

Supporting information

Buried Interface Modification for High Performance and Stable Perovskite Solar Cells

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Materials:

Formamidinium iodide (FAI, $\geq 99.9\%$), lead iodide (PbI_2 , $\geq 99.99\%$), methylammonium chloride (MACl , $\geq 99.9\%$), Cesium iodide (CsI , 99.9%), methylammonium bromide (MABr , $\geq 99.9\%$), Lead(II) bromide (PbBr_2 , 99.9%), 2,2',7,7'-tetrakis(N,N-dimethoxyphenylamine)-9,9'-spirobifluorene (Spiro-OMeTAD), dimethyl sulfoxide (DMSO, 99.9%), N, N-dimethylformamide (DMF, 99.8%), Chlorobenzene (CB, 99.8%), and isopropanol (IPA, 99.5%) were purchased from Advanced Electron Technology Co., Ltd. Lithium bis(trifluoromethanesulfonyl)imide (Li-TFSI), 4-tert-butylpyridine (tBP, 99.0%), , and Tin (IV) oxide (SnO_2) colloidal precursor, 15% in H_2O colloidal dispersion were obtained from Alfa Aesar. Titanium tetrachloride (TiCl_4) and ethyl ether were purchased from China National Pharmaceutical Group Corporation.

Device fabrication

FTO was cleaned with glass cleaning agent, deionized water and ethanol, dried, and treated with ozone for 15 min. SnO_2 colloidal dispersion was diluted with deionized H_2O in a volume ratio of 1:3 before use. The SnO_2 precursor was spin-coated onto the FTO at 5000 rpm for 40 s, followed by thermal annealing in air at $150\text{ }^\circ\text{C}$ for 30 min. Then, the substrate is treated with UV for 15min, and the coated with PC bottom modification layer (the solvent is water). It is spin-coated on the SnO_2 layer at 4000 rpm for 30 s and annealing at $100\text{ }^\circ\text{C}$ for 10 min. The 1.3 M perovskite precursor solution was prepared by mixing CsI , FAI, MABr , PbI_2 and PbBr_2 in 1 ml DMF:DMSO (5:1/v:v) mixed solvent with a chemical formula of $(\text{FA}_{0.98}\text{MA}_{0.02})_{0.95}\text{Cs}_{0.05}\text{Pb}(\text{I}_{0.95}\text{Br}_{0.05})_3$, 5 mol% of excess PbI_2 was needed to improve the device performance. Then 15.5 mol% MACl was added to the perovskite precursor solution and stirred for 6h. 60 μL perovskite solutions were spin-coated onto FTO/ SnO_2 at 1000 rpm for 10 s, subsequently at 5000 rpm for 40 s. 900 μL ethyl ether was dripped onto the center of film at 12 s before the end of spin-coating. The as-prepared perovskite films were subsequently annealed on a hotplate at $120\text{ }^\circ\text{C}$ for 20 min. Subsequently, the PEAI solution (2.0 mg/mL in IPA) was dropped on the annealed perovskite films during a spin-coating procedure at 5000 rpm for 30 s. The Spiro-OMeTAD layer was deposited

on the top of perovskite films by spin-coating the solution (90 mg Spiro-OMeTAD in 1 mL CB with 22 μ L Li-TFSI solution (520 mg/mL in ACN), and 36 μ L tBP) at 5000 rpm for 30 s. Finally, an 80 nm gold electrode was thermally evaporated under vacuum. For the preparation of F-PSCs, a flexible substrate was obtained by sputtering 120nm ITO onto PET using magnetron sputtering, and the annealing condition for SnO₂ was 120 °C for 45 min. The other preparation methods were consistent with the above process. The preparation of FAPbI₃-based PSCs was consistent with what we reported before.^[1]

Device characterization

X-ray diffraction (XRD) patterns were measured using a DX-2700BH diffractometer and grazing incidence X-ray diffraction (GIXRD) measurements were conducted using a PANalytical Empyrean diffractometer, which both equipped with Cu K α radiation. Grazing incidence X-ray scattering (GIWAXs) pattern at an incidence angle of 0.4° was conducted by Shanghai Synchrotron Radiation Facility at the BL17B1 line station. The in situ UV–Vis absorption spectroscopy was recorded with a F20 spectrometer (Filmetrics, Inc.) equipped with tungsten halogen and deuterium light sources in transmission mode. A Newport light source, 2400 digital source meter was used to test the perovskite cell J-V curve (scan voltage: 1.5 V -- 0.1 V, scan step: 0.02 V), V_{oc} variation with light intensity text, dark J-V curves text and stability text. In situ UV spectra was recorded by a QE-Pro (Ocean Optics) spectrometer with an RPB-785 optical probe located ca. 8 mm above the substrate. The EQE of the PSCs was measured using an Enlitech QE-R system. Photoluminescence (PL) and time-resolved PL (TRPL) measurements of the perovskite films were made using a PicoQuant FluoTime 300 with 510 nm laser excitation. FTIR profiles were obtained using a Bruker Vertex 70. The Mott-Schottky curves of PSCs were measured in the dark using a ModuLab XM CHAS08 electrochemical work station in a voltage range of –0.1 to 1.5 V with a scan step of 10 mV. EIS was measured in the dark using a ModuLab XM CHAS08 with a frequency range of 0.1 Hz to 100 MHz. Atomic force microscopy (AFM) images were acquired on a Bruker Dimension Icon system. Young's modulus was obtained through a Dimension ICON SPM system. Scanning electron microscopy (SEM) images were

measured by a Hitachi SU8020, with a secondary electron detector at an emission current of 10 μA and an accelerating voltage of 3 keV. The peeling method of perovskite film is as follows: A small amount of UV glue was dropped onto the perovskite thin film, and another piece of FTO was placed on the glue. After irradiating with a UV lamp for 30 s, the two were separated to peel off the perovskite thin film.

Figures and tables:

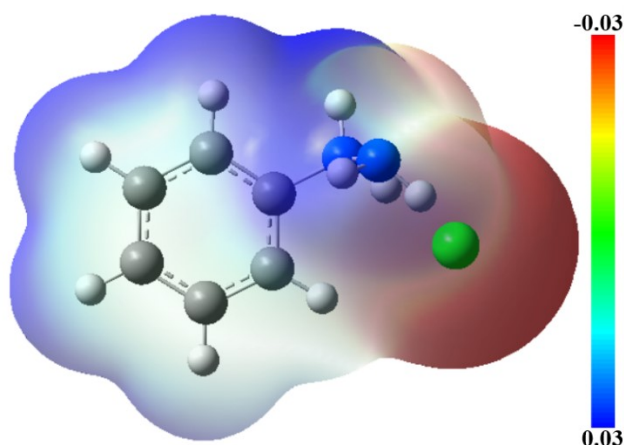


Figure S1 The electron cloud density distribution of PC.

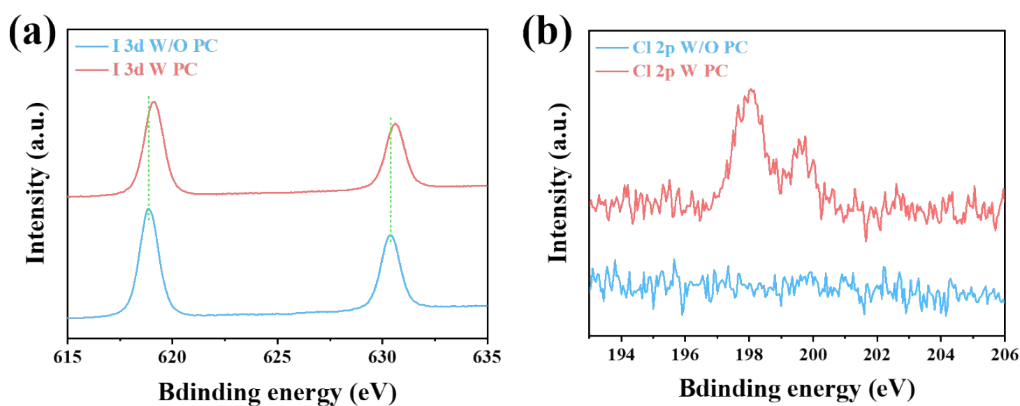


Figure S2 The XPS spectra of (a) I 3d and (b) Cl 2p of the perovskite film prepared without and with PC buried interface modification.

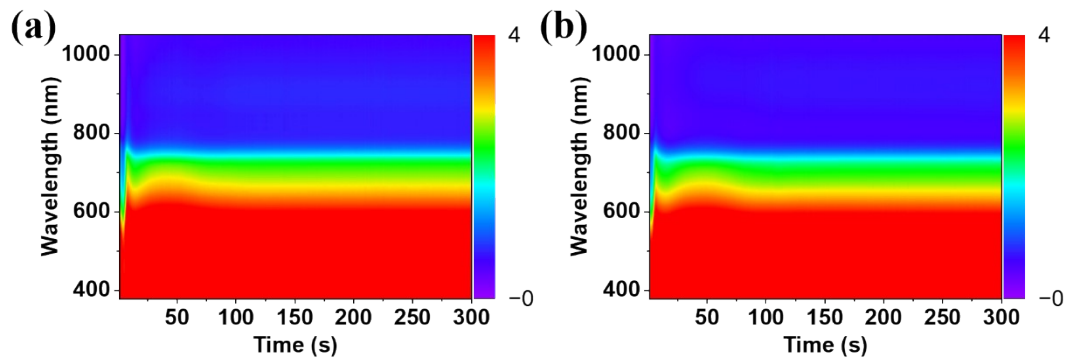


Figure S3 The in situ UV-vis results of the annealing process of perovskite films prepared (a) without and with (b) PC buried interface modification.

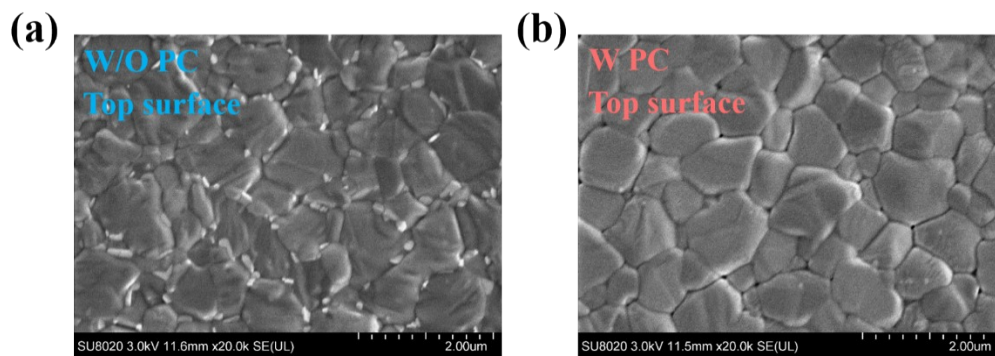


Figure S4 The SEM results of the top surface of perovskite films prepared (a) without and with (b) PC buried interface modification.

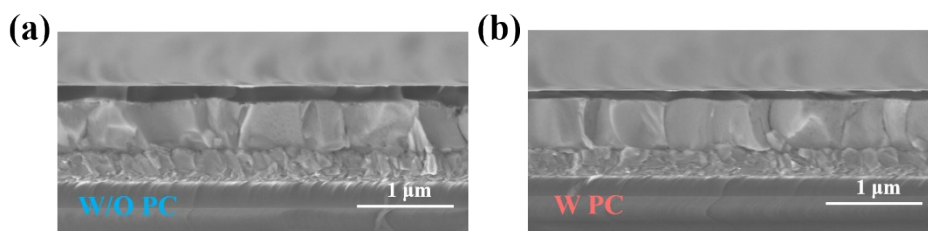


Figure S5 The SEM cross-sectional view of the device prepared (a) without and (b) with PC buried bottom modification material.

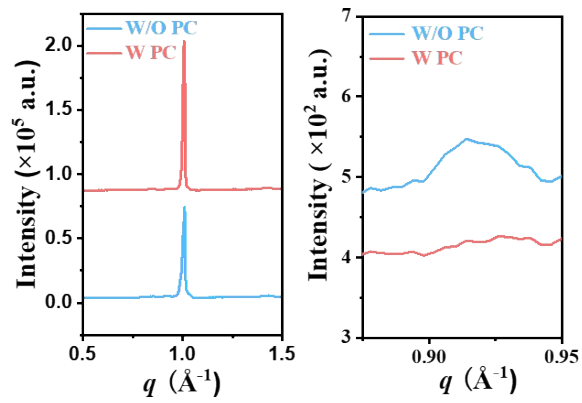


Figure S6 The curve of (a) the (100) crystal plane ($q = 1.0$) and (b) the PbI_2 peak ($q=0.92$) of the perovskite film prepared without and with PC buried interface modification.

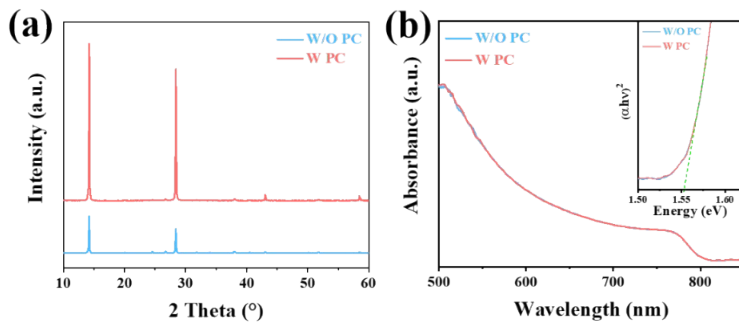


Figure S7 (a) The XRD results and (b) UV-vis results of the perovskite film prepared without and with PC buried interface modification.

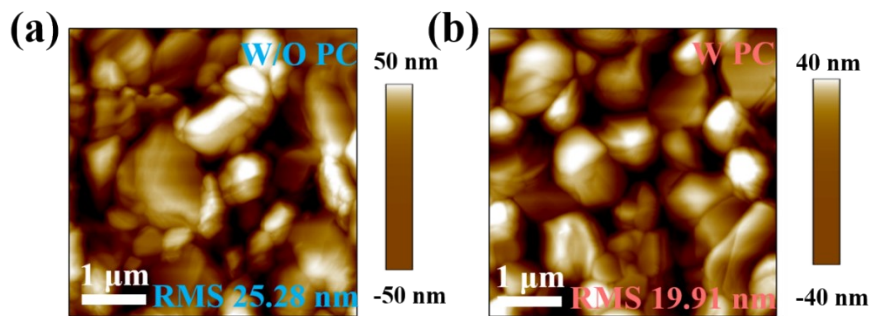


Figure S8 The AFM results of the perovskite film prepared (a) without and (b) with PC buried interface modification.

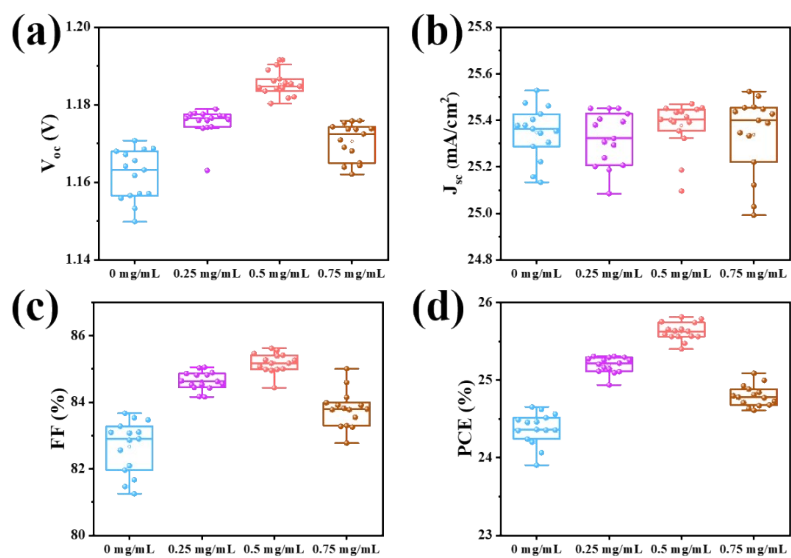


Figure S9 The distribution of V_{oc} , J_{sc} , FF and PCE for the PSCs prepared with different concentrations PC buried interface modification.

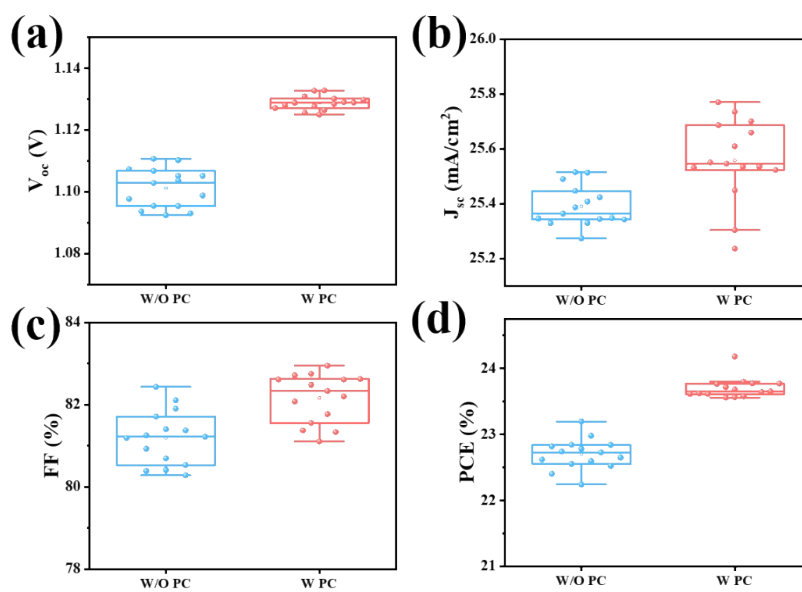


Figure S10 The distribution of V_{oc} , J_{sc} , FF and PCE for the FAPbI₃ PSCs prepared without and with PC buried interface modification.

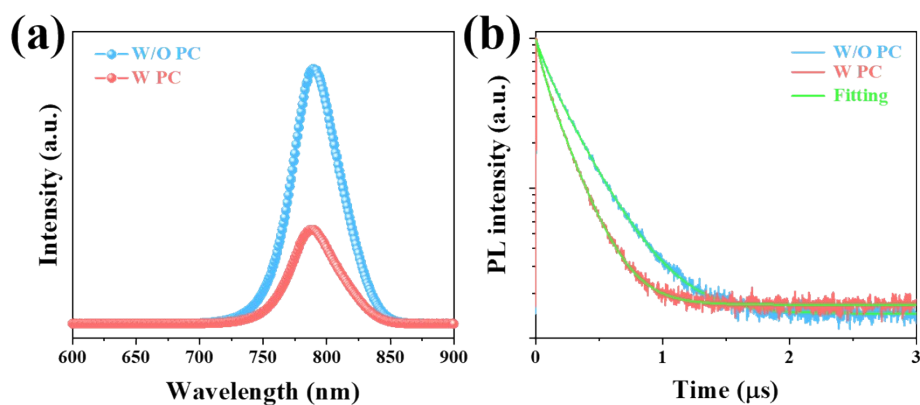


Figure S11 The (a) PL and (b) TRPL results perovskite films prepared with and without PC buried bottom modification materials.

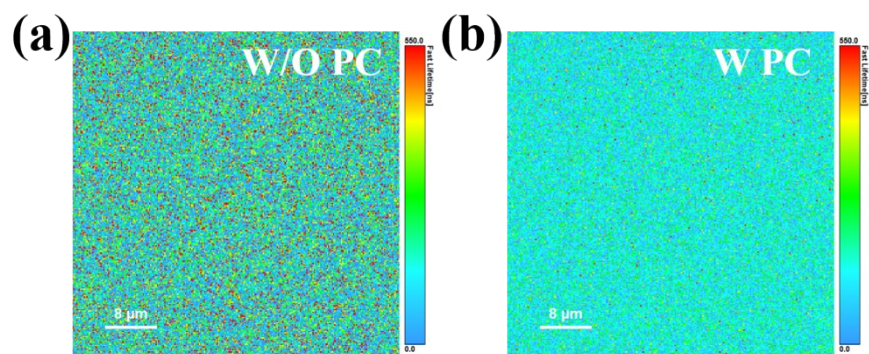


Figure S12 The PL mapping results perovskite films prepared (a) without and (b) with PC buried bottom modification materials.

Table S1 The parameters of PSCs prepared with different concentrations PC buried interface modification.

Simple	V_{oc} (V)	J_{sc} (mA/cm ²)	FF (%)	PCE (%)
0 mg/mL	1.162 ± 0.006	25.34 ± 0.11	82.66 ± 0.79	24.37 ± 0.20
0.25 mg/mL	1.176 ± 0.004	25.32 ± 0.12	84.64 ± 0.28	25.20 ± 0.10
0.5 mg/mL	1.185 ± 0.003	25.38 ± 0.10	85.18 ± 0.30	25.62 ± 0.12
0.75 mg/mL	1.171 ± 0.005	25.34 ± 0.17	83.79 ± 0.55	24.79 ± 0.14

Table S2 The parameters of FAPbI₃ PSCs prepared without and with PC buried interface modification.

Simple	V _{oc} (V)	J _{sc} (mA/cm ²)	FF (%)	PCE (%)
W/O PC	1.101 ± 0.006	25.39 ± 0.07	81.19 ± 0.66	22.70 ± 0.23
W CP	1.129 ± 0.002	25.54 ± 0.15	82.17 ± 0.60	23.70 ± 0.15

Table 3 The fitting data of the Nyquist plots for PSCs prepared without and with PC buried bottom modification.

Simple	R _s (Ω)	R _{tr} (Ω)	R _{rec} (Ω)
W/O PC	168.8	665.7	1548
W PC	143.7	556.6	2136

Table S4 The fitting data of TRPL spectra for perovskite films prepared without and with PC buried interface modification.

Simple	A ₁	τ ₁ (μs)	A ₂	τ ₂ (μs)	τ _{ave} (μs)
Top surface W/O PC	7114.40	0.27	2167.00	0.08	0.25
Top surface W PC	7019.40	0.18	2465.00	0.04	0.17
Buried surface W/O PC	5527.70	0.47	3008.30	0.21	0.42
Buried surface W PC	5800.30	1.10	2124.10	0.30	1.02

References

- [1] Y. Cao, N. Yan, M. Wang, D. Qi, J. Zhang, X. Chen, R. Qin, F. Xiao, G. Zhao, Y. Liu, X. Cai, K. Zhao, S. Liu, J. Feng, *Angew. Chem., Int. Ed.*, **2024**, 63, e202404401.