

Materials and Designs for Wearable Photodetectors

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Photodetectors (PDs), as an indispensable component in electronics, are highly desired to be flexible to meet the trend of next-generation wearable electronics. Unfortunately, no in-depth reviews on the design strategies, material exploration, and potential applications of wearable photodetectors are found in literature to date. Thus, this progress report first summarizes the fundamental design principles of turning “hard” photodetectors “soft,” including 2D (polymer and paper substrate-based devices) and 1D PDs (fiber shaped devices). In short, the flexibility of PDs is realized through elaborate substrate modification, material selection, and device layout. More importantly, this report presents the current progress and specific requirements for wearable PDs according to the application: monitoring, imaging, and optical communication. Challenges and future research directions in these fields are proposed at the end. The purpose of this progress report is not only to shed light on the basic design principles of wearable PDs, but also serve as the roadmap for future exploration in wearable PDs in various applications, including health monitoring and Internet of Things.

1. Introduction

As great progress has been made on the developments of highly intelligent and integrated devices, science and technologies are expected to live in and onto our bodies in different forms of wearables. Drug delivery,^[1] health monitors,^[2,3] power supplies,^[4] optoelectronic devices,^[5] various sensors,^[6] are all examples of wearables changing our daily life. Strategies to realize “wearable” have been proposed over the past few years, including electronic skins,^[7,8] textiles,^[9] wristbands,^[10] helmets,^[11] patches,^[1] lenses,^[12] etc., all fitting well with the body and clothes without interrupting or restricting physical activities of users.^[2] A great challenge concerning the design principles of wearables is the requirement of both high performance and favorable mechanical properties such as strength and flexibility. By careful selection of materials, well-designed device structures, and elaborately planned integration process, considerable efforts have been made in the above aspects, such as employing functional polymers as active layers,^[13] flexible conductive materials as electrodes,^[14–16] and novel material structures like nanomesh.^[9] Other features like transparency,^[17] stretchability,^[16] and self-healability^[7] have also

been extensively studied. Some new architectures except for conventional flexible planar devices and techniques are adopted to further optimize the performance and simplify fabrication process (like helix,^[10] core–sheath,^[4,14] and 3D printing^[18]).

Photodetectors (PDs), which transform light into other signals, have been widely used in ozone sensing, flame detection, medical imaging, memristor, astronomical exploration, and so forth.^[19–23] The most commonly and widely used PDs are the photoelectrical detectors, which convert electromagnetic radiation into electrical currents,^[24–29] and so far high-performance photodetectors have been fabricated based on that.^[30–35] The application of such devices on wearables utilizes the photodetection principles to meet the practical requirements and also provides opportunities for the realization of smart monitoring.

For traditional PDs, the device performance is evaluated by the optoelectronic properties, for example, “5S” key parameters: sensitivity, signal-to-noise ratio, speed, selectivity, and stability.^[36] However, when it comes to wearable PDs, more features need to be considered, such as portability, flexibility, and strength as they will significantly influence the performance of devices under mechanical stress.^[37] In addition, PDs-based wearable functional systems usually require additional units such as power sources, amplifiers, processors, potential memory modes, and data displays to realize the complete function. Thereby, the miniaturization and wearability of the external units are as important as that of PDs for a complex integrated system.

The research so far has been focused on exploring suitable materials, designing novel device structures, adopting efficient fabrication methods, and developing new applications. Herein, we present a progress report on different kinds of wearable photodetectors according to the material selections, device configurations, and applications. An overview of examples of wearable photodetectors is given with the detailed parameters presented in **Table 1**. The first part elaborates on the main strategies for realization of the flexibility and portability of photodetectors according to different substrates: polymer which includes planar, crumpled, and 3D substrates; paper substrates such as origami, fiber shaped substrates including metal and polymer fibers, which sometimes even serve both purposes of substrates and electrode, and other substrates based wearable PDs. In the second part, we summarize recent applications of wearable photodetectors, which are classified into three categories—monitoring, imaging, and communication—in accordance with different usages. Finally, in part three we

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DOI: 10.1002/adma.201808138

Table 1. Summary of representative examples of wearable PDs.

Design Principle	Substrate	Active material	Electrode	Strain/bending	I_{on}/I_{off}	Responsivity	Rising/decay time	EQE	Wavelength	Ref.
Small size/low consumption	Glass	TiO ₂ /SiO ₂ /ZnO	Pt	–	9.3×10^6 at 1 V	–	–	–	UV	[40]
Polymer substrate/nanomaterial	Glass	ZnO/NiO nanostructure	Pt	–	4×10^5 at 0.2 mV	>12 AW ⁻¹ at 1 V	5/9 s	–	UV	[39]
	Elastomer	PbS quantum dots	Graphene–Au	70%	1.3×10^4 at 0 V	0.13 AW ⁻¹ at 0 V	0.087/0.129 s	12%	Near IR	[65]
Polymer substrate/organic materials	PI	ZnO granular nanowires	Nanosilver	$R < 333 \mu\text{m}$	$\approx 10^5$ at 1 V	7.5×10^6 AW ⁻¹ at 1 V	0.56/0.32 s	–	UV	[76]
	PET	Porphyrin–SWNT	Graphene	50%	–	3.1×10^{-3} AW ⁻¹ at $V_{DS} = -1$ V	Both > 100 s	–	UV	[62]
Fiber substrate/nanomaterials	PET	P3HT:PCBM	PEDOT:PSS/EGaIn	$R \approx 2.1$ mm	$\approx 10^2$ at -1 V	Max = 0.086 AW ⁻¹	–	25.3% at -1 V	Near-UV to visible	[81]
	Ti microfiber	p-CuZnS/n-TiO ₂	Ti/graphene	60°	Photocurrent \approx mA at 3 V	640 AW ⁻¹ at 3 V	Both < 0.2 s	2.3×10^5 at 3 V	UV	[37]
Paper substrate	Carbon fiber/Cu fiber	Perovskite	Cu/carbon	–	393 at 0 V	0.5629 AW ⁻¹ at 0 V	Both < 0.3 s	–	Broadband	[101]
	Paper	PMMA LA	–	–	–	–	–	–	UV with color change	[92]
Other substrate	Paper	ZnO nanowire	Carbon	1000% strain/30° bending/360° twist	–	–	1.1/0.3 s	–	UV	[94]
	Ni textile	ZnO nanrod array	Ni/graphene	120°	$\approx 10^2$ at 1 V	0.27 AW ⁻¹ at 1 V	6/18 s	–	UV	[106]
	Mica	reduced graphene oxide-ZnO	Ag	180°	–	3.24 AW ⁻¹ at 1 V	17.9/46.6 s	–	UV	[107]

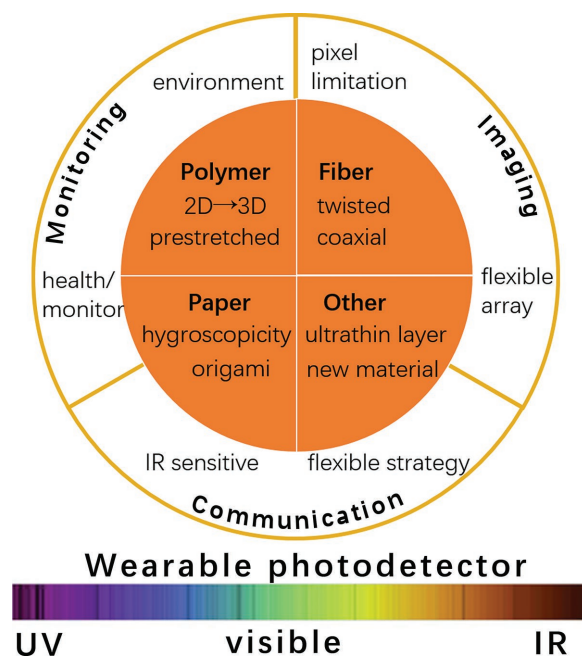


Figure 1. The design principles and applications of wearable photodetectors.

give an overall summarization and discussion about the future development directions of wearable photodetectors, as well as the challenges ahead. **Figure 1** schematically illustrates the outline of this review.

2. Materials and Architecture Design of Wearable PDs

The past decade has witnessed great progress on the developments in the design and fabrication of wearable PDs. In some work, the small size and the resulting portability of the devices are defined as the sign of “wearable,” while in others PDs exhibiting high optoelectronic performance under low external applied voltage are regarded to be wearable as large power supply units are not needed.^[38–41] The self-powered property is thus attracting increasing attention in order to get rid of external power sources to make the whole systems portable and wearable.^[42–44] Generally, there are two design routes to achieve self-powered PDs. One route is to integrate PDs with miniaturized energy-harvesting/storage components like solar cell,^[45] supercapacitor,^[46] and nanogenerator.^[47] The energy-harvesting component, which is typically integrated with photoconductive PDs, usually operates under an external bias to efficiently convert environmental energy into electricity. Energy-storage components can store different forms of energy for future usage. The other route is to develop self-powered photodiodes based on the photovoltaic effect, where the incident signals intrinsically power the device due to the built-in potential.^[48] The first strategy, which is compatible with various PDs and wearable energy-harvesting/storage devices, provides abundant choices for the system constructions. Thus, the high-performance wearable systems are realized at the cost of

increased complexity. The advantages of the second method lie in the simplicity of its structure and the maintenance-free feature, but the photovoltaic working mechanism sets bottleneck to the self-powered photoelectric performance.^[49] The self-powered ability is generally limited by the open-circuit voltage (V_{oc}), and the photocurrent is limited by the zero responsivity gain in photovoltaic devices. Future self-powered photodetectors should focus on these challenges, while novel flexible power sources^[50,51] may also be a promising approach in solving the problems. PDs with a high photocurrent also exhibit the potential to provide enough electric signals without amplifiers, which plays an important role in wearable devices. The illumination area is obviously limited in miniaturized photodetectors, and a high photogain is extremely desirable to generate decent photocurrents. The traditional photoconductive responsivity gain is often observed in photoconductive devices like metal–semiconductor–metal (MSM) structure^[52] and photo field-effect transistors (photo-FETs). The photoconductive gain is based on the unbalanced trapping of electrons or holes in photoconductive materials, which prolongs the lifetime of the trapped carriers and provides additional photoconductivity at the sacrifice of response speed.^[53] Apart from the photoconductive gain, the novel photovoltage field-effect transistors have enabled the photovoltage and transconductance gain, which delivers ultrafast response speed along with high responsivity gain.^[54]

Even though a small size and a low requirement for applied voltages are essential for wearable PDs, currently, a majority of scholars in the relevant field believe that the key features of truly wearable PDs are the intrinsic flexibility and the ability to maintain a stable performance under a certain range of bending, stretching, compressing or twisting.^[55,56]

Three approaches/(fundamental design principles) are generally adopted to achieve wearable/flexible PDs.^[57] 1. Selecting intrinsically flexible materials such as polymers, paper, fiber, and ultrathin semiconductor layers. 2. Making modifications to the materials to form flexible structures or increase the flexibility of the material, such as crumpled and helical structures. 3. Forming rigid island structures to turn rigid components to a flexible integer. Each of these approaches has its own advantages and drawbacks. PDs based on intrinsically flexible materials are more conformal and comfortable in terms of wearable and the fabrication process is less complicated in comparison with the other two approaches. However, the number of intrinsic flexible materials suitable for the construction of wearable PDs is rather limited. Besides, the stretchability of the devices based on intrinsic flexible materials is still far from satisfactory and the performance of the devices is susceptible to strain. There is a significant improvement of stretchability, deformability, and light absorption on crumpled structures, but it also means that the photocurrents are sensitive to strain. Rigid island approach holds the ability to turn inflexible components into a flexible integer and has a fundamental tradeoff between active area and stretchability with a stable current. The requirements for materials in each component are less in rigid island structures but the connection between components is a huge challenge.

In order to be intrinsically wearable, the substrate, active materials and electrodes of PDs and the other electronic parts

in the systems have to be elaborately selected and designed based on the aforementioned design approaches. To date, a variety of materials have been used as the substrates of wearable PDs, among which polymer, paper, and fiber are the three most common types due to their facile preparation process, great abundance, and high deformability. In Section 2, we will discuss about the construction of wearable PDs from the perspective of substrates while exploring their fundamental design principles.

2.1. Polymer Substrate Based PDs

Polymers, such as polyimide (PI),^[58–61] polyethylene terephthalate (PET),^[62–64] polyethylene naphthalate (PEN),^[65] and polydimethylsiloxane (PDMS),^[66,67] have been widely utilized as substrates for wearable PDs for the inherent flexibility.

For the photoactive layers, nanomaterials, including 0D materials, 1D materials, and 2D materials, are extensively used thanks to the micromechanical properties brought by the nanostructures. Deformable and bendable nanomaterials such as ZnO nanoparticles,^[68] ZnO nanorods,^[69] perovskite nanowires,^[70] MoO₃ nanosheets,^[71] graphene,^[72] and monolayer WS₂,^[73] have thus been constructed on the polymer substrates for wearable PDs. For the device fabrication process, directly placing the nanomaterials on the polymer substrates, such as spin coating,^[69] suspension dropping,^[64,68] printing,^[59] magnetron sputtering,^[74] pulsed-laser deposition,^[61,75] transferring,^[58,71] etc., is the main strategy to fabricate polymer substrate based wearable PDs. Adopting the above-mentioned fabrication strategies, a flexible all-printable ZnO granular nanowire-based PD using Ag nanowires as the electrodes was presented by Liu et al. A moderate bendability with a bending curvature of $>30\text{ cm}^{-1}$ was demonstrated, as shown in **Figure 2a**.^[76]

Apart from nanostructured materials, organic materials such as poly(3-hexylthiophene) (P3HT):[6,6]-phenyl C₆₁-butyric acid methyl ester (PCBM) blend, *N,N'*-bis(2-phenylethyl)-perylene-3,4:9,10-tetracarboxylic diimide (BPE-PTCDI), poly[2,5-bis(3-tetradecylthiophen-2-yl)thieno[3,2-b]thiophene](PBTTT):[6,6]-phenyl-C₆₁-butyric acid methyl ester (PCBM), polyin-dacenedithiophene-pyridyl[2,1,3]thiadiazole-cyclopentadithiophene (PIPCP), etc. are also built on polymer substrate to fabricate wearable PDs to take the advantage of their elasticity.^[77–82] A 3D printed organic PD was successfully fabricated with a stacking structure, using P3HT:PCBM as the active layer and poly(ethylenedioxythiophene):polystyrene sulfonate (PEDOT:PSS)/eutectic gallium indium (EGaIn) liquid metal as electrodes, as shown in **Figure 2b**.^[81] This PD shows a relatively stable optoelectronic performance while the bending radius is 2.1 mm (**Figure 2f**). In addition, the PD can be integrated into sensing arrays for future applications.

Based on polymeric substrates, certain modifications have been made to these substrates to amplify the flexibility, bendability, and light absorption by forming crumpled structures.^[67,78,83–85] Kim et al. proposed a stretchable photodetector based on a crumpled hybrid structure of graphene–gold nanoparticle with exceptional optoelectronic performance and mechanical stretchability at a tensile strain of $\approx 200\%$.^[72] A polymer substrate was first biaxially prestrained

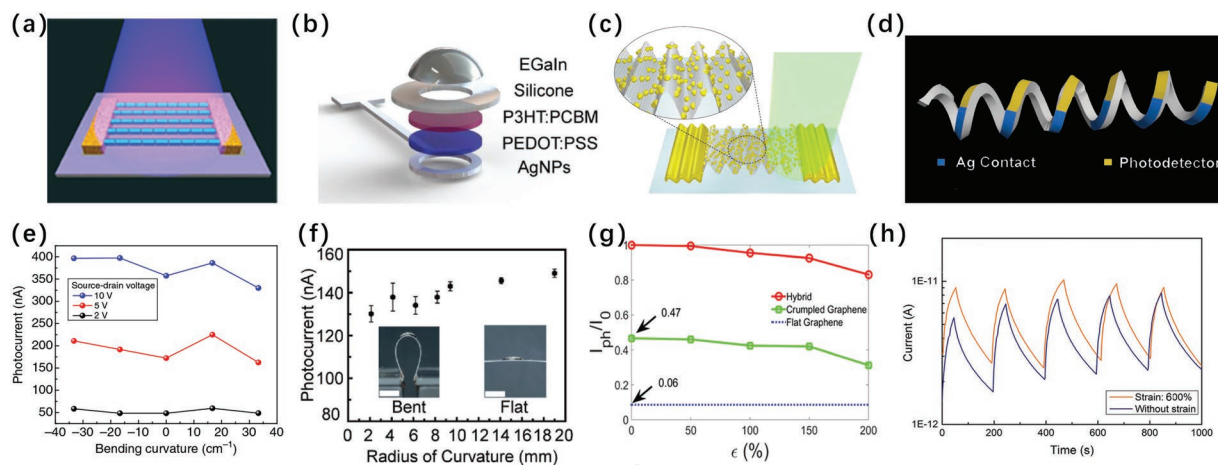


Figure 2. Polymer substrate based photodetectors. Architecture and deformability of a,e) ZnO granular nanowire-based PD using nanosilver electrodes on polyimide substrate. Reproduced with permission.^[76] Copyright 2014, Springer Nature. b,f) P3HT:PCBM based organic PD using PEDOT:PSS/EGaIn electrodes on a PET substrate. Reproduced with permission.^[81] Copyright 2018, Wiley-VCH. c,g) Graphene–Au nanoparticles based PD using gold electrodes on a prestrained polymer substrate. Reproduced with permission.^[72] Copyright 2016, Royal Society of Chemistry. d,h) ZnO nanowires based PD using Ag electrodes on a helical PDMS substrate. Reproduced with permission.^[86] Copyright 2018, Royal Society of Chemistry.

before graphene–Au hybrid was transferred onto it. After hybrid transferring, the strain was released and a crumpled active-material layer was formed along with a crumpled substrate as shown in Figure 2c. This structure contributed to a highly enhanced photocurrent of the PD compared with the flat counterpart as a result of higher light absorption. (Figure 2g).

Polymer substrate based wearable PDs with a higher stretching capacity (up to 600%) has been reported by using a unique helical structure.^[86] ZnO nanoparticles were coated on the PDMS substrate precoated on a screw as the seed layer for growth of ZnO nanobranched arrays by hydrothermal synthesis. Then half side of the screw was coated using Ag paste as the contacts. A helical structure device would take shape when peeling the whole substrate from the screw thread, as illustrated in Figure 2d. This PD possesses a high stretchability of $\approx 600\%$ strain (see Figure 2h). Such a high stretchability mainly benefits from the unique helical structure that rotates to accommodate deformation and thus it is very promising for construction of other wearable devices. With the same design principle of turning 2D shape into 3D architectures, configured polymer films served as the substrate to support graphene and MoS₂ active material for constructions of 3D flexible PD.^[87]

Polymer substrates are frequently employed in wearable PDs thanks to their own advantages: 1. Easy access to polymer. 2. The thickness and the shape of the substrate can be precisely controlled. 3. The intrinsic flexibility of polymer. 4. The possibility for large-scale applications. However, there still exist challenges. Even though polymer substrates exhibit certain intrinsic flexibility, such flexibility is unsatisfactory without geometrical design. Besides, the surface of polymer is generally slippery, which hampers the adherence of materials.

2.2. Paper Substrate

Paper substrate is another commonly used substrate for the fabrication of wearable PDs due to its adequate availability, low

cost, mechanical flexibility, and biocompatibility.^[88–90] The fundamental design principles of paper substrate based PDs are partially similar to that of polymer substrate based PDs. Works related to paper substrate based wearable PDs shared some similarities in the efforts on exploring nanomaterials and organic materials with polymer substrate based PDs, by taking advantage of the intrinsic flexibility of these materials or structures.^[88,91]

However, there is a better adhesion between paper and materials due to the porous structure of paper. Thus, some PDs taking advantage of the hygroscopicity have been proposed. In addition, the bendability of paper being better than that of polymer leads to structural designs such as origami. Moreover, paper substrate is extremely favorable for the construction of environment-friendly and disposable devices.

Taking advantage of the hygroscopicity and porous structure of paper, ZnO and MoS₂ were synthesized as the photoactive material on cellulose paper presaturated with seed solution via hydrothermal growth as shown in Figure 3a. Recently, a paper substrate based ultraviolet (UV) PD using a photoelectrochromic ink has been presented by Zou et al.^[92] This work made full use of the moisture absorption property of paper by adopting a photoelectrochromic ink as the photoactive material to indicate the type (UV-A, UV-B, or UV-C) and intensity of the ultraviolet light. Phosphomolybdic acid (PMA), which is light yellow (almost colorless) with the absorption peak at about 310 nm on its pristine stage and turns heteropoly blue when it is reduced under UV illumination with changed absorption edge, was selected as the photoactive material with lactic acid (LA) as the e[−] donor. The invisible aqueous solution containing PMA and LA was drawn on paper substrate with transparency films as the coating filters as shown in Figure 3b. When exposed to UV light, the color of smileys changed and the color-changing degree depended on the dosage of UV light. Thus, this paper substrate based UV PD not only was naked-eye observable but also could be tuned according to the specific need of individuals, for the UV light dosage absorbed by photoactive materials can be modulated by changing the

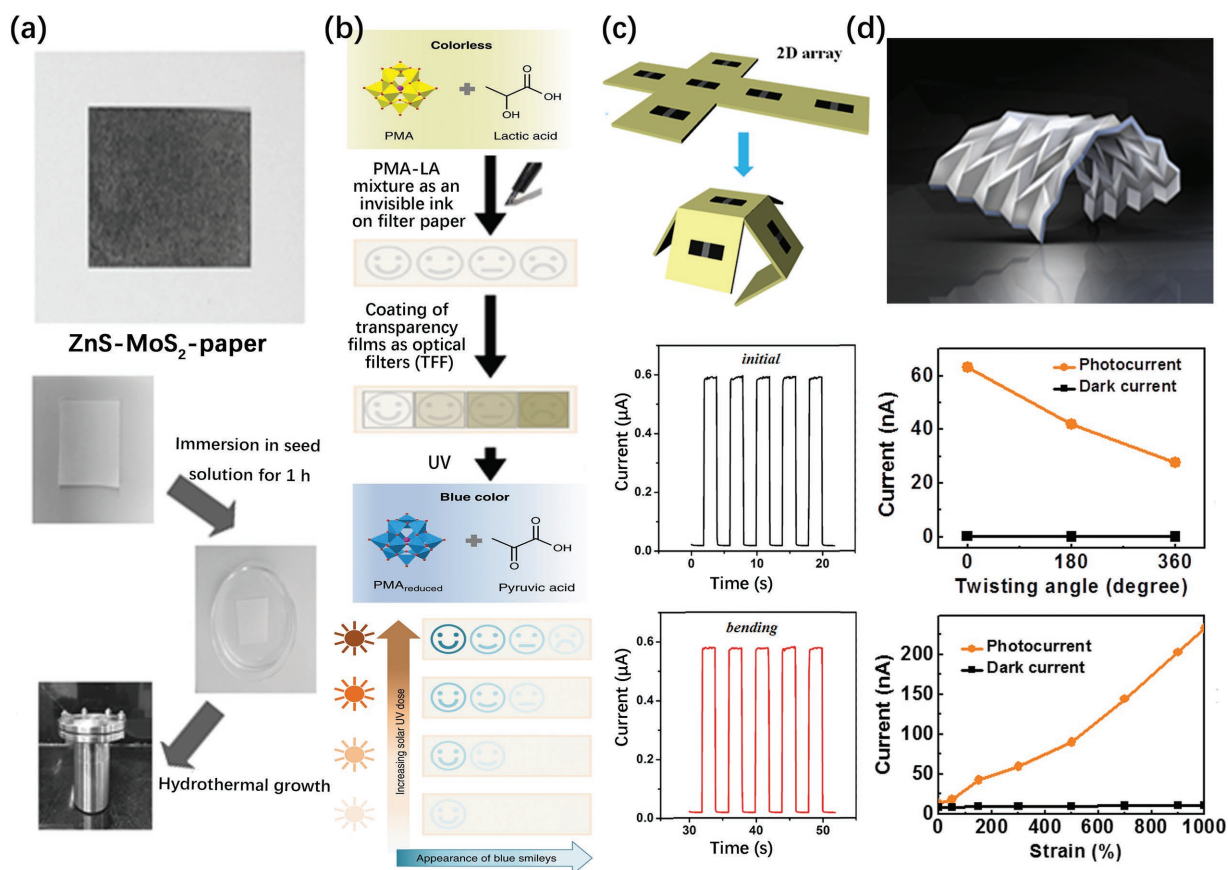


Figure 3. Paper substrate based photodetectors. Architecture and deformability of a) ZnS–MoS₂ paper PD. Reproduced with permission.^[91] Copyright 2017, Wiley-VCH. b) Naked-eye monitoring PD with paper substrate. Reproduced with permission.^[92] Copyright 2018, Nature Publishing Group. Copyright 2017, American Chemical Society. c) Origami perovskite PD based on paper substrate. Reproduced with permission.^[93] Copyright 2017, American Chemical Society. d) Miura-origami structure PD. Reproduced with permission.^[94] Copyright 2017, American Chemical Society.

number of transparency films above the ink. Furthermore, the absorptions of the active material for UV-A, UV-B, and UV-C were discrepant, which helped to differentiate the illuminated light. This distinguishing capacity was highly favorable for discerning three types of UV lights which hold remarkable difference in effect on human body.

Strategies such as applying origami to the paper substrate, would greatly enhance the stretchability and spatial recognition of a PD. Fang et al. proposed a spatially discernible wearable PD as shown in Figure 3c.^[93] CH₃NH₃PbI₃ and pencil trace were used as the active material and electrodes, respectively. The as prepared PD showed stable optoelectronic properties even after 1000 bending cycles. Also inspired by origami technology, a wearable PD that could be stretched up to 1000% strain, twisted up to 360°, and bended up to ±30° while still maintaining a decent detecting capacity was presented.^[94] Carbon pastes were first screen-printed onto a piece of paper followed by the screen-printing of ZnO nanowire ink. The whole system was then folded into a Miura-origami structure with silver paste to reinforce the foldlines as shown in Figure 3d.

2.3. Fiber Substrate Based PDs

Fiber, as an intrinsically flexible and wearable structure, is a promising candidate for developing wearable PDs. Several

eye-catching work of wearable PDs based on fiber substrates has been reported recently where nickel wires, titanium wires, Kevlar fibers, carbon nanotube (CNT) fibers, ZnO fibers, etc. were employed.^[95–99] According to current fiber based PDs, the most extensively used fibers for wearable PDs are metal fibers (for it can serve as an electrode as well as the reactant for photoactive material synthesis) and polymer fibers (for it can be reformed into any shape with little effort and serve as encapsulation material).^[100] Semiconductor fibers and CNT fibers are also used as substrates for wearable PDs but relevant studies are not as abundant as the former ones. On the basis of bendable fibers, the close and robust interface between active materials and fiber substrate is a primary requisite for wearable PDs.

A typical fiber based PD has a twisting structure, with one fiber electrode incorporated with photoactive materials, and entwined by the other fiber electrode, as demonstrated by the p-CuZnS/n-TiO₂ photodetector from Xu et al.^[37] The Ti micro-fiber played three roles here. First, it worked as the substrate to provide the device with strength and flexibility. Second, it functioned as the electrode with a high electrical conductivity. Third, it served as the source for growth of TiO₂ nanotube arrays. A transparent p-type conducting CuZnS film was then conformably deposited on TiO₂ nanotube arrays to form a heterojunction. The other electrode, carbon nanotubes fiber, was twisted

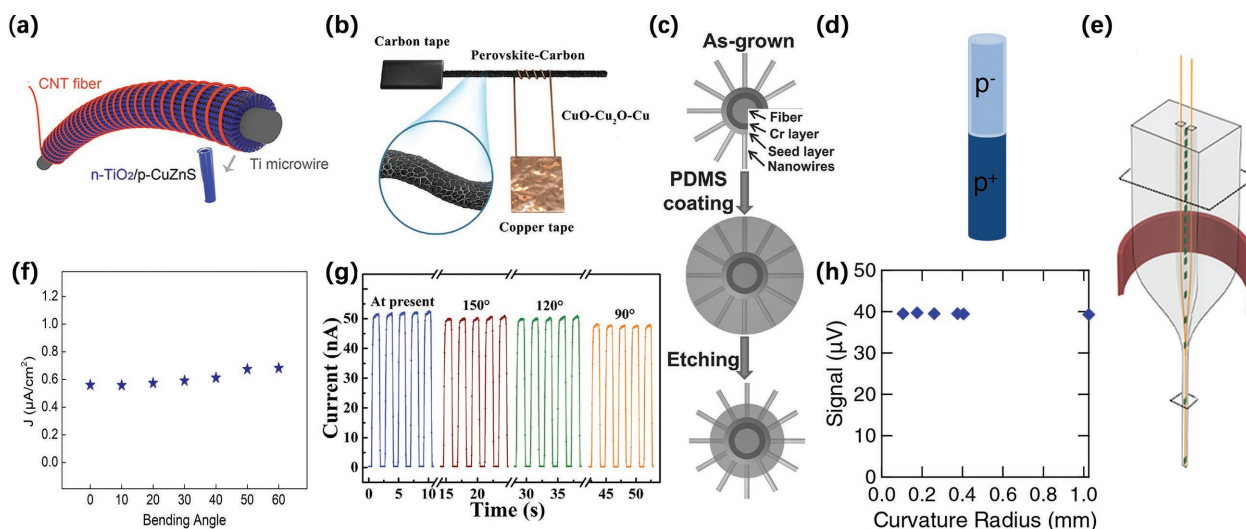


Figure 4. Fiber substrate based photodetectors. Architecture and deformability of a,f) p-CuZnS/n-TiO₂ composite based PD on Ti fiber. Reproduced with permission.^[37] Copyright 2018, Wiley-VCH. b,g) Double-twisted fibrous PD. Reproduced with permission.^[101] Copyright 2018, Wiley-VCH. c) Kevlar fiber substrate based PD with ZnO nanowires as photoactive material. Reproduced with permission.^[95] Copyright 2015, Wiley-VCH. d,h) Carbon fiber based PD. Reproduced with permission.^[96] Copyright 2018, American Physical Society. e) Drawing process of the fabric-based PD. Reproduced with permission.^[100] Copyright 2018, Springer Nature.

on the p-CuZnS/n-TiO₂, as shown in Figure 4a. Mechanical test indicated that this fiber shaped PD was bendable (see Figure 4f) and held considerable robustness as its optoelectronic properties remained stable after 200 bending cycles. Similar to the Ti wire based PD, a self-powered PD was successfully fabricated using Zn wire as the substrate as well as one electrode while ZnO nanowires were synthesized as the photoactive material.^[99]

Apart from the single-twisting fiber device, both fiber electrodes can be incorporated with photoactive materials and twisted together for a dual-wavelength PD. For example, Sun et al. presented a double-twisted broadband perovskite PD based on fiber substrates.^[101] A carbon fiber sequentially coated with a TiO₂ and a perovskite layer was then twisted with air-annealed Cu wire whose surface was covered with Cu₂O and CuO, as shown in Figure 4b. The double-twisted PD showed a high deformability (Figure 4g) while the facile but well-designed fabrication process makes the effective charge separation and collection possible.

Other than the twisting structure, coaxial structure is also typically employed in designing fiber-shaped devices. Using Kevlar fiber as the substrate, a self-powered coaxial fiber-shaped UV PD was synthesized by Zhang et al.^[95] A Cr layer was deposited on the surface of the fiber as one electrode and ZnO nanowires were grown on it as the active material. PDMS, which was coated on the bottom part for the ZnO nanowires, was used to improve the mechanical performance of the device. Finally, a Pd layer was sputtered on ZnO nanowires as the other electrode. The as synthesized PD (Figure 4c) showed high stability in bending, twisting and durability test. Other attempts have used polymer (specifically polycarbonate) fibers for support and protection, while miniature semiconductor devices and metallic wires were embedded into the polymer fiber substrate by a thermally drawing process, as shown in Figure 4e.^[100] This device could still function normally after being washed by a

household washing machine, exhibiting an excellent robustness and bendability.

Without any complex structure, single carbon fiber based PD was synthesized via doping and silver coating as shown in Figure 4d.^[96] The as fabricated PD exhibited no signal decaying even with a bending curvature of 100 μm (see Figure 4h).

Fiber-based PDs generally exhibit show excellent optoelectronic performances due to their compact structures and the ability to absorb light from 360°. Fiber-based PDs also show an advantage for the future integration with other applications. For example, fibrous PDs could be woven with textiles even though the choice of suitable fiber substrate is currently limited to metals.

2.4. Other Substrates

Apart from polymer, paper, and fiber, there are some other materials used as substrates for wearable PDs such as silicon,^[102,103] mica,^[104] etc.^[105]

Silicon substrate based PD was turned to “soft” from “hard” by reducing the thickness of silicon.^[102] Etching the silicon layer to 30 μm dramatically enhances the bendability of the substrate compared with the one with larger thickness. Based on such soft silicon layer, active materials, and metal electrodes were sequentially deposited to form a PD with a decent bendability, as shown in Figure 5a,e.

Ni textile was also employed as the substrate to form a highly bendable PD as shown in Figure 5b.^[106] ZnO nanowire arrays were grown on Ni textile by a hydrothermal synthesis. Ag nanowires were dispersed on part of the ZnO nanowire arrays to facilitate electron-hole separation. Then the poly(methyl methacrylate) (PMMA)-supported graphene film was assembled on the surface of Ag nanowires as one electrode while the Ni textile acted as the other. The as fabricated PDs exhibited no current degeneration under 120° bending (see Figure 5f), showing a decent wearability.

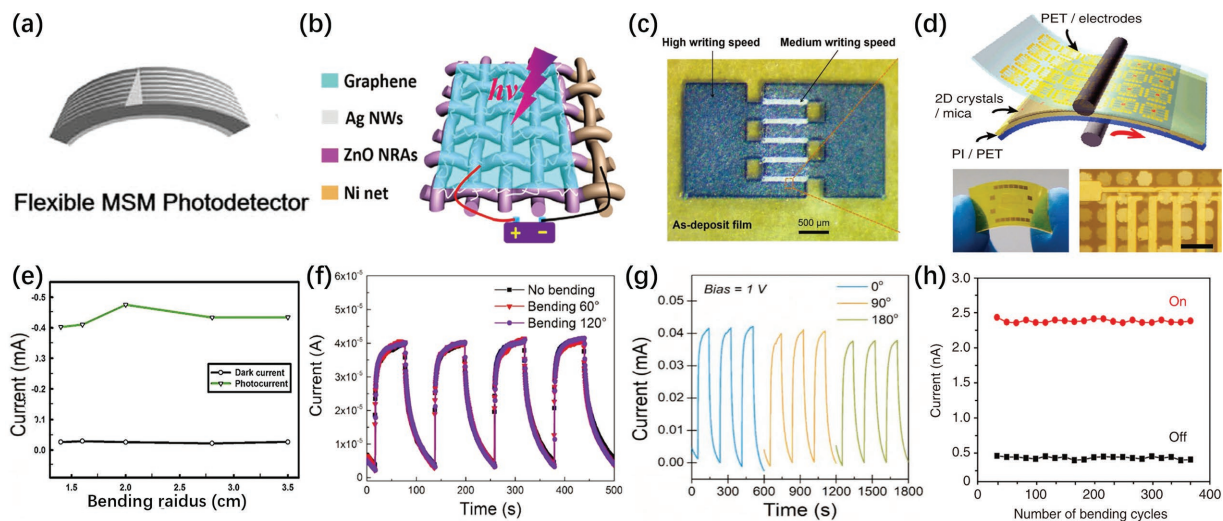


Figure 5. Wearable photodetectors based on other substrates. Architecture and deformability of a,e) flexible PD based on 30 μm thick silicon substrate. Reproduced with permission.^[102] Copyright 2016, Elsevier B.V. b,f) Ni textile substrate based PD. Reproduced with permission.^[106] Copyright 2017, Wiley-VCH. c,g) Mica substrate based PD using single-step selective laser writing. Reproduced with permission.^[107] Copyright 2018, Wiley-VCH. d,h) Mica substrate based PD fabricated through lamination. Reproduced with permission.^[104] Copyright 2015, Macmillan Publishers.

Bendable mica substrate was combined with one-step selective laser writing strategy for the fabrication of wearable PDs as shown in Figure 5c.^[107] The mixture of graphene oxide nanosheets and zinc acetate solution was coated on the mica substrate and femtosecond laser direct writing was conducted to reduce graphene oxide and synthesize ZnO, which served as the photoactive material. This device was able to maintain a stable performance under 180° bending angle as shown in Figure 5g. Besides, the morphology of active materials can be controlled by adjusting the writing speed, which offered a convenient way for property control. Another mica substrate based PD using 2D In_2Se_3 crystal arrays as photoactive material, metals as electrodes and PET as packaging material showed a decent bendability and a commendable optoelectronic performance, as shown in Figure 5h. In this work, lamination process was adopted to combine materials together as shown in Figure 5d.^[104]

In the architecture of wearable PDs, the intrinsic flexibility of materials, including substrates, photoactive materials, and electrodes is fully utilized. Moreover, mechanical designs to the substrate such as prestretching and origami have been widely adopted to increase the deformability as well as the light absorption of the device.

Polymer, paper, and fiber are the three most commonly adopted substrate materials. Polymer and paper share some similarities on fundamental design principles for both of them show a certain degree of flexibility and are suitable for large-scale fabrication. However, there is a better adhesion between paper and materials thanks to the porous structure of paper. Moreover, the higher bendability of paper makes spatial designs such as origami possible, which turns 2D structures into 3D configurations. Fiber substrate based PDs, apart from being breathable, can also absorb light from 360° and can be woven with textiles for further applications.

Designs to improve optoelectronic performances of the wearable PDs are also studied. Resonant optical cavity device

architecture,^[79] plasmonic enhancement,^[72] charge carrier transfer engineering,^[81] single-mode waveguides,^[108] etc. have been explored and employed for desirable device properties such as more efficient electron–hole separation and a higher responsivity.

Due to the limitation of space, here we mainly focus on the materials selections, especially substrates, of wearable PDs, while performances of devices are also of vital significance. Table 1 shows the materials selection, mechanical properties, and optoelectronic properties of some typical wearable PDs for consultation.

3. Applications of Wearable Photodetector Systems

The typical applications of photodetector systems can be divided into three types: monitoring, imaging, and optical communications. Wearable photodetector systems inherit the basic functions of these applications but set additional challenges due to the required flexibility. Monitoring system generally requires a precise output of the intensity or intensity variation of the incident light, which contains the target information. Thus, the requirements of photoresponsivity and detectivity are high for the photodetectors in monitoring systems to guarantee a sufficient sensitivity. The flexibility of wearable PDs actually limits the device structures and decreases the optoelectronic performances to some extent. Therefore, more attention should be paid to the device fabrication for wearable monitoring. Although some monitoring applications like UV monitors and light power meters are based on the direct response to external light sources, plenty of other applications like smoke detectors, biosensors, and locators rely on the integration with other electronic devices, such as light sources (light emitting diode (LED) etc.), where the target information hides in the scattering and absorption data. However, the integration of LEDs with PDs on flexible structures may bring

new problems in these occasions. Photoimagers are based on photodetector arrays, where each photodetector serves as an individual pixel. Traditional commercial imagers are based on silicon technology, where the substrates are “hard.” While for flexible substrates, high-pixel integration, and the relevant electric circuits are difficult to achieve. Thus, this is a significant challenge that high-performance wearable imagers would need to solve in the future. Optical communication systems have a strict limitation on wavelength, thus two things should be taken into consideration in designing wearable systems. 1. Avoiding the interference from environmental light. 2. Diminishing the signal attenuation during signal transmission. To meet these requirements, color buffers and complicated three-electrode device architectures, which are rather difficult to realize on flexible structures, are needed. Apart from the aforementioned contents, the complex signal treatments also rely on additional electric components to provide logic and memory function, setting up additional obstacles to the wearable applications. Fortunately, great progress has been made on the related territory, and we will give a detailed description about the wearable applications of photodetector systems in the following part, discussing the progress and pointing out the unsolved problems.

3.1. Wearable Monitoring Systems

Photodetectors are particularly promising candidates for health monitors due to their essential function of light detection. To gain a better understanding of the realization of wearable monitors based on photodetection, here we sort these wearables into two categories according to different applications: (1) monitoring internal signals involving heart rates, pulses, blood oxygen levels, etc.; (2) monitoring ambient light, for instance, UV intensity.

Several reports introducing photodetectors into health monitoring have attracted increasing attention. As can be seen from **Figure 6b**, the near-field communication (NFC) technology, which powers up the photodetector and LEDs as well as other electronics, is used to fabricate a miniaturized, wireless and battery-free device for monitoring signals of photoplethysmograms and reflectance pulse oximetry.^[109] The commercial photodetector collects the backscattering or transmitting light (red and infrared) signals, which are related to the blood oxygenation, heart rate, and heart rate variability. The ultrasmall system can be mounted on soft skin or hard surfaces, presenting the possibilities of achieving highly integrated wearables. Other interesting findings related to photoplethysmograms sensors utilized phototransistors with lower power consumption.^[110] The integration of LEDs and PDs continues to be a great strategy for health monitoring. One groundbreaking work successfully achieved an ultrathin flexible system (optoelectronic skins) with a total thickness of just 3 μm using red polymer LEDs and organic photodetectors (**Figure 6a**).^[111] The device could not only work as a pulse oximeter but also display visualized results. It is noteworthy that the stretchable nature results from the encapsulation layer consisting of SiON and parylene. In addition to organics PDs, inorganic PDs owning reliable electrical and mechanical properties are also used for attachable and flexible pulse and heart rates measuring.^[112]

Excess UV exposure is one of the most common causes of skin cancers. Recently, a real-time wearable UV monitor system based on a fiber-shaped p–n junction was reported, as shown in **Figure 6c**.^[37] Particular emphasis is placed on its ultrahigh photocurrent, which originates from the well-chosen materials and elaborately designed device layout. The conformal coating of p-typed transparent CuZnS film perfectly maintains the structure of TiO₂ nanotube arrays, and the p–n junction effectively introduces the self-powered property and suppresses the combination of photo-induced carriers. The high current reaches the electrical requirements of integrating with other commercial electronics to realize a monitoring system with a smartphone through Wi-Fi. Much work so far has focused on other wide band gap semiconductors such as ZnO for high photocurrents and spectral selectivity other than TiO₂.^[40] Optimized hierarchical structure of ZnO photodetectors is proposed to obtain ultrahigh milliamperere photocurrents and very low dark currents, featuring a high sensitivity and an excellent selectivity to very low ultraviolet light intensities.^[38] The specific focus of the device is its ultraporous nanoparticle networks with the particle size below twice the ZnO Debye length. Apart from UV photoelectrical detectors, other wearable sensors have proved promising and very useful. Here are two examples: Zou et al. applied an ink composed of multiredox active photoelectrochromic molecules for spectrally selective UV monitoring, and developed a paper-based bracelet meeting the needs of people with different skin phototypes.^[92] Qiu et al. utilized a photoelectrochemical cell on the basis of TiO₂ nanotubes and Prussian blue. The device can change from dark blue to transparent when exposed to UV light.^[113]

Photosensitive materials also go beyond light detection for other wearable applications, such as UV-assisted gas sensors^[114] and biomolecule detection.^[115] For wearable sensors, synthesis process and responsive properties must accommodate demands of superior sensitivity, flexibility, and stability for integration.

Particular attention should be paid to the two critical characteristics of wearable monitors: (1) self-powered property, as the current power sources are not miniature enough to meet the requirements of functional wearable electronic systems; (2) flexibility and robustness, which allow devices to maintain stable output signals under dynamic motions.^[37] Constructing p–n junctions or MSM structures are common ways to endow a PD with self-powered features. Strategies to make a PD device wearable include careful selection of substrates and elaborate design of the device layout, which can be referred to the discussion in Section 2.

The research about wearable monitoring based on photodetectors has continued to grow in numbers, indicating that there is an urgent need, but it is still a significant challenge to realize the integration of real-time wearable data acquisition, processing, and outputting. In addition, to transform rigid device into a flexible and portable one is the essence of the problem, which has already been thoroughly discussed in Section 2.

3.2. Wearable Imaging System

Flexible imaging, mainly in visible range but also including other wavelength ranges, is one of the most important

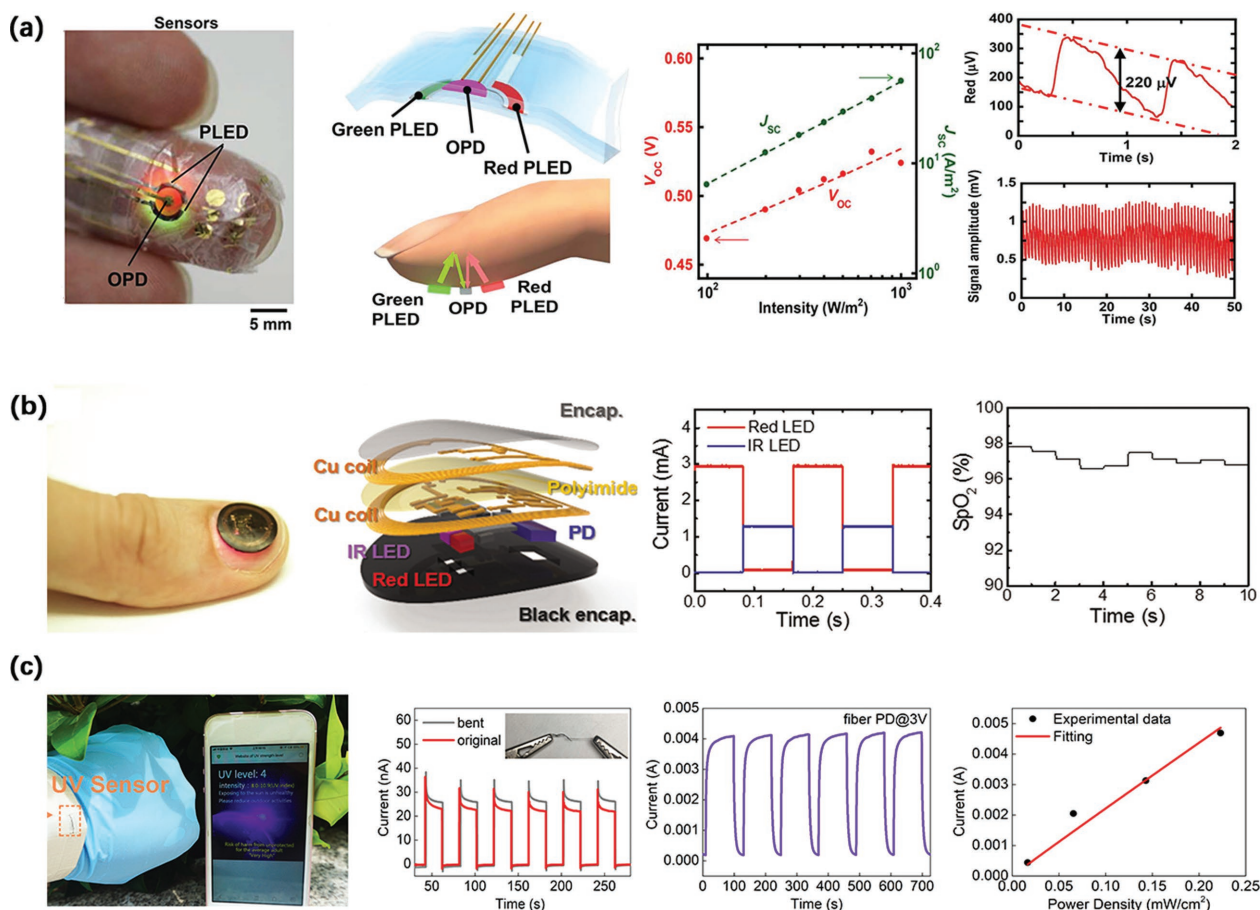


Figure 6. Different wearable health/environment monitors. a) The optoelectronic skins possessing health-monitoring sensors, displays, and ultraflexible polymer LEDs. The device structure and operation principle are shown in schematic diagram. The pulse wave measurements are based on the results of light intensity-dependent V_{oc} (red) and light intensity-dependent J_{sc} (green) of the photodetector. Reproduced with permission.^[111] Copyright 2016, American Association for the Advancement of Science. b) Situations when using wearable pulse oximetry based on NFC operating on different body locations. The schematic illustration presents the millimeter-scale, NFC-enabled pulse oximeter device. The time-resolved current implies the excellent photoresponse of the organic photodetector. The SpO_2 is calculated from the fingernail. Reproduced with permission.^[109] Copyright 2017, Wiley-VCH. c) A real-time wearable UV monitoring system. Its on-off switching testing results under 0 and 3 V bias demonstrate self-powered properties and ultrahigh photocurrent, respectively. The real-time monitoring is based on the fitted relationship of experimental and calculated photocurrents as a function of power density. Reproduced with permission.^[37] Copyright 2018, Wiley-VCH.

applications of wearable photodetector systems, which enables the detection of objects and environments. Wearable imaging system is getting increasingly important. On the one hand, the ability of quick response (QR) code recognition of wearable imagers can meet the needs of the information age. On the other hand, wearable imagers play an important role in special cases like evidence collection for police and wearable navigation for blind people. Commercial display screens like cathode ray tube, plasma display panel, and liquid crystal display are currently incompatible with wearable systems due to the bulk size as well as the complicated electronic control. Fortunately, wireless signal transmission techniques avoid the physical contact between photodetectors and external components, which have catalyzed mature wearable photodetector systems based on remote display equipment like mobile phones.^[37]

A great number of wearable applications like artificial eyes have been developed based on wearable imaging systems. Flexible or bendable photodetectors are usually based on

nanostructured materials with photoconductive effect or photovoltaic effect. The nanostructures not only enable a large specific surface area for a high light absorption, but also enhance the flexibility due to the micromechanically compatible structures. Moreover, unlike the flat photodetectors, flexible photodetectors offer excellent facilitation for curve design and show obvious advantages in omnidirectional photodetection.^[116]

Plenty of considerable work has been reported in flexible imaging field, with frequency ranging from UV to THz. In the research of Li et al., ZnO quantum dot (QD) decorated Zn_2SnO_4 nanowire array displayed excellent flexible UV imaging performance (Figure 7a).^[68] The nanowires could stand severe bending on PET substrate and the flexible 10×10 device array maintained nearly the same photocurrent at a bending angle of 150° . The type II ZnO/ Zn_2SnO_4 heterojunction delivered little hindered effect and the photoconductive property was fully guaranteed with a responsivity gain of 1.1×10^7 , along with a high specific detectivity of 9.0×10^{17} Jones and a fast

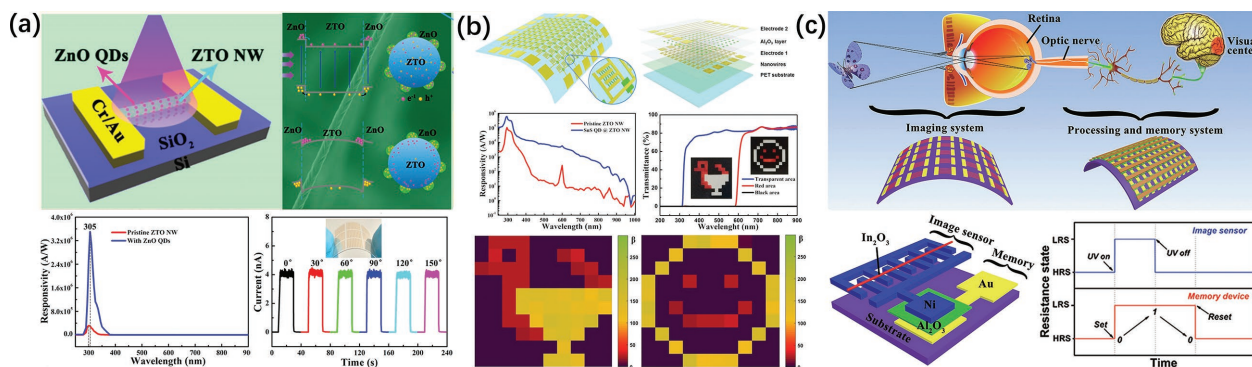


Figure 7. Wearable optical imaging systems. a) Wearable UV imager based on ZnO₂ decorated Zn₂SnO₄ nanowires. Reproduced with permission.^[68] Copyright 2017, American Chemical Society. b) SnS₂/Zn₂SnO₄ UV-to-NIR photoimagers recognizing pattern with red light and white light. Reproduced with permission.^[117] Copyright 2018, Wiley-VCH. c) In₂O₃ photodetectors integrated with memristors to form a visual memory system. Reproduced with permission.^[119] Copyright 2018, Wiley-VCH.

response time of 47 ms. Similarly, low-bandgap p-type SnS₂ QD was introduced to replace the ZnO QD for the decoration on Zn₂SnO₄ nanowires (Figure 7b),^[117] which extended the photosensitive range to near infrared (NIR) wavelength and generated broadband flexible photodetectors. The SnS₂ QD decorated photodetector was also reported to exhibit excellent sensitivity, flexibility, and stability. The UV-to-NIR broadband photosensitivity was further utilized in a flexible 10 × 10 photodetector array to form a 100-pixel imaging system. The flexible imaging system could recognize targets with both white light and red light, showing promising applications in flexible broadband imaging technology.

Wearable THz imaging technology has also gained increasing attention due to its superb performance in biochemical probing and damage detection. Recently, Suzuki et al. reported a wearable carbon-nanotube-related THz imager based on chemical Fermi-level-controlled methods,^[118] which was sufficiently miniaturized to be attached at fingertips and delivered decent imaging performances.

As stated above, great progress has been made in flexible imagers, both in device design and material innovation. However, the current flexible imagers still cannot meet the commercial needs. Current wearable imagers with merely 10 × 10 pixels cannot satisfy the recognition of the simplest QR codes, not to mention other applications. Typical QR codes contain 24 × 24 to 117 × 117 pixels, and wearable imagers should at least exceed this level to achieve basic applications. As for wearable objects and environment observation, we regard 72 dpi as the lowest acceptable resolution, which is commonly set as the lowest figure quality in commercial requirements with 600 × 480 pixels. Taking the fingertip area (≈2 cm²) as the proper device size, the first-generation commercial wearable imagers should at least reach a pixel density of about 10⁴ cm⁻² to satisfy the QR code recognition, and the next-generation products should reach 1.5 × 10⁵ cm⁻² for basic object photographing. The synthesis of photosensitive nanostructures is no longer a bottleneck, but the fabrication of highly integrated detector arrays along with the electric circuits is still a huge challenge, which is the key to the scanning process in fast imaging. Real-time imaging is necessary in some cases, but the perceived image immediately fades away as the incident signal stops, which is

inconvenient for signal processing. Combining visual memory mode with imaging enables a multifunctional integration to achieve electronic visions like human eyes. As showed in Figure 7c,^[119] a memristor was integrated with In₂O₃ nanowire photoconductor to store the perceived data, and a biomimetic visual memory system was developed on flexible substrates. The stored information could be erased under reverse bias, indicating a controllable vision memory function for further signal processing. Apart from the device structures, more attention should be paid to the electric insulation and signal interference among different pixels during the bending process. Advanced fabrication strategies with an improved geometry accuracy may contribute to the commercial application of flexible imagers for wearable photonic-electronic systems.

3.3. Wearable Optical Communication

Wearable optical communication can serve as the bridge of wearable systems, which connects various wearable devices for further data processing, human interaction, and result display. In a word, wearable optical communication catalyzes advanced applications on the basis of wearable devices. Wearable visible light communication has been achieved in previous reports. In the research of Fink and co-workers,^[100] fiber-shaped photodiodes were integrated with fiber-shaped LEDs to construct a communication system with the ability to remotely transmit data at a distance of 1 m from each other (see Figure 8b). Although visible light and UV communication have embraced great progress in the past decade, IR light (typical wavelengths: 850, 1310, and 1550 nm) still dominates the optical communication owing to their low signal attenuation. Massive related applications have been developed including remote control, distance measuring, data communication, and radar systems.^[120] Thereby, researchers in this field still pay dominant attention to IR photosensitivity to facilitate distance communication and cater to the wearable requirements of “Internet of Things.” As stated above, two-electrode device architectures like photodiodes and photoconductors play an important role in UV or visible light detection, which are well adapted to lateral structures. However, the photosensitive materials in IR photodetectors are

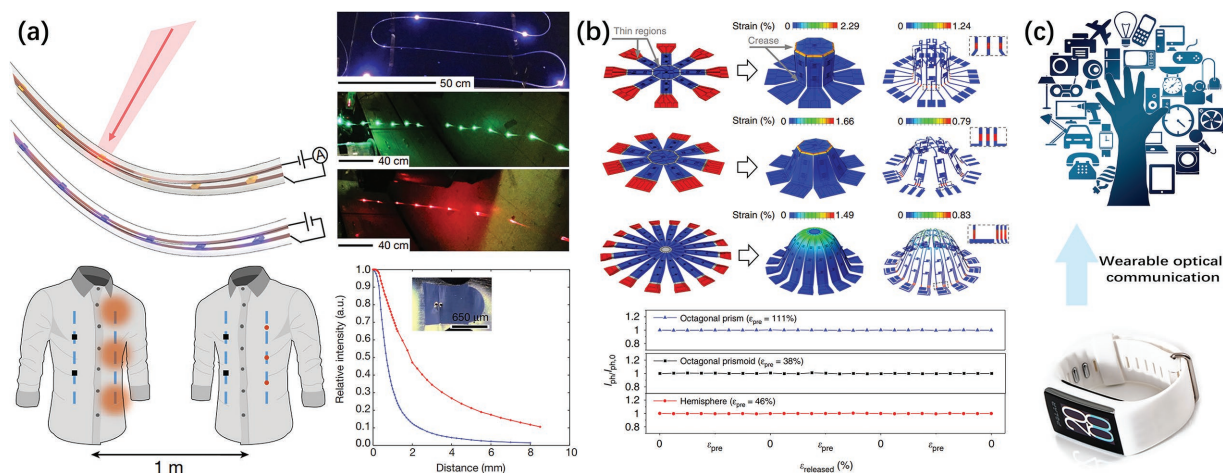


Figure 8. a) A wearable visible light communication system based on a fiber-like LED and photodetectors. Reproduced with permission.^[100] Copyright 2018, Springer Nature. b) The architecture optimization of flexible structures to fully reduce the bending strain. Reproduced with permission.^[87] Copyright 2018, Springer Nature. c) A schematic diagram of the potential applications of wearable optical communication equipment.

usually narrow-bandgap semiconductors with a high intrinsic conductivity, and they often deliver a high dark current in two-electrode devices. Thus, current IR photodetectors are generally based on three-electrode architectures, where additional vertical gate electrode is introduced to provide the gate bias voltage. The gate bias suppresses the lateral dark current due to the field-effect modulation and thus guarantees an adequate on/off ratio for signal resolution.

However, the introduction of gate electrode is incompatible with most flexible photodetectors in previous reports. Lateral designs no longer support three-electrode devices and vertical multilayer structures are necessary to realize the device architecture of IR photosensors. The multilayer structures not only complicate the device fabrication but also introduce potential problems to the photoelectric properties. Therefore, special attention should be paid to the film quality, vertical gate contact, interlayer insulation, and dielectric interlayer. Although highly miniaturized inflexible devices may fulfill the wearable requirements, flexibility is still a preferred property for wearable systems, especially for large-area occasions.

Rigid island device is a promising route to achieve flexibility with inflexible components, which has the potential to inherit the merits of inflexible structures simultaneously. This strategy has been reported in some flexible photodetector structures. In report of Zhang et al.,^[121] miniaturized silicon p-i-n photodiodes are spliced to form a flexible origami silicon hemispherical electronic eye systems, which delivered an excellent omnidirectional photoresponse. In the work of Rogers and co-workers,^[87] the flexible rigid island structures was systematically designed to diminish the warping strain (Figure 8b). The rigid island photodetector remains inflexible while delivering a general flexibility, avoiding the performance deterioration from deformation and bending stress. Similar flexible method could also be applied to various inflexible three-electrode photodetectors, including traditional photo-FETs and novel gate-related devices like hybrid photo-FETs^[53] and photovoltage field-effect transistors.^[54] Therefore, rigid island approach provides abundant flexibility for the device construction, which holds the potential for further usage in IR photodetectors.

Despite the existing problems, the development in flexible transistors still inspires a potential route apart from the rigid island approach to construct flexible optical communication systems.^[122] Unlike the inflexible phototransistors, the photo-sensitive channel suffers strong bending stress in flexible transistors, and traditional inorganic films become unsuitable due to the increased fracture risk and possible vertical short-circuit. Thus, flexible phototransistors require semiconducting materials with excellent bending resistance, such as conductive polymers^[123–126] and 2D materials.^[127] Although few IR-photo-sensitive phototransistors have been reported, the advances of novel materials^[128,129] may effectively contribute to considerable achievements in this field.

In brief summary, the wearable optical communication systems are still underdeveloped with potential routes and existing problems. The reliance on gate-based field-effect modulation sets difficulties to the flexible requirements, but rigid island approach and flexible transistor have been reported as potential solutions. With the help of the developing novel materials, wearable optical communication systems will eventually lead promising future applications as the development of “Internet of Things” as shown in Figure 8c.

4. Conclusions and Outlook

To date, transformation from rigid bulky photodetectors to flexible and even stretchable and deformable devices has been explored by researchers worldwide through careful selection of materials, elaborated design of device configuration and assembly technique.

For 2D wearable PDs, including polymer and paper substrate-based devices, geometrical forms such as wave, spring, hinges, etc., have been effectively applied to substrates or electrodes to endow the devices with flexibility. Those ideas masterly leverage the relation between micromechanics and materials to turn “hard” devices to “soft.” However, a lack of theoretical guidance, which may estimate the mechanical robustness of each layer in a device under deformation and

help build the ideal device layout for designated applications, becomes the current bottleneck. Only through a richer database of the physical properties of materials in any form and a more mature model between microstructural mechanic properties of device and materials, can 3D printing and artificial intelligence be easily cooperated in the process of developing versatile low-cost high-performance wearable PDs.

For 1D wearable PDs, a twisting structure is typically employed in the fiber-shaped devices, that is, a core metal wire electrode incorporated with photoactive materials and entwined by the other electrode. Carbon nanotube fibers/graphene fibers have been demonstrated as good candidates as the outer electrodes, however, their conductivities are less satisfying ($\approx 10^3 \text{ S cm}^{-1}$), especially in meters-long. Moreover, neither metal nor carbon-based electrodes are transparent. As transparent electrodes are always desired in optoelectronics, developing transparent flexible highly conducting materials are of great importance in wearable photodetectors. Attempts like Ag nanowires/PDMS composite materials have shown some potentials, with Ag network as the conducting phase and polymer as the flexible matrix, future studies may further improve the conductivities of the composite materials while keeping the transparency through nanostructure and microstructure design.

For a wearable PD, other than flexibility, more stringent requirements need to be met. To effectively operate a wearable PD and acquire useful information, the integration of PDs with other electronic devices in the system is indispensable. On the one hand, as the current wearable power supplies cannot fulfill the need of portable electronic systems, self-powered features are highly desired for PDs. Strategies such as integration with solar cells and construction of a p–n junction have been demonstrated successfully. On the other hand, the commercially miniature data processing and transmitting chips typically require a threshold current of $\approx 1 \text{ mA}$ at a low bias ($< 5 \text{ V}$), while the photocurrent of most PDs in literature are far behind. To get a higher photocurrent, MSM structure and photo-FETs are two main approaches to obtain a high photogain.

As for the applications, wearable photodetectors can be mainly categorized into three groups: monitoring, imaging, and optical communications.

In wearable monitoring systems, internal signals (human body) and external signals (ambient environment) are the two primary directions. For health monitoring, photodetectors are generally integrated with LEDs with constant power densities to detect biological signals of human body, such as heart rate, heart rate variability, and oxygenation, which are primary indicators of the health condition of a person. Conformal contact with the skin or nails is generally required for the wearable devices to obtain a stable and reliable measurement. And the environment light needs to be deducted for the accuracy of results. Thus, ultraflexibility and a high signal-to-noise ratio are particularly important for a wearable PD in this scenario. For monitoring the real-time UV intensity in ambient environment, as UV-B is believed to be a major etiologic agent in most skin cancers or diseases, a few reports have shown potential as real-time wearable UV alerts through visible color changes or intensity variations read on cell phones. However, for the read-through-color, it is not reliable especially one wants to know

the exact UV intensity. For the read-through-numbers on smart phones, the bulky and heavy data transmitter and processor are not compatible with the thin, flexible and wearable UV PDs. On the one hand, further improvement of the photocurrent of UV PDs and self-powered characteristics will free the devices from amplifiers and additional power sources. On the other hand, with the advance of flexible electronic circuits, a fully portable, flexible, and wearable UV monitoring system may be envisioned.

For wearable imaging systems, visible and NIR PDs are particularly important. A few examples have demonstrated the preliminary wearable imaging recognition system with the size of 10×10 pixels, or tried to explore new functions such as biomimetic visual memory system by integrating PDs with memristors. However, it is still hard to realize a more sophisticated and accurate imaging recognition system due to the possible signal interference and short circuits, especially under bending or stretching. On the other hand, color display, as an indispensable component in display, has witnessed some progress in wearable systems. That is direct color display, which directly switches the generated electrical signals as color changes and avoids additional electronic components mostly based on electrochromic effect.^[130] The electrochromic display is well adapted to low-cost flexible substrates like paper.^[92] Thus, it not only simplifies the wearable system fabrication but also becomes very suitable for special working environments like gaseous mine, where electric output is highly limited for safety. Despite of these developments, the wearable data display still suffers from monolithic image output, which requires advances in compatible integrated circuits and flexible display screens.^[131] Further advances on the flexible control circuits, device layouts, compatible and effective fabrication methods, and encapsulation techniques may aid to improve the geometry accuracy of wearable imaging and realize full color flexible display.

For wearable optical communication, IR PDs play a dominant role in versatile applications, including remote control, data communication, radar systems, and even to future wearable “Internet of Things.” As IR PDs typically have a three-terminal device structure, elaborated design is needed when introducing the gate electrode in the flexible electronic systems. In other words, for flexible IR PDs, both the preparation methods and integration techniques with other electronic devices become more complicated in contrast to the stacking or planar two-terminal PDs. Further exploration of organic IR sensitive materials and utilization of the interlaced textile structure in the fiber-shaped IR device may serve as a way to address those challenges.

Acknowledgements

S.C. and X.J.X. contributed equally to this work. The authors would like to thank Xiang Wu for his help with figures and language polish. The authors acknowledge the support from the National Key Research and Development Program of China (2017YFA0204600), the National Natural Science Foundation of China (Grant Nos. 51721002, 11674061, 51872050, and 11811530065), and Science and Technology Commission of Shanghai Municipality (18520710800, 18520744600, and 17520742400).

Conflict of Interest

The authors declare no conflict of interest.

Keywords

design strategy, flexible electronics, material exploration, wearable photodetectors

Received: December 17, 2018

Revised: January 20, 2019

Published online: February 20, 2019

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