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Supporting Information

Achieving a High-quality Acitive Film Through Surface Passivation to Enhance Stability of Inverted Perovskite Solar Cells

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Materials:

Lead(II) iodide (PbI2, 99.999%), methylamine hydrochloride (MACl), formamidinium iodide (FAI), cesium iodide (CsI), methylammonium bromide (MABr), lead(II) bromide ($PbBr_2$), phenyl-C61-butyric acid methyl ester (PC61BM, 98%), N4, N4'-bis(4-ethenylphenyl)-N4, N4'-di-1-naphthalenyl-[1,1'-biphenyl]-4,4'-diamine (VNPB), and Bathocuproine (BCP, >99%) were procured from Xi'an Polymer Light Technology Corp. N, N-dimethylformamide (>99.9%), dimethyl sulfoxide (>99.9%), anhydrous ethyl acetate, and isopropanol were obtained from Alfa Aesar Chemical Co., Ltd. The (Cyanomethyl)triphenylphosphonium Chloride (CTPC) was acquired from Aladdin Biochemical Technology Co., Ltd.

PSC Fabrication:

The FTO glass substrates were pre-patterned and subjected to a sequential ultrasonic cleaning process involving detergent, deionized water, acetone, and isopropanol. Subsequently, the substrates were passivated with UV-ozone for 15 minutes. A layer of NiOx was deposited onto the FTO glass substrate using an electron beam (E-beam) and subsequently passivated with oxygen plasma for 10 minutes. Prior to use, the substrates were transferred into a N_2-filled glovebox. A thin layer of VNPB (1 mg/ml in chlorobenzene) was spin-coated onto the substrates at 6000 rpm for 30 seconds, followed by annealing at 120 °C for 10 minutes. The perovskite precursor solution, with a concentration of 1.5 M, was formulated by blending CsI, FAI, MABr, PbI_2, and $PbBr_2$ in a mixed solvent of 1 ml DMF:DMSO (5:1/v:v), following the chemical composition of $(FA_{0.98}MA_{0.02})_{0.95}Cs_{0.05}Pb(I_{0.95}Br_{0.05})_3$. To enhance PSC performance, an additional 5 mol% of PbI2 was incorporated. Subsequently, 15.5 mol% of MACl was introduced into the precursor solution and stirred until fully dissolved. Prior to application, the precursor solutions were filtered through a 0.22 μm PTFE membrane. The resulting solutions were then spin-coated onto the VNPB layer at 5000 rpm for 35 s. For the glass/ITO/hole transporting layer (HTL) substrate, 60 μL of perovskite solution was first spin-coated at 1000 rpm for 10 s, followed by 5000 rpm for 50 s. At 10 s before the spin-coating process ended, 300 μL of EA was slowly dripped onto the film's center. The as-prepared perovskite films were annealed on a hotplate at 110 ℃ for 20 min. Subsequently, a PC61BM layer was spin-coated onto the perovskite film using a 20 mg/mL chlorobenzene solution at 2000 rpm for 30 s. A BCP solution (0.5 mg/mL in isopropanol) was then spin-coated onto the PC61BM layer at 4000 rpm for 30 s. Finally, Ag (~80 nm) was evaporated onto the films through a shadow mask under high vacuum (approximately Pa).

Surface Passivation Strategy:

Following the preparation of the initial perovskite films, a rotational coating of 100 μL of CTPC isopropanol solution was applied onto the perovskite substrate at 4000 rpm for 35 seconds. Subsequently, the substrate underwent annealing at 110°C for 5 minutes through a dynamic spinning drop step. Other processes followed the description outlined for the fabrication of control PSCs.

PSC Characterizations:

Current density-voltage (J-V) curves were measured in ambient air conditions utilizing a Keithley 2400 source under simulated AM1.5G illumination (100 mW/cm^2) provided by a xenon-lamp-based solar simulator, with the active area of the PSC is set at 0.09 cm^2. EQE measurements were conducted using a QTest Station 2000 ADI system (Crowntech, Inc.). Trap densities and carrier mobilities were assessed employing a Keithley 2400 source through the space-charge-limited current (SCLC) method. Capacitance-voltage (C-V) measurements were acquired utilizing a ModuLab XM CHAS 08, operating within a frequency range spanning from 0.1 Hz to 100 MHz in the absence of light.

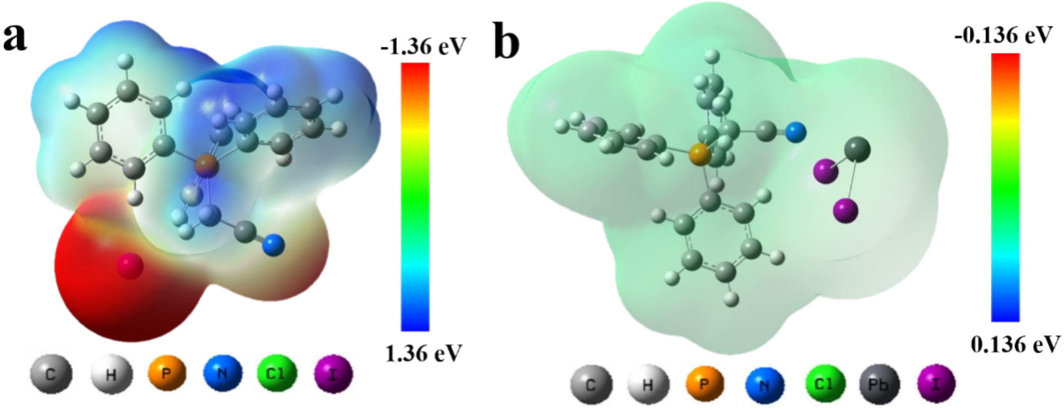

Figure S1 Electrostatic potential (ESP) of (a) CTPC with I$^-$ (b) CTPC with PbI$_2$

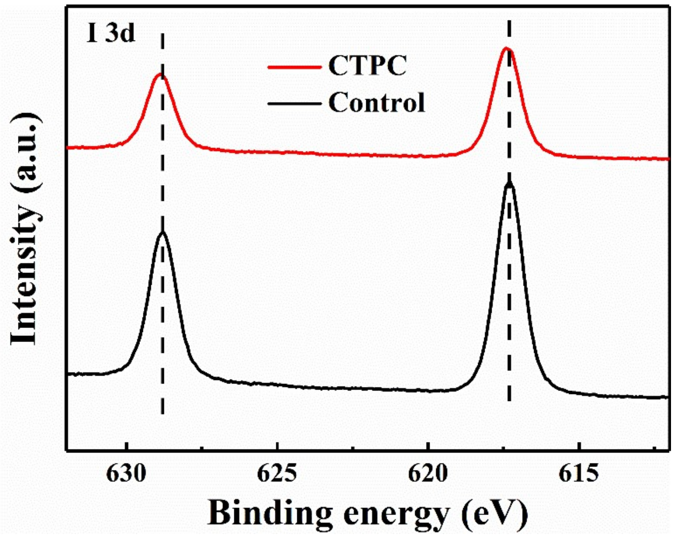

Figure S2 I 3d XPS spectra of the control perovskite and perovskite with CTPC films

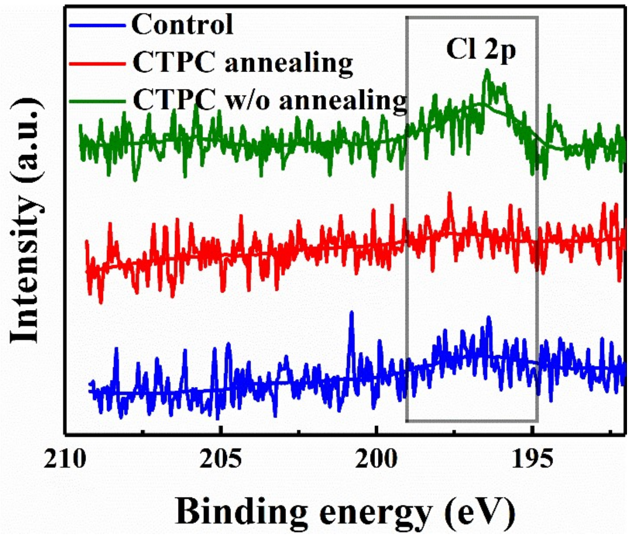

Figure S3 Cl 2p XPS spectra of the control perovskite and perovskite with CTPC films.

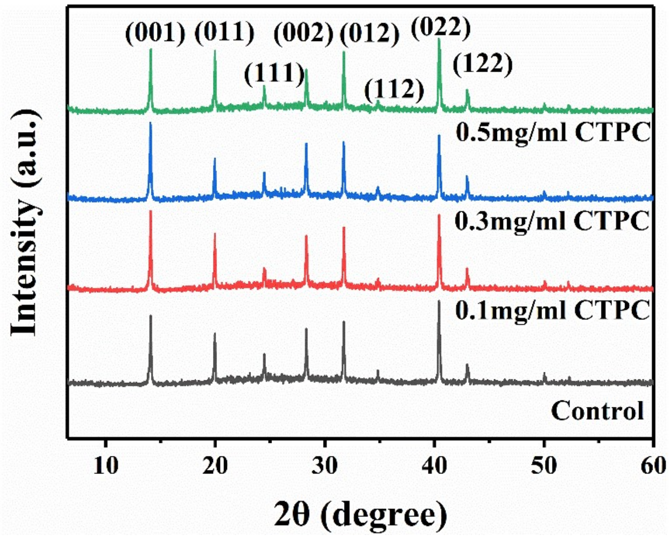

Figure S4 XRD characterization of perovskite films passivated with different concentrations of CTPC

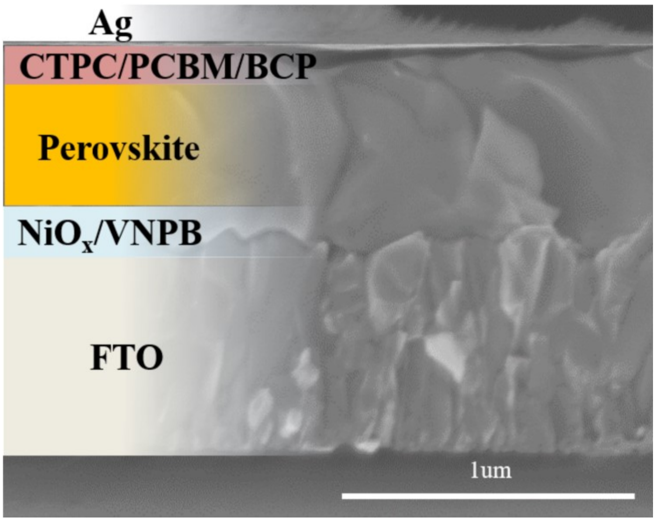

Figure S5 CTPC passivation treatment of PSCs with cross-sectional SEM image

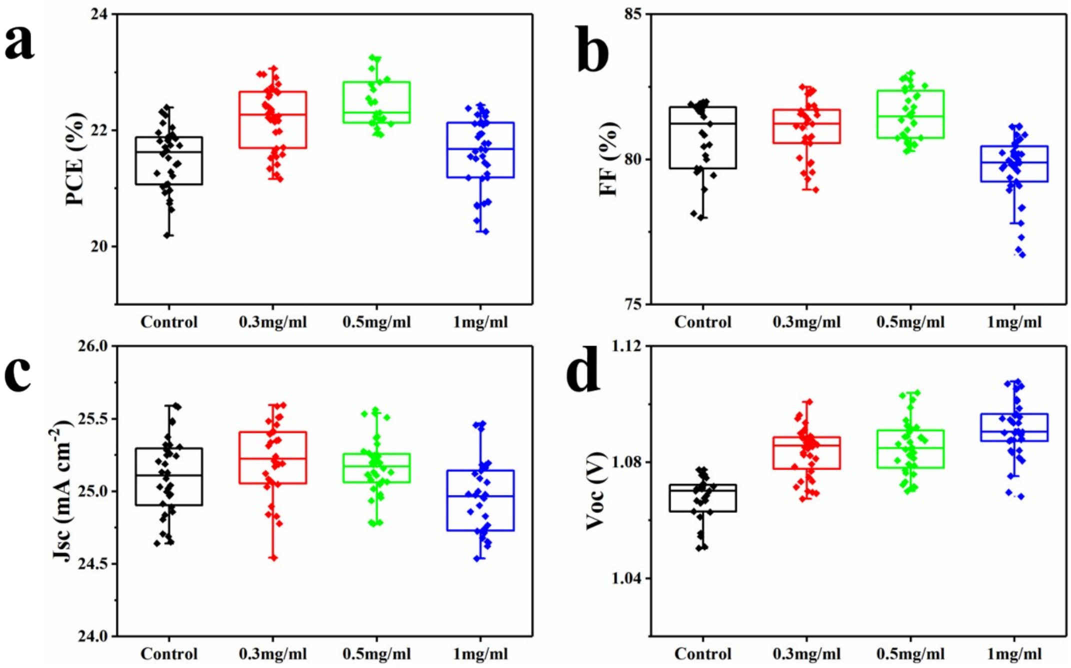

Figure S6 Statistical distribution of PSC photovoltaic parameters passivated by different concentrations of CTPC (a) PCE (b) FF (c) Jsc (d)Voc

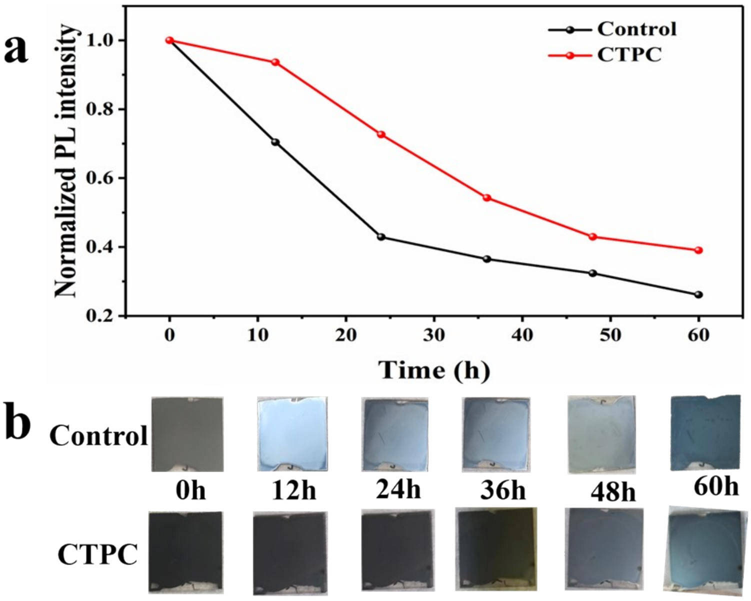

Figure S7 (a) PL peak intensity over time (b) electronic photos of perovskite films.

Table S1 Fitting parameters of TRPL spectra of the control perovskite and perovskite with CTPC films.

Sample	A_1	τ_1 (μs)	A_2	τ_2 (μs)	τ_{ave} (μs)
PVK	679.47	1.29	582.35	1.29	1.29
PVK/CTPC	137.05	0.22	430.30	3.83	3.76

Table S2 EIS fitting parameters for control and CTPC passivated PSCs.

Sample	RS (Ω)	RT (kΩ)	RR (kΩ)
Control	47.89	11.02	58.86
CTPC	12.45	40.06	155.40