Efficient SWIR Organic Photodetectors with Spectral Detection Extending to 1.4 µm Using a Benzobisthiadiazole-Based Acceptor

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Organic photodetectors (OPDs) offer significant advantages in biomedical applications, including medical imaging, heart rate monitoring, and tumor therapy. Despite advancements in OPD technology, the efficiency of these devices in the short-wave infrared (SWIR) region remains considerably lower than that of inorganic semiconductors. To tackle this challenge, this study developed an ultra-narrow bandgap acceptor of CS-1, featuring an A-D-A1-D-A structure where benzobisthiadiazole (BBT) serves as the electron-deficient unit A1, which exhibits a wide absorption range from 300 to 1550 nm. This molecular design not only enhances the absorption properties of the material but also improves the overall performance of the OPD device. It is worth noting that the optimal PTB7-Th:CS-1 device realizes a specific detectivity (D_n^*) of 2.96 \times 10¹⁰ Jones at 1.30 μ m, making it one of the most efficient devices at this wavelength to date. Additionally, it demonstrates the high linear dynamic range (LDR) of 91.9 dB even at 1300 nm. These results indicate that the PTB7-Th:CS-1 device significantly enhances detection efficiency in the SWIR region, surpassing most commercially available silicon-based photodetectors. This highlights the significant potential of the BBT unit for achieving high-performance SWIR OPDs.

absorption properties, cost-effective fabrication, and lightweight characteristics.^[1-6] Materials with the first near-infrared region (NIR-I; 700-1000 nm) absorption and second near-infrared region (NIR-II; 1000-1700 nm) radiation are particularly valuable in medical applications like imaging, heart rate monitoring, and tumor ablation due to their deep penetration, high resolution, and efficient photothermal conversion, making them a research focus in biomedicine.[7-10] Despite many important advances in shortwave infrared (SWIR) OPDs, their device performance still lags far behind that of inorganic semiconductors. A key challenge is developing ultra-narrow-band-gap organic semiconductors for efficient SWIR absorption. Extending the absorption range and adjusting the absorption window are particularly important.[11-16] Narrow-bandgap non-fullerene acceptors (NFAs) are emerging as promising materials, offering flexible structures, high absorption coefficients, adjustable energy levels, and controllable crystallinity, with great potential

1. Introduction

Organic photodetectors (OPDs) have attracted considerable attention because of their ease of processing, tunable

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in photovoltaic and OPD applications.^[17–19]

Enhancing the intramolecular charge transfer (ICT) effect is an effective strategy to fine-tune the absorption range of narrow-bandgap organic semiconductors, which is based on the

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-1.74

(a)

(c)

(d)

Energy Level (eV)

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Strong electron affinity

Low lying LUMO level

2.22 Å 2.22 Å

-2.89





Figure 1. a) The calculated LUMO energy levels and chemical structures of benzothiadiazole and its derivatives; b) chemical structure of CS-1; c) the optimal conformation and HOMO/LUMO distribution for CS-1; d) normalized absorption spectra of CS-1; e) UPS curves of CS-1; f) CV curves of Py1, Py6, and Py7 in thin films.

electron push-pull effect between the electron-donating and electron-withdrawing units (the D-A effect).^[20-22] The most common ICT configuration in non-fullerene acceptors is the A-D-A structure, where an electron-donating core (D) is flanked by two electron-accepting end groups (A). By selecting units with varying electron-donating or electron-accepting strengths, the absorption window can be precisely adjusted.^[23–26] On this basis, introducing an electron-withdrawing unit into the central core unit to form the A-D-A₁-D-A configuration can significantly reduce the bandgap, enabling absorption beyond 1000 nm, and even exceeding 1200 nm.^[27–30]

Benzothiadiazole (BT) and its derivatives are widely utilized in the design of NFAs.^[31–34] As shown in **Figure 1**a, BT and its derivatives exhibit low lowest unoccupied molecular orbital (LUMO) energy level (E_{LUMO}), with benzobisthiadiazole (BBT) possessing the lowest at -3.67 eV. A low LUMO energy level is advantageous for electron filling and enhances the ICT effect. BBT is a well-known acceptor unit, featuring a 14π electron conjugated structure, along with the inclusion of a tetravalent sulfur atom, ensuring sufficient electron delocalization, which results in a high electron affinity and reduced bandgap. It has been extensively employed as a building block for synthesizing NIR-II region molecules, showcasing exceptional performance in both biomedical and organic optoelectronic applications. For example, Fan and colleagues synthesized a narrow bandgap polymer, PBBTTT, using BBT as the strong acceptor unit and a thiophene unit as the electron donor.^[35] This polymer features an ultranarrow bandgap and demonstrates excellent hole and electron mobility, making it suitable for high-performance bipolar transistors. In another study, Gurudutt Bhat and co-workers incorporated the ultra-narrow bandgap bipolar polymer of PBBTCD into an organic photosensitized device, effectively detecting nearinfrared (NIR) light.^[36] PBBTCD significantly extends the absorption range up to 1.2 µm and enhances the device's photocurrent responsivity to 0.4 mA/W. Notably, while BBT has been widely used for polymer synthesis, the application of BBT-based small molecules in OPDs remains unexplored.

In this study, we have developed a new ultra-narrow bandgap NFA named CS-1. The absorption onset (λ_{onset}) of the film is 1550 nm using a tangent method. Based on the formula for the optical bandgap, $E_g^{opt} = 1240/\lambda_{onset}$, the bandgap of CS-1 is calculated to be 0.80 eV. The molecular structure of CS-1 follows the A-D-A₁-D-A configuration, incorporating a strong electron-absorbing unit of benzobisthiadiazole as A₁ component, a cyclopentadithiophene as the electron-donating unit, and an IC-2F as the end group. The resulting CS-1 film demonstrates an ultra-broad absorption spectrum ranging from 300 to 1550 nm. The potential mechanism for the narrow bandgap of CS-1 can be attributed

to the A-D-A₁-D-A structure, which enhances the ICT effect between the donor and acceptor units. This charge transfer induces π -electron delocalization, reducing the energy gap between the highest occupied molecular orbital (HOMO) and the LUMO. Additionally, the BTT unit, with its deep LUMO energy level, further stabilizes the LUMO, contributing to the narrowing of the bandgap. Furthermore, the incorporation of quinoidal structures extends the π -conjugation across the molecule, thereby increasing the overlap of molecular orbitals-particularly the HOMO and LUMO-leading to an effective reduction in the bandgap. We improved the morphology of the active layer through successive device optimizations, transitioning from pull-coating to solution dripping and heating to 100 °C. Specifically, under pullcoating conditions, excessive aggregation of BBT resulted in poor external quantum efficiency (EQE) and responsivity. By replacing these conditions with solution drip and applying heat, phase separation was obviously improved, resulting in a high EQE of 1.47% at 1300 nm. Of particular interest, the specific detectivity (D_n^*) of the device exceeds 10^{10} Jones in the wavelength range of 0.78 to 1.35 μ m, reaching a high value of 2.96 \times 10¹⁰ Jones at 1.30 µm, making it one of the most efficient devices reported to date. The device also exhibits a high responsivity of 15.48 mA W⁻¹ at 1.3 nm at 0 V. Furthermore, at 1.3 µm, PTB7-Th:CS-1 device achieves a linear dynamic range (LDR) of 91.9 dB at zero bias, surpassing the commercial PTTQ:PCBM system, which recorded an LDR of 78.7 dB at the same wavelength. Additionally, this device demonstrated a higher photocurrent density ($J_{\rm ph}$) of 1.25×10^{-5} A cm⁻² under this illumination and zero bias. Our results indicate device performance can be significantly enhanced through a rapid film formation process and reveal the great potential of BBT-based NFAs for a wide range of applications in SWIR OPD.

2. Results and Discussion

The synthetic route for CS-1 is provided in Scheme S1 (Supporting Information). Compound 3 was synthesized by Migita-Kosugi-Stille Coupling between [4,4-bis(2-ethylhexyl)-4Hcyclopenta [2,1-b:3,4-b']dithien-2-yl]trimethyl-stannane and dibrominated BBT.^[35,37,38] Subsequently, compound 4 was obtained using the Vilsmeier-Haack reaction. The final target compound, CS-1, was synthesized via Knoevenagel condensation of the intermediate dialdehyde and IC-2F, which features an A-DA₁DA conjugate motif as a non-fullerene acceptor. The intermediates and CS-1 were characterized using ¹H/¹³C NMR and high-resolution mass spectrometry (HRMS). To further broaden the molecular absorption, we attempted to replace the F atom on the terminal group of CS-1 with Cl. However, due to the poor solubility of the chlorinated acceptor, we were unable to obtain a pure product. As exhibited in Figure S1 (Supporting Information), the thermal decomposition temperature of CS-1 at a 5% weight loss is 347 °C, demonstrating its good thermal stability.

Theoretical calculations by using Gaussian 16 at the B3LYP-D3(BJ)/def2-SVP level were conducted to explore the molecular conformation and frontier orbital energy levels of CS-1. As presented in Figure 1c, CS-1 exhibits multiple intramolecular non-covalent interactions, including strong N…H interactions of 2.22 Å and S…N interactions of 2.78 Å. These interactions contribute to the high degree of molecular planarity of CS-1. The calculated HOMO and LUMO energy levels are –5.43 and –4.38 eV, respec-

tively. Furthermore, both the HOMO and LUMO are delocalized across the molecular skeleton, which is advantageous for charge transport.

The UV-vis-NIR absorption spectra of CS-1 in solution and as thin films are presented in Figure 1d. In chlorobenzene solution, CS-1 shows an absorption peak at a wavelength of 1043 nm. In contrast, the CS-1 film exhibits broad absorption across the 300-1550 nm range, with a red-shifted maximum absorption peak at 1288 nm with an absorption coefficient of 1.02×10^5 cm⁻¹ (Figure S2, Supporting Information), compared to the solution, indicating an ultra-narrow bandgap of 0.80 eV based on the absorption edges (1550 nm). Interestingly, different from common A-D-A₁-D-A, CS-1 exhibits a strong absorption spectrum in the 400-900 nm range, which may be attributed to the local ICT effect induced by the strong electron-withdrawing nature of the BBT unit. It is important to highlight that CS-1 represents the A-D-A type NFAs with the smallest band gap reported to date. Notably, the PTB7-Th:CS-1 blend exhibits a broad absorption spectrum ranging from 300 to 1550 nm, with high absorption coefficients of 4.8×10^4 cm⁻¹ at 705 nm and 4.7×10^4 cm⁻¹ at 1130 nm. We further evaluated the stability of PTB7-Th:CS-1 BHJ films under two conditions: dark storage in a nitrogen atmosphere and exposure to continuous 1-sun illumination in ambient air (Figure S2, Supporting Information). The absorption behavior of the films remained largely unchanged, with only slight variations observed in the shortwave infrared range (900-1400 nm). These results indicate that the films exhibit excellent intrinsic stability and photostability.

In addition, ultraviolet photoelectron spectroscopy (UPS) was employed to investigate the energy levels of CS-1 (Figure 1e). The work function and ionization potential were determined to be 0.87 eV and 4.49 eV, respectively. Based on the optical band gap and the ionization potential, the calculated LUMO energy level is 3.69 eV. Furthermore, we also conducted cyclic voltammetry (CV) measurements to investigate the E_{IUMO} and HOMO energy level (E_{HOMO}) of CS-1 and PTB7-Th. As shown in Figure 1f, the E_{LUMO}/E_{HOMO} values are estimated to be -4.26/-5.29 eV for CS-1, and -3.26/-5.41 eV for PTB7-Th, respectively. The observation that the LUMO energy level of the acceptor is higher than that of the donor from CV curves is consistent with the test results obtained from ultraviolet UPS. It is worth noting that the HOMO level of CS-1 is higher than that of PTB7-Th, which could result in hole accumulation at the donor-acceptor interface, thereby reducing the charge extraction efficiency and ultimately lowering the EQE of the device.

Considering the narrow bandgap and good molecular planarity of CS-1, we adopted a device structure comprising ITO/ZnO/PTB7-Th:CS-1/MoO₃/Ag (**Figure 2a**). To optimize the active layer morphology and enhance device performance, thin films were fabricated using three different methods: static spin coating or called pull method, dynamic spin coating, or called solution dripping, and heated dynamic spin coating (Figure 2b). We noted that the heated dynamic spin coating method was performed by preheating the solution to 100 °C and then directly depositing it by spin coating at 3000 rpm to prepare the active layer film. This process yielded three distinct thin film samples: PTB7-Th:CS-1 (pull), PTB7-Th:CS-1 (drip), and PTB7-Th:CS-1 (100 °C). The photodetection performance of the OPDs is illustrated in Figure 2c–h, which highlights the influence of fabrication www.advancedsciencenews.com

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Figure 2. a) A schematic diagram of the device structure; b) a schematic diagram of device optimization process; c) EQE curves, d) responsivity, e) characteristic *J*--V curves in the dark condition, f) EIS curves g) the noise current spectrum under zero bias for PTB7-Th:CS-1 (100 °C) device, and h) detectivities for PTB7-Th:CS-1 device under three different conditions.

techniques on key parameters such as photocurrent, responsivity, and detectivity. The detailed device optimization process, including the details of 1-chloronaphthalene (CN) and 1,8-diiodooctane (DIO) as additives, is presented in Figures S3–S5 and Tables S1 and S2 (Supporting Information). The optimal thickness of the active layer is 100 nm. It should be noted that the other specific detectivity calculated by dark current, $D_{\rm sh}^{**}$, in this context is overestimated and is therefore provided solely as a reference to illustrate the trend in device condition optimization.

The device based on PTB7-Th:CS-1 (pull) achieves a maximum EQE of 0.08% across the broad wavelength range of 300–1400 nm. For the PTB7-Th:CS-1 (drip) device, a significantly higher maximum EQE of 0.35% is observed at 1300 nm. Most notably, the PTB7-Th (100 °C) device exhibits the best performance, reaching a high EQE of 1.47% at 1300 nm, with an EQE exceeding 3% in the NIR range between 930 and 1220 nm. These results highlight the broad spectral response of the PTB7-Th:CS-1 based devices, covering the range from 300 to 1400 nm, which notably surpasses the typical response limit of \approx 1100 nm observed in conventional silicon-based photodetectors.^[39–41] It is worth men-

tioning that the relatively low EQE of the PTB7-Th:CS-1 device may be attributed to the misalignment of the donor-acceptor energy levels, which can result in hole accumulation within the device.

The responsivity (R), as an important index of photodetector performance, is defined as the ratio of generated photocurrent to incident light intensity, which can be calculated as:

$$R = EQE \times \lambda / 1240 \times 100\%$$
(1)

where λ is the wavelength of incident light. Compared to the PTB7-Th:CS-1 (pull) based device,^[42,43] the devices utilizing PTB7-Th:CS-1 (drip) and PTB7-Th:CS-1 (100 °C) demonstrate higher R values over a broad spectral range from 300 to 1400 nm, primarily due to their enhanced EQE. Under zero bias conditions, the PTB7-Th:CS-1 (drip) based device achieves a high responsivity of 3.74 mA W⁻¹ at 1300 nm, with responsivity exceeding 10 mA W⁻¹ in the 920–1190 nm range. In contrast, the device based on PTB7-Th:CS-1 (100 °C) exhibits a higher responsivity of 15.4 mA W⁻¹ at 1300 nm and maintains responsivity above 20.8

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mA W^{-1} across the 910–1280 nm range. High responsivity indicates that these devices possess strong photoelectric conversion capabilities, showcasing significant potential for various applications.

The current density–voltage (*J*–*V*) characteristics of OPDs based on PTB7-Th:CS-1 processed by different methods— specifically, PTB7-Th:CS-1 (pull), PTB7-Th:CS-1 (drip), and PTB7-Th:CS-1 (100 °C) at zero bias—reveal distinct dark current values (Figure 2e). The corresponding dark current density (*J*_d) values for the three devices are 2.53×10^{-9} , 3.51×10^{-9} , and 4.32×10^{-9} A cm⁻², respectively. Due to the low *J*_d value, the PTB7-Th:CS-1 (pull) devices exhibit a low specific detectivity. Electrochemical impedance spectroscopy (EIS) exhibits a similar trend in noise behavior, as shown in Figure 2f. As a fact, the *D*_{sh}^{*} calculated solely based on *J*_d as a source of noise is often inaccurate and frequently overestimated.^[44] In contrast, using white noise for the calculation yields a specific detectivity that is closer to the true value. The equation defining *D*_n^{*} is as follows:

$$D_n^* = \left(R\sqrt{A}\Delta f \right) / \left(I_{n,rms} \right)$$
⁽²⁾

where A represents the device's response area, Δf denotes the measurement system's bandwidth (0.1 Hz), and $I_{n rms}$ (100-1000 Hz) refers to the white noise current. The noise current spectrum at zero bias is presented in Figure 2g. The I_{n.ms} of the PTB7-Th:CS-1 (pull) device at zero bias voltage is 1.66×10^{-12} A Hz^{-1/2}, which is also significantly lower than that of the pull configuration. Furthermore, under the reverse bias conditions (-1, -2, -2)and -4 V), there is a significant increase in noise current power across all spectral ranges when compared to the device operating at zero bias, as shown in Figure S6 and Table S3 (Supporting Information). This rise in noise current, compared to the zero-bias conditions, is unfavorable for specific detectivity as it increases the total noise floor and reduces the detector's sensitivity to weak signals. The noise current of the reference device exhibits a significant increase, rising from 1.66 \times 10⁻¹² A Hz^{-0.5} to 2.17 \times 10^{-7} A Hz^{-0.5}. In contrast, the noise current of the device with dynamic heating increases from 3.31 \times 10 $^{-14}$ A Hz $^{-0.5}$ to 3.29 \times 10^{-8} A Hz^{-0.5}. This trend is consistent with findings in the literature, where the application of reverse bias, often leads to higher noise levels due to increased dark current and charge injection from the electrodes. The charge injection at reverse bias voltages is predominantly attributed to energy-level misalignment, leading to a significant reduction in D_n^* at -4 V compared to 0 V, with a decrease of \approx 3–4 orders of magnitude. Nonetheless, the impact of noise current on energy-level misalignment remains minimal at 0 V. As indicated in Figure 2h and Table S4 (Supporting Information), the corresponding ${D_{\rm n}}^*$ are calculated to be 3.40×10^7 Jones for PTB7-Th:CS-1 (pull) device, 1.18×10^{10} Jones for PTB7-Th:CS-1 (drip) device, and 2.96×10^{10} Jones for PTB7-Th:CS-1 (100 °C) device, respectively. It is worth mentioning that the detectivity of 2.96×10^{10} Jones at a wavelength of 1300 nm is considered to be an excellent performance among current SWIR OPDs (Figure S7 and Table S5, Supporting Information). The enhanced D_n* of the PTB7-Th:CS-1 (100 °C) device can be attributed to its superior J_d and higher EQE, achieved through precise optimization of the processing conditions. These findings show that thermal annealing and spin-coating processes have a significant

impact on noise and responsivity, directly improving the detection performance of PTB7-Th-based OPDs in both the visible and near-infrared regions.^[45,46]

In OPD applications, the J_d and photocurrent density (J_{ph}) are critical factors influencing the device performance of OPDs, with a high switching ratio ($J_{\rm ph}/J_{\rm d}$, on-off ratio) being is essential for achieving optimal results.^[43] The switching ratio of the material was evaluated as illustrated in Figure 3a-d. The switching ratios of the three OPDs were 1.46×10^1 , 2.94×10^2 , and 2.94×10^3 at 1300 nm light and zero bias, respectively, demonstrating a significant increase in that order. Notably, the device based on PTB7-Th:CS-1 (100 °C) exhibited a high $J_{\rm ph}$ of 1.25 × 10⁻⁵ A cm⁻² at 1300 nm light and zero bias, indicating high exciton dissociation efficiency and charge collection efficiency. Both the low dark current and high photocurrent further corroborated the high EQE and R observed over a broad spectral range of 300-1400 nm, underscoring the device's enhanced performance. Apart from studying the OPD performance, we also evaluated the device's operational stability and the changes in EQE under conditions of dark storage and continuous illumination (Figure S8, Supporting Information). After 24 consecutive measurements, the dark current density of the devices showed no obvious increase. Furthermore, we tested the EQE stability under two conditions: dark storage in a glove box and continuous 1-sun illumination in ambient air. The results indicated that after 400 h of continuous illumination in air, the devices maintained 88% of their initial EQE. Meanwhile, devices stored in the dark retained nearly unchanged EQE after 400 h, demonstrating excellent photostability and dark storage stability.

The response time and linear dynamic range (LDR) of photodetectors under SWIR light irradiation were investigated, as these factors reflect the ability of photodetectors to accurately detect and process rapidly changing and complex optical signals. The LDR represents the region where the electrical output of a photodetector maintains a linear relationship with light intensity. Figure 3e-h illustrates the LDRs of three devices under 1300 nm illumination.^[47,48] Specifically, the LDR for the PTB7-Th:CS-1 (pull) device was 43.4 dB, and for the PTB7-Th:CS-1 (drip) device, it was 73.9 dB at 1.3 µm. In contrast, the LDR of the PTB7-Th:CS-1 (100 °C) device at 1.3 µm was significantly enhanced to 91.9 dB, surpassing that of the commercial PTTQ:^[49,50]PCBM system, which had an LDR of 78.7 dB at the same wavelength. We have supplemented the analysis of PTTQ:PCBM with data on noise current, EQE, and D_n^* , as shown in Figure S9a-c (Supporting Information). The noise current of PTTQ:PCBM system is 5.92 \times 10⁻¹⁴ A Hz^{-0.5} at 0 V, increasing to 2.76 \times 10⁻¹¹ A Hz^{-0.5} at -1 V. These results indicate the ease of carrier injections under reverse bias, even in an energy-level- matched active layer system. At a wavelength of 1300 nm and 0 V, the EQE of the device was determined to be 2.19%, with a responsivity of 23.1 mA W^{-1} . The D_n^* reached 2.46 $\times 10^{10}$ Jones. However, the active layer system consisting of low bandgap polymer and fullerene derivative may exhibit greater structural disorder and defects, which contribute to an increase in dark current. This improvement in LDR can be attributed primarily to the reduced dark current, which helps minimize noise.

Furthermore, we calculated the noise-equivalent power (NEP) and detectivity from the LDR, using the NEP limit to define the threshold at which the LDR signal fades into noise



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αV⁽ⁿ



Figure 3. Characteristic *J*–V curves for the devices based on a) PTB7-Th:CS-1 (pull), b) PTB7-Th:CS-1 (drip), c) PTB7-Th:CS-1 (100 °C) and d) PTTQ:PCBM; linear dynamic range (red lines represent linear fitting) for the devices based on e) PTB7-Th:CS-1 (pull), f) PTB7-Th:CS-1 (drip), g) PTB7-Th:CS-1 (100 °C) and h) PTTQ:PCBM; i) response time of the OPD based on PTB7-Th:CS-1 (100 °C); j) the electron mobility of three PTB7-Th:CS-1 devices; k) *J*–V curves of the electronic devices.

(Figure 3e-h; Figure S10, Supporting Information). The calculation of the NEP is based on the formula: $NEP_{limit} = I_{shot}/R_{1300}$, where I_{shot} represents the shot current and R₁₃₀₀ is the responsivity at 1300 nm. The detectivity is calculated as: $D_n^* = (AB)^{0.5}/NEP$, where A is the device area and B = 1 Hz. Figure S11a (Supporting Information) shows the NEP of the standard Ge detector and the samples at 1300 nm. The NEP of Ge was measured as 4.02 \times 10^{-13} W Hz^{-0.5}, while the NEP trends of the samples are consistent with those of the LDR. The optimized NEP values were 1.49 \times 10 $^{-10}$ W Hz $^{-0.5}$ for PTB7-Th:CS-1 and 2.18 \times 10 $^{-10}$ W Hz $^{-0.5}$ for PTTQ:PCBM. It is worth noting that the D_n^* calculated from noise currents and NEPs differ by approximately half an order to one order of magnitude (Figure S11b, Supporting Information). This discrepancy may be attributed to environmental constraints during measurements, instability under low light intensities, and the higher sensitivity of organic thermal noise compared to inorganic systems. Despite these challenges, the optimized devices maintain a robust linear dynamic range, demonstrating the feasibility of the novel small-molecule system for short-wave infrared imaging applications.

The experimental results demonstrate that these self-powered OPDs are capable of maintaining signal fidelity across an exceptionally broad spectrum of light intensities. The current-voltage curves of the OPDs were analyzed with a 1310 nm laser source (MDL-H-1310, CNI), and the response and -3dB bandwidth were measured with a 1300 nm LED (M1300L4, THORLABs) light

source. A Ge detector (Enlitech RC-G108018-E) was employed as a standard reference for source intensity calibration and comparative measurements. The on-off ratio was evaluated at various bias voltages under an intensity of 10 mW, and the performance was compared to that of the Ge detector, as shown in Figure S12a,b and Table S6 (Supporting Information). Notably, the onoff ratios for both the Ge detector and PTB7-Th:CS-1 decreased beyond -0.1 V. This observation reaffirms that the low on-off ratios are associated with the injection of an external electric field, predominantly influenced by thermal carriers at 0 V.^[51] To further investigate the response rate of the OPDs, the -3 dB bandwidth of the Ge detector was measured to be \approx 70 kHz, as shown in Figure 3i and Figure S12c,d (Supporting Information), with the EQE of the standard devices presented in Figure S12e (Supporting Information). In comparison, the bandwidth of the PTB7-Th:CS-1 system was ≈19.6 kHz, and that of the PTTQ:PCBM system was \approx 14.2 kHz. The rise time and fall time of the PTB7-Th:CS-1 system were calculated to be 5.8 and 7.4 µs, respectively. The slower response of the OPDs can be primarily attributed to their low carrier mobility. Furthermore, the -3dB bandwidth of the Ge detector was measured to be \approx 2.8 kHz, while the PTB7-Th:CS-1 system exhibited a bandwidth of \approx 1.5 kHz. These supplementary results confirm that the response time of the PTB7-Th:CS-1 system is comparable to that of the Ge detector.

To investigate the effect of charge transport on SWIR photodetection, we measured the electron mobilities of blend films using the space-charge-limited current (SCLC) method (Figure 3j).^[52] The electron mobility($\mu_{\rm e}$) was measured as 1.02×10^{-4} cm² V⁻¹·s⁻¹ for the PTB7-Th:CS-1 (pull) film, and 1.31×10^{-4} cm² V⁻¹·s⁻¹ for the PTB7-Th:CS-1 (drip) film. Notably, the PTB7-Th:CS-1 (100 °C) film exhibited significantly higher electron mobility, with $\mu_{\rm e} = 1.65 \times 10^{-4}$ cm² V⁻¹·s⁻¹. This enhancement in mobility explains why the PTB7-Th:CS-1 (100 °C) based organic OPD demonstrates superior EQE and D_n^* , which can be attributed to improved charge transport achieved through the optimization of processing conditions.

The defect states (N_t) were calculated from the J-V curves of the electronic devices using the equation $N_t = 2\epsilon_r \epsilon_0 V_{TFL}/(qL^2)$, where L is the thickness of the active layer and V_{TFL} is the trap filling limit voltage (Figure 3k).^[53,54] The V_{TFL} values for PTB7-Th:CS-1 (pull) and PTB7-Th:CS-1 (drip) films were determined to be 1.31 and 1.25 eV, respectively, with the corresponding N_t values calculated as 6.27×10^{16} and 5.16×10^{16} cm⁻³. After thermal annealing at 100 °C, the V_{TFL} of the device based on PTB7-Th:CS-1 (100 °C) decreased to 1.02 eV, resulting in a low N_t value of 3.95 $\times 10^{16}$ cm⁻³, which is significantly reduced compared to the other two devices. This reduction in defect states, combined with the improved charge transport, contributes to the enhanced specific detectivity of PTB7-Th:CS-1 (100 °C) device. Furthermore, we analyzed PTB7-Th:CS-1 (100 °C) and PTTQ:PCBM using deep-level transient spectroscopy (DLTS), a technique designed to characterize trap states within a diode. In this study, the current-DLTS method implemented in Paios was used to record the filling and release of traps in response to a reverse voltage pulse, as shown in Figure S13a,b (Supporting Information). The rapid rise in current observed within 1~100 µs is attributed to the release of carriers from shallow traps. These traps are located near the conduction or valence band, requiring lower energy for carrier release, which occurs within a short timeframe. Conversely, in the 1-100 µs range, the release of carriers from deeper traps dominates, taking longer due to the slower thermal excitation process associated with deeper energy levels. We measured the devices at bias voltage from -1 to -3 V and observed distinct current-DLTS responses for devices fabricated from PTB7-Th:CS-1 and PTTQ:PCBM. Prior to 100 µs, carriers are predominantly generated from shallow traps, as evidenced by variations in bias voltage. This timescale corresponds to a relatively low energy barrier for trapping carriers, allowing their efficient release back into the conduction or valence band. For PTB7-Th:CS-1 system, the maximum release occurs at $\approx 0.1 \,\mu$ s, while for PTTQ:PCBM system, it peaks at around 0.07 µs. This suggests that the energyscale mismatch in PTB7-Th:CS-1 system results in shallow traps with higher energy barriers. Interestingly, in the $1 \sim 100 \,\mu s$ range, no significant current change is observed for PTTQ:PCBM system under varying bias voltages, indicating that carrier release is governed by deep traps, leading to slower release rates. In contrast, PTB7-Th:CS-1 system shows no notable carrier release during this period, possibly due to its non-fullerene architecture, which appears to reduce the influence of deep traps despite the energy-scale mismatch. Fullerene systems, however, exhibit pronounced deep trap effects, contributing to slower responses, increased dark current, and reduced stability. We further analyzed carrier dynamics and trap states through open-circuit voltage decay (OCVD) measurements, which track the change in opencircuit voltage (V_{OC}) under varying light intensities. As illustrated

in Figure S13c,d (Supporting Information), higher light intensities reduce the rate of voltage decay due to the filling of shallow traps by photogenerated carriers, thereby extending carrier lifetimes. Conversely, at lower light intensities, the prolonged voltage decay in PTTQ:PCBM system suggests the influence of deep traps. Additionally, the energy-level mismatch in PTB7-Th:CS-1 system leads to energy losses, resulting in a smaller $V_{\rm OC}$ compared to PTTQ within the same energy-gap system. In conclusion, our analysis of trap state distribution and carrier dynamics reveals that the low bandgap polymer with small-molecule (non-fullerene) system (PTB7-Th:CS-1) effectively mitigates the impact of deep traps compared to the conventional low bandgap polymer with fullerene system (PTTQ:PCBM). This improvement underscores the potential of non-fullerene architectures in enhancing device stability and performance.

In addition to analyzing trap distribution and carrier dynamics through DLTS and OCVD, Figure S14a,b (Supporting Information) presents injection charge extraction by linearly increasing voltage (CELIV) measurements, which were employed to investigate the effects of charge injection and extraction as well as to calculate the CELIV mobility. The observed oscillations and decreases are primarily attributed to variations in the electric field distribution. The results indicate that PTB7-Th:CS-1 exhibits a higher plateau current compared to PTTQ:PCBM at 0 V, suggesting that PTB7-Th:CS-1 facilitates better charge extraction. Under bias conditions, PTTQ:PCBM shows a substantial increase in extracted charge, likely due to the influence of deep traps that lead to higher device loading. The peak carrier mobility was calculated using the equation: $\mu = 2d^2/3Vt_{max}$, where d is the active layer thickness (m), V is the applied voltage, and t_{max} is the time to peak. The calculated CELIV mobilities were \approx 4.48 × $10^{-5} \text{ cm}^2 \text{ (V} \cdot \text{s})^{-1}$ for PTTQ:PCBM and $3.64 \times 10^{-5} \text{ cm}^2 \text{ (V} \cdot \text{s})^{-1}$ for PTB7-Th:CS-1. The lower mobility of PTB7-Th:CS-1 is attributed to charge accumulation and its inherently low hole mobility, which reduces the overall device mobility. Furthermore, the imbalance between electron and hole mobilities contributes to the lower EQE observed in PTB7-Th:CS-1.

The morphology of the active layer is a key factor in determining the performance of the SWIR OPDs, which is closely related to exciton dynamics, carrier transport, and recombination. Grazing-incidence wide-angle X-ray scattering (GIWAXS) was first utilized to study the molecular stacking and crystallization characteristics of pure and blended films.^[55] As shown in Figures 4 and S15 (Supporting Information), the CS-1 (pull) film exhibits a microstructure with multiple crystalline phases, likely arising from lattice defects induced by the slow film formation process. In contrast, altering the spin coating conditions significantly alleviated the strong diffraction signals and multiple diffraction peaks observed in the CS-1 (drip) film. Furthermore, after thermal annealing at 100 °C, the microstructures of multiple crystalline phases are basically eliminated, and the CS-1 (100 °C) film displayed a distinct (010) diffraction peak at 1.85 $Å^{-1}$ in the out-of-plane (OP) direction and a (100) diffraction peak at 0.27 Å⁻¹ in the in-plane (IP) direction. Similarly, we also used GIWAXS to analyze the diffraction signals of the donor PTB7-Th under the same three conditions (Figure S16, Supporting Information). Notably, PTB7-Th exhibited fewer diffraction signals during slow film formation, likely due to its intrinsic weak aggregation characteristics. When the donor and acceptor were

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Figure 4. a) 2D GIWAXS patterns of neat CS-1 films; b) 2D GIWAXS patterns and c) in-plane and out-of-plane line-cut profiles from 2D GIWAXS data of PTB7-Th:CS-1 blend films.

blended, the resulting mixed film retained the properties of the pure acceptor film. Specifically, during the slow film formation process, the mixed film displayed characteristics of multiphase crystallization. Following thermal annealing at 100 °C, the microstructures associated with multiple crystalline phases were eliminated, resulting in an improved crystalline coherence length (pull: 32.4 Å; drip: 33.2 Å; 100 °C: 35.3 Å) in the OP direction. The GIWAXS results indicate that a rapid film formation process effectively suppresses multiphase crystallization and enhances the crystalline order of the material.

Then, we performed a detailed investigation of the active layer morphology using atomic force microscopy (AFM) and optical microscopy (OM) (Figure 5). In the AFM images, the PTB7-Th:CS-1 (pull) film exhibited a significantly high root mean square (RMS) roughness of 21.3 nm, which can be attributed to the excessive aggregation of CS-1 during the slow film formation process. This excessive aggregation leads to poor film uniformity and suboptimal device performance. In contrast, the blend film prepared via drip-in spin coating and subsequently annealed at 100 °C showed a drastically reduced RMS roughness of 3.83 nm, indicating that the over-aggregation of CS-1 was effectively suppressed. This reduced roughness may be a key factor in the enhanced performance of the OPD devices. The OM images further corroborate these findings. The PTB7-Th:CS-1 (pull) film suffers from severe aggregation, which negatively impacts film uniformity and device functionality. In comparison, the PTB7-Th:CS-1 (100 °C) film exhibited much better uniformity, a critical parameter for optimizing OPD performance. These OM observations are consistent with the AFM results, highlighting the importance of processing conditions in controlling the morphology and ultimately improving device efficiency. By reducing aggregation and achieving a smoother, more uniform film, the device's overall performance, especially in terms of photoresponse, can be significantly enhanced.

To demonstrate the feasibility of our OPD for SWIR detection, we conducted a single-pixel imaging process to reconstruct a target image projected by a SWIR light source (MDL-H-1310, CNI laser). The experimental setup is schematically illustrated in **Figure 6a**. A mask pattern, defined by the "MCUT" letters imprinted on a transparent substrate, was placed between the



Figure 5. The OM images (left) and AFM height images of three PTB7-Th:CS-1 blend films.

photodetector and a collimated laser beam with a wavelength of 1310 nm. The mask was mounted on a dual-axis translation stage (Thorlabs MTS50-Z8) that allowed precise movement in both the horizontal (X-axis) and vertical (Y-axis) directions, with a resolution of 200×100 pixels (step size of 0.05 mm). As the transmitted SWIR light passed through the "MCUT" pattern, we recorded the photocurrent from the source meter (Keithley 2636B) for image reconstruction. It is important to note that the response photocurrent of the photodetector was collected without applying a bias voltage. More details about the system setup can be found in Figure S17 (Supporting Information). As shown in Figure 6b,c, we successfully reconstructed intact images of the "MCUT" using an indium gallium arsenide (InGaAs) photodetector (Newport 818-IG/DB) and the proposed SWIR OPD under 1310 nm illumination. Although our proposed OPD exhibited a lower photoresponse under the same incident light intensity (0.24 µW cm⁻²), the reconstructed image reveals welldefined boundaries and excellent fidelity to the original pattern,

highlighting its capability to capture intricate details. These results underscore the significant potential of OPDs for SWIR optical imaging, positioning them as promising candidates for advancing next-generation low-cost imaging technologies within the SWIR spectral range.

In conclusion, we propose a novel narrow bandgap nonfullerene acceptor, CS-1, which incorporates a benzobisthiadiazole (A₁) as the central electron-withdrawing unit, combined with dithiophene-cyclopentadiene (D) and IC-2F end groups to form an A-D-A₁-D-A architecture. This molecular design achieves an ultralow bandgap of 0.8 eV and a broad absorption range of 300– 1550 nm. Through continuous optimization of the device preparation process, optimized devices exhibit a significant improvement in detectivity compared to unoptimized ones. At zero bias, the PTB7-Th:CS-1 (100 °C) device exhibits a D_n^* exceeding 10^{10} Jones within the wavelength range of 0.78 to 1.35 µm, achieving a high D_n^* of 2.96 × 10^{10} Jones and responsivity of 15.48 mA W⁻¹ at 1.30 µm, ranking among the most efficient devices reported. www.advancedsciencenews.com

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SWIR laser

(a)

(b)

om Mask Photodetector (InGaAs or Cs-1) (C)



Figure 6. a) The schematic illustrations of the single-pixel SWIR imaging setup; The reconstructed SWIR images based on the photocurrent reading from b) a commercial InGaAs photodetector and c) the OPD proposed in this work under 1310 nm illumination.

Furthermore, a linear dynamic range of 91.9 dB, along with a rapid response time is recorded, highlighting its substantial potential for optoelectronic communication. These findings indicate that the CS-1-based devices possess a broad optical response range and excellent detection capabilities, making them well-suited for advanced biomedical and optoelectronic applications. This study underscores the great potential of acceptors incorporating BTT units for achieving high-performance OPDs.

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 from the corresponding author upon reasonable request.

Keywords

benzobisthiadiazole, detectivity, non-fullerene acceptor, organic photodetector, short-wavelength infrared region

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