Supporting Information

Unveiling and overcoming the interfacial degradation between CuSCN and metal electrodes in perovskite solar cells

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Figure S1. Energy band diagram of CuSCN with (a) Au, (b) C, (c) Ag and (d) Al electrodes before and after contact.



Figure S2. UPS measurement of CuSCN films.



Figure S3. Atomic Force Microscope (AFM) images of metal stages with the

thickness of around 10 nm.



Figure S4. Angle resolved XPS test for CuSCN/Au sample under 0.8 V bias for one

week.



Figure S5. Polar intensity profiles along the q range of 0.2-3.0 Å⁻¹ for Au, CuSCN and CuSCN/Au thin films.



Figure S6 XRD patterns of 10 nm Ag/Al and with CuSCN films with applied 0.8 V BV.



Figure S7. A cross-section TEM image of a whole perovskite solar cell.



Figure S8. The *J-V* curve of the device with Cu electrode.



Figure S9. The *J-V* curves of CuSCN-based devices with Au electrode under applied 0.8V BV.



Figure S10. XPS spectra of Cu 2p for CuSCN, CuSCN in carbon with and without applied BV.



Figure S11. V_{OC} as a function of ln (J_{SC}) with ideality factors of 1.42 and 1.64 for devices with carbon and Au electrodes, respectively.



Figure S12. Energy band diagram of CuSCN/Au interface with AuCu alloy.



Figure S13. The stabilized state current density and efficiency for CuSCN-based device with carbon electrode.

Electrodes	$V_{OC}(\mathbf{V})$	J_{SC} (mA/cm ²)	FF (%)	PCE (%)
Au	1.1036	24.909	80.26	22.06
Ag	0.9625	22.401	51.80	11.17
Al	0.9074	19.507	32.92	5.83

Table S1. Photovoltaic parameters of devices in Figure 1a.

 Table S2. The summary of reported perovskite solar cells combining CuSCN with carbon electrodes.

Configurations of perovskite solar cells	РСЕ (%)	V _{OC} (V)	J_{SC} (mA cm ⁻²)	FF (%)	Stability (conditions, lifetime)	Reference
FTO/TiO2/CsPbIBr2/CuSCN/C	7.3	1.13	10.43	62	storage in ambient air, $T_{94} = 1100 \text{ h}$	1
FTO/SnO ₂ /CsFA _{0.83} MA _{0.17} PbI _{2.53} Br _{0.47} /CuSCN/ C	14.7	1.01	20.6	70.1	storage in ambient air, $T_{93} > 1920 \text{ h}$	2
FTO/SnO2/CsFAMAPbIBr/CuSCN/C	15.81	1.08	22.09	66.4	storage in ambient air, $T_{93} = 2000 \text{ h};$ thermal stress, $T_{83} = 300 \text{ h}$	3
FTO/SnO ₂ /CsFA _{0.83} MA _{0.17} PbI _{2.53} Br _{0.47} /CuSCN/ C	13.6	1.02	18.49	72.1	storage in dry air, $T_{100} = 2160$ h; humidity stress, $T_{98} = 240$ h	4
FTO/c-TiO2/mp-TiO2/FAMACsPb(IxBr1-x)3/CuSCN/C	15.5	1.02	22.0	69.3	humidity stress, $T_{80} = 1000 \text{ h}$	5

FTO/c-TiO ₂ /mp-TiO ₂ / (FAPbI ₃) _{0.83} (MAPbBr ₃) _{0.17} /CuSCN/C	17.58	1.01	23.7	73.4	humidity stress, $T_{95} = 100$ h; irradiation stress, $T_{80} = 1000$ h	6
FTO/c-TiO2/mp-TiO2/ MAPbI3/CuSCN/C	12.41	0.96	18.90	68	humidity stress, $T_{68} = 4800$ h; thermal stress, $T_{92} = 175$ h	7
ITO/SnO ₂ /FA _{0.95} MA _{0.05} Pb(I _{0.95} Br _{0.05}) ₃ /CuSCN/C	20.86	1.142	24.5	74.58	thermal stress, $T_{80} = 300 \text{ h};$ MPP tracking, $T_{80} = 1000 \text{ h}$	This work

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