

## Supporting Information

### Universal Energy Level Tailoring of Self-Organized Hole Extraction Layers in Organic Solar Cells and Organic-inorganic Hybrid Perovskite Solar Cells

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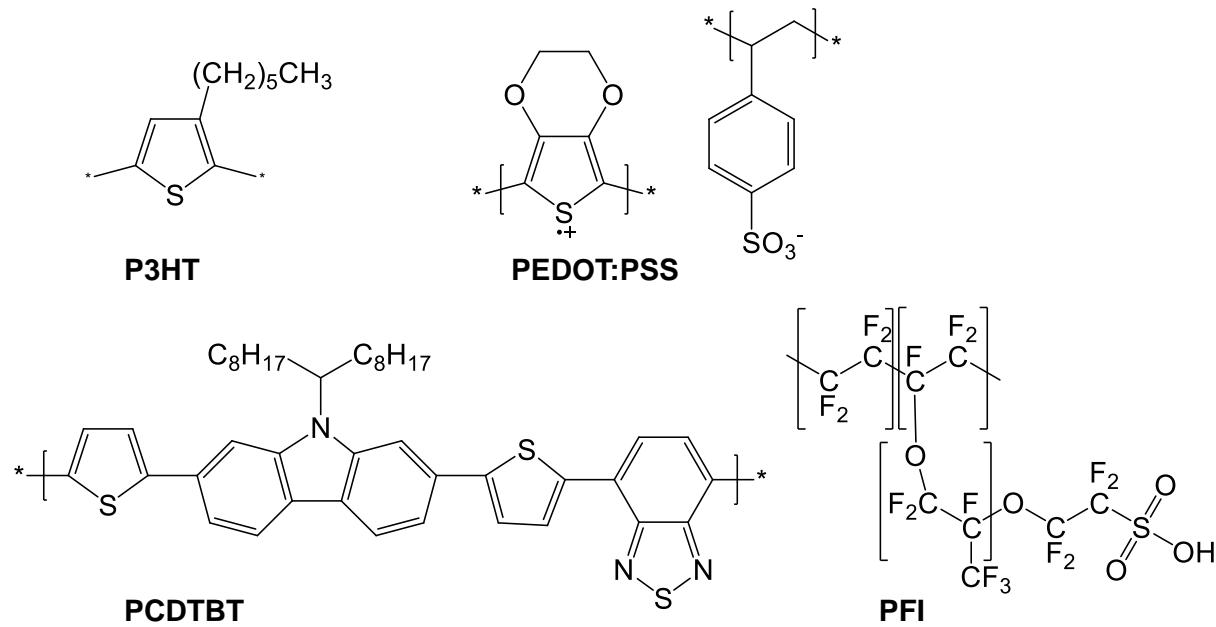


Figure S1. Molecular structures of the conjugated polymers, PEDOT:PSS, and PFI

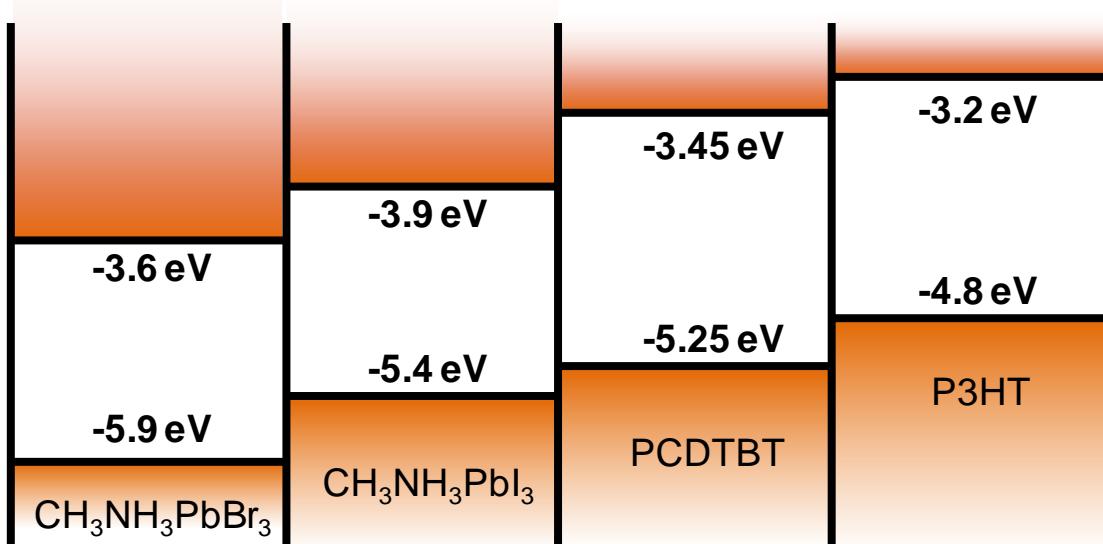


Figure S2. Schematic diagrams of energy levels for P3HT, PCDTBT,  $\text{CH}_3\text{NH}_3\text{PbI}_3$ , and  $\text{CH}_3\text{NH}_3\text{PbBr}_3$ .

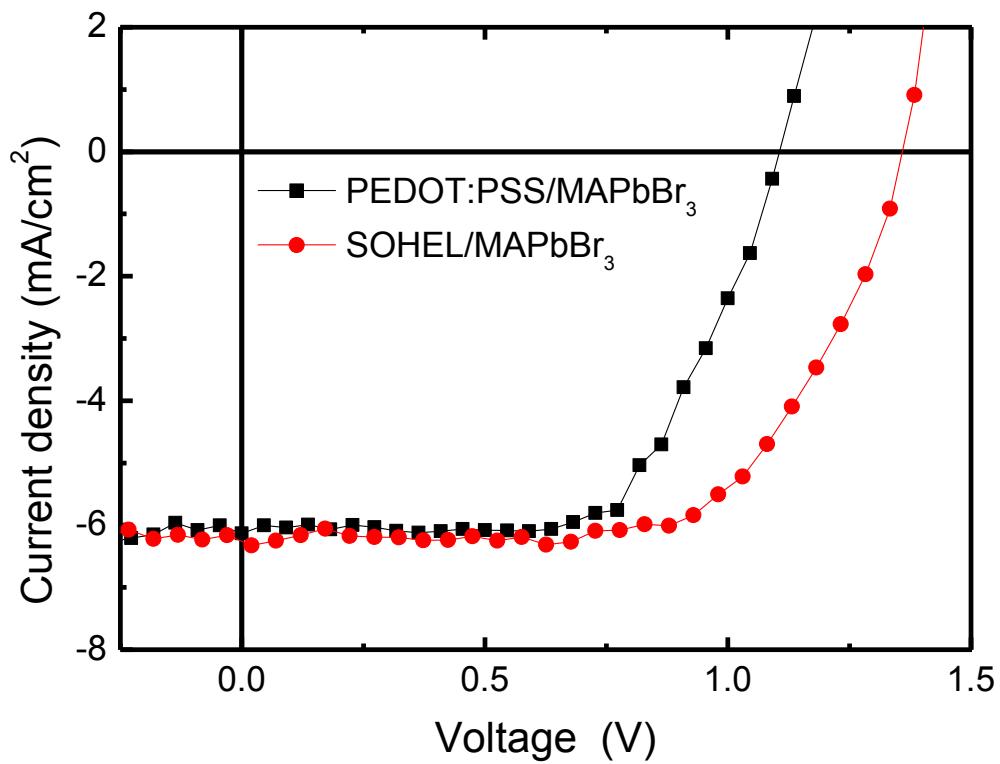


Figure S3. The J–V characteristics of MAPbBr<sub>3</sub> perovskite photovoltaics

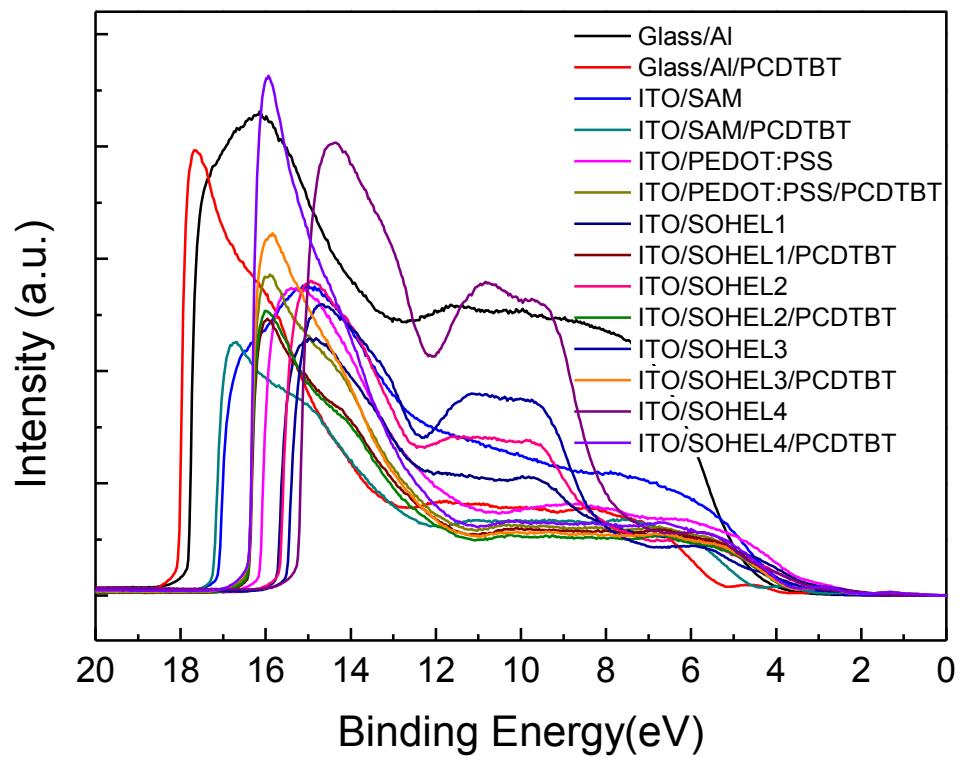


Figure S4. Ultraviolet photoelectron spectroscopy spectra of PCDTBT with various substrates.

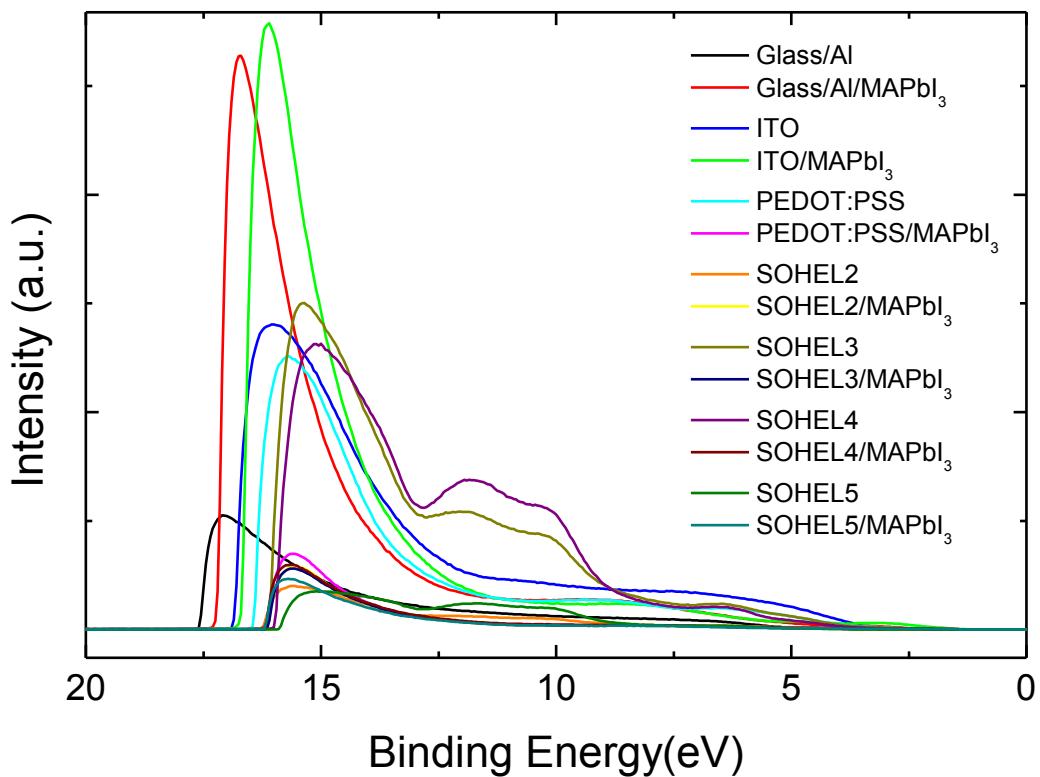


Figure S5. Ultraviolet photoelectron spectroscopy spectra of MAPbI<sub>3</sub> with various substrates.

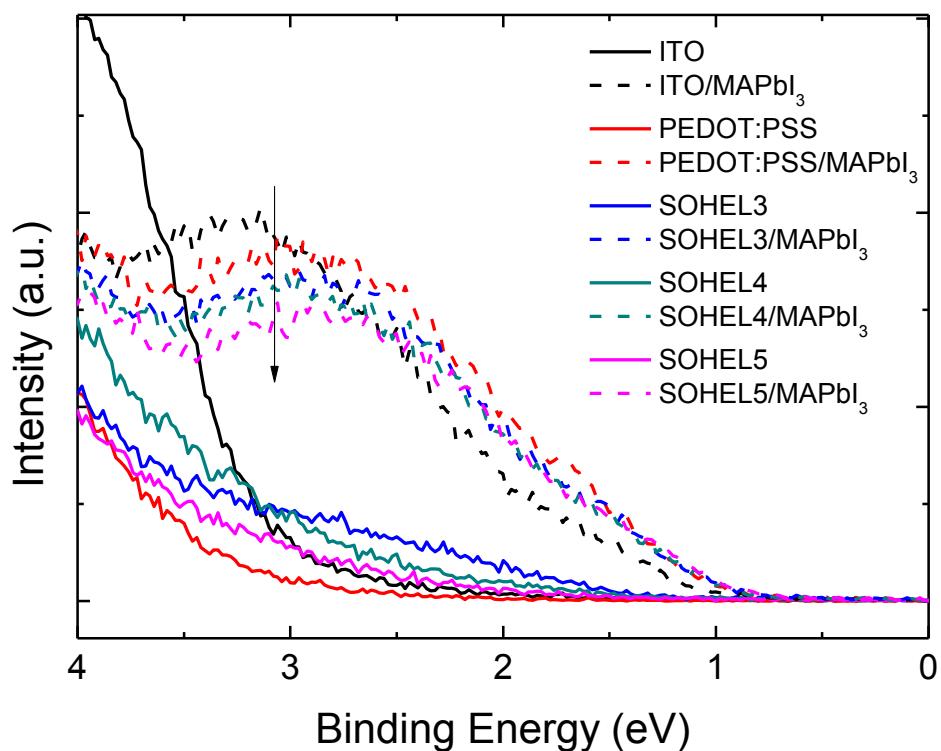


Figure S6. Ultraviolet photoelectron spectroscopy spectra of MAPbI<sub>3</sub> perovskite thin films depending on the various substrates.

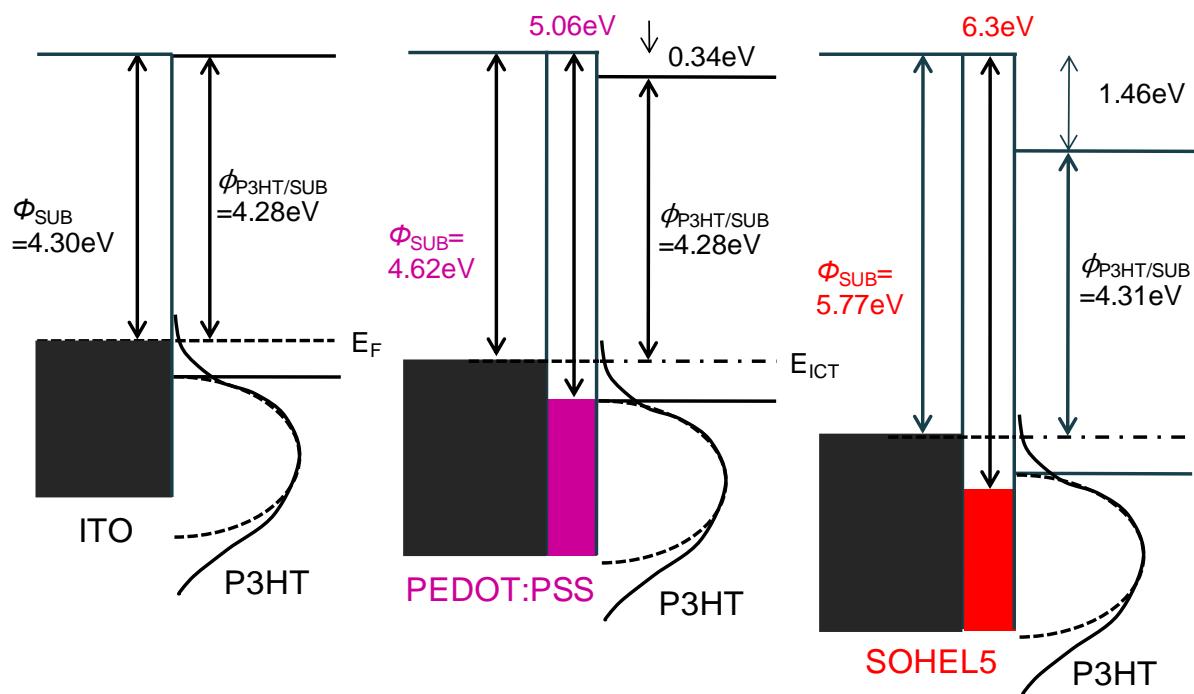


Figure S7. Schematic diagrams of energy level at the SUB/P3HT interface. (a) ITO/P3HT, (b) PEDOT:PSS/P3HT, and (c) SOHEL5/P3HT.

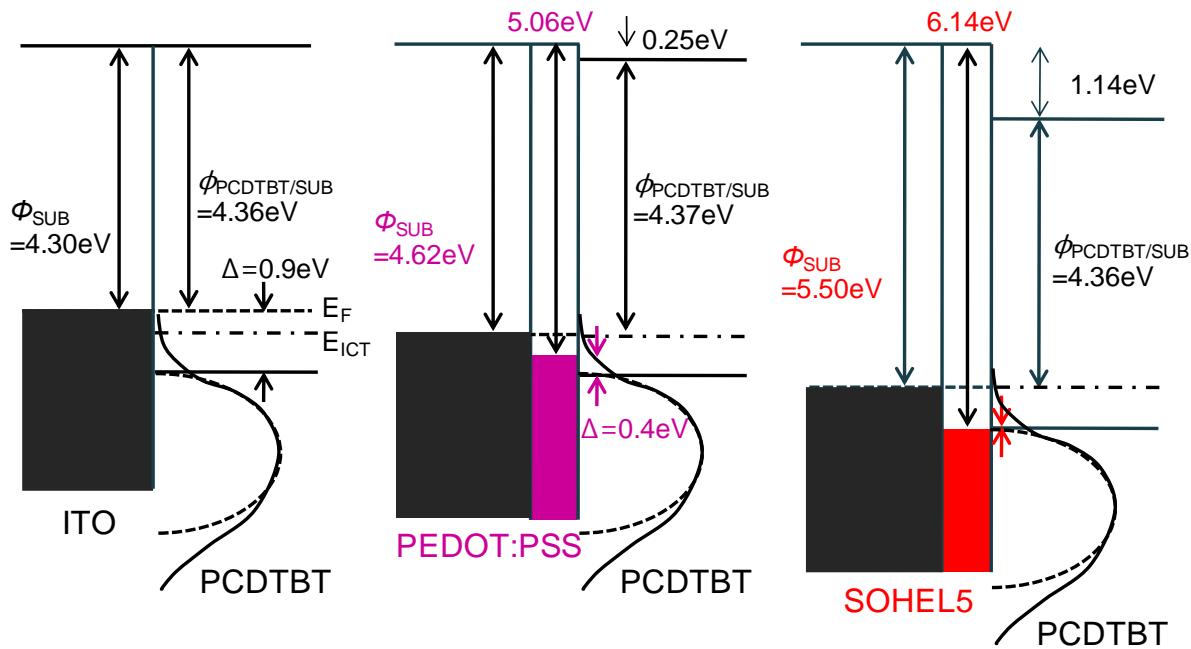


Figure S8. Schematic diagrams of energy level at the SUB/PCDTBT interface. (a) ITO/PCDTBT, (b) PEDOT:PSS/PCDTBT, and (c) SOHEL5/PCDTBT. The Fermi level ( $E_F$ ) pinning to the integer charge transfer ( $E_{\text{ICT}}$ ) level of the perovskite occurs when  $\text{WF}_{\text{SUB}}$  is larger than  $E_{\text{ICT}}$  value.  $\Delta$  is potential loss between HEL and PCDTBT.

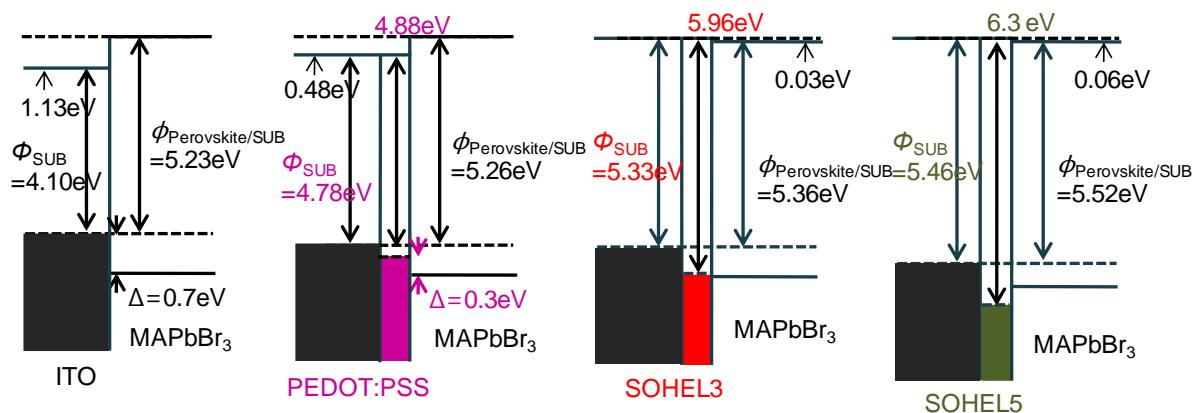


Figure S9. Schematic diagrams of energy level at the SUB/MAPbBr<sub>3</sub> interface. (a) ITO/MAPbBr<sub>3</sub>, (b) PEDOT:PSS/MAPbBr<sub>3</sub>, (c) SOHEL5/MAPbBr<sub>3</sub>, and (d) SOHEL5/MAPbBr<sub>3</sub>.

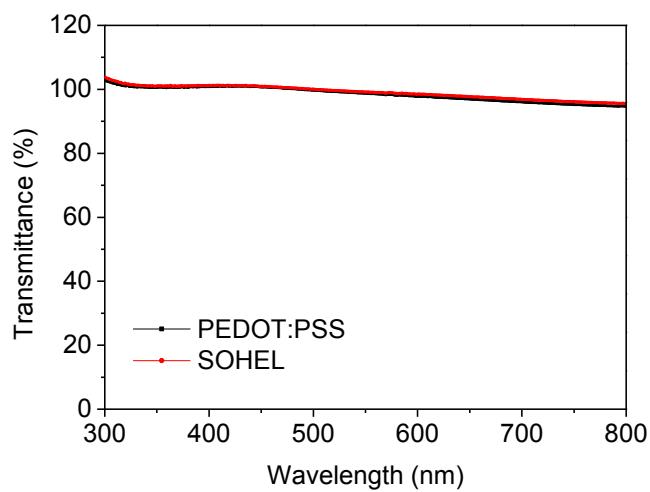


Figure S10. Transmittance of PEDOT:PSS and SOHEL4 films

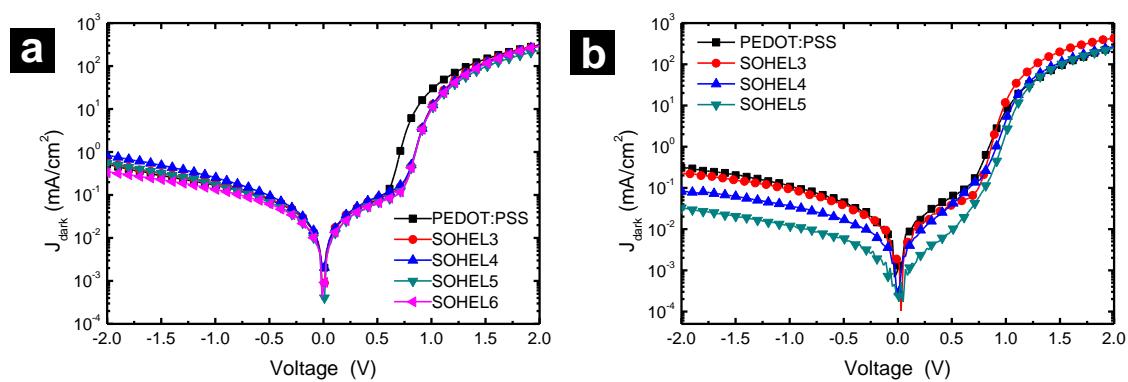


Figure S11.  $\log(J_{\text{dark}})$  vs  $V$  of (a) PCDTBT:PC<sub>70</sub>BM organic solar cells and (b) MAPbI<sub>3</sub> perovskite solar cells.

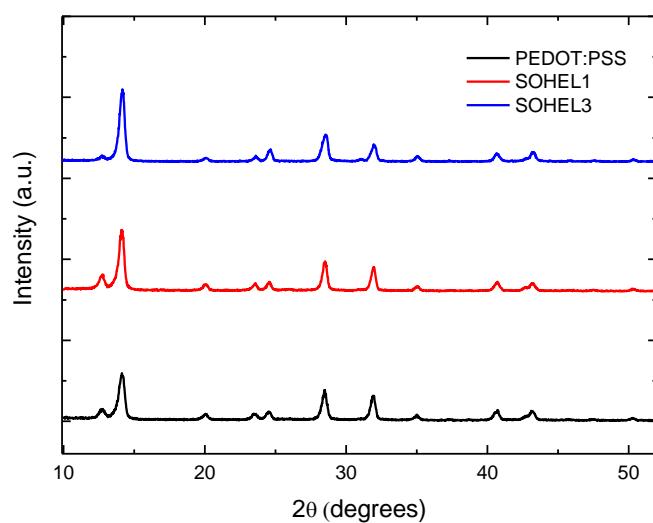


Figure S12. XRD patterns of MAPbI<sub>3</sub> films deposited on PEDOT:PSS, SOHEL1, or SOHEL3.

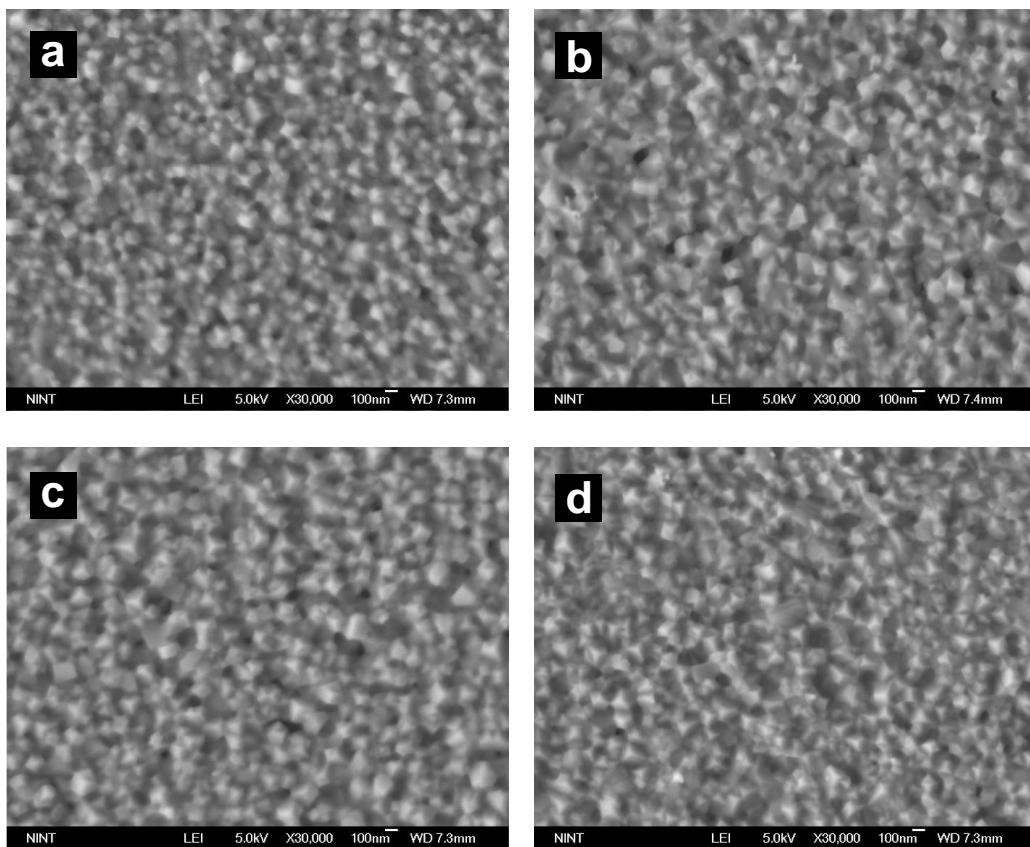


Figure S13. SEM images of MAPbI<sub>3</sub> films in high magnification (x30k). MAPbI<sub>3</sub> films deposited on (a) PEDOT:PSS; (b) SOHEL1; (c) SOHEL3; and (d) SOHEL5.

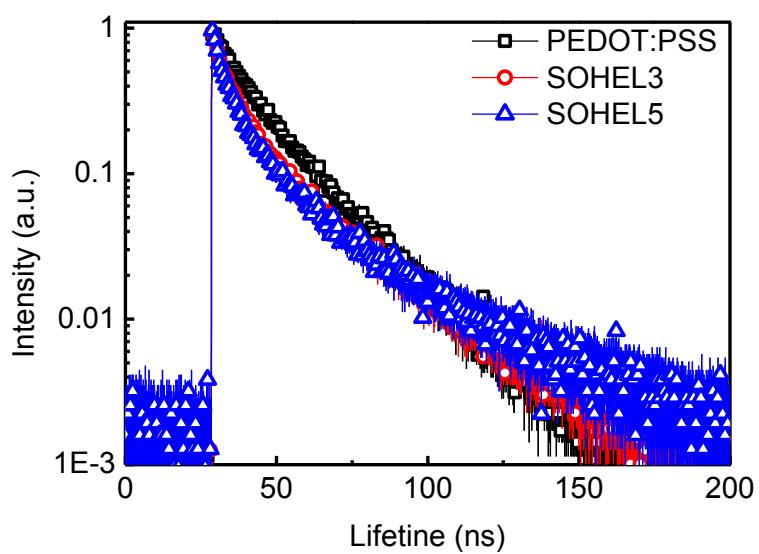


Figure S14. Transient photoluminescence (Tr-PL) spectroscopy of PEDOT:PSS, SOHEL3, and SOHEL5.

Table S1.  $V_{OC}$ ,  $J_{SC}$ ,  $FF$ , and  $PCE$  of the PCDTBT:PC<sub>70</sub>BM photovoltaic cells with PEDOT:PSS and SOHELs.

	$V_{OC}$ (V)	$J_{SC}$ (mA/cm <sup>2</sup> )	$FF$ (%)	$PCE$ (%)
PEDOT:PSS	0.738	10.2	56.4	4.2
SOHEL1	0.810	11.5	53.3	5.0
SOHEL2	0.880	11.9	59.7	6.3
SOHEL3	0.897	11.7	60.3	6.3
SOHEL4	0.904	11.6	58.3	6.1
SOHEL5	0.901	11.6	57.4	6.0
SOHEL6	0.901	11.5	57.8	6.0

Table S2. Average and standard deviation of  $V_{OC}$ ,  $J_{SC}$ ,  $FF$ , and  $PCE$  of MAPbI<sub>3</sub> perovskite solar cells with PEDOT:PSS and SOHELs

		HELs			
		PEDOT:PSS	SOHEL3	SOHEL4	SOHEL5
$V_{OC}$ (V)	Average	0.903	0.988	1.022	1.005
	Standard Deviation	±0.060	±0.033	±0.023	±0.020
$J_{SC}$ (mA/cm <sup>2</sup> )	Average	13.15	14.65	15.29	14.24
	Standard Deviation	±2.64	±2.19	±1.26	±1.61
$FF$ (%)	Average	62.8	70	70.9	73.4
	Standard Deviation	±5.34	±9.68	±3.41	±1.96
$PCE$ (%)	Maximum	8.1	10.3	11.7	11.4
	Average	7.03	8.7	10.90	10.55
	Standard Deviation	±1.88	±2.54	±0.77	±1.06

Table S3. The peak position and FWHM for MAPbI<sub>3</sub> perovskite films deposited on PEDOT:PSS, SOHEL1, or SOHEL3.

Crystal face	Substrate			
	PEDOT:PSS	SOHEL1	SOHEL3	
[110] of MAPbI <sub>3</sub>	Peak position(°)	14.1	14.1	14.1
	FWHM	0.43	0.45	0.44
[001] of PbI <sub>2</sub>	Peak position(°)	12.7	12.7	12.7
	FWHM	0.51	0.45	0.48