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Supporting Information

A Naphthodithiophene-Based Nonfullerene Acceptor for High-Performance

Polymer Solar Cells with a Small Energy Loss

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Experimental Section

Instruments and Measurements

¹H NMR and ¹³C NMR spectra were measured in CDCl₃ on Bruker AV 400 MHz FT-NMR spectrometer. Elemental analysis was carried out on a flash EA1112 analyzer. Thermogravimetric analysis (TGA) was performed on a Perkin-Elmer TGA-7. UV-vis absorption spectra were taken on an Agilent Technologies Cary Series UV-Vis-NIR Spectrophotometer. The electrochemical cyclic voltammetry (CV) was performed on a Zahner Ennium IM6 Electrochemical Workstation with glassy carbon disk, Pt wire, and Ag/Ag⁺ electrode as working electrode, counter electrode, and reference electrode respectively, in a 0.1 M tetrabutylammonium hexafluorophosphate (Bu_4NPF_6) acetonitrile solution. Photoluminescence (PL) spectra was performed on an Edinburgh Instrument FLS 980. The GIWAXS measurements were performed at beamline 7.3.3 at the Advanced Light Source (ALS). Samples were prepared on a Si substrate under the same conditions as those used for device fabrication. The 10 KeV X-ray beam was incident at a grazing angle of 0.11°-0.15°, which maximized the scattering intensity from the samples. The scattered x-rays were detected using a Dectris Pilatus 2M photon counting detector. RSoXS transmission measurements were performed at beamline 11.0.1.2 at the Advanced Light Source (ALS). Samples for R-SoXS measurements were prepared on a PSS modified Si substrate under the same conditions as those used for device fabrication, and then transferred by floating in water to a $1.5 \text{ mm} \times 1.5 \text{ mm}$, 100 nm thick Si₃N₄ membrane supported by a 5 mm \times 5 mm, 200 μ m thick Si frame (Norcada Inc.). 2-D scattering patterns were collected on an in-vacuum CCD camera (Princeton Instrument PI-MTE). The sample to detector distance was calibrated from diffraction peaks of a PS300 sample. The beam size at the sample is approximately 100 μm by 200 μm.

The current density-voltage (*J-V*) characteristics of the OSCs were recorded with a keithley 2450. The power conversion efficiencies of the OSCs were measured under 1 sun, AM 1.5G (air mass 1.5 global) (100 mw cm⁻²) using a SS-F5-3A (Enli Technology CO., Ltd.) solar simulator (AAA grade, 50 mm x 50 mm photo-beam size). 2×2 cm⁻² Monocrystalline silicon reference cell (SRC-00019) was purchased from Enli Technology CO., Ltd. The EQE was measured by Solar Cell Spectral Response

Measurement System QE-R3011 (Enli Technology CO., Ltd.). Sensentive EQE measurements were done using a chopped monochromatic light source (chopping frequency 173 Hz), a current preamplifier (Stanford Instrument, SR570), a lock-in amplifier (Stanford Instrument, SR830), a reference Si diode. EQE-EL measurements were done using a home built setup. In this measurment, electric current was injected into the solar cell by a forward voltage bias, using a Keithly 2400 source meter, and emission from the solar cell was recorded by a Si diode and a Keithly picoammeter. The light intensity at each wavelength was calibrated with a standard single-crystal Si photovoltaic cell. Atomic force microscopy (AFM) measurements were performed on a Dimension 3100 (Veeco) Atomic Force Microscope in the tapping mode. Transmission electron microscopy (TEM) was performed using a TePNai G2 F20 S-TWIN instrument at 200 kV accelerating voltage, in which the blend films were prepared using a processing technique, as following: first, the blend films were spincast on the PFN/ZnO/ITO substrates; second, the resulting blend film/PFN/ZnO/ITO substrates were submerged in deionized water to make these blend films float onto the air-water interface; finally, the floated blend films were taken up on unsupported 200 mesh copper grids for a TEM measurement.

Device fabrication and characterization:

The OSCs devices were fabricated with an conventional device structure of ITO/PEDOT:PSS/Active layer/PFN-Br/Al, which were fabricated and characterized in a N₂-filled glovebox. The ITO-coated glass substrate was cleaned with deionized water, acetone, and isopropanol, respectively. Subsequently, the pre-cleaned ITO-coated glass

substrate was treated by UV-ozone for 20 min. Then, the PEDOT:PSS anode buffer layer was spin-coated onto the pre-cleaned ITO glass at 6000 rpm for 45 s. The active layer was deposited by spin-coating a chloroform solution of PM6:NT-4F with a blend concentration of 16 mg/mL inside a nitrogen box containing less than 5 ppm oxygen and moisture. Then, the PFN-Br as cathode interlayer was deposited on the active layer at 2500 rpm for 40 s. Finally, the whole device was completed by vacuum evaporating Al metal electrodes (100 nm) to acquire an area of 0.04 cm² cell.

Mobility measurement

The hole and electron mobilities were calculated by using the space-charge-limited current (SCLC) method.

$$J = 9\varepsilon_0\varepsilon_{\rm r}\mu(V_{\rm appl} - V_{\rm bi} - V_{\rm s})^2/8L^3$$

Where *J* is the current density, ε_0 is the permittivity of free space (8.85×10⁻¹² F m⁻¹), ε_r is the relative permittivity of the material (assumed to be 3), μ is the mobility of hole or electron, V_{appl} is the applied voltage, V_{bi} is the built-in voltage (0V), V_s is the voltage drop from the substrate's series resistance ($V_s = IR$) and L is the thickness of the active layer.

Energy loss calculation

The V_{oc} of any type of solar cells is determined by the following equation¹:

$$V_{oc} = \frac{kT}{q} ln \left(\frac{J_{sc}}{J_0} + 1 \right) \tag{1}$$

Where k is the boltzmann constant, T is the temperature, q is the elementary charge, J_{sc} is the short circuit current and J_0 is the dark saturation current. J_{sc} and J_0 is determined by the following equations:

$$J_{sc} = q \cdot \int_{0}^{\infty} EQE_{PV}(E) \cdot \phi_{AM1.5}(E)dE$$

$$J_{0} = \frac{q}{EQE_{EL}} \cdot \int_{0}^{\infty} EQE_{PV}(E) \cdot \phi_{BB}(E)dE$$

$$(3)^{2}$$

Where $^{EQE_{EL}}$ is radiative quantum efficiency of the solar cell when charge carriers are injected into the device in dark, and $^{\emptyset_{BB}}$ is the black body spectrum.

When all the recombination is radiative $(^{EQE_{EL}=1})$, J_0 is minimized, and V_{oc} is maximized:

$$J_{0}^{rad} = q \cdot \int_{0}^{\infty} EQE_{PV}(E) \cdot \phi_{BB}(E)dE$$

$$V_{oc}^{rad} = \frac{kT}{q} ln \left(\frac{J_{sc}}{J_{0}^{rad}} + 1 \right) = \frac{kT}{q} ln \left(\frac{q \cdot \int_{0}^{\infty} EQE_{PV}(E) \cdot \phi_{AM1.5}(E)dE}{q \cdot \int_{0}^{\infty} EQE_{PV}(E) \cdot \phi_{BB}(E)dE} + 1 \right)$$
(5)

The general quantum efficiency $EQE_{PV}^{SQ}(E)$ can be defined as follow in the Shockley-Queisser theory³:

$$EQE_{PV}^{SQ}(E) = 1, \qquad E > E_{gap} \quad ; \qquad EQE_{PV}^{SQ}(E) = 0, \qquad E < E_{gap} \quad . \tag{6}$$

Substituting equation 6 into equation 4, we can deduce the saturation current in the SQ limit, J_0^{SQ}

$$J_{0}^{SQ} = q \cdot \int_{E_{gap}}^{\infty} EQE_{PV}^{SQ}(E) \cdot \phi_{BB}(E)dE = q \cdot \int_{E_{gap}}^{\infty} \phi_{BB}(E)dE$$
(7)

$$V_{oc}^{SQ} = \frac{kT}{q} ln \left(\frac{J_{sc}}{J_0^{SQ}} + 1 \right) = \frac{kT}{q} ln \left(\frac{q \cdot \int_0^\infty EQE_{PV}(E) \cdot \phi_{AM1.5}(E) dE}{q \cdot \int_{E_{gap}}^\infty \phi_{BB}(E) dE} + 1 \right)$$
(8)

The difference between V_{oc}^{SQ} and V_{oc}^{rad} is due to that in the SQ theory, the band edge of the absorber is totally abrupt when calculating V_{oc}^{rad} , the band gap will be smeared out for the existence of charge transfer state absorption.

Therefore, we can calculate the $\Delta V^{rad, below gap}_{oc}$ (voltage loss of radiative recombination below the gap) and the $\Delta V^{non-rad}_{oc}$ (voltage loss due to non-radiative recombination) by the following equations:

$$\Delta V^{rad, \ below \ gap}_{oc} = V^{SQ}_{oc} - V^{rad}_{oc}$$
(9)
$$\Delta V^{non - rad}_{oc} = V^{rad}_{oc} - V_{oc}$$
(10)

Thus, we can calculate the ΔE_1 , ΔE_1 and ΔE_1 by the previous discussions.

Materials and synthesis

Materials: Commercially available reagents were used without further purification. Solvents were dried and distilled from appropriate drying agents prior to use. The compound 1 was synthesized according to reported method,⁴ **NTO-4F** was synthesized as follows:



Compound 3. To a two-necked round bottom flask were added compound 1 (1.32 g, 1.6 mmol), Compound 2 (0.89 g, 3.8 mmol), and toluene (50 mL). The mixture was deoxygenated with argon for 30 min. $Pd(PPh_3)_4$ (127 mg, 0.1 mmol) was added under argon. The mixture was refluxed at 110 °C for 24 h and then cooled down to room temperature. Water (100 mL) was added and the mixture was extracted with

dichloromethane (2×100 mL). The organic phase was dried over anhydrous MgSO₄ and filtered. After removing the solvent from filtrate, the residue was purified by column chromatography on silica gel using dichloromethane/petroleum ether (1:1) as eluent yielding an orange solid (1.03 g, 80%). ¹H NMR (400 MHz, CDCl₃), δ (ppm): δ 7.91 (s, 2H), 7.56 (d, 2H), 7.41 (s, 2H), 7.28 (d, 2H), 4.31 (m, 4H), 4.23 (d, 4H), 2.12 (m, 2H), 1.55 (m, 4H), 1.4 (m, 4H), 1.31 (m, 6H), 1.03 (m, 8H), 0.91 (t, 12H). ¹³C NMR (100 MHz, CDCl₃), δ (ppm): 163.24, 161.99, 152.74, 143.67, 137.51, 135.38, 130.73, 129.39, 128.35, 125.75, 124.94, 124.47, 102.08, 72.01, 60.98, 39.59, 30.76, 29.31, 24.11, 23.14, 14.26, 11.29. Matrix-Assisted Laser Desorption/Ionization Time of Flight Spectrometry (MALDI-TOF) MS: calculated. For C₄₄H₅₂O₆S₄ m/z = 805.14; found, 804.35.



Compound 3. To a two-necked round bottom flask were added compound 3 (0.97 g, 1.2 mmol) and THF (20 mL). The mixture was deoxygenated with argon for 30 min. A THF solution (15 mL) of fresh 4-hexylphenyl-1-magnesium bromide which prepared from magnesium turnings (356mg, 14.4mmol) and 1-bromo-4-hexylbenzene (2.89 g, 12 mmol) was added dropwise to the mixture. The mixture was refluxed for 12 h and then cooled down to room temperature. A saturated NH₄Cl aqueous solution was added

and the mixture was extracted with dichloromethane (2×50 mL). The combined organic layer was dried over MgSO₄. After removing the solvent, the yellow residue was added into a two-necked round bottom flask with solvent of octane (100ml). Acetic acid (20mL) and sulfuric acid (0.2 mL) were added slowly, the mixture was deoxygenated with argon for 30 min. The resulting solution was stirred at 65 °C for 6 h, then cooled down to room temperature and filtered. After removing the solvent from filtrate, the residue was purified by column chromatography on silica gel using petroleum ether as eluent yielding a light yellow crude product, which was used directly for the next step.

Compound 4. DMF (5 mL) was slowly injected into phosphorus oxychloride (4 ml) at 0 °C. After stirring for 1 hour, crude product of compound 3 (520 mg) in chloroform (25 mL) was added. The mixture was refluxed at 75 °C for 12 h, then the reaction was quenched with aqueous sodium succinate, and extracted with dichloromethane. After removing the solvent from filtrate, the residue was purified by column chromatography on silica gel using petroleum ether/dichloromethane (1:1) as eluent yielding a yellow solid (400 mg, 24%).1H NMR (400 MHz, CDCl3), δ (ppm): 9.81 (s, 2H), 7.74 (s, 2H), 7.17 (d, 8H), 7.07 (d, 10H), 4.00 (d, 4H), 2.53 (t, 8H), 2.02 (m, 2H), 1.56 (m, 12H), 1.36 (m, 12H), 1.26 (m, 24H), 0.95 (t, 12H), 0.85 (t, 12H). 13C NMR (100 MHz, CDCl3), δ (ppm): 182.50, 161.07, 154.13, 152.47, 148.10, 144.40, 142.21, 138.19, 137.99, 131.80, 131.71, 128.53, 128.15, 128.12, 100.82, 72.1, 62.41, 38.90, 35.54, 31.67, 31.30, 30.59, 29.70, 29.13, 29.07, 23.89, 23.08, 22.56, 14.21, 14.04, 11.04. MALDI-TOF-MS: calculated. For C₉₀H₁₀₈O₄S₄ m/z = 1382.08; found, 1381.87.



NTO-4F. To a two-necked round bottom flask were added compound 4 (207 mg, 0.15 mmol), 2-(5,6-difluoro-3-oxo-2,3-dihydro-1H-inden-1-ylidene)malononitrile (103 mg, 0.45 mmol), chloroform (40 ml), and 0.9 ml pyridine. The mixture was deoxygenated with argon for 30 min and then refluxed for 12 h. After cooling down to room temperature, the mixture was poured into methanol (200 mL) and filtered. The residue was purified by column chromatography on silica gel using dichloromethane/ petroleum ether (1:1) as eluent yield a blue solid (200 mg, 74%). ¹H NMR (400 MHz, CDCl₃), δ (ppm): 8.86 (s, 2H), 8.51 (m, 2H), 7.76 (s, 2H), 7.65 (t, 2H), 7.17 (d, 8H), 7.08 (d, 10H), 4.00 (d, 4H), 2.56 (t, 8H), 2.05 (m, 2H), 1.57 (m, 12H), 1.29 (m, 12H), 1.27 (m, 24H), 0.95 (t, 12H), 0.85 (t, 12H). ¹³C NMR (100 MHz, CDCl₃), δ (ppm): 185.20, 161.72, 157.82, 157.36, 156.29, 154.51, 151.91, 141.56, 138.85, 138.33, 137.98, 137.40, 136.32, 134.13, 133.40, 131.06, 127.69, 127.16, 121.03, 118.74, 113.99, 113.56, 100.02, 71.09, 67.27, 61.27, 37.80, 34.54, 30.65, 30.31, 29.49, 28.68, 28.05, 22.93, 22.06, 21.54, 13.17, 13.04, 9.93. MALDI-TOF MS: calculated. For $C_{114}H_{112}F_4N_4O_4S_4$ m/z = 1806.39; found, 1805.82.



Fig. S1 TGA curve of NTO-4F with a heating rate of 10 °C min⁻¹ under nitrogen atmosphere.



Fig. S2 The LUMO and HOMO distribution of NTO-4F and NT-4F (alkyl chains are simplified to methyl) calculated at B3LYP/6-31G** level.



Fig. S3 The UV-vis absorption spectra of NTO-4F and NT-4F (alkyl chains are simplified to methyl) calculated at B3LYP/6-31G** level.



Fig. S4 UV-vis absorption spectra of NTO-4F in chloroform.



Fig. S5 UV-vis absorption spectra of PM6:NTO-4F-based blend films.



Fig. S6 Cyclic voltammetry curves for PM6 and NTO-4F.



Fig. S7 J-V (a) and EQE (b) curves of solar cells based on PM6:NTO-4F blends with

different D/A ratios.



Fig. S8 J-V curves of solar cells with different TA temperature.



Fig. S9 J-V curves of solar cells with different TA time.



Fig. S10 Plots of PCE against V_{oc} in PSCs with PCE $\ge 10\%$ and $V_{oc} \ge 0.9$ V.



Fig. S11 PCE trends of PM6:NTO-4F-based devices versus TA treatment time under 180 °C.



Fig. S12 Photoluminescence (PL) quenching efficiency of PM6:NTO-4F blends compared to PM6 (a) and NTO-4F (b) pure films.



Fig. S13 (a) Hole mobilities of corresponding devices, (b) Electron mobilities of corresponding devices.

Table S1 Photovoltaic performance of the PSCs based on PM6:NTO-4F with different

D/A ratios	V _{oc} (V)	$J_{\rm sc}$ (mA cm ⁻²)	FF	PCE (%)
1:1	1.02	14.4	0.40	5.9
1.25:1	1.02	14.9	0.40	6.1
1.5:1	1.02	13.2	0.40	5.4

TA temperature	TA time	V _{oc} (V)	J _{sc} (mA cm ⁻²)	FF	PCE (%)
140°C	5 min	1.01	17.9	0.57	10.3
150°C	5 min	0.99	18.2	0.57	10.3
160°C	5 min	0.99	17.6	0.61	10.6
170°C	5 min	0.99	18.0	0.61	10.9
180°C	5 min	0.99	19.1	0.61	11.5
190°C	5 min	0.98	18.1	0.62	10.9
200°C	5 min	0.98	18.3	0.57	10.2

Table S2 Photovoltaic performance of the PSCs based on PM6:NTO-4F (1.25:1, w/w) with different TA treatment temperature under the illumination of AM 1.5 G, 100 mW cm⁻².

Table S3 Photovoltaic performance of the PSCs based on PM6:NTO-4F (1.25:1, w/w)

TA temperature	TA time	V _{oc} (V)	$J_{\rm sc}$ (mA cm ⁻²)	FF	PCE (%)
180°C	1 min	0.99	18.2	0.57	10.3
180°C	3 min	0.99	17.7	0.62	10.9
180°C	5 min	0.99	19.1	0.61	11.5
180°C	7 min	0.99	19.0	0.59	11.0
180°C	15 min	0.98	19.3	0.54	10.2
180°C	30 min	0.98	18.9	0.52	9.6
180°C	45 min	0.97	18.8	0.51	9.3
180°C	60 min	0.97	18.4	0.50	8.9

with different TA treatment time under the illumination of AM 1.5 G, 100 mW cm⁻².

Donor	NFA	E_g^{opt} (eV)	V _{oc} (V)	E _{loss} (eV)	J_{sc} (mA cm ⁻²)	PCE _{max} (%)	Ref.
SM1	IDIC	1.62	0.905	0.71	15.18	10.11	5
PBT1-C	TPTT-IC	1.63	0.96	0.67	15.6	10.5	6
PBDB-T	FDICTF	1.63	0.94	0.69	15.81	10.06	7
J71	BDT-IC	1.53	0.92	0.61	17.3	10.5	8
PBT1-C	TPTTT-2F	1.56	0.92	0.64	17.63	12.1	9
J71	BT-IC	1.43	0.90	0.53	17.75	10.46	10
FTAZ	IOIC2	1.55	0.90	0.65	19.7	12.3	11
PBT1-EH	ITCPTC	1.58	0.95	0.63	16.5	11.8	12
PBDB-T	FDNCTF	1.6	0.93	0.67	16.5	11.2	13
PBDB-T	DF-PCIC	1.59	0.91	0.68	15.66	10.14	14
PBDB-T	F-M	1.65	0.98	0.67	14.56	10.08	15
DRTB-T	F-2C1	1.54	0.969	0.57	17.24	10.76	16
PBDB-T	ITCC-M	1.68	1.03	0.65	14.8	10.1	17
PBDB-T	IT-M	1.60	0.94	0.66	17.44	12.1	18
PTB7-Th	6TBA	1.52	0.98	0.54	15.2	10.13	19
PDTB-ET-T	IT-4F	1.54	0.90	0.63	20.73	14.2	20
J71	ITCF	1.57	0.91	0.666	18.48	13.3	21
PBT1-C	ITCPTC	1.58	0.94	0.64	17.0	12.7	22
J71	MeIC	1.65	0.92	0.73	18.41	12.5	23
PFBDB-T	C8-ITIC	1.55	0.94	0.61	19.6	13.2	24
PDTB-ET-T(P ₂)	IT-4F	1.54	0.90	0.64	20.7	14.2	25
T1	IT-4F	1.54	0.9	0.64	21.5	15.1	26
PBDB-TF	AT-4Cl	1.60	0.90	0.70	19.52	13.3	27

Table S4 Photovoltaic results of recent high performance PSC devices (PCE $\geq 10\%$ and $V_{oc} \geq 0.9$ V) based on polymer donor and non-fullerene acceptor (NFA).

PBQ-4F	ITIC	1.57	0.95	0.62	17.87	11.34	28
FTAZ	ITIC2	1.53	0.925	0.61	18.88	11	29
PBDB-T	ITC6-IC	1.6	0.97	0.63	16.41	11.61	30
PBDB-T	ITOIC-2F	1.45	0.90	0.55	21.04	12.2	31
PPBDB-TF	IDIC-C4Ph	1.62	0.94	0.68	19.06	14.0	32
PBDB-T	IT-DM	1.63	0.97	0.66	16.48	11.29	33
PBDB-T	IT-OM-2	1.59	0.93	0.66	17.53	11.9	34
PBDB-T	ITCC	1.67	1.01	0.66	15.9	11.4	35
J61	m-ITIC	1.58	0.912	0.67	18.31	11.77	36
PBDB-T	IDTTIC	1.51	0.919	0.59	16.9	11.2	37
PBDB-T	NIBT	1.63	1.04	60	17.34	10.8	38
TBD-3F	IT-4F	1.55	0.92	72.3	21.6	14.4	39
PM6	IT-DOH	1.53	0.96	73	17.78	12.5	40
PBDB-T	IDTCN-O	1.53	0.91	73.2	19.96	13.28	41
PTQ10	ZITI-S	1.61	0.96	65.75	16.94	10.69	42
PBDB-T	a-BTTIC	1.43	0.904	74	20.31	13.6	43
PBDB-T	BDSePh-Cl	1.41	0.92	73.1	20.35	13.68	44
PBDB-T	o-F-ITIC	1.72	0.918	66.97	18.07	11.11	45
PM6	NTO-4F	1.55	0.99	0.56	19.1	11.5	This Work

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