

Supporting Information

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Experimental Section

Materials

MAI, Butylamine iodide (BAI), Guanidine thiocyanate (GUTS) and Methylazanium iodide (MAI) were bought from Xi'an Polymer Light Technology Corp. PbI₂ was purchased from TCI. Titanium diisopropoxy diacetylacetonate, N, N-dimethylsulfoxide (DMSO), N, N-dimethylformamide (DMF), Chlorobenzene (CB), Ethyl acetate (EA) were brought from Sigma-Aldrich. Isopropanol (IPA) was purchased from Macklin. Titanium dioxide (TiO₂) was purchased from Great Cell Solar. All materials were prepared without further purification.

Device fabrication

The perovskite solar cells were fabricated with the following the mesoporous structure: glass/FTO/c-TiO₂/m-TiO₂/MAPbI₃/C. FTO glasses was conducted by detergent, diluted water, acetone and sonicated respectively, and then shift to an ultrasonic bath

with full of ethanol to sonicate for 20 min. The compact TiO₂ layer prepared by spray pyrolysis method of decomposition titanium diisopropoxy diacetylacetonate soluble in isopropanol was deposited on FTO glasses, followed by annealed at 450 °C for 1 h. Subsequently, the mesoporous TiO₂ layer which was employed via spin-coating titanium dioxide solution with 3000rpm, 30s procedure was further sintering at 500 °C for 1h. The substrate was treated for 20 min prior to use. Then, the precursors of MAPbI₃ consisting of 1.4 M PbI₂, 1.4 M MAI in mixed solution of DMF and DMSO with the volume ratio of 1:4 was further deposited substrate at 4500 rpm for 25 sec and 105ul of EA was slowly dripped on the film of rotating in 18s before this film turned to be fuzzy. The transparent film was transferred to the heated platen at 70 °C for 5min in order to form MAI-PbI₂-DMSO adduct and 100 °C for 10 min. For devices based on X% M GUTS, the varied concentration (1% M , 3% M , 5% M , 7% M , 9% M) of GUTS in IPA was covered on substrate at 4000rpm for 30 sec, and then was sintered at 100 °C for 10 min for the MAI-PbI₂-DMSO adduct. For films based on X mM-M BAI, the various concentration (5 mM, 10 mM, 15 mM, 20 mM) of BAI in IPA was covered on MAI-PbI₂-DMSO film at 4000rpm for 30 sec, and then was sintered at 100 °C for 10 min. For films based on X mM-H BAI, the various concentration (5 mM, 10 mM, 15 mM, 20 mM) of BAI in IPA was covered on MAPbI₃ film at 4000rpm for 30 sec, and then was sintered at 100 °C for 10 min. For devices based on X% M GUTS & X Mm-H BAI, the BAI was further deposited with 5000 rpm for 30min and then was sintered at 100°C for 10 min after depositing GUTS. Eventually, the carbon electrode was deposited by screen printing method, and then was annealed at 100 °C for 30 min.

Characterization

The morphologies of samples were measured via SEM (JSM-6700F). X-ray diffraction (XRD) patterns were adopted by an Empyrean diffractometer. UV-Vis absorption spectra were employed by a Lambda 750S absorption spectrophotometer. X-ray photoelectron spectroscopy (XPS) was performed by Thermo Scientific K-Alpha⁺ device. Time-resolved PL decay curves were executed by DeltaFlex. Atomic force

microscope (AFM) images were used by Innova SPM P700. Steady-state PL spectra were recorded on LabRAM HR800. An electrochemical workstation (CHI 760) (EIS) was employed to measure impedance of solar cells. The Fourier transform infrared (FT-IR) spectra were recorded by using Nicolet N10. The current density-voltage ($J-V$) characteristics of PSCs were evaluated by SS150. Atomic force microscopy (Innova SPM P700) (AFM) was adopted to research the surface roughness of perovskite layer. The Transmission electron microscopy (TEM) was used to study the internal structure of perovskite. UV photoelectron spectroscopy was employed to explore the energy level of films.

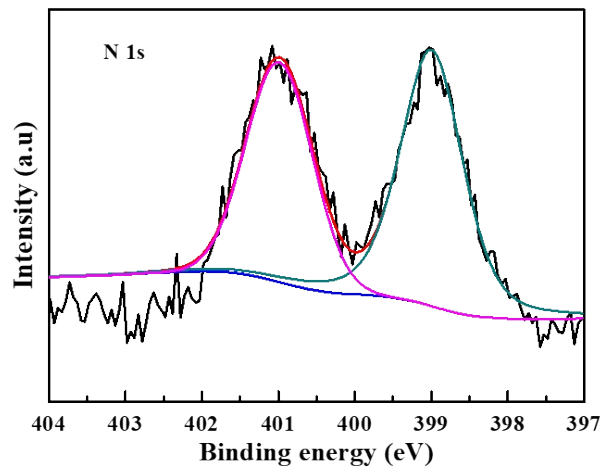


Figure S1. The N 1s curve of XPS for 5% M GUTS passivation treatment

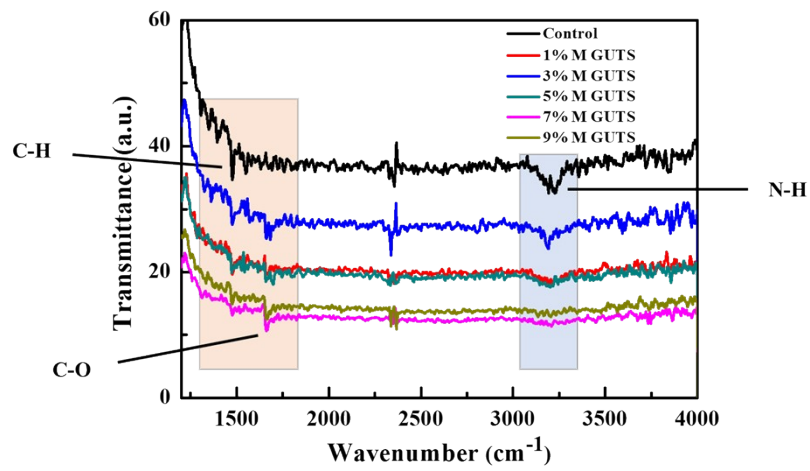


Figure S2. The FTIR spectra of the pristine MAPbI₃ film and X% M GUTS film

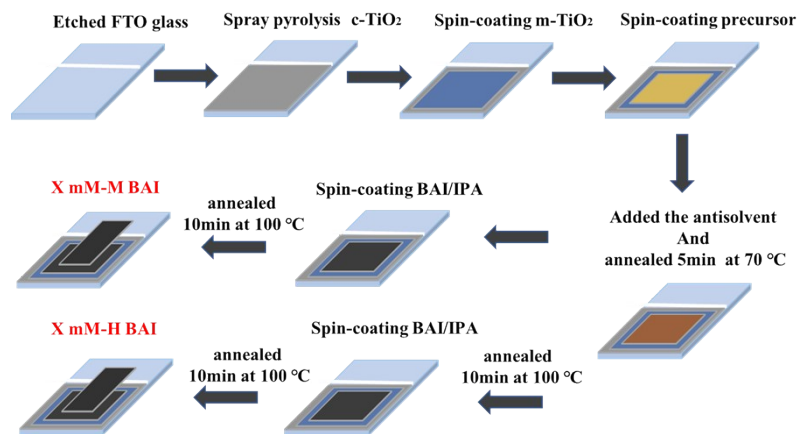


Figure S3. The flow chart of experimental preparation of device based on X mM-M BAI and X mM-H BAI

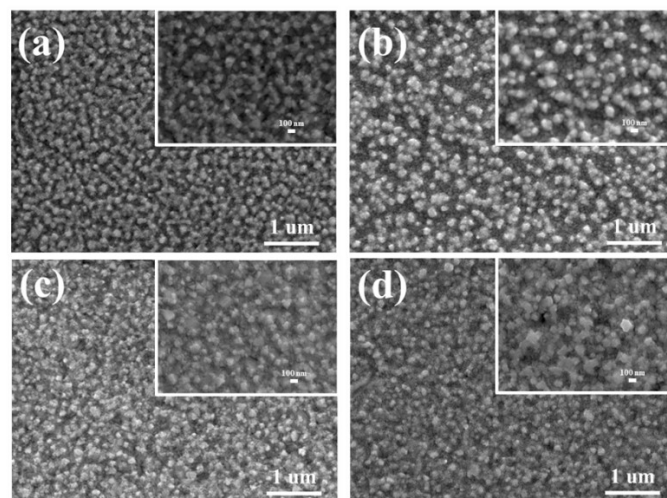


Figure S4. The images of SEM of (a) the 5 mM-M BAI film, (b) the 10 mM-M BAI film, (c) the 15 mM-M BAI film, and (d) the 20 mM-M BAI film

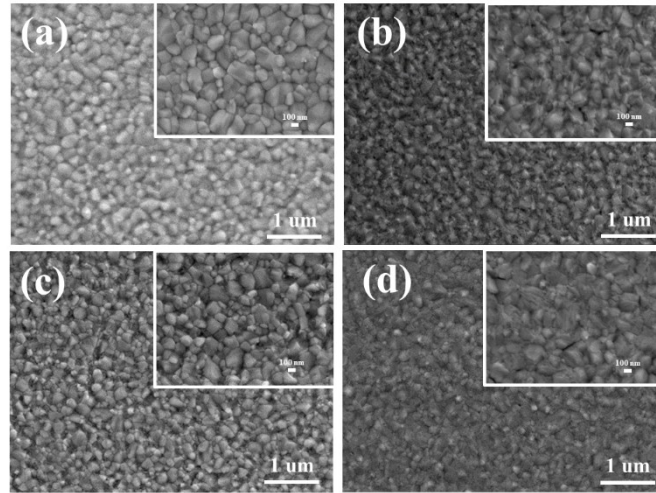


Figure S5. The images of SEM of (a) the 5 mM-H BAI film, (b) the 10 mM-H BAI film, (c) the 15 mM-H BAI film, and (d) the 20 mM-H BAI film

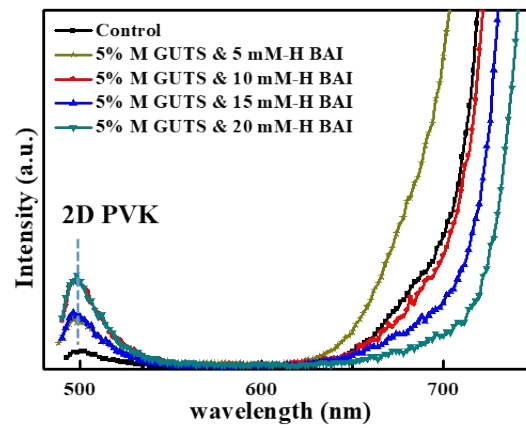


Figure S6. The PL spectra in the low wavelength range of 5% M GUTS & X mM-H BAI film

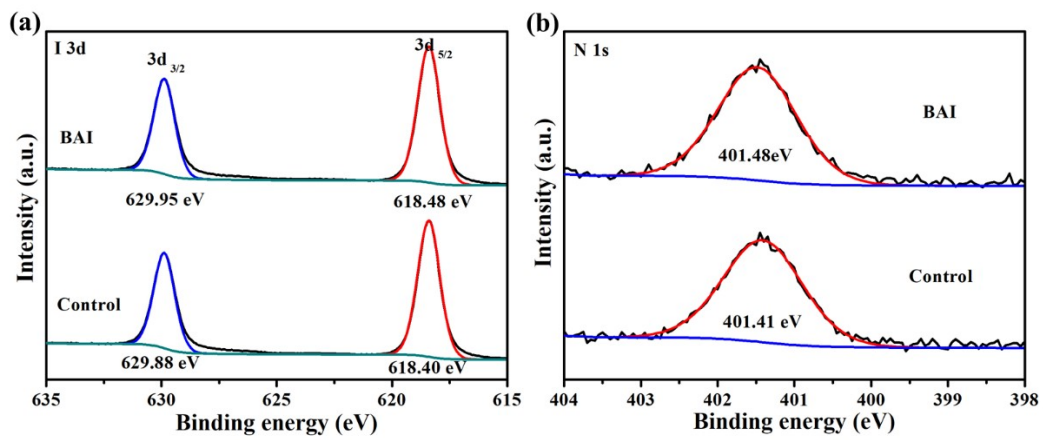


Figure S7. The I 3d and N 1s curve of XPS for BAI passivation treatment

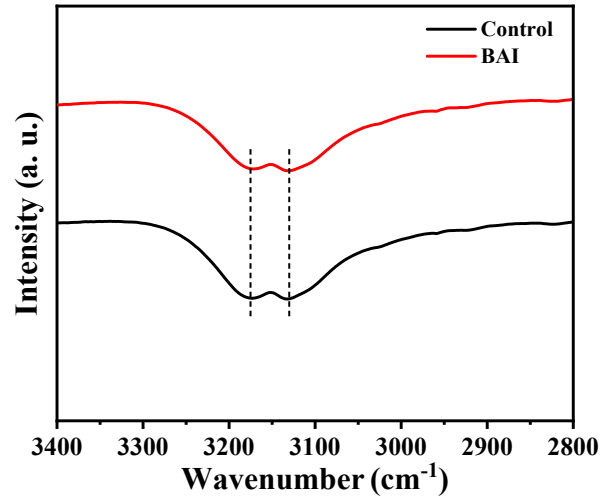


Figure S8. The FTIR spectra of the pristine MAPbI₃ film and BAI-based film

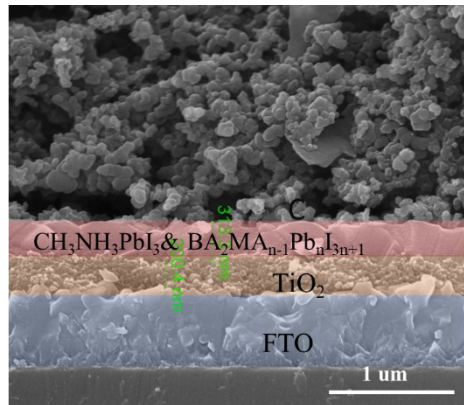


Figure S9. The cross-sectional SEM image of the CH₃NH₃PbI₃ & BA₂MA_{n-1}Pb_{n-1}I_{3n+1} PSCs

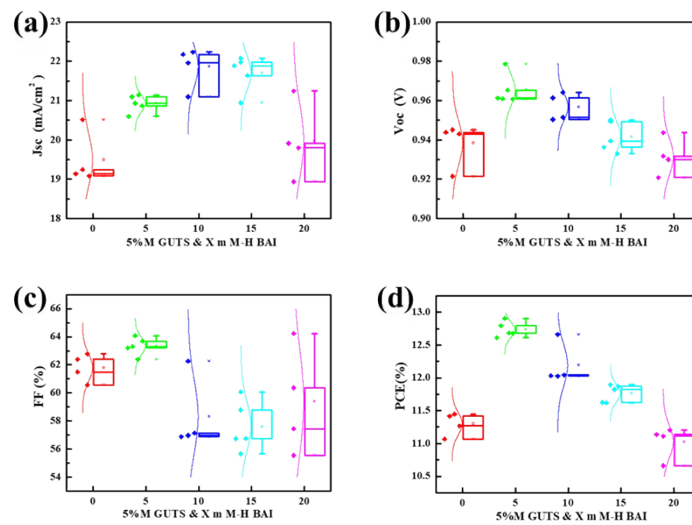


Figure S10. The statistical distribution of the J_{sc} (a), V_{oc} (b) FF (c) and PCE (d) of 5%M GUTS& X m M-H BAI.

Table S1. AFM test data of 5% M GUTS passivation treatment.

Sample	Rq (nm)	Ra (nm)	Rz (nm)
Control	12.218	9.580	160.327
5% M GUTS	19.133	15.127	128.341

Table S2. The UPS data of 5% M GUTS passivated perovskite film

Sample	$E_{\text{cut-off}}$ (eV)	E_{edge} (eV)	W_f (eV)	VB (eV)	E_g (eV)	CB (eV)
5%MGUTS	17.236	1.677	3.984	5.661	1.570	5.634

Table S3. The TRPL fitting data for perovskite films treated with different GUTS concentrations

Sample	τ_1 (ns)	τ_2 (ns)	A_1 (%)	A_2 (%)	τ_{ave} (ns)
Control	23.40	9.84	43.40	56.60	18.63
1% M GUTS	14.81	42.98	57.44	42.56	34.03
3% M GUTS	31.28	62.76	34.31	65.69	56.26
5% M GUTS	48.43	75.25	19.72	80.28	71.59
7% M GUTS	20.45	6.81	24.31	75.69	13.51
9% M GUTS	22.43	4.58	14.51	85.49	12.68

Table S4. The TRPL fitting data for perovskite films treated with 15 Mm-M BAI and 5% M GUTS & 5 Mm-H BAI

Sample	τ_1 (ns)	τ_2 (ns)	A_1 (%)	A_2 (%)	τ (ns)
TiO ₂ /PVK-1	30.03	2.54	7.36	92.64	15.85
TiO ₂ /PVK-2	53.77	12.00	17.55	82.45	34.03
PVK-1	0.25	136.13	33.43	66.57	123.73
PVK-2	14.98	82.08	31.81	68.19	76.82

Table S5. The impedance spectrum fitting data of the reference device and the device treated with 5% M GUTS & 5 Mm-H BAI

Sample	Rs	Rtrans	Ctrans	Rrec	Crec
Control	79.87	390.50	1.2636E-7	4459	5.4935E-8
5% M-GUTS & 5 mM-H BAI	37.65	447.20	2.1206E-7	20677	4.8945E-8