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## **Supporting information**

## Well-balanced Hole and Electron Charge Transport in an Organic P-type-Insulator-N-type Layered Sandwich Structure

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**Fig.** S1 (a) Transfer curve of PD transistor with gate leakage current in p-type mode operation, gate-source voltage ( $V_{GS}$ ) ranging from -50 V to 20 V, drain-source voltage ( $V_{DS}$ ) at -50 V. (b) Transfer curve of PPD transistor with gate leakage current in p-type mode operation,  $V_{GS}$  ranging from -50 V to 20 V,  $V_{DS}$  at -50 V.



Fig. S2 Dual sweep transfer curve of (a) PD and (b) PPD transistor with gate leakage current in p-type mode operation,  $V_{GS}$  ranging from -50 V to 20 V,  $V_{DS}$  increasing from -10 V to -50 V



Fig. S3 Transfer curve of a PD transistor in n-type mode with  $V_{GS}$  increasing from -20 V to 50 V and  $V_{DS}$  increasing variations.



Fig. S4 Output curve of a PD transistor in n-type mode with  $V_{DS}$  from 0 V to 50 V and  $V_{GS}$  variations.



**Fig.** S5 50 transfer curves measured to evaluate the uniformity of PD transistors and extract reliable electrical parameters





Fig. S6 Transfer curve of a PPD transistor in n-type mode with  $V_{GS}$  increasing from -20 V to 50 V and  $V_{DS}$  increasing variations.



Fig. S7 Output curve of a PD transistor in n-type mode with  $V_{DS}$  from 0 V to 50 V and  $V_{GS}$  variations.



**Fig.** S8 50 transfer curves measured to evaluate the uniformity of PPD transistors with balanced hole and electron currents due to the parylene interlayer and to extract reliable electrical parameters.





Fig. S9 The transfer curves of four PPD transistors located at the four edges of a 4-inch wafer, the voltage bias conditions of  $V_{GS}$  from -50 V to 20 V and  $V_{DS}$  from -10 V to -50 V.



**Fig.** S10 (a) actual images of PPD transistor with banded state, flat state. Transfer curve of (b) PPD and (c) PD transistor realized on the flexible substrate (Business card) with  $V_{GS}$  from -20 V to 20 V,  $V_{DS}$  from -10 V to -30 V.



**Fig.** S11 Histogram of drain current extracted from the (a) PD and (b) PPD transistor, the voltage bias condition was  $V_{GS}$  at -50 V,  $V_{DS}$  increasing from -20 V to -50 V.



**Fig.** S12 (a) Transfer curve of parylene/PTCDI-C13 transistor at  $V_{DS} = 50 \text{ V}$  (b) Transfer curve of parylene/PTCDI-C13/parylene transistor at  $V_{DS} = 50 \text{ V}$ .



**Fig.** S13 (a) Transfer curve of parylene/DNTT transistor at  $V_{DS}$  = -50 V (b) Transfer curve of parylene/DNTT transistor at  $V_{DS}$  = -50 V.



**Fig.** S14 XPS results for compositional analysis of organic small molecules (a) PTCDI-C13 and (b) DNTT



**Fig.** S15 Analysis of (a) PTCDI-C13 and (b) DNTT UPS examined to extract energy band diagrams.



**Fig.** S16 UV-visible results of (a) PTCDI-C13 and (b) DNTT analyzed to investigate the energy bandgap.



Fig. S17 Energy band diagram of DNTT, PTCDI-C13, Au.



**Fig.** S18 Voltage transfer curve of a Type 2 inverter with PD transistor as pull-up device and PPD transistor as pull-down device.

Semiconductor	V <sub>DS</sub> (V)	I <sub>hole</sub> /I <sub>electron</sub> (A·A <sup>-1</sup> )	V <sub>TS</sub> (V)	Reference
PDPP-Pya (ambipolar)	-60	≈ 10	-22.2	[1]
4HNi (ambipolar)	-60	≈ 10	-10	[2]
PDITTTT (n-type) / C8-BTBT (p-type)	-180	N/A	-25	[3]
ZnO (n-type) / PDPPBTT (p-type)	-30	≈ 10	-10	[4]
C6-DPA (p-type) / F16CuPc (n-type)	-60	≈ 10	-5	[5]
MoS <sub>2</sub> (n-type) / BTBT (p-type)	-3	N/A	-6	[6]
DNTT (p-type)/ PTCDI-C13(n-type)	-10	0.004	-11	This work
DNTT (p-type)	-10	1.08	0	This work
/parylene (dielectric)/ PTCDI-C13(n-type)				

**Table S1** Table for the recent fabrication methods and performance trends ofambipolar transistors.

PD transistor	I <sub>Hole</sub> (V <sub>GS</sub> = -50 V)	I <sub>Electron</sub> (V <sub>GS</sub> = 20 V)	V <sub>TS</sub>
V <sub>DS</sub> = -20 V	$6.61 \pm 2.16 \times 10^{-10}$ A	$3.45 \pm 1.04 \times 10^{-7} \mathrm{A}$	-23 ± 2.83 V
V <sub>DS</sub> = -30 V	$8.65 \pm 6.67 \times 10^{-10}$ A	$6.73 \pm 1.74 \times 10^{-7} \text{ A}$	-29 ± 6.04 V
V <sub>DS</sub> = -40 V	9.90 ± 7.83 × 10 <sup>-10</sup> A	$1.09 \pm 0.25 \times 10^{-6} \text{ A}$	-35 ± 5.20 V
V <sub>DS</sub> = -50 V	$1.08 \pm 0.87 \times 10^{-9} \text{ A}$	$1.57 \pm 0.32 \times 10^{-6} \text{ A}$	-42 ± <sup>4</sup> .56 V

**Table S2** Specifying the average values of  $I_{Hole}$  when  $V_{GS}$  is -50 V and  $V_{DS}$  is increased from -20 V to -50 V,  $I_{Electron}$  when  $V_{GS}$  is 20 V and  $V_{DS}$  is increased from -20 V to -50 V, and  $V_{TS}$  when  $V_{DS}$  is increased from -20 V to -50 V in 50 PD transistor.

PPD transistor	I <sub>Hole</sub> (V <sub>GS</sub> = -50 V)	I <sub>Electron</sub> (V <sub>GS</sub> = 20 V)	V <sub>TS</sub>
V <sub>DS</sub> = -20 V	$2.00 \pm 0.34 \times 10^{-7}$ A	$1.37 \pm 0.84 \times 10^{-8} \text{ A}$	-2.18 ± 2.83 V
V <sub>DS</sub> = -30 V	$2.74 \pm 0.39 \times 10^{-7}$ A	$3.45 \pm 0.46 \times 10^{-7} \text{ A}$	-5.98 ± 2.62 V
V <sub>DS</sub> = -40 V	$3.39 \pm 0.56 \times 10^{-7}$	$6.04 \pm 0.71 \times 10^{-7} \text{ A}$	-9.54 ± 2.56 V
V <sub>DS</sub> = -50 V	3.86 ± 0.86 × 10 <sup>-7</sup> A	$9.45 \pm 1.07 \times 10^{-7} \text{ A}$	-14 ± 2.45 V

**Table S3** Specifying the average values of  $I_{Hole}$  when  $V_{GS}$  is -50 V and  $V_{DS}$  is increased from -20 V to -50 V,  $I_{Electron}$  when  $V_{GS}$  is 20 V and  $V_{DS}$  is increased from -20 V to -50 V, and  $V_{TS}$  when  $V_{DS}$  is increased from -20 V to -50 V in 50 PPD transistor.

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- [1]. . Z. Chen, M. Li, M. Hu, S. Wang, Z. Miao, S. Xu, C. Chen, H. Dong, W. Huang and R. Chen, *Journal of Materials Chemistry C*, 2020, **8**, 2094-2101.
- [2]. T. Kitamori, D. Yoo, K. Iijima, T. Kawamoto and T. Mori, *ACS Applied Electronic Materials*, 2019, **1**, 1633-1640.
- [3]. X. Zhao, T. Liu, H. Liu, S. Wang, X. Li, Y. Zhang, X. Hou, Z. Liu, W. Shi and T. J.
  S. Dennis, ACS Appl. Mater. Interfaces, 2018, 10, 42715-42722.
- [4]. S. Guo, J. Yao, Y. Wang, L. Zhang, F. Zhai, X. Zhang, Y. Feng, W. Feng, X. Zhang and J. Jie, *Journal of Materials Chemistry C*, 2021, **9**, 5758-5764.
- [5]. M. Yoon and J. Lee, Journal of Materials Chemistry C, 2021, 9, 9592-9598.
- [6]. Y. Wang, Q. Liao, D. She, Z. Lv, Y. Gong, G. Ding, W. Ye, J. Chen, Z. Xiong and G. Wang, ACS Appl. Mater. Interfaces, 2020, 12, 15370-15379.