

Regulating the Electron Transporting Property of the Indacenodithiophene Derivatives for
Perovskite Solar Cells with PCEs up to 19.51 %

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Experimental Section

Materials

In this work, all major materials were purchased from commercial suppliers and used without further purification, including P3CT (Rieke Metals), PbI₂ (*p*-OLED, >99.99 %), MAI (*p*-OLED, ≥99.5 %), PCBM (*p*-OLED, ≥99 %), C60 (*p*-OLED), BCP (*p*-OLED), Rhodamine 101 (Sigma-Aldrich), DMF (Sigma-Aldrich, 99.8 %), DMSO (Sigma-Aldrich, 99.8 %) and CB (Sigma-Aldrich, 99.8 %). IDTCN, IDT6CN and ITCPTC were synthesized according to the previous work [1].

Device Fabrication

The indium tin oxide (ITO) glass substrates were sequentially washed by sonication using detergent, deionized water, ethanol and acetone. The hole transporting layer P3CT-Na [2] was spin-coated on

ITO substrates at 4000 rpm for 60 s followed by annealing at 140 °C for 30 min. Then the samples were transferred into a N₂-filled glovebox. A CH₃NH₃PbI₃ precursor solution (1.4 M in DMF:DMSO mixed solution with a v/v of 4:1) was spin-coated in a two-step program at 400 and 5000 rpm for 3 s and 30 s, respectively. During the second step, 200 μL of chlorobenzene was dropped onto the spinning substrate at 10 s after the start-up. Next, the as-spun perovskite layer was annealed on a hot plate at 60 °C for 1 min and at 80 °C for 2 min to remove the solvent and form the perovskite phase. Then, PCBM, IDTCN, IDT6CN and ITCPTC solutions in chlorobenzene were spin-coated onto the perovskite layer. To fabricate ITO/P3CT-Na/Perovskite/ETL/Rhodamine 101/LiF/Ag device, Rhodamine 101 was subsequently deposited by spin coating Rhodamine 101 solution (0.05 wt% in isopropanol) at 1000 rpm for 40 s [3]. Afterwards, 1 nm thick LiF was thermally evaporated, and an approximately 100 nm thick Ag counter electrode was deposited on top to finish the device fabrication. In this device architecture, Rhodamine 101 and LiF layers work as the double interlayers between the ETL and the Ag electrode. To fabricate ITO/P3CT-Na/Perovskite/interlayer/C60/BCP/Ag device, C60 (20 nm) and BCP (8 nm) were evaporated under high vacuum on top of the ETL sequentially. Finally, a 100 nm thick Ag electrode was deposited through a shadow mask. The active area of our device is 0.09 cm².

Characterization

UV-vis absorption spectra were measured on a Shimadzu UV-2450 absorption spectrophotometer. The morphology of perovskite films were characterized by FE-SEM images (JSM-7800F). AFM images were collected in air on a Bruker AFM using a tapping mode. The current-voltage (*J-V*) curves were measured under 100 mW cm⁻² (AM 1.5 G) simulated sunlight using Keithley 2400 in

conjunction with a Newport solar simulator (94043A). The external quantum efficiency (EQE) was calculated from the photocurrent measurement under monochromatic illuminations at different wavelengths, using a 150 W xenon lamp and a monochromator. Steady-state PL spectra were recorded on a Hitachi F-4600 FL Spectrophotometer. Time-resolved PL decay curves were measured by a single photon counting spectrometer from Edinburgh Instruments (FS5). The impedance spectra were measured with CHI in the dark at a bias of 0.9 V.

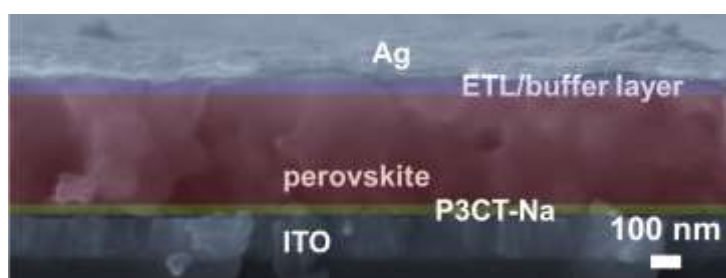


Figure S1. Cross-sectional SEM images of inverted perovskite solar cells with IDT-based ETLs.

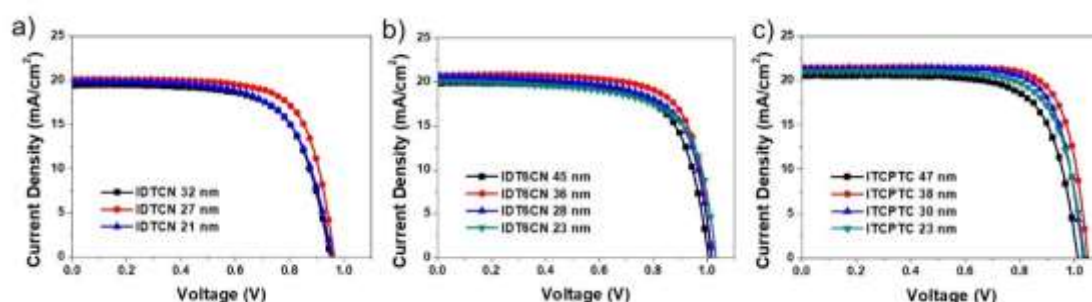


Figure S2. J - V curves of PSCs using the three ETLs in different thicknesses.

Table S1. Photovoltaic data of PSCs using IDTCN ETL in different thicknesses.

IDTCN	V_{oc} (mV)	J_{sc} (mA cm ⁻²)	FF	PCE (%)
32 nm	950	19.45	0.68	12.56
27 nm	964	20.13	0.71	13.86
21 nm	957	19.89	0.66	12.59

Table S2. Photovoltaic data of PSCs using IDT6CN ETL in different thicknesses.

IDT6CN	V_{oc} (mV)	J_{sc} (mA cm ⁻²)	FF	PCE (%)
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45 nm	1006	19.87	0.70	14.07
36 nm	1018	20.80	0.73	15.56
28 nm	1017	20.64	0.70	14.61
23 nm	1033	20.09	0.69	14.23

Table S3. Photovoltaic data of PSCs using ITCPTC ETL in different thicknesses.

IDT6CN	V_{oc} (mV)	J_{sc} (mA cm ⁻²)	FF	PCE (%)
47 nm	1012	20.52	0.72	14.90
38 nm	1049	21.49	0.77	17.42
30 nm	1032	21.33	0.76	16.72
23 nm	1037	21.01	0.73	15.96

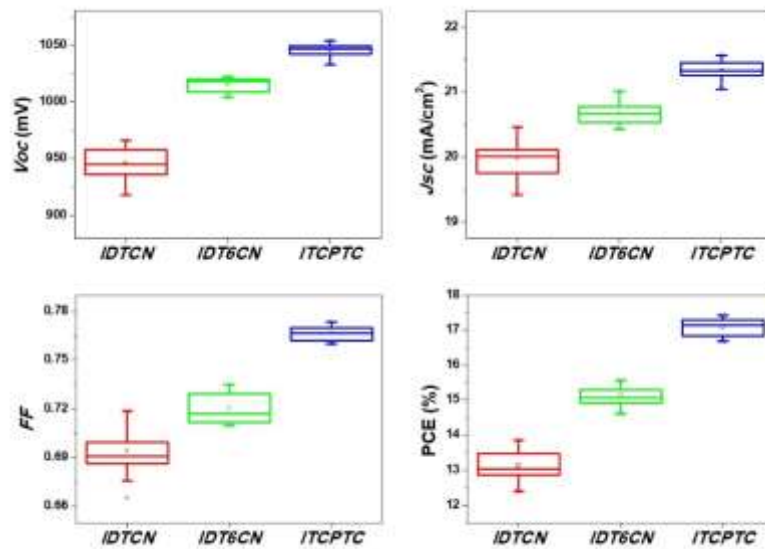


Figure S3. Photovoltaic parameters statistic for devices using different ETLs (ITO/P3CT-

Na/Perovskite/IDT series/Rhodamine 101/LiF/Ag).

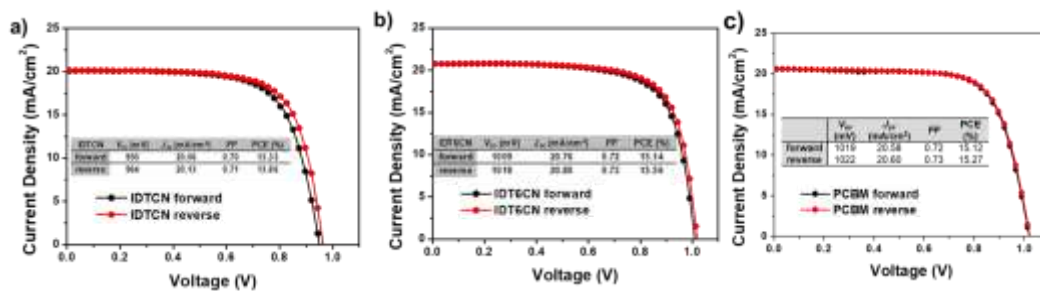


Figure S4. J - V curves scanned from forward and reverse directions in a) IDTCN, b) IDT6CN and c) PCBM-based devices.

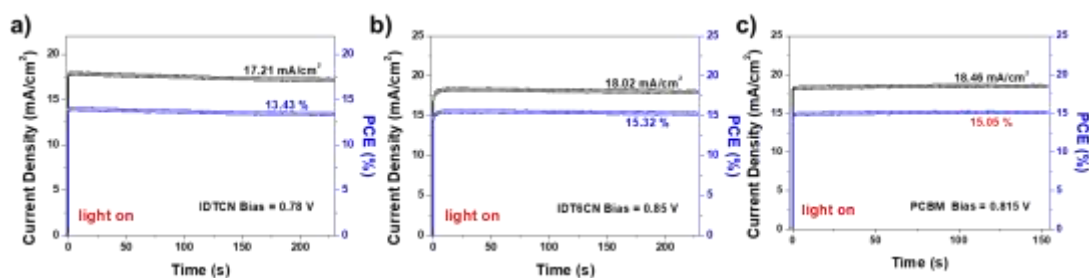


Figure S5. Steady J_{sc} and PCE measured at the maximum power output point of the optimized devices with a) IDTCN, b) IDT6CN and c) PCBM ETLs.

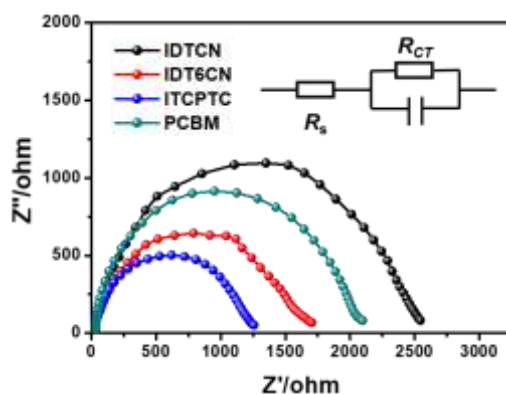


Figure S6. Nyquist plots of PSCs with different ETLs measured in the dark condition. Inset is the equivalent circuit model.

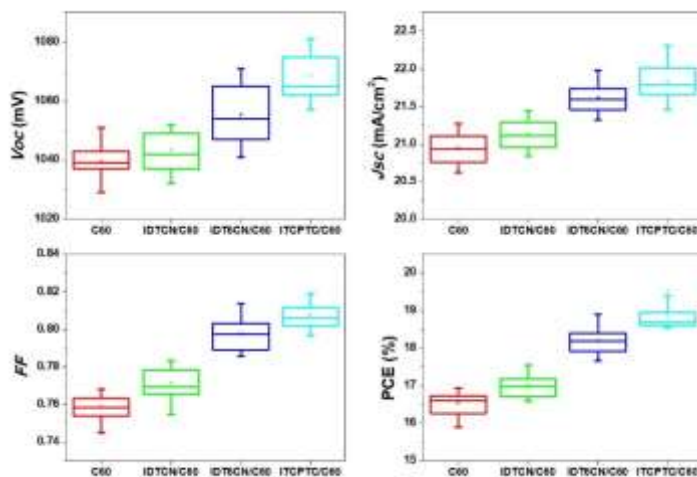


Figure S7. Photovoltaics parameters statistic for devices using different interlayers (ITO/P3CT-Na/Perovskite/IDT series/C60/BCP/Ag).

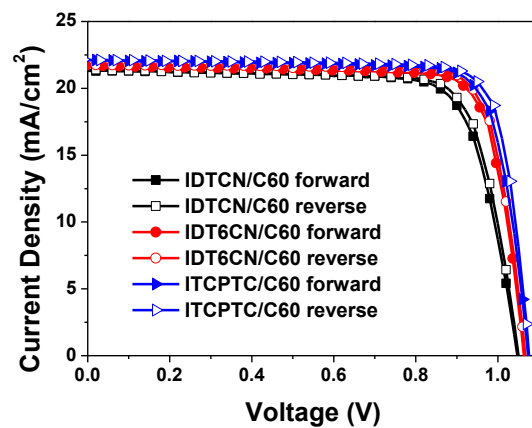


Figure S8. *J-V* curves scanning from forward and reverse directions of PSCs adopting the three IDT-based interlayers.

Table S4. Photovoltaic data of PSCs using the three IDT-based interlayers when scanning from different directions.

Device		V_{oc} (mV)	J_{sc} (mA cm ⁻²)	FF	PCE (%)
IDTCN/C ₆₀	Forward	1047	21.32	0.77	17.16
	Reverse	1050	21.36	0.78	17.54
IDT6CN/C ₆₀	Forward	1063	21.69	0.80	18.50
	Reverse	1067	21.78	0.81	18.89
ITCPTC/C ₆₀	Forward	1074	22.10	0.81	19.14
	Reverse	1076	22.14	0.82	19.51

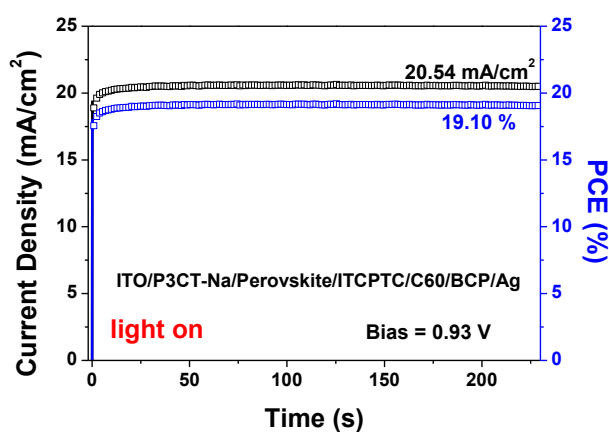


Figure S9. Steady J_{sc} and PCE measured at the maximum power output point of the optimized devices with ITCPTC interlayer. The device architecture is: ITO/P3CT-Na/Perovskite/ITCPTC/C60/BCP/Ag.

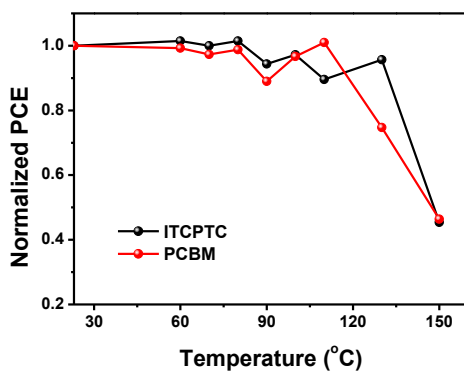


Figure S10. The thermal stability test of PSCs with ITCPTC or PC₆₁BM ETL measured in a glove box without encapsulation (the heating time is 30 minutes).

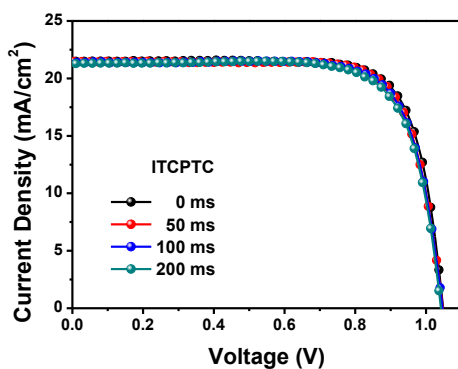


Figure S11. J - V curves of inverted PSCs using ITCPTC ETL scanning at different delay times.

Table S5. Photovoltaic data of PSCs using ITCPTC ETL scanning at different delay times.

Delay time	V_{oc} (mV)	J_{sc} (mA cm ⁻²)	FF	PCE (%)
0 ms	1049	21.49	0.77	17.42
50 ms	1048	21.48	0.77	17.31
100 ms	1046	21.40	0.76	17.07
200 ms	1042	21.01	0.76	16.82

References:

- [1] W. Gao, M. Zhang, T. Liu, R. Ming, Q. An, K. Wu, D. Xie, Z. Luo, C. Zhong, F. Liu, F. Zhang, H. Yan, C. Yang, *Adv. Mater.* 2018, 1800052.
- [2] X. Li, X. Liu, X. Wang, L. Zhao, T. Jiu, J. Fang, *J. Mater. Chem. A* 3 (2015) 15024-15029.
- [3] K. Sun, J. Chang, F. H. Isikgor, P. Li, J. Ouyang, *Nanoscale* 7 (2015) 896-900.