

Supplementary Information

5 Modified deposition process of electron transport layer for efficient inverted planar perovskite solar cells

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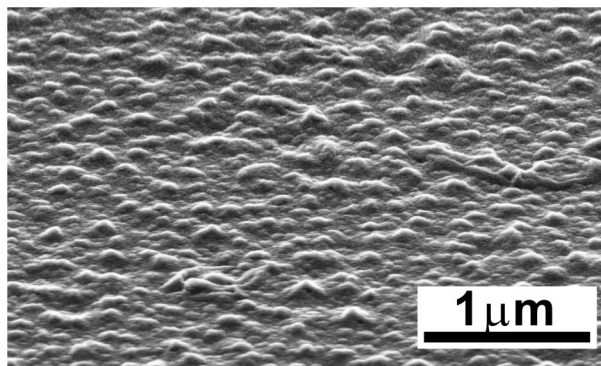
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20 **Table S1** Photovoltaic performances of the hybrid perovskite solar cell based on one-step MAPbI₃ layer.

Structure	PCBM (nm) spin-coating	PCBM (nm) Vaporizing	J _{sc} (mA/cm ²) (ave±s.d.)	V _{oc} (V) (ave±s.d.)	FF (ave±s.d.)	PCE(%) (ave±s.d.)
DEVICE A	40	-	14.78±1.67	0.81±0.05	0.52±0.04	6.23±1.32
ITO/PEDOT/ MAPbI ₃ (one-step)	60	-	15.98±0.78	0.84±0.04	0.55±0.03	7.37±0.92
/PCBM(spin-coating)/Ag	80	-	15.92±0.95	0.83±0.07	0.54±0.02	7.14±1.01

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Fig. S1 Tilt-angle SEM image of the structure B: ITO/PEDOT/ MAPbI₃(two-step)/PCBM(spin-coating, 100 nm)

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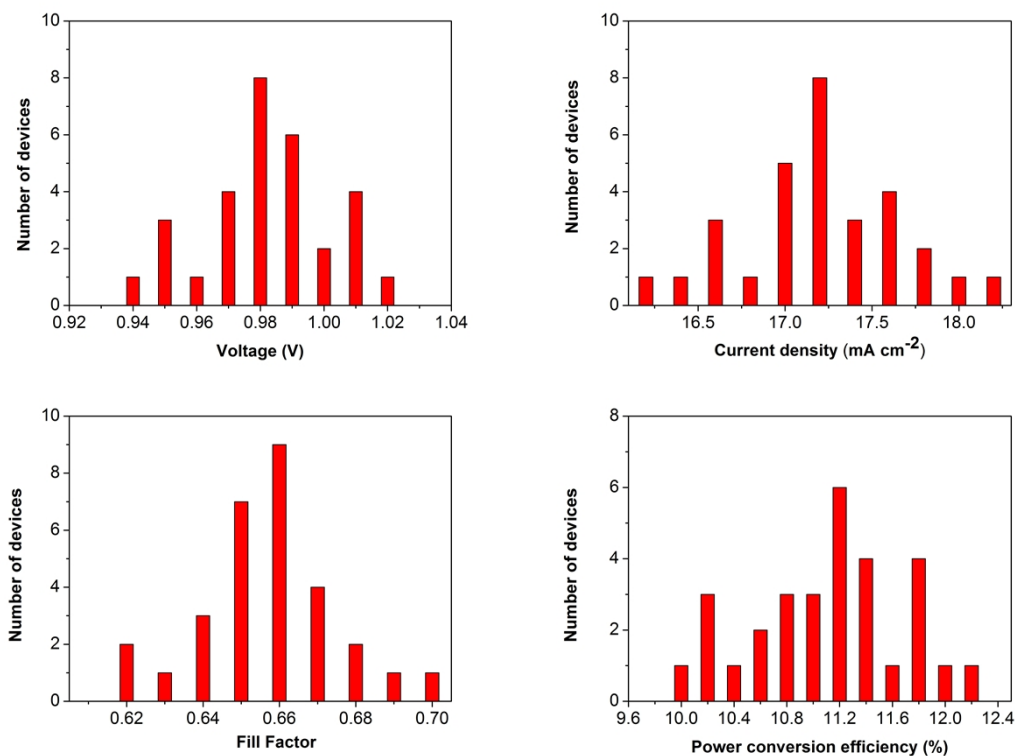
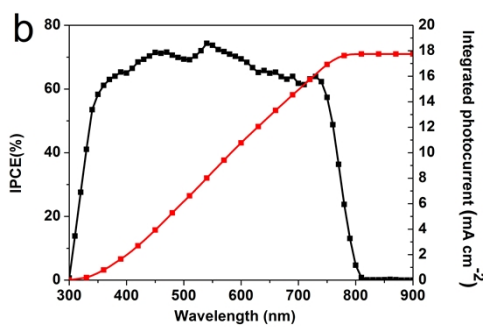
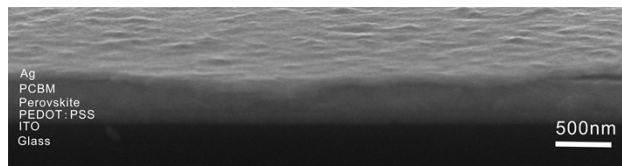


Fig. S2 Histograms of device parameters (V_{oc} , J_{sc} , Fill Factor and PCE.) measured for a bare of 30 cells: ITO/PEDOT/MAPbI₃(two-step)/PCBM(spin-coating 60 nm)/PCBM(Vaporing 30nm)/Ag.



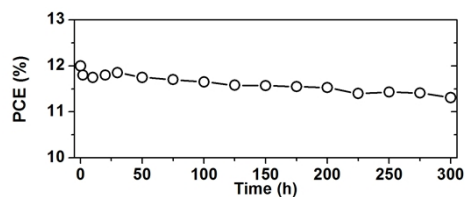
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Fig. S3 IPCE of the Device D with the highest efficiency and the corresponding integrated short circuit current over the spectrum



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Fig. S4 SEM cross-section of the optimized device D.



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Fig. S5 The evolution of the PCE about the best device D in the air (PCBM: 60 nm + 30 nm)

Experimental Section

Preparation of Materials:

Methylammonium iodide ($\text{CH}_3\text{NH}_3\text{I}$) was purchased from Dyesol. PbI_2 (99.999 wt%) was purchased from Alfa Aesar. PEDOT:PSS (CLEVIOS PH 1000) solution was acquired from CLEVIOS, and PCBM were acquired from Haraeus, respectively.

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Fabrication of solar cells:

ITO glass substrates were cleaned sequentially by detergent, pure water, acetone for 20 min. The dried substrates were treated with ultraviolet ozone plasma for 5 min for further clean. PEDOT:PSS solution (dissolved with deionized water in a mixture ration of 1:3) were spun at 1000 rpm for 30 s and annealed at 120 °C for 20 min.

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One-step $\text{CH}_3\text{NH}_3\text{PbI}_3$ layer:

A 1:3 ratio of $\text{PbI}_2/\text{CH}_3\text{NH}_3\text{I}$ was mixed in DMF solution. Specifically, the concentration of the PbI_2 and $\text{CH}_3\text{NH}_3\text{I}$ were 0.8 and 2.4 M. The solution was spin-coated onto the PEDOT:PSS layer at 2000 rpm and then annealed at 100 °C for 2 h.

Two-step $\text{CH}_3\text{NH}_3\text{PbI}_3$ layer:

PbI_2 film was prepared by spin-coating a PbI_2 DMF solution onto the PEDOT:PSS layer at 5000 rpm for 60 s, baked on a 70 °C hot plate for 1 h, then dipped into a 10 mg/mL $\text{CH}_3\text{NH}_3\text{I}$ anhydrous isopropyl alcohol solution for 30 s, and finally flushed with anhydrous isopropyl alcohol for 10 s. This film was further annealed on a 70 °C hot plate for 20 min to finish the formation of $\text{CH}_3\text{NH}_3\text{PbI}_3$.

20 As for devices A, B and D, 40 nm, 60 nm, 80 nm, 100 nm and 120nm n-type films were deposited by spin-coating PCBM solution (15 mg/ml in chlorobenzene, 2000 rpm), (20 mg/ml in chlorobenzene, 2000 rpm), (30 mg/ml in chlorobenzene, 2000 rpm), (30 mg/ml in chlorobenzene, 1500 rpm), (30 mg/ml in chlorobenzene, 1000 rpm) for 60s, respectively. As for device C and D, different thickness of PCBM films were obtained by vaporeing with different deposition time at the rate of 1Å/s. Eventually, the devices were completed by consecutively vacuum deposited Ag electrodes (120 nm) under 10^{-5} mbar, through a shadow mask. The processes of preparing

25 PEDOT:PSS layer, $\text{CH}_3\text{NH}_3\text{PbI}_3$ layer and PCBM layer (spin-coating) were achieved in the glovebox with nitrogen atmosphere.

Here it is noted that all the thickness of the PCBM was calibrated by the ellipsometer on the planar glass substrates before preparing solar cells.

Characterization:

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The morphologies were investigated by a scanning electron microscopy (SEM) (Quanta 250, FEI). The roughness was determined using an atomic force microscopy (AFM) (NT-DMT, solver P74H-PRO, Russia). The crystalline structures were performed by a X-ray diffraction (XRD) (D/MAX-2400, Rigaku, Japan) with Cu K α radiation. Device characteristics were evaluated in ambient under a AAA solar simulator (XES-301S, SAN-EI Electric. Co. Ltd.), AM 1.5G illumination with an intensity of 100 mW/cm² (1 sun, calibrated by a

35 NREL-traceable KG5 filtered silicon reference cell). Meanwhile, the current density-voltage (J-V) curves were measured by a Keithley digital source meter (Model 2602), and the scan rate is 0.05V/s for both reverse and forward. The Incident Photon-to-current Conversion Efficiency (IPCE) spectra were obtained by the solar cell quantum efficiency measurement system (SolarCellScan 100, Zolix instruments. Co. Ltd).

The active area of the cells was defined by a metal mask with square aperture of the area of 0.09 cm². The premasked active area of the

40 solar cells was approximately 0.16 cm² nominally defined by the overlap area of the Ag and ITO electrodes. Devices were masked for all the current voltage measurements. The reference cells and test cells are located in the same spot under the solar simulator during measurement.