Electronic Supplementary Information

2-Amino-5-chlorobenzophenone passivating perovskite films using multiple functional groups towards high-performance solar cells

Yuxin Wu [a], Chenrui Lu [a], Fei Gao [a], Yuanrui Li [a], Bonan Shi [a], Xuediao Cai [b*], Fei Yang [a], Junqi Zhang [a], and Shengzhong (Frank) Liu [a,c*]*

[a] *Key Laboratory of Applied Surface and Colloid Chemistry, National Ministry of Education; Shaanxi Key Laboratory for Advanced Energy Devices; Shaanxi Engineering Lab for Advanced Energy Technology, School of Materials Science and Engineering, Shaanxi Normal University, Xi'an 710119, China.*

[b] *School of Chemistry & Chemical Engineering, Shaanxi Normal University, Xi'an 710119, China.*

[c] *Dalian National Laboratory for Clean Energy; Dalian Institute of Chemical Physics, Chinese Academy of Sciences, Dalian 116023, China*

[*]Corresponding author.

E-mail address: feigao@snnu.edu.cn (F.Gao), xdcai@snnu.edu.cn (X.D.Cai), liusz@snnu.edu.cn (S.F.Liu)

Films Preparation and Solar Cell Fabrication

Fluorine-doped tin oxide (FTO) glass was immersed in glass cleaning solution (The ratio of glass cleaning agent and ultrapure water is 1:100) and ultrasonically cleaned for 30 min, followed by ultrasonic cleaning with ultrapure water twice, each time for 30 min. The cleaned substrate was dried by nitrogen flow. TiO_2 as the electron transport layer was deposited on the FTO glass substrate via chemical bath approach. First, the dried FTO glass substrate was treated in a UV-ozone cleaner for 15 min, and then soaked in 0.2 M $TiCl_4$ aqueous solution at 70℃ for 1 h. Finally, the TiO_2/FTO substrate was washed with deionized water, dried with N_2 and annealed at 200℃ for 30 min, and then UV-ozone treated for 15 min to increase surface wettability. $(FAPbI_3)_{0.95}(MAPbBr_3)_{0.05}$ perovskite precursor solution was prepared by dissolving a mixture of 1.23 mmol PbI_2, 1.23 mmol FAI, 0.07 mmol $PbBr_2$, 0.07 mmol MABr, and 4.5 mmol MACl in 1 mL of solvent of DMF and DMSO (9:1, v/v), and stirred at 25℃ for 6 h. ACB was dissolved in chlorobenzene (CB) with different concentrations (3.0, 6.0, and 9.0/mg mL). 90 mg Spiro-OMeTAD was dissolved in 1 mL CB mixed with 22 μL Li-TFSI (520mg/mL in acetonitrile) and 36 μL 4-tert-butylpyridine. 50 μL perovskite precursor solution was spin-coated onto the TiO_2/FTO glass substrates by one-step spin coating (5000 rpm for 50 s). 200 μL CB was dropped onto the surface at 10 s after the start of spin coating with CB as the anti-solvent, and then the films were annealed at 150℃ for 20 min. The ACB solution was spin-coated onto the annealed perovskite films at 4000 rpm for 30 s and then annealed at 90℃ for 10 min. The Spiro-OMeTAD was deposited on the perovskite films as hole transport layer (HTL) by spin coating (5000 rpm for 30 s). Finally, 80 nm thick gold electrode was deposited on the surface of HTL by vacuum thermal evaporation. The active area of each PSC is 0.09 cm^2.

Characterization and Measurement

X-ray diffraction (XRD) patterns of perovskite films were obtained by a Bruker D8 GADDS diffractometer using Cu Kα radiation (λ = 0. 154 nm). Scanning electron microscopy (FESEM, HITACHI, SU-8020) were applied to acquire the surface morphology of perovskite films and cross-section structures of PSCs. Atomic force microscopy (AFM) images were obtained by a Bruker Dimension Icon instrument. Absorption spectra of the perovskite films were measured via UV-Vis spectrometer. Steady-state photoluminescence (PL) and time-resolved PL (TRPL) spectra were measured using Edinburgh Instruments FLS920 fluorescence spectrometer. The space-charge limited current (SCLC) measurements and dark J-V test were performed through Keithley 2400 source. Mott-Schottky analysis and the electrochemical impedance spectroscopy (EIS) were measured in dark using an electrochemical workstation (AMETEK-Modulab XM). Kelvin probe force microscopy (KPFM) images were collected by BRUKER-NanoScope. X-ray photoelectron spectroscopy (XPS, ESCALAB250Xi, Thermo Fisher Scientific), Fourier transform infrared spectroscopy (FTIR, Bruker Vertex 70), and 1H and 13C nuclear magnetic resonance (NMR, Bruker AVANCE III 600) were used to study the interaction between ACB and perovskite. The J-V curves of the PSCs were measured with a Keithley 2400 under AM 1.5 G illumination under 100 mW/cm^2 conditions. External quantum efficiency (EQE) spectra were obtained through a Q Test Station 500TI system (Crowntech, Inc. USA) and the intensity of monochromatic light is calibrated by a reference silicon photodiode. Water contact angles of perovskite films were measured using a DataPhysics OCA 20.

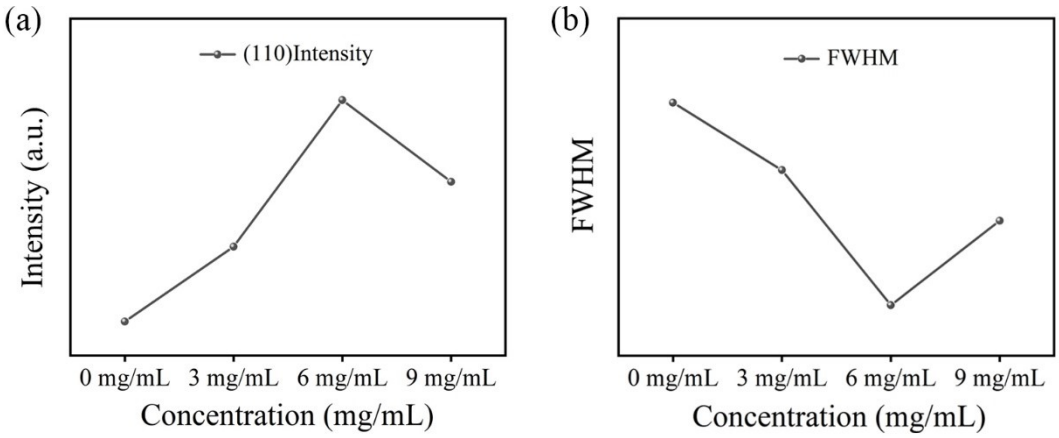

Figure S1 (a) Intensity and (b) full width at half maximum of the (110) peak of perovskite films treated with different concentrations of ACB.

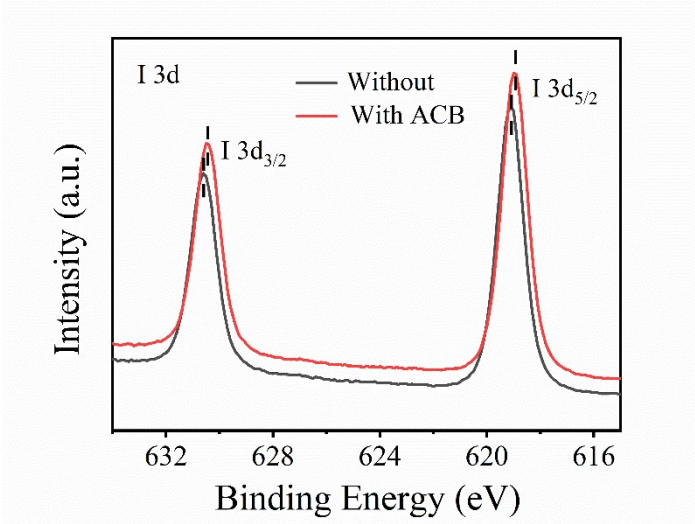

Figure S2 XPS spectra of I 3d of the untreated and ACB treated perovskite.

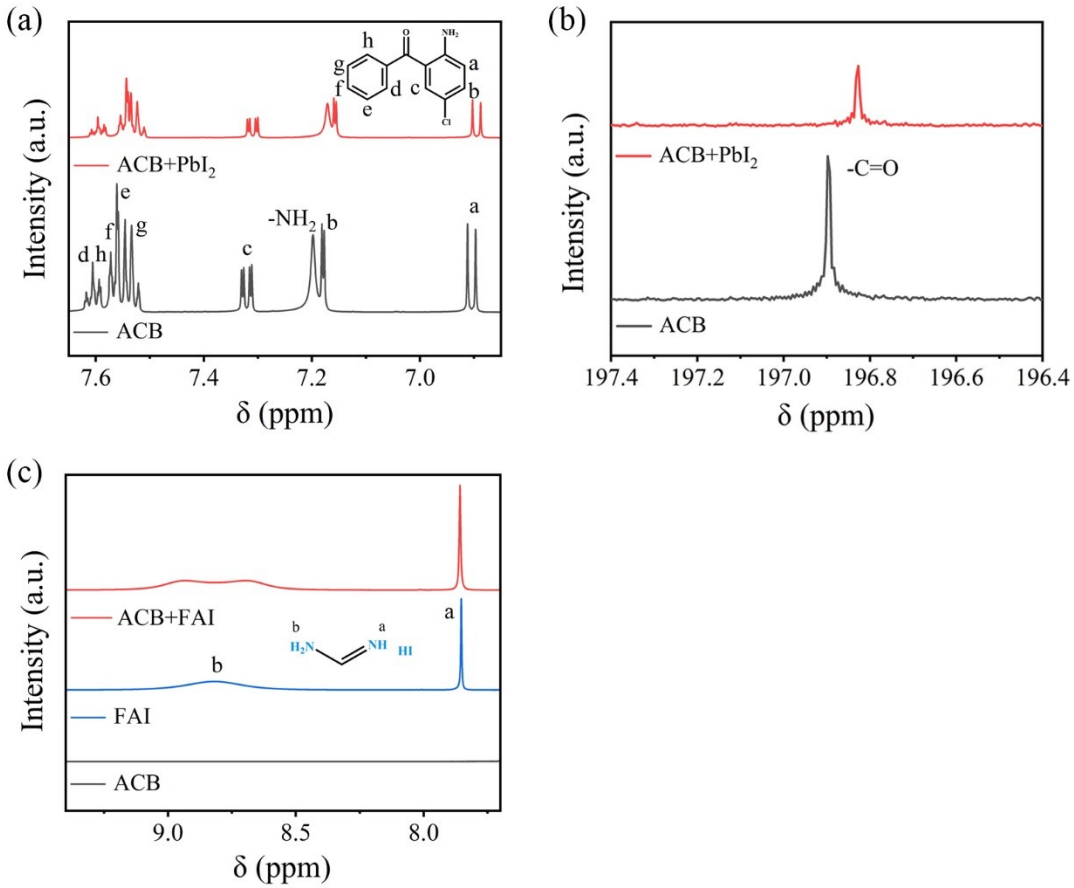

Figure S3 ^{1}H NMR spectra of (a) ACB and ACB+PbI_2. (b) ^{13}C NMR spectra of ACB and ACB+PbI_2. (c) ^{1}H NMR spectra of ACB and ACB+FAI.

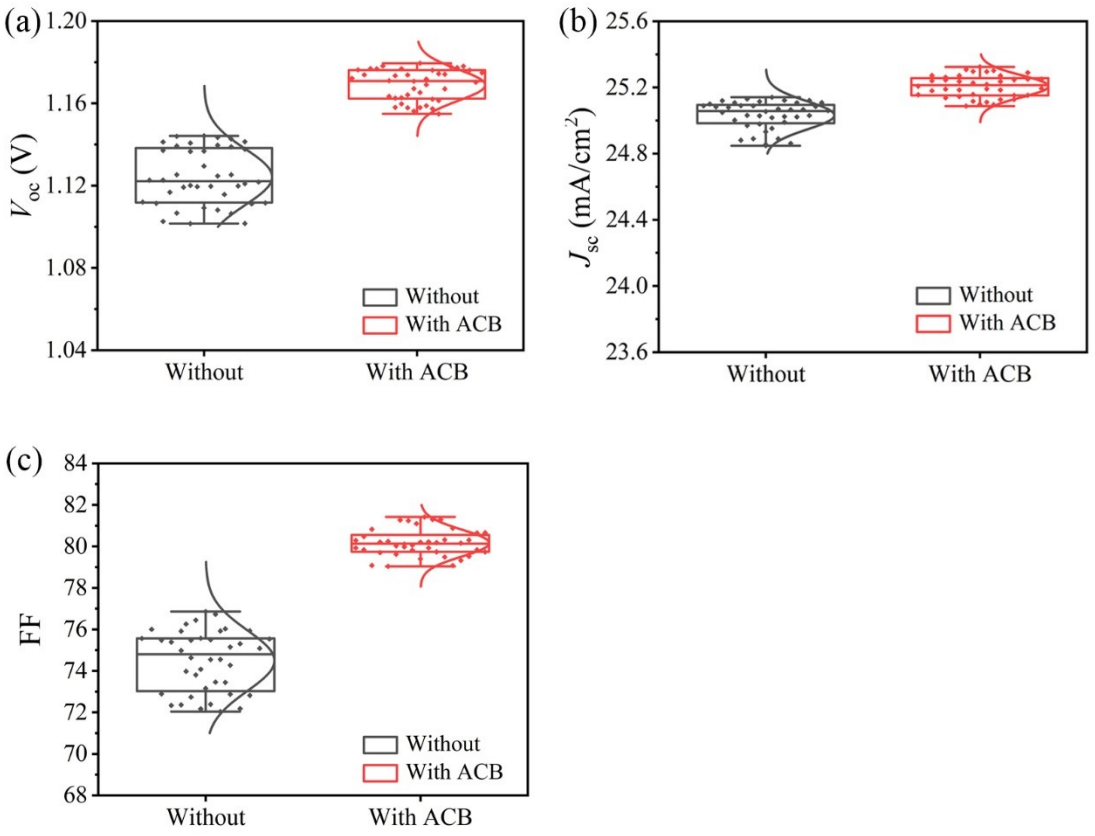

Figure S4 Statistical distributions of (a) V_{oc}, (b) J_{sc} and (c) FF of the PSCs without and with ACB treatment.

Table S1 Fitted values of τ_1, τ_2, A_1, A_2 and calculated τ_{ave} for TRPL of perovskite films without and with ACB treatment.

	τ_{ave} (ns)	τ_1 (ns)	A_1 (%)	τ_2 (ns)	A_2 (%)
Without	290.57	164.92	3351.1	317.19	1365.4
6 mg/mL	419.49	171.2	3545.9	446.45	1003.8

Table S2 Photovoltaic parameters of PSCs treated with different concentrations of ACB.

Concentration	V_{oc} (V)	J_{sc} (mA/cm^2)	FF (%)	PCE (%)
0 mg/mL	1.14	25.14	76.86	22.11
3 mg/mL	1.16	25.30	78.71	23.07
6 mg/mL	1.18	25.32	81.42	24.32
9 mg/mL	1.17	25.30	80.11	23.80

Table S3 Photovoltaic parameters of PSC without ACB treatment in forward and reverse scanning.

Without	V_{oc} (V)	J_{sc} (mA/cm^2)	FF (%)	PCE (%)	H-index
Reverse scan	1.14	25.14	76.86	22.11	0.064
Forward scan	1.14	25.05	72.71	20.74	

Table S4 Photovoltaic parameters of ACB treated PSC in forward and reverse scanning.

Without	V_{oc} (V)	J_{sc} (mA/cm^2)	FF (%)	PCE (%)	H-index
Reverse scan	1.18	25.32	81.42	24.32	0.025
Forward scan	1.17	25.31	79.99	23.72	

Table S5 Fitted parameters of PSCs without and with ACB treatment from Nyquist plots.

	R_s (Ω)	R_{tr} (Ω)	C_{tr} (F)	R_{rec} (Ω)	C_{rec} (F)
Without	19.92	21.26	6.213×10^{-8}	2281	9.479×10^{-9}
6 mg/mL	15.33	19.25	9.581×10^{-8}	3617	1.041×10^{-8}