

Novel Materials for Semi-Transparent Organic Solar Cells

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The rapid development of photovoltaic technology has driven the search for novel materials that can improve the cost-effectiveness and efficiency of solar cells. Organic semiconductors offer unique optical tunability and transparency, allowing customization for the absorption of specific optical spectra like near-infrared radiation. Through the molecular engineering of electron donors and acceptors, these materials can be optimized for targeted optical selectivity. This adaptability enables the development of efficient energy-harvesting devices tailored for specific spectral regions. Consequently, organic semiconductors present a promising avenue for specialized applications such as semi-transparent organic solar cells.

Keywords: semi-transparent organic solar cells (OSCs) ; advanced energy materials ; power conversion efficiency (PCE)

1. Introduction

In the modern era, the world is grappling with intensifying environmental issues alongside a growing demand for sustainable energy solutions ^{[1][2][3][4]}. Solar energy, a clean and inexhaustible resource, stands at the forefront of resolving these challenges ^{[5][6]}. High-performance photovoltaic technologies have the potential to not only substantially augment global energy production but also to lessen environmental impacts ^[7]. In the past few years, there has been remarkable advancement in power conversion efficiency (PCE) for various solar cell technologies. Some of the record PCEs are 26.7% for mono-crystalline silicon ^[8], 24.4% for multi-crystalline silicon ^[9], 10.3% for hydrogenated amorphous silicon (a-Si:H) ^[10], 23.6% for CIGS ^[11], 22.3% for CdTe ^[12], 26.1% for Perovskite-single junction ^[13], 33.7% for Perovskite-2 Terminal (perovskite/Si) tandem solar cells ^[13], 20.6% for organic solar cells (OSCs) ^{[14][15][16]}, and 14.2% for dye-sensitized solar cells ^[17]. Despite these advances, silicon-based photovoltaics (PV) currently dominates the market, accounting for around 95% of total commercial production. This dominance suggests that there is ample room to enhance and expand alternative PV technologies. Among various PV technologies, organic photovoltaics (OPV) emerges as a particularly promising innovation characterized by solution-based processing methods, semi-transparency, lightweight attributes, flexibility, and optical tuneability by molecular engineering ^[18]. OPV offers a versatile approach to solar energy conversion. In recent years, OPV technologies have witnessed groundbreaking advances, propelling their PCE above the 20% mark ^[14]. This impressive achievement is largely due to advances in organic semiconductor materials and device architectures. Unlike their silicon-based counterparts, which often require specialized conditions for installation, OPV devices offer greater semi-transparency, which makes them more adaptable for integration with other applications ^{[19][20]}. With their unique capability for optical semi-transparency, organic semiconductors have led to the advent of specialized OSCs that are not just confined to traditional power generation but have versatile applications, extending from Building-Integrated Photovoltaics (BIPV) to car shelters, greenhouses, automobile sunroofs, and even wearable electronics, which can be seamlessly integrated into windows, rooftops, and façades, generating electricity and contributing to the aesthetics of the built environment as well ^{[20][21]}.

The notable potential of organic semiconductors is their optical tunability feature and hence transparency, which allows for targeted optical selectivity to specific spectral ranges—ultraviolet (UV), visible, or near-infrared (NIR)—through molecular engineering of the active materials, such as electron donors and electron acceptors. This tunability paves the way for customized, efficient energy-harvesting materials for specific spectral regions ^[22]. In contrast to the UV-visible range, the IR absorption spectrum encompasses a notable 52% of the solar spectrum's total energy. The IR spectrum can be broadly categorized into three segments: NIR, mid-IR, and far-IR. The energy of photons within the IR range varies from 1.6 to 0.025 eV, which is considerably lower than that of photons in the UV-visible region. As a result, capturing these plentiful yet lower-energy IR photons can enhance the short-circuit current density (Jsc) and, hence, the performance of OSCs ^[23].

2. Novel Materials for Semi-Transparent OSCs

In exploring high-performing ST-OSCs, the trade-off between PCE and average visible transmittance (AVT) has long been a challenging hurdle. Devices' overall efficiency is defined by Light Utilization Efficiency (LUE), which is the product of PCE and AVT [24].

The recent innovations in ST-OSCs are particularly noteworthy, especially the novel donor materials that absorb more efficiently in the NIR region. To synthesize donor material useful for ST-OSCs by narrowing the bandgap of the donor polymer, Kini et al. [22] described two primary strategies. The first strategy involves the synthesis of polymers using a donor–acceptor (D–A) molecular architecture. This is a unique push–pull molecular architecture featuring alternating electron-rich and electron-deficient units, known as “D” and “A”, respectively [25][26][27][28]. This configuration results in an efficient intramolecular charge transfer (ICT), effectively narrows the polymer's bandgap, and thus enhances its ability to absorb NIR radiation [26][29][30]. This approach not only reduces the bandgap of polymer donors through the hybridization of HOMO and LUMO between the “D” and “A” components but also facilitates electron delocalization along the polymer backbone, thanks to a strong push–pull effect. The second strategy focuses on stabilizing the structure through quinoidal resonance along the conjugated backbone. This method is effective because quinoidal structures tend to have a lower bandgap compared to aromatic structures.

Over the last few years, a variety of materials have been commonly used as the D units in copolymers for OSC devices. These include benzo [1,2-b:4,5-b']dithiophene (BDT) [31], carbazole [32], thiophene [33][34], and benzo [1,2-b:4,5-b']difuran [35][36]. As for the A units, the most frequently employed materials are benzothiadiazole [37][38][39], benzotriazole [40], and benzodithiophene-4,8-dione (BDD) [41]. Researchers have experimented with several structural modifications to enhance the photovoltaic performance of these copolymers. Approaches to fine-tuning their properties have included fluorination, chlorination, and side-chain engineering. By strategically deploying this push–pull architecture, researchers are making strides in elevating both the PCE and the LUE, thereby inching closer to a more balanced and efficient semi-transparent photovoltaic system. PTB7-Th, which is commonly utilized in OSCs due to its semi-transparency [42][43], exemplifies this with a D-A backbone comprising benzodithiophene (BDT) and fluorinated thienothiophene (FTT). Unique to PTB7-Th is its “regioregularity”, an attribute influenced by the asymmetry in the FTT unit, which in turn significantly impacts both physicochemical properties and device performance [44]. Researchers have experimented with modifying the molecular architecture to a D-A-A configuration to further enhance ICT by incorporating two FTT units. This led to the development of the polymers 44-PTB and 66-PTB. Impressively, 66-PTB exhibited robust NIR absorption between 650 nm and 850 nm, owing to the dual FTT units at the 6-position. In contrast, 44-PTB, having FTT units at the 4-position, manifested absorption profiles akin to those of PTB7-Th. Differences in backbone planarity due to FTT positioning account for this variability. A blend of 66-PTB with a narrow-bandgap acceptor, IEICO-4F, achieved an optimized PCE of 7.84%, despite the absorption of radiation only in the NIR region [31].

In research conducted by Huang et al. [45], a new approach was taken to improve the compatibility of PTB7-Th (also called PCE10) with Y6, two materials commonly used in OSCs. By introducing a third type of unit into the PTB7-Th matrix, specifically a BDT unit that was modified by a fluorine or chlorine group, two new series of terpolymer donors were created: PCE10-BDT2F and PCE10-BDT2Cl. Notably, the solar cell device made with PCE10-BDT2F-0.8 and Y6 achieved an impressive PCE of 13.80%. This represents a nearly 40% improvement over devices made with the original PCE10 and Y6 combination. The terpolymer design strategy can effectively be used to tailor the properties of the polymer donor, achieving a significantly better performance.

Chen et al. [46] synthesized a unique dimeric porphyrin-cored small molecule, CS-DP, featuring an A– π 2–D– π 1–D– π 2–A architecture. This molecule is distinguished by its narrow bandgap (E_g) of 1.22 eV, allowing it to have an extended absorption spectrum in the 700–1000 nm range. When coupled with PC₇₁BM as an acceptor in Bulk Heterojunction (BHJ) OSCs, the deeper energy levels of the molecule's Highest Occupied Molecular Orbital (HOMO) and Lowest Unoccupied Molecular Orbital (LUMO) contribute to achieving an impressive PCE of 8.29% [46]. In a separate study, Lee et al. developed OSCs based on DPP2T:IEICO-4F that are efficient at converting NIR light into electricity with a PCE of 9.13% while remaining transparent in the visible spectrum, making them ideal for applications where visible-light transparency is desired, such as in transparent windows that can also generate power [47].

Xie et al. [48] explored using a fibril network strategy to achieve both a high AVT and PCE in semi-transparent OSCs. Utilizing wide-bandgap (WBG) PBT1-C-2Cl and Y6 as their electron donor and electron acceptor devices, respectively, they discovered that PBT1-C-2Cl possesses a unique property of self-assembling into fibrillar nanostructures. Adding even a small quantity of PBT1-C-2Cl to the blend resulted in a dramatic increase in hole mobility (μ_h) by creating charge pathways. This, in turn, also led to an improvement in optoelectronic properties [48].

Hu et al. [49] were pioneers in fabricating semi-transparent OSCs that utilized Y6 as an electron acceptor and PM6 as an electron donor. This blend achieved an exceptional AVT of 50.5% at a thickness of 100 nm and achieved a PCE of 14.2% [49].

Yoon et al. [50] advanced the field by developing a new polymer donor with a cyclopentadithiophene (CPDT) core. This polymer exhibits powerful absorption in the near-infrared (NIR) spectrum without compromising visibility. The deep energy levels of its HOMO also align well with low-bandgap NFAs. Consequently, they engineered an OSC achieving a notable PCE of 9.91% and an AVT of 40.4%, leading to a high LUE of 4% for the PL2 polymer donor with Y6 and PC₆₁BM as acceptors. A PTB7-Th-based system with NFAs demonstrated an LUE of around 3% in contrast to PL2 blends, which achieved a higher LUE. PL2, with its lower HOMO levels compared to PTB7-Th, offers better compatibility with NFAs [50]. Jiang et al. [51] developed an OSC using a PM2:Y6-BO system with a PCE of over 11%. When compared with PM6-based systems, the PCE of the PM6 system is higher than that of the PM2-based system. However, the AVT of the PM2-based system (an AVT of 32.7%) is higher than that of the PM6-based system (an AVT of 20.9%). This research also identified challenges with the PM2-based semi-transparent OSCs, as they grappled with charge recombination and energy loss issues, indicating avenues for further refinement and optimization in this promising OSC system.

In recent times, the focus of OPV research has also increasingly moved toward NFAs, which offer several advantages over traditional fullerene-based materials. NFAs are typically structured using architectures like “A–D–A”, “A-DA'D-A”, or “A–(π -spacer)–D–(π -spacer)–A”, where a core-electron-donating fused-ring heterocycle, marked as “D” and “DA'D”, is linked to terminal “A” groups through either vinyl linkages or π -spacer units [52]. By tweaking these core-donating units, side groups, π -spacers, and terminal acceptor units through chemical modification, it becomes possible to selectively optimize their optoelectronic characteristics, molecular arrangement, charge mobility, and overall PCEs. One notable example of this process is the development of Y-series NFAs, beginning with the Y1 molecule. This innovation in the field of organic photovoltaics has been a pivotal factor in the increased PCEs observed in Y-NFA-based OSCs. Building upon this, Yang [53] outlined strategies to further improve the efficiency of Y-NFA, as illustrated in **Figure 1**. These modifications may involve integrating sp^2 -hybridized heteroatoms and inserting π -bridges to improve the π -conjugation system. Optimizing the side chains in different positions with a variety of chemical structures is another strategy, which aims to enhance the active layer's morphology. Furthermore, the central “A” unit and end group can also be modified. With the high performance already achieved in OPVs utilizing Y-NFAs, such structural adjustments to the Y1 molecule are poised to lead to the creation of new, high-performing acceptor molecules. This approach underscores the dynamic potential of NFAs in elevating the performance of OSCs [53].

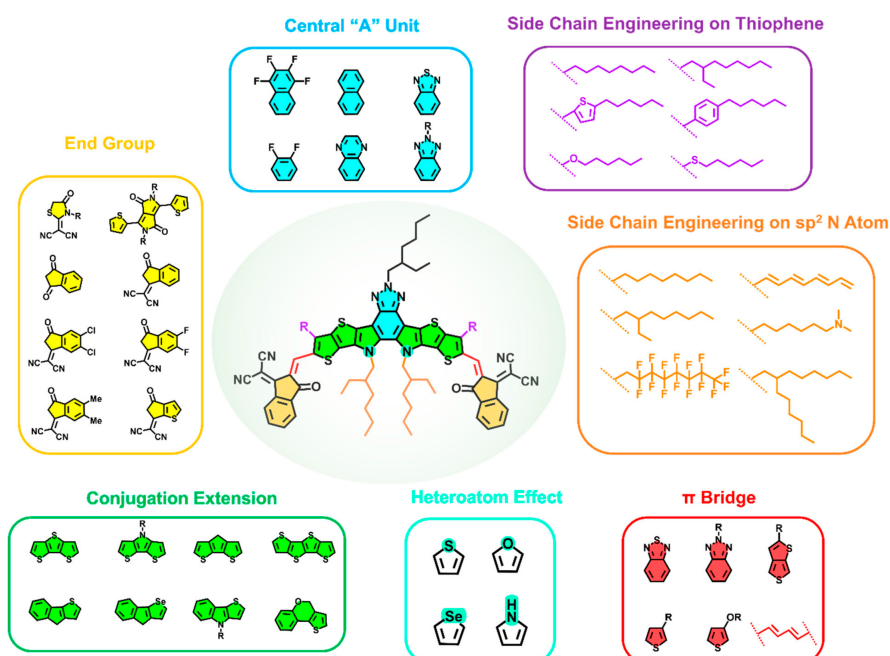


Figure 1. Strategies for exploring innovative Y-Series non-fullerene acceptors [53].

A common strategy to enhance NIR absorption in NFAs is to include terminal “A” units featuring varied functional groups, which increases the electron-withdrawing effect. Recent research highlights that these terminal “A” units impact photophysical characteristics and the energy bandgap (E_g) and affect molecular orientation and overall morphology due to non-covalent interactions [22]. The chemical structures of recently used highly efficient acceptor materials for ST-OSCs.

Incorporating a π -spacer into the “A–D–A” molecular architecture to create “A– π –D– π –A”-type NFAs is a widely used strategy for extending π -conjugation and fine-tuning photochemical characteristics and solubility. Introducing heteroatoms or appending suitable soluble alkyl chains to these units often achieves this. These π -spacers have added benefits, including increased charge transport and solid-state packing through non-covalent conformational locking, due to enhanced coplanarity. They also have lower synthetic complexity in comparison to highly conjugated fused-ring cores. Frequently used electron-rich units for π -spacers include thiophene, selenophene, and benzene derivatives, while electron-deficient units often consist of benzothiadiazole (BT) or ester-substituted thiazoles [22].

Yao et al. [54] presented a modified material known as IEICO, in which they replaced the alkyl-substituted thiophene spacer in IEIC with an alkoxy-thiophene unit. This change led to a significant increase in coplanarity and a longer π -conjugation due to non-covalent S...O interactions, resulting in strong NIR absorption with an absorption edge at approximately 925 nm ($E_g = 1.34$ eV). The modification also led to slightly higher HOMO levels and comparable LUMO values when compared with IEIC. By leveraging these unique properties of IEICO with the complementary absorbing material PBDTTT-ET, Xie et al. [49] developed semi-transparent OSCs that achieved a PCE of 9%, a high J_{sc} of 16.3 mA cm^{-2} , and an AVT of 17% through efficient NIR photon harvesting. These devices also demonstrated PCEs above 7%, along with an AVT and Incident Photon-to-Current Efficiency (IPCE) of 26.2% and 80.6%, respectively [48]. In a subsequent study, researchers enhanced the IEICO structure by replacing the DIC end groups with more potent electron-withdrawing 2F-DIC units, resulting in a new acceptor unit called IEICO-4F. This modification led to an exceptionally narrow energy bandgap (E_g) of 1.24 eV, thanks to the combined effects of an extended π -conjugation length and a stronger ICT effect induced by the 2F-DIC units. This resulted in an absorption onset situated at 1000 nm in the NIR region, which is 0.1 eV lower than the original IEICO, optimizing the material for effective NIR radiation harvesting. To test the capabilities of this new material, Hu et al. [55] formulated semi-transparent OSC devices using a blend of IEICO-4F and PTB7-Th (in a 1.2:1.5 weight ratio). The devices achieved a remarkable J_{sc} of 20.59 mA cm^{-2} and a PCE of approximately 9.5%, along with an AVT of 23.7%, J_{sc} of 20.59 mA cm^{-2} , V_{oc} of 0.718 V, and FF of 9.48. They strategically reduced the content of WBG-PD in the active layer to achieve these results. Pure PTB7-Th exhibits its main absorption peak at 700 nm, which is complementary to IEICO-4F's NIR absorption. By reducing the PTB7-Th content in the PTB7-Th:IEICO-4F blend, the researchers successfully minimized photon harvesting in the visible spectrum while enhancing it in the NIR spectrum. This adjustment enabled the researchers to achieve an optimal balance between PCE and AVT [55].

Chen et al. [57] modified the IEICO-4F structure by substituting the alkoxy-thiophene linker with an alkylthio-thiophene unit, creating a new material known as ACS8. The solid-state aggregation of alkylthio-alkyl chains in ACS8 resulted in both NIR absorption capabilities and an energy gap (E_g) of 1.3 eV, which is 0.06 eV higher than that of its predecessor, IEICO-4F, having a bandgap of 1.24 eV. Moreover, ACS8 demonstrates an electron mobility of $2.65 \times 10^{-4} \text{ cm}^2/\text{Vs}$, outperforming IEICO-4F, which shows a mobility of only $1.14 \times 10^{-4} \text{ cm}^2/\text{Vs}$. This new molecule also exhibited impressive PCE values ranging from 9.4% to 11.1%, alongside a varying AVT between 43.2% and 28.6% depending upon the thickness of the silver (Ag) layer [57].

To further understand the influence of alkylthio-alkyl side chains, the researchers compared the absorption spectrum of A078, which has a partially fused core, to that of its fully fused analog, SBT-FIC. Intriguingly, A078 demonstrated a substantial bathochromic shift and more compact molecular packing than SBT-FIC. This finding highlights the importance of non-covalent S...S interactions in facilitating the planarization and rigidification of the π -conjugated structure, which in turn narrows the energy gap. Taking advantage of ACS8's compatibility with semi-transparent OSCs due to its transparency in the visible spectrum, the team achieved a PCE of 11.0% and a 25% AVT [58].

In a study conducted by Zhang et al. [59], a novel NIR material called the BFIC acceptor was developed. Utilizing an A– π –D– π –A molecular architecture, the material incorporates FTT as a π -bridge. This dual-function inclusion extends the molecular conjugation and stabilizes its resonance structure. Furthermore, the weak electron-withdrawing capacity of FTT enhances the ICT, effectively narrowing BFIC's bandgap. The BFIC material exhibits a wide and intense NIR absorption spectrum that ranges from 700 to 1050 nm. This is further enhanced by the non-covalent interactions triggered by the fluorine atom in FTT, which optimizes molecular packing in the solid state. BFIC is highly compatible with the donor polymer PTB7-Th due to well-aligned energy levels. Regarding the thin-film absorption of BFIC, there is a shift toward longer wavelengths, peaking at 925 nm. This behavior indicates strong intermolecular packing and is a testament to the efficacy of its molecular architecture. Importantly, BFIC features an ultra-narrow E_g of 1.18 eV, which was confirmed by its absorption onset extending to 1050 nm on the optical spectrum. These properties contribute to the notable PCE of 10.38% of OSCs that are constructed using PTB7-Th and BFIC. In a semi-transparent OSC variant, a PCE of 6.15% was obtained, along with an AVT of 38.79%. This study also revealed that FTT incorporation is beneficial for the improvement of PCE [59].

In a research study conducted by Xu et al. [60], a novel A-DA'D-A acceptor named BZO-4Cl was developed. This material boasts an exceptionally low optical bandgap of 1.26 eV and an absorption that is shifted by approximately 60 nm toward longer wavelengths when compared to the material Y6. When examining the films, PTB7-Th combined with BZO-4Cl demonstrated good π - π stacking. Additionally, a pronounced backbone peak was observed, likely arising from an extended backbone ordering. This feature is key to augmenting the Jsc and fill factor (FF) of OSCs. Further, the blended film of PTB7-Th and BZO-4Cl exhibited an optimal morphology, which led to several benefits: increased charge generation and transport and decreased charge recombination. A significant factor behind the enhanced efficiency of the PTB7-Th: BZO-4Cl device is its minimized energy loss, which is attributed primarily to a drop in radiative recombination loss when compared to the Y6-integrated device. As a testament to its efficiency, the non-transparent variant of the PTB7-Th: the BZO-4Cl device achieved an impressive PCE of 14.12%. After further refinement for semi-transparency, the device achieved a PCE of 9.33%, an AVT of 43.08%, an LUE of 4.02%, and remarkable color rendering, with a color rendering index (CRI) exceeding 90% [60].

Recently, there has been significant interest in non-fused-ring Small-Molecule Acceptors (SMAs) because of their straightforward synthesis, diverse structures, and adjustable morphology. These SMAs have achieved an impressive PCE exceeding 15% due to their molecular designs, which utilize conformational lock strategies. These “locks” are often realized through non-covalent bonds, which help stabilize the molecule's structure. For instance, past research identified two non-covalent interactions, $S\cdots O$ and $C-H\cdots O$, which keep the planar structure of conjugated polymers stable, thereby enhancing carrier mobility. Other interactions, like $C-H\cdots F$, $S\cdots N$, and $S\cdots Cl$, have also been employed in crafting SMAs, resulting in improved PCEs between 13 and 15%. Specifically, Zhu et al. [61] designed two asymmetric dual-donor SMAs named IOEH-N2F and IOEH-4F. While both shared a common central unit, they had different terminals. The IOEH-N2F variant, with its specific terminal structure, displayed a broader planar structure, better crystallinity, and enhanced NIR absorption compared to IOEH-4F. As a result, devices using IOEH-N2F achieved a remarkable PCE of 14.25%, an 18% improvement over those utilizing IOEH-4F. These findings underscore the potential of tweaking electron-withdrawing terminals in asymmetric acceptors to enhance both performance and cost-efficiency in OSCs [61].

Innovative molecular architecture designs are driving remarkable advances in the field of organic semiconductors, paving the way for new levels of efficiency and versatility. While challenges persist, the architecture of these materials holds immense promise for applications such as BIPV. **Table 1** summarizes the optoelectronic properties of novel materials.

Table 1. Optoelectronic properties of novel materials.

Donor	Donor $E_g^{Opt.}$ (eV)	E_{LUMO}/E_{HOMO} (Donor)	Acceptor	Acceptor $E_g^{Opt.}$ (eV)	E_{LUMO}/E_{HOMO} (Acceptor)	V_{oc} (V)	J_{sc} (mA cm ⁻²)	FF (%)	PCE (%)	AVT (%)	Ref.
PM6	1.81	3.50/5.56	Y6	1.33	4.10/5.65	0.83	25.3	76.1	15.7	-	[62]
PCE10-BDT2F	1.59	3.82/5.42	Y6	-	4.10/5.66	0.751	20.73	69.74	10.85	41.08	[45]
CS-DP	1.26	3.74/4.96	PC ₇₁ BM	-	-	0.796	15.19	70.0	8.29	-	[46]
PL2	1.51	3.98/5.49	Y6:PC ₆₁ BM	-	-	0.69	24.07	59.74	9.91	40.4	[50]
PM2	1.41	3.89/5.30	Y6-BO	1.30	4.38/5.68	0.71	16.2	68.9	7.9	32.7	[51]
PM6	1.84	3.70/5.54	Y6-BO	1.30	4.38/5.68	0.81	19.1	73.5	11.4	20.9	[51]
PM6	[62]	-	Y6	1.33	4.10/5.65	0.855	25.39	72.93	15.83	50.5	[49]
PBT1-C-2Cl	1.85	-	Y6	1.33	-	0.83	15.71	67.7	9.1	40.1	[48]
PBDTTT-E-T	-	-	IEIC	1.50	-	0.90	11.7	47	4.9	-	[54]
PBDTTT-E-T	-	-	IEICO	1.34	3.95/5.32	0.82	17.7	58	8.4	-	[54]
PTB7-Th	-	-	IEICO-4F	-	-	0.718	20.59	64.0	9.48	23.7	[55]
PTB7-Th	-	3.33/5.30	ACS8	1.30	4.05/5.54	0.74	19.9	63.8	9.4	43.2	[57]
PTB7-Th	-	-	A078	1.40	4.06/5.58	0.75	21.2	73	11.3	47.8	[58]
PTB7-Th	-	-	SBT-FIC	1.65	4.15/5.81	0.71	18.7	65	8.2	-	[58]

Donor	Donor $E_g^{Opt.}$ (eV)	E_{LUMO}/E_{HOMO} (Donor)	Acceptor	Acceptor $E_g^{Opt.}$ (eV)	E_{LUMO}/E_{HOMO} (Acceptor)	V_{OC} (V)	J_{SC} (mA cm ⁻²)	FF (%)	PCE (%)	AVT (%)	Ref.
PTB7-Th	-	3.64/5.24	BFIC	1.18	4.00/5.26	0.66	13.82	65.4	6.15	38.79	[59]
PTB7-Th	-	3.04/5.34	BZO-4Cl	1.26	3.97/5.71	0.708	19.73	66.69	9.33	43.08	[60]
PM6	-	3.64/5.45	IOEH-N2F	1.34	3.98/5.44	0.846	22.63	74.41	14.25	-	[61]
PM6	-	3.64/5.45	IOEH-4F	1.38	3.99/5.46	0.874	17.81	75.55	11.77	-	[61]

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