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Supplementary Information

Microstructure Modulation of $CH_3NH_3PbI_3$ Layer in Perovskite Solar Cell by 2-propanol Pre-wetting and annealing in Spray-assisted Solution Process

Faming Li[a], Chunxiong Bao[a], Weidong Zhu[a], Bihu Lv[b], Wenguang Tu[a], Tao Yu*[abc], Jie Yang[a], Xiaoxin Zhou[a], Yangrunqian Wang[a], Xiaoyong Wang[b], Yong Zhou[a] and Zhigang Zou[abc]

[a] National Laboratory of Solid State Microstructures & Eco-Materials and Renewable Energy Research Center (ERERC) at Department of Physics, Nanjing University, Nanjing 210093, P. R. China. *E-mail: yutao@nju.edu.cn*

[b] Collaborative Innovation Center of Advanced Microstructures, Nanjing University, Nanjing 210093, P. R. China.

[c] Jiangsu Key Laboratory for Nano Technology, Nanjing 210093, P. R. China.

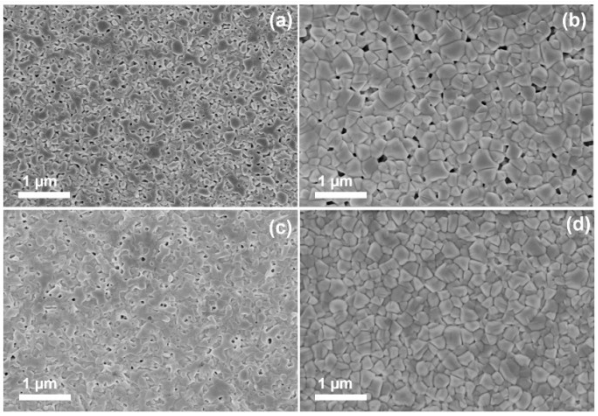

Figure S1. SEM images of PbI_2 films were pre-wetted by same volume ethanol (a) and IPA (c) with spray-coating, and the $CH_3NH_3PbI_3$ film fabricated by spray-assisted solution process (b) and (d), respectively.

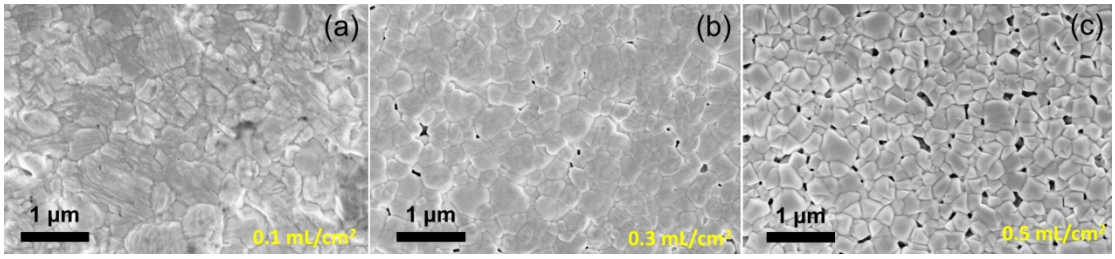

Figure S2. The surface SEM images of perovskite films prepared under different amount ethanol pre-wetting of 0.1 (a), 0.3 (b), 0.5 mL/cm^2 (c).

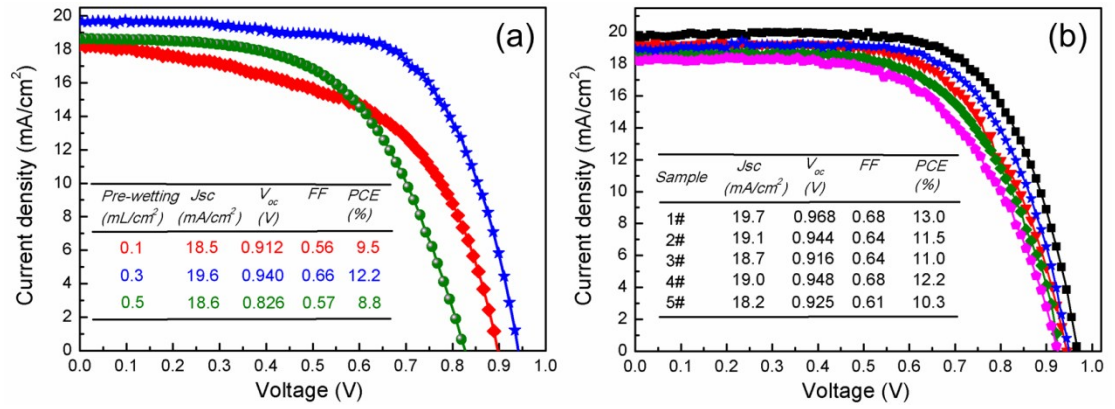

Figure S3. The J-V curves and performance parameters of PSCs for light absorption layer prepared under different amount ethanol pre-wetting (a), and the J-V curves of five cells prepared by pre-wetting of 0.3 mL/cm^2 (b).

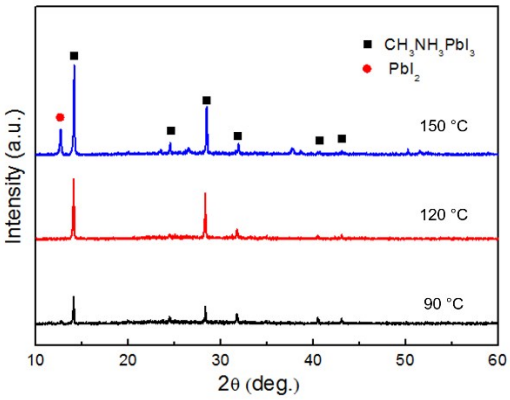

Figure S4. XRD patterns of perovskite films at different annealing temperature.

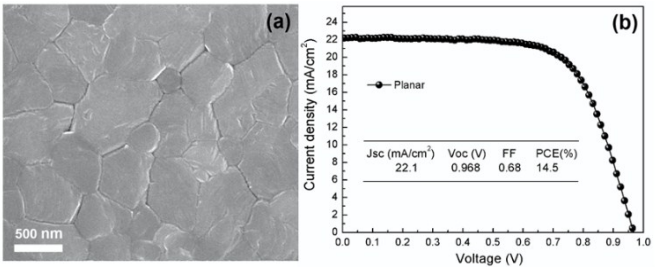

Figure S5. SEM image of perovskite film was fabricated on the FTO/TiO_2 hole-blocking layer substrate (a) and the optimal J-V curve of the planar structured PSCs (b).

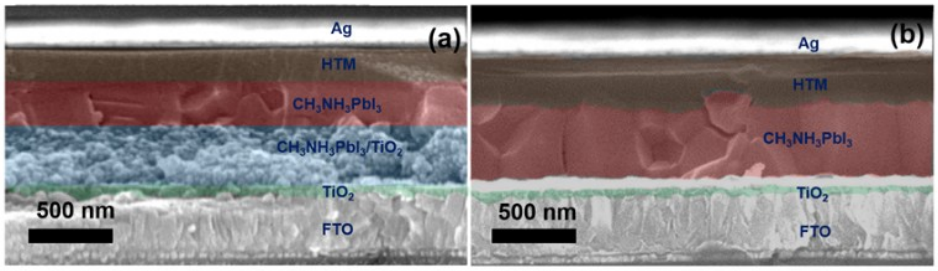

Figure S6. Cross-sectional SEM images of mesoporous structure PSCs (a) and planar structure PSCs (b).

Table S1. Summary of the performance parameters of the perovskite photovoltaic devices were fabricated under different annealing time (AT). The SD and HI represent scan direction and the hysteresis index, respectively.

AT (˚C)	SD	J_{sc} (mA/cm2)	V_{oc} (V)	FF	PCE (%)	HI
90	FB-SC	15.3	0.904	0.55	7.6	0.156
	SC-FB	15.8	0.829	0.44	5.8	
120	FB-SC	19.1	0.948	0.69	12.5	0.015
	SC-FB	19.1	0.952	0.67	12.2	
150	FB-SC	18.5	0.914	0.64	10.8	0.193
	SC-FB	18.1	0.843	0.55	8.4	

Table S2. Summary of the biexponential fit decay parameters for the time-resolved PL decays shown in Figure 9.

AT(˚C)	τ_1 (ns)	Fraction 1	τ_2 (ns)	Fraction 2	k_{et}/s^{-1}
90	13.1	57%	48	43%	0.5×10^8
120	1.3	91%	30	9%	7.3×10^8
150	4.5	71%	39	29%	1.2×10^8

Generally, the resulting curves can be fitted by biexponential decay, which is consisted of a fast decay and a slow decay process. The fast decay component, τ_1, is generally corresponding to the quenching of free carriers in the $CH_3NH_3PbI_3$ film through transport to TiO_2 layer. The slow decay component, τ_2, could be attributed to radiation recombination. The fraction 1 and 2 represent the weight percentage of fast and slow decay, respectively.

Table S3. The performance parameters of the perovskite photovoltaic device, the absorbers were annealed at different time for 120 °C.

AT Time (min)	J_{sc} (mA/cm^2)	V_{oc} (V)	FF	PCE (%)
10	18.2	0.890	0.55	8.9
30	19.1	0.948	0.69	12.5
60	20.1	0.982	0.72	14.2
90	18.9	0.930	0.61	10.7