Supporting information

High performance perovskite solar cell via multi-cycle low temperature process of lead acetateprecursor solutions

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Materials Method

Experimental:

The FTO (10 Ω/□, NSG Group, Japan) conducting glass substrates were patterned using zinc powder and 3 M HCl aqueous solution and cleaned with commercial detergent (2% Hellmanex in water), deionized water (DI), acetone and ethanol in a sonication bath sequentially, and dried with nitrogen. TiO_2 CL was spin-coated onto cleaned FTO glass by using 0.15 M titanium dissopropoxidebis-(acetylacetonate) (Tiacac) (75 wt% in isopropanol, Sigma-Aldrich) in a 2-propanol (99.9%, Wako) solution at 3000 rpm for 30 seconds, followed by drying at 125°C for 5 minutes and cooled down to room temperature. Further two successive coating (3000 rpm, 30 s) from 0.3 M Tiacac solution were carried out and dried at 125°C for 5 minutes. The TiO_x coated samples were further treated with 40 mM $TiCl_4$ aqueous solution at 70°C for 60 minutes. After thoroughly washing the treated substrates with DI water, the samples were annealed at 500°C for 30 minutes in a muffle furnace. The TiO_2 compact layer was treated with UV-Ozone for 10 minutes and these samples were transferred into nitrogen filled globe box for perovskite deposition.

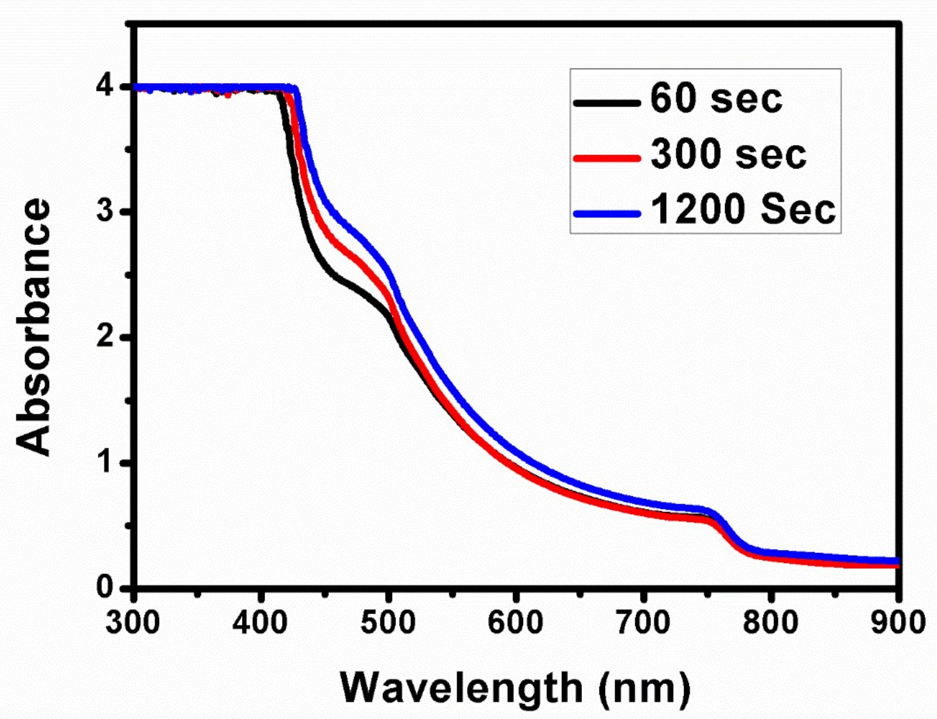
45 *wt%* solution of anhydrous lead acetate and MAI in DMF were heated at 50°C for 30 minutes prior to the coating. The perovskite solution was spin coated on UV-ozone treated CL layer at 2000 rpm for 30 s (this refers first coating). After spin coating the samples were directly transferred to hot plate which is at 90°C and heated for 60-1200 seconds. Further for

double coating, second coating was carried out immediately after first coating with 3000 rpm for 30 seconds. The hole transporting layer was spin coated at 2000 rpm from 8 *wt%* solution of spiro-OMeTAD in chlorobenzene solution containing additives of 24.66 μL lithium bis(trifluoromethanesulfonyl) imide and 5.75 μL 4-tert-butylpyridine and after coating the sample were kept overnight for ageing in nitrogen filled glove box. Finally, Au metal electrode of 100 nm was thermally evaporated on top of the HTM layer to complete the device.

Characterization

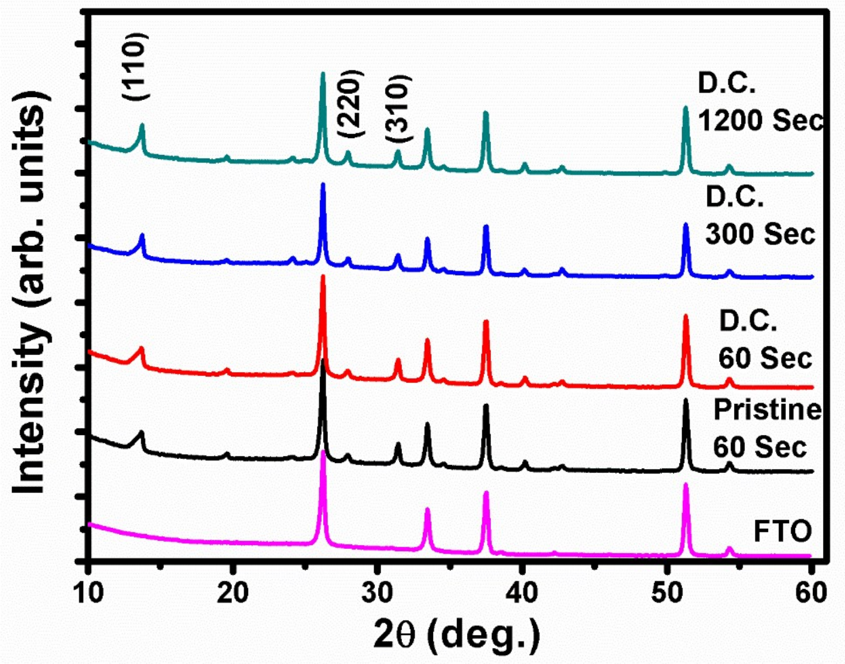
The solar cell characteristics of all devices with an active area of 0.09 cm^2 were measured with a Keithley 2400 source meter under 1 sun illumination using a Peccell Technologies PEC-L01 solar simulator (AM 1.5 G, 100 mW cm^{-2}). The EQE spectra of devices were measured in air at room temperature via Peccell Technologies, PEC-S20 action spectrum measurement setup. The structural, optical and morphological analysis were carried out using X-ray diffractometer (D8 Discover, Brucker) with CuKα radiation source, UV-Vis spectrometer (lambda 950, Perkin elmer) and scanning electron microscope (SU8000, HITACHI) respectively.

Supporting Data



S1: UV-Vis Spectrum of perovskite layer with various (60sec. to 1200 sec.) annealing times at 90 ^{o}C.

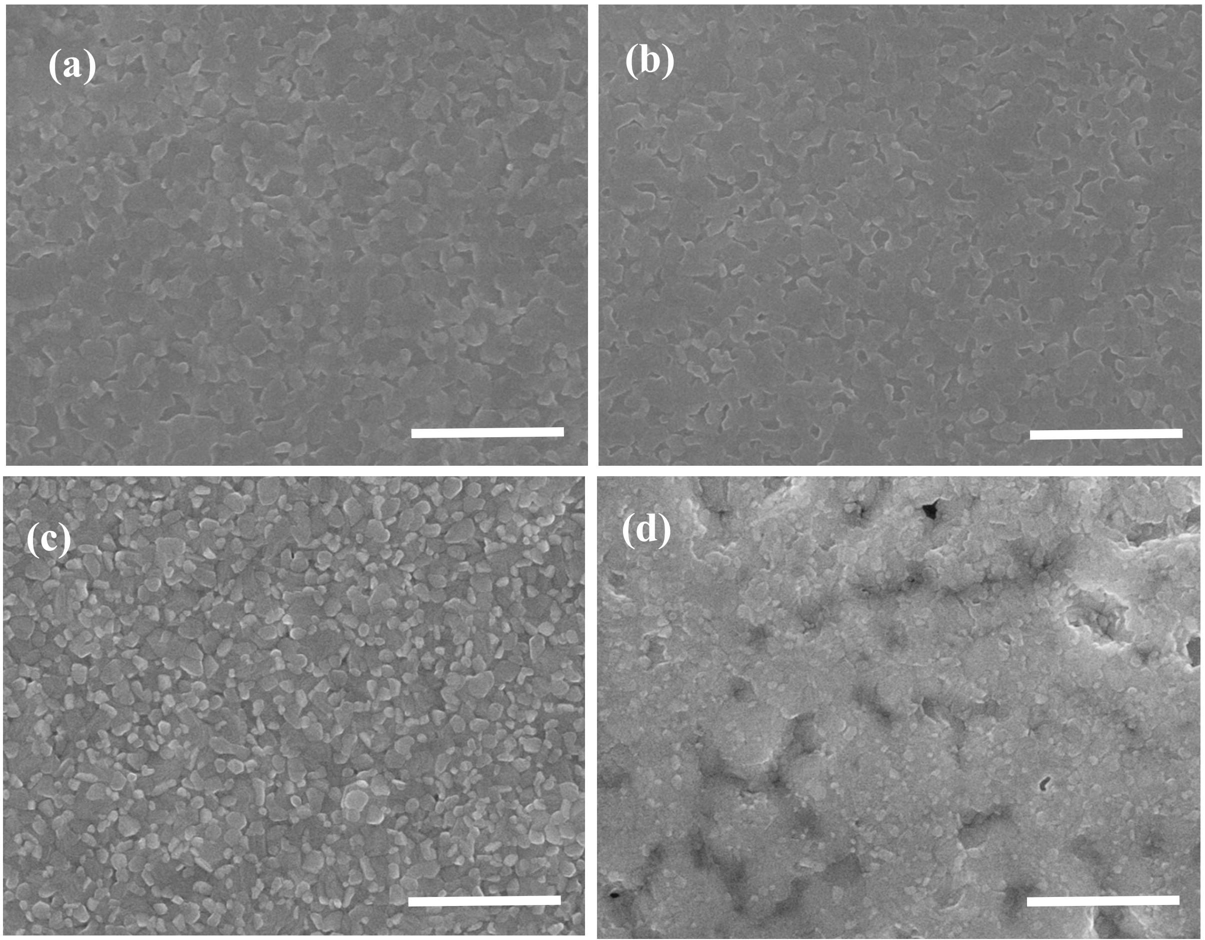
The optical absorption spectra of perovskite films showed an absorption centred around 775 nm for all the samples whereas the optical absorption of longer time annealed perovskite samples also showed almost same absorption behaviour which show a very fast formation of perovskite using lead acetate precursor.



S2: XRD of perovskite absorber layer annealed at 90^{o}C for various post annealing time (60-1200 sec). (Pristine is single coating and DC refers to double coating)

The crystal structure of perovskite films has been determined from X-ray diffraction measurements as shown in ESI 2. The perovskite films formation using lead acetate showed very fast crystallization with pure perovskite formation. The crystal quality slightly improved on long time annealing of perovskite films (1200 seconds).

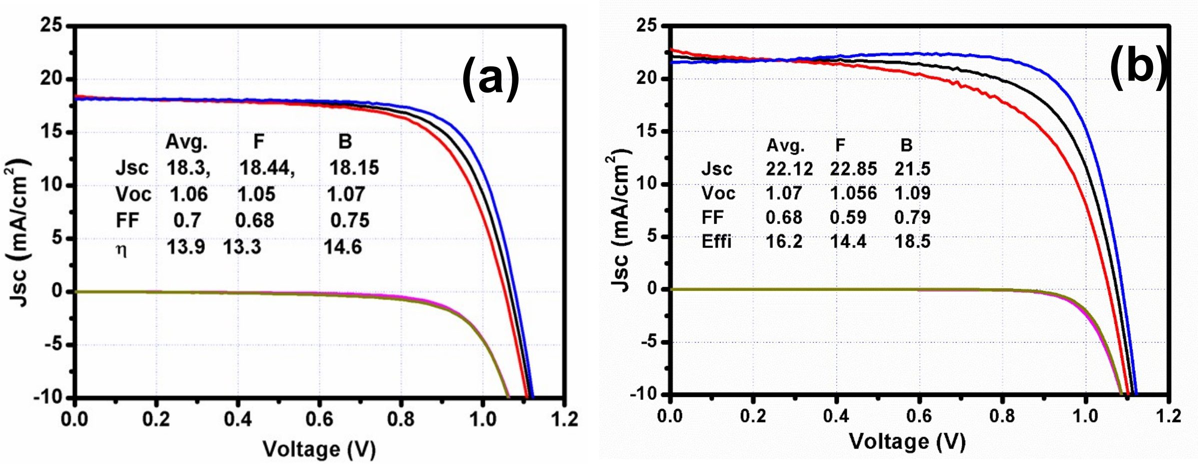
S 3



S3. SEM top view of perovskite films with (a) Single coating. (b) double coating, (c) 3 cycle coating and (d) 5 cycle coating. The scale bar is 2µm.

S3 shows the top view of various cycle coating of perovskite and in the present case the films remain smooth up to two coating cycles and further repeating the coating cycles films become rough and some pin holes appeared. In multi - cycle coating the low performance of perovskite device can be attributed to the non -uniform films and pin holes.

S4



	Avg.	F	B
Jsc	18.3,	18.44,	18.15
Voc	1.06	1.05	1.07
FF	0.7	0.68	0.75
η	13.9	13.3	14.6

	Avg.	F	B
Jsc	22.12	22.85	21.5
Voc	1.07	1.056	1.09
FF	0.68	0.59	0.79
Effi	16.2	14.4	18.5

S4: J-V curve of (a) single and (b) double coated perovskite devices.

S4 shows the J-V curve of devices measured under forward (short-circuit to open-circuit) and reverse (open-circuit to short-circuit) scans with 50 ms delay time under standard conditions in light (AM 1.5 G, 100 mW cm^{-2}) and in dark. In case of single coating cycle devices only small hysteresis is observed (S4 a), However, for multi-coating devices exhibits larger hysteresis.