Supporting Information

Templated Growth of $FASnI_3$ Crystals for Efficient Tin Perovskite Solar Cell

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Materials

The following chemicals were used as received form commercial sources, including SnI_2 (99.99%, Sigma-Aldrich), $CH(NH_2)_2I$ (FAI) (>98%, Tokyo Chemical Industry Co., Japan), $CH_3(CH_2)_2NH_3I$ (PAI) (>99%, Sigma-Aldrich), SnF_2(>99%, Sigma-Aldrich), C_{60} (99.5%, Sigma-Aldrich), bathocuproine (>99%, Wako), All solutions were filtered with a 0.2 μm PTFE filter before using.

Solar cell fabrication

Patterned ITO glass substrates were cleaned in the order of detergent, deionized water, acetone, and isopropanol for 15 min by ultra-sonicated. Then, the cleaned ITO substrates were treated by ultraviolet-ozone for 30 min before the deposition of PEDOT:PSS layer. The perovskite precursor solution composed of 1 M SnI_2, 1 M FAI and 0.1M SnF_2 in DMSO was stirred for 2 hours at room temperature.

The perovskite solar cells were fabricated according to our previous work.[1] In short, perovskite films were spin-coated on the PEDOT:PSS layer at 1000 rpm for 12 s and 5000 rpm for 48 s in the glove box. 80 μL Chlorobenzene was in-situ dripped onto the perovskite film during the second step at 30 s. In order to construct the templated growth structure, the PAI (1-5 mg/mL) dissolved in a mixed solvent of CF and DMSO (100:0.5-2 v/v) were spin-coated on perovskite films at the speed of 2000-6000 rpm. Then the perovskite films were annealed at 60 °C for 10 s and 100 °C for 12 min. Finally, 50 nm C_{60} and 8 nm BCP and 70 nm Ag electrode were evaporated under high vacuum ($< 2 \times 10^{-7}$ Torr). The device areas were defined by a mask with

an aperture area of 0.10 cm^2. For the control of oxygen concentration, an accurate oxygen volume was injected into the glovebox.[2]

Characterization:

The XRD and rocking curves were measured by Rigaku Smart lab thin-film x-ray diffractometer using Cu K_a radiation. The SEM image were measure by HITACHI-SU8000 field-emission scanning electron microscope. The UV-vis spectra were obtained by a Shimadzu UV/vis 3600 spectrophotometer. The XPS spectra were measured by PHI Quantera SXM (ULVAC-PHI) with X-ray source of Al Kα (mono), the incident angle and take-off angle are 90 ^{o} and 45 ^{o}, respectively. The steady state PL and TRPL were measured with a Hamamatsu C12132 fluorescence lifetime spectrometer using a laser at an excitation wavelength of λ=402 nm. The *C-V* measurements and the TPV were measured by a platform for all-in-one characterization of solar cells and OLED (PAIOS) of FLUXim Company. *C-V* measurements were taken at 1 kHz with voltage amplitude of 10 mV under dark at room temperature. TPV were measured at open-circuit condition, the pulse length of a white light is 1 ms, background light of 0.09 Sun (9 mW cm^{-2}) was applied.

The *I-V* curves were measured under forward scan (-0.1 V to 0.8 V) or reverse scan (0.8 V to -0.1 V) by a fixed step voltage of 10 mV and delay time of 50 ms by a solar simulator with standard AM 1.5G sunlight (100 mW cm^{-2}, WXS-155S-10, Wacom Denso) according to our previous report.[3,4] The aperture area was defined by a mask of 0.09 cm^2, Monochromatic IPCE spectra were measured by a

monochromatic incident light of 1×10^{16} photons cm^{-2} in director current mode (CEP-2000BX, Bunko-Keiki). The light intensity of the solar simulator was calibrated by a standard silicon solar cell. The solar cells were firstly encapsulated in nitrogen filled glovebox by cavity glass and UV-curable glue. All the cells were encapsulated according to our previous reports for operation stability test[5].

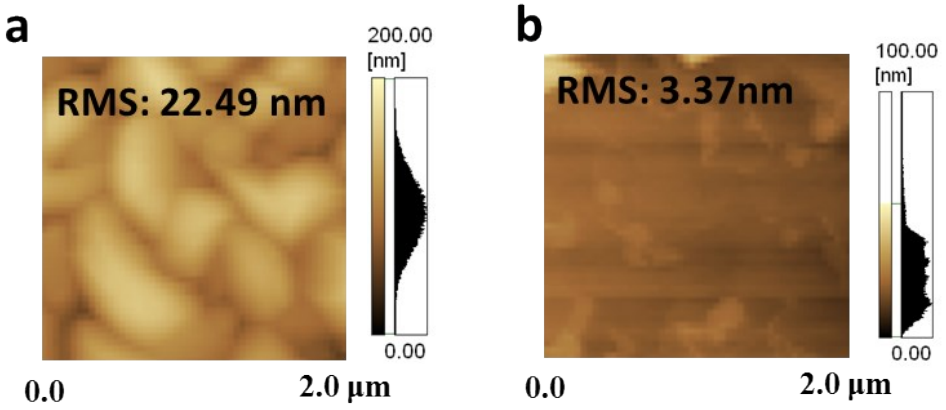


Figure S1. AFM images of (a) $FASnI_3$ and (b) TG-$FASnI_3$ films.

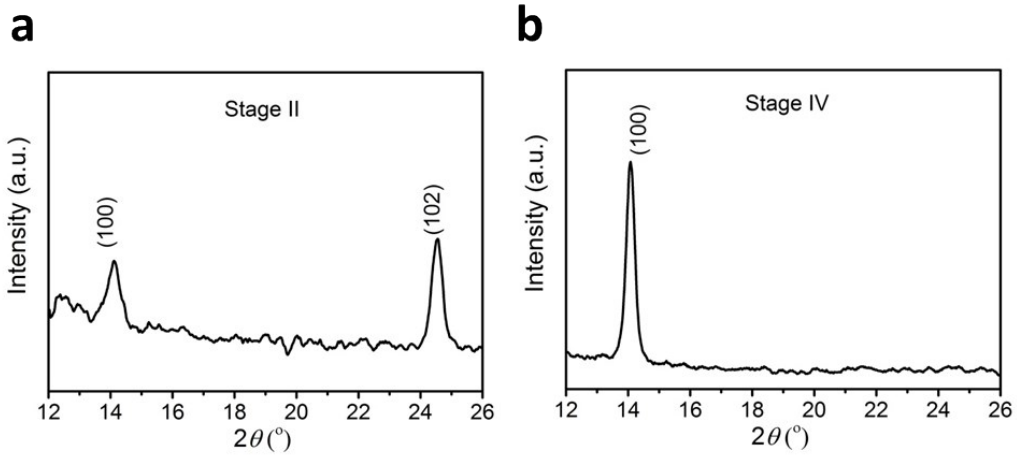


Figure S2. The GIXRD patterns for the tin perovskites in Fig. 1a stage II the treatment of PAI) and stage IV (after the treatment of PAI) with the incident angle of 0.2^{o}.

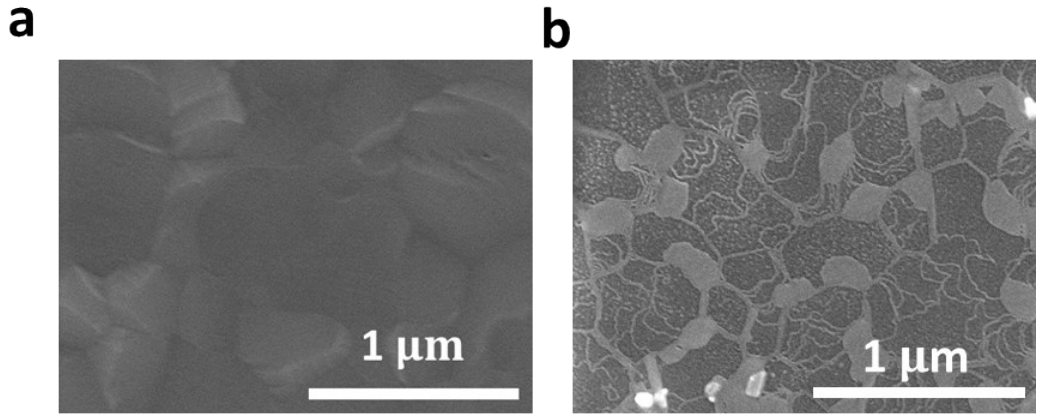


Figure S3. SEM images of (a) 1 mg mL^{-1} PAI and (b) 5mg mL^{-1} PAI treated TG-$FASnI_3$ films.

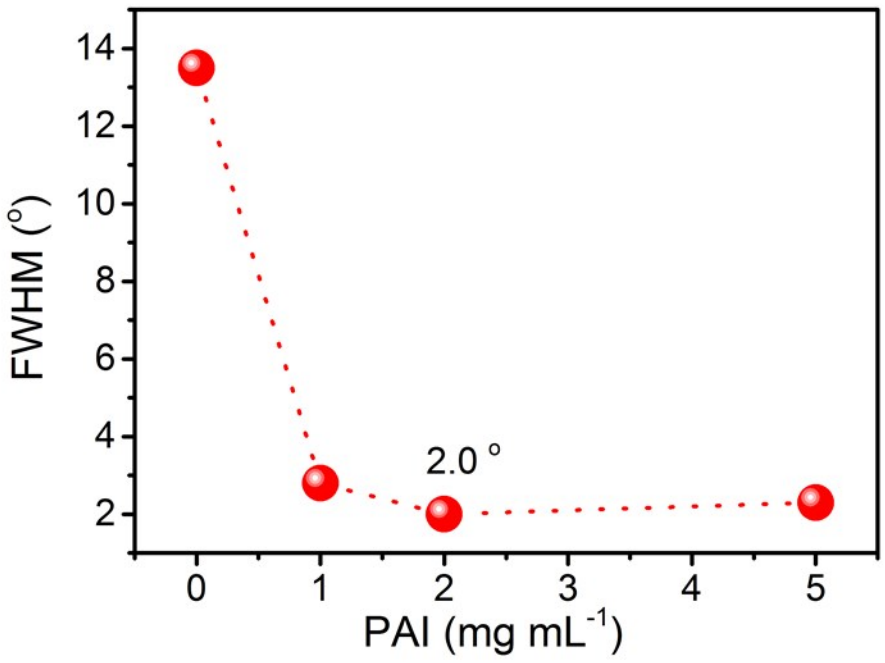


Figure S4. FWHM of the rocking curve XRD for $FASnI_3$ film and 1 mg mL^{-1}, 2 mg mL^{-1} and 5 mg mL^{-1} PAI treated TG-$FASnI_3$ films.

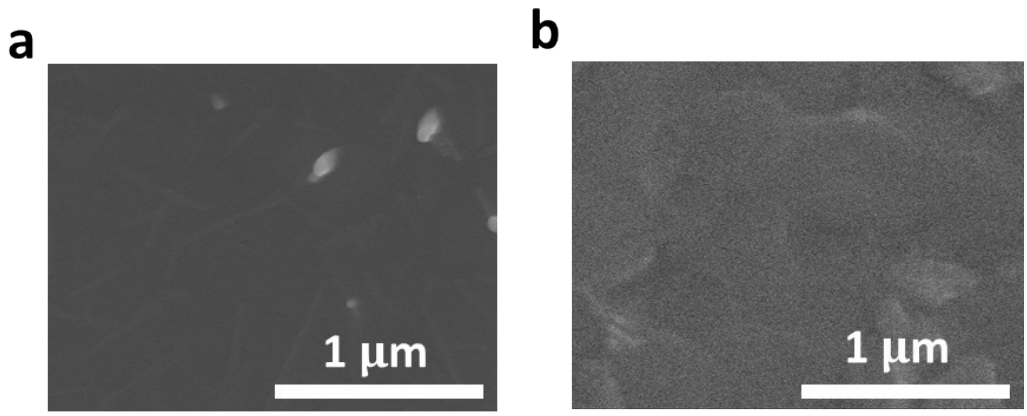


Figure S5. SEM images of 2 mg mL^{-1} PAI treated TG-$FASnI_3$ with (a) 2000 rpm/s and (b) 6000 rpm/s.

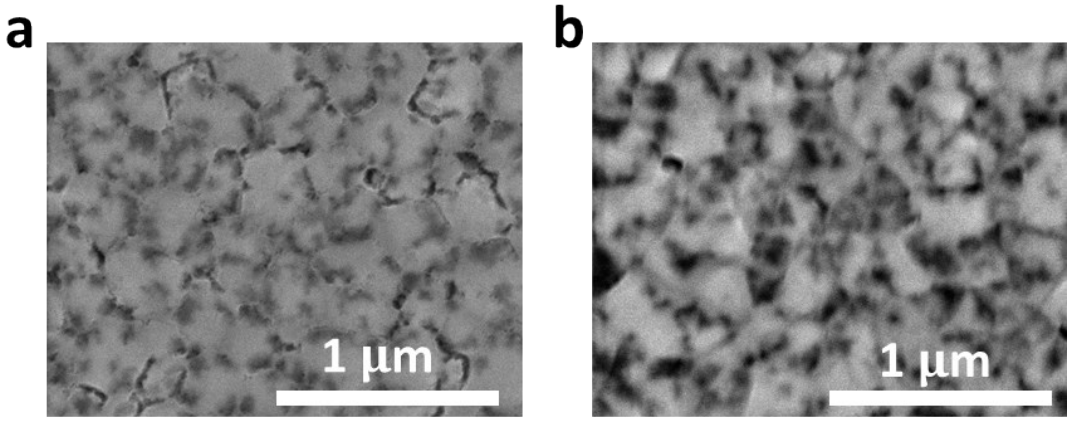


Figure S6. SEM images of 2 mg mL^{-1} PAI treated TG-$FASnI_3$ with the PAI dissolved in (a) CF/DMSO (100:0.5 v/v), (b) CF/DMSO (100:2 v/v).

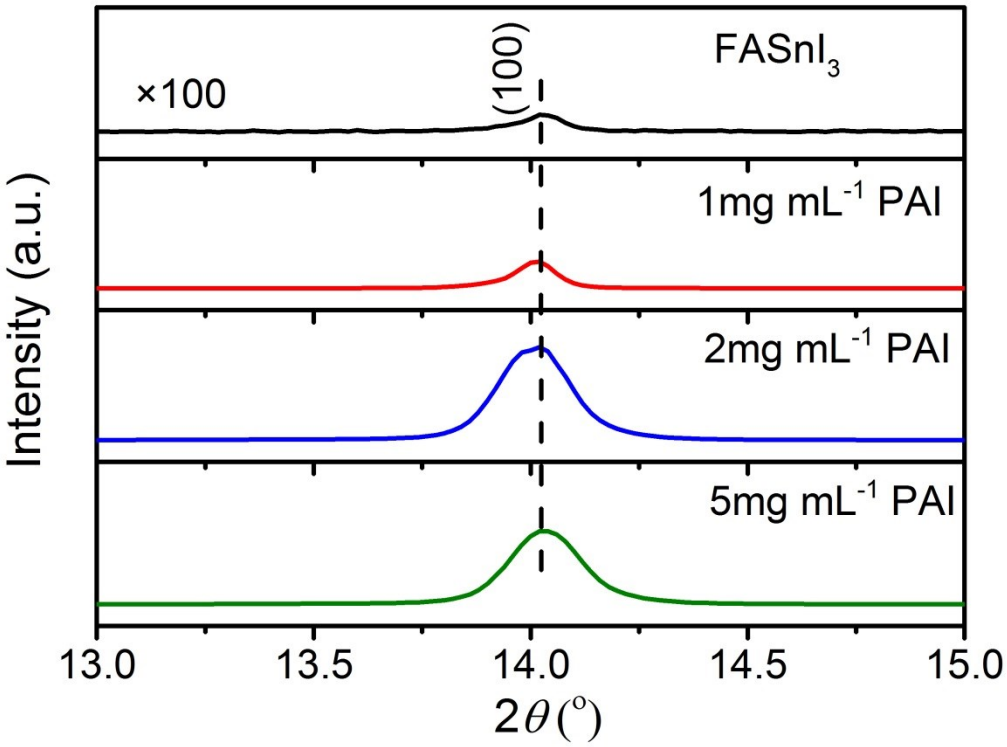


Figure S7. Amplified the signals of XRD (100) peak of $FASnI_3$, 1 mg mL^{-1}, 2 mg mL^{-1} and 5 mg mL^{-1} PAI treated TG-$FASnI_3$.

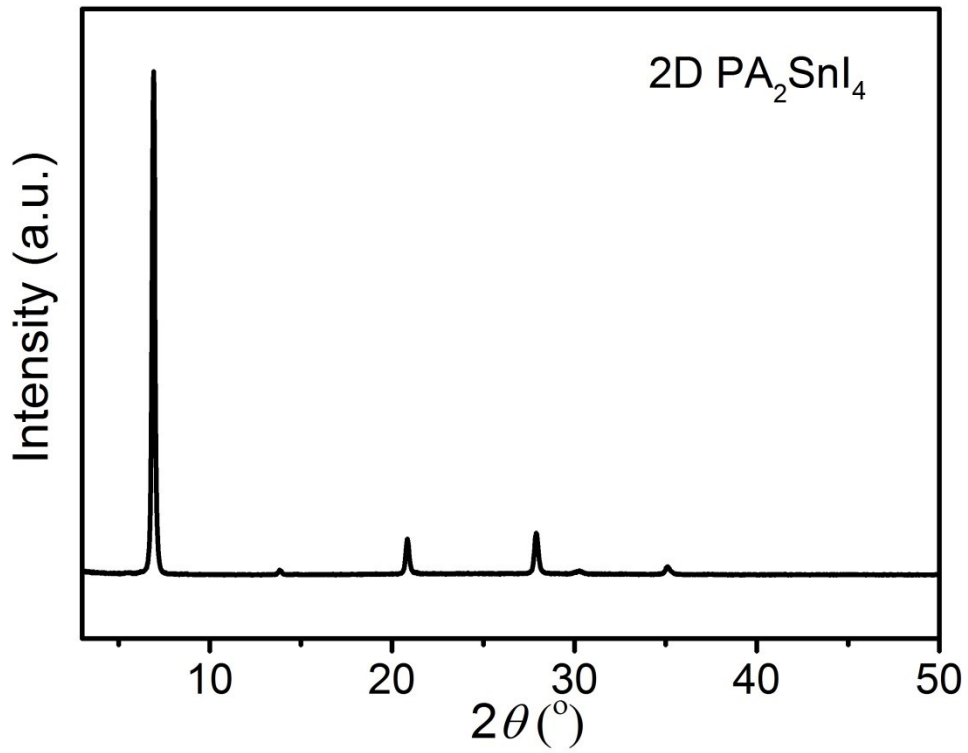


Figure S8. XRD pattern of pure 2D PA_2SnI_4.

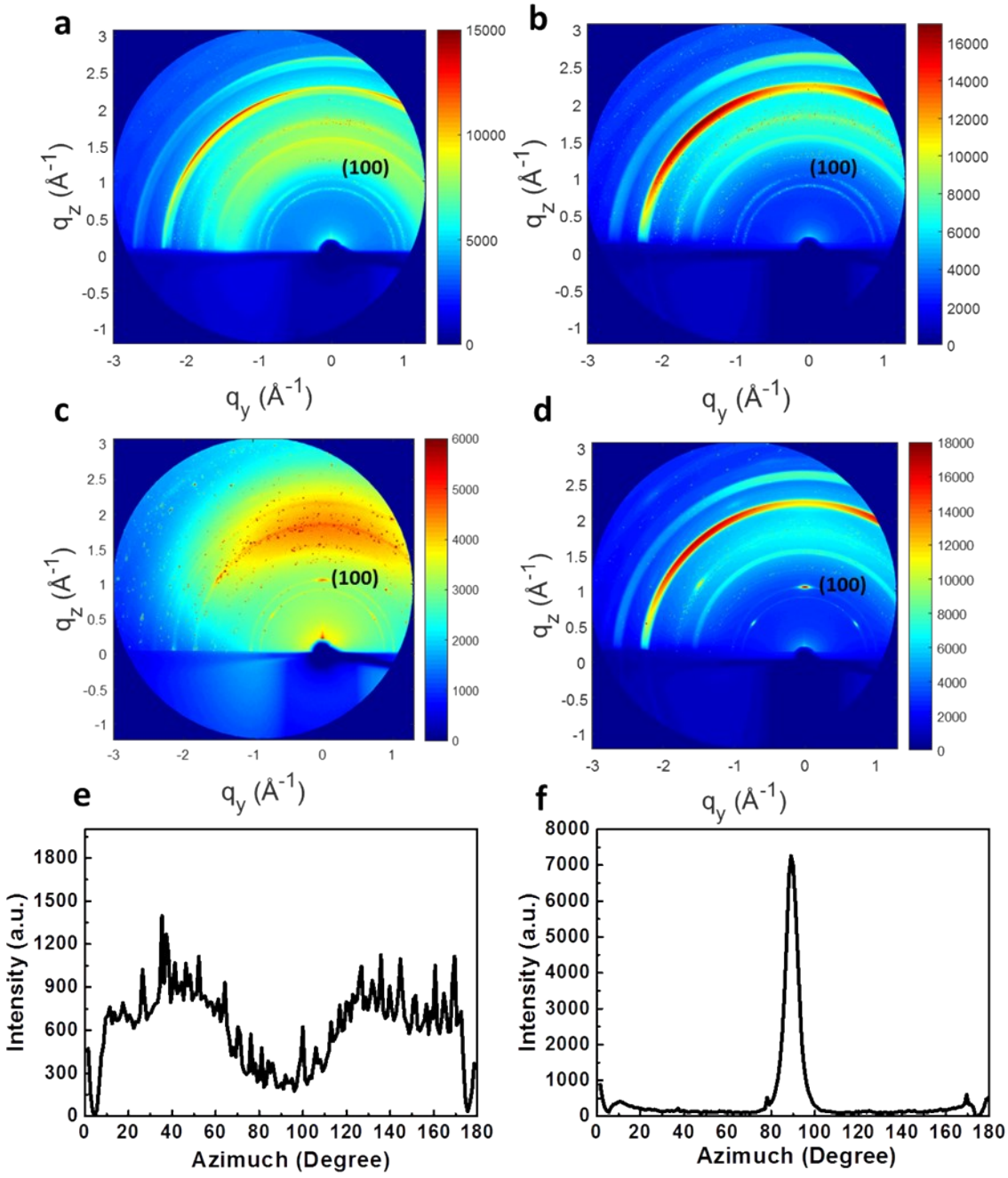


Figure S9. (a) GIWAXS images of $FASnI_3$ and (b) TG-$FASnI_3$ with incident angle of 0.1 ^{o} and (c) GIWAXS image of $FASnI_3$ and (d) TG-$FASnI_3$ with incident angle of 1 ^{o}, (e) Azimuthal degree pattern of q ≈ 1 Å^{-1} in (b) and (f) Azimuthal degree pattern of q ≈ 1 Å^{-1} in (d).

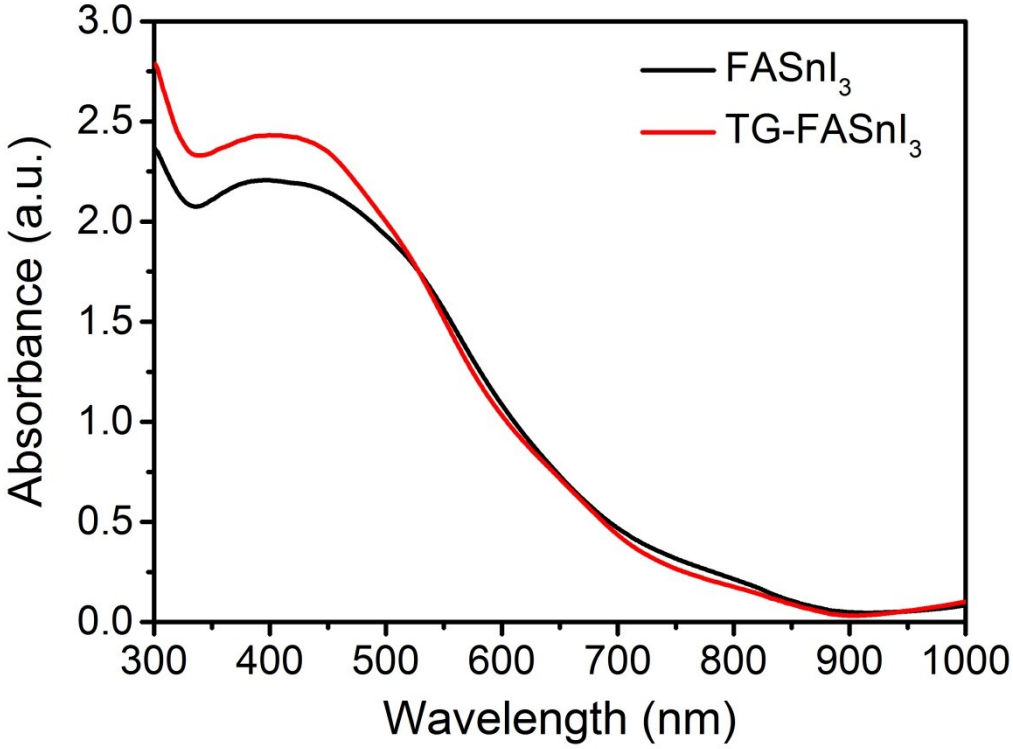
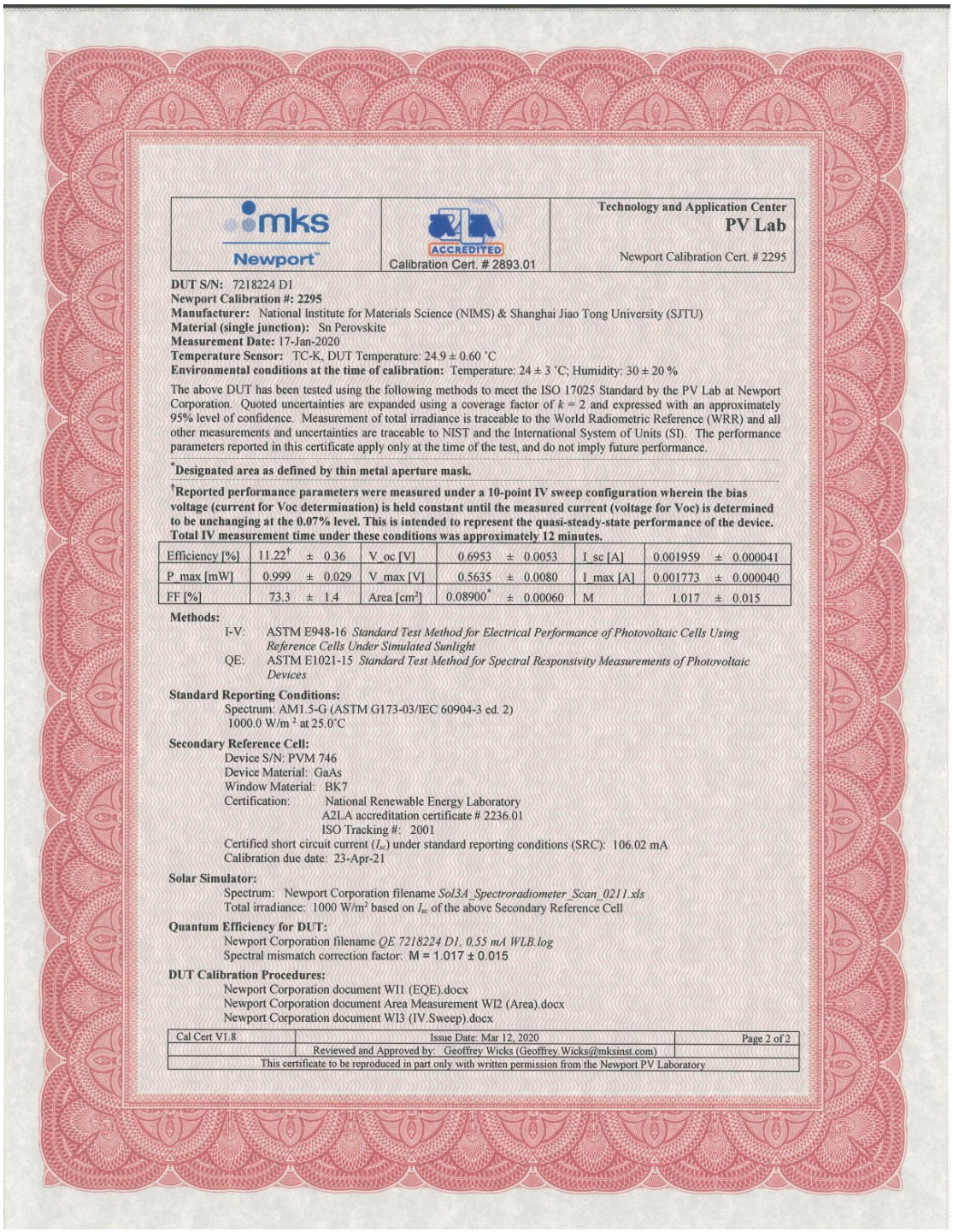


Figure S10. UV-vis spectrum of $FASnI_3$ and TG-$FASnI_3$.



mks Newport

ACCREDITED Calibration Cert. # 2893.01

Technology and Application Center
PV Lab
Newport Calibration Cert. # 2295

DUT S/N: 7218224 D1
Newport Calibration #: 2295
Manufacturer: National Institute for Materials Science (NIMS) & Shanghai Jiao Tong University (SJTU)
Material (single junction): Sn Perovskite
Measurement Date: 17-Jan-2020
Temperature Sensor: TC-K, DUT Temperature: 24.9 ± 0.60 °C
Environmental conditions at the time of calibration: Temperature: 24 ± 3 °C; Humidity: 30 ± 20 %

The above DUT has been tested using the following methods to meet the ISO 17025 Standard by the PV Lab at Newport Corporation. Quoted uncertainties are expanded using a coverage factor of $k = 2$ and expressed with an approximately 95% level of confidence. Measurement of total irradiance is traceable to the World Radiometric Reference (WRR) and all other measurements and uncertainties are traceable to NIST and the International System of Units (SI). The performance parameters reported in this certificate apply only at the time of the test, and do not imply future performance.

***Designated area as defined by thin metal aperture mask.**

†Reported performance parameters were measured under a 10-point IV sweep configuration wherein the bias voltage (current for Voc determination) is held constant until the measured current (voltage for Voc) is determined to be unchanging at the 0.07% level. This is intended to represent the quasi-steady-state performance of the device. Total IV measurement time under these conditions was approximately 12 minutes.

Efficiency [%]	11.22† ± 0.36	V_oc [V]	0.6953 ± 0.0053	I_sc [A]	0.001959 ± 0.000041
P_max [mW]	0.999 ± 0.029	V_max [V]	0.5635 ± 0.0080	I_max [A]	0.001773 ± 0.000040
FF [%]	73.3 ± 1.4	Area [cm²]	0.08900* ± 0.00060	M	1.017 ± 0.015

Methods:
I-V: ASTM E948-16 *Standard Test Method for Electrical Performance of Photovoltaic Cells Using Reference Cells Under Simulated Sunlight*
QE: ASTM E1021-15 *Standard Test Method for Spectral Responsivity Measurements of Photovoltaic Devices*

Standard Reporting Conditions:
Spectrum: AM1.5-G (ASTM G173-03/IEC 60904-3 ed. 2)
1000.0 W/m ² at 25.0°C

Secondary Reference Cell:
Device S/N: PVM 746
Device Material: GaAs
Window Material: BK7
Certification: National Renewable Energy Laboratory
A2LA accreditation certificate # 2236.01
ISO Tracking #: 2001
Certified short circuit current (I_{sc}) under standard reporting conditions (SRC): 106.02 mA
Calibration due date: 23-Apr-21

Solar Simulator:
Spectrum: Newport Corporation filename *Sol3A_Spectroradiometer_Scan_0211.xls*
Total irradiance: 1000 W/m² based on I_{sc} of the above Secondary Reference Cell

Quantum Efficiency for DUT:
Newport Corporation filename *QE 7218224 D1, 0.55 mA WLB.log*
Spectral mismatch correction factor: M = 1.017 ± 0.015

DUT Calibration Procedures:
Newport Corporation document WI1 (EQE).docx
Newport Corporation document Area Measurement WI2 (Area).docx
Newport Corporation document WI3 (IV.Sweep).docx

Figure S11. The certified results of a typical TG-FASnI$_3$ TPSC obtained from an accredited test center (Newport，USA). A quasi-steady-state PCE of 11.22% was obtained on an aperture area of 0.08900 cm^2.

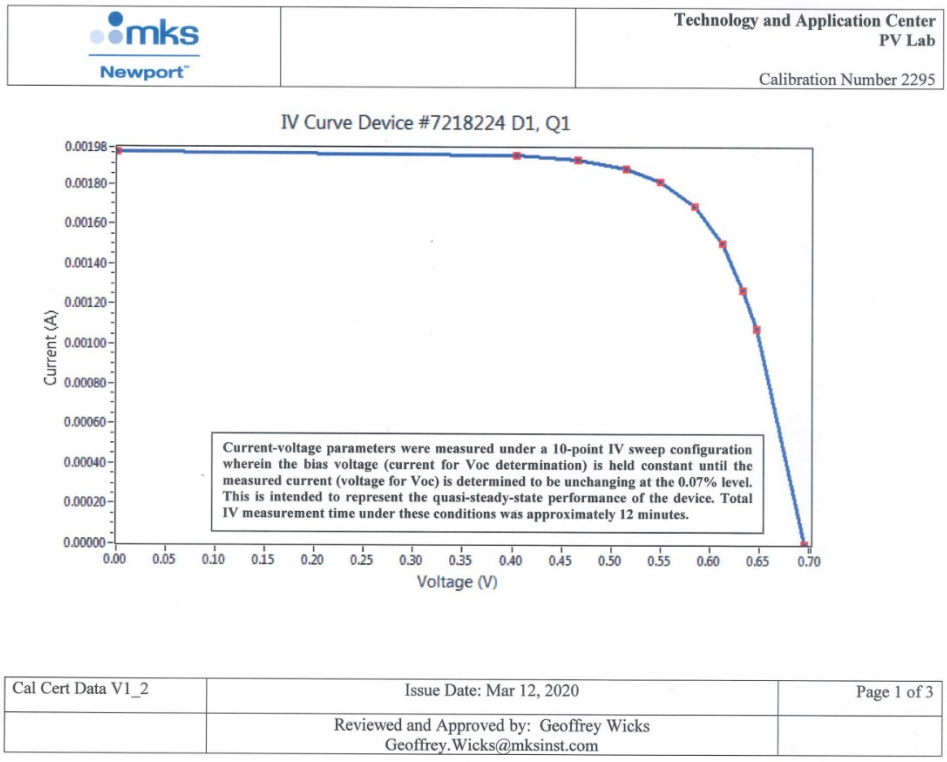


Figure S12. Certified results of *I-V* curve.

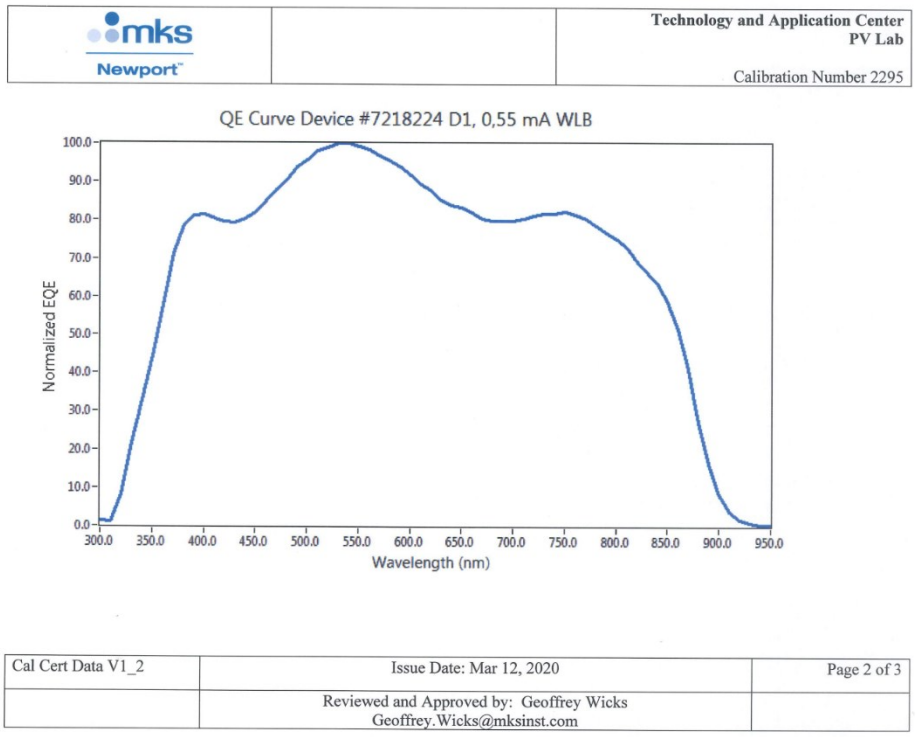


Figure S13. Certified results of normalized external quantum efficiency.

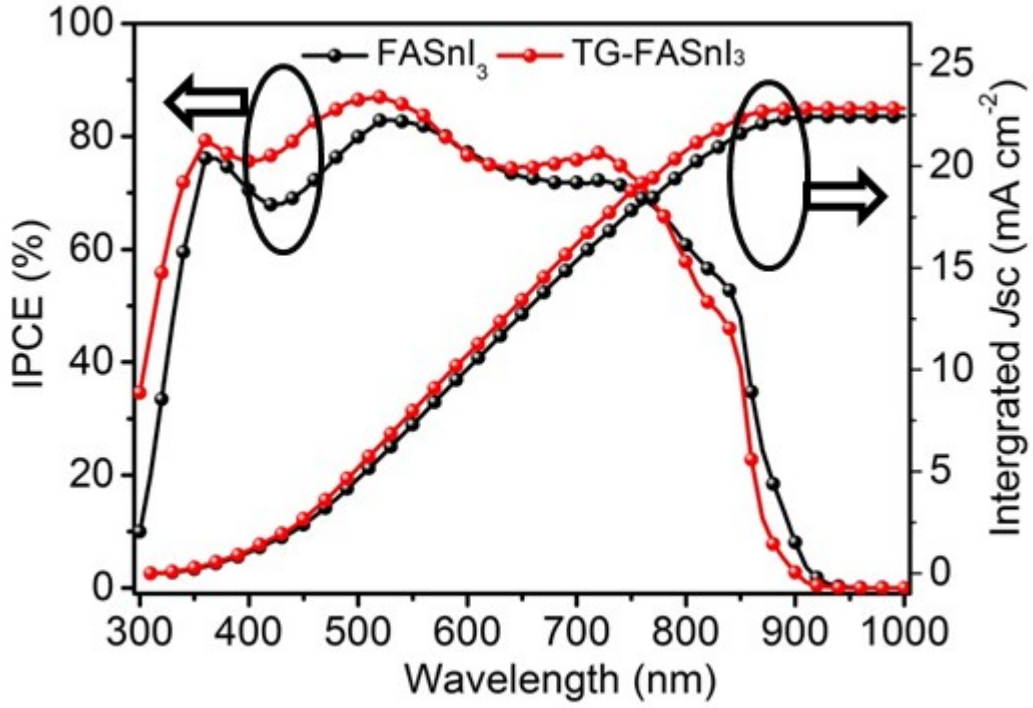


Figure S14. IPCE of the of $FASnI_3$ and TG-$FASnI_3$ devices.

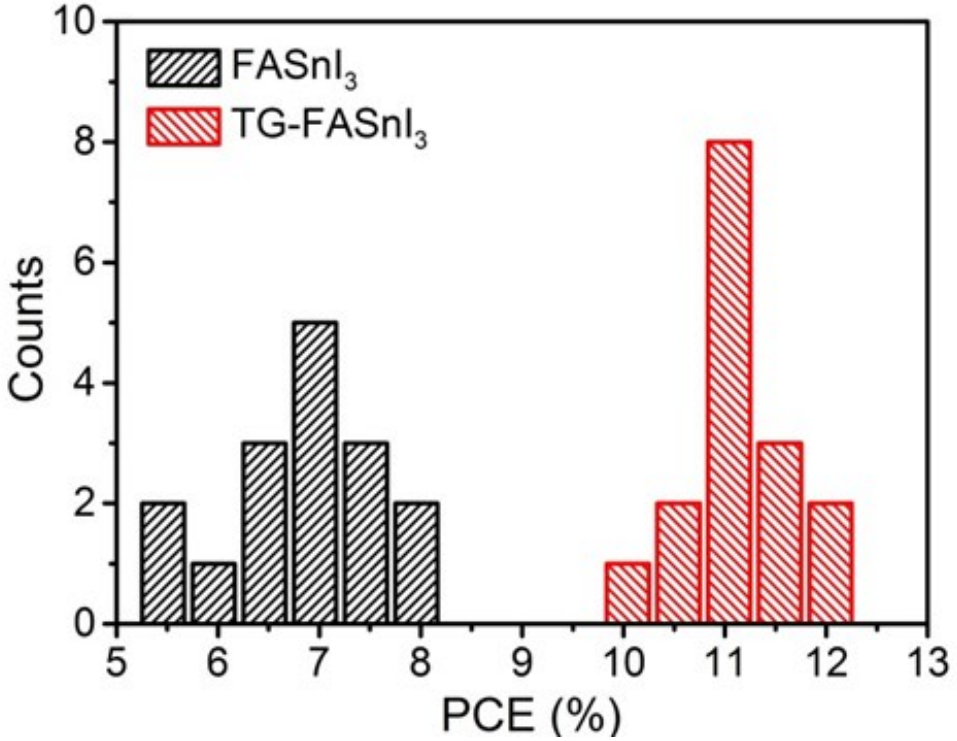


Figure S15. Histogram of efficiency statistics of sixteen devices of $FASnI_3$ and TG-$FASnI_3$ devices.

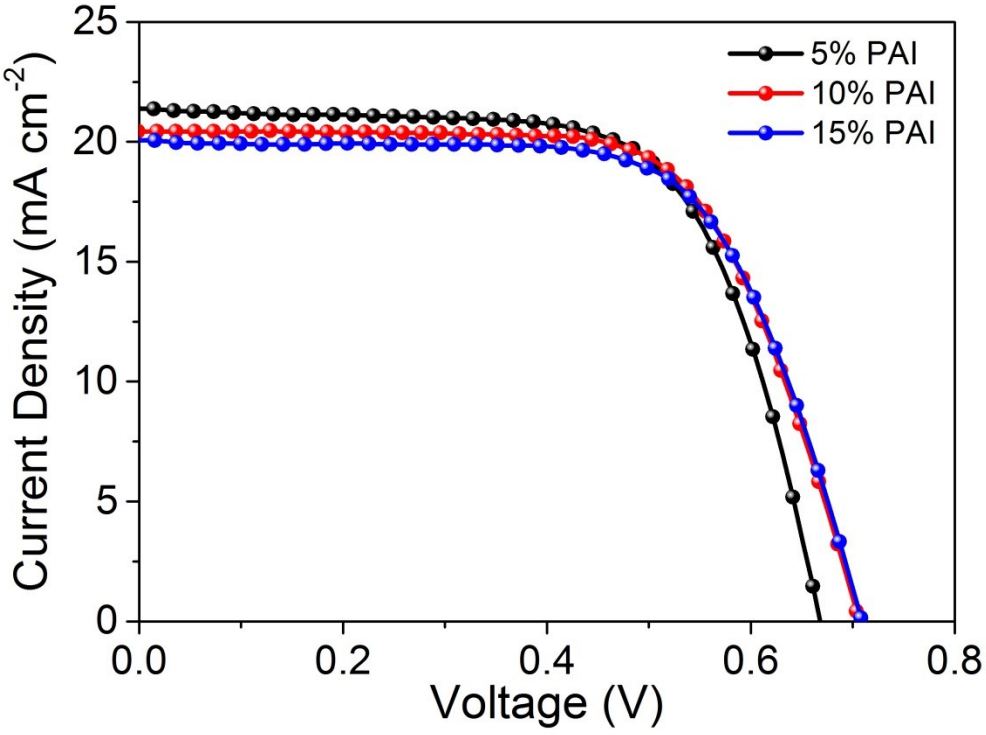


Figure S16. *I-V* curve of $FASnI_3$ with PAI as additives in precursor solution

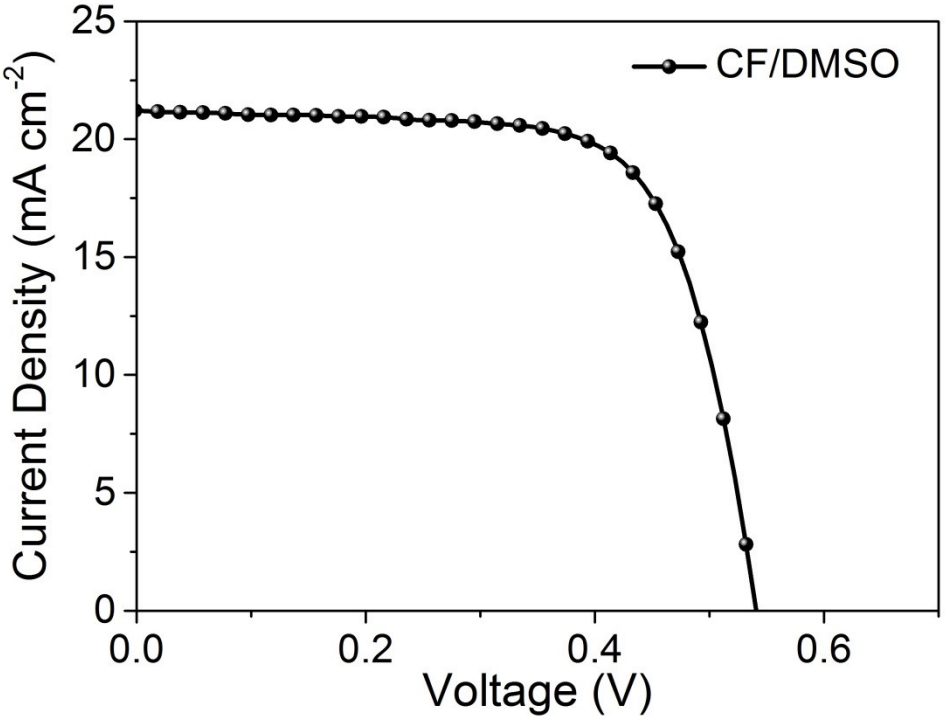


Figure S17. *I-V* curve of $FASnI_3$ with CF/DMSO pretreatment

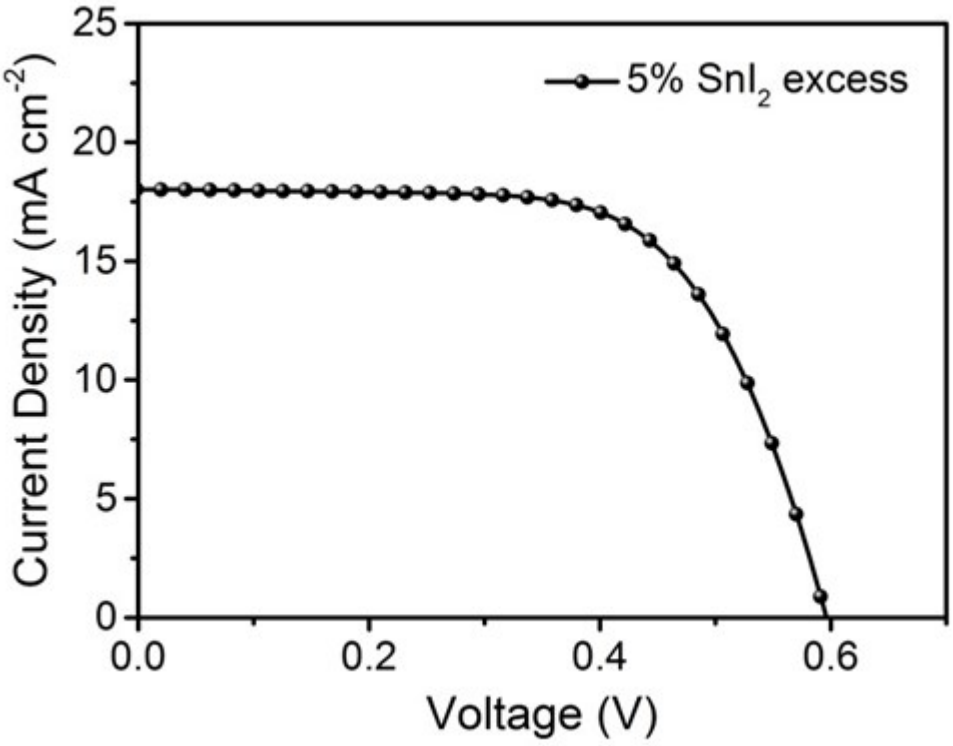


Figure S18 I-V curve of 5% SnI_2 excess $FASnI_3$ with PAI pretreatment

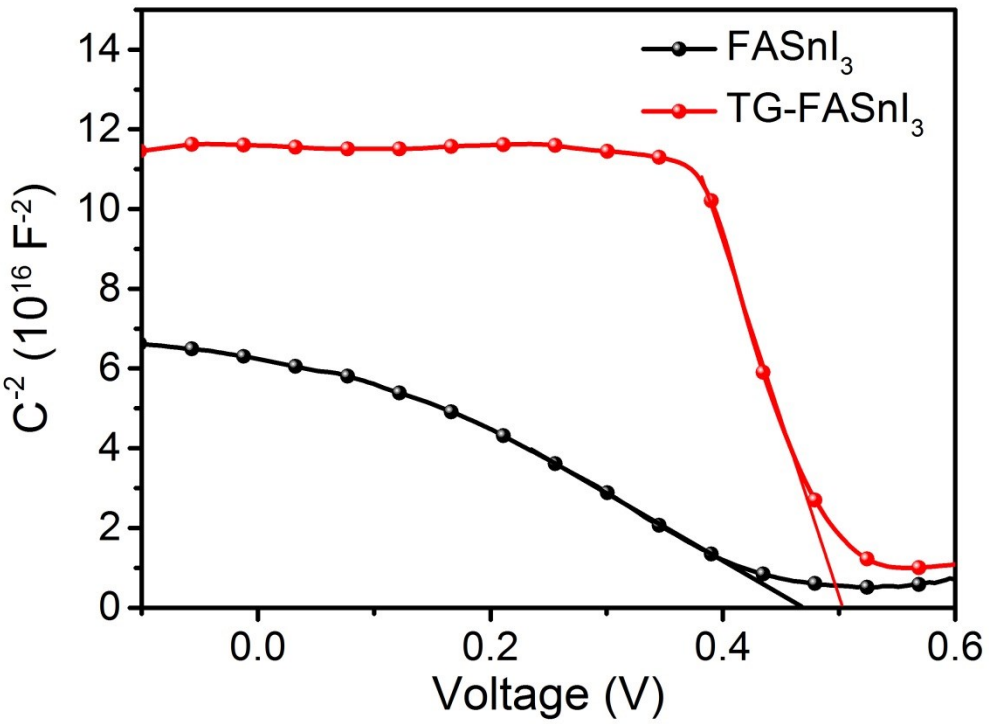


Figure S19. *C-V* curves of $FASnI_3$ and TG-$FASnI_3$ devices.

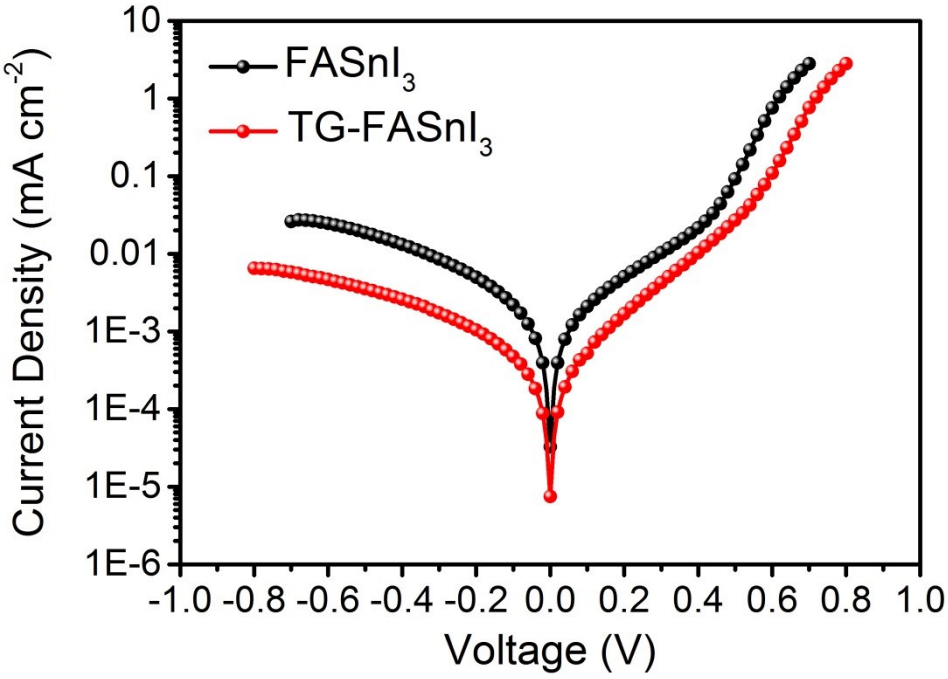


Figure S20. Dark current densities of $FASnI_3$ and TG-$FASnI_3$ devices.

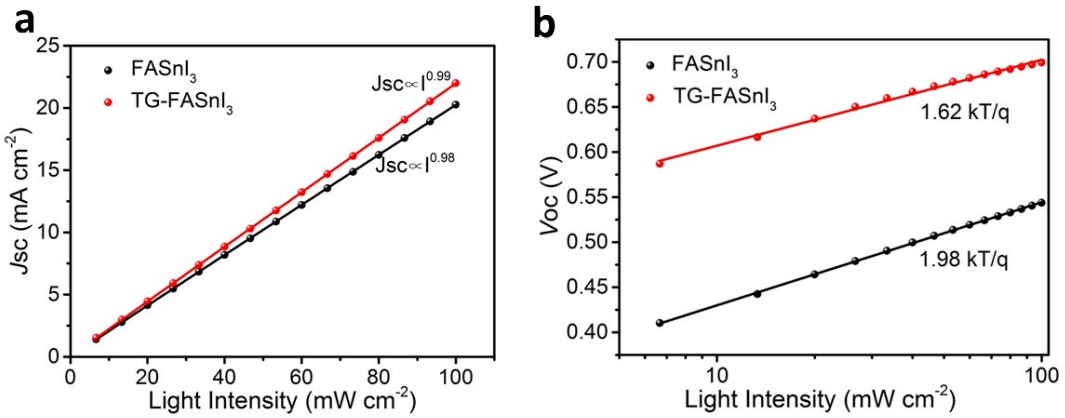


Figure S21. J_{SC} and V_{OC} versus illumination intensity of $FASnI_3$ and TG-$FASnI_3$ devices.

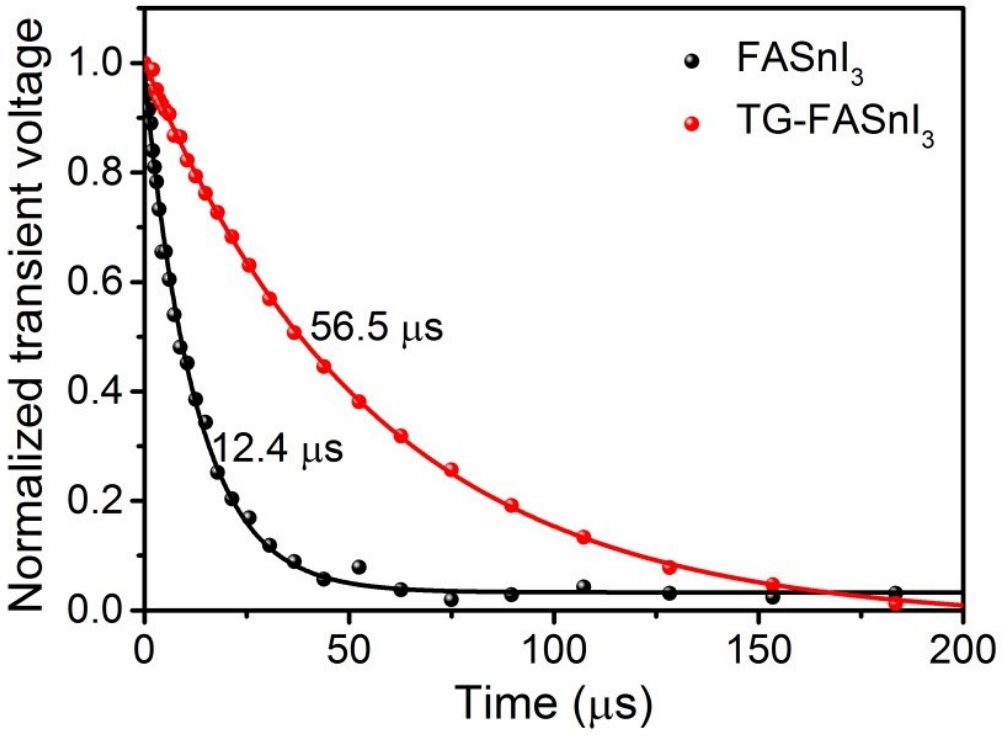


Figure S22. TPV of $FASnI_3$ and TG-$FASnI_3$ devices.

Table S1: Device parameter for typical $FASnI_3$ and TG-$FASnI_3$ based TPSCs.

Perovskite	Scan direction	J_{SC} (mA cm^{-2})	V_{OC} (V)	FF (%)	PCE (%)
$FASnI_3$	Forward	21.97	0.52	67.2	7.68
	Reverse	22.11	0.53	67.6	7.93
TG-$FASnI_3$	Forward	22.10	0.72	73.0	11.62
	Reverse	22.37	0.73	72.0	11.78

References

1. X. Liu, Y. Wang, F. Xie, X. Yang and L. Han, *ACS Energy Lett.*, 2018, **3**, 1116-1121.
2. X. He, T. Wu, X. Liu, Y. Wang, X. Meng, J. Wu, T. Noda, X. Yang, Y. Moritomo, H. Segawa and L. Han, *J. Mater. Chem. A*, 2020, **8**, 2760-2768.
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5. W. Chen, Y, Wu, Y. Yue, J. Liu, W. Zhang, X. Yang, H. Chen, E. Bi, I. Ashraful, M. Grätzel and L. Han, *Science*, 2015, **350**, 944-948.