

**Supporting information**

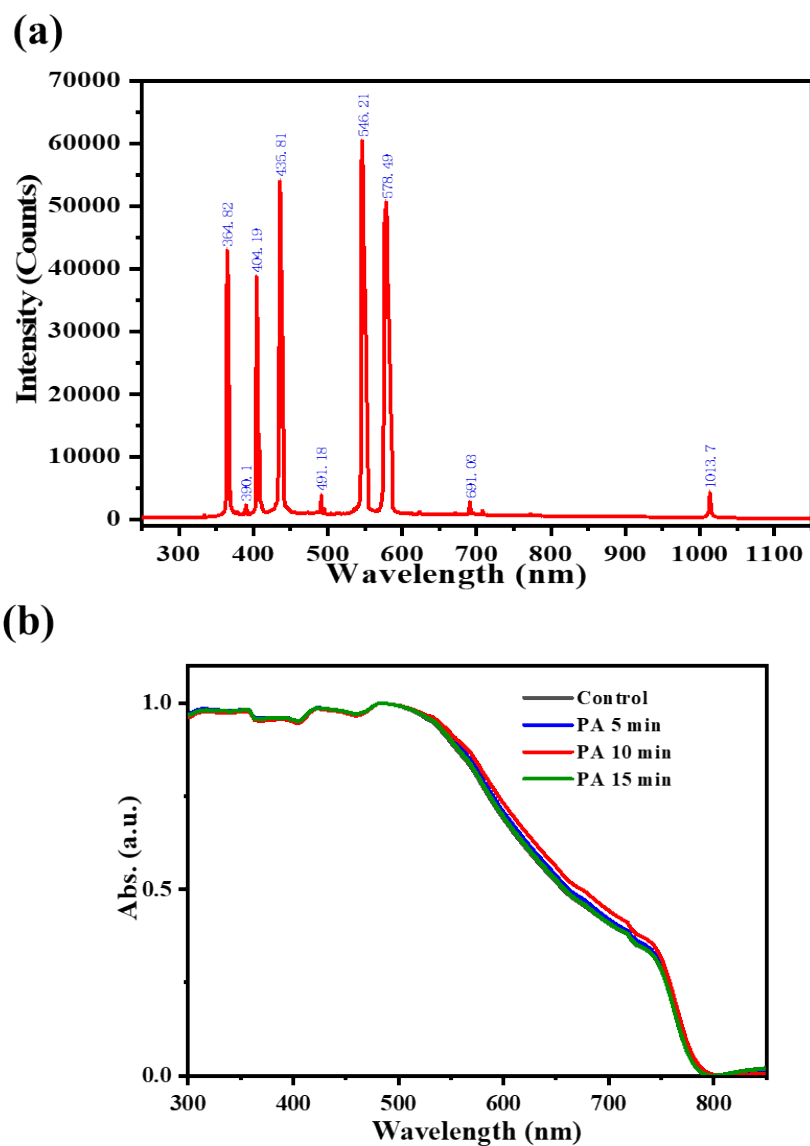
# **To Greatly Reduce Defect via Photo-Annealing for High Quality Perovskite Film**

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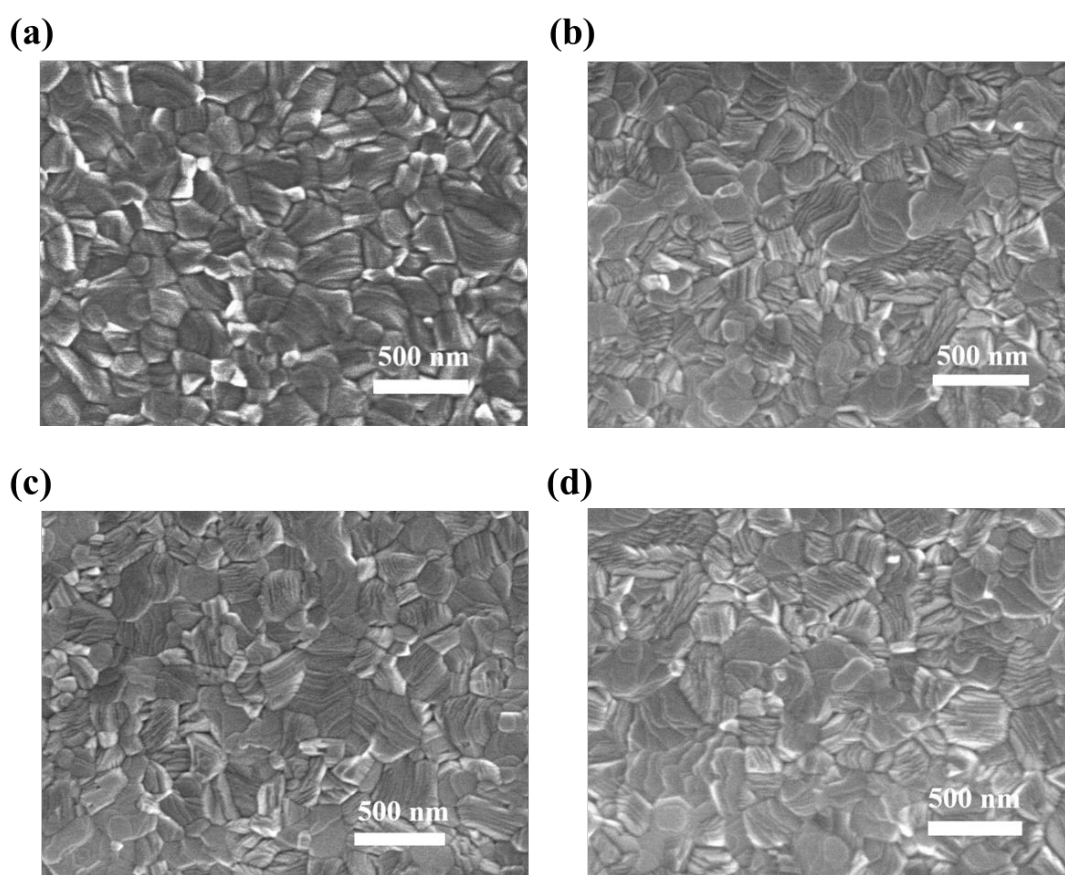
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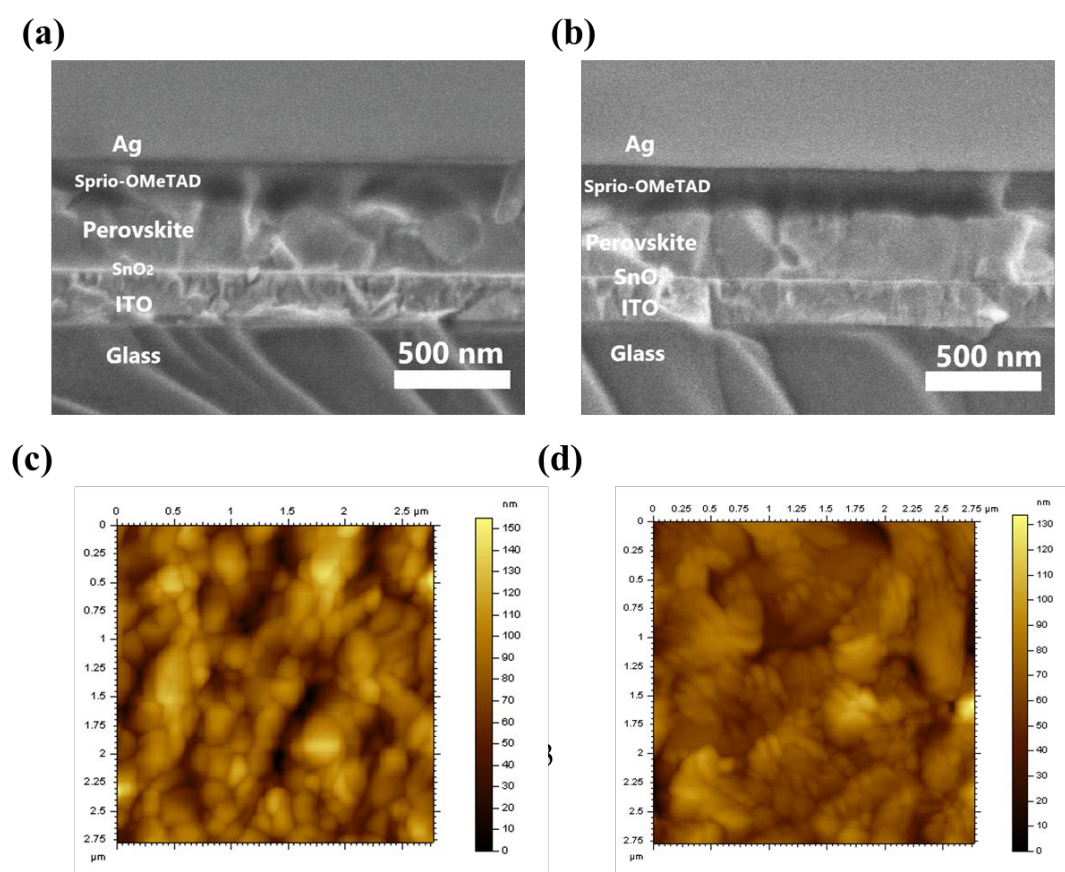
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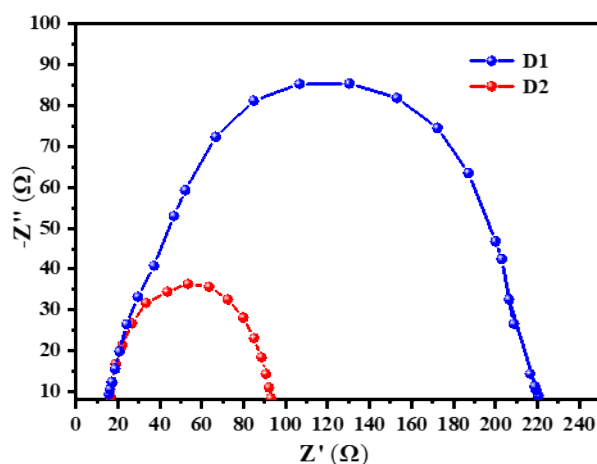
**Fig. S1.** (a) Spectrogram of the halogen lamp (Philips QVF135). (b) UV-Vis spectra of perovskite films based on TA and PA.



**Fig. S2.** SEM images of ITO/SnO<sub>2</sub>/MAPbI<sub>3</sub> film (a) based on TA. (b) based on PA 5 min. (c) based on PA 10 min. (d) based on PA 15 min.



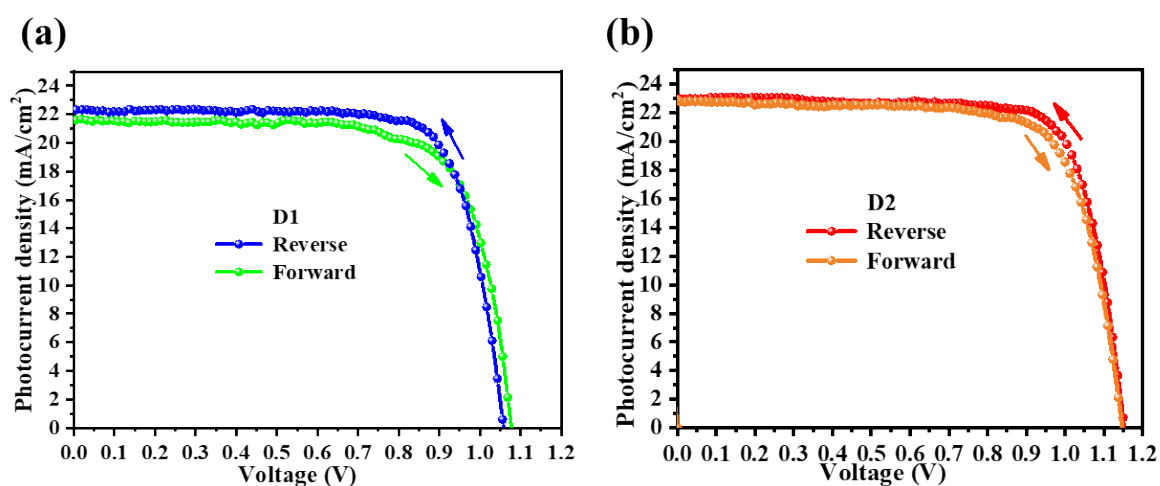
**Fig. S3.** Cross-section SEM of the device with the structure of ITO/SnO<sub>2</sub>/MAPbI<sub>3</sub>/Spiro-OMeTAD/Ag based on **(a)** TA **(b)** PA. AFM topography images of ITO/ SnO<sub>2</sub>/MAPbI<sub>3</sub> films based on **(c)** TA **(d)** PA.



**Fig. S4.** Electronic impedance spectrum in the form of Nyquist plots for D1 and D2.

**Table S1.** Transient PL spectroscopy results of MAPbI<sub>3</sub> films based on TA and PA 10min on glass.

Sample	$\tau_1$ (ns)	$\tau_1$ ratio (%)	$\tau_2$ (ns)	$\tau_2$ ratio (%)	Average life time (ns)
Control	30.53	72.75	98.95	27.25	49.17
PA 10 min	43.36	11.32	90.32	88.68	85.01



**Fig. S5.** J-V curves of the **(a)** D1 and **(b)** D2 measured at reverse and forward scans.

**Table S2.** The best photovoltaic parameters of D1 and D2 measured at reverse and forward scans.

Device	Scanning direction	Jsc(mA·cm <sup>-2</sup> )	Voc(V)	FF	PCE(%)
D1	Forward	21.21	1.08	0.733	17.03
	Reverse	22.31	1.07	0.757	18.08
D2	Forward	22.86	1.13	0.755	19.51
	Reverse	22.97	1.14	0.779	20.41

## 1. Experimental Section

*Materials.* 180 nm of indium tin oxide (ITO) coated glass substrates with a sheet resistance of 8  $\Omega$ /sq were purchased from Huayulianhe Co., Ltd. SnO<sub>2</sub> colloid precursor (tin(IV) oxide, 15% in H<sub>2</sub>O colloidal dispersion), anhydrous N,N-dimethylformamide (DMF), anhydrous dimethyl sulfoxide (DMSO), and chlorobenzene were obtained from Alfa Aesar. PbI<sub>2</sub> (99.9985%) and spiro-OMeTAD were purchased from Xi'an Polymer Light Technology Co., Ltd. CH<sub>3</sub>NH<sub>3</sub>I (99.5%, denoted as MAI) was acquired from Borun New Material Technology Co., Ltd. Lithium bis(trifluoromethylsulfonyl) imide (Li-TFSI) and 4-tertbutylpyridine (TBP) were obtained from Aldrich. All these commercially available materials were used as received without any further purification.

*Device Fabrication.* ITO-coated glass was sequentially cleaned in deionized water, acetone, and ethanol under ultrasonic each treatment for 30 min<sup>1</sup>. The cleaned ITO-coated glass substrates were then dried with a nitrogen stream and treated by oxygen plasma cleaning for 15 min immediately.

The SnO<sub>2</sub> colloid precursor was ultrasonically diluted by deionized water (1:6 volume ratio) for 60 min. The acquired SnO<sub>2</sub> solution was spin-coated onto glass/ITO substrates at 3000 rpm for 30 s and then baked on a hot plate at 150 °C for 30 min<sup>2</sup>.

The perovskite film was deposited by a modified one-step spin coating method. 1.25 mmol MAI and PbI<sub>2</sub>, 89 μL DMSO was mixed in 1 mL DMF at room temperature and to form a precursor solution. The perovskite CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> (denoted as MAPbI<sub>3</sub>) film was fabricated by a homemade low-pressure-assisted method as reported in literature<sup>3-4</sup>. The perovskite precursor solution was spin-coated onto the cooled glass/ITO/SnO<sub>2</sub> substrate at 4000 rpm for 6 s. The coated film was quickly moved to a small sample chamber that was pumped immediately for 70 s (below 15 Pa). A brown and transparent film was then obtained<sup>5</sup>.

After these, for control devices, the gas-pump dried film was baked on a hot plate at 100 °C for 10 min with the conventional thermal annealing method<sup>6</sup>. For the optimized devices, the gas-pump dried film was placed in a position of about 5 cm under the halogen lamp (Philips QVF135 500W) for 5 min, 10 min, and 15 min, with the aim of illumination and crystallization, while the temperature was about 75 °C ( as seen in **Fig. 1**).

Spiro-OMeTAD solution, which was prepared by mixing 72.3 mg Spiro-OMeTAD, 39 μL of TBP and 35 μL of Li-TFSI solution (260 mg Li-TFSI in 1 mL acetonitrile) in 1 mL chlorobenzene, was spin-coated on the perovskite layer at 4000 rpm for 30 s<sup>7</sup>. All of the above, all processes were performed in ambient atmosphere. Finally, a 100 nm Ag cathode was deposited by thermal evaporation at a pressure of  $< 2.5 \times 10^{-3}$  Pa and a evaporation rate of  $< 0.3$  nm/s. The circular solar cells with the area was 0.09 cm<sup>2</sup> were shaped by mask.

*Characterization.* The morphology of samples was measured by a scanning electron microscope (SEM) (Hitachi S-4800). The current density–voltage (J–V) curves were measured with a scan rate of 20 mV/s under 100 mW/cm<sup>2</sup> AM 1.5G simulated illumination of Newport solar simulator using a Keithley 2611 Semiconductor Characterization System.. The X-ray diffraction (XRD) patterns were obtained by using a D/MAX-2000 X-ray

diffractometer with monochromatic Cu K $\alpha$  irradiation (1 1/4 1.5418 Å) at a scan rate of 6°/min. The absorption spectrum was recorded with a UV-visible spectrophotometer (Agilent 8453). The electrochemical impedance spectroscopy (EIS) measurements were performed on the Zahner Zennium electrochemical workstation in the dark. Photoluminescence (PL) (excitation at 485 nm) was measured with NaonLog infrared fluorescence spectrometer (Nanolog FL3-2Ihr). Transient PL measurement was measured using UltraFast lifetime Spectrometer (Delta flex). The incident photon-to-current conversion efficiency (IPCE) spectrum was observed using a lock-in amplifier (model SR830 DSP) coupled with a 1/4 m monochromator (Crowntech M24-s) and 150 W tungsten lamp (Crowntech). The halogen lamp was Philips QVF135 with the power of 500 W. All of the measurements of the solar cells were performed in an ambient atmosphere at room temperature without encapsulation.

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