

Towards energy-autonomous wearables: Multimodal harvesting technologies and sustainable applications

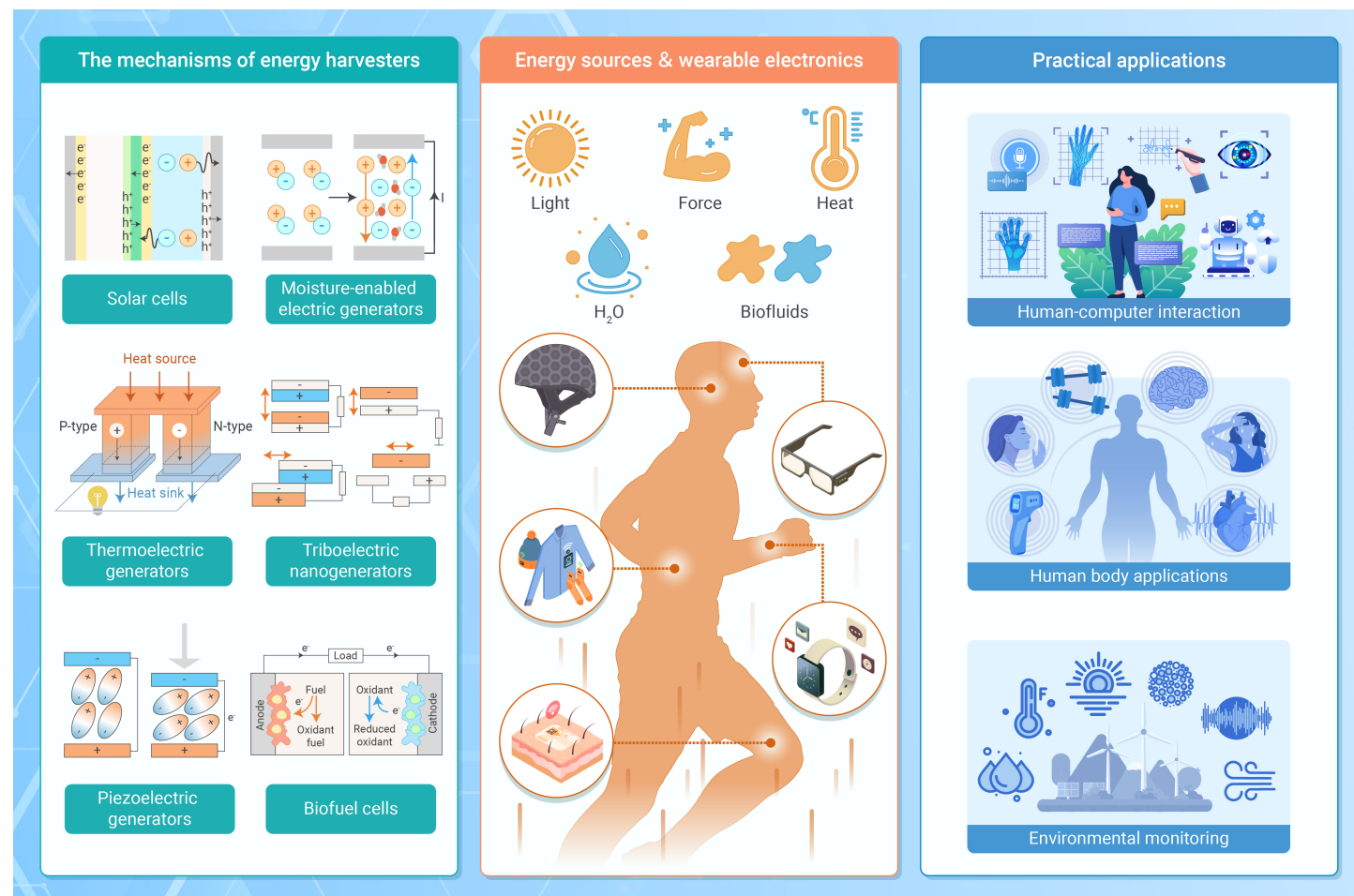
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GRAPHICAL ABSTRACT



PUBLIC SUMMARY

- This review summarizes the principles of self-powered technologies.
- The challenges and solutions faced by self-powered wearable electronics are introduced.
- The latest applications of self-powered wearable electronics are showcased in the fields.
- The development prospects of self-powered wearable devices are discussed.

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The integration of wearable electronics into daily life is revolutionizing personalized healthcare, environmental sensing, and human-machine interfaces. The global market for wearable electronics was valued at approximately \$70–80 billion in 2023. By 2029, the scale will reach \$138.5 billion. However, the reliance on rigid and short-lifespan batteries severely limit their adaptability and sustainability. Self-powered systems capable of harvesting ambient energy from both environmental sources (light and moisture) and human biomechanics (thermal gradients, motion, and biofluids) represent a paradigm shift toward autonomous and eco-friendly wearable devices. This review systematically classifies and compares six key energy harvesting technologies, i.e., solar cells, moisture-enabled/thermo-electric/triboelectric/piezoelectric generators, and biofuel cells, summarizing their working principles and analyze the issues they face when used in wearable devices, such as improving energy output, flexible manufacturing, and mechanical stability. In addition, we focus on three application scenarios: environmental monitoring, human applications, and human-computer interaction, introducing emerging applications spanning such as real-time PM2.5 detection, self-powered cardiac pacemakers, and deep-sea exploration robots, etc., demonstrating how energy autonomy intersects with global sustainability goals. This review provides a roadmap for transforming laboratory innovation into market ready solutions. Ultimately, the convergence of Adv. Mater., AI-driven design, and circular economy principles outlined here will accelerate the development of next-generation wearable electronics that seamlessly adapt to both human and environmental dynamics.

INTRODUCTION

The rapid advancement of the “Internet of Things” (IoT) has catalyzed significant interest in wearable electronic devices, which are increasingly being integrated into diverse applications such as human health monitoring,^{1–3} intelligent textiles,^{4,5} thermal management,^{6,7} and human-computer interaction interfaces, etc.^{8,9} Despite their good momentum, a major challenge impeding the broad adoption of lightweight and flexible wearable electronics is the energy supply. Traditional chemical batteries are inherently rigid while offering high energy density, which constrains the flexibility and adaptability of integrated systems.¹⁰ Furthermore, the limited lifespan of these batteries and the associated environmental concerns are at odds with principles of sustainable development.¹¹ To address these issues, self-powered technologies have emerged as a promising solution. These systems convert ambient energy sources, such as light, moisture, heat, mechanical force, and biofluids into electrical energy, thus enabling wearable devices to operate independently without reliance on conventional power sources.¹² By incorporating self-powered systems, wearable electronics can advance towards greater intelligence and miniaturization.^{13,14} Consequently, self-powered flexible wearable devices represent a highly promising direction for the future of wearable technology.

Recent efforts have led to the development of various self-powered energy systems designed to ensure continuous operation of wearable electronic products.¹² These systems often employ composite technologies that cater

to a wide range of scenarios. However, challenges remain, including high costs, complex manufacturing processes, and immature design approaches, which hinder their commercialization.¹⁵ Moreover, while some reviews have addressed specific aspects of wearable devices, comprehensive discussions on self-powered strategies, material design, and application scenarios are still lacking.¹⁶ There is a pressing need for a detailed review that consolidates progress and identifies the challenges faced by wearable self-powered electronics, covering advancements in power generation, material design, and multi-scenario applications. Overall, we need to study more in-depth self-powered mechanisms, develop more advanced material systems, and explore more diverse application scenarios to promote the development of wearable self-powered electronics and achieve large-scale production and market applications.

In this review, we provide a summary of recent advancements in wearable self-powered electronics. Self-powered strategies are classified based on their energy sources: energy harvesting from environmental sources (solar cells and moisture-enabled electric generators (MEGs)), energy harvesting from the human body (thermoelectric generators (TEGs), triboelectric nanogenerators (TENGs), piezoelectric generators (PENGs), biofuel cells (BFCs)). We first introduced and compared these six types of self-powered energy harvesters, summarized the challenges and optimization measures while they used in wearable electronic devices, such as improving output performance, flexible manufacturing, mechanical stability, etc., and then we introduced the synergetic energy harvesting system. Next, three primary application scenarios for wearable self-powered devices are reviewed: environmental monitoring, human body applications, and human-computer interaction. Finally, we identified existing challenges and pointed out possible future development directions of wearable self-powered devices, focusing on mechanisms understanding, materials development, device fabrication, systems integration, application prospects.

ENERGY SOURCE FOR THE POWER GENERATIONS

The primary function of a wearable self-powered system is to convert collected and stored energy into electrical energy.¹² This can be achieved through various means such as converting light, thermal energy, mechanical energy, bioenergy and moisture into electrical energy.^{17,18} The corresponding technologies for these energy conversions include solar cells, thermoelectric generators, triboelectric nanogenerators, piezoelectric generators, biofuel cells, and moisture-enabled electricity (Figure 1). Although the overall principles are similar in that they all aim to collect energy from the environment or human activities, the mechanisms for converting different types of energy into electrical energy vary. In this section, we categorized energy sources into those harvesting from the environment, harvesting from the human body, and harvesting from the human body-environment, then introduce the principles and latest developments (common materials, flexible manufacturing measures, methods for optimizing efficiency) of different self-powered generators under different energy sources. Finally, we pointed out the challenges and prospects currently faced by each wearable self-powered generator.

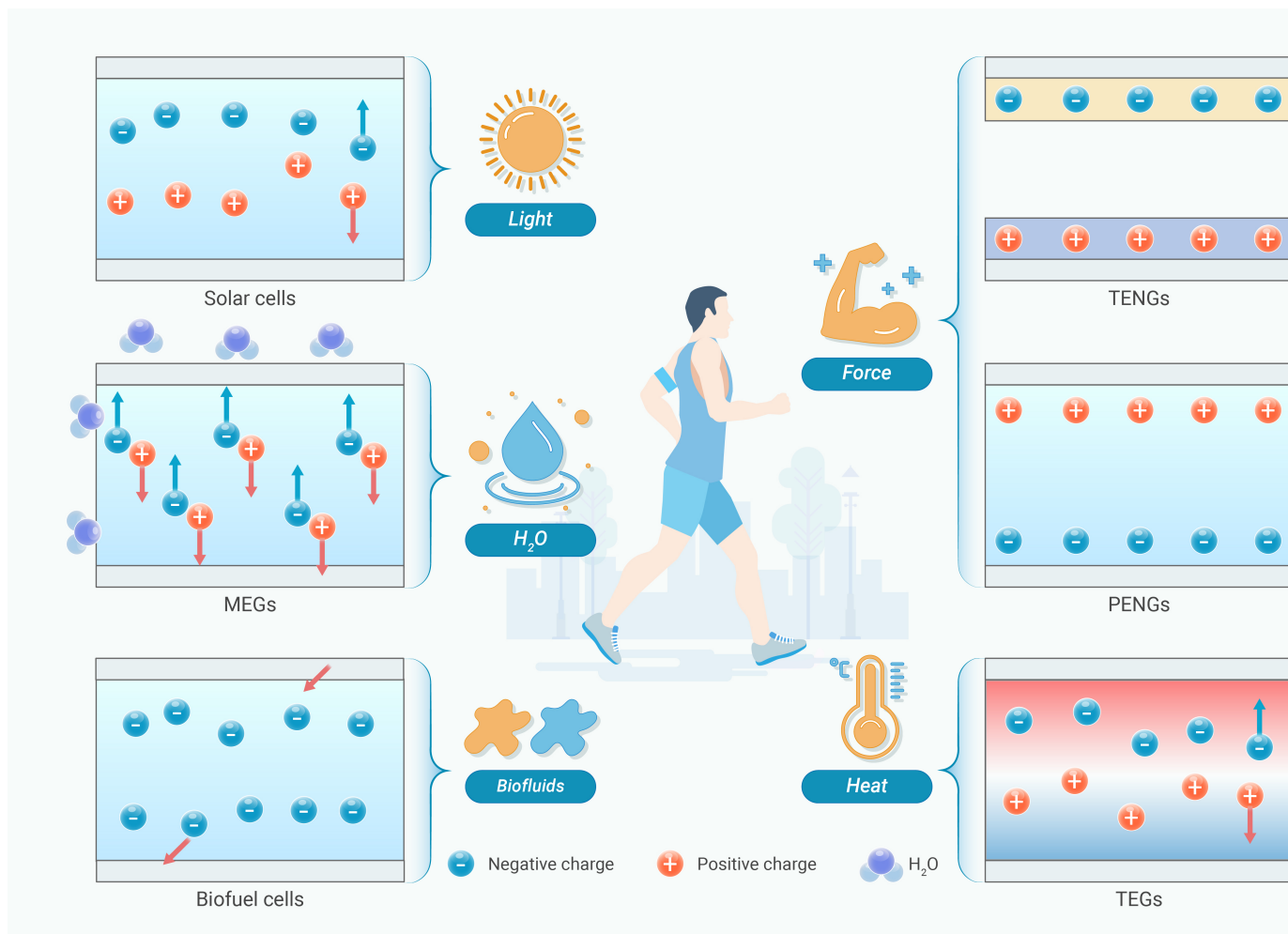


Figure 1. Various energy sources for the power generations.

Harvest the energy from the environment for self-power generation Solar cells

Solar cells convert light energy into electricity through photovoltaic effects. They typically consist of three parts: active layers, carrier-selective layers, and electrodes. When sunlight hits the solar cells, the active layers absorb the photons, creating electron-hole pairs or excitons. These pairs are then separated by the built-in potential and collected by the electrodes through carrier-selective layers, generating a photo-generated electromotive force. At last, when the load is connected to the electrodes, the photo-generated electromotive force drives electrons and holes to flow in the circuit, creating a photo-generated current. Figure 2A shows the working principle of a silicon-perovskite tandem solar cell.

Solar cells offer the benefits of a compact structure and high conversion efficiency, making them an ideal source of green and clean energy. Common solar cells include silicon solar cells (SSCs), perovskite solar cells (PSCs), organic solar cells (OSCs), and dye-sensitized solar cells (DSSCs) (Figure 2B). The thickness of the active layer ranges from a few hundred nanometers to a few micrometers. Reducing the thickness of the substrate can improve the power density of the device, making it flexible and lightweight. With the emergence of new functional materials, the advancement of manufacturing technologies, and the increasing demand in the wearable market, flexible solar cells have seen rapid progress.

The thickness of the active layer in solar cells has a significant impact on their performance, directly affecting light absorption, exciton dissociation, and carrier transport efficiency. When the active layer becomes thicker, the number of photo generated excitons increases, the built-in electric field weakens, and the distance of carrier transport increases, resulting in a decrease in carrier collection efficiency and a decrease in material flexibility.

On the contrary, reducing the thickness of the active layer can improve the performance and flexibility of the device. Of course, decreasing the thickness may lead to lower PCE of the device due to lower open-circuit voltage (V_{oc}) of the material. Therefore, the thickness of the active layer should be controlled appropriately. For instance, Erkan Aydin et al.²³ enhanced the photovoltaic coupling of chalcogenide-silicon tandem solar cells by employing ultrathin (5 nm) amorphous indium-zinc oxides (IZOs) as interconnecting transparent conductive oxides (TCOs). The researchers systematically reduced the thickness of the interconnecting TCO layer in the chalcogenide-silicon tandem cell from 20 nm to 2 nm to measure its effect on the device performance, and it was found that the tandem performance could be improved with the gradual reduction of the thickness of the TCO layer. However, when the thickness is less than 2 nm, it leads to lower V_{oc} due to film discontinuities, and when the thickness is about 5 nm, the device achieves the best performance due to the induced higher net dipole and work function (WF) uniformity as well as consistency with the coverage factor, which improves the PCE from 29.6 to 32.6%. Besides, compared to single-junction solar cells, tandem solar cells with specific structures have the best PCE for the device due to their wide photon absorption range, which absorbs high energy photons from the top cell through a wide bandgap, and uses a relatively narrow bandgap absorbing material at the bottom of the cell to absorb the low energy photons, which in turn minimises the thermalisation and the loss of transparent energy. For example, Nikhil Shrivastav et al.²⁴ designed a tandem solar cell consisting of a chalcogenide-based top cell and a CIGS-based bottom cell to achieve an overall device PCE of 29.7%.

In recent decades, the comprehensive performance of solar cells has been greatly improved,²⁵ especially in the case of PSCs. Sargent et al.²⁶ achieved a power conversion efficiency (PCE) of 26.15% in a 0.05 square centimeter illu-

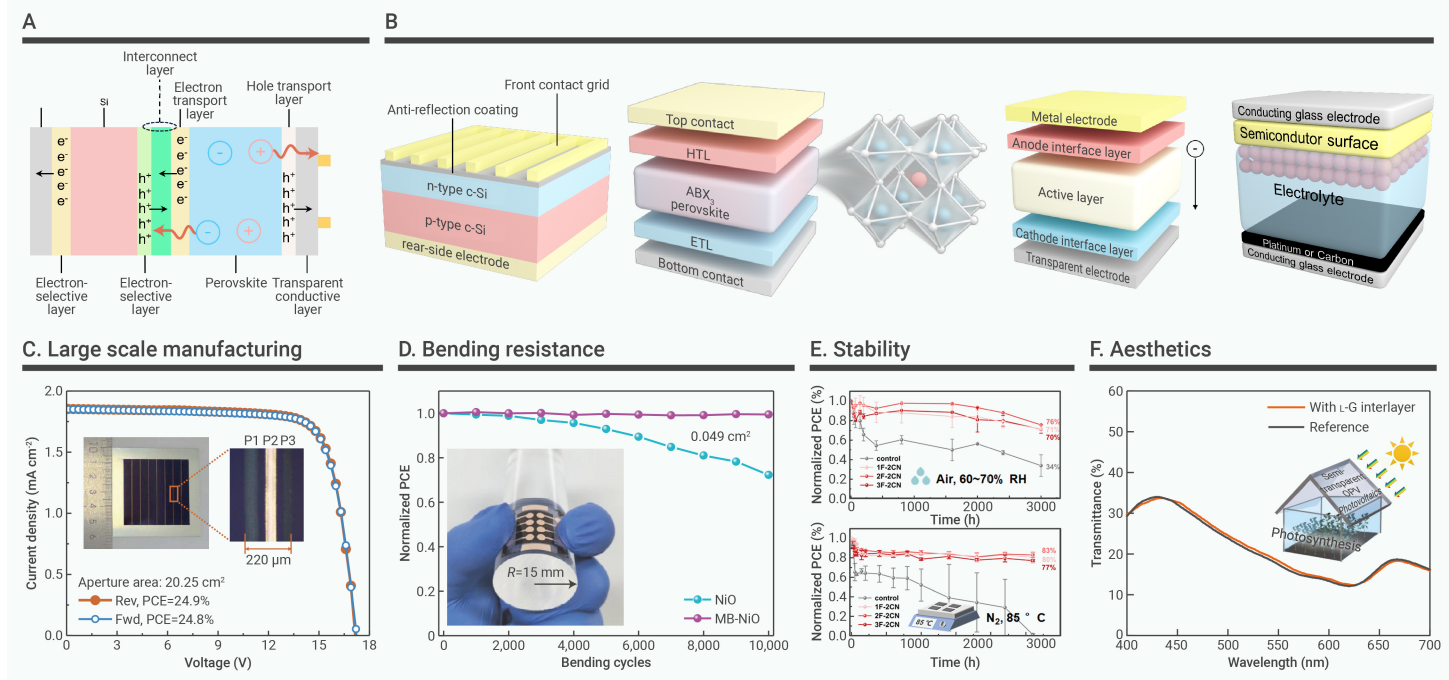


Figure 2. The principle, structure, challenges, and solutions of solar cells (A) Working principle of a silicon-perovskite tandem solar cell. (B) Different types of solar cells. From left to right are silicon solar cells, perovskite solar cells, organic solar cells and dye-sensitized solar cells. (C) Photo of the 20.25 cm² module (left) and an optical microscopy image (right). Reproduced with permission.¹⁹ Copyright 2024, AAAS. (D) Bending tests of flexible tandem cells. Reproduced with permission.²⁰ Copyright 2022, Springer Nature. (E) PSC operates steadily in harsh environments. Reproduced with permission.²¹ Copyright 2023, Royal Society of Chemistry. (F) Transmittance measurements of the devices. Reproduced with permission.²² Copyright 2023, Springer Nature.

minated area and 24.74% in a 1.04 square centimeter illuminated area for an inverted (pin) PSC. Researchers have made breakthroughs in the large-scale preparation of perovskite solar cells. Tan et al.¹⁹ used aminoacetamide hydrochloride to homogenize the crystallization of perovskite and selectively passivate the defects at the buried perovskite interface. The steady-state PCE of the prepared large-area perovskite module reached 24.5% (Figure 2C). Flexible solar cells have also achieved leapfrog development, with improvements in flexible PSCs, flexible SSCs and OSCs have also seen significant development. Liu et al.²⁷ improved the flexibility of silicon wafers through structural design, allowing SSC to be rolled to over 360° with large-area (>240 cm²) preparation and high PCE (>24%). Additionally, Liu et al.²⁸ developed a flexible OSC with micrometer-sized surface topologies which achieved a PCE of 19.06%. The PCE of different solar cells in recent years is shown in Table 1.

PSCs are considered an ideal material for solar self-powered equipment due to their advantages such as lightweight, thinness. The morphology of perovskite layers, such as grain size and grain boundary density, has a significant impact on charge transfer and battery performance. For example, by optimizing the crystal quality and interface engineering of perovskite layers, defect states at grain boundaries can be significantly reduced, thereby improving carrier mobility and battery efficiency. For commercial wearable self-powered devices, solar cells not only need to have good perovskite layer quality, but also need to have good flexibility, as well as good mechanical properties and stability.³⁹ Tan et al.²⁰ bridged perovskite with NiO nanocrystalline films, which endowed flexible perovskite solar cells maintain 100% performance even after 10000 bending cycles with a radius of 15 mm (Figure 2D). Ge et al.²¹ effectively optimize defects at grain boundaries of perovskite

Table 1. The PCE of different solar cells in recent years.

Type	Type of technology	PCE	Ref.
Tandem solar cells	Perovskite/silicon tandem solar cells	32.60%	23
Tandem solar cells	Perovskite/silicon tandem solar cells	31.30%	29
Silicon solar cells	Monocrystalline silicon solar cell	27.30%	30
Silicon solar cells	Silicon heterojunction solar cell	26.70%	31
Silicon solar cells	Silicon heterojunction solar cell	24.50%	27
Perovskite solar cells	All-perovskite tandem solar cell	28.20%	32
Perovskite solar cells	Single junction perovskite (inverted structure)	26.70%	33
Perovskite solar cells	Single junction perovskite	26.15%	34
Organic solar cells	Single-junction organic solar cell	20.82%	35
Organic solar cells	Binary organic solar cell	19.20%	36
Organic solar cells	Micro-nano surface all-polymer solar cell	19.06%	28
Dye-sensitized solar cells	Co-sensitized organic dyes and cobalt-based electrolyte	14.30%	37
Dye-sensitized solar cells	Organic dye and cobalt-based electrolyte	13.00%	38

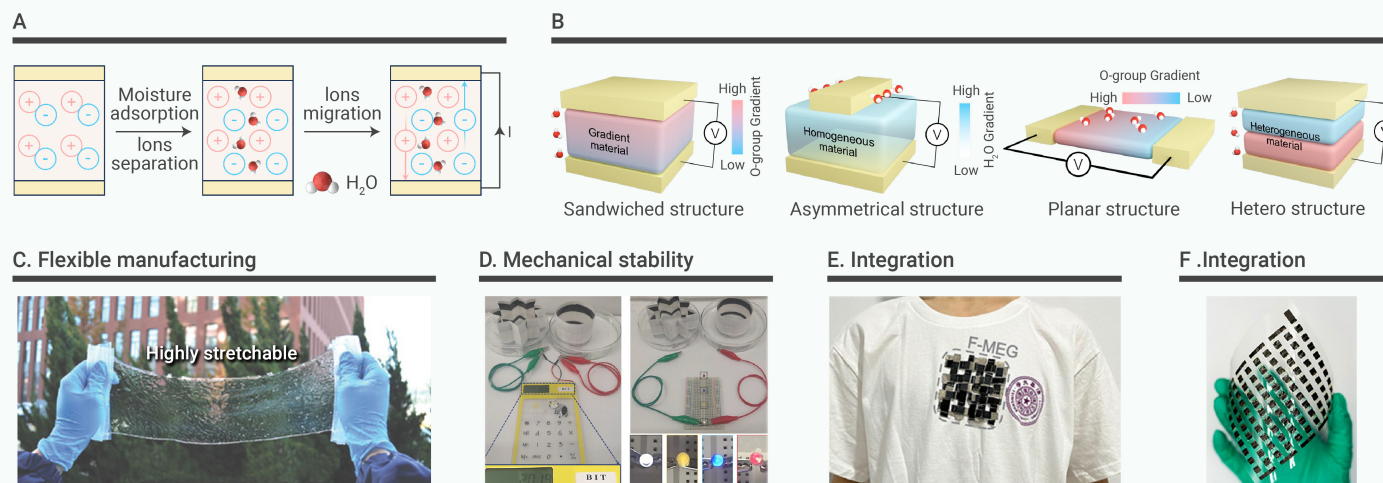


Figure 3. The principle, structure, challenges, and solutions of MEGs (A) Diagram illustrating a MEG operating based on ion diffusion mechanisms. (B) Different structures of MEGs. (C) Highly stretchable MEG. Reproduced with permission.⁴⁵ Copyright 2023, Wiley-VCH. (D) Stable output of MEG in different folding forms. Reproduced with permission.⁴⁶ Copyright 2019, American Chemical Society. (E, F) Different integrations of MEGs. Reproduced with permission.^{47,48} Copyright 2024, 2023, Cell Press.

cell, resulting in it can maintain about 80% of the initial PCE after 3000 hours of operation in harsh environments (Figure 2E). In addition to flexibility and stability, aesthetics is also important for wearing experience. For example, semi-transparent solar cells (Figure 2F), Yang et al.²² developed a semi-transparent perovskite solar cell with good transparency and stability, with a retention rate of 84.8% after 1008 hours of continuous illumination. These cells can be used not only for rooftops, but also for wearable devices.

Despite continuous breakthroughs in the research of solar cells, they still encounter numerous challenges when used in self-powered wearable devices. For instance, solar cells have unstable output due to natural sunlight discontinuity, making them unreliable for providing power to electronic devices in all weather conditions. Therefore, developing stable energy storage devices is crucial for creating a powerful solar-powered self-powered energy system. Compared to rigid solar cells, flexible solar cells have a lower PCE, and most stability reports have focused on bending tests. We need to conduct stability tests under real operating conditions. What's more, considering the limitations of energy density, we not only need to further improve PCE, but also optimize the collection and storage devices of solar energy so that they do not take up too much space on wearable devices, otherwise, it will contradict the principle of miniaturization for flexible wearable devices.

Moisture-enabled electric generators (MEGs)

As an emerging energy harvesting technology, MEGs have a wide range of application prospects. At present, the MEGs are based on mechanisms such as ion diffusion, streaming potential, and surface charge potential,⁴⁰ all of which are influenced by interactions at the liquid-solid interface. Taking ion diffusion mechanism for example, when the moisture around MEGs reacts with these hygroscopic materials, it causes the ionization of functional groups leading to the release of a large number of positively (negatively) charged ions. This process will generate an ion concentration gradient, resulting in directional migration of ions from high concentration to low concentration (Figure 3A), thereby generating an electric potential between the two electrodes.^{41,42} The ion diffusion mechanism involves three main processes: the uptake of water molecules by the material, the ionisation or hydrolysis of surface groups to produce ions and form an ion concentration gradient, and the directional diffusion of the formed ions. Generally speaking, there are two strategies for introducing ionic concentration gradients. The first one is to construct the gradient of functional groups directly in the power generating materials, mainly by preparing materials with oxygen-containing functional groups, and prompting the ionisation of the oxygen-containing functional groups (e.g. -COOH) when the material absorbs water molecules. Free-moving H⁺ is generated, and the gradient of the oxygen-containing functional groups inside the material produces an ion concentration gradient that induces the directional migration of H⁺.⁴³ The second is to introduce

water into the material in a directional manner, for example, by placing the PSSA membrane between two electrodes, perforating one of the electrodes and leaving the other electrode intact, so that the two electrodes have different permeability to water, prompting different concentrations of H⁺ produced by ionisation of the two ends of the PSSA membrane to form an ionic concentration gradient.⁴⁴ According to device structure classification, MEGs have four types: sandwich structure, asymmetric structure, planar structure, and heterostructure (Figure 3B).

The energy output of MEGs directly affects its application effect in wearable devices. Several strategies have been proposed, including materials optimization, (such as surface modification,⁴⁹ structure engineering,⁵⁰ ion doping,⁵¹ etc.), device design (such as device configuration,⁵² electrode/active material interface,⁵³ etc.), and controlling kinetics of moisture adsorption.⁵⁴ These mainly focus on improving the hygroscopicity and water capacity of the material; enhancing the directional migration of ions and improving the transmission efficiency of ions; improving ion dissociation ability, increase the number of mobile ions, thereby increasing current density. Wei et al.⁵⁵ successfully prepared MEGs based on FG DY-AIMC using Al ions coordinated with F. Due to the macroporous molecular structure in FG DY, the migration energy barrier of Al ions can be effectively reduced, and the performance can be significantly improved during which a stable output of high power can be achieved. Meanwhile, based on its excellent flexibility, it can be widely used in wearable electronics and IoT devices and in various humid environments. Duan et al.⁵⁶ proposed a method to construct an ion concentration gradient by coupling the photocatalytic hydrogen precipitation reaction and the hydro-electric effect, which enhanced the power density by 500% and greatly improved the power generation of MEG. Meanwhile, due to the restoration of the ion concentration gradient by photocatalytic hydrogen precipitation, the continuous and sustained output of the MEG can be achieved for more than 600 hours, which provides a design idea for the design of long-life MEGs with high sustained stability. Gao et al.⁵⁷ designed and developed a yarn-based MEG with a unique internal structure that maintains a persistent humidity gradient between the inner and outer electrodes, which further improves the performance through proton diffusion of PSSA and Al ions. This innovative design provides ideas for the development of self-powered e-textiles capable of utilising sweat as energy for applications such as warning waistcoats and hair bands.

Besides improving output, to use MEGs in flexible self-powered wearable devices, it's necessary to consider their flexibility, mechanical stability and integration. By adopting flexible materials and optimized structural design, MEGs can achieve high flexibility and stretchability without sacrificing performance.^{58,59} For example, through molecular engineering of hydrogels, Li et al.⁴⁵ have developed a high-performance, highly stretchable MEG with a

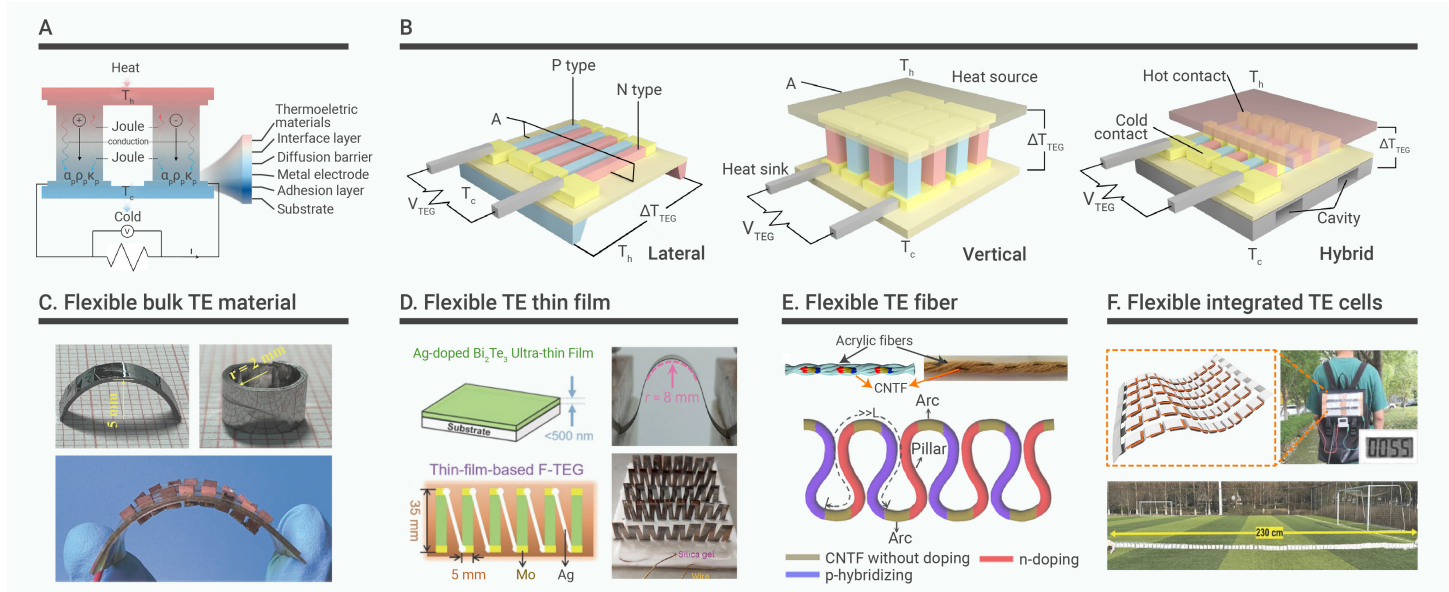


Figure 4. The principle, structure of TEGs and different flexible TE materials (A) Working principle of a thermoelectric generator based on the Seebeck effect. (B) The typical structures of thermoelectric devices. (C) Photo of plastic Bi₂Te₃ bulk material. Reproduced with permission.⁶⁸ Copyright 2024, AAAS. (D) Photo of a thin-film-based wearable F-TEG. Reproduced with permission.⁶⁹ Copyright 2022, Springer Nature. (E) The stretchable 3D thermoelectric generator woven from thermoelectric fibers. Reproduced with permission.⁷⁰ Copyright 2020, Springer Nature. (F) Photographs of flexible thermocells integrated on a bag and an integrated thermocells device of 80 units with a length of 230 cm. Reproduced with permission.⁷¹ Copyright 2023, Wiley-VCH.

tensile ability of up to 506% (Figure 3C). Mechanical stability of MEGs is critical for their application in wearable devices. Qu et al.⁴⁶ prepared a flexible hydropower film based on Al₂O₃ that can be mass-produced, which is resistant to mechanical deformation and has three-dimensional scalability (Figure 3D). With modular design and integrated manufacturing, MEGs can be conveniently integrated into various wearable devices. Wan et al.⁴⁷ developed an all-fabric MEG based on a self-gradient structure that enables energy conversion through a self-humidity gradient in the open while wearing the required breathability and comfort (Figure 3E). Qu et al.⁴⁸ prepared a planar MEG unit through printing technology, which can generate an open circuit voltage of 1.1 V and a power density of 2.6 mW cm⁻². It can form a fully printed functional integrated system with energy storage devices and functional devices (Figure 3F).

The research on MEGs is still in progress, and several challenges need to be solved. Firstly, in terms of mechanism research, there is a need for a guiding principle for design to further study the interactions between moist-electric materials and different forms of water (liquid, vapor). Secondly, to improve the power density of MEGs, it is necessary to develop suitable new moist-electric materials to reduce internal resistance and increase electrical power. Next is the development of electrode materials, using active materials as electrodes can accelerate the collection speed of water, thereby increasing power output. Finally, efforts need to be focused on miniaturization, integration, and the improvement of device stability to meet commercial requirements.

Harvest the energy from the human body for power generation

Thermoelectric generators (TEGs). Thermoelectric technology is considered the next generation of clean energy technology,^{60,61} which operates based on the Seebeck effect, generating electricity by utilizing the temperature difference (ΔT) between the two ends of the thermoelectric material.⁶² The Seebeck effect refers to the ability of thermoelectric materials to produce voltage when subjected to a ΔT . When a ΔT between the two ends of a thermoelectric material, the charge carriers inside the material will move from the high-temperature end to the low-temperature end under the effect of temperature gradient (Figure 4A), thereby forming a potential difference inside the material. In the application of thermoelectric wearable devices, heat can be directly collected from the human body to achieve self-powered.⁶³ The typical structures of thermoelectric devices include lateral structures, vertical structures, hybrid structures,⁶⁴ (Figure 4B) etc. By combining radiative cooling and photothermal absorption materials, the temperature difference between the two sides of the thermoelectric material can be further improved, thereby increasing the output.⁶⁵⁻⁶⁷

Thermoelectric materials can be classified based on their working temperature into low-temperature (< 400 K) thermoelectric materials, medium-temperature (500 K-900 K) thermoelectric materials, and high-temperature (> 1000 K) thermoelectric materials.^{72,73} Low-temperature thermoelectric materials (such as Bi₂Te₃) are widely used in the field of thermoelectric cooling.⁷⁴ Medium temperature thermoelectric materials (such as PbTe) have potential application prospects in industrial waste heat recovery.^{75,76} High-temperature thermoelectric materials (such as SiGe alloy, and half-Heusler alloy) can be

Table 2. The highest zT value, advantages and limitations of typical thermoelectric material systems.

Material system	Highest zT value (temperature range)	Advantage	Limitation	Ref.
Bi ₂ Te ₃ -based	1.7 (450 K)	Excellent room temperature performance and high module efficiency	High brittleness, rapid performance degradation at high temperatures	82
PbTe-based	~2.2 (800 K)	High efficiency in the medium-high temperature region	Lead toxicity and high processing cost	83
SnSe-based	2.6 (913 K)	Intrinsic low thermal conductivity, environmentally friendly	Poor mechanical performance, difficult to prepare on a large scale	84
Organic-inorganic complex	~1.0 (300 K)	Good flexibility and compatibility, capable of solution processing	Low conductivity and insufficient thermal stability	72

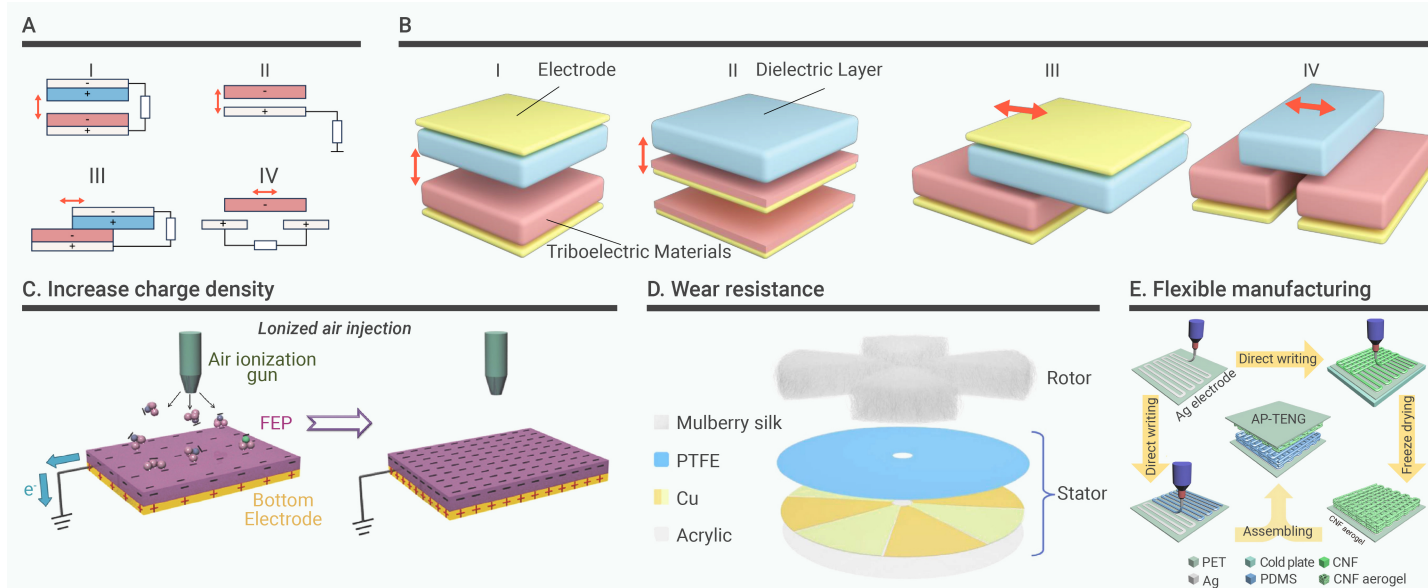


Figure 5. The principle, structure, challenges, and solutions of TENGs (A, B) Different types of TENGs models. I–IV: vertical contact-separation model, single-electrode model, contact-sliding model, freestanding model. (C) Surface functionalization to increase surface charge density by injecting single-polarity ions onto surfaces. Reproduced with permission.¹⁰⁸ Copyright 2014, Wiley-VCH. (D) Structure of MS-TENG. Reproduced with permission.¹⁰⁹ Copyright 2023, Elsevier. (E) Schematic fabrication process of AP-TENG. Reproduced with permission.¹¹⁰ Copyright 2019, Elsevier.

applied to integrated devices and deep space exploration.^{77,78} The efficiency of thermoelectric materials in converting heat into energy can be evaluated using a dimensionless factor zT . $zT = S^2\sigma T/\kappa$, where S , σ , T , and κ denote the Seebeck coefficient, electrical conductivity, absolute temperature, and thermal conductivity, respectively. A high zT value means high conversion efficiency. To improve zT , it can be achieved via increasing S^2 , σ and reducing κ . The current strategies mostly focus on optimizing electron transport properties by controlling carriers' concentration and band engineering, as well as reducing κ through structural design and defect engineering.^{79–81} Table 2 shows the zT value, advantages and limitations of typical thermoelectric material systems.

High zT materials face rigidity limitations, for example, high-performance materials such as Bi_2Te_3 and PbTe are mostly brittle crystals, which are difficult to directly use for flexible substrates such as textile fibers. The fracture toughness of Bi_2Te_3 bulk material is only $\sim 1 \text{ MPa}\cdot\text{m}^{1/2}$, and the bending radius needs to be greater than 10 mm to avoid cracking.⁷² However, material flexibility will lead to a decrease in performance. Flexible TEGs often use thin film or fiber structures, but a decrease in thickness and volume will significantly increase interface resistance. For example, the zT value of Bi_2Te_3 thin films is usually 30%–50% lower than that of bulk materials.⁸⁵ Flexible bulk thermoelectric materials, flexible thermoelectric films, flexible thermoelectric fibers, etc., which can achieve power outputs of approximately 100 μW ,^{61,86–88} 10 μW ,^{89,90} and 5 μW ,^{87,91,92} respectively. Therefore, limited flexibility and inadequate performance are the main factors limiting the commercialization of flexible thermoelectric materials. Developing new methods for preparing large-area flexible thermoelectric materials and preparing advanced flexible thermoelectric materials can effectively overcome constraints.⁹³ For instance, Chen et al.⁹⁴ developed a printing technology to produce inorganic flexible thermoelectric thin films, and constructed a flexible thermoelectric device assembled from n-type Bi_2Te_3 based and p-type $\text{Bi}_{0.4}\text{Sb}_{1.6}\text{Te}_3$ thin films through printing technology, achieving a normalized power density over $3 \mu\text{W}/\text{cm}^2\cdot\text{K}^2$.

In the practical application of wearable devices, the development of thermoelectric materials should balance flexibility and thermoelectric performance. The current methods include defect engineering,⁹⁵ interface design,⁷² fibrous structures,⁹⁰ composite materials, etc.⁹¹ They can synergistically modulate the carrier concentration, Seebeck coefficient, mobility, and propagation of local stress, or enhance the bonding force between rigid materials and flexible substrates and electrodes, reducing contact resistance and thereby improving the flexibility and thermoelectric output performance of the

device.^{96–98} Shi et al.⁵⁸ utilized anti site defects to generate high-density and diverse microstructures, thereby plasticizing brittle Bi_2Te_3 (Figure 4C), whose quality factor at 300 K is 1.05. Zheng et al.⁶⁹ induced uniform lattice distortion through silver doping in the Bi_2Te_3 film, and the material can achieve a zT value similar to 1.2 at room temperature and withstand 2000 bending tests at an 8 mm bending radius (Figure 4D). Snyder et al.⁷⁰ fabricated a stretchable 3D thermoelectric generator woven from thermoelectric fibers by wrapping alternately doped carbon nanotube fibers with acrylic fibers (Figure 4E). Besides, there are other forms of TEGs, for instance, Yan et al.⁷¹ designed a thermogalvanic ionogel with a Seebeck coefficient of 32.4 mV/K, which can be applied to the human body (Figure 4F).

In addition, phonon engineering is also an effective method, especially for thermoelectric thin film materials. For instance, through combining high electron concentration metals with thermoelectric materials to construct superlattice nano multilayer thin films, the multilayer structure can overcome the inherent contradiction between conductivity and Seebeck coefficient, exhibiting very high electrical conductivity. The energy filtering effect of the semiconductor/metal interface can improve the Seebeck coefficient, thereby enhancing the power factor (PF).⁹⁹ Moreover, superlattice nanostructures can reduce the mean free path of phonons, enhance phonon scattering, and thus lower the thermal conductivity of materials.¹⁰⁰ Shen et al.¹⁰¹ achieved a PF of 1785 $\mu\text{W}/\text{m}\cdot\text{K}^2$ by constructing a superlattice nano multilayer structure of $\text{Bi}_2\text{Te}_3/\text{W}$. In summary, the superlattice structure material system based on phonon transport can effectively enhance the power factor of thermoelectric materials and has application prospects in wearable self-powered devices.

TEGs still face some challenges when used in self-powered wearable devices, such as efficiency issues, miniaturization issues, system integration, and thermal management issues. In wearable devices, TEGs need to be miniaturized and integrated to meet the size requirements of the device, but this may lead to a decrease in power density. In the future, the application potential of TEG should be enhanced by optimizing material properties, improving structural design, and strengthening thermal management.

Triboelectric nanogenerators (TENGs)

The TENGs can generate electrical energy through slight movements or friction between materials, which are mainly based on the coupling of contact electrification and electrostatic induction effects, converting mechanical energy into electrical energy through physical motion.¹⁰² In this process, the charge is transferred through electrons, the direction of electron transfer depends on the material involved, whose driving force is Maxwell displace-

Table 3. The charge density and key mechanisms under different modification strategies.

Modification strategy	Material system	Charge density	Key mechanisms	Ref.
Optimization of fluorinated polymers	RRF-FEP	352 $\mu\text{C}/\text{m}^2$	Molecular chain orientation and crystallinity Regulation	125
Micro-nano patterning	Honeycomb FEP	200 $\mu\text{C}/\text{m}^2$	Increased contact area	126
Chemical functionalization	Ion irradiation Kapton	332 $\mu\text{C}/\text{m}^2$	Introduction of polar functional groups	127
Charge injection	Corona polarized FEP	240 $\mu\text{C}/\text{m}^2$	Dipole oriented arrangement	128

ment current, finally, the charge is maintained in electrostatic balance through an external circuit.¹⁰³ TENGs typically consist of conductive electrodes and dielectric materials. Common conductive electrodes include metals and other conductive materials, which act as free charge collectors, allowing free and movable charges to flow between two electrodes. Dielectric materials, such as polytetrafluoroethylene and fluorinated ethylene propylene (FEP), are used to generate triboelectric charges. Overall, researchers have developed TENGs models such as contact separation,¹⁰⁴ single electrode,¹⁰⁵ contact sliding,¹⁰⁶ and freestanding, etc.¹⁰⁷ (Figures 5A-B), which can be applied to corresponding situations.

In the wearable design of TENGs, three factors should be considered: high charge density, high durability, and good flexibility. The following methods have been used to increase charge density. Manufacturing micro/nano surface structures to increase contact area.¹¹¹⁻¹¹⁴ Improving the effective surface contact area or surface roughness of dielectrics can greatly enhance surface charge density. Introducing micro nano scale structures such as micro/nano pyramids and wrinkled structures can improve surface contact area and increase surface roughness, making it easier for frictional charges to separate in micro structured thin films and form larger dipole moments between electrodes. In addition, reproducible and cost-effective technologies for large-scale production have also been widely studied, including template method, laser surface patterning, photolithography technology, electrospinning and other micro/nano preparation methods. For instance, Liu et al.¹¹⁵ effectively increased the contact charged area of the interface by constructing a pyramid structured TENG. While under lighter forces, the contact area and open circuit voltage of TENG increased with the increase of pyramid structure spacing, while under heavier forces, the contact area and open circuit voltage of TENG decreased with the increase of pyramid structure spacing, revealing the correlation between the contact area and electrical performance of TENG and array structure. Gu et al.¹¹⁶ constructed a nano layered folded structure on frictional materials to achieve self-healing TENG. Through using a heterostructure of ultra-thin ZnO nanowires/self-healing PDMS, more charge capture sites were achieved, resulting in a charge density of 130 $\mu\text{C}/\text{m}^2$ and self-healing functionality.

Chemical functionalization to increase surface charge density or facilitating triboelectric charge transfer:^{108,117-119} the chemical structure and electrical properties of dielectrics are closely related. The surface functional groups of polymers can determine the charge density and polarity of materials. Starting from the physical properties such as material work function, interface potential barrier, electron affinity, and band structure, the correlation between molecular structure and macroscopic contact electrification can be elucidated for solid-solid and solid-liquid interfaces.

Charge injection to increase charge density, the triboelectric output capability of materials depends on the surface charge density on the dielectric. During the material preparation process, the use of charge injection or charge excitation can to some extent alter the surface charge density of dielectric materials. Common charge injection methods can be achieved through ionization guns, corona discharge, and electrospinning. For instance, when the fluorinated ethylene propylene (FEP) substrate is exposed to negatively ionized air, the transfer of electrons from aluminum to the FEP surface significantly increases (Figure 5C).¹⁰⁸

Fluorinated polymer (e.g., PTFE, FEP, and PVDF) occupies a central position in triboelectric materials. Fluorine atoms have high electron affinity (3.98 eV) and are more likely to capture electrons from paired materials (such as nylon and polyimide) during contact charging, forming stable negatively charged surfaces;¹²⁰ The high dielectric constant of fluorinated polymers (e.g., PVDF's $\epsilon \approx 10$) enhances interfacial polarization and suppresses charge dissipation.¹²¹

However, fluorinated polymers generally have high brittleness (e.g., PTFE fracture toughness $< 1 \text{ MPa}\cdot\text{m}^{1/2}$), making it difficult to directly use for flexible substrates.¹²² The current solutions include nanocomposite and surface functionalization, which enable fluorinated polymers to maintain high charge density while achieving good mechanical stability.¹²³ For instance, Zhou et al.¹²⁴ used PVDF multi walled carbon nanotubes nanofiber film as the tribo-positive and corona charged fluorinated ethylene propylene (FEP) solid film as the tribo-negative layer, resulting in 32 times electrical output than single-layer friction nanocomposite fibrous membrane-TENG, which can generate a maximum surface power density of 3.78 W/m^2 under acoustic excitation of 200 Hz and 116 dB and achieve good wearable applications. Table 3 introduces the charge density and key mechanisms under different modification strategies.

The wear and tear of triboelectric materials during operation inevitably affects the durability of TENGs, which is a key issue limiting the practical application of TENGs. The current methods mainly focus on material selection, structural design, and other aspects in order to improve the durability. For example, He et al.¹⁰⁹ proposed a TENG based on mulberry silk (MS-TENG) (Figure 5D), which has a fluffy structure and good frictional electrical properties. This enables the MS-TENG to achieve high charge density and low friction coefficient. In addition, some new materials, such as diamond-like carbon films, are very promising materials with unique mechanical and physical properties, such as low friction coefficient, high hardness, and corrosion resistance.^{129,130} Structural design can also ensure ultra durability of TENGs. For example, planar structures,^{131,132} cylindrical structures,¹³³ spherical structures, etc.¹³⁴ have been proven to significantly improve the wear resistance of TENGs.¹³⁵ Besides output performance and wear resistance, the flexibility of TENGs is crucial for wearable devices. Researchers have developed advanced methods for preparing flexible TENGs, such as all-printing, weaving, etc.¹³⁶ For instance, Song et al.¹¹⁰ used an all-printing method to manufacture flexible TENG with 3D micro/nano-layered patterned structures (Figure 5E). Wang et al.¹³⁶ used a double needle bed flat knitting machine technology to fabricate a 3D double-faced interlock fabric TENG, which can serve as a bending sensor for detecting the degree of arm bending, a 3D tactile sensor, and an energy harvesting device.

The development of TENGs is still in the early stages before large-scale commercialization. From the theoretical perspective, we need to study the electron transfer mechanism from the microscopic perspective and propose new strategies to improve the output capacity, stability, and durability of TENG sensors. From the perspective of material design, we need to develop advanced triboelectric materials with characteristics such as lightweight, low cost, ecological friendliness, good flexibility, and good biocompatibility.

Piezoelectric generators (PENGs)

The piezoelectric effect refers to the phenomenon of generating electricity by applying pressure to specific materials. Some materials, such as tourmaline and quartz, can generate surface charges under mechanical stress and are called piezoelectric materials (Figures 6A-B). The working principle of piezoelectric nanogenerators (PENGs) is based on two effects: the first originates from the direct piezoelectric effect, and the second is the polarization change caused when the symmetry of the crystal structure center of non-centrosymmetric piezoelectric materials is disrupted. The polarization change caused by stress will generate an internal potential difference throughout the materials, which is used to drive the flow of charges in the external circuit, thereby generating alternating current.

Piezoelectric materials can be categorized into piezoelectric ceramics, piezoelectric polymers, and piezoelectric composite materials. Piezoelectric

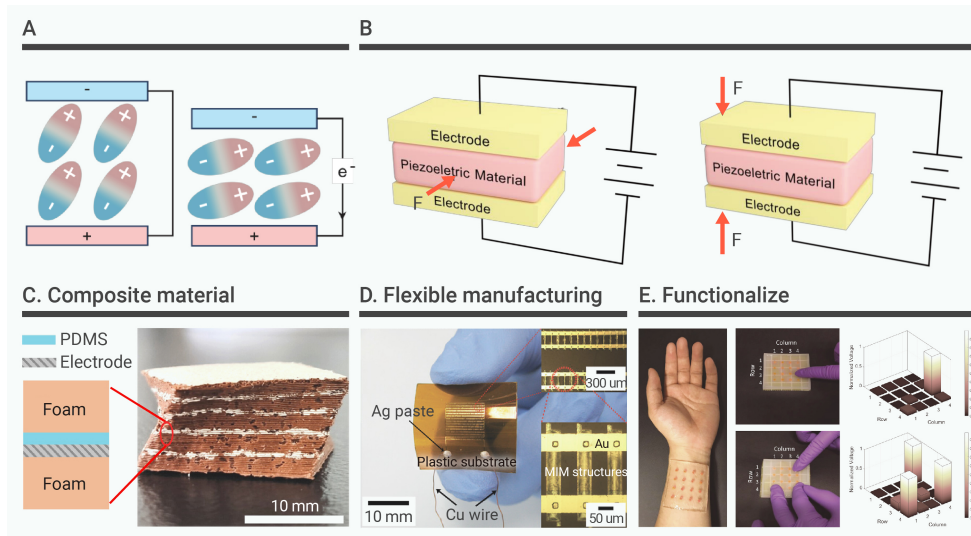


Figure 6. The principle, structure, challenges, and solutions of PENGs (A, B) Working principle and structure of piezoelectric nanogenerators. (C) Schematic diagram of composite foam material. Reproduced with permission.¹⁴⁵ Copyright 2023, AAAS. (D) Picture of BaTiO₃-based PENG based on flexible substrate (PDMS). Reproduced with permission.¹⁵⁰ Copyright 2010, American Chemical Society. (E) The gesture recognition function of piezoelectric electronic skin under single-touch and multi-touch. Reproduced with permission.¹⁵¹ Copyright 2021, Springer Nature.

ceramics (e.g., PbZrTiO₃ (PZT),¹³⁷ BaTiO₃,¹³⁸ ZnO,¹³⁹ etc.) exhibit high piezoelectric constants, sensitivity, and resistance to acid and alkali. However, their mechanical properties are brittle and cannot undergo extensive deformation. Furthermore, bulk piezoelectric ceramics are rigid with high resonant frequencies, making them unsuitable for collecting energy from low-frequency environmental vibrations.¹⁴⁰ As a result, directly application of piezoelectric ceramics to wearable self-powered electronic devices is challenging; Piezoelectric polymers (e.g., poly(vinylidene fluoride) (PVDF),¹⁴¹ Polyvinyl alcohol (PVA),¹⁴² polyvinylidene fluoride-trifluoroethylene (P(VDF-TrFE)), etc.¹⁴³) are flexible, lightweight, and inexpensive, which can be widely used to manufacture wearable devices.¹⁴⁴ However, their energy conversion rate is low. Consequently, piezoelectric composite materials, which are a combination of piezoelectric ceramics and piezoelectric polymer materials, have the advantages of lightweight, good flexibility, and high processability.^{145,146} Typical synthetic combinations include: PVDF-PZT,¹⁴⁷ BaTiO₃/PVDF,¹⁴⁸ ZnO-PVDF,¹⁴⁹ etc. Shao et al.¹⁴⁵ designed a composite foam based on PDMS and CaCu₃Ti₄O₁₂ nanoparticles, which can achieve ultra-high flexible electrical effect. This foam material shows extremely strong deformation characteristics under compression (Figure 6C), its density-specific equivalent piezoelectric coefficient is 120 times of the lead zirconate titanate.

Researchers have developed many optimization methods for flexible PENG manufacturing based on piezoelectric composite materials,¹⁵² such as direct writing,¹⁵³ electrospinning,¹⁵⁴ etc. Lee et al.¹⁵⁰ first prepared BaTiO₃ thin films on flexible substrates (PDMS) through microfabrication and soft lithographic techniques (Figure 6D). The combination of composite materials and flexible manufacturing has put PENG on a solid path towards functionalization. Petritz et al.¹⁵¹ developed a flexible energy harvesting device consisting of PENG, diode rectifier, and storage capacitor. It can be connected and stacked with various materials and surfaces, and operates stably at a bending radius of 40 μ m. Based on this, they prepared a self-powered ultra soft piezoelectric electronic skin, demonstrating the gesture recognition function of piezoelectric electronic skin under single touch and multi touch (Figure 6E). Babita Sharma et al.¹⁵⁵ prepared PENGs by casting method using PDMS as a matrix and KNN as a nanofiller to improve the dielectric and piezoelectric properties of the material by adjusting different concentration gradients, to achieve stable output voltages and currents, and to successfully apply them to low-power flexible electronic devices. Sayan Chakraborty et al.¹⁵⁶ achieved high performance PENGs by doping MoSe₂ nanosheets into PVDF to form a nanocomposite friction layer, utilising DFM as a solvent to achieve an optimised concentration of MoSe₂ in the internal structure, which significantly improves the collection efficiency. The PENGs were shown through testing to output stable voltages during numerous biomechanical activities such as wrist or elbow movements and walking, etc. Therefore, the PENGs can be widely used in portable wearable electronics and IoT applications.

Owing to the frequent mechanical stress and strain experienced by piezoelectric materials in practical applications, it is important to improve their mechanical properties (tensile and wear resistance) in a low-cost and mass-produced manner. Further improvements are required in the piezoelectric

properties of the materials to optimize energy harvesting efficiency and ensure efficient operation of the devices. What's more, the long-term reliability of PENG devices is crucial, it's necessary to improve the durability and long-term performance of piezoelectric materials, especially in applications where consistency and lifespan are crucial, such as biomedical applications. Wang et al.¹⁵⁷ designed a transient PENGs that can be converted into a high amplitude waveform output, which has a faster charging efficiency due to greatly increasing its electrical energy. In addition, the PENGs can be applied in the area of powering implantable biomedical devices since the power they provide is capable of providing stimulation signals for a properly functioning commercial pacemaker.

Biofuel cells (BFCs)

BFCs are mainly categorized as either microbial BFCs or enzymatic BFCs. Microbial BFCs utilize microorganisms as biocatalysts for related reactions, while enzymatic BFCs utilize enzymes for oxidation and reduction reactions. For wearable self-powered devices, enzymatic BFCs have more application prospects. Enzymatic BFCs can convert biochemical energy into electrical energy through redox reactions of various biological fluids in the human body, such as tears, blood, and sweat. The oxidation and reduction catalysts in the human body accelerate this redox reaction. The oxidation catalyst promotes the oxidation reaction of fuel at the biological anode, while the reduction catalyst promotes the reduction of fuel to water at the biological cathode (Figures 7A-B).¹⁵⁸ Ultimately, electrons in the oxidation-reduction reaction flow in the external circuit to generate an electric current, the magnitude of which depends on the concentration of chemical fuel in the biological fluid and the efficiency of the biocatalyst in promoting electron transfer between electrodes.

BFCs electrodes need to possess characteristics such as elasticity, conductivity, and biocompatibility, the combination of carbon-based materials (graphene, carbon nanotubes, etc.) and flexible substrates (polymer films, hydrogels, etc.) can meet most needs.¹⁶³⁻¹⁶⁵ Gao et al.¹⁵⁹ reported an integrated electronic skin driven by human sweat, which obtains energy from human sweat through lactic acid BFCs. It can provide a power density of 3.5 mW/cm² in human sweat and run stably for 60 hours. In addition, the integrated electronic skin can monitor specific metabolites and real-time skin temperature and muscle contraction during human movement (Figure 7C). This work has greatly promoted the application of biofuel cell self-powered electronic skin in the fields of health monitoring and exercise health care.

The improvement of biomass management and electrode mechanical performance can ensure the stable operation of BFCs. Liao et al.¹⁶⁰ used polydimethylsiloxane (PDMS) to prepare a microfluidic device for storing sweat from human skin (Figure 7D). This device can store and release sweat at different flow rates under different bending degrees. Based on this, a hybrid microfluidic device can generate an average of 7.2 mJ and 80.3 μ W per charge. In practical applications, besides intermittent sweating, the wear resistance of flexible biological batteries is also crucial, especially when wearable devices adhere to human skin for a long time, requiring consideration of varying degrees of device strain. Zhou et al.¹⁶¹ designed the electrode array of flexible BFCs into a serpentine structure, which has a better stress distribution than traditional straight-line structures (Figure 7E), solving the efficiency decline problem happened when long-term strain in practical applications.

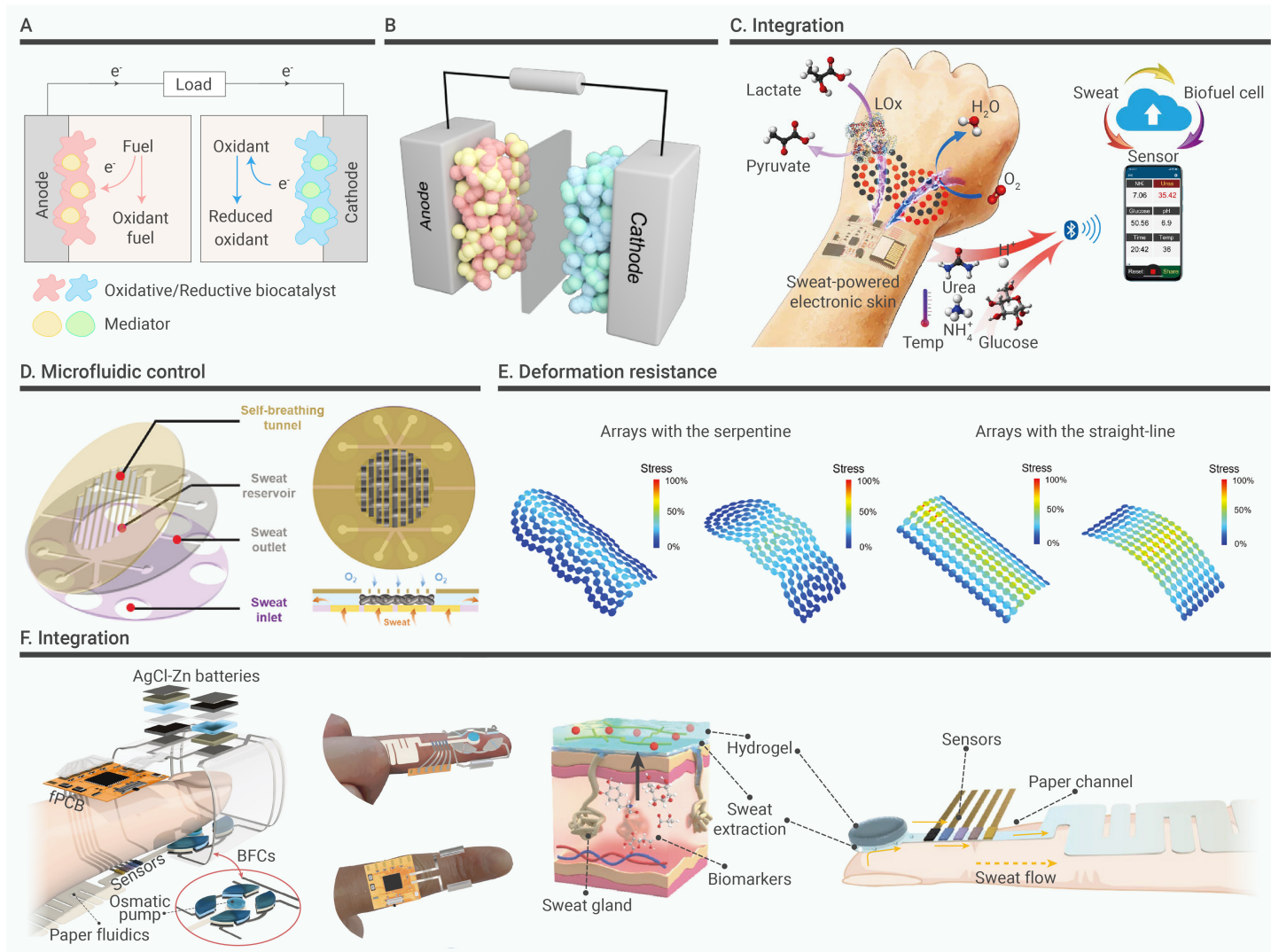


Figure 7. The principle, structure, challenges, and solutions of BFCs (A, B) Working principle and structure of a biofuel cell. (C) Schematic of a biofuel-powered e-skin performs multiplexed biosensing and wirelessly transmits data to a mobile user interface through Bluetooth. Reproduced with permission.¹⁵⁹ Copyright 2020, AAAS. (D) Layered diagram of sweat control device. Reproduced with permission.¹⁶⁰ Copyright 2023, Wiley-VCH. (E) Simulations of the stress distributions of the LIFE-G electrode arrays with the serpentine and the straight-line designs under mechanical deformation. Reproduced with permission.¹⁶¹ Copyright 2023, Wiley-VCH. (F) Schematic of the microgrid system and sweat excretion from human fingertips and sensor array on fingertips. Reproduced with permission.¹⁶² Copyright 2024, Springer Nature.

In addition, take enzyme biofuel cells for instance, researchers can optimize enzyme stability, electron transfer kinetics, electrode materials, and biofuels to enhance the output of enzyme fuel cells. The issue of enzyme activity is a difficult problem that hinders the stability of BFCs. Researchers have currently developed immobilization techniques, nano interface modifications, etc. to optimize enzyme activity. For example, the immobilization of glucose oxidase on the electrode was successfully achieved by combining multi walled carbon nanotubes with small biomolecules chitosan and crosslinking agent glutaraldehyde. After a period of cycling, the oxidation peak current density of the electrode only decreased by 9.8%, maintaining the stability and activity of the enzyme well.¹⁶⁶

To solve the problem of low output power, researchers mainly focus on promoting electron transfer and optimizing electrodes. For instance, nano conductive materials can promote interface electron transfer between electrodes and catalysts, reducing the activation energy of electron transfer in glucose oxidation reactions.¹⁶⁷ Jeeran et al.¹⁶⁸ modified the anode of BFCs with carbon nanotubes, which results in the BFC yield a V_{oc} of 0.45 V and a maximum power density of 266 $\mu\text{W}/\text{cm}^2$. Liao et al.¹⁶⁹ designed a bio anode with a layered structure of MXene/single-walled carbon nanotubes/lactate oxidase, which achieved a V_{oc} of 0.48 V and a power density of 220.9 $\mu\text{W}/\text{cm}^2$.

In terms of system integration and practical applications, Wang et al.¹⁶²

developed a continuous sweat sensing system based on enzyme biofuel cells and AgCl-Zn batteries. The system integrates a complete sensor array, energy module, and microcontroller unit system on one finger, and achieves energy capture and stable output without physical activity in the human body (Figure 7F). It relies on osmosis to continuously provide sweat to the sensor array, which can achieve long-term detection of glucose, vitamin C, lactate, and levodopa. This BFC can generate nearly 500 mJ of energy during the day and 300 mJ of energy at night, and can generate 125 μAh of energy during 8 hours of daytime activity and 90 μAh of energy during nighttime sleep to charge the battery.

BFCs still face key challenges such as low power output, the lifespan of biocatalysts, and stable fuel supply. For systemic stability issues, the lifespan of catalysts can be improved through bioengineering and optimization and modification of biocatalysts, and a stable supply of biofuels can be ensured by constructing microfluidic channels. The device itself, should have good mechanical properties, and its adhesion and breathability to the skin are very important.

Synergetic system

Based on the previous discussion, we have summarized and compared the characteristics of different energy harvesters, as shown in Table 4. Currently, most self-powered systems rely on a single input source, which has limitations in terms of sustainability and stable power supply. The synergetic

Table 4. The index comparison of six energy harvesters.

Index	Solar cells	MEGs	TEGs	TENGs	PENGs	BFCs
energy source	sunlight	Humidity gradient	Temperature gradient	mechanical friction	mechanical stress	Biofuels
Output voltage	0.5-1.0 V	0.3-1.2 V	10-100 mV/K	100-5000 V	1-50 V	0.2-0.8 V
power density	10-100 mW/cm ²	0.1-10 μ W/cm ²	1-100 μ W/cm ²	0.1-50 mW/cm ²	0.01-1 mW/cm ²	1-100 μ W/cm ²
Conversion efficiency	15-33%	<5%	5-15%	10-60%	10-30%	20-50%
Environment dependency	Light intensity	Humidity (>60% RH)	Temperature difference	mechanical movement	Vibration frequency	Biofuel concentration
Cost	Moderate to high	Extremely low	High	Low	Moderate	Moderate to high
Advantage	Mature technology, high efficiency, and scalability	Green, renewable, portable, and flexible	No moving parts, capable of working in harsh environments	High power density, lightweight, easy to manufacture	High power density, flexibility, and lightweight	Biocompatibility, and ability to work in a biological environment
Disadvantage	Dependent on sunlight, requiring a large area	Low output power, limited by humidity	Low efficiency, expensive materials, limited by temperature differences	Intermittent power generation, limited by mechanical input	Intermittent power generation, limited by mechanical input	Low efficiency, limited by biochemical reactions

energy harvesting system can utilize the synergistic effect of different types of energy collectors in the system to simultaneously collect multiple types of energy, thereby generating high output current and voltage, greatly improving the efficiency and stability of the entire module,¹⁷⁰ especially in situations where energy is weak or unavailable. The first synergetic energy harvesting device was reported in 2009, which utilizes solar cells and PENG to collect solar and ultrasonic energy.¹⁷¹ These two energy-harvesting devices with different working mechanisms can work simultaneously or independently, and they can also be integrated in parallel or series to improve overall energy-harvesting efficiency and output power. Since then, research on synergetic energy harvesters has rapidly developed, for instance, the synergy between TENGs and BFCs,¹⁷² TENGs and solar cells, TENGs and PENGs, as well as TENGs and TEGs.¹⁷³⁻¹⁷⁵

In a synergetic energy harvester system, it is easy to improve the efficiency of each component in the system through integrating macroscopic physical phenomena. For example, photo-thermal-electric synergetic systems,¹⁷⁶ hybrid mechanical energy harvesting systems,¹⁷⁷ integrated biological environment systems,^{172,178} all-weather energy matrices.¹⁷³⁻¹⁷⁵ Wang et al.¹⁷² adopted the concept of microgrid design, integrating BFCs and TENGs into an electronic textile microgrid system, and developed self-powered e-textiles with synergetic energy harvesting effects. Microgrid-integrated systems compensate for the limitations of BFCs due to delayed sweating and TENGs due to lack of movement, which can be easily installed on shirts, collecting energy through the sliding motion of the TENGs and sweat on the chest by the BFCs. Lin et al.¹⁷⁸ have developed a wearable self-powered wound dressing that can be activated by various stimuli from the patient's body and used for on-demand wound treatment. This self-powered dressing is composed of Bi₂Te₃ nanoplates, which are triggered by the surrounding temperature difference to control the production of hydrogen peroxide and effectively inhibit bacterial growth at the wound site. The integrated electrode of the wound dressing is connected to wearable TENGs, providing electrical stimulation to enhance cell proliferation, migration, and angiogenesis, thereby accelerating wound healing.

In addition to integrating macroscopic physical phenomena, different energy harvesters can also boost device performance from a theoretical perspective.^{179,180} For instance, Wang proposed piezo-phototronic effect can effectively combine the piezoelectric and photoelectric effects at the interface.^{181,182} While under stress, the strain piezoelectric charge at the interface would regulate the generation, separation, transportation, and recombination of charge carriers.^{183,184} Pan et al.¹⁸⁴ utilized two piezoelectric semiconductor materials: GaN and ZnO to construct a heterostructure-based self-powered flexible ultraviolet photodetector. While under strain induction, electrons will

move towards the n-ZnO side and holes will move towards the p-GaN side, resulting in the energy band on the ZnO side bending downwards and the energy band on the GaN side bending upwards. An increase in the height of the potential barrier can enlarge the built-in electric field, expand the depletion region, facilitate the separation and extraction of photo-generated electrons and holes, and repel their recombination.^{179,180} Yan et al.¹⁸⁵ combined moist-electric and thermoelectric mechanisms to prepare a flexible moist-thermoelectric generator. In this flexible synergetic generator, the polyelectrolyte membrane undergoes spontaneous adsorption of water molecules and heat, leading to rapid dissociation and diffusion of Na⁺ and H⁺, resulting in high power generation. Therefore, the assembled flexible MTEG (effective area = 1 cm²) can generate a V_{oc} of 1.81 V and a power density of 4.75 μ W/cm².

When designing a synergetic energy harvesting system, it is necessary to consider the nonlinear coupling effects of the hybrid system, the impact of interface engineering on multi physics field collaboration, and the comparative bias caused by the lack of standardized testing protocols.

In many applications related to wearable self-powered devices based on synergetic energy harvesting systems, researchers have achieved a "1+1>2" effect. To efficiently and reliably construct a multi-source energy harvesting system, we need to guide the selection of components with complementary characteristics and performance based on the design principles of "compatible external factors, proportional performance, and complementary functionality". The theoretical research on multi-source hybrid energy harvesting still needs to be further explored. We need systematic and in-depth theoretical research to select the most suitable synergetic mechanisms and optimal integration strategies, for achieving the best power output in the miniaturization process of wearable self-powered devices.

PROGRESSIVE APPLICATION SCENARIOS

Over the past few decades, with advances in materials design as well as processing technology, much of the work is beginning to focus on expanding the applications of flexible self-powered-generators. In this section, we discussed current applications of flexible self-powered generators for environmental monitoring, human monitoring and human-computer interaction, and looked forward to the future development of applications.

Environmental monitoring

Wearable environmental monitoring devices make it more convenient for users to obtain environmental data, greatly optimizing user experience. However, in some extreme environments, the battery system of traditional environmental monitoring equipment cannot withstand it, resulting in the

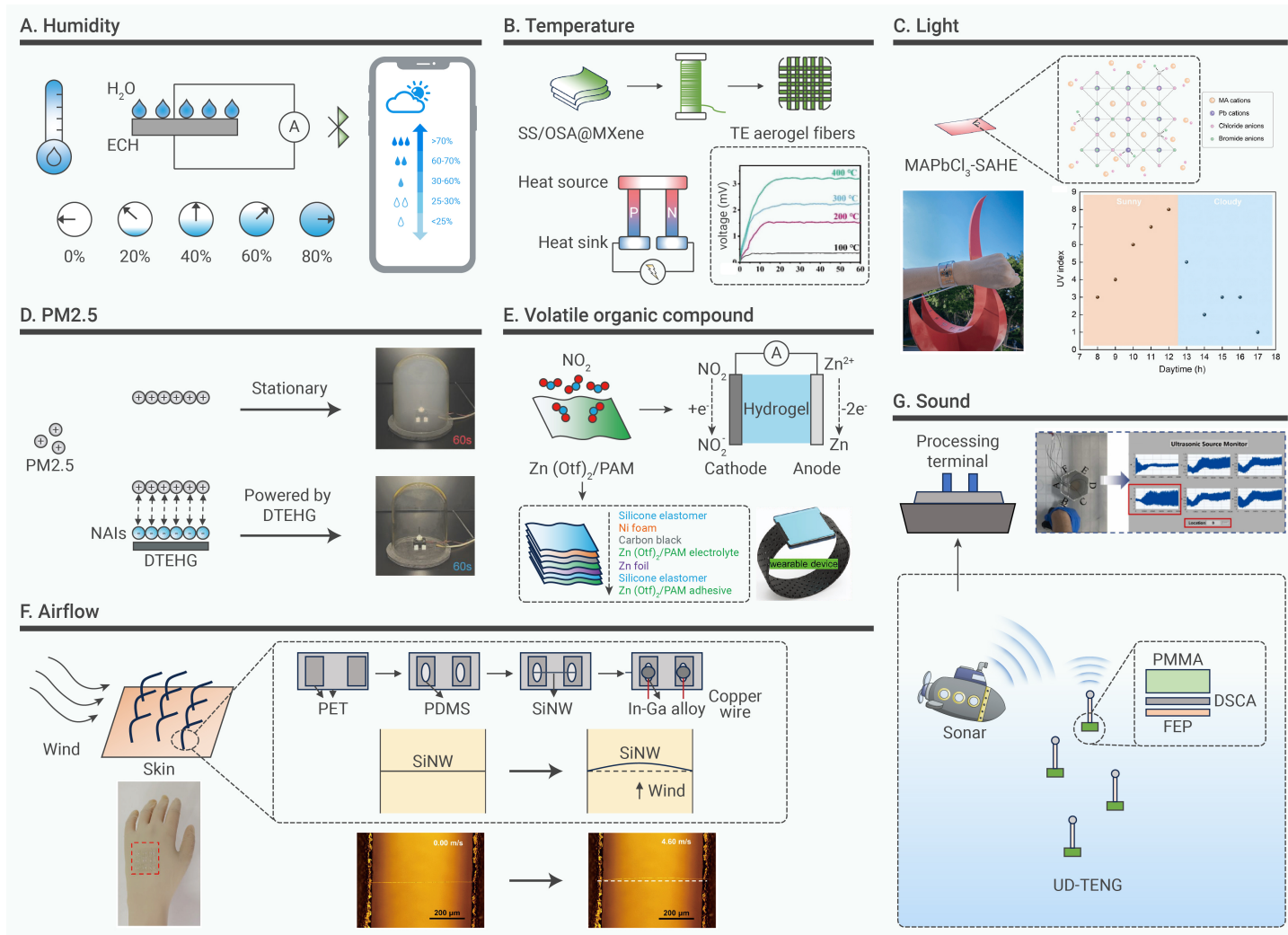


Figure 8. Application of self-powered wearables in environmental monitoring field (A) Self-powered electrochemical humidity detection system. Reproduced with permission.¹⁹⁸ Copyright 2023, Elsevier. (B) Self-powered, self-healing thermoelectric fibers for temperature detection. Reproduced with permission.¹⁹⁹ Copyright 2024, Springer Nature. (C) Wearable halide-exchanged chalcogenide photodetectors. Reproduced with permission.²⁰¹ Copyright 2022, Elsevier. (D) Self-powered air quality testing and purification. Reproduced with permission.¹⁹⁷ Copyright 2022, Wiley-VCH. (E) Self-powered, rechargeable volatile organic compounds detection. Reproduced with permission.²⁰² Copyright 2023, Wiley-VCH. (F) Ultra-fast, highly sensitive airflow detection. Reproduced with permission.²⁰³ Copyright 2021, American Chemical Society. (G) Self-powered generator for underwater positioning. Reproduced with permission.²⁰⁴ Copyright 2023, Elsevier.

device not functioning properly.^{186–188} Self-powered generators enable smaller, smarter and, most importantly, more efficient devices than active devices, harvesting ambient energy and converting it into electricity without the need for an external power source.¹⁸⁹ These features enable them to detect various environmental factors in real-time in complex environments.^{190–197}

Humidity sensors have important applications in environmental and physiological monitoring. Combining humidity sensors with bluetooth wireless transmission devices enables the design of integrated humidity sensing devices.¹⁹⁸ This humidity sensing device has a special humidity sensing electrolyte that generates its own voltage when the humidity changes (Figure 8A), eliminating the need for an external power supply. Integrating sensors into clothing is certainly a very attractive way to do this. However, sensing units integrated into clothing can easily fall off or inconvenience the body's own movement. Self-healing thermoelectric fiber-based temperature sensors (Figure 8B) can be prepared using a coaxial wet-spinning strategy.¹⁹⁹ These fiber-based sensors exhibit accurate temperature sensing between 200°C and 400°C, and the fiber structure is well suited for integration into firefighting suits to ensure the safety of firefighters when operating at fire scenes.

Smart bracelets are a very suitable application scenario for wearable device integration.²⁰⁰ It has become very convenient and trendy to integrate and process some sensing units (light-sensitive units, gas-sensitive units, etc.) into a bracelet. A UV-detecting bracelet has been designed by integrating a chalcogenide film with excellent UV-detecting properties and commer-

cially available soft circuits.²⁰¹ The bracelet is capable of detecting indoor and outdoor UV radiation around the clock and incorporates the necessary environmental information (Figure 8C), which can be displayed and recorded on a smartphone, to prevent prolonged UV exposure. Or a multilayered gas-sensitive film designed in the shape of a watch that can be worn on the wrist to detect ambient NO₂ levels (Figure 8E).²⁰² Some of the sensors even have the ability to purify the air. By combining the high-voltage output of TENGs and the high-current output of electromagnetic generators (EMGs), an all-in-one sensing device with the dual functions of air quality monitoring and air purification at the same time (Figure 8D) can be achieved.¹⁹⁷ Driven by airflow, the designed hybrid nanogenerator can release negative air ions and continuously power an air quality detector, which is important for sustainable environmental management.

With the rapid development of environmental sensor devices, sensors are beginning to require smaller detection limits and faster response times to accommodate the need for fast and accurate detection. Inspired by hairs, an array of ultra-long silicon nanowires (Figure 8F) can rapidly and accurately detect the flow rate of gases with record response time (< 40 ms) and extremely low detection limit (< 0.15 m/s).²⁰³ In addition, more extreme application environments are beginning to be considered by researchers. An angle-resolved triboelectric nanogenerator (AR-TENG)-based device for precise underwater positioning has been developed.²⁰⁴ This device has a vibration

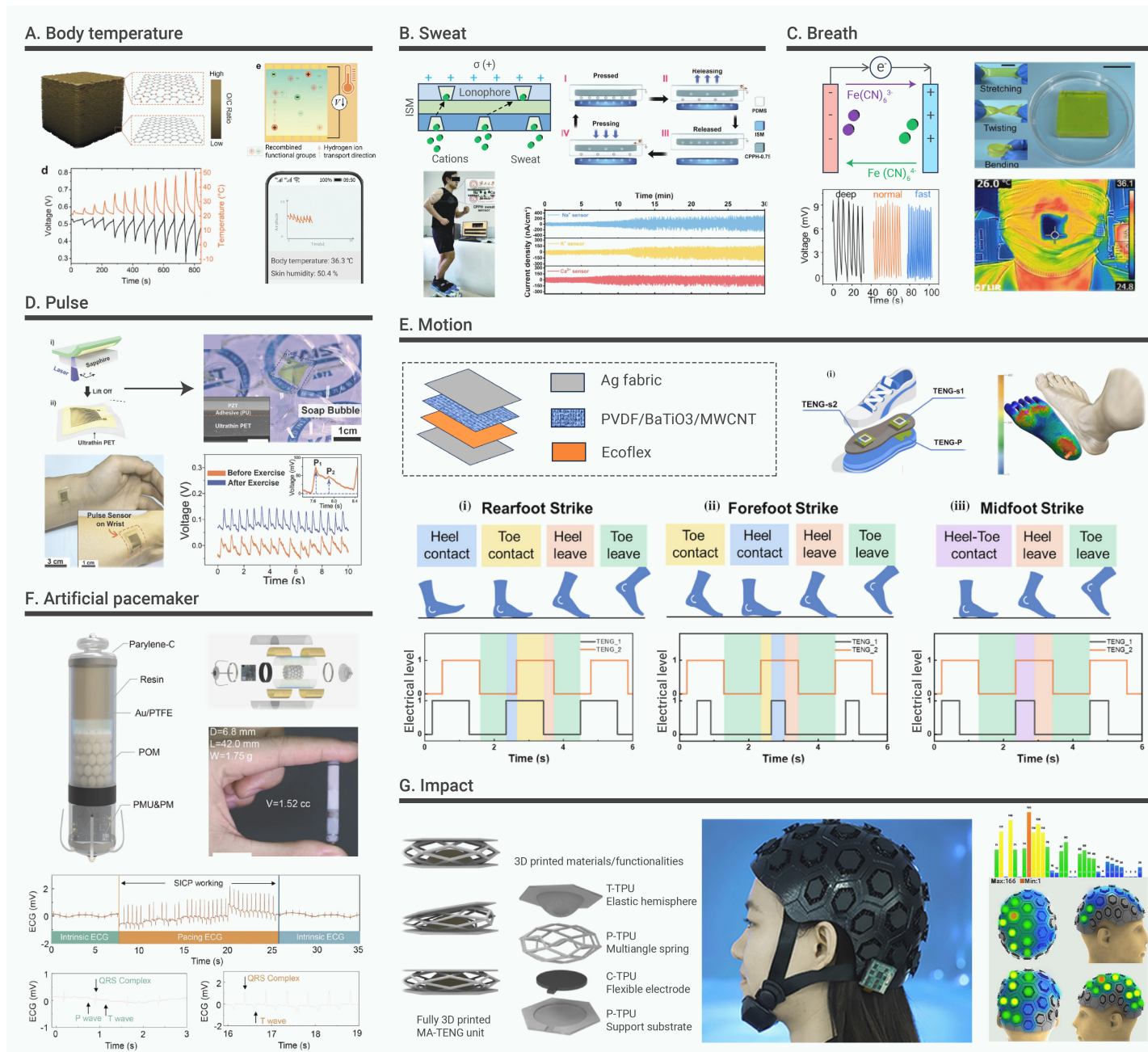


Figure 9. Application of self-powered wearables in human body field (A) Wet electrically driven graphene oxide for monitoring body temperature changes. Reproduced with permission.²¹⁸ Copyright 2022, Wiley-VCH. (B) Stretchable self-powered sweat sensor made of nanocellulose hydrogel. Reproduced with permission.²¹⁹ Copyright 2022, Wiley-VCH. (C) Dual network thermogalvanic hydrogels for self-powered respiratory monitoring. Reproduced with permission.²²⁰ Copyright 2022, American Chemical Society. (D) Self-powered ultra-thin piezoelectric sensors for pulse detection. Reproduced with permission.²²¹ Copyright 2017, Wiley-VCH. (E) Self-powered gait sensing analysis system for motion detection. Reproduced with permission.²²² Copyright 2023, Wiley-VCH. (F) Self-powered miniature artificial pacemaker. Reproduced with permission.²²³ Copyright 2023, Springer Nature. (G) Sensors array for head impact monitoring. Reproduced with permission.²²⁴ Copyright 2023, AAAS.

direction recognition capability with an angular resolution of 15° . Thus, underwater vibration localization can be achieved by simply intersecting the vibration directions determined by two parallel aligned AR-TENGs respectively, with a localization accuracy of 95%, which is very attractive in the field of deep-sea exploration (Figure 8G).

Self-powered sensors are growing rapidly in the field of environmental detection. With the discovery of new materials, engineering techniques and sensing mechanisms, it is possible not only to extend the sensing capabilities of self-powered sensor-based sensors, but also to consolidate the real-world strength and stability of their output signals for detection and applications in more extreme environments, such as the deep ocean and space. How to achieve more stable power generation, more efficient signal transmission, and lower cost for commercialization are pressing issues for self-

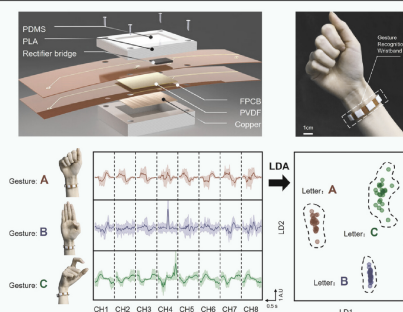
powered devices.

Human body applications

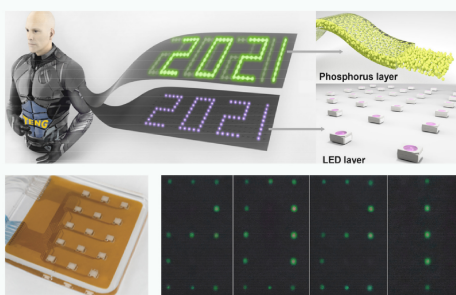
Early warning and treatment of many diseases require continuous physiological monitoring and intervention. Multi-modal, non-invasive access to the body can be achieved through a dermal electronic/optical/fluidic interface.^{205–208} Wearable electronic devices are devices that can be worn or work with the human skin to closely monitor an individual's activity on a continuous basis without interrupting or restricting the user's movement.^{209,210} This technology is essential to enable everyday physical healthcare as well as personalized medicine.

Self-powered technology takes remote healthcare enabled by sensing devices to another level.²¹¹ Recent exciting advances have demonstrated the

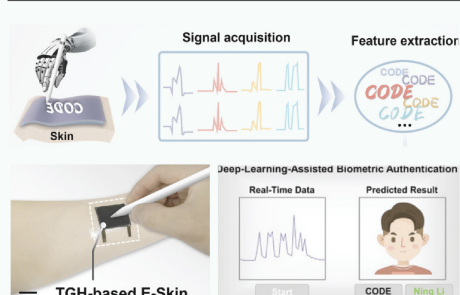
A. Gesture recognition



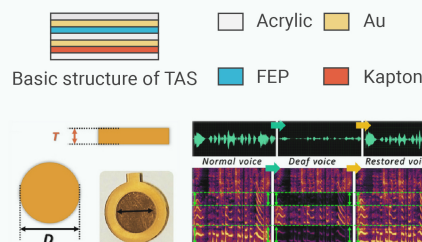
B. Afterglow display



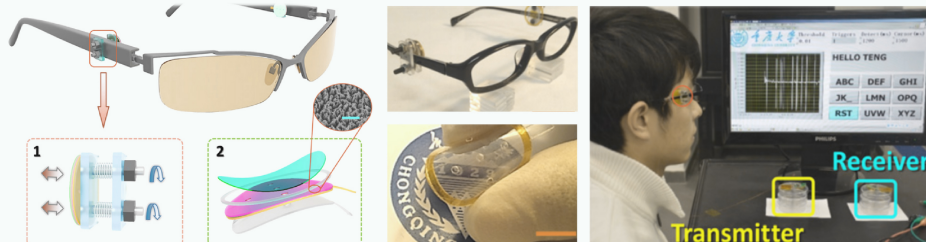
C. Signature recognition



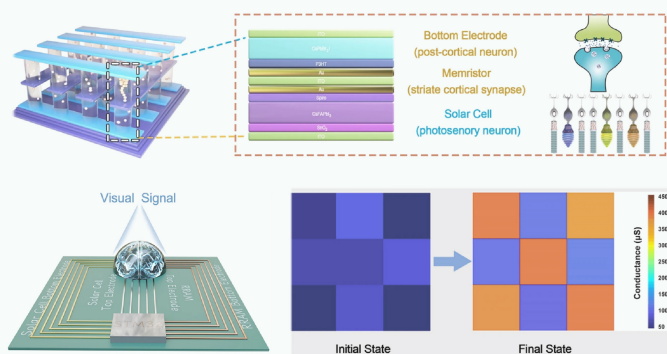
D. Speech recognition



E. Electroocular system



F. Memristor



G. Robotic

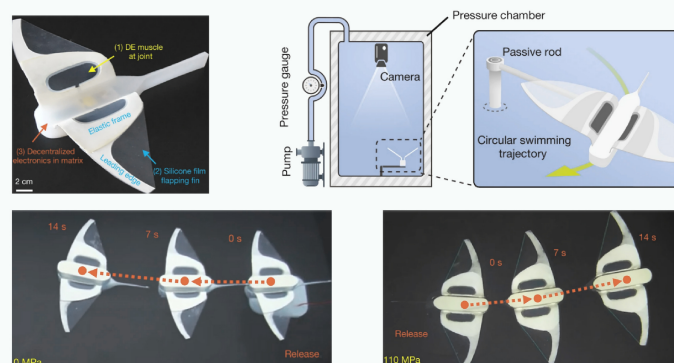


Figure 10. Application of self-powered wearables in human-computer interaction field (A) Gesture recognition wristbands enabled by machine learning. Reproduced with permission.²³⁴ Copyright 2022, Wiley-VCH. (B) Self-powered afterglow emitter for display. Reproduced with permission.²⁴⁰ Copyright 2021, American Chemical Society. (C) Deep learning-assisted e-skin for self-powered signature recognition. Reproduced with permission.²³⁵ Copyright 2023, Wiley-VCH. (D) High sensitivity self-powered auditory sensors. Reproduced with permission.²³⁸ Copyright 2018, AAAS. (E) Electric eye sensor communication system. Reproduced with permission.²³⁹ Copyright 2017, AAAS. (F) Self-powered array memristors for optically encoded pattern recognition. Reproduced with permission.²⁴³ Copyright 2022, Springer Nature. (G) Self-powered soft bionic robot for deep-sea exploration. Reproduced with permission.²⁴⁴ Copyright 2020, Springer Nature.

unique benefits of battery-free sensing in wireless wearables on a number of well-designed devices that accurately and consistently monitor all types of physical conditions in the human body, including, but not limited to basic physiological parameters such as body temperature,^{212,213} perspiration,^{214,215} and heart rate.^{216,217} But sadly, the efficiency, stability and sustainability of the power generated by these devices are hardly satisfactory.²¹⁸ By adsorbing water molecules in the air, the developed graphene oxide sensor can work continuously (Figure 9A). Water molecules have a unique response behavior when the sensor is exposed to different stimulations. With the aid of machine learning, the body temperature, as well as a variety of other physiological parameters, can be continuously monitored. A flexible, self-healing, highly conductive hydrogel electrode was prepared as a self-powered sweat sensor by encapsulation followed by assembly with an ion-selective membrane (ISM).²¹⁹ This self-powered sensor, driven by a TENG, is able to visualize during exercise and can quantify the ionic concentration of sweat (Figure 9B). As a low-grade sustainable heat source, the residual heat from the body's exhaled breath produces temperatures higher than the ambient temperature. Converting this heat into electricity to be used as a power source or to detect signals is extremely important in cutting-edge wearable medicine. The use of thermoelectric hydrogel patches allows for simultaneous thermoelectric

conversion and respiratory monitoring, and the dual-network polymerized hydrogel is resistant to freezing and drying, which is not the case with ordinary hydrogels (Figure 9C), and works well in low temperatures.²²⁰ Pulse signals are usually detected by piezoelectric sensors. The prepared piezoelectric modules (Figure 9D) are integrated onto ultrathin plastics that can be fully adhered in human skin in response to small pulsed changes occurring on the surface of the epidermis.²²¹

In addition to some basic physiological signals, self-powered wearable sensors are capable of detecting morphological changes in the human body during movement. It is worth mentioning that such sensors, by introducing some elastomer units, not only do not have an impact on the person's own movement, but are also able to increase the comfort level of the human body. For example, by combining a generator with an insole, not only is it able to detect pace and power through the combined TENGs (Figure 9E), but the additional introduction of Ecoflex elastomers also adds to the human experience and comfort level.²²² Self-powered devices still have a bright future in the biomedical field. Harvesting biomechanical energy from cardiac motion is an attractive power source for implantable bioelectronic devices. Combining the coupling effects of friction electricity and electrostatic induction has led to the design of a battery-free, transcatheter, self-powered intracardiac pace-

maker (Figure 9F) that can be used to treat cardiac arrhythmias in large animal models.²²³ This self-powered pacemaker will have a great opportunity for human application. External impacts are often unforeseen, and minor concussions can easily occur after receiving a blow to the head, with possible long-term cognitive, emotional, and physical sequelae. Zu et al. have designed a self-powered sensor that can prevent minor concussions.²²⁴ The sensor array is designed in the form of a helmet (Figure 9G), which can also protect the head to some extent and mitigate the effects of impact.

For now, most wearable for body monitoring sensors do serve a purpose. However, the human body is in a very complex environment, and incorrect signal acquisition and processing can even cause some danger to the human body. How to accurately collect, analyze and process signals is an important issue that needs to be addressed in the commercialization of wearable sensors.

Human-computer interaction

With the advent of the artificial intelligence era, generators have been given more cross-generational significance, and many futuristic applications have emerged, which are mainly reflected in human-computer interaction (HCI).²²⁵⁻²²⁸

Virtual reality (VR) provides users with an immersive experience by simulating specific scenes in reality, is a brand-new technology that can be applied extensively.²²⁹⁻²³¹ VR technology relies on human-computer interaction devices, these active devices are usually large in size, single-function, and require strong arithmetic and energy support for complex scene processing.^{232,233} Combining generators and machine learning techniques can give rise to unexpectedly small passive VR devices. Specifically, the system includes a smart wristband, an eight-channel Bluetooth module, and a mobile computer terminal (Figure 10A).²³⁴ This integrated device uses an integrated array of PENGs and TENGs sensors as inputs. Gesture inputs were recognized using linear discriminant analysis (LDA) model training with a prediction accuracy of 92.6%. This self-powered small HCI device has a very high potential for future applications. Similar to wearable HCI wristband systems, this construction is also suitable for gesture recognition, voice recognition and other scenarios. Passivation and flexibility allow the system to fit more closely to the human skin and adapt to more complex and changing scenarios. Ionic hydrogels with high electrical conductivity and resistance to freezing are preferred for the construction of novel electronic skins (Figure 10C). Interestingly, the inherent thermo-current effect of this hydrogel receives pressure and changes, and handwritten signature features can be recognized with high accuracy by assisted techniques of deep learning.²³⁵ This advanced technology will play an important role in the field of information security.^{236,237} For human-computer interaction, the ear is one of the most prominent interaction channels. Guo et al. developed a self-powered auditory sensor (Figure 10D) for building external hearing aid architectures for electronic hearing systems and intelligent robotics applications.²³⁸ This TAS sensor is prepared layer by layer based on TENGs technology. The devices are small in size and can even be transparent in appearance, and can be well suited for use in the electronic hearing systems of intelligent robots. For the visual construction of HCI, the electro-ocular system is extremely attractive.²³⁹ By fixing flexible self-powered electro-ocular sensors to glasses, home appliances and wireless typing can be controlled by simple signal processing circuits (Figure 10E), showing great potential for application in HCI.

Self-powered devices can be found everywhere in a number of displays, memory devices, and even robotics, as opposed to the classic exploration of human-computer interaction devices. Combining generators and LEDs can light up afterglow materials with 100% exciton utilization.²⁴⁰ Whereas TENGs currents normally produce transient light and thus lead to incomplete information, the afterglow material system achieves a complete display of information, broadening the use of TENGs in displays and visualization tracking (Figure 10B). Unlike a display system, a photoreceptor system is a photoelectric conversion device, similar to the sensory field of the human eye. Inspired by it, many machine vision systems have been researched, but still cannot process every part of the visual pathway in depth.^{241,242} Stacking solar cells on top of a memristor (Figure 10F) to form a cross-processor leads to the construction of a self-powered bionic optical synaptic system.²⁴³ Such systems can overcome the challenges faced by conventional computer

vision and contribute to future fast edge detection. Both sensing and display devices are currently struggling to play a significant role in extreme environment detection. Extremely high pressures in the deep ocean, the largest uncharted territory on the planet, make it difficult to explore with existing equipment. Inspired by deep-sea snailfish, Li et al. developed a self-powered soft robot for deep-sea exploration.²⁴⁴ Incredibly, the self-powered robot, which does not require any rigid container, was successfully activated in the Mariana Trench at a depth of 10,900 meters and was able to swim freely under high pressure (Figure 10G). This design can be extended to a wide range of soft devices, facilitating applications in many directions.

In the future, more self-powered devices are expected to be used in the field of HCI. However, there are still several issues that need to be focused on. Firstly, a more stable and efficient energy collection device needs to be designed. Secondly, increase the transmission efficiency of HCI. Finally, optimize the experience in the actual process of HCI.

THE BRIGHT FUTURE OF THE WEARABLE SELF-POWERED ELECTRONICS

Wearable self-powered electronics, which harness energy from environmental and physiological sources, have garnered significant attention as an emerging technology with immense potential. In this review, we have discussed three aspects of this topic: (1) Different principles of self-powered generators (solar cells, MEGs, TEGs, TENGs, PENGs, and BFCs as well as synergetic energy harvester systems) and their latest developments (structure type, common materials, and optimization strategies); (2) Application scenarios for wearable self-powered devices in environmental monitoring, human body applications, and human-computer interaction; (3) Challenges and possible solutions for wearable self-powered electronics in practical applications.

Wearable self-powered electronics development is still in the early stages, on the road of future development, opportunities and challenges coexist. We propose the following prospects for different directions:

Mechanisms understanding: The core mechanism of wearable self-powered electronics is to effectively obtain energy from the environment or human body using different materials. Starting from the source of energy, we need to enrich the variety of self-powered harvesters available for selection. From the mechanism of energy conversion, it's necessary to further explore in order to maximize the efficiency of energy conversion. Furthermore, exploring effective synergistic complementary power generation mechanisms is also important for the uninterrupted operation of wearable self-powered electronics.

Materials development: For wearable self-powered electronics, it's crucial to develop the soft materials with high stability, excellent mechanical performance, long service life, low cost and large-scale production, while ensuring that they can be used for high-power energy harvesting devices. For instance, perovskite has become a popular material for solar cells due to its low cost and wide spectral absorption range, but there are still some challenges to large-scale production. In terms of piezoelectric materials, because of high piezoelectric coefficient and flexibility, PVDF has become a hot material for PENG, we need to ensure its flexibility and stability while further enhancing its piezoelectric properties. In addition, the biocompatibility and comfort between materials and the human body should also be considered, including stretchability, environmental friendliness, and breathability.

Device fabrication: For device processing, we need to ensure the consistency of the devices during the processing, which requires standardized and scaled manufacturing processes and testing flow. Some emerging device manufacturing technologies, such as advanced printing, laser writing, and roll to roll processing, are crucial for the large-scale fabrication of miniaturized and integrated devices. In the structural design of devices, most wearable self-powered electronics adopt a multi-layer integrated structure design. In this structure, the relatively thin functional layer will bear greater strain than other parts, which may lead to interface failure or film rupture. Besides, different energy harvesters have different requirements for the thickness of active materials. Therefore, the design of the devices must take into account the differences in materials.

System integration: Due to the fact that the power output generated by self-powered systems in practical applications is usually discontinuous and fluctuating,

tuating, we need to attach great importance to power management circuits, which play a crucial role in adjusting output, energy storage, and providing stable power to the entire system. Furthermore, transmission circuits and signal processing circuits are also important for the effective operation of the entire system. However, it's a challenge to design compatible flexible circuits and we need to consider the power consumption of the whole circuit, and internal resistance and leakage should also be minimized as much as possible. Finally, we should also develop new system integration and packaging methods.

Application prospect: In the future, the combination of AI and wearable self-powered electronics has a very broad prospect. For instance, wearable self-powered electronics can monitor real-time physiological data of users, such as heart rate, blood pressure, blood sugar, etc., and combine AI algorithms for data analysis to provide health guidance and advice to users. It can also obtain user's exercise data, which can be processed and analyzed by AI algorithms to monitor the user's step count, running distance, calorie consumption, etc. in real time, and provide corresponding exercise suggestions and training plans.

In short, with the advancement of new processing technologies and micro/nano electronics, wearable self-powered electronic products have a bright future. We need to make efforts from mechanism understanding, material design, and system integration to ensure that the electronics can operate stably and efficiently in practical applications, providing users with a more reliable wearing experience.

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AUTHOR CONTRIBUTIONS

Z. Yao and Z. Guan contributed equally to this work. Z. Wu and Z. Hu supervised the project. Z. Yao and Z. Guan co-wrote the manuscript. Z. Wu and Z. Hu conceptualize, review and edit the manuscript. S. Zhang, J. Zeng, and X. Zhang gave advice on the manuscript. All authors reviewed and approved the final manuscript.

DECLARATION OF INTERESTS

The authors declare no competing interests.

DATA AND CODE AVAILABILITY

No new data and code were created or analyzed in this review.