Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.


# Supplementary Information

## Enhancement of open circuit voltage for the CuSCN-employed perovskite solar cells by controlling perovskite/CuSCN interface with functional molecules

In Seok Yang, Soomin Lee, Juseob Choi, Min Tai Jung, Jeongho Kim* and Wan In Lee*

Department of Chemistry and Chemical Engineering, Inha University, Incheon, 22212 Korea

*Corresponding authors.

*E-mail address*: jkim5@inha.ac.kr (J. Kim), wanin@inha.ac.kr (W. I. Lee).

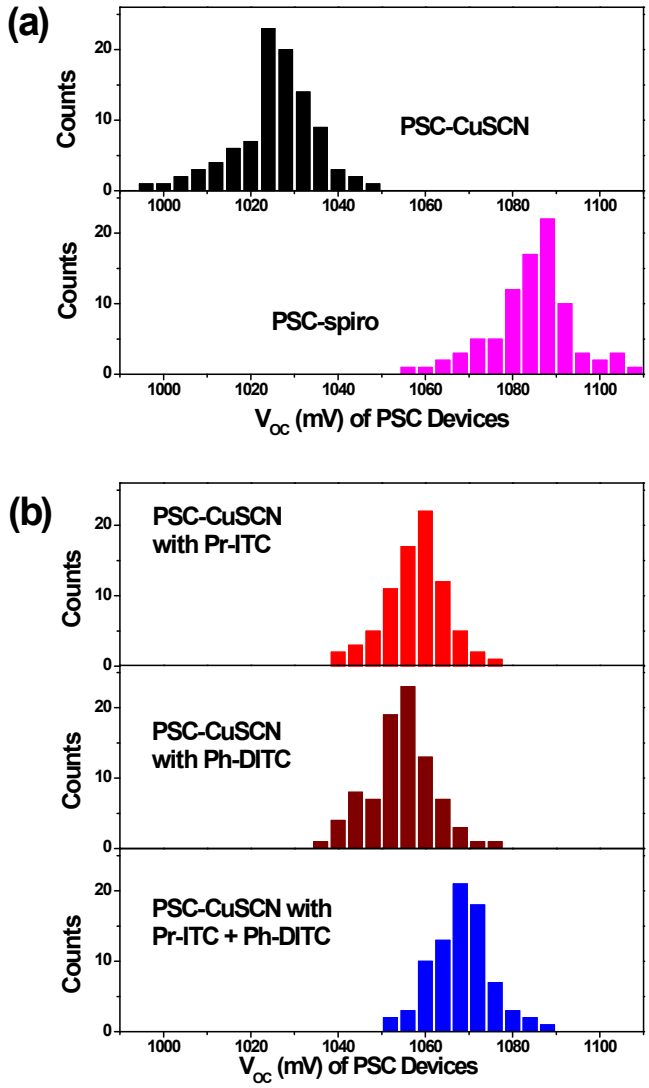



**Fig. S1**. $V_{OC}$ distributions observed from J-V curves of PSC devices with CuSCN and spiro-OMeTAD (a) and those of PSC-CuSCN devices employing Pr-ITC, Ph-DITC, and 50:50 mixture of Pr-ITC and Ph-DITC as IML (b).

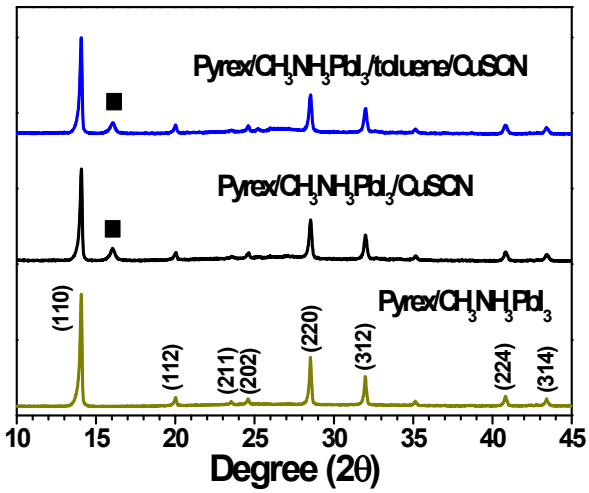
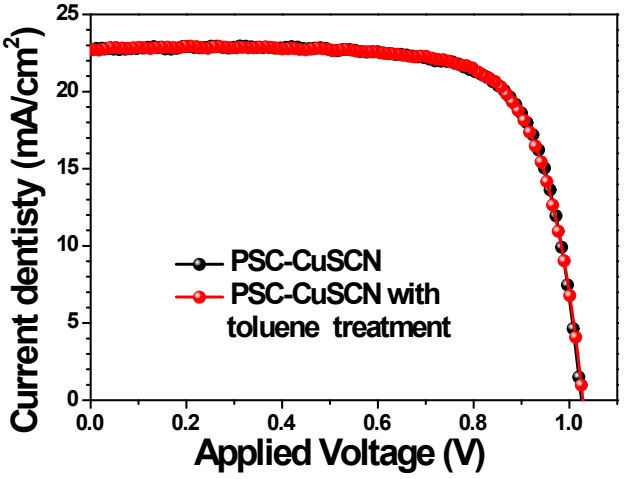



**Fig. S2**. XRD patterns of bare $CH_3NH_3PbI_3$, $CH_3NH_3PbI_3$/CuSCN and $CH_3NH_3PbI_3$ /toluene/CuSCN films coated on Pyrex substrate. In the last sample, toluene was applied as anti-solvent for the as-prepared $CH_3NH_3PbI_3$ film before depositing CuSCN layer. ■ denotes the (003) facet of CuSCN.




| PSCs | $V_{OC}$ (V) | $J_{SC}$ (mA cm$^{-2}$) | FF (%) | PCE (%) |
|---|---|---|---|---|
| PSC-CuSCN | 1.025 | 22.77 | 74.45 | 17.38 |
| PSC-CuSCN with toluene treatment | 1.028 | 22.71 | 74.61 | 17.42 |

**Fig. S3**. *J-V* curves for the bare and toluene-treated PSC-CuSCN devices with average PCE values. In the latter device, toluene was applied as anti-solvent for the as-prepared $CH_3NH_3PbI_3$ film before depositing CuSCN.

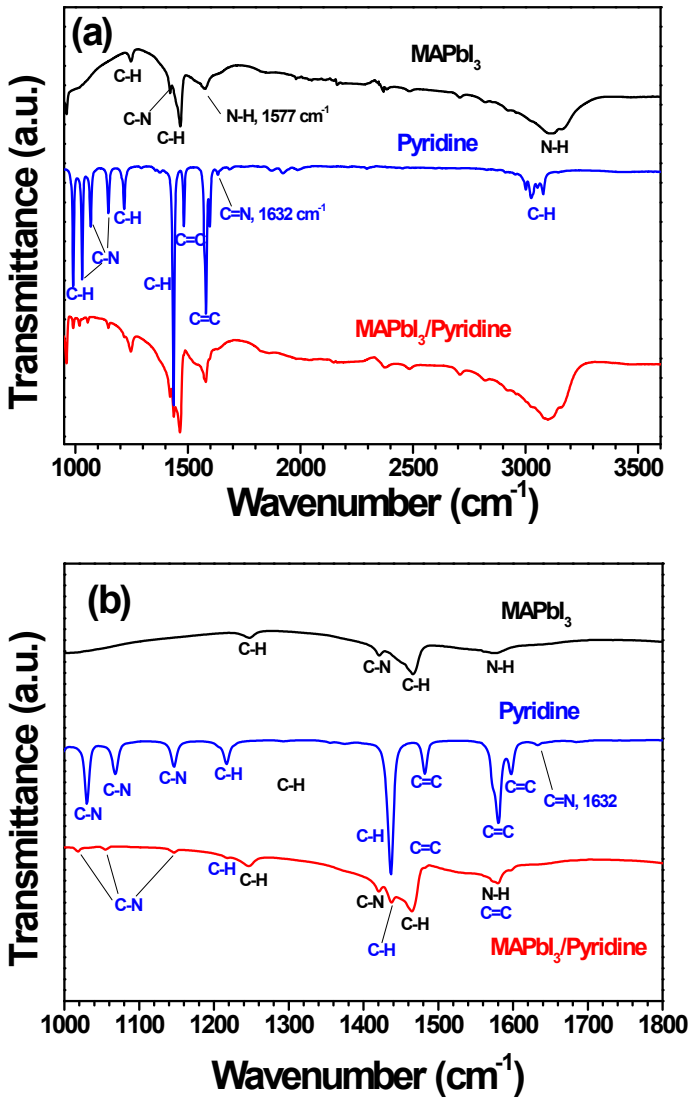



**Fig. S4**. FTIR spectra (a,b) of $MAPbI_3$, pyridine, and $MAPbI_3$/pyridine. The $MAPbI_3$ powders treated by pyridine ($MAPbI_3$ /pyridine) were prepared by following procedure. 50 μL anhydrous pyridine was dissolved in 7 mL toluene in Ar atmosphere. 1.0 g $MAPbI_3$ powders was then added to this solution and stirred for 20 min in Ar atmosphere. The suspension was filtered, washed by toluene several times, and dried in a vacuum oven at 70°C.

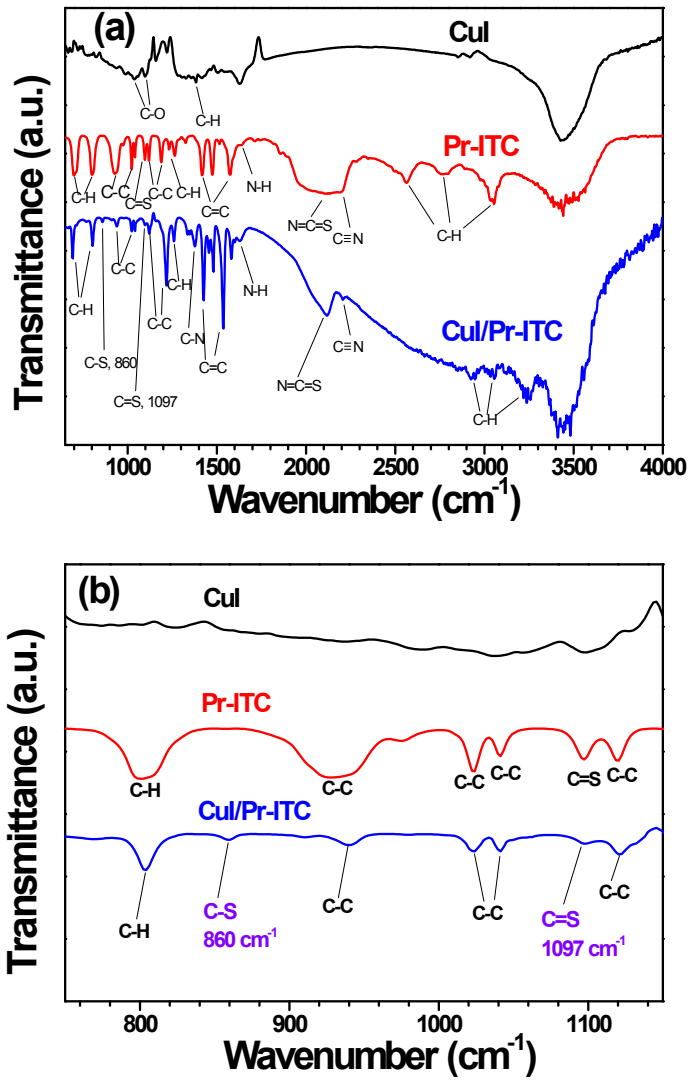
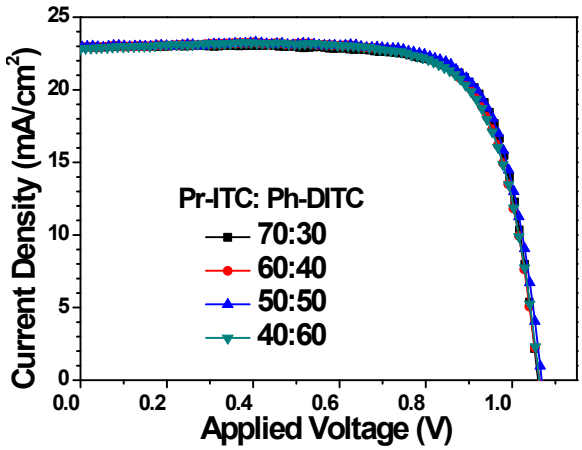



**Fig. S5**. FTIR spectra (a, b) of CuI, Pr-ITC, and CuI/Pr-ITC. The CuI powders treated by Pr-ITC (CuI/Pr-ITC) were prepared by following procedure. 25 μL Pr-ITC was dissolved in 5 mL anhydrous methanol in Ar atmosphere. 0.5 g CuI fine powders were then added to this solution, and the suspension was stirred for 20 min in Ar atmosphere. The suspension was filtered, washed by methanol several times, and dried in a vacuum.




| Pr-ITC : Ph-DITC (molar ratio) | $V_{OC}$ (V) | $J_{SC}$ (mA/cm$^2$) | FF (%) | PCE (%) |
|---|---|---|---|---|
| 100:0 | 1.057 | 22.68 | 75.94 | 18.20 |
| 70:30 | 1.059 | 22.94 | 75.38 | 18.31 |
| 60:40 | 1.064 | 22.92 | 75.76 | 18.48 |
| 50:50 | 1.068 | 22.85 | 76.08 | 18.57 |
| 40:60 | 1.059 | 22.83 | 75.23 | 18.18 |
| 0:100 | 1.054 | 22.72 | 74.91 | 17.94 |

**Fig. S6.** *J-V* curves of PSC-CuSCNs introducing the mixed IMLs of Pr-ITC + Ph-DITC with various ratios.

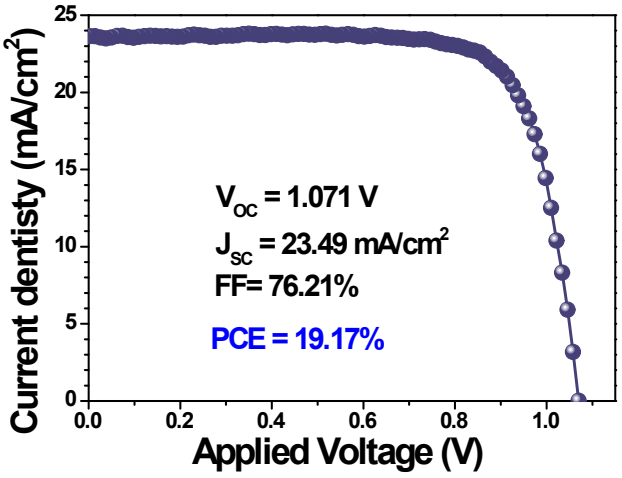
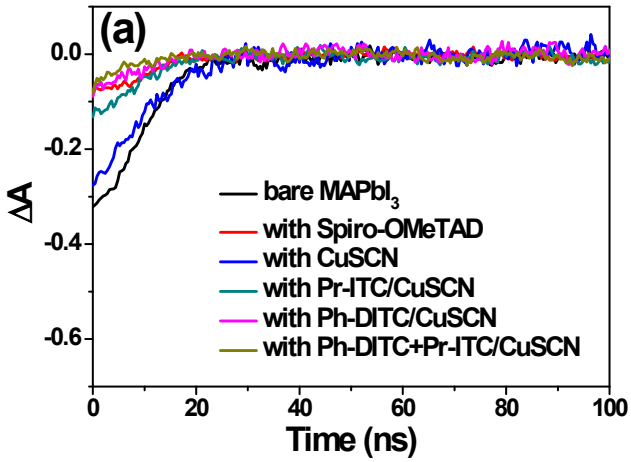
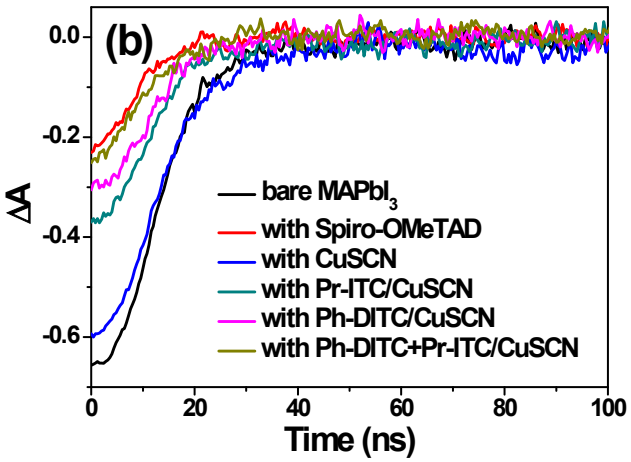


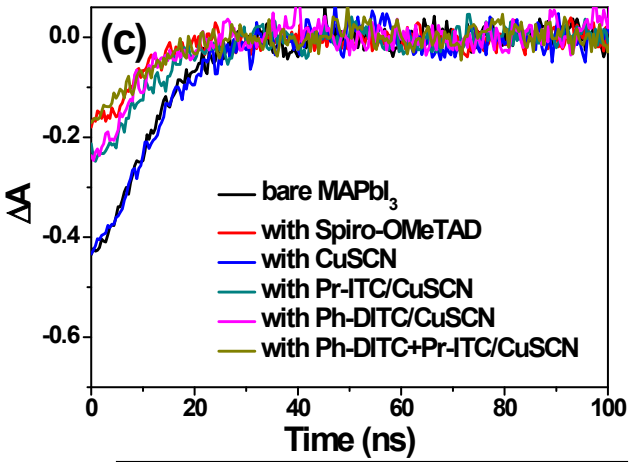

**Fig. S7**. J-V curves of the champion PSC-CuSCN device employing 50:50 mixture of Pr-ITC and Ph-DITC as IML.




| Devices | $\tau_1$ (ns) | $A_1$ |
|---|---|---|
| $CH_3NH_3PbI_3$ | 12.24 | -1.36 |
| $CH_3NH_3PbI_3$/Spiro-OMeTAD | 9.88 | -0.69 |
| $CH_3NH_3PbI_3$/CuSCN | 11.55 | -1.17 |
| $CH_3NH_3PbI_3$/Pr-ITC/CuSCN | 10.81 | -1.00 |
| $CH_3NH_3PbI_3$/Ph-DITC /CuSCN | 10.77 | -0.94 |
| $CH_3NH_3PbI_3$/Ph-DITC + Pr-ITC/CuSCN | 10.55 | -0.90 |

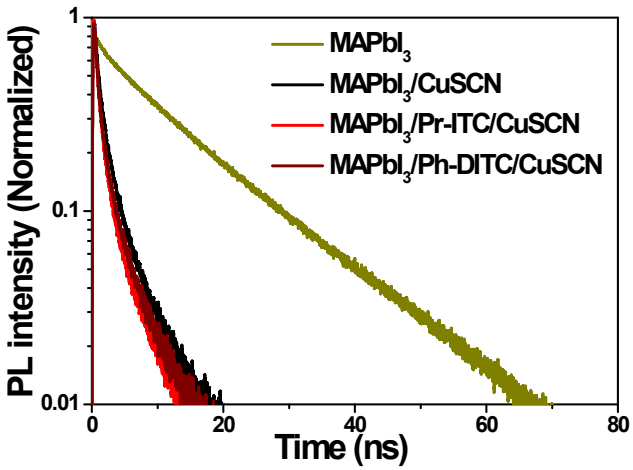



| Devices | $\tau_1$ (ns) | $A_1$ |
|---|---|---|
| $CH_3NH_3PbI_3$ | 13.89 | -3.52 |
| $CH_3NH_3PbI_3$/Spiro-OMeTAD | 8.74 | -0.49 |
| $CH_3NH_3PbI_3$/CuSCN | 12.42 | -2.04 |
| $CH_3NH_3PbI_3$/Pr-ITC/CuSCN | 9.91 | -1.13 |
| $CH_3NH_3PbI_3$/Ph-DITC /CuSCN | 10.56 | -1.35 |
| $CH_3NH_3PbI_3$/Ph-DITC + Pr-ITC/CuSCN | 9.73 | -0.66 |



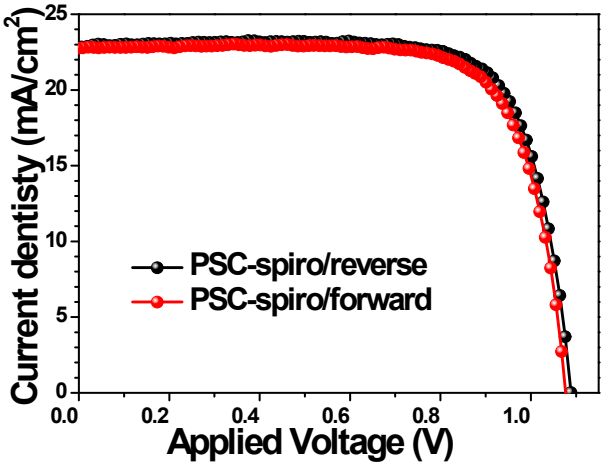

| Devices | $\tau_1$ (ns) | $A_1$ |
|---|---|---|
| $CH_3NH_3PbI_3$ | 11.68 | -0.44 |
| $CH_3NH_3PbI_3$/Spiro-OMeTAD | 8.87 | -0.59 |
| $CH_3NH_3PbI_3$/CuSCN | 12.38 | -0.46 |
| $CH_3NH_3PbI_3$/Pr-ITC/CuSCN | 10.33 | -0.23 |
| $CH_3NH_3PbI_3$/Ph-DITC /CuSCN | 10.55 | -0.19 |
| $CH_3NH_3PbI_3$/Ph-DITC + Pr-ITC/CuSCN | 8.03 | -0.15 |

**Fig. S8**. Temporal decays of TA signals at 730 nm (a), 750 nm (b), and 800 nm (c) of bare $MAPbI_3$, PSC-spiro, bare PSC-CuSCN, and PSC-CuSCNs with Pr-ITC + Ph-DITC and the corresponding fit parameters.




| Devices | $\tau_1$ (ns) | $A_1$ (%) | $\tau_2$ (ns) | $A_2$ (%) | $\tau_{avg}$ (ns) | $\tau_{CT}$ (ns) | CTE (%) |
|---|---|---|---|---|---|---|---|
| Bare $MAPbI_3$ | 16.588 | 53.920 | 2.863 | 46.080 | 10.264 | -- | -- |
| $MAPbI_3$/CuSCN | 3.877 | 19.880 | 0.623 | 80.120 | 1.270 | 1.449 | 87.63 |
| $MAPbI_3$/Pr-ITC/CuSCN | 3.967 | 12.450 | 0.554 | 87.550 | 0.978 | 1.082 | 90.47 |
| $MAPbI_3$/Ph-DITC/CuSCN | 3.409 | 18.920 | 0.484 | 81.080 | 1.037 | 1.154 | 89.89 |

**Fig. S9**. Temporal decays of TR-PL signal of bare $MAPbI_3$, PSC-CuSCN, and PSC-CuSCN with Pr-ITC or Ph-DITC. TR-PL Fit parameters are shown in the Table. The PL lifetime of each sample corresponds to the amplitude-weighted average lifetime of a multi-exponential decay fit.




| Scan direction | $V_{OC}$ (V) | $J_{SC}$ (mA cm$^{-2}$) | FF (%) | PCE (%) |
|---|---|---|---|---|
| backward | 1.088 | 22.83 | 76.60 | 19.03 |
| forward | 1.077 | 22.76 | 74.90 | 18.36 |

**Fig. S10**. *J-V* curves of PSC-spiro with average PCE values, acquired by forward and backward scans with a scan rate of 50 mV s$^{-1}$.

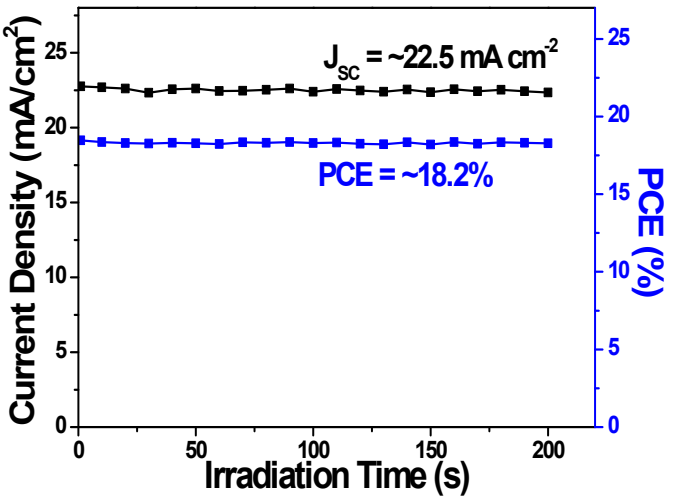



**Fig. S11**. Steady-state photocurrent and power output at the maximum power point for PSC-CuSCN with Pr-ITC + Ph-DITC.