Electronic Supplementary Material (ESI) for Materials Chemistry Frontiers.

Supporting Information

$CsPbI_{2.69}Br_{0.31}$ Solar Cells from A Low-Temperature Fabrication

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Experimental section

Materials characterization

Absorption spectra for perovskite films were recorded on a Shimadzu UV-1800 spectrophotometer. Wide angle X-ray diffraction (XRD) patterns were obtained on D/MAX-TTRIII (CBO) with Cu Kα radiation (λ = 1.542 Å) operating at 40 kV and 200 mA. Field emission scanning electron microscopy (FESEM) was performed on Hitachi SU-8220 operating at 5.0 kV. Atomic force microscopy (AFM) was performed on a Multimode microscope (Veeco) using tapping mode and a XE-7 scanning probe microscope in noncontact mode (Park Systems, Korea). Film thicknesses were measured with a profilometer (KLA Tencor D-120). XPS was measured on ESCALAB 250Xi.

Preparation of the precursor solution

Inorganic cesium lead iodide perovskite ($CsPbI_3$) precursor solution was prepared by dissolving 260 mg of cesium iodide (99.999%, Strem) and 461 mg of lead iodide (99%, Acros) in 1 mL of N,N-Dimethylformamide (DMF). The solution was stirred at room temperature for 12 hours in the nitrogen glovebox. Then different amounts of hydrobromic acid (25 μL/mL, 30 μL/mL, 35 μL/mL, 40 μL/mL, and 45 μL/mL, 48 wt.% in water, Aladdin) or hydroiodic acid (33 μL/mL, 57.0 wt.% in water, Aladdin)[1] were added into the solution with continuously stirring at room temperature for 1 hour before using.

Device fabrication and measurements

Patterned ITO glass with a sheet resistance of 15 Ω sq^{-1} was cleaned by ultrasonics in detergent, deionized water, acetone, isopropanol sequentially and then treated with UV-ozone for 10 min. The poly(3,4-ethylenedioxythiophene)-polystyrene sulfonate (PEDOT:PSS, Clevios™ PVP Al 4083) layer was formed by spin coating an aqueous dispersion onto ITO glass (4000 rpm for 30 s). PEDOT:PSS coated substrates were dried at 150 °C for 10 min, and then transferred into a N_2 glovebox. The perovskite

precursor solution was spin-coated onto PEDOT substrates at 3000 rpm for 30 s, and then annealed at different temperature (60°C, 80°C, 100°C, 120°C, and 140°C). $PC_{61}BM$ solution (20 mg/mL in chlorobenzene) was then spin-coated onto perovskite layer at 1500 rpm for 30 s. Finally, aluminum (100 nm) was deposited onto $PC_{61}BM$ layer through a shadow mask under vacuum (ca. 10^{-4} Pa). The effective area for the devices is 4 mm^2. *J-V* curves were measured by using a computerized Keithley 2400 SourceMeter and a Xenon-lamp-based solar simulator (Enli Tech, AM 1.5G, 100 mW/cm^2). The illumination intensity of solar simulator was determined by using a monocrystalline silicon solar cell (Enli SRC2020, 2 cm×2 cm) calibrated by NIM. The external quantum efficiency (EQE) was measured by using a QE-R3011 measurement system (Enli Tech).

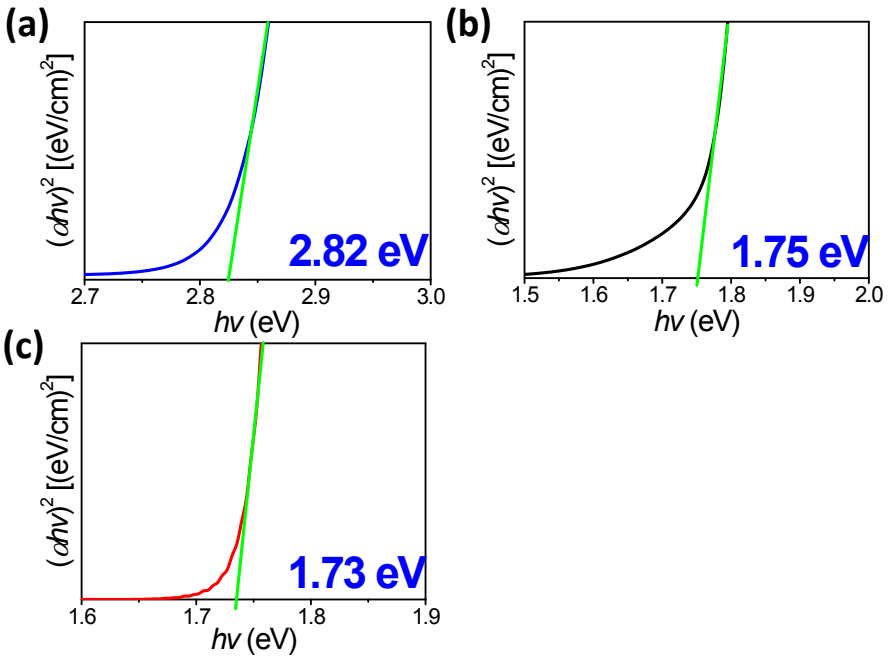

Figure S1 Tauc plots for perovskite films made without additive (a), with HBr additive (b) and with HI additive (c).

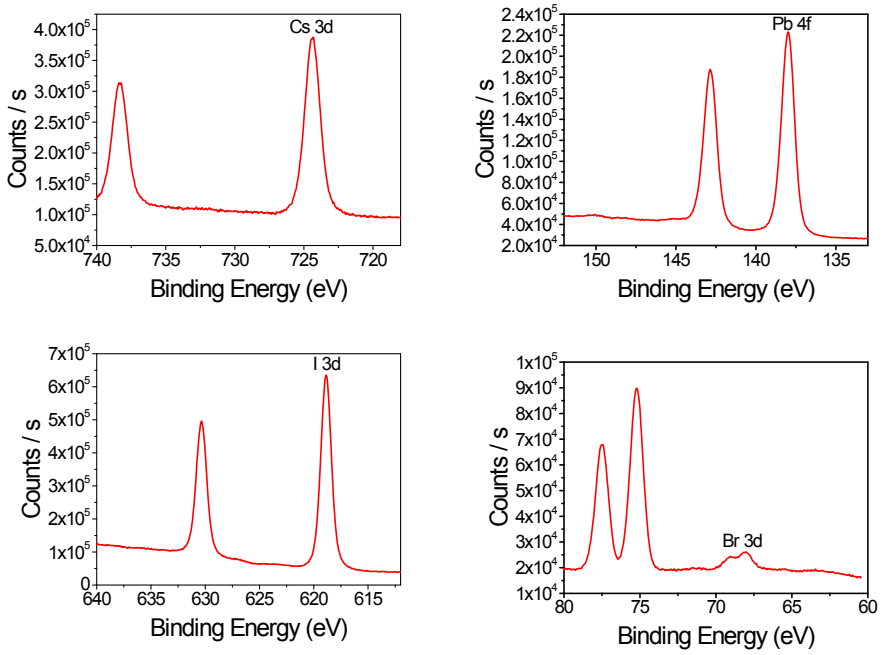

Figure S2 XPS spectra of $CsPbI_{2.69}Br_{0.31}$ film.

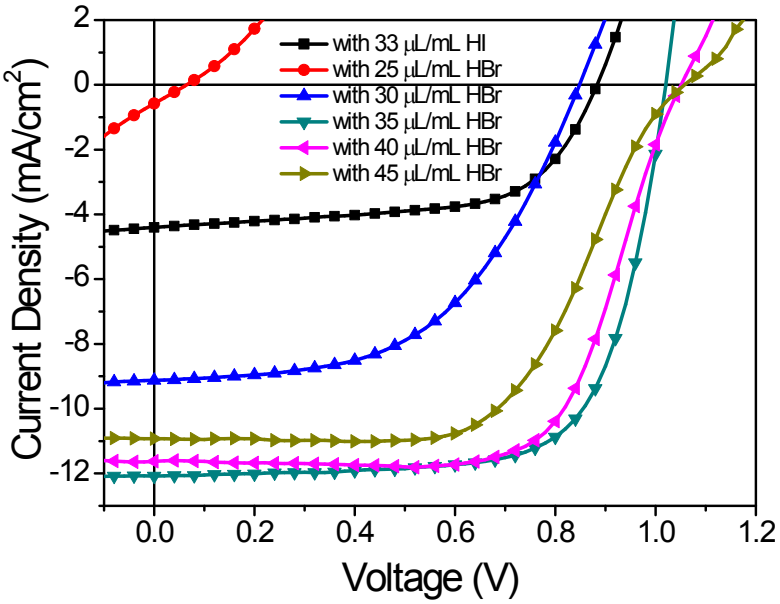

Figure S3 *J-V* curves for PSCs made with different additive concentrations (annealing temperature 100 °C, annealing time 10 min).

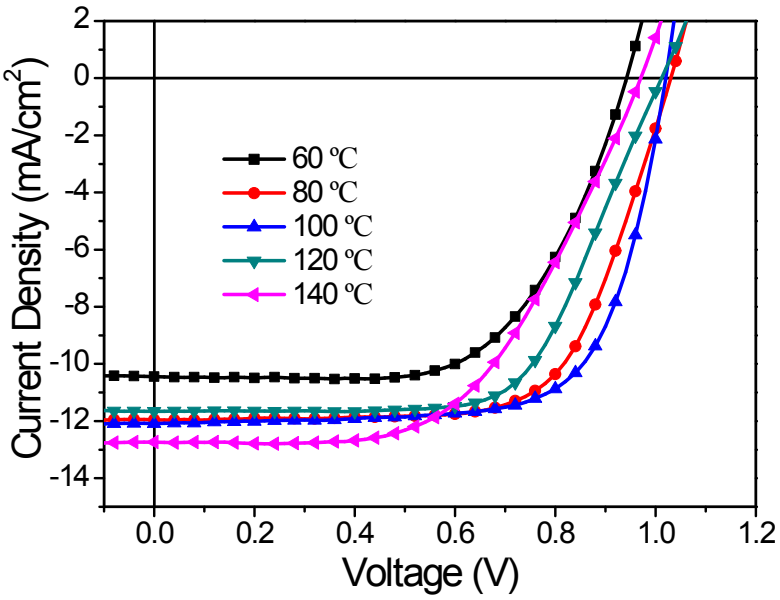

Figure S4 *J-V* curves for PSCs with different annealing temperature (35 μL/mL HBr, annealing time 10 min).

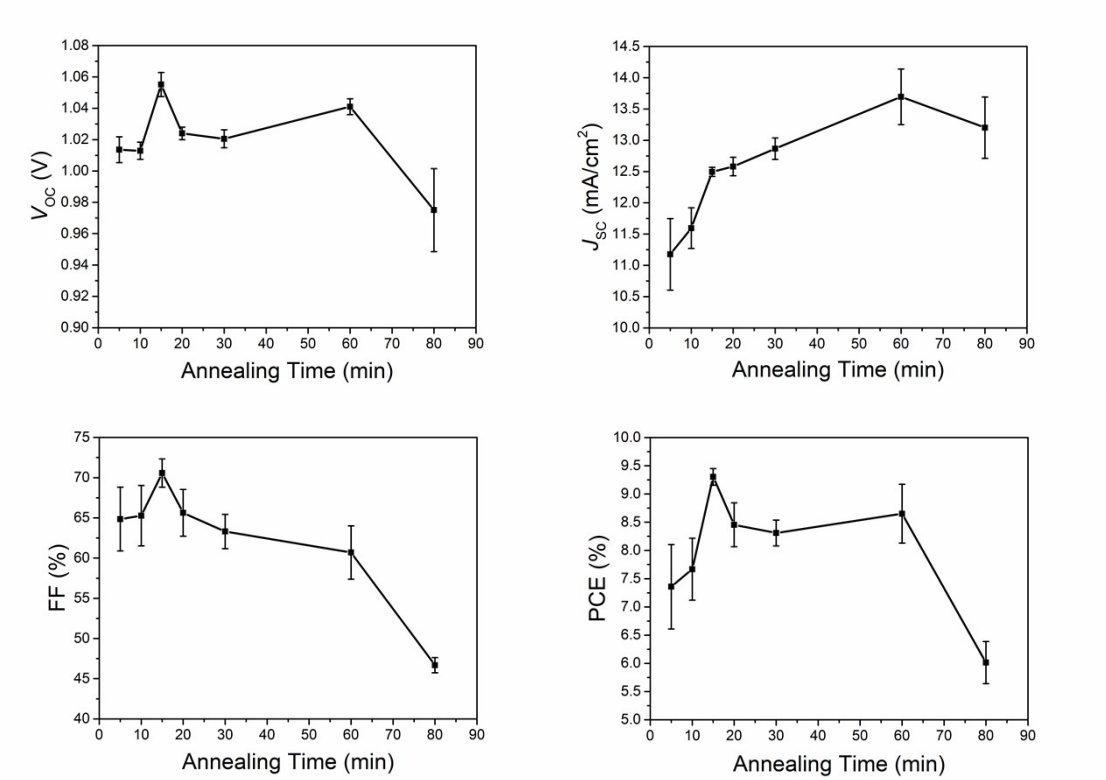

Figure S5 The performance parameters (V_{OC}, J_{SC}, FF, PCE) of $CsPbI_{2.69}Br_{0.31}$ solar cells plotted against the annealing time.

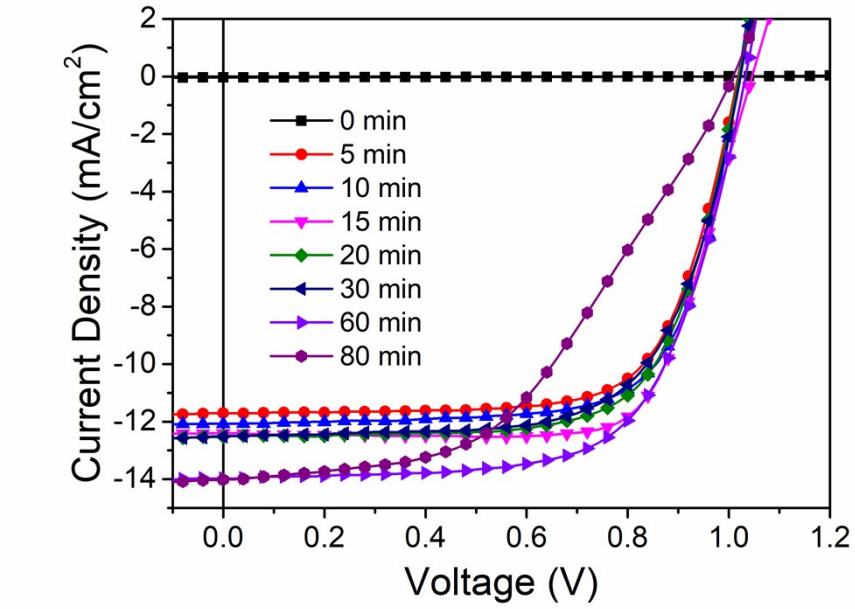

Figure S6 The *J-V* curves for devices with different annealing time (35 μL/mL HBr, annealing temperature 100 °C).

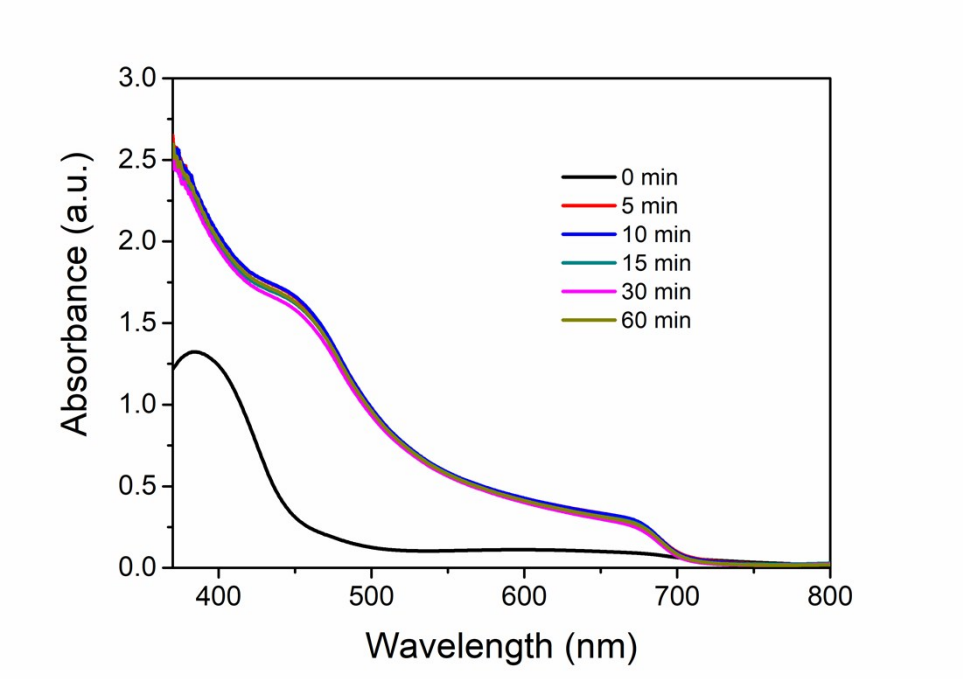

Figure S7 The absorption spectra for $CsPbI_{2.69}Br_{0.31}$ films with different annealing time.

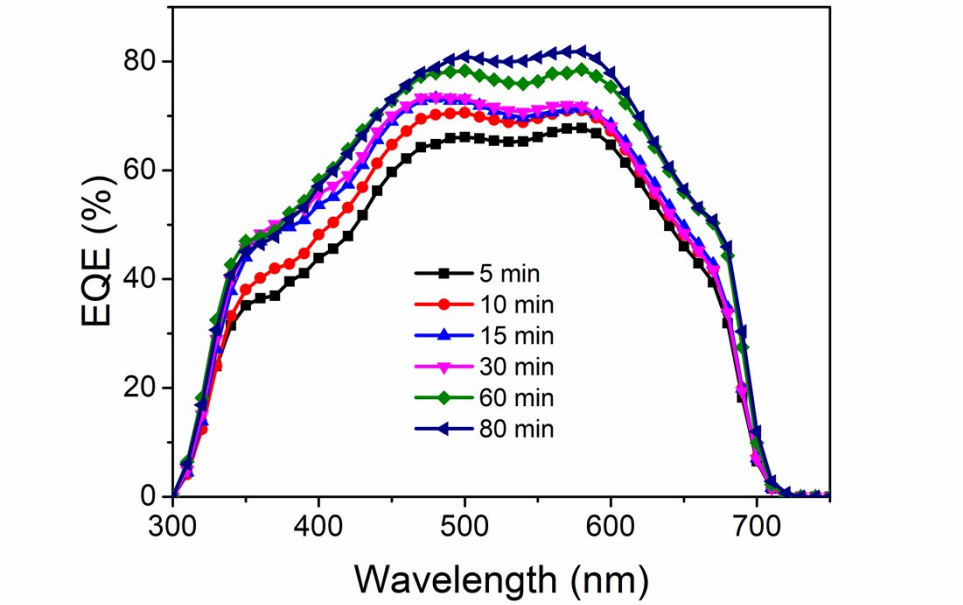

Figure S8 EQE spectra for $CsPbI_{2.69}Br_{0.31}$ solar cells with different annealing time.

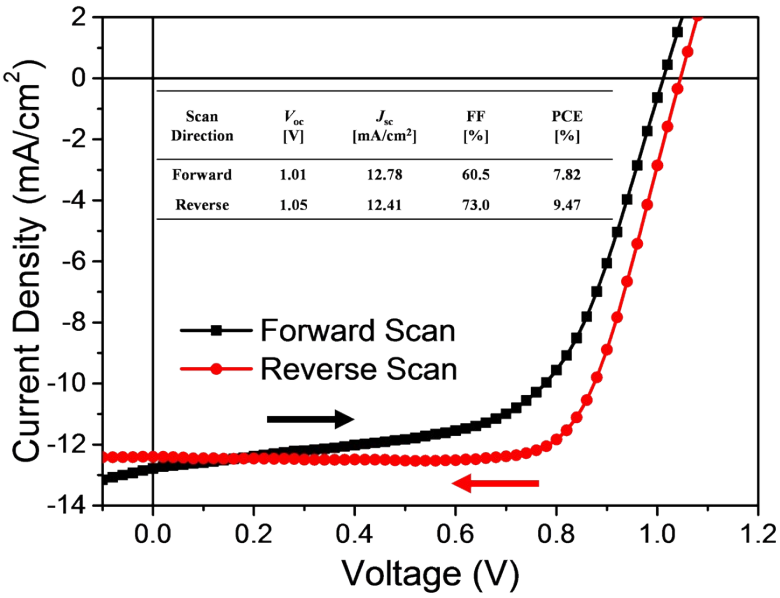

Figure S9 *J-V* curves (forward and reverse) and performance data (inset) for the $CsPbI_{2.69}Br_{0.31}$ solar cell.

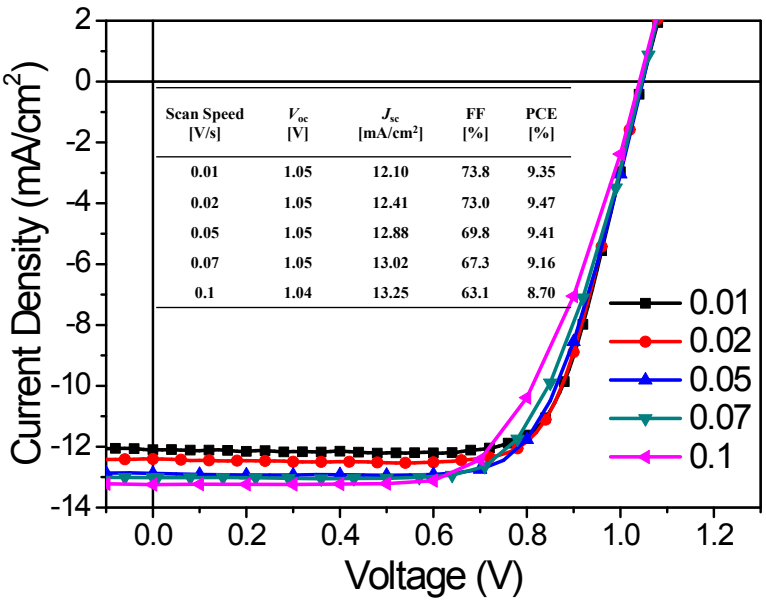

Figure S10 *J-V* curves and performance data (inset) under different scan speeds (0.01 V/s; 0.02 V/s; 0.05 V/s; 0.07 V/s; 0.1 V/s) for the $CsPbI_{2.69}Br_{0.31}$ solar cell.

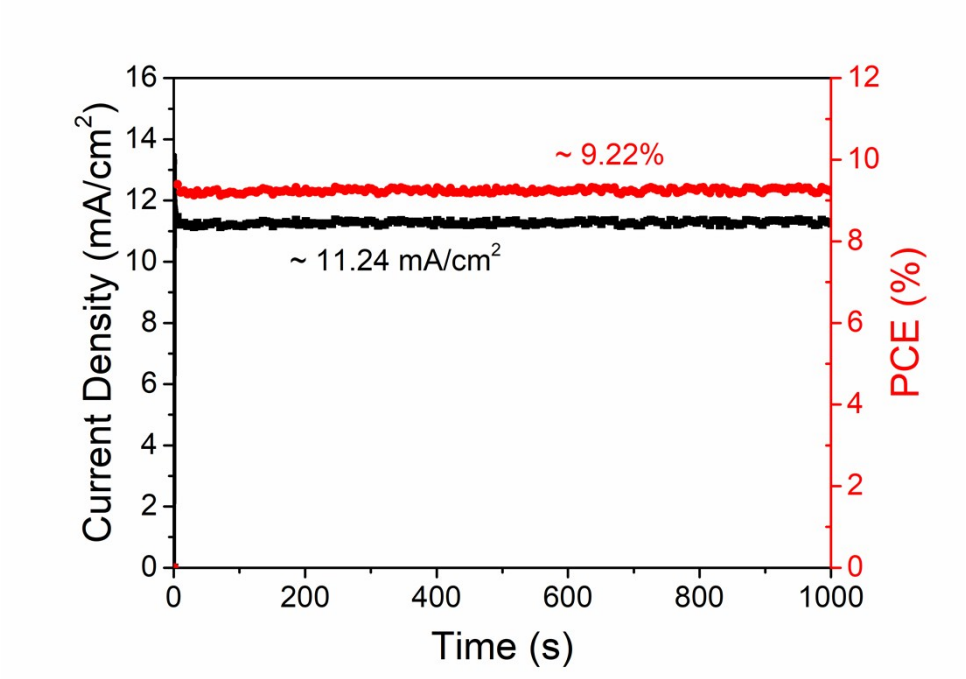

Figure S11 Photocurrent measured under 0.82 V bias near the maximum power point and PCE (active area 4 mm^2).

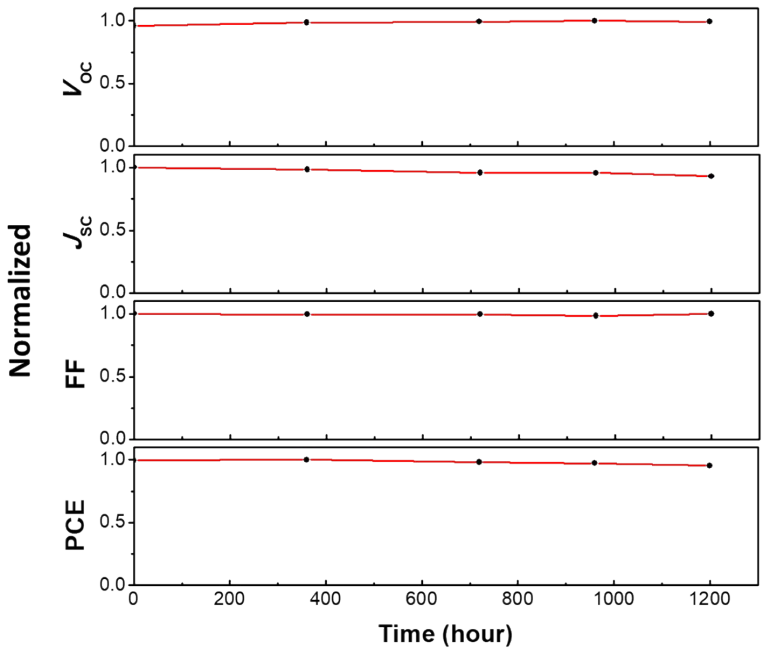

Figure S12 Tracking the device performance in a N_2 glovebox for 1200 hours.

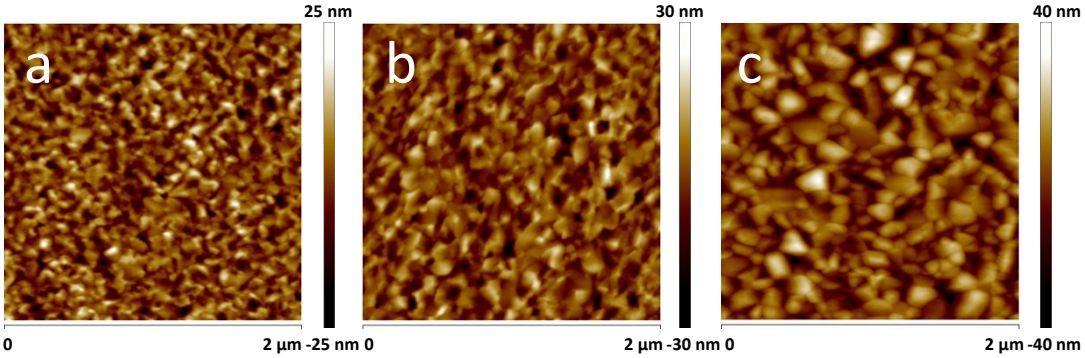

Figure S13 AFM height images for $CsPbI_3$ films with HBr additive annealed at 100 °C for 20 s (a), 15 min (b) and 60 min (c).

Table S1 Optimization of HBr concentration (annealing temperature 100 °C, annealing time 10 min).

HBr [μL/mL]	V_{oc} [V]	J_{sc} [mA/cm^2]	FF [%]	PCE [%]
25	0.06	0.58	24.3	0.01 (0.00)[a]
30	0.85	9.12	52.8	4.08 (3.61)
35	1.02	12.08	70.7	8.71 (7.67)
40	1.05	11.63	68.5	8.37 (7.44)
45	1.06	10.93	59.2	6.85 (6.85)

[a] Data in parentheses stand for the average PCEs for 8 cells.

Table S2 Optimization of perovskite film thickness (35μL/mL HBr, annealing temperature 100 °C, annealing time 10 min).

Thickness [nm]	V_{oc} [V]	J_{sc} [mA/cm^2]	FF [%]	PCE [%]
102	0.99	9.95	67.1	6.63 (6.32)[a]
167	1.02	12.08	70.7	8.71 (7.67)
223	0.88	7.79	60.0	4.12 (3.84)

[a] Data in parentheses stand for the average PCEs for 8 cells.

Table S3 Optimization of annealing temperature (35 μL/mL HBr, annealing time 10 min).

T [°C]	V_{oc} [V]	J_{sc} [mA/cm^2]	FF [%]	PCE [%]
60	0.94	10.45	62.8	6.18 (5.63)[a]
80	1.03	11.95	67.7	8.34 (7.62)
100	1.02	12.08	70.7	8.71 (7.67)
120	1.01	11.66	65.1	7.67 (6.46)
140	0.97	12.73	55.7	6.89 (6.31)

[a] Data in parentheses stand for the average PCEs for 8 cells.

References

[1] G. E. Eperon, G. M. Paternò, R. J. Sutton, A. Zampetti, A. A. Haghighirad, F. Cacialli and H. J. Snaith, *J. Mater. Chem. A*, 2015, **3**, 19688-19695.