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A boosting carrier transfer passivation layer for achieving efficient perovskite solar cells

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Experimental Section

Materials

Formamidinium iodide (FAI, 99.5%), methylammonium chloride (MACl, 99.5%), lead iodide (PbI_2, 99.99%), 2,2',7,7'-Tetrakis (N,N-di-p-methoxyphenylamino)-9,9'-spirobifluorene (spiro-OMeTAD, 99.8%), 4-terttertbutylpyridine (TBP, 96%), lithium bis(trifluoromethanesulphonyl)imide (Li-TFSI, 99%) and tris(2-(1H-pyrazol-1-yl)-4-tert-butylpyridine)-cobalt(III) Tris(bis(trifluoromethylsulfonyl)imide)) (Co-TFSI, 99%) were purchased from Xi'an Polymer Light Technology Corporation. Tin oxide colloidal dispersion (SnO_2, 15% in water) was purchased from Alfa Aesar. Chlorobenzene (CB, 99.5%), N,N-dimethylformamide (DMF, 99.9%), dimethyl sulfoxide (DMSO, 99.9%), and acetonitrile (ACN, 99.9%) were purchased from Sigma-Aldrich. Isopropanol (IPA, 99.9%), 4-methoxyphenylethylammonium iodide (CH_3O-PEAI, 99.5%) and laser-etched indium-doped SnO_2 on glass (ITO, 7-9 Ω) were purchased from Advances Election Technology CO, LTD. 2,3,5,6-tetrafluoro-7,7,8,8-tetracyanoquino-dimethane (F4-TCNQ, 97%) was purchased from Macklin.

Devices Fabrication

The transparent conductive ITO was washed sequentially with deionized water, ethanol, acetone, and isopropanol in an ultrasonic bath. ITO substrates were blown dry with nitrogen and treated in an ultraviolet ozone system for 30 min. The electron transport layers were prepared by spin-coating a SnO_2 solution (1:6, V/V, commercial

SnO_2 colloidal dispersion in ultra-pure water) onto the ITO substrate at 3000 rpm for 30 s and annealing at 150 °C for 30 min. For preparing $FAPbI_3$ solution, 1.4 M FAI, 1.47 M PbI_2, and 0.5 M MACl powder were mixed in DMF and DMSO (8:1, V/V). The perovskite solution was spin-coated at 1000 rpm for 5 s and 5000 rpm for 30 s. A 0.1 ml CB was dripped on the film 15 s after the start of spin coating. Then the wet film was annealed at 150 °C for 10 min in the air (30%–40% RH). For surface treatment, CH_3O-PEAI solution (15 mM in IPA with or without 0.5 mg/mL F4-TCNQ) was spin-coated on the perovskite film at 5000 rpm for 30 s and annealed at 100 °C for 5 min. Spiro-OMeTAD solution was obtained by mixing 90 mg spiro-OMeTAD powder, 39 μL TBP, 23 μL Li-TFSI solution (520 mg Li-TFSI in 1 mL ACN) and 10 μL Co-TFSI solution (375 mg Li-TFSI in 1 mL ACN) in 1 mL CB. The hole transport layers were prepared by spin-coating spiro-OMeTAD solution on the perovskite layer at 3000 rpm for 30 s. Finally, an 80 nm Ag electrode was deposited on the hole transport layer by thermal evaporation under 3.5×10^{-5} Pa.

Characterization

The current density–voltage (*J–V*) curves were measured by a Keithley 2400 Source-Meter under AM1.5G irradiation, which was calibrated using a Standard Si solar cell (Newport 91150 V). The monochromatic incident photon-to-electron conversion efficiency (IPCE) curves were measured by an IPCE measurement system. The x-ray diffraction (XRD) was measured by an X-ray diffractometer with a Cu Ka radiation source. The atomic force microscope (AFM) was measured by an atomic force microscope (Veeco). The scanning electron microscope (SEM) was measured by a scanning electron microscope (S4800, Hitachi). The absorption spectra and optical transmission spectra were measured by an ultraviolet-visible (UV-vis) spectrophotometer (UV-3600, Shimadzu). The steady-state photoluminescence (PL) spectra and the time-resolved photoluminescence (TRPL) spectra were measured by a fluorescence spectrometer (FLS980, Edinburgh). The space charge limited current (SCLC) curves were measured under dark conditions by a Keithley 2400 Source-Meter. The X-ray photoelectron spectroscopy (XPS) was measured by Thermo Scientific K-

Alpha XPS System. The ultraviolet photoelectron spectroscopy (UPS) was measured by PHI5000 Versa Probe III (Scanning ESCA Microprobe) SCA (Spherical Analyzer). The contact angles were measured by a measurement system (JY-82B Kruss DSA).

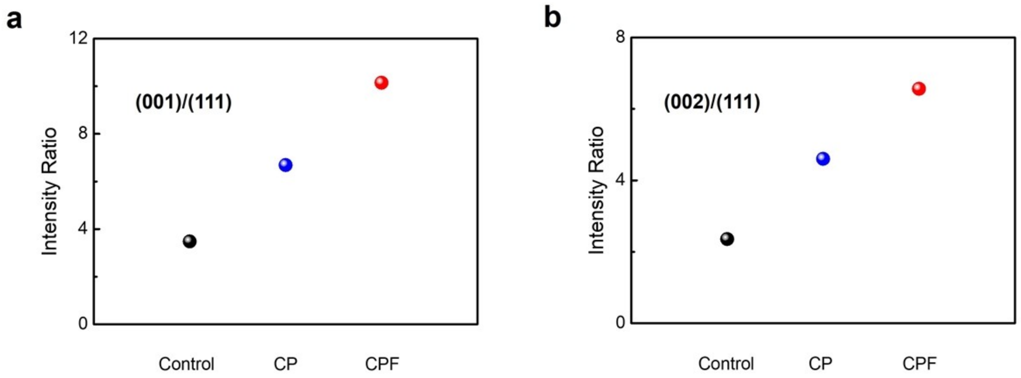

Fig. S1. The peak intensity ratio of perovskite films with different surface treatments. (a) (001)/(111). (b) (002)/(111).

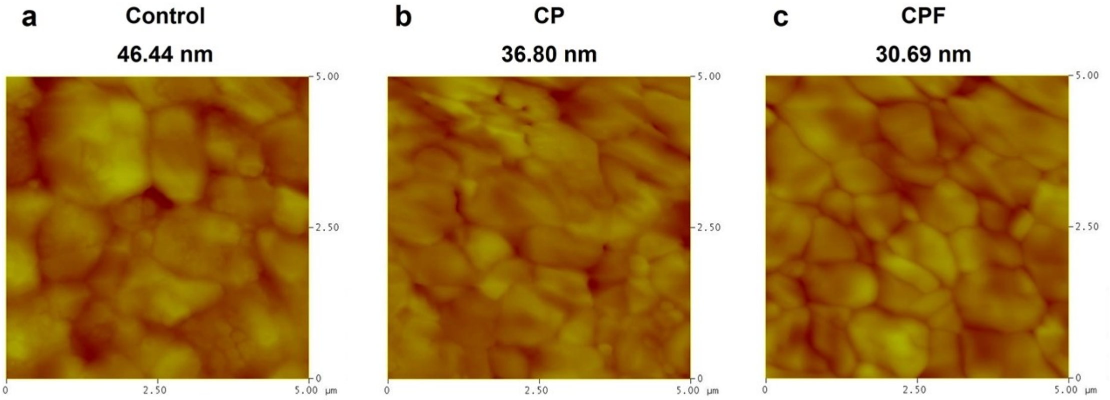

Fig. S2. AFM images of perovskite films with different surface treatments. (a) Control. (b) CP. (c) CPF.

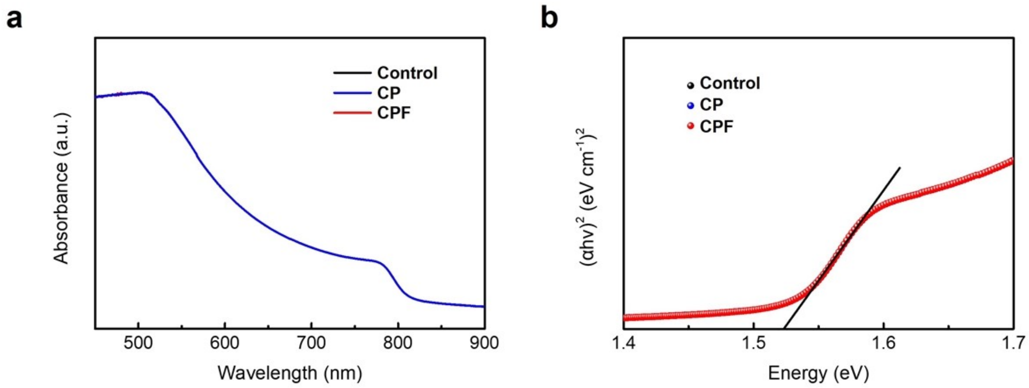

Fig. S3. (a) UV-vis absorption spectra. (b) Tauc plots of perovskite films with different surface treatments.

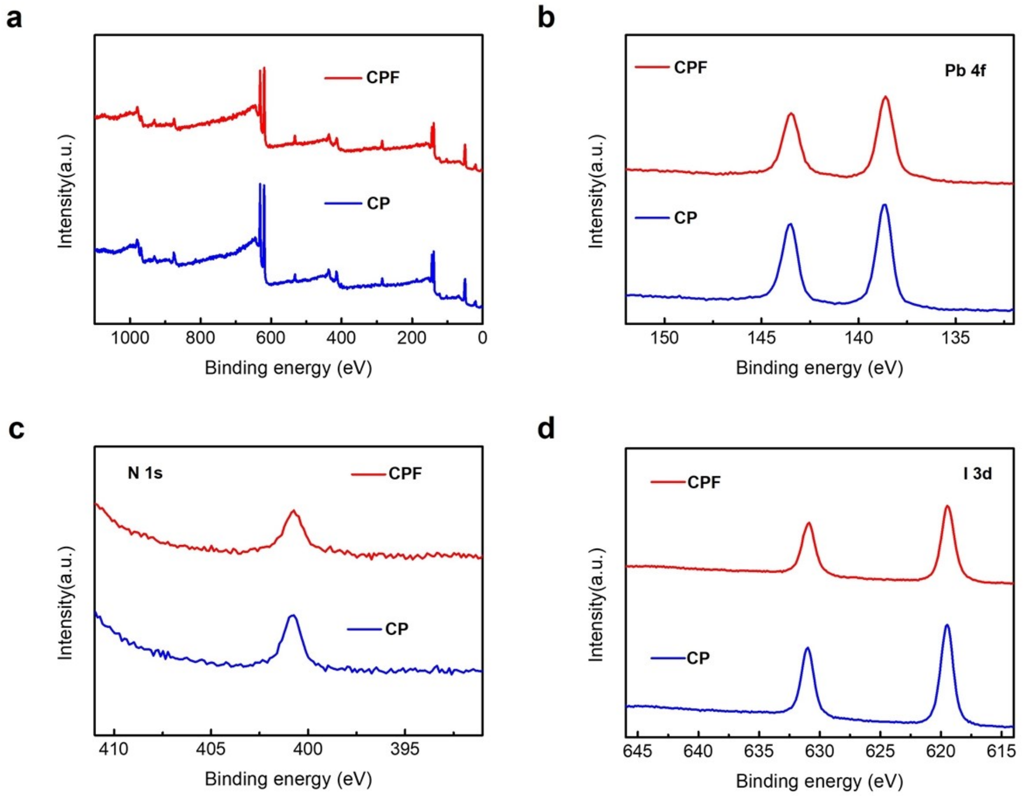

Fig. S4. (a) XPS spectra. (b) XPS of Pb 4f core-level spectra. (c) XPS of N 1s core-level spectra. (d) XPS of I 3d core-level spectra.

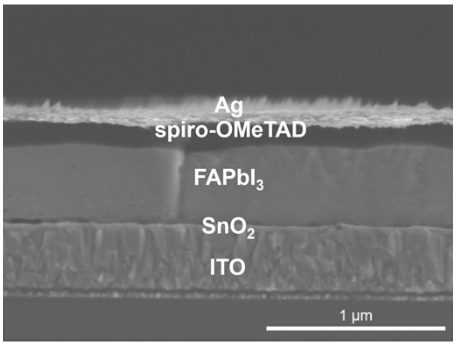

Fig. S5. Cross-sectional SEM image of perovskite solar cell.

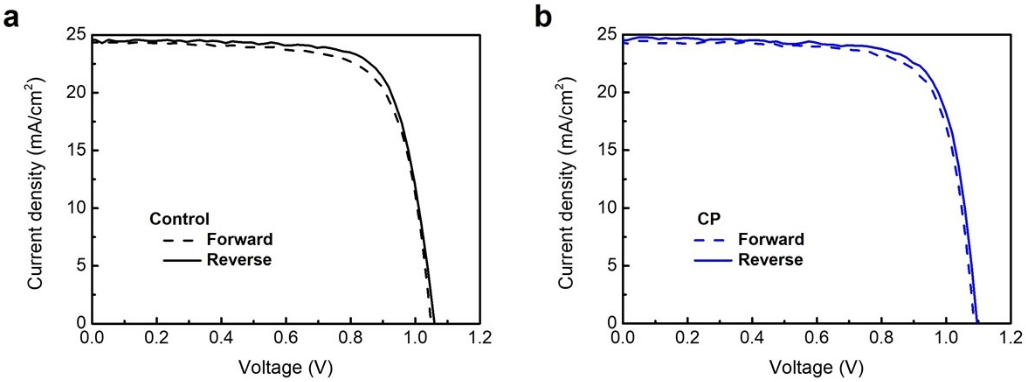

Fig. S6. The *J*–*V* curves of devices. (a) Control. (b) CP.

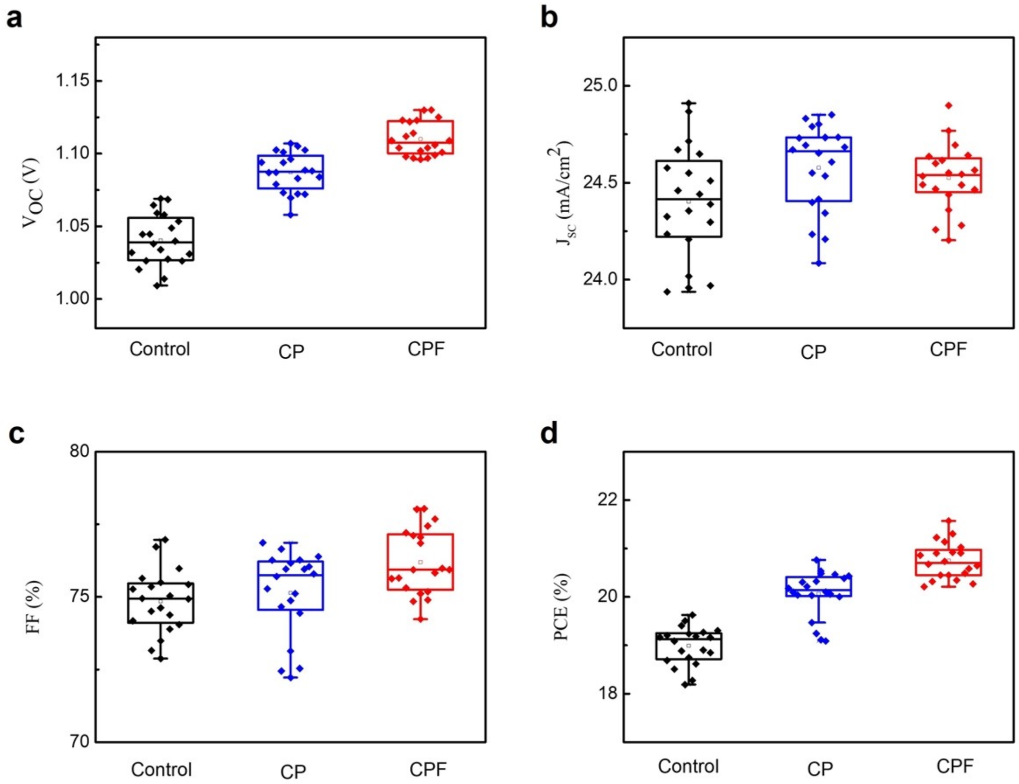

Fig. S7. Device parameters of devices. (a)V_{OC}. (b) J_{SC}. (c) FF. (d) PCE.

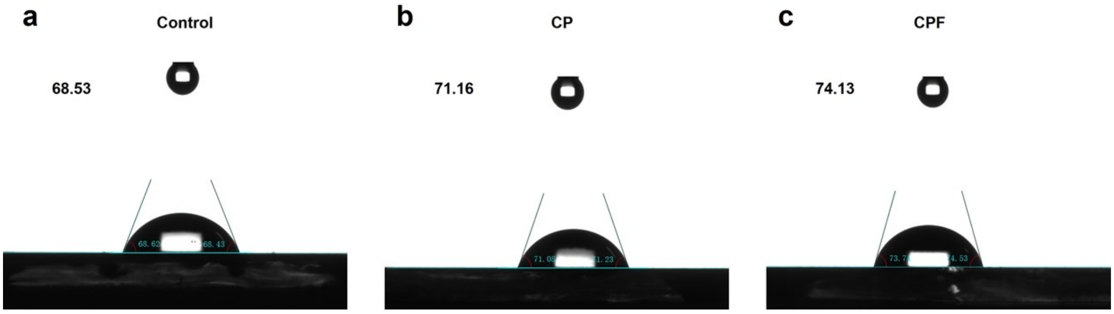

Fig. S8. Contact angel of perovskite films. (a) Control. (b) CP. (c) CPF.

Table S1. Device parameters of *J*–*V* curves.

	Scan direction	V_{OC} (V)	J_{SC} (mA/cm^2)	**FF (%)**	**PCE (%)**
Control	Forward	1.049	24.36	72.88	18.61
	Reverse	1.059	24.58	74.50	19.40
CP	Forward	1.087	24.28	75.14	19.81
	Reverse	1.096	24.54	76.12	20.48
CPF	Forward	1.124	24.55	75.83	20.92
	Reverse	1.130	24.64	77.42	21.57