Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Supporting Information

Nb-doped amorphous titanium oxide compact layer for formamidinium-based high efficiency perovskite solar cells by low-temperature fabrication

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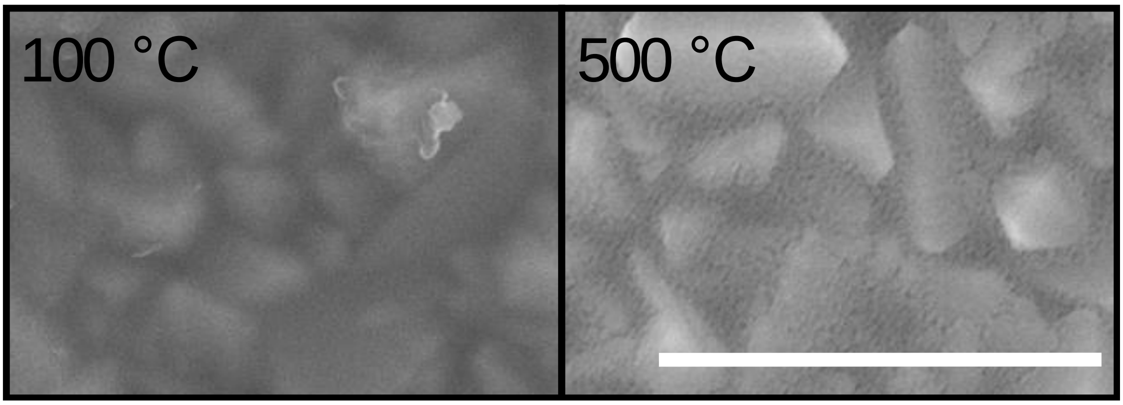

Figure S1. Surface SEM images of TiO_x CLs dried at 100 °C (left) and sintered at 500 °C (right), respectively. Scale bar = 1 μm.

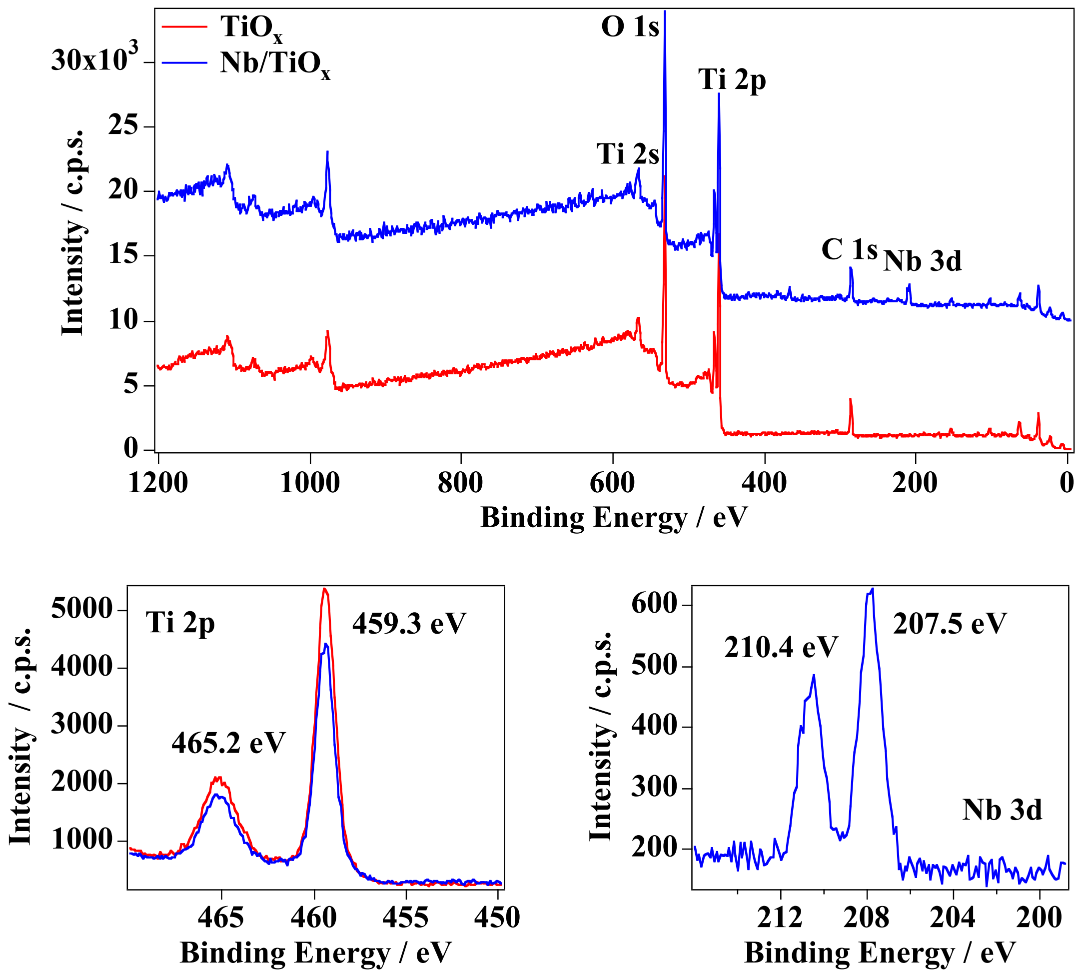

Figure S2. XPS spectra of TiO_X and (5%) Nb/TiO_X CL annealed at 150 °C.

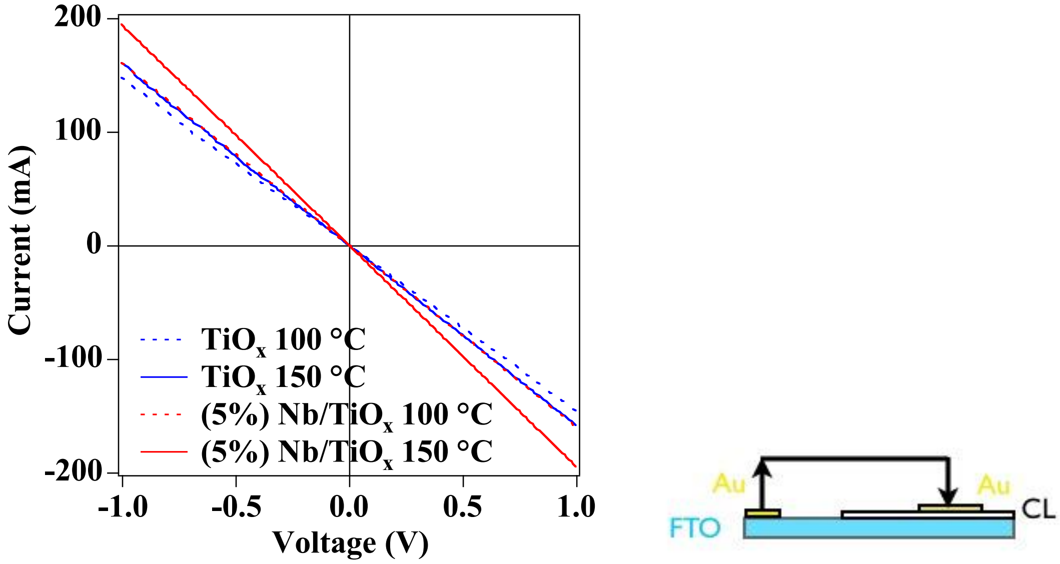

Figure S3. *I-V* curves of FTO/CL (TiO_X and (5%) Nb/TiO_X)/Au device; CL thickness is approximately 40 nm, and schematic illustration of the measured device.

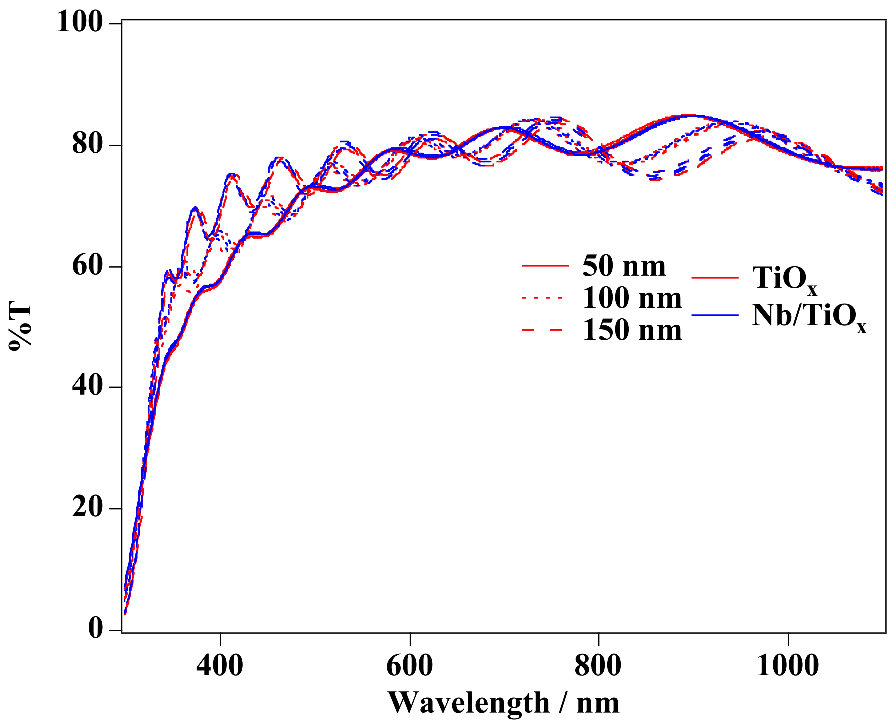

Figure S4. Transmittances of TiO_x and (5%) Nb/TiO_x CL on FTO substrates dried at 100, 130, and 150 °C with the thickness of 50, 100, and 150 nm, respectively.

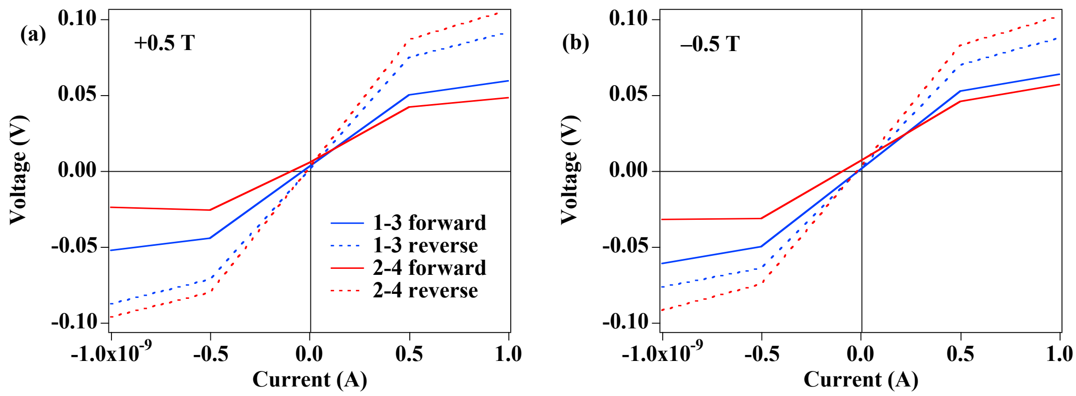

Figure S5. Hall-effect measurement data of (5%) Nb/TiO_X CL (film thickness: 7 nm) on a 1.25 × 1.25 cm^2 glass substrate measured under (a) +0.5 T and (b) –0.5 T magnetic field, respectively. Electrodes contacted at diagonal corners (1-3 and 2-4).

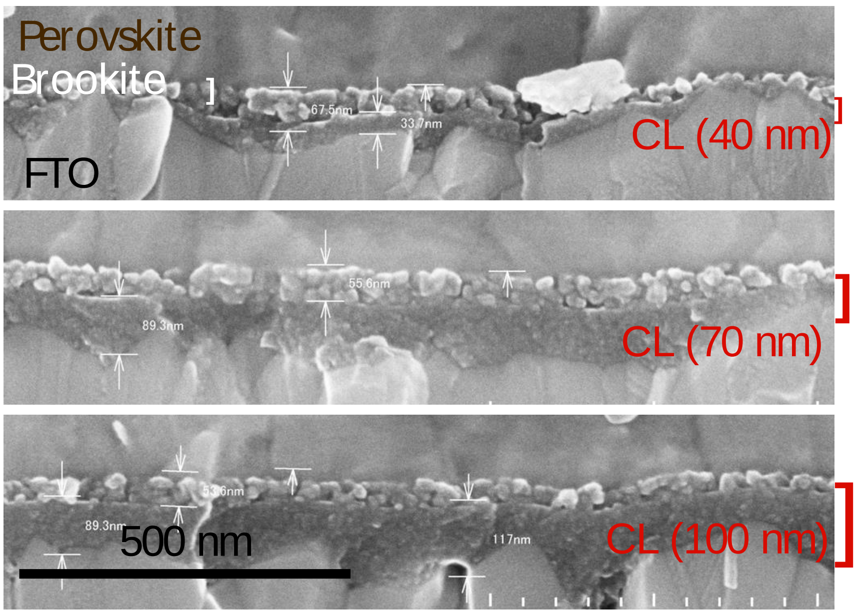

Figure S6. Cross-sectional SEM images of $(FAPbI_3)_{0.85}(MAPbBr_3)_{0.15}$ PSCs based on (5%) Nb/TiO_x CLs with different thickness; 40, 70, and 100 nm. Black scale bar: 500 nm.

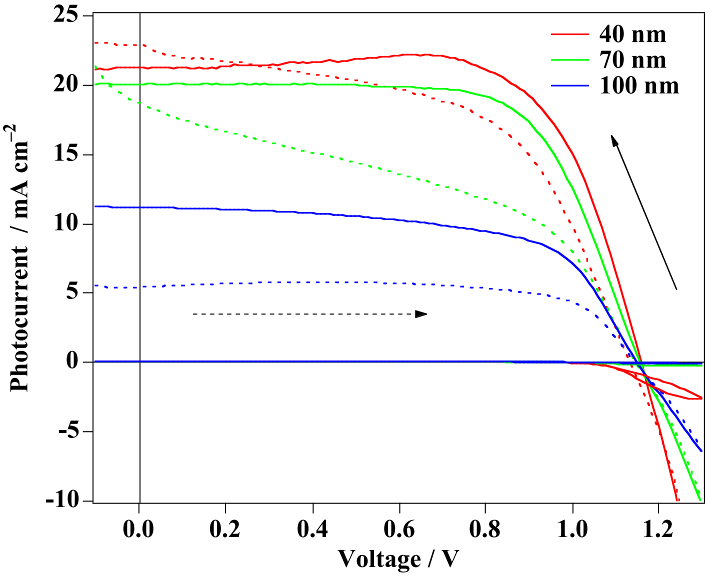

Figure S7. *J-V* curves of PSCs of $(FAPbI_3)_{0.85}(MAPbBr_3)_{0.15}$ based on (5%) Nb/TiO_x CLs with different thickness; 40 (red), 70 (green), and 100 nm (blue). Broken line: forward scans and line: reverse scans.

Table S1. Photovoltaic parameters in reverse scans ($V_{OC} \rightarrow J_{SC}$) of PSCs based on (5%) Nb/TiO_x CL with different thickness

CL thickness (nm)	scan direction	J_{SC} (mA cm^{-2})	V_{OC} (V)	*FF*	η (%)	No. of cells
40	forward	22.71±0.28 (22.78)	1.14±0.01 (1.14)	0.50±0.04 (0.54)	13.0±0.9 (14.1)	6
	reverse	20.56±1.06 (21.26)	1.16±0.01 (1.16)	0.69±0.02 (0.70)	16.5±0.6 (17.3)	
70	forward	18.25±1.57 (20.46)	1.13±0.01 (1.14)	0.41±0.04 (0.45)	8.5±1.4 (10.4)	6
	reverse	17.10±2.53 (20.00)	1.15±0.01 (1.16)	0.73±0.07 (0.68)	14.2±1.3 (15.7)	
100	forward	5.29±0.13 (5.38)	1.13±0.03 (1.15)	0.62±0.15 (0.73)	3.7±1.1 (4.5)	2[a)]
	reverse	9.93±1.68 (11.12)	1.13±0.02 (1.13)	0.60±0.02 (0.62)	6.8±1.6 (7.9)	

Scan direction: forward $J_{SC} \rightarrow V_{OC}$ and reverse: $V_{OC} \rightarrow J_{SC}$. In parentheses, the parameters of the best cell. Cell active area is 5 × 5 mm^2 defined by black mask. [a)] Because of high resistance, few cells cannot be estimated their conversion efficiencies, e.g., *FF* > 1.

Table S2. Photovoltaic parameters of $(FAPbI_3)_{0.85}(MAPbBr_3)_{0.15}$ PSCs based on Nb/TiO_x CLs with different Nb doping ratio dried at 150 °C

Compact layer	scan direction[a)]	J_{SC} (mA cm^{-2})	V_{OC} (V)	*FF*	η (%)	No. of cells
TiO_x	forward	21.48±0.94 (21.63)	1.10±0.02 (1.13)	0.54±0.15 (0.67)	12.7±3.4 (16.4)	20
	reverse	20.30±0.49 (20.51)	1.13±0.02 (1.16)	0.74±0.05 (0.79)	17.3±1.4 (19.1)	
Nb 1%	forward	22.20±0.74 (23.11)	1.08±0.01 (1.08)	0.56±0.04 (0.63)	13.5±1.2 (15.6)	14
	reverse	21.79±0.23 (22.30)	1.12±0.01 (1.13)	0.74±0.02 (0.76)	17.9±0.8 (19.2)	
Nb 2%	forward	22.06±0.60 (22.28)	1.05±0.02 (1.04)	0.53±0.07 (0.63)	12.3±1.8 (14.6)	16
	reverse	21.68±0.69 (22.31)	1.10±0.02 (1.13)	0.68±0.07 (0.74)	16.3±2.1 (18.4)	
Nb 3%	forward	21.68±0.52 (21.66)	1.07±0.02 (1.09)	0.60±0.04 (0.64)	13.9±1.1 (15.0)	18
	reverse	21.27±0.61 (21.90)	1.11±0.02 (1.12)	0.73±0.04 (0.75)	17.2±1.2 (18.4)	
Nb 4%	forward	22.08±0.56 (22.26)	1.09±0.02 (1.12)	0.59±0.07 (0.68)	14.2±2.0 (17.0)	15
	reverse	21.75±0.64 (22.36)	1.13±0.01 (1.14)	0.73±0.04 (0.77)	17.9±1.2 (19.6)	
Nb 5%	forward	21.29±0.89 (22.33)	1.08±0.01 (1.09)	0.49±0.07 (0.57)	11.5±1.9 (13.9)	14
	reverse	20.64±0.97 (20.71)	1.12±0.01 (1.13)	0.73±0.04 (0.77)	16.8±1.3 (18.1)	

Scan direction: forward $J_{SC} \rightarrow V_{OC}$ and reverse: $V_{OC} \rightarrow J_{SC}$. In parentheses, the parameters of the best cell. Cell active area is 5 × 5 mm^2 defined by black mask.

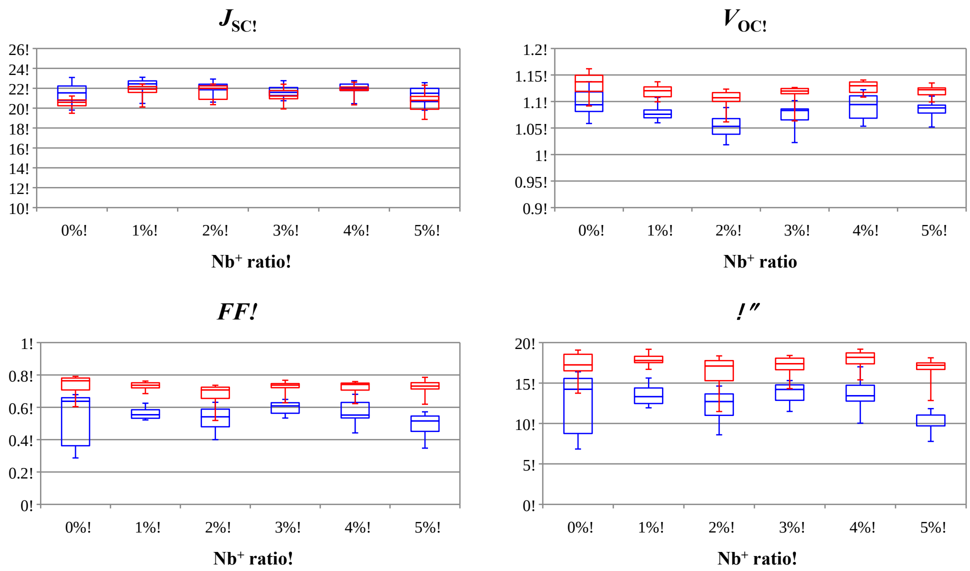

Figure S8. Nb-doping ratio dependency of photovoltaic parameters of $(FAPbI_3)_{0.85}(MAPbBr_3)_{0.15}$ PSCs based on Nb/TiO_x CLs dried at 150 °C.

Table S3. Photovoltaic parameters in reverse scans ($V_{OC} \rightarrow J_{SC}$) of PSCs based on $FA_{0.85}Cs_{0.15}PbI_3$ perovskite annealed at different temperatures.

Anneal temp.	No. of cells	J_{SC} (mA cm^{-2})	V_{OC} (V)	*FF*	η (%)
100°C	6 cells	19.56±0.22	0.93±0.02	0.44±0.02	8.1±0.5
	best eff.	19.70	0.95	0.48	9.0
110°C	6 cells	19.43±0.29	0.98±0.02	0.52±0.04	10.0±1.0
	best eff.	19.12	1.00	0.59	11.3
120°C	6 cells	21.25±0.27	0.97±0.04	0.52±0.03	10.8±1.0
	best eff.	21.52	1.01	0.55	11.9
130°C	12 cells	21.43±0.66	1.00±0.04	0.58±0.04	12.6±1.4
	best eff.	22.21	1.03	0.64	14.6
140°C	6 cells	20.33±0.76	1.01±0.03	0.61±0.04	12.6±1.3
	best eff.	20.01	1.04	0.64	14.0
150°C	6 cells	19.53±0.27	1.03±0.02	0.61±0.07	12.3±1.6
	best eff.	19.81	1.04	0.66	13.5
160°C	6 cells	17.96±1.12	0.97±0.05	0.53±0.10	9.2±2.0
	best eff.	19.59	1.03	0.62	12.4
170°C	6 cells	17.44±0.47	0.99±0.02	0.57±0.03	10.0±0.8
	best eff.	18.08	1.01	0.60	10.9

The PSCs are prepared on a substrate: FTO/high-temperature sintered TiO_2 CL/low-temperature dried brookite mesoporous layer. Scan direction: forward $J_{SC} \rightarrow V_{OC}$ and reverse: $V_{OC} \rightarrow J_{SC}$. In parentheses, the parameters of the best cell. Cell active area is 5 × 5 mm^2 defined by black mask.

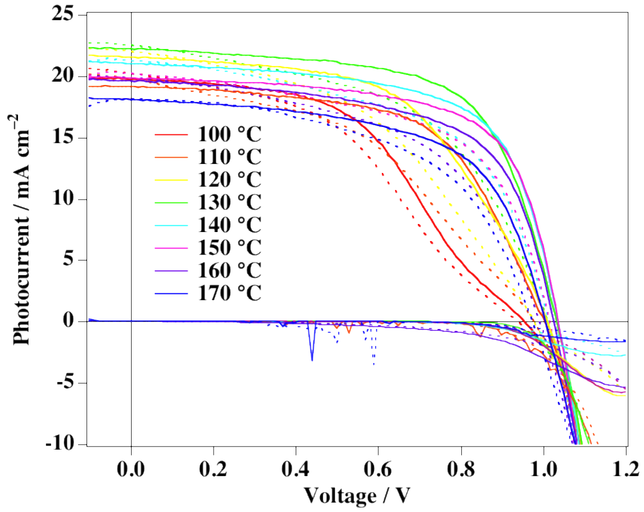

Figure S9. *J-V* curves of $FA_{0.85}Cs_{0.15}PbI_3$ based PSCs annealed at different temperatures. Forward (broken line) and reverse (solid line) scans.

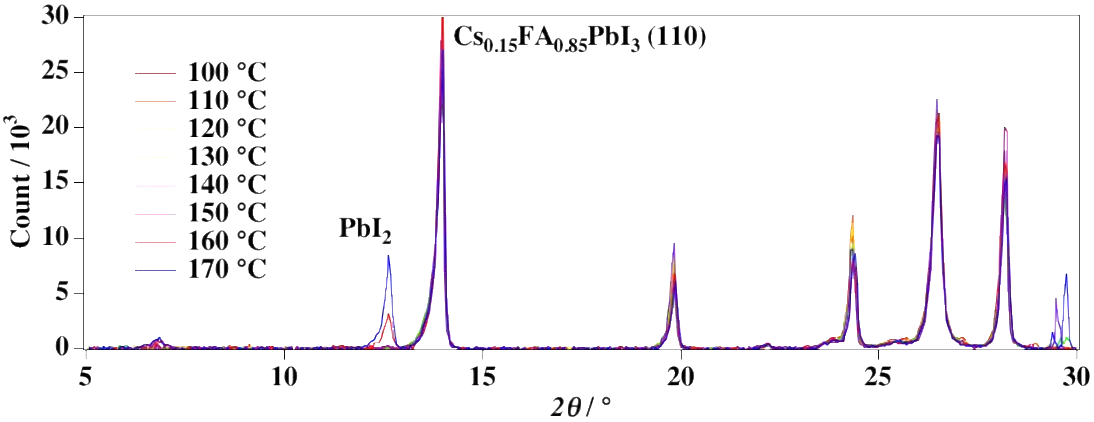

Figure S10. XRD chart of $FA_{0.85}Cs_{0.15}PbI_3$ perovskite film on mesoporous substrate prepared by different anneal temperatures.

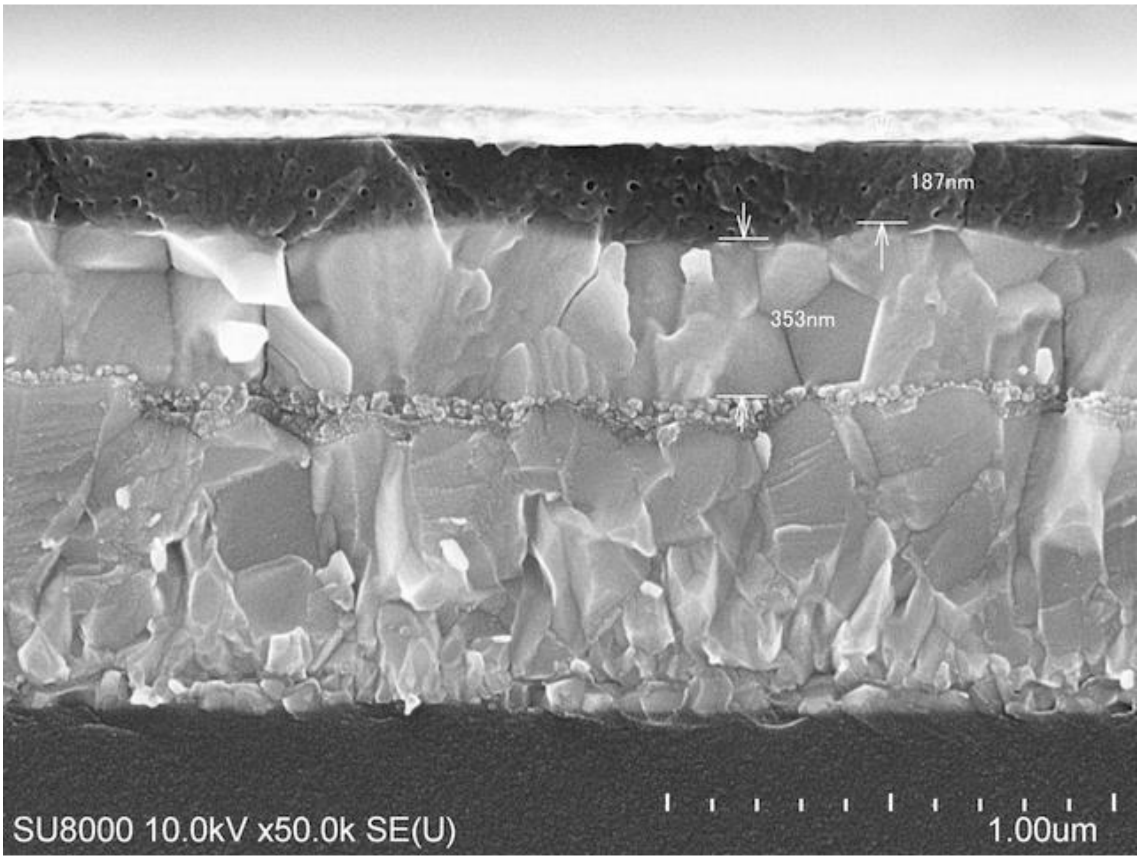

Figure S11. Cross-sectional SEM image of $FA_{0.85}Cs_{0.15}PbI_3$ PSC based on (4%) Nb/TiOx CL. For pinholes in the HTL, it is expected that they are formed by thermal stress during gold evaopration.[1]

Reference

1 A. K. Jena, M. Ikegami, and T. Miyasaka, *ACS Energy Lett*., 2017, **2**, 1760-1761.