## Supporting Information

## Efficient and Stable Carbon-based Perovskite Solar Cells Enabled by Inorganic Interface of CuSCN and Carbon Nanotubes

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	$J_{SC}$ (mA/cm <sup>2</sup> )	$V_{OC}(\mathbf{V})$	FF(%)	PCE(%)
with CuSCN	$21.72 \pm 0.53$	$1.00 \pm 0.01$	68.96 ± 1.73	$14.95\pm0.30$
w/o CuSCN	$22.16 \pm 0.82$	$0.84\pm0.01$	$62.33 \pm 6.68$	$11.70 \pm 1.14$

**Table S1** *J*–*V* parameters of the C-PSCs with and without CuSCN HTL under the illumination of AM 1.5G, 100 mW cm<sup>-2</sup>.

**Table S2** *J*–*V* parameters of the C-PSCs based on CuSCN HTLs deposited from different CuSCN/DES concentration under the illumination of AM 1.5G, 100 mW cm<sup>-2</sup>. The champion values are shown in the brackets.

Concentration of CuSCN (mg mL <sup>-1</sup> )	$J_{SC}$ (mA cm <sup>-2</sup> )	$V_{OC}$ (V)	FF(%)	PCE(%)
10	19.75 (19.9)	1.01 (1.02)	69.81 (72.15)	13.90 (14.60)
20	20.28 (20.2)	1.02 (1.02)	73.00 (73.60)	15.07 (15.16)
30	19.95 (20.0)	1.01 (1.02)	70.91 (72.40)	14.25 (14.83)
40	19.95 (19.1)	1.00 (1.01)	62.83 (70.69)	12.43 (13.63)
50	19.42 (19.5)	0.96 (0.96)	62.75 (65.60)	11.68 (12.31)



**Figure S1.** Contact angles test results of water on the surfaces of (a) perovskite, (b) CuSCN, (c) CNT, and (d) Au.



**Figure S2.** *J*–*V* curves of CuSCN inserted C-PSCs fabricated by dipropyl sulfide and diethyl sulfide solvent.



**Figure S3.** The surface topography images of CuSCN deposited from precursor solution with concentration of (a) 10 mg mL<sup>-1</sup>, (b) 20 mg mL<sup>-1</sup>, (c) 30 mg mL<sup>-1</sup>, (d) 40 mg mL<sup>-1</sup> (e) 50 mg mL<sup>-1</sup> deposited on perovskite.



**Figure S4.** AFM topography of perovskite on FTO/compact  $TiO_2$ /mesoporous  $TiO_2$  substrate with a scan area of (a) 5  $\mu$ m<sup>2</sup> and (b) 1  $\mu$ m<sup>2</sup>.



**Figure S5.** AFM topography images of CuSCN deposited on perovskite (a) before and (b) after annealing at 50 °C for 10 min. The scan area is 1  $\mu$ m<sup>2</sup>.



Figure S6. Full range XPS spectra of CuSCN film on FTO/TiO<sub>2</sub>/Perovskite substrate.



**Figure S7.** UV-visible spectroscopy and PL spectra of perovskite with (black line) and without CuSCN (red line)

Table S3 The extracted XPS characteristics of CuSCN film on FTO/TiO<sub>2</sub>/Perovskite substrate,

and the original data shown in Figure S6.

Name	Start BE (eV)	Peak BE (eV)	End BE (eV)	FWHM (eV)	Atomic (%)
Pb4f	152.6	138.42	132.8	0.91	0.48
C1s	297.6	284.66	278.8	1.16	48.63
N1s	409.6	398.58	391.8	0.85	12.94
<b>O1s</b>	544.6	532.48	524.8	2.15	1.5
I3d	639.6	619.35	609.8	1.21	1.65
S2p	174.6	163.35	156.8	1	18.27
Cu2p	964.6	932.6	924.8	1.23	16.55



**Figure S8.** (a) *J-V* curves measured in the forward and reverse scanning directions, (b) corresponding stabilized power output measured at 0.78 V under AM 1.5G illumination of 100  $mW/cm^2$ .



Figure S9. (a)  $V_{OC}$ , (b)  $J_{SC}$  and (c) FF histogram of one batch of devices (15 in total).



Figure S10. (a) *J*–*V* curve and (b) IPCE result measured at Xiamen University.