Recent progress on flexible nanogenerators toward self-powered systems

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Abstract
The advances in wearable/flexible electronics have triggered tremendous demands for flexible power sources, where flexible nanogenerators, capable of converting mechanical energy into electricity, demonstrate its great potential. Here, recent progress on flexible nanogenerators for mechanical energy harvesting toward self-powered systems, including flexible piezoelectric and triboelectric nanogenerator, is reviewed. The emphasis is mainly on the basic working principle, the newly developed materials and structural design as well as associated typical applications for energy harvesting, sensing, and self-powered systems. In addition, the progress of flexible hybrid nanogenerator in terms of its applications is also highlighted. Finally, the challenges and future perspectives toward flexible self-powered systems are reviewed.

KEYWORDS
flexible systems, hybrid nanogenerator, mechanical energy harvesting, piezoelectric nanogenerator, self-powered system, triboelectric nanogenerator

1 | INTRODUCTION

With the increasing issues of the energy crisis, global warming, and environmental pollution, it is essential to explore renewable energy resources to fulfill the growing demands of the human being to maintain sustainable development of the society. Green energy sources, including solar, wind, biomass, and hydrogen have been used as alternative energy sources for traditional fossil fuels. Mechanical energy, as another energy form that widely exists in our environment, such as water flow, wind, vibrations, and human motions, is abundant, clean, and sustainable, but usually being wasted in our daily life. Since 2006, the emergence of zinc oxide (ZnO) nanowire-
based nanogenerators have demonstrated for efficient conversion of mechanical energy into electricity,\(^1\) which has drawn great attention among researchers that various nanogenerators were developed ever since then. With the main focus on mechanical energy harvesting, they are broadly classified into two categories, namely piezoelectric nanogenerator (PENG) and triboelectric nanogenerators (TENG), which are based on their working principles of piezoelectric and triboelectric effects, respectively. Hybrid nanogenerators (HBNG) mainly refer to nanogenerators with the combination of these two and/or other effects such as thermoelectric, pyroelectric, etc.

Nowadays, the blossoming of portable and wearable electronics has triggered the tremendous desires of flexible power sources, where the traditional batteries show limitations of lifespan, disposable issue, and safety problems.\(^2\) However, the dramatic advances of nanogenerators have demonstrated their promising potential as efficient power supplies for the next-generation electronics, owing to the merits of flexibility, environmentally friendliness, cost-effectiveness, and high output.\(^3,4\) In the past decades, great efforts have been devoted by researchers to develop nanogenerators with high output and high conversion efficiency from materials’ and structures’ points of view, with the applications as flexible/wearable power sources toward self-powered systems for various sensing applications.

Here, this review focuses on the recent development of flexible mechanical energy harvesters, including piezoelectric and TENGs through three aspects, namely basic operating mechanisms, materials, and structural designs as well as applications. Besides, HBNGs toward their recent applications are also highlighted. To differentiate from previously published reviews on flexible nanogenerators,\(^5,6\) this article mainly covers the progress in recent 5 years.

2 | FLEXIBLE PENG

Since the first piezoelectric related work was reported in 1880 by the brothers Pierre Curie and Jacques Curie,\(^7\) piezoelectric materials have attracted great attentions.\(^8-10\) The unique properties and exceptional advantages of piezoelectric materials allow broad of applications in the fields of robotics and metrology.\(^11-13\) Recently, soft, and skin-integrated electronics, sometime also known as “epidermal electronics” have been of great interest due to their flexibility, stretchability, and lightweight.\(^14-17\) In the past several years, integration of functional materials into a flexible platform has become a hot research topic.\(^18-21\) Owning to the advances in materials science and mechanics, construction of piezoelectric materials in flexible and stretchable formats provides the foundations for numerous applications in wearable and implantable systems.\(^22-25\) The piezoelectric coefficient \(d_{33}\), ratio of open circuit charge density to applied stress (in unit of \(CN^{-1}\)), is typically used to quantify the performance of piezoelectric materials. Among the existing piezoelectric materials, lead zirconate titanate (PZT) has been widely applied as the active materials of self-powered electronics for its high \(d_{33} \sim 130 \text{ pmV}^{-1}\).\(^26\) However, high lead concentration in PZT renders the fast development of new lead-free piezoelectric materials, such as barium titanate (BaTiO\(_3\)), sodium niobite (NaNbO\(_3\)), potassium niobite (KNbO\(_3\)), and ZnO.\(^1,20,27-29\) In the past decades, explosive researches and efforts have been devoted to self-powering technologies that convert mechanical energies to electricity. Currently, flexible piezoelectric-based nanogenerators have been recognized as a competitive energy harvesting approach for their high power density output.\(^8-10\) This section highlights these advances, with an emphasis on operating mechanisms of piezoelectricity, following unusual mechanical attributes and newly developed materials. Various applications derived from those foundations demonstrate the potential of PENG for energy harvesting.

2.1 | Operating mechanisms of piezoelectricity

Piezoelectric materials that convert mechanical deformations to electricity are labeled as a direct piezoelectric effect, while inducing a mechanical strain by an applied electrical field is labeled as converse piezoelectric effect. The piezoelectric effect is governed by the piezoelectric constitutive equation\(^30\) as follows:

\[
\begin{bmatrix}
\delta \\
D
\end{bmatrix} = \begin{bmatrix}
s & d \\
 d & \epsilon^T
\end{bmatrix} \begin{bmatrix}
\sigma \\
E
\end{bmatrix}
\]

(1)

where \(\delta\) and \(\sigma\) represent strain and stress; \(D\) and \(E\) refer to electric displacement and electric field; \(s\), \(\epsilon\), and \(d\) are the elastic compliance, the dielectric constant, and the piezoelectric coefficient, respectively; \(E\) and \(T\) are evaluated at the constant electric field and constant stress, respectively; and \(t\) represents the transpose. Originated from the electric dipole moments, piezoelectric effect is partially induced by ions on crystal lattice sites (like ZnO), or by molecular groups directly (like cane sugar).\(^31\) To clearly describe the operating mechanisms of piezoelectricity, ZnO crystal of wurtzite-structured is selected as an example, as shown in Figure 1.\(^8\) Figure 1A presents...
the tetrahedrally coordinated Zn\(^{2+}\) and O\(^{2-}\) that accumulate along the c-axis. Figure 1B shows the working principle of piezoelectricity induced by ZnO as an external force applied on it. At the initial state, positive and negative charge centers cohere with each other. When an external stress (compressing or stretching) is applied on ZnO, charge centers of anions and cations separate, resulting in an electrical potential, as shown in Figure 1B. The operating mechanism of other widely applied piezoelectric materials can be also explained as mentioned above.

2.2 | Piezoelectric materials and structural design

Since the first report about the piezoelectric material,\(^7\) many researchers have put remarkable efforts to develop innovative piezoelectric materials, generally being divided into two types, inorganic and organic materials. Compared to the organic piezoelectric materials, inorganic piezoelectric materials exhibit higher piezoelectric constant and coefficient, and thus are widely used for energy harvesting and mechanical sensing. Traditionally, inorganic piezoelectric materials are divided into two parts: piezoelectric crystals and piezoelectric ceramics.\(^{32-34}\) Piezoelectric crystal, like ZnO nanowires,\(^{33}\) have a single crystal structure with natural piezoelectricity. Unlike piezoelectric crystals, piezoelectric ceramics, such as Ba(Zr\(_t\)Ti\(_{1-x}\))O\(_x\) (BZT)\(^{35}\) and PZT,\(^{36}\) only show piezoelectricity after poling processes, because of their random dipole orientations before poling. However, the brittle and rigid nature of inorganic ceramics limits their applications in flexible electronics. To solve this problem and improve the flexibility and stretchability, these rigid materials have been developed into thin film, nanowires, nanoparticles, and nanofibers, as shown in Figure 2.\(^{37-42}\)

Referring to the deposition method for those piezoelectric thin film or nanocomposite, sol-gel chemistry is no doubt the best route.\(^{43}\) Taking the most used piezoelectric material, PZT for example,\(^{44-46}\) precursors of lead, zirconium, and titanium are mixed together and dissolved in solvent. The mixed solution is spin-coated on substrates to form a layer of thin film after hydrolysis, followed up with pre-baking for organic residual removal. To achieve the desired thickness of the film, this process can be repeated multiple times. Eventually a high temperature annealing is performed on the film to form the crystal structures. Besides the solution processing deposition method, physical vapor deposition, for example sputtering, is another deposition method to form thin film of piezoelectric ceramics.\(^{47}\) Despite the low-processing temperature of physical vapor deposition compared to sol-gel chemistry, the deposition duration and chamber space restrict the film thickness and film area.\(^{48}\) In addition, the physical vapor deposited films usually suffer from poor dielectric constant and coefficients, compared to solution-processed films.\(^{47}\) Both organic and inorganic-based piezoelectric materials are able to be formed as nanoscale fibers by particular technique, such as electrospinning. The fiber-based piezoelectric composites show great dipole orientation and separation and therefore afford improved piezoelectric effects.\(^{49}\) Meanwhile, the fiber-based piezoelectric materials also show good flexibility under various mechanical strains.\(^{50}\) Transfer printing, as a key success to flexible piezoelectric ceramics, involves a liftoff process of the piezoelectric thin film that cast and cured at high temperature on oxidized silicon wafer, where silicon dioxide (SiO\(_2\)) serves as
a pre-buried sacrificial layer (removed by chemical wet etching). A pre-buried sacrificial layer (removed by chemical wet etching).51 Another liftoff process of piezoelectric thin films called laser liftoff is associated with exposures to high energy laser for inducing interface failure between PZT film and sapphire substrate, and thus for transfer printing onto flexible substrates.52

![FIGURE 2 Scanning electron microscope (SEM) images of inorganic materials with various structures. Image of (A) PZT thin film, Copyright 2019, Elsevier, (B) PZT nanowires, Copy 2019, IOP Publishing, (C) PZT nanoparticles, Copyright 2016, IOP Publishing, (D) PZT nanofibers, Copyright 2019, American Chemical Society, (E) BaTiO3 thin film, Copyright 2019, Wiley-VCH, and (F) BZT nanoparticles, Copyright 2019, Elsevier.](image-url)
Organic-based piezoelectric materials have been considered as a great candidate flexible electronics due to their inherent flexibility. Polyvinylidene difluoride (PVDF) is the most widely used organic-based piezoelectric material that has five different crystal structures: $\alpha$, $\beta$, $\delta$, $\gamma$, and $\epsilon$. The $\beta$ crystal phase is an electroactive phase which is mainly responsible for piezoelectricity. Therefore, generating stable state of $\beta$ phase in the polymers can lead to better piezoelectric effect. According to the previous reports, various structures of PVDF and its copolymers have been developed, including fiber arrays, nanowires, thin film, and nanotube arrays. Thin film poly(vinylidene fluoride-co-trifluoroethylene) (P[VDF-TrFE]) owns a significant value for large-area deposition, excellent uniformity, and remarkable surface morphology. However, the piezoelectric constant of the fiber-based composites exhibits about four times greater than the controlled thin film-based ones. Additionally, PVDF in the format of woven textiles has an exceptional advantage in wearable applications.

A key challenge to realize flexible nanogenerator is the development of strategies in mechanics that allows large elastic mechanical deformations and high areal coverages of active devices, even integrating with highly rigid and brittle materials. Therefore, optimizing device structure to enhance the flexibility and power density is desirable. Common piezoelectric-based energy generators adopt sandwich structures, including a functional layer (piezoelectric thin film) with two electrode layers attached at bottom and top, as shown in Figure 3A. However, the large electrode area limits mechanical deformations, and thus leads to poor stretchability. To increase the stretchability of the flexible generators, in-plane electrodes can be considered as a great solution. Compared to the sandwich structure, in-plane electrode structure has advantages of thinner thickness, simple fabrication process, and much greater stretchability. Recently, we reported a skin-integrated rubbery electronics with in-plane electrode structure (Figure 3B) that can survive under various mechanical displacement and rotational boundary condition: stretching up to 15.2%, bending over 160° at a radius of ~30 mm, and twisting 90°. The overall thickness of such generator is less than 1.2 mm, affords great stretchability. Another popular approach involves an island-bridge device design, in which the working area of the electronics at the “islands,” and the electrical interconnects (like serpentine and ribbons) form the “bridges.” Based on the island-bridge layout design, the rigid working areas undertake negligible mechanical deformations whereas the interconnections provide the majority of strain level. In-plane mechanical design of electrodes, such as serpentine shape, is another powerful method, which allows the stretchability increases up to hundreds of percentage.

2.3 | Applications

PENGs show the great potential in various fields, ranging from microelectromechanical systems to biomedical engineering. Recently, immense research efforts have been
made in the application of piezoelectric systems in the biomedical engineering. Rogers's team reported a thin flexible electronics integrating PZT nanoribbons that can be mounted on the surface of internal organs for power generating from mechanical deformations of the organs (Figure 4A). The energy harvester consisted of multiple PZT nanoribbons connected in series to increase the output voltage. The maximum strain level in these PZT ribbons is under 0.1% at a bending radius of 2.5 cm, exhibiting excellent flexibility. Owing to the advances in the materials science and engineering, the PZT-based devices exhibit great open-circuit voltage and short-circuit current, and the voltages and currents increase frequency with the same load amplitudes. Output voltage can reach up to ~3.7 V under a strain of 0.35%. The voltage of a micro battery, charged by the device under 7500
cyclic bending load, saturates at a value of 3.8 V, indicating an electrical energy of ~0.164 J. Additionally, the time-averaged power density of five devices can reach as large as 1.2 μW cm⁻², sufficient to power a cardiac pacemaker. Besides the reported PZT-based device, other similar nanogenerators have also been demonstrated to harvest energy from internal organs of animals.73

Recently, we reported a skin-integrated device incorporating PZT/polydimethylsiloxane (PDMS)/graphene composite for mechanical sensing and energy harvesting, as shown in Figure 4B.65 Owning to the low elastic modulus of the composite and PDMS (as encapsulation layer), the flexible electronics can stretch up to ~15.2%, bend over 160° at a radius of ~30 mm, and twist 90°. Moreover, the self-powered nature in the piezoelectric materials allowed energy harvesting from fist striking, with an open-circuit voltage and short-circuit current of 3 V and 1 μA, respectively. The voltage of 1 nF capacitor, charging by the PZT rubbery device, can reach to 1 V under repeated mechanical beating. To harvest the mechanical energy at human joint areas, the device is mounted onto a wrist, reaching to a maximum voltage of 0.8 V and a current of 58 nA at a bending angle of 15° and a frequency of 4 Hz. As attaching the energy harvester onto a human heel, the device can yield an average voltage and current of 5 V and 52.9 μA, contributing a remarkable power density of 972.43 μW cm⁻². Besides the PZT rubbery device, there are many nanogenerators based on piezoelectric materials incorporating PDMS, exhibiting excellent ability in harvesting energy from human daily motions.20,74,75

In 2014, Lee et al reported the fabrication of large-area PZT thin films via laser liftoff technique. The commercialized liftoff technique enables a high-quality piezoelectric thin film transfer from bulk sapphire substrates to plastic substrates by XeCl excimer laser. As shown in Figure 4C,52 the lightweight and flexible device based on the PZT film yield an open-circuit voltage and short-circuit current of 200 V and 1.5 μA under periodical bending and unbending motions with a strain of ~0.386% at a straining rate of ~2.32% s⁻¹. Moreover, these devices were robust with the test of 9000 bending cycles. As the PZT thin film nanogenerator is connected to an external load resistance (200 MΩ), the instantaneous power density reaches up to 17.5 mW cm⁻². To further demonstrate the energy conversion from biomechanical movements, a large-area PZT thin film nanogenerator (5 × 5 cm²) can yield a high current of ~8.7 μA under irregular and slight bending motions by a human finger. The nanogenerator can directly light up 105 commercial light emitting diode (LED) arrays without rectifier and charge circuit under slightly bending by a human finger. Other PENGs based on an inherently high piezoelectric perovskite thin film on a plastic substrate are reported.26,27,76 Among these attempts, a thin film PENG can output an extremely high power density (~7 mW cm⁻³) under periodic mechanical deformations.27

(Na₀.₅K₀.₅)NbO₃ (KNN) is another popular piezoelectric material, due to its high piezoelectric coefficient, large electromechanical coupling factor and good heat stability. Recently, a flexible nanogenerator based on Ag/KNN, multi-walled carbon nanotubes, and PDMS composite was reported by Wang et al (Figure 4D).68 Additional conductive materials are introduced to enhance the degree of polarization of the piezoelectric material (KNN), resulting in ultrahigh open-circuit voltage (~240 V) and short-circuit current of ~23 μA under external mechanical stress of 0.1 MPa. Due to the flexibility of composite layer, the output voltage keeps stable after over 1000 times cycling. Nine white LEDs can be illuminated by the device without any external storage. Figure 3D shows the output performance as the device was deformed by finger bending, an open-circuit voltage of ~0.75 V. Under slight foot squeezing, the open-circuit voltage by the device can reach up to ~35 V. To further enhance the polarization degree of piezoelectric materials, many researchers attempt to add conductive materials into the piezoelectric composites, like carbon nanotubes.77 The nanocomposite generator based on PZT and carbon nanotubes can yield an output voltage and current of ~100 V and ~10 μA, powering 12 commercial LEDs.

ZnO is an excellent candidate for PENG. In 2011, Kim et al reported a ZnO nanorod-based nanogenerator with the first use of cellulose paper as a substrate for its thermal stability, as shown in Figure 4E.69 The average diameter and height of ZnO nanorods were 80 nm and 2 μm, respectively. The output current density was up to 2 μA cm⁻², under the applying external force of 0.8 kgf. Benefited from the thermal stability of cellulose paper, the current output by the device was very stable in a broad temperature range and up to 200°C. To increase the output power density for energy converters, a new single crystalline (1-x)Pb(Mg₁/₃Nb₂/₃)O₃-xPbTiO₃ (PMN-PT) has been reported, exhibiting an extremely high piezoelectric charge constant of d₃₃ up to 2500 pC N⁻¹ (Figure 4F).70 Single crystalline rhombohedral PMN-PT ingots were grown directly from the melt by a modified Bridgman method near the morphotropic phase boundary. Periodic bending/unbending motions of the PMN-PT based flexible PENG yielded the output voltage and current of 8.2 V and 145 μA (~411.42 μW cm⁻²). Under 30 000 continuous bending cycles at a radius of 16.5 mm, the output current by the device presents negligible degradation, demonstrating its mechanical and electrical stability. Connecting the device to a coin battery through a
rectifier bridge, the voltage of the battery saturates at 1.7 V in 3 hours. As mounting the device on the heart of a rat, the generated energy (2.7 μJ) from the heartbeat is sufficient to trigger the action potential for artificially contracting the heart.

In a recent report, a new piezoelectric material, boron nitride nanotubes (BNNTs), was developed by Sung et al that exhibited excellent electrical, mechanical, and thermal properties (Figure 4G). Benefit from the characteristics of the piezoelectric material, the transparent, flexible, and biocompatible device can be utilized in various applications in energy harvesting. The output voltage and current of the reported generator were 22 V and 75 nA, contributing an output power density of 106 μW cm$^{-3}$ under an external force of 80 kgf. Under 36 000 periodic mechanical beating cycles, the output voltage by the energy harvester still exhibited good stability. Owning the instinctive nature of the piezoelectric material, the device with a large working area ($7 \times 6.5 \times 0.02 \text{ cm}^3$) was attached to a cellphone display, acting as a touch sensor, and the open-circuit voltage was 4 V under the finger touch. As attached to the different parts of human body, the output voltage can stabilize at 2.5 V for the foot, 1.98 V for the elbow, 0.48 V for the neck, 0.75 V for the wrist, and 1.05 V for the knee under different human movements. Up to now, many reports about the BNNTs have been published, presenting its good high-temperature resistance and piezoelectric capabilities.

PVDF is a typical organic piezoelectric material with inherent flexibility. Many researchers have made much effort to utilize PVDF with various formats in harvesting mechanical energy. It is reported that a PENG based on PVDF nanofibers can yield ~6.5 μW with an external resistance load of 5.5 MΩ. Recently, a PENG based on PVDF that incorporated with hygroscopic rare earth ytterbium salt (Yb-PVDF) was developed by Mandal et al, as shown in Figure 4H. The device could generate a voltage output of ~85 mV at continuous bending, and ~70 mV during repeated twisting. Additionally, the device can produce output voltage of 100–200 mV under the sound pressure ranging from 80 to 100 dB, illustrating its high sensitivity in detecting minuscule stress. As an energy harvester, an open-circuit voltage of ~7 V was measured by connecting a full wave bridge rectifier during finger imparting (Figure 3H). Connecting the device to an external resistance load (~20 MΩ), an instantaneous output power density of 1 μW cm$^{-2}$ is achieved, sufficient to power 50 blue LEDs. More reported PENGs can be found in Table 1.

3 | FLEXIBLE TENG

Triboelectric effect, a contact-induced electrification phenomenon occurs ubiquitously in our daily life, is normally regarded as a negative effect. Since 2012, the invention of TENG has made full utilization of this effect, converting mechanical energy in the ambient environment into electricity, such as vibrations, wind, water waves, and human motions. Owing to various advantages of TENG, such as a broad range of material selection, simplicity of structure design, cost-effectiveness, and high output, TENG has been intensively explored in the application of flexible power sources and self-powered environmental sensors, which exhibits the great potential for the future development of wearable electronics and the Internet of Things.

3.1 | Operating mechanisms of trboelectricity

Figure 5A demonstrates the construction of the first flexible TENG with vertical contact-separation (CS) mode. The basic working principle of TENG is through a conjunction of triboelectrification and electrostatic induction, which is illustrated in Figure 5B in detail. Generally, a TENG consists of two tribo-materials with different electronegativity, and when the two materials are brought into physical contact, opposite static charges with an equal amount will be generated on the surfaces due to the contact electrification (Figure 5B[II]). Once the two materials are separated, the electrostatic induction induces charges on the two electrodes at the back of two materials (Figure 5B [III]); consequently, a potential difference is established, resulting in electron transfer through the external circuit until an equilibrium state is reached when the two parts are fully separated (Figure 5B [V]). When the two materials approach to each other again, electrons flow in a reverse direction through the external circuit to neutralize the opposite charges on the electrode (Figure 5B [VI]), thus, upon the CS cycles, alternative current signals are generated.

3.2 | Materials and structural design

3.2.1 | Materials

When the two dissimilar tribo-materials are at the two ends of the triboelectric series, higher output is yielded, which is due to the largest difference of electronegativity of these two materials. Thus, from the material point of view, the electron affinity, surface function groups, as well as work function are important factors affecting the output performance of TENG. A vast number of advanced materials have been explored, including
Due to the uniqueness of mechanical flexibility, chemical stability, and high electron mobility, graphene has been regarded as a promising candidate for energy-related applications. Recently, a crumpled graphene (CG)-based TENG was reported as shown in Figure 6A.108 The CG layer was formed on pre-strained VHB tape (3M, VHB 4910), endowing its high stretchability and flexibility. The output performance of the device was enhanced with the increase of the crumple degree, which is ascribed to the increased contact area, surface roughness, and larger work function difference. Another graphene-based stretchable TENG is reported as a self-powered touch sensor with an overall thickness around 18 μm (Figure 6B). It can conformally contact on the uneven surface of the human palm and the sensitivity was 0.274 V kPa$^{-1}$ in the pressure range of 1–40 kPa.109

MXenes, a recently emerging family of two-dimensional materials, whose electrical and mechanical properties can be tuned by adjusting the composition and surface functional groups such as −O, −OH, and −F, was found more tribo-negative than polytetrafluoroethylene (PTFE).110 Dong et al reported that a layer of MXenes (Ti$_3$C$_2$Tx) film fabricated by spray coating its aqueous suspension on glass had a neutral state when contacting against PTFE, while a voltage of 650 V can be delivered when contacted with polyethylene terephthalate (PET) (Figure 6C). This is mainly because Ti$_3$C$_2$Tx has the same terminal groups with PTFE.110 Flexible TENG can be realized by coating the MXenes on flexible substrates, such as ITO/PET. Furthermore, an all electrospun TENG was fabricated comprising poly(vinyl alcohol) (PVA) and Ti$_3$C$_2$Tx composite nanofiber film and silk nanofibers film, which can output 1087 mW m$^{-2}$ power density.120

### Table 1

<table>
<thead>
<tr>
<th>Materials</th>
<th>Type</th>
<th>$d_{33}$</th>
<th>Performance</th>
<th>Power density</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>ZRS-PNG</td>
<td>Composite</td>
<td>NA</td>
<td>An output voltage of −85 V and a short-circuit current of −2.2 μA under repeating bending and relaxation with a frequency of −4 Hz</td>
<td>35.62 μW cm$^{-2}$</td>
<td>82</td>
</tr>
<tr>
<td>Annealed La-doped ZnO-PDMS</td>
<td>Composite</td>
<td>NA</td>
<td>A p-p voltage of 23 V and a p-p current of 150 nA at 2 N force</td>
<td>1.53 μW cm$^{-2}$</td>
<td>83</td>
</tr>
<tr>
<td>Li-doped ZnO-PDMS</td>
<td>Composite</td>
<td>−15 pmV$^{-1}$</td>
<td>The highest output of −20 V and −18 μA cm$^{-2}$ under finger bending conditions</td>
<td>5.33 mW cm$^{-2}$</td>
<td>82</td>
</tr>
<tr>
<td>ZnO/Spiro-MeOTAD</td>
<td>Nanowire</td>
<td>NA</td>
<td>An output current density about 500 nA cm$^{-2}$ under vertical compressive force of 1 kgf</td>
<td>N.A.</td>
<td>84</td>
</tr>
<tr>
<td>PZT-solid silicon rubber</td>
<td>Composite</td>
<td>NA</td>
<td>The p-p voltage and current of −65 V and −1 μA under the aforementioned periodic stretching stimulation.</td>
<td>−81.25 μW cm$^{-3}$</td>
<td>44</td>
</tr>
<tr>
<td>PMN-PZT</td>
<td>Film</td>
<td>1527 pCN$^{-1}$</td>
<td>Open-circuit voltage and short-circuit current of 100 V and 20 μA, respectively, through biomechanical bending and unbending motions</td>
<td>333.33 μW cm$^{-2}$</td>
<td>85</td>
</tr>
<tr>
<td>PZT particles-Cu@Ag branch</td>
<td>Nanofibers</td>
<td>NA</td>
<td>The p-p voltage and current of 61 V and 1.1 μA under stretching and releasing deformations (strain of 50%, strain rate of 6.4 cm s$^{-1}$ and frequency of 0.7 Hz)</td>
<td>22.36 μW cm$^{-3}$</td>
<td>84</td>
</tr>
<tr>
<td>KNN-BTO-PDMS</td>
<td>Composite</td>
<td>NA</td>
<td>An electrical output of 58 V and 450 nA</td>
<td>2.9 μW cm$^{-2}$</td>
<td>75</td>
</tr>
<tr>
<td>PVDF-KNN</td>
<td>Composite</td>
<td>NA</td>
<td>An electrical output of 3.7 V and 0.326 μA</td>
<td>N.A.</td>
<td>86</td>
</tr>
<tr>
<td>PVDF-MAPbI$_3$</td>
<td>Composite</td>
<td>NA</td>
<td>An electrical output of 17.8 V and 2.1 μA cm$^{-2}$ under an applied mechanical force 50 N</td>
<td>37.38 μW cm$^{-2}$</td>
<td>87</td>
</tr>
<tr>
<td>P(VDF-TrFE)-BNNTs</td>
<td>Composite</td>
<td>14 pCN$^{-1}$</td>
<td>The output voltage and current of 22 V and 640 nA and a sensitivity of 55 V/MPa under the pressure of 0.4 MPa</td>
<td>11.3 μW cm$^{-2}$</td>
<td>18</td>
</tr>
<tr>
<td>BSFTO-PDMS</td>
<td>Composite</td>
<td>18 pCN$^{-1}$</td>
<td>An electrical output of 16 V and 2.8 μA under the vertical force of 35 N</td>
<td>3.11 μW cm$^{-2}$</td>
<td>19</td>
</tr>
</tbody>
</table>

Graphene, MXenes, hydrogels, aerogels, black phosphorus (BP), etc.

Due to the uniqueness of mechanical flexibility, chemical stability, and high electron mobility, graphene has been regarded as a promising candidate for energy-related applications. Recently, a crumpled graphene (CG)-based TENG was reported as shown in Figure 6A.108 The CG layer was formed on pre-strained VHB tape (3M, VHB 4910), endowing its high stretchability and flexibility. The output performance of the device was enhanced with the increase of the crumple degree, which is ascribed to the increased contact area, surface roughness, and larger work function difference. Another graphene-based stretchable TENG is reported as a self-powered touch sensor with an overall thickness around 18 μm (Figure 6B). It can conformally contact on the uneven surface of the human palm and the sensitivity was 0.274 V kPa$^{-1}$ in the pressure range of 1–40 kPa.109

MXenes, a recently emerging family of two-dimensional materials, whose electrical and mechanical properties can be tuned by adjusting the composition and surface functional groups such as −O, −OH, and −F, was found more tribo-negative than polytetrafluoroethylene (PTFE).110 Dong et al reported that a layer of MXenes (Ti$_3$C$_2$Tx) film fabricated by spray coating its aqueous suspension on glass had a neutral state when contacting against PTFE, while a voltage of 650 V can be delivered when contacted with polyethylene terephthalate (PET) (Figure 6C). This is mainly because Ti$_3$C$_2$Tx has the same terminal groups with PTFE.110 Flexible TENG can be realized by coating the MXenes on flexible substrates, such as ITO/PET. Furthermore, an all electrospun TENG was fabricated comprising poly(vinyl alcohol) (PVA) and Ti$_3$C$_2$Tx composite nanofiber film and silk nanofibers film, which can output 1087 mW m$^{-2}$ power density.120
Recently, hydrogels as ideal ionic conductors have been widely studied as efficient current collectors in TENG applications. For example, Wang et al reported hydrogels with hybrid electronic/ionic conductivity consisting of silver nanowires and chitosan (CS) cross-linked by metal ions (Ag+/Cu2+) as current collectors for harvesting human motion energy (Figure 6D).\textsuperscript{115} The output performance of the TENG was associated with the concentration of silver nanowires as well as the complexation type of metal ions. In addition, ionogel-based TENG was also reported with a wider application temperature range ($-20^\circ\text{C}$–$100^\circ\text{C}$) comparing to that of hydrogel-based.\textsuperscript{121} Moreover, cross-linked polyethyleneimide (PEI)/PVA was developed by researchers as positive tribo-materials, and the composite-based TENG shows a much higher output when embedding Au nanoparticles in the polymer matrix, which was due to a 2.4-fold enhancement of dielectric constant of the composite bulk.\textsuperscript{122}

Due to the highly porous structures and super lightweight, aerogels have been developed in various applications. Mi et al reported a cellulose nanofibrils/PEI porous aerogel serving as positive material and in contact with PVDF in TENG\textsuperscript{117} as shown in Figure 6E. The output power density of the device improved 14.4 times due to the enhanced tribo-positivity. In addition, other materials

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure5.png}
\caption{Construction and working principle of the first flexible triboelectric nanogenerator (TENG). A, The construction and digital photos of the flexible TENG.\textsuperscript{88} Copyright 2012, Elsevier. B, Working principle of the flexible TENG.\textsuperscript{93} Copyright 2012, American Chemical Society}
\end{figure}
are also reported as the various choices of tribo-materials or electrodes for TENG application. For example, cashmere-based TENG,\textsuperscript{123} modified textile-based TENG by BP, and cellulose-derived hydrophobic nanoparticles (Figure 6F),\textsuperscript{119} wrinkled PEDOT: PSS film,\textsuperscript{124} ultrathin paper-based self-powered system,\textsuperscript{125} patterned Ag nanofiber electrodes,\textsuperscript{126} and so forth. A summary of the output characteristics of TENGs based on various materials was listed in Table 2.

### 3.2.2 Structural design

Basically, TENG has four working modes, including CS mode, lateral sliding mode, single-electrode (SE) mode, and sliding freestanding triboelectric-layer (SFT) mode.\textsuperscript{3} The basic structure for all these four working modes composes at least one pair of triboelectric surfaces as well as two electrodes. For SFT mode, there are two pairs of triboelectric surfaces and the ground serves as the other electrode for SE mode.\textsuperscript{103} A lot of structures have been designed in line with these four modes. Here, we mainly review the representative structures of each working mode.

The CS mode is vastly been studied due to its simple structure, easy fabrication, and high instantaneous power density. Surface functionalization,\textsuperscript{133,134} micro/nanostructure patterning,\textsuperscript{135,136} and bulk property engineering\textsuperscript{137,138} of the tribo-materials are frequently adopted by researchers to improve the output power. In the meantime, a lot of new designs have emerged. Figure 7A shows an ultraflexible three-dimensional TENG (3D-TENG) fabricated by hybrid 3D printing technique.\textsuperscript{139} This 3D-TENG uses printed resin composites as the tribo-electrification layer and ionic hydrogel as electrodes. Each unit undergoes CS cycles when the entire structure...
is deformed and released by an external force. Also, it is found that the output performance is associated with compression ratio, the structure, and parameters \((D, d)\) of the unit. A higher compression ratio enhances the effective contact surface area, endowing the increase of electrostatic charges per unit volume. Besides, the increase of \(D\) and \(d\) in each unit largely decreases the distance \(h\), which results in decreased output voltage while the output current and power are in an opposite trend, owing to the improvement of effective contact area. This 3D-TENG can produce a peak power per unit volume of 10.98 W m\(^{-3}\) under a low frequency of 1.3 Hz. Similarly, knit\text{-}ed fabric/textile-based TENGs usually adopt the CS mode.\textsuperscript{144,145}

Recently, an interesting snow-based TENG was developed for harvesting energy in harsh snowy environment with triboelectrification of snow as shown in Figure 7B.\textsuperscript{140} They used a 3D printing technique for the triboelectric layer of silicone and the electrode of poly(3,4-ethylene dioxythiophene)-poly(styrene sulfonate) (PEDOT: PSS) in a single electrode mode. The snow, as crystallized water, carries positive charges and will have contact electrification with silicone when falling or sliding on the surface of silicone. Consequently, the device generates an instantaneous power density of 0.2 mW m\(^{-2}\). Besides, the device shows the potential application as an arctic weather station for measurement of various meteorological parameters, including snowfall rate/direction, wind speed, and snow depth. Similar structures are frequently employed for wind energy and rain\-drop energy harvesting.\textsuperscript{146}

TENGs with SFT structure can harvest energy from moving objects while the system can still mobile without grounding wires,\textsuperscript{147} which is suitable for harvesting rotating motion energy, human walking, and air flow, etc. Guo et al reported a TENG consisting of checker-like interdigital metal electrodes (aluminum [Al]) and a sandwiched PET film, which can convert translational kinetic energy in all directions\textsuperscript{141} as shown in Figure 7C. When the PTFE squares and acrylic of the sliding pane move in certain direction, electricity is generated upon the separation and contact. The sandwiched PET film between the sliding panel and metal electrodes can be worn freely, thus, the operation lifetime of the device is greatly increased. The resultant TENG can yield an open-circuit voltage of 210 V in the \(X\) or \(Y\) sliding direction, a maximum output power density of 1.9 W m\(^{-2}\) and has the advantages of light weight, durable, flexible to fold, and portable.

To further enhance the output performance, various stacked-layer TENGs are proposed. Figure 7D shows a concept of electrical muscle stimulation powered by a zigzag TENG. In this TENG, a PET layer is firstly folded in a zigzag structure and Al layers are stacked to each surface of the folded PET layer as electrodes. PTFE serving as a triboelectric layer against Al film is assembled on top of another counterpart Al film. Finally, the stacked TENG is wrapped by another PET layer to confine the

| Table 2 | Summary of output characteristics of triboelectric nanogenerators (TENGs) with different triboelectric materials |
|---|---|---|---|---|
| Materials | Working mode | Open-circuit voltage and short-circuit current (density) | Power density | Reference |
| Crumpled graphene | CS | Contact against PDMS, 83 V, 25.78 \(\mu\)A | 2.5 W m\(^{-2}\) | 108 |
| \(\text{Ti}_3\text{C}_2\text{T}_x\) | CS | Contact against paper, ~119 V, ~22 \(\mu\)A | ~609.1 mW m\(^{-2}\) | 111 |
| Au/PEI/PVA | CS | Contact against PET, 161.1 V, 20 mAm\(^{-2}\) | 17.73 W m\(^{-2}\) | 122 |
| Porous CTS/PI aerogel | CS | Contact against porous PDMS, 60.6 V, 7.7 \(\mu\)A | 2.33 W m\(^{-2}\) | 116 |
| Paper | CS | Contact against FEP, 90 V, 6 mAm\(^{-2}\) | 285.6 mW m\(^{-2}\) | 125 |
| Textile modified with black phosphorus | SE | Contact against skin, ~1860 V, 1.1 \(\mu\)A cm\(^{-2}\) | 5.2 W m\(^{-2}\) | 119 |
| Zeolitic imidazole framework (ZIF) | CS | Contact against Kapton, 164 V, 7 \(\mu\)A | 392 mW m\(^{-2}\) | 127 |
| Nylon | CS | Contact against silicone rubber, 1.17 kV, 138 \(\mu\)A | 11.2 W m\(^{-2}\) | 128 |
| Polyacrylonitrile/polyamide 6 | CS | Contact against PVDF/PDMS, 540 V, 110 \(\mu\)A | 14.8 W m\(^{-2}\) | 129 |
| BNO-sulfonated polyimide | CS | Contact against PTFE, 75 V, 1 \(\mu\)A | 178.5 mW m\(^{-2}\) | 130 |
| Polytetrafluoroethylene foam | CS | Contact against PTFE, 12 V, 8.5 nA | NA | 131 |
| Graphene oxide | SE | Contact against skin, 1100 V, 55 \(\mu\)A | 3.13 W m\(^{-2}\) | 132 |
Thus, during pressing and releasing operation, the zigzag TENG can automatically return to the original position before next pressing and the mechanical deformation energy is converted to electricity with a short-circuit current of 35 μA. In addition, another stacked TENG with the open-book-like structure is reported.
by Zhong et al as shown in Figure 7E. The device can integrate multiple TENG units in a limited space and greatly enhanced the volume density. For a device with 50-unit TENG, the transferred charges and the short-circuit current are 26 μC and 0.45 mA, respectively. 143

3.3 Applications

Ever since the invention of TENG, it has been vastly explored as efficient power sources by scavenging different types of mechanical energy in the ambient environment and as self-powered sensors for various sensing applications. Comprehensive reviews about applications of TENG have been published. In this section, some representative examples will be reviewed including driving different actuators, TENG-based E-skin, motion detector, tactile mapping, chemical sensors, and so on.

Conventionally, biomechanical energy such as typing or pressing buttons is wasted in our daily life. Li et al developed an all-elastomer TENG-based keyboard cover for typing energy harvesting, as shown in Figure 8A. The keyboard cover consists of five elastomer layers, including structural elastomer (two layers), carbon black and carbon nanotubes-based elastomeric electrodes (two layers), and elastomeric dielectric layer (one layer). Through the optimization of material and structural aspects, the cover shows a high transferred charge density of 140 μC m⁻². Furthermore, the fully packaged keyboard was integrated with a supercapacitor as a self-powered system. Continuously typing the keyboard for 1 hour under normal speed, around 0.8 mJ of electricity can be stored to drive an electronic thermometer. This TENG-based keyboard cover successfully demonstrates the conversion of typing energy into electricity and is useful to further research the typing behavior of different people.

Since the high output voltage characteristics of TENG, it has been employed by researchers to drive various kinds of actuators. Recently, Li et al reported a novel application of TENG for sensitive nano-coulomb molecular mass spectrometry, as shown in Figure 8B. In this system, the total amount of ionization charges in mass spectrometry can be quantitatively regulated by the output of TENG. The authors show that plasma discharge ionization and nanoelectrospray ionization (nanoESI) are successfully induced by single-polarity or alternating-polarity ion pulses, which are generated by the high output voltage of two modes of TENG. This system opens a door for quantitative and highly sensitive mass spectrometric analysis using charge amount as well as demonstrates a facile and effective approach of TENG-driven ionization.

In addition, another application of high output voltage of TENG is for driving dielectric elastomer actuator (DEA) as reported by Chen et al (Figure 8C). Dielectric elastomers are similar with human skins in terms of low elastic modulus and large strain capability. Besides, they are actuator materials and their deformation can be controlled by an external high voltage. In this work, the authors delivered a self-powered actuation system composing of a SE TENG and a DEA. When the TENG (100 cm²) working at a CS velocity of 0.1~10 cm s⁻¹, an expansion strain of 14.5% can be induced for the DEA device with electrode diameter of 0.6 cm. The findings show its promising applications in artificial muscles and soft electronics.

Recently, soft skin-like TENGs with multiple functionalities such as high transparency and flexibility/stretchability for biomechanical energy harvesting and sensing received great interest among researchers. Figure 8D exhibits three examples of TENG-based E-skins with a sandwich structure compose of two elastomers (PDMS/VHB) and one layer of ionic hydrogel. The first skin-like TENG reported by Pu et al shows an ultra-high stretchability (1160%) and high transparency (96.2% in visible-light range). This device can yield an open-circuit voltage of 145 V and a peak power density of 35 mW m⁻². Meanwhile, the E-skin sensing pressure as low as 1.3 kPa was demonstrated. Due to the easy dehydration of hydrogels, toughly bonded elastomer/hydrogel hybrids through interfacial modification to ensure the stable mechanical and electrical performance of the E-skin device was reported by Liu et al. After the tough interfacial bonding, the dehydration of the hybrids is greatly alleviated with an average dehydration decreases by over 73%. Furthermore, Wang et al reported a transparent and flexible polyionic-skin TENG (PS-TENG) with resistance to dehydration. The output of the PS-TENG shows no significant degradation after storage in a desiccator under vacuum (RH of 23% at 23.8°C) for 5 days. Besides, the device demonstrates the output characteristics under tapping, bending, and curling.

Alternatively, another TENG-based E-skin toward practical applications of human-machine interfaces was developed by Wang et al as shown in Figure 7E. In this work, a flexible self-powered high-resolution (5 dpi) triboelectric sensor matrices (TESM) with 16 × 16 pixels based on a SE mode TENG is implemented, which can real-time map single-point and multi-point tactile stimuli with a remarkable pressure sensitivity of 0.06 kPa⁻¹. PDMS serving as electrification layer in TESM was dry-etched to micro/nanostructure morphology to enhance the effective contact area. Besides, the side length of each pixel is 2.5 mm and more precise tactile sensing can be realized by reducing the pixel size to micron-size level. The study demonstrates the capabilities of the large-scale potential application of real-time triboelectric tactile mapping in touch sensing, motion tracking, and human-machine interfaces.

To overcome the deterioration of TENG-based human-motion detection performance, which may be
induced by repeated stretching/releasing cycles with mechanical fracture, a stretchable and durable self-powered human-motion detector was reported by Lim et al (Figure 8F).\textsuperscript{152} Au nanosheet (NS)-embedded PDMS was employed as electrode and bottom triboelectric layer with patterned PDMS serving as the counter tribo-layer. This Au NS-TENG-based sensor was demonstrated for index finger, knuckle, and wrist bending and relaxation motion detection, demonstrating good bendability, stretchability, and durability.

Lastly, TENG-based chemical sensors are explored for different chemical sensing applications. Figure 8G shows an ultrasensitive flexible self-powered NH\textsubscript{3} sensor based on polyaniline-multiwalled carbon nanotubes (PANI-MWCNTS) composite thin film driven by a CS mode TENG.\textsuperscript{154} It is found that the output of NH\textsubscript{3} sensor has a proportional correlation with the concentration of NH\textsubscript{3}. This gas sensing system shows a response of 10% at a low concentration of NH\textsubscript{3} (0.001 ppm) and 225% at a higher concentration (100 ppm). Besides, the practical sensing application is extended to testing the human exhaled NH\textsubscript{3}. This study shows the potential application of TENG-based self-powered chemical sensor for environmental detector and human kidney health sensing.

4 | FLEXIBLE HBNG

Piezoelectric and triboelectric HBNGs are widely utilized to convert raw energies into usable electricity. TENGs transform mechanical energy to electricity by a coupling effect of triboelectrification and electrostatic induction,
while PENGs convert mechanical energy to electricity by electric dipole movements. To maximize the energy conversion efficiency, a concept of HBNGs has been built up that associates with the combination of various mechanisms of electromagnetics, electrostriction, pyroelectricity, piezoelectricity, and triboelectricity. For example, hybrid tribo-piezoelectric nanogenerators can yield great electrical power derived from both triboelectric and piezoelectric effects at the same time.\textsuperscript{157} Although the electromagnetic-based generators are widely applied in daily life, the HBNGs involving electromagnetic effect are relatively bulk due to their intrinsic operation mechanism.\textsuperscript{158-160}

Herein, we review the recent works of flexible HBNGs and their applications. As shown in Figure 9A, a fully encapsulated piezoelectric-triboelectric HBNG was reported by Fuh et al, where organic piezoelectric thin film PVDF was used as the piezoelectric functional layer and the copper and printed circuit board were adopted as the triboelectric functional layers.\textsuperscript{161} This hybrid generator could yield an output voltage of ~130 V and a current of 4 μA under hands-induced mechanical deformation. The authors applied the HBNG in sports for detecting different basketball dribbling heights (Figure 9A). Figure 8B presents a self-powered sock that consisted of poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) nanowires) on the top, an electromagnetic nanogenerator (planar coil and magnet) in the middle, and a TENG (Nylon 6.6 and PTFE) at the bottom (Figure 9D). The maximum open-circuit voltage of ~75 V and short-circuit current of ~45 mA were obtained, which could be used for charging capacitors. To scavenge mechanical and thermal energies, flexible HBNGs based on piezoelectric (or triboelectric) and pyroelectric effects is also discussed here. In 2018, Long et al reported a flexible pyroelectric-piezoelectric HBNG that adopted thermoplastic polyurethane nanoparticle membrane-carbon nanotubes composites as pyroelectric materials and substrates, electrospun PVDF nanofiber membrane as piezoelectric functional layer, and an electrospun poly (3,4-ethylenedioxythiophene)-poly(styrenesulfonate)-polyvinyl pyrrolidone conductive nanofiber membrane (PEDOT:PSS-PVP) as electrodes (Figure 9E).\textsuperscript{164} The device was able to harvest energy from the human daily motions, yielding a short-current as high as 320 nA. Recently, Choi et al reported a tribo-pyro HBNG that used bismuth telluride and PDMS as the pyroelectric and triboelectric functional materials (Figure 9F).\textsuperscript{165} The n-type and p-type are connected by series. Therefore, as heat is delivered to the device by finger touching, electrons from n-type and holes of p-type will be driven to the cold side, generating current in the circuit. Meanwhile, an electrical potential was generated on PDMS and skin due to the triboelectric effect. This nanogenerator could effectively work under lower frequency (<5 Hz), and afforded a maximum power density of 3.27 μW cm\textsuperscript{-3}.

5 CONCLUSION AND PERSPECTIVE

In summary, we systematically summarize the recent progress of flexible piezoelectric and TENGs in terms of their basic working principles, materials and structural designs, and applications. Furthermore, recent development of HBNGs is also reviewed. Since the invention of piezoelectric and TENGs, substantial efforts have been devoted to enhance the output performance of the two types of nanogenerators, achieving several orders of magnitude enhancement. The advances of flexible mechanical energy harvesters have already demonstrated the great potential in the next-generation electronics. It can be anticipated that more significance will be achieved by further development of this technology and dramatic impact will be received not only on the future progress of flexible electronics but also on our daily lives.

Nevertheless, some research aspects need to be considered toward further development of flexible nanogenerators and more work should be done in tackling the potential issues and constraints. First, with more new materials developed for mechanical energy harvesting
applications, the fundamental understanding of the mechanism in terms of charge transfer will benefit the optimization of the output characteristics to maximize the energy conversion efficiency. The impact of environmental conditions on the output performance is one of the key factors that are required to be considered in order to fully understand the working mechanism. Second, in addition to flexibility, novel structural design with multifunctionalities, such as stretchability, transparency, durability, degradability, and so on, are highly desirable to meet the different working scenarios toward real applications. Besides, consideration of the effective package of the device cannot be neglected to implement an efficient flexible power source. Finally, fully integration of the flexible power sources with other flexible/wearable electronics to realize self-powered systems relies on efficient power-management systems as well as energy-storage devices.

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CONFLICT OF INTEREST
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