Electronic Supplementary Information

A One-Step Low Temperature Processing Route for Organolead Halide Perovskite Solar Cells

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This paper describes a unique low temperature processing technique for lead halide perovskite solar cells incorporating an Al_2O_3 scaffold. Instead of applying a perovskite precursor solution to a mesoporous alumina scaffold (pre-sintered at 550 °C). We show that it is possible to add alumina nanoparticles directly to the perovskite precursor solution and to co-deposit a single Al_2O_3/perovskite layer followed by a low-temperature heating step (<110 °C). This ESI includes details of the methods used and data not shown in the communication submission.

Experimental

$CH_3NH_3PbI_2Cl$ perovskite synthesis

Methylamine (CH_3NH_2) solution (33 wt% in absolute ethanol, Sigma Aldrich) was reacted with hydroiodic acid (57 wt % in water, Sigma Aldrich) with excess methylamine under nitrogen atmosphere in ethanol at room temperature for 2 hours. Typical quantities were 50 mL methylamine, 20 mL hydroiodic acid and 200 mL ethanol. Crystallization of methylammonium iodide (CH_3NH_3I) was achieved using a rotary evaporator. A white crystalline solid was obtained and dried in a vacuum oven overnight.

Methylammonium iodide (CH_3NH_3I) and lead (II) chloride ($PbCl_2$, Sigma Aldrich) were dissolved in anhydrous N,N-Dimethylformamide (DMF, Sigma Aldrich) at a 3:1 molar ratio of CH_3NH_3I to $PbCl_2$, to produce a mixed halide perovskite precursor solution at 40 % total weight concentration.

Alumina nanoparticle suspension in DMF

50 ml of an alumina nanoparticle suspension (<50 nm particle size, 20 wt. % in isopropanol, Sigma Aldrich) was mixed with an equal volume of anhydrous N,N-Dimethylformamide in a round bottom flask. The lower boiling point isopropanol was evaporated in a rotary evaporator leaving the alumina nanoparticles suspended in DMF.

Alumina/perovskite co-deposition suspension

The 20 % wt. alumina nanoparticle suspension in DMF was added to the perovskite precursor solution in volume percentages of 5 %, 10 %, 15 %, 20 %, 25 %, 30 % and 40 %, yielding final weight percentages of 1 %, 2 %, 3 %, 4 %, 5 %, 6% and 8 % of alumina nanoparticles in the perovskite precursor solution respectively.

Compact TiO_2 precursor solution

A solution of titanium isopropoxide in ethanol (0.5 mM) and 40 mM solution of HCl in ethanol were mixed together slowly, in equal parts. This was made fresh before every spin coating deposition.

Device fabrication

Fluorine-doped tin oxide coated glass (Pilkington TEC 15) was patterned by etching with Zn powder and 4M HCl. The etched substrate was then cleaned with surfactant and rinsed with acetone and IPA. A compact TiO_2 layer (c-TiO_2) was deposited *via* spin coating the c-TiO_2 precursor solution (1 ml per 122 mm x 112 piece of glass) at a spin speed of 2000 rpm for one minute. The compact layer was then sintered at 500 °C for 30 minutes.

The glass was then broken along pre-scored lines into 28 mm x 28 mm pieces. The Alumina/perovskite co-deposition suspension (120 µl) was applied to the c-TiO_2 side of the glass and spin coated at 2000 rpm for 1 minute, followed by a heat treatment at 105 °C for 45 minutes to crystalize the perovskite and evaporate the DMF solvent.

After cooling, 100 µl of a chlorobenzene solution containing 68 mM Spiro-OMeTAD, 55 mM *tert*-butylpyridine (*t*-BP) and 9 mM lithium bis(trifluoromethylsyfonyl)imide salt was applied to the perovskite/alumina coated substrate and spun at a rate of 2000 RPM for 60 seconds. The 28 mm x 28 mm glass substrates were broken further (along pre-scored lines) into four 14 mm x 14 mm devices. These were left in the dark in air overnight prior to to thermal evaporation of patterned gold electrodes using a bell jar evaporator.

Solar cell device characterisation

The photovoltaic performance of assembled solar cell devices was tested using an Oriel Sol 3A solar simulator (equipped with a xenon arc lamp), under normal illumination at 1 sun (1000 W m^2, AM 1.5). The lamp output was calibrated with a reference silicon solar cell, traceable to NREL. UV-Vis spectroscopy measurements were carried out on a Perkin Elmer Lambda 750S UV-Vis-NIR spectrophotometer. Profilometry was carried out using Veeco Dektak 150 profilometer.

Scanning Electron Microscope (SEM) images were obtained using a S-4800 FEG-SEM from Hitachi (beam parameters: 12 keV, 10 µA; 8.8 mm working distance). The EDX point analyses were performed using an Oxford Instruments X-Ray detector (beam parameters: 12 keV, 10 µA; 12 mm working distance).

Figures and Tables

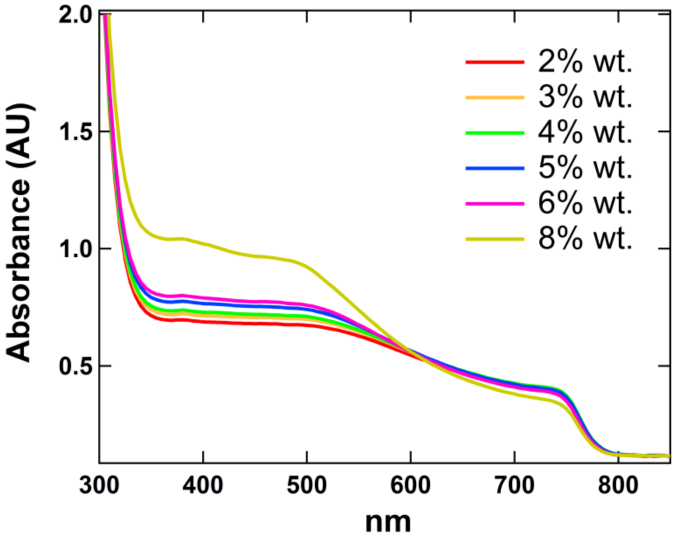

Fig. S1 UV-Vis absorption spectra of Al_2O_3/perovskite co-deposited film, spin coated onto microscope slides. It can be seen that absorbance increases as the % wt. of nanoparticles suspended in the precursor solution increases.

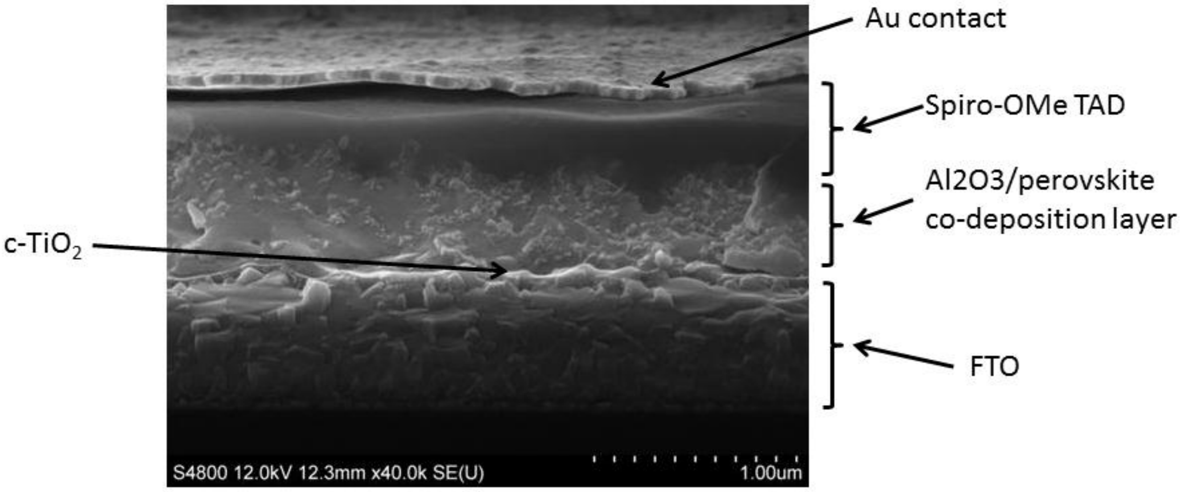

Fig. S2. Cross-sectional SEM image of one the highest efficiency devices made with the low temperature co-deposition method

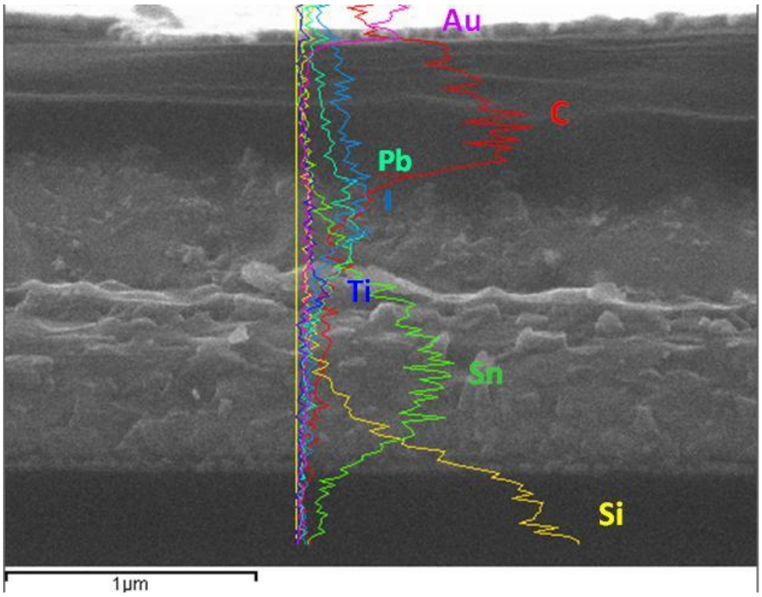

Fig. S3. Cross-sectional SEM image of one the highest efficiency devices overlain with EDX data

Table S1. Summary of 8 best performing devices: A comparison of high temperature sintering method *vs.* low temperature co-deposition method

High T Devices	V_{OC} (V)	J_{SC} (mA/cm^2)	Fill Factor (%)	η (%)	Low T Devices	V_{OC} (V)	J_{SC} (mA/cm^2)	Fill factor (%)	η (%)
Device 1	0.825	10.92	51.6	4.79	**Device 1**	0.849	11.05	47.0	4.41
Device 2	0.914	8.97	48.1	4.07	**Device 2**	0.856	9.93	50.3	4.27
Device 3	0.988	12.37	28.4	3.47	**Device 3**	0.829	10.33	49.6	4.25
Device 4	0.830	7.29	49.3	2.98	**Device 4**	0.810	10.45	48.0	4.06
Device 5	0.972	5.35	51.3	2.67	**Device 5**	0.861	10.20	44.8	3.93
Device 6	0.902	8.56	34.2	2.65	**Device 6**	0.848	10.06	44.4	3.79
Device 7	0.964	8.68	28.4	2.37	**Device 7**	0.821	9.92	45.4	3.70
Device 8	0.953	7.65	32.1	2.34	**Device 8**	0.821	7.63	55.2	3.46
Mean	**0.919**	**8.725**	**40.4**	**3.168**	**Mean**	**0.837**	**9.94**	**48.1**	**3.98**
STDEV	**0.063**	**2.165**	**10.5**	**0.880**	**STDEV**	**0.019**	**1.00**	**3.6**	**0.33**

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