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## Supplementary Information Crystallinity and grain boundaries control of TIPS-pentacene in organic thin-film transistors for ultra-high sensitivity NO<sub>2</sub> detection

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**Fig. S1** Transform curves ( $V_{DS} = -40$  V) of transistors based on a-b) ITO/PMMA substrate; d-e) Si/SiO<sub>2</sub> substrate. And the gate current curves ( $V_{DS} = -40$  V) of transistors based on c) ITO/PMMA substrate; f) Si/SiO<sub>2</sub> substrate.



Fig. S2 Transfer curves for the CB, Toluene and O-xylene devices for various  $NO_2$  concentrations at room temperature ( $V_{DS} = -40$  V) on the wafers. a) Square root drain-source curves versus voltage plots. b) Log plots of drain-source current versus gate voltage.



Fig. S3 XRD patterns of TIPS-pentacene films spun from different solvents on PMMA.



**Fig. S4** Optical microscopy images of four kinds of TIPS-pentacene films on Si/SiO<sub>2</sub> substrate.



**Fig. S5** The 3D AFM images of a) CB, b) 1,2-DCB, c) Toluene, d) O-xylene processed TIPS-pentacene films grown on PMMA dielectric.



Fig. S6 AFM images for the indicated solvents of the PMMA films.



Fig. S7 FT-IR spectroscopy of PMMA treated with the four bare solvents.



**Fig. S8** (a) Real-time sensitivity ( $V_{DS} = V_{GS} = -40$  V) of the four kinds of TIPS-pentacene films deposited on the ITO/PMMA substrate responding to dynamic switching between NO<sub>2</sub> concentrations. (b) Real-time drain current curves ( $V_{DS} = V_{GS} = -40$  V) of four devices under continuous applied bias to dry air.