

## Quantifying the energy loss for the perovskite solar cell passivated by acetamidine halide

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### Experiment

**Materials:** FAI (99.5%), MABr (99.5%), MACl (99.5%), PbI<sub>2</sub> (anhydrous, 99.99%), Spiro-OMeTAD (99.5%), lithium bistrifluoromethane-sulfonimide (LiTFSI, 99.95%) and 4-tert-butylpyridine (tBP) were purchased from Xi'an polymer light technology Corp. AAI and AABr were purchased from GreatCell Solar Ltd. Isopropanol (IPA, 99.5%), *N,N*-dimethylformamide (DMF, anhydrous, 99.8%), dimethyl sulfoxide (DMSO, anhydrous, 99.8%), chlorobenzene (CB, anhydrous, 99.8%) and acetonitrile (anhydrous, 99.9%) were acquired from Sigma-Aldrich and used as received without further purification. The SnO<sub>2</sub> colloid precursor (tin(IV) oxide, 15% in H<sub>2</sub>O colloidal dispersion) were acquired from Alfa Aesar.

**Fabrication of devices:** ITO glass substrates (Advanced Election Technology Co., Ltd.) were cleaned sequentially by sonication in detergent, deionized water, acetone, ethanol and isopropanol, twice for each liquid and 15 min for each time, followed by

ultraviolet-ozone treatment for 20 min. For SnO<sub>2</sub> ETL, the diluted SnO<sub>2</sub> nanoparticles solution (2.67%) was spin coated on glass/ITO substrates in ambient air, and then annealed at 150 °C for 30 min.<sup>1</sup> After depositing the electron transport layer, the perovskite layer was deposited by a two-step spin coating method; first, 1.3 M of PbI<sub>2</sub> in DMF :DMSO (v/v=95:5) was spin coated onto the electron transport layer at 3000 rpm for 50 s, during this process, the mixture solution of FAI:MABr:MACl (60 mg:6 mg:6 mg in 1 ml isopropanol) was adding before the end of 30 s, and then the film was annealed in at 150 °C for 15 min. For post-treatment condition, when the perovskite film cool down to the room temperature, acetamidine iodide/acetamidine bromide solution (0.5, 1.0, 2.0 mg mL<sup>-1</sup> in IPA) was spin-coated onto the perovskite surface with 5000 rpm for 60 s. Spiro-OMeTAD HTLs were deposited by spin-coating at 4000 rpm for 30 s. Ag electrode (80 nm) was eventually deposited by vacuum evaporation (pressure < 2 × 10<sup>-4</sup> Pa). For the thermal stability and humidity stability test, the Ag electrode is replaced by Au electrode (80 nm).

**Characterization:** *J-V* characteristics of photovoltaic cells were taken using a Keithley 2400 source measurement unit under a simulated AM 1.5G spectrum. With a SS-F5-3A solar simulator (Enli Technology CO., Ltd.), the light intensity was calibrated by standard single-crystal silicon solar cells. The absorption spectrum of perovskite films was measured on a Cary 5000 (Agilent) spectrophotometer. The top-view and cross-sectional SEM images of the samples were characterized using a high-resolution scanning electron microscope (Zeiss Merlin). An electron beam accelerated to 5 kV was used with an SE2 detector. Absorption spectrum was tested using a Cary Series UV-vis-NIR Spectrophotometer. X-ray diffraction (XRD) were measured on D2 PHASER (Bruker, Germany) diffractometer. Time-resolved photoluminescence (TRPL) spectroscopy was measured on Lifespec (Edinburgh Instrument, UK). The samples were excited by a pulsed laser, with a wavelength and frequency of 373 nm and 2 MHz. The Mott-Schottky plot and corresponding IPCE spectra were measured by an IM6 electrochemical workstation (Zahner Zennium, Germany) at room temperature in ambient conditions. Electroluminescence was performed by applying different voltages to the device with Keithley 2400 Source Measure Unit.

Simultaneously, the luminance was measured with a luminance meter (Konica Minolta, CS-200), and electroluminescence (EL) spectra were recorded by a Flame spectrometer (Ocean Optic). The current efficiency was calculated by dividing the luminance by current density, and the EQE was calculated using Lambertian emission profiles and obtained the EL spectrum.

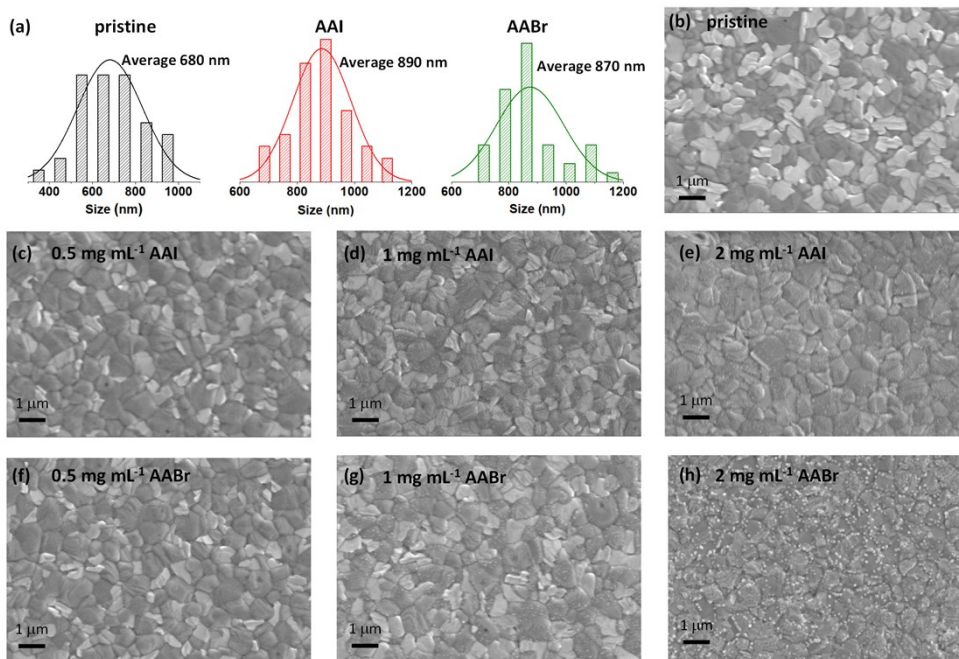


Figure S1 (a) The grain size distribution of pristine, AAI and AABr perovskite film SEM image of perovskite films. (b) Pristine perovskite film, treated perovskite perovskite with (c) 0.5mg mL<sup>-1</sup> AAI, (d) 1.0 mg mL<sup>-1</sup> AAI, (e) 2.0 mg mL<sup>-1</sup> AAI. (f) 0.5mg mL<sup>-1</sup> AABr, (g) 1.0 mg mL<sup>-1</sup> AAI, (h ) 2.0 mg mL<sup>-1</sup> AAI. The bar is 1  $\mu\text{m}$ .

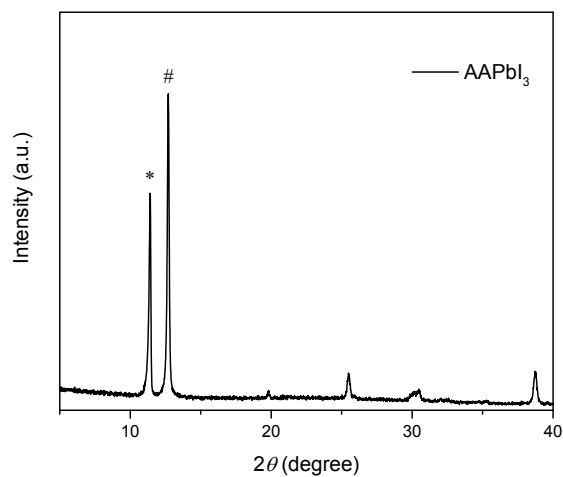


Figure S2. XRD of AAPbI<sub>3</sub> perovskite film. The film fabricated by two step method and only AAI in the IPA solution (\*: AAPbI<sub>3</sub>, #: PbI<sub>2</sub>).

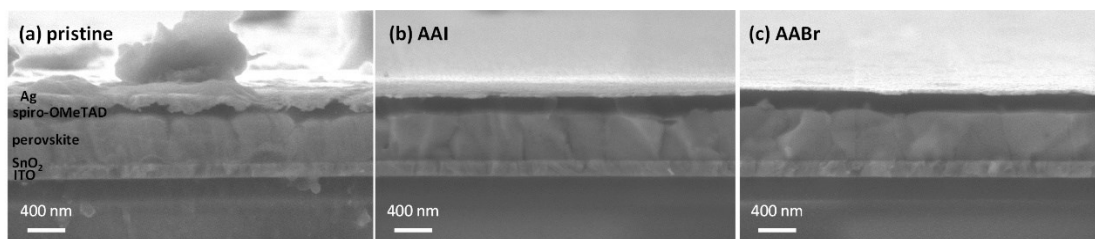


Figure S3. Cross-sectional SEM images of PSCs. (a) pristine, (b) AAI, (c) AABr. The bar is 400 nm.

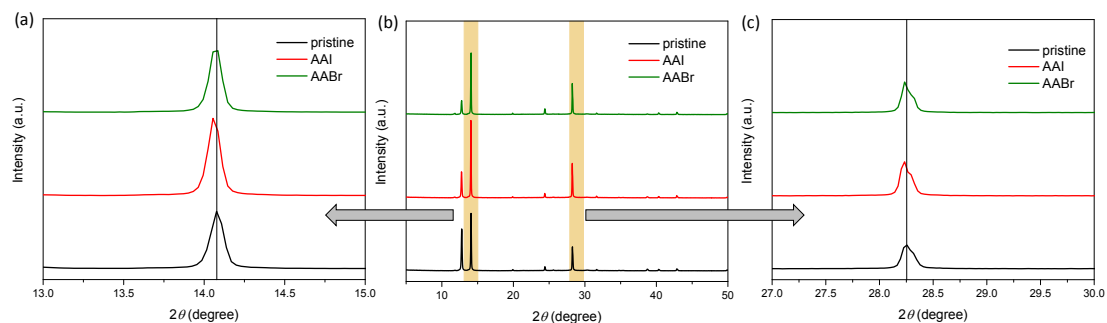


Figure S4. XRD of pristine perovskite film, AAI and AABr perovskite film, respectively. (a) and (c) are enlarged XRD pattern showing the (110) and (220) facet, respectively.

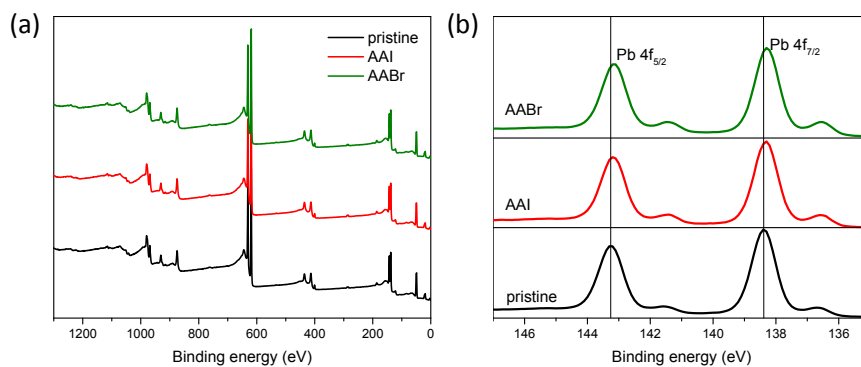


Figure S5. (a) XPS surface analysis of pristine, AAI and AABr treated films. (b) High-resolution deconvoluted Pb 4f peak for pristine, AAI and AABr treated films.

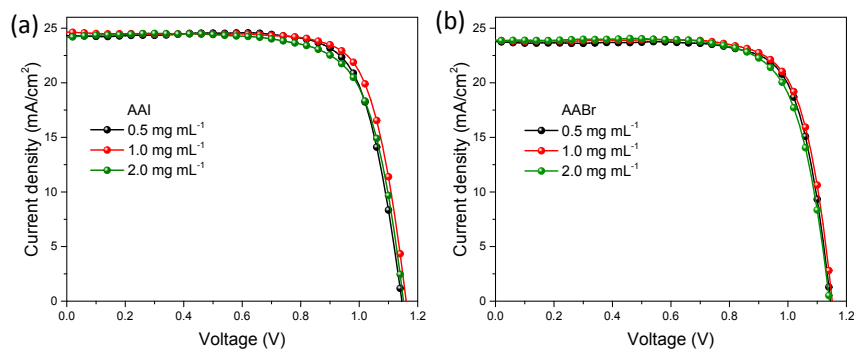


Figure S6.  $J-V$  curve of PSCs with different concentration of (a) AAI and (b) AABr treatment.

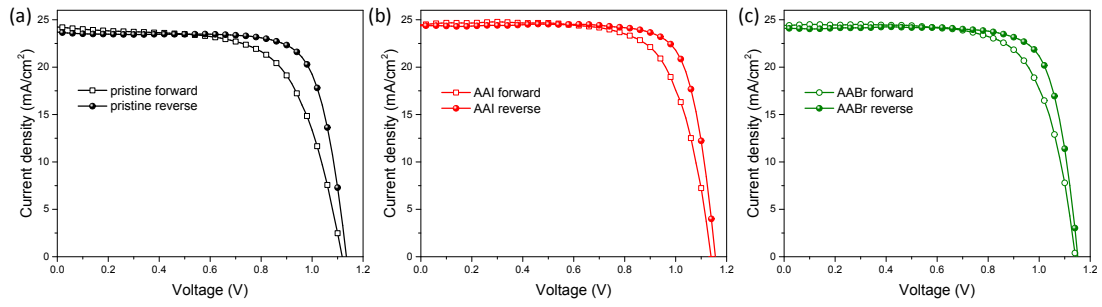


Figure S7.  $J$ - $V$  curve of forward and reverse scan for (a) pristine, (b) AAI and (c) AABr devices. (forward scan: 0 V to 1.2 V, scan rate 20 mV/s, delay time 1 ms and reverse scan: 1.2 V to 0 V, scan rate 20 mV/s, delay time 1 ms).

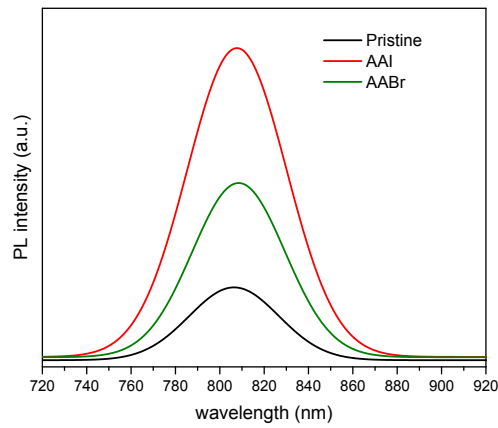


Figure S8. The PL spectra of pristine perovskite films, AAI treated perovskite film and AABr treated perovskite film on  $\text{SnO}_2/\text{ITO}$  substrate.

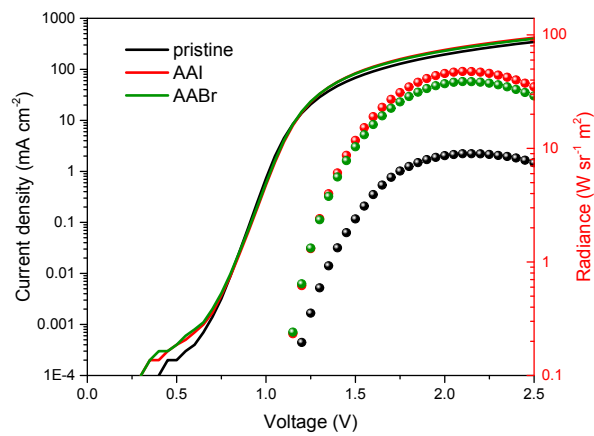


Figure S9.  $J$ - $V$  and radiance versus voltage ( $L$ - $V$ ) characteristics of the pristine, AAI and AABr PSCs.

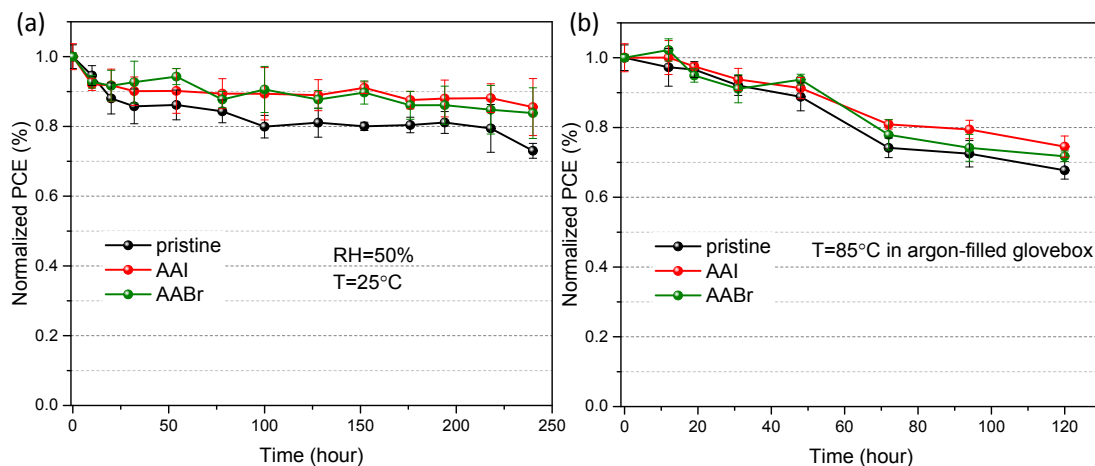


Figure S10. Stability of the unencapsulated PSCs for pristine, AAI and AABr PSCs in the dark. (a) Humidity stability (temperature is 25 °C, relative humidity around at 50%), (b) Thermal stability (temperature is 85 °C, storage at argon-filled glovebox).

Table S1. Photovoltaic parameter of PSCs with different concentration of (a) AAI and (b) AABr treatment.

Passivation layer	Concentration (mg mL <sup>-1</sup> )	$V_{oc}$ (V)	$J_{sc}$ (mA cm <sup>-2</sup> )	FF (%)	PCE (%)
AAI	0.5	1.15	24.31	75.38	21.00
	1.0	1.16	24.64	75.45	21.58
	2.0	1.15	24.15	73.55	20.44
AABr	0.5	1.15	23.78	75.69	20.61
	1.0	1.16	23.89	75.65	20.81
	2.0	1.14	23.87	73.81	20.13

Table S2. Photovoltaic parameter of forward and reverse scan for pristine, AAI and AABr treated PSCs.

Passivation layer	Scan mode	$V_{oc}$ (V)	$J_{sc}$ (mA cm <sup>-2</sup> )	FF (%)	PCE (%)	HI (%)
pristine	Forward	1.12	24.23	64.83	17.57	13.9
	reversed	1.14	23.76	75.13	20.41	
AAI	Forward	1.14	24.46	71.46	19.90	9.5
	reversed	1.16	24.47	78.04	21.98	
AABr	Forward	1.14	24.40	70.53	19.65	8.4
	reversed	1.15	24.08	77.37	21.45	

Table S3. Photovoltaic parameter of PSCs with PEAI and GAI treatment.

Passivation layer	$V_{oc}$ (V)	$J_{sc}$ (mA cm <sup>-2</sup> )	FF (%)	PCE (%)
pristine	1.11	24.00	75.97	20.35
PEAI	1.13	24.22	78.21	21.44
GAI	1.13	24.00	77.72	21.05

Table S4. Values for TRPL characteristics for pristine, AAI and AABr perovskite film coated on SnO<sub>2</sub>/ITO substrate.

	$\tau_1$ (ns)	$f_1$ (%)	$\tau_2$ (ns)	$f_2$ (%)	$\tau_{avg}$ (ns) <sup>1</sup>
pristine	0.69	61.47	29.00	38.53	29.69
AAI	1.42	16.27	83.73	64.28	85.15
AABr	0.99	29.49	75.40	70.51	76.40

<sup>1</sup> $\tau_{avg}$  were calculated  $\tau_i$  and  $f_i$  using the equation of  $\tau_{avg} = \frac{\sum f_i \tau_i^2}{\sum f_i \tau_i}$

Table S5 The defects of hole-only devices for pristine, AAI and AABr treated PSCs.

	$V_{TFL}$ (V)	$n_t$ (10 <sup>15</sup> ×cm <sup>-3</sup> )
pristine	0.77	10.9
AAI	0.65	9.20
AABr	0.68	9.62

Table S6 The FF loss of devices for pristine, AAI and AABr treated PSCs.

	FF <sub>S-Q limit</sub> (%)	FF <sub>max</sub> (%)	FF <sub>J-V</sub> (%)	FF loss (%)
pristine	90.1	82.5	75.1	7.4
AAI	90.1	84.3	78.0	6.3
AABr	90.1	84.4	77.4	7.0

## Reference

1. Q. Jiang, L. Zhang, H. Wang, X. Yang, J. Meng, H. Liu, Z. Yin, J. Wu, X. Zhang and J. You, *Nature Energy*, 2016, **2**, 16177.