

Supporting Information

Ionic Liquid-regulated PbI₂ Layer and Defect Passivation for Efficient Perovskite Solar Cells

Experimental Section

Materials: ITO glass substrates (sheet resistance of ca. 9 Ω sq⁻¹) were purchased from OPVTECH Inc. PbI₂ (99.8%), CsI (99.99%), 4-tert-butylpyridine (tBP, 96%) and bis(trifluoromethane) sulfonimide lithium salt (Li-TFSI, 99%), Ionic liquids (ILs) formamidinium acetate (FAAc) was supplied from Sigma-Aldrich. Methylammonium chloride (MACl), methylammonium bromide (MABr) and Formamidinium Iodide (FAI) were purchased from Xi'an Polymer Light Technology Crop. Spiro-OMeTAD (purity. 99.5%) was purchased Feiming Science and Technology Co., Ltd. IPA, DMF and DMSO were purchased from TCI. Tin (IV) oxide was purchased from Alfa Aesar. All materials were used as received without any further treatment.

Devices Fabrications: ITO substrates were sequentially rinsed by sonication in detergent, deionized (DI) water and iso-propanol for 15 min, respectively, and then dried under nitrogen gas. Cleaned ITO substrates were treated with ultraviolet-ozone for 30 min, followed by deposition of a SnO₂ electron transporting layer on the substrate by spin-coating the SnO₂ QDs precursor (1.3 M) at 4000 r min⁻¹ for 30 s, and subsequently annealed at 150 °C for 30 min on a hotplate. The perovskite layer was fabricated in the glovebox through a modified two-step sequential method according to

the literature. First, 1.4 M PbI_2 precursor was dissolved in 950 mL DMF and 50 μL CsI solution (1.5 M, 390 mg CsI was dissolved in 1 mL DMSO) , And add x mol FAAC to mix, x is the molar ratio of the ionic liquid to PbI_2 , x = 0, 0.5, 1,3. The above solution was then spin-coated on the SnO_2/ITO substrate at 1500 r min^{-1} for 30 s, and dried at $70 \text{ }^\circ\text{C}$ for 1 min. Thereafter, a mixture solution of FAI:MABr:MACl (60 mg: 6 mg: 6 mg in 1 mL IPA) was dropped on the PbI_2 film at 1500 r min^{-1} for 30 s. The as-cast perovskite film was annealed at $150 \text{ }^\circ\text{C}$ for 15 min. The spiro-OMeTAD solution was composed of 72.3 mg Spiro-OMeTAD, 30 μL TBP, and 35 μL Li-TFSI solution (260 mg in 1 mL acetonitrile) in 1 mL chlorobenzene, and then spin-coated on perovskite film at 4000 r min^{-1} for 30 s. Finally, a 100 nm Au electrode was deposited by thermal evaporation.

Characterization: The $J-V$ characteristics of the PSCs devices were carried out using IVS-KA6000 Enlitech sunlight simulator equipped with an AM 1.5 filter at 100 mW cm^{-2} and Keithley SMU source after correcting the light intensity with a standard calibration cell. The corresponding EQE spectrum was acquired in air by a QE-R system from Enli Technology Co. Ltd. Static PL from 600 nm to 900 nm and TRPL measurements of perovskite film (2000 ns) were recorded by Edinburgh FLSP1000 spectrophotometer equipped with an excitation source of 445 nm picosecond pulsed diode laser. Scanning electron microscopy (SEM) measurements of perovskite thin films were conducted to obtain the morphology on JSM-IT 800. X-ray diffractometer spectrum of perovskite film with scanning angle from 3 to 40° (2θ) were measured by Bruker D8 Advance. UV-vis absorption spectra of the perovskite films were obtained

by a UV-3600 Plus spectrophotometer. ^1H nuclear magnetic resonance spectroscopy (NMR) spectra were recorded on a Bruker 400 MHz spectrometer. X-ray photoelectron spectroscopy (XPS) of the perovskite films were carried out using the Thermo Kalpha XPS. Fourier-transform infrared spectra of thin films were recorded from the wavenumber 4000 to 500 cm^{-1} via Thermo Fisher Scientific Nicolet iS50. Atomic force microscope (AFM) characterizations were conducted on an OXFORD instruments MFP-3D Origin.

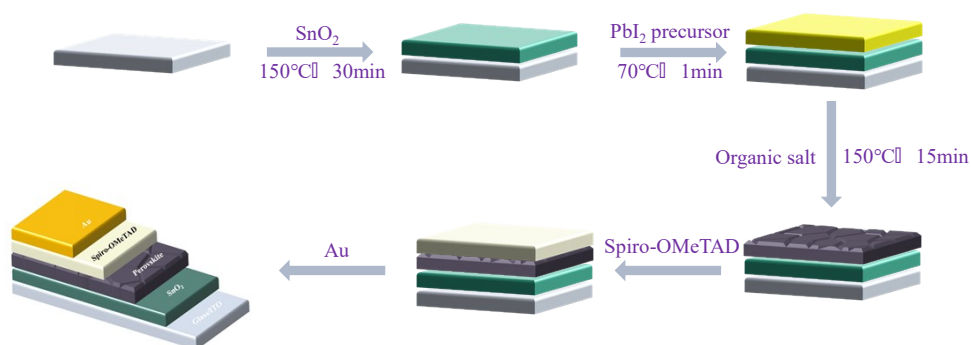


Figure S1. Schematic diagram of Two-step preparation of perovskite films.

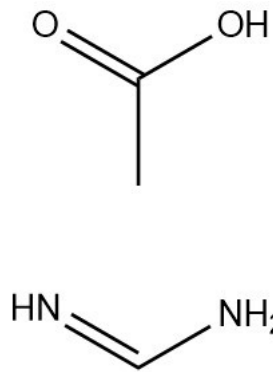


Figure S2. Schematic diagram of the structure of FFAc.

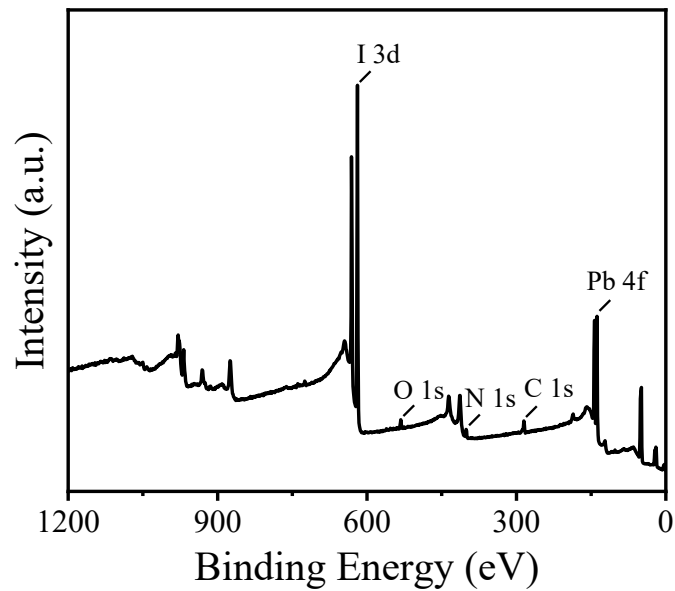


Figure S3. The full XPS spectrum of the control perovskite film.

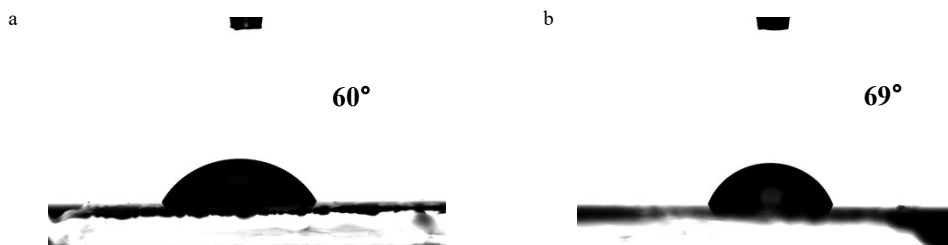


Figure S4. Contact angle to H₂O of the control (a) and FFAc-treated perovskite film(b).

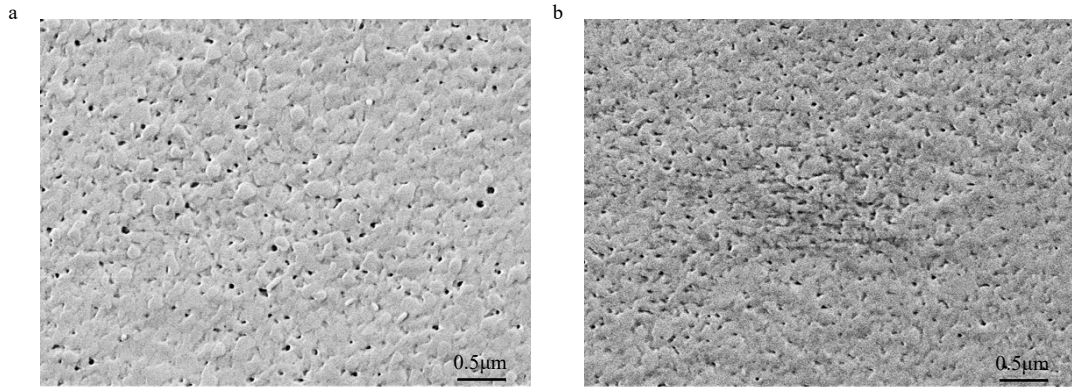


Figure S5. SEM image of the control (a) and FAAC-treated PbI_2 film(b).

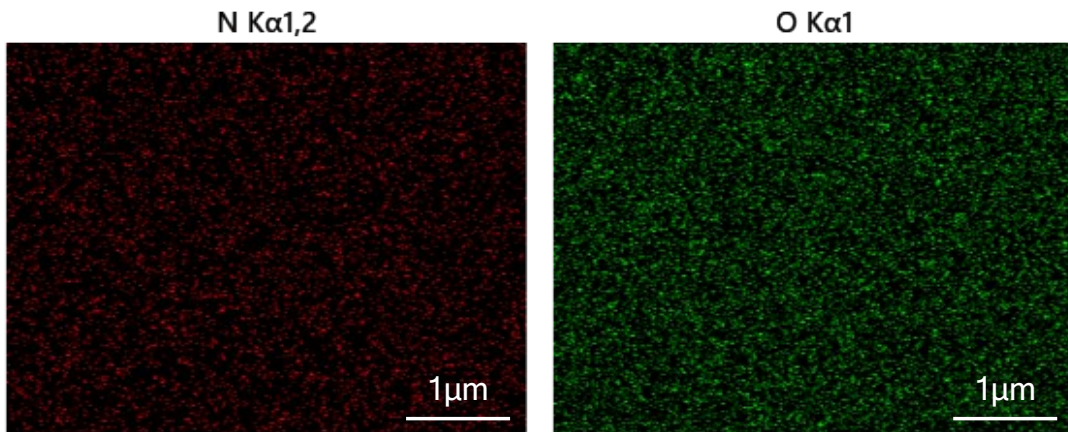


Figure S6. EDS images of N and O elements in FAAC-treated PbI_2 films.

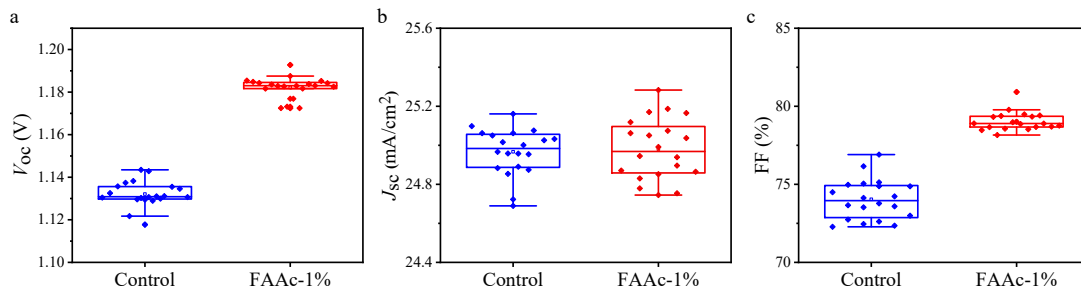


Figure S7. Statistical analysis of performance parameters for the control and FAAC-treated device acquired from 40 devices: (a) V_{oc} , (b) J_{sc} and (c) FF.

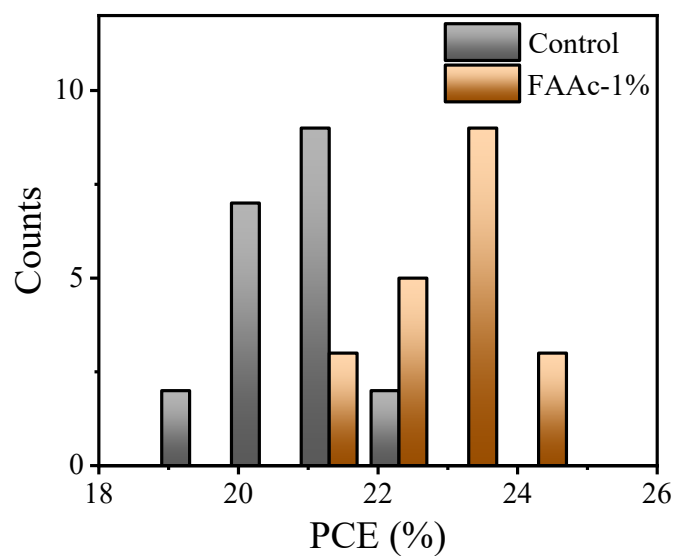


Figure S8. The PCEs histogram of the control and FFAc-treated devices.

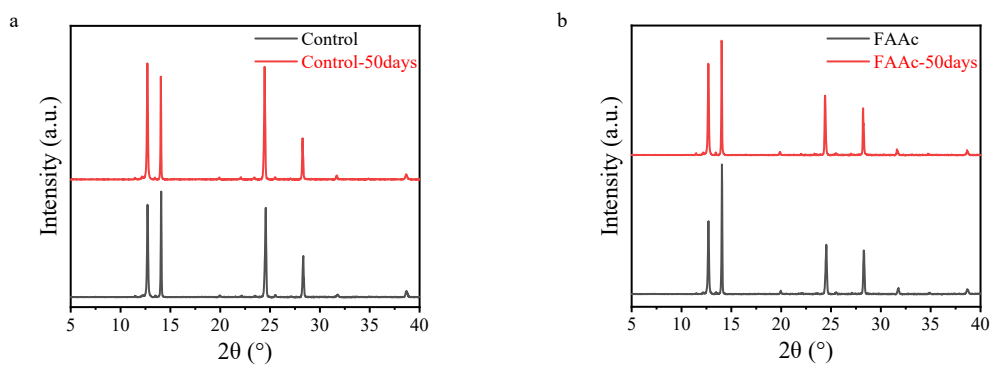


Figure S9. XRD patterns of (a)control and (b)FAAc-treated perovskite films at 25°C under 50% RH condition for fresh and 50 days, respectively.

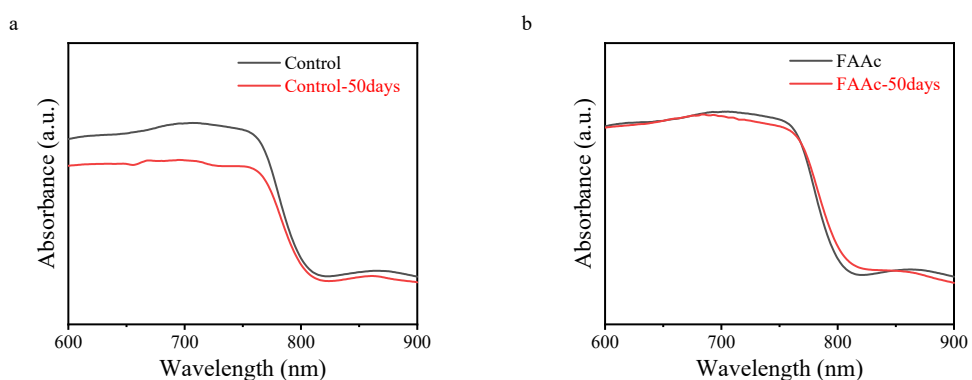


Figure S10. UV-Vis absorption spectra of (a) control and (b) FFAc-treated perovskite films at 25°C under 50% RH condition for fresh and 50 days, respectively.

Table S1. Photovoltaic parameters of different concentrations of FAAc-treated PSCs under AM 1.5G illumination at 100 mW cm⁻²

	V_{oc} [V]	J_{sc} (mA/cm ²)	FF (%)	PCE (%)
Control	1.143	25.16	76.91	22.12
FAAc-0.5%	1.188	25.34	78.52	23.64
FAAc-1%	1.193	25.28	80.92	24.41
FAAc-3%	1.184	25.12	76.75	22.83