Electronic Supplementary Information (ESI)

Boosting the Performance and Stability of Quasi-Two-Dimensional Tin-Based Perovskite Solar Cells Using a Formamidinium Thiocyanate Additive

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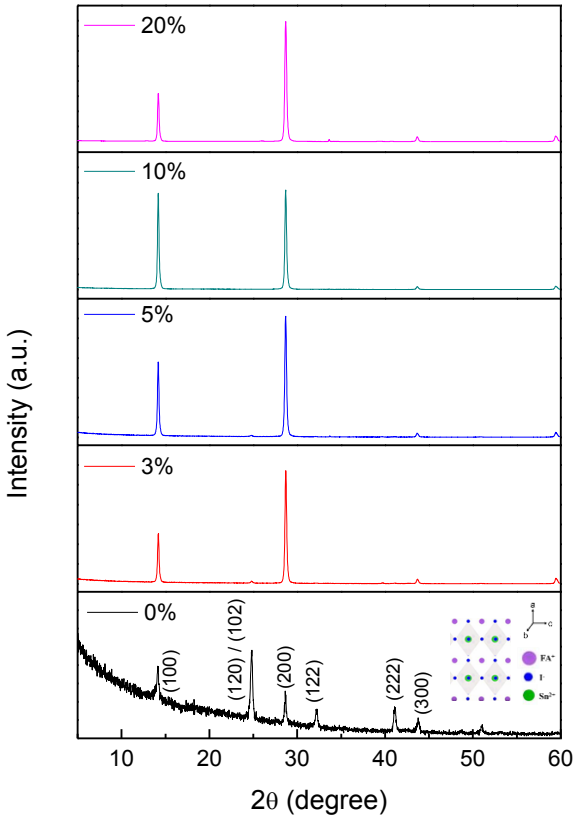

Fig. S1 XRD diffractograms of quasi-2D tin-based perovskite films with different molar ratios of PEAI to FAI: 0% ($FASnI_3$), 3%, 5%, 10%, and 20%.

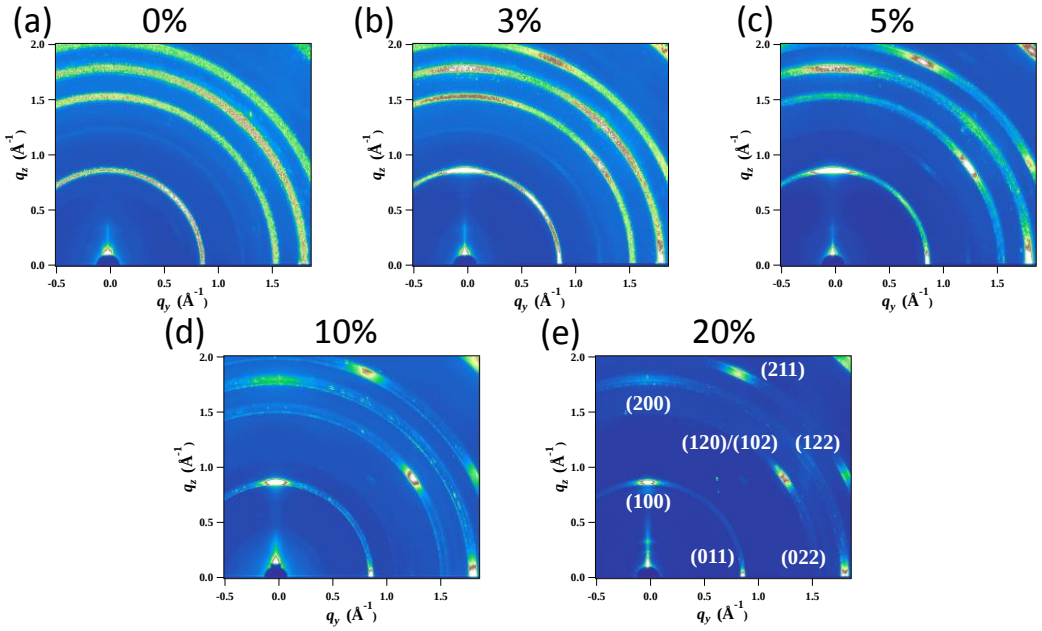

Fig. S2 GIWAXS images of quasi-2D tin-based perovskite films with different molar ratios of PEAI to FAI: (a) 0% ($FASnI_3$), (b) 3%, (c) 5%, (d) 10%, and (e) 20%.

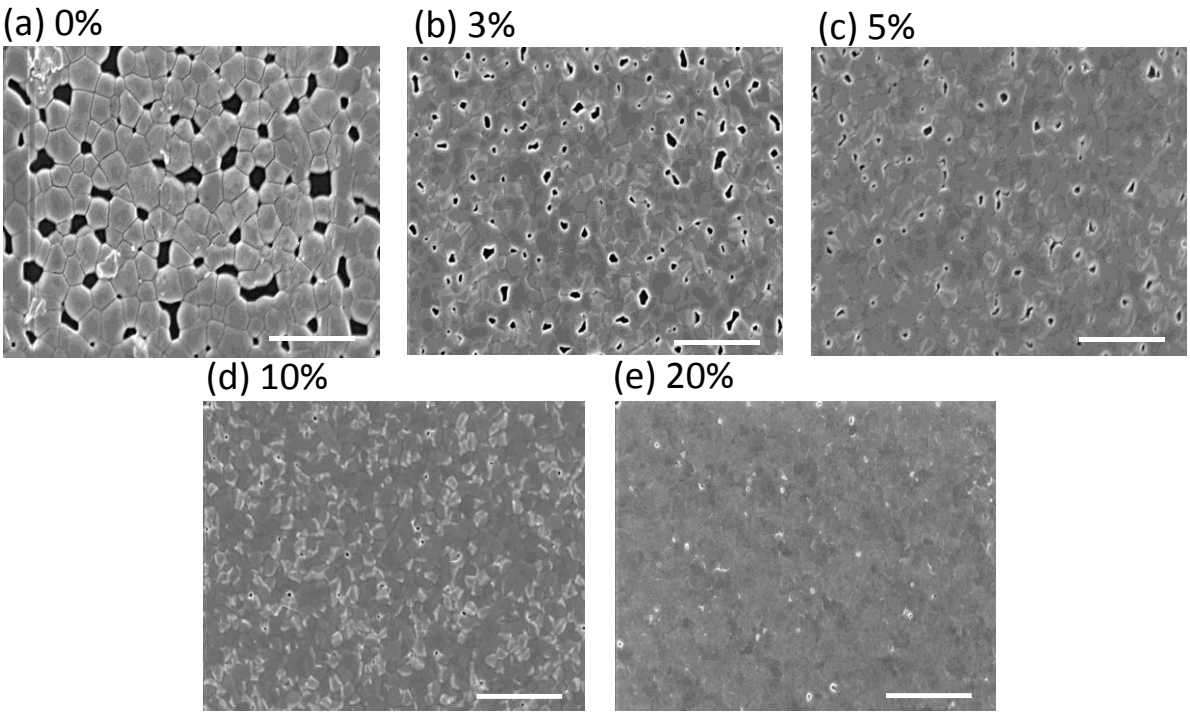

Fig. S3 SEM images of quasi-2D tin-based perovskite films with different molar ratios of PEAI to FAI: 0% ($FASnI_3$), 3%, 5%, 10%, and 20%. Scale bar in all images indicates 2.5 μm.

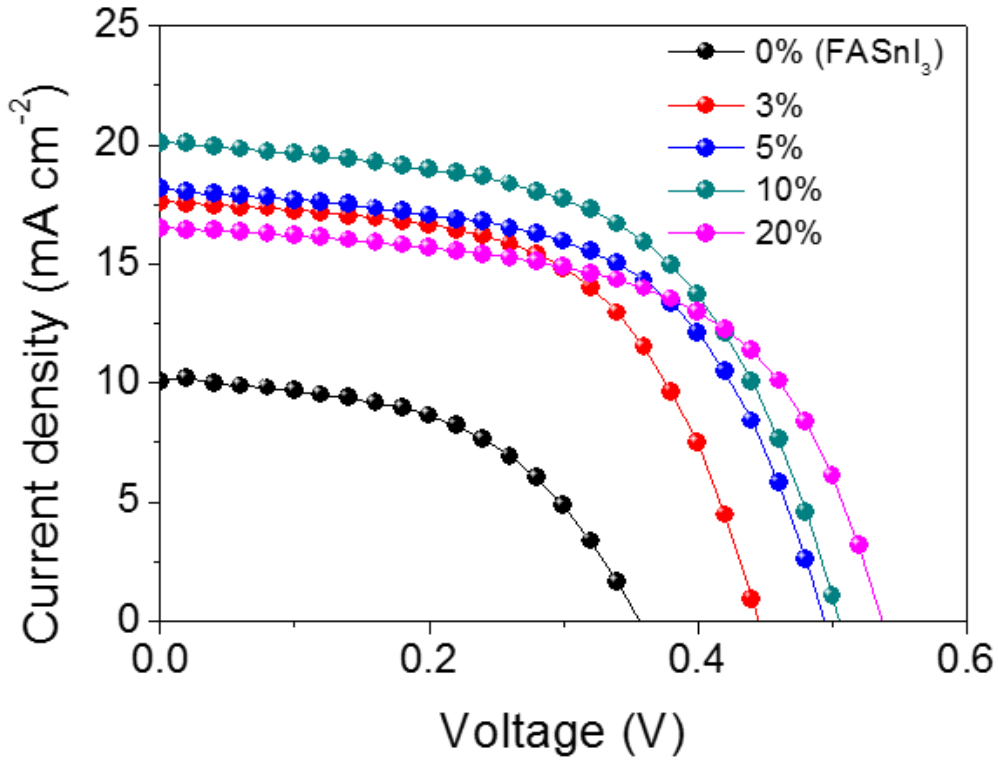

Fig. S4 *J*–*V* curves of 3D $FASnI_3$ and quasi-2D tin-based perovskite solar cells with different molar ratios of PEAI to FAI under AM 1.5 G illumination.

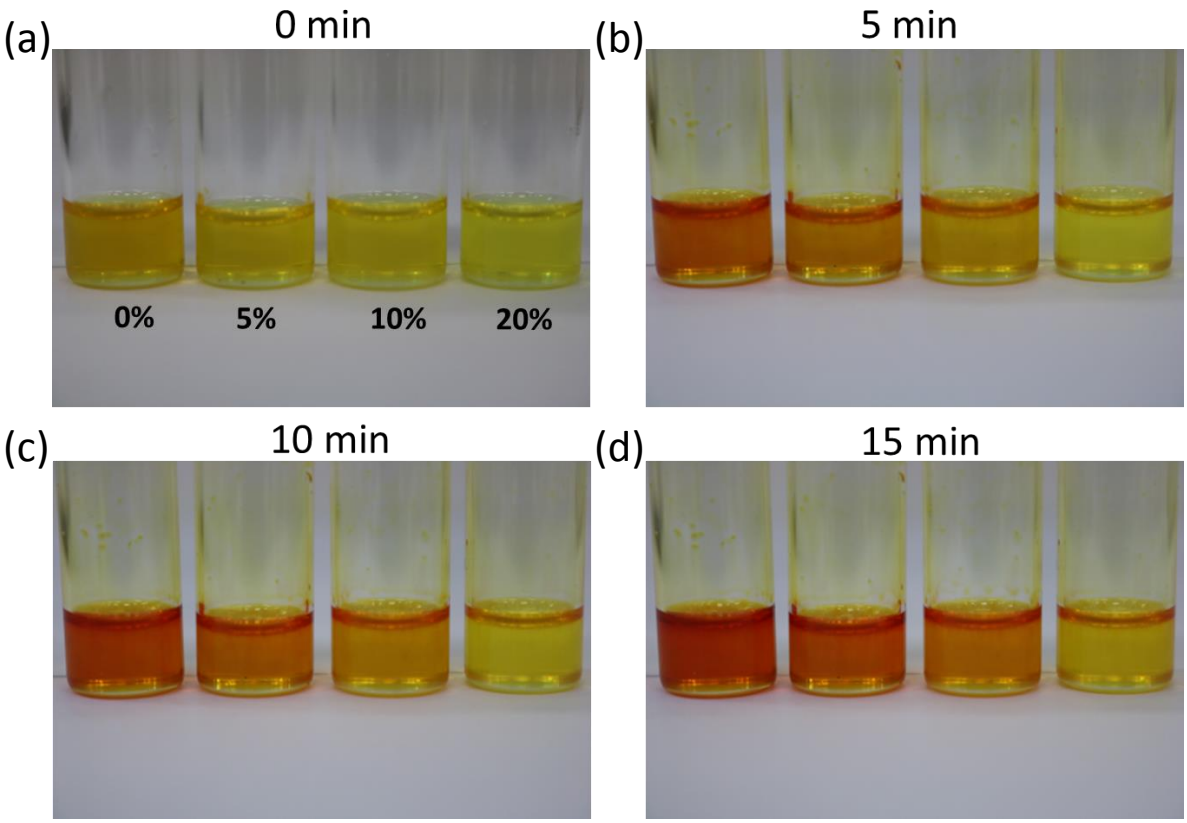

Fig. S5 Photographs of SnI_2 solutions in DMF:DMSO (8:2 volume ratio) with different concentrations of the FASCN additive (0, 5, 10, and 20 mol%) without SnF_2 incorporation as functions of exposure time to air: (a) 0, (b) 5, (c) 10, and (d) 15 min.

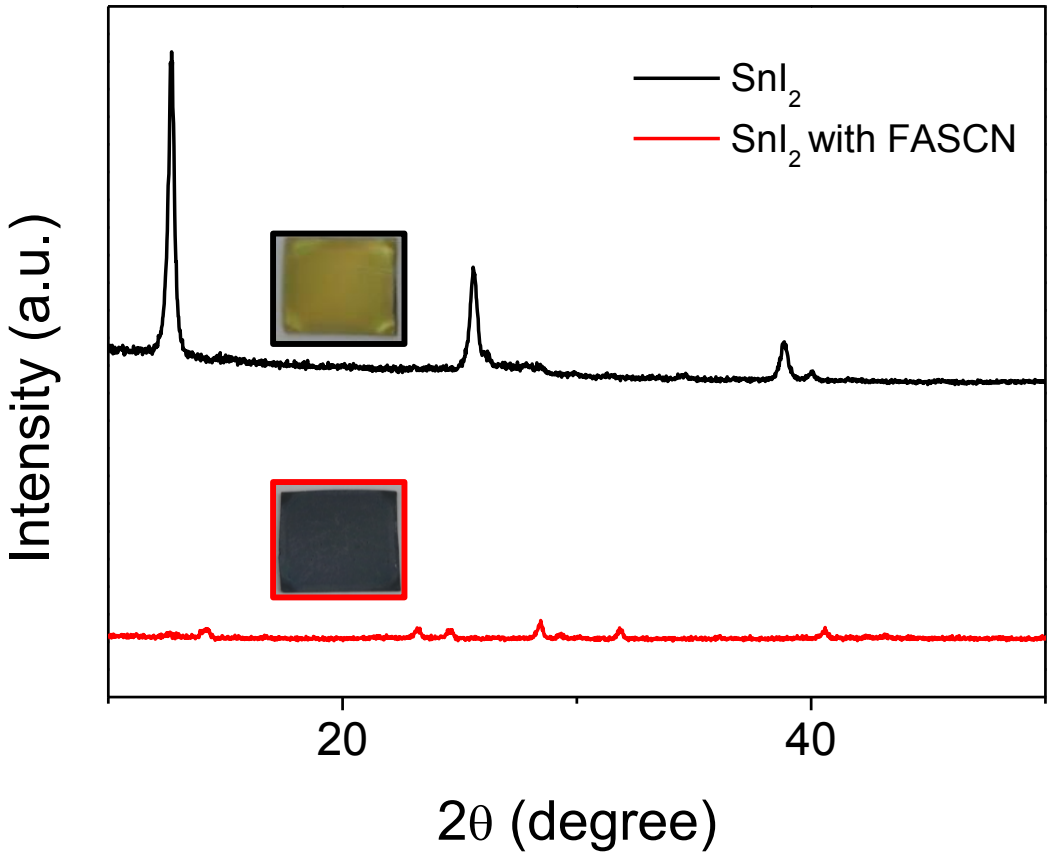

Fig. S6 XRD diffractograms of SnI_2 films with or without FASCN additive. Inset photographs display SnI_2 films coated on Si wafers corresponding to the XRD results. SnI_2 films were fabricated in a nitrogen-filled glove box and exposed to ambient air for 30 minutes for XRD analysis.

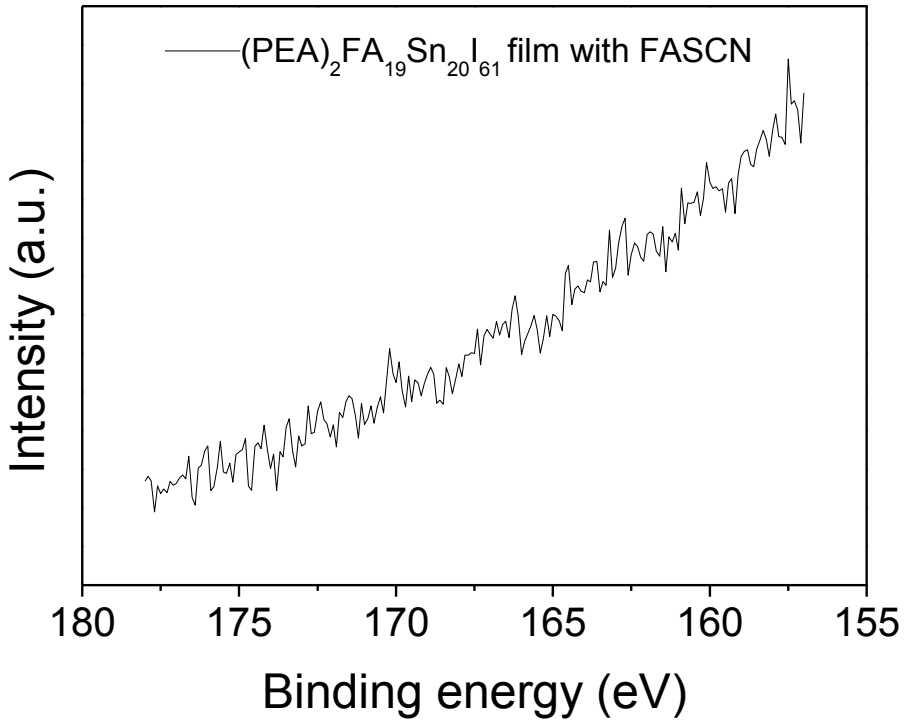

Fig. S7 XPS spectrum showing the absence of S (2p) peak in the final 10% PEAI film with FASCN additive. The S (2p) peak is expected to appear at the binding energy near 165 eV.

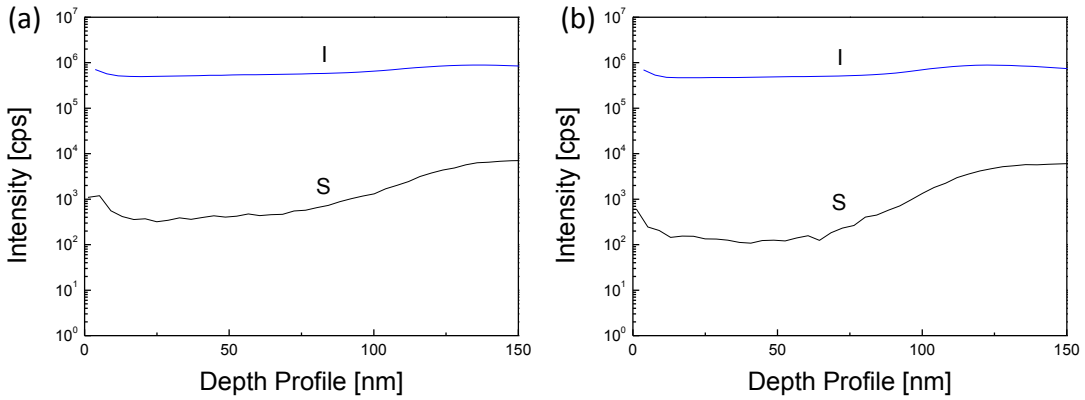

Fig. S8 ToF-SIMS depth profiles for quasi-2D tin-based perovskite films with or without FASCN additive (5 mol%) (a) before annealing and (b) after annealing.

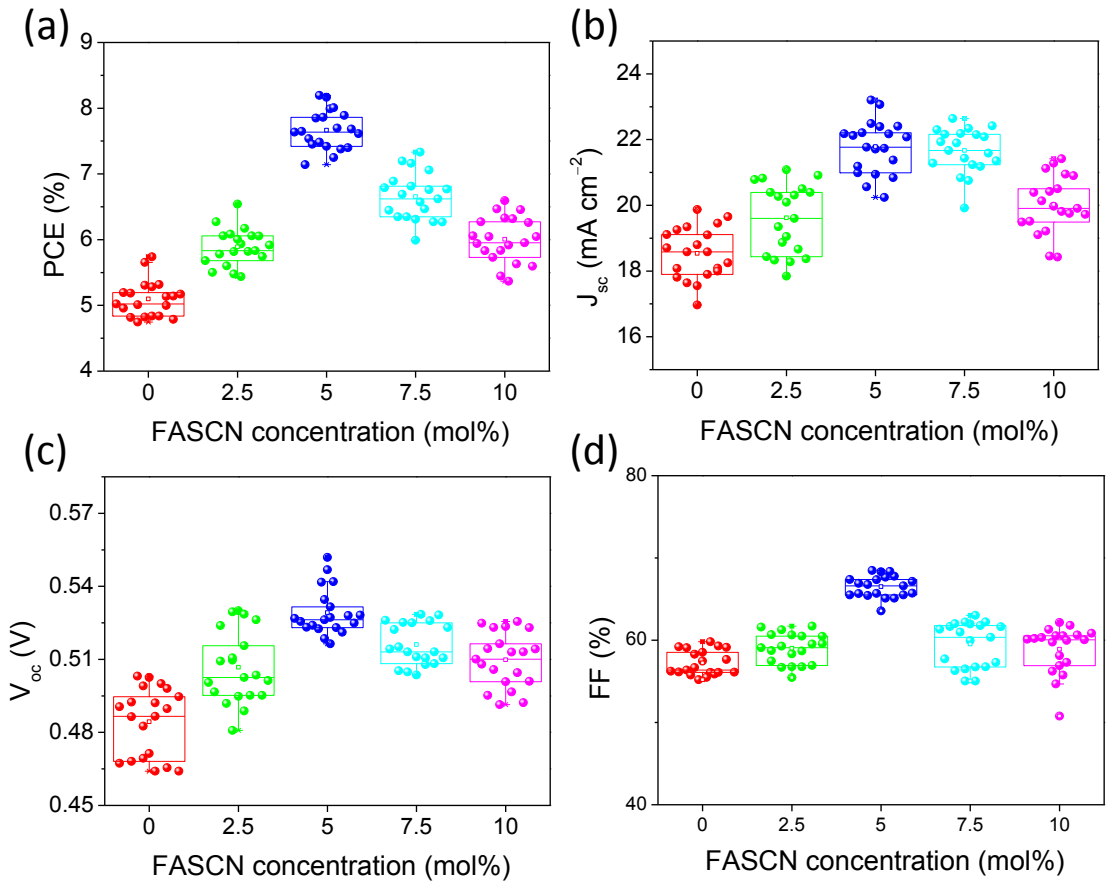

Fig. S9 Statistics of photovoltaic parameters including (a) PCE, (b) J_{sc}, (c) V_{oc}, and (d) FF of the best-performing 20 devices with different amounts of FASCN additive.

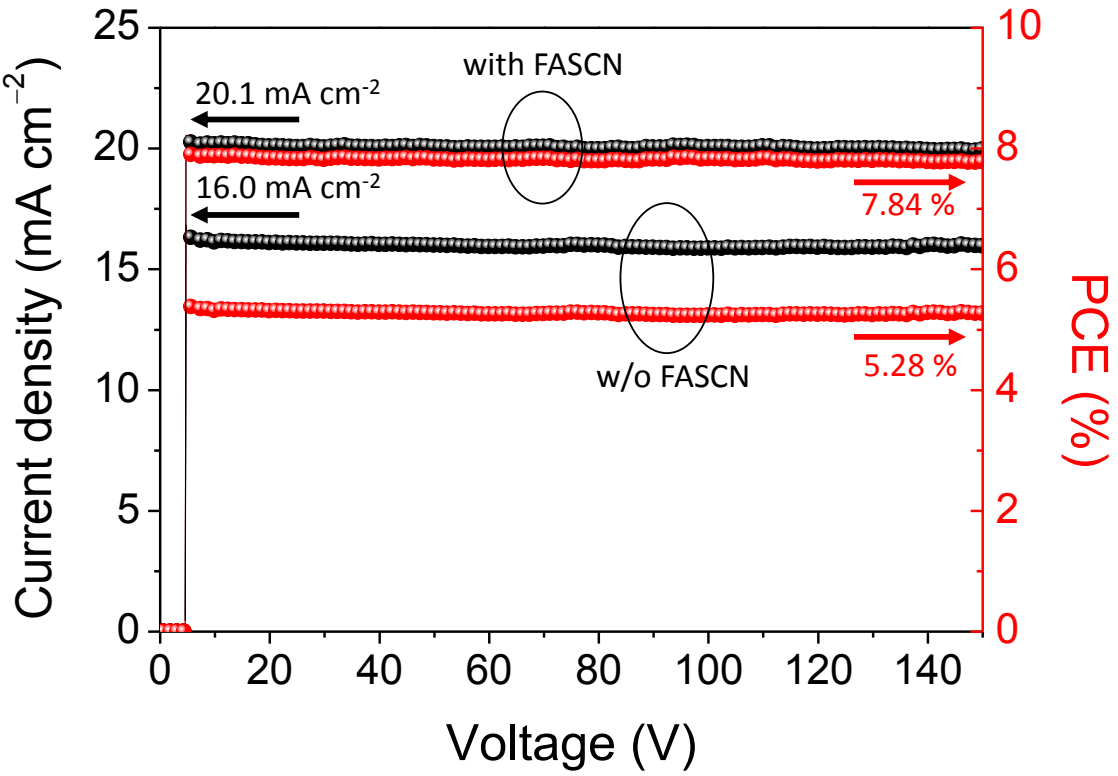

Fig. S10 Steady-state photocurrents and PCEs of the best-performing devices without or with FASCN additive measured under a constant bias of 0.33 V (without FASCN) and 0.39 V (with FASCN) near their MPP.

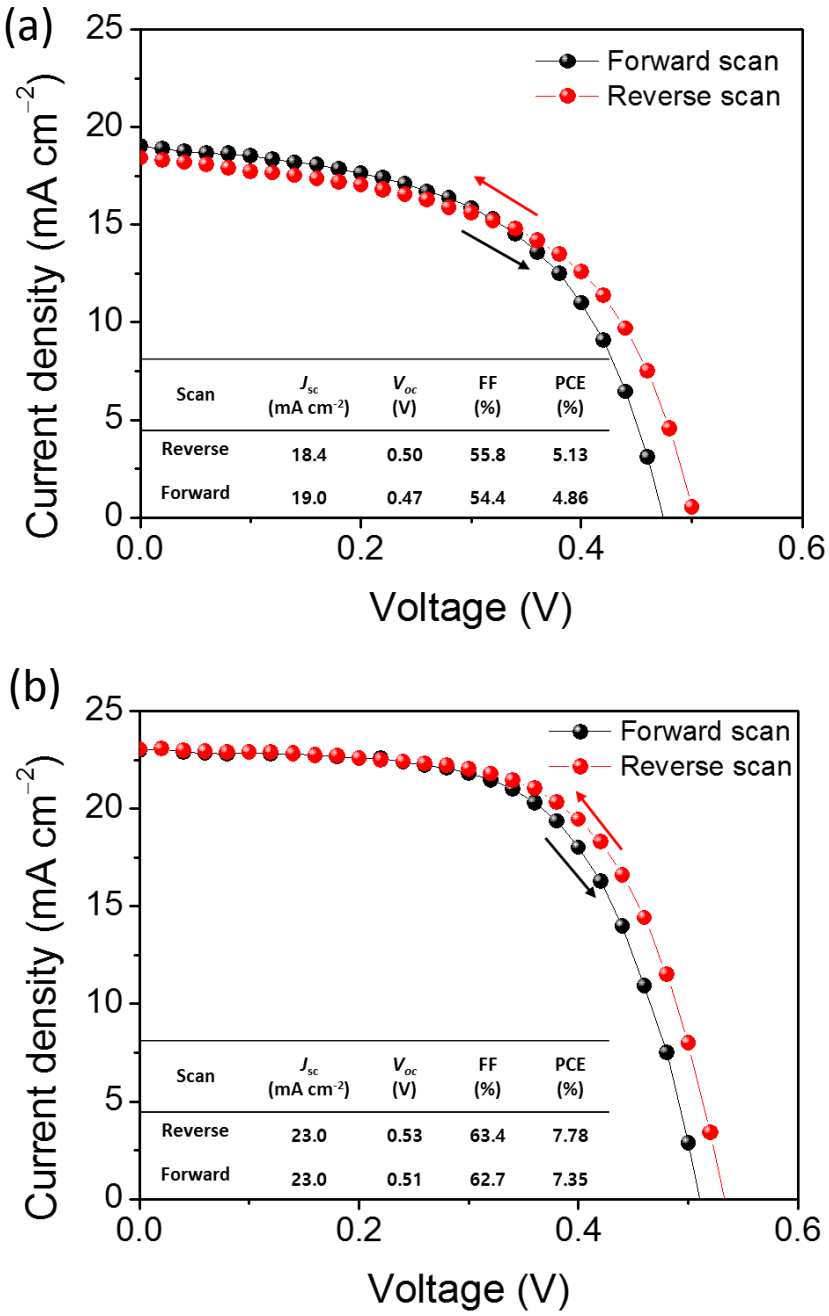

Fig. S11 *J*–*V* curves of the optimized devices (a) without or (b) with FASCN additive in forward and reverse scans.

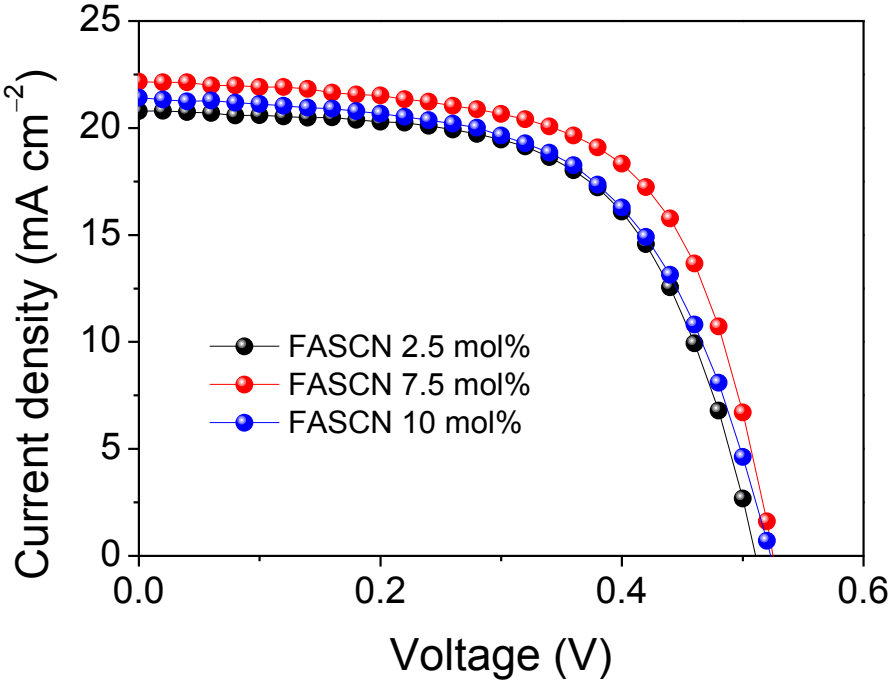

Fig. S12 *J–V* curves of the best-performing devices with different amounts of FASCN additive.

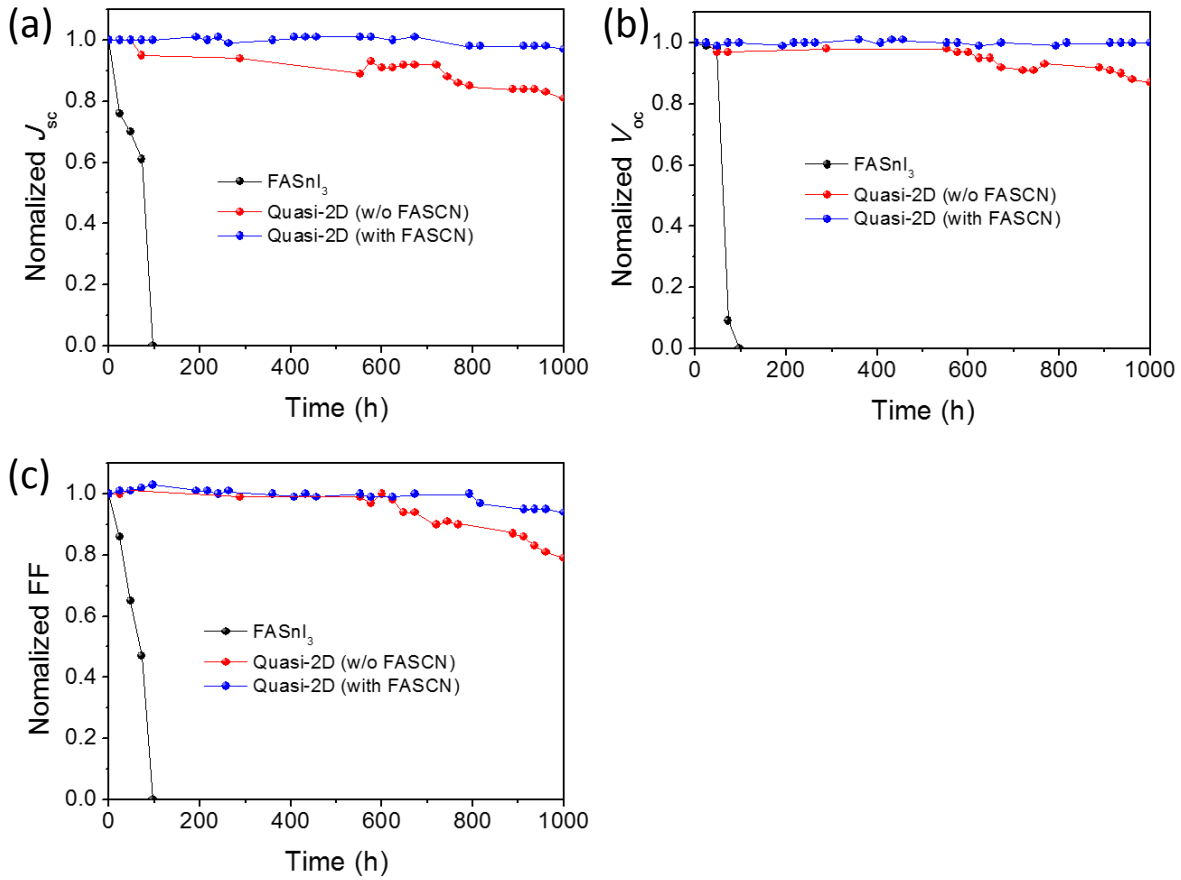

Fig. S13 Variations in photovoltaic parameters including (a) J_{sc}, (b) V_{oc}, and (c) FF for stability tests over 1000 hours in a nitrogen-filled glove box for $FASnI_3$ and 10% PEAI devices with or without FASCN additive. Initial photovoltaic parameters: $FASnI_3$ (J_{sc}: 10.0 mA cm^{-2}, V_{oc}: 0.35 V, FF: 52.0%, PCE: 1.81%); Quasi-2D (w/o FASCN) (J_{sc}: 18.8 mA cm^{-2}, V_{oc}: 0.50 V, FF: 57.7%, PCE: 5.40%); Quasi-2D (with FASCN) (J_{sc}: 22.2 mA cm^{-2}, V_{oc}: 0.53 V, FF: 67.4%, PCE: 7.86%)

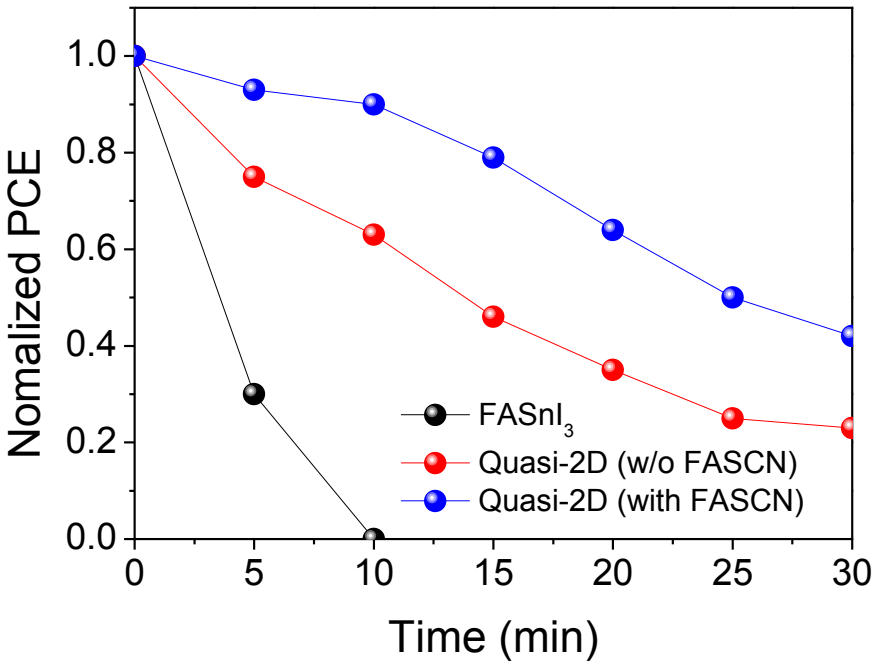

Fig. S14 Stability tests for $FASnI_3$ and 10% PEAI devices with or without FASCN additive in ambient condition with a 40% relative humidity at room temperature. Initial photovoltaic parameters: $FASnI_3$ (J_{sc}: 9.7 mA cm^{-2}, V_{oc}: 0.34 V, FF: 51.3%, PCE: 1.68%); Quasi-2D (w/o FASCN) (J_{sc}: 19.5 mA cm^{-2}, V_{oc}: 0.50 V, FF: 56.7%, PCE: 5.53%); Quasi-2D (with FASCN) (J_{sc}: 21.7 mA cm^{-2}, V_{oc}: 0.52 V, FF: 67.8%, PCE: 7.70%)

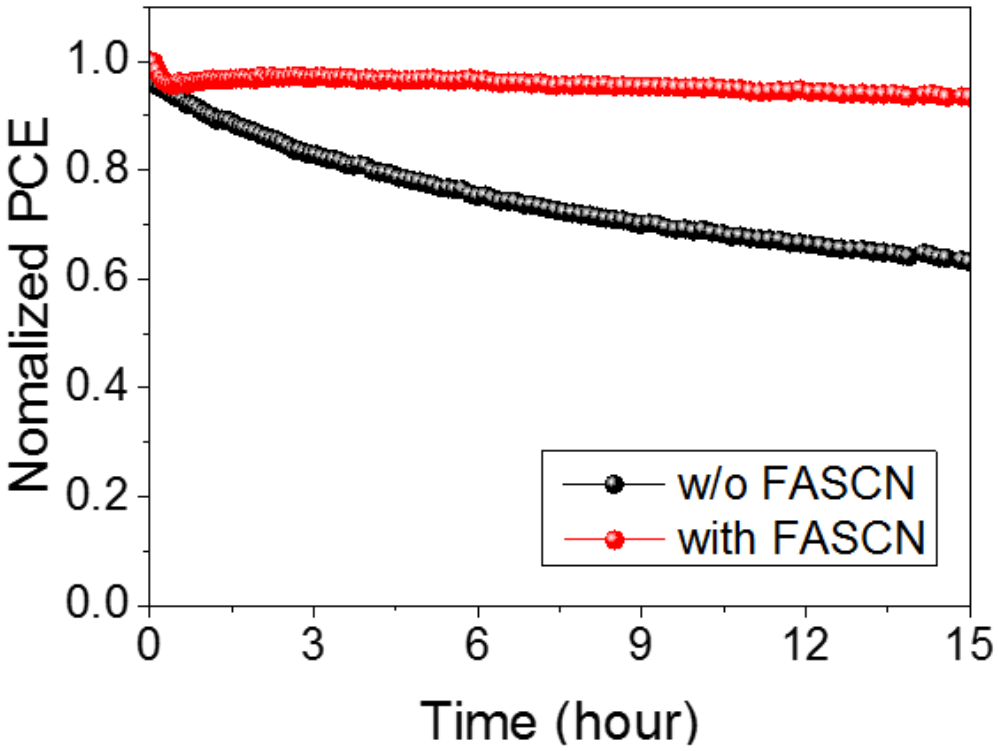

Fig. S15 PCE tracking for 10% PEAI devices with or without FASCN additive under continuous 1 sun illumination at their MPP, which was measured in a nitrogen-filled glove box at 25 °C. (Initial photovoltaic parameters: Quasi-2D (w/o FASCN) (J_{sc}: 17.6 mA cm^{-2}, V_{oc}: 0.49 V, FF: 57.9%, PCE: 5.01%, V_{mpp}: 0.36 V); Quasi-2D (with FASCN) (J_{sc}: 22.4 mA cm^{-2}, V_{oc}: 0.53 V, FF: 67.7%, PCE: 7.99%, V_{mpp}: 0.40 V)

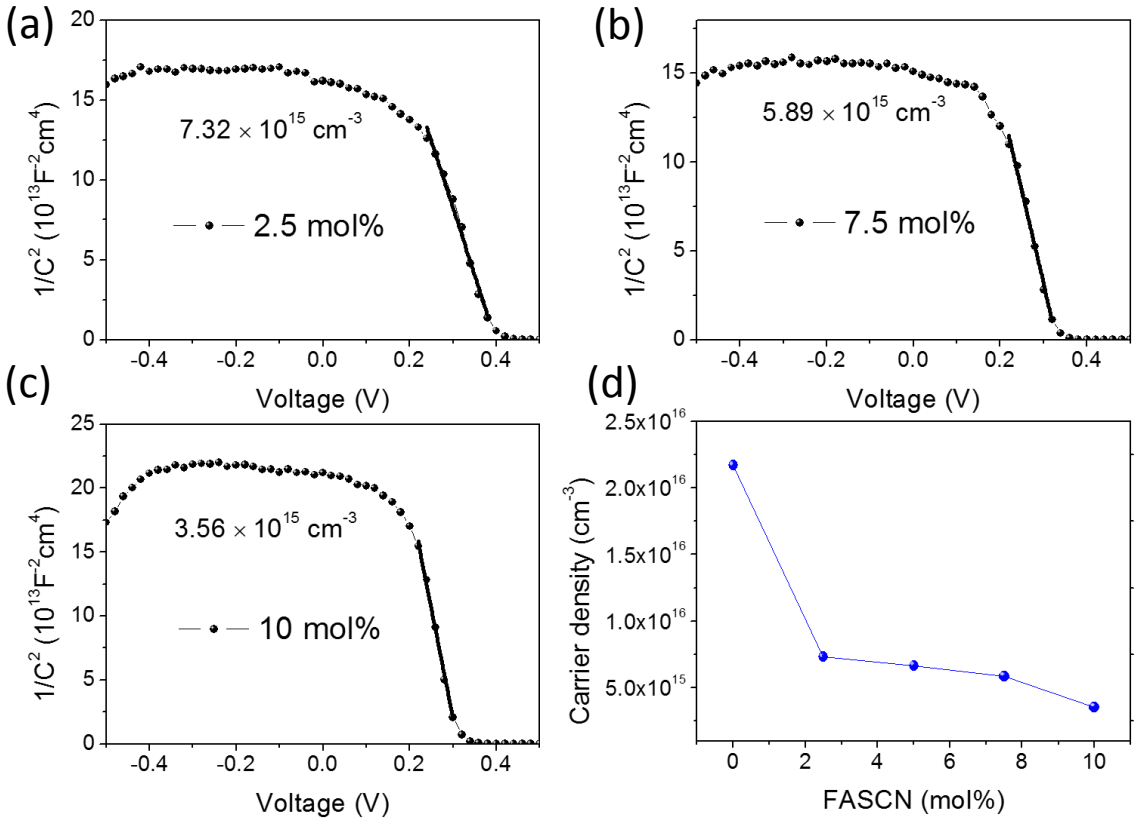

Fig. S16 (a-c) *C–V* measurement results of the 10% PEAI devices with different amounts of FASCN where the calculated carrier density for each device is noted in the inset, (d) Carrier density profile as a function of FASCN concentration.

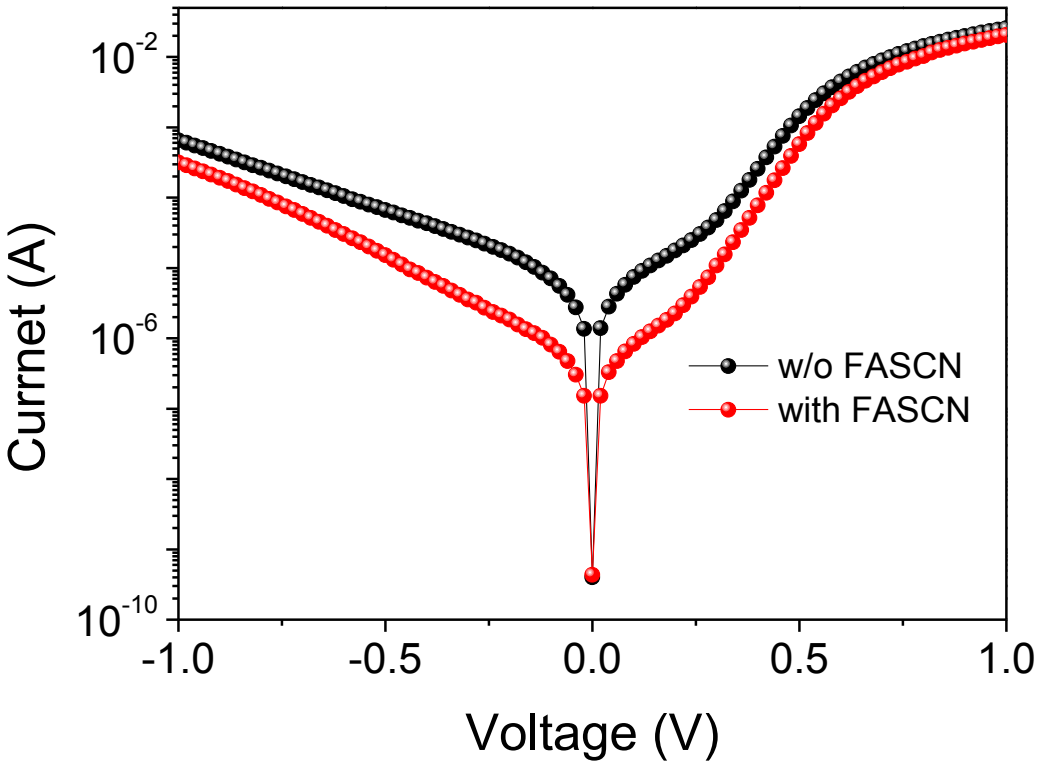

Fig. S17 Dark *I*–*V* curves of the 10% PEAI devices with or without FASCN additive.

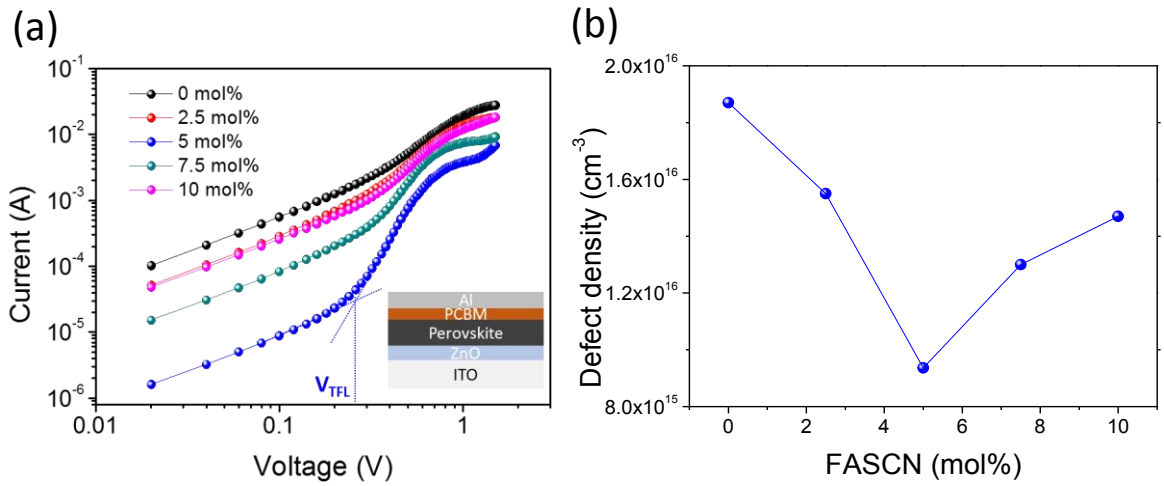

Fig. S18 (a) Dark *I*–*V* plots of electron-only devices with different amounts of FASCN additive where the configuration of electron-only device is shown in the inset, (b) Calculated trap densities as a function of amounts of FASCN additive.

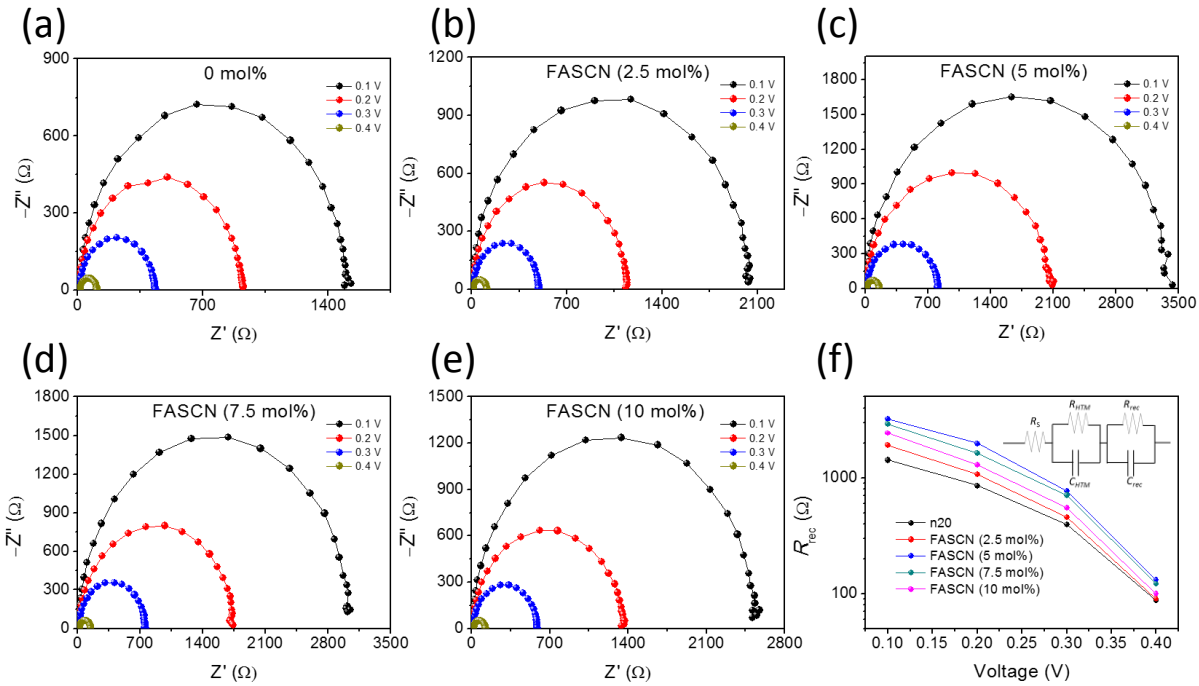

Fig. S19 EIS studies under AM 1.5 G illumination at 100 mW cm^{-2} at different forward voltages. (a-e) The resulting Nyquist plots with different amounts of FASCN additive and (f) their corresponding R_{rec} as a function of applied forward voltage where the equivalent circuit used for fitting is shown in inset. (R_s is the series resistance, R_{HTM} is the hole transporting material (HTM) resistance, R_{rec} is the recombination resistance, C_{HTM} is the HTM capacitance, and C_{rec} is the recombination capacitance, respectively.)

Table S1. Photovoltaic parameters of 3D $FASnI_3$ and quasi-2D tin-based perovskite solar cells

SnF_2 Concentration [mol%]	Molar ratio of PEAI to FAI [%]	J_{sc} [mA cm^{-2}]	V_{oc} [V]	FF [%]	PCE [%]
10	0	10.1	0.36	51.2	1.84
	3	17.6	0.45	57.3	4.53
	5	18.1	0.49	57.4	5.15
	10	19.9	0.50	57.5	5.74
	20	16.5	0.53	58.0	5.07

Table S2. Photovoltaic parameters of the best-performing devices with different amounts of FASCN additive

FASCN Concentration [mol%]	J_{sc} [mA cm^{-2}]	V_{oc} [V]	FF [%]	PCE [%]
2.5	20.8	0.51	61.7	6.54
7.5	22.2	0.53	63	7.33
10	21.4	0.52	58.9	6.59

Table S3. Summary for comparing the performances of recently reported $FASnI_3$-based perovskite solar cells and our work

Absorber	Device structure	Coating method	J_{sc} [mA cm^{-2}]	V_{oc} [V]	FF [%]	PCE [%]	Glove box level [ppm]	Ref.
$FASnI_3$ + pyrazine	Conventional	One-step	23.7	0.32	63	4.8 (2.8)[a]	N/A	(34)
$FA_{1-x}MA_xSnI_3$	Inverted	One-step	21.2	0.61	62.7	8.12 (6.6)	$O_2 < 1.0$ $H_2O < 0.02$	(22)
$(PEA)_2FA_{n-1}Sn_nI_{3n+1}$ (20% PEAI)	Inverted	One-step	14.44	0.59	69	5.94 (1.21)	$O_2 < 3$ $H_2O < 1$	(30)
$(PEA)_2FA_{n-1}Sn_nI_{3n+1}$ (8% PEAI)	Inverted	One-step	24.1	0.525	71	9 (6)	N/A	(31)
{en}$FASnI_3$	Conventional	One-step	22.54	0.48	65.96	7.14 (2.34)	N/A	(27)
$FASnI_{1-x}Br_x$	Conventional	One-step	19.8	0.414	66.9	5.5 (3.46)	N/A	(23)
$FASnI_3$ + SnF_2 + TMA	Inverted	Two-step	22.45	0.47	67.8	7.09 (4.2)	N/A	(24)
$FASnI_3$ + SnF_2 + TMA	Conventional	Two-step	21.65	0.31	64.7	4.34 (2.26)	N/A	(24)
$(PEA)_2FA_{n-1}Sn_nI_{3n+1}$ (10% PEAI)	Inverted	One-step	22.5	0.53	68.3	8.17 (1.84)	$O_2 < 5$ $H_2O < 2.2$	Our work

[a]The efficiency of the reference $FASnI_3$ device presented in the corresponding paper.