

Supporting Information

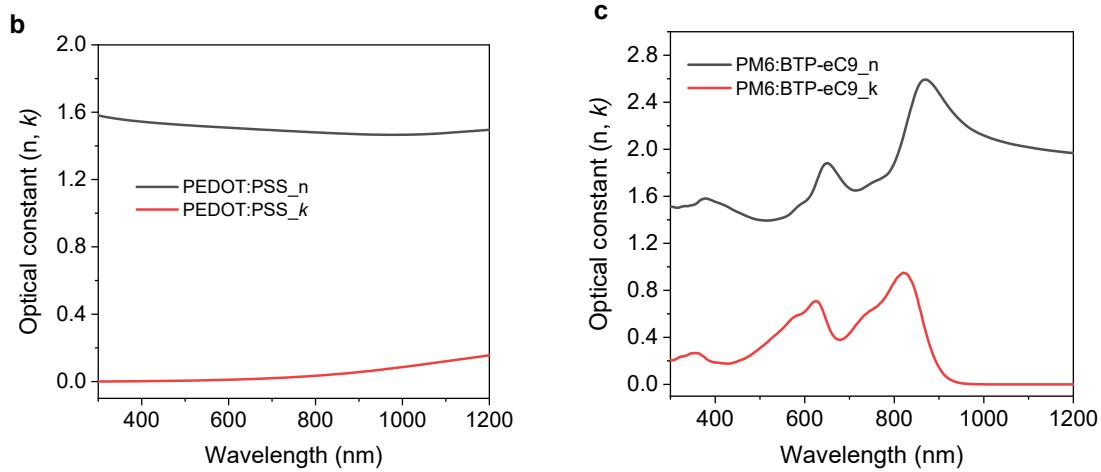
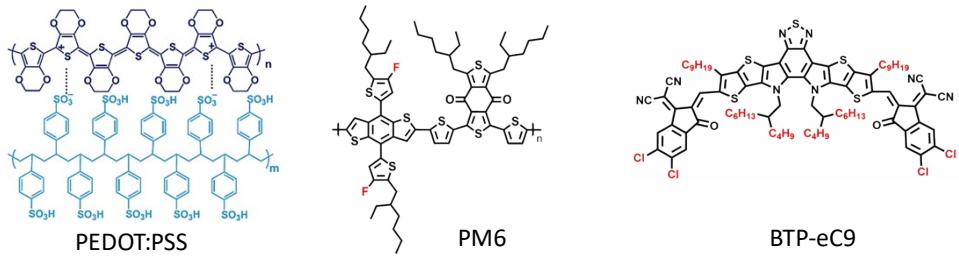
**Fully evaporated interfacial layers for high-performance and batch-to-batch reproducible organic solar modules**

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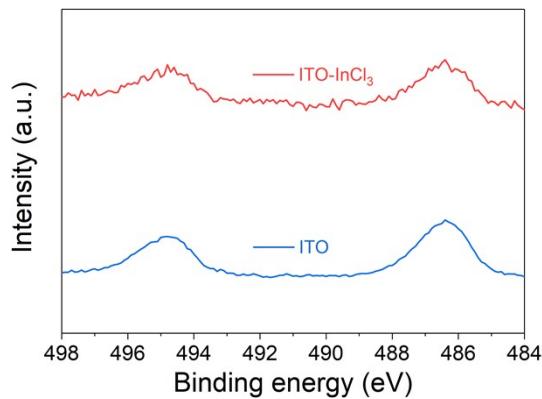
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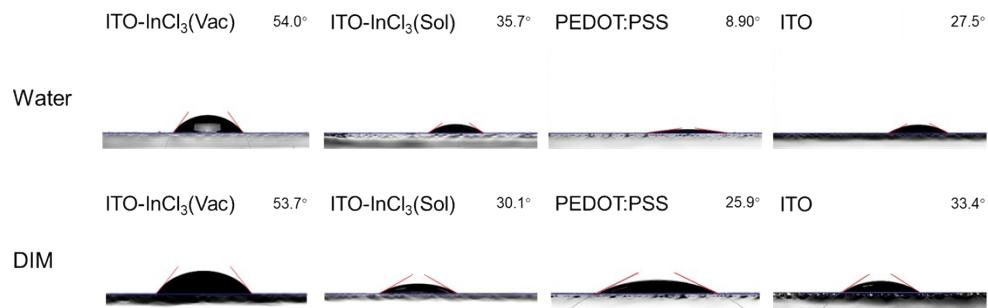
Corresponding authors E-mail: [liuquan@nimte.ac.cn](mailto:liuquan@nimte.ac.cn), [geziyi@nimte.ac.cn](mailto:geziyi@nimte.ac.cn).



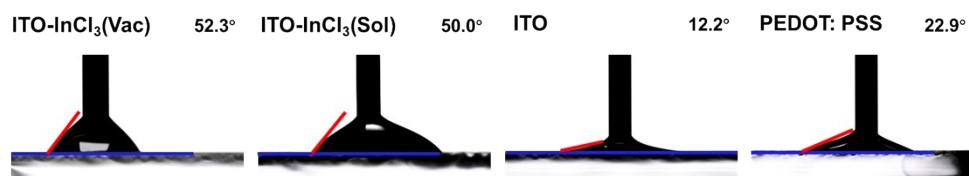
**Figure S1.** (a) Chemical structures of PEDOT:PSS, PM6 donor and BTP-eC9 acceptor. (b) and (c) are optical constants of PEDOT:PSS hole-transporting material and PM6:BTP-eC9 active material, respectively.



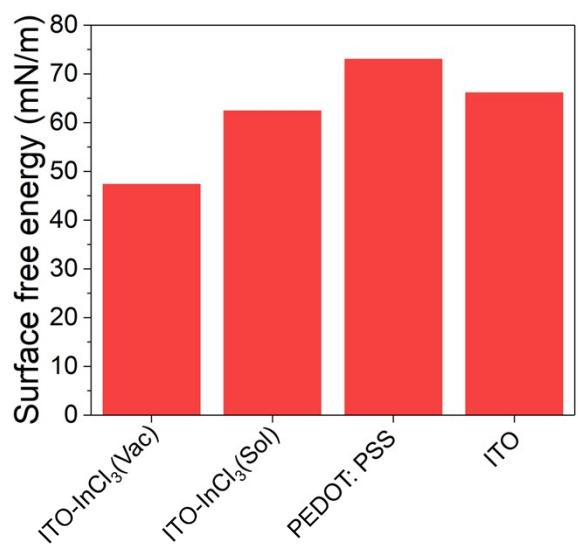
**Figure S2.** XPS spectra of Sn 3d orbitals for bare ITO and ITO-InCl<sub>3</sub>.



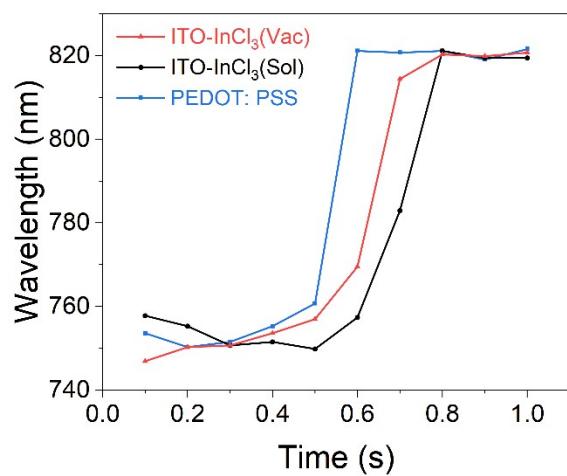
**Figure S3.** Contact angle images of water and DIM.



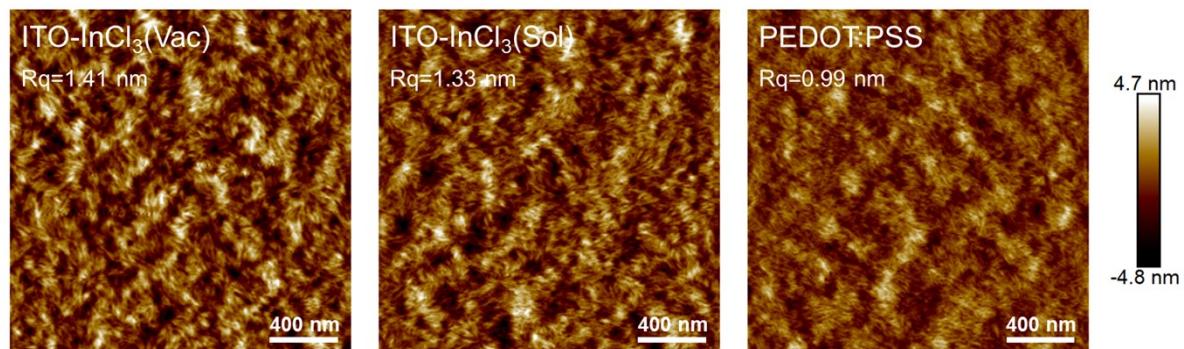
**Figure S4.** The receding contact angle images of water on different HTL-coated ITO substrates.



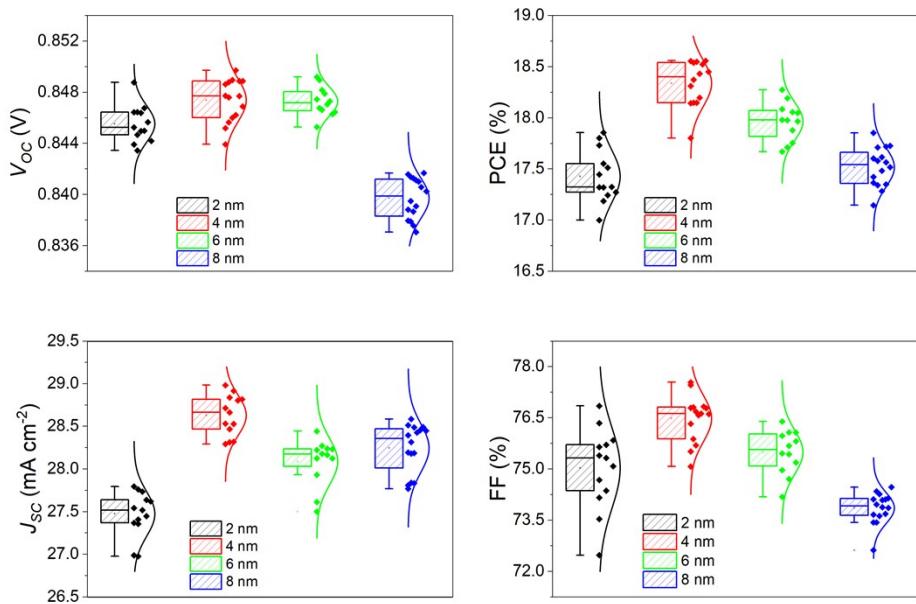
**Figure S5.** Surface free energy of different substrates.



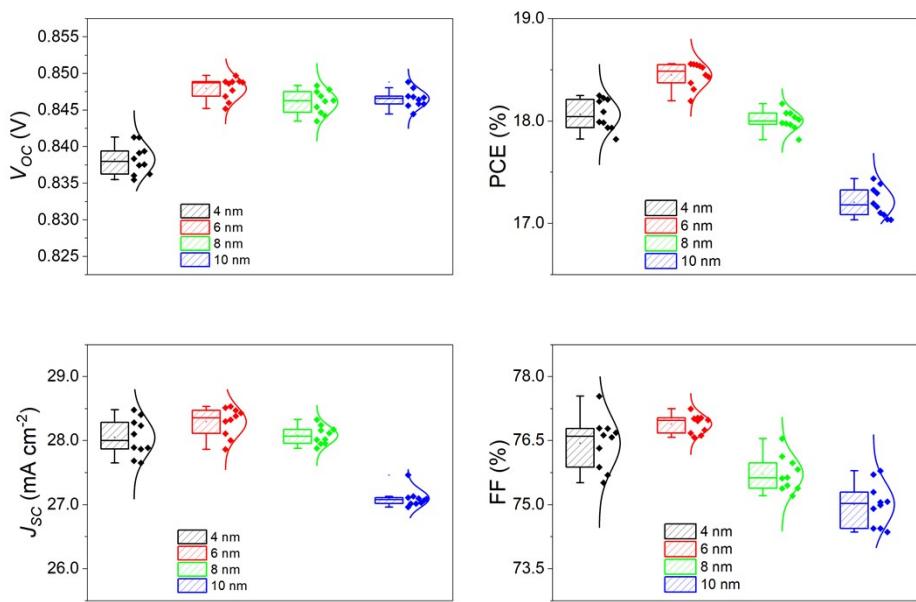
**Figure S6.** Time evolution of peak location and intensity of acceptor.



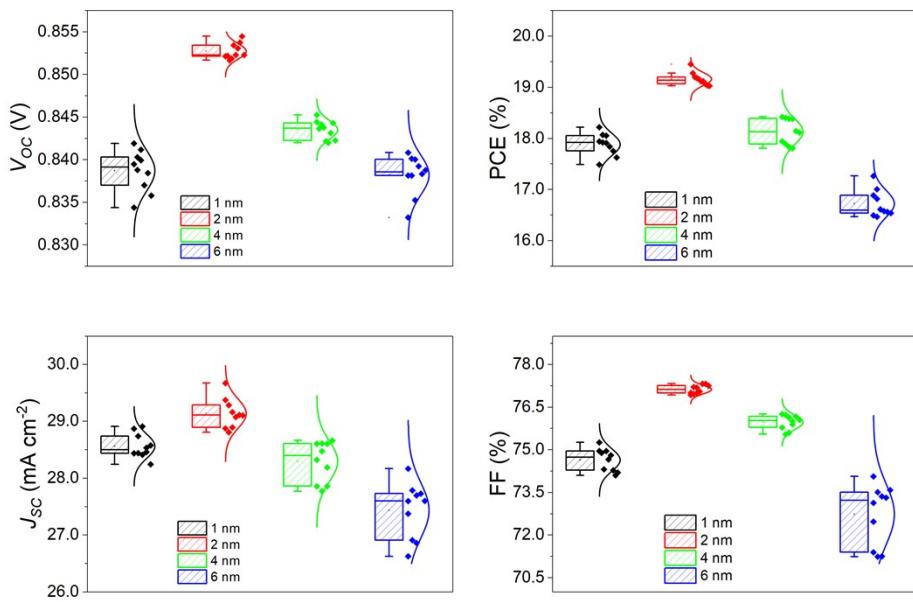
**Figure S7.** AFM images of PM6: BTP-eC9 blend films on different HTL coated ITO substrates.



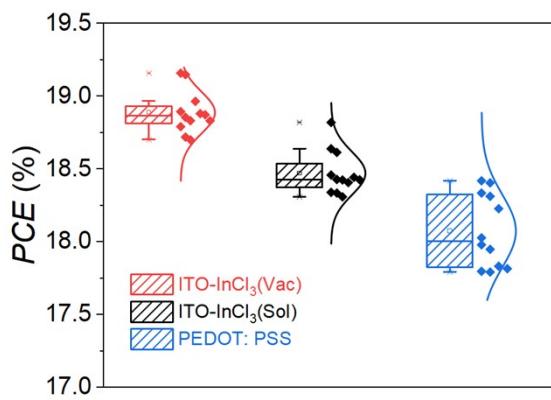
**Figure S8.** The effect of different thickness of  $\text{C}_60$  on the performance of organic solar cells.



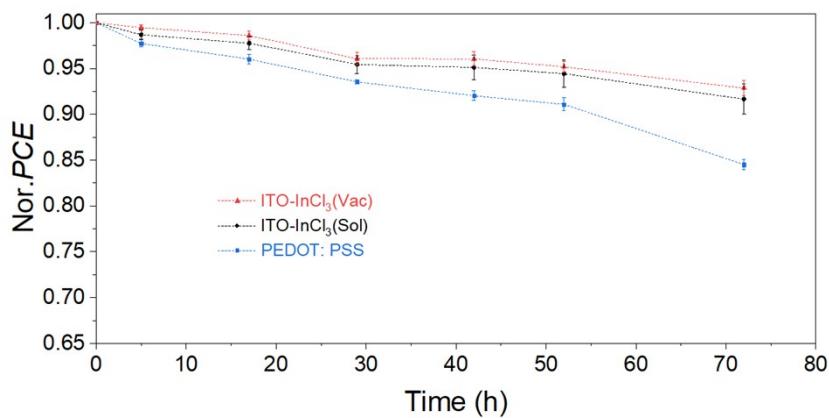
**Figure S9.** The effect of different thickness of BCP on the performance of organic solar cells.



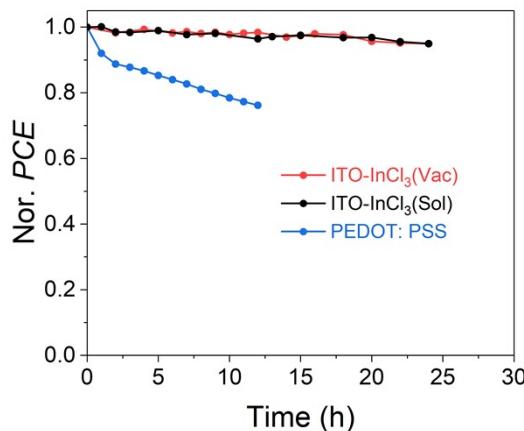
**Figure S10.** The effect of different thickness of  $\text{InCl}_3$  on the performance of organic solar cells.



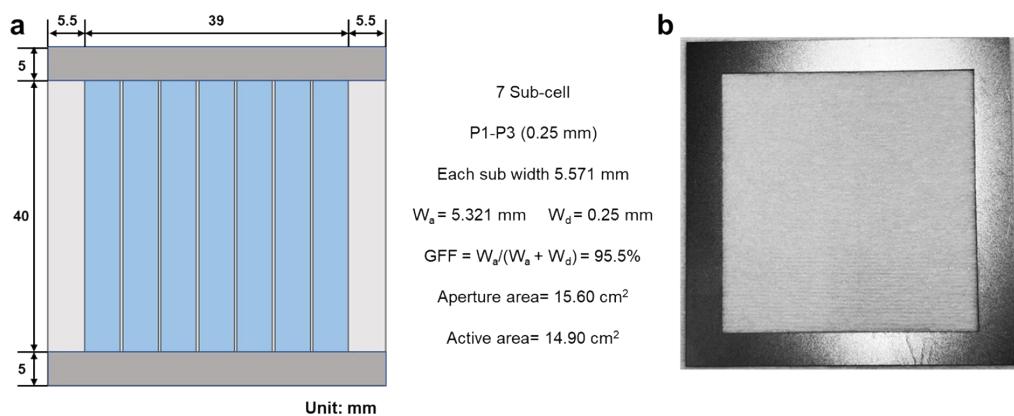
**Figure S11.** Statistical distributions of the photovoltaic parameters for OSC devices, using PM6: L8BO as active layers, are presented for devices with different hole interfacial layers.



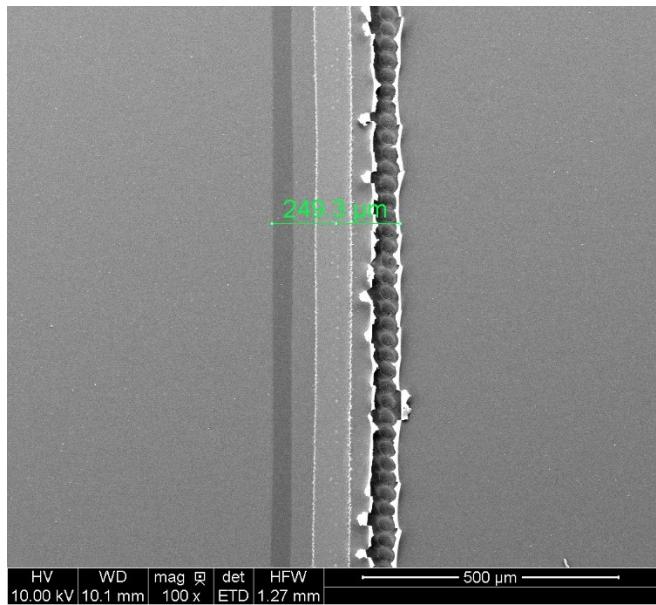
**Figure S12.** Normalized PCE of OSCs with different hole transport layers under continuous illumination. (Data from at least three independent devices)



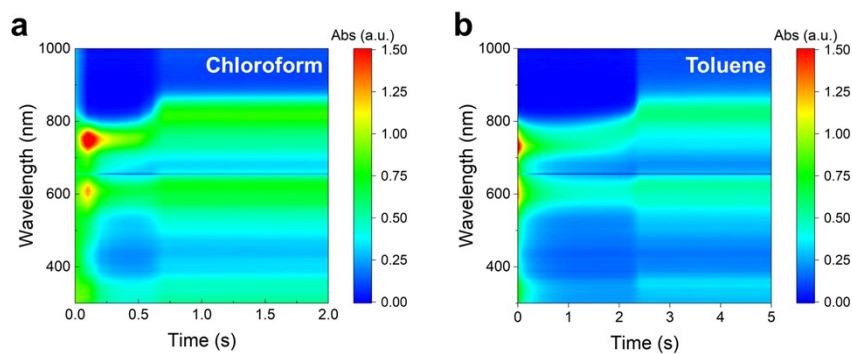
**Figure S13.** Thermal stability of OSCs with different hole transport layers.



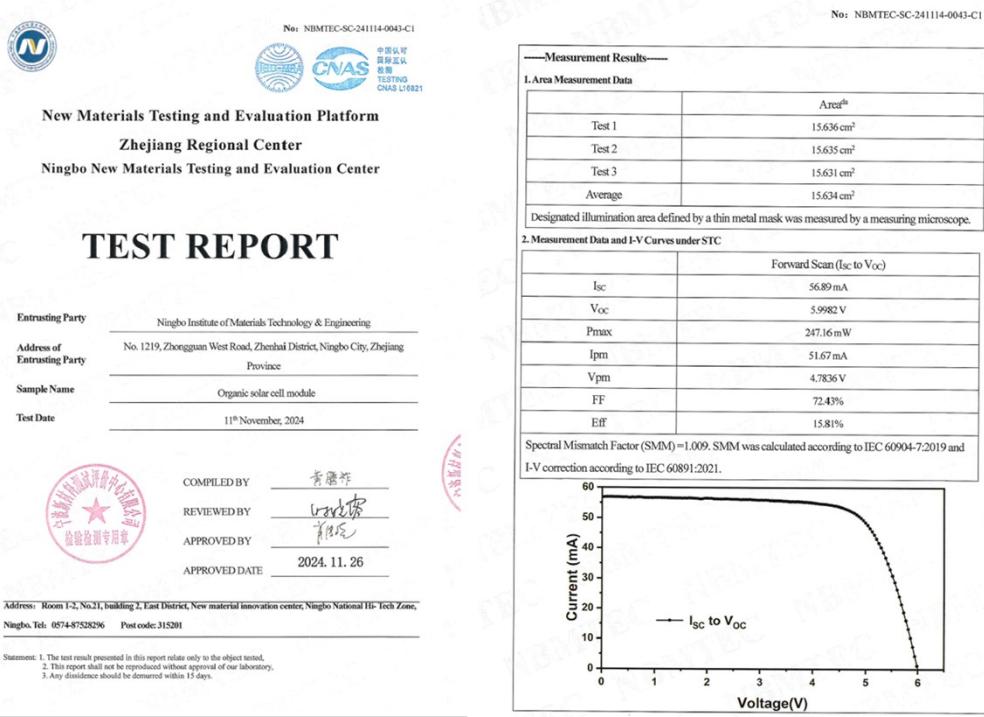
**Figure S14.** (a) The details for the OPV modules. (b) Practicality picture of the mask used for testing large-area modules.



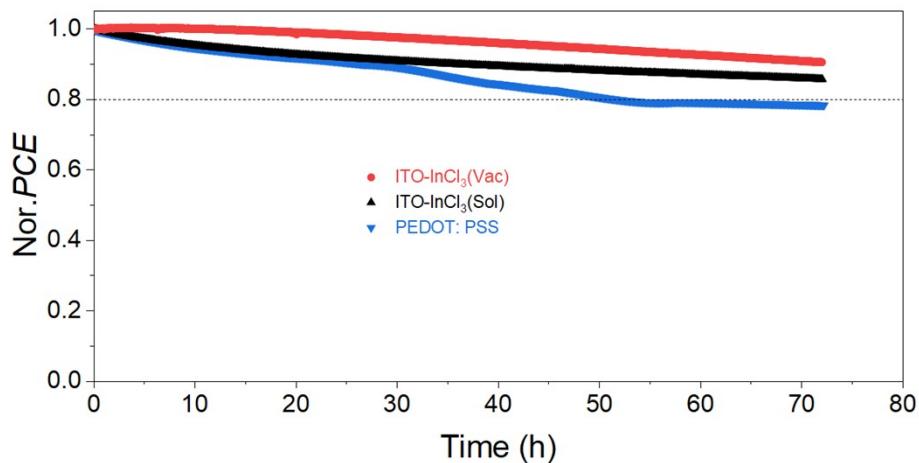
**Figure S15.** SEM image showing dead zone width.



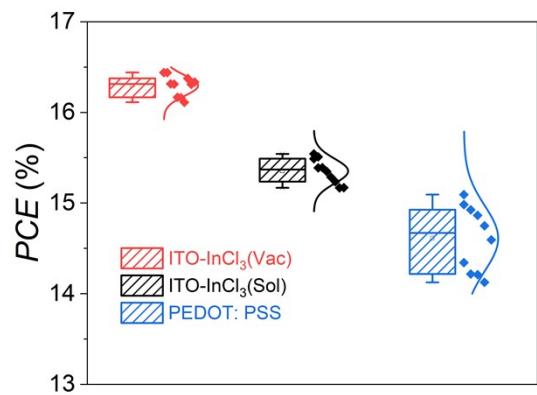
**Figure S16.** Time-dependent contour maps of in situ UV-vis absorption spectra of the two active layers during spin coating.



**Figure S17.** The certification report of the champion OPV module.



**Figure S18.** Normalized PCE under maximum power point (MPP) tracking for OSC modules with different hole transport layers, measured under continuous 1-sun illumination (LED light source: 300-1200 nm spectral range).



**Figure S19.** Statistical distributions of the photovoltaic characteristics for OPV modules with sublimed interface layers.

**Table S1** Contact angle of water and DIM on various hole transport layers, receding angle of DIM on distinct hole transport layers, and surface free energy of different hole transport layers.

Hole transport layers	Contact angle of water	Contact angle of DIM	Receding angle of DIM	Surface free energy (mN/m)
ITO	27.5°	33.4°	22.9°	66.110
PEDOT: PSS	8.90°	25.9°	12.2°	72.993
ITO-InCl <sub>3</sub> (Sol)	35.7°	30.1°	50.0°	62.388
ITO-InCl <sub>3</sub> (Vac)	54.0°	53.7°	52.3°	47.336

**Table S2** Detailed photovoltaic parameters of organic solar cells based on various hole transport layers.

	HTL	V <sub>oc</sub> (V)	J <sub>sc</sub> (mA/cm <sup>2</sup> )	FF (%)	PCE (%)
Device	ITO-InCl <sub>3</sub> (Vac)	0.880 (0.88±0.002)	27.87 (27.48±0.39)	78.3 (77.9±0.7)	19.16 (18.89±0.27)
	ITO-InCl <sub>3</sub> (Sol)	0.873 (0.88±0.007)	27.42 (27.20±0.22)	78.3 (77.1±1.2)	18.82 (18.47±0.25)
	PEDOT: PSS	0.881 (0.84±0.011)	26.93 (26.53±0.40)	77.6 (77.0±0.6)	18.42 (18.07±0.27)

**Table S3** Detailed photovoltaic parameters of modules based on varied HTLs.

Active layer	Aperture area	Active area	Active area	Aperture area	Ref.
	PCE (%)	PCE (%)	(cm <sup>2</sup> )	(cm <sup>2</sup> )	
PM6: L8BO: PC <sub>61</sub> BM	15.43	16.100	11.080	11.3	1
D18: L8BO	14.98	15.440	19.300	19.9	2
PM6: BO-4Cl	15.740	16.061	18.924	19.310	3
PM6: D18: L8BO	/	16.030	15.640	/	4
PM6: BO-4Cl	14.350	14.790	18.730	19.300	5
PM6: PC <sub>71</sub> BM: BTP-eC9	/	13.070	12.200	/	6
PM6: DTY6	13.98	14.400	18.000	18.05	7
PM6: Y6: BTO: PC <sub>61</sub> BM	/	14.260	36.000	/	8
PM6: BTP-eC9	11.35	14.070	25.210	31.24	9
TPD-3F: IT-4F	/	10.080	20.400	/	10
PM6: L8BO	/	15.200	18.730	/	11
PM6: PBQx-TCl: PY-IT	15.75	16.260	18.700	19.3	12
PM6: T8	/	15.380	7.500	/	13
PBDB-TFCI: D18-Cl: PY-IT	12.7	15.100	36.200	43	14
PBQx-TF: eC9-2Cl	15.1	16.100	21.000	23.6	15
D18-Cl: BTP-4F-P2EH	/	17.000	17.600	/	16
PM6: D18: BTP-eC9	15.55	16.700	15.600	16.8	17
PBQx-TCl: eC9-2Cl	15.5	16.500	22.170	23.6	18
PM6: Y6-C12: PC <sub>61</sub> BM	14.5	15.000	200	204	19

## Supporting information reference

- 1 E. Feng, C. Zhang, J. Chang, Y. Han, H. Li, Q. Luo, C.-Q. Ma, H.-L. Yip, L. Ding, J. Yang, *Cell Rep. Phys. Sci.* 2024, **5**, 101883.
- 2 Y. H. Li, Z. Y. Jia, P. H. Huang, T. Y. Liu, D. Q. Hu, Y. L. Li, H. Liu, X. H. Lu, S. R. Lu, X. X. Yin, Y. Yang, *Adv. Energy Mater.* 2024, **14**, 2304000.
- 3 Z. Y. Jia, J. N. Pan, X. Chen, Y. H. Li, T. Y. Liu, H. B. Zhu, J. Z. Yao, B. Y. Yan, Y. Yang, *Energy Environ. Sci.* 2024, **17**, 3908.
- 4 B. Zhang, W. J. Chen, H. Y. Chen, G. Zeng, R. Zhang, H. X. Li, Y. F. Wang, X. D. Gu, W. W. Sun, H. Gu, F. Gao, Y. W. Li, Y. F. Li, *Energy Environ. Sci.* 2024, **17**, 2935.
- 5 J. Y. Fan, Z. X. Liu, J. Rao, K. R. Yan, Z. Chen, Y. X. Ran, B. Y. Yan, J. Z. Yao, G. H. Lu, H. M. Zhu, C. Z. Li, H. Z. Chen, *Adv. Mater.* 2022, **34**, 2110569.
- 6 Y. Y. Jiang, X. Y. Dong, L. L. Sun, T. F. Liu, F. Qin, C. Xie, P. Jiang, L. Hu, X. Lu, X. M. Zhou, W. Meng, N. Li, C. J. Brabec, Y. H. Zhou, *Nat. Energy* 2022, **7**, 352.
- 7 S. Dong, T. Jia, K. Zhang, J. H. Jing, F. Huang, *Joule* 2020, **4**, 2004.
- 8 H. Y. Chen, R. Zhang, X. B. Chen, G. Zeng, L. Kobera, S. Abbrent, B. Zhang, W. J. Chen, G. Y. Xu, J. Oh, S. H. Kang, S. S. Chen, C. Yang, J. Brus, J. H. Hou, F. Gao, Y. W. Li, Y. F. Li, *Nat. Energy* 2021, **6**, 1045.
- 9 X. Y. Dong, Y. Y. Jiang, L. L. Sun, F. Qin, X. M. Zhou, X. Lu, W. Wang, Y. H. Zhou, *Adv. Funct. Mater.* 2022, **32**, 2110209.
- 10 C. Y. Liao, Y. Chen, C. C. Lee, G. Wang, N. W. Teng, C. H. Lee, W. L. Li, Y. K. Chen, C. H. Li, H. L. Ho, P. H. S. Tan, B. H. Wang, Y. C. Huang, R. M. Young, M. R. Wasielewski, T. J. Marks, Y. M. Chang, A. Facchetti, *Joule* 2020, **4**, 189.
- 11 J. Xiang, Z. X. Liu, H. Chen, C. Z. Li, *Adv. Mater.* 2023, **35**, 2303729.
- 12 T. Y. Chen, X. J. Zheng, D. Wang, Y. X. Zhu, Y. N. Ouyang, J. W. Xue, M. T. Wang, S. L. Wang, W. Ma, C. F. Zhang, Z. F. Ma, S. X. Li, L. J. Zuo, H. Z. Chen, *Adv. Mater.* 2024, **36**, 2308061
- 13 R. Sun, T. Wang, X. R. Yang, Y. Wu, Y. Wang, Q. Wu, M. J. Zhang, C. J. Brabec, Y. F. Li, J. Min, *Nat. Energy* 2022, **7**, 1087.
- 14 J. Q. Wang, Y. F. Wang, K. H. Xian, J. W. Qiao, Z. H. Chen, P. Q. Bi, T. Zhang, Z. Zheng, X. T. Hao, L. Ye, S. Q. Zhang, J. H. Hou, *Adv. Mater.* 2024, **36**, 2305424.
- 15 J. Q. Wang, Y. F. Wang, P. Q. Bi, Z. H. Chen, J. W. Qiao, J. Y. Li, W. X. Wang, Z. Zheng, S. Q. Zhang, X. T. Hao, J. H. Hou, *Adv. Mater.* 2023, **35**, 2301583.
- 16 L. Zhu, M. Zhang, G. Zhou, Z. Wang, W. Zhong, J. Zhuang, Z. Zhou, X. Gao, L. Kan, B. Hao, F. Han, R. Zeng, X. Xue, S. Xu, H. Jing, B. Xiao, H. Zhu, Y. Zhang, F. Liu, *Joule* 2024, **8**, 3153.
- 17 H. Gu, J. Zhu, H. Chen, G. Zeng, X. Chen, X. Tang, J. Xia, T. Zhang, B. Zhang, J. Zhang, J. Ding, Y. Li, Y. Li, *Giant* 2024, **18**, 100286.
- 18 J. Wang, Y. Wang, M. Du, Y. Yu, C. Wang, W. Wang, Q. Guo, Y. Cui, S. Zhang, J. Hou, *Energy Environ. Sci.* 2024, **17**, 8368.
- 19 R. Basu, F. Gumpert, J. Lohbreier, P. O. Morin, V. Vohra, Y. Liu, Y. H. Zhou, C. J. Brabec, H. J. Egelhaaf and A. Distler, *Joule*, 2024, **8**, 970.