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Electronic supplementary information

Tailoring a dynamic crystalline process during the conversion of lead-halide perovskite layer to achieve high performance solar cells

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1. Energy-dispersive X-ray spectroscopy

In order to identify the fraction of halide anions, the perovskite film surfaces were evaluated using energy-dispersive X-ray spectra (EDS) as illustrated in **Figure S1** and the compositions were calculated as shown in **Table S1**. It can be seen that no chloride component left in the prepared perovskite films. Similar to early report in the literatures [1-3]. Therefore, further composition calculation considers only the reaction between the FAI, MABr and PbI_2.

From EDS data, an average composition can be calculated to determine fraction of halide anions in the perovskite films, which are 93% for I^- and 7% for Br^-. The final perovskite formula of anion portion can be written as $FA_{1-x}MA_xPb(Br_{0.07}I_{0.93})_3$. These are approximately within our expectation. Supposing all feed-in compositions, FAI (60 mg, 0.35 mole) and MABr (6 mg, 0.05 mole) were reacted with PbI_2 fully. The final perovskite would expect to have 96% I^- and 4% Br^-. If it is not fully reacted, the fraction of I^- anion would be reduced.

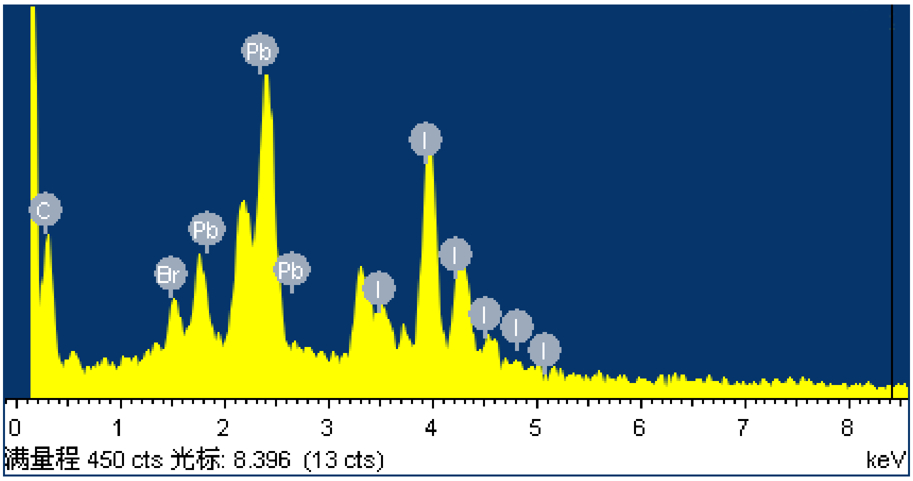

Figure S1. Energy-dispersive X-ray spectroscopy of perovskite film prepared using 1% DMSO in the second-step precursor of a sequential process.

Table S1. Atomic ratio of Br and I calculated from energy-dispersive X-ray spectra of the perovskite films

Specimen (DMSO fraction)	Atomic ratio (%)		Br/(Br+I)	I/(Br+I)
	Br	I		
0%	4.48	61.13	0.068	0.932
0.5%	4.52	56.71	0.074	0.926
1%	4.43	60.71	0.068	0.932
2%	5.15	61.97	0.077	0.923
3%	6.02	73.37	0.076	0.924
Average =			0.07 ± 0.01	0.93 ± 0.01

2. Proton nuclear magnetic resonance (^{1}H NMR)

The mixed cation composition in perovskite films prepared in this work was identified using proton nuclear magnetic resonance (^{1}H NMR) spectra, which were recorded on a 500 MHz nuclear magnetic resonance spectrometer (Bruker, AVANCE III HD 500 MHz NMR) using deuterated dimethyl sulfoxide, $(CD_3)_2SO$, as a solvent and tetramethylsilane (TMS) as an internal standard. About 15-20 perovskite films were scratched off into the powder as one specimen which was then dissolved into the $(CD_3)_2SO$. ^{1}H NMR spectra of the perovskite specimen is shown in **Figure S2**. The composition can be calculated by integrating peak at δ_H = 9.00 (N-H), 8.65 (N-H), and 7.86 (C-H) ppm for formamidinium group and δ_H = 7.49 (N-H) and 2.37 (C-H) for methylamine group. Therefore, the complete formula of the perovskite in this work is $FA_{0.85}MA_{0.15}Pb(Br_{0.07}I_{0.93})_3$.

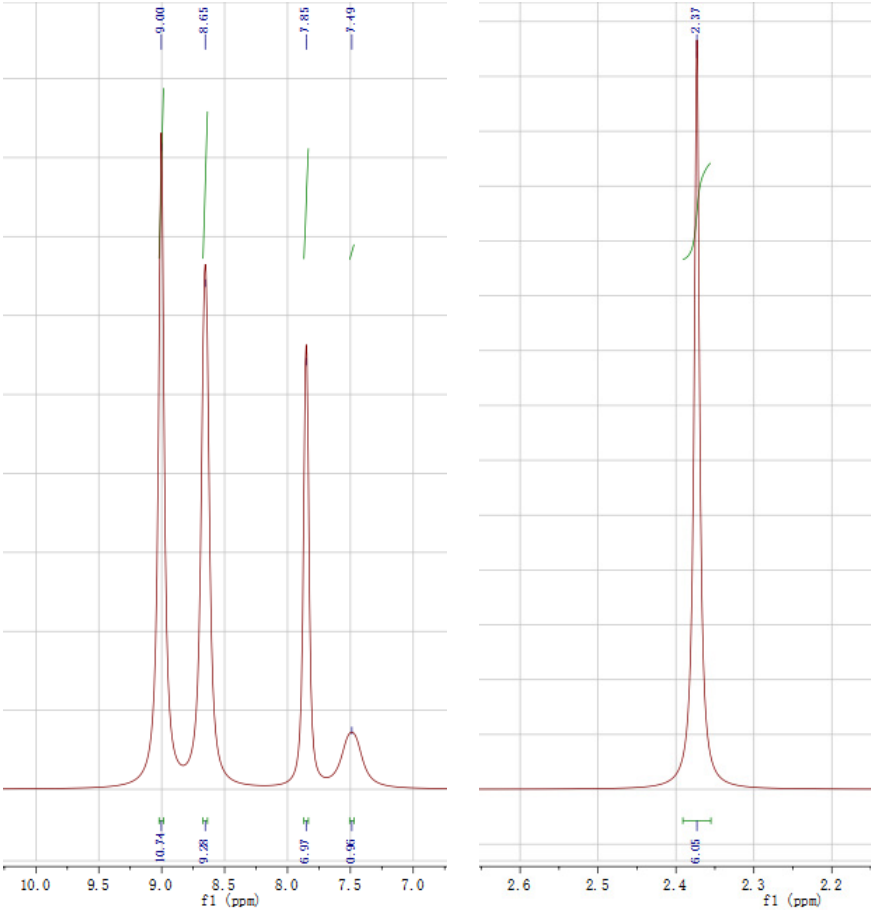

Figure S2 ^{1}H NMR spectra of perovskite specimen.

3. X-ray diffractions

The raw data of X-ray diffraction of perovskite specimen are listed in Table S2. The crystal structure was analyzed and the Miller indices were calculated carefully using crystal analyzing package. The data were found to fit well with a cubic unit cell of $Pm\bar{3}m$ (221) space group, a = b = c = 6.327 Å and cell volume of 253.3 Å, where those from PbI_2 and ITO substrate can be identified.

Table S2. X-ray data and simulation results

Simulation of Perovskite (1% DSMO)				**X-ray diffraction experimental data 2θ [°]**						**Notes**		**PbI_2 XRD data JCPDS 07-0235**		
d-spacing [Å]	**hkl**	**2θ [°]**		**0%**	**0.50%**	**1%**	**2%**	**3%**	**Intensity**			**2θ [°]**	**d-spacing [Å]**	**hkl**
				12.685	12.691	12.679	12.674	12.691	vs	PbI_2		12.672	6.980	001
6.327	100	13.986		14.007	13.994	13.988	13.978	13.964	vs					
4.474	110	19.829		19.878	19.854	19.848	19.832	19.812	m					
3.653	111	24.347		24.423	24.391	24.373	24.355	24.330	s					
												22.520	3.945	100
				25.517	25.475				w	PbI_2		25.510	3.489	002
												25.918	3.435	101
3.164	200	28.186		28.199	28.179	28.174	28.149	28.124	s					
2.830	210	31.594		31.641	31.612	31.597	31.577	31.546	m					
												34.277	2.614	102
2.583	211	34.701		34.753	34.769	34.731	34.704	34.661	w					
				37.896	37.903	37.903	37.894		vw	ITO				
				38.667	38.640	38.617	38.781		w	PbI_2		38.662	2.327	003
												39.528	2.278	110
2.237	220	40.284		40.360	40.330	40.314	40.287	40.241	m					

4. Hysteresis and of perovskite solar cells

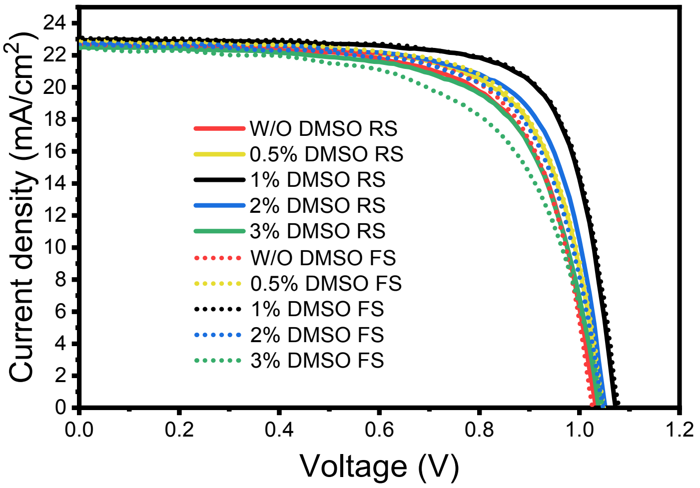

Figure S3 Hysteresis and *J–V* curves from reverse scan (V_{OC} to 0V) and forward scan (0V to V_{OC}) of perovskite solar cells prepared using different levels of DMSO additives in the second step FAI-MAX precursors.

References

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