

## Supporting Information

Enhancing efficiency and stability of perovskite solar cells via in-situ incorporation of lead sulfide layer

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## Experimental Details

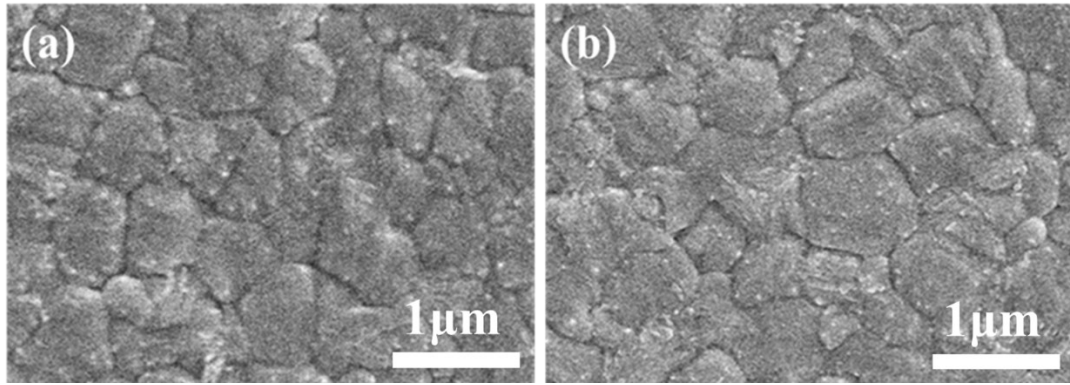
**Materials.** Lead (II) iodide (99.999%) was purchased from Sigma Aldrich. Methylammonium chloride, methylammonium bromide (>99.5%), methylammonium iodide (>99.5%), formamidinium iodide and Spiro-OMeTAD were purchased from Xi'an Polymer Light Technology in China. Thioacetamide was obtained from TCI. Anhydrous dimethylformamide, anhydrous dimethylsulfoxide, chlorobenzene and acetonitrile were purchased from Sigma-Aldrich. SnO<sub>2</sub> colloid precursor (tin (IV) oxide, 15% in H<sub>2</sub>O colloidal dispersion) was purchased from Alfa Aesar.

**Solar cell fabrication.** ITO glass (TECA7 7 Ω sq<sup>-1</sup>, Advanced Election Technology CO.,Ltd.) was cleaned by sequentially washing with deionized water, acetone, and isopropyl alcohol for 20 min, respectively, and then further treated by UV-ozone (UVO) for 15 min for device fabrication. The SnO<sub>2</sub> colloid precursor was diluted by DI water at volume ratio of 1:5, and then spin-coated on the clean substrate at 3,000 r.p.m. for 30 s, then it was dried at 150 °C for 30 min. The perovskite solution was prepared by dissolving 1105 mg ml<sup>-1</sup> of formamidinium lead iodide, 33 mg ml<sup>-1</sup> of methylammonium chloride in N,N-dimethyl formamide/dimethyl sulfoxide (8:1 v/v) mixed solvent. The solution was deposited by two consecutive spin-coating steps of 1,000 r.p.m. and 6,000 r.p.m. for 5 s and 25 s. During the second spin-coating step (6,000 r.p.m.), 1 ml of anhydrous chlorobenzene was quickly poured onto the substrate after 15 s. Then the film was put on a hotplate at 150 °C for 10 min. For the TAA treatment, the perovskite film was then immersed in TAA solution in mixed solvent (chlorobenzene:isopropanol = 95:5 v/v) (1mg/ml) for 1 min at room temperature. A hole-transporting layer of spiro-OMeTAD was deposited by spin-coating at 4,000 r.p.m. for 30 s in a glovebox. 1 mL spiroOMeTAD/chlorobenzene (80 mg/mL) solution was employed with addition of 17 μL lithium bis(trifluoromethanesulfonyl)imide (Li-TFSI)/acetonitrile (520 mg/mL) and 28.8 μL of 4-tert-butyl pyridine. Finally, an 80 nm Au electrode was deposited via thermal evaporation.

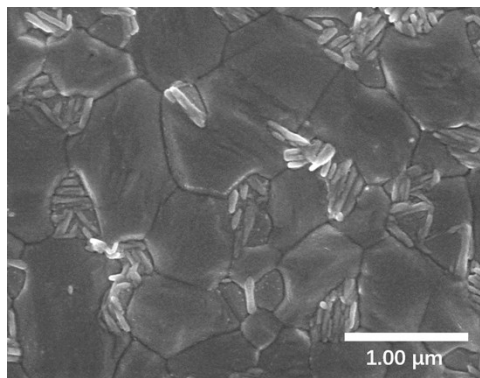
**Measurement and characterization.** The absorption spectra were collected using a UV/Vis spectrometer (SHIMADZU, UV-1800 UV/Vis Spectrophotometer) in the

wavelength range of 300–900 nm. Steady-state photoluminescence (PL) was measured using Edinburgh FLS980 system with an excitation at 485 nm. The powder XRD patterns was measured using a PANalytical Empyrean X-ray powder diffractometer equipped with a 2.2 kW Cu K $\alpha$  radiation (1.54 Å). The SEM images were taken from a Hitachi SU8020 SEM operated at 5 kV. The ToF-SIMS measurements were performed in a ToF-SIMS V instrument (ION-TOF GmbH, Münster, Germany). During analysis, dual-beam depth profiling in an interlaced mode was used. A pulsed 30 keV Bi<sup>+</sup> ion beam (10 kHz, 1.08 pA current) was used for scanning on an area of 100×100  $\mu\text{m}^2$  on the sample surface. A 1 keV Cs<sup>+</sup> with a 68 nA beam current was used for the sputtering with a crater size of 350×350  $\mu\text{m}^2$ . A flood gun with a current of ~2  $\mu\text{A}$  was used during analysis for charge compensation.

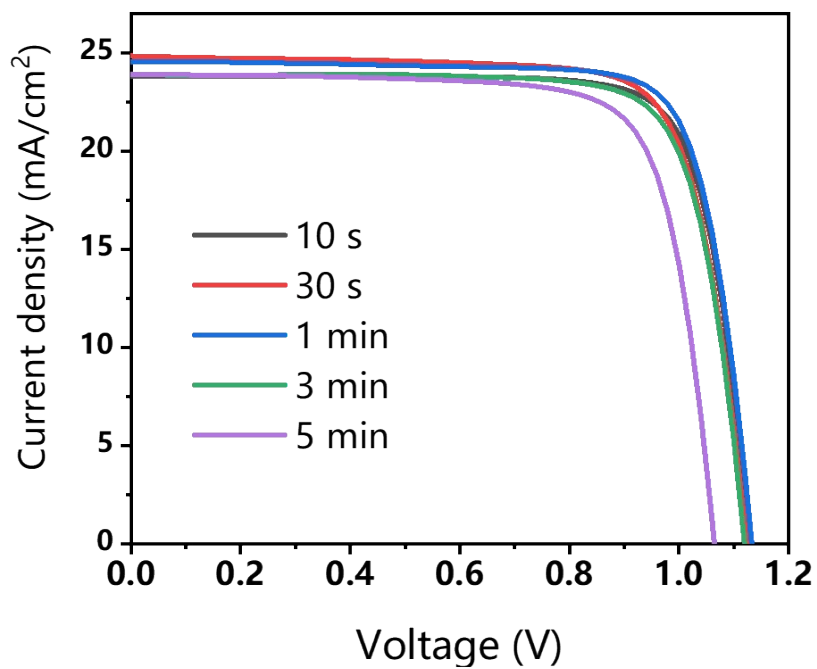
Current–voltage (J–V) characteristics were recorded by applying an external potential bias to the cell while recording the generated photocurrent with a Keithley model 2400 digital source meter. The light source was a 300 W collimated xenon lamp (Newport) calibrated with the light intensity to 100 mW cm<sup>-2</sup> under AM 1.5G solar light conditions by a certified silicon solar cell. The J–V curve was recorded by the reverse scans with a rate of 100 mV s<sup>-1</sup>. The active area was determined by metal shadow mask with an aperture of 0.0725 cm<sup>2</sup>. The incident photon-to-current conversion efficiency (IPCE) for solar cells was performed using a commercial setup (PV-25 DYE, JASCO). A 300 W Xenon lamp was employed as a light source for the generation of a monochromatic beam. IPCE spectra were recorded using monochromatic light without white light bias. Calibrations were performed with a standard silicon photodiode.



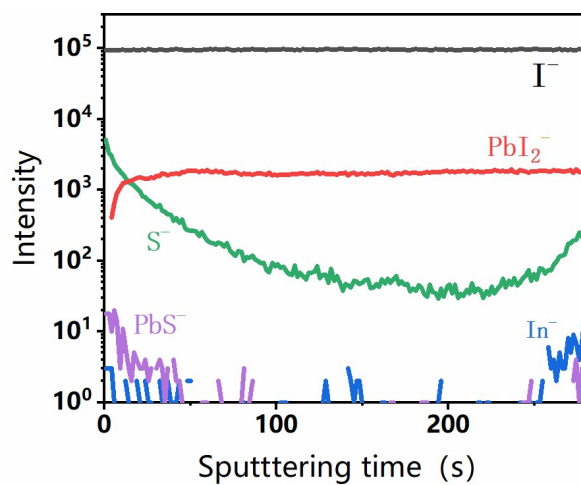
**Fig. S1.** The top-view SEM images of the perovskite films treated by TAA for 3 min (a) and 5min (b).



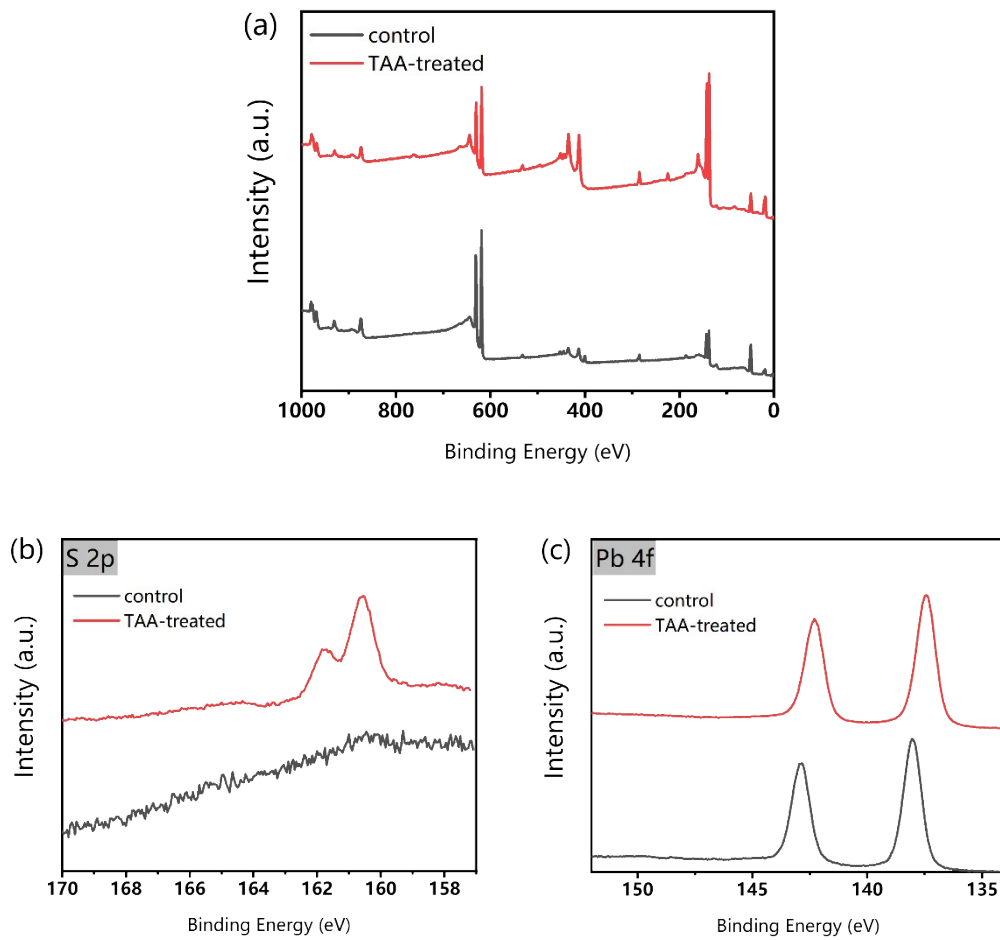
**Fig. S2.** The top-view SEM images of the perovskite film treated by  $\text{Na}_2\text{S}$  via spin coating spin without additional waiting time.



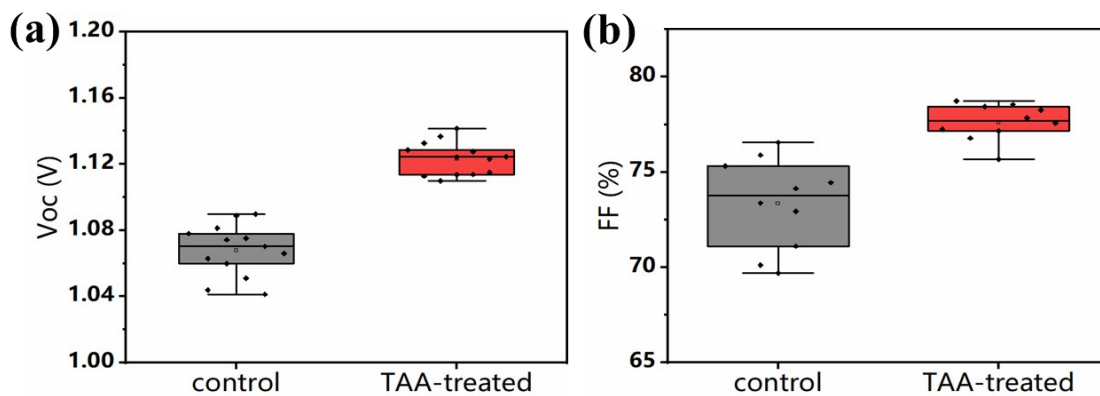
**Fig. S3.** Influence of the reaction time of TAA treatment on the  $J$ - $V$  characteristics of perovskite solar cells.



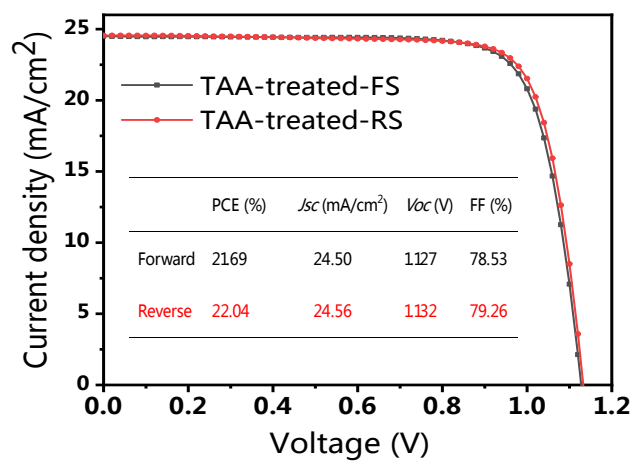
**Fig. S4.** ToF-SIMS elemental depth profiles for the film with TAA treatment.



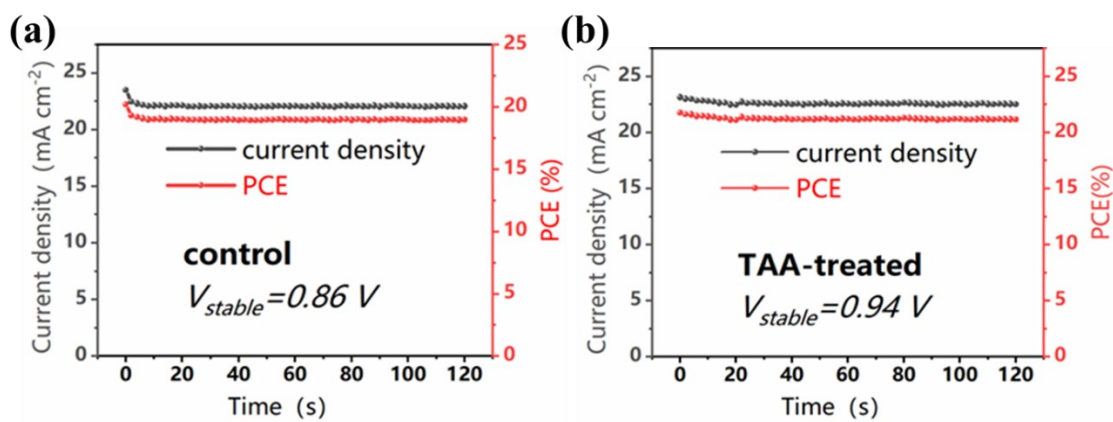
**Fig. S5.** The XPS full scan (a), XPS S 2p (b) and Pb 4f (c) spectra of the control and TAA-treated perovskite films.



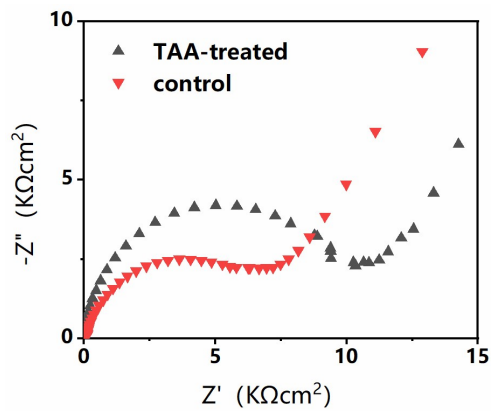
**Fig. S6.**  $V_{oc}$  (a) and FF (b) statistics of the control and the TAA-treated devices.



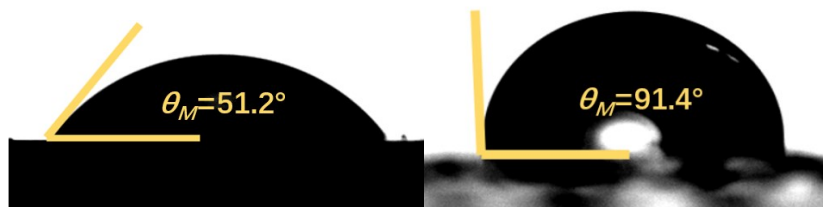
**Fig. S7.** The characteristic  $J-V$  curves of the TAA-treated device.



**Fig. S8.** Steady state photocurrent and efficiency output of the control (a) and TAA-treated (b) devices.

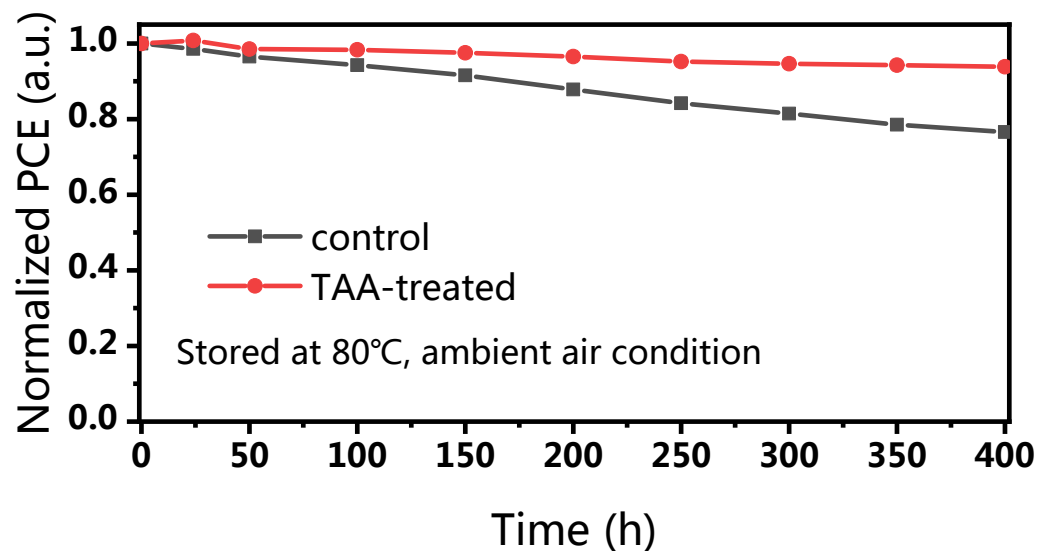


**Fig. S9.** Nyquist plots of the devices with and without TAA treated in the dark.



**Fig. S10.** Contact angles of the control and TAA-treated films.





**Fig. S11.** Long-term thermal stability of the control and the TAA-treated devices at 80°C without encapsulation.