Supporting Information

Importance of interface engineering between the hole transport layer and the indium-tin-oxide electrode for high-efficient polymer solar cells

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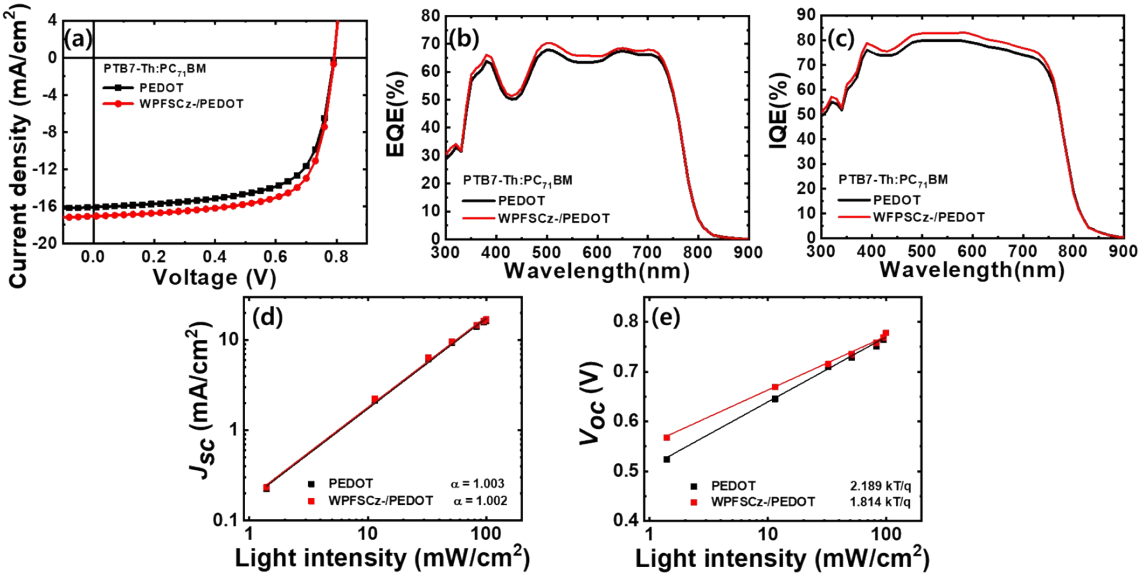


Figure S1. Photoelectric measurements of PTB7-Th:$PC_{71}BM$ devices made with and without a WPFSCz- layer. (a) *J-V* characteristics. (b) External quantum efficiency (EQE) curves. (c) Internal quantum efficiency (IQE) curves. (d) J_{sc} versus light intensity. (e) V_{oc} versus light intensity.

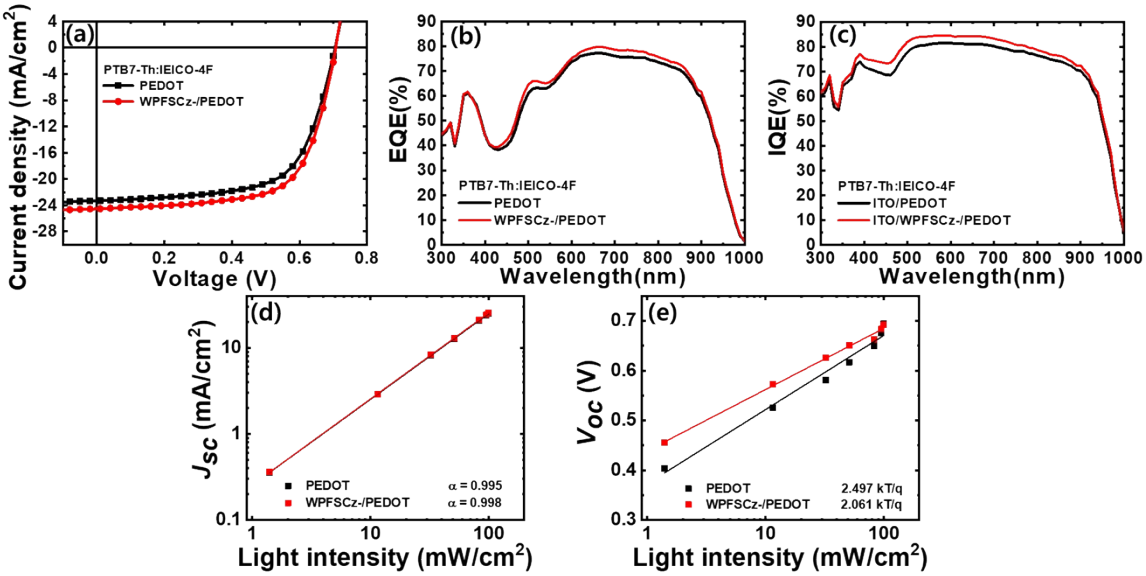


Figure S2. (a) The *J-V* characteristics, (b) EQE, and (c) IQE curves of PTB7-Th:IEICO-4F devices with and without a WPFSCz- layer. (d) J_{sc} and (e) V_{oc} versus light intensity for PTB7-Th:$PC_{71}BM$ with and without a WPFSCz- layer.

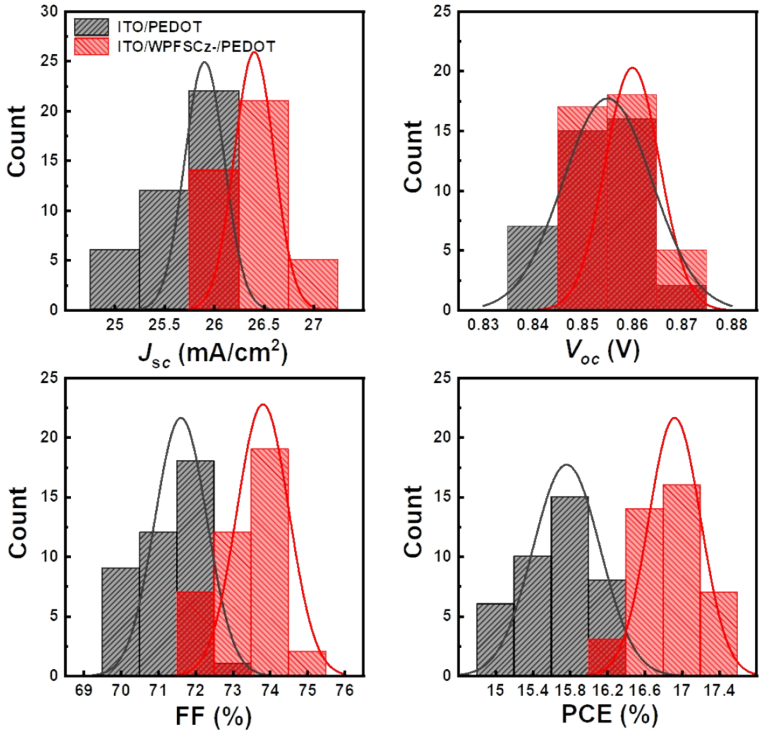


Figure S3. Histograms of PBDB-T-2F:Y6 solar cell performance parameters.

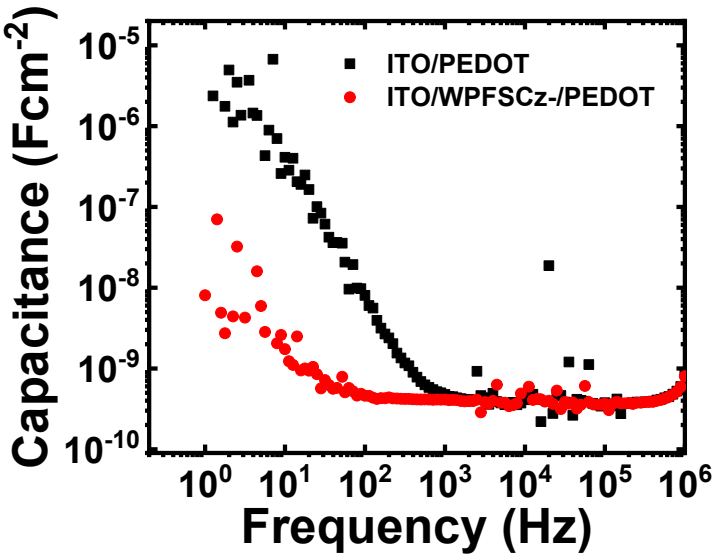


Figure S4. Frequency-dependent capacitance measurement of PBDB-T-2F:Y6 with and without a WPFSCz- layer.

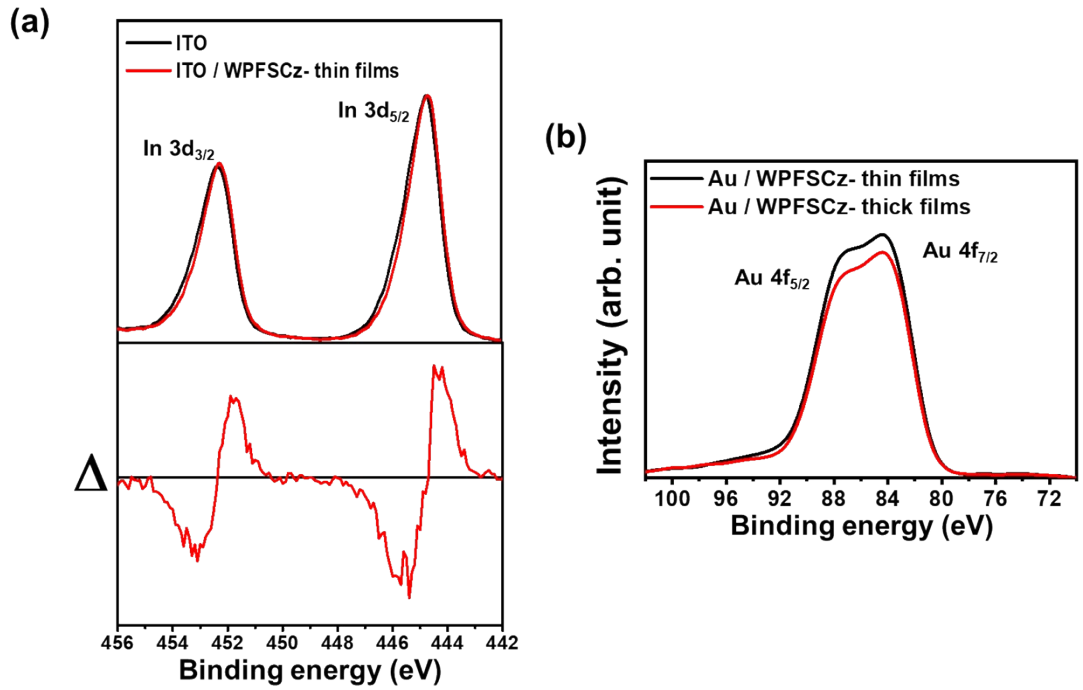


Figure S5. (a) In 3d XPS spectra and (b) Au 4f XPS spectra of WPFSCz- films on ITO substrates and Au substrates, respectively

Table S1. Relative area ratio according to O 1s XPS spectra.

	O - metal	O-H	O-C
ITO	1.000	1.000	1.000
WPFSCz- thin films	1.189	0.881	0.622
WPFSCz- thick films	1.249	0.664	0.699

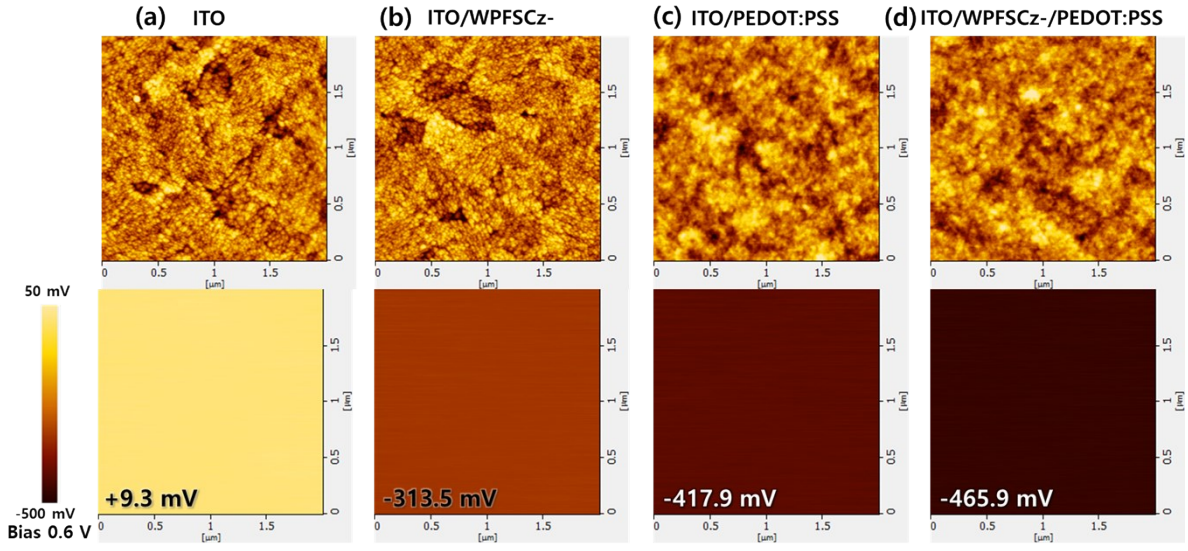


Figure S6. Surface potential images of (a) ITO, (b) ITO with WPFSCz-, (c) PEDOT:PSS on ITO, and (d) PEDOT:PSS on ITO with WPFSCz-.

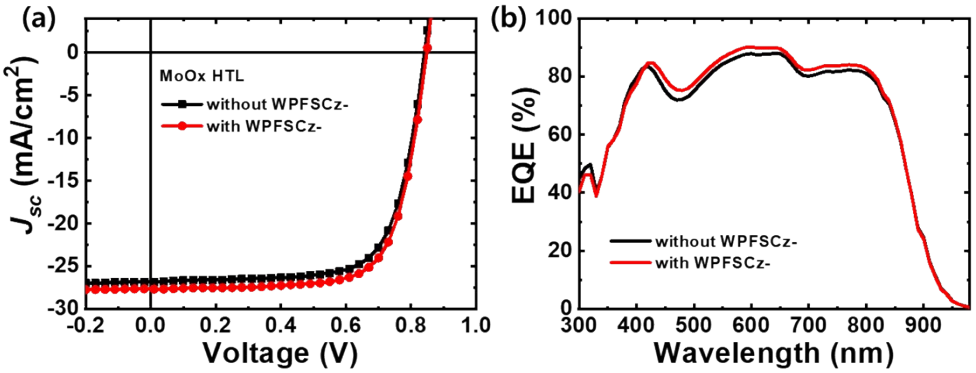


Figure S7. (a) The *J-V* characteristics and (b) EQE curves of PBDB-T-2F:Y6 devices with and without a WPFSCz- layer based on MoOx as the HTL.

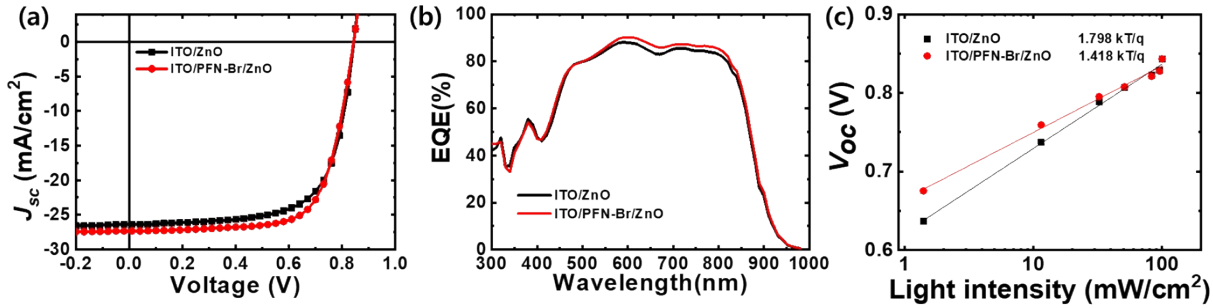


Figure S8. (a) The *J-V* characteristics and (b) EQE curves of PBDB-T-2F:Y6 inverted devices with and without PFN-Br layer. (c) V_{oc} versus light intensity for PBDB-T-2F:Y6 inverted devices with and without PFN-Br layer.

Table S2. Summary of photovoltaic parameters of PBDB-T-2F:Y6 solar cells with MoOx as the HTL.

	V_{OC} (V)	J_{SC} (mA/cm^2)	*FF*	η (%)
ITO/MoO_x	0.841	26.839	71.23	16.08
ITO/WPFSCz-/MoO_x	0.848	27.661	73.25	17.18

Table S3. Summary of photovoltaic parameters of PBDB-T-2F:Y6 inverted devices with ZnO HTL. In this inverted device, an n-type polyelectrolyte, PFN-Br, was utilized.

	V_{OC} (V)	J_{SC} (mA/cm^2)	*FF*	η (%)
ITO/ZnO	0.844	26.372	68.33	15.21
ITO/PFN-Br/ZnO	0.843	27.341	70.49	16.22

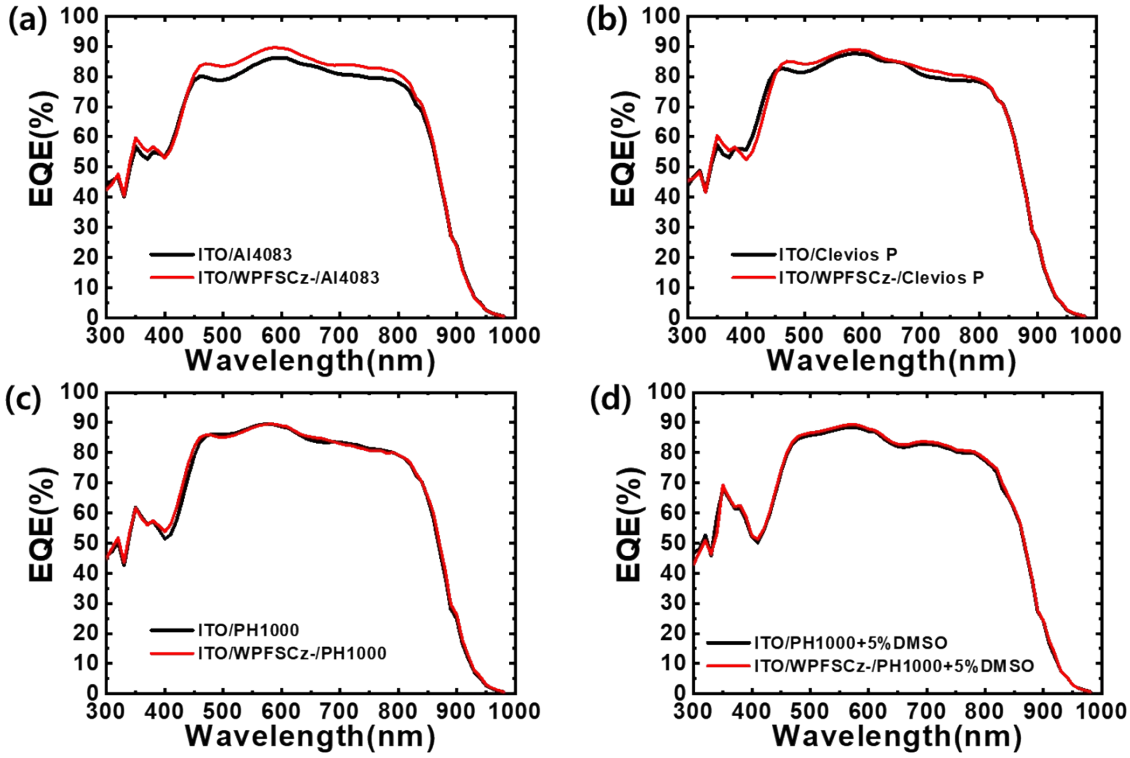


Figure S9. EQE spectra of PBDB-T-2F:Y6 solar cells made with and without a WPFSCz- layer and based on different types of PEDOT:PSS: (a) AI4083, (b) Clevios P, (c) PH1000, and (d) PH1000 + 5% DMSO.

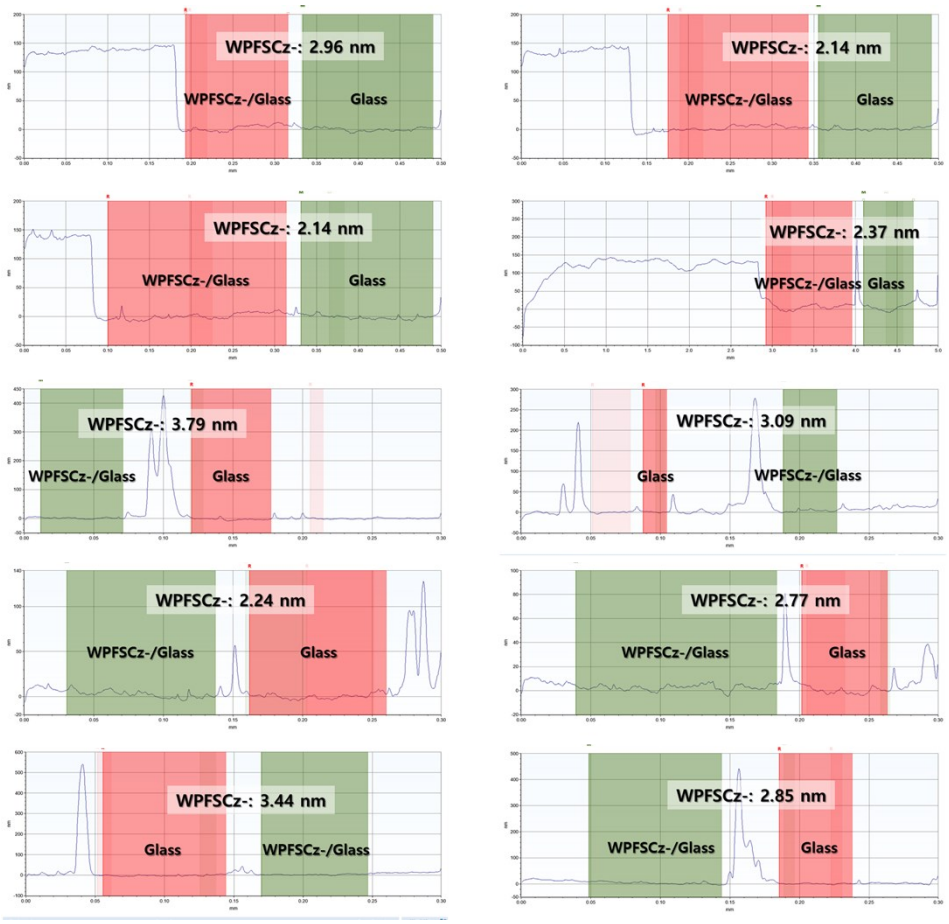


Figure S10. Film thickness measurements of WPFSCz- using a Bruker Dektak XT surface profiler. The average thickness of WPFSCz- is 2.78 nm.

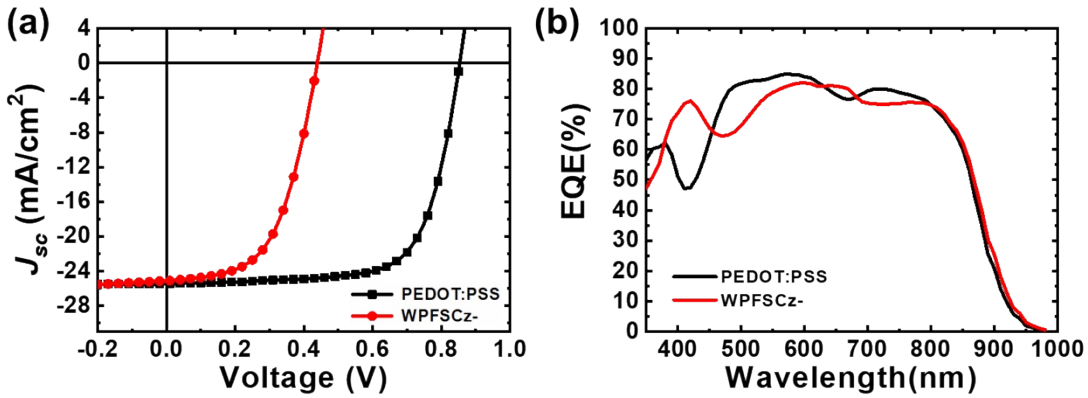


Figure S11. (a) The *J-V* characteristics and (b) EQE curves of PBDB-T-2F:Y6 devices PEDOT:PSS and WPFSCz- only as HTL.

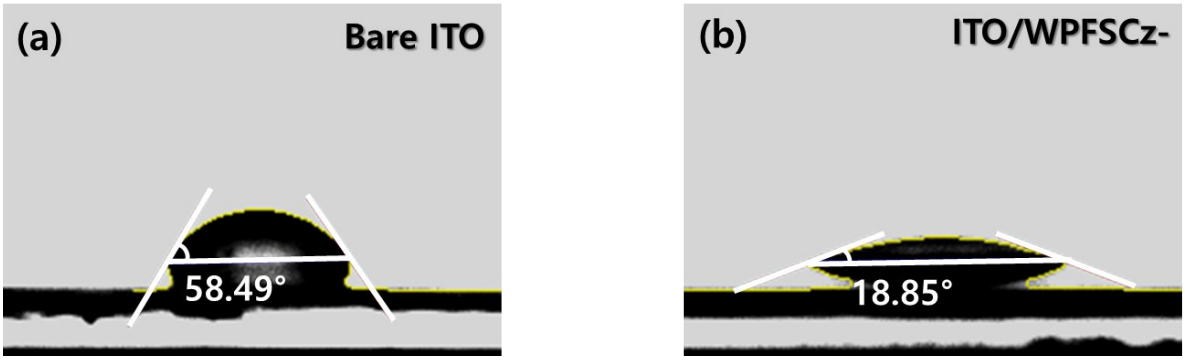


Figure S12. The water contact angle of (a) bare ITO and (b) ITO with a WPFSCz- layer.

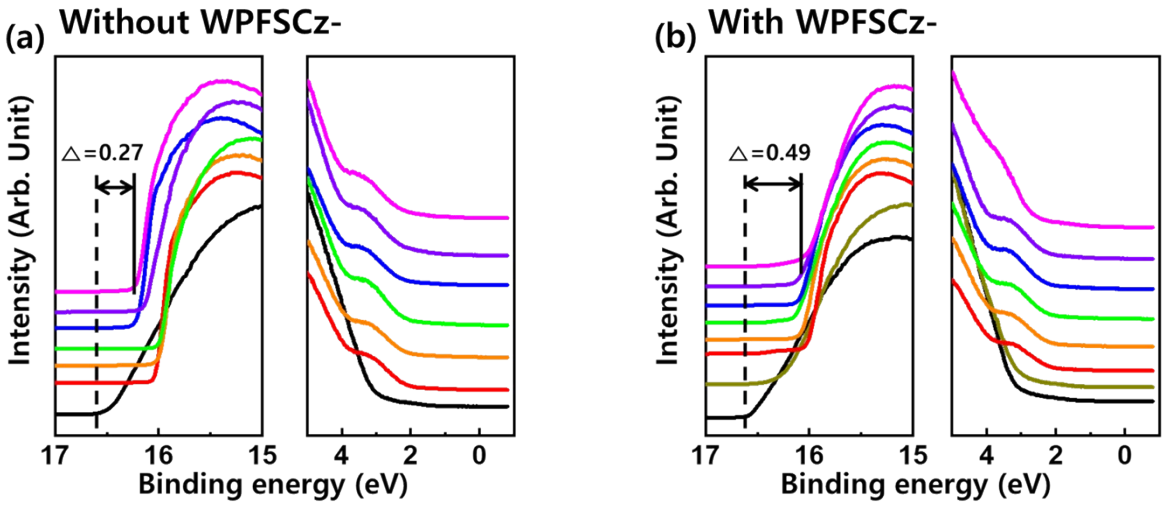
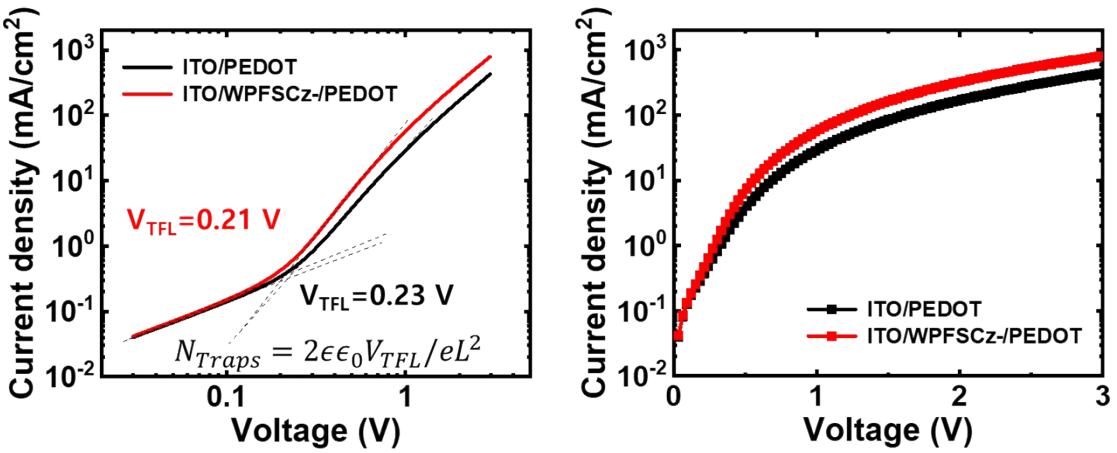


Figure S13. Secondary electron cut-off and HOMO onset UPS spectra of increasing thickness of PEDOT:PSS (a) without WPFSCz- and (b) with WPFSCz-.



	Hole mobility (cm^2/Vs)	N_{traps} (cm^{-3})
ITO/PEDOT	1.65 ×10^{-5}	8.65 ×10^{21}
ITO/WPFSCz-/PEDOT	1.94 ×10^{-5}	7.89 ×10^{21}

Figure S14. Hole mobility and trap density analysis.