Electronic Supplementary Material (ESI) for Sustainable Energy & Fuels.

Supporting Information

Layering technique for achieving pinhole-free organic-inorganic halide perovskite thin films through vapor-solid reaction

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Keywords: *Lead halide perovskite; Pinhole-free; Vapor–solid reaction; thin film; Layering technique*

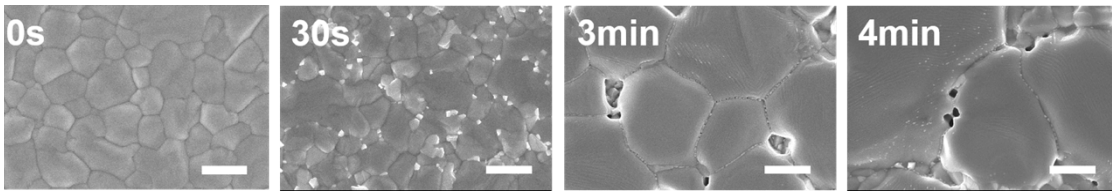

Figure S1. Scanning electron microscope (SEM) images of the perovskite thin films with different reaction times in FASCN vapor. Scale bar, 1 μm.

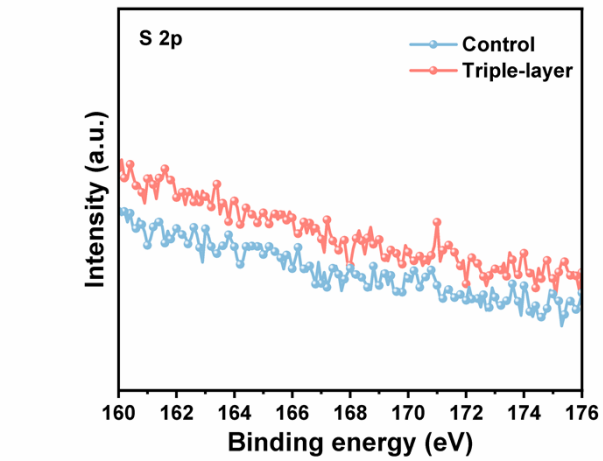

Figure S2. XPS spectra of the S 2p signal in the perovskite thin film with control and triple-layer.

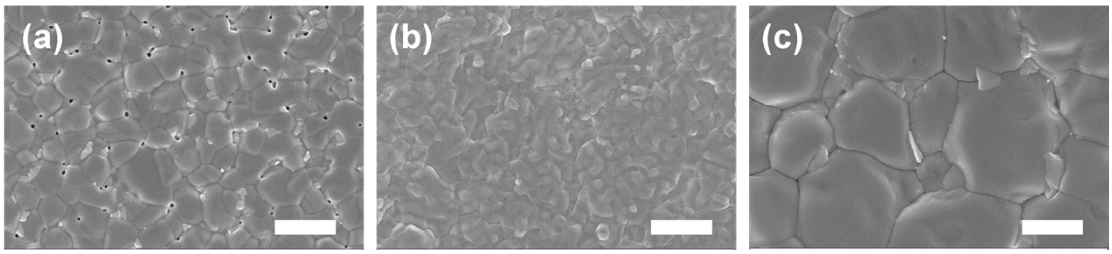

Figure S3. Scanning electron microscope (SEM) images during the preparation of double-layer perovskite thin films. (a) 300 nm perovskite thin film treated with FASCN vapor, (b) 300nm FASCN-perovskite/ 300 nm vapor-solid perovskite, (c) 300 nm FASCN-perovskite/300 nm FASCN-perovskite. Scale bar, 2 μm.

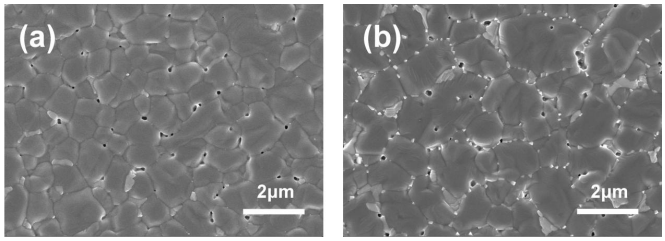

Figure S4. SEM images of the FASCN vapor-treated films (a) unannealed and (b) annealed.

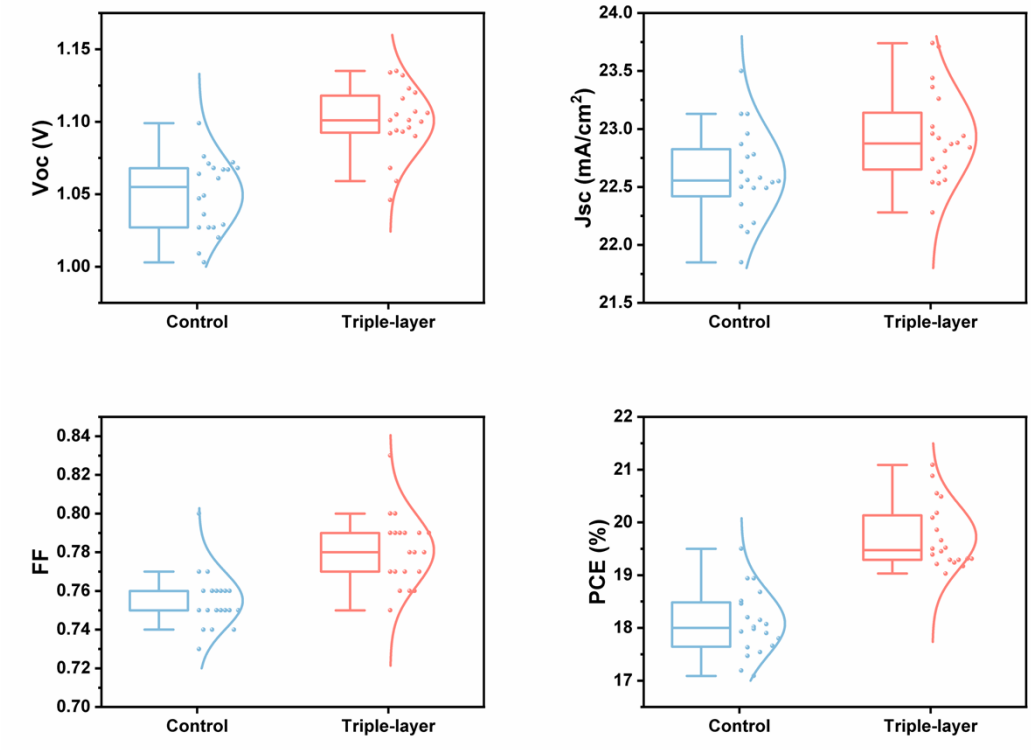

Figure S5. Statistics of the photovoltaic parameters distribution for solar cells based on control and triple-layer perovskite, 20 independent cells are fabricated for each type.

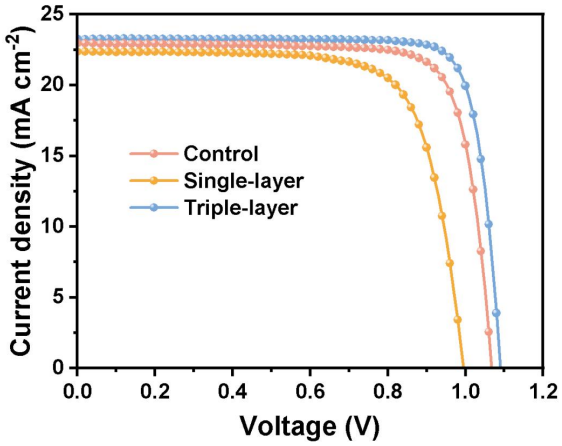

Figure S6. J-V curves of devices based on different layers of thin films

Table S1. Biexponential fitted TRPL parameters of the control and triple-layer samples.

Samples	A_1	τ_1 (ns)	A_2	τ_2 (ns)	τ_{avg} (ns)
Control	0.503	7.30	0.423	154.26	146.43
Triple-layer	0.510	7.85	0.424	318.48	309.54

Table S2. J-V parameters of the champion devices measured from forward scan (FS) and reverse scan (RS).

Champion device		V_{oc} (V)	J_{sc} (mA cm^{-2})	FF	PCE (%)
Control	RS	1.068	22.96	0.80	19.50
	FS	1.047	22.76	0.76	18.20
Triple-layer	RS	1.090	23.26	0.83	21.09
	FS	1.088	23.15	0.80	20.06

Table S3 Photovoltaic parameters of devices based on different layers of films.

Device	V_{oc} (V)	J_{sc} (mA cm^{-2})	FF	PCE (%)
Control	1.068	22.96	0.80	19.50
Single-layer	0.995	22.35	0.74	16.42
Triple-layer	1.090	23.26	0.83	21.09

Table S4 EIS Fitting parameters of solar cells based on control and triple-layer modified devices. The series resistance (R_s) and charge composite resistance (R_{rec}) can be obtained by data fitting of equivalent circuit diagram of PSCs by EC-Lab software.

Samples	R_s **(Ω)**	R_{rec} **(kΩ)**
Control	9.962	2.988
Triple-layer	9.379	6.978