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Supporting information

Pinhole-Free Perovskite Films for Efficient Solar Modules

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Device Fabrication

The glass substrates with patterned ITO electrodes were purchased from Colorado Concept Coatings. The ITO coated glass was cleaned with ultrasonic baths of detergent, deionized water, acetone, and iso-propanol each for 10 minutes, and then transferred into an Angstrom Engineering evaporation system, equipped with an electron beam source. The TiO_2 pellets purchased from Prof. Feierabend GmbH were evaporated at a rate of 1 Å/s onto ITO substrates, using a partial O_2 pressure of 1.7×10^{-4} Torr during the deposition, to maintain the stoichiometry of the film. To make the perovskite precursor solution, $Pb(CH_3CO_2)_2{\cdot}3H_2O$, $PbCl_2$, and CH_3NH_3I were dissolved in N,N-dimethylformamide (DMF). The concentration of lead source, $Pb(CH_3CO_2)_2{\cdot}3H_2O$ together with $PbCl_2$, was 0.8 M, while the concentration of CH_3NH_3I was 2.4 M (Pb : CH_3NH_3I = 1:3). The optimal $PbCl_2$ molar fraction in the lead source was around 20%-30%. The precursor solution was stirred at room temperature for 10 min, and then spin coated at 3000 rpm for 60 s onto the TiO_2 layer. The obtained films were annealed on a hot plate at 130 °C for 10 min to form a perovskite crystal structure. After that, 80 mg/ml Spiro-OMeTAD solution doped with 17.5 μl lithium bis(trifluoromethanesulfonyl) imide (520 mg/ml in acetonitrile) and 28.5 μl 4-tert-butylpyridine was spin-coated onto the perovskite films. All of the spin-coating processes mentioned above were performed in an N_2 filled glove box. The perovskite films coated with Spiro-OMeTAD were then exposed to air overnight for Spiro-OMeTAD oxygen doping. The small area devices were completed by depositing an 80 nm Au layer onto the Spiro-OMeTAD through shadow masks, defining an active area of 0.13 cm^2. The P1 patterning of the module was done by laser before TiO_2 evaporation. The P2 and P3 patterning (Fig. 1b) was performed within nitrogen atmosphere by using a knife mounted to a computer controlled xyz-stage. The P2 patterning shown was carried out before the deposition of Au. The pressure was set in order to fully remove the TiO_2 layer ensuring a low contact resistance at the interconnect. Then, after Au deposition, the P3 patterning finishes the device fabrication.

Characterizations

The X-ray diffraction pattern was recorded on a PANalyticalX'Pert Pro Materials Research Diffractometer using Cu Kα radiation. The top-view SEM images were obtained from FEI Nova 200 scanning electron microscope. Photovoltaic characteristics of the devices were measured under a nitrogen atmosphere using a Keithley 2602A Source-Measure Unit and an Abet solar simulator with 100 mW/cm^2 simulated AM1.5G illumination, calibrated with an KG5 band pass equipped ISE Fraunhofer certified Si photodiode. The devices were measured from forward to reverse bias, or from reverse bias to forward bias, with a scan speed of 1 V/s. The EQE was measured with a photospectrometer setup (Bentham PVE300) by illuminating the solar cell with a modulated monochromatic light (Xe and quartz halogen lamps). It is carried out with a chop frequency of 377 Hz but without light bias. The spectral resolution is 5 nm. The change of MPP over illumination time was recorded using a MPP tracker. To do this, the initial maximum power point was first identified in the initial *J-V* scan. This value was then used as the initial set point in the MPP tracker. Then, the MPP tracker compared the PCEs near the MPP every 5 s (± 0.01 V) to find the new MPP.

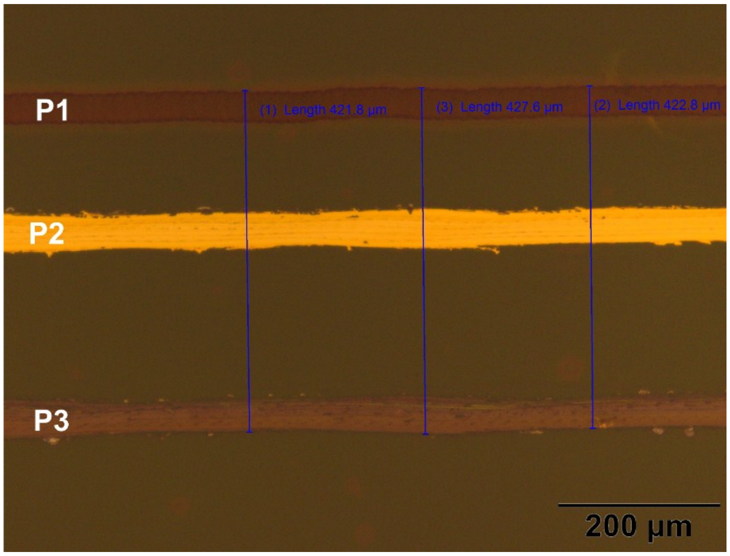

Fig. S1 Digital microscope image of the three patterning lines, resulting in an average loss spacing of around 0.43 mm between adjacent active areas of 4.57 mm width.

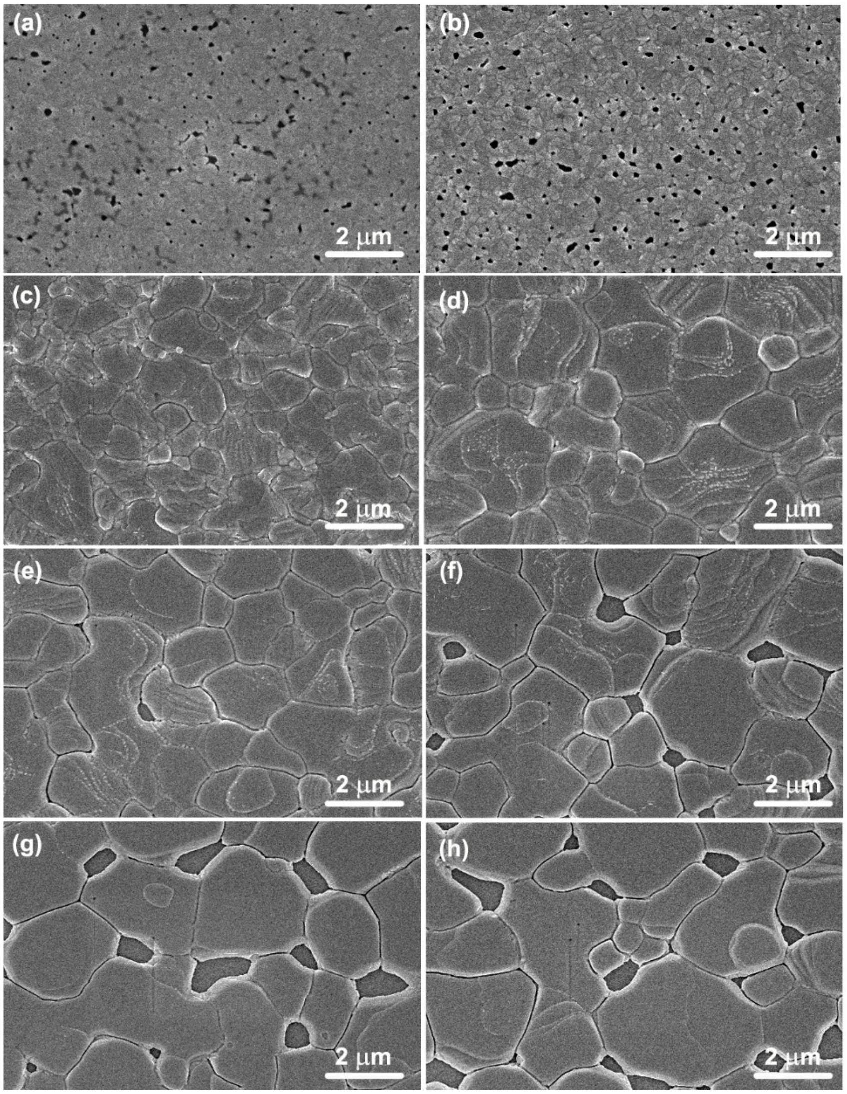

Fig. S2 Top-view SEM images of perovskite films made with different $PbCl_2$ molar fractions in the mixed lead source: (a) 0%; (b) 10%; (c) 20%; (d) 30%; (e) 40%; (f) 50%; (g) 60%; (h) 70%.

Table S1 The detailed photovoltaic parameters of the devices shown in Fig. 4d: *J-V* scanned from forward bias to reversed bias.

$PbCl_2$ fraction (%)	J_{sc} (mA/cm^2)	V_{oc} (V)	FF (%)	PCE (%)
0	21.0	0.91	68	13.2
20	21.4	0.97	79	16.7
40	20.8	0.92	78	15.1
60	20.5	0.88	74	13.5