Electronic Supplementary Material (ESI) for RSC Applied Interfaces.

Supporting information

to:

Solution-treatment controls charge-transfer states and energy-level alignment at hybrid CuSCN-organic interfaces

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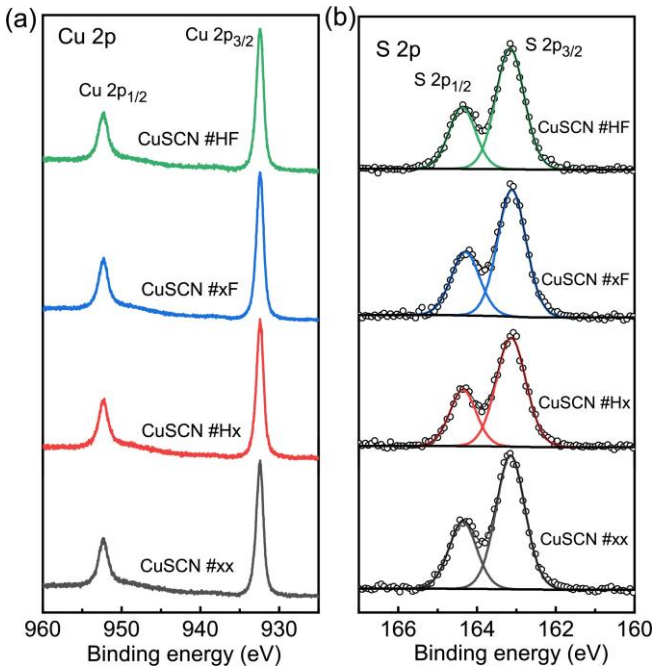

Figure S1. XPS measured Cu 2p (a) and S 2p (b) spectra of CuSCN thin films on ITO.

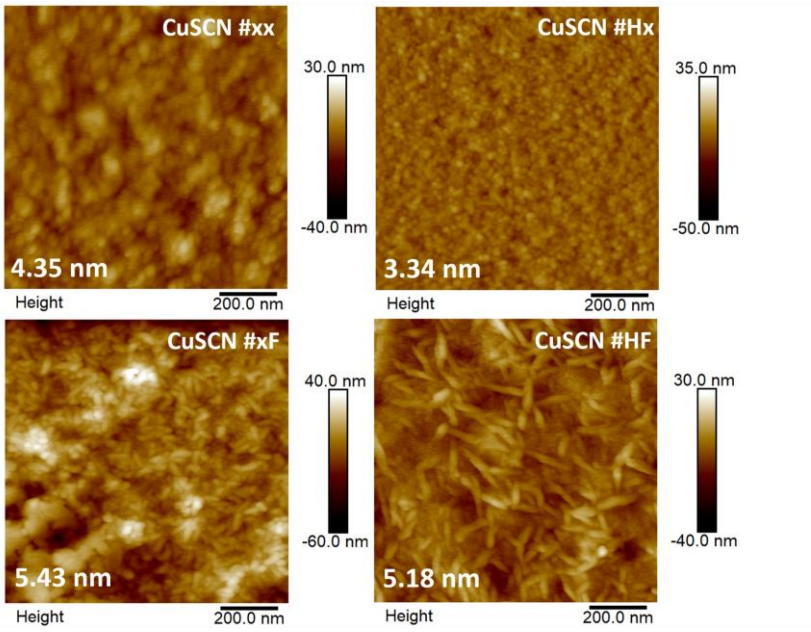

Figure S2. AFM topography of spin-coated CuSCN thin films on PEDOT:PSS/ITO substrates with their respective surface roughness.

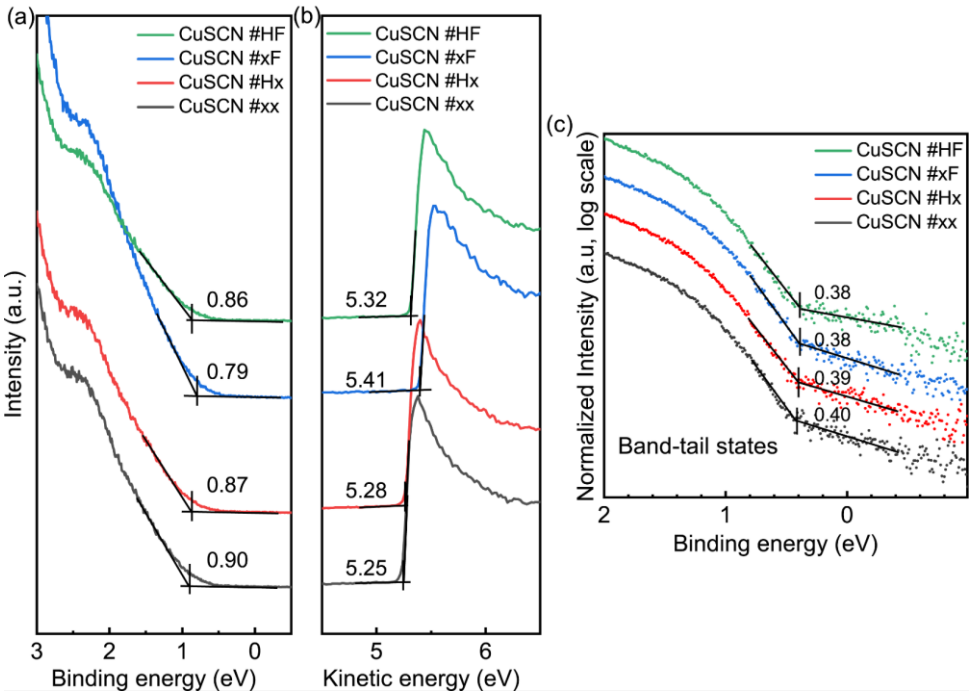

Figure S3. UPS-measured (a) valence band region, (b) secondary electron, and (c) valence band regions plotted on a semi-log scale of CuSCN thin films on PEDOT:PSS/ITO substrates.

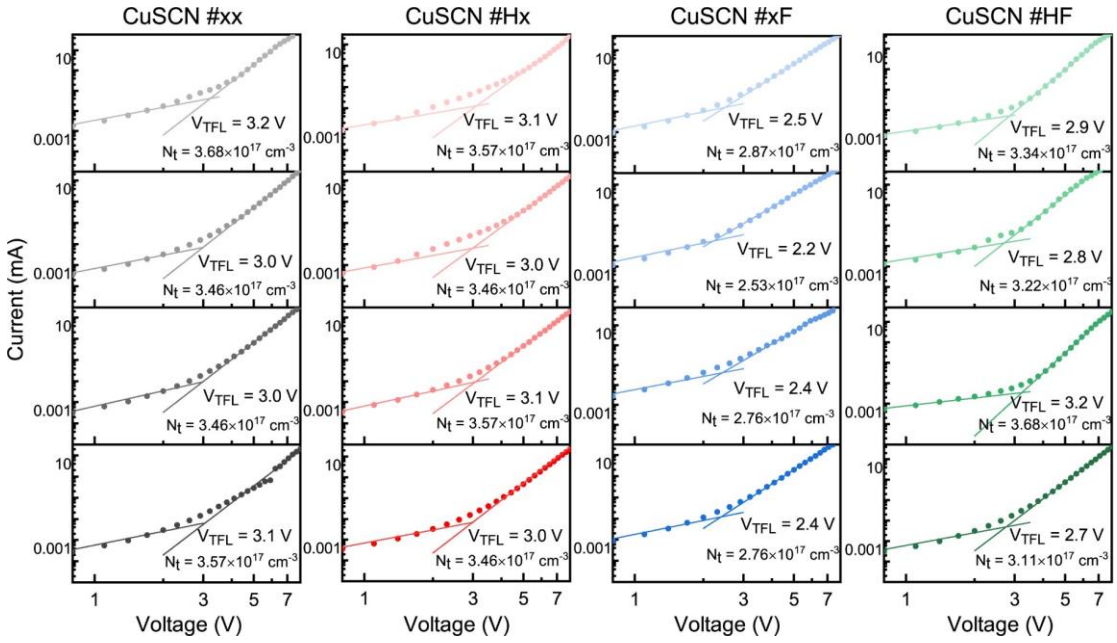

Figure S4. Characteristics of trap-filled-limit voltage and corresponding trap density by I–V curves of a single carrier device based on different CuSCN films.

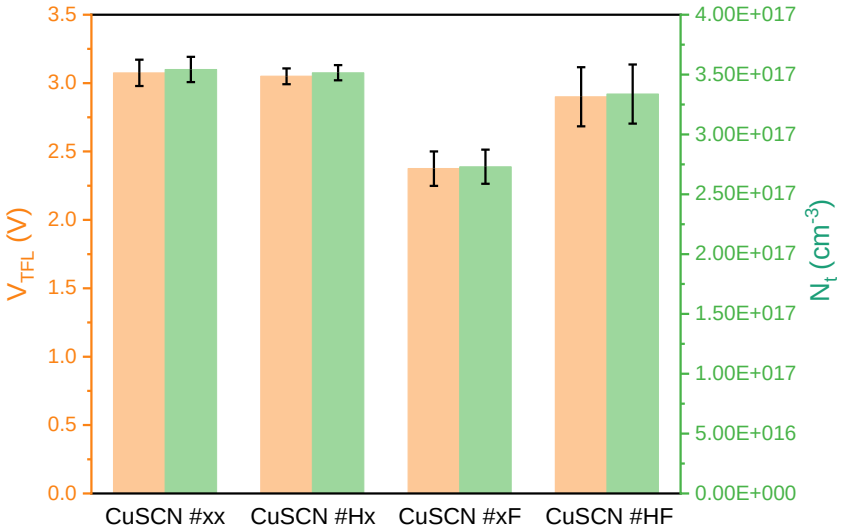

Figure S5. Trap-filled-limit voltage and corresponding trap density with standard deviation of different CuSCN-film-based hole-only devices.

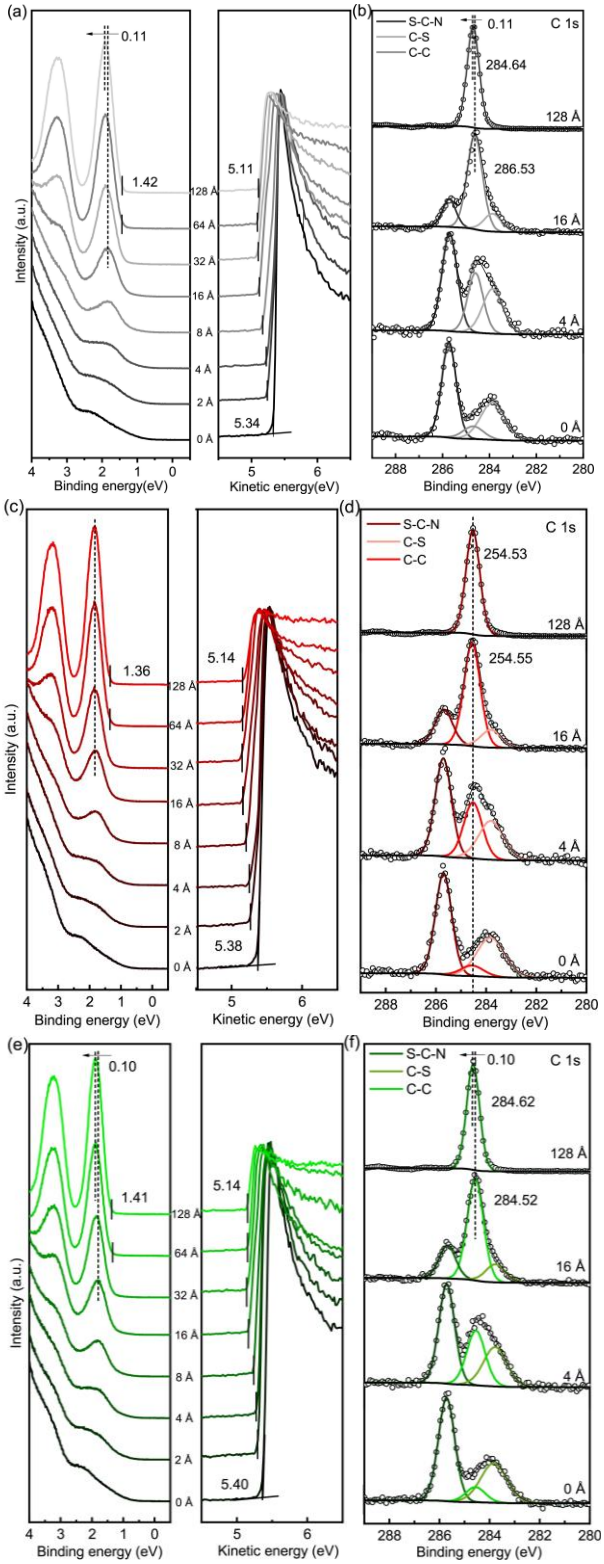

Figure S6. Thickness-dependent UPS spectra of (a) C_{60}/CuSCN #xx, (c) C_{60}/CuSCN #Hx, and (e) C_{60}/CuSCN #HF and XPS measured C 1s core level spectra of (b) C_{60}/CuSCN #xx, (d) C_{60}/CuSCN #Hx, and (f) C_{60}/CuSCN #HF, respectively. For C_{60}/CuSCN #xx the HOMO level onset of 1.6 nm C_{60} is at 1.31 eV BE and shifts gradually toward higher BE by 0.11 eV from 1.6 nm to 12.8 nm thickness, this shift is parallel to the shift of the C 1s core-

level shift, indicating energy-level bending in the C_{60} layer. For C_{60} on CuSCN #HF the HOMO and C 1s level shift both by 0.10 eV.

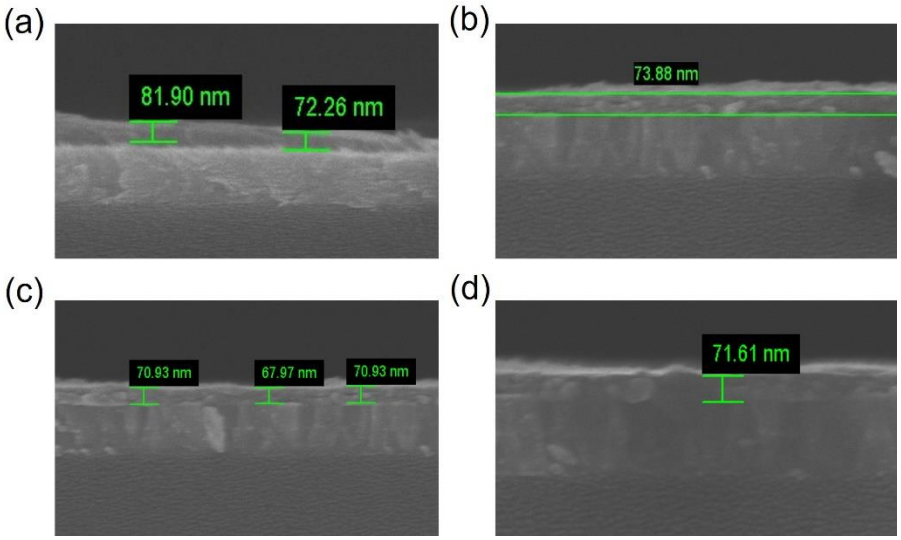

Figure S7. Cross-section scanning electron micrographs of CuSCN #xx (a), CuSCN #Hx (b), CuSCN #xF (c), CuSCN #HF (d).

Table S1. Properties of CuSCN films.

Substrate	CuSCN	RMS (nm)	VBM (eV)	Trap state density (cm^{-3})	CT state intensity (a.u.)
ITO (RMS=3.78)	#xx	6.81	0.83		
	#Hx	4.21	0.89		
	#xF	5.84	0.77		
	#HF	5.37	0.84		
PEDOT:PSS/ITO (RMS=1.82)	#xx	4.35	0.90	$3.54*10^{17}$	1.9
	#Hx	3.34	0.87	$3.51*10^{17}$	0.9
	#xF	5.43	0.79	$2.73*10^{17}$	10.6
	#HF	5.18	0.86	$3.34*10^{17}$	2.1