

RESEARCH ARTICLE

A Narrow-Bandgap Fused-Ring Acceptor for Organic Short-Wavelength Infrared Imager

 Wenliang Chen^{1,2} | Xiaoyu Zhu^{1,2} | Junhui Miao¹ | Jun Liu^{1,2}  | Lixiang Wang^{1,2} 
¹State Key Laboratory of Polymer Science and Technology, Changchun Institute of Applied Chemistry, Chinese Academy of Sciences, Changchun, P. R. China |

²School of Applied Chemistry and Engineering, University of Science and Technology of China, Hefei, P. R. China

Correspondence: Junhui Miao (jhmiao@ciac.ac.cn) | Jun Liu (liujun@ciac.ac.cn)

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ABSTRACT

Organic short-wavelength infrared (SWIR) imaging is a promising technology for consumer electronics and a broad range of other applications. The bottleneck of organic SWIR imaging is the lack of electron acceptors with ultrasmall bandgaps. Herein, we develop ultrasmall-bandgap electron acceptors by cyclizing the exocyclic vinylene linkers of a typical acceptor–donor–acceptor (A–D–A) electron acceptor with tetrafluorobenzene moieties. This tetrafluorobenzene cyclization redshifts the absorption spectrum by 224 nm and reduces the trap density of states of the neat film by nearly one order of magnitude. The resulting organic photodetector exhibits a peak responsivity of 0.36 A W^{−1}, a noise current of 1.41 × 10^{−11} A Hz^{−1/2} at 1 kHz, a specific detectivity exceeding 10⁹ Jones at 1140 nm under −1 V bias, and a linear dynamic range of 82 dB. Monolithic integration of this organic photodetector with thin-film transistor arrays yields a 256 × 256 pixel imager capable of high-contrast SWIR imaging. This work establishes tetrafluorobenzene cyclization as a powerful molecular paradigm for ultrasmall bandgap organic semiconductors, paving the way for low-cost, heavy-metal-free SWIR imagers in consumer electronics.

1 | Introduction

Short-wavelength infrared (SWIR) light in the spectral range of 1.0–1.7 μm can penetrate through smoke, fog, and biological tissues. SWIR imaging has received increasing attention due to its widespread applications in remote sensing, machine vision, medical imaging, and artificial intelligence [1–7]. The commercial SWIR imaging technique adopts inorganic semiconductors, such as indium gallium arsenide (InGaAs), as photoactive layers and suffers from high production cost. They are mainly used in cost-insensitive military applications and are not suitable for the widespread domestic applications. The newly emerging SWIR imaging technology is based on quantum dots (QDs) and solution-processing, which has the advantage of low cost and is on the early stage of commercialization. However, the heavy metal toxicity concerns of QD-based SWIR imaging technology

precludes their use in consumer electronics. Organic SWIR imaging using organic photoactive layers is an ideal candidate for consumer electronics because of its low-cost solution processing and no heavy metal toxicity [8–14]. Nevertheless, the development of organic SWIR imagers remains in its infancy because of the scarcity of organic materials capable of SWIR light absorption [15–19]. Moreover, aiming at high-quality organic SWIR imaging, noise current density must be substantially suppressed and responsivity need to be significantly enhanced for sensitive photodetection.

Classic electron acceptors, which are used in organic photoactive layers of organic solar cells (OSCs) or organic photodetectors (OPDs), possess small bandgap and exhibit light absorption in the near-infrared (NIR) region. To decrease their bandgap and redshift their absorption into SWIR region, the general strategy

is to introduce quinoidal moieties to the core, terminals, or the bridges of these electron acceptors. For synthesis availability, such SWIR-absorbing electron acceptors typically adopt non-fused-ring chemical structures. Notably, non-fused-ring molecular skeletons give rise to increased rotational degrees of freedom, leading to enhanced static conformational disorder and higher trap densities, which are detrimental to low noise current and high detectivity in photodetection [20–31]. Therefore, to advance the development of organic SWIR imagers, it is essential yet challenging to design electron acceptors that simultaneously feature ultrasmall bandgaps and suppressed molecular rotation.

All-fused-ring molecular architectures have emerged as a promising approach to simultaneously achieve narrow bandgaps and low trap densities. Zheng et al. reported the all-fused-ring non-fullerene acceptor with an electron-rich 4,8-dihydropentaleno[1,2-*b*:4,5-*b'*]dithieno[2,3-*d*]-thiophene (PDTT) core and two 2-(5,6-difluoro-2,3-dihydro-1*H*-inden-1-ylidene)-malononitrile terminal units for NIR OPDs, achieving a shot-noise-limited specific detectivity (D_{sh}^*) of 1.35×10^{13} Jones at 890 nm [32]. Our group developed an n-type all-fused-ring ADA'DA molecule (FM2) with 14 π -conjugated rings, electron-deficient benzotriazole core, electron-rich fused thiophene-pyrrole bridges, and malononitrile terminals, achieving a dark current density as low as 2.01×10^{-10} A cm $^{-2}$, and a D_{sh}^* exceeding 10^{13} Jones [33]. Recently, a fully fluorinated all-fused-ring acceptor (FM-8F) was designed to improve photoinduced charge generation [34]. Despite these advances, all-fused-ring acceptors reported to date have been confined to the NIR region; extending the photoresponse into the SWIR range (>1000 nm) while preserving low trap density remains an outstanding challenge.

Herein, we adopt a tetrafluorobenzene cyclization strategy to address this challenge. By cyclizing the two exocyclic vinylene linkers of a classical electron acceptor with tetrafluorobenzene moieties, we develop a new fused-ring electron acceptor with SWIR light absorption. This tetrafluorobenzene cyclization leads to not only redshifted absorption spectrum by 224 nm for SWIR light absorption, but also minimized trap density of states by one order of magnitude for suppressed noise current in OPDs. As a result, the OPDs exhibit a SWIR photoresponse extending to 1300 nm, with a peak responsivity (R) of 0.36 A W $^{-1}$, a noise current of 1.41×10^{-11} A Hz $^{-1/2}$ at 1 kHz, and a specific detectivity (D^*) of 7.17×10^9 Jones at 1140 nm under -1 bias. This excellent device performance enables us to fabricate 256×256 pixel organic SWIR imagers by monolithically integrating the SWIR OPDs with thin-film transistor (TFT) panels.

2 | Results and Discussion

The state-of-the-art electron acceptors adopt acceptor–donor–acceptor (A–D–A) architecture, in which a donor (D) core is connected to two acceptor (A) terminals via exocyclic vinylene linkers. These electron acceptors exhibit light absorption in the visible and NIR region (wavelength: $\lambda < 1000$ nm) and have been widely used in OSCs and NIR OPDs [33–35]. Figure 1 shows an example, 2,2'-((2*Z*,2'*Z*)-((6-(2-butyloctyl)-12,13-bis(2-hexyldecyl)-12,13-dihydro-6*H*-thieno[2'',3'':4',5']thieno[2',3':4,5]pyrrolo[3,2-*g*]thieno[2',3':4,5]thieno[3,2-*b*][1,2,3]triazolo[4,5-*e*]indole-2,10-diyl)bis(methanylylidene))bis(5,6-dichloro-3-oxo-

2,3-dihydro-1*H*-indene-2,1-diylidene))dimalononitrile (FM7-N). To develop electron acceptors with SWIR light absorption, we cyclize the two exocyclic vinylene linkers with two tetrafluorobenzene moieties and design a new molecule, 2,2'-((2*Z*,2'*Z*)-(10-(2-butyloctyl)-4,5,6,7,13,14,15,16-octafluoro-1,19-bis(2-hexyldecyl)-10,19-dihydro-1*H*-indeno[1''',2''':4'',5'']thieno[2'',3'':4',5']thieno[2',3':4,5]pyrrolo[3,2-*g*]indeno[1'',2'':4',5']thieno[2',3':4,5]thieno[3,2-*b*][1,2,3]triazolo[4,5-*e*]indole-3,17-diylidene))bis(5,6-dichloro-3-oxo-2,3-dihydro-1*H*-indene-2,1-diylidene))dimalononitrile (FM7, Figure 1).

The tetrafluorobenzene cyclization is a multifaceted modification that simultaneously rigidifies the backbone, extends π -conjugation, and enhances electron-withdrawing character. Covalently locking the rotatable vinylene linkers into a fused-ring system suppresses conformational disorder, while the tetrafluorobenzene ring extends the conjugated framework and delocalizes the lowest unoccupied molecular orbitals (LUMOs) (Figure 2a). Concurrently, the four fluorine substituents increase the electron-deficient nature of the fused bridge (Figure 2b), deepening the LUMO and strengthening intramolecular charge transfer (ICT) character. These effects are inherently coupled—the same tetrafluorobenzene bridge provides both the rigidity that suppresses trap density and the electronic modulation that enables SWIR absorption.

The synthetic route of FM7 is outlined in Scheme 1 and Figure S1. The fully fused-ring central core 1 was synthesized according to reported procedures. Starting from 1, a Friedel–Crafts acylation with 2-bromo-3,4,5,6-tetrafluorobenzoyl chloride, followed by a Pd(0)-catalyzed intramolecular C–H activation coupling, afforded the all-fused-ring precursor 3. The target molecule FM7 was then obtained via Knoevenagel condensation of 3 with 2-(5,6-dichloro-3-oxo-2,3-dihydro-1*H*-inden-1-ylidene)malononitrile. The chemical structure of FM7 has been confirmed by ^1H NMR, ^{13}C NMR, and matrix-assisted laser desorption/ionization time-of-flight mass spectrometry (MALDI-TOF MS) (Figures S20–S25). FM7 is readily soluble in common organic solvents, for example, chloroform and chlorobenzene, and is well-suited for solution processing.

To elucidate the effect of tetrafluorophenyl cyclization on electronic structure of FM7, we performed density functional theory (DFT) calculations at the B3LYP/6-31G(d, p) level (Figure 2a) on the skeletons of FM7 and FM7-N. The two tetrafluorophenyl moieties of FM7 predominantly contribute to the LUMO and negligibly affect the highest occupied molecular orbital (HOMO), indicating that the electron-deficient tetrafluorophenyl moieties would mainly lower the LUMO energy level (E_{LUMO}) of FM7. The cyclic voltammetry measurement results corroborate this trend. Compared to FM7-N, FM7 exhibits a downshifted E_{LUMO} by 0.36 eV and a downshifted HOMO energy level (E_{HOMO}) by 0.09 eV, reducing the electrochemical bandgap by 0.27 eV (Figure 3a). The electrostatic potential (ESP) maps (Figure 2b) further show strongly negative ESP localized on the tetrafluorobenzene-fused rings of FM7, highlighting their enhanced electron-withdrawing nature relative to the exocyclic vinylene linkers in FM7-N. The ground-state dipole moment of FM7 (7.05 Debye) is much larger than that of FM7-N (1.12 D) (Figure 2c), indicating that the cyclization with tetrafluorobenzene strengthens ICT.

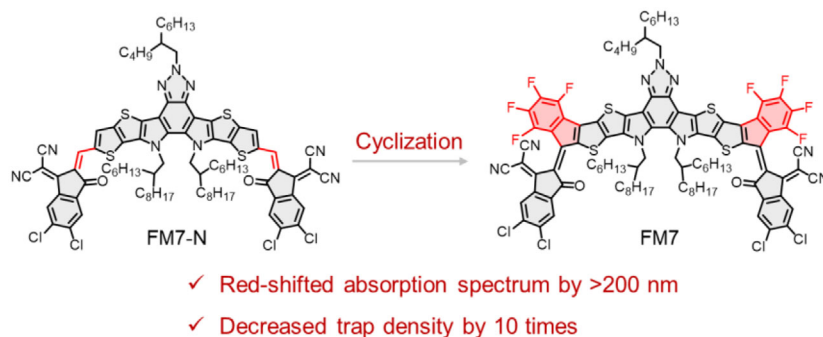


FIGURE 1 | Chemical structures of FM7 and FM7-N.

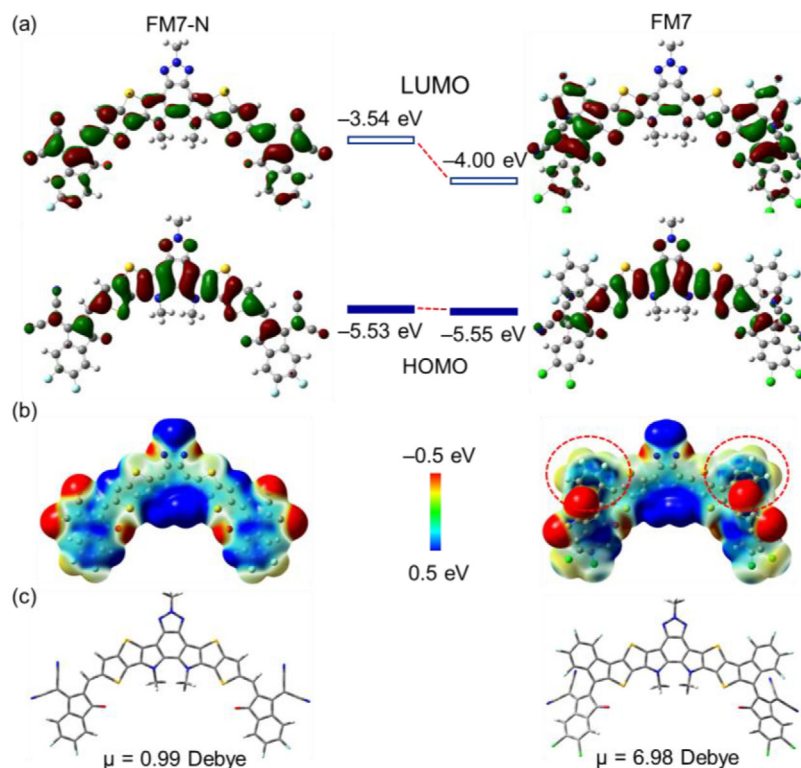
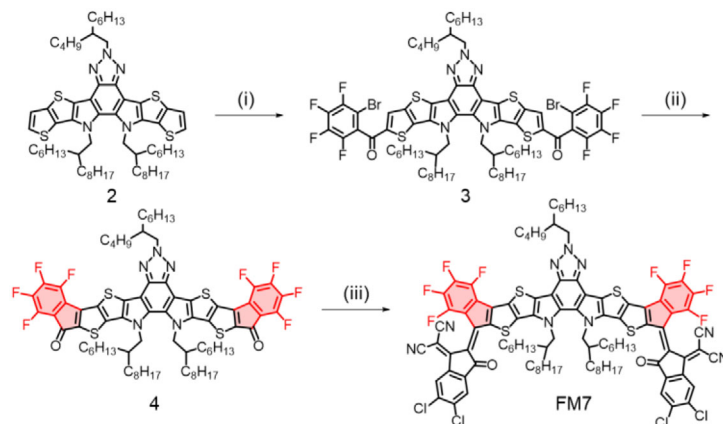


FIGURE 2 | (a) Kohn–Sham LUMO and HOMO of the framework of FM7-N and FM7 obtained by DFT calculations. (b) Electrostatic potential of FM7-N and FM7. (c) Ground-state dipole moments of FM7-N and FM7.

A trade-off in ultranarrow-bandgap organic semiconductors is that strong ICT often reduces HOMO–LUMO electronic coupling, diminishing transition dipole moment and light absorption intensity [36–38]. Our tetrafluorobenzene cyclization strategy circumvents this limitation. Despite deepened LUMO and narrowed bandgap, the HOMO–LUMO overlap integral even slightly increases from 0.637 of FM7-N to 0.653 of FM7 (Table S1), indicating preserved electronic coupling. Hole–electron analysis of the first excited state of FM7 reveals a remarkably small centroid separation of 0.27 Å, indicating a predominantly delocalized π – π^* character with minimal charge redistribution. Such a strongly coupled excited state maintains a large transition dipole moment, which indeed increases from 0.99 D (FM7-N) to 6.98 D (FM7) (Figure 2c). These theoretical calculation results are validated by the experimental absorption spectra. In

dilute chloroform solution (Figure S4), FM7 exhibits a maximum absorption at 1020 nm with a molar absorption coefficient of $1.80 \times 10^5 \text{ L mol}^{-1} \text{ cm}^{-1}$, obviously surpassing that of FM7-N ($1.10 \times 10^5 \text{ L mol}^{-1} \text{ cm}^{-1}$ at 775 nm). In thin film (Figure 3b), the absorption band of FM7 broadened to 700–1300 nm, with an absorption maximum at 1105 nm, an absorption coefficient of $1.18 \times 10^5 \text{ cm}^{-1}$, and an optical onset at 1265 nm (0.98 eV). Compared with FM7-N, FM7 exhibits a remarkable red-shift of 224 nm. This pronounced bathochromic shift demonstrates that tetrafluorobenzene cyclization effectively extends the absorption into the SWIR region. Notably, this tetrafluorobenzene cyclization strategy also preserves a high absorption coefficient, addressing the trade-off between bandgap narrowing and absorption intensity in ultranarrow-bandgap organic semiconductors. The electron mobilities of FM7 and FM7-N were measured using the



SCHEME 1 | Synthetic routes of FM7 and FM7-N. Reagents and conditions are indicated: (i) 2-Bromo-3,4,5,6-tetrafluoro-benzoyl chloride, aluminum chloride, dichloromethane, r.t.; (ii) tris(2-methylphenyl)phosphine, tris(dibenzylideneacetone)dipalladium, pivalic acid, cesium carbonate, toluene, 120°C; (iii) 2-(5,6-dichloro-2,3-dihydro-3-oxo-1H-inden-1-ylidene), pyridine, chlorobenzene, titanium(IV) chloride, 50°C.

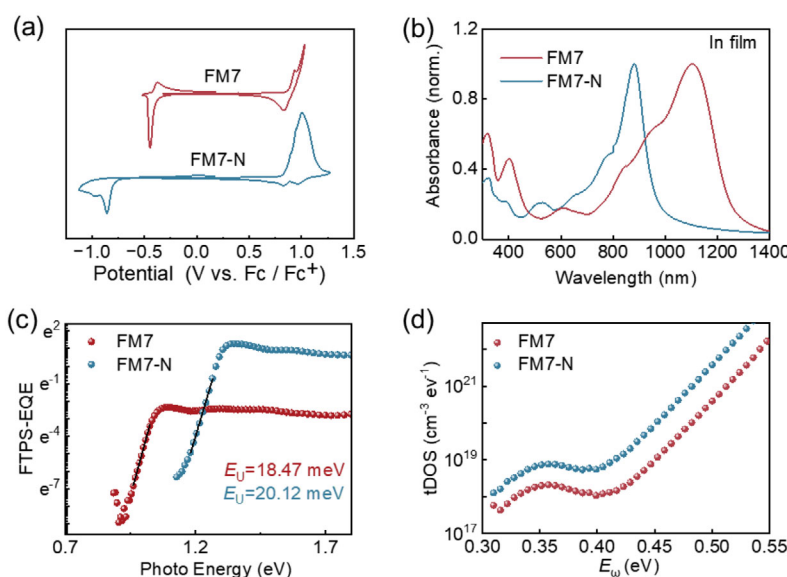


FIGURE 3 | (a) Cyclic voltammograms of FM7 and FM7-N. (b) UV-vis-IR absorption spectra of FM7 and FM7-N in thin films. (c) Extraction of Urbach energy from $\ln(\text{EQE})$ in the long-wavelength edge of FM7 and FM7-N. (d) Trap density of states (tDOS) spectra of the FM7 and FM7-N films.

space-charge limited current (SCLC) method as shown in Figure S11b. Both molecules exhibit electron transport capabilities, with mobilities of $1.28 \times 10^{-4} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ for FM7-N and $2.83 \times 10^{-4} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ for FM7.

In addition to strong SWIR absorption, suppressing energetic disorder and trap density is equally critical for achieving high detectivity in SWIR OPDs. The Urbach energy (E_U), which reflects the width of band tail states and serves as a key metric for energetic disorder, was extracted from Fourier-transform photocurrent spectroscopy external quantum efficiency (FTPS-EQE) measurements [39] (Figure 3c). The FM7-based single-component device exhibits an E_U of 18.47 meV, considerably lower than that of the FM7-N-based device (20.12 meV), indicating that tetrafluorobenzene cyclization reduces energetic disorder by rigidifying the molecular backbone and minimizing conformational fluctuations. To further evaluate the impact on trap

states, thermal admittance spectroscopy (TAS) was performed on neat films of FM7 and FM7-N [40] (Figure 3d). Over the entire measured energy depth range, FM7 consistently exhibits a lower trap density of states (tDOS) than FM7-N. More specifically, at an energy depth of 0.40–0.45 eV, FM7 shows a tDOS of $1.21 \times 10^{17} \text{ cm}^{-3}$, nearly an order of magnitude lower than that of FM7-N ($1.02 \times 10^{18} \text{ cm}^{-3}$). The reduced energetic disorder and suppressed trap density are expected to minimize trap-assisted recombination and thermal generation currents, thereby mitigating dark currents in FM7-based OPD devices.

The photodetection performance of FM7 and FM7-N was evaluated with a device structure of ITO/CuSCN/PM6:FM7 or PM6:FM7-N/PFN-Br/Ag (see Figure 4a). We chose a commercial available polymer PM6 (see Figures S2 and S19) as the electron donor. The energy level diagram of the devices is presented in Figure S3. The energy level offsets between PM6 and FM7

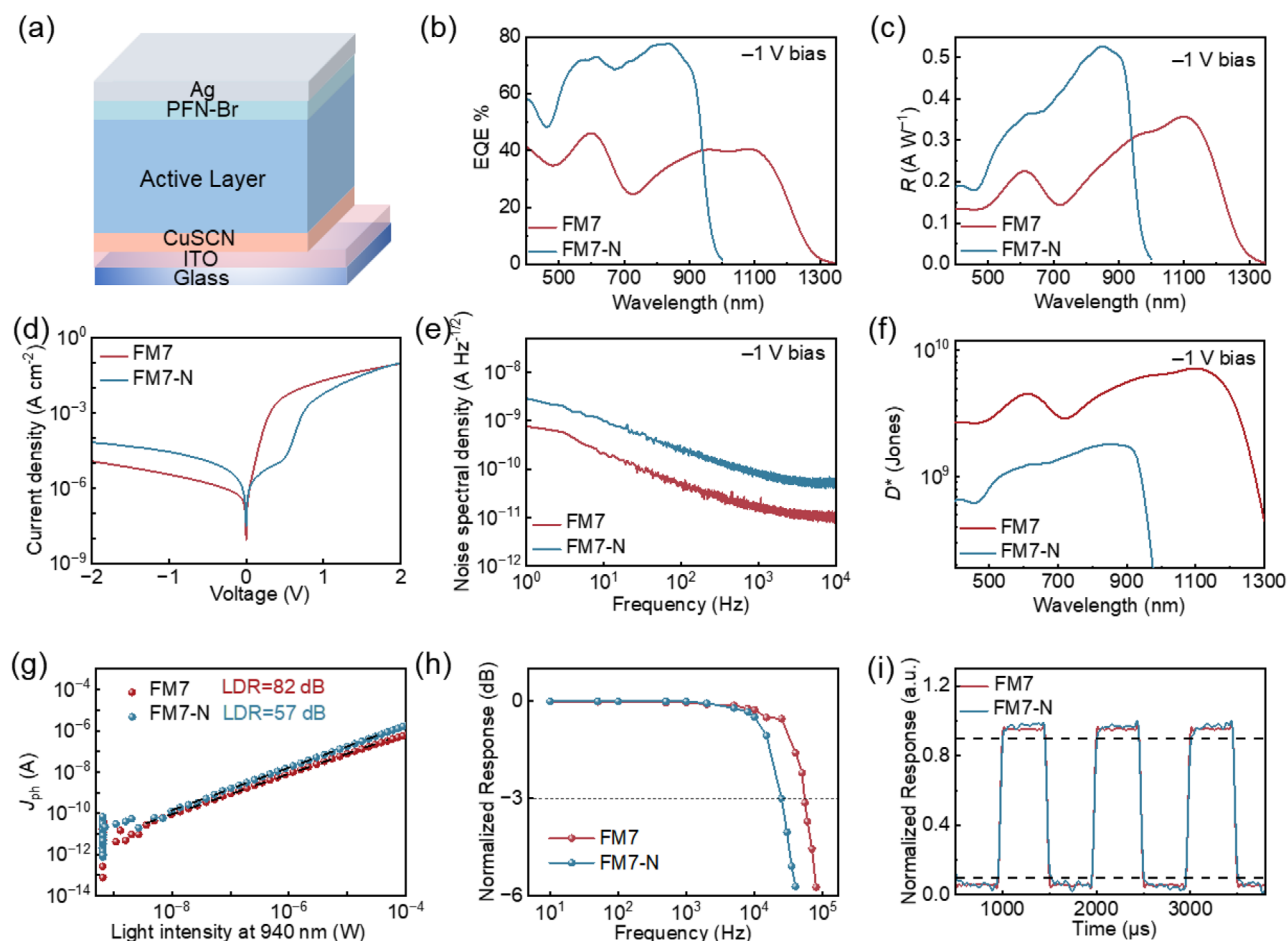


FIGURE 4 | (a) The schematic diagram of the OPD devices structure. (b) The EQE spectra curves and (c) the R curves under -1 V bias. (d) The J - V curves and (e) the measured device noise current spectra under -1 V bias in the dark condition. (f) The D^* curves of PM6:FM7-N and PM6:FM7-based devices under -1 V bias. (g) The linear dynamic range (LDR) measurements under 940 nm LED illumination, (h) the cut-off frequency of the optimized devices and (i) transient photo response characteristics under 940 nm 100 mW cm^{-2} .

or FM7-N are sufficient for efficient photo-induced hole and electron transfer. As shown in Figure 4b, the FM7-N-based device shows a spectral response spanning 400 nm $\sim$ 1000 nm, with a maximum external quantum efficiency (EQE) of 77.5% at 840 nm in NIR region. In comparison, the FM7-based device exhibits an extended response spectral range from 400 to 1300 nm with a maximum EQE of 40.3% at 1110 nm in SWIR region. The spectral responsivity of the two OPDs under -1 bias is depicted in Figure 4c. Notably, the FM7-based device achieves a peak R of 0.36 A W^{-1} at 1140 nm, which is among the highest for organic SWIR photodiodes reported to date.

Dark current density (J_d) is a critical parameter that impacts the sensitivity of OPD devices. According to the current density-voltage (J - V) curves of the two devices (see Figure 4d), at -1 V bias, the J_d values are 2.80×10^{-5} A cm^{-2} for the FM7-N-based device and 3.53×10^{-6} A cm^{-2} for FM7-based device. Compared to the FM7-N-based device, the FM7-based device exhibits an approximately eightfold reduction in J_d . Figure 4e illustrates the total noise current (i_n) of the two devices measured by frequency-dependent current noise spectrum at -1 V bias. At 1 kHz, the i_n of the FM7-N-based device and the i_n of the FM7-based device are 8.18×10^{-11} $\text{A Hz}^{-1/2}$ and 1.41×10^{-11} $\text{A Hz}^{-1/2}$, respectively. The i_n of the FM7-based device is approximately

six times lower than that of the FM7-N-based device, which is consistent with the J - V scanning results. The J - V curve was fitted using the well-established Shockley diode model (Figure S6), resulting in the diode ideality factor (n_{id}) of 1.34 for the FM7-based device and 2.36 for the FM7-N-based device. Under identical fabrication conditions, the FM7-based devices exhibit n_{id} closer to 1, indicating a lower density of interface states and traps. The suppression of noise current originates from the lower trap density and reduced energetic disorder in the tetrafluorobenzene-cyclized system, confirming our molecular design strategy's effectiveness in enhancing detector performance.

The noise current of the device was measured over a 1-s time window, and the root mean square noise current (I_{RMS}) was subsequently calculated, as depicted in Figure S7. The device based on FM7 exhibits an I_{RMS} of 2.80×10^{-12} A, whereas the FM7-N-based device has a value of 5.94×10^{-12} A. Using these values, the noise equivalent power (NEP) at peak response was calculated for both devices: the FM7-based device has an NEP of 7.97×10^{-12} W, while the FM7-N-based device shows an NEP of 1.49×10^{-11} W. The specific detectivity (D^*) of the two OPDs, which is calculated from the i_n and R under -1 V bias, are shown in Figure 4f. The FM7-N-based device shows D^* of above 10^9 Jones in the range of from 540 to 940 nm with the highest value of $1.82 \times$

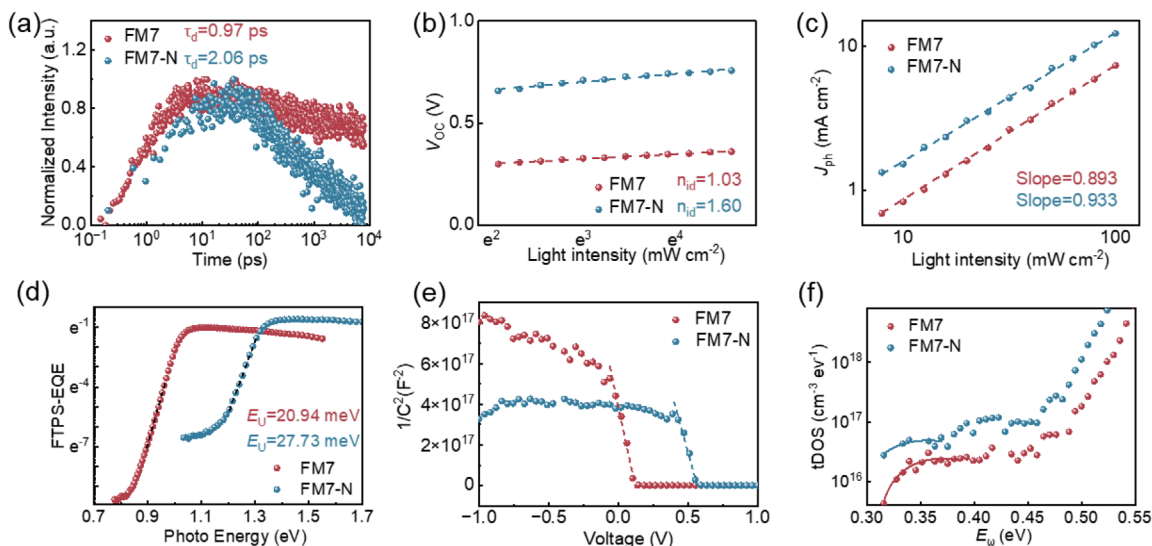


FIGURE 5 | (a) The extracted decay dynamics of PM6:FM7 and PM6:FM7-N films probed at 640 nm. (b) The dependence of the open-circuit voltage on light intensity. (c) The dependence of the short-circuit current density on light intensity. (d) Extraction of Urbach energy from $\ln(\text{EQE})$ in the long-wavelength edge. (e) The C^{-2} - V curves and (f) tDOS spectra and Gaussian fitted curves of FM7 and FM7-N-based devices.

10^9 Jones at 880 nm. Remarkably, the FM7-based device achieved D^* of exceeding 10^9 Jones across the range of from 400 to 1270 nm with the highest value of 7.17×10^9 Jones at 1140 nm in the SWIR region. The device metrics under different bias are provided in Figure S5.

To elucidate reason behind the high R of the FM7-based device, we studied the exciton dissociation, carrier transport, and charge collection of the two OPD devices. First, femtosecond transient absorption (TA) spectroscopy was used to unravel the ultrafast exciton dissociation dynamics in the two active layers [41–43]. We use a 920 nm beam to selectively excite the electron acceptors in the blend film and monitor the ground-state bleach (GSB) of the electron donor at 640 nm to study the photo-induced hole transfer dynamics (see Figure S9). The TA GSB decay kinetics were extracted and fitted to an exponential decay function (Figure 5a). The rising phase reflects the kinetics of hole transfer, while the decay component is attributed to charge recombination. The τ_d values of the hole transfer kinetic processes are 0.97 ps for the FM7-based blend film and 2.06 ps for the FM7-N-based blend film. This suggests that the FM7-based blend film has a faster charge transfer process and more efficient exciton dissociation than those of the FM7-N-based blend film. The tetrafluorobenzene cyclization in FM7 leads to improved hole transfer at the donor–acceptor interface of the active layers. This is attributed to the increased average electrostatic potential (see Figure S10) and downshifted E_{HOMO} with tetrafluorobenzene cyclization, which increase the driving force for exciton dissociation. Both active layers exhibit high and balanced charge carrier mobilities. The electron mobility/hole mobility of the PM6:FM7 film and PM6:FM7-N film are $1.40 \times 10^{-4} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ / $6.20 \times 10^{-4} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ and $1.76 \times 10^{-4} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ / $2.49 \times 10^{-4} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, respectively (Figure S11). The dependence of open-circuit voltage (V_{OC}) on light intensity (P_{in}) of the two OPD devices is fitted by the equation $V_{\text{OC}} = n_{\text{id}} kT/q \ln(P_{\text{in}})$, where k is the Boltzmann constant, n_{id} is the ideality factor, T is the thermodynamic temperature, and q is the elementary charge. As

shown in Figure 5b, an n_{id} of 1.03 was obtained for the FM7-based device and 1.60 for the FM7-N-based device. The n_{id} of the FM7-based OPD device is ultrasmall for SWIR OPDs, suggesting negligible trap-assisted recombination. The dependence of short-circuit current density (J_{sc}) on P_{in} follows the relation $J_{\text{sc}} \propto P_{\text{in}}^S$, where S represents the exponential factor. The S values are 0.893 for the FM7-based device and 0.933 for the FM7-N-based device (Figure 5c). The relatively low S value of the FM7-based device suggests severe bimolecular charge recombination, which is commonly observed in NIR or SWIR OPDs. These results suggest that the superior R of the FM7-based SWIR OPDs can be attributed to the improved hole transfer at the donor–acceptor interface and the suppressed trap-assisted charge recombination.

Despite the decreased bandgap of FM7, the FM7-based device exhibits lower noise currents than that of the FM7-N-based device. In photodiode-type OPDs, noise current primarily arise from thermal noise (S_{thermal}), shot noise ($S_{\text{shot-diode}}$), and $1/f$ noise. Among them, thermal noise is the dominant noise source in narrow-bandgap OPDs at high frequency under self-powered operation [44–46]. Electrochemical impedance spectroscopy measurements (Figure S15) yield shunt resistances of 132 k Ω (FM7-based device) and 163 k Ω (FM7-N-based device). According to Ng's physical model, thermal noise can be mitigated by increasing effective bandgap ($E_{\text{g}}^{\text{eff}}$) and inhibiting band tail disorder spread. This motivates us to study the energetic disorder and trap density of the two OPD devices. At first, we measure the E_{U} of the photoactive layers of the two devices using FTSP-EQE. As shown in Figure 5d, the E_{U} of the FM7-based and FM7-N-based devices are 20.94 and 27.73 meV, respectively. The smaller E_{U} of the FM7-based device is indicative of lower energetic disorder. This is attributed to the rigid molecular backbone and suppressed conformational disorder of FM7 with tetrafluorobenzene cyclization. Second, to assess the trap density, we perform capacitance–voltage measurement of the two devices for Mott–Schottky analysis to obtain the charge doping density (N_{A}) [46]. The N_{A} is $2.24 \times 10^{15} \text{ cm}^{-3}$ for the FM7-

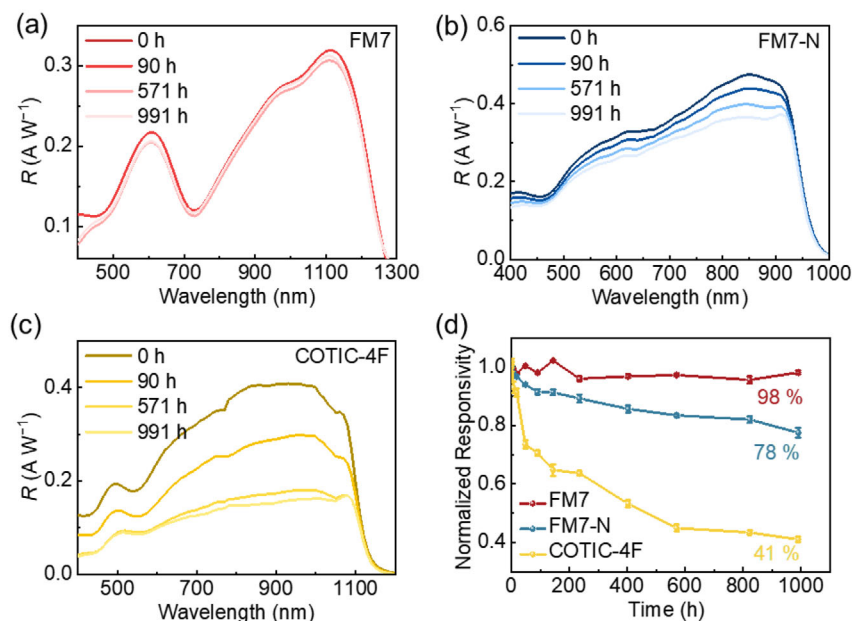


FIGURE 6 | Storage stability curves of R of packaged devices based on (a) PM6:FM7, (b) PM6:FM7-N, and (c) PTB7-Th: COTIC-4F in air. (d) The normalized responsivity decay curves. The error bars with average values were obtained from four individual devices.

based device and $3.42 \times 10^{15} \text{ cm}^{-3}$ for the FM7-N-based device (Figure 5e). Third, TAS measurement was employed to assess the trap density of the two OPD devices. As depicted in Figure 5f, at shallow energy states ranging from 0.35 to 0.45 eV, the trap density is $2.69 \times 10^{15} \text{ cm}^{-3}$ for the FM7-based device and $7.58 \times 10^{15} \text{ cm}^{-3}$ for the FM7-N-based device. Both the Mott-Schottky analysis and TAS measurement suggest lower trap density of the FM7-based device compared to the FM7-N-based device. Moreover, the reduced trap density in the film was corroborated by crystallinity and morphological analyses (see Figures S12–S14 and Tables S2, S3). Therefore, the tetrafluorobenzene cyclization in FM7 leads to minimized energetic disorder, decreased trap density, and consequently suppressed noise current in OPDs.

The linear dynamic range (LDR) was evaluated in accordance with the consensus guidelines for emerging photodetectors [47]. The photocurrent $I_{\text{ph}} = I_{\text{light}} - I_{\text{dark}}$ was measured under 940 nm LED illumination with incident optical power ranging from 3.6×10^{-9} to 6.0×10^{-5} W. At this wavelength, both devices show a measurable photoresponse, enabling a meaningful LDR comparison. The LDR was determined as $\text{LDR} = 20 \times \log(P_{\text{max}} / P_{\text{min}})$, where P_{max} and P_{min} are the maximum and minimum optical powers within which the slope α of the $\log(I_{\text{ph}}) - \log(P)$ fit falls within (0.99, 1.01), satisfying the strict linearity criterion. The FM7-based device exhibits an LDR of 82 dB ($\alpha = 1.00$), which is significantly wider than that of the FM7-N-based device (57 dB, Figure 4g). The higher LDR of the FM7-based device enables it to resolve a broader range of light intensities, which is essential for high-dynamic-range SWIR imaging. The cut-off frequencies ($f_{-3 \text{ dB}}$) of the FM7-N-based and FM7-based devices are 25 kHz and 55 kHz, respectively (Figure 4h). The rise and fall times ($t_{\text{rise}}/t_{\text{fall}}$) are 52/54 μs for the FM7-N-based device and 40/50 μs for the FM7-based device (Figure 4i). The $f_{-3 \text{ dB}}$ and $t_{\text{rise}}/t_{\text{fall}}$ of the FM7-based OPDs are adequate for typical 30 frames-per-second (fps) megapixel infrared imaging.

Storage stability is another crucial parameter for evaluating the practical applicability of devices. The storage stability of PM6:FM7, PM6:FM7-N, and PTB7-Th:COTIC-4F devices was assessed under ambient conditions of 25°C and 60–65% relative humidity. As shown in Figure 6a–c, the responsivity evolution of the three packaged devices was monitored over time. Notably, the PM6:FM7-based device retained 98% of its initial responsivity after approximately 1000 h (991 h), whereas the PM6:FM7-N-based and PTB7-Th:COTIC-4F-based devices exhibited considerably faster performance decay under identical conditions. The normalized responsivity decay curves in Figure 6d further highlight this contrast, with the PM6:FM7-based device showing a markedly slower attenuation trend. The superior storage stability of the PM6:FM7-based device can be attributed to the elimination of the highly reactive exocyclic vinyl linkers in the cyclized acceptor FM7, which imparts both enhanced chemical stability and improved photostability to the material itself. This result underscores that the tetrafluorobenzene cyclization strategy not only extends the photoresponse into the SWIR region but also imparts excellent long-term stability, which is essential for practical imaging applications.

The high photosensitivity of the FM7-based SWIR OPDs enables us to fabricate organic SWIR imaging arrays by monolithically integrating SWIR OPDs with TFT chip. As shown in Figure 7b, the OPD is directly fabricated on top of the TFT array panel to give the SWIR imager, the pixel array is defined by the underlying TFT architecture. The imager features a spatial resolution of 256×256 pixels with a pixel pitch of 50.8 μm , resulting in a total of 65,536 pixels distributed over a 13×13 mm active area. Figure 7c illustrates the longitudinal structure of the imager. The TFT drain is connected via an SiN_x layer to the indium tin oxide (ITO) plate, which serves as the anode of the OPD device.

The semitransparent TFT panel was fabricated using Tianma's standard G4.5 6-mask process. Solution processing was employed

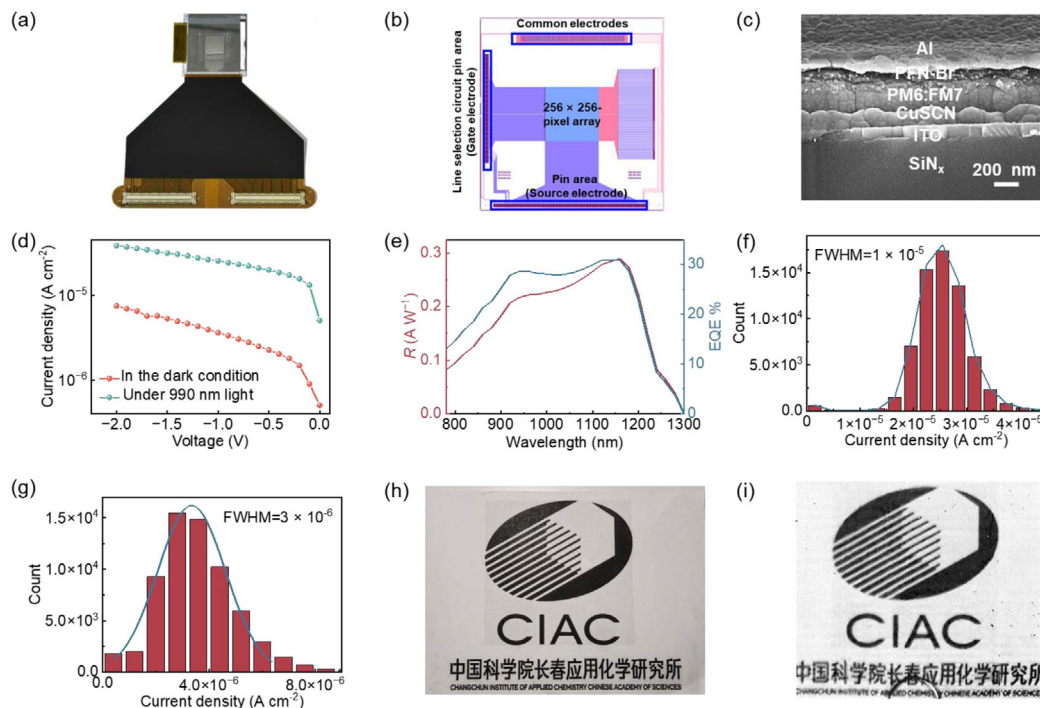


FIGURE 7 | (a) Photograph of an organic shortwave infrared imager. (b) The circuit layouts of 256 × 256 pixel TFT panel. (c) Photograph of the imager's cross-sectional SEM scans in the local magnification SEM image of the photodiode section. (d) The J – V curves of the imager in the light and dark states. and (e) The R and the EQE curves of the imager in the infrared region. (f) The distribution of the dark and (g) the light currents of all pixels measured at an external bias of -1 V. The data are fitted assuming a Gaussian distribution. The images of the emblem of the Changchun Institute of Applied Chemistry, Chinese Academy of Sciences, captured by (h) a smartphone camera under natural light conditions, and (i) images captured by our organic imaging array under 1064 nm illumination only.

to sequentially deposit the hole transport layer (CuSCN), the photosensitive layer (PM6:FM7), and the electron transport layer (PFN-Br) onto the TFT panel. An aluminum electrode was then deposited via vacuum evaporation to serve as the common cathode. Notably, the photosensitive layer does not require pixel-level patterning, significantly simplifying the fabrication process. Figure 7a presents a photographic image of the organic imaging array, comprising an organic photodiode-coated TFT panel bonded to a flexible printed circuit. The cross-sectional scanning electron microscopy (SEM) image of the active area (see Figure S16) shows the multilayered structure, including the gate electrode, the source/drain electrode, and photodiode layer. A magnified view of the photodiode layer is provided in Figure 7c, revealing the uniform deposition of the ITO layer on the SiNx layer, followed by sequential deposition of the CuSCN, active layer, PFN-Br, and aluminum electrode. The corresponding circuit layout is depicted in Figure S17. In imaging process, pixel selection is conducted via row-wise scanning by applying a gate voltage of 10 V to the array. Simultaneously, a source voltage of 0.1 V is applied column by column through the bit lines, enabling the readout of pixel current response via the aluminum co-electrode.

To evaluate the imaging capability under weak light intensity, we measured the J – V curve of the imager over a voltage range of -2 V $\sim$ 0 V under an incident light intensity of $15 \mu\text{W cm}^{-2}$ at the wavelength of 990 nm. As shown in Figure 7d, the photocurrent of the imager under the weak light illumination is an order of magnitude higher than the dark current, demonstrating its

capability for weak-light imaging. To assess the infrared photore-sponse of the imager, we measured the EQE and R across the infrared spectrum (780–1300 nm) and the results are presented in Figure 7e. The R remains above 0.20 A W^{-1} in the 1000 nm $\sim$ 1200 nm range with a peak value of 0.29 A W^{-1} at 1140 nm. The R value of the imager is slightly lower than that of the aforementioned single-element device, which is attributable to optical losses caused by the TFT panel of the backside-illuminated diode array.

For array format imagers, the uniformity of both dark current and photocurrent across all pixels is necessary [48–54]. At -1 V bias, the dark current density across 65,536 pixels shows near-normal distribution with the average value of $3.6 \times 10^{-6} \text{ A cm}^{-2}$ and the full-width at half maximum (FWHM) of $3 \times 10^{-6} \text{ A cm}^{-2}$ (Figure 7f). The photocurrents at the array pixel sites followed a normal distribution with the average value of $2.5 \times 10^{-5} \text{ A cm}^{-2}$ and an FWHM of $1 \times 10^{-5} \text{ A cm}^{-2}$ (Figure 7g). In addition, a total of 768 pixels exhibits photocurrents below 50% of the average, yielding a blind element rate of as low as 1.17%. The solution-processed organic SWIR imager exhibits excellent homogeneity, enabling high-fidelity and reliable imaging. Further characterization of the spatial resolution and signal-to-noise ratio (Figure S18) affirmed the practical promise of the organic imager.

To evaluate the imaging performance of the organic imager, we first acquired a reference image of the logo of the Changchun Institute of Applied Chemistry, Chinese Academy of Sciences,

under visible-light illumination using a commercial smartphone camera (Figure 7h). Upon removal of the visible-light source and subsequent illumination solely with a 1064 nm infrared light source, neither the human eye nor the smartphone camera was capable of capturing a discernible image. In marked contrast, under identical conditions, the organic imager produced an image with clarity comparable to that obtained by the smartphone under visible light (Figure 7i). Additionally, video footage of a car in motion was captured using a 1:64 scale model under 940 nm LED illumination (Supporting information). The white car overtakes two stationary reference vehicles from right to left, with its outline clearly delineated throughout the maneuver, effectively illustrating the motion sequence. These results highlight the potential of organic image sensors for nighttime driver assistance applications.

3 | Conclusion

In conclusion, we have developed an ultrasmall bandgap electron acceptor by cyclizing the exocyclic vinylene linkers with tetrafluorobenzene moiety on a typical A–D–A type electron acceptor. In this tetrafluorobenzene cyclization, backbone rigidification and electronic modulation are accomplished through a single structural modification, in which the rigid fused-ring framework suppresses conformational disorder and reduces the tDOS, while the electron-deficient tetrafluorobenzene bridge extends π -conjugation and delocalizes the LUMO, shifting the absorption spectrum by over 200 nm into the SWIR region. The resulting OPDs exhibit a broad photoresponse from 400 nm to 1300 nm, with a peak responsivity of 0.36 A W^{-1} , a specific detectivity exceeding 10^9 Jones at 1140 nm under -1 V bias, and an LDR of 82 dB. On the basis of the excellent OPD device performance, high-resolution organic SWIR imagers are demonstrated. According to these results, low-cost nontoxic SWIR imagers based on organic semiconductors for consumer electronics can be anticipated.

Author Contributions

Wenliang Chen: methodology, data curation, and writing – original draft. **Xiaoyu Zhu:** methodology and data curation. **Junhui Miao:** conceptualization, methodology, data curation, investigation, project administration, funding acquisition, writing – original draft, writing – review and editing, and validation. **Jun Liu:** conceptualization, writing – review and editing, project administration, funding acquisition, supervision, and resources. **Lixiang Wang:** project administration.

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Conflicts of Interest

The authors declare no conflicts of interest.

Data Availability Statement

The data that supports the findings of this study are available in the Supporting Information of this article.

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Supporting Information

Additional supporting information can be found online in the Supporting Information section.

Supporting File 1: anie74289-sup-0001-SuppMat.docx.

Supporting File 2: anie74289-sup-0002-Video.mp4.