

## Supporting information

# Perovskite Solar Cells with PCE over 19% Fabricated under Air Environment by Using Dye Molecule Additive

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### Experimental Section

**Materials:** Unless stated otherwise, solvents and chemicals were obtained commercially and used without further purification. Fluorine-doped tin oxide (FTO) substrate was purchased from Advanced Election Technology Co. N, N-dimethylformamide (DMF), dimethyl sulfoxide (DMSO), ethyl acetate (EA), isopropanol (IPA), acetonitrile (ACN), Titanium (diisopropoxide) bis (2, 4-pentanedionate) were purchased from Shanghai Macklin Biochemical Co., Ltd. PbI<sub>2</sub>, tBP, Li-TFSI were obtained from Sigma-Aldrich Inc. Methylammonium iodide (MAI), silver (Ag), FK209 and Spiro-OMeTAD were purchased from Xi'an Polymer Light Technology Corp.

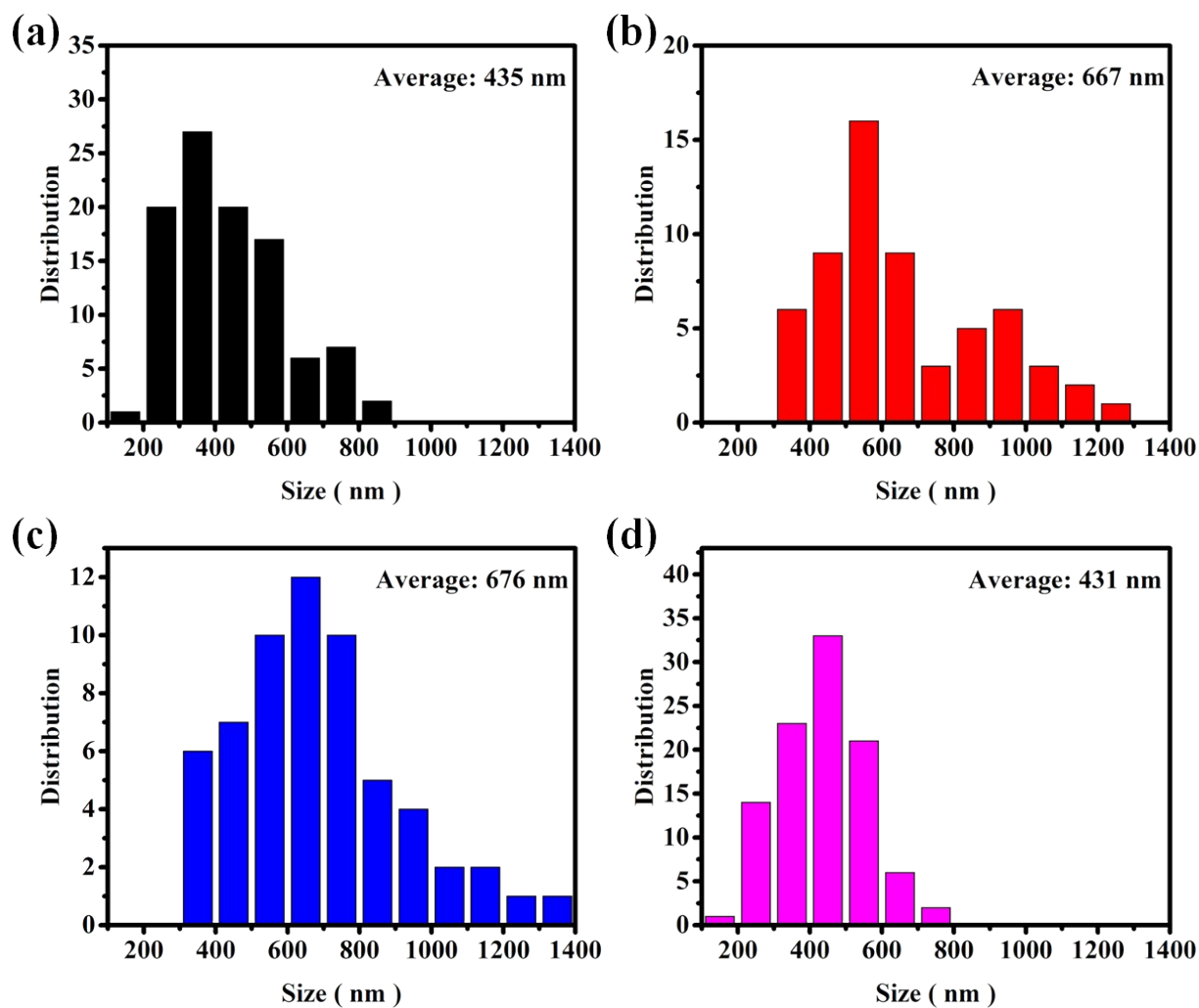
**Device Fabrication:** The planer Perovskite solar cells were fabricated in air environment with the following structure: FTO/TiO<sub>2</sub>/MAPbI<sub>3</sub>/Spiro-OMeTAD/Ag. The FTO glass substrates were sequentially cleaned by ultrasonic bath with acetone and alcohol for about 15 min, and followed by an ultraviolet ozone treatment for 30 minutes. TiO<sub>2</sub> precursor solution was prepared by dissolving the Titanium (diisopropoxide) bis (2, 4-pentanedionate) in absolute ethanol. The low-

concentration solution ( $70 \text{ mg mL}^{-1}$ ) was spin-coated at 2000 rpm for 20 s, and baked at  $150 \text{ }^\circ\text{C}$  for 30 min. Another high- concentration solution ( $150 \text{ mg/mL}$ ) was spin-coated at 4000 rpm for 20 s and baked at  $450 \text{ }^\circ\text{C}$  for 30 min and then cooled down to room temperature. Perovskite solution was prepared by dissolving 159 mg MAI, 461 mg  $\text{PbI}_2$  and 71 mL DMSO in 600 mL DMF. For the best 8GFF containing PVSK, 0.2 mg 8GFF was added into the anti-solution. The precursor solution was spin-coated on the substrate at 2000 rpm for 10 s, and 4000 rpm for 25 s, to which 250 mL of EA was rapidly dropped after 10 s. The spin-coated film was heated at  $100 \text{ }^\circ\text{C}$  for 30 min. The Spiro-OMeTAD solution was prepared by dissolving 65 mg Spiro-OMeTAD, 70  $\mu\text{L}$  lithium salt/acetonitrile solutions, 20  $\mu\text{L}$  tBP and 50  $\mu\text{L}$  FK/acetonitrile solution in 1 mL chlorobenzene. Then, the prepared solution was spin-coated on the perovskite layer at 4000 rpm for 30 s. The devices were completed by evaporating 100 nm Ag in a vacuum chamber. All the steps are kept in the ambient air environment with about 40% humidity except Ag deposition.

### **Characterization**

The morphology of the samples was characterized by Field-emission scanning electron microscope (FE-SEM, Ultra 55, Zeiss, Germany). Fourier transform infrared spectroscopy (FTIR) measurements were performed using the power mode on the infrared spectrometer (VERTEX 70V, Bruker, Germany). The XPS data were measured by using X-Ray photoelectron spectroscopy (XPS, K-Alpha, USA). The crystal structures of  $\text{MAPbI}_3$  were characterized by X-ray diffraction analysis (XRD, Thermo ARL-X'TRA, USA). The absorption spectra were recorded by UV-vis Spectrophotometer S-4 (UV1102II6812011, China). Monochromatic incident photon-to-current conversion efficiency (IPCE) spectra were measured with a monochromatic incident light of  $1 \times 10^{16} \text{ photons cm}^{-2}$  in director current mode (CEP-2000BX, Bunko-Keiki). The photocurrent density–voltage (J-V) characteristics of the cells were measured

using a Keithley 2400-SCS source meter under AM 1.5 illumination with an intensity of 100 mW cm<sup>-2</sup>. PL measurement was performed with Spectrofluorometer (FS5, Edinburgh instruments). Here, the time-resolved PL measurement was employed 465 nm pulsed laser as excitation source. And the EIS measurements were characterized by using a potentiostat (Im6ex/Zahner) under 10 mV AC amplitude, and frequency scanning range was 0.01-100 kHz.



**Figure S1** The size distribution of perovskite film with different concentration of 8GFF: (a) the control, (b) 0.1mg mL<sup>-1</sup>, (c) 0.2mg mL<sup>-1</sup> and (d) 0.3mg mL<sup>-1</sup>.

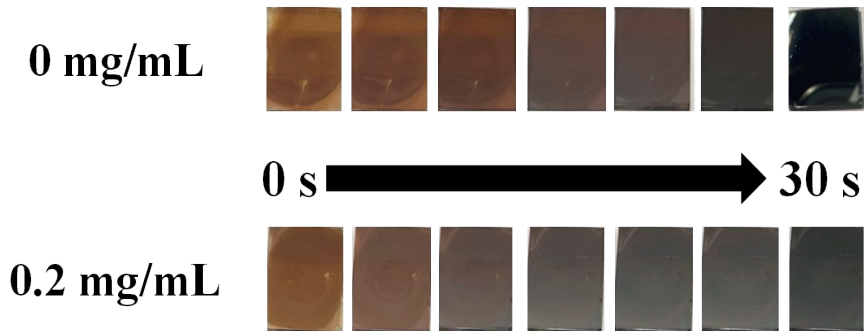


Figure S2 The corresponding photographs of perovskite film during the crystallization process at 100 °C.

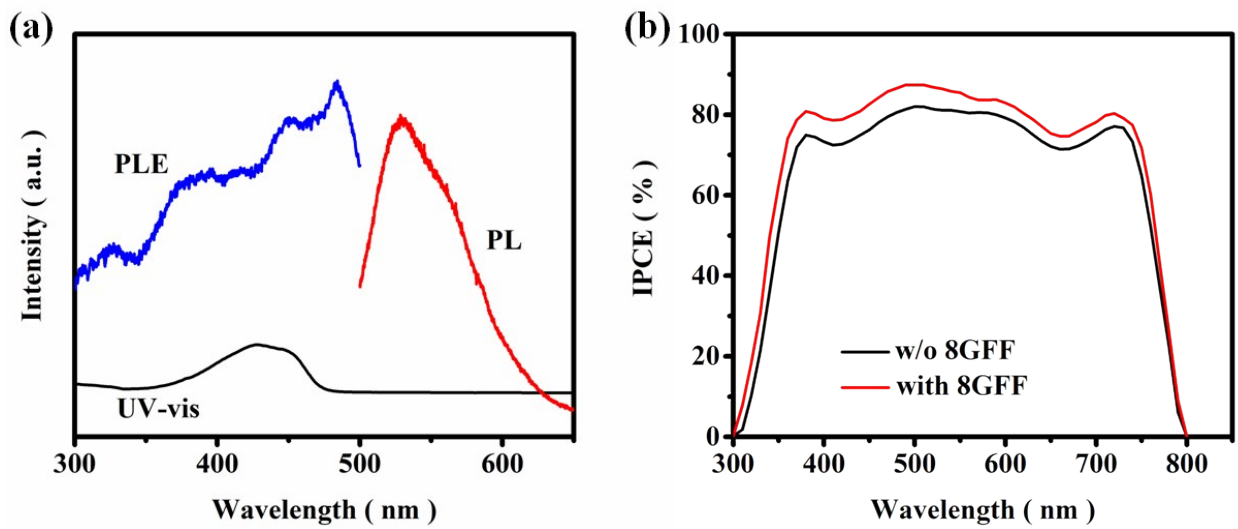


Figure S3 (a) UV-vis absorption (dissolved in EA), PL, and excitation (PLE) spectra of 8GFF, (b) IPCE spectra.

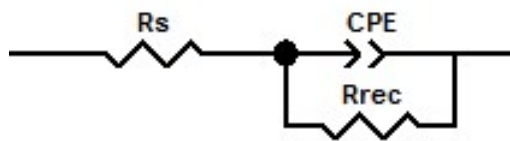
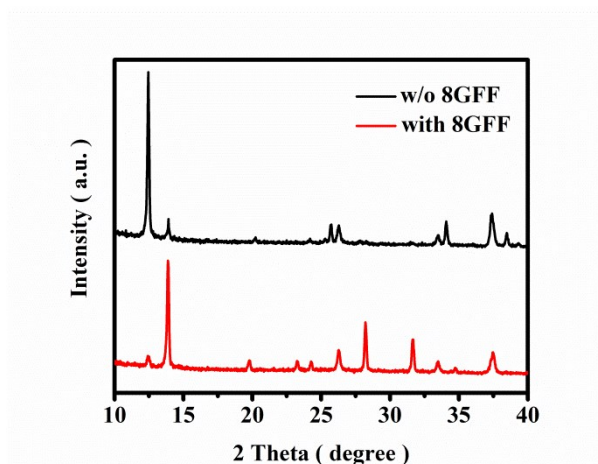
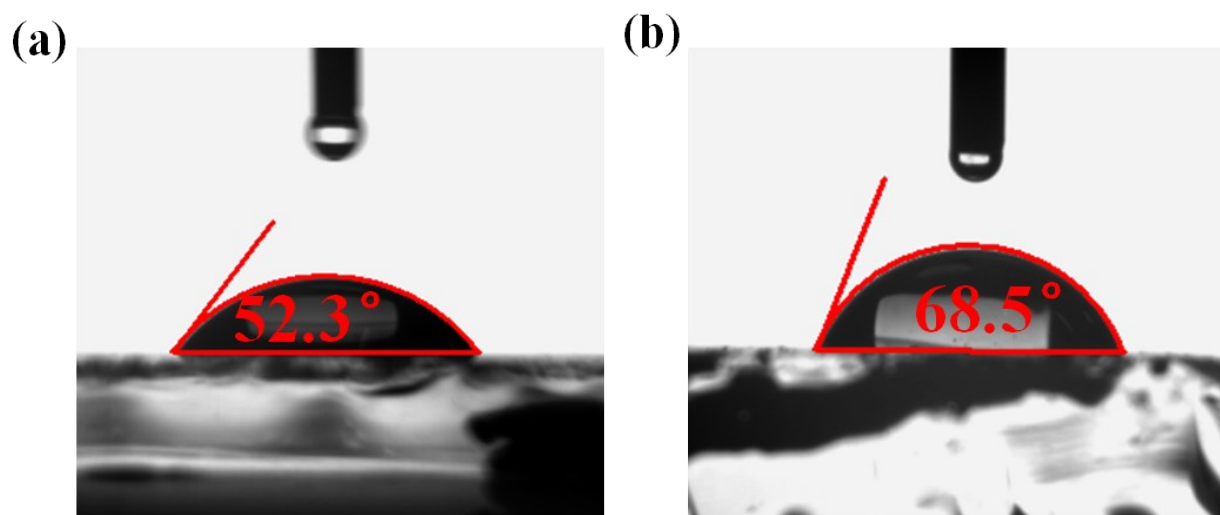


Figure S4 the equivalent circuit model of PSCs for EIS.



**Figure S5** XRD patterns of corresponding perovskite films after aging in air with relative humidity of about 80%.



**Figure S6** Images of water contact angle on the surface of the corresponding perovskite films (a) w/o 8GFF, and, (b) with 8GFF.

**Table S1** Detail FWHM value of the strong peak at 14.2°.

Concentration(mg mL <sup>-1</sup> )	FWHM
0	0.203
0.1	0.186
0.2	0.157
0.3	0.218

**Table S2** Detailed photovoltaic parameters of the devices in different scan directions.

PSCs	Scan direction	$J_{sc}$ (mA/cm <sup>2</sup> )	$V_{oc}$ (V)	FF(%)	PCE(%)
w/o 8GFF	Reverse	20.14	1.07	72.43	15.61
	Forward	19.37	1.05	70.16	14.27
with 8GFF	Reverse	23.10	1.10	75.40	19.16
	Forward	22.73	1.09	74.14	18.37

$$H = \frac{PCE_{reverse} - PCE_{forward}}{PCE_{reverse}}$$

The hysteresis index is defined by the equation: <sup>1</sup> Herein, the H value is

proportional to the degree of hysteresis.<sup>1</sup>

**Table S3** The fitted parameters of TRPL spectra.

Samples	$A_1$	$\tau_1$ (ns)	$A_2$	$\tau_2$ (ns)	$\tau_{avg}$ (ns)
w/o 8GFF	2531	4.17	2434	64.85	33.91
w 8GFF	2917	3.48	1986	43.92	20.06

$$\tau_{avg} = \frac{\sum A_i \tau_i^2}{\sum A_i \tau_i}$$

$\tau_{avg}$  represents the average PL decay time fitted by the equation:

**Table S4** EIS parameters of the PSCs.

Device	$R_s$ ( $\Omega$ )	$R_{rec}$ ( $\Omega$ )
w/o 8GFF	20.10	9448
with 8GFF	30.85	19517

## References

1. Q. He, M. Worku, L. Xu, C. Zhou, S. Lteif, J. B. Schlenoff and B. Ma, *J. Mater. Chem. A*, 2020, **8**, 2039-2046.
2. L. Xiao, J. Xu, J. Chen, J. Luan, Y. Yang, B. Zhang, J. Yao and S. Dai, *Sustainable Energy. Fuels*, 2017, **1**, 1358-1365.

