

## **Clothe CsPbI<sub>3</sub> perovskite in robust phase-pure armor to boost the thermal stability**

Xue Tan<sup>a</sup>, Shubin Wang<sup>a\*</sup>, Qixian Zhang<sup>a</sup>, Hailiang Wang<sup>a</sup>, Huicong Liu<sup>a</sup>, Weiping Li<sup>a</sup>, Liqun Zhu<sup>a</sup>, Tinglu Song<sup>b</sup>,  
Zhenhua Cui<sup>c</sup>, Yang Bai<sup>c</sup>, Haining Chen<sup>a\*</sup>

a. School of Materials Science and Engineering, Beihang University, No. 37 Xueyuan Road, Haidian District, Beijing 100083, People's Republic of China.

b. Experimental Center of Advanced Materials, School of Materials Science & Engineering, Beijing Institute of Technology, Beijing 100081, People's Republic of China.

c. Beijing Key Laboratory of Construction Tailorable Advanced Functional Materials and Green Applications, MIIT Key Laboratory for Low-dimensional Quantum Structure and Devices, Experimental Center of Advanced Materials, School of Materials Science & Engineering, Beijing Institute of Technology, Beijing 100081, People's Republic of China.  
\*E-mail: shubinwang@buaa.edu.cn; chenhaining@buaa.edu.cn

## Supporting information

**Materials:**

CsI (99.9%), PbI<sub>2</sub> (99.99%) and DMAI (99.5%) was purchased from Xi'an Polymer Light Technology Corp. DMF (99.8%, extra dry) was purchased from Acros Organics. Titanium diisopropoxide bis(acetylacetonate) (75 wt. % in isopropanol) were purchased from TCI (Shanghai) Development Co., Ltd. Commercial TiO<sub>2</sub> paste (30 NR-D) was purchased from Dyesol company. 1-Butanol (99%) and ethanol (99.5%) were purchased from Shanghai Aladdin Biochemical Technology Co. Ltd. The carbon paste (DD-20) was purchased from Guangzhou Seaside Technology Co. ltd. All chemicals were used as-received without further purification.

**Deposition of TiO<sub>2</sub> films:**

FTO glass was cleaned with successive sonication in deionized water, ethanol and isopropanol. TiO<sub>2</sub> blocking layer was then spin coated onto FTO glass at 2000 rpm for 20 s, using a titanium diisopropoxide bis(acetylacetonate) solution in 1-butanol (0.15 M), and then heated at 120 °C for 5 min. TiO<sub>2</sub> mesoporous scaffolds were deposited by spin coating at 5000 rpm for 30 s using a commercial TiO<sub>2</sub> paste dispersed in ethanol, followed by sintering at 100 °C for 5 min and then at 550 °C for 30 min.

**Deposition of perovskite films:**

Precursor solutions were prepared by mixing DMAI, PbI<sub>2</sub> and CsI in DMF. The molar ratio of DMAI: PbI<sub>2</sub>: CsI was 1.5:1.5:1 and the concentration of CsI in the solution was set at 1 M. The intermediate films were deposited by spin coating the precursor solution on substrate at 2000 rpm for 20 s, followed by heating at 100 °C for 30s. For the CsPbI<sub>3</sub> film, the intermediate film was directly annealed at 220 °C for 5 min without other treatments. For the IT CsPbI<sub>3</sub> film, the intermediate film was treated with the isopropanol solution of QCDI and then annealed at 220 °C for 5 min. The PT CsPbI<sub>3</sub> film was obtained by treating the surface of the final CsPbI<sub>3</sub> film with the QCDI solution and then annealed at 100 °C for 5 min. For the QCdPbI<sub>3</sub> film, the solution of QCDI and PbI<sub>2</sub> with equal moles dissolved in DMF/DMSO (1:1) was spin-coated on FTO substrate and then annealed at 150 °C for 5 min. For the DMAPbI<sub>3</sub> film, the solution of DMAI and PbI<sub>2</sub> with equal moles dissolved in DMF was spin-coated on FTO substrate and then annealed at 150 °C for 5 min. For the Cs<sub>4</sub>PbI<sub>6</sub> film, the solution of CsI and PbI<sub>2</sub> with molar ratio of 4:1 dissolved in DMF was spin-coated on FTO substrate and then annealed at 150 °C for 5 min.

**Deposition of carbon electrodes:**

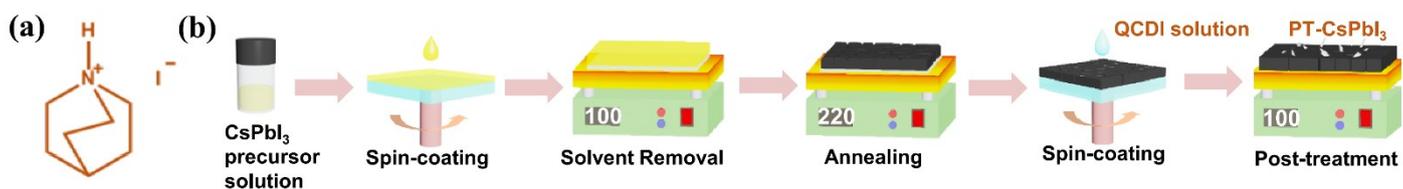
For fabricating C-PSCs, carbon electrode was directly painted on the perovskite films at room temperature, followed by annealing at 100 °C for 20 min. The whole procedure was conducted in dry air atmosphere.

**Characterizations:**

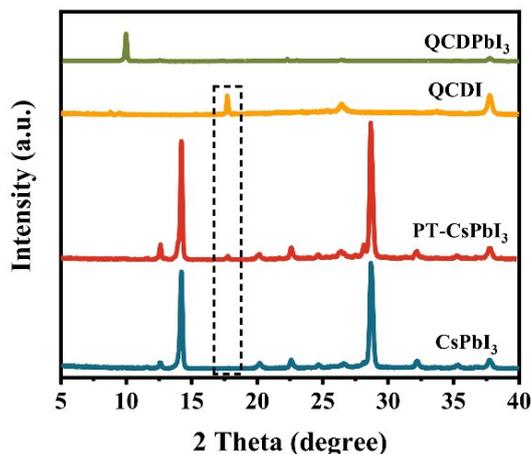
X-ray diffraction (XRD) patterns were obtained by a Rigaku D/MAX-2500 X-ray diffractometer with an X-ray tube Cu K<sub>α</sub> radiation. ( $\lambda = 1.5406 \text{ \AA}$ ). Chemical states of film surface were evaluated by an X-ray photoemission spectroscopy (XPS, ESCALab250Xi). Ultraviolet-visible-infrared spectrophotometer (UV-vis) absorption spectra were recorded on a Shimadzu UV-3600 ultraviolet-visible spectrometer. Scanning electron microscopy (SEM) images and EDS were obtained on a SUPRA55 SEM at an accelerating voltage of 5 kV. Surface roughness of these films were characterized by a Bruker Dimension ICON atomic force microscopy (AFM). TOF-SIMS measurement was performed using a PHI NanoTOF II instrument (ULVAC-PHI, Inc.), where an Argon gas beam was used for erosion and a 30 keV Bi<sup>+</sup> pulsed primary ion beam was used for the analysis.

Steady-state photoluminescence (PL) spectra were recorded on a Nanolog FL3-2iHR (Horiba Jobin Yvon Ltd) with an excitation wavelength of 340 nm. Time-resolved PL (TRPL) spectra taken on an ultrafast lifetime Spectrofluorometer (Delta flex) and a 475 nm ultrafast laser was used as the excitation light source. Confocal photoluminescence (PL) maps were recorded on a laser scanning confocal microscope (A1R-si).

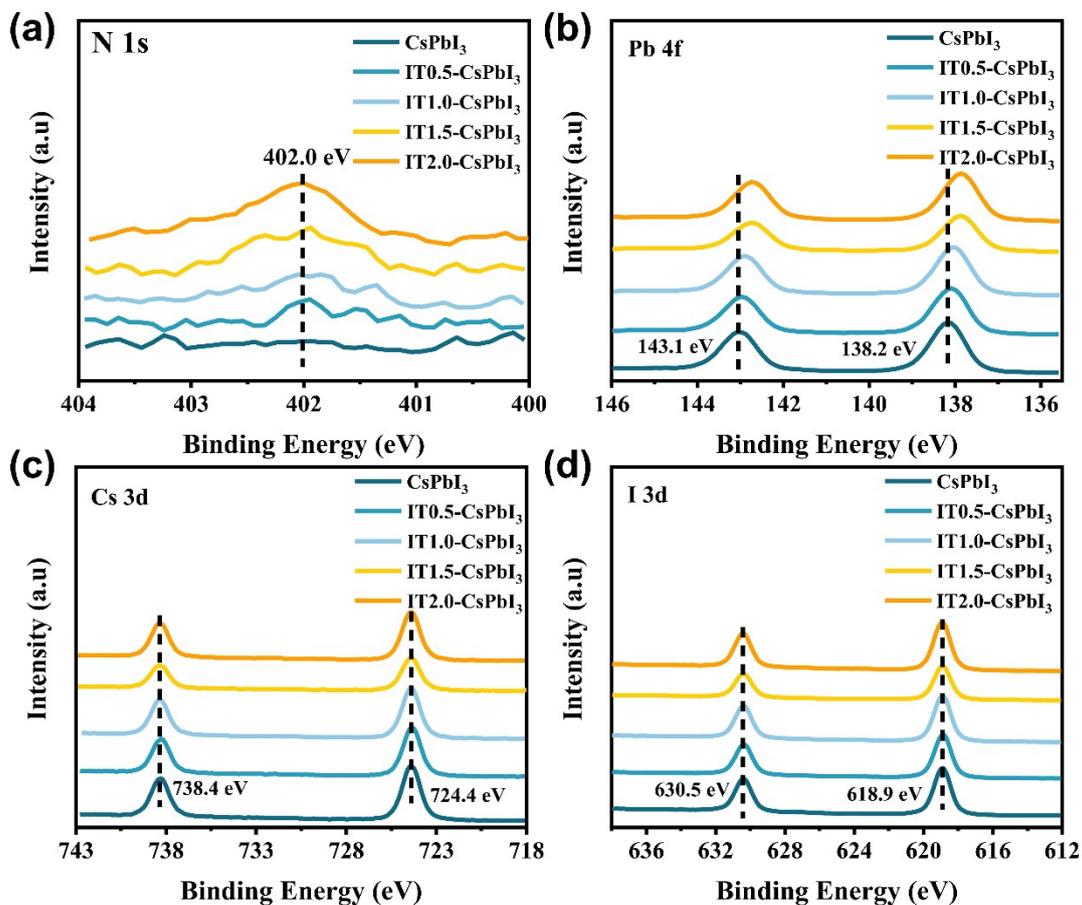
The photovoltaic performance was tested under a solar light simulator (Newport Oriel Sol 3A, model number 94063A, AM 1.5 global filter) in ambient air. The light intensity was calibrated to 1 Sun (100 mW/cm<sup>2</sup>) using an Oriel reference solar cell (monocrystalline silicon) and meter. The active area of cells was masked at around 6.25 mm<sup>2</sup>. Current density-voltage (*J-V*) curves, dependence of *J<sub>sc</sub>* and *V<sub>oc</sub>* on light intensity, Dark *J-V* measurement of the electron-only device and Mott-Schottky curves were measured on ZENNIUM pro electrochemical workstation (ZAHNER-Elektrik GmbH & Co., KG, Germany). The film stability tests were performed in air atmosphere (RH 10~20 %) at room temperature (~25 °C), in humid air (RH 80~90 %) at room temperature (~25 °C), and in N<sub>2</sub> (RH < 10%) at 85 °C. The device stability tests were performed by storing non-encapsulated devices in air atmosphere (RH 10~20 %) at room temperature (~25 °C) and in N<sub>2</sub> (RH < 10%) at 85 °C. *J-V* curves were measured periodically in ambient air atmosphere (RH 30~80 %) to track the device performance.



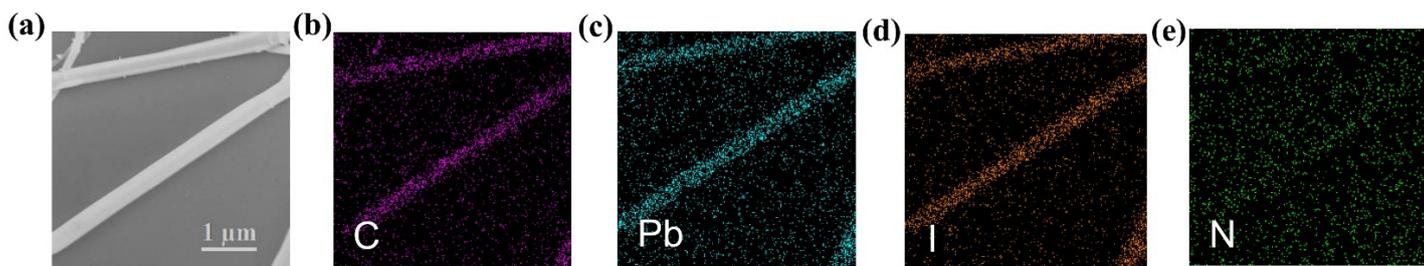
**Figure S1.** (a) Molecular structure of QCDI; (b) Schematic diagram of the fabrication procedure for the PT-CsPbI<sub>3</sub> film.



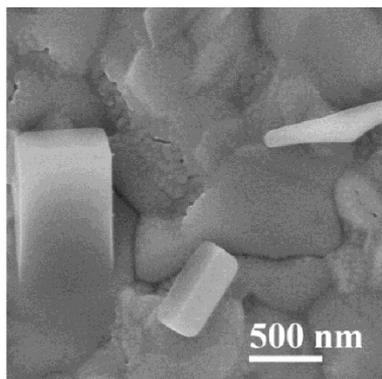
**Figure S2.** XRD patterns of the QCDPbI<sub>3</sub>, QCDI, CsPbI<sub>3</sub> and PT-CsPbI<sub>3</sub> films.



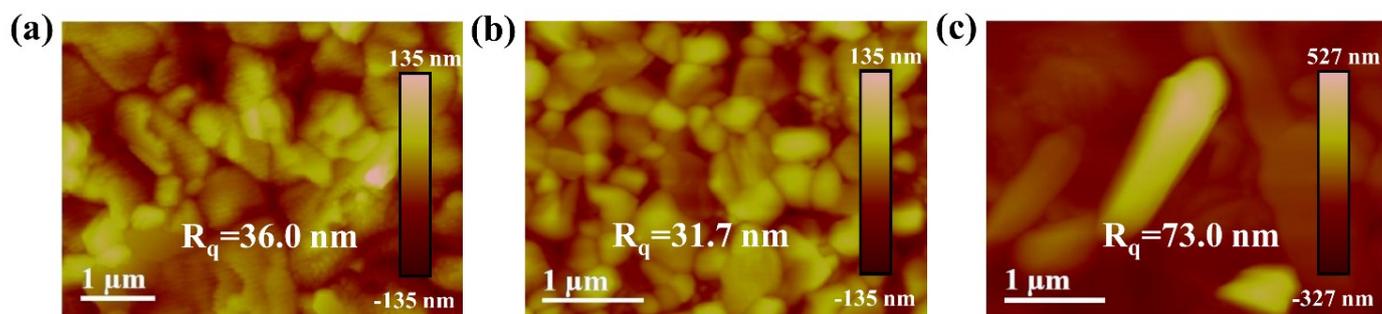
**Figure S3.** XPS spectra of (a) N 1s, (b) Pb 4f and (c) Cs 3d, and (d) I 3d of the IT-CsPbI<sub>3</sub> films with increasing amount of QCDI.



**Figure S4.** (a) Top-view SEM image and EDS mapping of (b) C element, (c) Pb element, (d) I element, and (e) N element distribution of the QCDPbI<sub>3</sub> film.



**Figure S5.** Top-view SEM image of the PT-CsPbI<sub>3</sub> film.



**Figure S6.** AFM images of (a) CsPbI<sub>3</sub>, (b) IT-CsPbI<sub>3</sub> and (c) PT-CsPbI<sub>3</sub> films.

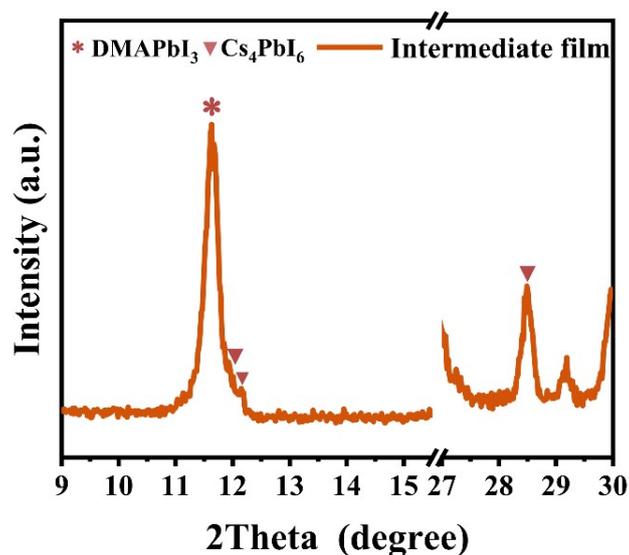


Figure S7. XRD patterns of the intermediate film.

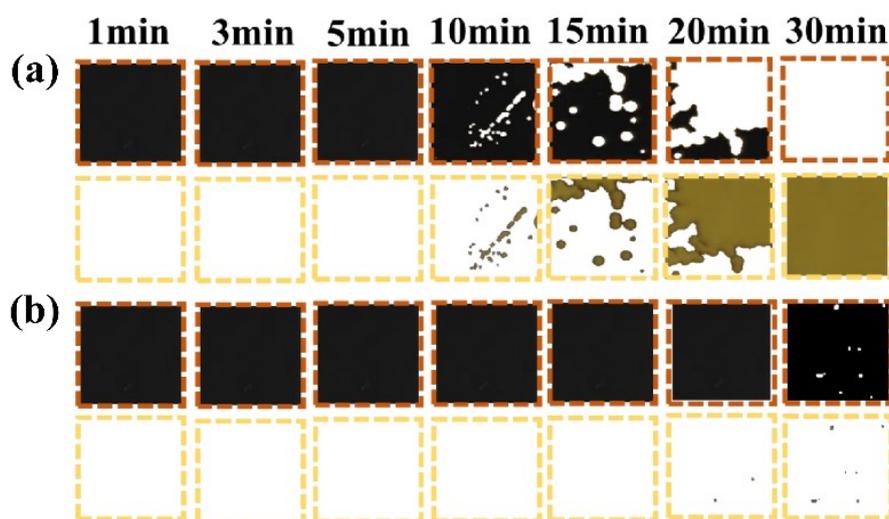


Figure S8. The separated black and yellow regions of photographs in Figure 3(a).

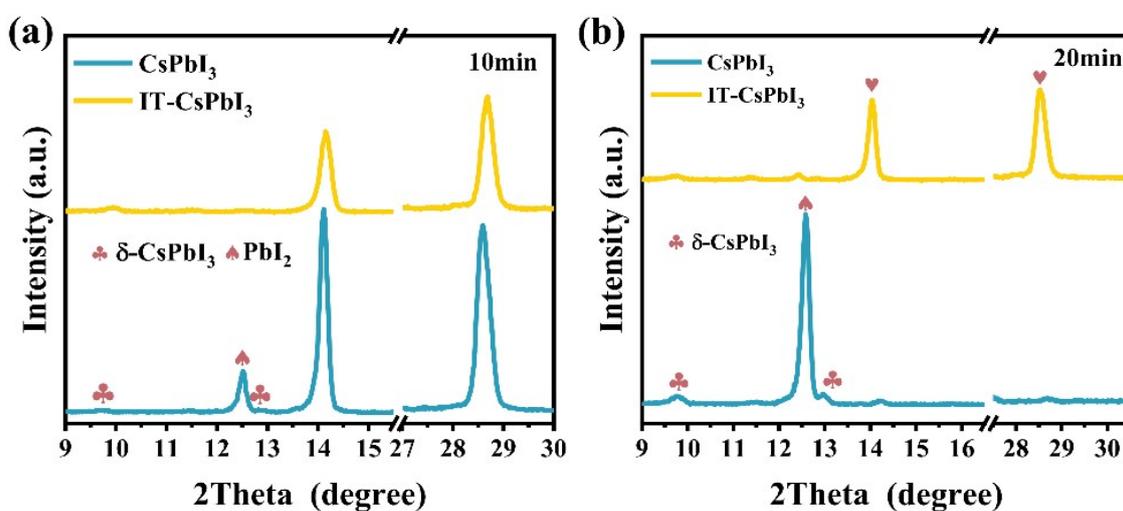
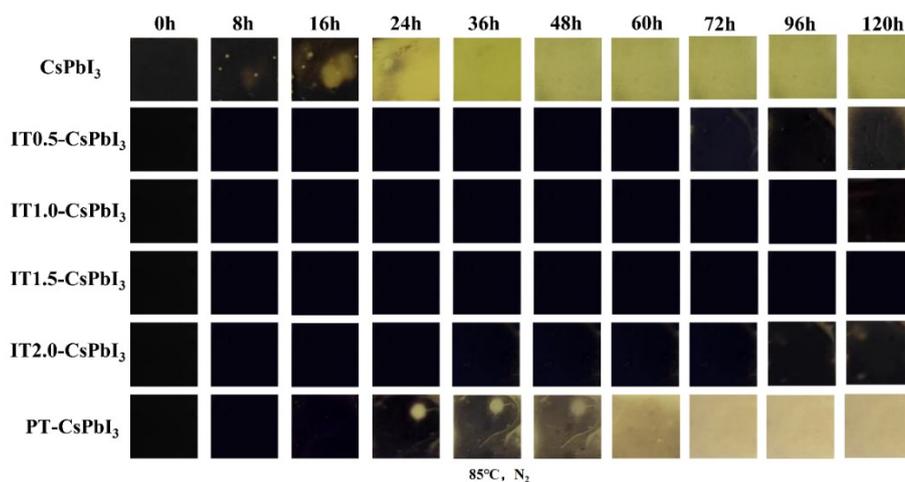
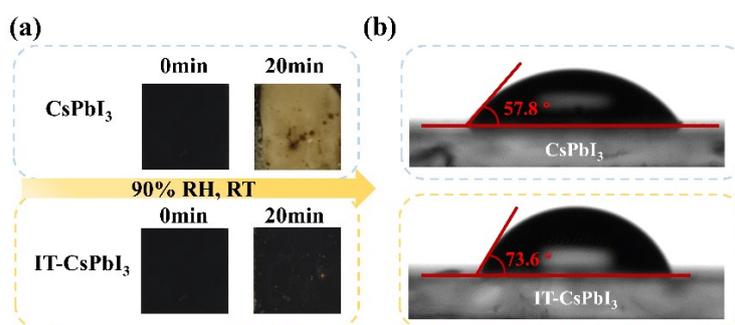


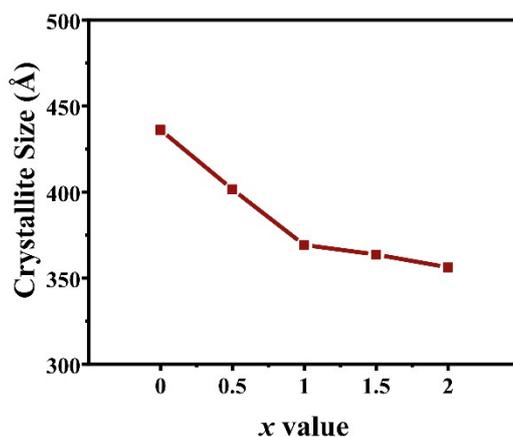
Figure S9. XRD patterns of the pristine CsPbI<sub>3</sub> and IT-CsPbI<sub>3</sub> film when annealing for (a) 10 min and (b) 20min.



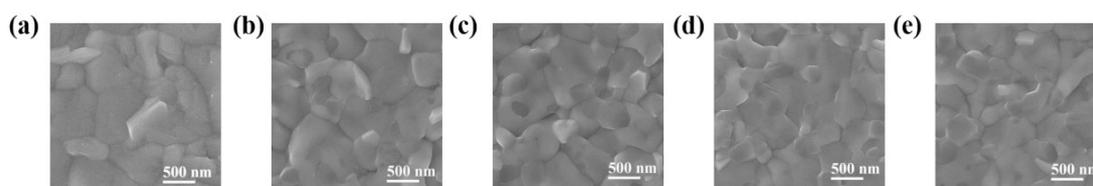
**Figure S10.** Photographs the CsPbI<sub>3</sub>, IT-CsPbI<sub>3</sub> and PT-CsPbI<sub>3</sub> films heated at 85°C in N<sub>2</sub> glovebox with increasing time.



**Figure S11.** (a) Photographs of the CsPbI<sub>3</sub> and IT-CsPbI<sub>3</sub> films before and after exposed to 90% RH at RT for 20 min; (b) Contact angles of the CsPbI<sub>3</sub> and IT-CsPbI<sub>3</sub> films.



**Figure S12.** Crystallite sizes of *p*-CsPbI<sub>3</sub> with increasing *x* value in the IT*x*-CsPbI<sub>3</sub> films.



**Figure S13.** Top-view SEM images of the (a) CsPbI<sub>3</sub>, (b) IT0.5-CsPbI<sub>3</sub>, (c) IT1.0-CsPbI<sub>3</sub>, (d) IT1.5-CsPbI<sub>3</sub>, and (e) IT2.0-CsPbI<sub>3</sub> films.

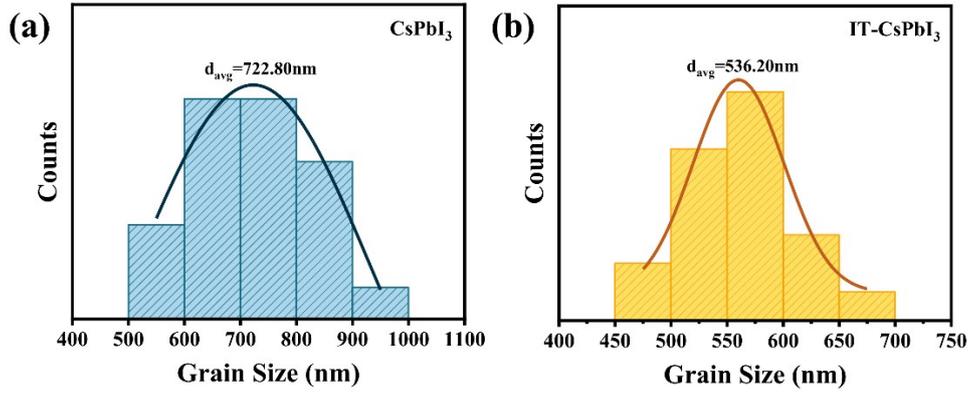


Figure S14. Counted grain sizes of the (a) CsPbI<sub>3</sub> and (b) IT1.5-CsPbI<sub>3</sub> films from corresponding SEM images.

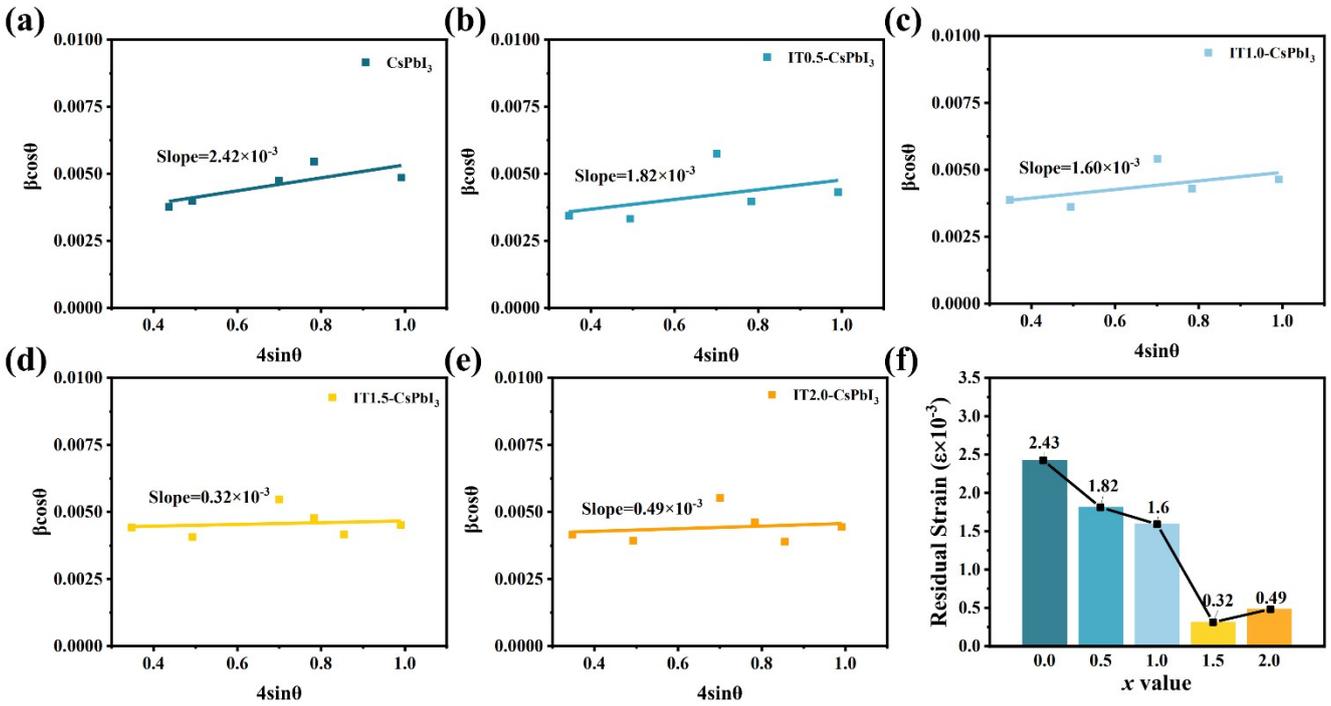


Figure S15. (a)-(e) Williamson-Hall plots and (f) the residual strain of the CsPbI<sub>3</sub> and IT<sub>x</sub>-CsPbI<sub>3</sub> films.

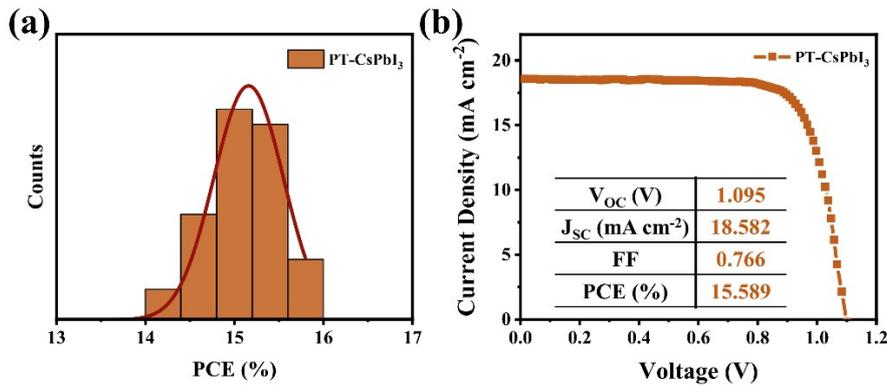
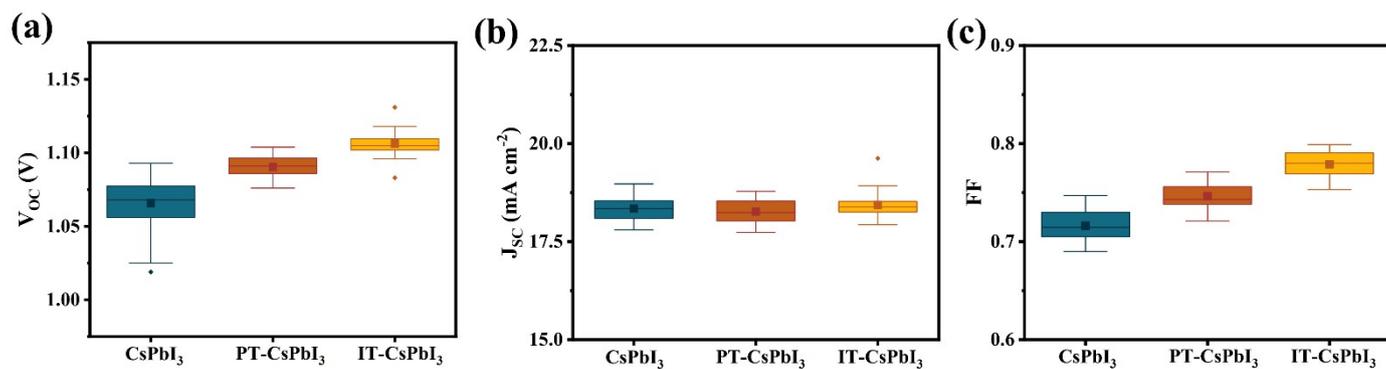
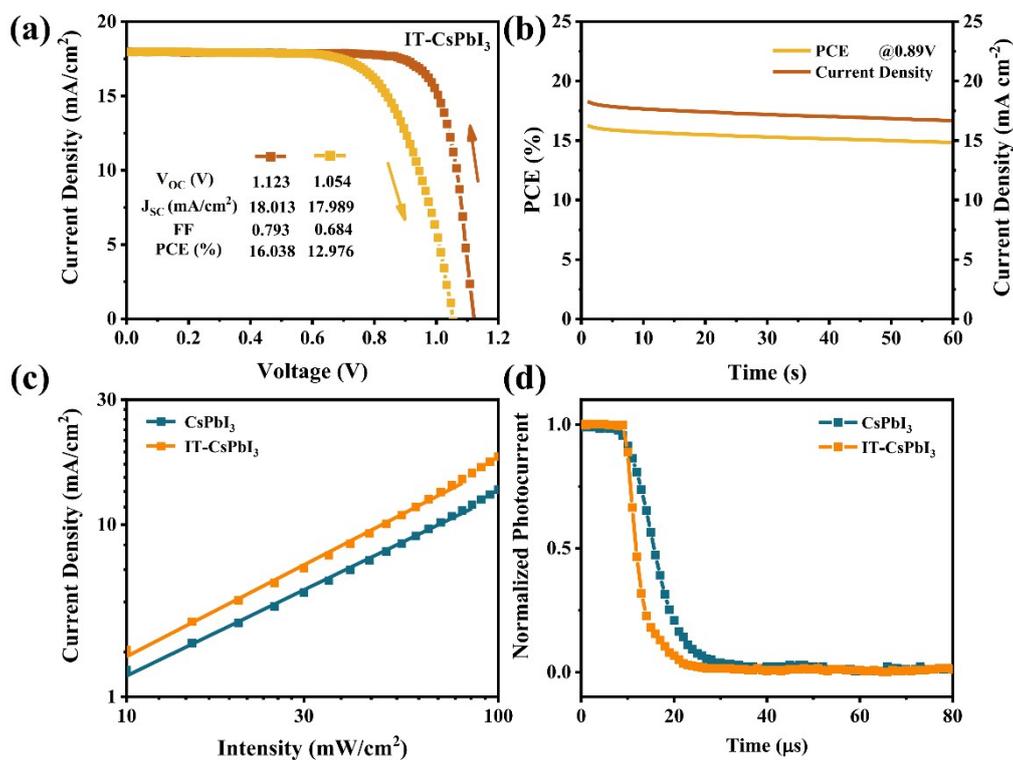


Figure S16. (a) Statistical data of PCEs and (b)  $J-V$  curves of the PT-CsPbI<sub>3</sub> devices.



**Figure S17.** Statistical data obtained from CsPbI<sub>3</sub>, PT-CsPbI<sub>3</sub> and IT-CsPbI<sub>3</sub> C-PSCs: (a)  $V_{OC}$ , (b)  $J_{SC}$ , (c) FF.



**Figure S18.** Other photovoltaic performance of C-PSCs. (a)  $J$ - $V$  curves under forward and reverse scans, (b) steady-state power output, and (c) TPC curves.

**Table S1.** The peak position of the characteristic peaks of *p*-CsPbI<sub>3</sub>.

Sample	CsPbI <sub>3</sub>	IT0.5-CsPbI <sub>3</sub>	IT1.0-CsPbI <sub>3</sub>	IT1.5-CsPbI <sub>3</sub>	IT2.0-CsPbI <sub>3</sub>
(110) plane	14.196°	14.252°	14.263°	14.255°	14.256°
(220) plane	28.756°	28.766°	28.763°	28.775°	28.775°

**Table S2.** The d-spacings (Å) of perovskite phase of the pristine CsPbI<sub>3</sub> and IT-CsPbI<sub>3</sub> films.

Sample	CsPbI <sub>3</sub>	IT0.5-CsPbI <sub>3</sub>	IT1.0-CsPbI <sub>3</sub>	IT1.5-CsPbI <sub>3</sub>	IT2.0-CsPbI <sub>3</sub>
(110) plane	3.141	3.129	3.127	3.128	3.128
(220) plane	1.601	1.601	1.601	1.600	1.600

**Table S3.** The grain sizes (nm) of the pristine CsPbI<sub>3</sub> and IT-CsPbI<sub>3</sub> films.

Sample	CsPbI <sub>3</sub>	IT0.5-CsPbI <sub>3</sub>	IT1.0-CsPbI <sub>3</sub>	IT1.5-CsPbI <sub>3</sub>	IT2.0-CsPbI <sub>3</sub>
Grain Size	722.80±57.63	614.85±37.55	560.74±51.88	536.20±34.07	572.62±41.27

**Table S4.** The fitting results of carrier lifetimes  $\tau$  (ns) for the pristine CsPbI<sub>3</sub> and IT-CsPbI<sub>3</sub> films.

	A <sub>1</sub>	$\tau_1$	A <sub>2</sub>	$\tau_2$	$\tau$
CsPbI <sub>3</sub>	0.668	5.513	0.429	5.514	5.513
IT-CsPbI <sub>3</sub>	1.662	4.061	0.117	63.127	34.972

**Table S5.** Performance comparison of existing C-PSCs based on CsPbX<sub>3</sub> inorganic perovskites.

Perovskite layer	Device structure	$V_{OC}$ (V)	$J_{SC}$ (mA/cm <sup>2</sup> )	FF	PCE (%)	Ref
CsPbI <sub>3</sub>	FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> / CsPbI <sub>3</sub> : Br /carbon	1.03	12.46	0.56	7.22	[1]
	FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CsPbI <sub>3</sub> /carbon	0.67	14.31	1.48	4.65	[2]
	FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CsPbI <sub>3</sub> /carbon	0.73	14.65	0.5	5.31	[3]
	FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CsPbI <sub>3</sub> /CuSCN/carbon	0.94	13.8	0.52	6.9	[4]
	FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CsPbI <sub>3</sub> /carbon	0.91	15.76	0.66	9.39	[5]
	FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CsPbI <sub>3</sub> /carbon	0.79	18.5	0.65	9.5	[6]
	FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> / Cs <sub>0.95</sub> Na <sub>0.05</sub> PbI <sub>3</sub> /carbon	0.92	16.5	0.703	10.7	[7]
	FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CsPbI <sub>3</sub> /PEI-CNT/carbon	0.80	18.58	0.71	10.55	[8]
	FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CsPbBr <sub>3</sub> / CsPbI <sub>3</sub> NCs/carbon	1.490	8.66	0.732	9.45	[9]
	FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CsPbI <sub>3</sub> : Br: InI <sub>3</sub> /carbon	1.20	15.68	0.64	12.04	[1]
CsPbI <sub>2</sub> Br	FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CsPbI <sub>3</sub> /carbon	1.056	17.47	0.79	14.60	[10]
	FTO/c-TiO <sub>2</sub> /CsPbI <sub>2</sub> Br/carbon	1.15	13.54	0.642	10	[11]
	FTO/TiO <sub>2</sub> /CsPbI <sub>2</sub> Br/P3HT-MWCNT/carbon	1.21	13.35	0.62	10.01	[12]
	ITO/SnO <sub>2</sub> /KOH/CsPbI <sub>2</sub> Br/carbon	14.24	1.20	0.687	11.78	[13]
	FTO/Ni <sub>2</sub> O <sub>5</sub> /Cs <sub>0.99</sub> Rb <sub>0.01</sub> PbI <sub>2</sub> Br/carbon	1.24	14.02	0.69	12.00	[14]
	FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CsPbI <sub>2</sub> Br/carbon	1.210	14.94	0.73	13.13	[15]
	FTO/Ni <sub>2</sub> O <sub>5</sub> /CsPbI <sub>2</sub> Br/carbon	1.20	13.96	0.66	11.02	[14]
	ITO/SnO <sub>2</sub> /Nb-CsPbI <sub>2</sub> Br/carbon	1.20	12.06	0.72	10.42	[16]
	FTO/SnO <sub>2</sub> /CsPbI <sub>2</sub> Br/carbon	1.23	15.46	0.64	12.19	[17]
	ITO/SnO <sub>2</sub> /CsPbI <sub>2</sub> Br/carbon	1.187	12.91	0.661	10.13	[18]
	ITO/SnO <sub>2</sub> /CsPbI <sub>2</sub> Br/carbon	1.14	14.25	0.641	10.44	[19]
ITO/SnO <sub>2</sub> /CsPbI <sub>2</sub> Br/PMMA/carbon	1.20	12.64	0.71	10.95	[20]	
ITO/SnO <sub>2</sub> /CsPbI <sub>2</sub> Br/Co <sub>3</sub> O <sub>4</sub> /carbon	1.19	13.09	0.82	11.21	[21]	

	FTO/c-TiO <sub>2</sub> /CsPbI <sub>2</sub> Br/carbon	1.207	16.62	0.74	14.84	[22]
	FTO/c-TiO <sub>2</sub> /CsPbI <sub>2</sub> Br/Carbon	1.15	13.87	0.64	10.21	[23]
CsPbIBr <sub>2</sub>	ITO/SnO <sub>2</sub> /CsPbIBr <sub>2</sub> /carbon	1.23	8.50	0.67	7.00	[24]
	FTO/ c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CsPbIBr <sub>2</sub> /carbon	0.99	13.15	0.57	7.36	[25]
	FTO/ c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CsPb <sub>0.9</sub> Sn <sub>0.1</sub> IBr <sub>2</sub> /carbon	1.26	14.30	0.63	11.33	[26]
	FTO/ c-TiO <sub>2</sub> /Li-CsPbIBr <sub>2</sub> /CuPc/carbon	1.22	10.27	0.74	9.25	[27]
	FTO/ ZnO <sub>2</sub> /CsPbIBr <sub>2</sub> /carbon	1.03	11.60	0.63	7.60	[28]
	FTO/ c-TiO <sub>2</sub> /CsPbIBr <sub>2</sub> /carbon	1.245	10.66	0.69	9.16	[29]
	FTO/ c-TiO <sub>2</sub> /CsPbIBr <sub>2</sub> /carbon	1.142	9.11	0.63	6.55	[30]
	FTO/ TiO <sub>2</sub> /CsBr/CsPbIBr <sub>2</sub> /carbon	1.284	10.31	0.52	6.88	[31]
	FTO/ TiO <sub>2</sub> /CsBr/CsPbIBr <sub>2</sub> /carbon	1.261	11.80	0.72	10.71	[20]
	CsPbBr <sub>3</sub>	FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CsPbBr <sub>3</sub> /carbon	1.29	5.7	0.68	5.00
FTO/c-TiO <sub>2</sub> /CsPbBr <sub>3</sub> /carbon		1.34	6.46	0.68	5.86	[33]
FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CsPbBr <sub>3</sub> /carbon		1.38	7.13	0.62	6.10	[34]
FTO/c-TiO <sub>2</sub> /CsPbIBr <sub>2</sub> /carbon		1.29	12.03	0.70	10.95	[35]
FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CsPbBr <sub>3</sub> /carbon		1.24	7.4	0.73	6.7	[36]
FTO/TiO <sub>2</sub> /CsPbBr <sub>3</sub> /carbon		1.19	7.48	0.688	6.12	[37]
FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CsPbBr <sub>3</sub> /P3HT/carbon		1.36	7.02	0.68	6.49	[38]
FTO/Ni-TiO <sub>2</sub> /CsPbBr <sub>3</sub> /CuPc/carbon		1.362	7.92	0.793	8.55	[39]
FTO/TiO <sub>2</sub> /SnO <sub>2</sub> /CsPbBr <sub>3</sub> /CuPc/carbon		1.310	8.24	0.814	8.79	[40]
FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CsPb <sub>0.995</sub> Zn <sub>0.005</sub> Br <sub>3</sub> /carbon		1.56	7.30	0.806	9.18	[41]
FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CsPbBr <sub>3</sub> /CsPbBr <sub>3</sub> NCs/carbon		1.348	7.85	0.744	7.87	[9]
FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CsPbBr <sub>3</sub> /P1Z1/carbon		1.578	7.652	0.830	10.03	[42]
FTO/TiO <sub>2</sub> /SnO <sub>2</sub> /CsPbBr <sub>3</sub> /carbon		1.610	7.80	0.844	10.60	[43]
FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CsPbBr <sub>3</sub> /carbon		1.594	7.48	0.851	10.14	[44]
FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CsPbBr <sub>3</sub> /MnS/carbon		1.52	8.28	0.83	10.45	[1]
FTO/SnO <sub>2</sub> /CsPbBr <sub>3</sub> /carbon		1.622	7.87	0.801	10.71	[45]
FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /Cu(Cr,Ba)O <sub>2</sub> /CsPbBr <sub>3</sub> /carbon		1.615	7.81	0.855	10.79	[46]
FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CsPbBr <sub>3</sub> /QDs/carbon		1.626	7.73	0.863	10.85	[47]
FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CsPbBr <sub>3</sub> /carbon		1.431	6.84	0.78	7.62	[48]
FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CsPbBr <sub>3</sub> /carbon		1.52	7.56	0.827	9.53	[49]
FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CsPbBr <sub>3</sub> /carbon		1.50	7.56	0.830	10.03	[50]
FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CsPbBr <sub>3</sub> /carbon		1.458	8.12	0.821	9.72	[51]
FTO/c-TiO <sub>2</sub> /m-TiO <sub>2</sub> /CsPb <sub>0.97</sub> Tb <sub>0.03</sub> Br <sub>3</sub> /SnS:ZnS/NiO <sub>x</sub> /carbon	1.57	8.21	0.796	10.26	[52]	

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