

Supplementary information

**Composition-controlled organometal halide
perovskite via CH₃NH₃I pressure in vacuum co-
deposition process**

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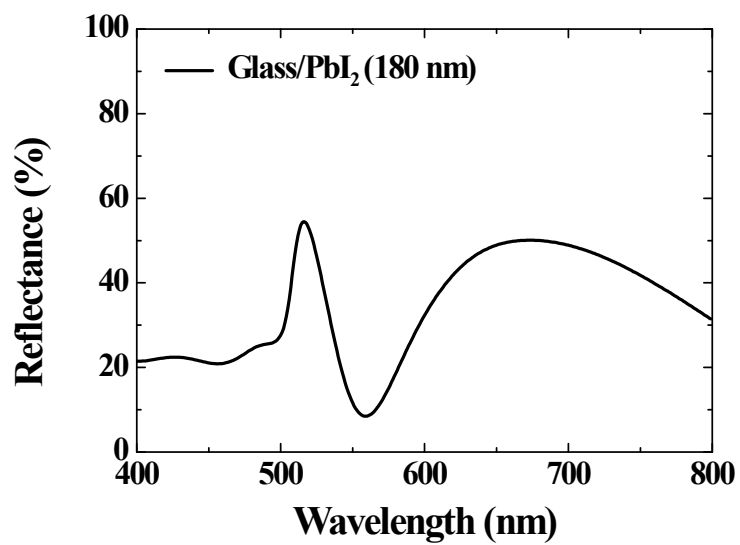


Fig. S1. Reflectance of PbI₂. The incident beam is tilted 7° toward the substrate.

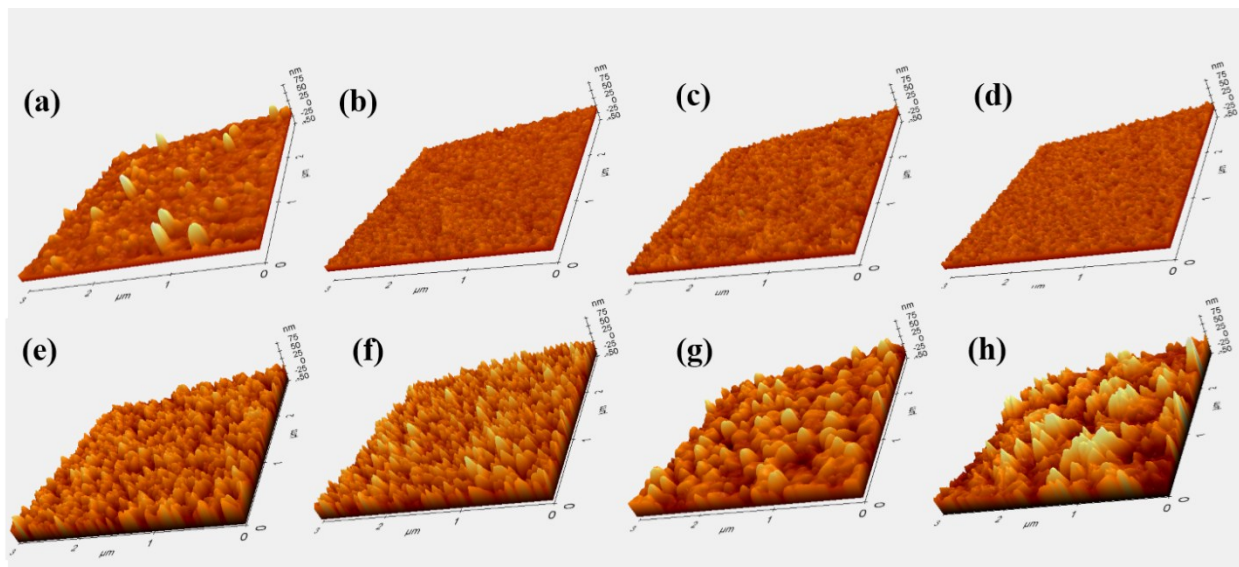


Fig. S2. Three-dimensional AFM image of the perovskite films shown in Fig. 4.

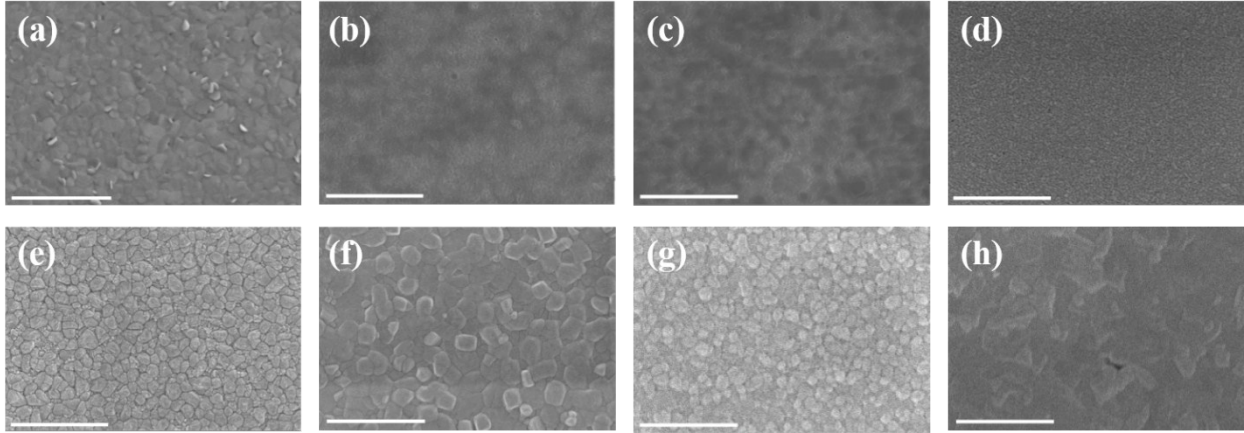


Fig. S3. Scanning electron microscopy image of the perovskite films shown in Fig. 4. The scale bars represent 1 micrometer.

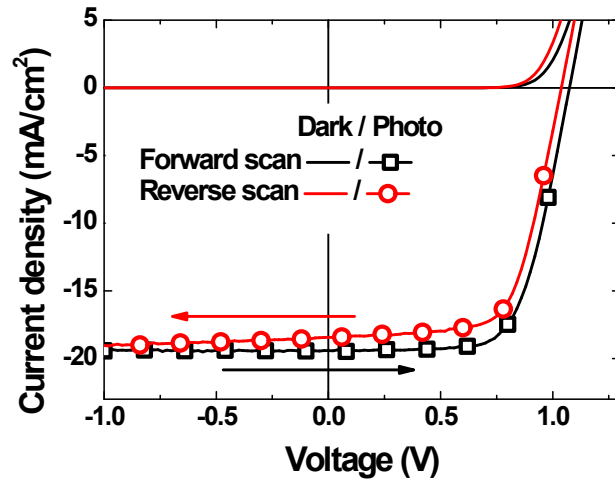


Fig. S4. Hysteresis of the solar cell fabricated under the optimized pressure, 5.1×10^{-5} torr.

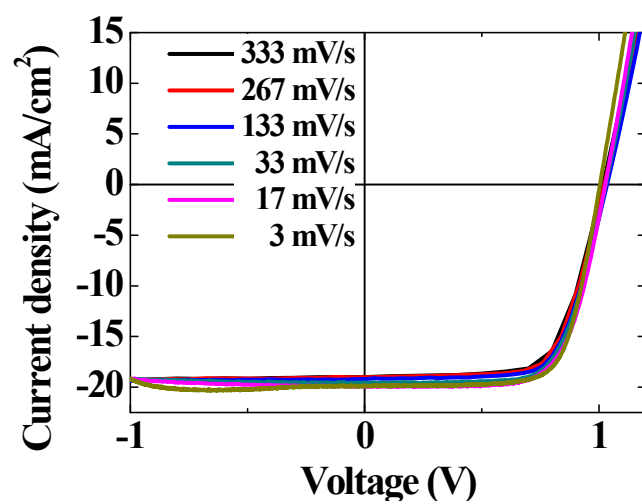


Fig. S5. J - V characteristics at the different scan rates of the device fabricated under optimized pressure, 5.1×10^{-5} torr .

Table S1. Extracted FWHMs and crystal sizes from the (110) peak of the XRD patterns shown in Fig. 3.

Pressure ($\times 10^{-5}$ torr)	0.6	1.7	3.2	4.1	5.1	6.5	8.3
FWHM (degree)	0.478	0.270	0.354	0.226	0.206	0.150	0.300
Crystal size (nm)	16.8	29.6	22.6	35.4	38.8	53.3	26.7

Table S2. Root-mean-square and peak-to-valley (R_{PV}) values of the perovskite films from the AFM images shown in Fig. 4.

Pressure ($\times 10^{-5}$ torr)	Only PbI₂	0.6	1.7	3.2	4.1	5.1	6.5	8.3
RMS (nm)	7.5	5.2	4.3	3.7	11.8	12.8	12.0	16.6
R_{PV} (nm)	118	68	39	33	85	96	90	127

Table S3. J_{SC} values obtained experimentally and J_{SC} values integrated from the IPCE spectra.

Pressure ($\times 10^{-5}$ torr)	0.6	1.7	3.2	4.1	5.1	6.5	8.3
Integrated J_{SC} (mA/cm²)	3.9	9.5	5.5	16.3	18.3	16.7	5.2
Experimental J_{SC} (mA/cm²)	3.8	8.5	9.4	17.7	19.1	12.6	1.2

Table S4. Photovoltaic parameters of the perovskite solar cell shown in Fig. S5at the different scan rates.

Scan rate (mV/s)	333	267	133	33	17	3
PCE	13.2	13.4	13.6	14.0	14.2	14.5
J_{SC} (mA/cm²)	19.0	19.0	19.2	19.53	19.90	19.93
V_{OC} (V)	1.08	1.06	1.01	1.02	1.03	1.00
FF	0.64	0.67	0.71	0.70	0.69	0.72

Table S5. Atomic percentages and ratios in the films from EDX data.

Pressure ($\times 10^{-5}$ torr)	At.% of Pb (%)	At.% of I (%)	Pb:I
Only PbI ₂	17.49	8.66	1:2.02
0.6	7.29	15.74	1:2.16
1.7	6.77	14.89	1:2.20
3.2	6.73	15.31	1:2.27
4.1	7.71	19.86	1:2.58
5.1	6.61	18.65	1:2.82
6.5	6.49	19.12	1:2.95
8.3	6.51	19.87	1:3.05