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Self-powered skin electronics for energy harvesting and healthcare monitoring

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ABSTRACT

Self-powered skin electronics capable of energy harvesting and health monitoring is being regarded as the next-generation wearable system, with broad applications both for academic research of artificial intelligence and clinical practice of healthcare medicine. Demonstrated examples of self-powered skin electronics involves various types of devices, associated with energy conversion from piezoelectricity, triboelectricity, biofuel cell, photovoltaics, and thermoelectricity into electrical power source. These systems are of particular interests because they can intimately conform to the surfaces of skin with great flexibility and stretchability, and serve as wearable electronics. In this context, intrinsically stretchable materials and advanced device designs are in development to establish functional, high-performance interfaces with the skin. This review summarizes the most recent advances in this field, with highlight on constituent materials, device configuration, system functionality and integration methods in the past 5 years. Besides, subsequent section includes the related applications of these electronic platforms, with emphasis on energy harvesting and healthcare for human body and animal models. Toward the end of the article, the challenges and opportunities for self-powered skin electronics are discussed, offering information and research ideas for readers. These advances establish the foundations for self-powered devices in electrical skin interfaces of the future, where these advanced technologies suggest broad relevance to diverse skin-integrated electronics.

1. Introduction

1.1. Background introduction

Skin, the largest and most visible organ in the human body, plays important roles in perceiving external signals (such as thermoregulatory, touch, pressure, and vibration) and protecting the internal organs that may suffer from pathogens of the environment. The utilization and mimicry of skin via modern electronics technologies is an innovative research topic called 'skin electronics'. As early as the 1980s, Hewlett-Packard Company launched the first personal computer with an integrated touch screen [1], as the first time that electrical devices were involved to achieve intuitive touch that simulated the human skin. Material selection for these electronics at early stage refers to flexible polymers that serve as device substrates, following with various soft materials used as electrodes (such as carbon-based, liquid metal, conductive hydrogel), active layers (such as piezoelectric ceramic and organic photovoltaics), and encapsulation layers (such as peucine and silicone). In the recent decades, the research of skin electronics has acquired prominence, because these and other related systems are distinguished relative to technologies of the past by their compliant architectures and low bending stiffness as minimally invasive interfaces to curved, soft, and dynamic skin surfaces [2,3]. Many innovations have also emerged in the sensing/actuating function of the skin electronics and sophisticated high-performance platforms of these types are increasingly well established. Embodiments

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range from precise measurements of pressure and temperature for skin function, to sweat sensing for human body and to continuous real-time monitoring of healthcare. Of particular interests are advanced platforms that establish large-area, high resolution and multifunctional flexible integrated interfaces intimately coupled to skin, with stable operational functions similar to or even beyond skins [4–7]. Rapid advances in materials science and microfabrication technologies are facilitating the development of skin electronics that provide highly comfortable interfaces on human skin, creating new opportunities in the research study of biomedical engineering, in physiological signals tracking for fitness applications, and in the examination and treatment of clinical medicine. The burgeoning skin electronics are changing the way we interact with the real world, opening new markets in artificial intelligence and personalized healthcare monitoring and advancing us understand of human physiology. In the future, development of skin electronics that is self-powered, self-healing, printable, transparent, wireless communication, and biocompatible are highly essential [8,9]. These works present imagination and creativity, and will greatly expand the application potential of skin electronics.

1.2. The advantages of self-powered skin electronics

The development of a sufficient and stable quantum of energy supply for skin electronics is vital to drive device operation reliably and continually. Primary cell and rechargeable battery are the most attractive choices in terms of their portable platforms [10]. However, challenges remain in applying these batteries in skin electronics due to the following reasons: (1) A minor explosion may occur when the battery is overheated, thus inducing injuries to human body. Any related potential hazards must preclude their use in human-related devices. (2) The thicker volume and rigid format is fundamentally mismatched to the soft, curved skin surfaces while undergoing mechanical deformation such as stretching and twisting. In this context, the electronic devices must be flexible, stretchable and lightweight, in ultrathin film forms that are similar to the epidermis and capable of seamlessly coupling with skin, offering electrical performance characteristics that can approach those of conventional wafer-based devices. To solve the problem for this purpose, the concept of a self-powered electronic system was proposed by Zhong Lin Wang et al. [11]. They proposed an aligned arrangement of zinc oxide nanowires (ZnO NWs) array that converted mechanical energy into electronic energy, enable supply energy source for nanoscale electronics and make the devices operate independently. Besides, mechanical energy [12,13], biomass [14], chemical [15], solar [16], thermal [17], wind [18] and tidal [19] energy are potential green energy sources that facilitate self-powered devices. Recent studies on self-powered electronics provide additional options for energy harvesting in devices and systems in a biocompatible, safe fashion, not only at the level of functional materials but also in terms of design architectures with mechanical compliances and energy conversion mechanisms [20,21]. The development of materials and devices for these self-powered devices are valuable, where these active systems demonstrate the rapid expansion of research emphasis in biomedical engineering. Based on the above discussion, the advantage of self-powered skin electronics is manifest and can be summed up as following: (1) Highly secure and environmentally friendly. Compared with intense chemical reaction and possible leakage in primary cell and rechargeable battery, self-powered skin electronics convert green energy harvested form surrounding environment or human motions into electrical energy in a mild way, which greatly improve the security of biosystem; (2) Durability and cost-effectiveness. It is well known that primary cells can only be used once, whereas the recharging cycle is limited in rechargeable batteries (on the order of tens to hundreds times). Studies have verified that self-powered electronics like triboelectric nanogenerator (TENG) and piezoelectric nanogenerator (PENG) can undergo more than thousands of times of deformation, demonstrating the long usage time and reduce the fabrication cost; (3) Flexibility and wearability: the thin volume and simple structure afford the flexibility and wearability in electronic systems; (4) Biocompatible and implantable. Most materials in self-powered skin electronics are biocompatible that made it possible to promote *in vivo* implantation.

1.3. Scope of the review

In this review, progress in respect of functional materials, device configuration, advantages and advanced applications for self-powered skin electronics are summarized in Fig. 1. The article begins with the background introduction of self-powered skin electronics. The following two to six sections describe the devices based on these concepts of energy conversion strategies, involving the usage of piezoelectricity, triboelectricity, biofuel cell, photovoltaics, and thermoelectricity. Representative examples highlight the key scientific issues tackled in these researches, with a focus on the relationship between electrical performance and mechanical flexibility performance. The review ended with an overview of key challenges and a summary of opportunities in the future directions for self-powered skin electronics.

2. Piezoelectricity

Piezoelectric nanogenerators (PENGs) that enable mechanical energy converted into electrical energy based on the piezoelectric effect, are a prominent candidate for energy harvesting and self-powered human-machine interface systems. In this section, we will first give a description of the working mechanism and then review the recent progress in development of piezoelectric materials according to their chemical structures, as well as the related applications in self-powered skin electronics including as energy source and biosensors.

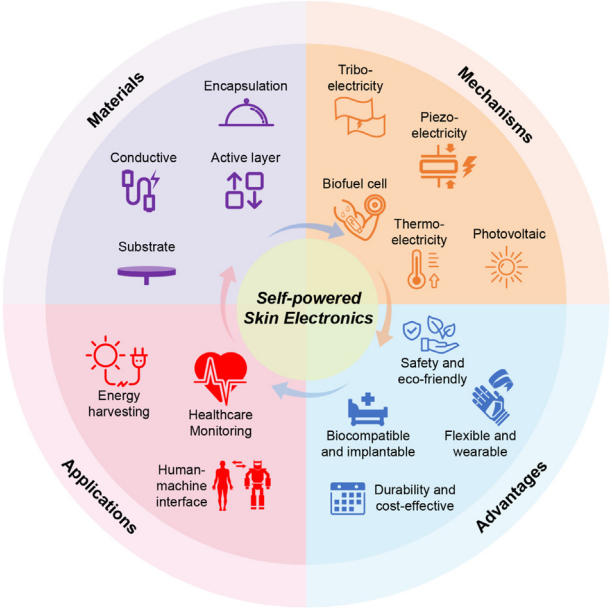

Fig. 1. Schematics of self-powered skin electronics in the aspects of materials science, device mechanism, practical applications, and outstanding advances.

2.1. Working mechanism of piezoelectric nanogenerators

The piezoelectricity refers to the mutual conversion of mechanical energy and electrical energy, where polarization of the piezoelectric materials requires high voltage as such they exhibit piezoelectric property (Fig. 2a). In 1880, Pierre Curie and Jacques Curie first discovered the piezoelectricity in tourmaline, and they verified the inverse piezoelectricity through experiments and obtained the forward and inverse piezoelectric constants. In essence, piezoelectricity is a manifestation of the piezoelectric potential generated after the central symmetry of the crystal structure destroyed by external mechanical force and a dipole polarization occurred inside. Therefore, the symmetry of the crystal structure determines whether the material possesses the piezoelectricity. As an example of wurtzite-structured zinc oxide (ZnO) [22], there is no polarization in the original state due to the overlap of the effective centers of tetrahedrally coordinated Zn^{2+} and O^{2+}. Application of external stress on the surface of ZnO yield that the two centers move in opposite directions, generating an electric dipole moment in the force direction. Then the consecutive accumulation of dipole polarization results in a macroscopic piezoelectric potential (Fig. 2a). For a deformed crystal connected with external loads, the free electrons are driven to flow through the circuit and screen the piezoelectric potential, regaining an electric equilibrium state, thereby yielding the process of energy conversion (Fig. 2b). Piezoelectric nanogenerators (PENGs) are based on the coupling of piezoelectric materials and flexible technologies, where sensitive material selections are the key factors for the development of PENG technology.

2.2. Functional materials and applications

The commonly used piezoelectric materials in skin electronics are one-dimensional (1D) ZnO nanomaterials [11], two-dimensional (2D) transition metal dichalcogenides (TMDCs) such as molybdenum disulfide (MoS_2) [23–25] and hexagonal form of boron nitride (h-BN) [26], three-dimensional (3D) piezoceramic such as lead zirconate titanate (PZT) [27,28], and polyvinylidene difluoride (PVDF) [29] and its similar copolymer poly(vinylidene fluoride-trifluoroethylene) P(VDF-TrFE) [30], as displayed in Fig. 2c.

(1) **ZnO**. In 2006, Zhong Lin Wang and his colleagues first developed an aligned arrangement of ZnO nanowires based-PENG due to the following three key advantages of ZnO among all the known 1D nanomaterials [11]:(1) ZnO exhibits both piezoelectricity and semiconductor characteristics, which lays the basis for electromechanically coupled transducers and sensors; (2) the abundant nanostructures of ZnO facilitate to meet various performance requirements; (3) the relative biocompatibility of ZnO enables the applications in biosensors. This report provided a new methodology for harvesting mechanical energy (i.e. joint movement, heartbeat, and pulse) and converting it into electrical energy. Thereafter, studies on ZnO-based PENG and their applications such as pressure sensors have been conducted. To obtain higher electric output, ZnO film was designed in a texture formation and coated on the surface of carbon fiber [31]. The flexibility of the carbon fiber induces the effective delivery of the deformation, and the textured-structure of ZnO enhance the electrical performance, so an output voltage of 3.2 V and a current density of 0.15 μA/cm^2 were obtained. To explore a wearable and sustainable device that works at low frequencies and can scavenge mechanical movements from human activities, ZnO nanowires (ZnO NWs)/PVDF hybrid piezoelectric material was combined around a conductive fiber to build a hybrid-fiber TENG. Attaching this hybrid-fiber generator with a 2 cm-length on a human arm, the current density of 10 nA/cm^2 and power density of 16 μW/cm^2 were obtained at folding-releasing the elbow about 90°. Besides energy harvesting, ZnO-based PENG also has been applied in health monitoring [32,33]. Li et al. implanted ZnO NWs-based PENG in a live rat to harvest the energy generated from its breathes and heartbeats, showing the great potential of sensing the low-frequency dynamic muscle motion in vivo [33].

(2) **MoS_2**. Recently, ultrathin 2D atomic-layer materials like transition metal dichalcogenides (TMDC) has aroused great research in the field of energy harvesting interest due to their unique electrical and physicochemical properties [23–25]. Taking the mostly used MoS_2 as an example, monolayer MoS_2 exhibits strong piezoelectricity owing to its broken

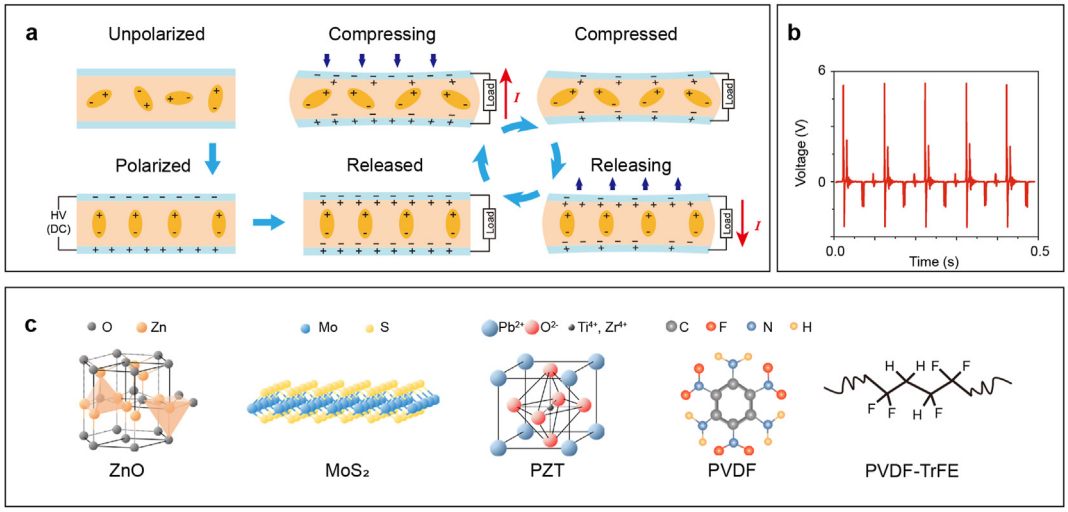

Fig. 2. Working mechanism of piezoelectric nanogenerator (PENG) and piezoelectric materials used in piezoelectricity. **(a)** Schematic illustrations of the orientation polarization of piezoelectric material and electric energy generation process of PENG. **(b)** The related electrical output performance of PENG under different deformation conditions, where positive signals are generated under compressing condition and negative signals are generated under releasing condition. **(c)** The chemical structures of common piezoelectric materials, including inorganic materials (zinc oxide [ZnO], molybdenum disulfide [MoS_2], and lead zirconate titanate [PZT]) and organic materials (polyvinylidene difluoride [PVDF] and copolymer poly(vinylidene fluoride-trifluoroethylene) (P(VDF-TrFE)).

inversion symmetry in the crystal structure, and the higher electron mobility that over than amorphous silicon and many other ultrathin semiconductors facilitates the charge transportation. In 2014, Zhong Lin Wang and his team reported the first experimental study of the MoS_2's piezoelectricity, and the experiments indicated that only MoS_2 with odd-numbered atomic layers can generate oscillating electric outputs at cyclic deformation, while even-numbered atomic layers cannot [34]. This monolayer MoS_2-based PENG could generate the electrical outputs with the power density of 2 mW/m^2 and energy conversion efficiency of 5.08%. And deep theoretical research can be found in their subsequent publication [35]. In the same year, Xiang Zhang et al. also got the same conclusion and measured the piezoelectric coefficient of $e_{11} = 2.9 \times 10^{-10}$ C/m [36]. The strong piezoelectricity and stretchability of monolayer MoS_2 show the great potential for applications in energy harvesting, pressure sensing, and self-powered wearable electronics. The chemical vapor deposition (CVD) is a better choice for fabricating large-area MoS_2, furthermore, the number of MoS_2 layers can be precisely controlled by the CVD deposition time [37]. But a large number of intrinsic point defects would generate in CVD-grown MoS_2 film that leads to reduced piezoelectric performance. To solve this problem, Sang-Woo Kim et al. proposed the sulfur (S) atom passivation strategy to fill up the point defects since the S-containing groups intend to form the covalent bonds with unsaturated S or molybdenum (Mo) vacancies [38]. Results demonstrated that the piezoelectric coefficient of the modified MoS_2 improved from 3.06 to 3.73 p.m./V that closes to the theoretical value, and the high power density of 73 $\mu W/m^2$ and mechanical stability of more than 5 h continuous operation were obtained. Moreover, all-solution-processed MoS_2-based PENG has been achieved and the long-term endurance comprising of 5000 cycles for 5 days was obtained [39]. In 2020, a new kind of TMDC material of tin disulfide nanosheets (SnS_2) grown by CVD was explored in PENG and realized the application in the smart sign language system [40]. Multiple flexible SnS_2 PENGs were assembled on a wired glove that could collect the electrical signals when fingers were in bending state, and the collected signals could trigger the robotic to repeat the motions of operator's finger in time via a designed human-machine interface system.

(3) **PZT**. PZT is a widely used piezoceramic material due to its high dielectric constant. Compared with randomly dispersed PZT piezoceramic particles, the gradient porous PZT could promote stress transfer and improve the energy conversion efficiency due to the high compressibility. Xin Cheng et al. applied the porous PZT to PENG, and obtained the output voltage up to 152 V, current up to 17.5 mA and sensitivity as high as 1.52 V/N [27]. Introducing high conductive materials such as graphene into PZT rubber is an efficient approach to realize high-performance flexible device [41]. A maximum power density of 972 $\mu W/cm^3$ when human walking and excellent deformation tolerance to stretching and twist more than thousands of cycles were obtained in this graphene-PZT-PENG. In addition to common energy harvesting, the application of tactile sensing which can be used in healthcare monitoring and physical treatment have been explored [28,42,43]. For instance, Xinge Yu et al. proposed a 10 × 10 PZT-based tactile sensor array by screen-printing method, which could rebuild the tactile information correctly and rapidly (Fig. 3a) [43]. As for healthcare examination, applications in vitro such as epidermal blood pressure and in vivo such as diseased tissue detecting have been developed. John A. Rogers et al. proposed a blood pressure measurement microsystem consisting of PZT pressure sensing elements and a SiNM *n*-MOSFET amplifier [44]. The device configuration and simplified equivalent circuit appears in Fig. 3b. When placed the sensors on the carotid artery and the lateral epicondyle vessel with a distance of 43 cm, the pulse wave velocity is calculated to be 5.4/ms with a time difference of 0.08 s. The skin-mounted pressure sensor reported here offers a promising way to long-term monitoring of cardiovascular diseases. Furthermore, they explored a needle-shaped piezoelectric microsystem for targeting biopsy *in vivo*, where the assembly of the probe refers to a simple, compact Au/PZT/Pt sandwich structure (Fig. 3c) [45]. Typically, the tissue tumor (e.g. liver suffered by malignancy) exhibit abnormally higher stiffness than surrounding healthy counterparts. Measurement examples from such probe indicate different moduli of 10 and 23 KPa obtained from human cirrhotic and healthy fresh livers by using piezoelectric microneedles This work demonstrates that PZT-based piezoelectric devices have great potential in clinical medicine and healthcare monitoring.

(4) **PVDF and its copolymer**. Polymer-based piezoelectric materials such as PVDF and its copolymer of P(VDF-TrFE) are commonly used in research study owing to their advantages of flexibility, sufficient mechanical strength, easy process and high chemical resistance [49–51]. To further improve the piezoelectric outputs, PVDF usually is fabricated with a porous structure or mixed with ZnO nanomaterials. For instance, Dayan Ban reported a porosity modulated ZnO nanoparticles/PVDF hybrid piezoelectric device, and a high peak voltage of 84.5 V and power density of 41.02 $\mu W/cm^2$ were achieved [52]. Polymer's chemical stability and mechanical flexibility are conducive to the applications in skin electronics in vitro or in vivo. Various strategies have been explored to fabricate flexible, wearable PVDF-based piezoelectric devices for applications in self-powered skin electronics. Zhongze Gu et al. integrated Cu/PVDF/Cu TENG with gecko-inspired nitrocellulose nanostructure for intimate attach and biosensing. Due to the structure of nanotentacles, the contact area increases and electrical performance strength four times than the planar one [53]. To enhance and extend the operation modes of PENG, as shown in Fig. 3d, complex 3D frameworks were fabricated by releasing the elastomer stretched beforehand that conventional planer 2D structures patterned by a lithography process and transfer process were deposited on it [46]. More than twenty different 3D geometrics were developed and demonstrated in various applications, such as energy harvesting from 2 mV to 790 mV, multifunctional sensors for human-machine interfaces with high sensitivity of 60 mV/N, biomedical implants in vivo to monitor the movement of the mouse. In aspect of human-machine interfaces, high sensitivity of 4.4 mV/degree bending from 44° to 122° and a fast response time of 76 ms were achieved by PVDF polymer [54]. Besides energy harvesting, piezoelectricity has been used to power the skin electronics, for instance, study on artificial sensory synapse developed by PENG with ion gel-gated transistor was reported. As shown in Fig. 3e, Qijun Sun et al. developed a piezoelectric artificial sensory synapse by integrating poly(-vinylidene fluoride-trifluoroethylene) [P(VDF-TrFE)]-based PENG and ion gel-gated transistor [47]. The piezo-potential acting as gate voltage modulates the current of conductive channel, meanwhile, the piezo-potential coupling is capable of linking the spatiotemporal strain information with the post-synaptic current. This work provides a new sight into skin-inspired neuromorphic computing based on piezoelectric synaptic devices. It is still a big challenge to detect and

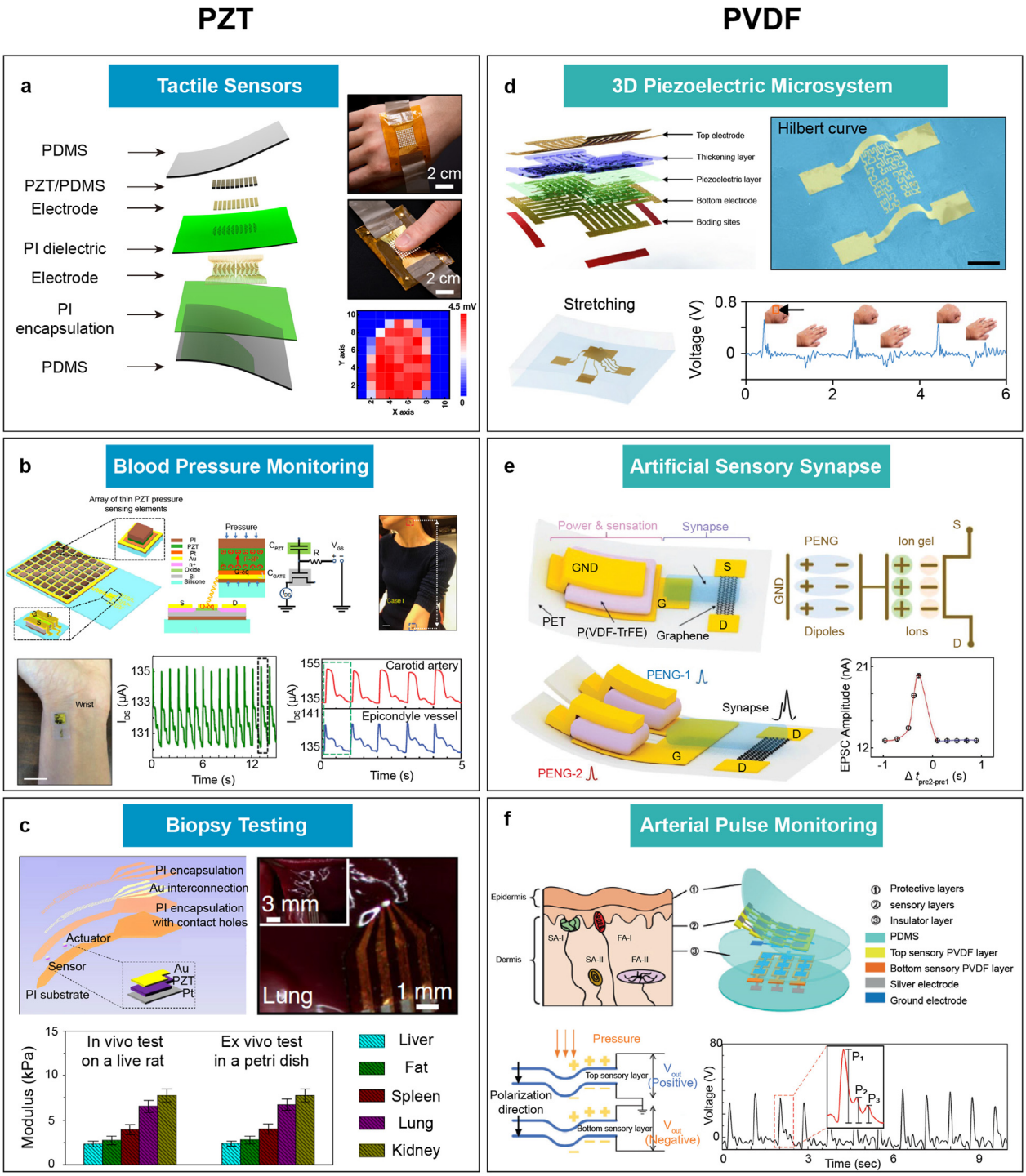

Fig. 3. Examples of applications of piezoelectricity in self-powered skin electronics. (a) The device configuration (Left), tactile sensor and related tactile signals mapping (Bottom Right) [43]. Reprinted by permission from Springer Nature. **(b)** Device construction and equivalent circuit (Upper Left), sensor placing on a human wrist and the measurement results (Bottom Left), and the applications of blood pressure (Right) [44]. Reprinted by permission from American Association for the Advancement of Science. **(c)** Device configuration (Upper Left), the device pierced into the lung of a rat (Upper Right), and the results of in vivo modulus test on a live rat and ex vivo test in a Petri dish (Bottom) [45]. Reprinted by permission from Springer Nature. **(d)** Device configuration (Upper Left), SEM image of PVDF mesostructure (Upper Right), stretching 3D piezoelectric device (Bottom Left), and the demonstration of harvesting energy from human skin (Bottom Right) [46]. Reprinted by permission from Springer Nature. **(e)** Illustration of the artificial sensory synapse and equivalent circuit (Upper), artificial neural network with two sensors (Bottom Left), and the excitatory postsynaptic current (Bottom Right) [47]. Reprinted by permission from Wiley-VCH Verlag. **(f)** Illustration of human skin structure and the skin-inspired tactile sensor array (Upper Left), the circuit diagram of the tactile sensor array (Bottom Left), and real-time detection of the neck arterial pulse (Bottom Right) [48]. Reprinted by permission from Wiley-VCH Verlag.

differentiate the external stimuli using just one single PENG sensor, which will limit the practical applications of piezoelectric tactile sensor. To solve this problem, Zhengbao Yang et al. explored a multilayer structure including two encapsulation layers (of polydimethyl siloxane [PDMS]), two sensory layers (of polyvinylidene difluoride [PVDF]) and one insulating

layer (of PDMS) to avoid the non-crosstalk with a whole device thickness less than 1 mm [48] (Fig. 3f). Consideration of press state, polarization direction and circuit connection, the electrical output of two sensory layers are opposite and the top sensor has a larger output signal, which provides a reference for different stimulus signals. Therefore, when the sensor array was attached onto human neck for detecting the subtle physiological signals, the incident wave P1, the tidal wave P2, and the diastolic wave P3 were recorded for arterial stiffness analysis. Besides, piezoelectric output signals have been used as the gate voltage in transistor for fabricating artificial sensory synapse, exhibiting great application potential in neuromorphic computing.

3. Triboelectricity

Triboelectric nanogenerators (TENGs), converting mechanical energy from frication into electrical energy based on triboelectricity effect, are another attractive example for self-powered skin electronics due to the advantages of low-cost, ease of fabrication, diverse range of material selection [12,55]. In this part, we will give a brief introduction for the energy conversion mechanism and review the latest progress in the functional materials (such as flexible electrodes and friction materials) and device structures. Furthermore, subsequent discussion for the applications of TENG in energy harvesters and tactile/physiological signal sensors follows.

3.1. Working mechanism of triboelectric nanogenerators

Triboelectrification is a universal phenomenon in our daily life, usually regarded as a negative effect to be avoided as far as possible. Although known for many years, operational mechanism has not been well established for nanogenerator technology until recent years due to more reasonable cognition and advancement of technical means. Development of TENG seeks to establish coupling effects of triboelectrification and electrostatic induction. Triboelectrification is contact-induced electrification where the material becomes electrically charged after contact with another material. The signed and the mount of charges lie on its relative polarity in comparison to the material contacted. It is generally believed that formation of chemical bond occurs on the contact surface (so called adhesion) and then charges start to transfer to equalize the electrochemical potential. Some bonded atoms tend to keep extra electrons while some tend to release electrons, then the triboelectric charges are generated on the contact interface. According to the device configuration and circuit connection methods, categorization for TENG refers to four fundamental modes (Fig. 4a): single-electrode mode, vertical contact-separation mode, in-plane contact-sliding mode, and freestanding triboelectric-layer mode. Single-electrode mode uses only one electrode and involves the ground electrode as the reference. Vertical contact-separation mode involves the relative movement of two triboelectric layers that move in a direction perpendicular to the contact interface, while the relative displacement is in the contact surface for in-plane contact-sliding mode. The freestanding triboelectric-layer mode consists of a triboelectric layer and interdigital electrodes. Each mode has various structural characteristics, corresponding advantages and disadvantages, as a result that the working mode can be selected according to the different application requirements [56].

3.2. Functional materials and devices

Many types of materials provide triboelectricity, in a broad range from skin, hair on human body to metal (such as gold, silver and copper), to inorganic materials (fabric, ceramics) to polymers (such as nylon, Kapton®, and rubber), all of which can be used as friction layers in TENG in theory. However, the high outputs of TENG strongly depend on the triboelectric polarity difference of two friction layers. In 1757, John Carl Wilcke reported triboelectric series following a trend from positive (i.e. easy releasing electrons) to negative (i.e. capturing electrons), and Zhong Lin Wang et al. also summarized the triboelectric series in their book [63]. We rebuild

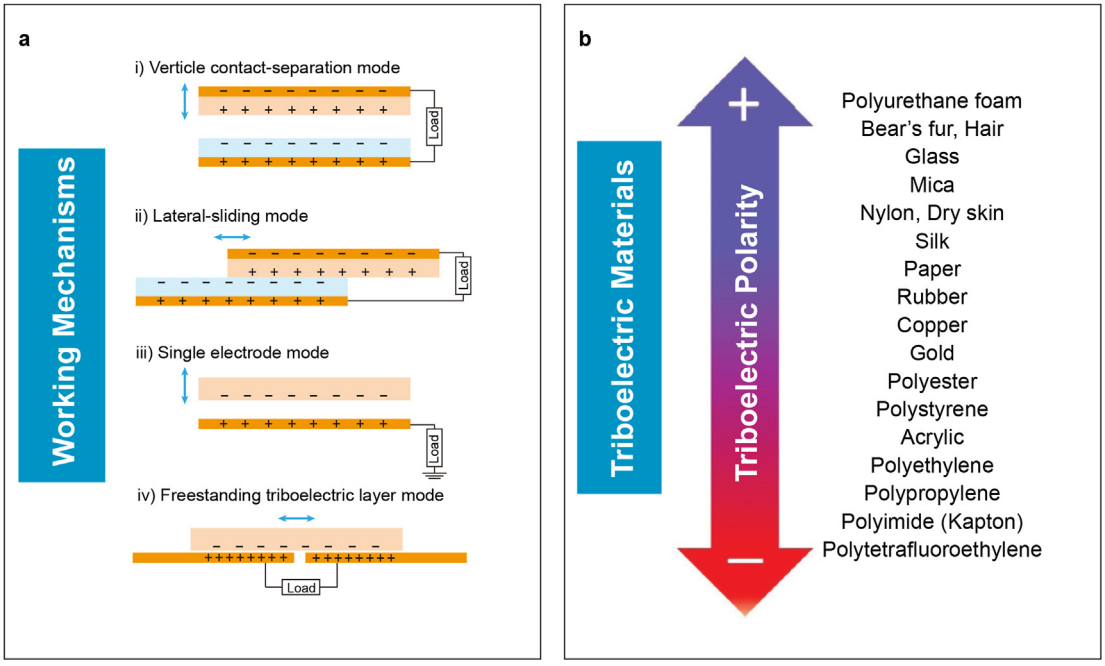

Fig. 4. Working mechanism of TENG and triboelectric materials in triboelectricity. (a) Schematic illustrations of TENG under four working modes, including vertical contact-separation mode, lateral-sliding mode, single electrode mode and freestanding triboelectric layer mode. **(b)** Common triboelectric materials listed in the manner of triboelectric polarization.

the common triboelectric series in Fig. 4b. Materials that have a strong triboelectric effect usually present low electrical conductivity or even are dielectric so that they can capture and retain the transferred charges for a longer duration. Polymers (such as polydimethyl siloxane [PDMS], polyimide [PI], and silicone) are commonly used as the triboelectric layer in soft skin electronics due to their excellent mechanical flexibility and stretchability.

To obtain higher electrical output of TENG for meeting the requirements of energy supply and sensory monitoring in skin electronics, surface modification engineering for polymer-based friction layer has been developed. Surface modification engineering refers to building micro/nanostructure to enhance the generation of triboelectric charges, including: (1) pyramids, cubes, and lines patterned by photolithography technology [64]; (2) constructing rough surface by chemical or physical methods, such as depositing nanowires on the surface of friction layer [65], or improving the aspect ratios by plasma-enhanced ion etching process [66] or laser line-scribing [67]; (3) constructing sponge/porous structures that exhibit an enhanced electrical output performance because of improved compressibility over the bulk materials [68–70]. For instance, Xinge Yu et al. proposed a water-soluble solid-template method to fabricate rough PDMS surface and sponge microstructure, and the electric output is boosted as expected [70].

The main factor limiting the flexibility of the TENG device is the conductive electrode, i.e. gold (Au) can be stretched up to 0.3% [57], which is far from satisfactory for the application in skin electronics. Tremendous efforts have been devoted to fabricating stretchable metal friction layer in TENG. As for intrinsically non-stretchable materials, the design of sophisticated geometric structure based on mechanics principles is an attainable approach [57,58,70,71]; Xinge Yu et al. designed a class of advanced cobweb-serpentine pattern for Au electrode and the TENG could achieve 30% stretch (Fig. 5a) [57]. In addition, they first systematically studied the relationship between effective working area and the electrical output performance of TENG, where the results demonstrated that electrical output reaches around saturation point when the effective working area ratio of Au electrode is above 55%. The results reveal that a balance can be achieved between output performance and mechanical stretchability, so long as the effective working area of the metal electrode is reasonably reduced and is designed as a meandering geometry. In their another research, serpentine Au electrode integrated with rough PDMS surface and cover layer for inhibiting cross-talk signals from surrounding environment to prepare highly sensitive TENG-based epidermal tactile sensor array (Fig. 5b) [58]. As for intrinsically stretchable materials, process compatibility and conductivity have been explored to meet the requirements of skin electronics. Intrinsically stretchable materials like: (1) liquid metal such as liquid gallium [72], or (2) conductive materials such as silver (Ag) Nanowires [70] or patterned gold (Au) combined [58] with elastomer are other viable options for flexible electrode in skin electronics. Furthermore, intrinsically stretchable conductive materials play a role of importance in self-powered skin electronics. For example, ionic conductors such as polyacrylamide hydrogel that contains lithium chloride acting as the ionic hydrogel and two commonly used elastomers [73], carbon grease [74], liquid metal Galinstan [75] and Mxene [76], metal elastomer [70] such as silver nanowires (Ag NWs), the semiconductor material of poly(3,4-ethylene dioxythiophene): poly(4-styrene sulfonate) (PEDOT:PSS) [77] and polymer of polypyrrole coated on cotton textile [78] have been explored for fabricating flexible and stretchable TENG.

3.3. Applications

Due to the commonness of friction in daily life and the high electrical output characteristics far exceeding other energy conversion devices, TENG involves different applications. For instance, TENG serves as the energy source to drive a microfluidic transport system and provides the control signals for the mini-vehicle [79]. Integrated TENG with the electrowetting technology, the microfluidic transport system could deliver some tiny objects whose weight less than 500 mg, demonstrating the huge applications potential in human healthcare such as controllable drug delivery. As displayed in Fig. 5c, Wei Gao et al. demonstrated the flexible interdigital freestanding TENG with a high power output of ~416 mW/m^2, sufficient for powering a multiplexed sweat biosensor and send data wirelessly to the user via Bluetooth module [59]. The freestanding TENG is based on the sliding between polytetrafluoroethylene (PTFE) and copper, and the structure consists of commercial flexible printed circuit board (FPCB) technique fabricated interdigital stator and slider. While subject human wearing this complex system is perspiring during exercise, integrated sweat sensors monitor the pH value and concentration of Na^+ in the sweat continuously without external power supply. Besides powering sensors, TENG device itself could act as a sensor as well. A biomimetic pressure sensor was constructed by sandwiching TENG within two supercapacitors, where the TENG's friction pair was composited of indium tin oxide (ITO) and polytetrafluoroethylene (PTFE). The biomimetic pressure sensor can not only store electrical energy via a wireless transfer mode but also imitate human skin to measure the dynamic pressure and static pressure via a self-powered mode and do not interfere with each other [80]. As for dynamic pressure, the detection was in a range of 1–4 N/cm^2 with a sensitivity of 17.5 $V/cm^2/N$ and limit of detection (LOD) of 0.3 N/cm^2. As a comparison, it is 0.5–4 N/cm^2 with a sensitivity of 12 $V/cm^2/N$ and LOD low to 0.07 N/cm^2 for static pressure. Furthermore, Geon-Tae Hwang et al. reported a converted magneto-mechano-TENG that convert alternating current into mechanical oscillation by magnetic force then further convert into electrical energy. The open-circuit voltage and short-circuit current reached 870 V and 145 μA when alternating current magnetic field is 7 Oe [81]. These results provided a reference for the application of TENG in artificial intelligence in terms of device configuration.

TENG-based pressure sensors also enable the monitoring of human physiological signals including arterial pulse and voice vibrations [82]. Recently, an all-textile pressure sensory array based on TENG was reported, in which nylon yarn and conductive yarn as friction pairs and the inner stainless steel were used to collect charges [60]. As shown in Fig. 5d, two sensory arrays were stitched into the cuff and chest of a shirt to monitor the pulse and respiratory signals wirelessly by Bluetooth, because of the high pressure sensitivity that was achieved. It could be useful in long-term monitoring in cardiovascular disease and sleep apnea syndrome. Furthermore, TENG has been used in self-powered highly sensitive electrotactile system for achieving haptic feedback interface in virtual reality (VR) and augmented reality (AR) [61]. As Fig. 5e displayed, the electrical signals generated from TENG were transferred into electrotactile interface, which further induce electrostatic discharge from electrotactile to skin; finally, the virtual tactile stimulations with given patterns were rebuild on skin. This system has great applications in human-machine interfaces (HMIs) such as virtual tactile displays and Braille instruction. Similarly, to improve the finger's resolution of multi-degree-of-freedom pressure, haptic-feedback smart glove combined with triboelectric nanogenerators (TENGs) and piezoelectric nanogenerators (PENGs) have been developed. Fig. 5f displays the designs of finger case of each phalanx, the positions of TENG sensors, and the related triboelectric output from 16 sensors when grabbing different objects. Through machine learning technique, this smart glove could realize precise artificial intelligence (AI)-based object recognition (96% accuracy)

Fig. 5. Examples of applications of triboelectricity in self-powered skin electronics. (a) Au electrodes with different effective working area ratios (Upper Left), device configuration (Bottom Left), optical image of device attaching onto human finger (Upper Right), and the output performance (Bottom Right) [57]. Reprinted by permission from Elsevier. **(b)** Tactile sensory array based on TENG (Upper Left), flexible PDMS film (Upper Right), scanning electron microscopy (SEM) image of rough PDMS surface (Bottom Right), and the demonstration of tactile information of City University of Hong Kong [CITYU] (Bottom Left) [58]. Reprinted by permission from Elsevier. **(c)** Schematic illustration of self-powered sweat sensor system (Left), the system wearing on human side torso (Upper Right), printed circuit board-based flexible TENG (Bottom Right) [59]. Reprinted by permission from American Association for the Advancement of Science. **(d)** Shirt with two sensory arrays assembled (Left), all-textile TENG (Right) [60]. Reprinted by permission from American Association for the Advancement of Science. **(e)** Schematic illustration of the electrotactile system (Left), exploded view of the electrode array (Upper Right), the process of electrotactile sense in system (Middle), and optical images of electrotactile array (Bottom Right) [61]. Reprinted by permission from American Association for the Advancement of Science. **(f)** Designs of finger case of each phalanx (Upper Left), demonstration of recognition six different objects using haptic feedback smart glove and machine learning technique (Bottom) [62]. Reprinted by permission from American Association for the Advancement of Science.

with triboelectric motion signals. This sensitive intelligent tactile glove shows potential in virtual reality surgical training, gaming or remote homecare/technical operations.

4. Biofuel cells

Fuel cells are green and environmentally friendly batteries that have attracted much attention in recent years. Fuel cells convert the chemical energy released from chemical reactions into electrical energy where hydrogen, natural gas, methanol, etc. acting as fuel and oxygen (O_2) acting as oxidant. The biofuel cell is a special kind of fuel cell, which uses natural microorganisms or enzymes as catalysts to complete the energy conversion process. Depending upon the type of catalyst, it can be divided into microbial fuel cells and enzymatic biofuel cells. Enzymatic biofuel cells provide access to large number of applications from the energy source to self-powered biosensors attributable to their unique advantages such as a wide range of fuel sources, mild operating conditions (room temperature and neutral pH value), high equipment safety, good biocompatibility, and low cost [83,84]. The first study on enzymatic

biofuel cell was reported in 1964, but the development was limited by the faulty electron-transfer mechanism and poor device stability. Until recent years, the rapid development of nanotechnology and biotechnology has provided a huge material foundation and reserve of knowledge and fabrication for the research of biofuel cells. In this part, we will overview the advance in electron-transfer mechanism, materials selection and output improvement strategy. Meanwhile, various applications (such as human healthcare monitoring and energy source) on self-powered skin electronics will also be summarized and analyzed, and therefore provide a fundamental understanding of the interplay between biological signals and sensor functions.

4.1. Working mechanism of biofuel cells

The enzymatic biofuel cell with bipolar chambers is composed of a cathode, an anode and a selective ion exchange membrane that use to separate cathode and anode [85,86]. As shown in Fig. 6a, the electrons generated by oxidation of these fuels under the catalysis enzyme pass through the external circuit to reach the cathode, and then the cathode accepts these electrons with the assistance of catalysis of the enzyme. This process does not involve combustion and is not limited by the Carnot Cycle, so the power conversion efficiency can be as high as 40%–60% [83,87,88]. Compared with conventional fuel cell, enzymatic biofuel cell present outstanding advantages: (1) wide range of biofuel sources and low cost materials; (2) environmentally friendly process, avoiding the introduction of precious metals, and the chemical reactions take place at

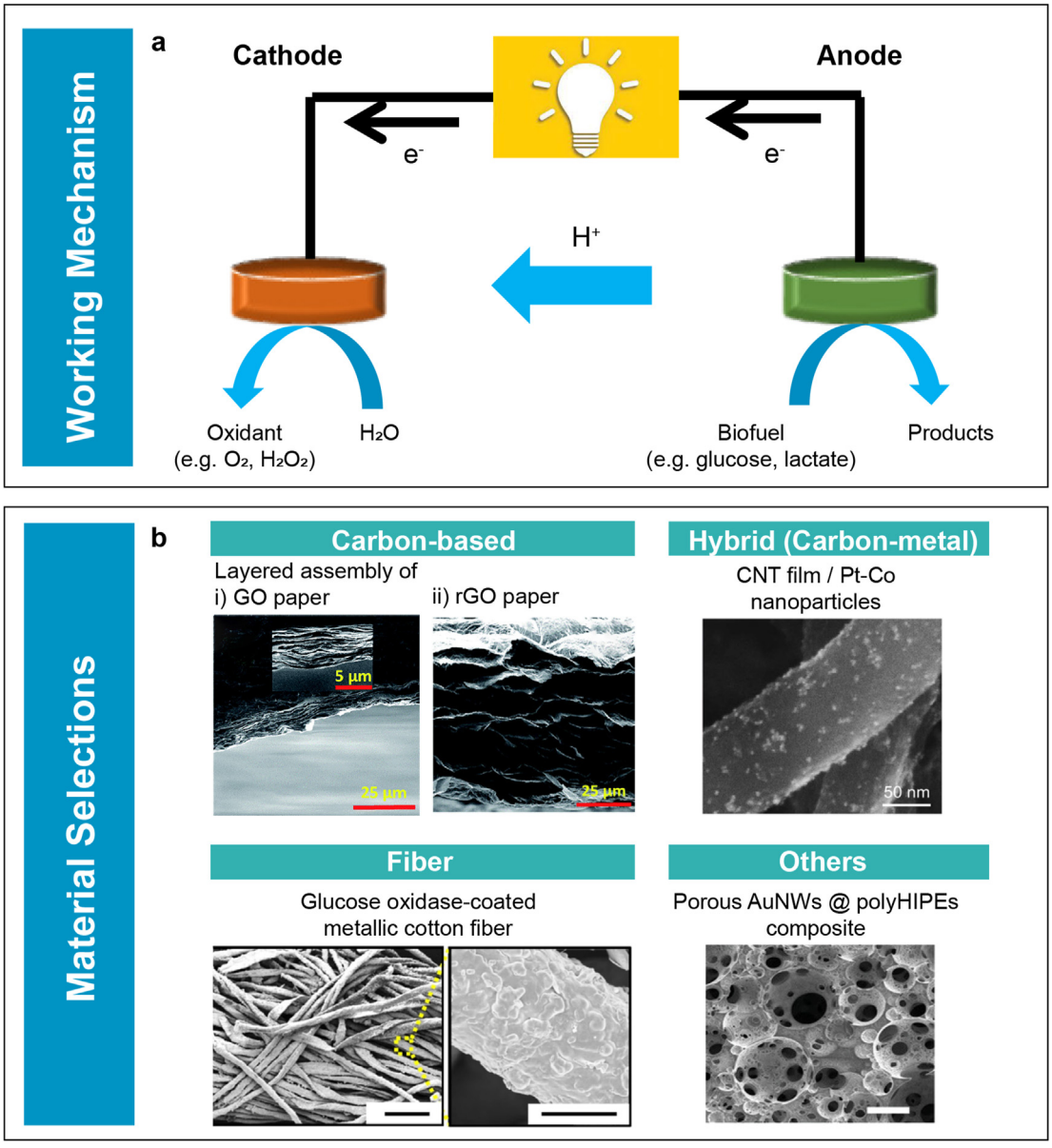

Fig. 6. Working mechanism and conductive support materials in Biofuel Cells. (a) Schematic illustration of enzymatic biofuel cell. **(b)** Common conductive support materials, including carbon-based materials (graphene oxide and reduced graphene oxide) [92] (reprinted by permission from Royal Society of Chemistry), carbon-metal hybrid materials [93] (reprinted by permission from American Association for the Advancement of Science), metallic cotton fiber [94] (reprinted by permission from Springer Nature) and others [95] (reprinted by permission from Wiley-VCH Verlag).

normal temperature and pressure, and pH value is close to 7. (3) good biocompatibility. Glucose-based biofuel cells are expected to be directly implanted in the human body as the power source for artificial organs such as cardiac pacemakers, miniature sensors and future molecular robots.

In general, electron transfer relies on the transfer rate of surface electrons and the catalytic conversion rate of enzymes. To achieve efficient electron transfer, the distance between enzymatic active sites and surface electrode should be less than 1.4 nm and the catalytic conversion rate should be in the range of $10-10^{-4}$/s [89]. Ideally, the electrons participated in the redox process can be directly transferred between active sites of enzymes and surface electrode, and the type of this electron-transfer is called 'direct electron-transfer' (DET) [90,91]. A limitation for DET is the selection of the material. When DET cannot be achieved due to the following reasons: (1) active sites are deeply buried inside the enzyme molecules, or (2) inadequate direction of enzymes, or (3) insufficient kinetics, mediators that exhibiting matched redox potential and activity are needed to assist the electrons transfer. Therefore, Biological enzymes act as catalysts and materials that widely present in nature such as alcohols, sugars, and amines act as biofuels in enzymatic biofuel cells. The commonly used catalysts for the anode are glucose oxidase, lactate oxidase, alcohol dehydrogenase, etc.

4.2. Functional materials and devices

In recent years, the development of nanoscience and nanotechnology has provided powerful platform for improving the performance of bio-electronic devices. Nanomaterials with excellent properties such as carbon nanotubes, nanospheres, graphene, conductive polymers, and nanoparticles with different sizes and compositions are used to construct the enzyme/electrode interface. Compared with traditional methods, the electrode interface modified by these materials presents high performance in terms of enzyme stability and activity. This is mainly because the introduction of nanomaterials provides a suitable microenvironment for enzyme activity and electrochemical activity and increases the electron transfer between the electrode surface and the active centers. Conductive nanomaterials can produce a relatively large specific surface area that is conductive to electron transmission, thereby improving the power density of the battery. Therefore, the nanoengineering of biocatalysts is crucial in the development of new enzymatic biofuel cells. The key point for the performance of biofuel cells is the immobilization of the enzyme for continuous catalytic reaction and efficient electron transfer. Therefore, the conductive supports in biofuel cells should have a larger surface area to provide more enzyme active-sites. In addition, the biocompatibility and mechanical properties of flexibility and stretchability should be considered to meet the requirements of applicability to wearable skin electronics. Based on the above two points, researchers have introduced carbon-based materials (such as carbon nanotubes [CNTs] [90,96–98], and graphene [92,99,100]), as conductive materials, and fiber materials (such as cotton [94], paper [101,102], and textile [103,104] as flexible support. Fig. 6b. To make it clear, typical types of biofuel cells have been summarized in Table 1, including detailed information of fuel materials, bioanode, biocathode, open circuit voltage (Voc), maximum power density, and flexibility.

4.3. Applications

Many advanced biofuel cells have been developed for powering [93,103–106,110] the integrated skin electronics and monitoring human health conditions [111,112]. Recently, Wei Gao et al. reported a flexible, wireless, integrated skin electronics for multiplexed metabolic monitoring *in situ* (such as simultaneous sensing urea and ammonia, glucose and pH), which are powered by lactate biofuel cells (Fig. 7a) [93]. To further achieve high power density and long-term device stability, assembly of bioanodes ranging from zero-dimensional (0D) to 3D nanomaterials include porous h-Ni, reduced graphene oxide (rGO), carbon nanotubes (CNTs), and Lox, and biocathode were assembled with CNT film and platinum (Pt) alloy nanoparticles. Results demonstrated that record-breaking power density of 3.5 mW/cm^2 and over 60 h continuous operation have been obtained. In 2017, Joseph Wang et al. proposed a stretchable, skin-electronic-based biofuel cells integrated with island-bridge array construction combination of screen-printed and lithographically technologies, where 3D carbon nanotube-based bioanodes and biocathodes were packed densely, which could power conventional electronic devices such as commercial light-emitting diodes and Bluetooth low energy radio [110]. The device exhibited a power density as high as 1.2 mW/cm^2 at 0.2 V without strain, and 1.0 mW under the repeatable stretching strains of 50%. Besides, they also reported the example of stretchable and wearable biofuel cell-supercapacitor system [104]. In this hybrid system, the lactate biofuel cell that was designed to harvest biochemical energy form user's sweat was screen-printed inside of the fabric, and MnO_2/carbon nanotubes composites-based supercapacitor designed as energy-storage module was screen-printed outside of the fabric (as Fig. 7b displayed). Considering from perspectives of both materials selection and geometric construction, the optimized elastomer-containing conductive ink and serpentine patterns enable the hybrid system to maintain a stable electrical performance during various mechanical deformations such as bending, folding, stretching, and twisting. And the successful demonstrations on human skin suggest the advantages of this fabrication route, which may satisfy both the power and mechanical requirements for skin electronics in a smart wearable fashion. In addition, biofuel cells have been assembled with textiles such as headband, wristband [103] and socks [106] to collect biochemical energy from human sweat.

Inspired by the rapid development of flexible biofuel cells, self-powered biosensors for monitoring physiological signals are being

Table 1
Summary of typical biofuel cell device and performance in skin electronics.

Fuel	Bioanode	Biocathode	Voc/V	$M_{power\ density}$/(mW/cm^2)	Flexibility	Ref.
Lactate	LOx/TTF/CNTs	Pt black	0.50	0.044	Epidermal	[105]
	LOx/NQ/CNTs	Ag2O/CNTs	0.46	0.25	Textile	[106]
	LOx/1,4-NQ/Buckypaper	BOx/PPIX/Buckypaper	0.74	0.52	20% stretching for 100 times	[101]
	LOx/NQ/CNTs pellet	Ag_2O/CNTs pellet	0.50	1.2	50% strain	[107]
	LOx/MDB-TTF-CNT/rGO/h-Ni	Pt–Co/MDB-CNT	0.60	3.5	/	[93]
Glucose	GDH/PolyMG/A-CNT/fibers	PTFE-CNT/BOD/A-CNT/fiber	0.36	0.22	Textile	[108]
	GOx/NQ/CNTs	Ag_2O/CNTs	0.44	0.16	Textile	[106]
	GOx/NQ/CNTs	Pt black	0.4	0.125	500% strain	[97]
Ethanol	AuNWs/polyHIPEs	Pt-AuNWs/polyHIPEs	0.57	0.48	a tensile strain of 40%	[95]
Fructose	FDH/CNTs/Carbon fabrics	BOx/CNTs/Carbon fabrics	0.75	0.06	/	[109]

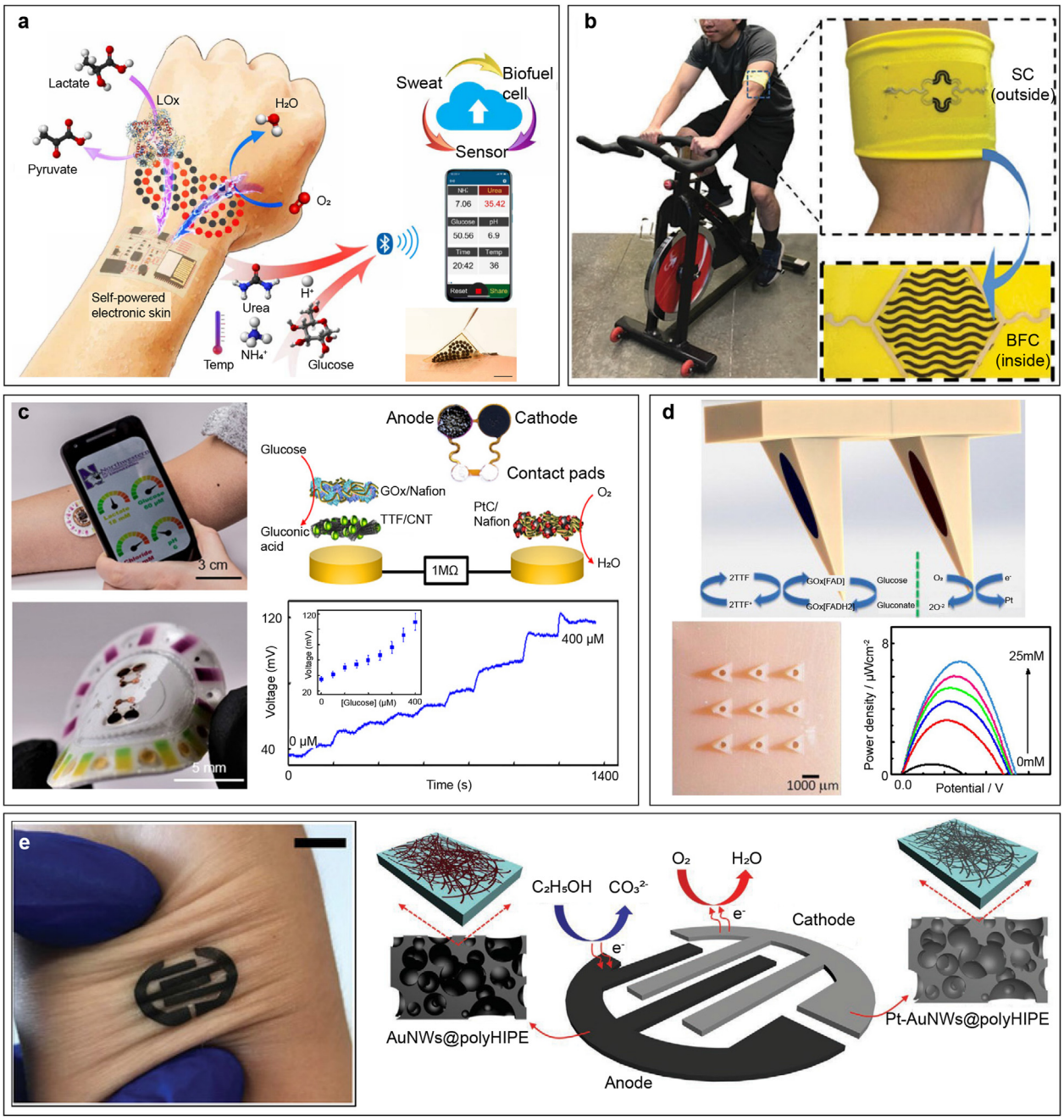

Fig. 7. Examples of applications of biofuel cells in self-powered skin electronics. (a) Schematic of the self-powered skin electronic device [93]. Reprinted by permission from American Association for the Advancement of Science. (b) Volunteer has worn an integrated self-powered hybrid system (Left), and the supercapacitor and biofuel cell were screen-printed on textile (Right) [104]. Reprinted by permission from Royal Society of Chemistry. (c) self-powered electronic system integrating microfluidic patch with sensors (Bottom Left), layer makeup of the biofuel cell-based lactate sensor (Upper Right), and the real-time sensing results (Bottom Right) [114]. Reprinted by permission from American Association for the Advancement of Science. (d) Carbon bio-electrodes paired on hollow microneedles (Upper), the microneedles array (Bottom Left) and power density from various glucose concentration (Bottom Right) [111]. Reprinted by permission from Elsevier. (e) Compressed biofuel cell on human forearm (Left), and device configuration (Right) [95]. Reprinted by permission from Wiley-VCH Verlag.

promoted [111,113]. For instance, John A. Rogers and his team proposed an advanced sensing platform that consists of biofuel cells-based biosensors module, near field communication module for data transfer and colorimetric module for data visualized presentation, which can monitor sweat rate/loss, pH value, the concentrations of lactate, glucose in sweat and blood simultaneously, laying the foundation for non-invasive and semiquantitative tracking of health-related parameters (Fig. 7c) [114]. Besides conventional in-plane structure, microneedle-shaped biofuel cell-based biosensor has been explored [111,112,115]. As shown in Fig. 7d, bioanode and biocathode were filled into a hollow microneedle array for continuous monitoring the subdermal glucose in a non-invasive way [111]. Furthermore, advanced biofuel cell-based tattoos with strain-insensitive electrodes have been studied for meeting the requirements of future wearable skin electronics 2.0 [95]. Recently, a skin-like biofuel cell-based tattoo with an overall

device thickness of 200 μm has been proposed, in which polymerized high internal phase emulsions (poly-HIPEs) were introduced as the 3D soft skeleton of ultrathin gold nanowires (Fig. 7e) [95]. Due to the high surface area of porous poly-HIPEs, biofuel cell with more catalytically active and mechanically stretchable features was obtained, resulting in a power density of 280 μW/cm^2 which are threefold higher than device with same thickness.

5. Photovoltaics

In addition to emerging energy conversion technologies mentioned above, other self-powered devices like photovoltaic and thermoelectric ones, which harvest energy from photic and heat also have been further studied for application to skin electronics. Compared with mechanical energy and other energies, solar energy is one of the most abundant and renewable green energy in the world. In this section, representative examples of photovoltaic cell will be discussed, including advances in functional materials, device configuration, and applications in self-powered skin electronics.

5.1. Working mechanism of photovoltaics

Light energy is especially promising as the power source for portable and wearable skin electronics due to the excellent power conversion efficiency (PCE) per area and the light from solar radiation. Although the pursuit of high-performance, large-area and long-term stable photovoltaics technologies has extended into silicon, dye-sensitized, organic materials and fledging perovskite solar cells, the research on flexibility and mechanical durability for these photovoltaic devices has just started to meet the energy supply needs of the new generation of wearable skin electronics. Taking the bulk-heterojunction solar cell as an example, the conversion process can be simply summarized in the following four steps (Fig. 8a) [116,117]: (1) Exciton formation; when light irradiates the surface of the device, the photoactive layer absorbs the incident light and converts photons into electron-hole pairs that are called as photo-induced excitons; (2) Exciton diffusion: excited by the external voltage, excitons diffuse to the heterojunction interface where they can form charge-transfer states; (3) Exciton dissociation: completing the decoupled process of electron pairs, the resultant free electrons and holes are moved to the interfaces of active layer/electron transporting layer and active layer/hole transporting layer, respectively; (4) Charge transport and collect: due to the matched energy level, electrons and holes injected into transporting layer via the highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO), and they are collected by cathode and anode, respectively. At this time, the conversion process from light into electricity is completed.

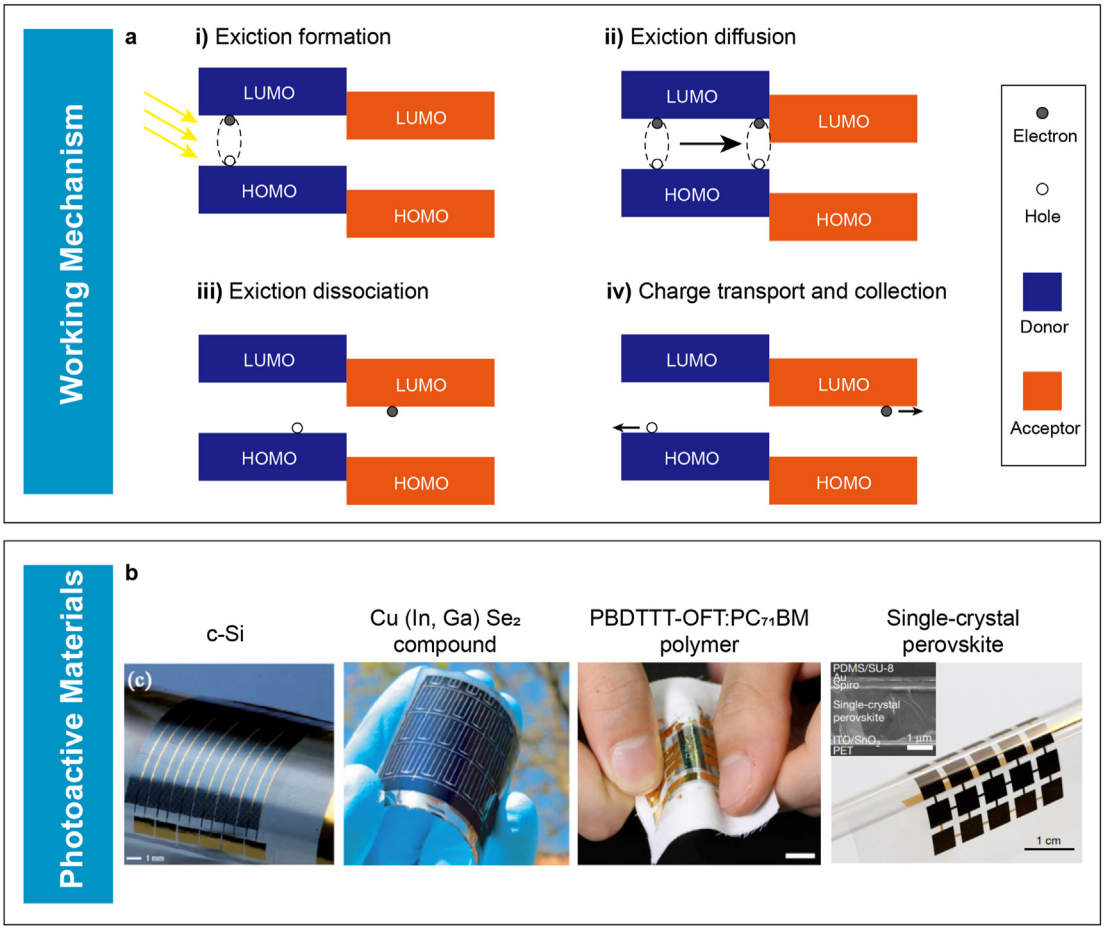

Fig. 8. Working mechanism and photovoltaic materials in photovoltaics. (a) Schematic illustration of the working mechanism of organic photovoltaic devices, which can be divided into four steps, including exciton formation, exciton diffusion, exciton dissociation, and charge transport and collect by electrodes. (b) Flexible photovoltaic devices based on different photoactive materials, including solar cell based on crystalline silicon (Si) (first generation) [122] (reprinted by permission from Springer Nature), copper (indium, gallium) diselenide [Cu(In, Ga)Se_2] compound (second generation) [123] (reprinted by permission from Springer Nature), and organic [124] (reprinted by permission from National Academic Sciences) and perovskite materials [125] (third generation) (reprinted by permission from Springer Nature).

5.2. Functional materials and devices

Active materials play important roles in light absorption and conversion, determining the PCE of the photovoltaics [118–120]. According to the photovoltaic materials, photovoltaic devices can be divided into silicon (Si)-based solar cells [121], compound thin film solar cells, dye-sensitized solar cells, organic solar cells and emerging perovskite solar cells (Fig. 8b).

Among various photovoltaic technologies, crystalline silicon (c-Si) solar cells hold the dominant market share due to the broad spectral absorption, high carrier mobility and mature fabrication technology. In skin electronics, the thickness of Si-based solar cell should be reduced to level of micrometers, and thin film c-Si solar cells have higher Voc that make them attractive for high-performance flexible photovoltaics. Conventional wire saw technique may create many kerfs. To solve this problem, different methods have been investigated: (1) epitaxial growth and layer transfer process, which enables the growth of high quality c-Si thin film on a foreign substrate [126]; (2) etch-release process, which is mature microfabrication technique in semiconductor industry can be used to fabricate flexible c-Si solar cell [122]; (3) exfoliating from bulk silicon wafer that is much more facile than abovementioned two methods has been developed in flexible c-Si solar cells [127]. The deposition of compound semiconductors, such as III-V semiconductors, on flexible substrates open new field of solar cells. Single crystalline III-V compound solar cells with power conversion efficiencies over 30% are very impressive, but their toxicity limit the applications in skin electronics.

When it comes to organic photovoltaics, the active layer consists of donor and acceptor materials where the donor used to release electrons and the acceptor used to harvest electrons. To achieve a flexible active layer, there usually are three methods that have been adopted into device fabrication: (1) Reducing the thickness of the active layer. This method can achieve the flexibility to some extent, but the degree of deformation is limited. A bending radius of a few millimeters is equivalent to less than 2% strain deformation on a substrate with a thickness of 100 μm [128]. (2) Adopting all-polymer materials as active layer both donor and acceptor [129–131]. For instance, Ergang Wang et al. proposed a highly efficient and mechanical robust solar cell based on PM6 polymer donor and PF2-DTSi polymer acceptor, which exhibited PCE up to 10.77%, toughness as high as 9.3 MJ/m^3 and a large elongation at a break of 8.6% [129]. Deformation durability test demonstrated that this all-polymer solar cell could maintain 90% of initial performance after undergoing 1200 times bend-relax cycles at a bending radius to 4 mm. (3) Synthesizing and applying the intrinsically flexible active materials. Studies showed that the length of the alkyl side chain in polymer determines the flexibility of conjugated polymers [132,133]. All these results demonstrate that the selection of active materials in flexible photovoltaics should consider the balance of electrical and mechanical properties. Recently, besides conventional polymer substrates (such as polyimide [PI], polyethylene terephthalate [PET], or polydimethyl siloxane [PDMS]), flexible material like textile fabrics has been proposed in flexible photovoltaics [134,135]. For instance, solar cells have been fabricated from polymer fibers into micro cables with the 320 μm-thick single layer, where the counter electrode was also made of Cu-coated fiber via a low-temperature wet process [136]. Integrated with fabric TENG, the hybrid power textile could charge a commercial capacitor (2 mF) up to 2 V in 1 min and continuously driving an electrical watch. In practical application, environmental stability such as waterproof and thermal stability should be considered. Takao Someya and his team proposed a stretchable and waterproof solar cell based on blend film of polymer with quaterthiophene and Naphtho[1,2-c:5,6-c']bis[1,2,5]thiadiazole (PNTz4T): [6,6]-phenyl-C71-butyric acid methyl ester ($PC_{71}BM$) by coating elastomer layers on the double sides of the device, further expanding the application with textile compatibility [137], as shown in Fig. 9a. On the other hand, heat generated inside of device during operation or from the surrounding environment may accelerate device aging and even failure. To solve this problem, thermally stable PI that has a high glass transition temperature of 265 °C as substrate and 1.36 μm-thick Teflon/parylene as encapsulation layer have been developed in flexible organic solar cell [156]. This photovoltaics could maintain 10% PCE when endured thermal stress at 100 °C for 4 h without any efficiency reduction, and it could retain 80% of initial PCE performance under accelerated testing conditions for more than 500 h in air.

The emerging perovskite materials have attracted enormous research interest due to the unique photoelectric properties, such as high absorption coefficient, high exciton mobility and long diffusion length, etc., and the mechanical flexibility, affording that the PCE values in flexible perovskite solar cell have surged to a record of 19.51% [138] and 21% [139] for single and tandem device, respectively. Among all perovskite materials, single-crystal perovskite is the most promising candidate for applications in skin electronics due to the improved charge carrier transportation and enhanced environmental stability. Most effort has been devoted to solve the heat intolerance of polymer substrate that is incompatible with the single-crystal growth process. Notably, Sheng Xu et al. proposed a solution-based lithography-assisted epitaxial-growth-and-transfer method to fabricate flexible perovskite solar cell (Figs. 8b and 9b) [125]. Devices based on this method show not only high stability against high temperature and humidity, but also exhibit excellent PV performance with an average PCE of 18.77%.

Based on the successful materials synthesis, the optimization of flexible electrodes and strategies for reducing trade-off between electrical and mechanical properties of photoactive materials, flexible photovoltaic devices with millimeter bending radius as well as excellent PCE have been developed [141–144]. In flexible solar cells, transparent electrode is critical factor to realize the high-performance photovoltaics acting as the incident light window. Traditionally, transparent conductive indium tin oxide (ITO) is one of the best options, but the brittleness of ITO limits the application in flexile devices due to there are cracks forming and leads to resistance increase when under bending state. Therefore, carbon-based materials with good mechanical flexibility and conductivity have been proposed, such as graphene [145–149], carbon nanotubes (CNT) [150–152], which exhibit two advantages as following: (1) The lower transmittance leads to higher light absorption; (2) The higher sheet resistance leads to inefficient charge transportation. Besides, solution-processable Ag NWs electrodes that process great electrical, optical, and mechanical properties have attracted huge attention in flexible photovoltaics. However, due to the high surface roughness, Ag NWs may penetrate the adjunct layers and cause larger leakage currents of the photovoltaics. To solve this problem, Ag NWs has been combined with conductive layers, such as ZnO [153] and Poly(3,4-ethylene dioxythiophene): poly(styrene sulfonate) (PEDOT:PSS) [154,155], or insulating substrate like PI [156]. Another transparent electrode is a metallic nanogrid based on Au [157,158], Ag [159,160], Cu [161], which has been realized by solution-processable of photolithography and lift-off processes. Alternatively, ultrathin metal films that the thickness is less than tens of nanometer can exhibit great conductivity, flexibility, and transparency, making them appealing to high-performance flexible solar cells. To obtain a fully percolating film, the conventional metal of Au, Ag and Cu are evaporated onto polymer substrate coated with a seed layer. The seed layer usually composited of metal oxides such as

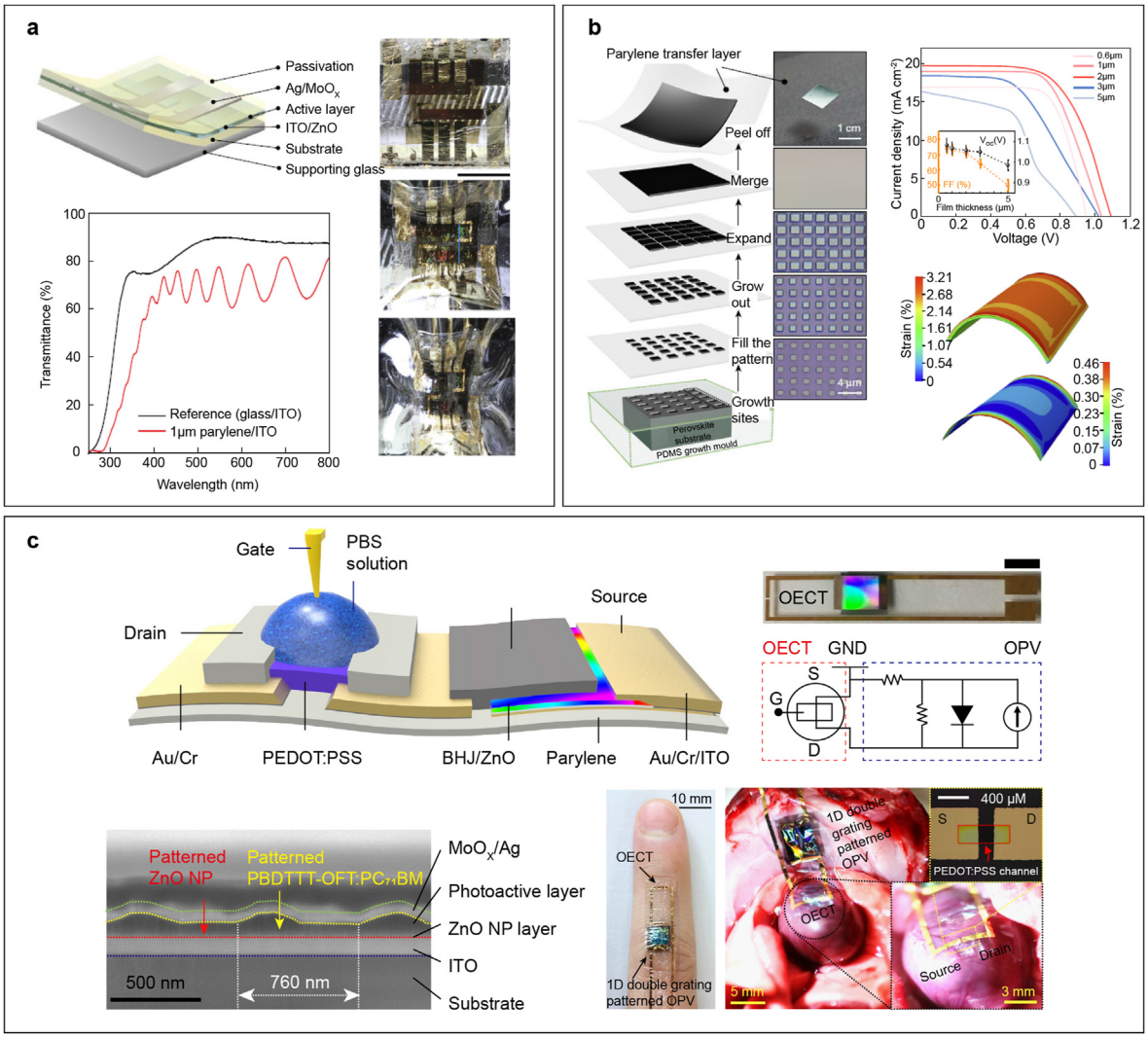

Fig. 9. Examples of flexible photovoltaics and application as energy source in self-powered skin electronics. (a) Device configuration (Upper Left), transmission performance (Bottom Left), and optical images of device under stretching (Right) [137]. Reprinted by permission from Springer Nature. (b) The procedural decomposition and corresponding SEM images (Left), current density-voltage curves of perovskite solar cells (Upper Right), and finite element analysis of the perovskite thin film at a bending radius of 2.5 mm (Bottom Right) [125]. Reprinted by permission from Springer Nature. (c) Device configuration (Upper Left), optical image and circuit diagram (Upper Right) Device configuration of organic photovoltaic device (Bottom Left), and demonstrations ex vivo attaching onto human finger and in vivo attaching onto the heart of a rat (Bottom Right) [140]. Reprinted by permission from Springer Nature.

titanium dioxide (TiO_2) [162], nickel oxide (NiO) [163], molybdenum trioxide (MoO_3) [164–166] and ZnO [167,168]. Although these ultrathin metal films offer more mechanical flexibility than normal ITO, the resulted photovoltaics showed lower PCE performance due to the limited electrical and optical properties. Therefore, advanced microcavity engineering [169,170] and insertion of a high refractive index layer were introduced to enhance the light absorption in the active layer aiming to offset the loss in optical energy. By coupling microcavity and plasmonic effect in flexible organic photovoltaics which that used ultrathin metal film and Ag nanogrid, PCE up to 9.4% was achieved which is 30% higher than ITO-based rigid devices. Besides, conducting polymer (such as PEDOT: PSS) is another promising option for flexible electrodes owing to the facile solution processability, mechanical flexibility, and optical transparency, but the conductivity is less than ITO. Ag nanomesh as a transparent electrode and PEDOT: PSS-PEIE as electron transporting layer that replaced brittle ZnO has been introduced to realize ultra-flexible photovoltaics [171]. Under different degrees of compressibility, natural buckling was formed with a bending radius less than 10 μm. And from the scanning electron microscopy (SEM) images, neither cracks nor delamination was generated in the peel-off process.

5.3. Applications

Nowadays, most effort is still devoted to the flexibility and stretchability of photovoltaics itself, and the study of application in an integrated system acting as an energy source is few. In the process of integrating self-powered photovoltaics module and sensor module into one microsystem, two issues should be taken into account: (1) Stable power output of photovoltaic module under various mechanical deformation conditions; (2) The fabrication temperature should be as low as possible to ensure the active material both photovoltaics and sensor can function. In 2018, Someya et al. reported a self-powered ultra-flexible electronic microsystem that could measure the electrophysical signals with a high signal-to-noise ratio of 40.02 dB on 1-μm-thickness parylene

substrate, and in which flexible organic photovoltaics used as an energy source to drive the electrochemical transistor-based biosensor (Fig. 9c) [140]. Further, the advanced 1D patterned charge transporting layers and photoactive layer with nanograting morphologies were replicated from a blank DVD-R, giving improved flexibility and a high PCE of 11.46 W/g. It is very meaningful to use photovoltaics for powering the implanted electric devices, which can promote the development of clinical medicine and enhance our understanding of biology. Rogers et al. proposed a fully biodegradable Si-based photovoltaics which was designed to operate at a long wavelength in the red and near-infrared bands, and it was able to generate about 60 μW when implanted into 4 mm-thick porcine skin and fat. This energy could power the blue light-emitting diodes that had been implanted in the rat's hypodermis, and it degraded completely after 4 months.

6. Thermoelectricity

6.1. Working mechanism of thermoelectric generators

Thermoelectric (TE) generators can directly convert heat energy into electrical power based on the material TE effect. Generally, we called TE module for power generation as Seebeck effect [172]. As shown in Fig. 10a [173], when the temperature difference exists between both ends of a material, the current is then generated in the circuit. TE materials can be classified as n-type and p-type. The generated current is actually the movement of charge carriers in a TE material under a temperature gradient. The specific source is the diffusion of electrons in the n-type material and holes in the p-type material from the high temperature side to the low temperature side [174]. To achieve excellent output voltages, n-type and p-type TE materials are usually used in combination.

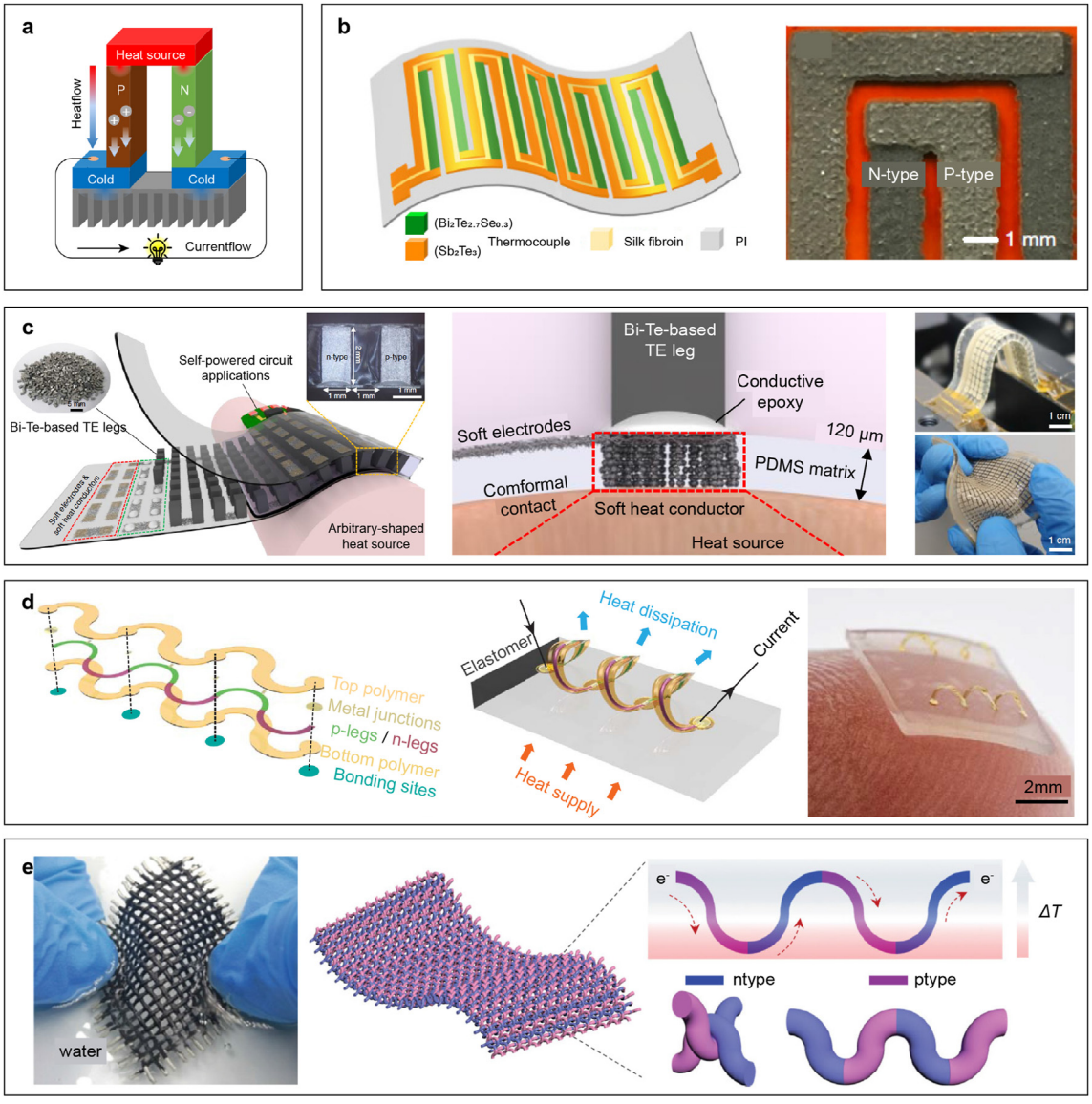

Fig. 10. Flexible thermoelectric generators for self-powered skin-integrated electronics. (a) Schematic illustrations of Seebeck effect for thermoelectric generators. (b) Coplanar TE generator. The n-type and p-type TE inks were printed on a polymeric substrate by the mature mass-fabrication technology of screen-printing [188]. Reprinted by permission from Springer Nature. (c) Flexible vertical-type TE device with TE legs [196]. Reprinted by permission from Springer Nature. (d) 3D TE coils as active components of flexible and deformable systems for harvesting electrical power [198]. Reprinted by permission from American Association for the Advancement of Science. (e) TE fibers for wearable textile-electronics [199]. Reprinted by permission from Springer Nature.

Thermoelectric figure of merit (ZT) is the most critical parameter for TE materials. ZT is defined as $S^2T\sigma/k$, where S, T, σ, and k are Seebeck coefficient, absolute temperature, electrical conductivity, and thermal conductivity respectively. High-performance TE materials are usually with a high ZT value. Thus, a high Seebeck coefficient, high electrical conductivity and low thermal conductivity are essential for high-performance TE materials. Currently, many inorganic materials exhibit good TE performance, such as bismuth-tellurium (Bi—Te) alloys [175] and silver-lead-antimony-tellurium (Ag—Pb—Sb—Te) compounds [176], whose values of ZT even exceed 100%.

Despite the good TE performance, large blocks or sheets of rigid inorganic materials are not suitable for processing into flexible TE generators. This is due to the fact that the inorganic TE materials are prone to fracture and deformation under external pressure, and the corresponding service life of the material will be reduced considerably. Moreover, the rigid TE devices cannot fit with the skin well to achieve effective heat utilization on the skin surface. In order to develop flexible TE devices, the conductive polymers have been introduced. Conductive polymers are organic polymers with intrinsic electrical conduction, good flexibility, relatively low σ, and low κ, which makes them very suitable for flexible TE applications. For example, Poly(3,4-ethylenedioxythiophene): poly(styrene sulfonate) [PEDOT: PSS] [177], poly[Cux(Cu-ett)] [178], polyvinylidene difluoride [PVDF] [179], carbon materials (single-walled carbon nanotube [SWCNT-], carbon nanotube [CNT-], reduced graphene oxide [RGO-], Graphene) [180—182] have been widely studied for flexible TE materials. In addition, The use of composite materials made from inorganic TE materials and organic materials to prepare hybrid TE materials on flexible substrates based on processes such as screen-printing to produce flexible TE devices is also a very good strategy [183,184].

6.2. Functional materials and applications

To be well used in skin-integrated electronics, the flexibility of TE devices is a necessary requirement. One approach to flex the device is based on the selection and optimization of materials [185—187]. Another approach for device optimization is from the point of view of device structural design. Here, we develop a description of skin-integrated TE devices from a structural point of view by classifying current flexible TE devices into coplanar, vertical, three-dimensional, and fiber types.

As shown in Fig. 10b [188], the n-type and p-type TE materials were fabricated on the same substrate and in the same plane. We refer to this type as a coplanar TE generator. The coplanar TE devices tend to be very thin, light and exhibit excellent TE characteristics. Many existing film fabrication methods can be used to form TE devices, such as screen-printing [189], inkjet printing [190], spray printing [191], deposition [192], sputtering [193], etc. Take the fabrication of device demonstrated in Fig. 10b for example, the TE powder was firstly mixed with prepared paste to form the TE ink [188]. Then, based on the mature mass-fabrication technology of screen-printing, n-type ($Bi_2Te_{2.7}Se_{0.3}$) and p-type (antimony telluride [Sb_2Te_3]) TE materials were fabricated on a polyimide (PI) substrate to form complementary double-chain thermocouples. Based on the Seebeck effect, when a heat source is applied to one side of the fabricated TE device, charge carriers' diffusion from the hot side to the cold will result in a current generation. Thus, when this thin thermoelectric device is applied to the human skin for self-power generation, the temperature difference comes from the different locations on the body surface where the thermoelectric device is located. Compared to the temperature difference between the body surface and the air, it is clear that the temperature difference between different locations on the body surface is very small, which greatly limits the self-power efficiency of TE devices.

To address this limitation of coplanar TE devices, most skin-integrated wearable TE devices are currently designed with a vertical structure [194,195]. As shown in Fig. 10c [196], the TE device is composed of many separated (Bi—Te)-based TE legs and flexible electrodes on the flexible substrate. The TE legs are in an approximately vertical position to the human skin, which is why we call it vertical TE devices. In this case, one end of the TE legs is attached to the skin surface and the other end is on the air side, so that the temperature difference between the skin and the air can be used to generate a TE voltage. Furthermore, these TE legs can also be made of rigid TE pillars. With the flexible substrate and electrode connection, the whole device can still be bent well without causing a breakage of the rigid TE material. With further material selection and structural optimization, it is certain that vertical TE devices will be easier to realize high performance for self-powered wearable applications. In Fig. 10c [196], Seoul National University's Byeongmoon Lee et al. proposed a comprehensive strategy to prepare high-performance compliant flexible TE generators. They developed a TE device with excellent flexibility, thereby allowing stable operation even when it is bent or stretched. In the device, bismuth telluride (Bi_2Te_3) TE legs functioned for TE generation. To lower the resistance of the TE device, a stretchable substrate composed of silver nanowires was designed as the electrode. The silver-nanowire-based soft electrodes interconnect TE legs allowed the TE generators to conform perfectly to curved surfaces. Meanwhile, elastomeric substrates with magnetically self-assembled Ag—Ni particles as soft heat conductors significantly enhanced the heat transfer to the TE legs and thereby maximized the energy conversion efficiency on three-dimensional heat sources. Due to the excellent conformability, the TE generator achieves a high TE performance on human skin and operates a self-powered wearable warning system. However, it is worth noting that for vertical TE devices, the thickness of the device is an issue due to the presence of the vertical TE pillars or legs. As too thick devices inevitably affect the fit and comfort when worn on the skin, new structural designs are necessary to address these issues. For example, a wearable TE device with Lego-like reconfiguring capabilities was proposed by combining modular thermoelectric chips, dynamic covalent PI, and liquid-metal electrical wiring in a mechanical architecture design of 'soft motherboard-rigid plugin modules.' [197] This is a typical example of combining a coplanar TE and a vertical TE design, incorporating the advantages of both.

To miniaturize the semiconductor devices, 3D TE coils were proposed as the active components of flexible and deformable systems for harvesting electrical power (Fig. 10d) [198]. This 3D structural design and preparation method offers an additional option for the design of TE devices. As shown in Fig. 1d, the fabrication method is based on 3D assembly. Firstly, the thin-film p-type and n-type materials were patterned into 2D serpentine shapes and transferred onto a layer of PI. Then, based on the photolithography and etching process, metal junctions and top patterned PI encapsulation were completed to form the final 2D structures. Finally, the 2D structure was chemically bonded to a silicone substrate uniaxially stretched beforehand. After the release of the stretch made beforehand, the final 3D architectures yield from a process of geometrical transformation. For the 2D structure, the TE p-legs and n-legs are coplanar. However, in the 3D TE coils, the 3D architectures multiplied the heat flow through the device and make sure that the TE device can maintain large temperature differences across the device terminals, thereby increasing the efficiencies for power conversion. The 3D architectures and scalable fabrication propose a good strategy for hard thin-film TE materials-based flexible TE generators integration into wearable systems.

Considering the advantages of textile electronics in wearable applications such as their wearing comfort and programmable

nature, developing fabric-based skin-integrated TE devices will be an important direction. Currently, many scientists are focusing their research on developing fiber-type TE generators with stable properties [199,200]. As shown in Fig. 10e [201], Tianpeng Ding et al., developed continuous p/n TE fibers based on a gelation extrusion strategy with high scalability and process efficiency. The p-type and n-type segments were successfully integrated into a continuous fiber. Thus, the carriers flowed in the same direction along the fiber to generate the voltage. Similar to the above 3D TE coils, the 3D structure of fibers emerged when the fiber is made into a textile. The TE textile harvested thermal energy in an out-of-plane direction. By controlling the segment length of the fiber, the pitch waves could realize successive p-n junctions alternate between hot and cold surfaces. With such alternating p/n-type TE fibers, the woven TE textiles successfully realized energy harvesting on a curved surface.

7. Conclusion and outlook

Over the past several years, work focused on energy harvesters has brought the promised high-performance, lightweight, mechanical robust self-powered skin electronics in fields of energy harvest and healthcare monitors. To enable these energy harvesters to meet the requirements of the skin electronic microsystems better and give full play to their functions, both electrical output that needs to provide continuous and stable energy supply and mechanical robustness that is built-in when attached to skin tissues seamlessly and ensure that the devices normally work in a coordinated manner and stable power and mechanical robustness should be realized simultaneously. According to this line of thought, advanced in working mechanism, functional materials, device configuration, and applications in skin electronics have been summarized and analyzed as outlined in the preceding sections. Energy supply devices based on piezoelectricity, triboelectricity, biofuel cell, photovoltaics, and thermoelectricity can harvest energy from human motions and ambient environment in a green way, thereby extending the device selection and application scenario. In general, the electrical output of these energy generators is transient that needs to combine with capacitors or supercapacitors to store energy. Furthermore, energy harvested from piezoelectricity or triboelectricity is not in a direct current form, thereby additional bridge rectifiers are needed to convert the harvested energy from alternating current to direct current.

However, several scientific and engineering challenges need to be overcome before these power supplies are fully exploited. For a start, with the development of science and technology, next-generation of self-powered skin electronics tend to be biocompatible and further biodegradable for meeting the needs of portability and implantation without the secondary surgery to remove, which is mainly to achieve upmarket healthcare applications, such as monitoring and preventing diseases in advance and helping doctors perform operations accurately. These can be realized by introducing biocompatible and biodegradable functional materials. Second, the permeation of environmental substances (e.g., water, oxygen, high-energy ultraviolet light, and others) into the device may result in a decreased performance or even fail completely by increasing the leakage current or corroding the device. Therefore, encapsulating and packaging the energy harvesting device and full skin electronic microsystem is necessary. The main difficulty in materials synthesis is that these encapsulation materials should offer stable environment stability such as insulating water, oxygen, and other ions, in the meanwhile, of a low Young's modulus and elastic mechanical properties. Thirdly, the great interface adhesion between epidermal skin and electronic devices for comfortable wear is critical. In nature, animals like gecko, cephalopods (octopus), and beetles (spider) exhibit outstanding wet/dry adhesion functionalities derived from their unique physiology structures, which can provide inspiration of advancement in epidermis-attachable electronic systems. Fourth, as part of an electronic microsystem such as energy harvesting or self-powered sensors, these devices need to be integrated with other elements to achieve a complete cycle from signal input to data analysis and conversion (sensors), and finally to data transmission and display (i.e. wireless Bluetooth and client-side). How to integrate all modules in a system that requires the reasonable design of the system distribution and wiring assembly? Considerations in size, cost, and manufacturing are additionally important for commercialize and large-scale production.

In summary, the emerging technologies and materials for self-powered skin electronics highlighted in the review can enable envisioned use in fundamental research studies for smart healthcare for human body. Critical challenges still remain for translating to real-world applications in clinical practice, including sophisticated capabilities for monitoring/treatments for associate diseases, with high-performance operation and stable biocompatibility in terms of chemistry and mechanics for specific application cases. Efforts to address these issues focus on the materials science, device configuration, fabrication technology, circuit design and biomedical engineering, as the basis for a next-generation of skin-electronic systems. These and other ongoing interdisciplinary efforts will shed light to the continued basic and applied research thereby promoting broad utility in human healthcare on the one hand and human-machine interfaces in artificial intelligence on the other.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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