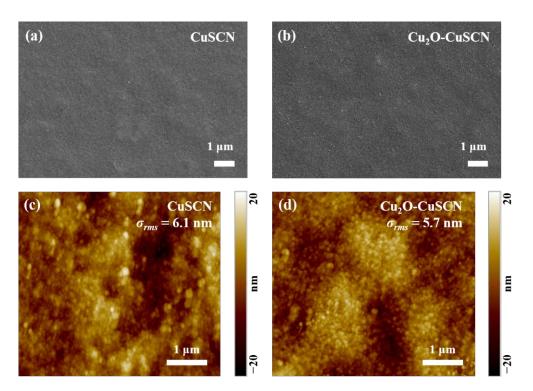
## **Supporting Information**

## Cu<sub>2</sub>O-CuSCN Nanocomposite as a Hole-Transport Material of Perovskite Solar Cells for the Enhanced Carrier Transport and Suppressed Interfacial Degradation

Jinhyun Kim,<sup>†</sup> Younghyun Lee,<sup>†</sup> Bumjin Gil,<sup>†</sup> Alan Jiwan Yun, Jaewon Kim, Hyungsub Woo,

Kimin Park, and Byungwoo Park<sup>\*</sup>

Department of Materials Science and Engineering, Research Institute of Advanced Materials, Seoul National University, Seoul 08826, Korea



**Figure S1. Surface morphologies and roughnesses of different HTLs.** (a) SEM image of each HTL depostied on CsFAMA. (b) Topography of each HTL as obtained by AFM.

<sup>\*</sup> E-mail: <u>byungwoo@snu.ac.kr</u>; Phone: +82-2-880-8319; Fax: +82-2-885-9671.

<sup>&</sup>lt;sup>†</sup> These authors contributed equally to this work.

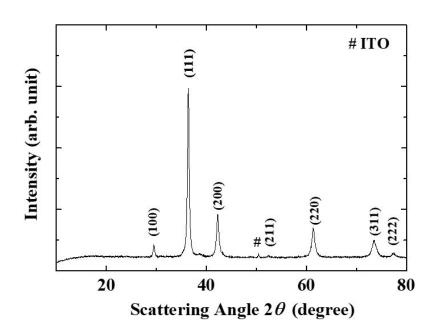


Figure S2. X-ray diffraction pattern of the  $\text{Cu}_2\text{O}$  nanoparticles.

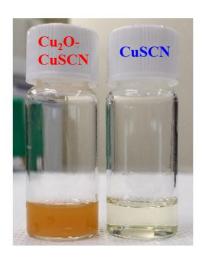


Figure S3. Optical images of the Cu<sub>2</sub>O-CuSCN and CuSCN solutions in diethyl sulfide.

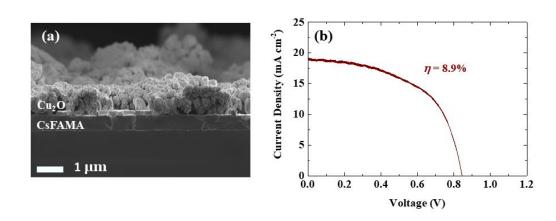
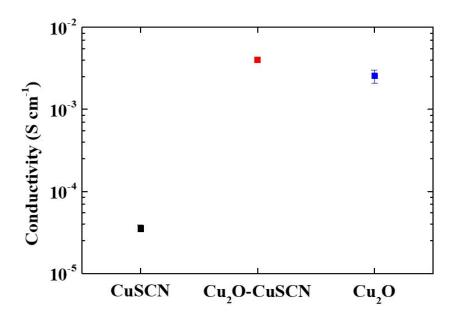


Figure S4. The film morphology and solar-cell performance with only Cu<sub>2</sub>O as an HTL. (a) Cross-sectional SEM image of the pure Cu<sub>2</sub>O film deposited on  $Cs_{0.05}(FA_{0.85}MA_{0.15})_{0.95}Pb(I_{0.85}Br_{0.15})_3$  (CsFAMA) film. (b) A *J-V* curve of the solar cell utilizing only Cu<sub>2</sub>O as an HTL. The device parameters are:  $V_{OC} = 0.846 \text{ V}$ ,  $J_{SC} = 18.9 \text{ mA/cm}^2$ , FF = 0.55, and  $\eta = 8.9\%$ .



**Figure S5. Electrical properties of various HTLs.** In-plane conductivity is measured by four-point probe for CuSCN, Cu<sub>2</sub>O-CuSCN nanocomposite, and Cu<sub>2</sub>O films deposited on glass substrates.

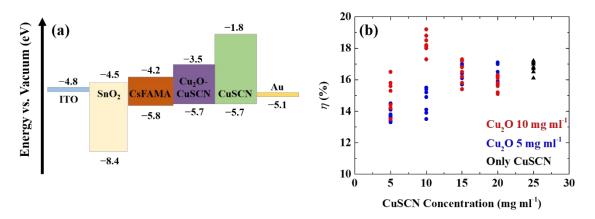


Figure S6. The energy level diagram and optimization processes of nanocomposite-based solar cells. (a) A schematic illustration of band structure of a solar cell. (b) Comparison of the solar-cell efficiencies at various Cu<sub>2</sub>O and CuSCN concentrations. The optimum condition is 10 mg/ml for both Cu<sub>2</sub>O and CuSCN.

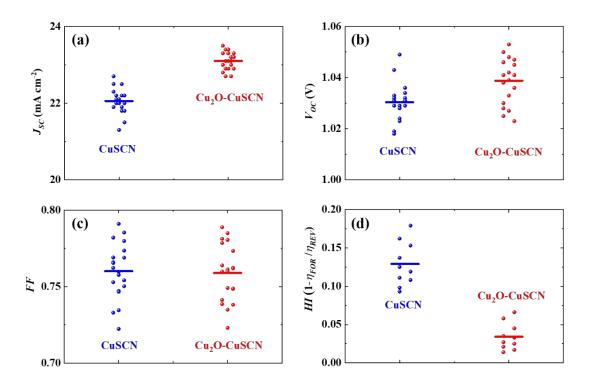


Figure S7. Device performance of solar cells with different HTLs. (a)  $J_{SC}$ , (b)  $V_{OC}$ , (c) FF, and (d) hysteresis index ( $HI = 1 - \eta_{FOR}/\eta_{REV}$ ) of the solar cells with Cu<sub>2</sub>O-CuSCN or CuSCN as an HTL, where lines represent average values.

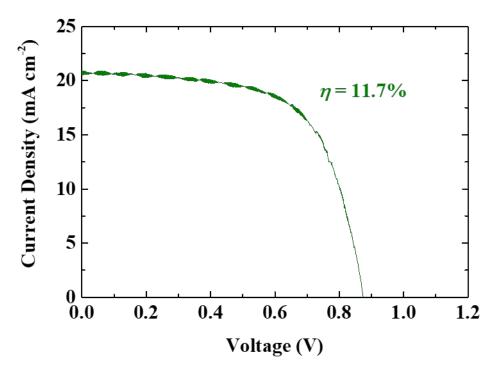


Figure S8. The *J-V* plot of the solar cell with a sequentially-deposited bilayer HTL. The perovskite solar cell was fabricated by the following sequence: 10 mg/ml of Cu<sub>2</sub>O-dispersion solution was spin coated on a perovskite film, then 10 mg/ml of CuSCN solution was deposited subsequently. The device parameters are:  $V_{OC} = 0.875 \text{ V}$ ,  $J_{SC} = 20.5 \text{ mA/cm}^2$ , FF = 0.652, and  $\eta = 11.7\%$ .

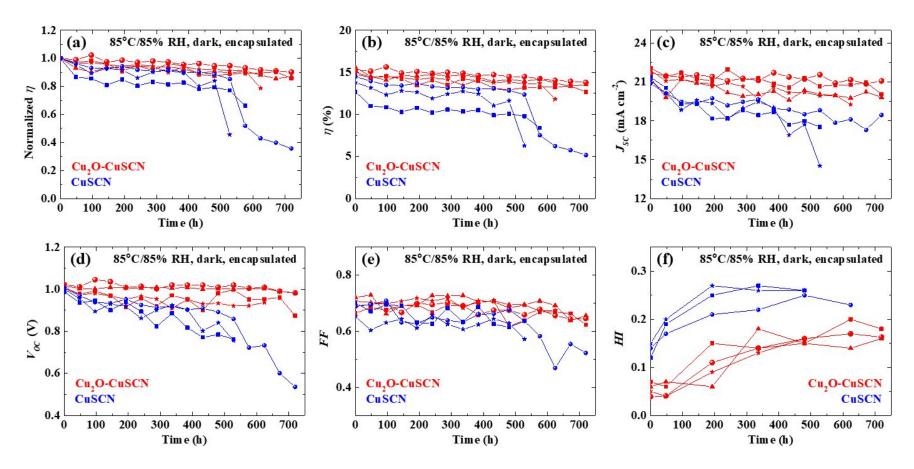


Figure S9. Long-term stability for several devices with different HTLs. (a) Normalized  $\eta$ , (b)  $\eta$ , (c)  $J_{SC}$ , (d)  $V_{OC}$ , (e) FF, and (h) HI of the solar cells stored under 85°C/85% RH with encapsulation in dark condition.