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An Electron Acceptor with Porphyrin and Perylene Bisimides for Efficient Non-Fullerene Solar Cells

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Abstract: A star-shaped electron acceptor based on porphyrin as a core and perylene bisimide as end groups was constructed for application in non-fullerene organic solar cells. The new conjugated molecule exhibits aligned energy levels, good electron mobility, and complementary absorption with a donor polymer. These advantages facilitate a high power conversion efficiency of 7.4% in non-fullerene solar cells, which represents the highest photovoltaic performance based on porphyrin derivatives as the acceptor.

 \mathbf{P} orphyrin molecules play important roles in biology such as in photosynthesis and oxygen transport. The porphyrin derivative chlorophyll a in plants absorbs photons from sunlight and converts these into chemical energy through a charge-transfer process.^[1] The properties of porphyrins also extend to their application as semiconductors in organic electronics. With their large π -conjugated macrocycles, porphyrins have the tendency to form structures through hierarchical self-assembly,^[2] thereby facilitating efficient charge transport in field-effect transistors (FETs).^[3] In addition, the absorption of porphyrins lies in both the blue (Soret band) and red (Q bands) parts of the visible spectrum, and can easily extend into the near-infrared (NIR) region when donor-acceptor structures are incorporated. These properties enable porphyrins to be successfully applied in photovoltaic devices, such as dye-sensitized solar cells with power conversion efficiencies (PCEs) over 13%.^[4]

Porphyrin-based semiconductors have also been widely reported as electron donors in bulk-heterojunction (BHJ) organic solar cells (OSCs).^[5] In particular, a series of molecules with porphyrin as the core and electron-deficient units as end groups have been developed to create materials with a broad absorption in the visible and NIR absorption regions.^[6] The incorporation of these materials as electron

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donors has resulted in increased PCEs of 8% in singlejunction solar cells^[6a] and 11% in ternary and tandem solar cells.^[7] The outstanding performance of porphyrin-based electron donors has also intrigued researchers to develop them as electron acceptors. A porphyrin-fullerene dyad was reported to function as an electron acceptor in OSCs, with PCEs of 3.35% obtained.^[8] In 2014, two porphyrin molecules end-caped with isoindigo were first applied as non-fullerene acceptors in solution-processed solar cells.^[9] The molecules show absorption spectra up to $\lambda = 800$ nm and aligned energy levels with poly(3-hexylthiophene), consequently providing an initial PCE of 0.57% when used in solar cells. In principle, porphyrin is an electron-donating unit and, hence, it seems a difficult task to design electron acceptors based on porphyrins as a component in non-fullerene organic solar cells (NFOSCs).

Herein, we are interested in the possibility of using porphyrin as an electron acceptor in solar cells, and exploiting its good tendency to crystallize as well as its excellent optical absorption. The design of the molecules is based on two principles: 1) Porphyrin is an electron-donating unit, so it is necessary to introduce strongly electron-deficient units to create low-lying energy levels that are close to those of fullerene derivatives,^[10] and 2) the aromatic porphyrin tends to aggregate into large domains that are detrimental to charge generation,^[11] but can be reduced in size by introducing functional groups that create steric hindrance.^[12] On the basis of these considerations, we designed PBI-Por (Scheme 1a), which has four pervlene bisimide (PBI) units connected at the meso-positions of a porphyrin ring through ethylnyl groups. Such star-shaped structures have proven to effectively reduce aggregation in BHJ morphologies and achieve high PCEs in non-fullerene solar cells.^[13] The strongly electron-withdrawing PBI unit, which has successfully been applied as an acceptor in NFOSCs,^[14] effectively tunes the energy levels of the porphyrin core. The new molecule exhibits three characteristic absorption bands in the visible and NIR regions that are perfectly complementary to the absorption profile of the donor polymer PBDB-T (Scheme 1b). PBDB-T and PBI-Por were combined in solar cells, thereby resulting in a promising PCE of 7.4% and featuring a photoresponse from $\lambda = 300$ to 850 nm. Our results demonstrate that porphyrin-based conjugated materials have great potential to be used as electron acceptors in high-performance NFOSCs.

The synthetic procedure for the porphyrin molecule PBI-Por is shown in Scheme 1. The key precursor zinc(II) 5,10,15,20-tetraethynylporphyrin (1) was prepared according to the literature method.^[15] PBI-Por was synthesized from 1 and monobromoperylene bisimide (2)^[12] through typical

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Scheme 1. Chemical structures of a) the electron acceptor PBI-Por and its synthetic route, b) donor polymer PBDB-T. i) Sonogashira coupling with Pd(PPh₃)₄/Cul in toluene/triethylamine, 80 °C, 72 h.

Sonogashira coupling reactions with Pd(PPh₃)₄/CuI as the catalyst and toluene/triethylamine as the solvent. After 72 h, PBI-Por was obtained in a reasonable yield of 63 %. The PBI units contained C₅ chains (R1, see Scheme 1) in a swallow-tail arrangement to ensure good solubility of PBI-Por in CHCl₃ and chlorobenzene (CB). The chemical structure of PBI-Por was confirmed by high-temperature ¹H NMR spectroscopy (Figure S1) and high-resolution MALDI mass spectrometry. The molecule shows good thermal stability, with a weight loss of only 5% at 383 °C (Figure S2), thus proving its suitability for fabrication in photovoltaic cells.

The absorption spectra of the polymer PBDB-T and the acceptor PBI-Por are shown in Figure 1; the absorption spectra of its precursors PBI and Por (Figure S3) are also shown for comparison. PBI has an absorption band at $\lambda =$ 450–550 nm and Por shows Soret and O bands at $\lambda = 400$ – 500 nm and 600-700 nm, respectively. The star-shaped molecule PBI-Por has a similar absorption band at $\lambda = 400-600$ nm, which arises from the absorptions from both the PBI and Por units. Interestingly, the Q bands of PBI-Por are shifted to the near-infrared region at $\lambda = 700-800$ nm and have an enhanced intensity (Figure 1a and Figure S4a), which indicates intramolecular charge transfer between PBI and Por. The temperature-dependent absorption spectra of PBI-Por in solution also showed a similar absorption band at $\lambda = 700-800$ nm at high temperature, which indicates that this band does not arise from the crystallization or aggregation of the Por core (Figure S4b,c). The absorption band of PBI-Por in a thin film is slightly red-shifted compared to that in solution, and yields an optical band gap (E_g) of 1.48 eV. Figure 1 a shows that the PBDB-T donor polymer has an absorption in the $\lambda = 500$ -700 nm range, perfectly complementary to that of PBI-Por.

The energy levels of PBI-Por were determined by cyclic voltammetry measurements (Figure 1b), which led to the highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) levels being assigned

as -5.46 eV and -3.68 eV. As a comparison, the donor polymer PBDB-T has HOMO and LUMO levels of -5.33 eV and -3.53 eV.^[16] Therefore, both the HOMO and LUMO offsets between PBDB-T and PBI-Por are less than 0.3 eV. Although some reports show that 0.3 eV is the lowest energy for efficient exciton dissociation into free charges because of the low dielectric constant of organic semiconductors,^[17] others reveal that an energy offset below 0.3 eV is also sufficient for charge separation.^[18] The low energy offset also indicates a low energy loss, which is helpful for achieving a high opencircuit voltage (V_{oc}) . We also applied density function theory (DFT) calculations to analyze

the frontier orbital energies of PBI-Por and found that the HOMO is mainly localized at the porphyrin core and the LUMO localized at the PBI units (Figure 1 c). This may be due to the strongly electron-deficient nature of PBI groups resulting in less orbital overlap with those of the donating porphyrin.^[19]

The charge-transport properties of PBI-Por were determined in FET devices using a bottom gate/bottom contact (BGBC) configuration (Figure S5 and Table S1). In the FETs, PBI-Por shows ambipolar transport with a hole mobility of $3.1 \times 10^{-4} \text{ cm}^2 \text{V}^{-1} \text{s}^{-1}$ and electron mobility of $1.0 \times 10^{-2} \text{ cm}^2 \text{V}^{-1} \text{s}^{-1}$. This ensures that PBI-Por has electron-



Figure 1. a) Absorption spectra of PBI-Por in CB solution and in a thin film, PBDB-T in a thin film, as well as PBI and Por in solution. b) Cyclic voltammogram of the PBI-Por thin film. Potential versus Fc/Fc⁺. c) DFT frontier molecular orbitals of PBI-Por.

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transport properties and so can be used as an electron acceptor in solar cells.

The complementary absorption, aligned energy levels, and good charge transport properties of the acceptor PBI-Por encouraged us to fabricate NFOSCs. Photovoltaic devices with an inverted configuration, with ITO/ZnO and MoO₃/Ag used as electrodes, were prepared by spin coating PBDB-T:PBI-Por photoactive blend layers from chlorobenzene. The concentration of 1,8-diiodooctane (DIO) as a high-boiling processing additive, the donor and acceptor ratio, and the thickness of active layers were carefully optimized (Table 1

Table 1: Characteristics of optimized inverted solar cells of PBDB-T:PBI-Por (1:1) fabricated from CB with DIO as an additive. The thickness of the active layers is around 70 nm.

Solvent	$\int_{sc}^{[a]} [mA cm^{-2}]$	V _{oc} [V]	FF	PCE [%]
СВ	13.9	0.80	0.61	6.7
CB/DIO (0.2%)	13.8	0.80	0.61	6.7
CB/DIO (0.5%)	14.4	0.81	0.62	7.3
CB/DIO (1%)	14.5	0.78	0.66	7.4
CB/DIO (2%)	14.1	0.79	0.64	7.1

[a] J_{sc} value calculated by integrating the EQE spectrum with the AM1.5G spectrum. FF = fill factor.

and Table S2). In general, PBDB-T:PBI-Por (1:1) blends with a thickness of 70 nm spin-coated from chlorobenzene with 1% DIO provided the best PCEs. The J-V characteristics of the optimized cells are shown in Figure 2a and the photovoltaic parameters is summarized at Table 1.

The optimized PBDB-T:PBI-Por cells showed a PCE of 7.4% with a high short-circuit current density (J_{sc}) of 14.5 mA cm⁻², V_{oc} of 0.78 V, and FF of 0.66. We fabricated 14 nominally identical cells, with an average PCE of 7.2% (Table S3). To the best of our knowledge, this represents the best PCEs among porphyrin-based NFOSCs at present.^[9] The high photocurrent is reflected in the high external quantum efficiency (EQE; Figure 2b) and the cells show a broad photoresponse from $\lambda = 300$ nm to 850 nm. In particular, the EQE in the range of $\lambda = 400$ –650 nm is almost 0.70, which can be attributed to the strong absorption of the Soret bands of the porphyrin, the PBI units, and the donor polymer. The high photocurrent and high FF indicates that PBDB-T:PBI-Por blends generate and transport charge efficiently.

The morphology of the PBDB-T:PBI-Por blends was further studied by atomic force microscopy (AFM) and 2D grazing-incidence wide-angle X-ray scattering (2D-GIWAXS; Figure 3). Large surface domains that exhibit fibrillar structures in the phase image are observed in the AFM height images of the blends. This provides the separation channel for charge (hole and electron) transport and contributed to the high FF in the solar cells.^[11] PBI-Por shows an out-of-plane and in-plane diffraction peak of low intensity in the (100) direction from 2D-GIWAXS, thus indicating its low crystallinity (Figure 3c), while the polymer PBDB-T shows a strong (100) diffraction peak in the in-plane direction and a (010) peak in the out-of-plane direction (Figure S6). The strong (100) peak and (010) peak in the out-



Figure 2. a) *J*–V characteristics in the dark (dashed line) and under illumination with white light (solid line). b) EQE of the optimized PBDB-T:PBI-Por (1:1) solar cells fabricated from CB/DIO (1%).



Figure 3. a) AFM height image (3 μ m×3 μ m) of optimized PBDB-T:PBI-Por thin films spin-coated from CB with 1% DIO. b) The corresponding phase image. 2D-GIWAXS images of c) a pure PBI-Por thin film and d) a PBDB-T:PBI-Por blended thin film fabricated from CB with 1% DIO.

of-plane direction in PBDB-T:Por-PBI thin films can still be observed. This observation reveals that the fibrillar structures in the blended thin films originated from the crystal PBDB-T.

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It is of interest to assess the potential of the PBI-Por nonfullerene electron acceptor in solar cells. First, although PBDB-T:PBI-Por cells provide a high J_{sc} value of 14.5 mA cm⁻², the EQE in the NIR region is still low (below (0.40) compared to that in the visible region (close to (0.70)). This may originate from the relatively low absorption coefficient in the NIR region as a result of intramolecular charge transfer between the PBI and porphyrin groups, as revealed by the weak overlapping of the frontier orbitals of PBI and Por in the DFT calculations (Figure 1c). We speculate that the introduction of relatively weak electrondeficient groups into porphyrin compounds will improve the absorption coefficient in the NIR and enhance the J_{sc} value. Second, the energy loss, that is calculated as the difference between $E_{\rm g}$ and $qV_{\rm oc}$, is 0.70 eV in PBDB-T:PBI-Por cells. This substantial energy loss can be reduced by finely tuning the energy levels of the donor and acceptors, thereby resulting in a higher $V_{\rm oc}$ value. Therefore, we believe that NFOSCs based on porphyrin-derived electron acceptors can achieve better performance by careful structure modification, and work towards that goal is in progress in our laboratory.

In conclusion, we have designed and synthesized a starshaped structure porphyrin-based non-fullerene electron acceptor with four PBI arms for use in solar cells. The acceptor shows three typical absorption bands between $\lambda =$ 300 and 850 nm. Solar cells based on this porphyrin acceptor provide a high PCE of 7.4%. Our structural design and photovoltaic performance indicates that porphyrins can be used to construct electron acceptors for use in high-performance NFOSCs.

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Conflict of interest

The authors declare no conflict of interest.

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An Electron Acceptor with Porphyrin and Perylene Bisimides for Efficient Non-Fullerene Solar Cells



Within arm's reach: A star-shaped porphyrin-based molecule with four perylene bisimide arms (PBI-Por) was designed as a non-fullerene electron acceptor for application in solar cells. The combination of a donor polymer with PBI-Por in a solar cell resulted in a photoresponse from $\lambda = 300$ to 850 nm, with a maximum external quantum efficiency (EQE) of almost 0.70, and a promising power conversion efficiency of 7.4%.

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