Recent Advances in Solution-Processable Organic Photodetectors and Applications in Flexible Electronics

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Organic photodetectors (OPDs) are promising for applications in flexible electronics due to their advantages of excellent photodetection performance, cost-effective solution-fabrication capability, flexible device design, and adaptivity to manufacturability. This review outlines the recent advances in the development of high-performance OPDs and their applications in flexible electronics. The approaches to developing different noise reduction methods, filter-free spectral selective detection, flexible OPDs, and scale-up production of flexible OPDs through solution-fabrication processes are discussed. Applications of the OPD technology ultimately result in the materialization of wearable units, flexible and compact information sensors at commercially viable costs, including wearable health self-monitoring devices, flexible optical communication systems, and flexible large-area image sensors.

1. Introduction

Existing silicon (Si), germanium (Ge), and indium gallium arsenide (InGaAs) based photodetectors (PDs) are dominated in the market. These inorganic semiconductor wafer-based PDs are rigid and fragile, having limitations in flexible electronic devices. The emerging solution-processable semiconductor materials, including π-conjugated organic semiconductors, colloidal quantum dots (QDs), perovskite,[1] and low-dimensional materials,[2–4] are promising for use in flexible PDs. For example, organic photodetectors (OPDs) can be prepared using solution-fabrication processes, offering an attractive opportunity to manufacture high-performance PDs through a low-temperature and vacuum-free solution-based process. The bandgap of the organic semiconductors, QDs, and perovskite semiconductors can be tailored for different spectral sensing, making them suitable for PD applications. Much progress has been reported for solution-processed PDs made with the QDs and perovskite. However, the use of toxic materials in these PDs for wearable electronics remains a concern. In comparison, the molecules in the organic semiconductors are carbon-based, such as the molecules of living things, making OPDs more biocompatible and environmentally friendly.

Organic semiconductors, including small molecules and polymers, have profoundly impacted various electronic and optoelectronic devices. Much progress has been made in the development of organic devices, including OPDs, organic light-emitting diodes (OLEDs), organic thin-film field-effect transistors (OTFTs),[5,6] and organic solar cells (OSCs).[7–9] A wide range of light detection from UV, visible, near-infrared (NIR), to short wave infrared (SWIR) range have been realized in OPDs. Great efforts have been made in material synthesis, morphologic control, and device engineering to improve the performance of OPDs, leading to impressive improvements in the performance of the solution-processable OPDs.[10,11] Fabrication of the conventional inorganic semiconductor-based PDs requires the high manufacturing cost and stringent conditions, e.g., a high processing temperature, a high vacuum environment, a complex epitaxial growth, and a sophisticated lithography procedure. OPDs can be fabricated via various methods, including vacuum deposition, spin coating, and solution-based printing fabrication processes. Thermal evaporation has been commonly applied to prepare patterned and multilayer OPDs. An OLED-OPD matrix reflection measurement unit has been fabricated by masked thermal evaporation.[12] In comparison, OPDs are more promising for large-area detection devices and scale-up production due to the solution-fabrication capability. The adaptivity to the manufacturability also enables the monolithic integration of OPD with other electronic devices. The photodetector electronic devices usually involve a readout integration circuit (ROIC) backplane, PDs, and the interconnects between ROIC and PDs. The OPDs can be deposited on top of ROIC, with the conductive component of the ROIC as the bottom electrode of the OPD. The direct integration of ROIC on the ROIC is free from external complex interconnects, compared to inorganic PDs that usually require lattice matching. Large-area OPD-based electronic devices can be scaled up using
solution-based coating techniques such as blade coating, spray coating, screen printing, stamping transfer processes, inkjet printing, and aerosol jet printing.[13]

The summaries on the properties of organic semiconductor materials,[13–15] efficiency, and device physics of OPDs[16,17] were reported by different groups. The developments of photodiodes,[10,18] phototransistors,[19] and photomultiplication (PM)-type OPDs[20,21] have been discussed. Yokota et al. have summarized the OPD designs and performance in flexible image sensors, while Giulio et al. have provided a quantitative analysis of the dark current in OPDs for use in imaging applications. Philip et al. have reported the optoelectronic and mechanical properties of the flexible OPDs. This review discusses the recent progress made in the OPDs, presenting different solution-processable fabrication methods, device designs, and the applications of the flexible OPDs.

First, the fundamentals and the critical figures of merit of OPDs are introduced. Following that, the recent advances of OPDs are addressed: 1) improving the performance of OPDs via suppressing noise current; 2) achieving filter-free spectral selectivity in OPDs, which is particularly important in future flexible and curved devices; and 3) developing flexible OPDs with flexible electrodes and substrates. In the fourth section of this review, the solution-processability of OPDs is highlighted, and the printing techniques are discussed. The fifth section presents the potential applications of OPDs in the field of flexible and wearable electronics. Finally, the challenges and future developments of OPDs are discussed.

2. OPDs

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

2.1. Organic Photodiodes

The organic photodiodes have a vertical two-terminal configuration, comprising an anode, hole-transporting layer (HTL), organic photoactive layer, electron-transporting layer (ETL), and a cathode (Figure 1a). The organic photoactive layers are usually made of an organic bulk heterojunction (BHJ), a blend of organic electron donor and electron acceptor materials. The photocurrent generation processes in the BHJ mainly involve exciton generation, exciton dissociation, charge transport, and charge collection.[18,22] The use of the HTL and ETL facilitates the charge extraction at the BHJ/electrode interfaces in an OPD, resulting in photocurrent generation. The photoresponse of a photodiode relies on the collection of photogenerated charge carriers so that they can be operated either in self-powered condition or under a reverse bias. The typical current–voltage ($I–V$) characteristics of an organic photodiode, obtained in the dark and under light illumination, are shown in Figure 1b. The light current in the reverse-biased region is significantly higher than the dark current, delivering a light-on signal and a light-off signal.

2.2. Organic Photoconductors

The organic photoconductors (OPCs) have a horizontal two-terminal configuration, having two symmetrical electrode contacts, an organic photoactive layer, and the substrate (Figure 1c). The photodetection in OPCs, operated typically under an external bias, is characterized by measuring the change in the electrical conductivity of the photoactive layer induced by the absorption of light. The typical $I–V$ characteristics of the OPCs measured in the dark and under illumination are shown in Figure 1d.

2.3. Organic Phototransistors

The organic phototransistors (OPTs) are a three-terminal device, having an organic photoactive channel layer, a dielectric layer,
and three electrodes, including the gate, drain, and source electrodes (Figure 1e). In OPTs, one type of charge carrier in the channel layer is conducted from the drain electrode and the source electrode, whereas the other type of charge carrier is trapped by the gate electrode. 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 advances in NIR OPTs have been summarized in a recent review article.[19] The performance of OPTs is characterized by the transfer curves measured in the dark condition and under illumination (Figure 1f). The advantage of the OPTs is their high gain due to a photogating effect. Therefore, one single photon incident on the OPT can induce many charge carriers in the channel layer, leading to a high photocurrent.

2.4. Performance Parameters of Photodetectors

2.4.1. External Quantum Efficiency and Responsivity

External quantum efficiency (EQE) is the ratio of the number of charge carriers circling across the PDs to the number of incident photons, which is a dimensionless parameter. Responsivity ($R$) is the ratio of photocurrent measured for the PDs to the power of incident light, with a unit of A W⁻¹. EQE and $R$ indicate the opto-electric conversion efficiency of the PDs. EQE and $R$ are described by

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

$$\text{EQE} = \frac{R \cdot h \cdot \nu}{q} \quad (2)$$

where $I_{ph}$ is the photocurrent measured for PDs, $P$ is the power of incident light, $h$ is the Plank constant, $\nu$ is the frequency of the light wave, and $q$ is the elementary charge. The EQE of the photodiode-type OPD is limited to 100%, while the EQE of the OPTs and PM-type OPDs can exceed 100%. The photoresponse amplification is enabled by light-triggered charge injection in the OPTs and PM-type OPDs.[23,24]

2.4.2. -3 dB Cutoff Frequency and Response Time

The response speed of PDs is evaluated by -3 dB cutoff frequency ($f_{-3\,\text{dB}}$) and response time. $f_{-3\,\text{dB}}$ is defined by the frequency of the modulated light under which the photocurrent, $I_{ph}$, reduces to 70.8% of the photocurrent, $I_0$, measured under continuous light.[18,25] Meanwhile, the relative photoresponse of the PDs, defined by $20 \times \log(I_{ph}/I_0)$, decreases to -3 dB. The $f_{-3\,\text{dB}}$ of the PDs is closely affected by the resistor–capacitor (RC) time constant ($\tau_{RC}$)-limited frequency ($f_{RC}$) of the equivalent circuit and carrier transit time ($\tau_{tr}$)-limited frequency ($f_{tr}$).

$$\frac{1}{f_{-3\,\text{dB}}} = \frac{1}{f_{RC}} + \frac{1}{f_{tr}} \quad (3)$$

$$\frac{1}{f_{RC}} = \frac{1}{2\pi \cdot \tau_{RC}} = \frac{1}{2\pi \cdot RC} \quad (4)$$

$$\frac{1}{f_{tr}} = \frac{3.5}{2\pi \cdot \tau_{tr}} = \frac{3.5 \cdot \mu \cdot E}{2\pi \cdot d} \quad (5)$$

where $\mu$ is the carrier mobility, $E$ is the electrical field, and $d$ is the carrier transit path.

The response time of a PD includes the rise time ($\tau_{r}$), defined by 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 ($\tau_{f}$), defined by the time for the photocurrent of the PD to drop from 90% to 10% of its maximum value upon the removal of light.

2.4.3. Noise

Noise ($S_n$) of OPDs is associated closely to shot noise ($S_{\text{shot}}$), thermal noise ($S_{\text{thermal}}$), 1/f noise ($S_{1/f}$). They are given by

$$S_n = \sqrt{S_{\text{shot}}^2 + S_{\text{thermal}}^2 + S_{1/f}^2} \quad (6)$$

$$S_{\text{shot}} = \sqrt{2q \times I_d \times B} \quad (7)$$

$$S_{\text{thermal}} = \sqrt{\frac{4kT B}{R_{sh}}} \quad (8)$$

where $I_d$ is the dark current measured for the PDs, $B$ is the detection bandwidth, $k$ is the Boltzmann constant, $T$ is the absolute temperature, and $R_{sh}$ is the shunt resistance of the PDs. $S_{\text{shot}}$ in the PDs originates from the electric charges, contributed by the statistical fluctuation of device current. $S_{\text{thermal}}$ is also named Nyquist noise or Johnson noise, is caused by the thermal activation of the charge carriers in the PDs. The sum of $S_{\text{shot}}$ and $S_{\text{thermal}}$ is denoted as the white noise, which is independent of the detection frequency. $S_{1/f}$ also called Flicker noise, depends on the frequency and mostly dominates in the low-frequency range. The sources of $S_{1/f}$ involve the trapping and detrapping of the charge carriers, charge generation–recombination process.

2.4.4. Specific Detectivity

Specific detectivity ($D^*$) is calculated by

$$D^* = \sqrt{A \cdot B} \quad (9)$$

where $A$ is the active area of PD, NEP is the noise equivalent power, a measure of the sensitivity of a PD indicating the power level of incident light that generates a signal-to-noise ratio of one in a 1 Hz output bandwidth.[27] The accurate determination of $R$ and $S_n$ is critical for the calculation of $D^*$. The $S_{\text{shot}}$ is usually considered the primary noise source in OPDs and is used to evaluate $D^*$. However, the measured $S_n$ can be much larger than $S_{\text{shot}}$ (Figure 2a). This can lead to an overestimated $D^*$ due to the thermal noise, especially in OPDs having photoactive layers made with narrow bandgap materials (Figure 2b).[28-30] As the photoresponse and the $S_n$ are frequency-dependent, the $D^*$ shows decays in the $S_{1/f}$-limited low-frequency range and
the response speed-limited high-frequency range (Figure 2c).\(^{31}\) Both the frequency-dependent \(R\) and \(S_n\) should be taken into consideration for calculating the \(D^*\).\(^{27}\)

2.4.5. Linear Dynamic Range

Linear dynamic range (LDR) presents the operational range of intensity of the incident light, which forms a linear relationship with the photoresponse in PDs. LDR is calculated by

\[
\text{LDR} = 20 \times \log \frac{P_{\text{max}}}{P_{\text{min}}} 
\]

where \(P_{\text{max}}\) and \(P_{\text{min}}\) are the maximum and minimum limits of the light intensity range where the photocurrent of PDs follows a linear dependence on the light intensity.\(^{26,32}\)

3. Advances in High-Performance OPDs

The state-of-the-art performance of an OPD requires high responsivity, low noise, and fast response speed. This section summarizes the recent advances on practical approaches for improving the performance of OPDs.

3.1. Noise Current Reduction

In contrast to the OSCs typically operating under strong light conditions, e.g., 100 mW cm\(^{-2}\), the OPDs are usually operated under weak light conditions, especially for applications in wearable health self-monitoring devices, portable detection systems, and flexible large-area image sensors. It becomes critical for the OPDs to have a low noise current for weak light detection. The suppression of noise current in OPDs is a prerequisite for attaining a high signal-to-noise ratio, a high \(D^*\), and a broad LDR.

The noise current in OPDs includes the intrinsic noise current and the extrinsic noise current such as injection current. Various approaches have been developed to reduce the noise current in OPDs, including reducing the leakage current using a thicker photoactive layer, morphological engineering of the organic photoactive layer,\(^{33}\) and suppressing charge injection current. The noise current in OPDs can be reduced using a thicker photoactive layer to increase the device resistance and prevent the pinholes in the photoactive layer.\(^{144}\) However, the use of a thicker photoactive layer usually results in a decrease in the responsivity and response speed of the OPDs.\(^{15,16}\) There is a trade-off between realizing the high performance of OPDs and using the thicker photoactive layer. The reduction of noise current without sacrificing the responsivity is desired for achieving high-performing OPDs.

The charge injection process and the suppression mechanisms of injection current in OPDs are illustrated in Figure 3a. The use of a BHJ photoactive layer is favorable for exciton dissociation in OPDs, contributing to the high responsivity. However, the charge injection in OPDs can occur at anode/organic side by electron injection or cathode/organic side by hole injection, when the OPDs are operated under a reverse bias. The injection energy barriers at electrode/organic interfaces play a critical role in the charge injection process in OPDs. The charge injection energy barriers include electron injection barrier, defined by the difference between the Fermi level of the anode and the lowest unoccupied molecular orbital (LUMO) level of acceptors, and hole injection barrier, defined by the difference between the Fermi level of cathode and the highest occupied molecular orbital (HOMO) level of donors. On the one hand, an anode with a higher work function and a cathode with a lower work function are usually beneficial to the charge injection suppression in OPDs. On the other hand, the charge injection in OPDs can be suppressed through different methods: 1) energetic barriers caused by planar heterojunction (PHJ) photoactive layer or the vertical phase segregation in the BHJ photoactive layer (left inset in Figure 3a); 2) using charge injection blocking layer, including electron blocking layer and hole blocking layer (right inset in Figure 3a).

The reduced dark current in conventional PHJ OPDs has been observed.\(^{137}\) The donor/acceptor bilayer PHJ has been proven to suppress the charge injection in OPDs, usually resulting in a lower dark current in PHJ OPDs than the BHJ counterparts. However, the responsivity of PHJ OPDs is limited by low exciton dissociation efficiency due to the limited donor/acceptor interfaces. An alternative solution is the donor/BHJ/acceptor-type vertical phase-segregated photoactive layer, as indicated in the left inset in Figure 3a.\(^{138,139}\) A clear donor/anode contact and a pure acceptor/cathode contact are formed to block charge...
injection. The vertical phase-segregated photoactive layer can be prepared by the sequential deposition of the donor layer and acceptor layer. All-polymer OPDs, with vertical phase separation of the donor and acceptor in the photoactive layer, were prepared using a sequential layer-by-layer solution deposition process using orthogonal solvents for depositing polymeric donor and acceptor materials, PTzBI-Ph and N2200, respectively.[40] The PTzBI-Ph/N2200 bilayer photoactive layer shows a gradient component distribution of PTzBI-Ph and N2200, with a PTzBI-Ph-rich region near to the anode side and an N2200-rich region near the cathode side (Figure 3b). It clearly shows that only PTzBI-Ph donors are contacted to the anode, and only N2200 acceptors are contacted to the cathode. On the contrary, the PTzBI-Ph:N2200-based BHJ exhibits a more homogenous vertical distribution of PTzBI-Ph and N2200 in the active layer, indicating that both the donors and acceptors have direct contacts with the anode and cathode. The performances of BHJ OPD prepared by a PTzBI-Ph:N2200 BHJ photoactive layer and the gradient BHJ OPD prepared by a bilayer PTzBI-Ph/N2200 photoactive layer were compared. It shows that the gradient BHJ OPD has a lower dark current and higher photocurrent (Figure 3c). The thickness-dependent characteristics were also studied, revealing a decrease in responsivity in a thicker OPD and an increase in dark current in the thinner OPDs. The
optimized PTzBI-Ph (80 nm)/N2200 (60 nm) gradient BHJ OPD shows a highest $D^*$ of $5.68 \times 10^{12}$ Jones. Furthermore, the gradient BHJ OPD shows excellent stability after aging for 160 days, suggesting that the sequential layer-by-layer approach is a reliable strategy for attaining low-noise and high-$D^*$ OPDs.

The vertical phase separation of the donor and the acceptor in the active layer is formed primarily via the interdiffusion of the organic donors and acceptors. The interdiffusion can be improved by adding a small portion of the nonorthogonal solvent in the solution precursor of the upper organic layer. For example, the OPDs fabricated from a layer-by-layer deposition process by sequentially depositing P3HT using a solvent of o-DCB, and PC$_{71}$BM using a solvent of 2-CP were studied.\(^{141}\) The layer-by-layer P3HT/PC$_{71}$BM OPDs exhibit a lower dark current density ($J_d$) of $1.39 \times 10^{-7}$ A cm$^{-2}$ than the P3HT:PC$_{71}$BM BHJ OPDs ($4.12 \times 10^{-6}$ A cm$^{-2}$). However, the P3HT:PC$_{71}$BM OPD also shows a reduced photocurrent compared to the P3HT: PC$_{71}$BM BHJ OPDs. Different portions of o-DCB were added in the PC$_{71}$BM precursor solution to assist the intermixing of P3HT and PC$_{71}$BM during the deposition process (Figure 3d). It was found that the 10% o-DCB in the PC$_{71}$BM precursor solution leads to an optimized OPD performance having a simultaneous low dark current, high photocurrent, high on/off current ratio, and the highest $D^*$. The results provide a method for optimizing vertical phase separation of the donor and acceptor in the photoactive layer for mitigating the noise current in OPDs, fabricated by the sequential layer-by-layer solution deposition.

The use of a charge blocking layer is an effective way to suppress the noise current in OPDs. It is challenging to find suitable charge blocking layers with appropriate energy levels and carrier mobility. Intense research efforts have been made to find the charge blocking materials for making low-noise OPDs.\(^ {28,42-45}\) Recently, the solution-processed CuSCN was used as the electron blocking layer in the NT40:IEICO-4F-based broadband OPDs, showing a significant function of blocking the electron injection in the OPDs.\(^ {146}\) The CuSCN interlayer was found to form a higher electron-injection barrier between the anode and organic photoactive layer, creating a higher shunt resistance in OPDs compared to that of the OPDs with a PEDOT:PSS interlayer due to its higher conduction band in CuSCN (Figure 3e). The OPDs with a CuSCN interlayer show a reduced $J_d$ of $2.7 \times 10^{-10}$ A cm$^{-2}$ as compared to that of the OPDs with a PEDOT:PSS interlayer ($2.3 \times 10^{-8}$ A cm$^{-2}$). Meanwhile, an increase in the depletion width was observed in the CuSCN-based OPD, leading to an improving responsivity. An order of magnitude increase in the $D^*$ was obtained for the OPD with a CuSCN HTL as compared to that of the control OPD with a PEDOT:PSS HTL. A room-temperature solution-processed p-type delafossite CuCrO$_2$ nanomaterial was used as an electron blocking layer in a large-area OPD.\(^ {147}\) The OPD has a low $J_d$ of $6.48 \times 10^{-8}$ A cm$^{-2}$ even when the device was operated under a reverse bias of $-5.0$ V. The low $J_d$ enables the OPD to fabricate a large-area image sensor with an active area of 75 mm $\times$ 81 mm. An ultralow $J_d$ of $\approx 0.5$ nA cm$^{-2}$ at $-2.0$ V and a high $D^*$ of over $10^{13}$ Jones at wavelengths up to 940 nm were obtained in NIR OPDs using a cross-linked PolyTPD electron blocking layer and a mixed C$_{60}$/LiF hole blocking layer.\(^ {148}\) The cross-linked PolyTPD layer was found to contribute to an excellent reproducibility and stability of the NIR OPDs. Meanwhile, the trap density and trap-induced carrier generation in the organic/cathode interface were effectively suppressed by the LiF doping C$_{60}$ layer, leading to the ultralow dark current in the NIR OPDs.

Recently, the reliability of ZnO ETL and its hole blocking function in low-noise and high-$D^*$ OPDs have been investigated.\(^ {149}\) It was found that the exposure to high-energy photons, such as UV light, would lead to a significant increase in the noise in OPDs having a ZnO ETL, e.g., increasing from $1.65 \times 10^{-14}$ A Hz$^{-0.5}$ to $5.52 \times 10^{-11}$ A Hz$^{-0.5}$ (Figure 3f). Correspondingly, the $D^*$ of the OPDs decreases from $10^{12}$ Jones to $10^{9}$ Jones, indicating a decrease of three orders of magnitude (Figure 3g). The phenomenon was attributed to the loss of selective electron-transporting and enhanced hole-transporting in the ZnO layer after exposure to UV light. In comparison, the performance of the OPDs having a SnO$_2$ ETL was more stable and not affected by the light exposure history (Figure 3f,g). The OPDs having a double-layer SnO$_2$ ETL show a low noise, a high $D^*$, and satisfied figures of merit without losing consistency after light illumination, proving an option for superior photodetection stability in OPDs.

### 3.2. Filter-Free Spectral Selective OPDs

In practice, spectral selective or narrowband photodetection is achieved by combining a broadband PD with a dedicated optical filter. The optical filters are usually rigid and not practical on flexible devices. The development of flexible optical filters remains a challenge.\(^ {50-52}\) The filter-free spectral selective OPD techniques are desired. Filter-free OPDs have been demonstrated via novel device designs and the dynamics of the photogenerated charge carriers. This section discusses the recent developments of filter-free spectral selective OPDs using charge collection narrowing (CCN), internal light depletion mechanism, and charge-injection PM effect. The performance characteristics of recent filter-free spectral selective OPDs are summarized in Table 1. The unique feature of the bias-switchable spectral selective OPDs that can detect incoming light selectively in different bands is discussed.

#### 3.2.1. Charge Collection Narrowing

The CCN-type narrowband OPDs were proposed using a thick photoactive layer and utilizing the electro-optical properties.\(^ {53}\) The CCN effect has the function of shaping the internal quantum efficiency to obtain spectral selective photoresponse due to the unbalanced charge-transporting behavior between the electrons and the holes. The dynamics of charge carriers in CCN-type OPDs are revealed in Figure 4a. The above-bandgap light is absorbed near the surface of the photoactive layer due to a high absorption coefficient. In contrast, the near-bandgap light is absorbed in the bulk region of the photoactive layer due to a modest light absorption coefficient. The photogenerated charge carriers, generated near the surface region of the photoactive layer, are lost due to the charge recombination induced by the highly unbalanced charge carrier transport. The photogenerated charges formed in the bulk of the photoactive layer can be
Table 1. A summary of recent progress made in the development of spectral selective OPDs.

| Photoactive layer | λ [nm] | FWHM [nm] | Bias [V] | R [mA W⁻¹] | D* [Jones] | Ref. |
|-------------------|--------|-----------|----------|-------------|------------|------|
| **Charge collection narrowing** |        |           |          |             |            |      |
| PCDTBT:PC71BM (2.0 μm) | ≈650  | ≈90       | 1.0      | ≈100        | ≈2.0 × 10¹²  | [53] |
| DPP-DTT:PC71BM (2.0 μm) | ≈950  | ≈90       | 1.0      | ≈60         | ≈5.0 × 10¹²  | [60] |
| F8BT:hPDI2 | 575  | 22        | 3.0      | 8.0         | 2.6 × 10⁰    |      |
| F8BT:hPDI3 | 600  | 19        | 3.0      | 7.3         | 1.7 × 10⁰    |      |
| F8BT:hPDI4 | 615  | 16        | 3.0      | 3.4         | 1.1 × 10⁰    |      |
| F8BT:hPDI3-Pyr-hPDI3 | 645  | 20        | −3.0     | 1.1         | 9.3 × 10⁹    |      |
| Internal light depletion |        |           |          |             |            |      |
| P3HT (708 nm)/PC61BM (70 nm) | 635  | ≈50       | 0        | 128         | 1.9 × 10¹²   | [60] |
| PTB7-Th:P3HT (8.5, 670 nm)/PCBM (100 nm) | 780  | 50–60    | 0.5      | 28          | 1.3 × 10¹²   | [67] |
| PPD2FBT:MEH-PPV (8.5, 670 nm)/PCBM (100 nm) | 705  | 50–60    | 1.0      | 70          | 3.0 × 10¹²   |      |
| P3HT (150 nm)/NTB812 (≈750 nm)/Y6 (≈50 nm) | 860  | 72       | 0.1      | 451         | 2.4 × 10²    | [62] |
| P3HT (150 nm)/DT-PDPP2T-TT/Y6 | 910  | 43       | 0.1      | ≈220        | –          |      |
| P3HT (150 nm)/DT-PDPP2T-TT/EICO-4 F | 940  | 66       | 0.1      | 240         | –          |      |
| PTB7-Th (800 nm)/PTB7-Th:CO,8DFIC:PC71BM (1:1:0.5, 100 nm) | 830  | ≈200    | 0        | 390         | 4.0 × 10¹¹   | [61] |
| P3HT (430 nm)/PBDB-T:m-ITIC (1:1, 120 nm) | 700  | ≈115    | 0        | 300         | –          |      |
| **Narrowband photomultiplication** |        |           |          |             |            |      |
| P3HT:PC71BM:CdTe QDs (3500 nm) | 660  | ≈120     | 6.0      | 1.06        | 7.3 × 10¹¹   | [72] |
| P3HT:PC71BM (100, 2.5 μm) | 650  | ≈28       | 10       | 0.26        | 1.3 × 10¹¹   | [71] |
| P3HT:PC71BM (100, 4 μm) | 665  | 23        | 10       | ≈0.37       | 4.7 × 10⁰    | [76] |
| P3HT:PTB7-Th:PC61BM (40:60:1, 3000 nm) | 800  | 40        | 10       | 0.03        | 2.4 × 10⁰    | [77] |
| F8T2:ZnO (1:6, 450 nm) | 358  | 16        | 3.0      | 2.26        | 8.5 × 10²    | [73] |
| F8T2:ZnO (1:3, 450 nm) | 360  | 19        | 15       | 6.30        | 1.2 × 10¹¹   | [105] |
| P3HT:PC71BM (100, 300 nm)/P3HT (320 nm)/P3HT:PTB7-Th:PC61BM (70:30:1, 500 nm) | 376  | ≈60       | 15       | 7.47        | 8.0 × 10²    | [78] |
| P3HT:PTB7-Th:PC61BM (70:30:1, 500 nm) | 654  | ≈280    | 35       | 5.48        | 5.3 × 10²    |      |

collected efficiently. As a result, the narrowband responsivity of CCN-type OPD located at the wavelength near the absorption edge of the photoactive layer (Figure 4a). Sub-100 nm full width at half maximum (FWHM) visible-blind red and NIR narrowband OPDs have been developed using a thick photoactive layer, e.g., a 2.0 μm-thick PCDTBT:PC71BM layer or a 2.0 μm-thick DPP-DTT:PC71BM layer. The PCDTBT:PC71BM layer has an absorption edge at ≈650 nm, and the PCDTBT:PC71BM-based CCN-type OPD shows narrowband responsivity near 650 nm with an FWHM of <90 nm. The absorption spectrum of the DPP-DTT:PC71BM layer extends to the NIR range and has a cutoff at ≈950 nm. Thus, the DPP-DTT:PC71BM-layer based CCN-type OPD exhibits NIR narrowband responsivity near 950 nm with an FWHM of <90 nm. The CCN-type OPD shows a redshifted response as the thickness of the photoactive layer increases. The detection window of the CCN-type OPD can be spectrally fine-tuned over a 100 nm range by adjusting the thickness of the photoactive layer. A thick junction reduced the defect density and suppressed the dark current, resulting in a high $D^*$ of $10^{12}$ Jones and a large LDR of 160 dB.

Various CCN-type narrowband PDs have been demonstrated using perovskite materials and perovskite/organic hybrid photoactive layers, showing a tunable narrowband photoresponse covering the visible and NIR wavelength range. The photoactive materials used in the CCN-type OPDs should fulfill the following requirement: 1) a high absorption coefficient above the optical bandgap and a sharp absorption edge; 2) the absorption edge should be tunable across different wavelength ranges. The bandgap tuning of the organic semiconductors is highly desired for designing CCN-type narrowband PDs. A set of helical ribbons were synthesized for the fabrication of CCN-type OPDs with a narrow FWHM. The synthesized helical ribbons, hPDI2, hPDI3, hPDI4, hPDI3-Pyr-hPDI4, show a sharp absorption edge and gradually redshift absorption (Figure 4b). The CCN-type OPDs, made using the helical ribbons as electron acceptors in photoactive layers, exhibit narrowband response in a wavelength range from 575 to 645 nm, with a narrow FWHM of 16–22 nm (Figure 4c). Such a narrow FWHM was realized by the sharp absorption edge of the helical ribbons, sufficient absorption coefficient above the absorption edge, and good carrier mobility. The rapid development of nonfullerene acceptors, having tunable NIR light absorption compared to the limited choices of fullerene counterparts, offers more choices for the development of spectral selective OPDs and their applications in flexible and wearable electronics.
3.2.2. Internal Light Depletion

Spectral selective photodetection can be achieved by regulating the optical profile and exciton dissociation in the OPDs, realized by the light depletion layer/light-absorption BHJ bilayer photoactive layer structure. A light depletion layer/light-absorption BHJ bilayer configuration OPD was demonstrated for achieving filter-free band-selective photodetection (Figure 4d–f). The light depletion layer has dual functions: 1) internal light depletion layer that depletes the shorter wavelength light; 2) efficient charge carrier transport layer. The dynamics of charge carriers in the bilayer spectral selective OPDs are illustrated in the schematic drawing in Figure 4d. The appropriate selection of depletion and BHJ layers allows the shorter wavelength light to be fully absorbed in the depletion layer and the longer wavelength light to be absorbed in the BHJ layer. The excitons created in the light depletion layer, due to absorption of shorter wavelength light, are lost because of charge recombination. The charge carriers generated in the BHJ layer, only by the absorption of longer wavelength light, are collected, forming a spectral selective photoreponse. As a result, the selective spectral response in the bilayer OPDs can be realized by adjusting the optical profile in the photoactive layer. For example, a visible-blind NIR-selective OPD was demonstrated using an 800 nm PTB7-Th light depletion layer and a 100 nm-thick PTB7-Th:CO8DFIC:PC71BM absorption layer and an 800 nm-thick PTB7-Th light depletion layer when operated in self-powered condition and under different biases of ~0.4 and ~1.0 V. The schematic diagrams showing the dynamics of the charge carriers in the PM-type narrowband OPDs, operated under reverse bias, in the presence of light source with wavelengths of g) 360 and 520 nm, h) 650 nm. i) The EQE spectra, measured under different biases, for the PM-type OPDs with P3HT:PC71BM (100:1) layer.

Figure 4. a) Schematic drawing illustrating the dynamics of the charge carriers, and the correlation between the absorption profile in photoactive layer and the response EQE spectrum in the CCN-type OPDs. b) The normalized absorption spectra of a set of organic materials: F8BT, hPDI2, hPDI3, hPDI4, hPDI3-Pyr-hPDI4. c) The normalized EQE spectra measured for the CCN-type OPDs fabricated using the above functional organic semiconductor materials. d) The schematic diagram shows the dynamics of the charge carriers in the bilayer spectral selective OPDs. e) The normalized absorption spectrum of a PTB7-Th:CO8DFIC:PC71BM ternary BHJ layer and the transmission spectrum of an 800 nm-thick PTB7-Th light depletion layer. f) The responsivity spectra measured for the visible-blind NIR selective OPD, having a 100 nm-thick PTB7-Th:CO8DFIC:PC71BM absorption layer and an 800 nm-thick PTB7-Th light depletion layer when operated in self-powered condition and under different biases of ~0.4 and ~1.0 V. The schematic diagrams showing the dynamics of the charge carriers in the PM-type narrowband OPDs, operated under reverse bias, in the presence of light source with wavelengths of g) 360 and 520 nm, h) 650 nm. i) The EQE spectra, measured under different biases, for the PM-type OPDs with P3HT:PC71BM (100:1) layer. (a–c) Reproduced with permission. Copyright 2017, American Chemical Society. (d–f) Reproduced with permission. Copyright 2020, John Wiley and Sons. (g–i) Reproduced with permission. Copyright 2017, American Chemical Society.
response at around 500 nm, a P3HT layer was introduced to the OPD as a HTL. Benefitting from the higher LUMO level of P3HT compared to the NT812 donor layer, the dark current of the OPD was further suppressed. With this strategy, a filter-free narrowband OPD centered at 860 nm with an FWHM of \(\approx 50 \, \text{nm}\), a peak EQE around 65\%, and a \(D^*\) over \(10^{13}\) Jones has been obtained. In this work, the underlying donor polymer NT812 was cross-linked to be a robust film by a cross-linker s-4PFA, for the sequential deposition of multilayer organic stack. Then, a thin layer of NT812 was deposited, followed by deposition of Y6 to realize interdiffusion BHJ layer, which guaranteed the efficient dissociation for the charge carriers generated by NIR light.

The depletion layer in the bilayer structure should have a high absorption coefficient in the shorter wavelength range and high charge carrier mobility to realize optical and electrical functions. The emerging organic–inorganic perovskite materials show a high absorption coefficient, solution processability, high charge mobility for holes and electrons, and tunable energy level. They are considered as a suitable candidate for the depletion layer in the bilayer spectral selective OPDs. A visible-blind NIR narrowband OPD, having a 1700 nm-thick CH\(_3\)NH\(_3\)PbI\(_3\) depletion layer and a 200 nm-thick PTB7-Th:PC\(_{71}\)BM BHJ layer, shows a narrowband response at 800 nm with an FWHM of \(<50 \, \text{nm}\)\(^{63}\). The narrowband OPD has an \(f_{\text{3dB}}\) of over 300 kHz, which is beneficial from the high charge mobility of the perovskite layer. The tunable absorption spectra of perovskite materials enable a tunable spectral response of the perovskite/organic bilayer OPDs spanning over visible to NIR ranges. The perovskite/organic bilayer OPDs also show suppressed \(J_d\) of \(1.85 \times 10^{-5} \, \text{mA cm}^{-2}\) and a high \(D^*\) of over \(10^{12}\) Jones\(^{64}\). A narrowband NIR OPD exhibiting an EQE of 74.26\% at 810 nm and an FWHM of 95 nm was realized using CH\(_3\)NH\(_3\)PbI\(_3\)/CuSCN light depletion layer and PM6:Y6 BHJ light-absorption layer\(^{65}\).

The spectral selective OPDs were fabricated by sequential deposition of polymer donor and acceptor using orthogonal solvents or evaporation deposition methods. An interdiffused BHJ layer would form at the interface of the donor layer and the acceptor layer. The donor layer itself acts as the internal light depletion layer in the bilayer OPD. This OPD configuration also benefits from a reduced dark current as compared to the BHJ-based OPDs. A green light selective OPD was developed using an F8T2 donor layer and a 70 nm-thick PC\(_{61}\)BM acceptor layer. A red light selective OPD was prepared using P3HT (708 nm) and a 70 nm-thick PC\(_{61}\)BM acceptor layer\(^{66}\). The red OPDs show a reduced dark current than the single BHJ layer-based OPD, an \(S_o\) of \(10^{-14} \, \text{AHz}^{-0.5}\), and a high \(D^*\) of over \(10^{12}\) Jones. The red light selective OPD exhibits a narrow FWHM of below 50 nm, while a high \(f_{\text{3dB}}\) of 15 kHz was retained. An NIR-selective OPD was fabricated using a p-type polymer blend of PTB7-Th:P3HT (8:5 by weight) as a light depletion layer\(^{67}\). The P3HT and PTB7-Th have complementary light absorption across 400–600 nm and 600–800 nm, and the light absorption of the PTB7-Th:P3HT blend layer entirely covers the visible range. As a result, the NIR-selective OPDs thus developed show narrowband NIR responsivity centered at 780 nm for the PTB7-Th:P3HT-based bilayer OPD, with a narrow FWHM and a well-suppressed response to visible light.

### 3.2.3. Narrowband Photomultiplication

The recently reported PM-type OPDs comprise a photoactive layer made by blending donor and acceptor at an unbalanced ratio. For example, a P3HT:PC\(_{71}\)BM blend layer, prepared using a weight ratio of P3HT to PC\(_{71}\)BM of 100:1, has been used to fabricate PM-type OPDs. The PC\(_{71}\)BM molecules spread in the organic photoactive layer form electron traps due to a lower LUMO level. The photocurrent in PM-type OPDs, e.g., comprising a layer configuration of ITO/PEDOT:PSS/P3HT:PC\(_{71}\)BM (100:1)/Al, is generated through a trap-assisted charge injection process. In the presence of light, the photogenerated electrons are trapped by the PC\(_{71}\)BM-induced traps in the photoactive layer. The accumulation of electrons near the vicinity of the P3HT/Al interface leads to a downward energy band bending. A narrow width of the energy barrier is formed, allowing the charge carriers tunneling injection at the P3HT/Al interface. Therefore, the holes can be effectively injected into the photoactive layer through the tunneling effect, generating a photocurrent. The PM-type OPDs with an EQE of \(>100\%\) can be achieved, delivering a high responsivity. However, the PM-type OPDs have a relatively slow response speed\(^{68,69}\). It has been found that the spectral response of the PM-type OPDs was strongly related to the optical profile in the devices\(^{70}\). The responsivity spectrum of the PM-type OPDs is primarily determined by the charge generation profile near the organic/electrode interface, where the charge injection process occurs. The narrowband photoresponses in the trap-assisted charge-injection PM-type OPDs are achieved through optimizing the optical profile to allow light with targeted wavelength to be absorbed near the charge-injecting organic/electrode interface. The light out of the targeted wavelength range is not absorbed near the organic/electrode interface. The reported PM-type OPDs have several advantages, including a narrow response window with an FWHM of \(<30 \, \text{nm}\), a high EQE of \(>100\%\), and a high spectral detection ratio.

A filter-free narrowband PM-type OPD, having a photoactive layer of P3HT:PC\(_{71}\)BM with a weight ratio of 100:1 and a thickness of 2.5 \(\mu\)m, was reported\(^{71}\). The narrowband PM-type OPD shows a peak EQE value of 53.5\% and an FWHM of \(<30 \, \text{nm}\) at the wavelength of 660 nm when operated under \(-60 \, \text{V}\). The charge injection narrowing mechanisms in PM-type narrowband OPDs are illustrated in Figure 4g,h. The electrons are difficult to move in the P3HT:PC\(_{71}\)BM (100:1) layer due to the lack of electron percolation channels. The charge injection is assisted by the trapped electrons in PC\(_{71}\)BM near the Al electrode under light illumination. The density of the electrons trapped in PC\(_{71}\)BM near the hole injection electrode plays a crucial role in determining the interfacial band bending for hole tunneling injection from the external circuit, thus creating photoresponse. The distribution of photogenerated electrons shows that only the incident light with a longer wavelength of around 650 nm can generate electrons near the Al side. The EQE measured for the PM-type OPDs with P3HT:PC\(_{71}\)BM (100:1) layer, operated under different biases, is shown in Figure 4i. The 2.5 \(\mu\)m-thick P3HT:PC\(_{71}\)BM (100:1)-based PM-type OPD exhibits a high EQE of over 50\%.
A highly sensitive narrowband red-to-NIR light OPD with a high gain was achieved by introducing CdTe QDs as electron traps in P3HT:PCBM photoactive layer with a thickness of 3.5 μm. The performance of OPDs with photoactive layers of P3HT:PCBM and P3HT:PCBM: CdTe was characterized. The OPD with CdTe QDs shows an EQE of around 200%, which was 20-folds of the EQE of the OPD without CdTe QDs. The gain in the OPD with CdTe QDs was realized by trap-assisted secondary hole injection when the OPD was operated under a reverse bias.

The broadband light absorption range of organic semiconductors makes it challenging to achieve a narrowband photodetection in the UV range. A UV narrowband PM-type OPD was demonstrated using a blend of a wide bandgap conjugated polymer F8T2 and a wide bandgap ZnO nanoparticles. The charge injection is realized by the trapped electrons in the ZnO particles that induce band bending of F8T2 and promote the hole tunneling process. The OPDs with an optimized F8T2:ZnO ratio of 1:3 show a responsivity of 0.224 A W⁻¹, a narrow FWHM of 16 nm, and a D* of 8.45 × 10¹² Jones at 358 nm under a bias of 3.0 V. Recently, a visible-blind UV narrowband PM-type OPD was demonstrated using 80 nm-thick TAPC:C60 (50:1) as the photoactive layer. The wide bandgap donor TAPC shows an absorption cutoff wavelength at around 370 nm and the small amount of C60 enables weak absorption to visible light in the photoactive layer. As a result, the UV OPD shows a narrowband response at 335 nm with an FWHM of 36 nm, an EQE of 1.08 × 10⁻⁶%, and a D* of 1.28 × 10¹⁴ Jones.

3.2.4. Bias-Switchable OPDs

The bias-switchable OPDs that respond to spectral selective light are attractive for multispectral photodetection and applications in flexible electronics. The bias-switchable spectral response has been achieved in PM-type OPDs with bidirectional charge injection. The PM-type OPDs show distinctly different spectral responses when operated under forward and reverse bias. A bias-switchable NIR and visible light dual-mode PM-type OPD was reported. The dual-mode OPD, having a trilayer visible light absorber/optical spacer/NIR light absorber photoactive layer configuration, exhibits a unique feature of a high NIR light response when operated under a reverse bias, and visible light response when operated under a forward bias. The profile of charge generation has revealed that the charge carriers generated by visible light were generated in the visible absorber, and the charge carriers generated by the absorption of the NIR light were created in the NIR absorber layer. In the presence of the NIR light, photocurrent is produced in the NIR absorber layer due to the trap-assisted charge injection at the organic/cathode interface at a reverse bias. Under visible light, photocurrent is produced in the visible absorber layer, enabled by the trap-assisted charge injection at the anode/organic interface at a forward bias. The dual-mode OPD shows an NIR responsivity of 8.66 A W⁻¹ when operated under −40 V and a visible responsivity of 7.47 A W⁻¹ when operated under 15 V.

Recently, an electrically switchable color-selective OPD, having a double-BHJ structure of ITO/ZnO (10 nm)/F8T2:

Figure 5. The schematic diagrams showing the charge carrier dynamics in the double-BHJ OPD when operated a) a forward bias, and b) a reverse bias. The responsivity measured for the double-BHJ OPD when operated c) in self-powered condition and under different forward biases, and d) under different reverse biases. (a,b,c,d) Reproduced with permission. Copyright 2021, American Chemical Society.
PC favouring metal oxides (MOs) (e.g., MoOx or MoOy, 10 nm)/Ag (22 nm) and conducting polymers such as conductive PEDOT:PSS (also named PH1000) are promising candidates for replacing the ITO. The deposition of the TCEs via a solution process is essential to the cost-effective manufacturing of flexible OPDs. The development of printable TCEs has been summarized in a review article. Recently, advances and performances of flexible OPDs are summarized in Table 2.

Metal thin films are the most commonly used conductive electrodes in optoelectronic devices, however, a metal film has limited transparency and is usually used as the rear reflective electrode in OPDs. Reducing the thickness of the metal electrode is a straightforward approach to realize flexible TCEs. For instance, the MoOx/Ag electrodes have been widely applied to fabricate semitransparent OSCs and flexible OPDs. The MoOx layer has a dual function of improving the coverage of the photoactive layer, and the adhesion of metal TCE to the substrates. A large-area flexible OPD with an area of 1.0 cm² was prepared using 700 nm-thick P3HT:ICBA as the photoactive layer, a MoOx (10 nm)/Ag (11 nm) TCE, and a flexible PES substrate. It shows a low dark current of around 10⁻¹⁰ A, a responsivity of 0.1 A W⁻¹, and a D* of 10¹² Jones. The flexible OPDs yielded compatible dark current, slightly reduced responsivity, and similar D* compared with the OPDs fabricated on rigid substrates and using ITO TCEs. A ring-shaped flexible OPD photoplethysmogram (PPG) sensor was demonstrated, indicating the ease of creating OPDs with complex shapes for applications in wearable electronics. A flexible narrowband OPD was prepared using a TCE made of MoOx, PES substrates and using ITO TCE. A large-area flexible PES substrate was used by the PI film, considering the incident light, with a wavelength of <0.500 nm, was fully depleted by the PI film, resulting in zero

3.3. Flexible OPDs

Organic semiconductors are inherently mechanically soft, providing the successful development of flexible OPDs on curved, conformable, flexible, and foldable substrates. The preparation of flexible OPDs raises the request for flexible substrates and flexible transparent conductive electrodes (TCEs). The available flexible substrates include a variety of plastic foils such as polyethylene terephthalate (PET), polyethylene naphthalate (PEN), polyethersulfone (PES), polyimide (PI),[80,81] and polydimethylsiloxane (PDMS).[82] The TCEs in flexible OPDs should meet the requirements of low sheet resistance, high optical transmittance, favorable work function, and robust flexibility. ITO layer is rigid and fragile, and is one of the most commonly used TCEs in the OPDs. The deposition of the ITO electrode usually requires a high temperature, which is not favored by the plastic foil substrates that usually have a lower glass transition temperature.

Table 2. A summary of recent progresses made in the development of flexible OPDs.

| Substrate | Transparent electrode | Manufacture | Photoactive layer | R [A W⁻¹] | D* [Jones] | Ref. |
|-----------|-----------------------|-------------|------------------|----------|------------|-----|
| PET       | ITO (150 nm)          | –           | P3HT:PC61BM      | 6.39     | 8.8 × 10¹¹ | [105]|
| SU-8/Parylene | ITO (100 nm)       | Sputter deposition | PIPCP:PC61BM     | 0.14     | 1.3 × 10¹³ | [114]|
| SU-8/Parylene | ITO                 | –           | PTzNTz-BOBO:PC61BM | 0.18     | –          | [115]|
| Parylene  | ITO (75 nm)           | Sputter deposition | P3HT:PCBM        | 0.21     | –          | [158]|
| PI        | ITO                   | –           | P3HT:PCBM        | 0.09     | –          | [45] |
| PI        | Ag (22 nm)            | Thermal evaporation | PCDTBT:PCBM      | 0.35     | 2.2 × 10¹² | [90] |
| PI        | MoOx (10 nm)/Ag (10 nm)/MoOx (30 nm) | Thermal evaporation | P3HT:PCBM       | 0.07     | –          | [95] |
| PES       | MoOx (10 nm)/Ag (11 nm) | Thermal evaporation | P3HT:ICBA       | 0.10     | 1.1 × 10¹² | [11] |
| PET       | MoOx (1 nm)/Ag (9 nm)/MoOx (2 nm) | Thermal evaporation | P3HT:PC71BM     | 388.4    | 2.0 × 10¹³ | [106]|
| PET       | Graphene/PEDOT:PSS (20 nm) | Spray coating | P3HT:PCBM       | 0.16     | 1.3 × 10¹² | [85] |
| PET       | PEDOT:PSS (200 nm)    | Slot-die coating | PBD8-T:PDNI:FT10 | 0.18     | > 1.0 × 10¹³ | [103]|
| PET       | PEDOT:PSS:CNT         | Spin coating | P3HT:PCBM       | 0.30     | –          | [87] |
| PET       | PEDOT:PSS             | Spin coating | D18/Y6           | 0.35     | 1.1 × 10¹² | [39] |
| PET       | PEDOT:PSS             | Slot-die coating | PTB7-Th:PDNI:FT10 | 0.18     | > 1.0 × 10¹³ | [104]|
| PET       | PEDOT:PSS (180 nm)    | Aerosol jet printing | PBT7:PC70BM      | 0.20     | 2.0 × 10¹¹ | [126]|
| PEN       | PEDOT:PSS             | Injet printing | P3HT:PCBM       | 0.20     | 3.4 × 10¹² | [101]|
| PEN       | PEDOT:PSS             | Blade coating | PVD4650:PC70BM   | ≈ 0.22   | –          | [120]|
| PEN       | CNT                   | Lamination | PBDTTT-EFT:PC70BM | ≈ 0.12   | 2.1 × 10¹⁴ | [97] |
| PEN       | CNT                   | Spray coating | P3HT:PCBM       | 0.32     | –          | [96] |
response to light with a wavelength below 500 nm. On the other hand, the transmittance of the MoO3 (10 nm)/Ag (10 nm)/MoO3 (30 nm) TCE decays rapidly at a wavelength above 600 nm. The flexible OPD shows a narrowband responsivity at 570 nm with an FWHM of 100 nm.

CNTs have been used as TCE for fabricating flexible OPDs.[96,97] A CNT TCE-based flexible OPD is illustrated in Figure 6a. The density of CNTs was optimized by considering the optical transmittance and the conductivity of the TCEs. The optimal flexible CNT electrode shows an average visible transmittance of 90% and a sheet resistance of 98 Ω cm². The CNT-based OPDs show a lower Jd of 9.62 A W⁻¹ and a higher D* of 2.07 × 10¹⁴ Jones than the flexible OPDs fabricated with an ITO or a PEDOT:PSS TCE, due to a deeper work function level of the CNT. The reliability of the CNT- and ITO-based flexible OPDs were compared by monitoring the D* after the OPDs were stressed with a cyclic flex test for 500 bending times at a bending strain of 0.8%. The D* of CNT-based OPDs only reduces to 80% of its initial value, while the D* of ITO-based OPDs dramatically decreases to 20% of the initial value. It reveals that the CNT-based flexible OPDs have a higher detectivity and better mechanical stability than the ITO-based flexible OPD.

The conducting polymer PEDOT:PSS layer is one of the commonly used TCEs in solution-processable flexible OPDs.[98,99] To fabricate flexible OPDs using PEDOT:PSS as anode and cathode electrodes, the work function of the PEDOT:PSS cathode needs to be modified. A PEI interlayer was deposited on the PEDOT:PSS layer to modify the work function of the cathode in flexible OPDs.[100,101] The PEDOT:PSS deposited on PET substrates was used as the TCEs for fabricating large-area, semitransparent flexible all-polymer OPDs using a roll-to-roll lamination process.[102–104] The OPDs were stacked using two sample specimens of PET/PEDOT:PSS/PEI/organic layer and PET/PEDOT:PSS/organic layer (Figure 6b). The organic photoactive materials function as the adhesive. The result flexible PBDB-T:PNDI-FT10 (2:1)-based OPD shows a constant responsivity of 0.18 A W⁻¹, a D* of over 10¹¹ Jones upon illumination from PEDOT:PSS and PEDOT:PSS/PEI sides, and a remarkable average visible transmittance of 23.2%.

Flexible PM-type OPDs with an ITO or a thin Ag-based TCE have been demonstrated. A flexible UV narrowband PM-type OPD was demonstrated using the F8T2:ZnO photoactive layer on the ITO-coated flexible PET substrate.[105] Only a slight decrease of 5% in the D* of the OPD was observed after the tensile and compressive bending test for 100 repetitions. A flexible

Figure 6. a) The schematic detector architecture, and D* measured for the flexible OPDs with different anodes of ITO, PEDOT:PSS, and CNT. b) The roll-to-roll fabrication process, the spectral responsivity, and D* measured for the flexible OPDs with a PEDOT:PSS TCE. c) The schematic diagram showing the nanofiber OPT on a PET/PDMS textile composite substrate. d) The layout, and e) a photograph taken for the ultrathin flexible OPDs. (a) Reproduced with permission.[97] Copyright 2021, Elsevier. (b) Reproduced with permission.[103] Copyright 2018, John Wiley and Sons. (c) Reproduced with permission.[110] Copyright 2016, John Wiley and Sons. (d,e) Reproduced with permission.[114] Copyright 2018, John Wiley and Sons.
PM-type OPD was demonstrated by replacing the glass/ITO substrate with a flexible PET/ZEOCOAT/MoO3 (1 nm)Ag (9 nm)/MoO3 (2 nm) substrate. The flexible PM-type OPD shows about two times higher responsivity and better flexibility over the rigid OPD. The device could maintain 51.4% of its initial EQE after bending 1000 times. The progress of flexible PM-type OPDs is attractive for applications in wearable devices detecting weak light. For example, a wearable UV light monitor was demonstrated using a visible-blind UV-sensitive PM-type OPD in series with a green OLED.[74]

The mechanical stability or robustness is an essential factor in the lifetime of flexible OPDs. The mechanical stability of flexible OPDs is evaluated by cyclic stressing tests, including bending, stretching, and compressing treatment. The flexibility of a flexible OPT array was studied by monitoring the electron mobility after cyclic bending tests.[107] The electron mobility stayed unchanged after the OPT array was bent for 6000 cycles over a 10 nm bending radius, and started to decay when bent over a radius of 2.0 mm. The bending process has a significant effect on the lifetime of flexible OPDs, leading to an irreversible degradation in the performance of the OPD. The X-ray-induced photocurrent in a flexible photoco conductor-type OPD reduced by 50% after the first-round bending and reached a stable performance, supported by a stable photoresponse after bending 100 times.[108] Similarly, a dramatic reduction of dark current was observed in a flexible OPD due to the deformation or partial damage of the organic photoactive layers under a bending condition. It thus shows a lower dark current at the tensile bending state than compressive bending or flat conditions, regardless of in dark condition or under light illumination. The OPD was found to have a dual function: 1) it is a PD when the device is under compressive bending and flat condition, and 2) it acts as a flexion sensor when the device is under proper tensile bending.[109] The reproducible cyclic bending changes indicate the applying of the flexible OPD on human elbow joints, hands, or other parts to detect mechanical movement.

Various efforts have been made to improve the mechanical stability and flexibility of OPDs. A flexible nanofiber OPT, fabricated on a PET/PDMS textile composite substrate as shown in Figure 6c, has a stable photoresponse of 82.3% after a bending test with a bending radius of 0.75 mm for 1000 cycles.[110] On the contrary, the OPTs made on a planar PET substrate or a PDMS substrate have poorer mechanical stability, obtaining only 35.6% and 21.5% of photoresponse after the bending test. The textile substrate-based flexible OPDs are promising for applications in wearable electronics.[111] The flexible OPDs using different ETLs of Cs2CO3, PEIE, and Cs2CO3-doped PEIE have been investigated.[45] The OPD with a Cs2CO3-doped PEIE ETL shows a better mechanical bending stability, remaining 90% performance after 10 000 cycles of tensile bending. The excellent stability was attributed to the improved adhesion between the organic photoactive layer and the Al electrode.

The reduced thickness of the OPD devices is preferred to realize higher tolerance of flexible and stretchable deformation.[112,113] A few μm thick flexible OPDs have been reported. An ultrathin OPD with a total thickness of <3.0 μm was demonstrated using a 1.0 μm-thick polymeric SU-8 substrate and passivated by a 1.0 μm-thick parylene layer (Figure 6d).[114] The flexible OPDs can be attached to human skin for conformal PPG sensing (Figure 6e). The flexible OPDs were laminated on a polymer elastomer for the mechanical deformation test, in which deformation from 200% stretching to 20% compression was applied to the flexible OPDs. The short-circuit current (Isc) of the OPDs decreases by 33.6%, while the open-circuit voltage (Voc) maintains a steady value when the deformation states of OPD change from 200% stretching to 20% compression. Meanwhile, the response time of the OPDs was less affected by the deformation states. The ultrathin OPD also shows a stable performance after 1000 cycles of repetition bending testing. A self-powered flexible device was developed for wearable PPG monitoring by integrating an OSC power source, an OLED light emitter, and an OPD sensor unit on an ultrathin flexible substrate.[115] The ultrathin flexible OPDs are promising for wearable electronic applications. The environmental stability and mechanical robustness of the flexible devices can be further improved by a double-side coating with a plastic elastomer.[116] The flexible devices thus demonstrated show an improved stretchability and water stability compared to the free-standing devices, resulting in more stable optoelectronic performance.

4. Solution-Processable Fabrication Strategies and Printing Techniques

The manufacture of present crystalline inorganic PDs requires high-temperature and high-vacuum processes. In contrast, OPDs can be fabricated by solution-processable deposition techniques under low temperature and ambient conditions. The spin coating method has been widely used to fabricate OPDs for realizing superior efficiencies. However, spin coating is not ideally suitable for the high-throughput and large-area fabrication of OPDs. It is also challenging to directly deposit organic thin films onto nonplanar substrates using the spin coating technique.[117] The developments of reproducible and scalable manufacturing techniques are essential for the eventual commercial deployment of OPDs.

Tremendous research efforts have been made to develop cost-effective solution-fabrication techniques for making large-area OPDs.[118] This section provides an overview of the available solution-processable fabrication strategies, including printing techniques. A high-throughput printing process is favorable for depositing multilayer OPDs, enabling monolithic integration of OPDs with ROIC for mass production. An illustration of the various industrially relevant solution-processed deposition techniques is shown in Figure 7. Different printing technologies and the performance of the printed OPDs are presented in the following discussion.

4.1. Blade Coating

In the blade coating, also known as the knife coating, the setup mainly involves the coating knife, the syringe, and the coating board. First, the formulated ink is released through the gap between a coating knife and the target substrate. Next, the coating knife travels with a desired speed across the substrate, spreading the deposited ink to form a uniform wet film. The substrate is usually heated to assist the removal of the excess solvent in the wet film. Appropriate processing parameters, e.g., the
moving speed of the coating knife, the volume flow rate of the syringe pump that releases the ink, the annealing steps, and the temperature of the substrate, can be regulated for controlling the roughness and morphology of the blade-coated thin films. Blade coating has the advantages of rapid thermal processing and low material consumption. It is suitable for coating large-area thin films with well-controlled morphology and uniformity.

Blade-coated OPDs on a plastic substrate have been reported. The blade coating method was used to deposit the functional layer in the OPD, including the bottom PEDOT:PSS electrode, PEIE interlayer, and organic photoactive layer. The work function of the PEDOT:PSS/PEIE electrode can be tuned by adjusting the concentration of PEIE ink to improve the hole blocking properties and reduce the dark current of the OPDs. The thickness and morphology of the photoactive layer were well controlled in the blade coating process to attain high OPD performance and low device variability. The results indicate that the blade coating technique is favorable for preparing multilayer OPD. The blade coating was applied to create an organic photoactive layer with gradient thickness from 100 to 200 nm. A set of microcavity-resonant NIR OPDs was fabricated to achieve wavelength-tunable photodetection, resulting in a miniaturized spectrophotometer.

4.2. Screen Printing

Screen printing allows 2D patterning and can be applied to deposit relatively thick films, e.g., with a thickness up to several hundred micrometers. The screen printing process for OPDs can be divided into several steps: 1) the printing inks are poured onto the screen; 2) printing inks are transferred to the substrate by the moving of the squeegee; and 3) the patterning deposition is realized by openings of the screen. The patterning deposition of Ag interconnects and dielectric layers in a flexible OPT array was demonstrated by screen printing. Flexible OPD arrays were constructed by screen coating the top PEDOT:PSS electrode. The all-printed OPDs exhibit a comparable performance as the OPDs with thermally evaporated metal electrodes. The dark current of all-printed OPDs is lower than the OPDs with evaporated Au anode. Though significant progress has been made in screen printing OPDs, the technique still faces some challenges, e.g., poor performance on high-viscosity ink, screen mesh size, adhesion/delamination issues, using solvents with a low evaporation rate, and postdeposition annealing treatment.

4.3. Spray Coating

Spray coating deposits thin film by utilizing pressurized gas. The spray coating includes several independent steps: liquid atomization, atomized droplet transport, and deposition. It has the advantage of high-volume production with only minimal loss of the formulated ink. The quality of the thin films can be regulated by several critical process parameters, e.g., the size of the atomized droplets, the distance between the spray nozzle and sample substrate, flow rate, and substrate conditions. The desired thickness can be controlled by the spray duration and number of spraying cycles. The 2D patterning can be realized by spray coating technique coupling with a mask. It can also be applied to fabricate morphologically uniform films on a curved or a 3D surface.

Fully sprayed OPDs with transparent CNT electrodes on flexible PET substrates have been reported. The wettability of the CNT/PEDOT:PSS inks has been extensively studied, resulting in a high process yield above 90% by using IPA-dilute PEDOT:PSS ink. The spray deposition condition has been optimized to obtain a uniform deposition of the active layers and enable fine-tuning of the film thickness. The sprayed P3HT:PCBM OPDs have a low J<sub>d</sub> of 10<sup>−4</sup> mA cm<sup>−2</sup> and an EQE of 65%. The performance of the spray-coated P3HT:PCBM OPDs has been improved through tuning the thickness of the photoactive layer and adjusting the thermal annealing process. The spray-coated OPDs, operated under a reverse bias of −1.0 V, have a low J<sub>d</sub> of 2.90 × 10<sup>−8</sup> A cm<sup>−2</sup>. It reveals that the control of large PCBM clusters in BHJ with a phase separation between donor and the acceptor in the active layer plays a significant role in suppressing the dark current and improving the photocurrent of the
sprayed OPDs. A spray-coated large-area OPD image sensor prepared on an amorphous silicon (a-Si) thin-film field-effect transistors (TFT) backplane containing 256 × 256 pixels has been demonstrated.\[135\] The robust spray coating process led to low dark currents and high responsivities in the large-area OPDs by reducing the probability for pinholes and shunt paths. The progress suggests that spray coating technique is promising for depositing homogenous photoactive layers for high-resolution and large-area image sensors.

### 4.4. Inkjet Printing and Aerosol Jet Printing

In general, inkjet printing is primarily a digital printing technique, which is promising for making OPDs, due to its advantages of low material usage, cost-effectiveness, and large-area formation. In this method, formulated functional ink is delivered to the nozzles of a print head from the ink reservoir and then ejected in the form of microdroplets via thermal or piezolectric actuators. The ink can be directly deposited in the desired locations on the substrate without applying the physical masks. The precise control of the droplets and deposition parameters in the inkjet printing has achieved a coating resolution of tens of micrometers.

Aerosol jet printing is similar to inkjet printing. The difference is that the formulated ink is released as an aerosol state in aerosol jet printing. The aerosol of the ink is usually generated by ultrasonication or by pneumatic atomization. Aerosol jet printing is a direct-write and single-point deposition approach, offering the digital, vacuum-free, and noncontact method for depositing a fine mist (or aerosol) on the substrate. The main processing parameters of aerosol jet printing, e.g., drop shape, drop size, ejection speed, and ejection frequency, could be adjusted for obtaining high-quality uniform and dense thin films.

A PCDTPT:PC\(_{70}\)BM-based NIR OPD, showing a spectral detection range of up to 1.1 μm and a \(D^*\) of \(\approx 3.2 \times 10^{10}\) Jones at 950 nm, was fabricated by an inkjet printing method. The thickness of the photoactive layer could be tuned via controllable layer-by-layer printing deposition.\[124\] The aerosol jet printing technique was used to deposit the electron blocking layers on the top of the organic photoactive layer in OPDs without requiring orthogonal solvents or cross-linking treatment of the underlayer.\[154\] Fully printed nonfullerene acceptors (NFAs)-based OPDs were demonstrated by inkjet-printing the ZnO and organic photoactive layer, and aerosol-jet-printing the conductive PEDOT:PSS electrode. The digitally printed OPDs have achieved excellent responsivities up to 300 mA W\(^{-1}\) in the visible and NIR spectrum.\[125\] An all-printed flexible OPD was developed using the aerosol jet printing technique. The 1.0 mm width of bottom and top PEDOT:PSS electrode strips has been achieved in the printing process, realizing an active area of 1.0 mm\(^2\).\[130\] The printed semitransparent OPD comprising two PEDOT:PSS electrodes exhibits a responsivity of 0.178 and 0.204 A W\(^{-1}\) with top illumination and bottom illumination. The various processing methods and combinations of different hybrid material/solvent systems allow high freedom in manufacturing flexible OPDs.

The quality of the printed functional layers is related to the ink formulation, substrate treatment conditions, and postdeposition processing. For example, the inks with different concentrations usually show different viscosity, surface tension, and drying rates. The ink formulation is one of the key factors that determine the performance of the OPDs. A color-selective OPD was inkjet-printed using an ink comprising a blend of a wide-bandgap polymer donor and a narrowband-absorbing NFA.\[31\] The spectral responsivity of the printed OPDs is governed by the absorption spectra of the NFAs, IDFBR, and ITIC-4F, achieving blue- and red-selective responses. The use of different solvents, e.g., CB, DCB, and chloroform, is important in the ink formulation. It was found that the inhomogeneous drying process in printing the DCB-based ink can be improved by adding mesitylene as a second solvent.\[31\] The additive, e.g., 3% DIO, in the ink also improves the morphology and crystallinity of the printed organic photoactive layers by slowing the drying process.\[127\]

### 4.5. Stamping and Transfer Printing Processes

The transfer printing includes several steps: 1) the target thin film is prepared on a media substrate, e.g., Si wafers; 2) the soft stamp is used to grab target thin film off from the media substrate; 3) the stamp with the target thin film is attached to the receiver substrate; and 4) the stamp is removed from the substrate, remaining the target thin film onto the receiver substrate.\[128\]

Compared to the OPDs with a spin-coated photoactive layer, the OPDs with a transfer-printed photoactive layer have shown improved stability. In the desirable solvent-free condition, the transfer process produces the photoactive layer with uniform and condensed morphology, thus enhance interfacial stabilization and suppress the burn-in loss of internal resistance.\[129\] A bilayer interdiffused heterojunction OPDs was fabricated using the transfer printing process. A P3HT layer was transferred on the PCBM coated substrate, followed by the interdiffused heterojunction formation upon thermal annealing. The OPD with an interdiffused P3HT/PCBM photoactive layer shows a low \(J_d\) of \(\approx 7.7\) nA cm\(^{-2}\), an EQE of \(\approx 60\%\), and a peak \(D^*\) of \(4.8 \times 10^{12}\) Jones.\[138\] The donor polymer, P3HT, was deposited as an electron blocking layer in OPDs to reduce the dark current using the transfer stamping process.\[38,130\] A smooth and compact organic layer was aligned on the top of the photoactive layer without contaminating the underlayer. The results confirm that the dry transfer printing technique is favored for constructing organic photoactive multilayer in OPDs.

Recently, a high-performance transparent self-powered NIR OPD was demonstrated using a PEDOT:PSS TCE, prepared by a lamination-transfer process.\[98\] The transparent self-powered NIR OPD exhibited a \(D^*\) of \(>10^{12}\) Jones at 850 nm, a high \(f_{-3dB}\) of 71 kHz, and an LDR of 154 dB. The dry transfer printing of the electrode layer offers a new opportunity to depositing an organic layer, polymer interlayer, and polymer TCEs. However, transfer processing is not applicable for viscoelastic receiver substrates because it is processed by the differences in surface energy between the stamp and the receiver substrates, limiting the selectivity of the receiver materials.\[131\]
5. OPDs for Applications in Flexible and Wearable Electronics

OPDs have attracted significant interest in flexible electronics due to the advantages of broad spectral response, mechanical flexibility, fabrication simplicity, and integration compatibility. Replacing the conventional inorganic PDs with OPDs represents a promising strategy toward cost-effective, lightweight, and flexible electronics. The solution process also enables OPDs to be easily integrated with the ROIC and other electronic components. This section discusses the perspectives of applying OPDs in future flexible and wearable electronic devices, e.g., wearable health self-monitoring devices, flexible optical communication systems, and large-area image sensors.

5.1. Wearable Health Self-Monitoring Devices

PDs are essential components in healthcare monitoring systems such as heart-rate measurement, pulse-oximetry sensor, and transcutaneous oxygen monitor. For example, OPDs have been used in noninvasive and low-cost PPG sensor technology. The PPG sensor detects the change in the volume of the blood flow in the microvascular bed of tissue, e.g., from fingers. The PPG signals are measured by monitoring the changes in the light intensity using OPDs. The PPG sensors can be designed in a reflection mode by aligning the light source and OPD side by side, or a transmission mode by putting the light source and OPD on two sides of a finger. An NIR PPG sensor was demonstrated using OPD for real-time heart-rate monitoring (Figure 8a).[36] The intensity of light transmitting the finger varies with the volume change in the blood vessels. Therefore, the photocurrent characteristics of the NIR OPD directly reflect the heart-beat waveform. The accuracy of PPG sensors has also been improved by matching algorithms to reduce noise issues.[136] The PPG sensor, equipped with the heart rate variability algorithm, can be integrated with machine-learning methods (e.g., Random Forest) to detect drowsiness in real-life applications.

As the elderly population grows, real-time and long-term health monitoring is preferred to improve the health condition of the generation. Wearable PPG sensors equipped with flexible electronics are desired. An epidermal NIR PPG sensor was developed by integrating a low-power flexible OPT and inorganic LED on a PDMS substrate (Figure 8b).[137] The flexible PPG sensor could be wrapped around the finger to reduce discomfort for real-time heart rate measurement. In addition, flexible OLEDs are available as the light source for PPG sensors and provide better mechanical conformability for skin-mounted applications. An all-organic flexible PPG sensor combining a red OLED and an OPD has achieved similar performance to a conventional rigid PPG sensor.[136] The geometric designs of the OLED and OPDs were optimized to improve the performance of PPG sensors. As shown in Figure 8c, three different sensor geometries, e.g., rectangular, bracket, and circular, were explored to acquire high-quality PPG signals and improve sensor performance.[138] Similarly, an “8” shaped OPD was constructed to wrap around two OLED light sources in a flexible PPG sensor.[139] The improved light collection approach enables the PPG sensor to operate at a low electrical power of tens of microwatts. The solution-processable OPDs offer full freedom to geometric designs to address the needs of specific applications. A self-powered PPG sensor was proposed by integrating OLED, OPD, and an OSC.

![Figure 8](https://example.com/figure8.png)

Figure 8. a) Mechanisms of a PPG data acquisition and a photography taken for the PPG sensor, b) Schematic process flow of fabricating an epidermal and flexible NIR PPG sensor, c) Schematic diagrams of reflectance PPG sensors with different layouts, d) Schematic diagram of self-powered PPG sensor attached on the human hand. (a) Reproduced with permission.[36] Copyright 2020, John Wiley and Sons. (b) Reproduced with permission.[137] Copyright 2017, John Wiley and Sons. (c) Reproduced under terms of the CC-BY license.[138] Copyright 2019, The Authors. Published by IEEE Access. (d) Reproduced under terms of the CC-BY license.[115] Copyright 2021, The authors. Published by Springer Nature.
energy-harvesting power source (Figure 8d). The OSC module with ten series connections has an active area of 4.5 cm² for providing a power source for the PPG sensor.

The PPG sensors have been used to measure arterial blood oxygen saturation (SpO₂), also called pulse oximeters. SpO₂ is the ratio of oxygenated hemoglobin (HbO₂) concentration to the total hemoglobin (Hb) concentration. It is defined as the percentage of oxygen saturation in the arterial blood. In general, traditional pulse oximeters employ two LEDs at different wavelengths (e.g., 660 and 940 nm) and a broadband PD to measure the actual difference between the absorption spectra of Hb and absorption spectra of HbO₂ [140]. The narrowband emission of OLEDs and visible-to-NIR broadband detection of OPDs make them ideal for fabricating wearable pulse oximeters. An all-organic transmission-mode pulse oximeter was developed by combining a green (532 nm) OLED, a red (626 nm) OLED, and a broadband OPD (sensitive from 450 to 750 nm) [141]. Compared to the inorganic sensor, negligible 1% error for pulse rate and 2% error for oxygenation were obtained by the all-organic pulse oximeter. A flexible reflection-mode pulse oximeter was developed by integrating green and red OLEDs with an OPD [112]. The on-skin pulse oximeter, including the substrate and encapsulation layer, has a total thickness of around 3.0 μm, which minimizes the discomfort associated with wearable electronics. A flexible organic oximeter array has been demonstrated to replace the single-point SpO₂ sensor for obtaining accurate and stable SpO₂ signals without being affected by the motion artifacts. Nine oximeter units, each comprising an array of 4 x 4 integrated OLED/OPD sensors, were used to measure the oxygen saturation signals on the human forehead and provide a 2D forearm oxygenation map. The 2D mapping capability of this array makes it possible to use as novel sensing schemes for various locations, e.g., tissues, skin grafts, wounds, and transplanted organs [142]. Similarly, an organic oximeter comprising an array of 16 x 16 OPD pixels was developed for monitoring accurate PPG waveform signals [143].

Another possible medical application for OPDs is to measure the tissue oxygen tension or oxygen partial pressure (pO₂) at the skin surface, which is crucial for achieving accurate diagnostic results, e.g., burns, limb injury, and surgical interventions. Based on the luminescent gas sensing mechanism, a novel wearable bandage-like sensor was demonstrated to monitor the transcutaneous oxygen pressure (tcpO₂) [144]. Various functional components, e.g., an oxygen-sensing film, an encapsulation layer, an array of micro-LEDs, an optical filter, an OPD, and an optical isolation layer, were integrated on flexible substrates for developing a wearable bandage-like tcpO₂ sensor, which can be attached on different parts of the body [145]. As more breakthrough emerges, the OPD is believed to become a practical wearable optic-based component for next-generation accurate diagnostic evaluation, e.g., blood pressure, cardiac output, and arterial aging.

5.2. Flexible Optical Communication Systems

Optical communication is an advanced optical wireless communication technology, which has less health concern and a broader bandwidth. An optical transmitter and a PD receiver are required for free-space communication links. OPDs have attracted much attention in replacing the traditional inorganic PDs in numerous impressive applications, e.g., optical communications, IoT, light-fidelity (Li-Fi), positioning systems, and automatic driving cars.

Figure 9. a) Schematic diagrams illustrating an experimental VLC setup comprising a white LED and a self-powered OPD. b) The photograph taken for an all-organic flexible VLC system. c) The operating mechanism of a VLC system comprising the narrowband OPDs and RGB OLEDs. d) Demultiplexing dual-channel optical communication using two color-selective OPDs. (a) Reproduced under terms of the CC-BY license. Copyright 2018, The Authors. Published by IEEE Access. (b) Reproduced under terms of the CC-BY license. Copyright 2018, The Authors. Published by MDPI. (c) Reproduced with permission. Copyright 2016, Elsevier. (d) Reproduced under terms of the CC-BY license. Copyright 2020, The Authors. Published by John Wiley and Sons.
OPD receivers are desired due to low fabrication cost, sizeable optical detection area, and inherent compatibility with flexible substrates. The existing light sources, such as LEDs, can be used as a light transmitter in visible light communication (VLC). A VLC system comprising a commercial white LED as a light transmitter and a visible OPD receiver was proposed (Figure 9a). The P3HT:PCBM-based visible OPD with an optical detection area of 9.0 mm² shows a modulation bandwidth of 790 kHz, and a responsivity of 0.18 A W⁻¹ without external bias for receiving a real-time audio signal. The self-powered OPD is promising for an energy-saving system. The use of lightweight and flexible OPDs offers new opportunities for the services of VLC in indoor environments, e.g., adequate indoor pedestrian support systems, commercial products integration. A flexible Li-Fi system, comprising a flexible white OLED, a flexible P3HT:PCBM-based OPD, and flexible printed circuit boards, was developed (Figure 9b).

The bandwidth of present optical communication is limited by the slow response speed in OPDs. In general, the response speed of OPDs is limited by device area and the low carrier mobility of organic semiconductors. Several approaches have been proposed to boost the response speed of OPDs, including improving the mobility of materials, balancing electron/hole mobility, and reducing the RC time. Several high-speed OPDs have been reported, showing a bright future in optical communication. The inject printing nonfullerene OPDs, having a device area of 1 mm², have shown f_{3dB} of 1.5 and 3.5 MHz. A nonfullerene OPD (1.0 mm²) shows a high NIR responsivity of 0.4 A W⁻¹ and f_{3dB} of >4 MHz when operated under a reverse bias of -2.0 V. The f_{3dB} of the OPD is limited by the transit time since an f_R of 9.0 MHz has been evaluated in the OPD with a photoactive layer thickness of 200 nm. An f_{3dB} of 2.38 MHz and response speed of 146.8 ns were realized in a tandem OPD. The tandem structure of OPDs allow an increase in the thickness of the photovoltaic layer to absorb more light and reduce dark current, while the RC time could be restrained at a low level. In this way, both the carrier transit time and RC time of the tandem OPD are reduced, resulting in a fast response speed. Furthermore, a predistortion scheme of pulse-amplitude-modulation signal was applied for improving the data rate of the OPD receiver. A 5 times improvement of data rate, increased from 25 to 125 kbit s⁻¹, was realized using the identical OPD receiver, thereby enhancing the VLC performance.

The spectral demultiplex using spectral selective OPDs is promising for filter-free flexible optical communication. A filter-free band-selective OPD was used in multichannel optical communication. Accurate NIR light information was received by the visible-blind NIR-selective OPD, revealing the potential for encryption communication. An all-organic two-channel VLC system was demonstrated using a red light-sensitive OPD and a blue light-sensitive OPD (Figure 9c). The two-channel VLC system has shown a direct demultiplexing function when receiving optical signals with different spectral information. Two spectral selective OPDs have been realized for filter-free optical communication (Figure 9d). The photostabilized layer was prepared by blending transparent donor (P1F) and narrowband-absorbing NFAs (IDFBR and ITIC-4F). The photosresponse of the OPDs is determined by the absorption of NFA materials in the photoactive layer. A 2 × 2 OPD array with selective responsivity was fabricated by inkjet printing. The results suggest the future application of filter-free OPDs receivers and white RGB array OLED light transmitters for multichannel indoor applications such as Li-Fi.

5.3. Flexible Large-Area Image Sensors

Rapid progress has been made in large-area image sensors for a diversity of applications, including machine vision, environmental surveillance, and NIR medical imaging. Benefitting from their tunable optoelectronic properties, OPDs are widely developed for imaging from UV, visible, NIR, to SWIR ranges. A highly responsive visible organic image sensor was developed using the PM-type OPD. A high responsivity of >40 A W⁻¹, a low dark current, and a high rectification were obtained in the PM-type OPD with an additional rectifying layer. SWIR OPDs offer versatile characteristics delivering additional information that visible or NIR OPDs could not detect. A high-performance SWIR OPD having a peak EQE of 30% and a D* of 10¹¹ Jones at 1100 nm was achieved by improving exciton dissociation. A spectroscopic image sensor comprising an array of 4 × 4 SWIR OPD pixels was demonstrated to analyze the difference between the muscle and fat in animal tissue. A flexible SWIR OPD array was successfully fabricated on a PET substrate to detect spatial light intensity distribution.

X-ray imaging has been significantly applied in various applications, e.g., medical imaging, nondestructive testing in industrial products, and public security inspection. The detection of X-rays using OPDs is realized by indirect-conversion X-ray detectors combining scintillator and visible OPDs. The scintillators are activated by the X-ray excitation and emit visible light. The spectral responsivity of OPDs is designed to match the luminescence from scintillators. The conventional X-ray detector has limitations for application in curved sensors due to its rigidity nature. A large-area active-matrix X-ray detector comprising an array of 32 × 32 solution-processed OTFT/OPD sensors was demonstrated. The solution-processed OPDs with Jfs of 1.0 pA mm⁻² and responsivity of 0.2 A W⁻¹ were prepared on a 25 µm-thick plastic substrate for indirect X-ray imaging. A flexible X-ray detector, fabricated by integrating the printed OPD with a plastic scintillator, enables 2D spatial detection and self-powered operation. As shown in Figure 10b, a plastic foil-based curved digital X-ray image sensor was developed for approaching cone beam computed tomography X-ray imaging. The curved X-ray image sensor has a large area of 6.0 cm × 8.0 cm size and contains 480 × 640 pixels. A full 3D volume image of a piece of bone was successfully captured using the curved X-ray image sensor, demonstrating the promising future of using curved X-ray image sensors in medical applications.

The OPD-based image sensors can be prepared through monolithic integration of the detectors with the ROIC without using the lithography processes. The available ROIC backplanes include the CMOS, a-Si TFT, OTFT, indium gallium zinc oxide (IGZO) TFT arrays. To fabricate the organic image sensor, the organic photoactive layer was directly deposited on the top surface of a CMOS backplane.
in the CMOS act as the bottom electrode for the OPDs. The photoresponse generated in the OPDs could be read out by the CMOS circuit via the Al electrode. The pixel size (active area of the OPD) is then defined by the Al pad (80 μm × 80 μm) on the top of the CMOS substrate. An NIR image sensor was developed by integrating the NIR-sensitive organic–QD composite photoactive layer with a commercially available a-Si active-matrix TFT backplane (Figure 11b).[169] The a-Si TFT substrate was coated with a prepatterned ITO layer to be the bottom electrode for the lateral OPD. The subsequent deposition of PEDOT:PSS and the photoactive layer was processed with spin coating technique. The result image sensor has an array of 256 × 256 pixels and an aperture ratio of 83.3%. The horizontal carrier mobility of the organic layer is much lower than that of crystalline semiconductors, minimizing the electric crosstalk between pixels. The distance between pixels can be reduced while

**Figure 10.** a) The photograph taken for an X-ray imager comprising an array of the OTFT/OPD sensors. b) The photograph taken for the curved digital X-ray detector comprising the solution-processed OPDs and an IGZO TFT backplane. (a) Reproduced with permission.[162] Copyright 2013, Elsevier. (b) Reproduced under terms of the CC-BY license.[165] Copyright 2020, The Authors. Published by Springer Nature.

**Figure 11.** Schematic diagrams of a) OPDs integrated on the top of a CMOS backplane, b) OPDs integrated on the top of an a-Si TFT ROIC, c) an imaging passive pixel sensor circuit comprising an OPD and a top-contact OTFT, d) an image pixel comprising a PBDDTTT-C:T-PC71BM OPD and a top-contact OTFT. (a) Reproduced under terms of the CC-BY license.[168] Copyright 2020, The Authors. Published by Springer Nature. (b) Reproduced with permission.[169] Copyright 2009, Springer Nature. (c) Reproduced with permission.[171] Copyright 2011, Elsevier. (d) Reproduced with permission.[172] Copyright 2016, AIP Publishing.
the crosstalk remains small. It is believed that the designs of the bottom electrodes in the ROIC substrate can further improve the aperture ratio of an organic image sensor. A large-area optical fingerprint sensor, having a total area of 3.2 cm × 2.4 cm and a resolution of 508 ppi, was demonstrated.\cite{170} The image sensor consists of an a-IGZO TFT backplane as ROIC and slot-die-coated PCDTBT:PCBM-based OPDs as sensing units. The solution-processed monolithic integrated OPDs show excellent performances, e.g., a $J_A$ of 1.0 × 10^{-7} mA cm^{-2}, a broad LDR, and a high $D^*$ of 7.1 × 10^{13} Jones. The monolithically integrated all-organic designs of image sensors have been extensively investigated. Usually, the OTFT matrix is constructed to replace the CMOS readout backplane. As shown in Figure 11c, an organic passive pixel image sensor was made via integrating a bottom-gate, top-contact pentacene OTFT and a top-illuminated OPD.\cite{171} A similar design of image pixel comprising a PBDDTTT-C-T:PC71BM OPD and a top-contact p-type OTFT with an Al_{2}O_{3}/SAM dielectric layer was reported (Figure 11d).\cite{172} A flexible image sensor was developed by integrating OPDs with the OTFTs addressing matrix on a PEN substrate through printing deposition.\cite{173}

The color sensing functions are challenging for the present flexible image sensors due to the lack of flexible color filters. A fully printed flexible OPD array with RGB full-color imaging capability was reported.\cite{174} The flexible OPD having a broadband PVD4650:PC_{70}BM photoactive layer could detect broadband light covering the whole visible range. The pixel detecting yellow and red light was achieved by color filters made of organic films of F8BT:PC_{60}BM and PCDTBT through spray coating. Therefore, a flexible image sensor comprising 32 × 32 array of 1.0 mm × 1.0 mm sized OPD pixels, with average color accuracy of 98.5%, was successfully demonstrated. A full-color image sensor was demonstrated using a green selective photoactive layer on the CMOS backplane to act as a green light detection unit and green-light-blocking filters.\cite{166,167} The blue and red color detection were realized by the bottom CMOS sensors. The full-color image sensor has been developed by vertically stacking three organic photoactive layers, each with a set of specific readout circuits.\cite{175,176} In this design, full-color detection can be realized by the single pixel in the image sensor, achieving a high spatial accuracy and high resolution. The device requires narrowband photoactive layers and a complex design of ROIC. The complex circuit integration is less favored in the monolithic process of the image sensor. Recently, we have demonstrated an electrically switchable color-selective OPD for the future full-color image sensor. Selective blue detection and green-to-red detection can be switched by adjusting the OPD bias.\cite{79} An electrically switchable multispectral selective OPD is ideal for full-color imaging. The properties allow the identification of different colors by addressing the signals from the OPD array with one set of ROIC, offering a more straightforward image sensor design.

The fabrication adaptability of OPDs enables monolithic integration of NIR OPD with visible LED for NIR visualization through NIR-to-visible upconversion.\cite{177,178} The upconverter image sensor has been introduced in a previous review article.\cite{179} The present review discusses the recent progress made with the upconversion devices prepared through monolithic integration of the OPD and the LED units using the solution-fabrication method. In the upconverters, the OPD unit and LED unit are connected in back-to-back series (Figure 12a).\cite{133} The upconverter is operated for forward-biasing the LED unit and reverse-biasing the OPD unit. The current flow in the LED unit is blocked by the OPD unit when it is dark. In the

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**Figure 12.** a) Schematic diagrams showing the configuration of an upconversion device and the equivalent circuit. b) Schematic diagram illustrating the NIR visualization setup using the upconverter. c) The optical readout image of a fingertip and blood vessels from a human hand. d) The electronic readout waveform of the PPG signals. (a–d) Reproduced with permission.\cite{133} Copyright 2021, John Wiley and Sons.
presence of NIR light, a photocurrent is generated in the OPD unit, driving the LED unit to emit visible light. Pixelization is no further needed in upconverter image sensors because only the region that receives NIR light could emit visible light. The low horizontal carrier mobility in organic semiconductors enables a high resolution of 400 or 600 dots per inch in all-organic upconverters.\(^{[180,181]}\) The upconverter has been used to visualize the blood vessels under the skin offering an optical readout image and an electronic readout of the heartbeat waveform signals (Figure 12b,c,d).\(^{[113]}\) The integration strategy of upconverters allows high freedom for configurational designs. The upconverters based on an OPD/OLED and an OLED/OPD designs have been studied, where either common anode or common cathode was used for connecting the OPD and OLED units.

The two different upconverters show comparable photon-to-photon conversion efficiency.\(^{[182]}\) Solution-processable upconverters have been developed by integrating NIR OPD and perovskite LED. The interconnection layer made of PEDOT:PSS prevents the erosion to the organic photoactive layer during the solution deposition of perovskite mitters.\(^{[112,183]}\) The emission color of upconverters has been tuned across the entire visible range by incorporating red, green, and blue OLED units.\(^{[184]}\) An upconverter with a tandem OLED unit was fabricated, showing a higher photon-to-photon conversion efficiency of 29.8\% compared to 8\% from the upconverter with a single OLED unit.\(^{[185]}\)

6. Conclusion and Outlook

This review discusses the recent progress in high-performance OPDs, including filter-free spectral selective photodetection, large-area solution-fabrication technologies, and the applications in different flexible electronic devices. In terms of OPD performance, the suppression of noise current is critical for attaining the high \(D^*\) of OPDs. Thus, the recent progress made in achieving low-noise OPDs is discussed. Recent efforts to achieve filter-free spectral selective OPDs are highlighted, e.g., using CCN effect, employing heterostructure for realizing internal light depletion, and PM-type narrowband detection in the OPDs. In particular, the proposed filter-free spectral selective OPD configuration offers an effective way for applications in flexible narrowband photodetection with facile integration and tunable responsive spectrum. To address the scale-up of the OPDs from laboratory to large-scale industrialization, solution-based printing techniques for fabricating large-area OPDs with merits, e.g., high efficiency, high throughput, and low cost, have been introduced. Finally, we outline the future of flexible and wearable electronic devices for different applications, e.g., healthcare, optical communication, and imaging.

The solution-processable flexible OPD technologies provide a significant cost-benefit and a functional superiority over the wafer-based PD techniques. However, the long-wavelength photodetection and lifetime of the solution-processable OPDs are not yet good enough for commercial viability. To optimize the performance and lifetime of the OPDs, it is important to develop low-bandgap organic photoactive materials, understand the charge transport properties under various operation conditions, develop solutions to suppress the noise current, and enhance the operation stability. Below is an outlook for the future development of OPDs.

6.1. Infrared OPDs

The organic semiconductors have the advantage of tunable bandgap through engineering the molecular structures. The development of narrow bandgap organic semiconductors and infrared OPDs is attractive to various biomedical imaging applications and optical communication. However, most of the reported OPDs respond to light with a wavelength <1000 nm, which is usually in the visible and NIR range. The development of long-wavelength infrared OPDs is still in the early stage, while the present infrared OPDs are limited due to the high noise current and a limitation in achieving high responsivity in the long-wavelength range.\(^{[30,186]}\) The exciton dissociation in narrow bandgap organic semiconductors is hindered by the exciton binding energy, limiting the photodetection efficiency. A dielectric screening strategy has been proposed to reduce the exciton binding energy to improve exciton dissociation, thus improving the EQE of a SWIR OPD to 30\% at 1100 nm and expanding the response edge to 1400 nm.\(^{[160]}\) The PM process has been utilized to enhance the responsivity to infrared light with a wavelength of >1000 nm. However, reducing dark current in the PM-type infrared OPDs remains challenging before achieving a high \(D^*\).\(^{[187]}\)

The improvement of infrared OPDs requires effort in advancing materials and device structure. The full understanding of design rules of organic semiconductors with a very small bandgap remains a challenge.\(^{[15]}\) An open-shell donor–acceptor conjugated polymer was recently used as a photoactive layer to absorb infrared light with small energy. The polymer-based conductor-type OPD shows a broadband response in the entire infrared range.\(^{[188]}\) Further attention should be paid to molecular engineering, trap states suppression, morphological control, and interfacial engineering to realize high \(D^*\) in infrared OPD. The PM-type infrared OPDs also remain an open topic in the field. The spatial distribution of the trap states in the device should be carefully controlled to improve responsivity and reduce dark current for attaining high-\(D^*\) infrared OPDs.\(^{[189,190]}\)

6.2. Device Integration through Solution-Fabrication Processes

Solution-processed technologies come with several exceptional advantages, e.g., low unit costs, large-area manufacturing, and high-volume rates of production, when compared with vacuum-processed technologies. They have also been favored for low-temperature processing on flexible plastic substrates. Meanwhile, solution-processed technologies have limitations, e.g., patterning resolution, morphological control, and interface erosion issues of different stacked layers. Few all-solution-processed examples of organic electronics have been reported. The full printing of the bottom electrode, organic photoactive layers, interlayers, and top electrodes have rarely been realized. An all-solution integration process is a promising fabrication approach for next-generation electronic systems. To date, the printable high-performance flexible OPDs are still in the development stage. The full-printing ROIC is another challenge, requiring a
high patterning resolution and reliability of the printing techniques. The future improvement of printing techniques involves improving thin film morphology, patterning resolution, in situ constructing multilayer structure, and the digital printing process.

6.3. Stability of Solution-Processable OPDs

The stability of OPDs is the prerequisite for commercialization. For example, the long-term reliability of OPDs is limited due to the decrease in responsivity.[129,191] The deterioration in the performance of OPDs is a complex process associating with the intrinsic degradation of the functional materials, change in morphology properties of the functional layers, thermal instability, light-induced degradation, electric stress, moisture, and oxygen encroachment due to the imperfection of the encapsulation.[192,193]

Various encapsulation methods, e.g., glass encapsulation employing thermally or optically curable resins, thin-film encapsulation, have been applied to prevent the penetration of moisture and oxygen.[194] The encapsulation techniques of rigid OPDs might not work for flexible devices. The encapsulation of flexible OPDs is still an open challenge. The vacuum technologies have been commonly used to deposit high-quality inorganic thin-film encapsulation layers, e.g., employing two different metal oxide materials, the aluminum oxide (Al2O3)/silicon oxynitride (SiON) bilayer for the thin-film encapsulation.[195] The Al2O3 and SiON layers were deposited by atomic layer deposition (ALD) and plasma-enhanced chemical vapor deposition (PECVD), respectively. Low-cost solution processing encapsulation is a promising alternative technology to bridge the flexible OPD design and solution-based fabrication. For example, the seamless organic–inorganic hybrid multilayer, e.g., PDMS/SiOx/SiNx/SiOxNy structure,[196] PDMS/SiOx alternating layer,[197] and PDMS/Al2O3 nanolaminates,[198] have the potential for use in hermetic sealing of the large-area flexible OPDs. A solution-processable encapsulation using a parylene layer deposited by chemical vapor deposition (CVD) after the deposition of the top electrode was reported.[112-114] The flexible OPD had excellent storage stability up to ≈100 000 min under ambient air conditions, e.g., with a temperature of ≈20°C and humidity of 30% RH.[115]

One should note that unlike OSCs and OLEDs operating under a high current flow in the devices, the OPDs are usually operated under mild conditions, such as room temperature, weak light condition, and low humidity. The intrinsic instability of the OPDs should be improved. The fullerene acceptors, e.g., PC61BM, PC71BM, and bis-PC61BM, have intrinsic photochemical instability such as photooxidation degradation. Recently, OPDs made from NFAs have shown significantly improved stability due to a stable morphology of photoactive later.[198] Besides, machine-learning approaches, e.g., kernel ridge regression,[199] Random Forest regression,[199,200] decision tree,[200,201] and sequential minimal optimization regression,[202] have been demonstrated for predicting efficiency, stability, and significant attributes of devices. The machine-learning methods can be applied to assist the time-consuming experimentation and optimization in the development of stable OPDs.

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Conflict of Interest

The authors declare no conflict of interest.

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coating techniques, flexible electronics, organic photodetectors, printing, solution-processable semiconductor materials, wearable electronics.

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