Supporting Information

High-Throughput Large-Area Vacuum Deposition for High-Performance Formamidine-based Perovskite Solar Cells

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Perovskite stoichiometry

The ratio of different elements within perovskite film was calculated by XPS (Fig. S13) as following equation:

$$\frac{n_i}{n_j} = \frac{I_i}{I_j} \times \frac{\sigma_j}{\sigma_i} \times \frac{E_{kj}{}^{0.5}}{E_{ki}{}^{0.5}}$$

where n is the atomic number, I is peak area of element, σ is the photoionization cross-sections of elements, and E_k is photoelectron kinetic energy. The stoichiometry for Cs:N:Pb:I is 0.15:1.69:0.99:3. Thus, the perovskite formula can be approximately described as $Cs_{0.15}FA_{0.85}PbI_3$.

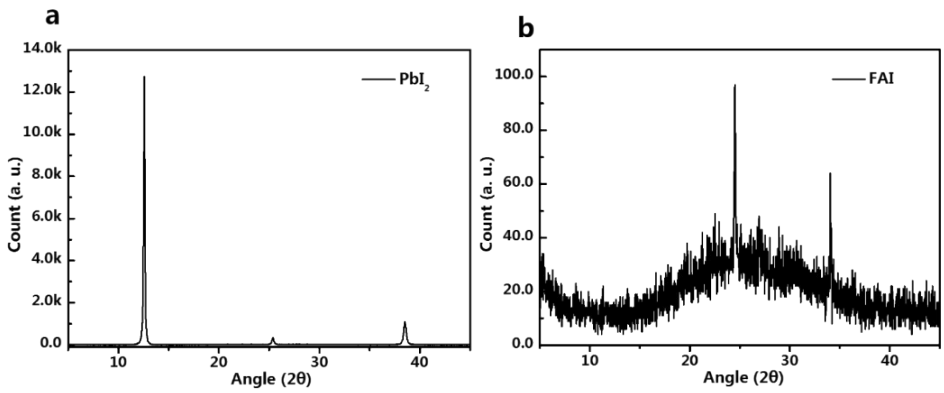

Fig. S1. The XRD patterns of (a) PbI_2 and (b) FAI deposited by vacuum evaporation.

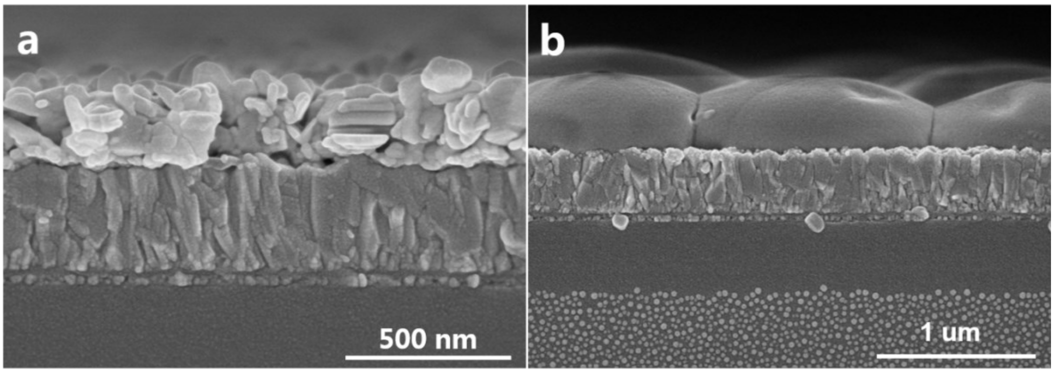

Fig. S2. Cross-sectional SEM images of (a) PbI_2 and (b) FAI.

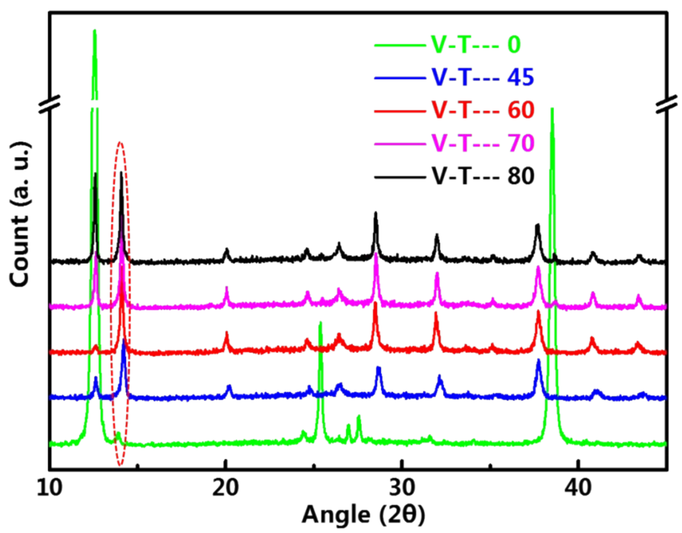

Fig. S3. XRD patterns of the precursor with different annealing temperatures.

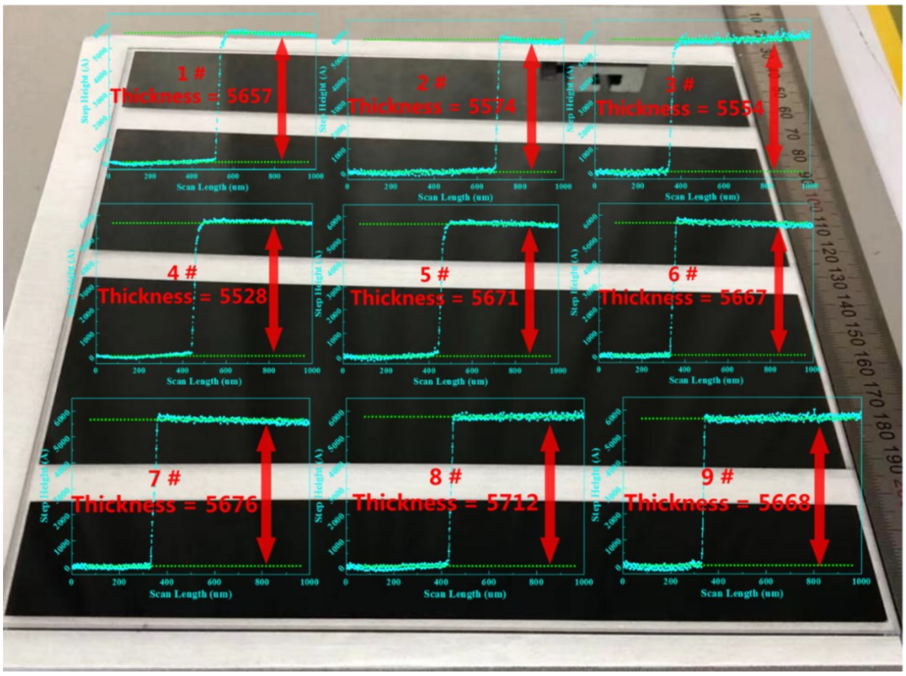

Fig. S4. The thickness of the large area perovskite film (400 cm^2) with 9 regions.

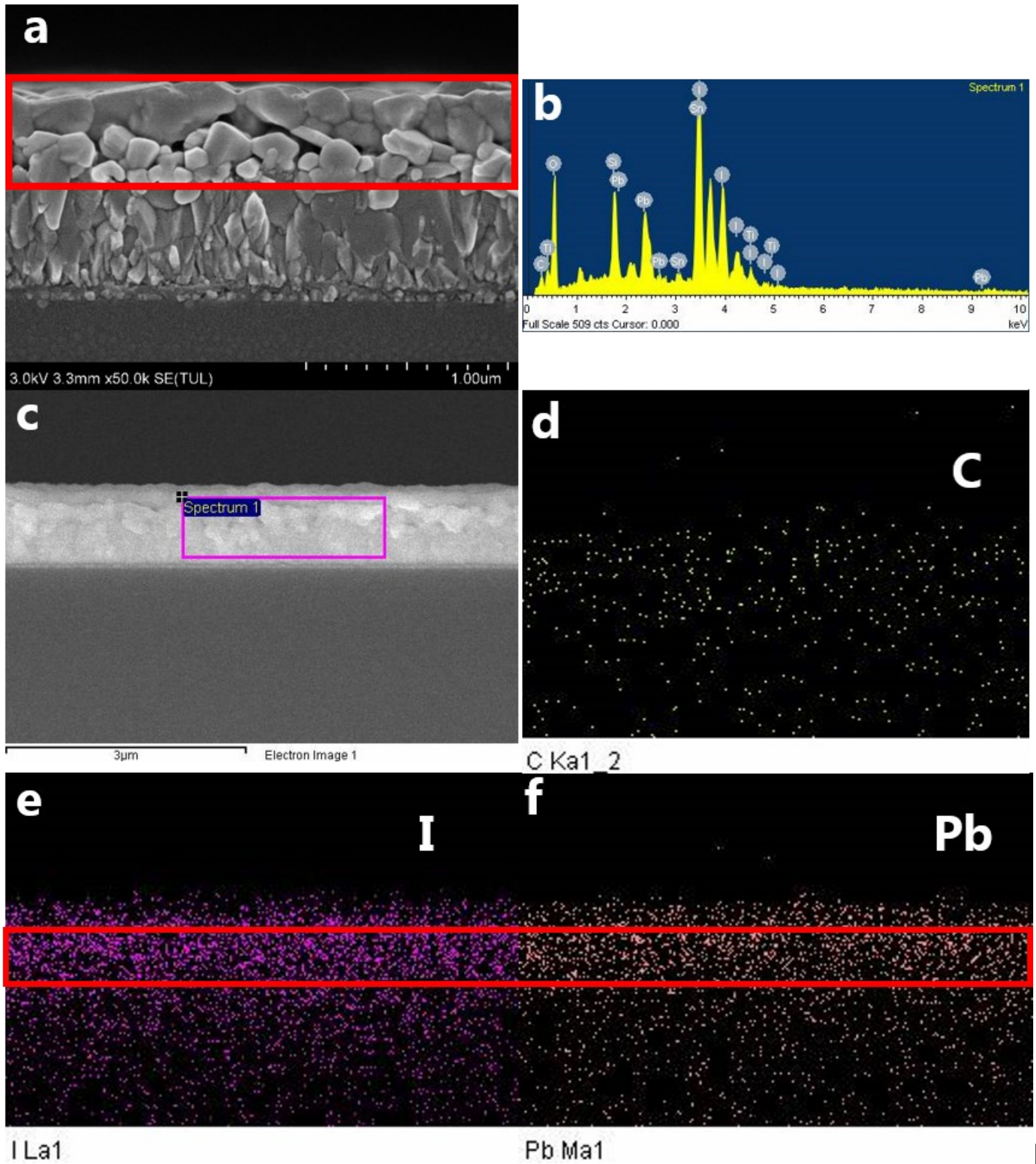

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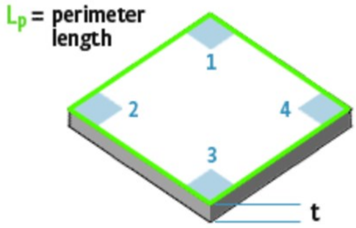
Fig. S5. (a) Cross sectional SEM of film at low annealing temperature, the red square area is unreacted PbI2 layer. (b) EDS spectrum and (c) EDS selection of film at low annealing temperature. EDS of (d) C, (e) I and (f) Pb elements at the selected area.

Sample	
ID:	
Type:	van der Pauw
Thickness	
t [nm]:	550
Other dimensions	
Lp [mm]:	28
Hall factor:	1
Max voltage [V]:	20
Max current [mA]:	20
Gate bias voltage [V]:	0
Comment:	



Final results

		Mean value	Limit
μ_H	Hall mobility [m²/V·s]	1.3265E-2	
	Carrier type	P	100 %
n	Carrier concentration [1/m³]	4.2162E14	
n_{sheet}	Sheet carrier concentration [1/m²]	2.3189E8	
R_H	Hall coefficient [m³/C]	1.4804E4	
R_{Hsheet}	Sheet Hall coefficient [m²/C]	2.6916E10	
ρ	Resistivity [Ω·m]	1.1160E6	
ρ_{sheet}	Sheet resistivity [Ω/□]	2.0290E12	
V_H	Hall voltage [V]	1.5833E-2	
	Phase [deg.]		
	Worst case Ohmic check correlation (2-3)	9.8927E-1	

Fig. S6. Hall measurement results of perovskite film deposited on glass substrate.

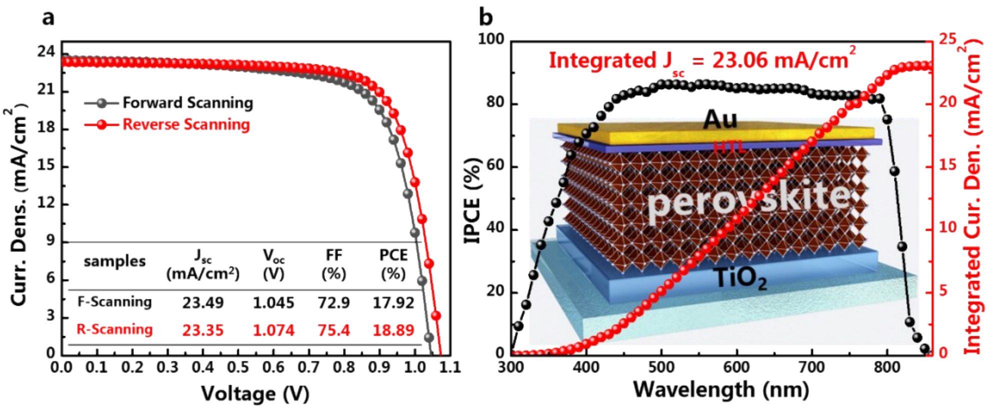


Fig. S7. (a) J-V curves of champion device scanned at reverse and forward directions. (b) IPCE and integrated photocurrent with the AM 1.5G photon flux for champion device.

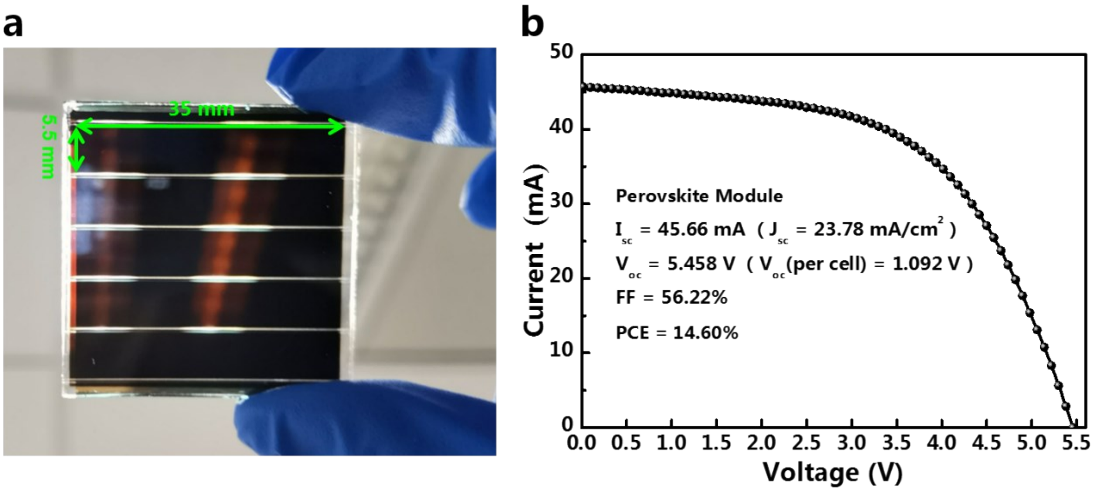


Fig. S8. (a) Photography and (b) J-V curve of perovskite module with the active area of 9.6 cm^2.

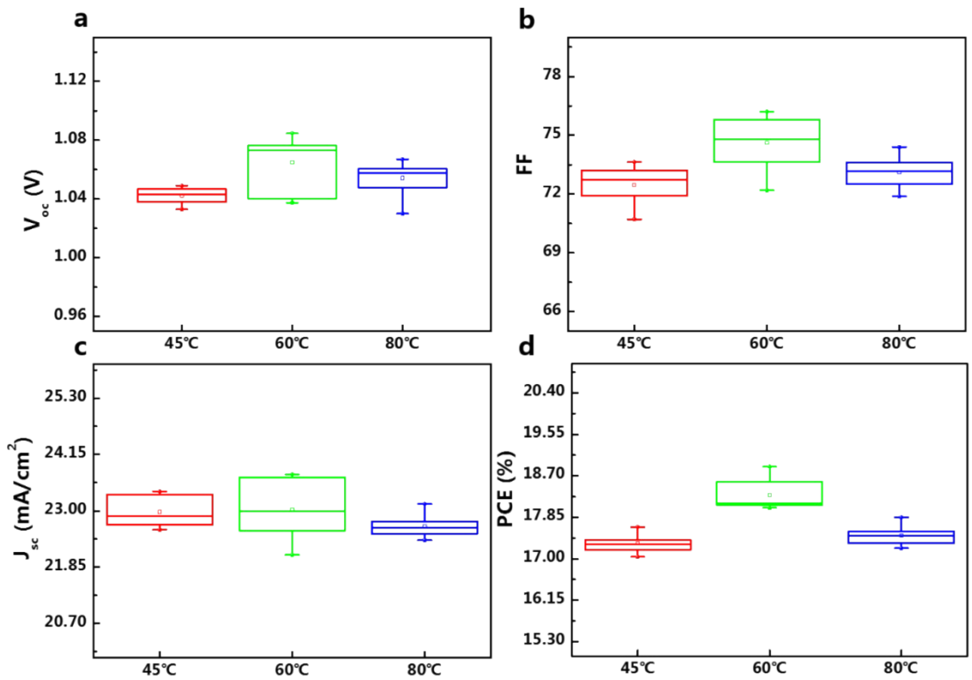


Fig. S9. Boxplot distribution of perovskite devices at annealing temperatures 45°C, 60°C and 80°C.

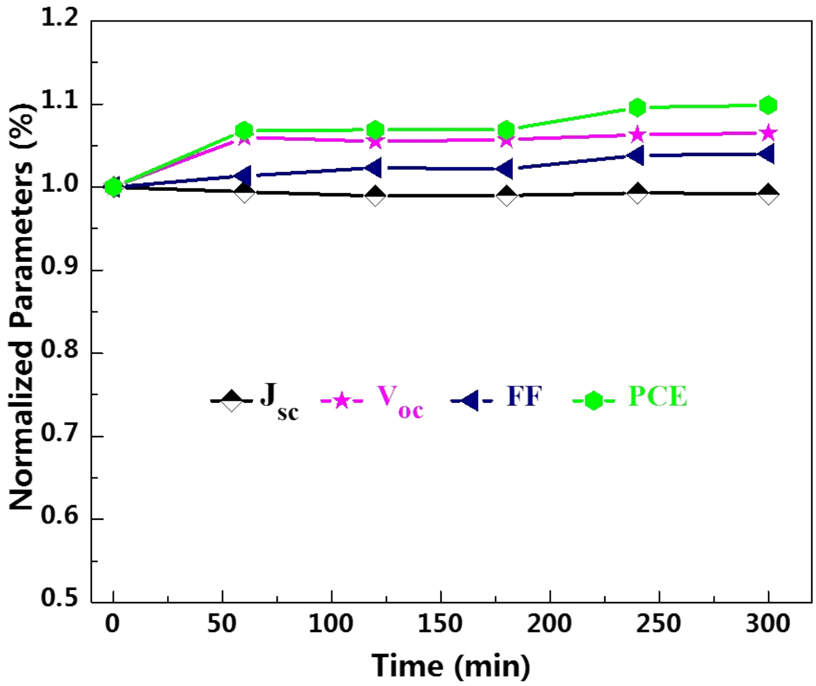


Fig. S10. The device parameters vs. test time under continuous AM 1.5G illumination.

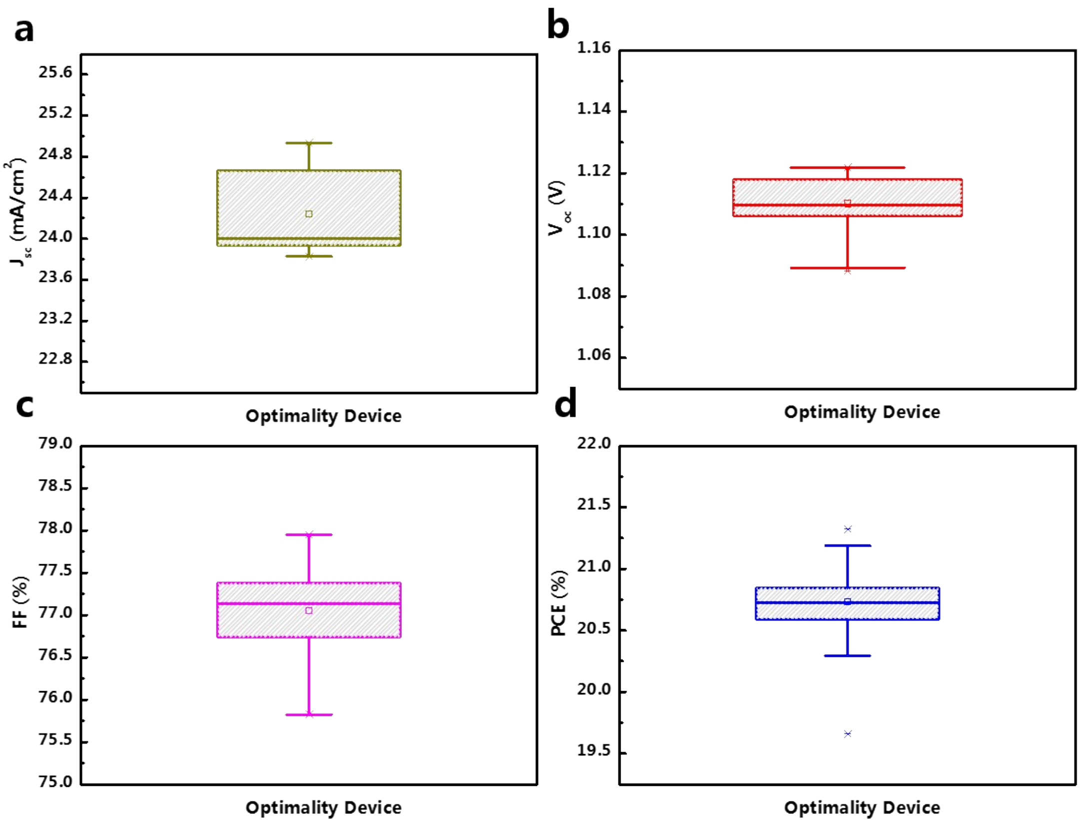


Fig. S11. Boxplot distribution of 34 perovskite devices.

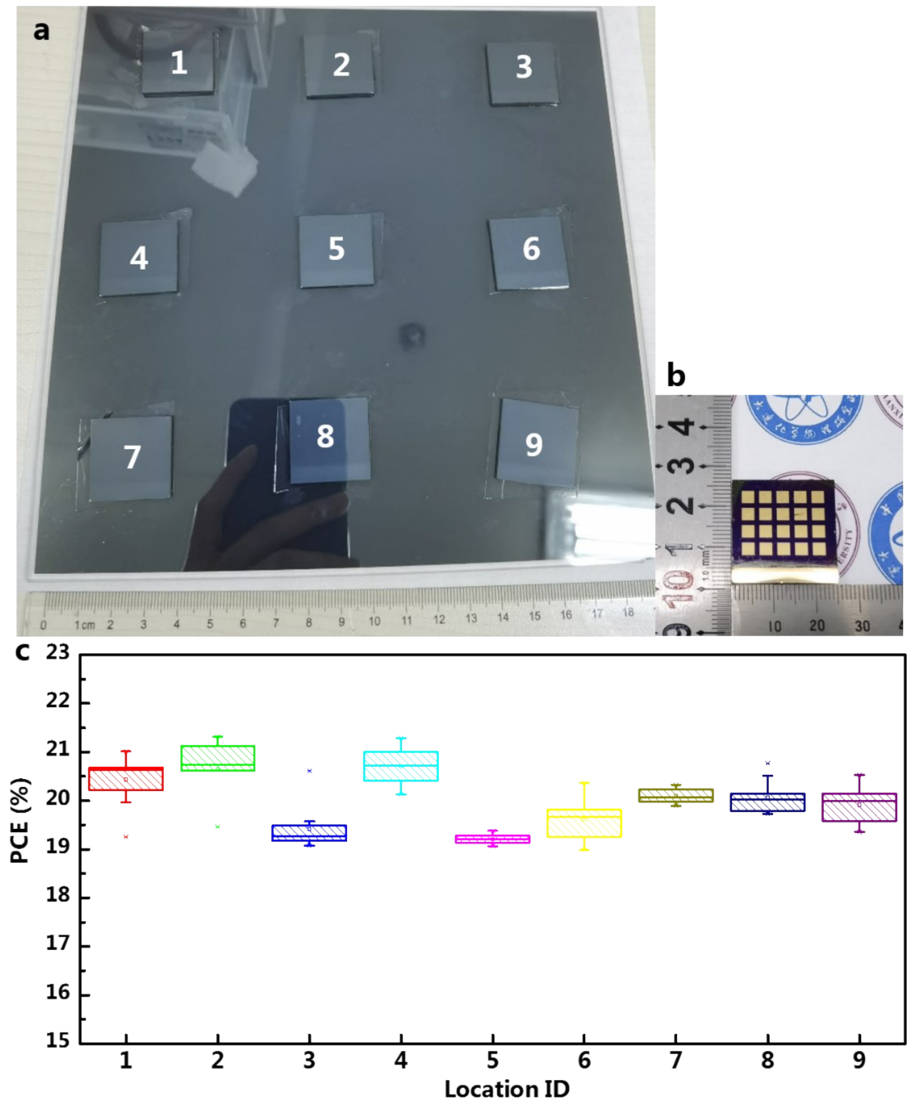


Fig. S12. The process of preparing large area perovskite films. (a) Formed perovskite films at different positions. (b) Fabrication of perovskite devices using 9 samples located at different positions. (c) Boxplot distribution of devices at 9 different locations.

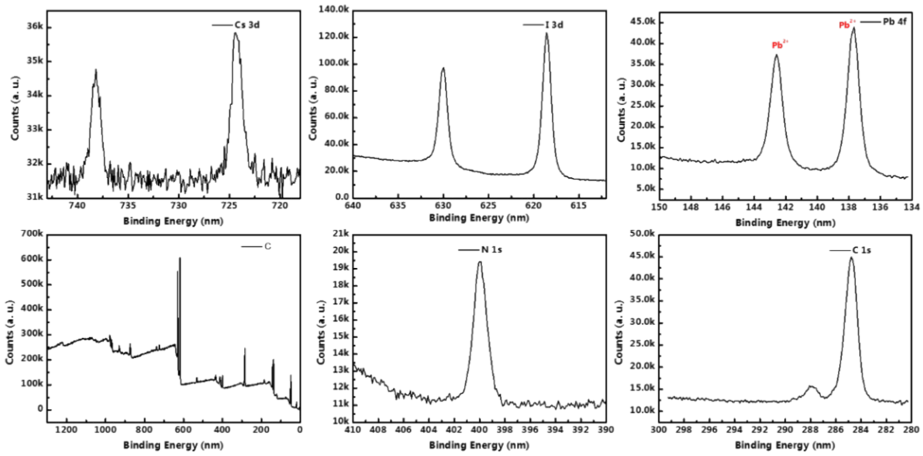


Fig. S13. XPS spectra of the perovskite film.

Table S1. Fitting parameters of TRPL of perovskite films with different annealing temperatures.

Samples	**τ_1 (ns)**	**A_1 of τ_1 (%)**	**τ_2 (ns)**	**A_2 of τ_2(%)**	**τ_{ave} (ns)**
V-T --- 45	44.1	22.4	3.3	77.6	12.5
V-T --- 60	145.7	78.9	5.6	21.1	116.2
V-T --- 80	121.2	39.2	2.01	60.7	48.7

Table S2. Fitting parameters of EIS for perovskite devices based on different annealing temperatures.

Devices	**R_s (Ω)**	**R_{rec} (Ω)**	**C_g (F)**	**R_{dr} (Ω)**	**C_{dr} (F)**
V-T --- 45	1.88	536	8.43×10^{-8}	113.6	8.87×10^{-7}
V-T --- 60	1.68	3715	7.91×10^{-8}	86.3	4.79×10^{-8}
V-T --- 80	1.67	1649	9.23×10^{-8}	586.1	7.3×10^{-8}