Advances in Flexible Organic Photodetectors: Materials and Applications

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Abstract: Future electronics will need to be mechanically flexible and stretchable in order to enable the development of lightweight and conformal applications. In contrast, photodetectors, an integral component of electronic devices, remain rigid, which prevents their integration into everyday life applications. In recent years, significant efforts have been made to overcome the limitations of conventional rigid photodetectors, particularly their low mechanical deformability. One of the most promising routes toward facilitating the fabrication of flexible photodetectors is to replace conventional optoelectronic materials with nanomaterials or organic materials that are intrinsically flexible. Compared with other functional materials, organic polymers and molecules have attracted more attention for photodetection applications due to their excellent photodetection performance, cost-effective solution-fabrication capability, flexible design, and adaptable manufacturing processes. This article comprehensively discusses recent advances in flexible organic photodetectors in terms of optoelectronic, mechanical properties, and hybridization with other material classes. Furthermore, flexible organic photodetector applications in health-monitoring sensors, X-ray detection, and imager devices have been surveyed.

Keywords: photodetectors; organic materials; flexible; hybrid material systems; wearable electronics

1. Introduction

Photodetectors (PDs) are critical components for various electronic applications such as image sensors, medical monitoring, optical communication, and manufacturing process monitoring. On the other hand, over the past decade, many efforts have been dedicated to the fabrication of flexible devices such as sensors [1,2], memory devices [3], power modules, and wearable electronics [4,5]. Incorporating optoelectronic functions into flexible [6] and soft surfaces, particularly human skin, is at the forefront of multidisciplinary research. For example, lightweight and conformal image sensors with different wavelength sensitivity will open up numerous applications in wearable electronics, robotics, and the automotive industry. Due to the rapid rise of the Internet of Things (IoT), there is an increasing demand for smart and unobtrusive PDs that can be adhered to curved surfaces or applied to the body (either via the skin or implanted). These devices are able to revolutionize the healthcare industry by enabling accurate, continuous detection of physiological signals without interfering with human daily activities. In recent years, several promising approaches have been utilized for flexible photodetector fabrication, such as the application of silicon devices incorporated by flexible interconnects on a patterned polymer substrate [7] and conformal thin-film devices adhering to malleable biological organs [8], skin [9], and textiles [10] via manipulation of inorganic PDs. However, the above approaches do not completely satisfy the requirements of flexible PDs. For example, in the case of silicon devices, the thickness must be reduced to tens or hundreds of nanometers, which reduces light absorption significantly. In addition, due to their stiffness, inorganic flexible devices fail to conform to curved surfaces.

An alternative approach involves the application of solution-processable semiconductor materials, including multi-conjugated organic semiconductors, colloidal quantum
dots (QDs) [11–13], perovskites [14–16], and two-dimensional materials [17,18]. In contrast to bulk materials, nanoscale materials (such as thin films with a thickness of a few micrometers) exhibit a much lower flexural rigidity [19]. Moreover, these nanomaterials and organic materials exhibit excellent optoelectronic properties, such as high photo-absorption coefficients or narrow-bandwidth light emission. There has been a great deal of progress reported with solution-processed PDs based on QDs and perovskites. However, toxic materials are used in these PDs for wearable electronics, which remain a cause for concern [20]. Additionally, PDs based on two-dimensional materials are limited in spectral response, response time, air stability, and fabrication for large areas [21]. Unlike other solution-processed materials, organic semiconductors (molecules and polymers) exhibit low toxicity, broad spectral response, relatively fast response times, and the ability to be deposited on soft, flexible substrates, including plastics and elastomers, using blade coating, roll-to-roll printing, or inkjet printing for large areas [22].

Recent developments in organic heterojunction structures have enhanced the charge photogeneration efficiency of organic thin films, achieving detectivity up to $10^{14}$ Jones and a photovoltaic efficiency equal to inorganic PDs under sunlight [23]. In addition to this, the absorption wavelength of organic photodetectors (OPDs) can be easily tuned by modifying the chemical structure of the active material [24], allowing a tunable spectral range including ultraviolet–visible (UV–vis), near-infrared (NIR), and even X-ray wavelengths. Several review articles have focused on OPDs, mainly analyzing the performance, figure of merits, and working principles of OPDs on glass substrates [22,25], and not the ones on flexible substrates. This paper presents an overview of recent advances in flexible OPDs. We begin with a summary of the fundamental operating principles of OPDs, a description of the various types of OPDs developed to date, as well as the key figure of merits relevant to the flexible applications. Next, we discuss various novel materials for active layers, including organic materials and their hybrids with other material systems developed to enhance the light-sensing performance and mechanical properties of OPDs. Finally, we explore applications enabled by flexible OPDs, such as flexible health-monitoring sensors, flexible image sensors, and X-ray detectors.

2. Flexible OPD Background

2.1. Flexible OPD Working Principle

In organic semiconductors, low dielectric constants ($\varepsilon_r = 3–4$) and the localized nature of the excited states give rise to coulombically bound electron–hole pairs called excitons. Typical binding energies of excitons in organic materials have been reported to range between 0.3 and 0.5 eV [26]; therefore, large photocurrents require efficient separation of excitons.

An effective way to separate electron–hole pairs in organic materials is to form a heterojunction between electron-donating (donor) and electron-accepting (acceptor) materials, where the differing electron affinity and/or ionization potentials provide a driving force for charge separation and send them towards their corresponding electrode, which has been shown in Figure 1a.

Robust harvest of excitons without compromising light absorption is enabled by the use of a bulk heterojunction (BHJ) structure [27,28]; the structure of a BHJ is shown in Figure 1b. A BHJ consists of a bicontinuous interpenetrating network of the donor and acceptor materials with nanoscale phase segregation. The large interfacial area significantly reduces the distance required for the excitons to travel before reaching an interface where they can separate into free charges. A photocurrent is created when the charge is successfully extracted at the electrodes without undergoing recombination [29]. Organic semiconductors exhibit low charge mobilities compared to their inorganic counterparts, ranging from $10^{-6}$ to $10^{-3}$ cm$^2$ V$^{-1}$ s$^{-1}$ to around 10 cm$^2$ V$^{-1}$ s$^{-1}$ in amorphous or disordered films [30]. Electron–hole pairs can also form intermolecular charge-transfer states at the heterojunction, typically with energy below the optical bandgap [31]. These charge-transfer states are weakly optically coupled to the ground state and may lead to a broadening of the absorption edge, as reported for a wide range of
systems [32]. Most OPDs developed to date are based on a donor–acceptor heterojunction, which can efficiently separate photogenerated electrons and holes. While some devices employ a planar heterojunction (bilayer) structure [33], most have a BHJ structure. Many of the state-of-the-art OPDs and organic solar cells are based on very similar material systems. This is most likely due to these material systems’ high charge generation yields. However, it is important to point out that systems with optimal solar cell efficiencies may not necessarily be the best systems for photodetection. For instance, while materials in organic solar cells are designed to maximize the absorption range across the solar spectrum, PDs used for wearable applications have a very different set of requirements regarding the ideal absorption range. Therefore, there exists a large amount of room for improvements in terms of material and device designs for optimizing the performance of OPDs for applications other than solar energy.

![Figure 1. Organic photodetectors. (a) Band diagram of an organic photodiode depicting charge separation of excitons at donor–acceptor interface. (b) Use of bulk heterojunction (BHJ) structure with nanoscale domains enables optimal harvesting excitons. Reprinted with permission from [20] ©2021 Wiley-VCH GmbH.](image)

The three most common configurations of today’s OPDs are organic photodiodes, photoconductors, and phototransistors. The general configurations, working mechanisms, and performance characteristics of OPDs are discussed in this section.

2.2. Flexible OPD Configuration

2.2.1. Organic Photodiodes

In vertical two-terminal organic photodiodes (Figure 1a), there is an anode, a hole-transporting layer (HTL), an organic photoactive layer, an electron-transporting layer (ETL), and a cathode. Organic photoactive layers are typically composed of an organic BHJ, a mixture of organic electron-donating and electron-accepting materials. Four main processes are involved in photocurrent generation in the BHJ: exciton generation, exciton dissociation, charge transport, and charge collection. The use of the HTL and ETL in an OPD facilitates charge extraction at the BHJ/electrode interfaces, resulting in photocurrent generation. A photodiode’s photoresponse is based on collecting photogenerated charge carriers, allowing it to operate either self-powered or with a reverse bias [20].

2.2.2. Organic Photoconductors

Organic photoconductors (OPCs) are horizontal two-terminal devices with two symmetrical electrode contacts, an organic photoactive layer, and a substrate (Figure 1b). OPCs are typically operated with an external bias, and photodetection is defined as the detection of light absorption by measuring the change in electrical conductivity of the photoactive layer [20].

2.2.3. Organic Phototransistors

The organic phototransistors (OPTs) are three-terminal devices, having an organic photoactive channel layer, a dielectric layer, and three electrodes, including the gate, drain, and source electrodes (Figure 1). In OPTs, one type of charge carrier in the channel layer is
conducted from the drain electrode and the source electrode, whereas the gate electrode traps the other type of charge carrier. The carrier concentration in the organic layer increases with the photogenerated charge carriers, thus contributing to the change in drain-source current \( I_{DS} \). The advantage of OPTs is their high gain due to the photogate effect. As a result, a single photon incident on the OPT can induce a large number of charge carriers in the channel layer, resulting in a large photocurrent [20].

2.3. Figure of Merits

2.3.1. Optoelectronic Performance

EQE and Responsivity

External quantum efficiency (EQE) is a dimensionless parameter that is defined as the ratio of the number of charge carriers circling across the PDs to the number of incident photons. Responsivity \( R \) is defined as the ratio of photocurrent measured for the PDs to incident light power with a unit of AW\(^{-1}\). In this case, EQE and \( R \) represent the conversion efficiency of an optical device to an electric device. EQE and \( R \) are defined as [22]:

\[
R = \frac{I_{ph}}{P} \quad (1)
\]

\[
EQE = \frac{R hv}{q} \quad (2)
\]

where \( I_{ph} \) is the photocurrent measured for PDs, \( P \) is the incident light power, \( h \) is Planck’s constant, \( v \) is the light-wave frequency, and \( q \) is the elementary charge, respectively. It is worth mentioning that the EQE of photodiode-type OPDs is limited to 100%, whereas the EQE of OPTs and photo multiplication (PM)-type OPDs can exceed 100%. Enhanced photoresponses are possible with light-triggered charge injection in OPTs and PM-type OPDs.

Response Time

The rise time \( r \) of a PD is defined as the time interval for the photocurrent of the PD to rise from 10% to 90% of its maximum value upon the arrival of light, and the fall time \( f \) is defined as the time interval for the photocurrent of the PD to drop from 90% to 10% of its maximum value upon the removal of light.

Noise

OPD noise \( S_n \) is closely related to shot noise \( S_{shot} \), thermal noise \( S_{thermal} \), and 1/f noise \( S_{1/f} \), which can be calculated as follows [22]:
Specific Detectivity

Specific detectivity ($D^*$) is calculated by [22]:

$$D^* = \frac{\sqrt{A \cdot B \cdot NEP}}{S_n} = \frac{R \cdot \sqrt{A}}{S_n}$$  \hspace{1cm} (6)

where $A$ is the active area of the PD, $NEP$ is the noise equivalent power, and $R$ is a sensitivity measure indicating the power level of incident light that produces a signal-to-noise ratio of one in a 1 Hz output bandwidth, respectively. The precise determination of $R$ and $S_n$ is essential for calculating $D^*$. In OPDs, the $S_{shot}$ is typically regarded as the primary noise source and is used to calculate $D^*$. However, the measured $S_n$ can be significantly greater than the $S_{shot}$. This can result in an overestimated $D^*$ due to thermal noise, particularly in OPDs with photoactive layers of narrow-bandgap materials. Due to the frequency dependence of the photoresponse and the $S_n$, the $D^*$ decays in the $S_{1/f}$-limited low-frequency range and the response speed-limited high-frequency range. To calculate the $D^*$, both the frequency-dependent $R$ and $S_n$ should be considered.

2.3.2. Stability and Durability

Although OPDs fabricated on thin plastic foil exhibit excellent mechanical compliance and durability, it remains difficult to achieve high performance, mechanical durability, and stability in air and water simultaneously. This is due to the reduced thickness of the plastic substrate and encapsulation layer, which results in an insufficient barrier to moisture and oxygen for wearable device applications [34]. While rigid OPDs can easily achieve long device lifetimes by encapsulation with epoxy and glass [35], new strategies for effectively protecting the active layer without introducing mechanical stiffness are required for wearable devices.

Jinno et al. [10] created the first washable and stretchable organic photodiodes by encapsulating the free-standing ultrathin organic photodiode between two pre-stretched 500-micrometer-thick acrylic elastomers. The organic photodiode achieved high solar cell efficiency (7.9%) and stretch ability (52%), even when completely submerged in water, using this method. Furthermore, it was discovered that after compression for 20 cycles with 100 min of water exposure, 80 percent of the initial photovoltaic efficiency was maintained.

3. Flexible OPDs Advances

Using flexible organic semiconductors, flexible OPDs can be designed for flexible applications on curved, conformable, and foldable substrates. Flexible OPD fabrication also necessitates using flexible substrates, flexible transparent conductive electrodes (TCEs), and in more advanced applications (stretchable OPDs), an elastic active layer. Cyclic stressing tests, which include bending, stretching, and compressing treatments, are used to assess the mechanical stability of flexible OPDs. The performance characteristics of recent flexible OPDs with different active materials, flexible substrates, and device performances are summarized in Table 1. In the following section, we will go over the significance and the impact of advances in each of the factors mentioned above.

| Ref | Active Materials | Substrate | Device Type | Flexibility | Responsivity | Detectivity | Response Time |
|-----|------------------|-----------|-------------|-------------|--------------|-------------|---------------|
| [36] | Tin-based perovskite/PEDOT:PSS | PI | PT | $R_b = 8$ mm 300 times | $\sim 8.7 \times 10^5$ A/W | - | - |
| [37] | P3HT:PC70BM | PET | PM | 1000 bending at $R = 8.54$ mm | 388.4 A/W | - | 210 ms |
| [38] | PCDTB:PCBM | PI | Photodiode | - | 0.21 A/W | $3 \times 10^{12}$ Jones | - |
| [39] | PBDB | PET | PC | 2000 bending at theta = 120 degree | 0.9 A/W | 1.8 $\times 10^{10}$ Jones | 900 ms |
| [40] | TQ1-PNDI2T10 | PET | Photodiode | - | 0.17 A/W | 10^{11} Jones | - |
| [41] | D18:Y6 and D18/Y6 | PET | Photodiode | Bending with $R = 11$ mm | 0.35 A/W | 9.39 $\times 10^{11}$ Jones | 0.013 ms |
Table 1. Cont.

| Ref | Active Materials | Substrate | Device Type | Flexibility | Responsivity | Detectivity | Response Time |
|-----|------------------|-----------|-------------|-------------|--------------|-------------|---------------|
| [42] | P3HT:o-IDTBR     | PET       | Photodiode  | 1000 bending at R = 1 cm | -            | -           | -             |
| [43] | PBDTTT-OFT:IEICO-4F | PI        | Photodiode  | 1000 bending at R = 25 micrometer | 11.9 A/W     | 1.7 × 10^13 Jones | 0.037 s       |
| [44] | DPP-DT:PC61 BM/cross-linked DPP-DT | PI        | PT          | Bending with R = 6 mm | 150 A/W      | -           | 0.15 s        |
| [45] | D18              | PET       | PT          | 2000 bending at R = 10 mm | 10 A/W       | 1.7 × 10^13 Jones | few milliseconds |
| [46] | IrO_2/PTPBT-ET   | PI        | PT          | 1000 bending with R = 5 mm | 20 A/W       | 1.4 × 10^12 Jones | 5 ms          |
| [47] | SQ-H:PC61BM MAPbD MWs covered by an assembled P3HT | PET       | Photodiode  | -            | 0.2 A/W      | 9 × 10^10 Jones | -             |
| [48] | In_2O_3/PTPBT-ET | PET       | PT          | 1000 bending with theta = 90 degree | 400 A/W      | -           | 12 ms         |
| [49] | SEBS: P3HT: ICBA | Silicone elastomer PDMS | Photodiode  | -            | 0.006 A/W    | 2.3 × 10^9 Jones | 0.06 ms       |
| [50] | P3HT:PCBM/3DG    | PET       | Photodiode  | 100 bending with R = 5 mm | 5.8 × 10^7 A/W | 3 × 10^15 Jones | 24 ms         |
| [51] | PTB7-Th:COTIC-4F | PEN       | Photodiode  | 100 bending with R = 7.5 mm | ≈0.42/A/W | 1.25 × 10^13 Jones | 0.021 ms   |
| [52] | PEDOT:PSS/SeSnSe_2 | ITO/PET | Photodiode  | 3000 bending | 0.73 A/W     | -           | 16 ms         |
| [53] | ZnO/P3HT         | PET       | Photodiode  | -            | 156 μA/W     | 0.74 × 10^9 Jones | 40 ms       |
| [54] | RNA              | PET       | Photodiode  | Bending with theta = 150 degree | 14.99 μA/W  | 5.23 × 10^7 Jones | 5 s          |

3.1. Flexible Substrate

The first step toward flexible OPDs is to replace rigid glass substrates with thin and flexible ones. Flexible substrates can be made by direct fabrication on plastic foil [55]. Plastic foils such as PET and poly (ethylene naphthalate) (PEN) are frequently used as substrates for flexible OPDs because of their high optical transparency and good barrier properties against gas and moisture. PET and PEN, on the other hand, deform at temperatures above 120–150 °C, making it difficult to deposit indium tin oxide (ITO) electrodes on them without damaging the substrate and this makes the replacement of the ITO electrodes a challenge.

In addition, a polymeric layer spin-coated on a rigid substrate can be peeled off after device fabrication [56]. Using materials with high mechanical strength [57–59] or fabricating electronics in a neutral strain position [60] makes it possible to attain very small bending radii (r = ~0.1 mm).

A third very promising route is the use of ultrathin substrates [61–64]. These substrates offer excellent elastic stretchability and mechanical durability, which are essential for smart skin [65], biological tissue sensing [66], and even solar cells [64]. Polyurethane (PU) [67], poly (styrene–butadiene–styrene) (SBS) [68], poly (styrene-ethylene–butylene–styrene) SEBS [69], Ecoflex [70], polydimethylsiloxane (PDMS) [71] and Parylene C [72] with thicknesses of less than hundreds of micrometers have typically been chosen because of their mechanical elasticity and high transparency.

3.2. Transparent Conductive Electrodes

For flexible OPDs to function properly, we need TCEs with low sheet resistance, high optical transmittance, and robust flexibility. ITO and fluorine-doped tin oxide (FTO) are currently used as transparent electrodes because of their high optical transparency (about 90% in the visible region) and electrical conductivity (10–25 Ω sq⁻¹). However, ITO is brittle and rigid. A number of materials have been developed to overcome this problem, including carbon nanotubes (CNTs) [73], silver nanowires (Ag NWs) [74], and semiconducting polymers such as poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS (also known as PH1000)) [75].

CNTs have relatively low electrical conductivity (~100 Ω sq⁻¹) [76,77]. Recently, CNT TCEs have been optimized by studying the density of CNTs for better optical transmittance and conductivity [23], as shown in Figure 2a. The ideal CNT electrode has a visible transmittance of 90% and a sheet resistance of 98 cm⁻². Because of the deeper work function, the CNT-based OPDs have a lower J_d of 9.62 × 10⁻¹³ Acm⁻² compared with...
ITO or PEDOT:PSS and a higher $D^*$ of $2.07 \times 10^{14}$ Jones. The $D^*$ after the OPDs were stressed with a cyclic flex test for 500 bending cycles at a bending strain of 0.8 percent only reduced to 80% of its initial value, whereas the $D^*$ of ITO-based OPDs reduced to 20% of its initial value.

Solution-processed networks of Ag NWs have a sheet resistance and transmittance comparable to those of ITO ($10-25 \ \Omega \ \text{sq}^{-1}$ at 80% transmittance), together with a relatively high work function of 4.5 eV [79]. Furthermore, they are promising for high-throughput roll-to-roll manufacturing of low-cost transparent conducting films. However, Ag NWs with an electrical conductivity of $\approx 20 \ \Omega \ \text{sq}^{-1}$ are required to be processed by nanoimprint lithography [80,81], and their high surface roughness limits their application [82]. Moreover, the Ag NWs that make up the film can easily penetrate the thin active layer (~100 nm) atop the Ag NW electrode, resulting in a short-circuited device. To address this challenge, Yu et al. transferred smooth Ag NW film from a glass substrate to a transparent cross-linked polymer overcoat; Ref. [83] and Gaynor et al. [84] embedded Ag NWs into the conducting polymer PEDOT:PSS by lamination.

PEDOT:PSS is advantageous because of its ease of processing on large areas. It can be deposited as a homogenous layer by spin- or slot-die-coating or by printing techniques [85]. However, the micro-patterning of this film is not trivial due to the physio-chemical properties of this material and the process’s influence on the TCE’s morphological and electrical properties. In particular, standard photoresists damage PEDOT:PSS films during deposition, development, and chemical removal. In addition, the acidity of PEDOT:PSS also affects the cross-linking or causes uncontrolled de-composition of widely used photoresists. Many different techniques for the photolithographic patterning of PEDOT:PSS have been explored [86]. The most common method is the so-called lift-off process, where a PEDOT:PSS layer is deposited on top of a previously structured photoresist, which is subsequently mechanically removed to give the desired pattern [85]. Another popular approach requires using a sacrificial layer (e.g., parylene or silver) to protect the PEDOT:PSS from the developer. In 2018, Rene Fischer et al. developed a simpler and more effective protocol to structure PEDOT:PSS that does not require any sacrificial layer or specially tailored material [87]. They presented the advantage of intermixing single-wall carbon nanotube (SWCNT) with PEDOT:PSS to form a dispersion for easy deposition. As a result of the low CNT charge, their SWCNT/FE-T (PEDOT:PSS formulation) films present a lower specific resistance ($0.26 \times 10^{-4} \ \text{m}$) than pristine FE-T films ($5.6 \times 10^{-5} \ \text{m}$) while maintaining high optical transparency (91% at 550 nm). The fabricated OPDs eliminate the need for an electron-blocking layer with improved mechanical properties and higher chemical stability than pure FE-T electrodes.

Furthermore, a polyethylenimine (PEI) interlayer was deposited on the PEDOT:PSS layer to modify the cathode’s work function. The use of aliphatic amine-rich polymers, such as PEI, as surface modifiers for electronic contacts was pioneered by Zhou et al. [88].
This helps reduce device dark currents and, therefore, improves the power detection limit and specific detection. In another publication, Matteo Cesarini et al. adopted non-toxic solvents for printing PEI different from the most common 2-methoxyethanol [78], as shown in Figure 2b. Their work studies the PEI interlayer deposition parameter’s heavy impact on the reproducibility of device performance in inkjet printing. Their multi-solvent approach drastically improves yield (from less than 20% to over 90%). In addition, dark currents are reduced to as low as 57 nA cm$^{-2}$. In addition, these devices exhibited a faster response time of up to 30 s, due to a better interface between the PEI interlayer and the photoactive layer.

3.3. Active Layers

3.3.1. Organic Materials

Organic semiconductors can be divided into polymers and small molecules. Although both classes of materials achieve similar levels of performance in thin-film transistors (i.e., charge carrier mobility) and solar cells (i.e., efficiency), each material class has a unique set of advantages and disadvantages [89]. For example, polymers can be easier to coat from solution than small molecules [89]. Small molecules, however, are monodisperse and, therefore, are less subject to batch-to-batch variability [89]. The advantage of polymers is that they offer superior mechanical resilience over small molecules since they are van der Waals solids that are not entangled and, therefore, have enhanced strength and toughness. Therefore, the amount of energy absorbed by conjugated polymers during either elastic or plastic deformation, as well as their resilience, tensile strength, or toughness, will likely exceed that of small-molecule semiconductors.

Small Molecules

In small molecules, crystallinity has been a key factor in enhancing (opto) electronic performance. In BHJs, crystallinity is linked to enhanced charge photogeneration efficiencies [90]. They typically participate in BHJs as donors, which include diketopyrrolopyrrole (T-DPP-T)-containing molecules, squarine derivatives, oligothiophenes, acenes, phthalocyanines, various push–pull molecules [90], and molecules with a complicated donor–acceptor (D–A) architecture; see Ref. [91] for a recent review.

The higher mobility of small molecules in single crystals makes them better suited for OPTs. On the other hand, OPT development is mainly limited by the following factors: (i) Si and SiO$_2$, as commonly used gate electrodes and dielectric layers, respectively, are rigid and fragile. (ii) Channel layers always pose the dilemma of balancing the trade-off between charge transport and light absorption. As a solution to these issues, H. Huang et al. [45] fabricated a phototransistor using high-mobility, unipolar polymer poly[(2,6-(4,8-bis(5-(2-ethylhexyl)-3-fluoro)thiophen-2-yl)-benzo[1,2-b:4,5-b’]dithiophene)-alt-5,5’-(5,8-bis(4-2-butyloctyl)thiophen-2-yl)dithieno[3′,2′,3,4;2″,3″:5,6]benzo[1,2-c][1,2,5]thiadiazole]) (D18). The device architecture was simplified with superior device stability because D18 worked as both the light absorption layer and the charge-transport channel, as shown in Figure 3a. The authors also replaced the SiO$_2$ dielectric layers with tantalum pentoxide (Ta$_2$O$_5$) and polyacrylonitrile (PAN) hybrid layers, which possess high flexibility and capacitance. The calculated photocurrent and mobility values as a function of the bending cycle indicated excellent stability, as shown in Figure 3b.

It is better to construct organic devices with stacked structures for industrial production. The sequential deposition (SD) of donor and acceptor films demonstrates little dependence on the ratio of donor and acceptor materials, solvents, additives, and vertical phase segregation structures, which can simplify device fabrication and achieve comparable power conversion efficiency (PCE) to blend-casting (BC) devices [92]. In 2021, Yanan Wei et al. fabricated a series of self-powered OPDs including D18/2,2′-(2Z,2’Z)-(12,13-bis(2-ethylhexyl)-3,9-diundecyl-12,13-dihydro-[1,2,5]thiadiazolo[3,4-e]thieno[2″,3″:4′,5′]thieno[2′,3′:4,5]pyrrolo[3,2-g]thieno[2′,3′:4,5]thieno[3,2-b]indole-2,10-diyl)bis(methanylylidene))bis(5,6-difluoro-3-oxo-2,3-dihydro-1H-indene-2,1-diylidene)dimalononitrile(Y6) with high $D^*$ and photocurrent stability with the SD method [41]. Due to the vertical phase segre-
In recent years, the development of conjugated polymers, including BHJs for PDs and flexible phototransistors fabricated with various methods, has been explored rigorously. In 2020, Canek Fuentes-Hernandez et al. demonstrated a flexible OPD based on a P3HT:ICBA active layer on a polyestersulfone (PES) substrate that achieved a level of performance in the visible spectrum that is statistically equivalent to that of state-of-the-art low-noise Si PDs (Hamamatsu S1133, for example; NEP~200 fW and $D^* \approx 2 \times 10^{12} \text{ cm-Hz}^{1/2} \text{ W}^{-1}$ at $B = 1.5 \text{ Hz}$) [93], except for the response time, which is 35 microseconds and remains compatible with video rates. In addition to operating in a large area, the device is capable of producing photodetectors that have complex shapes, such as a ring-shaped flex-OPD, a shape that is better suited to maximizing SNR and minimizing power consumption for PPG sensors. Despite this, the flexible device had lower values of $R$ as a result of series resistance, absorption, and reflection losses (~30%) caused by the semi-transparent Ag/MoOx electrode. In addition, it yielded $D^*$ values that were at least two times smaller for flexible OPDs compared to those on glass.

In BHJs, a large number of additional acceptors easily form scattering centers for hole transport and percolating pathways for electron transport. This results in reduced hole mobility and increased dark current [94]. On the other hand, bilayer heterostructure (BHS) phototransistors have been verified as promising device architectures [94–97], where the BHS is combined with a BHJ film as the light absorption layer to generate and separate excitons, and a high-mobility semiconductor layer for carrier transport. However, fabricating BHS OPTs in the solution process is challenging because the underlying layer is prone to be destroyed or even washed off during the continuous solution-deposition process. In 2021, Q. Li et al. [44] fabricated NIR polymeric phototransistors with solution-processable BHS, and cross-linked polymeric semiconductors (CLPS) were used as the bottom conducting channel. Additionally, BHJs comprised of low-bandgap polymer donors and fullerene acceptors were used as the NIR photoactive layer; the schematic of the structure is shown in Figure 4a [44]. The active layer, including diketopyrrolopyrrole (DPP)-based polymer (PDPP-DTT), was used to make the phototransistors highly responsive to 808 nm light. They also used indanone-condensed thiadiazolo[3,4-g]quinoxaline-based polymer (PBTQCN-TT) as the NIR light absorber, which the phototransistors had a good response to, namely 808 nm, 1064 nm, and 1550 nm NIR light. Both devices exhibited improved performance compared with their controlled devices. This strategy enabled the continuous
solution processing of multilayer devices and eliminated the limitations of low mobility and high off-current on the performance of phototransistors based on low-bandgap polymers (especially bandgap <1 eV). The flexible NIR phototransistor array was fabricated on a polyimide (PI) substrate. The devices remained relatively stable at different bending radii (as low as 1 mm). Its dark current remained below 0.2 nA even after folding, and its maximum output power (P) was above $10^4$ at a bending radius of 1 mm.

OPTs with conformable channels have utilized p-channel materials due to the lack of n-channel organic materials with air stability and good performance [98–100]. The development of conformable OPTs based on n-channel organic material is extremely important for the fabrication of complementary electronics and optoelectronic circuits, which provide various advantages such as high operational stability, easily controlled photo-switching voltages, and high photosensitivity and R [101–103]. In 2018, M. Liu et al. [104] presented an ultrathin conformable OPT array on the polyvinyl alcohol (PVA) supporting layer, in which the air-stable n-type N,N'-Diditracyl-3,4,9,10-perylenedicarboximide (PTCDI-C13H27) thin film serves as the active layer and the thickness of the entire OPT is only ~830 nm, as shown in Figure 4b. From the view of molecular design, PTCDI-C13H27 is a parylene derivative favorable for good stability in n-type organic transistors [10]. The large-area OPT array shows excellent electrical properties in the dark state with mobility as high as 0.58 cm$^2$ V$^{-1}$ s$^{-1}$, an extremely high on/off ratio of over $10^9$, and high stability in the air atmosphere. When the OPT surfaces are bent parallel and vertical to channel length, it is found that the performance changes. The on-state current, off-state current, ON/OFF ratio, and mobility are similar, although the conductive channel region presents a different bending deformation. With the decreased bending radius on different curved surfaces, both the dark current and light current of the conformal device present a weak decrease. However, the synchronous decrease in the dark and light current keeps the photosensitivity unchanged and remains at $>10^4$ when the device adheres to objects with different curved radii, showing good photosensitivity consistency under strain.

![Figure 4](https://example.com/figure4.png)

**Figure 4.** (a) Schematics of the flexible OPT reprinted with permission from [44] ©2020 Elsevier. and (b) the conformal device reprinted with permission from [104] ©2018 Nature Portfolio.

The majority of organic materials absorb visible light and do not have an effective wide-bandgap donor–acceptor material combination. This is why there are no reports of visible-blind UV PM-type OPDs prepared with fully organic materials. To address this challenge, Dechao Guo et.al introduced a novel design for high-performance photomultiplication-type (PM-type) visible-blind flexible UV OPDs based on wide-bandgap organic semiconductor materials (TPAC: C60 blend) [105]. A wide-bandgap absorber was selected as a donor, and a small amount of C60 was used as an acceptor. A device with a ratio of 50:1 displayed a narrowband response, an ultrahigh external quantum efficiency of $1.08 \times 10^6 \text{%}$, and a remarkable specific detectivity of $1.28 \times 10^{14} \text{ Jones}$ at 335 nm under a bias of 14 volts. The flexible device on a PET substrate integrated with a flexible OLED to work as a wearable UV monitor. A key advantage of this configuration is that the flexible visible-blind UV PM-type OPD is capable of driving the OLED without the need for an additional current amplifier since it is capable of generating sufficient photogenerated current under the applied bias.
voltage and UV illumination. Additionally, the device exhibits UV wavelength-selective response characteristics, so other wavelengths of light will not activate the UV monitor. Furthermore, the wearable UV monitor has excellent weak-light detection capabilities, which are mainly due to the PM characteristics of visible-blind UV OPDs.

Dyes

Organic dyes are gaining popularity as alternative photoactive materials due to their tunable optical properties via molecular design and crystal engineering and their inherent high absorption coefficient. Furthermore, their thin active layers are often solution-processable and, thus, flexible and lightweight, and suitable for low-cost wearable applications. J. Kim et al. [47] reported a highly efficient short-wave infrared (SWIR) OPD based on the J-type dicyanovinyl-functionalized squaraine dye (SQ-H) mediated by intermolecular charge transfer; a diagram schematic and the flexible fabricated device are shown in Figure 5a,b [47]. The optimized BHJ OPDs have a maximum EQE value of 12.3% and a full width at half maximum (FWHM) of 85 nm (815 cm\(^{-1}\)) at 1050 nm under 0 V due to a favorable combination of absorption and charge-transport properties. The flexible devices at both 690 and 1050 nm had a slightly lower performance of 9% EQE.

Elastomers

Since the surface of a soft object experiences strains in normal conditions, stretchable PDs might provide an ideal platform for implementing wearable health-monitoring devices such as photoplethysmography (PPG) sensors, [106,107] or sensors mounted onto living organisms for smart agriculture [108]. Furthermore, they can be applied to artificial skin [9,108] and soft robotics [109–111], electronic eyes on curvilinear surfaces [112], and asset tracking, gesture, and motion recognition sensors [112]. Elastomeric semiconductors can be used to fabricate stretchable optoelectronics. An elastomeric semiconductor layer has a low Young’s modulus (E) and a large strain at break. Currently, there is no stretchable OPD used in photovoltaics that maintains its performance beyond 50% strain, with one notable exception that sustains 100% strain but has a high E of 5.5 GPa. Recently, a ternary blend of polydimethylsiloxane (PDMS), a donor polymer, and a non-fullerene acceptor yielded a high E value of 990 MPa, maintaining a normalized power conversion efficiency of 86.7% up to a strain of 20% [112]. These devices were tested for photovoltaic applications, so their characteristics were not reported at low irradiance or in the dark. In 2021, Youngrak Park reported combining the elastomer SEBS, the donor polymer poly(3-hexylthiophene-2,5-diyl) (P3HT), and the acceptor Indene-C60 bisadduct (ICBA) to yield a skin-like e-BHJ with a low E and a high strain at break, similar to SEBS [49]. The fabrication process is shown in Figure 5c. Tensile testing on e-BHJ films yields a tensile modulus of 2.4 MPa, with a strain at break of 189%, within the range of values found in human tissue. Further, stretchable OPDs displayed dark current density values smaller than 600 pA cm\(^{-2}\) under reverse bias, measured root mean square electronic noise values in the tens of femtoamperes range, and measured NEP values at 653 nm between 13 and 24 pW at strain values up to 60%, resulting in a \(D^*\) value in the 10\(^{10}\) Jones range. The reason was that adding SEBS at 50 weight percent (wt. %) reduces the absorption of light in a pristine BHJ by at least 50%.

3.3.2. Hybrid Active Layers

Polymers and Perovskites

Various types of perovskite materials have recently been used for PDs due to their tunable band gaps, long carrier lifetimes, and high light absorption coefficients [113–116]. Moreover, many studies indicated that flexible perovskite devices could be easily realized on plastic substrates [117–120] for next-generation optoelectronic devices. However, several challenges must be addressed in this direction, such as developing lead-free perovskite material PDs and finding a better strategy for using metal halide perovskites (MHPs).

So far, lead-free perovskite PDs have limited R, and their performance is far below the requirement of sensitive detection. Chun-Ki Liu reported a PD based on a lead-
free perovskite/organic semiconductor with the structure of [(NH2)2CH] SnI3/poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate)(FA5SnI3/PEDOT:PSS) in order to enhance lead-free PD properties with vertical heterostructure as shown in Figure 6a [36]. The device exhibited an excellent photoresponse over a wide range, 300 to 1000 nm, due to a photogate effect. Furthermore, the device’s maximum R and gain are $2.6 \times 10^6$ A/W and $4.7 \times 10^6$, respectively. Due to the absence of back-scattered light from the substrate, their fabricated device on flexible PI had a slightly lower photoresponse. The device bendability test was conducted 300 times by curving the flexible device to an 8 mm radius on the surface of a bottle. The R at low intensity and the duration of the responses were nearly unchanged, as shown in Figure 6b.

![Figure 5](image-url)  
Figure 5. (a) Energy diagram of the SQ-H:PC61BM blend-based OPDs, (b) photographs of a flexible SQ-H-based OPD, reprinted with permission from [47] ©2021 Wiley, and (c) schematics of the fabrication process of the elastomeric OPD, reprinted with permission from [49] ©2021 American Association for the Advancement of Science.

![Figure 6](image-url)  
Figure 6. (a) Schematic diagram of the FASnI3/PEDOT:PSS PD, (b) R vs. light intensity for the flexible device before and after the bending test, reprinted with permission from [36] ©2020 ACS Publications, (c) schematic illustrating the energy-level diagram of the perovskite insulator organic (PIO) heterojunction and photo-induced charge generation mechanism, and (d) the I−V curves of the original flexible PIO device and the device subjected to 1 and 100 bending cycles at 90° and then followed by 1 cycle at 180° under the illumination of white light, reprinted with permission from [48] ©2021 Wiley.
Polycrystalline-built three-dimensional (3D) MHPs suffer from unfavorable charge recombination at grain boundaries and low carrier mobility [121–123]. The large numbers of grain boundaries in polycrystalline 3D MHPs act as charge-trapping defects, deteriorating their optoelectronic properties [124]. Moreover, because they are vulnerable to moisture, the grain boundaries are the key factors in the instability of polycrystalline 3D MHPs in the air [124]. As compared to polycrystalline 3D MHPs, 1D single-crystalline MHPs have attracted great interest due to their low light/dark current on/off ratio (>2 × 10^5), a high R (>400 AW^−1), defect density, fewer grain boundaries, efficient 1D carrier-transport path, anisotropic 1D geometry, and high flexibility. Despite their promising characteristics, PDs based on pure MHPs have a number of inherent limitations. The most serious problems are the high binding energy and limited carrier mobility [125]. Researchers have considered hybridizing MHPs with other materials to solve these problems to form heterostructures. Such hybridization can improve exciton dissociation, carrier separation, and transportation. Using this approach, various high-performance hybrid MHP PDs have been developed, ranging from perovskite–CNTs [126,127] to perovskite–graphene, perovskite–transition metal dichalcogenides (TMDs) [128], perovskite–Mxene, perovskite–organic heterostructures [120], etc. In 2021, J. Zhang et al. [48] proposed a novel design of radial heterostructure hybrid perovskite microwires (MWs) based on perovskite and an organic semiconductor by a self-assembly process. The charge-transport schematic is shown in Figure 6c. The structure of the radial heterostructure was core MAPbI\(_3\) MWs fully or partially covered by an assembled P3HT layer with an ultrathin insulating dielectric interface between them. Owing to the efficient exciton dissociation and charge transfer at the huge radial heterojunction interface, the photoresponsivity of the PD based on MAPbI\(_3\)/P3HT hybrid MWs was improved dramatically over that of pristine MAPbI\(_3\) MWs. The photoresponse of the flexible device was stable under bending conditions, demonstrating the excellent bendability of our perovskite MW-based flexible PDs, as shown in Figure 6d.

**Polymers and Inorganic Materials**

Inorganic semiconductors exhibit higher mobility, a higher optical absorption coefficient, and the ability to be widely tuned by doping, stoichiometry control, size tuning, strain engineering, etc. [52]. However, they require elevated temperatures during device fabrication, require high-cost manufacturing, have limited solution processability, and have less compatibility with lightweight and flexible substrates.

In order to circumvent both organic and inorganic semiconductor limitations, hybrid organic–inorganic junctions are highly desirable. Diverse research groups have used inorganic–organic hybridization for various applications, including UV PDs, piezophototronic effects, and IR PDs on flexible substrates.

In 2018, Xiaoyu Zhang fabricated flexible PM-type narrowband UV PDs using a blend of poly[{9,9-dioctylfluorenyl-2,7-diyl}-alt-co-(bithiophene)] (F8T2) and ZnO nanoparticles (NPs) as the active layer; the structure is shown in Figure 7a [129]. The flexible PDs exhibited EQE spectra with two narrow response peaks under a reverse bias, and a very low dark current density of 1.3 × 10^{-5} mA cm\(^{-2}\), even under a strong reverse bias of −15 V. The EQE value produced by the PDs was 2170% under 360 nm illumination and 220% under 510 nm illumination. Moreover, the PDs were bent under both tensile and compressive stresses. The peak D* decreased only by about 5% in total, from 8.8 × 10^{11} to 8.5 × 10^{11} jones, after tensile bending and to 8.3 × 10^{11} jones after compressive bending, as shown in Figure 7b. In both tensile and compressive bending, the response speed remained almost constant.

Fibrous devices are gaining popularity thanks to their light weight, flexibility, wearability, and even stretchability, making them ideal for integrating multifunctional systems such as smart textiles, energy-autonomous electronics, and soft robotics. Fiber-shaped PDs have received little attention despite their low photoresponsivity, slow response speed, and their potential applications. Because of its excellent photoelectric properties, direct bandgap (3.37 eV), and large excitation binding energy (60 meV), 1D ZnO has become the preferred
structure for fabricating ultraviolet PDs. Furthermore, the piezoelectric property of 1D ZnO provides a novel strategy for modulating photo-induced charge carrier interfacial transfer. Xinxin Du demonstrated a series of p–n junctions for efficient carrier separation and rapid carrier transport in 2022, using flexible tungsten wire and conductive alginate fiber as the inner and outer electrodes, respectively, with the well-oriented architecture of coaxial multilayers [53]. They reported a self-powered UV PD made from a vertical ZnO/P3HT heterostructure. The structure is shown in Figure 7c. The device was fixed on a flexible PET film to study the bend-induced piezo-phototronic effect. The PD’s output current under lighting and bending (0.19 µA) is significantly higher than that under only lighting (0.14 µA) and only bending (1.35 nA). The bend-induced piezo-phototronic effect due to ZnO’s noncentral symmetric wurtzite structure significantly boosts the photoresponse by increasing the photoresponse by 81.2 percent for the bent device with 1.96 percent strain compared to the device without strain as shown in Figure 7d.

Figure 7. (a) Schematic illustration of the flexible narrowband UV PD structure and molecular structure of F8T2, (b) EQE spectra of the PD before and after tensile and compressive bending at −15 V bias, reprinted with permission from [129] ©2018 ACS Publications, (c) Light-induced carrier transport in fiber-shaped PD based on vertical heterostructure, and (d) output current signals of the photodetector under bending, lighting, and lighting + bending, reprinted with permission from [53] ©2021 Elsevier.

In recent years, increasing demand for photodetection in the SWIR and NIR spectra for applications such as optical communication, environmental gas sensing, medical diagnostics, night vision, light detection and ranging (LIDAR) [130], and long-term wearable monitoring has been utilized. However, many IR-responsive materials are rigid and unstable in air, which limits their use. Conjugated semiconducting polymers have aroused much interest for their application in low-cost photodetection [130]. While there is continuous progress toward synthesizing low-bandgap polymers, it is still difficult to have SWIR photodetectors entirely based on semiconducting polymers exhibiting R beyond λ = 1.2 µm [131–134]. One possible strategy to reach that point is to apply photon upconversion to convert low-energy photons into high-energy ones. Various upconversion systems in the form of solution-processed NPs have been synthesized, [135–137] mostly based on the doping of trivalent lanthanide cations in low-phonon-energy hosts, which perform upconversion through a two- or multi-photon mechanism [138]. Recently, Zhang et al. fabricated novel erbium silicate nanosheets to upconvert λ = 1.5 µm photons to visible ones [139]. In 2019, Hengyang Xiang et al. demonstrated high-performance flexible photodetectors sensitive to λ = 1.5 µm photons based on the formation of a solution-processed organic (a donor–acceptor BHJ blend of diketopyrrolopyrrole (DPP)-based copolymer and [6,6]-phenyl-C71-butyric acid methyl ester (PC71BM))/inorganic
hybrid composed of conjugated polymer/small-molecule bulk heterojunctions (BHJs, host) together with Er3-doped upconversion nanoparticles (UCNPs, guest); its structure is shown in Figure 8a [140]. Under the illumination of $\lambda = 1.5 \mu m$ SWIR photons, optimized hybrid BHJ/UCNP photodetectors exhibit a clear photoresponsivity of 0.73 and 0.44 mA/W for devices built on glass substrates and flexible PET substrates, respectively. The rise time of the device was 80 $\mu s$ under the illumination of $\lambda \approx 1.5 \mu m$ photons, which is faster by more than one order of magnitude than previous photodetector studies applying Er3+-doped upconversion systems [139] and by two to four orders than most other non-avalanche semiconductor photodetectors at SWIR wavelengths [141–143]. Mechanical bending tests showed a bending angle greater than 120°, and a photocurrent with minimal photocurrent change (~5%) after 500 bending cycles, as shown in Figure 8b. In another publication, Xinyu Zhao et al. reported, for the first time, a flexible SWIR photodetector with excellent photosensitivity to eye-safe light at 1532 nm using a SWIR-responsive rare earth doped-nanoparticle semiconducting polymer (RENP–SCP)(RE–SCP) composite; its schematic shown in Figure 8c [144]. The RE–SCP composite maximizes photon-to-electron conversion efficiency and minimizes optical scattering losses through (1) spectral matching of the properties of SWIR-responsive nanophotonic converters with that of an SCP and (2) sophisticated control of nanophotonic converter size and interactions within the SCP matrix. The SWIR upconversion characteristics of the NaYF4 core–shell NPs (N-NPs) that dictate the success of SWIR detection of these RE–SCPs were controlled by tuning the dopant chemistries to match the SCP’s peak absorption properties. Subsequently, an optimal photoactive layer of SWIR-responsive N-NPs dispersed within a diketopyrrolopyrrole-based SCP was fabricated. The decrease in photocurrent density $J$ was observed for the photodetectors with different bending curvatures because of the reduction in incident illumination power density due to an increase in the illumination area for the bent photodetector. The calculated R and EQE, with minimal differences for different curvatures, are shown in Figure 8d.

![Figure 8](image-url)
n-type metal oxide/polymer semiconductor heterostructures: indium oxide (In$_2$O$_3$) and n-type polymerizing small molecular acceptors (PSMA), a polymer semiconductor of poly5,5′-bis[3,5-bis(thienyl)phenyl]-2,2′-bithiophene-3 ethylesterthiophene] (PTPBT-ET) [46]. By virtue of rational energy band alignment, the hybrid phototransistor combined the advantages of both In$_2$O$_3$ transistors and the high NIR-response of PTPBT-ET polymers, exhibiting a high saturation mobility of 7.1 cm$^2$ V$^{-1}$ s$^{-1}$ and a large current on/off ratio of >10$^7$. The phototransistor performed well in NIR light sensing, with an R of 200 A W$^{-1}$, a $D^*$ of 1.2 × 10$^{13}$ Jones, and a fast photoresponse time of 5/120 ms. Moreover, even after 160 days in the air, their device maintained excellent electrical stability with no performance degradation. The device bending tests showed that with a bending radius of 5 mm, device characteristics remained unchanged for up to 1000 bending/releasing cycles.

Polymer and Two-Dimensional-Based Materials

Two-dimensional materials and their three-dimensional derivatives show extraordinary optoelectronic and photodetection properties [145–147]. On the other hand, due to their stiffness and instability under air exposure, they need to hybridize with other soft semiconductor material systems to be applicable in flexible applications.

Among these two-dimensional semiconductors are layered MoS$_2$, WS$_2$, MoSe$_2$, InSe, black phosphorous, SnS$_2$, SnSe$_2$, and GaSe, which are being investigated extensively for optoelectronics as they exhibit high absorption coefficients and tunable properties. Among these layered materials, SnSe$_2$ is a potential candidate for optoelectronic applications owing to its high intrinsic absorption coefficient [148] and being an earth-abundant and environmentally benign compound. SnSe$_2$ exhibits a hexagonal CdI$_2$ crystal structure with n-type conductivity and a direct bandgap of around 1.2 eV. In 2022, B. Reddy et al. [52] fabricated a bulk heterojunction (BHJ) of SnSe$_2$–PEDOT:PSS by simple sonication-assisted mechanical mixing for broadband photodetection. The flexibility tests showed that the measured R values exhibit minimal degradation in performance due to bending. The response time was estimated to be 16 ms, 221 ms, and 274 ms for UV, Vis, and NIR spectra, respectively, and it was higher for the UV light due to the band transition in ITO, which has a bandgap of 3.38 eV. Furthermore, the device showed detectivity values of 6.51 × 10$^{10}$ Jones, 6.23 × 10$^{10}$ Jones, and 6.08 × 10$^{10}$ Jones in the UV, visible, and NIR regions, respectively. Moreover, the device exhibited good stability after 4 weeks and retained ~90% of its initial performance.

The most appropriate three-dimensional topological insulators (3D TIs), Bi$_2$Te$_3$, with a suitable narrow bandgap (0.145 eV), have drawn considerable attention [149–153]. Although some progress has been made, photodetection based on Bi$_2$Te$_3$ is usually hindered by the large dark current and dilatory response time due to the lack of inherent differences. Moreover, the above Bi$_2$Te$_3$-based photodetectors are in the IR region, specifically in the mid-IR range. In 2019, Ming Yang et al. prepared a photodetector with a 3D TI Bi$_2$Te$_3$/organic heterostructure using n-type Bi$_2$Te$_3$ and p-type organic thin films, and the structure is shown in Figure 9a [154]. This photodetector possesses a wide-waveband response from VL to mid-IR (450–3500 nm) with an ultrahigh R up to 14.89 AW$^{-1}$ and an ultrafast response time of 1.89 ms at room temperature. More importantly, the R reaches 1.55 AW$^{-1}$ at 3500 nm, several orders of magnitude higher than previous photodetectors based on 3D TIs. At the same time, the response time is one order of magnitude faster than that of the previous Bi$_2$Te$_3$-based photoelectric devices. Devices constructed on a flexible mica substrate exhibited a photocurrent diagram at 650 nm in the self-powered mode when a bending force was applied at two sides of the device. As the force increases, the magnitude of the photocurrent decreases as the distance between the two sides d decreases. When the device is at its maximum bending (4 mm), the photocurrent decreases by only 9.8%, indicating the flexibility and stability of the device, as shown in Figure 9b.
P3HT-based BHJ materials; the structure is shown in Figure 9c [50]. Flexible 3DG film/organic hybrid detectors are compatible with flexible substrates, and the dark current of the flexible device is slightly lower than that of the rigid device due to the higher resistivity of PET/ITO. The response current of the flexible device can maintain more than 90% of that of the rigid device under 520 nm illumination. Furthermore, after 100 bending cycles, the flexible device’s photocurrent still maintains more than 80% of the initial response current, as shown in Figure 9d.

4. Applications

This section discusses the application of OPDs to wearable electronic devices, such as health-monitoring devices, image sensors, and X-ray detectors. In Figure 10, a pictorial description of the flexible OPDs is shown.
P3HT-based BHJ materials; the structure is shown in Figure 9c [50]. Flexible 3DG film/or-ganic hybrid detectors are compatible with flexible substrates, and the dark current of the flexible device is slightly lower than that of the rigid device due to the higher resistivity of PET/ITO. The response current of the flexible device can maintain more than 90% of that of the rigid device under 520 nm illumination. Furthermore, after 100 bending cycles, the flexible device’s photocurrent still maintains more than 80% of the initial response current, as shown in Figure 9d.

Figure 9. (a) The structure diagram of Bi$_2$Te$_3$/pentacene heterostructure self-powered PD, (b) the distance between two ends and photocurrent diagram of PDs under irradiation of 650 nm, reprinted with permission from [154] ©2019 ACS Publications, (c) schematic of the flexible 3DG film/OPD, and (d) normalized photocurrent for flexible 3DG film/OPD as a function of cycle number of re-peated bending to a radius of 5 mm reprinted with permission from [50] ©2022 ACS Publications.

4. Applications

This section discusses the application of OPDs to wearable electronic devices, such as health-monitoring devices, image sensors, and X-ray detectors. In Figure 10, a pictorial description of the flexible OPDs is shown.

Figure 10. Schematic illustration of representative applications of organic material flexible photodetectors.

4.1. Health Monitoring

Heart rate monitors, pulse oximetry sensors, and transcutaneous oxygen (oxygen level of tissues beneath the skin [155]) monitors all use PDs. OPDs have been used in noninvasive and low-cost PPG sensor technology, for example. The PPG sensor detects changes in blood flow volume in a microvascular bed of tissue, such as in the fingers. The PPG signals are measured by using OPDs to track changes in light intensity. PPG sensors can be designed in two modes: reflection (aligning the light source and OPD side by side) or transmission (placing the light source and OPD on two sides of a finger). For the optical implementation of heart rate sensing, a highly detective photodetector is required because human tissue (bones, blood, fat, water, and melanin) scatters and absorbs a large portion of light. In 2018, G. Ryu et al. [156] reported a printed flexible optoelectronic sensor composed of red organic LEDs (OLEDs) and organic PDs (OPDs) for fabricating flexible PPG devices and the subsequent detection of various biological signals. Three OPD structures were investigated. PPG signals were successfully detected from the fabricated flexible PPG sensor and driving circuit. A human study was conducted to evaluate the flexible PPG sensor’s performance in a real-world application, where subject drowsiness was estimated from the PPG signals. Heart rate variability (HRV) was extracted from the PPG signals using machine learning algorithms to classify drowsiness. The flexible PPG sensor produced 79.2% accuracy and 72.1% area under the receiver (AUC) to predict drowsiness, which are meaningful results compared to conventional PPG sensors (83.3% accuracy and 69% AUC).

The all-organic devices provide great flexibility and conformity to the skin; however, they currently only operate in the visible spectral range (e.g., green and red). This imposes a limitation on the penetration depth of light into the skin tissue and limits the ability to detect signals. In addition to penetration depth, light attenuation also needs to be taken into account. To engineer an OPD with high EQE in the red-NIR spectrum, the photosensitive organic semiconductors (OSCs) must have a low optical bandgap, ideally ≈1.20 eV or lower [157]. In 2017, H. Xu et al. [158] demonstrated epidermal and flexible PPG sensors that could operate in the NIR regime with high sensitivity and low power consumption. The configuration of the PPG sensor is shown in Figure 11a. The PPG sensor hybridizes an organic phototransistor (OPT) with an inorganic light-emitting diode. This LED offers much higher power conversion efficiency (14.3%) compared to NIR OLEDs, and it can be easily bonded to flexible substrates through transfer printing.

In 2022, João Simões et al. [51] demonstrated a new solution-processed flexible OPD architecture based on a BHJ of poly[4,8-bis(5-(2-ethylhexyl)thiophen-2-yl)benzo[1,2-b:4,5-b’]dithiophene-2,6-diyl-alt-(4-(2-ethylhexyl)-3-fluorothieno[3,4-b]thiophene)-2-carboxylate-
2-6-diyl)] (PTB7-Th) and 2,2′-(((2Z,2′Z)-(((4,4-bis(2-ethylhexyl)-4H-cyclopenta[2,1-b:3,4-b′]
dithiophene-2,6-diyl)bis(4-(heptan-3-yloxy)thiophene-5,2-diyl))bis(methanylidene))bis(5,6-
difluoro-3-oxo-2,3-dihydro-1H-indene-2,1-diylidene))dimalononitrile (COTIC-4F). The nar-
row optical bandgap of the NFA COTIC-4F allowed good photoresponse beyond 800
nm, outperforming the majority of fullerene-based OPDs. Blood oxygen saturation was
monitored successfully using a custom PPG sensor integrating the fabricated OPD, which
achieved a low error of 2% compared to a commercial PPG sensor. The configuration is
shown in Figure 11b. The resultant PPG waveforms, between 40 and 50s for 660 and 940
nm, showed from ≈21 to ≈22 mV with a ≈1 mV

peak to peak, and from ≈16.5 to 18 mV

with a ≈1.5 mV

peak to peak, respectively. They mentioned that since the HbO₂ absorbs
less red light than NIR light, in cases where SpO₂ is high, blood volume changes have less
influence on the detected red signal.

In 2018, Sungjun Park et al. reported skin-conformal NIR OPDs that simultaneously
achieve a high cutoff frequency over 1 kHz at −3 dB and mechanical conformability
sufficient for health-monitoring applications [107]. The configuration is shown in Figure 11c.
The ultrathin (total thickness < 3 µm) organic photodetector showed exceptional operational
durability under conditions of extreme mechanical deformation (10^3 cycles of operation
at a bending radius of 3 µm), with stable charge carrier transport under repetitive motion.
The precise and fast near-IR switching behavior with low dark current could be controlled
by modulation of resistance and capacitance of the active layer. In addition, the long-term
near-IR responsive operation is maintained even when the device is attached to a curved
and/or folded part of the human skin by locating devices at a neutral plane within 3 µm of
device’s total thickness.

Noninvasive 2D oxygenation mapping capability has the potential to transform real-
time and post-surgery management as well as the monitoring of wounds, tissues, and
organs [159–163]. An in vivo spatial oxygenation mapping device can aid in assessing
tissue damage and injury susceptibility. One such application scenario, where a flexible
optoelectronic sensor array is used to map the 2D oxygenation of a skin graft, is illustrated
in Figure 11d. In 2018, Y. Khan et al. [164] reported a reflectance oximeter array (ROA): a
flexible and printed electronic system realized by printing and integrating arrays of organic
optoelectronics with conventional silicon integrated circuits for blood and tissue oximetry.
The ROA is composed of four red and four NIR printed organic light-emitting diodes
(OLEDs) and eight organic photodiodes (OPDs). The fabricated sensor on flexible plastic
substrates is comfortable to wear and increases the SNR by establishing a high-fidelity
sensor–skin interface. With a 0.7 × 0.7 cm² active area for both OLEDs and OPDs and
0.5 cm spacing between the OLEDs and OPDs, the dimensions of the complete ROA became
4.3 cm in both length and width. The cutoff frequency was measured at over 5 kHz for
OPDs. Since the operation frequency of the pulse oximeters is generally less than 1 kHz,
this bandwidth is sufficient for oximetry. Additionally, they explored the placement of
the sensor on the body. The forehead provided the strongest pulsatile signal, and their
reflectance oximeter monitored the oxygen saturation of a volunteer on the forehead with a
mean error of 1.1%.

Most organic-based pulse oximeters focus more on the feasibility and/or on such form
factor advantages. Little attention, on the other hand, has been paid to the potential of
organic-based pulse oximeters in terms of ultra-efficient operation to meet the challenging
power demand of wearables applications. In 2018, Hyeonwoo Lee et al. showed a reflective
patch-type OPO sensor with ultralow power consumption based on flexible OLEDs and
OPDs [165]. The configuration is shown in Figure 11e. Through an optical simulation, an
ideal OPD wraparound layout has been proposed where an OPD resembling the numeral 8
wraps around small circular OLEDs for each red and green emission. With this approach,
the proposed monolithically integrated OPO sensors exhibited successful operation at
electrical powers as low as a few tens of microwatts on top of various body parts. Moreover,
the conformal, index-matched contact between the substrate and the skin makes it possible
to access and use a significant amount of light that would otherwise be confined within the
substrate. It was also shown that this index matching is important for the suppression of direct coupling of light between an OLED and an OPD. The results presented here illustrate that organic devices not only have form factor advantages but also hold great promise as enablers for all-day wearable health-monitoring systems.

Figure 11. (a) Photograph of a finger covered with the epidermal hPPG sensor (scale bar, 5 mm). (b) Illustration of the testing setup used for PPG waveform acquisition. (c) skin conformal near-IR photoplethysmogram sensor. (c) (i, left image) Photograph of fingerprint-conformal near-IR PDs; inset indicates position of skin-conformal near-IR PD on finger, (ii, right top image) enlarged image of (i), and (iii, right bottom image) microscope image of top silver electrode and skin surface, indicating high skin conformability, Reprinted with permission from [158] ©2017 Wiley. (d) Schematic of an application scenario of the ROA: 2D oxygenation mapping of a skin graft on the forearm. After surgery, the ROA is placed on the skin graft to map oxygenation of the reconstructed skin and ROA sensor configuration, reprinted with permission from [164] ©2018 American Association for the Advancement of Science. (e) Schematic of the proposed OPO sensor with enlarged cross-sectional view to depict device arrangement and light-receiving process through the skin medium, reprinted with permission from [163] ©2018 National Academy of Sciences.

Opportunities in the seamless, continuous assessment of health/wellness and advanced functions in wound monitoring/care and human–machine control systems motivate research in this field. Babilas et al. [166] measured transcutaneous \( O_2 \) pressure (tcpO\(_2\)) under variations in the microcirculatory system using a luminescent \( O_2 \) sensor [166]. However, the tcpO\(_2\) value was recorded using a luminescence lifetime imaging method, which
requires expensive devices apart from the sensing film on the skin \cite{166}. Additionally, Liu et al. \cite{166} demonstrated a structurally integrated luminescent O$_2$ sensor composed of OLEDs, an OPD, and sensing film \cite{166}. However, the proof of concept was limited to the detection of dissolved [O$_2$] \cite{166}. In 2018, Chang-Jin Lim et al. presented a new concept for a wearable tcpO$_2$ sensor utilized in the early detection of diseases based on O$_2$ partial pressure by combining the technologies of luminescent gas-sensing, wearable, and biocompatible devices. The sensor was a bandage-like O$_2$ sensor based on the luminescent sensing mechanism and consisted of three components on a plane: a sensing film, an OLED, and an OPD as a detector. As the sensor facilitates the time-resolved, real-time monitoring of patients, the number of medical staff members required to care for a patient can be drastically reduced, facilitating the immediate care and rescue of patients. In addition, there is little need for patients to be hospitalized for this real-time monitoring, which will result in significantly reduced medical costs. The I$_{10}$/$I_{30}$ (2.19) of the sensor was sufficient for measuring tcpO$_2$ variations in tissues of the lower arm and a thumb after pressure-induced occlusion, and due to its noninvasive and flexible features, it could be used in any part of the body, even when a person is exercising or working.

4.2. Image Sensors

One advantage of OPDs in comparison with conventional inorganic devices for imaging is the ease of achieving full-color detection. For conventional photodetectors, which have a broad absorption range covering the entire visible spectrum, color selectivity is typically achieved using red, green, and blue filters. Filters, however, reduce the amount of light, limiting detectability \cite{167}.

Increased pixel densities are an important feature of sensing arrays, and to date, minimizing pixel footprints has been challenging. The ratio of the sensor’s active area to the total pixel area is called the pixel fill factor \cite{112}. For image sensors, fill factors of less than unity result in poorer sensitivity and increased cost, requiring the use of micro-lenses to direct light to the sensor \cite{168}. Thus, monolithic integration is preferred since it would allow for pixel fill factors of unity—pushing achievable pixel densities to their limit. The major challenge in manufacturing these types of monolithic architectures is maintaining the process compatibility of all layers during the fabrication process. In 2018, Peter Zalar et al. demonstrated a passively addressed photodiode matrix that takes advantage of the monolithic integration of photodiodes and diodes \cite{169}. Their structure permits high pixel densities shown at up to 262 pixels per inch (ppi), with pixel areas of just $2.5 \times 10^{-3}$ mm$^2$ and pixel pitches of 50 µm. Imaging capability is shown in Figure 12a. This is possible because a pixel fill factor of unity can be maintained by virtue of this structure. The demonstrated structure allows for real-time imaging with a satisfactory resolution for a variety of applications such as real-time spatial PPG measurements on the ball of a human foot and heart rate measurements matching simultaneous electrocardiography (ECG) measurements.

Due to limited registration accuracy, variation in the printing process stability, and highly sensitive drying effects \cite{170,171}, the imager device integration and packing density, in combination with consistent device performance, have remained challenging. In 2018, Ralph Eckstein et al. overcame these challenges by exploiting a self-alignment process induced to fabricate a fully digitally printed image sensor based on organic photoactive materials with high performance, reproducibility, and lab-scale fabrication yields of 100% \cite{172}. The passive matrix image detector is composed of 256 micro-pixels with individual active areas of ~250 µm $\times$ 300 µm for a total footprint of 64 mm$^2$; the imaging capability is shown in Figure 12b. Characterization of the single OPD pixels demonstrated state-of-the-art performances. The robust fabrication method was based on printed dewetting patterns, which improves registration accuracy and reduces film-forming defects.
Figure 12. (a) Optical microscope image of a 16 × 16 (active area, 2.40 mm$^2$; pixel pitch, 50 µm, left panel) array on a glass substrate, Reprinted with permission from [168] ©2018 Wiley, (b) photograph, microscope image, and topographical image (white light interferometric microscope image) of the completed pixel array, reprinted with permission from [171] ©2018 Wiley, (c) schematic illustration of the image sensor irradiated by 1342 nm light and the corresponding output images of the bird by the flexible SWIR image sensor, reprinted with permission from [39] ©2020 Elsevier, and (d) schematic diagram of multi-pixel image sensor imaging and the deer pattern composed of 1024-pixel output by Ti$_3$C$_2$Tx-RAN PD image sensor reprinted with permission from [54] ©2022 Wiley.

Lei Lv used their single-polymer PD to create IR imaging sensors in another study [39]. The polymer PDs were incorporated into a 256-pixel (16 × 16) flexible image sensor with the number of pixels easily increased to a million-scale array using an industrial micromachining technique. One pixel has a sensor area of 900 × 900 µm and a periodicity of 700 µm. With irradiation of 1342 nm (26.3 mW/cm$^2$) at 8 V, the corresponding patterns were placed on the array’s surface to display the expected images. Imaging capability is shown in Figure 12c.

In some works, researchers tried to use hybrid heterostructures to enhance the performance of the imagers. As an example, Chuqiao Hu reported a high-pixel imaging system with 1024 pixels (32 × 32) selected from the Ti$_3$C$_2$Tx-RAN PD array and integrated into the flexible image sensor; the imaging capability is shown in Figure 12d [54]. The incident
light (1064 nm, 159 mW cm\(^{-2}\)) integrated into the flexible image was irradiated from the backside of the substrate during the image-sensing measurement, and the deer-shaped mask was placed on one side of the device array. The pixels were all responsive to the 1064 nm light, except for the area covered by the mask. The image is clearer, and the deer pattern is more vivid and exquisite, thanks to the thousands of pixels. The high imaging capability of Ti\(_3\)C\(_2\)Tx-RAN PDs suggests that they have a lot of potential in the field of high-pixel image sensing.

4.3. X-ray Detectors

Medical imaging, nondestructive testing in industrial products, and public security inspection are just a few examples of where X-ray imaging is used. Indirect conversion X-ray detectors that combine scintillator and visible OPDs are used to detect X-rays using OPDs. The X-ray excitation activates the scintillators, which then emit visible light. OPDs have been designed to match the luminescence of scintillators in terms of spectral response. However, the absorption of light by the organic semiconductor forms excitons, which need to be dissociated, resulting in significant losses and limiting detector sensitivity as opposed to a direct conversion process. In 2018, H.M. Thirimanne et al. introduced a broadband, direct, X-ray detector concept based on a thin-film hybrid semiconductor diode consisting of an organic BHJ–bismuth oxide (Bi\(_2\)O\(_3\)) NP composite [173]. The flexible detector offers a high sensitivity of 280 \(\mu\)C mGy\(^{-1}\) cm\(^{-3}\). More importantly, these sensitivities are achieved at \(-10\) V. The improved X-ray sensitivity is a result of impact ionization and an enhanced path length due to Mie scattering and the efficient separation and transport of these by the BHJ–NP architecture, resulting in high charge collection efficiencies (>60%). Because of its rigidity, the traditional X-ray detector is limited in its application in curved sensors. Two-dimensional spatial detection and self-powered operation are enabled by a flexible X-ray detector made by combining the printed OPD with a plastic scintillator. A plastic foil-based curved digital X-ray image sensor developed for approaching cone-beam computed tomography X-ray imaging is shown in Figure 13a [38]. The curved X-ray image sensor has a large surface area of 6.0–8.0 cm and 480 × 640 pixels. The X-ray image sensor successfully captured a full 3D volume image of a piece of bone. Over the entire area of the digital detector, the images show good and homogeneous contrast. Because the lateral spread of optical photons is lower in the thinner scintillator layer than in the thicker scintillator, the spatial resolution is clearly higher. For the 700 and 400 \(\mu\)m thick scintillators, the modulation transfer function (MTF) was 31 and 37 percent at 1 lp/mm, respectively. These figures match the MTF predicted for an X-ray detector with a 40 \(\mu\)m thin-film barrier between the image sensor and the scintillator.

Fabricated flexible OPDs have been reported to show fluorescence under the high-intensity X-ray radiation utilized in medical applications [174]. This radiation-induced
fluorescence produces a strong non-linearity in the R of the OPDs due to their high sensitivity to optical photons and prevents the full realization of tissue-equivalent printed polymer dosimeters. M.J. Large et al., in 2021, provided the first study of a non-fullerene acceptor (o-IDTBR) in combination with a donor polymer fabricated onto polyimide (Kapton) as a mechanically flexible substrate that is known to exhibit high transmission to X-rays and negligible radiation-induced fluorescence; the structure is shown in Figure 13b [42]. To enhance the sensitivity of the organic detectors, they utilized a plastic scintillator with a spectral window where the organic semiconductors have a much higher absorption coefficient. Full characterization of the flexible OPD device response to both visible light and X-rays indicated acceptable tissue equivalence, good sensitivity to X-rays, and also demonstrated a large improvement in stability under high radiation doses when utilizing the non-fullerene-based o-IDTBR in place of the more widely studied PCBM. This sample preservation method resulted in negligible variations in the photoresponse of the detectors over a time period of 17 days. Detectors employing photoactive layer P3HT:o-IDTBR displayed an extremely linear response with increasing radiation dose with indirect X-ray detection sensitivities 114.2 ± 0.7 pC/cGy. In another publication, in 2021, Jessie A. Posar et al. reported the first-ever demonstration of a printed X-ray detector that is fully tissue-equivalent, has a rapid temporal response, and exhibits good sensitivity at zero-bias operation [175]. This performance is achieved by coupling an RP400 plastic scintillator with a photodiode composed of donor polymer P3HT and NFA (o-IDTBR) to create an indirect X-ray detector. The X-ray-detecting material system was printed into flexible devices with pixel sizes of 60 µm that exhibit exceptional stability against degradation due to aging, repeated bending, and high-irradiation doses. PET films are known to exhibit scintillation with a deep-blue photon emission, potentially overlapping with that of the RP400 plastic scintillator employed in the devices [176]; however, based on their investigation, these barrier films have minimal effects on both sensitivity and radiation hardness [176]. The device showed no degradation after encapsulation for up to 1000 h within the measurement error margin of 4%.

5. Discussion

Flexible OPDs based on solution-processable technologies provide a significant cost benefit and functional advantage over wafer-based PD techniques by surpassing their natural issues such as strong exciton binding and non-crystalline structures. These advantages are obtained from the chemical composition of the photoactive layer (novel organic materials or hybridization) or from modifying the device structure.

A flexible OPD’s bending radius and $D^*$ are both key figures of merit which can be considered for classification when evaluating their applicability. Among the studied flexible OPDs, the one with D18:Y6 BHJ and on PET substrate [41] shows the highest bending radius of 11 mm due to the more flexible nature of PET in comparison to other flexible substrates. However, it exhibited a moderate $D^*$ of $1 \times 10^{11}$ Jones. In terms of $D^*$, both advanced organic active materials and their hybrid counterparts perform very well by reaching values more than $1 \times 10^{13}$ Jones [43,50,51]. Moreover, utilizing novel functional layers such as new electrodes and interlayers has resulted in the highest detectivity of $1 \times 10^{14}$ Jones for flexible organic photodiodes based on CNT electrodes [23]. In another class of flexible OPDs, conformal devices show promising stretchability by tolerating ~60% of applied strain and losing minimal performance after bending [49]. Nonetheless, their $D^*$ still needs to improve for further feasibility since they show a relatively low value of $1 \times 10^{10}$ Jones due to their elastomeric active materials with lower absorption coefficients.

On the other hand, several critical challenges regarding the real-world commercialization of flexible OPDs remained unsolved. First, although flexible OPDs with good resistance to water [159], heat [177], and mechanical deformation [78] have now been demonstrated, even better output stability and device lifetime are likely preferred. Thus, further development of scalable and low-cost encapsulation strategies and ultrathin passivation layers that do not compromise the mechanical flexibility of the device are needed. Second, the
integration of flexible OPDs with other electronic components (such as batteries, wireless transmitters, and read-out integrated circuits) is essential for fully integrated electronic systems. Ultimately, developing OPDs that can be integrated into textiles and polymer fibers would be a promising route for application in health monitoring and telecommunications.

6. Conclusions

The recent progress toward the realization of high-performance and mechanically flexible OPDs has been highlighted. The combination of efficient charge generation, high detectivity, broad and tunable absorption range, and ease of fabrication on thin and flexible plastic foils make OPDs a very promising technology for conventional inorganic photodetector substitution. Moreover, the ability to use scalable fabrication techniques as well as organic materials’ biocompatibility open up a variety of applications for OPDs in next-generation wearable electronics, such as sensors for the continuous monitoring of health signals and imaging.

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