

## Supporting Information

# Organic Ammonium Salts-Assisted Pinhole-free CuSCN Film for Carbon-based Perovskite Solar Cells

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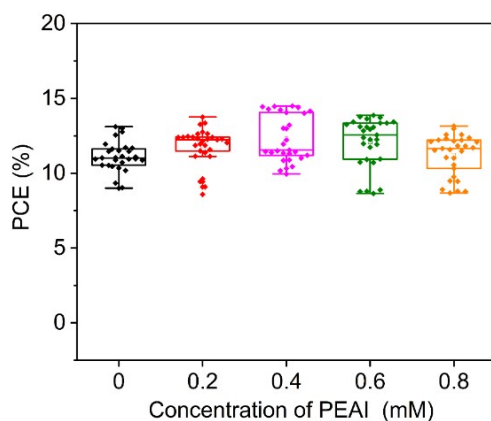


Figure S1. PCE statistical distribution of the cells fabricated using the modified CuSCN at different concentration of PEAI.

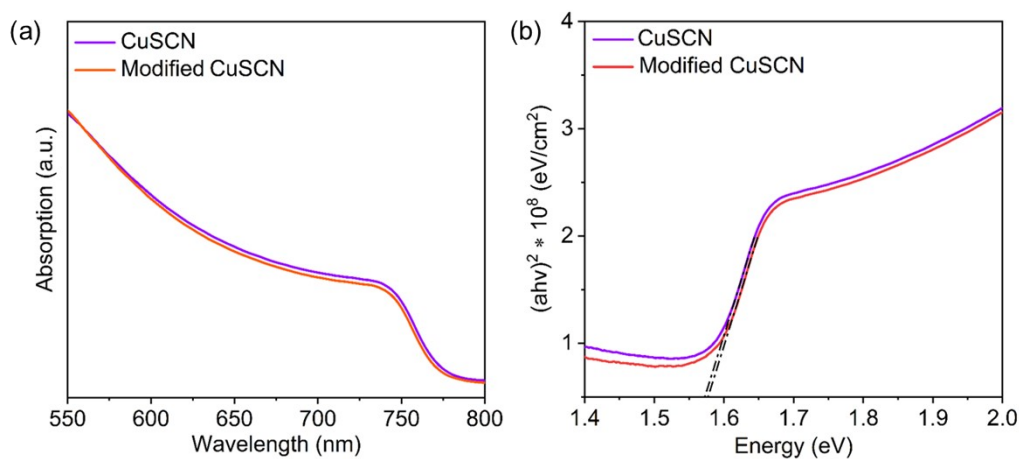


Figure S2. Characterization of the FTO/SnO<sub>2</sub>/perovskite/(pristine or modified) CuSCN samples: (a) UV-Vis spectra and (b) the Tauc plot curves.

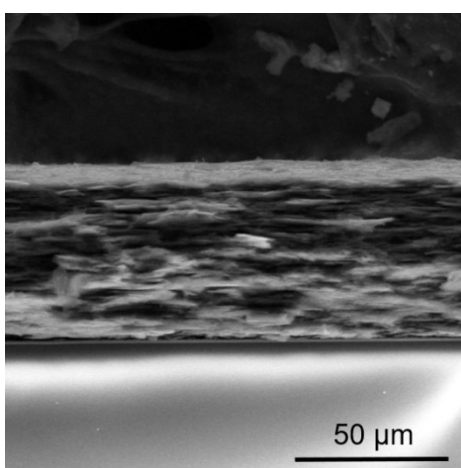


Figure S3. Cross-sectional SEM image of the whole C-PSC, where the thickness of carbon layer is 50  $\mu\text{m}$ .

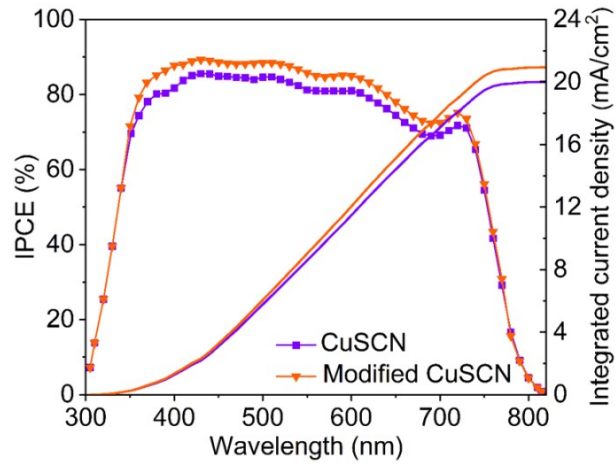


Figure S4. IPCE spectra of C-PSCs based on pristine and modified CuSCN.

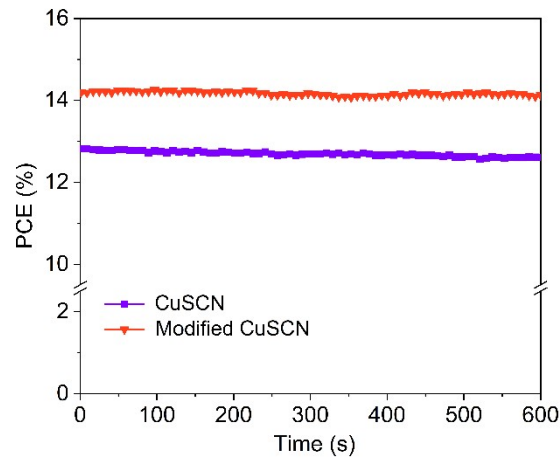


Figure S5. The output tracking of the device based on pristine and modified CuSCN in the maximum power point under AM 1.5G illumination.

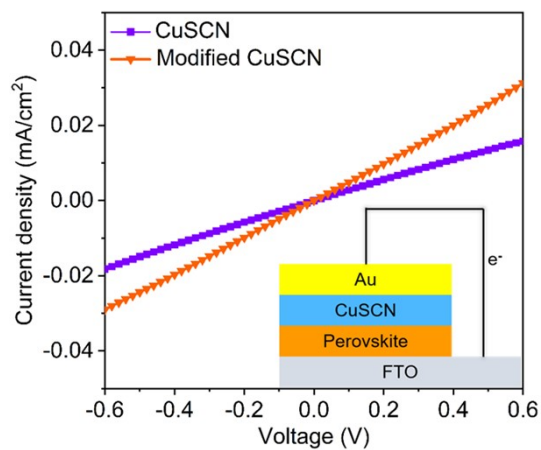


Figure S6. LSV curves of the devices based on pristine and modified CuSCN, and the inset is structure of the device.

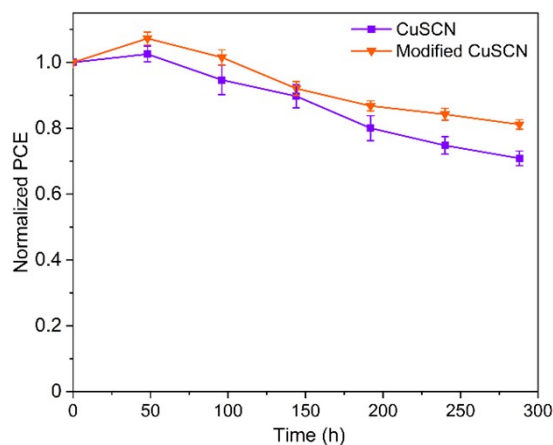


Figure S7. Thermal stability of unencapsulated devices based on pristine and modified CuSCN under 85 °C in environment, and error bars represent statistical data from 5 cells at the same batch.

Table. S1 Photovoltaic parameters of C-PSCs based on pristine and modified CuSCN

Devices		$V_{oc}$ (V)	$J_{sc}$ (mA·cm <sup>-2</sup> )	FF	PCE (%)
CuSCN	Champion	1.04	20.87	0.61	13.20
	Average	1.02 ± 0.02	19.09 ± 1.08	0.58 ± 0.03	11.49 ± 0.93
Modified-CuSCN	Champion	1.09	21.62	0.62	14.62
	Average	1.07 ± 0.02	20.22 ± 0.66	0.59 ± 0.03	12.64 ± 1.12

Table S2. TRPL parameters of half-devices with FTO/perovskite/(pristine or modified) CuSCN.

Devices	$\tau_1$ (ns)	$\tau_2$ (ns)
CuSCN	0.53	27.86
Modified CuSCN	0.41	23.75