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Supplementary Information

Enhanced Charge Collection with Passivation of the Tin Oxide Layer in Planar Perovskite Solar Cells

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Experimental methods

Device Fabrication

Chemically etched FTO glass (Nippon Sheet Glass) substrates were sequentially cleaned with detergent solution, water and ethanol. A PTO layer was prepared by spin-coating a precursor solution of $SnCl_4$ (Acros) dissolved in water. The solution was spin-coated on the UV-ozone treated FTO substrate at 5,000 rpm for 10 s. 0.1 M and 0. 5 M $SnCl_4$ aqueous solutions were used to get ~20 nm and 100 nm thick LTO layers. Then the PTO film was transferred onto a hotplate and dried at 100 ^oC for 1 min, and post-annealed at 180 ^oC for 1 h and cooled down before deposition of perovskite. ALD SnO_2 films were prepared with tetrakis-dimethyl-amine tin as a Sn precursor and ozone as an oxygen reactant.[22] The deposition was carried out at 100 ^oC, and films are post-annealed with the same condition. A compact TiO_2 layer was coated on the cleaned FTO substrate by spray pyrolysis deposition at 450 °C with a precursor solution prepared by diluting titanium diisopropoxide (Sigma-Aldrich) in ethanol. The $(FAPbI_3)_{0.85}(MAPbBr_3)_{0.15}$ precursor solution was prepared by mixing FAI (1.05 M, Dyesol), PbI_2 (1.10 M, TCI), MABr (0.185 M, Dyesol) and $PbBr_2$ (0.185 M, TCI) in a mixed solvent of DMF:DMSO = 4:1 (volume ratio). The solution was spin-coated at 1,000 rpm for 10 s and, continuously at 5,000 rpm for 30 s. During the second step, 100 μL of chlorobenzene was poured on the film at 15 s. Films are post-annealed at 100 °C for 60 min. The HTM solution was prepared by dissolving 10 mg of PTAA (Emindex) with additives in 1 mL of toluene. As additives, 7.5 μL of Li-bis(trifluoromethanesulphonyl) imide (Aldrich) from the stock solution (170 mg in 1 mL of acetonitrile), and 4 μL of 4-tert-butylpyridine were added. The HTM layer was formed by spin-coating the solution at 3,000 rpm for 30 s, and followed by the deposition of the 70 nm thick Au electrode by a thermal evaporation. All the preparative work to deposit perovskite and PTAA were done inside the glovebox under N_2 atmosphere.

Film and Device Characterization

X-ray diffraction (XRD) analysis was carried out using Bruker D8 Advance diffractometer in an angle range of $2\theta = 10°$ to $30°$, or $20°$ to $60°$. Scanning electron microscopy (SEM) measurements were carried out on a high-resolution ZEISS Merlin at 5 kV, and images were acquired with the in-lens detector. Transmission electron microscopy imaging and energy-dispersive X-ray (EDX) analysis were performed on a FEI Tecnai Osiris at 200 kV. This microscope is equipped with a high brightness X-FEG gun and silicon drift Super-X EDX detectors and Bruker Esprit acquisition software. Samples for TEM imaging were prepared via depositing the LTO film on a TEM grid with holey carbon support film. The TEM grids were then heat-treated at 100 °C and 180 °C. Absorbance and reflectance were measured with an integrating sphere using the UV/Vis/NIR spectroscopy (PerkinElmer Lambda 950S). The photoluminescence emission was measured with the Fluorescence spectrometer (PerkinElmer LS 55). The solar cell measurement was done using Oriel solar simulator (450W Xenon, AAA class). The light intensity was calibrated with a Si reference cell equipped with an IR-cutoff filter (KG3, Newport), and it was recorded prior to measurement. Current-voltage characteristics of the cells were obtained by applying an external voltage bias while measuring the current response with a digital source meter (Keithley 2400). The voltage scan rate was 50 mV s^{-1} and no device preconditioning such as light soaking or forward voltage bias applied for long time, was applied before starting the measurement. The cells were masked with the active area of 0.16 cm^2 to fix the active area and reduce the influence of the scattered light. EQE was measured with IQE200B (Oriel) without bias light.

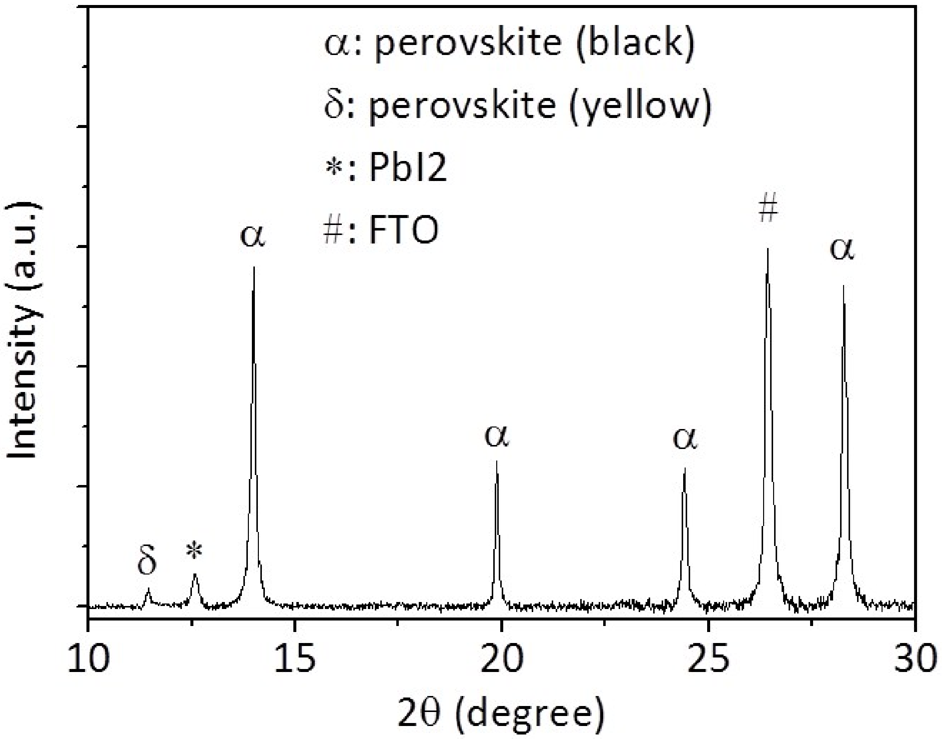

Fig. S1 X-ray diffraction (XRD) pattern of the 5 mol.% PbI_2-rich $(FAPbI_3)_{0.85}(MAPbBr_3)_{0.15}$ perovskite film.

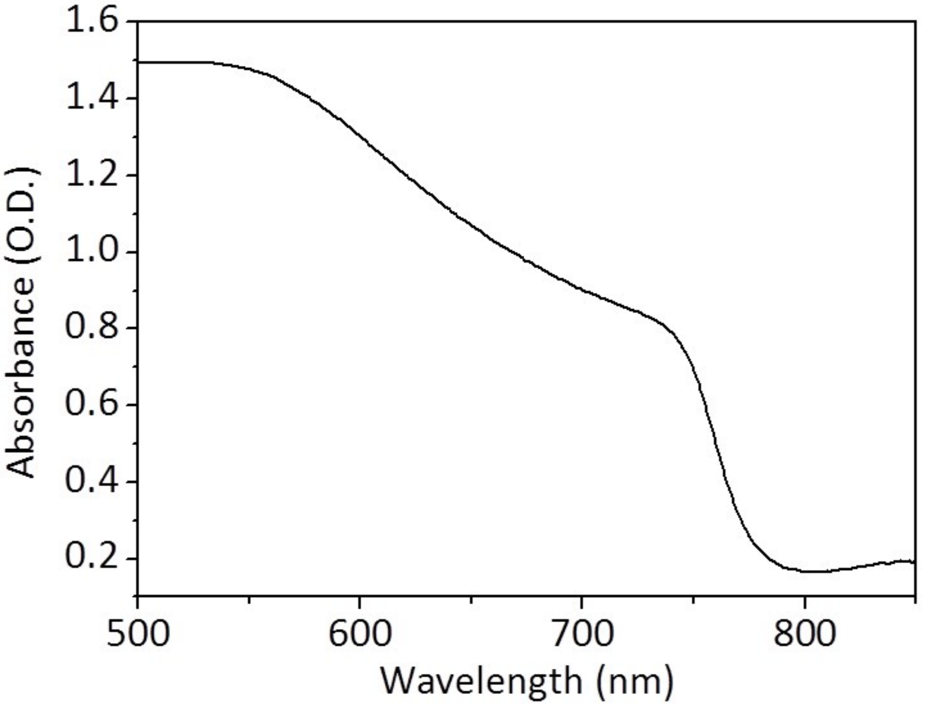

Fig. S2 Absorbance of $(FAPbI_3)_{0.85}(MAPbBr_3)_{0.15}$ perovskite on the FTO/c-TiO_2/PTO substrate.

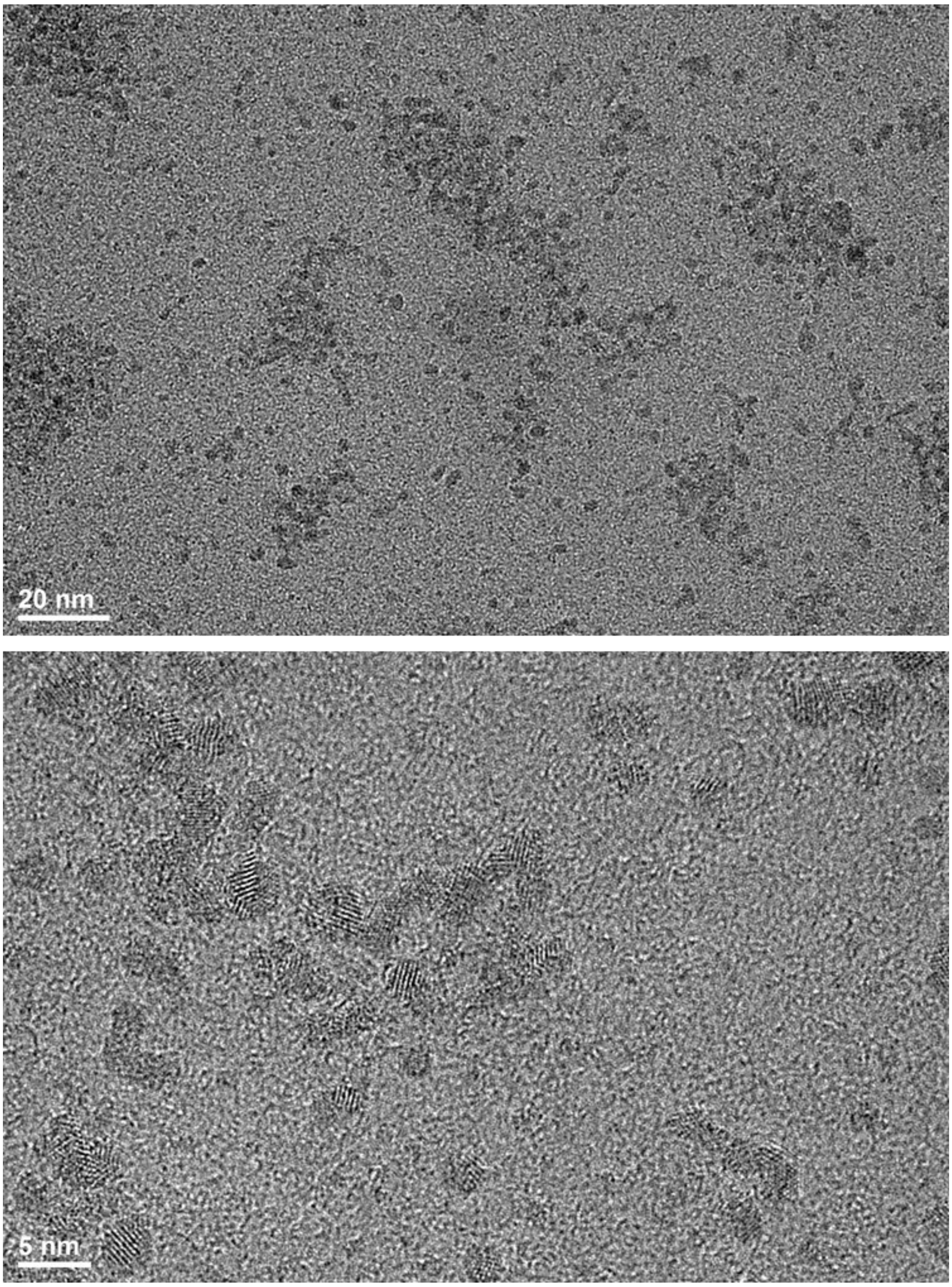

Fig. S3 TEM images of the PTO film annealed at 100 ^{o}C for 1 h.

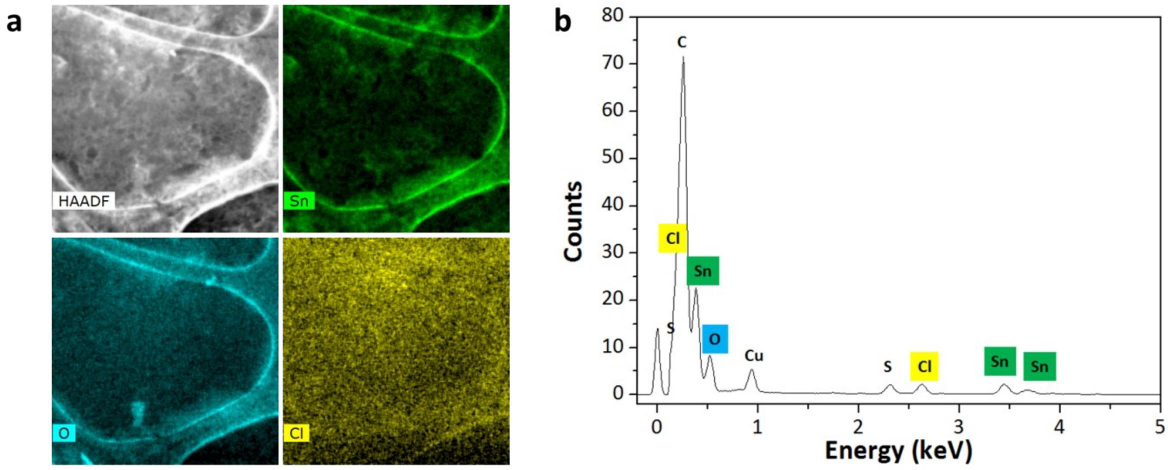

Fig. S4 Energy dispersive X-ray (EDX) analysis. a) High-angle annular dark-field (HAADF) scanning TEM image and corresponding elemental maps of Sn, O and Cl of the PTO film deposited on a holey carbon TEM grid. b) EDX spectrum from the region marked in (a). Films are annealed at 180 ^{o}C for 1 h.

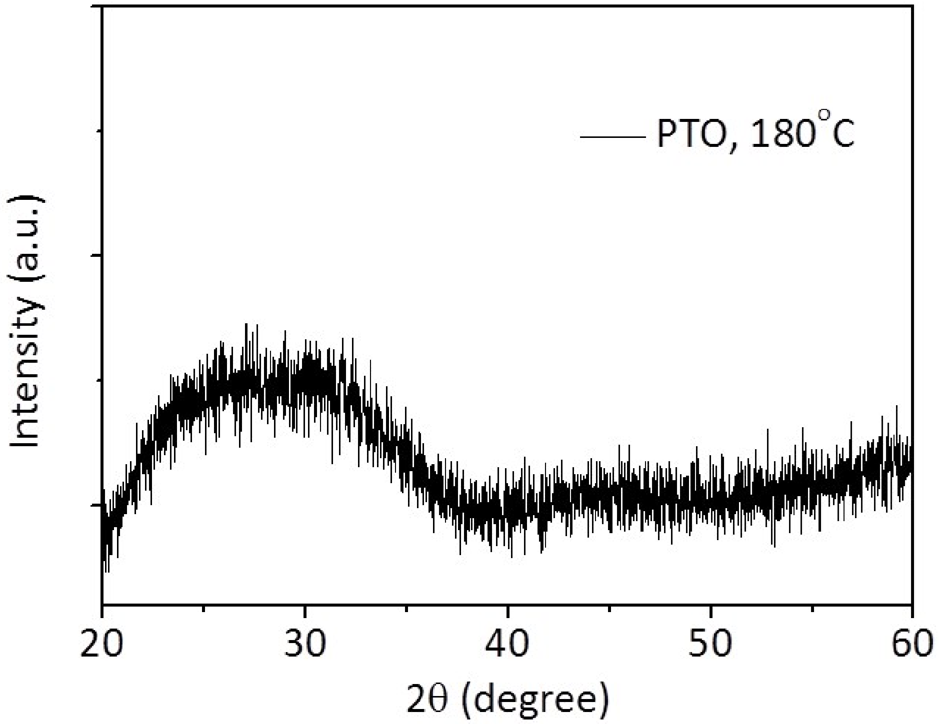

Fig. S5 XRD pattern of the PTO film annealed at 180 °C for 1 h. A ~150 nm thick film was used for the analysis.

Table S1 Summarized PCEs of the perovskite solar cells. Average values are obtained from 5 cells at the same batch, with backward scanning under 1 sunlight illumination.

ETL	J_{sc} [mA cm^{-2}]	V_{oc} [V]	FF	PCE [%]
TiO2 (20 nm)	15.59 ± 2.02	1.10 ± 0.02	0.65 ± 0.11	11.13 ± 1.85
PTO (100 nm)	21.48 ± 0.18	1.10 ± 0.03	0.69 ± 0.04	16.36 ± 1.35
TiO2/PTO (5/20 nm)	22.63 ± 0.16	1.13 ± 0.01	0.76 ± 0.01	19.44 ± 0.36

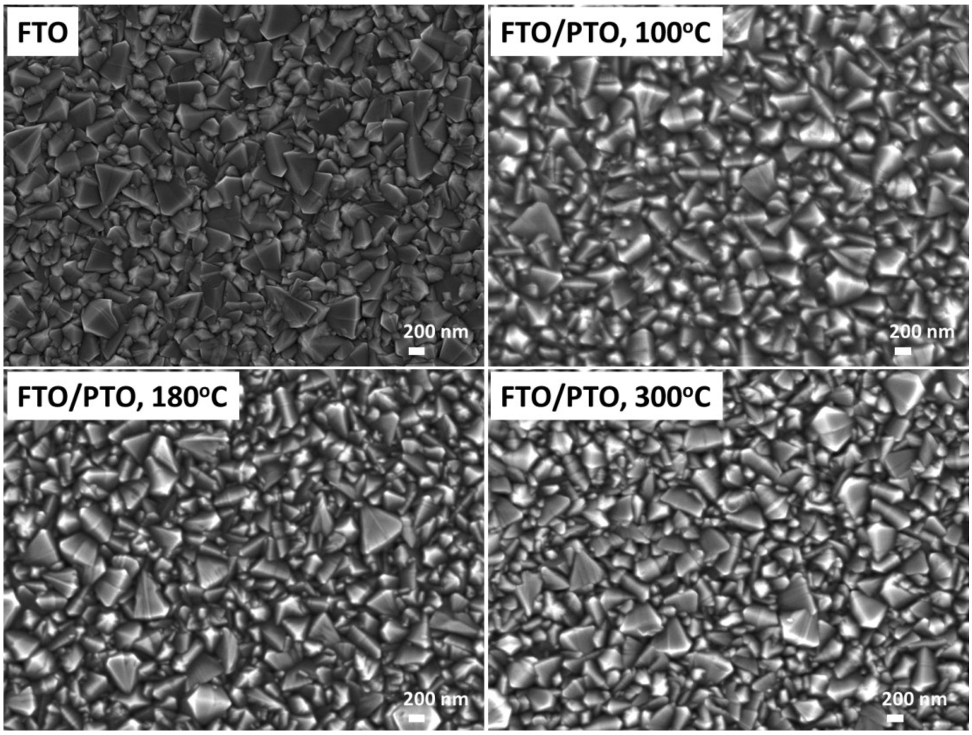

Fig. S6 SEM top-view images of FTO and PTO films. Films heat-treated at 100, 180 and 300 °C for 1 h show similar morphologies.

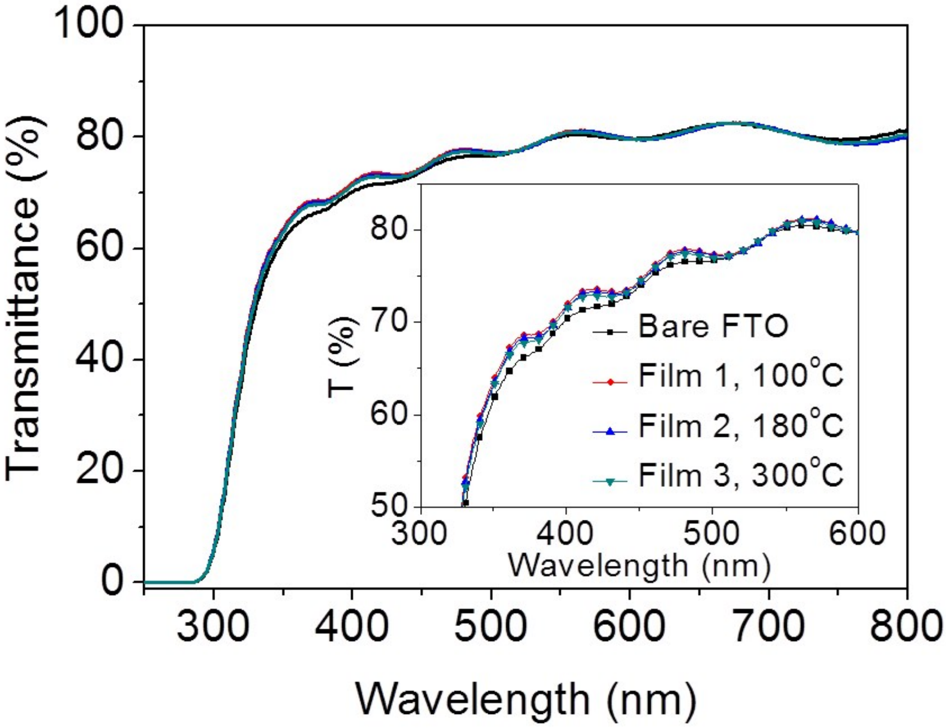

Fig. S7 Transmittance of PTO films as a function of post-annealing temperature.

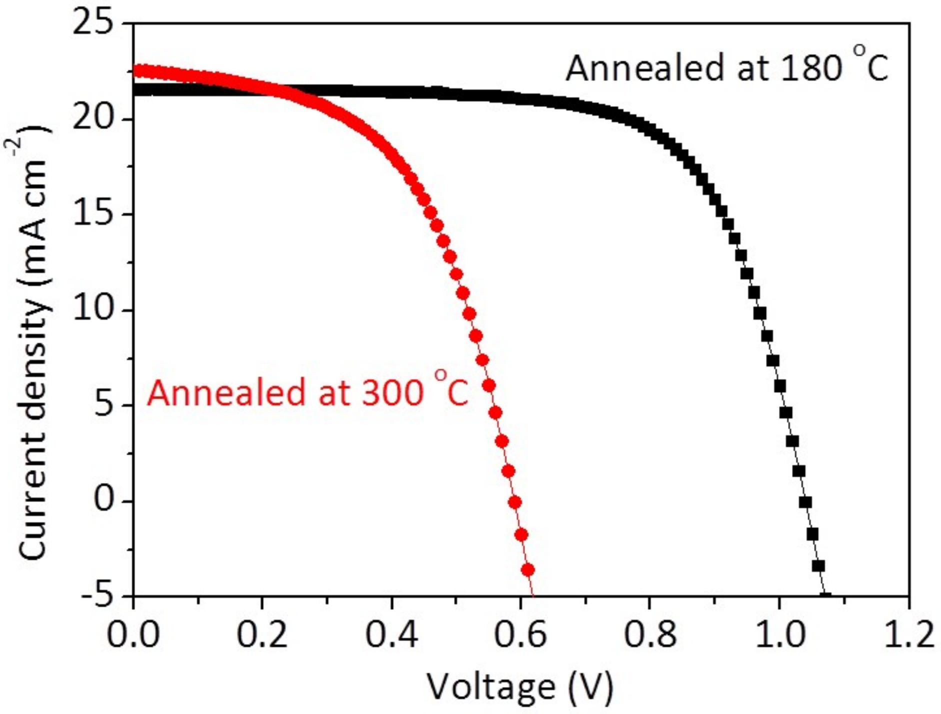

Fig. S8 *J*–*V* curves of ALD SnO_2 based perovskite solar cells. As-deposited films are annealed at 180 °C and 300 °C for 1 h, respectively.

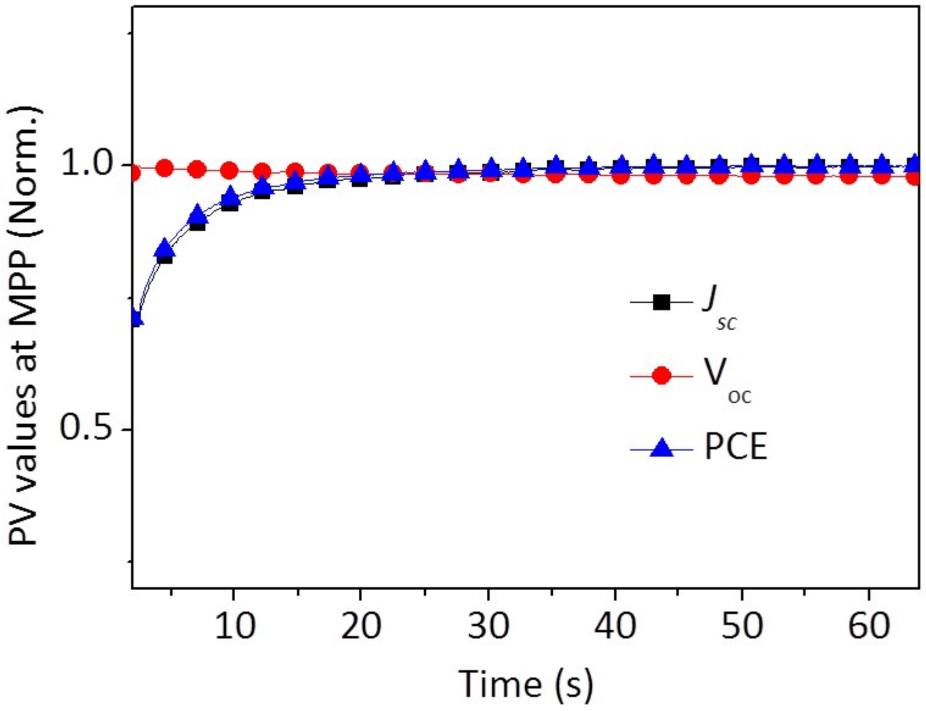

Fig. S9 Stabilized power output. The values are obtained at a maximum power point (MPP) tracking condition under 1 sunlight illumination.