

Silicon-Rivalling Large-Area Flexible Broadband Organic Photodetectors

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Organic photodetectors (OPDs) have demonstrated significant advantages as a promising next-generation photodetection technology, especially for application in flexible and curved image sensing. However, there is still a big gap in device performance between OPDs and commercial silicon photodiodes, which hinders their practical application. Herein, using a novel near-infrared electron acceptor featuring acceptor–donor–acceptor molecular skeleton and electron-withdrawing cyano-substituted terminal groups, the study reports an OPD that can fully compete with the commercial silicon photodiodes. Benefiting from the strong electron push-pull effect and large molecular dipole moment, the OPD exhibits a broad detection range from 300 to 1100 nm and a peak external quantum efficiency of 70% at 940 nm. Moreover, owing to the reduced energetic disorder and trap density, the OPD exhibits a picoampere-level dark current density ($1.30 \times 10^{-10} \text{ A cm}^{-2}$) at -0.1 V bias. Such high responsivity and low dark current density endow the OPD with a detectivity surpassing 10^{13} Jones across the whole spectral range. More importantly, this superior photodetection performance can be maintained in large-area flexible device (80 mm^2). These results demonstrate the huge potential of OPDs in addressing the limitations of silicon photodiodes for application in flexible and wearable electronics.

1. Introduction

The convergence of photonics and micro-electronics has entrenched photodetection as a linchpin of modern information technology, which is ubiquitous across various domains of contemporary society, including telecommunications, artificial intelligence, and bioimaging, among others.^[1–4] In a photodetection system, the photodetector is the key component, and its response range and device performance play a decisive role in determining practical application scenarios. Over the past few decades, significant advancements have been made in inorganic photodetectors such as silicon (Si),^[5–7] germanium (Ge),^[8] and indium gallium arsenic (InGaAs),^[9] leading to their widespread commercial utilization. Among them, monocrystalline silicon photodiodes (SiPDs) have emerged as the most commonly utilized photodetectors.^[10,11] This can mainly be attributed to their broad photodetection range, covering ultraviolet,

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visible, and near-infrared (spanning from 300 to 1100 nm), thus meeting the demands of various application scenarios. Moreover, the outstanding semiconductor properties and low trap density of monocrystalline silicon confer SiPDs excellent responsivity (R), low dark current density (J_d), and rapid response. However, manufacturing SiPDs requires epitaxial growth to deposit monocrystalline silicon on the readout circuit, involving intricate and costly processes.^[12] Moreover, the rigidity of Si precludes the realization of flexible and custom-shaped devices. Recently, large-area curved photodetectors have attracted substantial attention due to their wide viewing angles and low data redundancy.^[13,14] These attributes enable the provision of more comprehensive and accurate image information in complex environments. Besides, flexible and wearable electronics have an urgent need for large-area flexible photodetectors.^[15–20] Therefore, the development of novel photodetection technologies is crucial to overcome the limitations of SiPDs.

Recently, organic photodetectors (OPDs) have received widespread attention due to their solution processability, which eliminates the need for complex and costly crystal growth.^[21–30] Additionally, the light weight and flexibility of organic semiconductors endow OPDs great potential for the fabrication of large-area curved devices.^[31–38] Impressively, OPDs have exhibited comparable responsivity and detectivity to SiPDs in visible region.^[39–42] However, a large gap still exists between OPDs and SiPDs for near-infrared (NIR) photodetection. For example, the response range of most OPDs is limited to 1000 nm, falling short of the response cutoff of SiPDs (1100 nm).^[43–47] Furthermore, the low external quantum efficiency (EQE) in NIR remains a significant challenge for OPDs.^[48–53] Besides, the thermally-generated carrier and external carrier injection are more serious for OPDs working in NIR region, resulting in higher J_d .^[54–59] Moreover, the immaturity of fabricating large-area devices on flexible substrates often causes performance degradation in OPDs.^[60] Therefore, it's a big challenge to achieve large-area flexible broadband OPDs that are fully comparable to SiPDs in device metrics.

Herein, a novel electron acceptor, BTPV-CN, featuring acceptor–donor–acceptor (A-D-A) molecular skeleton and electron-withdrawing cyano-substituted terminal groups (CN-IC) is reported. Benefiting from the strong electron push-pull effect, the optical bandgap equivalent to that of silicon was achieved. Additionally, BTPV-CN exhibited deep energy levels, large molecular dipole moment, and pronounced electrostatic potential (ESP) distribution, which facilitates efficient exciton diffusion and dissociation. Thus, the optimized OPDs achieved a competitive responsivity with commercial SiPDs from 300 to 1100 nm and a peak external quantum efficiency (EQE) of 70% at 940 nm. Additionally, the J_d at picoampere level (1.30×10^{-10} A cm⁻² at -0.1 V) was achieved, which can be attributed to the reduced energy disorder and trap state density of BTPV-CN. As a result, BTPV-CN-based OPDs exhibited excellent detectivity above 10^{13} Jones in the whole spectral response range. Moreover, this superior performance can be maintained in large-area flexible device (80 mm²). To the best of our knowledge, this is the first case where a flexible OPD can rival the commercial SiPDs. These results demonstrate the huge potential of OPDs in addressing the limitations of SiPDs for application in flexible and wearable electronics.

2. Results and Discussion

The chemical structure of BTPV-CN is shown in **Figure 1a**. The BTPV core, based on the efficient fused-ring core of Y-series BTP derivatives,^[61–63] incorporates a vinylene bridge to extend π -conjugation, aiming to ensure efficient charge transport and broaden absorption. Furthermore, CN-IC was chosen as the terminal unit due to its stronger electron-withdrawing capability than the conventional halogenated ones, inducing greater lowest unoccupied molecular orbital (LUMO) downshift and enhanced intramolecular charge-transfer (ICT) effects.^[64] The molecule was synthesized through acid-catalyzed Knoevenagel condensation with a yield of 86% (Figure S1a, Supporting Information). It is worth pointing out that BTPV-CN would exist as three isomers, namely BTPV-CN-in, BTPV-CN-out, and BTPV-CN-m (Figure S1b, Supporting Information), due to the fact that the terminal unit CN-IC contains two isomers. According to the analysis of the ¹H NMR spectrum, the molar ratio of these two isomers is close to 1:1 (Figure S2, Supporting Information). For comparison, the reference molecule BTPV-IC with 2-(3-oxo-2,3-dihydro-1H-inden-1-ylidene)malononitrile (IC) as the terminal group was also prepared (Figure S1a, Supporting Information). The chemical structures of both acceptors were characterized by Fourier-transform infrared, ¹H, ¹³C nuclear magnetic resonance (NMR), and matrix-assisted laser desorption/ionization time-of-flight (MALDI-TOF) mass spectroscopy (MS) (Figures S3–S6, Supporting Information). Both acceptors demonstrated favorable solubility in common solvents such as chloroform and chlorobenzene, indicating their good solution processability. The decomposition temperatures (T_d) of exceeding 300 °C allow both molecules to maintain stability during the device processing procedure (Figure S7, Supporting Information).

The molecular geometries and dipole moments of BTPV-CN and BTPV-IC were investigated by density functional theory (DFT) calculation. As shown in Figure S8 (Supporting Information), all acceptors exhibited planar conjugated backbones, which is beneficial to intra- and intermolecular charge transport. According to the optimized molecular geometries, the ground-state dipole moments of BTPV-CN and BTPV-IC in three-dimensional directions and the total ones were extracted (Table S1, Supporting Information). As shown in Figure 1b, the reference BTPV-IC exhibited a small dipole moment of 0.64 Debye, while the CN-IC endows BTPV-CN isomers with a tenfold increase in magnitude (above 10.0 Debye) at opposite direction. Based on the optimized molecular geometries, we also found more negative electrostatic potential (ESP) values around CN-IC unit, thereby resulting in more positive ESP values within the BTPV core (Figure 1c). The area distribution of ESP values further verified that BTPV-CN exhibited larger regions with higher ESP values (Figure 1d). Given the identical BTPV core in both molecules, these distinct ESP characteristics predominantly originate from their different terminal units (Figure S9, Supporting Information). Such ESP distribution characteristics of BTPV-CN not only lead to enhanced intermolecular interaction, thereby reducing trap density, but also strengthen the intermolecular electric field (IEFs) between the donor and acceptor, promoting exciton dissociation.^[65,66]

For a more in-depth investigation of the exciton dissociation process, the distribution and separation characters between holes and electrons in both acceptors were further investigated via

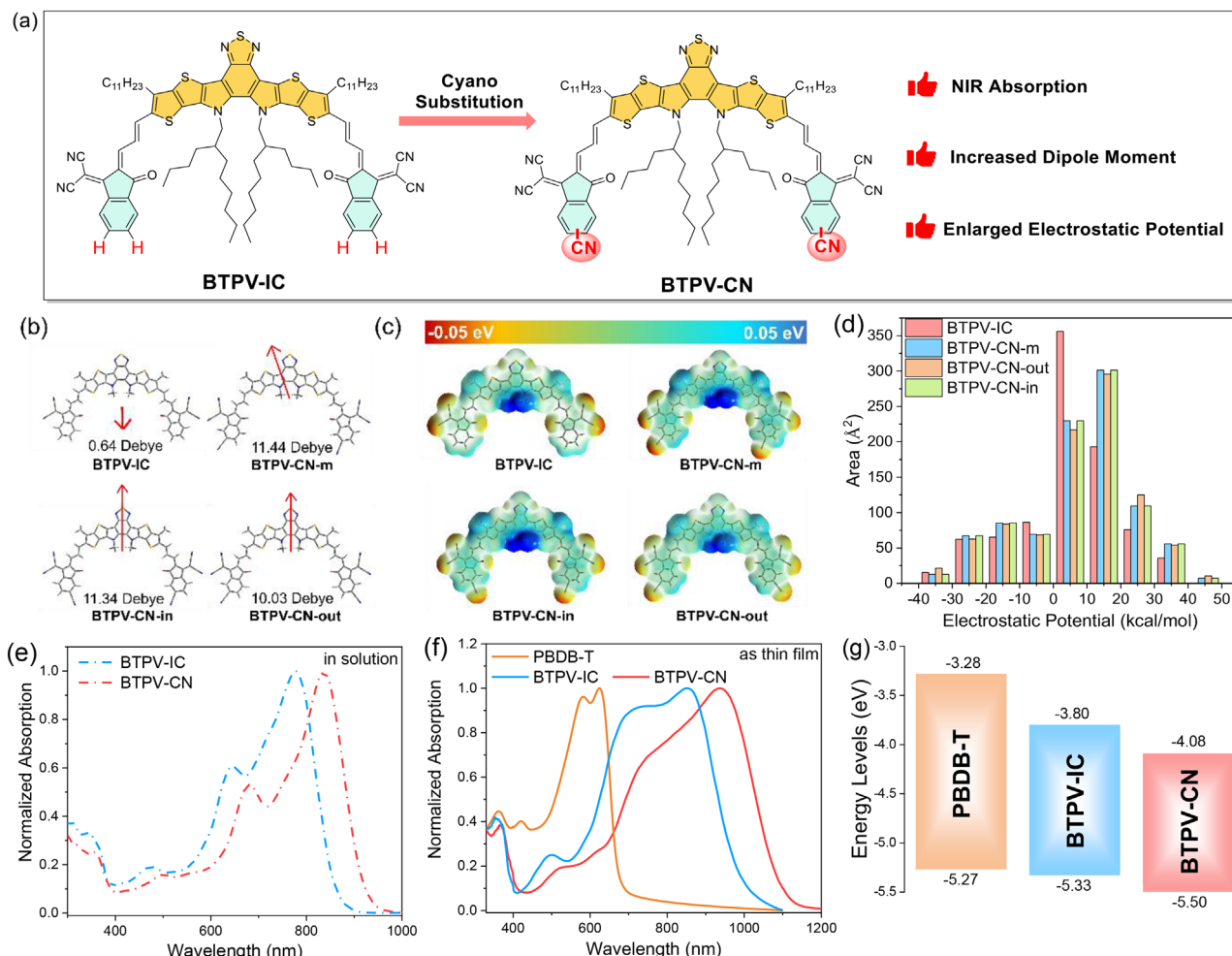


Figure 1. a) The chemical structures, b) molecular dipole moment, c) electrostatic potential, and d) corresponding area distribution of BTPV-IC and BTPV-CN. e–g) The UV-vis–NIR absorption spectra in chloroform solution (e) and as thin film (f), as well as (g) energy levels of PBDB-T, BTPV-IC, and BTPV-CN.

time-dependent DFT calculation. First of all, the difference between the ground- and excited-state dipole moments ($\Delta\mu_{ge}$) was calculated by the Equation (1), which reflects the degree of photoinduced intramolecular charge transfer.^[67–69]

$$\Delta\mu_{ge} = \sqrt{(\mu_{gx} - \mu_{ex})^2 + (\mu_{gy} - \mu_{ey})^2 + (\mu_{gz} - \mu_{ez})^2} \quad (1)$$

where μ_{gx} , μ_{gy} , and μ_{gz} refer to the ground-state dipoles in the x, y, and z directions, and μ_{ex} , μ_{ey} , and μ_{ez} refer to the excited-state dipoles in the x, y, and z directions, respectively. The $\Delta\mu_{ge}$ values were calculated to be 7.74 for BTPV-IC, 8.35 for BTPV-CN-in, 12.27 for BTPV-CN-out, and 9.92 for BTPV-CN-m (Table S1, Supporting Information). The larger $\Delta\mu_{ge}$ value for BTPV-CN isomers indicates the stronger intramolecular charge transfer, which is beneficial for increasing the separation efficiency of the electron–hole pairs and lowering the exciton binding energy. Based on the time-dependent DFT calculation, the hole–electron distributions of BTPV-IC and BTPV-CN isomers are illustrated in Figure S10 (Supporting Information), where both holes (blue

area) and electrons (green area) are predominantly delocalized over the intermediate cores with less distribution on the end-groups. Hole and electron delocalization indices (HDI and EDI) were employed to accurately characterize the degree of charge delocalization. As presented in Table S2 (Supporting Information), both BTPV-CN-in and BTPV-CN-out exhibited larger HDI and EDI than those of BTPV-IC, indicating effective charge delocalization. However, due to its asymmetrical end-group, BTPV-CN-m possessed a smaller HDI compared to that of BTPV-IC. To evaluate the separation between holes and electrons, acceptors' overlap degree of hole–electron (S_r index) as well as centroid distance between them (D index) were calculated (Table S2, Supporting Information). Simultaneously, the majority of BTPV-CN isomers exhibited a smaller S_r index and a larger D index compared to BTPV-IC, with BTPV-CN-m displaying the smallest S_r and largest D index. The smoothed hole/electron distributions of acceptors clearly demonstrate distinct spatial segregation between holes and electrons in BTPV-CN-m, thereby elucidating its reduced S_r and enlarged D index (Figure S11, Supporting Information). These observations suggest a more efficient charge

Table 1. Optical and electrochemical properties of PBDB-T, BTPV-IC, and BTPV-CN.

Sample	$\lambda_{\text{sol,max}}$ [nm]	$\lambda_{\text{sol,onset}}$ [nm]	$\lambda_{\text{film,max}}$ [nm]	$\lambda_{\text{film,onset}}$ [nm]	$E_{\text{g}}^{\text{opt}}$ [eV]	HOMO [eV]	LUMO [eV]
PBDB-T	—	—	624	672	1.85	−5.27	−3.28
BTPV-IC	776	851	829	986	1.26	−5.33	−3.80
BTPV-CN	837	937	923	1100	1.13	−5.50	−4.08

separation process would be found for BTPV-CN, which is favorable to exciton dissociation in active layer.

The UV–vis–NIR absorption spectra of BTPV-IC and BTPV-CN in solution and as thin film are shown in Figure 1e,f, respectively. The corresponding numerical data can be found in Table 1. In chloroform solution, BTPV-CN exhibited an obviously red-shifted absorption maximum ($\lambda_{\text{sol,max}}$) compared to BTPV-IC (837 vs 776 nm), which is consistent with the strong electron-withdrawing ability of CN-IC. In thin film, the absorption onset of BTPV-CN redshifts further to 1100 nm, resulting in an optical bandgap ($E_{\text{g}}^{\text{opt}}$) of 1.13 eV, whereas a significantly larger $E_{\text{g}}^{\text{opt}}$ of 1.28 eV was found for BTPV-IC. Notably, BTPV-CN also exhibited a prominent, sharpened (0-0) aggregation absorption peak at 923 nm, suggesting the formation of J-aggregates, which can be further evidenced by the intermolecular displacement angle (Figure S12, Supporting Information) and the negative total excitonic coupling energies (Table S3, Supporting Information) of the BTPV-CN dimer, as calculated by the Spano model.^[70,71] Additionally, as shown in Figure 1f, BTPV-CN exhibited excellent spectral complementarity with the polymer donor PBDB-T.^[72] The broad absorption range from 300 to 1100 nm indicates that the resulting OPDs would be comparable to SiPDs in terms of response range.

Additionally, the LUMO and highest occupied molecular orbital (HOMO) levels of BTPV-CN, BTPV-IC, and PBDB-T were calculated by cyclic voltammetry measurements (Figure S13, Supporting Information). As shown in Figure 1g, BTPV-CN demonstrated significantly downshifted energy levels relative to BTPV-IC, which can be ascribed to the strong electron-withdrawing nature of the CN-IC. These energy level shifts were corroborated by ultraviolet photoelectron spectroscopy (UPS), which measured HOMO levels of −5.37 eV for BTPV-IC and −5.45 eV for BTPV-CN (Figure S14, Supporting Information). The frontier molecular orbital energy levels obtained from DFT calculations also support this trend (Figure S15, Supporting Information). Importantly, this downshift in the energy level of BTPV-CN enables a larger HOMO–HOMO and LUMO–LUMO energy offset with PBDB-T. This would promote more efficient exciton dissociation, thereby contributing to a high EQE in the resulting OPDs.

The photodetection performance of BTPV-CN and BTPV-IC was evaluated in a device with a structure of indium tin oxide (ITO)/zinc oxide (ZnO)/active layer/molybdenum oxide (MoO_x)/Ag. To reduce trap density, the layer-by-layer (LbL) processing method was used for fabricating active layer by sequentially depositing donor and acceptor.^[73] Notably, this spin-coating sequence would result in the deposition of the donor material on the cathode and proximity of the acceptor material to anode in principle, which could adversely affect charge transport and ex-

traction. However, many studies have shown that the donor and acceptor can interpenetrate each other and ultimately form a vertically mutually penetrating morphology, thereby fulfilling the requirements for efficient charge transport.^[74,75] To verify this in the active layer, depth analysis using X-ray photoelectron spectroscopy (XPS) was conducted to investigate the vertical composition distribution. N 2p and O 1s were selected as characteristic elements for indicating acceptors and PBDB-T, respectively. Additionally, Zn 2p and In 3d were chosen as characteristic elements for indicating ZnO and ITO, respectively. As shown in Figure S16 (Supporting Information), the vertical composition distribution of both active layers exhibited a similar and uniform pattern with consistent concentrations of N 2p and O 1s throughout the vertical direction. This suggested that within the two active layers, the donor and acceptor have achieved full vertical interpenetration (Figure 2a). The vertical distribution of PBDB-T and acceptor within the active layers was further investigated using film-depth-dependent light absorption spectroscopy (Figure S17, Supporting Information). Notably, $\approx 59\%$ of BTPV-IC and 46% of BTPV-CN were detectable at the bottom of the active layers, confirming intimate interdiffusion between the acceptor and PBDB-T throughout the vertical direction. For a direct comparison, we fabricated control devices using the conventional bulk heterojunction (BHJ) for both active layers. As detailed in Figure S18 (Supporting Information), the BHJ devices showed inferior performance compared to the LbL devices. This performance gap confirms the critical advantage of the vertically interpenetrated donor–acceptor network achieved by LbL processing for efficient charge transport and superior photodetection.

As shown in Figure 2b and Table 2, the BTPV-IC-based OPD exhibited EQE values below 50% from 300 to 1000 nm at −0.1 V bias, with only 43.7% at 850 nm, indicating moderate device performance. By contrast, the BTPV-CN-based OPD not only achieved a broader response range spanning from 300 to 1200 nm, but also demonstrated higher EQE values, particularly an EQE of 70% at 940 nm under a −0.1 V bias. Moreover, the negligible bias-dependence of EQE further corroborates the efficient charge generation and collection within the device (Figure S19, Supporting Information). The R values of both devices were obtained according to the following equation:

$$R = \frac{\text{EQE}}{100\%} \times \frac{\lambda}{1240} \quad (2)$$

where λ is the wavelength of the incident light. As shown in Figure 2c, the OPD based on BTPV-CN demonstrated excellent R values in NIR region, with a peak value of 0.53 A W^{-1} at 940 nm. However, due to the limited EQE, the R values of BTPV-IC-based OPD were constrained to less than 0.3 A W^{-1} , with a maximum R

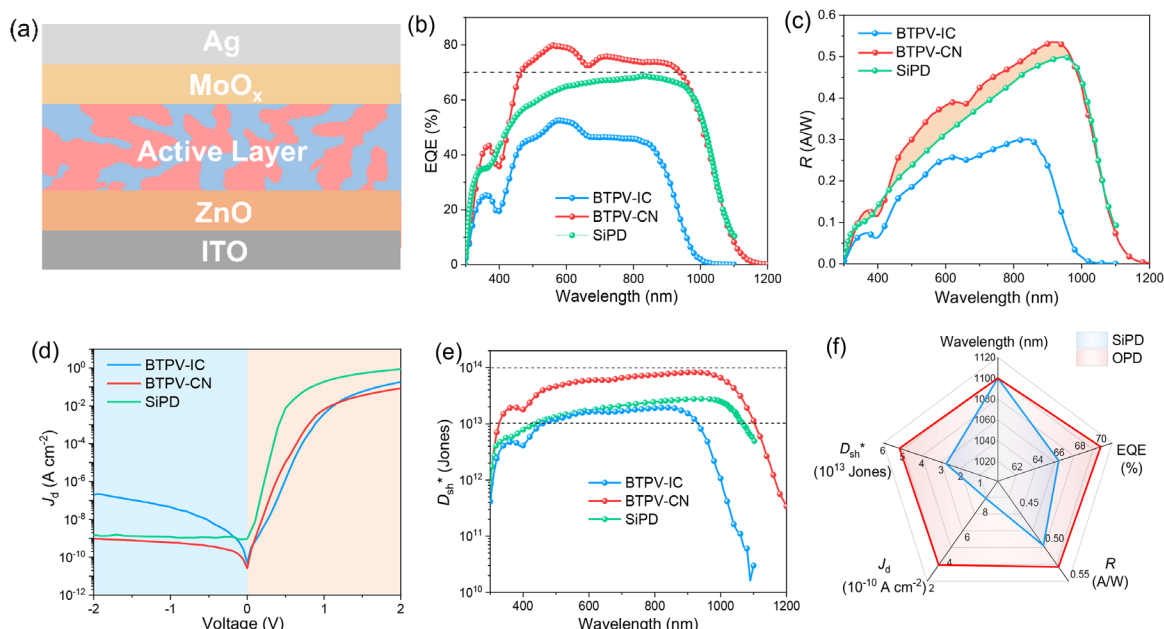


Figure 2. a) The device structure, b) external quantum efficiency spectra, c) responsivity spectra, d) J_d - V curves in dark, and e) shot-noise-limited specific detectivity of photodetectors based on PBDB-T/BTPV-IC, PBDB-T/BTPV-CN, and silicon under -0.1 V bias. f) Device performance matrix (@ 940 nm, at -0.1 V bias) of our OPD (PBDB-T/BTPV-CN) and SiPD (Hamamatsu S1787-08).

of 0.29 A W^{-1} at 850 nm. Additionally, we found that the R spectrum of BTPV-CN-based OPD exhibited a striking resemblance to, and even surpassed, that of SiPD (Hamamatsu S1787-08). To our best knowledge, this is the first instance where OPD exhibits a comparable response range and responsivity with commercial SiPDs.

The BTPV-CN-based OPD achieved ultralow J_d of $1.3 \times 10^{-10} \text{ A cm}^{-2}$ at -0.1 V and 9.6×10^{-10} at -2 V, which ranks among the lowest values reported for state-of-the-art NIR OPDs (Table S4, Supporting Information). Its performance also surpasses that of the reference commercial SiPD (Hamamatsu S1787-08) across a wide bias range up to -2 V, suggesting its potential to compete with SiPD in low-power applications. In comparison, the BTPV-IC-based OPD showed a J_d of $7.74 \times 10^{-10} \text{ A cm}^{-2}$ at -0.1 V but exhibited a more significant increase, reaching $1.99 \times 10^{-7} \text{ A cm}^{-2}$ at -2 V. This stark contrast highlights the efficient suppression of dark current in the BTPV-CN-based device, particularly at operational biases relevant to flexible applications.^[76,77] The suppressed J_d in BTPV-CN devices can be attributed to the deeper frontier molecular orbital energy levels, which raise the barrier for external carrier injection, coupled with a reduced trap-state density. By fitting these J - V curves with the ideal generalized

Shockley diode equation (Figure S20, Supporting Information), we can obtain the ideality factor (n), dark saturation current density (J_0), and shunt resistance (R_{sh}).^[78] Among them, n reflects the charge recombination mechanism in the device. The BTPV-CN-based device exhibited a smaller n value than that of BTPV-IC-based device (1.19 vs 1.49), suggesting suppressed trap-assisted recombination. Additionally, the BTPV-CN-based OPD also exhibited smaller J_0 ($1.10 \times 10^{-10} \text{ A cm}^{-2}$) and larger R_{sh} ($5.93 \times 10^{10} \Omega \text{ cm}^2$) than those of BTPV-IC-based OPD ($J_0 = 1.47 \times 10^{-7} \text{ A cm}^{-2}$, $R_{sh} = 1.03 \times 10^7 \Omega \text{ cm}^2$), which confirms that it has a greater carrier injection barrier under reverse bias.

The shot-noise-limited specific detectivity (D_{sh}^*) of the devices were calculated according to the below equation:

$$D_{sh}^* = \frac{R\sqrt{A}}{\sqrt{2qI_d}} = \frac{R}{\sqrt{2qJ_d}} \quad (3)$$

where A and i_d represent the active area and dark current, respectively. As shown in Figure 2e, the BTPV-IC-based device obtained D_{sh}^* exceeding 10^{12} Jones under a -0.1 V bias within the response range of 350–1000 nm. By virtue of its higher R value and broader response range, the D_{sh}^* of the BTPV-CN-based device can be

Table 2. Performance parameters of the OPDs based on PBDB-T/BTPV-IC and PBDB-T/BTPV-CN and SiPD (Hamamatsu S1787-08) measured under -0.1 V bias.

Photodetector	EQE ^{a)} [%]	R^a [A W ⁻¹]	J_d [A cm ⁻²]	$D_{sh}^{*a)}$ [Jones]	S_n [A Hz ^{-0.5}]	$D^{*a)}$ [Jones]
PBDB-T/BTPV-IC	43.7	0.29	7.74×10^{-10}	1.93×10^{13}	3.65×10^{-14}	1.86×10^{12}
PBDB-T/BTPV-CN	70.0	0.53	1.30×10^{-10}	8.21×10^{13}	5.01×10^{-15}	2.41×10^{13}
Silicon	65.7	0.34	8.79×10^{-10}	2.36×10^{13}	3.29×10^{-13}	6.33×10^{11}

^{a)} Obtained from the OPDs at 850 nm for PBDB-T/BTPV-IC and at 940 nm for PBDB-T/BTPV-CN, and from SiPD at 940 nm, under -0.1 V bias.

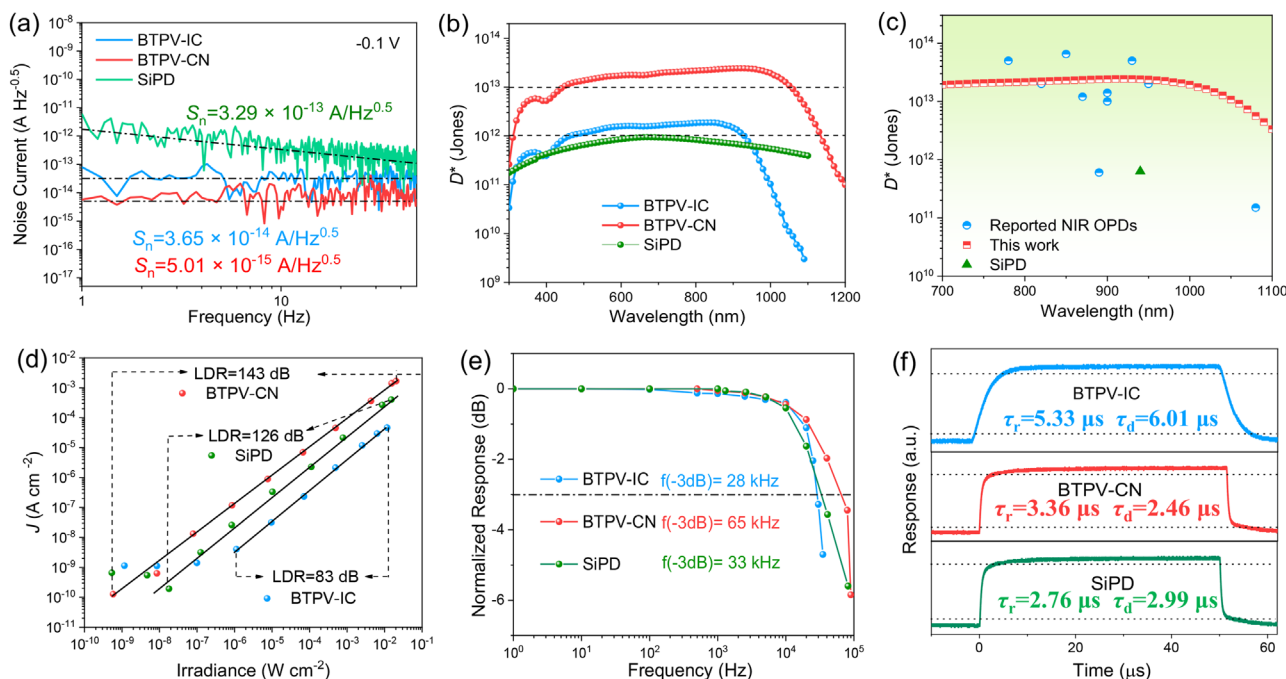


Figure 3. a) Noise current density S_n and b) specific detectivity D^* of SiPD (Hamamatsu S1787-08), BTPV-IC and BTPV-CN OPDs at -0.1 V bias. c) D^* versus wavelength for SiPD, state-of-the-art NIR OPDs, and PBDB-T/BTPV-CN OPD (reference data sources in Table S4, Supporting Information). d) Linear dynamic range. e) Cut-off frequency under -0.1 V bias with 100Ω load. The SiPD reference (Hamamatsu S1787-08), operating under identical low-bias conditions required for direct OPD comparison, may exhibit bandwidth constraints as this configuration deprives it of optimal depletion characteristics. f) Response speed at 10 kHz light modulation for OPDs. All measurements used 850 and 940 nm LEDs for BTPV-IC and BTPV-CN OPDs, respectively, and 940 nm LED for SiPD.

elevated to above 10^{13} Jones spanning from 350 – 1100 nm, with a maximum value of 8.21×10^{13} Jones at 940 nm, which even surpasses Hamamatsu S1787-08 (Table 2). Consequently, we can conclude that the performance of BTPV-CN-based device is comparable to commercially available SiPDs in significant device performance parameters (Figure 2f).

To more accurately evaluate the detectivity, the specific detectivity (D^*) was calculated according to the equation below:

$$D^* = \frac{R\sqrt{A\Delta f}}{S_n} \quad (4)$$

where Δf is the detection bandwidth, and S_n is the noise current. As shown in Figure 3a, the average S_n of BTPV-IC- and BTPV-CN-based devices under a -0.1 V bias were determined to be 3.65×10^{-14} and 5.01×10^{-15} A Hz^{-0.5}, respectively. This remarkable reduction in noise current enables the BTPV-CN-based device to achieve excellent D^* above 10^{13} Jones within 450 – 1100 nm and an outstanding D^* of 2.41×10^{13} Jones at 940 nm (Figure 3b). More importantly, the close proximity between D_{sh}^* (8.21×10^{13} Jones) and D^* (2.41×10^{13} Jones) for the BTPV-CN-based device indicates effective suppression of thermal noise, consistent with the higher R_{sh} . Subsequently, we summarized the D^* for thus-far reported NIR OPDs (Table S4, Supporting Information), and found that our results are in the top tier (Figure 3c). Notably as well, the performance of BTPV-CN device is even superior to SiPD (Figure 3a–c).

Linear dynamic range (LDR) represents the range of light intensity to which OPD responds linearly, which followed an equation:

$$LDR = 20 \log \frac{I_{upper}}{I_{lower}} \quad (5)$$

where I_{upper} and I_{lower} are the maximum and minimum photocurrent densities in the linear range. The LDR of BTPV-IC-based devices was restricted to 83 dB (850 nm LED) due to their limited performance under low light intensity conditions. In contrast, the BTPV-CN-based device achieved a higher LDR of 143 dB (under 940 nm LED) (Figure 3d), enabled by its lower J_d and higher R that maintained photocurrent linearity at ultra-low irradiance. Notably, the LDR of BTPV-CN-based device is even higher than that of SiPD. The cutoff frequency (f_{-3dB}) is a critical parameter for assessing photodetectors' effective bandwidth, representing the frequency at which the output amplitude decreases by 3 dB. The BTPV-CN-based device achieved 65 kHz, outperforming BTPV-IC (28 kHz) due to enhanced charge transport (Figure 3e). For reference, the SiPD offered an f_{-3dB} of 33 kHz under the same testing conditions. The response speed is quantified by the rise time and fall time, which represent the duration for the signal to ascend from 10% to 90% and descend from 90% to 10% , respectively. In the case of OPD based on BTPV-IC and BTPV-CN, their respective rise times (τ_r) were measured as 5.53 and 3.36 μ s, while corresponding fall times (τ_d) were recorded as 6.01 and 2.46 μ s (Figure 3f). The shorter rise and fall times indi-

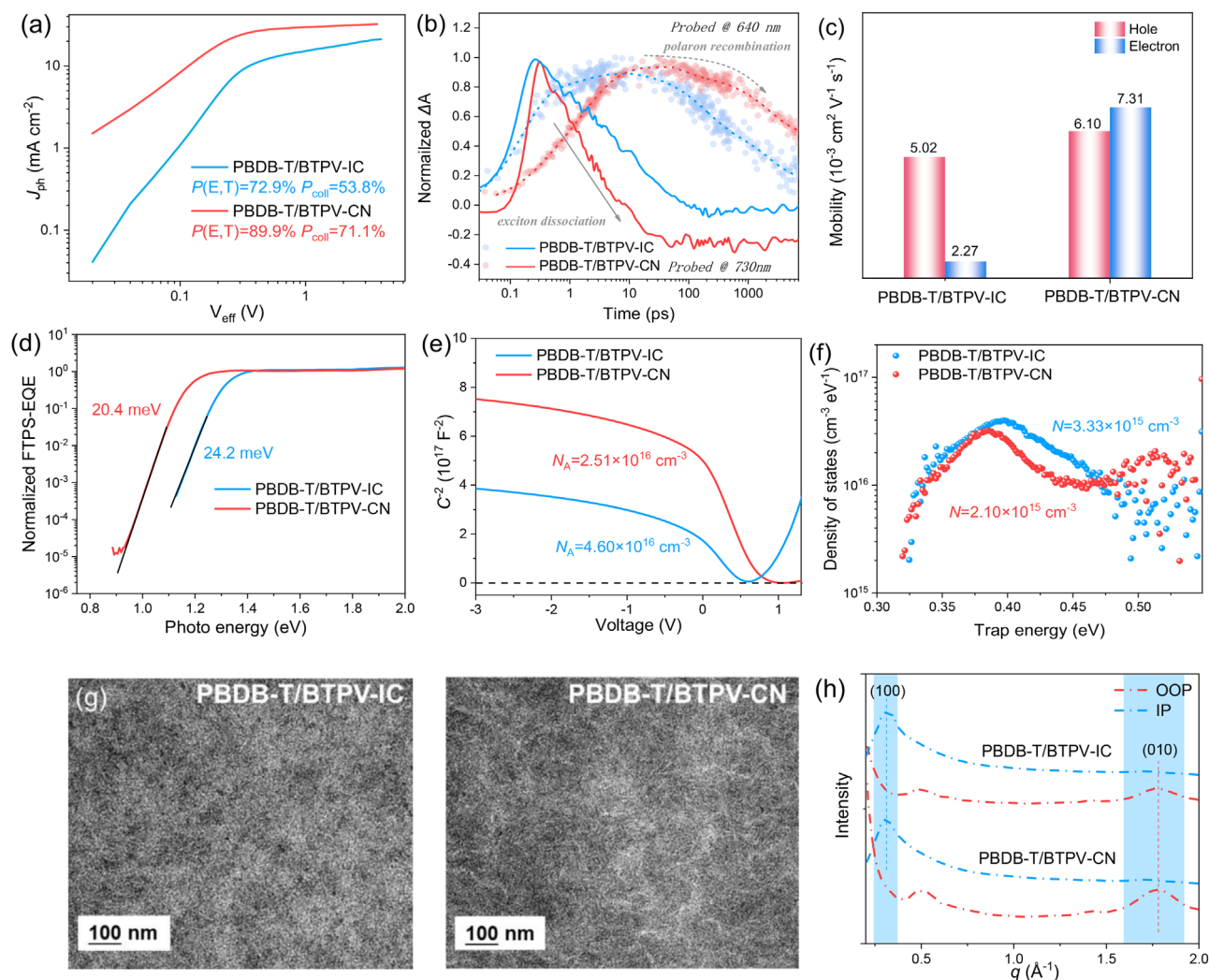


Figure 4. a) The $P_{(E,T)}$ of the OPDs based on PBDB-T/BTPV-IC and PBDB-T/BTPV-CN. b) fs-TA spectra of the PBDB-T/BTPV-IC and PBDB-T/BTPV-CN films. c) Summary of the charge carriers mobilities of PBDB-T/BTPV-IC and PBDB-T/BTPV-CN-based devices. d) The normalized FTPS-EQE spectra, e) Mott–Shockley plot, and f) trap DOS of the OPDs based on PBDB-T/BTPV-IC and PBDB-T/BTPV-CN. g) The TEM image and h) 2D GIWAXS patterns of PBDB-T/BTPV-IC and PBDB-T/BTPV-CN films.

cate faster response for the OPDs of BTPV-CN. For comparison, the reference SiPD exhibited τ_r of 2.76 μ s and τ_d of 2.99 μ s.

The steady-state photoluminescence (PL) measurements were performed to further investigate the charge transfer process. Compared to BTPV-IC (61.67%), the higher PL quenching efficiency of BTPV-CN (81.73%) in active layer suggested a more efficient hole transfer, which can be attributed to the increased HOMO–HOMO energy offset between the electron donor and acceptor (Figure S21, Supporting Information). Furthermore, based on the photocurrent density (J_{ph}) versus effective voltage (V_{eff}) plot, the exciton dissociation efficiency $P_{(E,T)}$ was determined to be 72.9% for BTPV-IC-based device and 89.9% for BTPV-CN-based device, which is consistent with the PL quenching efficiency (Figure 4a). Additionally, the BTPV-CN-based device exhibited a higher carrier collection efficiency (P_{coll}) compared to that of the BTPV-IC-based device (71.1% vs 53.8%). These advantages confirmed the more efficient exciton dissociation

and carrier collection within BTPV-CN-based device, which aligns with the high R.

The exciton dissociation and polaron recombination kinetics in blend films were investigated by femtosecond transient absorption (fs-TA). As shown in Figures S22 and S23 (Supporting Information), upon selective excitation of the acceptors using a pump wavelength of 700 nm, the ground state bleach (GSB) signal peaks from the acceptors rapidly emerge within the range of 675–750 nm within 0.2–0.5 ps, indicating the formation of the photogenerated exciton. Subsequently, the exciton diffuses to the donor/acceptor interface where it undergoes dissociation, concurrent with the emergence of the GSB signal of donor (500–675 nm). As shown in Figure 4b, the exciton in PBDB-T/BTPV-CN film undergoes complete dissociation within 10 ps, whereas the exciton in PBDB-T/BTPV-IC film requires 10 times longer to fully dissociate. This suggests that the exciton dissociation process in PBDB-T/BTPV-CN film is significantly more rapid

compared to that in PBDB-T/BTPV-IC film. Additionally, Figure 4b indicates that the PBDB-T/BTPV-IC film exhibits a slightly faster hole transfer rate, with a lifetime of 1.00 ps. In contrast, the hole transfer lifetime for PBDB-T/BTPV-CN blends is 2.48 ps (Table S5, Supporting Information). However, it is noteworthy that this 2.48 ps hole transfer rate remains substantially faster than the exciton diffusion timescale within 10 ps. Moreover, the polaron associated with hole transfer in PBDB-T/BTPV-CN film has a longer lifetime, suggesting suppressed polaron recombination. These results suggest that despite the slower hole transfer in PBDB-T/BTPV-CN blends, efficient exciton dissociation and inhibited polaron recombination contribute significantly to the generation of photogenerated current, consistent with the reports that balanced charge extraction dynamics govern performance in organic photodiodes.^[79,80]

The carrier transport characteristics of the neat and blend films were further investigated using the space-charge-limited current (SCLC) method. It was observed that the BTPV-CN neat film shows a higher electron mobility (μ_e) compared to BTPV-IC (Figure S24 and Table S6, Supporting Information). This higher μ_e can also be found in the PBDB-T/BTPV-CN film (Figures S25 and S26 and Table S6, Supporting Information). Additionally, PBDB-T/BTPV-CN thin film exhibited higher hole mobility (μ_h) and balanced hole/electron transport ($\mu_h/\mu_e = 0.83$) (Figure 4c). Comparatively, measurements of the corresponding BHJ devices revealed significantly lower electron and hole mobilities than those in the LBL devices (Figures S27 and S28 and Table S7, Supporting Information), demonstrating the advantage of the LBL structure for charge transport. The bimolecular recombination can be investigated by extracting the power index from $J_{sc} \propto P_{light}^\alpha$. As shown in Figure S29a (Supporting Information), the BTPV-CN-based device has an α value closer to 1, indicating more efficient suppression of bimolecular recombination. The trap-assisted recombination can be studied by extracting the slope from $V_{oc} \propto nkT/q \ln(P_{light})$. The smaller slope of BTPV-CN-based device (1.17 vs 1.46 kT/q) suggests reduced trap-assisted recombination (Figure S29b, Supporting Information). Additionally, the BTPV-CN-based OPD exhibited shorter decay time (0.18 vs 0.51 μ s) in transient photocurrent measurements (Figure S30, Supporting Information), which further demonstrates efficient carrier transport and extraction.

To elucidate the origin of low J_d , we initially measured the Urbach energy (E_U) to evaluate the energy disorder within the OPDs. As shown in Figure 4d, by fitting the Fourier transform photocurrent spectroscopy external quantum efficiency (FTPS-EQE) onset, the E_U values of 20.4 and 24.2 meV were determined for PBDB-T/BTPV-CN- and PBDB-T/BTPV-IC-based OPDs, respectively. The lower E_U indicates reduced energetic and structural disorder, leading to a decrease in trap density and subsequent suppression of J_d in PBDB-T/BTPV-CN-based OPD. Subsequently, the capacitance–voltage ($C-V$) measurements and corresponding Mott-Shockley analysis were employed to investigate the trap density (N_A). As shown in Figure S31a (Supporting Information), the PBDB-T/BTPV-IC- and PBDB-T/BTPV-CN-based devices both exhibited a rapid increase in capacitance near 0 V, which is consistent with the steep absolute slopes in Mott-Shockley analysis (Figure 4e).^[81] Furthermore, the maximum

slope within this voltage range was extracted to calculate trap density N_A according to the equation

$$N_A = \frac{-2}{q\epsilon_r\epsilon_0 A^2} \left(\frac{dV}{dC^{-2}} \right) \quad (6)$$

As listed in Table S8 (Supporting Information), the N_A of PBDB-T/BTPV-IC- and PBDB-T/BTPV-CN-based devices were determined to be 4.60×10^{16} and $2.51 \times 10^{16} \text{ cm}^{-3}$, respectively. The lower N_A suggests less trap-mediated dark current in PBDB-T/BTPV-CN-based OPD. Additionally, the depletion layer width (W) was calculated by Mott-Shockley analysis using the formula:

$$W = \sqrt{\frac{2\epsilon_r\epsilon_0 (V_{bi} - V)}{qN_A}} \quad (7)$$

in which V_{bi} and V represent the built-in potential and applied DC voltage (0 V), respectively. As listed in Table S8 (Supporting Information), the W of PBDB-T/BTPV-CN-based device was found to be twice of BTPV-IC-based device (101 vs 59 nm), which suggests that the entire sensing layer in the former is fully depleted even under zero bias, and aligns with the weak bias-dependent EQE observed under reverse bias.

To conduct an in-depth analysis of the density distribution of trap states, capacitance–frequency ($C-\omega$) measurements were further performed. As shown in Figure S31b (Supporting Information), there is a rapid change in capacitance within the frequency range of 10^4 – 10^5 Hz observed for both devices, indicating the presence of deep-energy trap.^[82,83] Following the methodology proposed by Walter et al.^[84] it is feasible to extract the state density distribution corresponding to different energy defects using the equation

$$DOS(E_w) = -\frac{V_{bi}}{qAW} \frac{\omega}{kT} \frac{dC}{d\omega} \quad (8)$$

As shown in Figure 4f, by performing Gaussian fitting on these two curves respectively, the trap state densities of PBDB-T/BTPV-IC- and PBDB-T/BTPV-CN-based devices were estimated to be 3.33×10^{15} and $2.10 \times 10^{15} \text{ cm}^{-3}$, respectively. Meanwhile, the PBDB-T/BTPV-CN-based device also exhibited shallower trap energy (E_t) and narrower Gaussian width (σ) (Table S8, Supporting Information). This trend was further validated by SCLC measurements (Figure S32, Supporting Information), which probe the total trap-limited carrier transport.^[85,86] For the PBDB-T/BTPV-IC blend, hole transport displayed a trap-filled limit voltage (V_{TFL}) of 0.28 V ($N_t = 4.13 \times 10^{16} \text{ cm}^{-3}$), while electron transport showed a V_{TFL} of 0.39 V ($N_t = 5.76 \times 10^{16} \text{ cm}^{-3}$). In contrast, the PBDB-T/BTPV-CN blend demonstrated significantly reduced V_{TFL} values: 0.27 V for hole transport ($N_t = 3.98 \times 10^{16} \text{ cm}^{-3}$) and 0.25 V for electron transport ($N_t = 3.69 \times 10^{16} \text{ cm}^{-3}$). This quantification confirms a substantial decrease in both hole and electron trap densities in the cyano substitution system. Consequently, the reduced trap density impedes carrier transition under dark conditions, accounting for the lower J_d in PBDB-T/BTPV-CN-based devices.

The morphology of the blend films was investigated by transmission electron microscopy (TEM) and grazing incidence

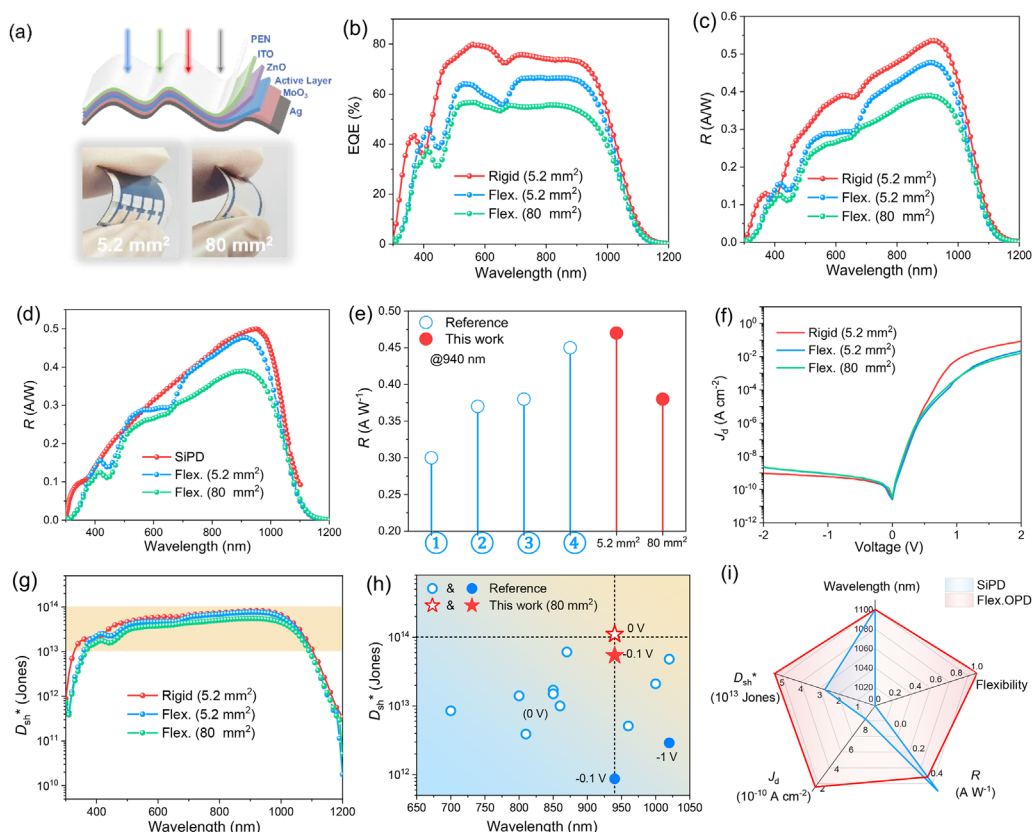


Figure 5. a) The device diagram of flexible OPDs. b,c) The EQE (b) and R spectra (c) of the rigid and flexible OPDs based on BTPV-CN. d) The R spectra of SiPD (Hamamatsu S1787-08) and our flexible OPDs. e) The comparison between the flexible OPDs reported in literatures and this work regarding R at 940 nm. f) The J - V curves in dark and g) D_{sh}^* under a -0.1 V bias of the rigid and flexible OPDs based on BTPV-CN. h) Plot of D_{sh}^* versus wavelength of flexible OPDs operated without bias (hollow) and under bias (solid). i) Device performance matrix (@ 940 nm, at -0.1 V bias) of flexible OPD (80 mm^2) and SiPD. The corresponding source references of insets (e,h) are provided in Table S11 (Supporting Information).

wide-angle X-ray scattering (GIWAXS). As shown in Figure 4g, clear phase separation between electron donor and electron acceptor was evident in both the PBDB-T/BTPV-IC and PBDB-T/BTPV-CN films. This morphological feature promotes the diffusion of excitons to the donor-acceptor interface and improves their dissociation. Moreover, the PBDB-T/BTPV-CN film exhibits a more distinct fibrous network structure, which not only supports efficient charge carrier transport but also suppresses recombination processes, thereby resulting in enhanced EQE and R . The microstructures of the neat acceptors and donor:acceptor active layer were further examined by GIWAXS. The figures depicting the 2D GIWAXS patterns and corresponding 1D line-cut profiles can be found in Figures S33 and S34 (Supporting Information) and Figure 4h. Detailed numerical data, such as peak positions (q), stacking distance (d), full width at half-maximum (FWHM), and crystalline coherence lengths (L_c), have been compiled in Tables S9 and S10 (Supporting Information). The neat films of both BTPV-IC and BTPV-CN exhibited a face-on orientation, characterized by the clear (010) diffraction peaks at out-of-plane (OOP) direction with q values ranging from 1.78 to 1.80 Å^{-1} , as well as a (100) diffraction peak at in-plane (IP) direction with q values ranging from 0.35 to 0.36 Å^{-1} . Additionally, the π - π stacking distances were determined to be 3.49 Å for BTPV-IC and 3.53 Å for BTPV-CN neat films (Figure S34, Supporting

Information), which aligns with the typical values observed in state-of-the-art electron acceptors. Although the π - π stacking distance of BTPV-CN is slightly larger than that of BTPV-IC, the enhanced electron mobility in its electron-only device suggests that charge transport is not solely governed by crystalline packing. This aligns with the emerging understanding that in molecular systems, long-range charge transport can be dominated by the electronic connectivity through amorphous regions, where efficient intermolecular coupling often outweighs the effect of precise packing distance.^[87] In donor:acceptor active layer, the obvious face-on orientation was also observed. This advantageous molecular orientation facilitates efficient electron and hole transport toward the respective electrode. Notably, the PBDB-T/BTPV-CN film exhibited larger L_c s than those of PBDB-T/BTPV-IC film, particularly in terms of the (100) diffraction peak at IP direction (42.84 vs 34.48 Å). The increased L_c is consistent with improved charge transport and suppressed recombination process in BTPV-CN-based device.

Having demonstrated the excellent performance of BTPV-CN-based OPD on rigid substrate, the flexible OPDs with active areas of 5.2 and 80 mm^2 were further fabricated on polyethyleneterephthalate (PEN) substrate (Figure 5a). As shown in Figure 5b,c, the flex-OPD (5.2 mm^2) also maintained excellent responsivity across 300 – 1100 nm , with an EQE of 61.9% and R of 0.47 A W^{-1} at

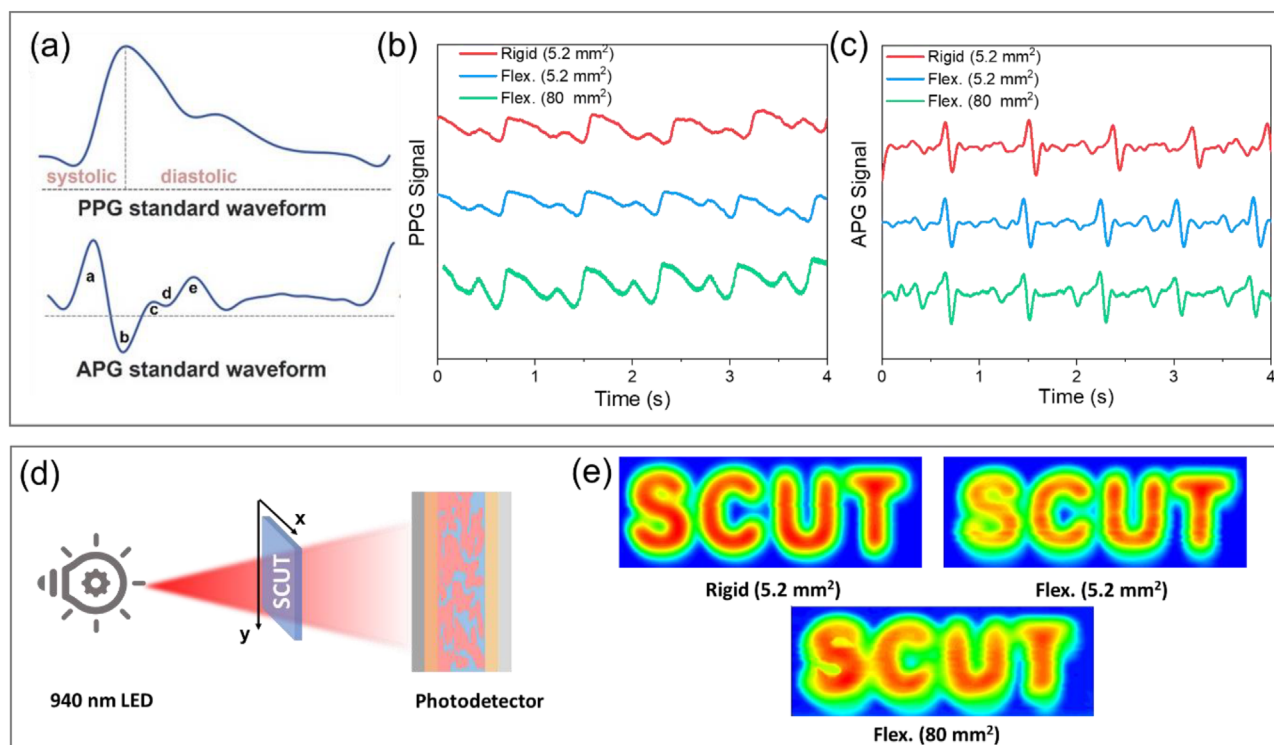


Figure 6. a) The schematic diagram of the PPG application. b,d) The standard PPG (b) and APG waveform (c) of BTPV-CN-based OPDs. d,e) Schematic diagram of NIR optical imaging (d) and captured image under illumination of 940 nm (e).

940 nm. Although this represents a slight reduction compared to the rigid device, the performance remains competitive with commercial SiPDs (Figure 5d). Further, when scaling the active area to 80 mm², the flexible device still retains 72% of the EQE of the rigid device at 940 nm (EQE = 50.5%, $R = 0.38 \text{ A W}^{-1}$). Comparative analysis with previously reported flexible OPDs demonstrates our device's superior responsivity at 940 nm (Table S11, Supporting Information), representing the highest value documented to date (Figure 5e).

Beyond maintaining excellent EQE and R , the flexible OPDs also achieved J_d s comparable to the rigid device. Specifically, the rigid OPD exhibited a J_d of $1.30 \times 10^{-10} \text{ A cm}^{-2}$ at -0.1 V bias, while the flexible devices demonstrated similarly low J_d s in the pA cm⁻² range ($1.20 \times 10^{-10} \text{ A cm}^{-2}$ for the 5.2 mm² device and $1.56 \times 10^{-10} \text{ A cm}^{-2}$ for the 80 mm² device) (Figure 5f), with the marginally higher values under higher reverse bias potentially linked to the reduced thickness of ITO electrodes on flexible substrates (Figure S35, Supporting Information). The preserved low J_d characteristics at -0.1 V bias enable the flexible OPDs to achieve D_{sh}^* exceeding 10^{13} Jones across 300–1100 nm (Figure 5g). Detailed analysis reveals the D_{sh}^* values of 7.6×10^{13} and 5.4×10^{13} Jones at 940 nm for the flexible OPDs with areas of 5.2 and 80 mm², respectively. Notably, the self-powered OPDs exhibited D_{sh}^* beyond 10^{14} Jones, representing one of the highest reported values for flexible OPDs to date (Figure 5h). To systematically evaluate performance parity, we constructed a radar chart comparing key metrics between large-area flexible OPDs and SiPDs (Figure 5i). The analysis demonstrates equivalent performance in both spectral response range and D_{sh}^* , while the

flexible OPDs maintain significant advantages over silicon counterparts in both scalable device dimensions and mechanical conformability.

The noise spectral density analysis reveals that, in comparison to rigid devices, the S_n of the flexible OPDs exhibits a slight increase, with values of $1.41 \times 10^{-14} \text{ A Hz}^{-0.5}$ for the 5.2 mm² device and $3.15 \times 10^{-14} \text{ A Hz}^{-0.5}$ for the 80 mm² device, respectively (Figure S36a, Supporting Information). Moreover, their superior responsivity allows the flexible OPDs to maintain LDR exceeding 120 dB (Figure S37, Supporting Information). Based on the corresponding R and S_n data, the D^* of the flexible OPDs remains above 10^{12} Jones within the range of 300–1100 nm (Figure S36b, Supporting Information), indicative of highly sensitive photodetection capabilities. Measurement of the noise-equivalent power (NEP) provided further insight, yielding values of 0.06, 0.07, and 0.12 pW Hz^{-0.5} for the rigid, flexible, and large-area flexible devices, respectively (Figure S38, Supporting Information). These correspond to D^* values of 2.8×10^{12} , 2.3×10^{12} , and 1.3×10^{12} Jones at 940 nm, which are lower than those calculated from the integrated noise spectral density (Figure S36b, Supporting Information), owing to the inherent sensitivity of low-frequency NEP measurements to potential residual noise components, which may not be fully resolved within the averaged noise level over the broader bandwidth of the spectral density measurement (Figure S36a, Supporting Information). Furthermore, the flex-OPD (80 mm²) exhibits a $f_{-3 \text{ dB}}$ of 11 kHz, whereas flex-OPD (5.2 mm²) achieves a $f_{-3 \text{ dB}}$ of 33 kHz (Figure S39, Supporting Information), which is adequate to satisfy the shutter requirements of standard cameras.

To evaluate the long-term operational stability, both rigid and flexible BTPV-CN-based OPDs were subjected to unencapsulated storage in ambient air. Remarkably, after 300 hours, both the EQE and R at 940 nm retained values approaching 90% of their initial levels for both device types. The calculated D_{sh}^* remained relatively stable, fluctuating within one order of magnitude of its starting value over this period (Figures S40 and S41, Supporting Information), demonstrating good environmental stability. Crucial for flexible applications, we further evaluated the operational stability under repeated mechanical deformation. The flexible OPD (5.2 mm² active area) was subjected to cyclic bending at different radii (5.50, 4.00, and 3.25 mm), with EQE and dark current (J_d) measured every 50 cycles (Figure S42, Supporting Information). Impressively, under a bending radius of 5.5 mm, the devices endured 350 cycles while preserving 80% of their EQE and an almost unchanged J_d , showcasing excellent bending resilience (Figure S43, Supporting Information). While performance degraded at more stringent radii (e.g., 3.25 mm), this establishes the mechanical operational boundary. In summary, these results underscore the potential of the BTPV-CN-based OPD for use in high-performance flexible photodetection systems.

To demonstrate the practical application potential of these OPDs, the photoplethysmographic (PPG) and single-pixel imaging were performed at 940 nm. The PPG technology is a non-destructive method for heart rate measurement.^[88,89] Its underlying principle involves monitoring the cardiac cycle by detecting variations in light absorption caused by different blood flow states during each heartbeat (Figure 6a). As shown in Figure 6b, all OPDs exhibit distinct diastolic and systolic signals, corresponding to a heart rate of 78 beats per minute. Furthermore, owing to the larger size and curved shape, the flex-OPD (80 mm²) demonstrates a superior signal-to-noise ratio. Based on PPG data, accelerated photoplethysmograph (APG) can be derived through secondary micro-differentiation processing, which can further investigate the parameters of blood circulation. The standard waveform of APG is shown in Figure 6a, where the signals a, b, c, d, and e represent the early systolic positive wave, early systolic negative wave, late systolic reincreasing wave, late systolic redecresing wave, and early diastolic positive wave, respectively. The APG signal pattern measured by our OPDs effectively captures all characteristic waveforms of a normal blood vessel state, indicating its promising potential in health monitoring (Figure 6c). For single-pixel imaging, we established an NIR optical prototype and employed our device as the detector (Figure 6d). An acrylic plate bearing the inscription hollowed-out “SCUT” is positioned between the light source and the detector. The acrylic plate is scanned within this plane, and each position induces the detector to generate a photocurrent signal. By recording both the position and its corresponding photocurrent, we are able to obtain a high-resolution image displaying a distinct “SCUT” word (Figure 6e). This result proved the huge potential of our device in NIR imaging.

3. Conclusion

In conclusion, we reported the first large-area flexible broadband OPDs rivalling silicon photodiodes. Specifically, a novel NIR electron acceptor featuring acceptor–donor–acceptor molec-

ular skeleton and electron-withdrawing cyano-substituted terminal groups, named BTPV-CN, was designed and synthesized. Benefiting from the strong electron push-pull effect, BTPV-CN exhibited a small bandgap of 1.13 eV, consistent with that of silicon, which establishes a spectral foundation for comparability with silicon photodiodes. Moreover, the large molecular dipole moment and electrostatic potential were found for BTPV-CN, which can promote charge generation and enhance intermolecular interaction. By combining with the wide-bandgap polymer donor PBDB-T, the resulting OPDs demonstrated excellent EQE within the spectral range of 300–1100 nm. At a bias of −0.1 V, the peak EQE at 940 nm reached 70%, with the corresponding R value of 0.53 A W^{−1}, marking the best results for OPDs. Additionally, this OPD also exhibited low energy disorder and trap-state density, thereby resulting in an ultralow J_d at picoampere level (1.30×10^{-10} A cm^{−2}) and high detectivity up to 10¹³ Jones. Remarkably, the large-area flexible OPDs can maintain an outstanding D_{sh}^* exceeding 10¹³ Jones, which represents the first large-area flexible OPD rivalling silicon photodiodes. These results demonstrate the huge potential of OPDs in addressing the limitations of silicon photodiodes for application in flexible and wearable electronics.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available in the supplementary material of this article.

Keywords

broadband photodetection, electron acceptor, large-area flexible device, organic photodetectors

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