

Supporting Information

Enhanced crystallization and stability of perovskites by a cross-linkable fullerene for high-performance solar cells

Meng Li,^{‡a} Yi-Hsiang Chao,^{‡b} Tin Kang,^b Zhao-Kui Wang,^{*a} Ying-Guo Yang,^c Shang-Lei Feng,^c Yun Hu,^a Xing-Yu Gao,^c Liang-Sheng Liao,^{*a} and Chain-Shu Hsu^{*b}

^a *Jiangsu Key Laboratory for Carbon-Based Functional Materials & Devices, Institute of Functional Nano & Soft Materials (FUNSOM), Soochow University, Suzhou 215123, China*

^b *Department of Applied Chemistry, National Chiao Tung University, 1001 Ta Hsueh Road Hsin-Chu, Taiwan 30010*

^c *Shanghai Synchrotron Radiation Facility, Shanghai Institute of Applied Physics, Chinese Academy of Sciences, Shanghai 201204, China*

Corresponding Authors:

zkwang@suda.edu.cn (Z.K.Wang); lsliao@suda.edu.cn (L.S.Liao); cshsu@mail.nctu.edu.tw (C.S.Hsu)

Keywords: perovskite solar cells; perovskite crystallization; stability; cross-linkable fullerene;

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Preparation of $\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}$: PCBSD Precursor Solution

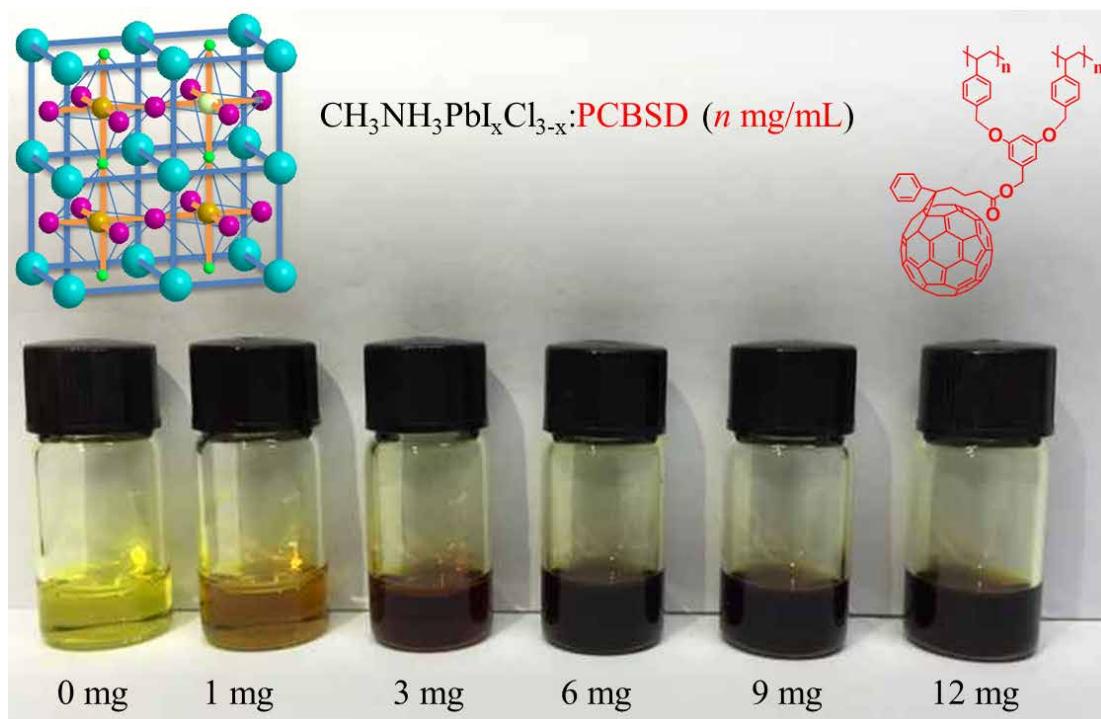


Figure S1. Molecular structures of $\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}$ and PCBSD, and photographies of prepared $\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}$:PCBSD precursor solutions with varied mass of PCBSD.

$\text{CH}_3\text{NH}_3\text{I}$ was synthesized and purified by following previously reported process.^[1-3] Pristine perovskite precursor solution was prepared by mixing PbCl_2 and $\text{CH}_3\text{NH}_3\text{I}$ powder with a molar ration of 3:1 in DMF solvent (30 wt %). The perovskite precursor solution containing C-PCBSD was prepared as follows: $\text{CH}_3\text{NH}_3\text{I}$ (256 mg), C-PCBSD (varied mass) and 149 mg PbCl_2 were dissolved in 1 ml DMF mixed solvent with stirring overnight at 60 °C. The mixture was kept stirred at 60 °C for 8h and filtered through 0.45 μm filter before usage. And then precursor solution of perovskite was spin coated onto the PEDOT:PSS layer at 4000 rpm/40s.

- [1] X. Gong, M. Li, X. B. Shi, H. Ma, Z. K. Wang, L. S. Liao, *Adv. Funct. Mater.* **2015**, 25, 6671.
- [2] Z. K. Wang, X. Gong, M. Li, Y. Hu, J. M. Wang, H. Ma, and L. S. Liao, *ACS Nano* **2016**, 10, 5479.
- [3] Z. K. Wang, M. Li, Y. G. Yang, Y. Hu, H. Ma, X. Y. Gao, and L. S. Liao, *Adv. Mater.* **2016**, DOI: 10.1002/adma.201600626

Performance of $\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}$: PCBSD (2 h annealing) Based PSCs

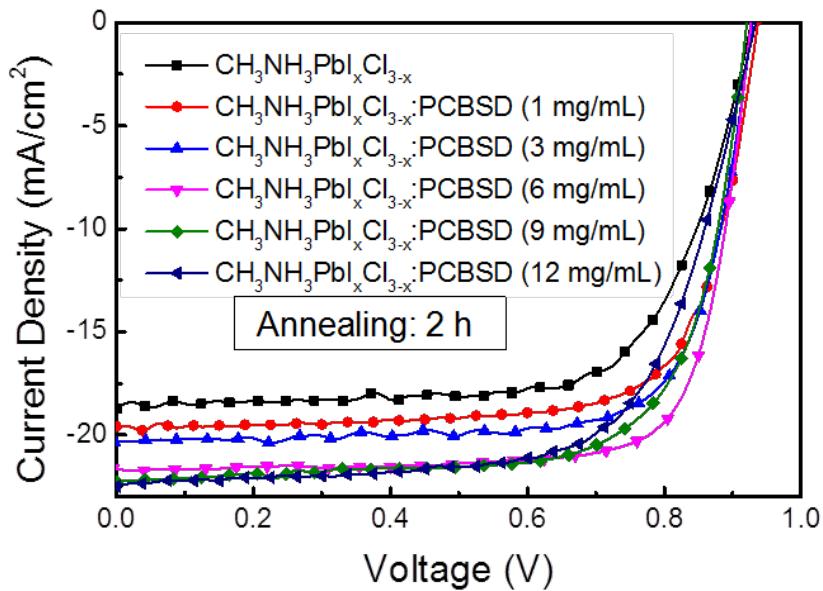


Figure S2. J - V curves of $\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}$: PCBSD (2 h annealing) based perovskite solar cells measured under simulated AM 1.5 sunlight of 100 mW/cm² irradiation.

Table S1. Cell parameters of $\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}$: PCBSD (2 h annealing) based perovskite solar cells.

Active Layer	V_{oc} (V)	J_{sc} (mA/cm ²)	FF (%)	PCE (%)	PCE _{AVE} (%)	R_s (Ω)
$\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}$	0.92	18.73	70	12.06	11.83 ± 0.25	57.6
$\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}$:PCBSD (1 mg/ml)	0.93	19.57	74	13.46	13.32 ± 0.17	52.3
$\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}$:PCBSD (3 mg/ml)	0.93	20.32	75	14.09	13.75 ± 0.36	33.4
$\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}$:PCBSD (6 mg/ml)	0.93	21.64	75	15.09	14.86 ± 0.42	20.1
$\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}$:PCBSD (9 mg/ml)	0.92	22.27	72	14.63	14.48 ± 0.22	25.8
$\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}$:PCBSD (12 mg/ml)	0.93	22.50	67	14.04	13.83 ± 0.62	18.3

Performance of $\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}$: PCBSD (3 h annealing) Based PSCs

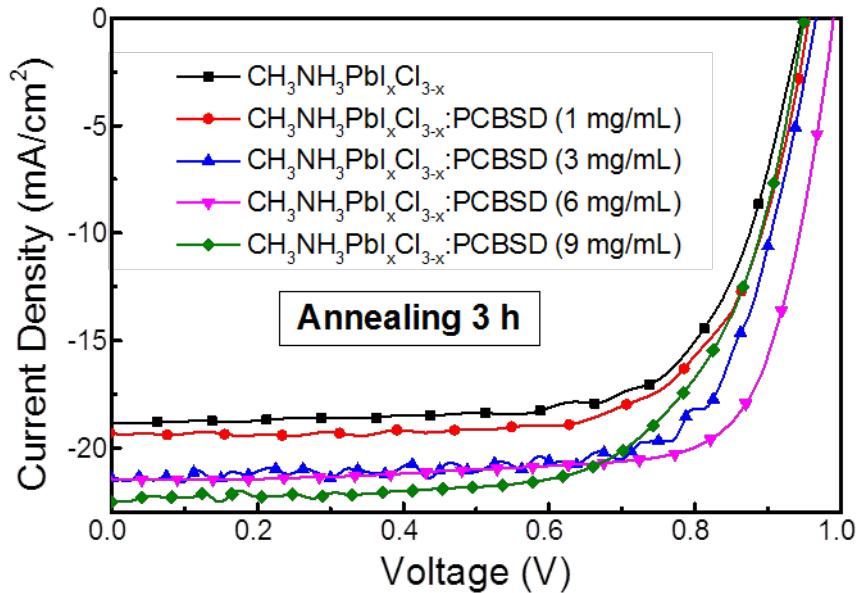


Figure S3. J - V curves of $\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}$: PCBSD (3 h annealing) based perovskite solar cells measured under simulated AM 1.5 sunlight of 100 mW/cm² irradiation.

Table S2. Cell parameters of $\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}$: PCBSD (3 h annealing) based perovskite solar cells.

Active Layer	V_{oc} (V)	J_{sc} (mA/cm ²)	FF (%)	PCE (%)	PCE _{AVE} (%)	R_s (Ω)
$\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}$	0.93	18.83	71	12.43	11.83 ± 0.25	51.3
$\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}$:PCBSD (1 mg/ml)	0.96	19.35	70	13.01	12.76 ± 0.27	38.9
$\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}$:PCBSD (3 mg/ml)	0.96	21.40	73	15.00	14.84 ± 0.23	29.2
$\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}$:PCBSD (6 mg/ml)	0.96	21.71	75	15.63	15.57 ± 0.17	33.0
$\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}$:PCBSD (9 mg/ml)	0.95	22.50	66	14.11	14.09 ± 0.14	37.6

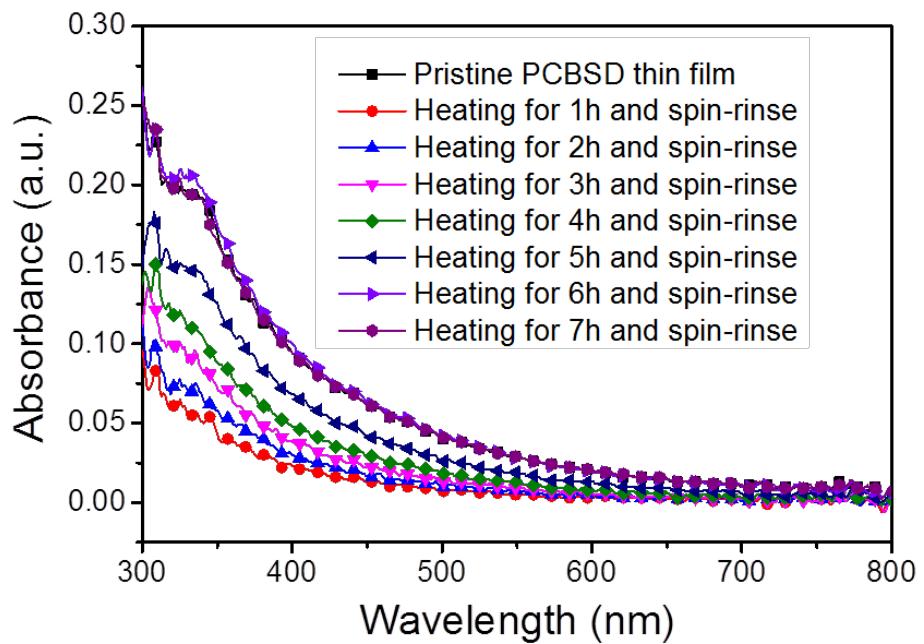


Figure S4. Thin film absorption spectra of pristine PCBSD and C-PCBSD thin films heating in 100 °C with different annealing time and then were washed by chlorobenzene.

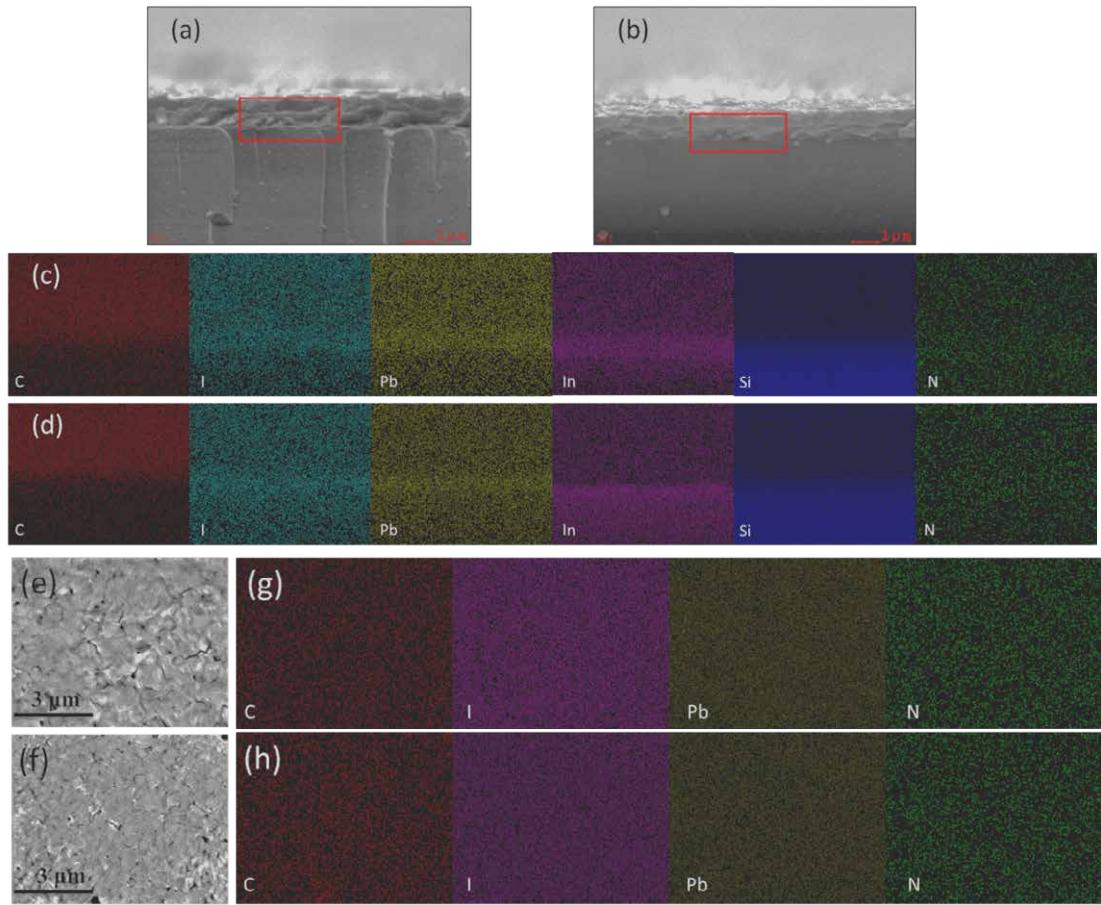


Figure S5. The EDAX spectra of perovskite film; SEM cross-sections image and top view in $\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}$ (a, e) and $\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}$: C-PCBSD (6 mg/ml) (b, f) samples, (c, e) I, Pb, In, Si, N mapping of $\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}$, (d, f) I, Pb, In, Si, N mapping of $\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}$: C-PCBSD (6 mg/ml).

Table S3. Cell parameters of $\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}$, $\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}:\text{PCBM}$ and $\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}:\text{PCBSD}$ based perovskite solar cells scanned in forward and reverse directions.

Device	V_{oc} (V)	J_{sc} (mA/cm ²)	FF (%)	PCE (%)
$\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}$ (forward)	0.95	18.67	71	12.59
$\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}$ (reverse)	0.95	19.58	57	10.06
$\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}:\text{PCBM}$ (forward)	0.95	22.05	73	15.29
$\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}:\text{PCBM}$ (reverse)	0.94	21.97	71	14.66
$\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}:\text{PCBSD}$ (forward)	0.98	22.08	79	17.09
$\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}:\text{PCBSD}$ (reverse)	0.98	22.65	77	17.09

Table S4. Electrical impedance spectroscopy (EIS) parameters of $\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}$ and $\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}$: C-PCBSD (6 mg/ml) based PSCs.

	$\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}$	$\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}$:C-PCBSD
R_s (Ω)	59.6	27.3
R_{CT} (Ω)	1269	891
C (F)	2.1×10^{-6}	2.4×10^{-6}

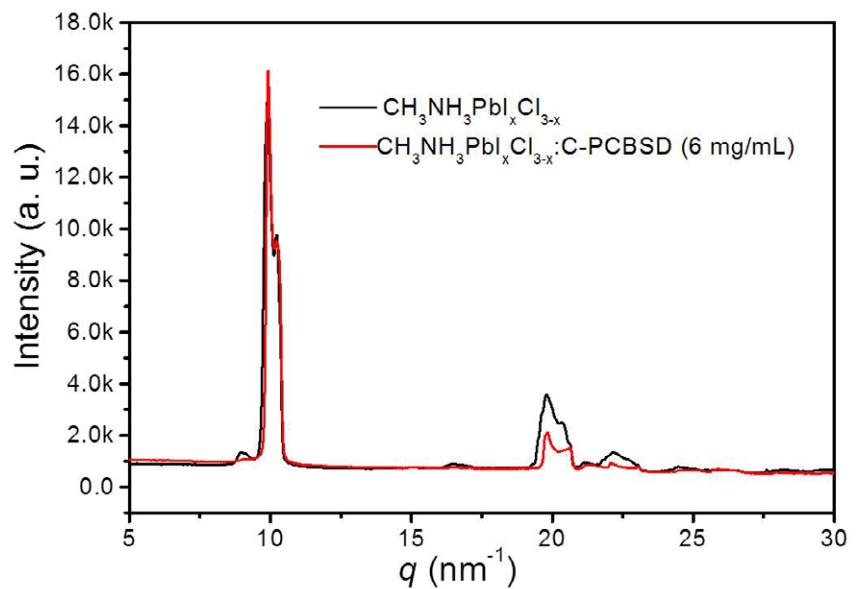


Figure S6. Azimuthally integrated intensity plots along the direction of outside surface (azimuth Angle of 90 degrees in the 2D GIXRD) of the corresponding GIXRD patterns of the two films in Figure 4 (d) and (e).

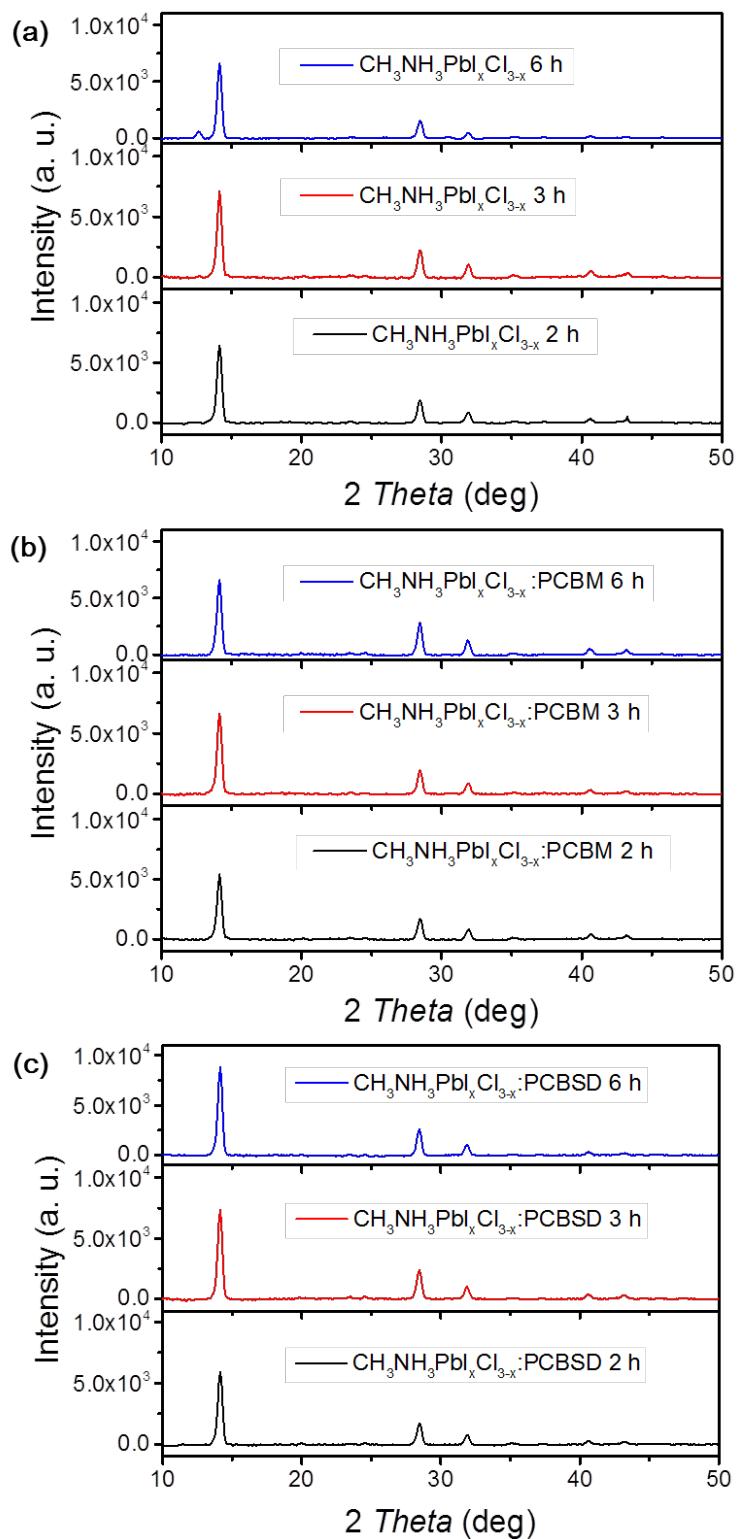


Figure S7. XRD patterns of (a) $\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}$ (Thickness: 282 nm), (b) $\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}:\text{PCBM}$ (6 mg/ml) (Thickness: 295 nm) and (c) $\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}:\text{PCBSD}$ (6 mg/ml) (Thickness: 305 nm) films with varied annealing time of 2 h, 3 h, and 6 h at 100°C.

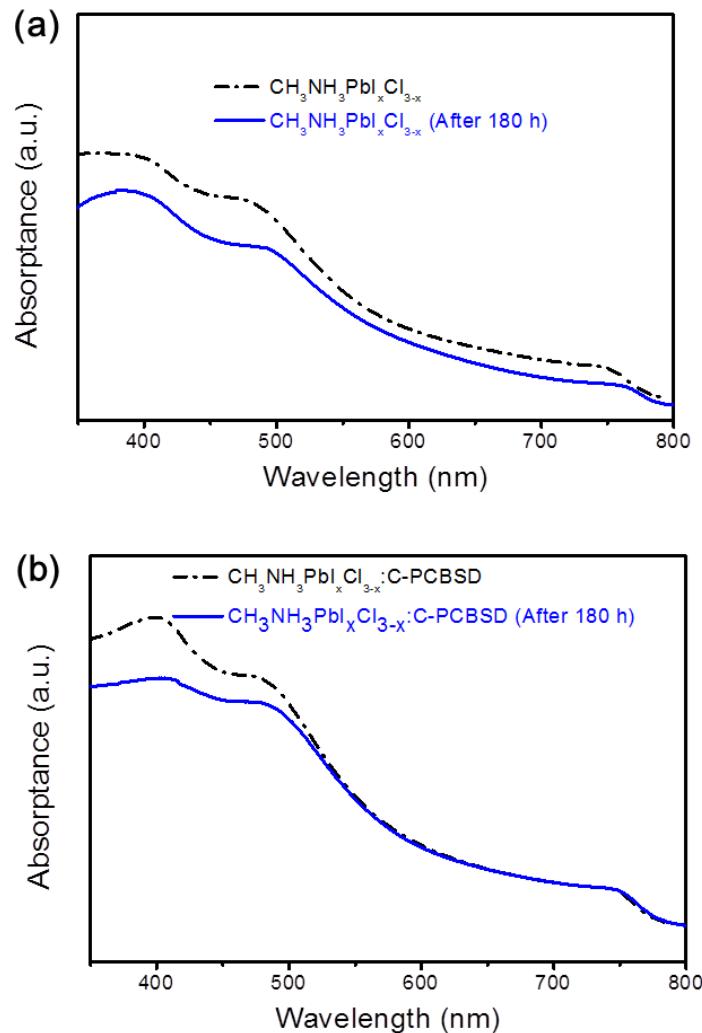


Figure S8. Absorption spectra of fresh and aged (after 180 h) $\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}$ (280 nm) and $\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}\text{:C-PCBSD}$ (6 mg/ml) (280 nm) films.

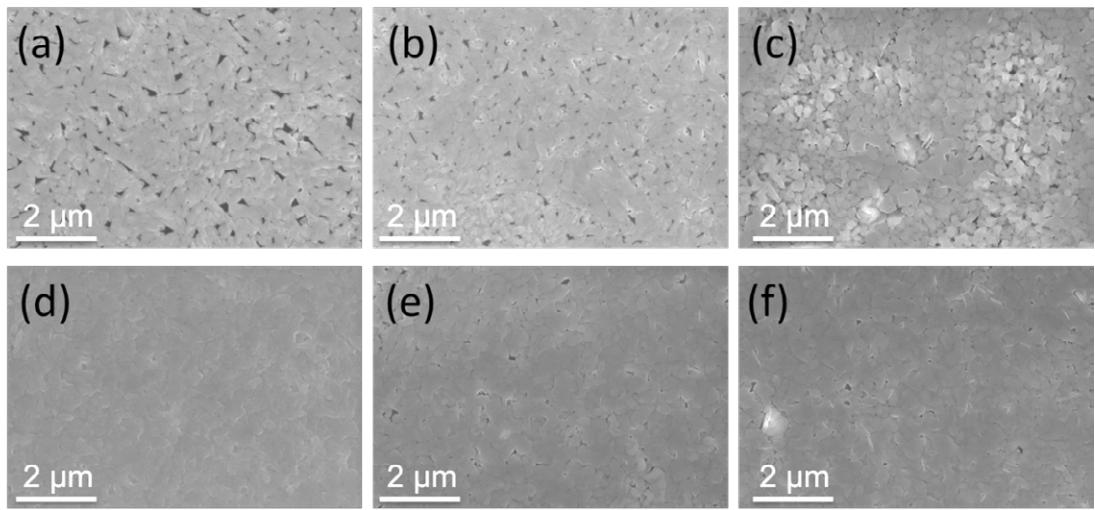


Figure S9. SEM images of (a) $\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}$ (aging time: 0h), (b) $\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}$ (aging time: 20h), (c) $\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}$ (aging time: 40h), (d) $\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}$: C-PCBSD (aging time: 0h), (e) $\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}$: C-PCBSD (6 mg/ml) (aging time: 20h) and (f) $\text{CH}_3\text{NH}_3\text{PbI}_x\text{Cl}_{3-x}$: C-PCBSD (aging time: 40h).