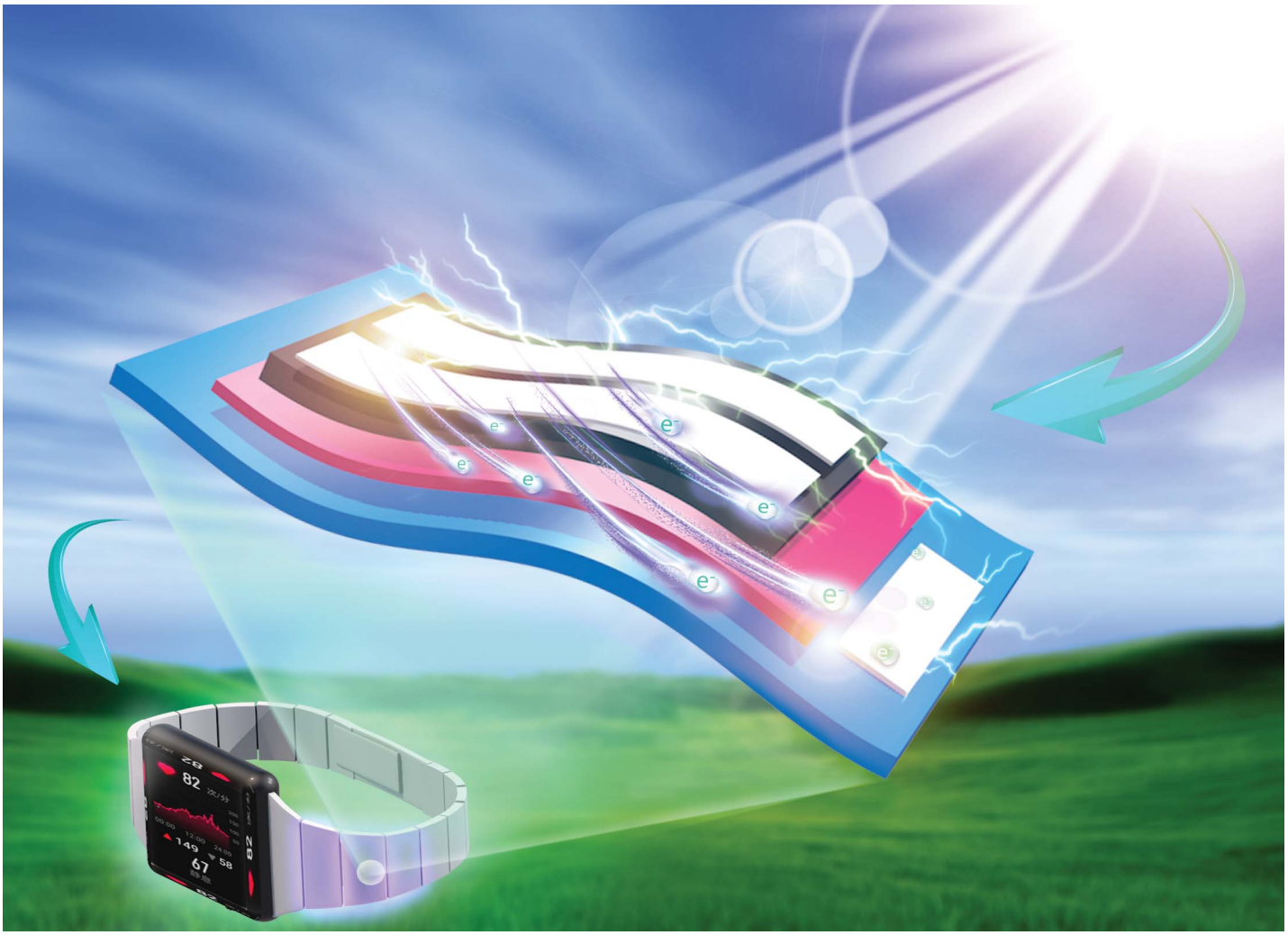

Highlighting a summary on the recent progress in the wearable self-powered energy systems by a group of researchers led by Dr Jiangqi Zhao from Sichuan University, Prof. Zhiyuan Zeng and Prof. Chaoliang Tan from City University of Hong Kong.

Recent advances in wearable self-powered energy systems based on flexible energy storage devices integrated with flexible solar cells

We first provide a comprehensive summary on the recent progress and future perspectives of wearable self-powered energy systems by categorizing different composed modules. Thereafter, we give a short discussion on the integration of flexible ESDs with flexible PVCs. Lastly, the main challenges and future perspectives of self-powered energy systems for wearable electronics are discussed.

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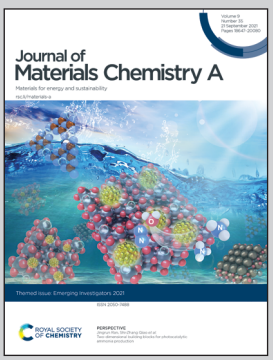

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Recent advances in wearable self-powered energy systems based on flexible energy storage devices integrated with flexible solar cells

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Wearable electronics are considered to be an important technology in next-generation smart electronics. Meanwhile, the ever-increasing energy consumption and the growing environmental awareness have highlighted the requirements of green and renewable energy. Integrating flexible photovoltaic cells (PVCs) with flexible energy storage devices (ESDs) to construct self-sustaining energy systems not only provides a promising strategy to address the energy and environmental issues, but also enables the entire system to be operated continuously without external charging, which is considered to be a promising direction for future wearable electronics. Herein, we summarize the recent progress in wearable self-sustaining energy systems based on flexible ESDs integrated with flexible PVCs. First, the recent developments in flexible PVCs and their characteristics are summarized. Following that, we discuss the advances in flexible ESDs used in wearable electronics. Then, we give a short discussion on the integration of flexible ESDs with flexible PVCs. Thereafter, various functional applications of these self-sustaining energy systems in wearable electronics are introduced. Finally, the main challenges and future prospects of self-powered energy systems for wearable electronics are discussed.

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1. Introduction

Wearable electronics have received increasing attention and experienced rapid growth in recent years.[1,2] These devices have been widely used in multifunctional entertainment, intelligent monitoring, personal healthcare and exercise management, and gradually change people's lifestyles.[3–5] At the same time, the huge consumption of energy caused by

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the rapid development of electronic devices along with the growing environmental awareness has highlighted the requirements for new technology to collect and store green and renewable energy.[6] Solar energy is the most abundant, sustainable and cleanest energy in the world.[7] Along with the continuous improvement of conversion technology from light to electricity, solar energy accounts for an increasing proportion of the energy system, and holds promise as an ideal alternative to conventional fossil fuel energy in the future.[8,9] However, because of the non-continuous and unstable nature of sunlight, it is not stable for solar cells to directly supply power to wearable electronics.[10] Combining photovoltaic cells (PVCs) with energy storage devices (ESDs) to realize a self-powered system provides a feasible solution to address the imposed restrictions of sunlight availability and acts as a reliable source of energy.[11] In this regard, self-sustaining energy technology is considered to be a highly promising direction for future wearable electronics since it can enable electronics to operate continuously without external power supply.[12] At the same time, building integrated self-powered systems enables the development of wearable electronic devices toward intelligence and miniaturization.[12]

Up to now, different types of self-powered energy systems with a great variety of flexible PVCs and flexible ESDs have been developed as reliable energy sources used in wearable electronics.[13–16] Although these wearable electronics with multiple functions have been demonstrated to be used in various applications, these demonstrations are still far from actual commercial products, which is caused by the complex fabrication process, high cost, single function, immature design, *etc.* In addition, in some previous review papers, the authors usually only focus on a certain part (energy harvest, energy storage, wearable sensors, *etc.*), and the discussion on their applications in the area of wearable devices is limited.[17–20] Overall, the development of the field of self-powered wearable energy systems is still in its infancy. It is quite urgent to explore more advanced functional materials and efficient integration strategies to promote the development of self-powered energy systems to realize large-scale production and market applications. The conventional concept of a "self-powered system" which has been proposed before only accounts for a small part of the full context, and especially ignores the energy conversion efficiency, reliability, durability and challenges of the entire systems.[17–20] The lack of a comprehensive review article, which summarizes the progress of self-powered systems and the problems faced, drives us to summarize the recent progress from the perspective of entire self-powered energy systems.

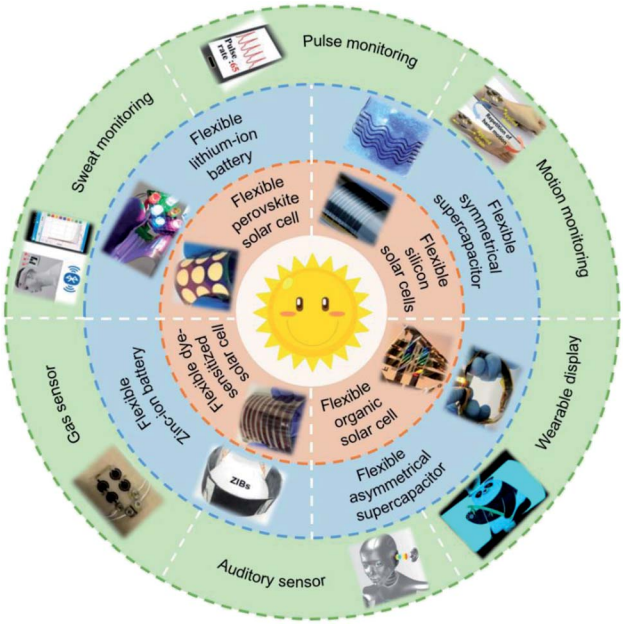

Fig. 1 Schematic illustration of the important applications of flexible ESDs integrated with PVCs.

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Herein, we provide a comprehensive summary of the recent progress and future prospects of wearable self-powered energy systems by categorizing different composed modules. In this regard, firstly, flexible PVCs, including silicon solar cells (SSCs), organic solar cells (OSCs), dye-sensitized solar cells (DSSCs) and perovskite solar cells (PSCs), are introduced based on different light conversion mechanisms. Then, according to different energy storage mechanisms, recently developed flexible ESDs, such as supercapacitors (SCs), lithium-ion batteries (LIBs) and zinc-ion batteries (ZIBs), and their application in wearable electronics are summarized. Thereafter, we give a short discussion on the integration of flexible ESDs with flexible PVCs. Following this section, various functional applications of self-powered energy systems in wearable electronics, such as pulse monitoring, sweat monitoring and gas detection, are presented. Finally, to facilitate further research and technology development, some significant challenges and further research directions of self-powered wearable systems are discussed. A brief summary is schematically shown in Fig. 1, which illustrates the main studies of wearable self-powered energy systems.

2. Flexible photovoltaic cells in self-powered wearable electronics

Photovoltaic cells have become ideal alternatives to common energy sources because of their advantages of light weight, compact structure and high conversion efficiency. With the advent of new functional materials and progress of fabrication technologies, as well as the growing demand in the wearable market, different types of flexible photovoltaic cells, including SSCs, OSCs, DSSCs and PSCs, have been continuously enriched. The performance of various kinds of flexible PVCs is summarized in Table 1.

2.1. Flexible silicon solar cells (SSCs)

Benefiting from the high natural abundance, excellent reliability and good power conversion efficiency (PCE), SSCs, the earliest developed solar cells, have long dominated the photovoltaic market.[21] According to the working mechanisms, silicon-based solar cells can be divided into monocrystalline, polycrystalline and amorphous SSCs.[22] In contrast to early rigid SSCs, flexible ones have been processed in recent years through depositing silicon on flexible substrates or embedding microscale SSCs into flexible substrates.[23,24] In 2011, Yoon *et al.* reported flexible concentrator photovoltaics fabricated by embedding microscale SSCs into luminescent waveguides.[25] As shown in Fig. 2a, the incident light could be reflected in the flexible matrix and redirected onto the microscale SSCs to increase the power of the obtained SSCs by more than 300%. This unusual design endows the resulting SSCs with ultrathin, mechanically bendable formats. Later on, Lee *et al.* prepared printable nanostructured SSCs from wafer-based materials directly (Fig. 2b).[26] This strategy can reduce the cost of materials and facilitate programmable, large-scale distribution. Ultrathin micro-SSCs integrated with flexible substrates (poly-ethylene terephthalate) exhibited a PCE of 12.4%. Compared with crystalline silicon, amorphous silicon costs less and is easier to be processed into thin films. In 1991, Kishi *et al.* demonstrated a flexible solar cell for the first time by coating amorphous silicon on a transparent plastic substrate with an output power of 275 mW g^{-1} at that time.[27] From then on, amorphous silicon has been extensively studied and used in mass-producing flexible solar cells (Fig. 2c).[28] In addition to the above flat flexible solar cells, flexible fiber-shaped SSCs, with the features of light weight, foldability and knittability, are also an important research direction. In 2009, Lee *et al.* fabricated a solar power wire by coating P3HT/PCBM on stainless wires (Fig. 2d), and the PCE was up to 3.27%.[29] Furthermore, Plentz *et al.* reported a flexible energy harvesting textile fabricated by coating amorphous thin-film SSCs on glass fiber fabrics (Fig. 2e).[30] The textile solar cell exhibits a PCE of up to 1.4% and a pseudo-efficiency of up to 2.1%. With the continuous development of flexible SSCs, their application studies in flexible wearable electronics are also increasing. So far, a variety of self-powered wearable devices based on flexible SSCs have been developed.[17,31,32] As illustrated in Fig. 2f, Ostfeld *et al.* designed a flexible power source by integrating commercial SSCs with lithium-ion batteries.[33] Under an irradiance of 100 mW cm^{-2}, the self-charging system can be charged to 4.1 V at a current of around 40 mA h, and demonstrates good stability during the cycles of charge and discharge. Moreover, the integrated device can be flexed in the hand and attached onto a jacket sleeve (Fig. 2g), to collect energy from the surrounding environment and power pulse oximeter components.

Table 1 Summary of the types, materials and capacities and flexibility of flexible ESDs

Types	Materials	Specific power	PCE	Ref.
SSC	Nanostructured silicon	—	12.4%	26
SSC	P3HT/PCBM	—	3.27%	29
SSC	Amorphous silicon	—	1.4%	30
OSC	PEDOT:PSS	10 W g^{-1}	4.2%	38
OSC	PBDTTT-OFT/PC71BM	11.46 W g^{-1}	10.5%	39
DSSC	N719 TNARs	—	4.54%	45
DSSC	TiO_2	—	3.27%	46
DSSC	TNAR/Ti N-719	—	3.82%	47
PSC	Perovskite	23 W g^{-1}	12%	56
PSC	Perovskite	—	14.78%	57
PSC	Perovskite	—	12.2%	59

2.2. Flexible organic solar cells (OSCs)

In the past few decades, flexible OSCs have received increasing attention because of their light weight, good flexibility and low cost.[34,35] Compared with SSCs, the good flexibility of organic materials makes OSCs more suitable for application in flexible electronics.[36,37] In general, flexible OSCs consist of a flexible substrate, transport electrode, active layer and metal electrode, as shown in Fig. 3a.[38] Kaltenbrunner *et al.* fabricated ultrathin

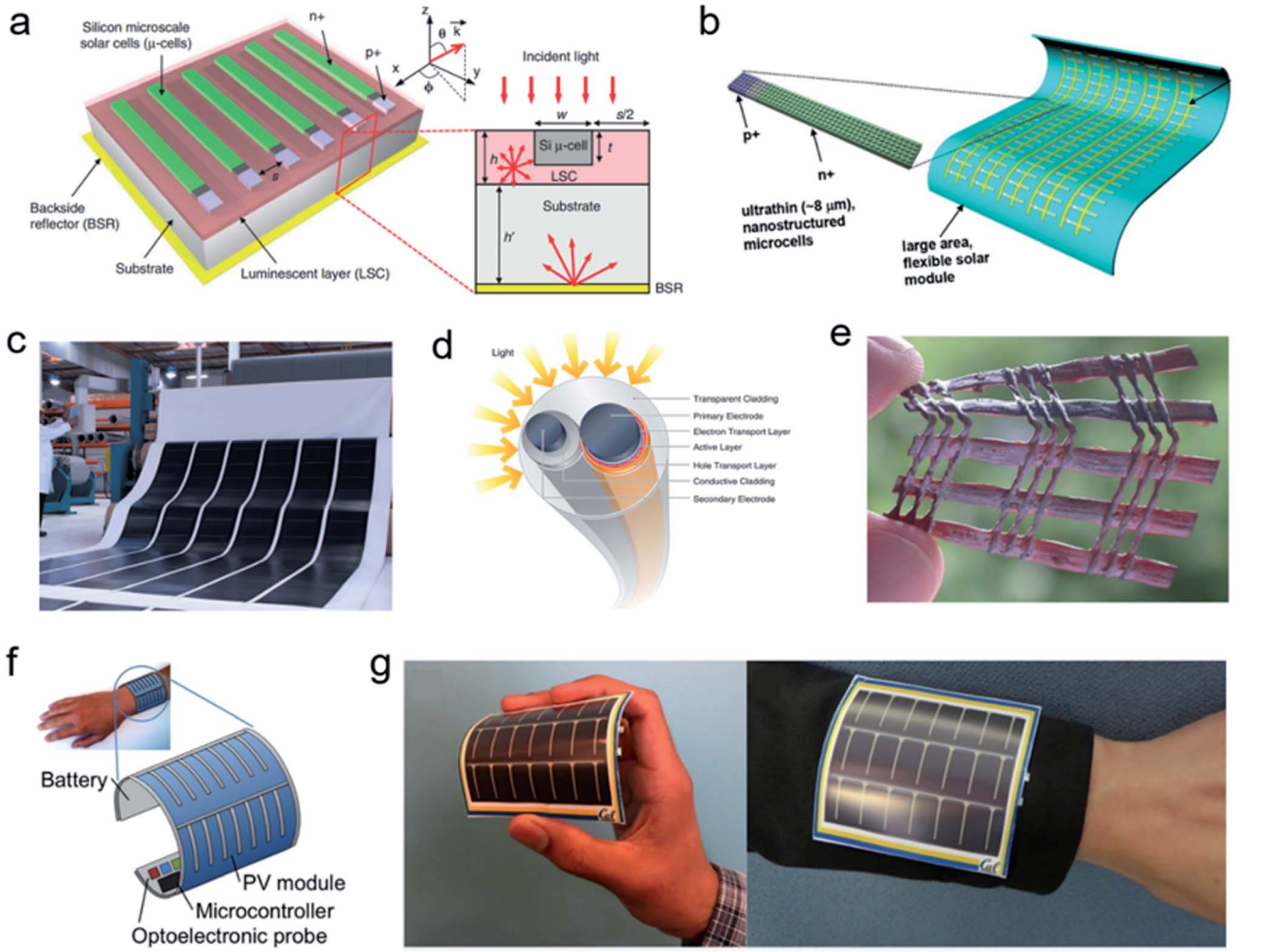

Fig. 2 (a) Schematic illustration of a flexible SSC consisting of a solar microcell array.[25] Copyright 2011, Springer Nature. (b) Schematic illustration of nanostructured silicon solar microcells.[26] Copyright 2014, American Chemical Society. (c) Photograph of mass-production of flexible SSCs.[28] Copyright 2008, Wiley-VCH. (d) Schematic of a fiber-shaped SSC.[29] Copyright 2009, American Association for the Advancement of Science. (e) Photograph of a textile glass fiber fabric.[30] Copyright 2016, Elsevier. (f) Illustration of a self-powered activity-tracking wristband. Photographs of a self-charging system being flexed in the hand (left) and on a jacket sleeve (right).[33] Copyright 2016, Springer Nature.

flexible OSCs (less than 2 μm thick) by coating a highly conductive PEDOT:PSS film on a PET substrate. As shown in Fig. 3b, the prepared OSCs have an extremely high specific power of 10 W g^{-1} and a PCE of 4.2%. More importantly, the OSCs can withstand elastic compression deformations of different amplitudes and withstand more than 300% tensile strain (Fig. 3b). Furthermore, Park *et al.* designed double-grating-OSCs on an ultra-thin and flexible substrate (Fig. 3c).[39] The obtained OSCs not only exhibit shiny outer surfaces and ultra-flexibility (Fig. 3d), but also possess a high power of 11.46 W g^{-1} and an outstanding PCE of 10.5%. In addition, the authors further integrated the OSCs with organic electrochemical transistors to fabricate self-powered ultra-flexible electronic devices. The fabricated self-powered ultra-flexible electronic devices can measure biometric signals with very high signal-to-noise ratios when located on skin or other tissues (Fig. 3e).

2.3. Flexible dye-sensitized solar cells (DSSCs)

Because of the low cost, abundant raw materials, and high photoelectrical conversion efficiency, DSSCs have caught more and more peoples' attention.[40] Compared with DSSCs based on rigid and brittle conductive glass, flexible DSSCs possess unique advantages of light weight, low production cost, and compatible with roll-to-roll manufacturing technology.[41] Consequently, considerable research work has been done on the development of flexible DSSCs on transparent conductive oxide-coated polymer substrates.[42–44] In the work done by Xu *et al.*, a flexible DSSC was prepared based on a CuS transparent conductive film (Fig. 4a),[45] in which the CuS film shows good electrocatalytic performance, resulting in a PCE of 4.54% for the prepared DSSC. This flexible DSSC exhibited superior mechanical robustness and kept a stable current density in the bending cycling measurement. In another attempt, Wu *et al.* reported a large-scale flexible DSSC module, fabricating by deposition TiO_2 on a plastic substrate.[46] The flexible DSSC exhibited a large

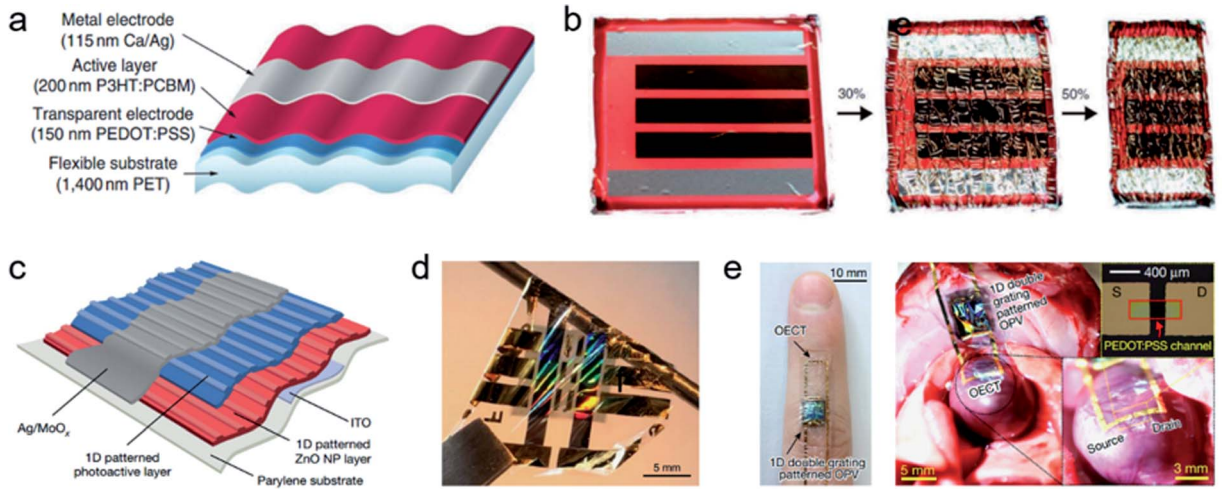

Fig. 3 (a) Schematic illustration of a flexible OSC. (b) Bending flexibility demonstrated by wrapping a flexible OSC.[38] Copyright 2012, Springer Nature. (c) Structure of a flexible OSC device. (d) Photograph of the flexible OSC wrapped over a spatula rod and pulled with tweezers. (e) Photograph of a self-powered electronic attached to a finger and attached to the heart of a rat (left), and enlarged images of the channel area (right, top) and of the source-drain electrode (right, bottom).[39] Copyright 2018, Springer Nature.

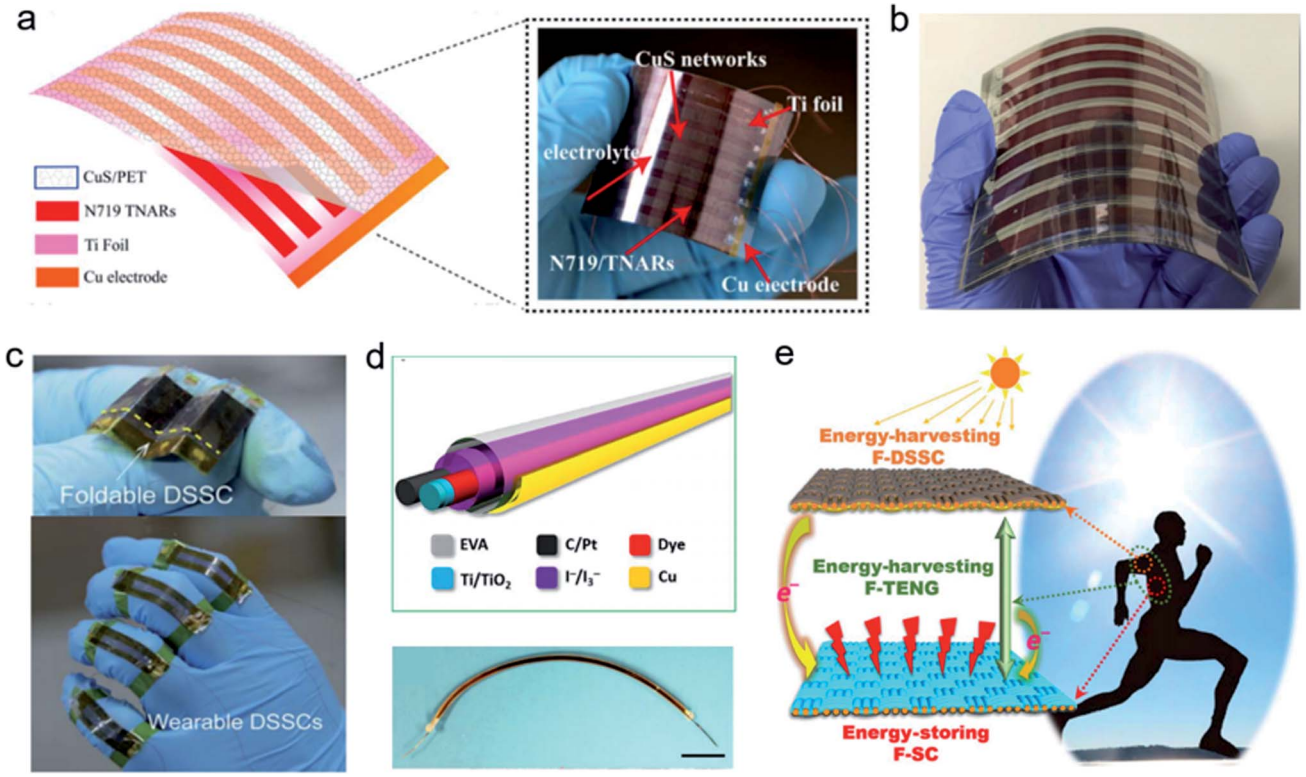

Fig. 4 (a) Schematic illustration (left) and corresponding photograph (right) of a flexible DSSC.[45] Copyright 2018, Elsevier. (b) Photograph of a flexible DSSC module with the dimensions of 100 mm × 100 mm.[46] Copyright 2016, Elsevier. (c) Photograph of wearable DSSCs affixed to human fingers.[47] Copyright 2015, Royal Society of Chemistry. (d) Schematic diagram (up) and photograph (down) of a single fiber-shaped DSSC. (e) A view of the application of a fiber-based DSSC in a self-powered system.[48] Copyright 2016, American Association for the Advancement of Science.

effective working area of 100 mm × 100 mm (Fig. 4b) with a PCE of 3.27%. In the work reported by Zhou *et al.*, a highly flexible, conductive and catalytic Pt network has been used as a transparent counter electrode for flexible DSSCs.[47] The Pt networks can be transferred onto flexible substrates and possess excellent mechanical flexibility in bending and twisting

tests. Therefore, the obtained wearable DSSCs with a PCE of 3.82% could be adhered onto human fingers with good working stability (Fig. 4c). Furthermore, Wen *et al.* demonstrated a fiber-shaped DSSC through coating dye-adsorbed TiO_2 nanotube arrays on a Ti wire (Fig. 4d).[48] Then they demonstrated the application of the fiber-shaped DSSC in wearable electronics by integrating with triboelectric nanogenerators to form an all-fiber-shaped energy harvesting module. The hybridized energy harvesting module was able to be conveniently woven into textiles to get wearable smart clothes (Fig. 4e).

2.4. Flexible perovskite solar cells (PSCs)

Thanks to their flexibility, light weight and excellent photovoltaic efficiencies, PSCs are regarded as a promising alternative to conventional photovoltaics.[49,50] In addition, the photoactive layer in PSCs can be processed at rather low temperature, making PSCs ideal platforms for fabricating flexible devices.[51,52] Planar PSC perovskite batteries generally have a sandwich structure that contains a photoactive layer between the two charge transport layers, as shown in Fig. 5a.[53] In addition, there are some reports about double-twisted PSCs based on conductive yarn (*e.g.*, carbon nanotube fibers).[54,55] Fig. 5b shows a schematic of this type of PSC structure.[55] Kaltenbrunner *et al.* prepared a flexible PSC by introducing a chromium oxide–chromium interlayer and a transparent polymer electrode.[56] The resulting PSCs achieve a PCE of 12% and a specific power as high as 23 W g^{-1}. In the work reported by Park *et al.*, solution-processed Li-doped SnO_2 has been developed at low temperature, which served as an effective electron transport layer in flexible PSCs.[57] The prepared PSCs exhibit a high PCE of 14.78% and good flexibility. Furthermore, the PSC could be fixed on the wrist and power a small fan successfully (Fig. 5d). Flexible PSCs with light weight and desirable efficiency are desirable for wearable power sources, giving them huge application potential in self-powered wearable electronics.[58] For instance, Kim *et al.* prepared a flexible PSC by depositing a TiO_x layer on a cheap polyethylene naphthalate (PEN) substrate.[59] Benefiting from the reasonable structural design and fast electron transport, the prepared PSC achieves a high PCE of 12.2%. As shown in Fig. 5e, the PSC shows excellent flexibility and exhibits outstanding stability during different bending tests. Based on the above advantages, the authors envisioned using this flexible PSC to construct a self-powered smart watch (Fig. 5f).

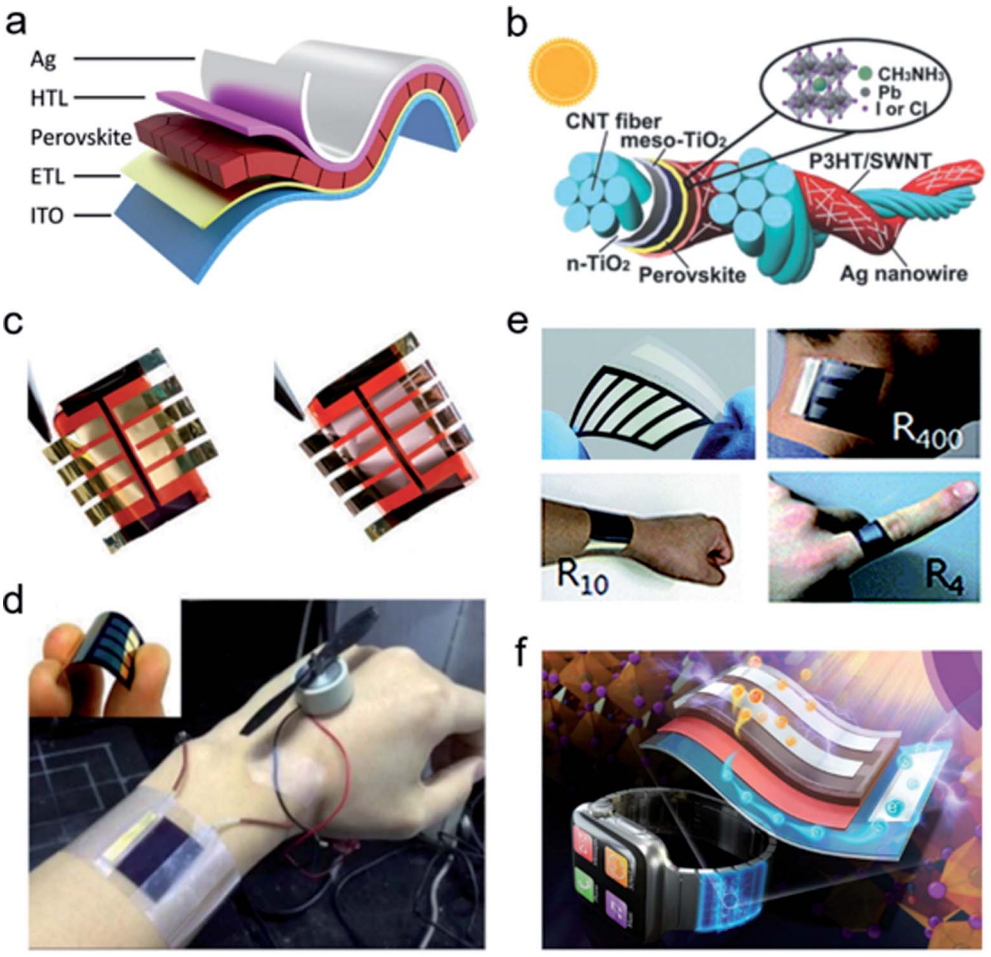

Fig. 5 (a) Schematic illustration of a planar PSC.[53] Copyright 2019, Wiley-VCH. (b) Schematic of a double-twisted fibrous PSC.[55] Copyright 2015, Wiley-VCH. (c) Freestanding 3 μm-thick solar cells with gold top metal.[56] Copyright 2015, Springer Nature. (d) Photographs of a flexible PSC fixed on the wrist and powering a small fan.[57] Copyright 2016, Elsevier. (e) The photographs of a flexible PSC attached on the human neck, wrist, and finger, respectively. (f) A view of the self-powered smart watch based on flexible PSCs.[59] Copyright 2015, Royal Society of Chemistry.

3. Flexible energy storage devices (ESDs) in self-powered wearable electronics

Limited by the non-continuous and unstable nature of sunlight, the output of PVCs is unstable and unlikely to power electronics directly. Thus, ESDs have also been introduced into self-powered energy systems. Combining PVCs with ESDs to construct a self-sustaining energy system not only breaks through the restrictions of the sunlight intermittency, but also enables the electronics without external charging to operate continuously. At the same time, the growing market of wearable and portable electronics stimulates huge demand for ESDs that are endowed with both high electrochemical performance and good flexibility. In the past decade, flexible ESDs including different kinds of batteries and SCs have already made great progress with the help of the development of new energy materials and device structures. The performance of various kinds of ESDs is summarized in Table 2.

3.1. Flexible supercapacitors (SCs)

Owing to their high power density, high charge and discharge rate and long cycle life, SCs have attracted considerable attention recently.[60] In particular, flexible SCs with a small volume, light weight, good safety and good mechanical durability are increasingly used in various wearable electronics.[61,62] According to whether the materials of the two electrodes are the same or not, SCs can be divided into symmetrical SCs and asymmetric SCs.

3.1.1. Flexible symmetrical supercapacitors (SSCs). For SSCs, the electrodes on both sides are made of the same material. Generally, the electrodes of SSCs are made of carbon-based materials, including activated carbon particles, carbon nanotubes (CNTs), carbon nanofibers (CNFs) and graphene.[63,64] As shown in Fig. 6a, the energy storage mechanism of SSCs is generally based on electric double layer capacitance (EDLC), and the electrochemical performances are mainly dependent on the electrical conductivity, pore structures and the specific surface area of the electrode materials.[20] By using flexible electrodes and quasi-solid electrolytes, flexible quasi-solid-state SCs can be prepared and used in a self-powered system (Fig. 6b).[65] In addition, two yarn/fiber electrodes can be assembled into yarn/fiber shaped SCs, which can be further weaved into textile SCs (Fig. 6c).[66] For instance, Kou *et al.* prepared core-sheath fibre electrodes consisting of polyelectrolyte-wrapped graphene/CNT by a coaxial wet-spinning assembly approach.[67] Then, these core-sheath fibres are used directly as electrodes to fabricated two-ply yarn SCs with an ultra-high capacitance of 269 mF cm^{-2} and an energy density of 5.91 mW h cm^{-2}. On this basis, a cloth SC is further interwoven from two individual core-sheath fibres (Fig. 6d). Furthermore, Du *et al.* prepared yarn-based SCs using composite electrodes, which are fabricated by depositing poly(3,4-ethyl-enedioxythiophene) (PEDOT) on stainless steel yarns.[68] The all-solid-state yarn-based SCs achieve an excellent areal specific capacitance of 158.2 mF cm^{-2} as well as good stability. Moreover, the authors incorporated the yarn-based SCs with linear sensors into an all-in-one wearable textile system (Fig. 6e). The self-powered and self-sensing wearable textile system displayed high flexibility and outstanding sensitive to detect human motion (Fig. 6f). In recent years, printing technology, with the features of enabling scalable and feasible fabrication, has aroused more and more research interest in the fabrication of SCs. For example, Lin *et al.* prepared a flexible all-solid-state planar symmetrical SC through an inkjet-printing method.[69] Benefiting from the unique nanocoral structure, the as-fabricated flexible planar SSCs exhibit an areal capacitance of up to 52.9 mF cm^{-2} and excellent versatility. Meanwhile, symmetrical SCs can be designed into different artistic symbols and further integrated with commercial polycrystalline silicon solar cells to form a self-powered T-shirt. As shown in Fig. 6g and h, the SCs, in the patterns of HKUST (the logo of the Hong Kong University of Science), are able to power 41 LED arrays arranged in the shape of three letters of "UST".

3.1.2. Flexible asymmetric supercapacitors (ASCs). For ASCs, the materials of the two electrodes are different. One of the electrodes is usually a carbon-based material, while the other is usually an electrochemically active material, such as conducting polymers and metal oxides/nitrides/sulfides.[70] Different from EDLCs, these electrochemically active materials can offer pseudocapacitance through reversible electrochemical reactions, which endows the asymmetric SCs with a higher

Table 2 Summary of the types, materials, capacities and flexibility of flexible ESDs

Types	Materials	Capacities	Energy density	Ref.
SSC	Polyelectrolyte-wrapped graphene/CNT	269 mF cm^{-2}	5.91 μW h cm^{-2}	67
SSC	PEDOT on stainless steel	158.2 mF cm^{-2}	10.3 μW h cm^{-2}	68
SSC	MnO_2 nanoflakes on Ni nanocoral	52.9 mF cm^{-2}	11.1 mW h cm^{-3}	69
ASC	Peptide–Co_9S_8 nanobricks	1.3 F cm^{-2}	79 W h kg^{-1}	74
ASC	Zinc–nickel–cobalt ternary oxides	564 mF cm^{-2}	53.33 μW h cm^{-2}	75
ASC	NiCo LDH@GSs@CF-NF	1729.413 mF cm^{-2}	109.6μW h cm^{-2}	78
LIB	CNT/LMO	2.2 mA h m^{-1}		87
LIB	CNT–Si	0.22 mA h cm^{-1}	4.5 mW h cm^{-2}	88
LIB	$LiFePO_4$	140.4 mA h g^{-1}		90
ZIB	α-MnO_2	306 mA h g^{-1}	6.18 mW h cm^{-2}	97
ZIB	CNT@MnO_2	302.1 mA h g^{-1}	53.8 mW h cm^{-3}	98
ZIB	COG@MnO_2	369.73 mA h g^{-1}	20.5 mW h cm^{-3}	100

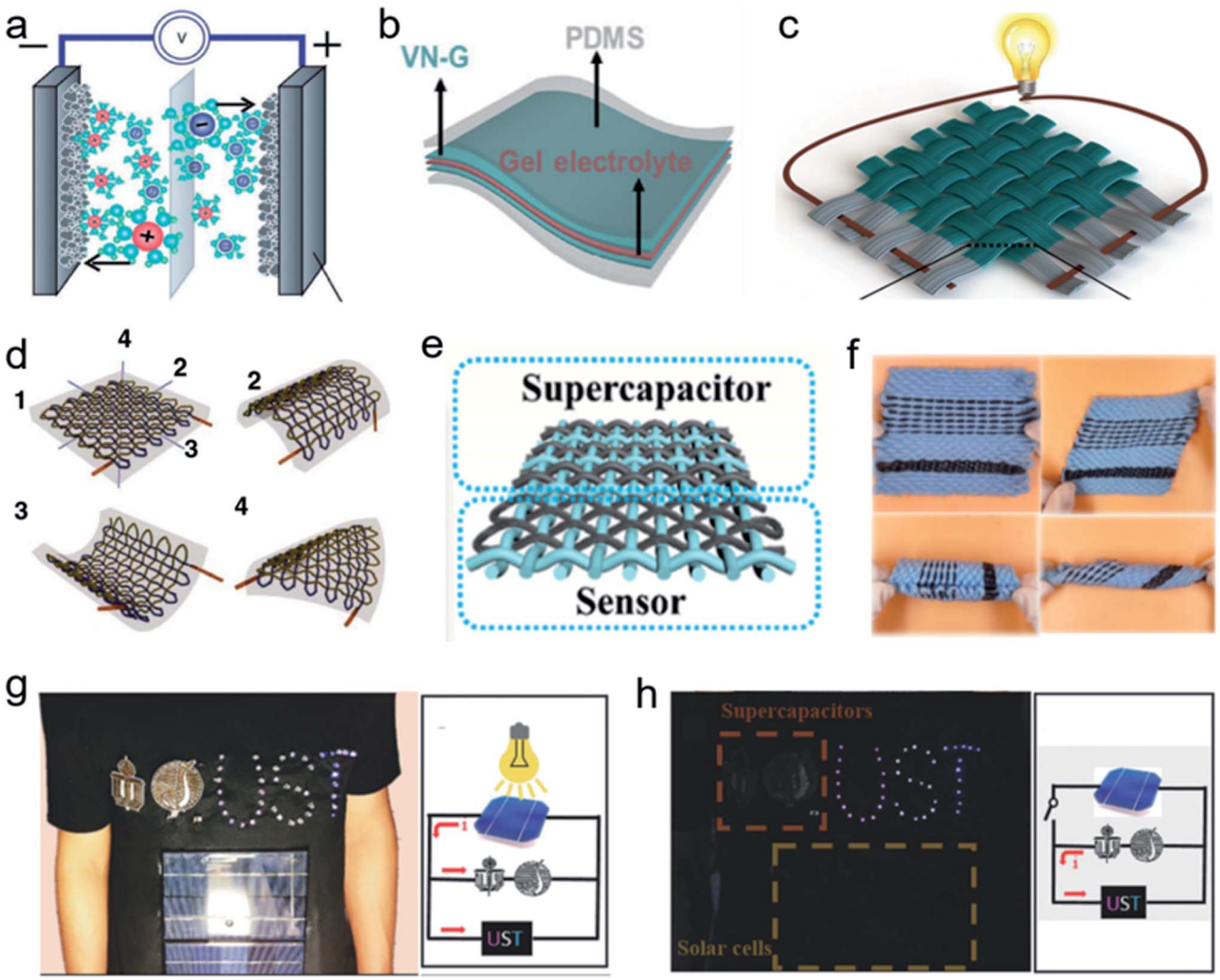

Fig. 6 (a) Schematics of an all-carbon EDLC.[20] Copyright 2018, Wiley-VCH. (b) Schematic illustration of a flexible SSC device.[65] Copyright 2019, Tsinghua University Press. (c) Schematic illustration of a woven SSC using activated carbon fiber threads.[66] Copyright 2018, Wiley-VCH. (d) Schematic illustration of a cloth woven from fiber shaped SCs with bending different angles.[67] Copyright 2014, Springer Nature. (e) Schematic illustration and (f) images of a yarn-based wearable system consisting of linear sensors and yarn-based SCs.[68] Copyright 2020, American Chemical Society. Photographs of a wearable self-powered system on a T-shirt that could be lit by: (g) solar cell panels under illumination or (h) SCs under dark conditions.[69] Copyright 2017, Wiley-VCH.

capacitance and energy density.[71–73] As illustrated in Fig. 7a, Xiong *et al.* fabricated a flexible ASC based on active carbon and core–shell structured peptide–Co_9S_8 nanobricks.[74] The obtained ASC shows a capacitance of 1.3 F cm^{-2} and good cycling stability. In addition, the ASC can be combined with a TENG to form a flexible self-powered TENG/SC system. Furthermore, Guo *et al.* prepared a fiber-shaped ASC based on yarn electrodes (Fig. 7b).[75] The obtained ASC exhibits high specific capacitance (564 mF cm^{-2}), outstanding energy density (53.33 μW h cm^{-2}) and excellent mechanical flexibility. Thanks to their higher energy density and lightness, flexible asymmetric SCs have wide application prospects in the field of self-powered wearable electronics. For instance, Tian *et al.* reported a magnesium ion quasi-solid-state ASC through printable technology. The printed ASC shows an excellent energy density of 13.1 mW h cm^{-3}.[76] On this basis, the ASCs are combined with amorphous Si solar cells to form a flexible self-charging integrated unit (Fig. 7c), which exhibits good mechanical robustness, excellent photo-charging cycling stability, and high overall energy conversion efficiency (Fig. 7d). Yun *et al.* reported an all-transparent stretchable ASC, consisting of Au/Ag embedded polydimethylsiloxane (PDMS) and WO_3 nanotube/PEDOT:PSS electrodes as well as a polyacrylamide (PAAm)-based hydrogel electrolyte.[77] The prepared ASC shows a maximum specific capacitance of 471.0 F g^{-1}. As exhibited in Fig. 7e, the fabricated ASC is further integrated into a wearable patch device to serve as an electrochromic coloration device plus an electrochemical ESD. Furthermore, Gao *et al.* fabricated a fiber-shaped ASC from 3D copper foam, graphene sheets and layered double hydroxides.[78] The graphene sheets act as bridges to increase the surface area and decrease the interface resistance, resulting in a high capacity (1729.413 mF cm^{-2}) and a rate capability (87.1% retention from 1 to 20 mA cm^{-2}). More importantly, the fiber-shaped ASCs with good

Fig. 7 (a) Schematic illustration of a flexible solid-state ASC.[74] Copyright 2019, Elsevier. (b) The schematic diagram of a fiber-shaped ASC device.[75] Copyright 2018, Elsevier. (c) Schematic illustration of an integrated solar-charging self-powered unit, and (d) digital photo showing its bending condition.[76] Copyright 2019, Springer Nature. (e) Schematic illustration of a wearable patch device based on transparent ASCs.[77] Copyright 2019, American Chemical Society. (f) A view of application of fiber-shaped ASCs in various electronic devices, including a watch, headphone and lab cloth.[78] Copyright 2018, American Chemical Society.

flexibility can be knitted into clothes or implanted in wearable electronic devices, as schematically illustrated in Fig. 7f.

3.2. Flexible batteries

Batteries can convert chemical energy into electrical energy. Typically, a battery consists of a cathode, an anode, a separator and an electrolyte. Compared with SCs, batteries (such as LIBs and ZIBs) generally have higher capacity and energy density, which makes them more widely used in wearable and portable electronics.

3.2.1. Flexible lithium-ion batteries (LIBs). Ascribed to their high energy density and working voltage and long cycle life, LIBs play a dominant role in the market of electric vehicles, portable electronics and large-scale energy-storage systems.[79,80] However, conventional LIBs are typically rigid and heavy, and have inherent safety hazards, which considerably restrict their extensive application in the area of wearable electronics.[81] In recent years, more and more flexible LIBs have been developed based on the usage of flexible electrodes as well as the improvement of electrolytes.[82–84] In the work reported by Gaikwad *et al.*, flexible electrodes were prepared by using a CNT coating porous membrane, as shown in Fig. 8a.[85] The prepared electrode possesses increasing tensile strengths of ≈5.5–7.0 MPa and good stability during bending tests. The final batteries exhibited enhanced mechanical performance and stability. In addition, several attempts have been made to fabricate fiber-

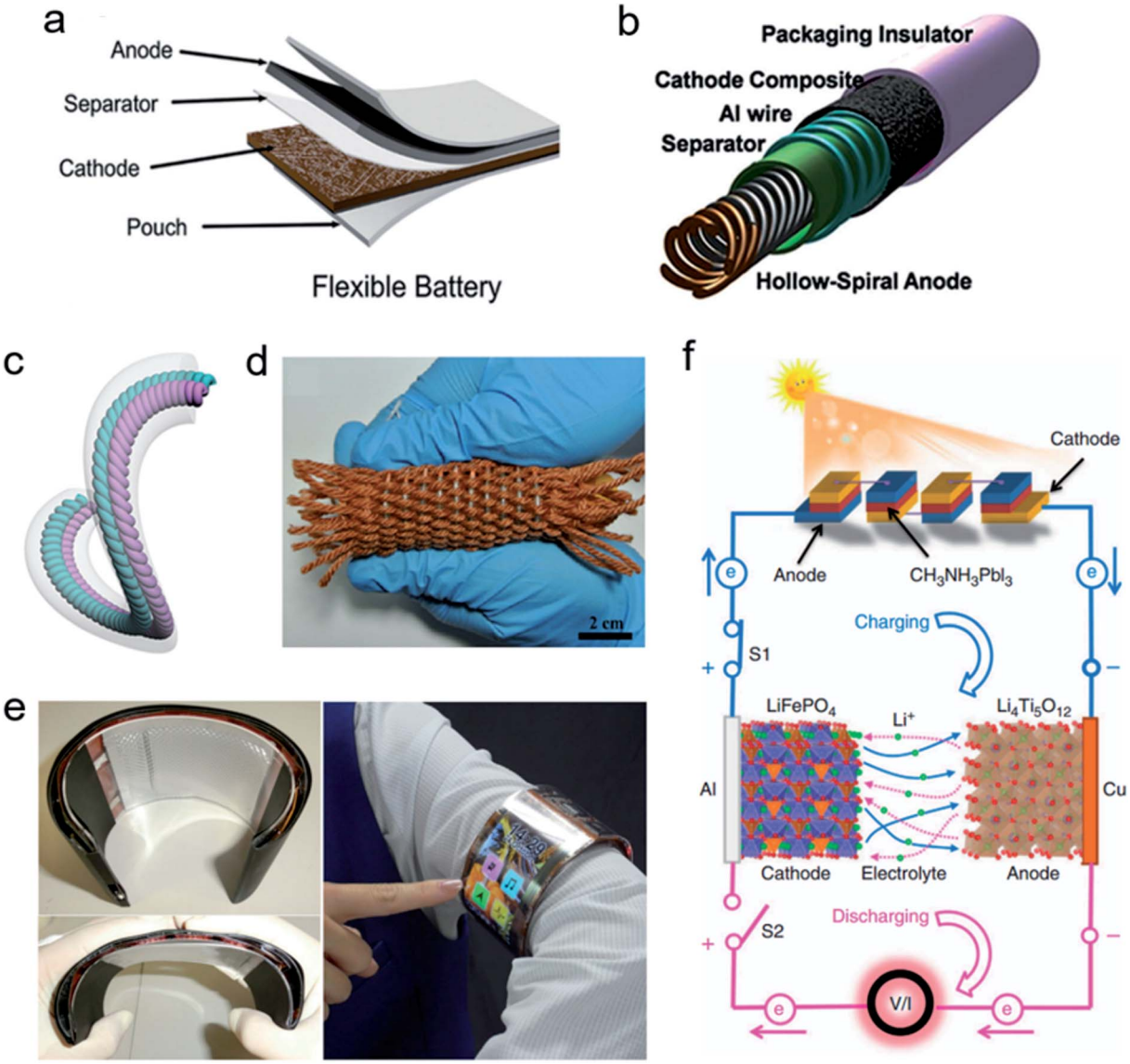

Fig. 8 (a) Schematic of a flexible LIB.[85] Copyright 2014, Wiley-VCH. (b) Schematic illustration of a cable LIB with a hollow anode system.[86] Copyright 2012, Wiley-VCH. (c) Schematic illustration of a fiber-shaped ZIB with an intertwined structure.[87] Copyright 2014, Wiley-VCH. (d) Photograph of fiber-shaped LIBs being woven into a textile.[88] Copyright 2014, American Chemical Society. (e) Photographs of a self-powered wearable display integrated with flexible LIBs and a flexible display.[89] Copyright 2014, Society for Information Display. (f) Schematic diagram of a PSC-LIB self-sustaining system.[90] Copyright 2015, Macmillan Publishers Limited.

shaped LIBs, which are more soft, flexible and durable. According to the electrode position, fiber-shaped batteries can be mainly categorized into core–shell (Fig. 8b) and intertwined structures (Fig. 8c).[86,87] Furthermore, fiber-shaped LIBs can be knitted into textiles (Fig. 8d), which is highly desirable for making wearable electronics.[88] Tajima *et al.* developed a flexible LIB and integrated it with a flexible organic light-emitting diode (LED) panel or a flexible printed circuit to construct a wrist-wearable display (Fig. 8e).[89] The flexible LIB can retain a stable electrochemical performance during a bending test, which helps to fabricate a wearable display. For self-sustaining systems composed of batteries and solar cells, the overall photo-electric conversion and storage efficiency are very important. Xu *et al.* demonstrated a self-charging device by using PSC-based direct photo-charging LIBs, as schematically shown in Fig. 8f.[90] The developed PSC-LIB exhibits a high overall photo-electric conversion and a storage efficiency of 7.80% as well as excellent cycling stability, holding promise for various potential wearable applications.

3.2.2. Flexible zinc-ion batteries (ZIBs). The inadequate energy density of SCs has hindered their applications in miniaturized electronics.[91] LIBs have a relatively higher energy density and dominate the commercial battery market, especially for portable electronic devices and electric vehicles. However, the inherent security issue of LIBs limits their application in wearable electronics.[92] Recently, rechargeable ZIBs, using non-toxic aqueous electrolytes, have attracted

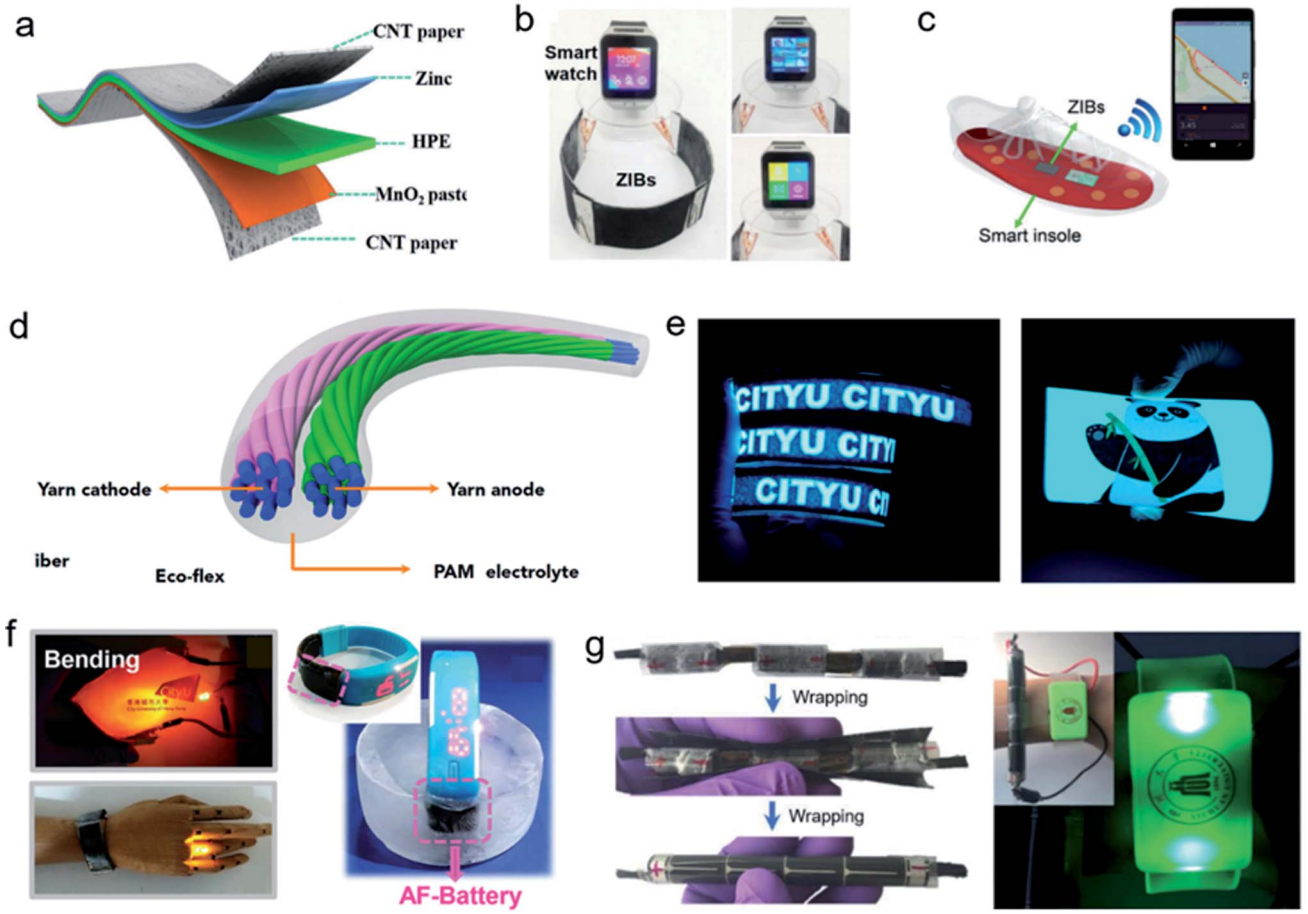

Fig. 9 (a) Schematic illustration of a flexible planer ZIB. (b) Photographs of three flexible ZIBs connected in series and powering a commercial smart watch. (c) Schematic diagram of the application of flexible ZIBs in a self-powered smart insole.[97] Copyright 2018, Royal Society of Chemistry. (d) Schematic diagram of a yarn ZIB. (e) Photographs of various electroluminescent panels powered by yarn batteries.[98] Copyright 2018, American Chemical Society. (f) Photographs of a yellow LED powered by flexible ZIBs in contact with a demonstrator's wrist (left), and a watch powered by flexible ZIBs while being sealed in solid ice (right).[99] Copyright 2019, Royal Society of Chemistry. (g) Photographs of a self-sustaining power bar consisting of a flexible SSC and three flexible ZIBs (left), and a watch powered by the self-sustaining power bar (right).[100] Copyright 2020, Royal Society of Chemistry.

significant attention due to their high safety, light weight, cost-effectiveness and eco-friendliness.[93,94] Particularly, quasi-solid ZIBs coupled with hydrogel electrolytes show high capacity and good flexibility, making them a promising alternative to LIBs.[95,96] Therefore, rechargeable ZIBs have developed rapidly in recent years, and begun to shine light on the wearable field. In 2018, Li *et al.* reported a solid-state ZIB using a polymer electrolyte, which is extremely safe (Fig. 9a).[97] The obtained ZIB not only delivers a high specific capacity (306 mA h g^{-1}), areal energy density (6.18 mW h cm^{-2}) and power density (148.2 mW cm^{-2}), but also displays an extremely safe performance after different kinds of damage, including cutting, bending, hammering, puncturing, sewing and washing in water. In addition, flexible ZIBs could power various wearable electronics, including smart watches (Fig. 9b) and smart insoles (Fig. 9c). In addition, Li *et al.* fabricated a high-performance yarn ZIB using fiber electrodes and a polymer electrolyte (Fig. 9d).[98] The PAM electrolyte endows the ZIB with a high specific capacity (302.1 mA h g^{-1}) and a high volumetric energy density (53.8 mW h cm^{-3}) as well as the advantages of being waterproof and stretchable. The obtained yarn ZIB can be further woven into a battery textile and power a LED belt and flexible electroluminescent panel (Fig. 9e). On this basis, Mo *et al.* developed an anti-freezing hydrogel electrolyte, which allows ZIBs to work under low temperature (−20 °C).[99] Thus, anti-freezing ZIBs have high potential to power portable and wearable electronics in extremely cold environments (Fig. 9f). The current electrode materials are mostly made of synthetic materials, which are usually expensive and not environmentally friendly. In response to this problem, Zhao *et al.* reported a quasi-solid-state ZIB battery derived from biomass garbage floating on the ocean.[100] The electrodes possess a special honeycomb-like structure with multi-level open channels, which contributes to electrolyte diffusion and ion transportation. As a result, the obtained ZIBs exhibit a high capacity of 306.7 mA h g^{-1}, good rate capability, and excellent cycling stability. Furthermore, as shown in Fig. 9g,

the obtained ZIBs were further combined with a flexible solar cell to form a self-sustaining power bar, which can harvest and store clean and sustainable energy from the surrounding environment and power wearable electronics, including a watch or a badge. This work provides a new strategy for the preparation of biomass-based ZIBs.

4. Integration of self-powered energy systems

With respect to self-powered energy systems, the integration process of PVCs and ESDs is quite vital. It not only affects the overall energy collection/storage efficiency of the fabricated self-powered energy systems, but also decides the appearance, flexibility, and durability of the final products. Based on the current fabrication methods, the integration of a self-powered system can be classified into two categories; they are a combination of two independent PVCs and ESDs or the integral production of PVCs and ESDs, respectively.

4.1. Combination of two independent PVCs and ESDs

In most self-powered energy systems, researchers only focus on PVCs or ESDs separately instead of studying the two parts as a whole item. For instance, Zhao *et al.* fabricated a high-performance ZIB based on defective MnO_{2-x} nanosheet-grown carbon cloth (MnO_{2-x}@CC), which not only showed excellent flexibility with an ultrahigh energy density of 5.11 mW h cm^{-2}, but also presented a high safety in a wide temperature range under various severe conditions.[101] Subsequently, they attached flexible PSCs to the surface of the obtained flexible ZIBs to construct a safe, self-powered wristband which could power a commercial smart bracelet (Fig. 10a). Moreover, cylindrical ZIBs could be prepared directly from ocean floating biomass garbage.[100] Furthermore, three obtained ZIBs were wrapped with a commercial flexible solar cell to form a self-sustaining power bar (Fig. 9g). In this kind of integration method, researchers usually choose suitable flexible PVCs to match the prepared ESDs. In this way, the resulting self-powered energy systems show better flexibility, which is suitable for fabricating wearable electronics. As shown in Fig. 10b, the self-powered wristband with desirable flexibility can be conveniently worn on the subjects' wrist during sports including outdoor running and indoor biking. However, as mentioned before, PVCs and ESDs are prepared independently, and are not specifically designed for each other. Therefore, the energy loss between these two parts will lead to a low energy transfer efficiency from PVCs to ESDs.[90,102]

4.2. Integral production of PVCs and ESDs

Another integration strategy is to fabricate PVCs and ESDs at the same time as a whole device, and they can even share

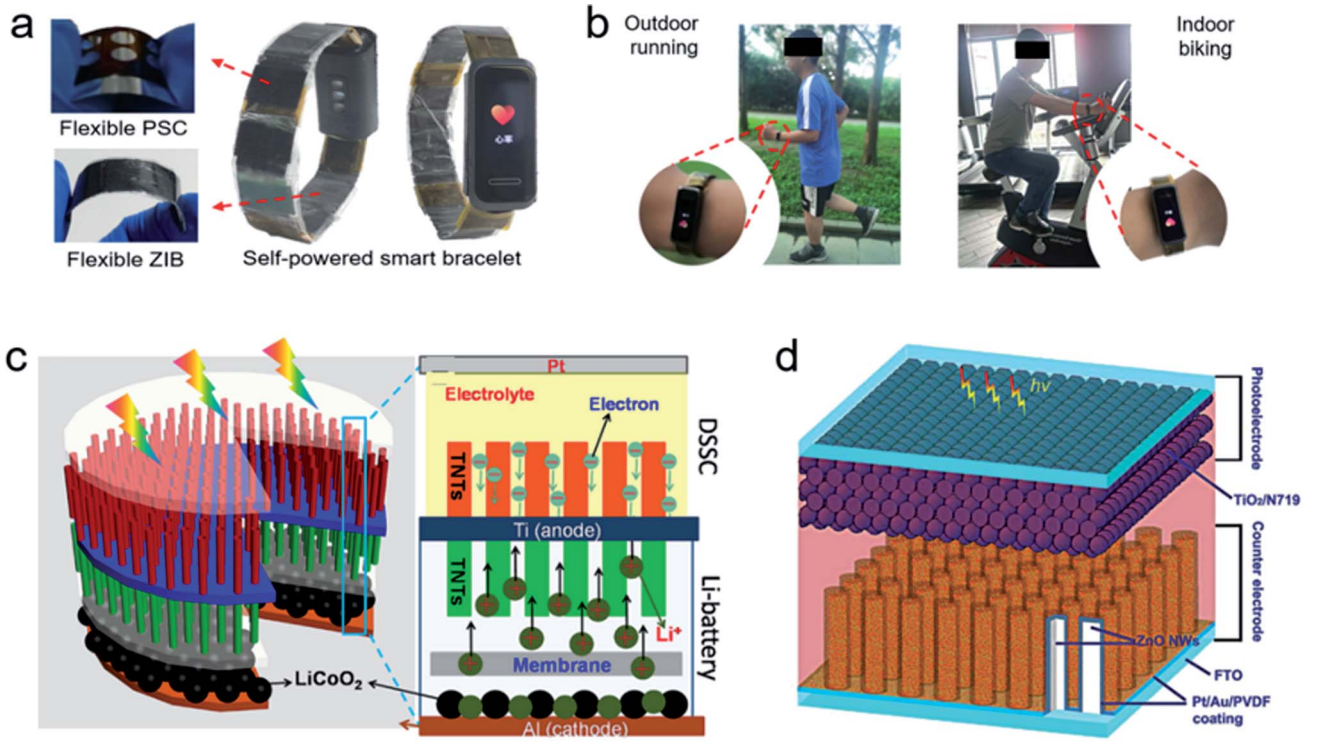

Fig. 10 (a) Digital photographs demonstrating the integration process of a self-powered smart bracelet from flexible ZIBs with flexible PSCs and a commercial bracelet. (b) Digital photographs showing a subject wearing the self-powered smart bracelet on the wrist during outdoor running and indoor biking.[101] Copyright 2021, American Chemical Society. (c) Detailed structure and working principle of the integrated self-powered energy system.[103] Copyright 2012, American Chemical Society. (d) Scheme of a TiO_2 dye-sensitized solar cell with energy storage function by modifying the CE with PVDF/ZNWA nanocomposites.[104] Copyright 2013, Wiley-VCH.

electrodes. In 2012, Guo *et al.* put forward a novel approach to fabricate an integrated power pack by hybridizing energy harvest and storage processes.[103] As shown in Fig. 10c, this power pack incorporates a series-wound DSSC and a LIB on the same Ti foil that has double-sided TiO_2 nanotube arrays. Attributed to this unique structural design, the integrated power pack was able to be charged to about 3 V in around 8 min, and the discharge capacity was about 38.89 μA h under a discharge density of 100 μA. Furthermore, in the work reported by Zhang *et al.*, an integrated self-powered system with dual functions of photocurrent output and energy storage has been developed.[104] As shown in Fig. 10d, the counter electrodes of DSSCs are modified with PVDF/ZnO nanowire array nanocomposites for the purpose of obtaining higher energy storage ability. The Pt/Au catalytic layer for a DSSC and the charge storage interface share the same PVDF surface coated with ZnO nanowires. In this integration strategy, since PVCs and ESDs are prepared as a whole device, the energy loss between the two parts is relatively small, which helps to improve the total energy conversion and storage efficiency of the whole system. However, at the time of writing, most of this class of all-in-one self-powered systems are rigid, which hinders their applications in the field of wearable electronics.[103,104]

5. Applications of self-powered energy systems in wearable electronics

Based on the various flexible PVCs and ESDs discussed above, numerous self-powered wearable electronics with different functions have been reported and are summarized in Table 3. According to different usage scenarios, the main application fields of self-powered wearable electronics include motion monitoring, pulse monitoring, sweat monitoring, gas monitoring and so on.

5.1. Motion monitoring

The real time monitoring of the motion information (including gait, motion state, position, *etc.*) is important for sports training and assessing the physical condition of the elderly or disabled.[105] With the rapid development of technology and the increase of people's demands, more and more smart wearable electronics, such as smart watches, smart bracelets, electronic skin and so on, have been developed and equipped with motion monitoring functions.[106–108] However, most of the built-in batteries for these wearable electronics are rigid and considerably bulky and require external charging or frequent replacement. Recently, Li *et al.* presented a self-powered wearable sensing system by integrating a flexible PSC, lithium-ion capacitor (LIC) and strain sensors (Fig. 11a).[109] The combined flexible PSC-LIC module achieves an overall energy conversion efficiency of 8.41% and a high output voltage of 3 V. In addition, the strain sensors in this study are prepared by coating of a thin reduced graphene oxide film onto the backbone of a PU sponge, with the advantages of cost-effectiveness, low-voltage operation, wearability, and ultrasensitivity. Thus, the self-powered sensing system can be placed on the wrist (Fig. 11b) and work for 5 h continuously and monitor physiological signals, such as finger motion (Fig. 11c).

5.2. Pulse monitoring

The heart rate is an unequivocal sign of a person's heart health.[110] Heart rate monitoring can provide the personal health status, early disease warning, and therapeutic treatments to patients.[111] Traditional heart rate monitoring needs electrocardiograph (ECG)/pulse monitoring devices and professional technical operations, resulting in inconvenience in daily life and restricting medical applications.[112] Recently, many research teams have developed flexible pressure sensors to monitor the pulse.[113–115] Furthermore, self-powered sensors can be obtained by integrating pressure sensors with energy harvesting and storage models, which is of great significance in promoting mobile health and telemedicine.[116–118] For example, Rajendran *et al.* presented a self-powered monitoring system by integrating a flexible PVC, a printed SC, a microcontroller, a pulse rate sensor and an OLED display (Fig. 11d).[119] With the help of a stretchable sweat band, the self-powered monitoring system can be worn comfortably on the wrist (Fig. 11e). In addition, the sensor is fixed on a finger for pulse rate monitoring and the results can be displayed real time. The test results illustrate that as the exercise intensity increases, the heart rate increases accordingly.

5.3. Sweat monitoring

Sweat is a type of body fluid secreted by the sweat glands of the human body.[120] It contains rich substances, such as electrolyte ions, metabolic small molecules, nutrient elements and medicine molecules.[121,122] Benefiting from easy access and rich composition, sweat detection holds great opportunities for monitoring of an individual's health conditions at the molecular level.[123–126] Zhao *et al.* developed a fully integrated and self-powered smart watch, which contains flexible PSCs and rechargeable ZIBs in the forms of a "watch strap", electrochemical glucose sensors, customized circuits, and display units integrated into a "dial" platform, as shown in Fig. 12a.[102]

Table 3 Summary of the types and performance of sensors involved in different self-powered systems

Functions	Sensors	Materials	Sensitivities	Ref.
Motion monitoring	Strain sensor	rGO@PU	25–30 kPa^{-1}	109
Pulse monitoring	Strain sensor			119
Sweat monitoring	Electrochemical sensor	Glucose oxidase	3.29 nA μM^{-1}	102
Gas detection	Gas sensor	SnO_2	13.2–1.54 wt%	134

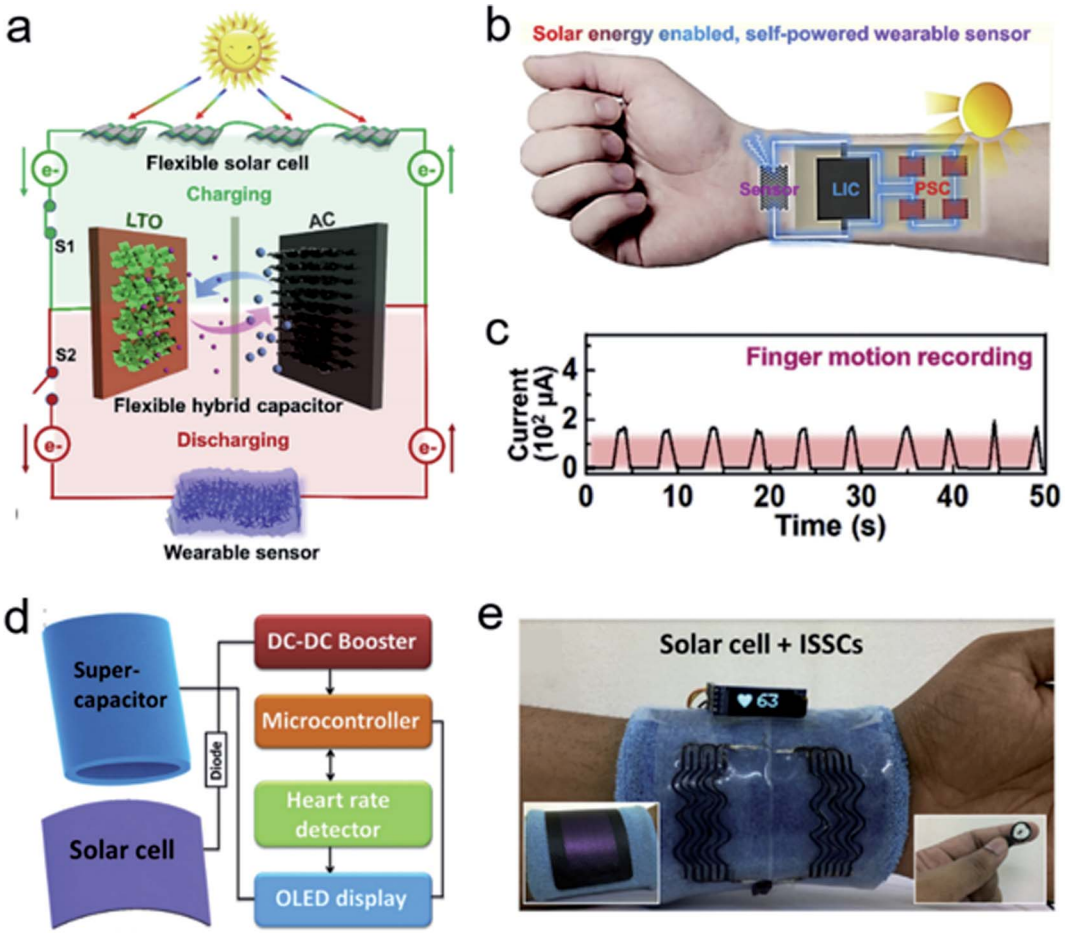

Fig. 11 (a) Schematic illustration of a self-powered sensing system consisting of PSCs, LICs and a wearable sensor. (b) Schematic diagram of the self-powered sensing system in contact with a wrist for motion monitoring. (c) The measurement results of finger motion.[109] Copyright 2019, Elsevier Ltd. (d) System-level block diagram of a self-powered sensor system consisting of a flexible PVC, a SC, a microcontroller, a pulse rate sensor and an OLED display. (e) Photographs of the self-powered sensor system for real time pulse monitoring.[119] Copyright 2019, Elsevier Ltd.

Thanks to the superior flexibility of the power devices and sensors, the integrated smart watch can be comfortably worn on the wrist (Fig. 12b). More importantly, the fully integrated smart watch can realize energy harvesting/storage, biosensing, and signal processing, integrated in a single platform. Under outdoor sunlight, the self-powered smart watch can be charged up to 6.0 V within 1 h, and achieves a cruising duration of up to 8 h. In addition, it takes around 2 h to charge the battery to 4.2 V under room light, and the stored energy can support the watch for around 1 h. As shown in Fig. 12c, volunteers can wear the self-powered smart watch during indoor cycling and outdoor running and get the real-time data of glucose levels in sweat. The as-developed fully integrated and self-powered smart watch provides a promising protocol for health monitoring and fitness management, and offers a new opportunity for the construction of wearable self-powered energy systems.

5.4. Gas detection

Some toxic gases exist in the environment where we live, such as formaldehyde, phenol and carbon monoxide.[127] The detection of these toxic gases in a simple and quick method is very important to protect our health and safety. In addition, some gases are important markers of the body.[128,129] By detecting these gases, we can learn about the health status of our body. Many research teams have tried to develop self-powered gas sensor systems, which allowed people to detect some special gases quickly and conveniently.[130–133] Lin *et al.* reported a self-powered smart sensor system integrated with printable SCs, printed interconnects, printed gas sensors and embedded SSCs, for ethanol and acetone gas detection.[134] The structure of this fully integrated self-powered platform is illustrated in Fig. 11d. More interestingly, the whole system is designed into a planar architecture and further packaged into a flexible and wearable wristband fashion, as shown in Fig. 11e. The prepared SnO_2 gas sensors deliver a sensitivity of 13.2–1.54 vt% of ethanol and 3.1–6.07 vt% of acetone. When the system is exposed to ethanol or acetone, the resistance of the SnO_2 sensor will decrease, leading to the increase of voltage applied on the LED. Thus, the LED can be turned on as a warning signal.

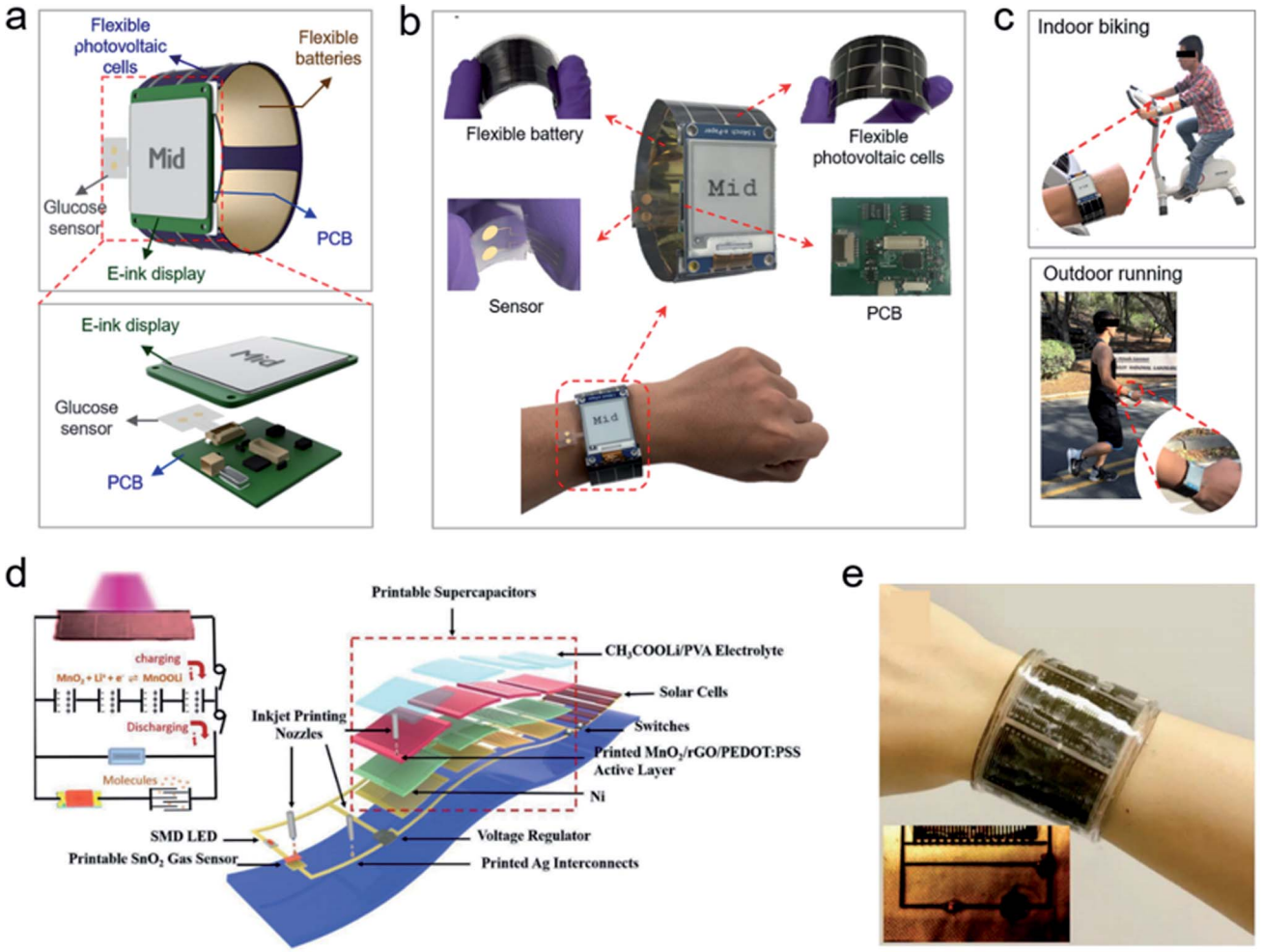

Fig. 12 (a) Schematic illustrations and (b) images of a self-powered smart watch consisting of flexible SSCs, ZIBs, a glucose sensor, a printed circuit board and an electronic ink display. (c) Photographs of a subject wearing the self-powered smart watch during indoor cycling (upper panel) and outdoor running status (lower panel).[102] Copyright 2019, American Chemical Society (further permissions related to the material excerpted should be directed to the ACS). (d) Schematic illustrations of a self-powered gas sensor system fabricated by a printable approach. (e) Photos of the self-powered gas sensor system in contact with a wristband.[134] Copyright 2018, Wiley-VCH.

6. Conclusions and prospects

Wearable electronics with good flexibility, safety and multiple functions have witnessed impressive growth and can find wide applications in motion monitoring, pulse monitoring, sweat monitoring, gas monitoring and so on. At the same time, the fast development of wearable devices has in turn promoted the development of flexible ESDs. In addition, the increasing consumption of energy and the growing environmental awareness have highlighted the demand for green energy harvesting from the environment, which spurred the development of self-powered energy systems that combine energy harvesting and storage modules. In this review, we have summarized the most recent progress in the fabrication of wearable self-powered energy systems based on flexible ESDs integrated with flexible PVCs. In this regard, firstly, recently developed flexible solar cells, which were categorized into four main different groups, including SSCs, OSCs, DSSCs and PSCs, were discussed. Afterward, recent advances in flexible ESDs, such as SCs, LIBs, ZIBs, and so on, were reported. In the following parts, various applications of current self-powered energy systems in wearable electronics have also been summarized.

The rapid advances and achievements in wearable self-powered energy systems have demonstrated their promising potential toward practical applications in future wearable electronics. However, several significant challenges still exist and further efforts are needed to achieve the commercial development of self-powered energy systems for wearable electronics. First, due to the limitation of energy density, ESDs, including various SCs and batteries, often occupy most of the space and weight of current wearable and portable electronics, which go against the miniaturization of wearables.[135] Second, most of the materials reported so far for energy harvest and storage devices are synthetic materials, such as CNTs, graphene, metal oxide nanoparticles, MXenes, metal–organic frameworks (MOFs), *etc.* These materials face many problems, such as a complex synthesis process, low yield, and environmental

incompatibility. Third, although there are many long-term cycling test standards and good results for individual parts, such as solar cells, batteries, and sensors of self-powered energy systems, there are rare reports on the long-term durability test for entire self-powered energy systems. Fourth, safety is a very critical issue for wearable electronics, especially under some extreme conditions. Most ESDs in current wearable electronics still pose serious security risks of toxicity and flammability to consumers. Fifth, the development in the field of self-powered wearable systems is still at the early stage of lab research. There are relatively few practical self-powered wearable electronics developed at present, and their costs are relatively high, which is a very big obstacle to their promotion in the consumer market.

In response to some of the challenges listed above, a lot of work can be done in the near future. Firstly, more efforts need to be continuously devoted to the miniaturization of wearable electronics. To achieve this goal, it is a must to improve the conversion efficiency of PVCs, the energy density of ESDs, and the overall energy storage efficiency to reduce the size of these components. Secondly, with the increasing awareness of environmental protection, future wearable electronic products must meet the environmentally friendly requirements. On the one hand, wearable electronics should give priority to using renewable green materials. Besides, new technologies need to be developed to process and recycle waste electronic devices to reduce environmental pollution. Thirdly, in order to be quickly recognized by consumers, the evaluation standards for self-powered wearable electronics, such as the overall energy conversion efficiency, comfort, durability, and safety, should be set up as soon as possible. It is a collaborative process which needs to be completed by the relevant enterprises and scientific research units. After that, the replaceable components of wearable electronics produced by different companies should be unified. In addition, for those wearable devices in direct contact with the human body, the safety issue of electronics is the priority among priorities in future research. First, nontoxic and non-flammable aqueous electrolytes or solid electrolytes should be developed and used in building ESDs to ensure the safety of wearable electronics. Moreover, the biocompatibility analysis of flexible substrates and active materials, including their long-term toxicity analysis, must be carried out simultaneously. Furthermore, the durability of self-powered wearable electronics should be concerned, which includes but not limited to the long-term working stability under different bending, rubbing and even washing conditions. It is highly recommended that the stability performance and fatigue life under dynamically mechanical deformations such as bending, folding, twisting, stretching, and compressing should be systematically tested. Finally, it is necessary to improve the preparation method of the materials as well as the assembly technologies and efficiency of integrated systems, to realize the large-scale production of wearable electronics. Along with the continuously increasing research and progress on energy harvest and storage devices as well as integration technology, we believe that self-powered wearable electronics will emerge in our daily lives and change our lifestyle in the near future.

Conflicts of interest

There are no conflicts to declare.

Acknowledgements

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