]

to TENG. Then, Xiao-Sheng Zhang et al[21]reported TENG using paper and Teflon as the triboelectric layer in 2017; Luca Valentini et al[18] reported a TENG adding alginate to the triboelectric layer. Nowadays, more plant-based biodegradable materials combined with TENG and achieved outstanding performance.

At present, there are currently six plant-based degradable materials used in TENG, including cellulose, paper, leaves, wood, rice paper (starch), and alginate. Alginate is a common food additive used for thickening, emulsification, and stability.[54] It is usually added to other mixed systems (such as salt solution, etc.) to form a film by cross-linking.[55] As a polysaccharide derivative, alginate materials are easily degraded and have good biocompatibility. In addition, ions increase the solubility of the material itself in water, so that it can be degraded in water or aqueous solutions containing ions. This property enables the controlled degradation of alginate-based TENG.[13]Rice paper is another plant-based degradable material widely used in TENG. Its raw material is starch, a common edible polysaccharide produced from crops such as rice and corn.[56]Leaves, paper, wood (wood fiber), pure cellulose, the main components of these four materials are cellulose, which is derived from the branches of plants in nature. The main component is the most common polysaccharide cellulose in nature, these materials can be degraded under the conditions of acid, alkali, and special enzymes, but the degradation rate is related to the specific structure of the material.[57,58] Specially modified molecules can endow cellulose with stronger abilities to transfer electrons and better physical and chemical properties (Figure 4).[59]

3.3 | TENG based on artificial degradable materials

The chemically synthesized artificial materials play an important role, such as PTFE, PET, and PI, in triboelectric materials for TENG.[7,32,35] Recent, some artificial degradable materials have been used as triboelectric materials to meet short-term applications. In 2016, Qiang Zheng et al first proposed the biodegradable TENG based on artificial degradable materials.[12] In 2018, Zhe Li et al applied artificial degradable materials and gold nanorods to TENG and used NIR light to regulate the degradation of TENG in vivo.[13] The artificial degradable materials are showing a promising prospect in applying TENG results in the ease of large-scale manufacturing (Figure 5).

There are currently five kinds of artificial degradable materials used in TENG, include polylactic acid (PLA), poly lactic-co-glycolic acid (PLGA), polyvinyl alcohol (PVA), polycaprolactone (PCL), and poly 3-hydroxybutyrate-co-3-hydroxyvalerate (PHB/V). As an emerging synthetic material, PLA has excellent material properties such as high strength and high transparency, which is known as one of the ideal substitutes for petroleum-based polymer materials.[65,66] In addition, both PLA and PLGA are polymerized by ester bonds, and degradation products are human metabolites. Thus, these materials have bioabsorbable properties and can be well used in implantable applications.[67,68]

The PCL and PVA are degradable materials derived from petroleum, while PVA is the only vinyl material among these five materials. Since vinyl alcohol is not very stable, ethyl acetate is generally used as a monomer for PVA.[69] Polyvinyl alcohol film has good mechanical properties, biodegradability, biocompatibility, but poor thermal stability.[70,71] The PCL generally has a larger overall molecular weight and relatively slow biodegradation compared with among other polymer materials.[72]

PHB/V, as a copolymer of hydroxybutyric acid and valeric acid, is also one of PHB derivative polymers. The synthesis of PHB/V mainly relies on chemical methods.[73] Like several other materials which are mentioned, PHB/V has good biocompatibility and biodegradability and has good mechanical properties after film formation.[74]

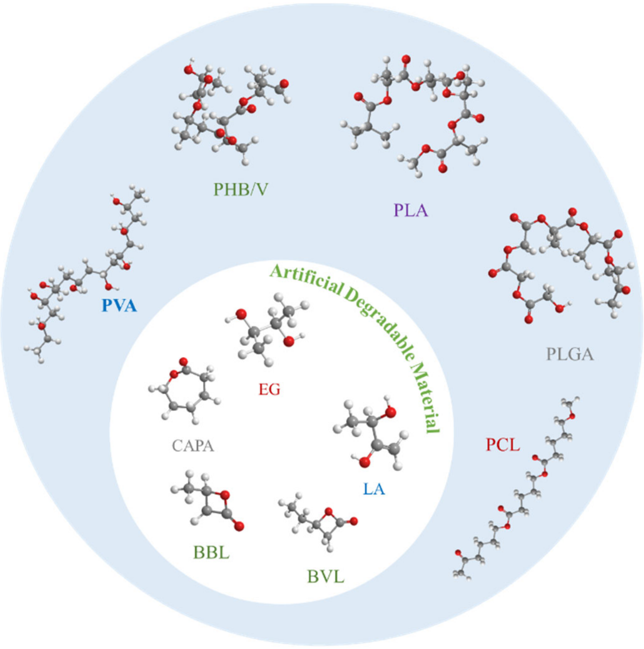

FIGURE 5 Artificial degradable material used in TENG and its monomer; BBL, β-butyrolactone; BVL, β-valerolactone; CAPA, caprolactone; EG, ethylene glycol; LA, lactic acid; PCL, polycaprolactone; PHB/V, poly1hydroxylbutyrate valerate; PLA, polylactic acid; PLGA, poly(lactic-co-glycolic acid); PVA, polyvinyl alcohol

4 | TENG APPLICATIONS BASED ON DEGRADABLE MATERIALS

DB-TENG has shown many excellent properties, such as eco-friendly, biocompatible, biodegradable, and so on. They are widely applied in energy harvesting, signal sensing, and implantable medical devices.

4.1 | DB-TENG for energy harvesting

Harvesting energy from the environment and living organisms have been an important goal of TENG since it was invented. The emergence of DB-TENG has brought dawn to the new eco-friendly and degradable energy supply. Luca Valentini firstly used the degradable material alginate for TENG and proving its output capability. Ruoxing Wang et al reported a TENG made by laser processing technology. The output of TENG with different chitosan doping during degradation has also been tested (Figure 6A).[15] Young Choi et al report a TENG based on silk fibroin with a unique curved structure to harvest energy from human motion. Its output is reaching 28.13 V/2.71 μA (Figure 6B).[75] Wei Yang et al demonstrated a washable and air-permeable TENG based on a paper that reaches an output of 230 V/9.5 μA (Figure 6C).[76] In 2018, Yange Feng reported a hybrid TENG to harvest wind energy in the environment. The leaves are directly used as the triboelectric layer. The maximum current of the TENG can reach 150 μA at 7 m/s wind speed.[78] In addition, Dongwhi Choia et al designed a liquid-solid contacted degradable TENG to harvest raindrop energy in external environments (Figure 6D).[22,61] Sai Sunil Kumar Mallineni fabricated TENG with PLA and achieved a high voltage of up to 2700 V (Figure 6E).[77] In addition, Yaokun Pang fabricated degradable TENG based on alginate film. The maximum output can reach 33 V/150 nA (Figure 6F).[62] In 2019, Kequan Xia proposal an environmentally friendly TENG with wasted tea leaves and packaging boxes. The output of the TENG also reached 792 V/42.8 μA.[79]

4.2 | DB-TENG for sensing

The TENG can be used not only as an energy harvesting device, but also as a sensor which is superior in mechanical sensing due to its high sensitivity and short response time. The eco-friendly, biocompatible, and biodegradable

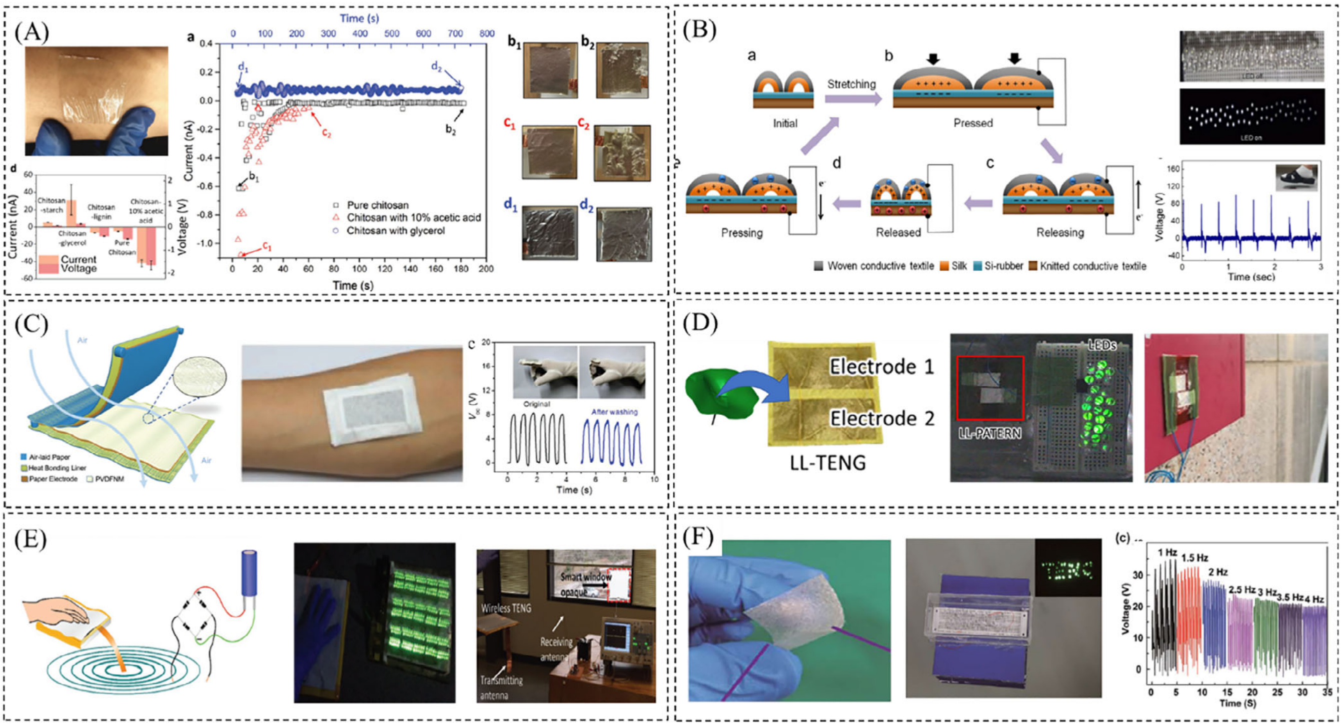

FIGURE 6 DB-TENG for energy harvesting. A, Performance and biodegradation test of chitosan–glycerol based TENG,[15] and, B, Schematic of working principle and applications for corrugated textile TENG.[75] C, Structure and applications of air- permeable TENG.[76] D, Schematic of working principle and applications for leaf-based TENG.[22] E, Schematic of a wireless TENG[77] based on PLA, and its application. F, Performance and applications of the alginate-based TENG.[62] The left one is the optical image of alginate film

materials also endow TENG more distinctive features in transient electronic device applications.

Xiao-Sheng Zhang et al first reported a paper-based TENG with an output of 85 V/3.75 μA. The graphene pencils are applied to make the electrode layer. The paper-based TENG can attract and control the movement of droplets on the platform by using the generated electric potential(-Figure 7A).[21] Jianjun Luo et al demonstrated a flexible and durable wood-based TENGs. This device has been applied in self-powered sensing with athletic big data analytics (Figure 7B).[80] Sheng Chen et al used paper and cellulose paper as the triboelectric layer to fabricated TENG and achieved 196.8 V and 31.5 μA output. This work reported a paper piano with the paper-based TENG's (Figure 7C).[81] In addition, Chaoxing Wu et al reported a thin sheet of TENG with paper and PVC. the TENG could distinguish the types of materials, such as common glass, cotton, wood by the efficiency of transferring electrons. Simultaneously, this work also tried multi-layer folded paper-based TENG to generate electricity. The space between the different thin layers of the TENG of this structure is relatively small, which can transform the sound vibration in the surrounding air to electrical output (Figure 7D).[60]

Flexible TENG is often applied in human health surveillance. Lingyun Wang et al used PDMS and PVA/PEI to make transparent TENG. This device can sense the slight movement of the palm and fingers.[82] Yun-Ting Jao et al reported a system for detecting human movement and sweat. This work adhered chitosan-glycerin film to different parts of the socks and demonstrated the TENG can be integrated with fabrics. The NaCl concentration in the sweat has also been detected (Figure 7E).[45] Wei Xu et al reported a TENG using hydrogel containing PVA, which can directly and effectively detect arm bending from 0° to 150°.[20] Besides, Yinben Guo et al proposed a hybrid TENG with PVDF fiber and silk fibroin to detect human motion. They showed the alarm of an emergency. For example, when the human body falls, the devices on the arm will generate short and high-output signals, thereby prompting the mobile phone to send emergency signals (Figure 7F).[83] Zhu Zhiyuan et al report a single-electrode TENG with rice paper to detect sweat produced on the arm.[84]

To improve the monitoring accuracy, some new materials and structures are applied in DB-TENG. Cuncun Qian et al fabricated multi-layer TENG with doped cellulose nanofibers and PDMS by laser processing technology. The human motion status monitoring has been archived.[86] Jong-Nam Kim et al reported a motion sensor based on DB-TENG with more compatible for skin. The degradable material gelatin mixed with different proportions of silica gel used as the triboelectric layer.[46]Faliang He et al used silk nanofibers and PVA/MXene nanofibers as the triboelectric

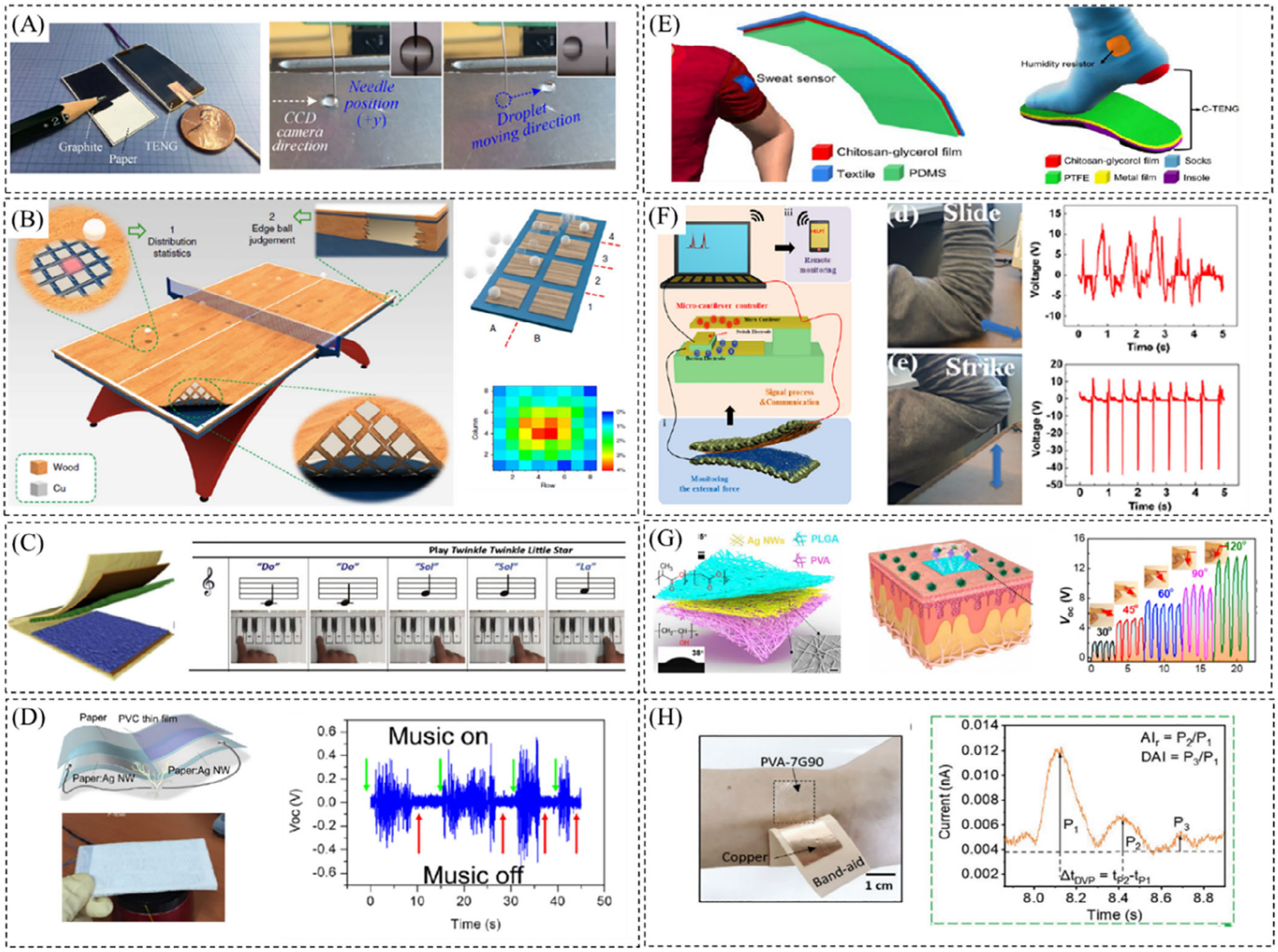

FIGURE 7 DB-TENG for sensing. A, The structure of the paper-based TENG[21] and its application. B, Schematic of a wood-based TENG[80] for sport training. C, The structure of the paper-based TENG[81] and its application of paper piano. D, A TENG[60] based on paper .E, The structure and application of the chitosan-glycerin based TENG.[45] F, Schematic of silk-fibroin based TENG and its application.[83]G, The structure and application of all-fiber TENG.[24]H, A DB-TENG[85] and its performance

layer to fabricate a small and transparent TENG for real-time healthcare detection.[87] In addition, DB-TENG can also effectively detect cardiovascular events. Peng Xiao et al report a band-aid-liked TENG with biodegradable materials PVA and PLGA, which can monitor different movements of the human body or pulse information (Figure 7G).[24] At the same time, Ruoxing Wang et al have produced a DBTENG by different doped PVA films and PI, which can sense the stress of 0.6 N/cm^2 on the skin and accurately monitor the pulse(Figure 7H).[85]

4.3 | DBTENG for implantable medical devices

Implantable medical devices improve patient care and disease therapy due their ability to vital physiological signals monitoring and electrical stimuli. However, most existing implantable medical devices must be removed or replaced via an invasive, complex surgery at the end of service life. Implantable biodegradable electronic device is a new trend to avoid invasive secondary surgery that can be absorbed or degraded in vivo.

In 2016, Qiang Zheng et al first reported the full biodegradable TENG as a life-time designed implantable power source which can convert mechanical energy of organism to kinetic energy.[12] In this work, PLGA, PHB/V, PVA, and PCL four kinds of artificial degradable materials were used as the triboelectric layer. The maximum output of this DB-TENG can reach about 40 V, 1 μA. The device can produce electricity stably for 2 weeks in a rat body. In addition, this work also proved that the electrical stimulation of TENG can effectively stimulate the directional growth of nerve cells. It is expected to be applied in nerve repair in the future. In recent years, there are key findings show that the voltage generated by TENG applied to wound treatment can effectively reduce infection and promote healing. Wen Jiang et al demonstrated a full bioabsorbable TENG with several natural degradable materials, such as animal-based materials chitosan, egg white, silk fibroin, plant-based materials cellulose, and rice paper.[16] The maximum output of 55 V/0.6 μA was reached when egg white and rice paper are used as the triboelectric layer. It can provide a stable electrical output within a few days when implanted in the rat. Besides, they also proposed treatment using methanol to reduce the degradation rate of the device in the body (Figure 8).

To regulating the degradation rate of implantable devices, Zhe Li et al proposed a degradable TENG based on Au nanorods and reached the maximum output of 28 V, 220 nA. They doped Au nanorods in the PCL, PLA, and PLGA film as the triboelectric layer. The NIR(near infrared) light can indirectly control the degradation of the device by the photothermal effect.[13] In the case of added NIR light, the output of the implanted TENG quickly decreases to 0 within 24 hours, while the control group (without NIR light) can remain output on the 28th day. This work verified that the electrical output of TENG can effectively promote cell proliferation. This device is expected to be applied in wound tissue recovery. Yujia Zhang et al proposed a thin-sheet TENG based on silk fibroin obtained by genetic engineering. The size of TENG is 2 cm × 3 cm, and the maximum electrical output can reach 145 V/4.28 μA. They implanted the device into the wound of the mouse for wound treatment and achieved an antibacterial rate of 93% for *Escherichia coli* and 58% for *Staphylococcus aureus*. Correspondingly, the control group for the electric energy not produced by TENG only had an antibacterial rate of about 20%.[93] In addition, DB-TENG can also be used as an energy supply device to power for implantable devices. Compared with conventional implantable electronic medical devices, small-sized TENG as an energy supply can replace batteries that occupy most of the device's volume, thereby reducing the overall volume of the implanted device.

5 | CHALLENGES AND OPPORTUNITIES

With the in-depth research of materials and structure design, degradable TENG has shown great development and promising potential in energy harvesting, signal sensing, and implantable medical treatment in the past few years. However, there are still some challenges required to overcome in the future as an emerging field. Here, from aspects of components and properties, we discussed

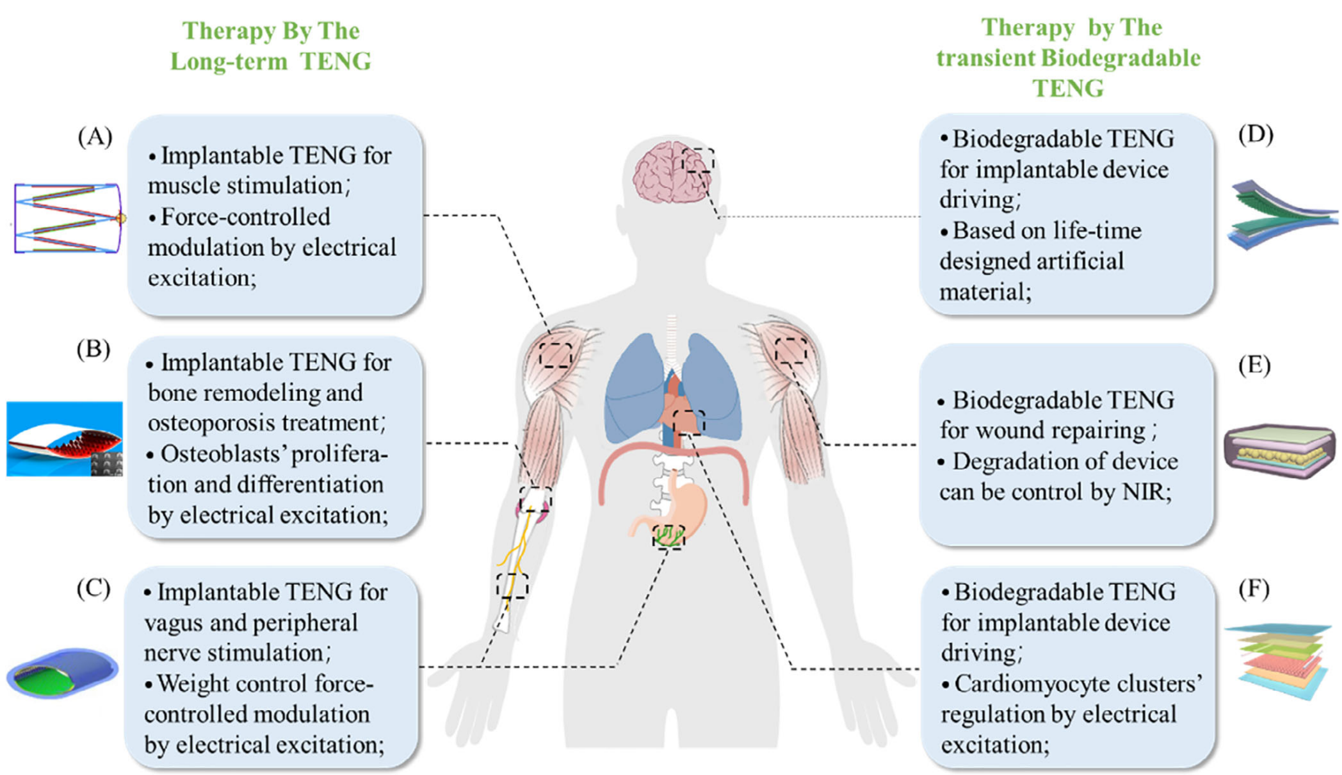

FIGURE 8 Implanted TENG-based devices used in medical applications. A, References from 88;B, References from 89,90; C, References from 91,92; D, References from 12; E, References from 13; F, References from 16

the direction of optimization for DB-TENG and its future prospects.

5.1 | Degradable materials for DB-TENG

Materials are an important part of TENG, which deserves further discussion. Materials' properties, such as surface microstructure, triboelectric, and mechanical properties, will influence the performance and application of DB-TENG.

5.1.1 | Molecular structure of materials

In 2018, Pro. Morten and Pro. Zhong Lin Wang[94] proposed an electron-cloud-potential-well model to explain contact electrification and charge transfer and release between two materials. In this model, the triboelectric effect between materials is the charge transfer caused by the overlap of electron clouds. The functional groups[95] have a great influence on the electron cloud of the material, while electronegative groups are more likely to be negatively charged in contact electrification.

For degradable materials, the animal-based materials mainly contain proteins and their derivatives, and have more electronegative groups than the plant-based materials, which mainly contain polysaccharides. According to the previous work, we list a triboelectric series table of these degradable materials.[12,13,16,96,97] In addition, modification methods such as nitration and amination can also change the surface potential of degradable materials, thus changing the charged state in contact electrification. Different materials and modification methods can improve the performance of DB-TENG effectively.

Besides, molecular structure of materials also plays an important role in biocompatibility and biodegradability, which could affect the in vivo or in vitro applications of DB-TENG. Specific molecular sites, such as the peptide bonds, are easily recognized and degraded by organisms. In contrast, some molecular structures can disrupt the normal life of organisms. In that sense, the modification of materials will have an impact on biocompatibility and biodegradability.

5.1.2 | Surface microstructure of materials

In addition, surface microstructure is also an important factor affecting the output of DB-TENG. Various process of material preparation and modification have been demonstrated to affect the surface microstructure. Water lithography, electrostatic spinning, and other materials preparation methods can effectively form a specific surface microstructure. Besides, ICP etching, plasma cleaning, and other methods can also form microstructure on the surface of materials and improve the performance of TENG.

5.1.3 | Mechanical properties of materials

The mechanical properties of materials are also the focus of researchers, while it is crucial to the devices based on DB-TENG, especially human integrated devices.[98] Some properties, such as young's module, will directly affect users' comfort level. Besides, some mechanical properties such as elasticity and tensile properties also play an important role in some special applications of DB-TENG. In recent years, there has been a great deal of work on mechanical modification of materials, but it is rarely used in DB-TENG. In the future, more and more material modification methods, such as doping and chemical modification, will be used in DB-TENG to improve the performance of devices (Figure 9).

5.2 | Output performance for DB-TENG

The output performance of the DB-TENG still needs to be improved. In Table 1, we summarize the TENG report in 5 years, which use degradable material as the

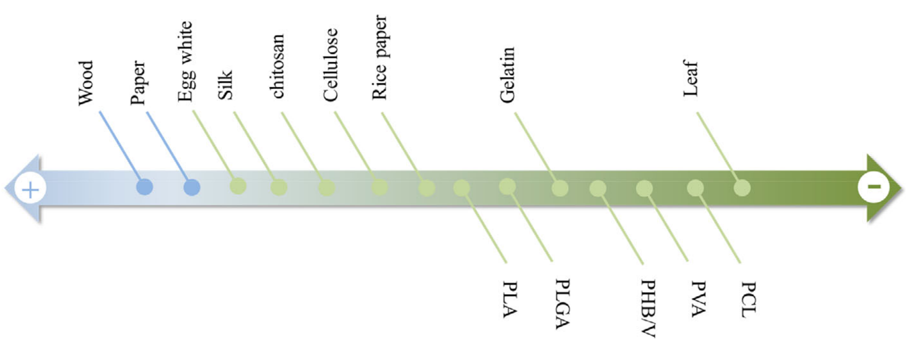

FIGURE 9 Triboelectric series[12,13,16,96,97] of degradable materials based on their ability to lose or gain electrons. The reference material is PDMS

TABLE 1 Performance comparison of DB-TENGs with different triboelectric materials

Degradable material type	Material	The other electrode	Degrade condition	Biodegradable/ Biocompatible	Electrical output	Size (area)	Application	References
Animal-based	Chitin/chitosan	PTFE		Biocompatible	130 V/15 μA	5 cm × 3 cm	Sweat sensor	[45]
Animal-based	Chitin/chitosan	PI	Water	Biocompatible	1.35 V/42 nA	2 cm × 1 cm	Energy harvesting	[15]
Animal-based	Chitin/chitosan	PI		Biocompatible	16.2 V/125 μA	~5 cm × 5 cm	Speed senor	[47]
Animal-based	Chitin/chitosan	FEP		Biocompatible	150 V/1.02 μA	3 cm × 4 cm	Behavior sensor	[46]
Animal-based	Egg white	Rice paper	In vivo & methanol	Both	55 V/0.6 μA	1 cm × 2 cm	Drive implantable device	[16]
Animal-based	Silk/silk fibroin	Rice paper & cellulose	In vivo & methanol	Both	~40 V/0.4 μA & ~25 V/0.3 μA	1 cm × 2 cm	Drive implantable device	[16]
Animal-based	Silk/silk fibroin	PI		Both	~15 V/~2.5 μA	~50 cm^2	Drive electronics	[19]
Animal-based	Silk/silk fibroin	Regenerative silk		Both	41.6 V/0.5 μA	1 cm × 2 cm	Body sensor	[99]
Animal-based	Silk/silk fibroin	PET		Both	268 V/5.78 μA	2 cm × 4 cm	Active sensor	[43]
Animal-based	Silk/silk fibroin	PET		Both	145 V/4.28 μA	2 cm × 3 cm	Antibacterial patch	[93]
Animal-based	Silk/silk fibroin	PET		Both	213.9 V/0.34 μA	3 cm × 3.6 cm	Energy harvesting	[52]
Animal-based	Silk/silk fibroin	PDMS		Both	12 V/0.6 μA		Behavior sensor	[100]
Animal-based	Silk/silk fibroin	PVDF		Both	500 V/12 μA	2 cm × 4 cm	Gesture sensor	[83]
Animal-based	Silk/silk fibroin	PVA/MXene		Both	117.4 V	1.5 cm diameter circle	Behavior sensor	[87]
Animal-based	Silk/silk fibroin	Si-rubber		Both	28.13 V/2.71 μA	5 cm × 3 cm	Wearable Energy Harvesting	[75]
Animal-based	Gelatin	PLA		Both	500 V/16 μA	4 cm × 4 cm	Energy harvesting	[101]
Animal-based	Gelatin	PTFE/PDMS		Both	130 V/0.35 μA	5 cm × 5 cm	Wearable Energy Harvesting	[51]
Animal-based	Polypeptide	PTFE		Both	~350 V/10 μA	2.15 cm × 5.16 cm	Energy harvesting	[102]
Animal-based	Polypeptide	PTFE		Both	65 V	10 cm × 10 cm	Energy harvesting	[44]
Plant-based	Cellulose	Egg white & silk fiber	In vivo & methanol	Both	45 V/0.4 μA & 32 V/0.3 μA	1 cm × 2 cm	Drive implantable device	[16]
Plant-based	Cellulose	PTFE		Both	7.925 V/1.095 μA	3 cm × 3 cm	Energy harvesting	[103]
Plant-based	Cellulose	Cu		Both	13 V/3.2 μA	5 cm × 5 cm	Energy harvesting	[23]
Plant-based	Cellulose	FEP & Cu		Both	8 V/9 μA & 0.8 V/0.8 μA	1 cm × 1 cm	Energy harvesting	[104]
Plant-based	Cellulose	FEP		Both	~30 V/90 μA	40 cm^2	Drive electronics	[63]
Plant-based	Cellulose	FEP		Both	21.9 V/0.17 μA	3 cm × 3 cm	Antibacterial patch	[64]

(Continues)

TABLE 1 (Continued)

Degradable material type	Material	The other electrode	Degrade condition	Biodegradable/ Biocompatible	Electrical output	Size (area)	Application	References
Plant-based	Cellulose	Al		Both	28 V/3.8 μA	3 cm × 3 cm	Wearable Energy Harvesting	[105]
Plant-based	Cellulose	Al		Both	320 V/11.25 μA	1.5 cm × 1.5 cm	Energy harvesting	[106]
Plant-based	Cellulose	Steel		Both	240 V/50 μA	800 cm^2	Drive electronics	[107]
Plant-based	Cellulose	FEP		Both	286.5 V	18 cm^2	Drive electronics	[108]
Plant-based	Cellulose	PDMS		Both	55.8 V/0.94 μA	3.2 cm × 3.2 cm	Human behavior sensor	[86]
Plant-based	Cellulose	PDMS		Both	45 V	2.5 cm × 2.5 cm	Drive electronics	[109]
Plant-based	Cellulose	PDMS		Both	181 V/21 μA		Human behavior sensor	[110]
Plant-based	Cellulose	Ni		Both	18 V/2.4 μA	1.5 cm × 1 cm	Drive electronics	[111]
Plant-based	Cellulose	phosphorene		Both	5.2 V/1.8 μA	1 cm × 1 cm	Energy harvesting	[112]
Plant-based	Paper	PTFE		Biocompatible	400 V/170 μA	10 cm × 15 cm	Drive electronics	[113]
Plant-based	Paper	PTFE		Biocompatible	85 V/3.75 μA	2 cm × 4 cm	Control droplet	[21]
Plant-based	Paper	PI		Biocompatible	~96 V/11 μA	6 cm × 7 cm	Drive electronics	[114]
Plant-based	Paper	PVC		Biocompatible	100 V	10 cm × 10 cm	Loudspeaker	[60]
Plant-based	Paper	PVDF		Biocompatible	197 V/16.2 μA	4 cm × 4 cm	Drive electronics	[76]
Plant-based	Paper	Nitrocellulose membrane		Biocompatible	196.8 V/31.5 μA	2.5 cm × 2.5 cm	Paper piano	[81]
Plant-based	Paper	Teflon		Biocompatible	~1000 V/42 μA	4 cm × 2 cm (stacked)	Drive electronics	[115]
Plant-based	Paper	PCL/GO		Biocompatible	120 V/4 μA	4 cm × 4 cm	Drive electronics	[116]
Plant-based	Leaf	PVDF		Biocompatible	1000 V/60 μA	4 cm × 4 cm	Environmental energy harvesting	[78]
Plant-based	Leaf	PTFE		Biocompatible	792 V/42.8 μA	5 cm × 5 cm	Behavior sensor	[79]
Plant-based	Leaf	PMMA		Biocompatible	230 V/9.5 μA	8 cm × 8 cm	Drive electronics	[61]
Plant-based	Leaf	Water (droplet)		Biocompatible	~0.3 V/18 nA		Environmental energy harvesting	[22]
Plant-based	Rice paper	Chitosan, egg white, & silk	In vivo & methanol	Both	28 V/0.2 μA & 45 V/0.4 μA & 48 V/0.42 μA	1 cm × 2 cm	Drive implantable device	[16]
Plant-based	Rice paper	Skin (hand)	Water	Both	11.2 V/~1 μA	4.4 cm × 4.4 cm	Human behavior sensor	[84]
Plant-based	Rice paper	PVC		Both	244 V/6 μA	3 cm × 3 cm	Human behavior sensor	[117]

TABLE 1 (Continued)

Degradable material type	Material	The other electrode	Degrade condition	Biodegradable/ Biocompatible	Electrical output	Size (area)	Application	References
Plant-based	Rice paper	Laver	Water	Both	23 V/315 nA	2 cm × 2 cm	Drive electronics	[118]
Plant-based	Wood	PTFE		Biocompatible	~80 V/~1.8 μA	4 cm × 4 cm	sport monitoring and assisting	[80]
Plant-based	Alginate	Al	NaCl (high temperature)	Both	30 V/150 nA	5 cm × 5 cm	Drive electronics	[62]
Plant-based	Alginate	PTFE	high temperature	Both	1.3 V/1 nA	2 mm^2	Energy harvesting	[18]
Artificial material	PLA	Au (nano rods)	PBS, 37°C (NIR)	Both	28 V/220 nA	1.2 cm × 1.2 cm	Tissue repairing	[13]
Artificial material	PLA	PTFE		Both	2.7 kV	~16 cm × 18 cm	Drive electronics	[77]
Artificial material	PLA	PP/PE		Both	202 V		Energy harvesting	[119]
Artificial material	PLGA	Au (nano rods)	PBS, 37°C (NIR)	Both	28 V/220 nA	1.2 cm × 1.2 cm	Tissue repairing	[13]
Artificial material	PLGA	Skin	PBS (PH 7.4), 37°C	Both	90 V/1.5 μA	2 cm × 2 cm	E-skin	[24]
Artificial material	PLGA	PCL, PVA, & PHB/V	PBS (PH 7.4), 37°C	Both	40 V/1 μA, 26 V/0.4 μA, & 15 V/0.3 μA	2 cm × 3 cm	Drive implantable device	[12]
Artificial material	PVA	PLGA, PCL, & PHB/V	PBS (PH 7.4), 37°C	Both	26 V/0.4 μA, 15 V/0.3 μA, & 13 V/0.2 μA	2 cm × 3 cm	Drive implantable device	[12]
Artificial material	PVA	PTFE/nylon		Both	210 V/27 μA	2 cm × 2 cm	Energy harvesting	[120]
Artificial material	PVA	Alginate	Water	Both	1.47 V/3.9 nA	30 cm^2	Energy harvesting	[121]
Artificial material	PVA	Al		Both	200 V/22.5 μA	8 cm × 8 cm	Human behavior sensor	[20]
Artificial material	PVA	PDMS		Both	70 V/12 μA	2 cm × 2 cm	Human behavior sensor	[82]
Artificial material	PVA	PS		Both	30 V/~1 μA	2.5 cm × 2.5 cm	Energy harvesting	[122]
Artificial material	PVA	PI		Both	~1.5 V/5 nA	~1 cm × 1 cm	Wearable cardiovascular monitoring	[85]
Artificial material	PCL	PLGA/PHB/V/PVA	PBS (PH 7.4), 37°C	Both	40 V/1 μA, 28 V/0.6 μA, 15 V/0.3 μA	2 cm × 3 cm	Drive implantable device	[12]
Artificial material	PHB/V	PCL/PVA/PLGA	PBS (PH 7.4), 37°C	Both	28 V/0.6 μA, 13 V/0.2 μA, 15 V/0.3 μA	2 cm × 3 cm	Drive implantable device	[12]

Abbreviations: PTFE, polytetrafluoroethylene; FEP, fluorinated ethylene propylene; PI,polymide; PDMS, polydimethylsiloxane; PET, polyethylene terephthalate; PVA, polyvinyl Acetate; PLA, polylactic acid; PVC, polyvinyl chloride; PP, polypropylene; PE, polyethylene; PCL, polycaprolactone; GO, graphene oxide; PMMA, polymethyl methacrylate; PHB/V, poly1hydroxylbutyrate valerate; PS, polystyrene.

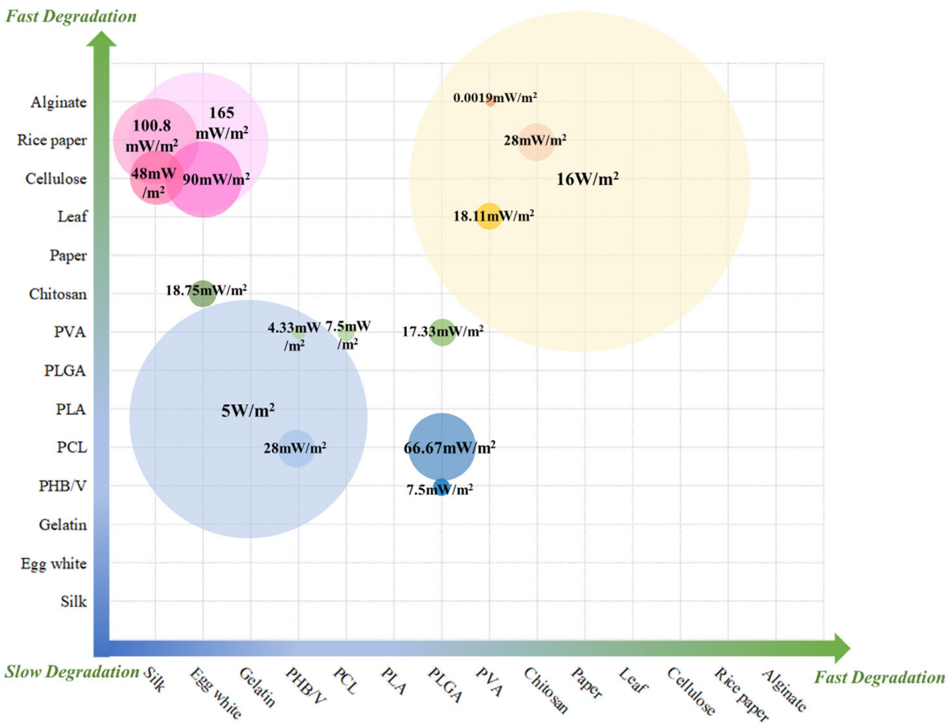

FIGURE 10 The performance comparison of TENGs with two degradable triboelectric layers

triboelectric layer and summarize the properties of these materials. The output of DB-TENG with two degradable triboelectric layers is shown in Figure 10. There are few DB-TENGs with high output, which may impede the clinical application of DB-TENG. It is challenging that drive electronic devices or achieves the efficient electrical stimuli to the DB-TENG.

There are some methods expected to improve the performance of TENG. Except for material modification, other processes, such as high-voltage charge injection can effectively increase the output of TENG. Besides, structural optimization can also greatly improve the output of DB-TENG.

5.3 | Encapsulation for DB-TENG

More efficient encapsulation is required for the implantable or wearable TENG devices based on degradable materials. The rapid corrosion of degradable materials will reduce the lifetime and stability of the device. A reliable encapsulation is essential to TENG for wearable, even implantation. However, the existing encapsulation materials such as, PDMS, silica gel, epoxy resin, and parylene all have low mechanical strength. And the encapsulating method is only to wrap the device in materials. It is significant for future DB-TENG that build flexible and even elastic structures.

5.4 | Controllable degradation for DB-TENG

The foremost challenge is the degradation control for TENG. If the degradation is too fast, the service time cannot meet the requirement of treatment. However, if the degradation is too slow, it may cause security risks. Until now, degradation control for TENG is limited. Methods such as material methanol treatment, NIR control have been used to regulate the degradation of DB-TENG. However, the precise control of material degradation cannot be achieved. In the future, Physical, biological, and chemical stimulation is promising methods to control degradation.

However, challenges and opportunities always complement each other. In the context of environmental pollution and climate warming, it is an important trend that green and eco-friendly biodegradable materials are applied in daily life. In the future, the combination of biodegradable materials and TENG will further promote the development of green and eco-friendly energy technology.

6 | CONCLUSION

In recent years, a large number of TENGs based on degradable materials have emerged and aim to provide green and eco-friendly energy. So far, with the development of biodegradable materials for TENG, a variety of degradable materials have been discovered, such as gelatin, chitosan, silk fibroin protein, cellulose, and alginate. There are many self-powered applications have been achieved, in particular, self-powered therapy and diagnosis, and so on. From intelligent electronics to health monitoring, the DB-TENG will affect our daily lives in the future.

In this article, the development of TENG based on degradable materials was concluded from the aspects of working mechanism, material sources, and application scenarios. According to sources, the degradable materials used in TENG are classified into three categories: Animal-based degradable material, such as silk protein, chitosan, gelatin, egg white and peptide materials; Plant-based degradable material, such as cellulose, leaves, wood, rice paper, alginate; Artificial degradable material, such as PVA, PLA, PLGA, PCL, and PHB/V. The characteristics of these materials and their application advantages in TENG are discussed. In addition, we have classified and discussed these DB-TENG application scenarios. Finally, the potential challenges and prospects of DB-TENG are introduced.

The development of TENG based on degradable materials strongly promote next-generation green energy technologies. It is expected to avoid pollution and hazards caused by metal and hardly degradable plastic materials. There are a dozen of degradable materials developed for TENG since the first application of degradable materials to TENG. DB-TENGs have been applied in energy harvesting, mechanics sensing, even implantable electric stimuli. Although it is full of challenges and requires more research and exploration, the DB-TENG future is promising and achievable.

ACKNOWLEDGMENTS

This study was supported by the National Natural Science Foundation of China (61875015), National Key R&D Project from Minister of Science and Technology, China (2016YFA0202703 and 2016YFC1102202), National Postdoctoral Program for Innovative Talent (BX20190026), China Postdoctoral Science Foundation (2019M660410), the 111 Project (B13003), Fundamental Research Funds for the Central Universities, and the National Youth Talent Support Program.

CONFLICT OF INTEREST

The authors declare no competing interests.

DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available from the corresponding author upon reasonable request.

ORCID

Zhou Li https://orcid.org/0000-0002-9952-7296

REFERENCES

1. Collivignarelli C, Riganti V, Urbini G. Recycling. Battery lead recycling and environmental pollution hazards. *Conservation*. 1986;9(1):111-125.
2. Ouyang H, Li Z. The first technology can compete with piezoelectricity to harvest ultrasound energy for powering medical implants. *Science Bulletin*. 2019;64(21):1565-1566. https://doi.org/10.1016/j.scib.2019.09.010.
3. Hinchet R, Yoon H-J, Ryu H, et al. Transcutaneous ultrasound energy harvesting using capacitive triboelectric technology. *Science*. 2019;365(6452):491-494.
4. Hwang BU, Lee JH, Trung TQ, et al. Transparent stretchable self-powered patchable sensor platform with ultrasensitive recognition of human activities. *ACS Nano*. 2015;9(9):8801-8810. https://doi.org/10.1021/acsnano.5b01835.
5. Jeong CK, Baek KM, Niu S, et al. Topographically-designed triboelectric nanogenerator via block copolymer self-assembly. *Nano Lett*. 2014;14(12):7031-7038. https://doi.org/10.1021/nl503402c.
6. Lee H, Lee HE, Wang HS, et al. Hierarchically surface-textured ultrastable hybrid film for large-scale triboelectric nanogenerators. *Adv Funct Mater*. 2020;30:2005610.
7. Fan F-R, Tian Z-Q, Lin Wang Z. Flexible triboelectric generator. *Nano Energy*. 2012;1(2):328-334. https://doi.org/10.1016/j.nanoen.2012.01.004.
8. Zhang C, Tang W, Han C, Fan F, Wang ZL. Theoretical comparison, equivalent transformation, and conjunction operations of electromagnetic induction generator and triboelectric nanogenerator for harvesting mechanical energy. *Adv Mater*. 2014;26(22):3580-3591. https://doi.org/10.1002/adma.201400207.
9. Niu S, Wang ZL. Theoretical systems of triboelectric nanogenerators. *Nano Energy*. 2015;14:161-192.
10. Jiang D, Shi B, Ouyang H, Fan Y, Wang ZL, Li Z. Emerging implantable energy harvesters and self-powered implantable medical electronics. *ACS Nano*. 2020;14(6):6436-6448. https://doi.org/10.1021/acsnano.9b08268.
11. Lai YC, Deng J, Zhang SL, Niu S, Guo H, Wang ZL. Single-thread-based wearable and highly stretchable triboelectric nanogenerators and their applications in cloth-based self-powered human-interactive and biomedical sensing. *Adv Funct Mater*. 2017;27(1):1604462.
12. Zheng Q, Zou Y, Zhang Y, et al. Biodegradable triboelectric nanogenerator as a life-time designed implantable power source. *Sci Adv*. 2016;2(3):e1501478.
13. Li Z, Feng H, Zheng Q, et al. Photothermally tunable biodegradation of implantable triboelectric nanogenerators for tissue repairing. *Nano Energy*. 2018;54:390-399. https://doi.org/10.1016/j.nanoen.2018.10.020.
14. Yang B, Yao C, Yu Y, Li Z, Wang X. Nature degradable, flexible, and transparent conductive substrates from green and earth-abundant materials. *Sci Rep*. 2017;7(1):4936. https://doi.org/10.1038/s41598-017-04969-y.
15. Wang R, Gao S, Yang Z, et al. Engineered and laser-processed chitosan biopolymers for sustainable and biodegradable triboelectric power generation. *Adv Mater*. 2018;30(11):1706267–1706274. https://doi.org/10.1002/adma.201706267

16. Jiang W, Li H, Liu Z, et al. fully bioabsorbable natural-materials-based triboelectric nanogenerators. *Adv Mater*. 2018;30(32):e1801895. https://doi.org/10.1002/adma.201801895.
17. Katz A. Studies in boundary theory: three essays in adjudication and politics. *Buff L Rev*. 1978;28:383.
18. Valentini L, Rescignano N, Puglia D, Cardinali M, Kenny J. Preparation of alginate/graphene oxide hybrid films and their integration in Triboelectric generators. *European Journal of Inorganic Chemistry*. 2015;2015(7):1192-1197. https://doi.org/10.1002/ejic.201402610.
19. Kim H-J, Kim J-H, Jun K-W, et al. Silk Nanofiber-networked bio-Triboelectric generator: silk bio-TEG. *Advanced Energy Materials*. 2016;6(8):1502329–1502334. https://doi.org/10.1002/aenm.201502329.
20. Xu W, Huang L-B, Wong M-C, Chen L, Bai G, Hao J. Environmentally friendly hydrogel-based triboelectric nanogenerators for versatile energy harvesting and self-powered sensors. *Advanced Energy Materials*. 2017;7(1):1601529–11601536. https://doi.org/10.1002/aenm.201601529.
21. Zhang X-S, Su M, Brugger J, Kim B. Penciling a triboelectric nanogenerator on paper for autonomous power MEMS applications. *Nano Energy*. 2017;33:393-401. https://doi.org/10.1016/j.nanoen.2017.01.053.
22. Choi D, Kim DW, Yoo D, Cha KJ, La M, Kim DS. Spontaneous occurrence of liquid-solid contact electrification in nature: toward a robust triboelectric nanogenerator inspired by the natural lotus leaf. *Nano Energy*. 2017;36:250-259. https://doi.org/10.1016/j.nanoen.2017.04.026.
23. Kim H-J, Yim E-C, Kim J-H, Kim S-J, Park J-Y, Oh I-K. Bacterial nano-cellulose triboelectric nanogenerator. *Nano Energy*. 2017;33:130-137. https://doi.org/10.1016/j.nanoen.2017.01.035.
24. Peng X, Dong K, Ye C, et al. A breathable, biodegradable, antibacterial, and self-powered electronic skin based on all-nanofiber triboelectric nanogenerators. *Science Advances*. 2020;6(26):eaba9624.
25. Parida K, Xiong J, Zhou X, Lee PS. Progress on triboelectric nanogenerator with stretchability, self-healability and biocompatibility. *Nano Energy*. 2019;59:237-257. https://doi.org/10.1016/j.nanoen.2019.01.077.
26. Torres FG, De-la-Torre GE. Polysaccharide-based triboelectric nanogenerators: a review. *Carbohydrate Polymers*. 2021;251:117055–117071. https://doi.org/10.1016/j.carbpol.2020.117055.
27. Yu A, Zhu Y, Wang W, Zhai J. Progress in triboelectric materials: toward high performance and widespread applications. *Advanced Functional Materials*. 2019;29(41):1900098–1900124. https://doi.org/10.1002/adfm.201900098.
28. Zheng Q, Tang Q, Wang ZL, Li Z. Self-powered cardiovascular electronic devices and systems. *Nat Rev Cardiol*. 2020;18:7–21. https://doi.org/10.1038/s41569-020-0426-4.
29. Zhu M, Sun Z, Zhang Z, et al. Haptic-feedback smart glove as a creative human-machine interface (HMI) for virtual/augmented reality applications. *Sci Adv*. 2020;6(19):eaaz8693.
30. Shi B, Liu Z, Zheng Q, et al. Body-integrated self-powered system for wearable and implantable applications. *ACS Nano*. 2019;13(5):6017-6024. https://doi.org/10.1021/acsnano.9b02233.
31. Niu S, Wang S, Lin L, et al. Theoretical study of contact-mode triboelectric nanogenerators as an effective power source. *Energy & Environmental Science*. 2013;6(12):3576–3583. https://doi.org/10.1039/c3ee42571a.
32. Wang S, Lin L, Xie Y, Jing Q, Niu S, Wang ZL. Sliding-triboelectric nanogenerators based on in-plane charge-separation mechanism. *Nano Lett*. 2013;13(5):2226-2233. https://doi.org/10.1021/nl400738p.
33. Niu S, Liu Y, Wang S, et al. Theoretical investigation and structural optimization of single-electrode triboelectric nanogenerators. *Adv Funct Mater*. 2014;24(22):3332-3340. https://doi.org/10.1002/adfm.201303799.
34. Meng B, Tang W, Too Z-H, et al. A transparent single-friction-surface triboelectric generator and self-powered touch sensor. *Energy Environ Sci*. 2013;6(11):3235-3240.
35. Wang S, Xie Y, Niu S, Lin L, Wang ZL. Freestanding triboelectric-layer-based nanogenerators for harvesting energy from a moving object or human motion in contact and non-contact modes. *Adv Mater*. 2014;26(18):2818-2824. https://doi.org/10.1002/adma.201305303.
36. Yang Y, Zhang H, Lin Z-H, et al. Human skin based triboelectric nanogenerators for harvesting biomechanical energy and as self-powered active tactile sensor system. *ACS Nano*. 2013;7(10):9213-9222.
37. Xu C, Wang AC, Zou H, et al. Raising the working temperature of a triboelectric nanogenerator by quenching down electron thermionic emission in contact-electrification. *Adv Mater*. 2018;30(38):e1803968. https://doi.org/10.1002/adma.201803968.
38. Brenckle MA, Cheng H, Hwang S, et al. Modulated degradation of transient electronic devices through multilayer silk fibroin pockets. *ACS Appl Mater Interfaces*. 2015;7(36):19870-19875.
39. Wang H, Zhu B, Ma X, Hao Y, Chen X. Physically transient resistive switching memory based on silk protein. *Small*. 2016;12(20):2715-2719.
40. Fu KK, Wang Z, Dai J, Carter M, Hu L. Transient electronics: materials and devices. *Chem Mater*. 2016;28(11):3527-3539.
41. Landi G, Sorrentino A, Iannace S, Neitzert H. Differences between graphene and graphene oxide in gelatin based systems for transient biodegradable energy storage applications. *Nanotechnology*. 2016;28(5):054005.
42. Hwang SW, Tao H, Kim DH, et al. A physically transient form of silicon electronics. *Science*. 2012;337(6102):1640-1644. https://doi.org/10.1126/science.1226325.
43. Zhang X-S, Brugger J, Kim B. A silk-fibroin-based transparent triboelectric generator suitable for autonomous sensor network. *Nano Energy*. 2016;20:37-47. https://doi.org/10.1016/j.nanoen.2015.11.036.
44. Chen C-H, Tsao Y-H, Lin Z-HJET. Development of biocompatible triboelectric nanogenerators by using polypeptides as the contact materials. *ECS Trans*. 2016;72(6):61-65.
45. Jao Y-T, Yang P-K, Chiu C-M, et al. A textile-based triboelectric nanogenerator with humidity-resistant output characteristic and its applications in self-powered healthcare sensors. *Nano Energy*. 2018;50:513-520. https://doi.org/10.1016/j.nanoen.2018.05.071.
46. Kim J-N, Lee J, Go TW, et al. Skin-attachable and biofriendly chitosan-diatom triboelectric nanogenerator. *Nano*

Energy. 2020;75:104904. https://doi.org/10.1016/j.nanoen.2020.104904.
47. Ma C, Gao S, Gao X, et al. Chitosan biopolymer-derived self-powered triboelectric sensor with optimized performance through molecular surface engineering and data-driven learning. *InfoMat*. 2019;1(1):116-125. https://doi.org/10.1002/inf2.12008.
48. Yue L, Sun D, Khan IM, Liu X, Jiang Q, Xia W. Cinnamyl alcohol modified chitosan oligosaccharide for enhancing antimicrobial activity. *Food Chem*. 2020;309:125513.
49. Liu N, Chen X-G, Park H-J, et al. Effect of MW and concentration of chitosan on antibacterial activity of *Escherichia coli*. *Carbohydr Polym*. 2006;64(1):60-65.
50. Tabata Y, Ikada Y. Protein release from gelatin matrices. *Adv Drug Deliv Rev*. 1998;31(3):287-301.
51. Han Y, Han Y, Zhang X, et al. Fish gelatin based triboelectric nanogenerator for harvesting biomechanical energy and self-powered sensing of human physiological signals. *ACS Appl Mater Interfaces*. 2020;12(14):16442-16450. https://doi.org/10.1021/acsami.0c01061.
52. Liu C, Li J, Che L, Chen S, Wang Z, Zhou X. Toward large-scale fabrication of triboelectric nanogenerator (TENG) with silk-fibroin patches film via spray-coating process. *Nano Energy*. 2017;41:359-366. https://doi.org/10.1016/j.nanoen.2017.09.038.
53. Zhong Q, Zhong J, Hu B, Hu Q, Zhou J, Wang ZL. A paper-based nanogenerator as a power source and active sensor. *Energy & Environmental Science*. 2013;6(6):1779–1784. https://doi.org/10.1039/c3ee40592c.
54. Lee KY, Mooney DJ. Alginate: properties and biomedical applications. *Prog Polym Sci*. 2012;37(1):106-126. https://doi.org/10.1016/j.progpolymsci.2011.06.003.
55. Augst AD, Kong HJ, Mooney DJ. Alginate hydrogels as biomaterials. *Macromol Biosci*. 2006;6(8):623-633.
56. Zhang L, Sun X, Hu Z, Yuan C, Chen C. Rice paper as a separator membrane in lithium-ion batteries. *J Power Sources*. 2012;204:149-154.
57. Bugg TD, Rahmanpour R. Enzymatic conversion of lignin into renewable chemicals. *Curr Opin Chem Biol*. 2015;29:10-17. https://doi.org/10.1016/j.cbpa.2015.06.009.
58. Arca HC, Mosquera-Giraldo LI, Bi V, Xu D, Taylor LS, Edgar KJ. Pharmaceutical applications of cellulose ethers and cellulose ether esters. *Biomacromolecules*. 2018;19(7):2351-2376.
59. Rol F, Belgacem MN, Gandini A, Bras J. Recent advances in surface-modified cellulose nanofibrils. *Prog Polym Sci*. 2019;88:241-264.
60. Wu C, Kima TW, Sung S, Park JH, Li F. Ultrasoft and cuttable paper-based triboelectric nanogenerators for mechanical energy harvesting. *Nano Energy*. 2018;44:279-287. https://doi.org/10.1016/j.nanoen.2017.11.080.
61. Jie Y, Jia X, Zou J, et al. Natural leaf made triboelectric nanogenerator for harvesting environmental mechanical energy. *Adv Energy Mater*. 2018;8(12):1703133–1703139. https://doi.org/10.1002/aenm.201703133.
62. Pang Y, Xi F, Luo J, Liu G, Guo T, Zhang C. An alginate film-based degradable triboelectric nanogenerator. *RSC Adv*. 2018;8(12):6719-6726. https://doi.org/10.1039/c7ra13294h.
63. Yao C, Hernandez A, Yu Y, Cai Z, Wang X. Triboelectric nanogenerators and power-boards from cellulose nanofibrils and recycled materials. *Nano Energy*. 2016;30:103-108. https://doi.org/10.1016/j.nanoen.2016.09.036.
64. He X, Zou H, Geng Z, et al. A hierarchically nanostructured cellulose fiber-based triboelectric nanogenerator for self-powered healthcare products. *Adv Funct Mater*. 2018;28(45):1805540–1805547. https://doi.org/10.1002/adfm.201805540.
65. Rhim J-W, Park H-M, Ha C-S. Bio-nanocomposites for food packaging applications. *Prog Polym Sci*. 2013;38(10–11):1629-1652.
66. Li HB, Huneault MA. Effect of nucleation and plasticization on the crystallization of poly(lactic acid). *Polymer*. 2007;48(23):6855-6866. https://doi.org/10.1016/j.polymer.2007.09.020.
67. Schwach G, Coudane J, Engel R, Vert M. More about the polymerization of lactides in the presence of stannous octoate. *J Polym Sci A Polym Chem*. 1997;35(16):3431-3440.
68. Kumari A, Yadav SK, Yadav SC. Biodegradable polymeric nanoparticles based drug delivery systems. *Colloids Surf B Biointerfaces*. 2010;75(1):1-18. https://doi.org/10.1016/j.colsurfb.2009.09.001.
69. Maksimova NI, Krivoruchko OP. Study of thermocatalytic decomposition of polyethylene and polyvinyl alcohol in the presence of an unsteady-state Fe-containing catalyst. *Chem Eng Sci*. 1999;54(20):4351-4357.
70. Liu B, Qiu D, Zhao CZ. Effect of mixture of plasticizer on the thermoplastics formability of polyvinyl alcohol (PVA). *Trans Tech Publ*. 2010;447:652-656.
71. Chiellini E, Corti A, D'Antone S, Solaro R. Biodegradation of poly (vinyl alcohol) based materials. *Prog Polym Sci*. 2003;28(6):963-1014. https://doi.org/10.1016/S0079-6700(02)00149-1.
72. GHA S. Non-medical biodegradable polymers: environmentally degradable polymers. *Handbook of Biodegradable Polymers*. Amsterdam: Hardwood Academic; 1997:473-511.
73. Agostini D, Lando J, Shelton JR. Synthesis and characterization of poly-β-hydroxybutyrate. I. Synthesis of crystalline DL-poly-β-hydroxybutyrate from DL-β-butyrolactone. *J Polym Science Part A-1: Polym Chem*. 1971;9(10):2775-2787.
74. Miguel O, Fernandez-Berridi M, Iruin J. Survey on transport properties of liquids, vapors, and gases in biodegradable poly (3-hydroxybutyrate)(PHB). *J Appl Polym Sci*. 1997;64(9):1849-1859.
75. Choi AY, Lee CJ, Park J, Kim D, Kim YT. Corrugated textile based Triboelectric generator for wearable energy harvesting. *Sci Rep*. 2017;7:45583. https://doi.org/10.1038/srep45583.
76. Yang W, Cao R, Zhang X, Li H, Li C. Air-permeable and washable paper-based triboelectric nanogenerator based on highly flexible and robust paper electrodes. *Adv Mater Technol*. 2018;3(11):1800178–1800186. https://doi.org/10.1002/admt.201800178.
77. Mallineni SSK, Dong Y, Behlow H, Rao AM, Podila RA. Wireless triboelectric nanogenerator. *Adv Energy Mater*. 2018;8(10):1702736–1702742. https://doi.org/10.1002/aenm.201702736.
78. Feng Y, Zhang L, Zheng Y, Wang D, Zhou F, Liu W. Leaves based triboelectric nanogenerator (TENG) and TENG tree for wind energy harvesting. *Nano Energy*. 2019;55:260-268. https://doi.org/10.1016/j.nanoen.2018.10.075.

79. Xia K, Zhu Z, Fu J, et al. A triboelectric nanogenerator based on waste tea leaves and packaging bags for powering electronic office supplies and behavior monitoring. *Nano Energy*. 2019;60:61-71. https://doi.org/10.1016/j.nanoen.2019.03.050.
80. Luo J, Wang Z, Xu L, et al. Flexible and durable wood-based triboelectric nanogenerators for self-powered sensing in athletic big data analytics. *Nat Commun*. 2019;10(1):5147. https://doi.org/10.1038/s41467-019-13166-6.
81. Chen S, Jiang J, Xu F, Gong S. Crepe cellulose paper and nitrocellulose membrane-based triboelectric nanogenerators for energy harvesting and self-powered human-machine interaction. *Nano Energy*. 2019;61:69-77. https://doi.org/10.1016/j.nanoen.2019.04.043.
82. Wang L, Daoud WA. Highly flexible and transparent polyionic-skin triboelectric nanogenerator for biomechanical motion harvesting. *Adv Energy Mater*. 2018;9(5):1803183. https://doi.org/10.1002/aenm.201803183.
83. Guo Y, Zhang X-S, Wang Y, et al. All-fiber hybrid piezoelectric-enhanced triboelectric nanogenerator for wearable gesture monitoring. *Nano Energy*. 2018;48:152-160. https://doi.org/10.1016/j.nanoen.2018.03.033.
84. Zhu Z, Xia K, Xu Z, Lou H, Zhang H. Starch paper-based triboelectric nanogenerator for human perspiration sensing. *Nanoscale Res Lett*. 2018;13(1):365. https://doi.org/10.1186/s11671-018-2786-9.
85. Wang R, Mu L, Bao Y, et al. Holistically engineered polymer-polymer and polymer-ion interactions in biocompatible polyvinyl alcohol blends for high-performance Triboelectric devices in self-powered wearable cardiovascular Monitorings. *Adv Mater*. 2020;32(32):e2002878. https://doi.org/10.1002/adma.202002878.
86. Qian C, Li L, Gao M, et al. All-printed 3D hierarchically structured cellulose aerogel based triboelectric nanogenerator for multi-functional sensors. *Nano Energy*. 2019;63:103885. https://doi.org/10.1016/j.nanoen.2019.103885.
87. Jiang C, Wu C, Li X, et al. All-electrospun flexible triboelectric nanogenerator based on metallic MXene nanosheets. *Nano Energy*. 2019;59:268-276. https://doi.org/10.1016/j.nanoen.2019.02.052.
88. Wang H, Wang J, He T, Li Z, Lee C. Direct muscle stimulation using diode-amplified triboelectric nanogenerators (TENGs). *Nano Energy*. 2019;63:103844–103853. https://doi.org/10.1016/j.nanoen.2019.06.040.
89. Tang W, Tian J, Zheng Q, et al. Implantable self-powered low-level laser cure system for mouse embryonic osteoblasts' proliferation and differentiation. *ACS Nano*. 2015;9(8):7867-7873.
90. Tian J, Shi R, Liu Z, et al. Self-powered implantable electrical stimulator for osteoblasts' proliferation and differentiation. *Nano Energy*. 2019;59:705-714. https://doi.org/10.1016/j.nanoen.2019.02.073.
91. Yao G, Kang L, Li J, et al. Effective weight control via an implanted self-powered vagus nerve stimulation device. *Nat Commun*. 2018;9(1):5349. https://doi.org/10.1038/s41467-018-07764-z.
92. Lee S, Wang H, Wang J, et al. Battery-free neuromodulator for peripheral nerve direct stimulation. *Nano Energy*. 2018;50:148-158. https://doi.org/10.1016/j.nanoen.2018.04.004.
93. Zhang Y, Zhou Z, Sun L, Liu Z, Xia X, Tao TH. "Genetically engineered" biofunctional triboelectric nanogenerators using recombinant spider silk. *Adv Mater*. 2018;30(50):e1805722. https://doi.org/10.1002/adma.201805722.
94. Willatzen M, Lin Wang Z. Theory of contact electrification: optical transitions in two-level systems. *Nano Energy*. 2018;52:517-523. https://doi.org/10.1016/j.nanoen.2018.08.015.
95. Li S, Nie J, Shi Y, et al. Contributions of different functional groups to contact electrification of polymers. *Adv Mater*. 2020;32(25):e2001307. https://doi.org/10.1002/adma.202001307.
96. Chen J, Wang ZL. Reviving vibration energy harvesting and self-powered sensing by a triboelectric nanogenerator. *Joule*. 2017;1(3):480-521. https://doi.org/10.1016/j.joule.2017.09.004.
97. Zou H, Zhang Y, Guo L, et al. Quantifying the triboelectric series. *Nat Commun*. 2019;10(1):1427. https://doi.org/10.1038/s41467-019-09461-x.
98. Ray TR, Choi J, Bandodkar AJ, et al. Bio-integrated wearable systems: a comprehensive review. *Chem Rev*. 2019;119(8):5461-5533. https://doi.org/10.1021/acs.chemrev.8b00573.
99. Niu Q, Huang L, Lv S, Shao H, Fan S, Zhang Y. Pulse-driven bio-triboelectric nanogenerator based on silk nanoribbons. *Nano Energy*. 2020;74:104837. https://doi.org/10.1016/j.nanoen.2020.104837.
100. He F, You X, Gong H, et al. Stretchable, biocompatible, and multifunctional silk fibroin-based hydrogels toward wearable strain/pressure sensors and triboelectric nanogenerators. *ACS Appl Mater Interfaces*. 2020;12(5):6442-6450. https://doi.org/10.1021/acsami.9b19721.
101. Pan R, Xuan W, Chen J, et al. Fully biodegradable triboelectric nanogenerators based on electrospun polylactic acid and nanostructured gelatin films. *Nano Energy*. 2018;45:193-202. https://doi.org/10.1016/j.nanoen.2017.12.048.
102. Park IW, Choi J, Kim KY, et al. Vertically aligned cyclo-phenylalanine peptide nanowire-based high-performance triboelectric energy generator. *Nano Energy*. 2019;57:737-745. https://doi.org/10.1016/j.nanoen.2019.01.008.
103. Sun Z, Yang L, Liu S, Zhao J, Hu Z, Song WA. Green Triboelectric Nano-generator composite of degradable cellulose, piezoelectric polymers of PVDF/PA6, and nanoparticles of BaTiO3. *Sensors (Basel)*. 2020;20(2):506–519. https://doi.org/10.3390/s20020506.
104. Yao C, Yin X, Yu Y, Cai Z, Wang X. Chemically functionalized natural cellulose materials for effective triboelectric nanogenerator development. *Adv Funct Mater*. 2017;27(30):1700794–1700800. https://doi.org/10.1002/adfm.201700794.
105. Chandrasekhar A, Alluri NR, Saravanakumar B, Selvarajan S, Kim S-J. A microcrystalline cellulose ingrained polydimethylsiloxane triboelectric nanogenerator as a self-powered locomotion detector. *J Mater Chem C*. 2017;5(7):1810-1815. https://doi.org/10.1039/c6tc05104a.
106. Peng J, Zhang H, Zheng Q, et al. A composite generator film impregnated with cellulose nanocrystals for enhanced triboelectric performance. *Nanoscale*. 2017;9(4):1428-1433. https://doi.org/10.1039/c6nr07602e.
107. Wang M, Li W, You C, Wang Q, Zeng X, Chen M. Triboelectric nanogenerator based on 317L stainless steel and ethyl cellulose for biomedical applications. *RSC Adv*. 2017;7(11):6772-6779. https://doi.org/10.1039/c6ra28252k.

108. Zhang C, Lin X, Zhang N, et al. Chemically functionalized cellulose nanofibrils-based gear-like triboelectric nanogenerator for energy harvesting and sensing. *Nano Energy*. 2019;66:104126. https://doi.org/10.1016/j.nanoen.2019.104126.
109. Šutka A, Ruža J, Järvekülg M, et al. Triboelectric nanogenerator based on immersion precipitation derived highly porous ethyl cellulose. *J Electrostat*. 2018;92:1-5. https://doi.org/10.1016/j.elstat.2018.01.003.
110. Shao Y, Feng C-P, Deng B-W, Yin B, Yang M-B. Facile method to enhance output performance of bacterial cellulose nanofiber based triboelectric nanogenerator by controlling micro-nano structure and dielectric constant. *Nano Energy*. 2019;62:620-627. https://doi.org/10.1016/j.nanoen.2019.05.078.
111. Li M, Jie Y, Shao L-H, et al. All-in-one cellulose based hybrid tribo/piezoelectric nanogenerator. *Nano Research*. 2019;12(8):1831-1835. https://doi.org/10.1007/s12274-019-2443-3.
112. Cui P, Parida K, Lin M-F, Xiong J, Cai G, Lee PS. Transparent, flexible cellulose nanofibril-phosphorene hybrid paper as triboelectric nanogenerator. *Adv Mater Interfaces*. 2017;4(22):1700651–1700657. https://doi.org/10.1002/admi.201700651.
113. Mao Y, Zhang N, Tang Y, Wang M, Chao M, Liang E. A paper triboelectric nanogenerator for self-powered electronic systems. *Nanoscale*. 2017;9(38):14499-14505. https://doi.org/10.1039/c7nr05222g.
114. Jang S, Kim H, Oh JH. Simple and rapid fabrication of pencil-on-paper triboelectric nanogenerators with enhanced electrical performance. *Nanoscale*. 2017;9(35):13034-13041. https://doi.org/10.1039/c7nr04610c.
115. Xia K, Du C, Zhu Z, Wang R, Zhang H, Xu Z. Sliding-mode triboelectric nanogenerator based on paper and as a self-powered velocity and force sensor. *Appl Mater Today*. 2018;13:190-197. https://doi.org/10.1016/j.apmt.2018.09.005.
116. Parandeh S, Kharaziha M, Karimzadeh F. An eco-friendly triboelectric hybrid nanogenerators based on graphene oxide incorporated polycaprolactone fibers and cellulose paper. *Nano Energy*. 2019;59:412-421. https://doi.org/10.1016/j.nanoen.2019.02.058.
117. Chi Y, Xia K, Zhu Z, et al. Rice paper-based biodegradable triboelectric nanogenerator. *Microelect Eng*. 2019;216:111059. https://doi.org/10.1016/j.mee.2019.111059.
118. Khandelwal G, Minocha T, Yadav SK, et al. All edible materials derived biocompatible and biodegradable triboelectric nanogenerator. *Nano Energy*. 2019;65:104016. https://doi.org/10.1016/j.nanoen.2019.104016.
119. Qiao H, Zhang Y, Huang Z, Wang Y, Li D, Zhou H. 3D printing individualized triboelectric nanogenerator with macropattern. *Nano Energy*. 2018;50:126-132. https://doi.org/10.1016/j.nanoen.2018.04.071.
120. Ryu H, Lee J-H, Kim T-Y, et al. High-performance triboelectric nanogenerators based on solid polymer electrolytes with asymmetric pairing of ions. *Adv Energy Mater*. 2017;7(17):1700289–1700295. https://doi.org/10.1002/aenm.201700289.
121. Liang Q, Zhang Q, Yan X, et al. Recyclable and green triboelectric nanogenerator. *Adv Mater*. 2017;29(5):1604961–1604967. https://doi.org/10.1002/adma.201604961.
122. Cui N, Liu J, Lei Y, et al. High-performance triboelectric nanogenerator with a rationally designed friction layer structure. *ACS Appl Energy Mater*. 2018;1(6):2891-2897. https://doi.org/10.1021/acsaem.8b00530.

How to cite this article: Chao S, Ouyang H, Jiang D, Fan Y, Li Z. Triboelectric nanogenerator based on degradable materials. *EcoMat*. 2021;3:e12072. https://doi.org/10.1002/eom2.12072