

## REVIEW

# Triboelectric nanogenerator based on degradable materials

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## 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.<sup>1</sup> Triboelectric nanogenerator (TENG), as a neotype green energy technology, can get the energy from the surrounding environment and organism.<sup>2-6</sup> It was

reported by Pro. Wang and co-workers in 2012,<sup>7</sup> 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.<sup>8</sup> 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.<sup>9</sup>

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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.<sup>10-13</sup> 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.<sup>14-16</sup>

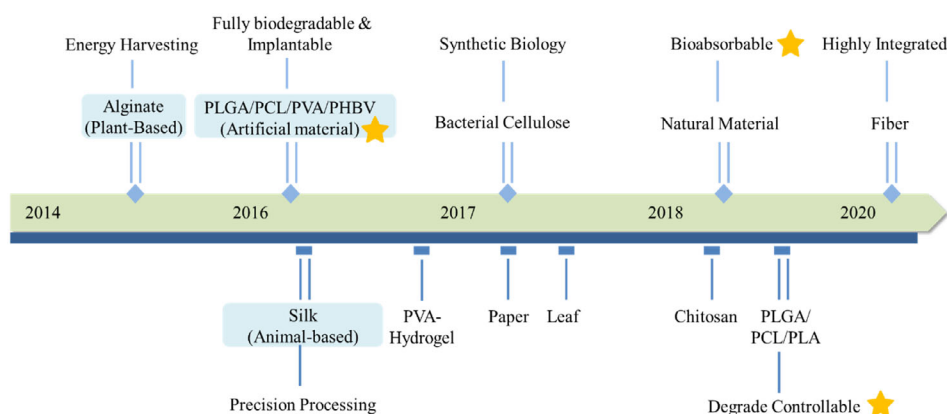
Here, we divide the degradable materials into three types by the source, including animal-based degradable material, plant-based degradable material, and artificial degradable material. Animal-based degradable materials are defined in degradable materials obtained from animal sources in this review, 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.<sup>17</sup> 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 (polyhydroxybutyrate 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.<sup>12</sup> 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.<sup>18</sup> 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.<sup>19</sup> In 2017, Xiao-Sheng Zhang et al began to use plant-based degradable material paper as a triboelectric layer to make TENG.<sup>21-33</sup> 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.<sup>16</sup> In October of the same year, Zhe Li et al applied gold nanorods to DB-TENG, completing the photothermal degradation regulation of implantable TENG.<sup>13</sup> Besides, some studies have reviewed DB-TENG from different prospects.<sup>25-28</sup> From the perspective of materials, Aifang Yu et al<sup>27</sup> introduce the biodegradable/bioabsorbable TENG with degradable materials and discuss the performance optimization of it. Kaushik Paridaa et al<sup>25</sup> 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,<sup>29</sup> and medical care.<sup>30</sup>

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,<sup>18</sup> 2016 to 2017,<sup>12,19,20</sup> 2017 to 2018,<sup>21-23</sup> 2018 to 2019,<sup>13,15,16</sup> 2019 to 2020<sup>24</sup>

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.

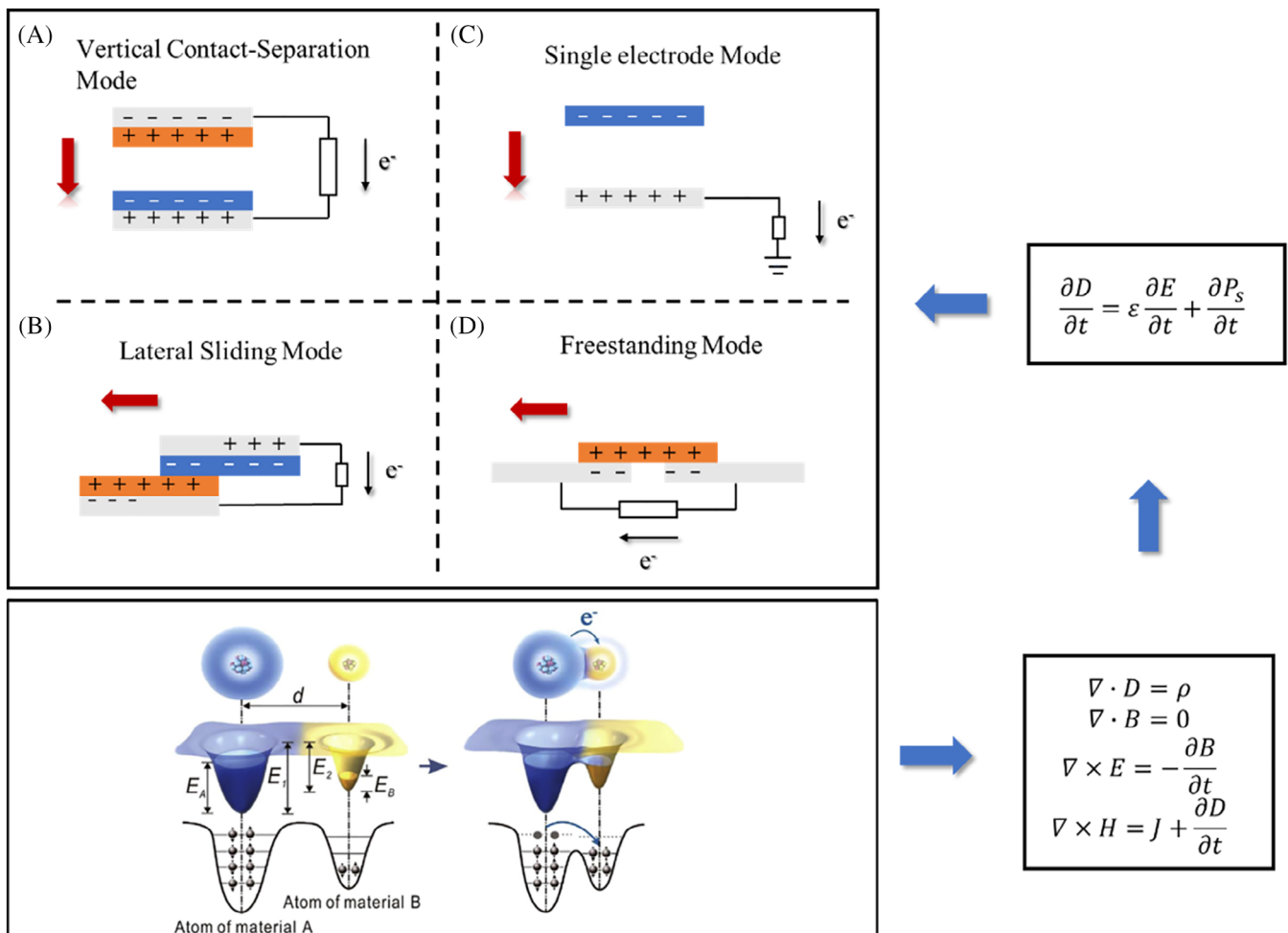
## 2 | TRIBOELECTRIC NANOGENERATOR

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<sup>31</sup> 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.<sup>31,32,35,36</sup> (e) CE mechanism explained by electron cloud-potential well model at high temperatures<sup>37</sup>

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<sup>32</sup> 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<sup>33,34</sup> 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.<sup>33</sup> 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.<sup>35</sup> 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,<sup>38,39</sup> gelatin,<sup>40,41</sup> and so on are served as substrates

and encapsulation layers and are widely used in transient electronic devices.<sup>42</sup> Recently, the animal-based degradable materials with excellent triboelectric effect have been applied to the triboelectric layer of TENG. Since 2015, The silk fibroin,<sup>19,43</sup> gelatin,<sup>44</sup> and chitin<sup>15,45-47</sup> 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.<sup>48</sup> Besides, chitin and chitosan have excellent antibacterial effects and have promising applications in the medical field.<sup>49</sup>

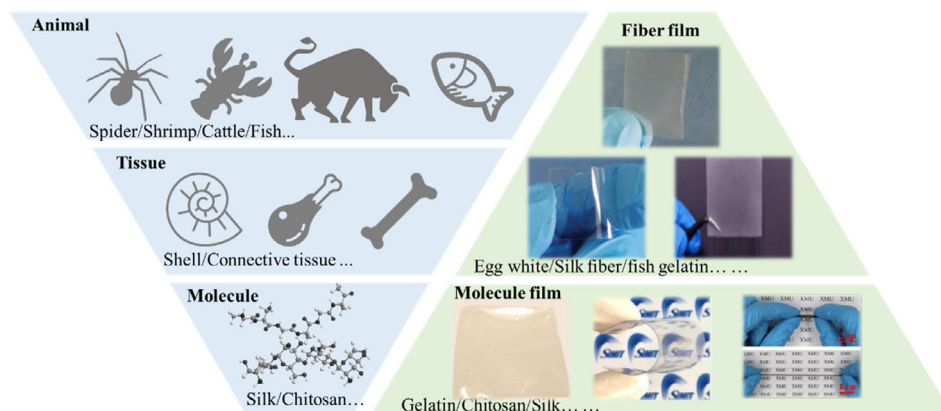
The protein and its derivatives, such as silk, gelatin, egg white, peptides, and so on can serve as a triboelectric layer in TENG. These triboelectric materials usually have a strong ability to transfer electrons due to the carboxyl group. Besides, these proteins can be quickly degraded in environments 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,<sup>50</sup> 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).

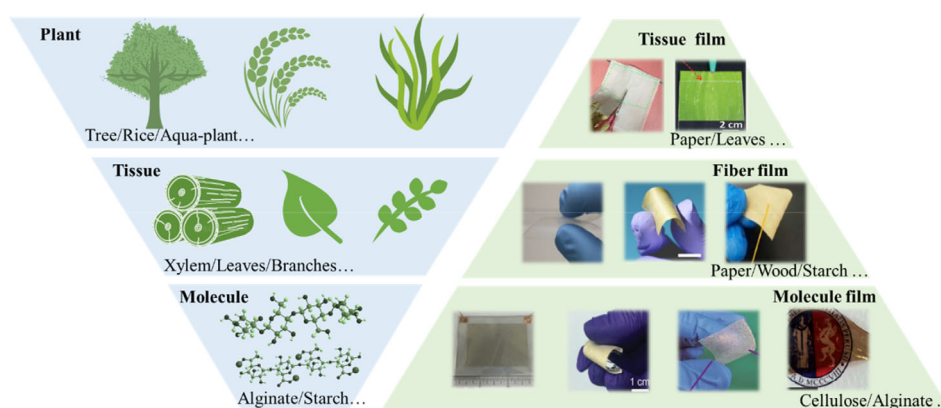
## 3.2 | TENG based on plant-based degradable materials

In 2013, Z. L. Wang's group began to explore an important plant-based biodegradable material—paper as a component of TENG.<sup>53</sup> 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<sup>12,15,16,51,52</sup>



**FIGURE 4** Hierarchy chart for plant-based degradable material used in TENG<sup>16,18,60-64</sup>



to TENG. Then, Xiao-Sheng Zhang et al<sup>21</sup> reported TENG using paper and Teflon as the triboelectric layer in 2017; Luca Valentini et al<sup>18</sup> 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.<sup>54</sup> It is usually added to other mixed systems (such as salt solution, etc.) to form a film by cross-linking.<sup>55</sup> 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.<sup>13</sup> 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.<sup>56</sup> 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.<sup>57,58</sup> Specially modified molecules can endow cellulose with stronger abilities to transfer electrons and better physical and chemical properties (Figure 4).<sup>59</sup>

### 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.<sup>7,32,35</sup> 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.<sup>12</sup> 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.<sup>13</sup> 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.<sup>65,66</sup> 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.<sup>67,68</sup>

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.<sup>69</sup> Polyvinyl alcohol film has good mechanical properties, biodegradability, biocompatibility, but poor thermal stability.<sup>70,71</sup> The PCL generally has a larger overall molecular weight and relatively slow biodegradation compared with among other polymer materials.<sup>72</sup>

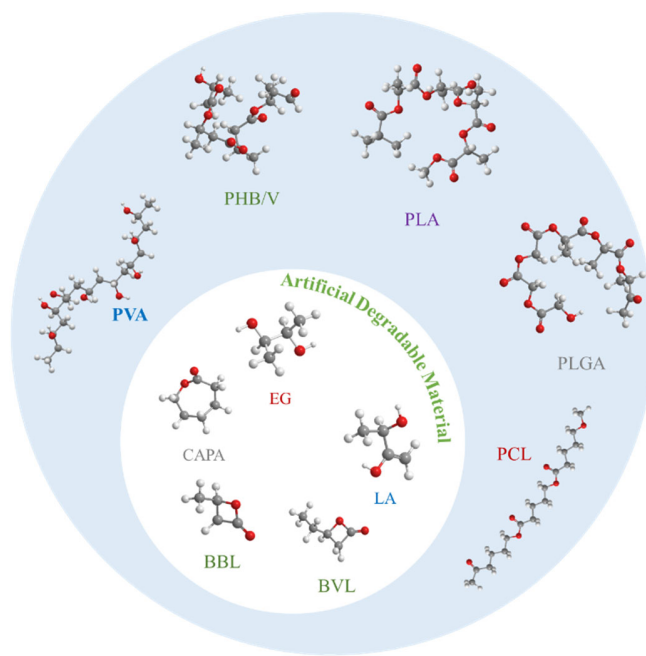
PHB/V, as a copolymer of hydroxybutyric acid and valeric acid, is also one of PHB derivative polymers. The synthesis of PHB/V mainly relies on chemical methods.<sup>73</sup> Like several other materials which are mentioned, PHB/V has good biocompatibility and biodegradability and has good mechanical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

## 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).<sup>15</sup> 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

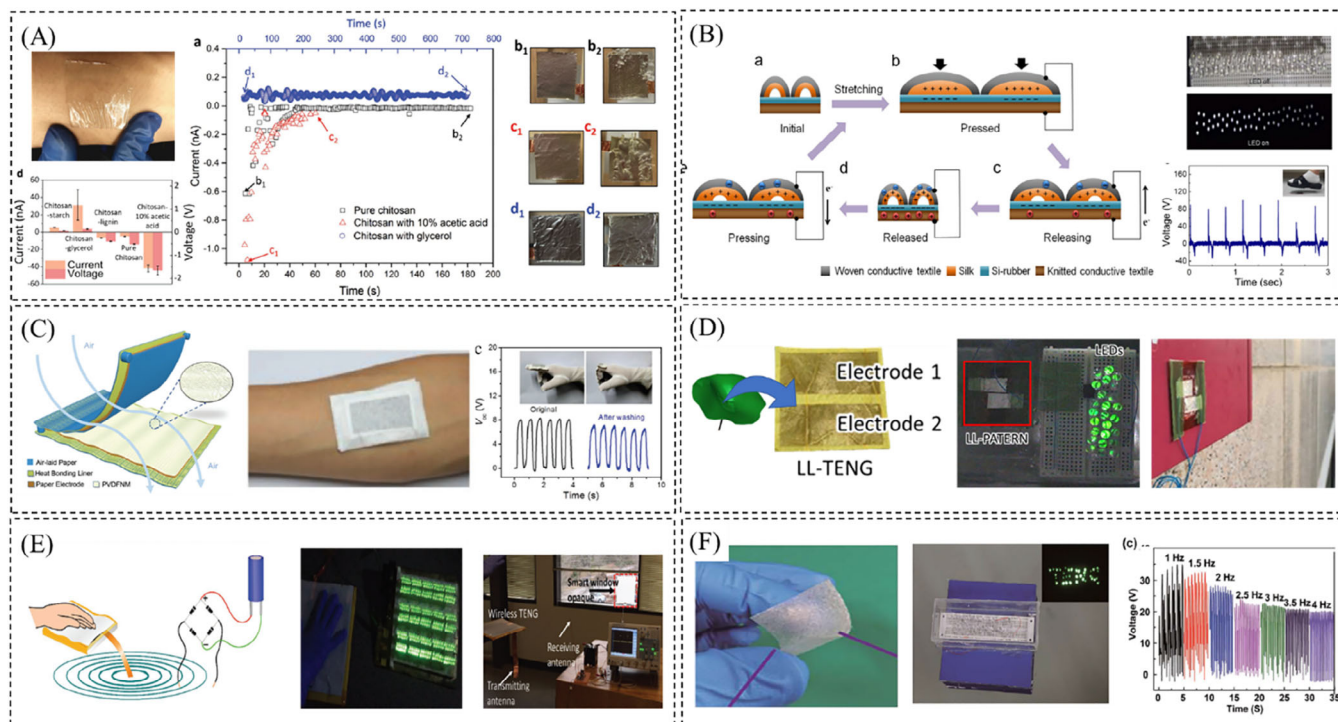


**FIGURE 5** Artificial degradable material used in TENG and its monomer; BBL, β-butyrolactone; BVL, β-valerolactone; CAPA, caprolactone; EG, ethylene glycol; LA, lactic acid; PCL, polycaprolactone; PHB/V, poly(1-hydroxybutyrate-co-3-hydroxyvalerate); PLA, polylactic acid; PLGA, poly(lactic-co-glycolic acid); PVA, polyvinyl alcohol

reaching 28.13 V/2.71 μA (Figure 6B).<sup>75</sup> 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).<sup>76</sup> 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.<sup>78</sup> In addition, Dongwhi Choia et al designed a liquid-solid contacted degradable TENG to harvest raindrop energy in external environments (Figure 6D).<sup>22,61</sup> Sai Sunil Kumar Mallineni fabricated TENG with PLA and achieved a high voltage of up to 2700 V (Figure 6E).<sup>77</sup> In addition, Yaokun Pang fabricated degradable TENG based on alginate film. The maximum output can reach 33 V/150 nA (Figure 6F).<sup>62</sup> In 2019, Kequan Xia proposed an environmentally friendly TENG with wasted tea leaves and packaging boxes. The output of the TENG also reached 792 V/42.8 μA.<sup>79</sup>

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

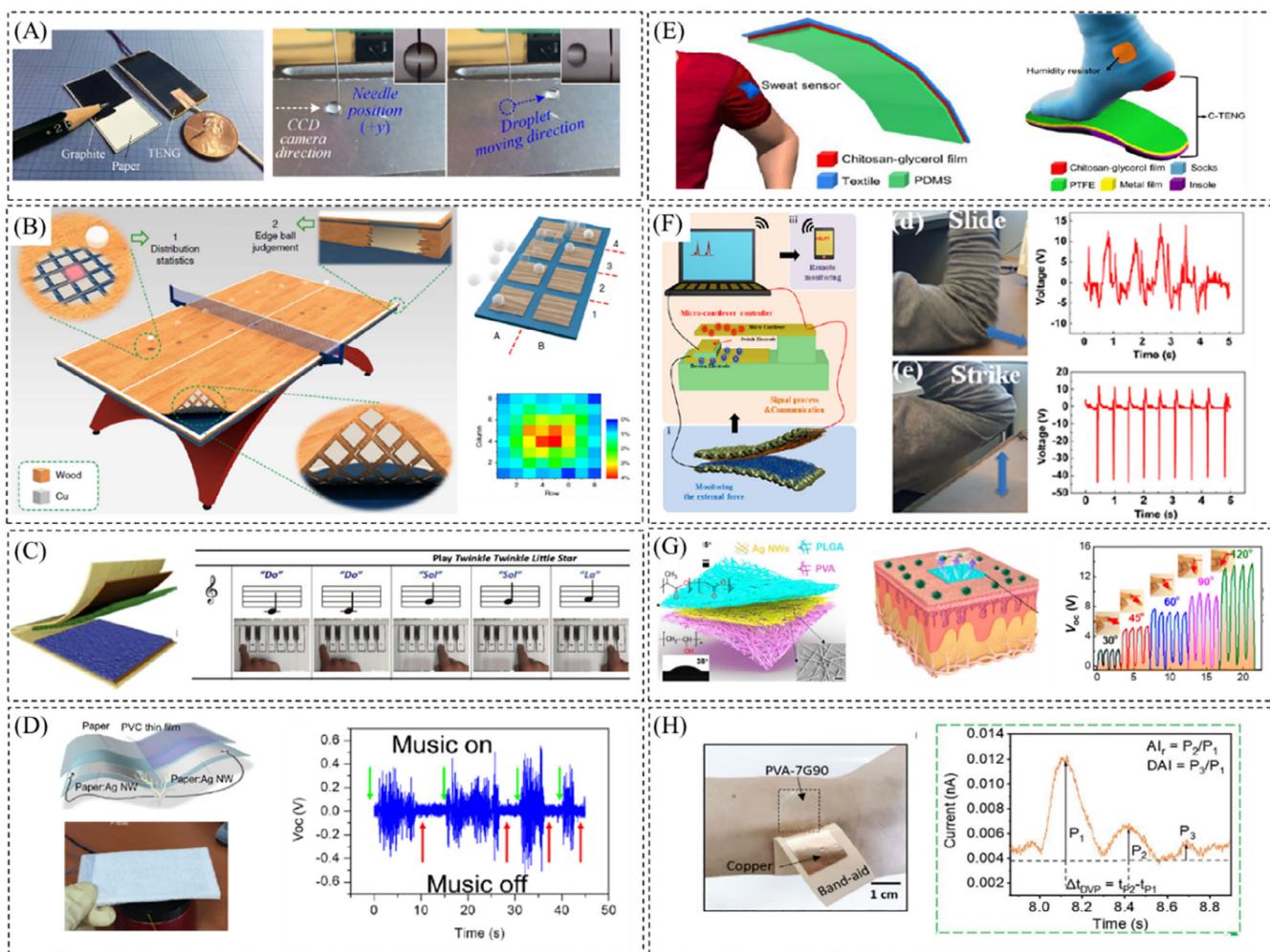
materials also endow TENG more distinctive features in transient electronic device applications.

Xiao-Sheng Zhang et al first reported a paper-based TENG with an output of 85 V/3.75  $\mu$ A. 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).<sup>21</sup> 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).<sup>80</sup> Sheng Chen et al used paper and cellulose paper as the triboelectric layer to fabricated TENG and achieved 196.8 V and 31.5  $\mu$ A output. This work reported a paper piano with the paper-based TENG's (Figure 7C).<sup>81</sup> In addition, Chaoping 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).<sup>60</sup>

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.<sup>82</sup> 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).<sup>45</sup> Wei Xu et al reported a TENG using hydrogel containing PVA, which can directly and effectively detect arm bending from 0° to 150°.<sup>20</sup> 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).<sup>83</sup> Zhu Zhiyuan et al report a single-electrode TENG with rice paper to detect sweat produced on the arm.<sup>84</sup>

To improve the monitoring accuracy, some new materials and structures are applied in DB-TEG. 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.<sup>86</sup> Jong-Nam Kim et al reported a motion sensor based on DB-TEG with more compatible for skin. The degradable material gelatin mixed with different proportions of silica gel used as the triboelectric layer.<sup>46</sup> 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<sup>21</sup> and its application. B, Schematic of a wood-based TENG<sup>80</sup> for sport training. C, The structure of the paper-based TENG<sup>81</sup> and its application of paper piano. D, A TENG<sup>60</sup> based on paper. E, The structure and application of the chitosan-glycerin based TENG.<sup>45</sup> F, Schematic of silk-fibroin based TENG and its application.<sup>83</sup> G, The structure and application of all-fiber TENG.<sup>24</sup> H, A DB-TENG<sup>85</sup> and its performance

layer to fabricate a small and transparent TENG for real-time healthcare detection.<sup>87</sup> 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).<sup>24</sup> 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<sup>2</sup> on the skin and accurately monitor the pulse (Figure 7H).<sup>85</sup>

### 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.<sup>12</sup> 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  $\mu$ A. 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

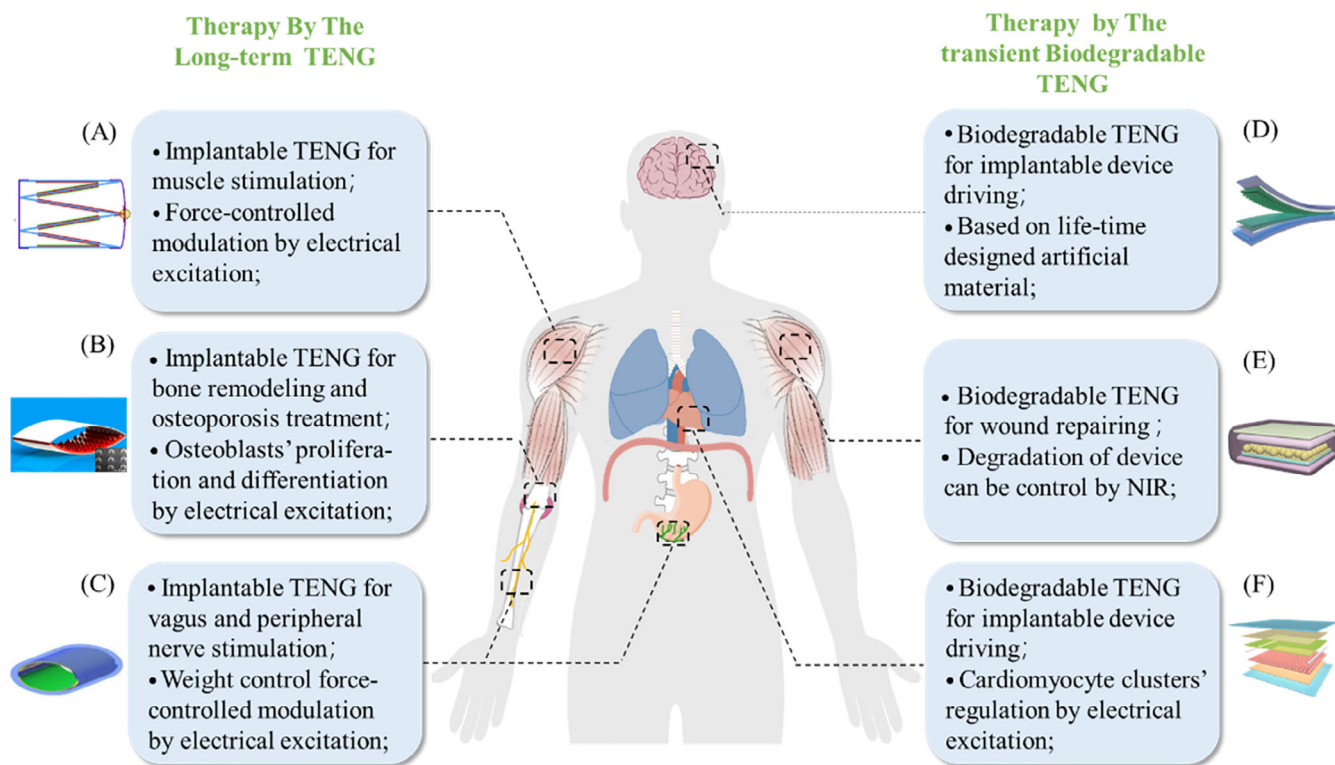
generated by TENG applied to wound treatment can effectively reduce infection and promote healing. Wen Jiang et al demonstrated a full bioabsorbable TENG with several natural degradable materials, such as animal-based materials chitosan, egg white, silk fibroin, plant-based materials cellulose, and rice paper.<sup>16</sup> The maximum output of 55 V/0.6  $\mu$ A 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.<sup>13</sup> 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  $\times$  3 cm, and the maximum electrical output can reach 145 V/4.28  $\mu$ A. 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%.<sup>93</sup> 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<sup>94</sup> 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<sup>95</sup> 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.<sup>12,13,16,96,97</sup> 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

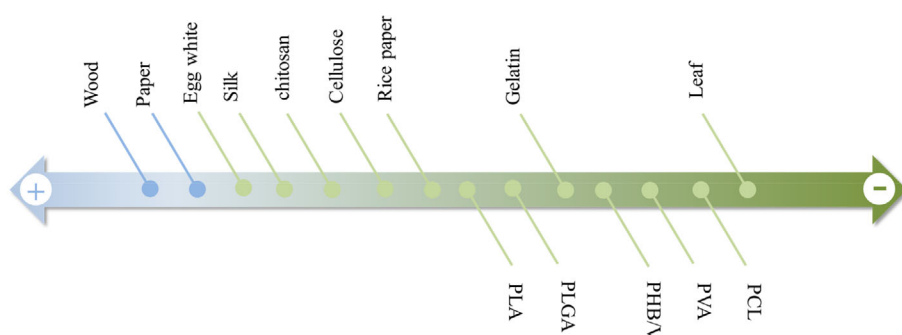
In addition, surface microstructure is also an important factor affecting the output of DB-TENG. Various process of material preparation and modification have been demonstrated to affect the surface microstructure. Water lithography, electrostatic spinning, and other materials preparation methods can 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.<sup>98</sup> 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<sup>12,13,16,96,97</sup> 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 $\mu$ A	5 cm $\times$ 3 cm	Sweat sensor	[45]
Animal-based	Chitin/chitosan	PI	Water	Biocompatible	1.35 V/42 nA	2 cm $\times$ 1 cm	Energy harvesting	[15]
Animal-based	Chitin/chitosan	PI		Biocompatible	16.2 V/125 $\mu$ A	$\sim$ 5 cm $\times$ 5 cm	Speed sensor	[47]
Animal-based	Chitin/chitosan	FEP		Biocompatible	150 V/1.02 $\mu$ A	3 cm $\times$ 4 cm	Behavior sensor	[46]
Animal-based	Egg white	Rice paper	In vivo & methanol	Both	55 V/0.6 $\mu$ A	1 cm $\times$ 2 cm	Drive implantable device	[16]
Animal-based	Silk/silk fibroin	Rice paper & cellulose	In vivo & methanol	Both	$\sim$ 40 V/0.4 $\mu$ A & $\sim$ 25 V/0.3 $\mu$ A	1 cm $\times$ 2 cm	Drive implantable device	[16]
Animal-based	Silk/silk fibroin	PI		Both	$\sim$ 15 V/ $\sim$ 2.5 $\mu$ A	$\sim$ 50 cm <sup>2</sup>	Drive electronics	[19]
Animal-based	Silk/silk fibroin	Regenerative silk		Both	41.6 V/0.5 $\mu$ A	1 cm $\times$ 2 cm	Body sensor	[99]
Animal-based	Silk/silk fibroin	PET		Both	268 V/5.78 $\mu$ A	2 cm $\times$ 4 cm	Active sensor	[43]
Animal-based	Silk/silk fibroin	PET		Both	145 V/4.28 $\mu$ A	2 cm $\times$ 3 cm	Antibacterial patch	[93]
Animal-based	Silk/silk fibroin	PET		Both	213.9 V/0.34 $\mu$ A	3 cm $\times$ 3.6 cm	Energy harvesting	[52]
Animal-based	Silk/silk fibroin	PDMS		Both	12 V/0.6 $\mu$ A		Behavior sensor	[100]
Animal-based	Silk/silk fibroin	PVDF		Both	500 V/12 $\mu$ A	2 cm $\times$ 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 $\mu$ A	5 cm $\times$ 3 cm	Wearable Energy Harvesting	[75]
Animal-based	Gelatin	PLA		Both	500 V/16 $\mu$ A	4 cm $\times$ 4 cm	Energy harvesting	[101]
Animal-based	Gelatin	PTFE/PDMS		Both	130 V/0.35 $\mu$ A	5 cm $\times$ 5 cm	Wearable Energy Harvesting	[51]
Animal-based	Polypeptide	PTFE		Both	$\sim$ 350 V/10 $\mu$ A	2.15 cm $\times$ 5.16 cm	Energy harvesting	[102]
Animal-based	Polypeptide	PTFE		Both	65 V	10 cm $\times$ 10 cm	Energy harvesting	[44]
Plant-based	Cellulose	Egg white & silk fiber	In vivo & methanol	Both	45 V/0.4 $\mu$ A & 32 V/0.3 $\mu$ A	1 cm $\times$ 2 cm	Drive implantable device	[16]
Plant-based	Cellulose	PTFE		Both	7.925 V/1.095 $\mu$ A	3 cm $\times$ 3 cm	Energy harvesting	[103]
Plant-based	Cellulose	Cu		Both	13 V/3.2 $\mu$ A	5 cm $\times$ 5 cm	Energy harvesting	[23]
Plant-based	Cellulose	FEP & Cu		Both	8 V/9 $\mu$ A & 0.8 V/0.8 $\mu$ A	1 cm $\times$ 1 cm	Energy harvesting	[104]
Plant-based	Cellulose	FEP		Both	$\sim$ 30 V/90 $\mu$ A	40 cm <sup>2</sup>	Drive electronics	[63]
Plant-based	Cellulose	FEP		Both	21.9 V/0.17 $\mu$ A	3 cm $\times$ 3 cm	Antibacterial patch	[64]

(Continues)

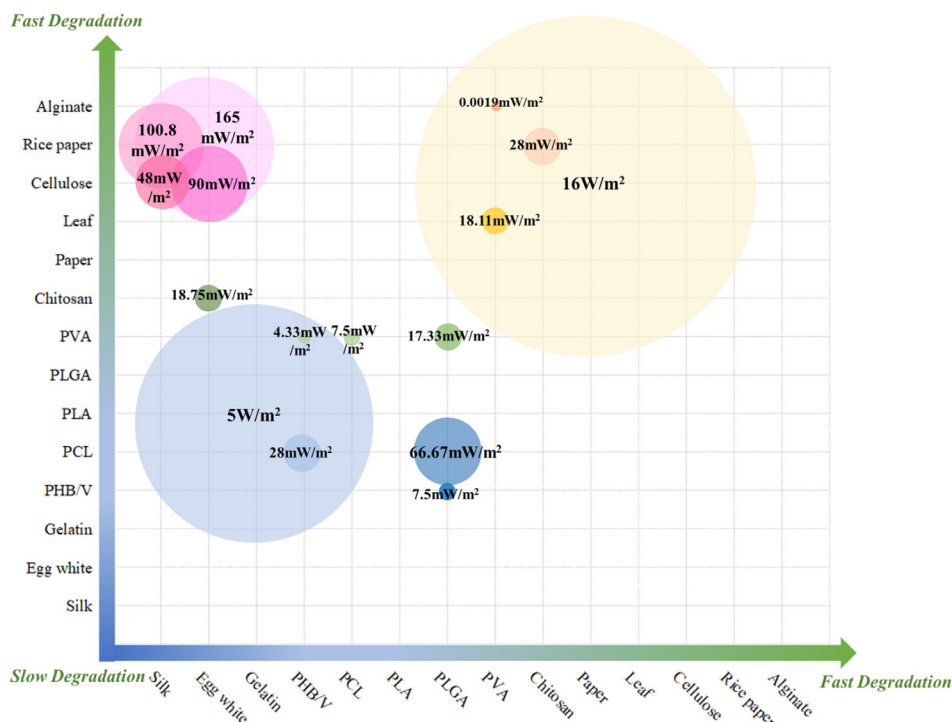
TABLE 1 (Continued)

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 $\mu$ A	3 cm $\times$ 3 cm	Wearable Energy Harvesting	[105]
Plant-based	Cellulose	Al		Both	320 V/11.25 $\mu$ A	1.5 cm $\times$ 1.5 cm	Energy harvesting	[106]
Plant-based	Cellulose	Steel		Both	240 V/50 $\mu$ A	800 cm <sup>2</sup>	Drive electronics	[107]
Plant-based	Cellulose	FEP		Both	286.5 V	18 cm <sup>2</sup>	Drive electronics	[108]
Plant-based	Cellulose	PDMS		Both	55.8 V/0.94 $\mu$ A	3.2 cm $\times$ 3.2 cm	Human behavior sensor	[86]
Plant-based	Cellulose	PDMS		Both	45 V	2.5 cm $\times$ 2.5 cm	Drive electronics	[109]
Plant-based	Cellulose	PDMS		Both	181 V/21 $\mu$ A		Human behavior sensor	[110]
Plant-based	Cellulose	Ni		Both	18 V/2.4 $\mu$ A	1.5 cm $\times$ 1 cm	Drive electronics	[111]
Plant-based	Cellulose	phosphorene		Both	5.2 V/1.8 $\mu$ A	1 cm $\times$ 1 cm	Energy harvesting	[112]
Plant-based	Paper	PTFE		Biocompatible	400 V/170 $\mu$ A	10 cm $\times$ 15 cm	Drive electronics	[113]
Plant-based	Paper	PTFE		Biocompatible	85 V/3.75 $\mu$ A	2 cm $\times$ 4 cm	Control droplet	[21]
Plant-based	Paper	PI		Biocompatible	$\sim$ 96 V/11 $\mu$ A	6 cm $\times$ 7 cm	Drive electronics	[114]
Plant-based	Paper	PVC		Biocompatible	100 V	10 cm $\times$ 10 cm	Loudspeaker	[60]
Plant-based	Paper	PVDF		Biocompatible	197 V/16.2 $\mu$ A	4 cm $\times$ 4 cm	Drive electronics	[76]
Plant-based	Paper	Nitrocellulose membrane		Biocompatible	196.8 V/31.5 $\mu$ A	2.5 cm $\times$ 2.5 cm	Paper piano	[81]
Plant-based	Paper	Teflon		Biocompatible	$\sim$ 1000 V/42 $\mu$ A	4 cm $\times$ 2 cm (stacked)	Drive electronics	[115]
Plant-based	Paper	PCL/GO		Biocompatible	120 V/4 $\mu$ A	4 cm $\times$ 4 cm	Drive electronics	[116]
Plant-based	Leaf	PVDF		Biocompatible	1000 V/60 $\mu$ A	4 cm $\times$ 4 cm	Environmental energy harvesting	[78]
Plant-based	Leaf	PTFE		Biocompatible	792 V/42.8 $\mu$ A	5 cm $\times$ 5 cm	Behavior sensor	[79]
Plant-based	Leaf	PMMA		Biocompatible	230 V/9.5 $\mu$ A	8 cm $\times$ 8 cm	Drive electronics	[61]
Plant-based	Leaf	Water (droplet)		Biocompatible	$\sim$ 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 $\mu$ A & 45 V/0.4 $\mu$ A & 48 V/0.42 $\mu$ A	1 cm $\times$ 2 cm	Drive implantable device	[16]
Plant-based	Rice paper	Skin (hand)	Water	Both	11.2 V/ $\sim$ 1 $\mu$ A	4.4 cm $\times$ 4.4 cm	Human behavior sensor	[84]
Plant-based	Rice paper	PVC		Both	244 V/6 $\mu$ A	3 cm $\times$ 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 <sup>2</sup>	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 <sup>2</sup>	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, polyimide; 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, poly(1-hydroxybutyrate 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.

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## 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.

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