

## Electronic Supplementary Information (ESI)

# High Performance Planar-Heterojunction Perovskite Solar Cells Using Amino-based Fulleropyrrolidine as the Electron Transporting Material

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### Experimental Section

#### Preparation for precursor solution:

All materials mentioned were purchased from *Alfa Aesar* or *Sigma-Aldrich*, and were used as received without any further purification. The  $\text{CH}_3\text{NH}_3\text{I}$  (MAI) was synthesized according to previous literature.<sup>1</sup> MAI and lead chloride ( $\text{PbCl}_2$ , 99.999%, *Alfa*) powder were mixed in anhydrous DMF (99.9%, *Alfa*) with a molar ratio of 3:1 for 40mg/100mg precursor solution. The mixture solutions were stirred at 70°C overnight in a glovebox with nitrogen atmosphere and filtered with a 0.45  $\mu\text{m}$  Polytetrafluoroethylene (PTFE) filter before spin coating. The  $\text{C}_{60}\text{-N}$  material was synthesized according to previous paper.<sup>2</sup>

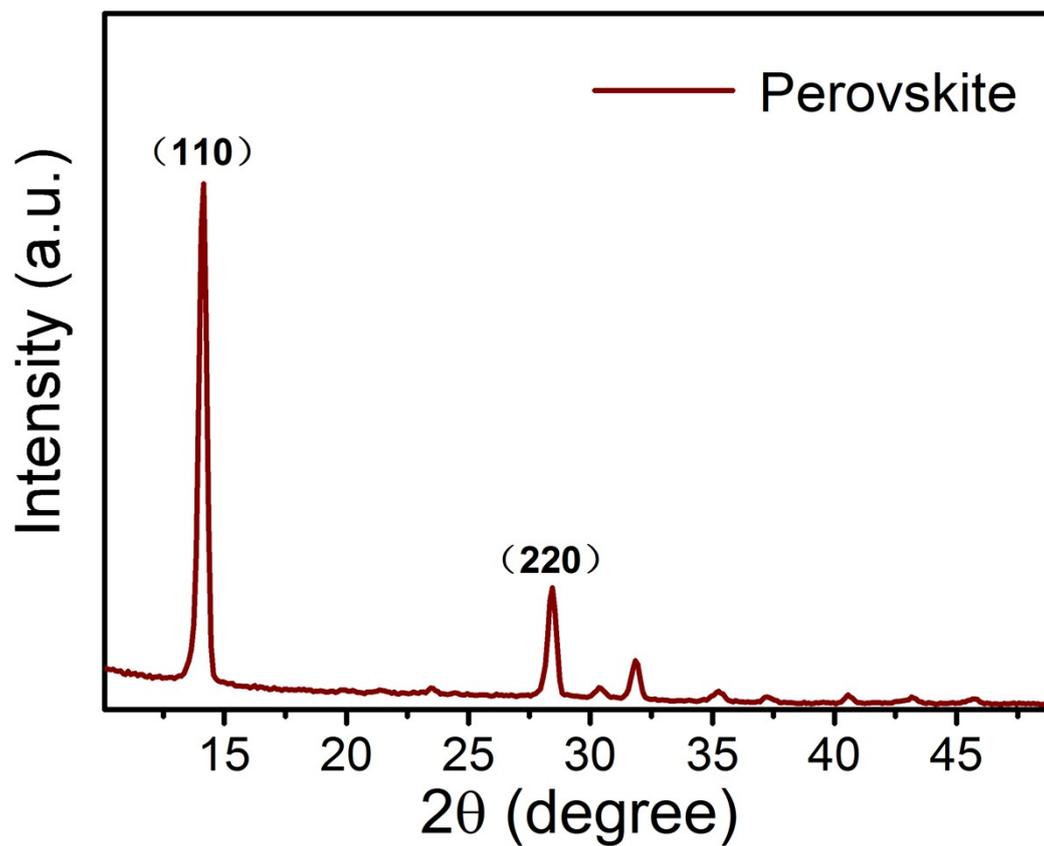
#### Device fabrication:

The device structure is: ITO/PEDOT:PSS/ $\text{CH}_3\text{NH}_3\text{PbI}_{3-x}\text{Cl}_x$ /ETM/Al. First the indium-tin oxide (ITO) was pre-cleaned and treated with ultraviolet-ozone (UVO) for 30 minutes. Then the poly(3,4-ethylene-dioxy-thiophene):polystyrene (PEDOT:PSS, Al 4083) was spin-coated on the ITO substrate at 4500 rpm for 40s and baked at 150 °C for 10 minutes in ambient atmosphere. A 40 wt%  $\text{CH}_3\text{NH}_3\text{PbI}_{3-x}\text{Cl}_x$  precursor solution was spin-coated on the PEDOT:PSS layer at the speed 4000 rpm for 40s in a nitrogen glove box with  $\text{O}_2$  and  $\text{H}_2\text{O}$  both < 2 ppm and left the as-deposited

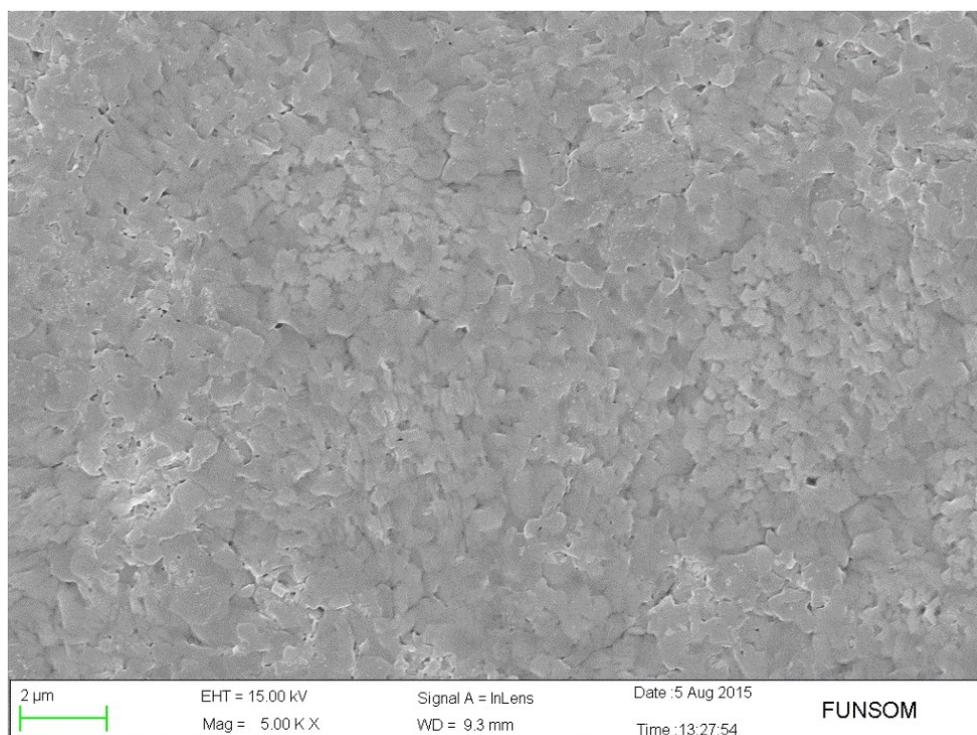
perovskite film for five minutes in a capped petri dish, and subsequently annealed at 90°C for 60 minutes, with the color of the films converted from light yellow to black. After being cooled to room temperature, the electron transport layer was deposited from the solution with a concentration of 20 mg/ml where PCBM was dissolved in a blend solvents of chloroform and chlorobenzene and C<sub>60</sub>-N was dissolved in trifluoroethanol and spin-coated at 2000 rpm for 40s. Finally, a thickness of 100 nm Al cathode was deposited by thermal vacuum evaporation through a shadow mask (active area 7.25 mm<sup>2</sup>) under 10<sup>-6</sup> Torr.

**Characterization :**

The current density-voltage (*J-V*) parameters of the devices were characterized by using a Keithley 2400 digital source meter under simulated AM 1.5G solar irradiation at 100 mW cm<sup>-2</sup> (Newport, Class AAA solar simulator, 94023A-U). The light intensity was calibrated by a certified Oriol Reference Cell (91150 V). The EQE was performed using a certified EQE instrument (Zolix Instruments, Inc. Solar Cell Scan 100). The photoluminescence (PL) measurements were carried out on a FluoroMax-4 Spectrofluorometer (HORIBA Scientific). A 625 nm laser source was used in the time resolved PL measurement. SEM images were obtained from a field emission scanning electron microscope (Quanta 200 FEG, FEI Co.). The ultraviolet-visible spectroscopy (*UV-vis*) spectra were recorded on a Perkin Elmer model Lambda 750 instrument. Conventional XRD measurement was conducted using a PANalytical 80 equipment (Empyrean, Cu K $\alpha$  radiation). Ultraviolet photo electron spectroscopy (UPS) were carried out on a Kratos AXIS UltraDLD ultrahigh vacuum (UHV) surface analysis system under a base pressure of 5 $\times$ 10<sup>-10</sup> torr with an unfiltered He (21.21 eV) gas discharge lamp.



**Fig. S1** XRD patterns of the perovskite films after thermal annealing.



**Fig. S2** SEM image of the perovskite's surface.

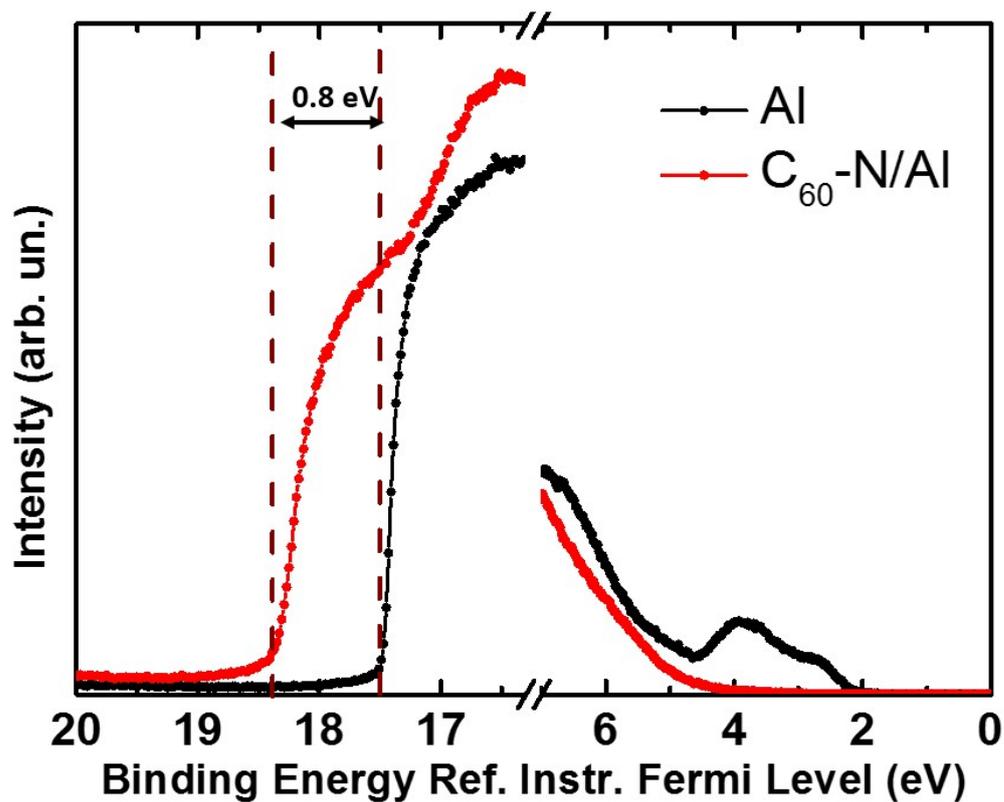


Fig. S3 Ultraviolet photoelectron spectra of C<sub>60</sub>-N/Al layer. The binding energy of Al is decreased by 0.8eV after the deposition of an ultra-thin C<sub>60</sub>-N layer.

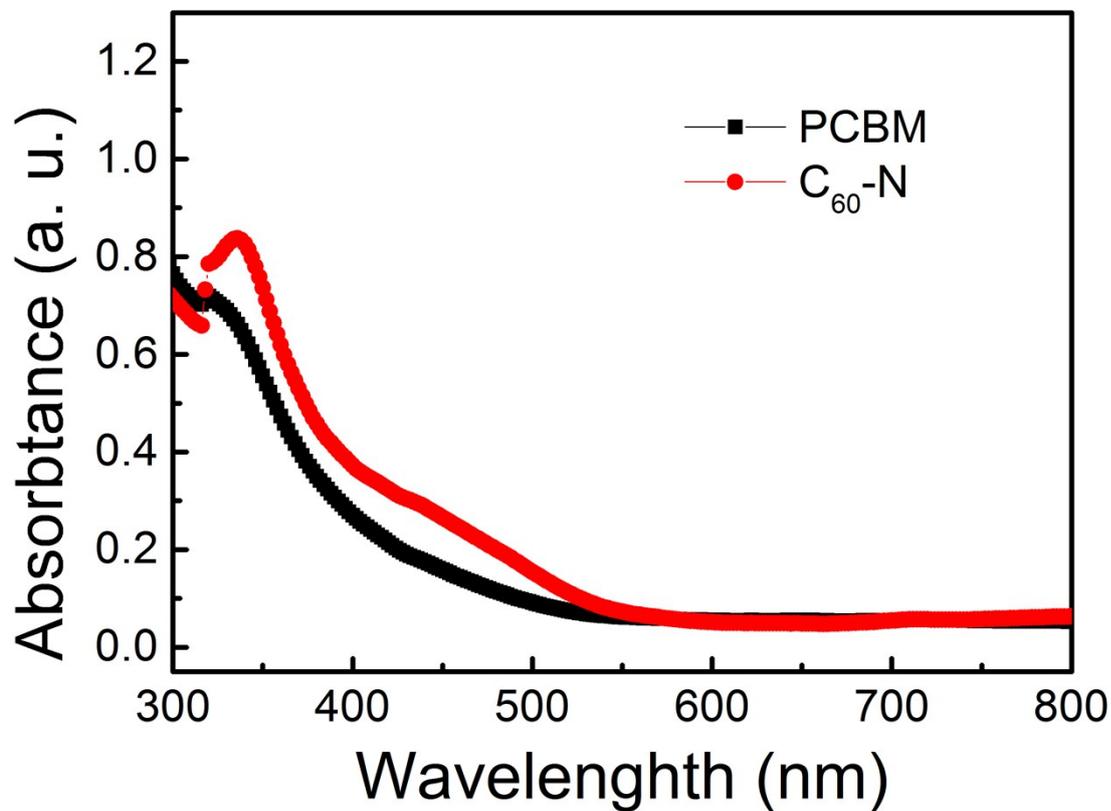


Fig. S4 UV-*vis* absorbance of devices based on PCBM and C<sub>60</sub>-N as the ETM.

**Table S1.** Device performances by using different ETMs.

ETM	$V_{oc}$ (V)	$J_{sc}$ (mA cm <sup>-2</sup> )	Fill Factor (%)	PCE (%)
PCBM	0.98	17.29	72.7	12.3
C <sub>60</sub> -N	1.02	21.24	76.4	16.6
C <sub>60</sub> -N (s-w <sup>*</sup> )/PCBM	1.00	20.96	72.6	15.2
Solvent <sup>**</sup> /PCBM	0.98	17.17	72.2	12.2

\* Used trifluoroethanol to wash off the C<sub>60</sub>-N layer and then deposited PCBM. \*\* Used trifluoroethanol to wash the perovskite film and then deposited PCBM.

### Supplementary References

- 1 H. S. Kim, C.-R. Lee, J.-H. Im, K.-B. Lee, T. Moehl, A. Marchioro, S.-J. Moon, R. Humphry-Baker, J.-H. Yum, J. E. Moser, M. Grätzel and N.-G. Park, *Sci. Rep.*, 2012, **2**, 591.
- 2 Z. A. Page, Y. Liu, V. V. Duzhko, T. P. Russell and T. Emrick, *Science*, 2014, **346**, 441-444.