



# High-temperature shaping perovskite film crystallization for solar cell fast preparation

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## ABSTRACT

Recent developments of PSCs have achieved great progress with power conversion efficiency reached to 22.1%. The crystal growth process as an important factor will significantly influence the quality of perovskite films and the device performance. In this paper, we demonstrate a simple approach for fast preparation based on the precursor compositions of (FAPbI<sub>3</sub>)<sub>0.85</sub>(MAPbBr<sub>3</sub>)<sub>0.15</sub> by regulating the temperature during annealing process for transition from FAI, PbI<sub>2</sub>, MABr, and PbBr<sub>2</sub> to perovskite. The film will be shaped in 15 s for one-step deposition at annealing temperature of 220 °C. It has been found that higher temperature will induce perovskite to form larger size and oriented crystal grains rapidly. This will contribute to longer carrier lifetime and smaller carrier recombination which, are beneficial for solar cell device. The champion cell with PCE of 17.1% had been obtained measured under conditions of AM 1.5 G, 100 mW/cm<sup>2</sup>. Furthermore, this method exhibits good stability which is benefited from large size crystal, the device will not be deteriorated and the PCE maintained over 90% after 3 months.

## 1. Introduction

Organic-inorganic perovskite and its derivatives as a new branch of solar cells have achieved rapid development since miyasaka and co-workers reported perovskite solar cells (PSCs) for the first time in 2009 [1–7]. In the past few years, PSCs have shown rapid progress due to the high power conversion efficiency (PCE) and relatively low potential cost. Park et al. reported Spiro-OMeTAD as HTM used in solid state organic/inorganic perovskite solar cells (PSCs) and got photoelectric conversion efficiency of 9.7% [8]. In last year, seok's team used intramolecular exchange process (IEP) technique to induce perovskite to form crystals by PbI<sub>2</sub>-DMSO reacting with FAI and got 20.1% power conversion efficiency. This certified record was refreshed up to 22.1% until today [9].

Despite such a rapid progress in perovskite solar cells [10–13], many issues about efficiency and stability in the light-harvesting process still remain unclear. The intrinsic property of the metal-halide perovskite was proposed as a fundamental factor influencing the performance. The poor morphology of the perovskite thin film, trap states at surface, interstitial defects in crystal lattice, recombination

between charge carriers, and the crystallization process during the growth of the perovskite thin films not only cause electrical shorting but also harm charge carriers transportation. In order to increase devices performance and avoid energy loss, researchers focus their attention on making dense and low defects perovskite layer with high quality crystallization.

A large number of recent successes on lead halide perovskite solar cells are observed in solution processed thin films due to their facile fabrication and several strategies have been used to control the crystallization processes. Adjustment of precursor solution [14], additives like HX [15], gas-assisted method [16], microwave irradiation process [17], and laser-assisted crystallization [18] have been used for nucleation and to promote growth of the perovskite crystals. The effect of crystallization conditions, especially temperature has an important impact on the morphology of the perovskite films. The effect of temperature and annealing time on the perovskite crystal formation have lot of reports, ranging from 90 °C to 130 °C and from 20 to 90 min respectively. Liu's team had reported that higher temperature will produce perovskite films (MAPbI<sub>3</sub>) not only got better crystallinity and larger grain size, but also better photovoltaic performance with a PCE

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of 12.9% prepared at 200 °C and heated for 6 min [19]. But the most commonly used lead halide perovskite of MAPbI<sub>3</sub> has a lower thermal decomposition temperature, and the decomposition process can be further accelerated by high temperature [20–22]. Due to the thermal stability obstacle, high temperature treatment of lead halide perovskite film crystallization for fast preparation only has scarcely studies [23–25]. In this aspect, a higher thermal decomposition material (FAPbI<sub>3</sub>)<sub>x</sub>(MAPbBr<sub>3</sub>)<sub>1-x</sub> is desired to resist heating destruction, which will enable perovskite films can be subjected to high temperature treatment owing to improved stability of intrinsic perovskite absorber layer and much higher formation energies [26,27].

The conventional thermal annealing process transfers energy into the as-prepared perovskite film is from bottom to up, which retards the evaporation of the solvent and makes the grains grow gradually. In contrast, high-temperature provides a simple and straight forward way to define the crystal orientation, unit cell volume and grain size. Compared with conventional annealing method which usually produces more defects with small crystal grain, perovskite films treated by high temperature always have slightly larger grain size which could render low trap density and excellent charge-carrier diffusion length. This can effectively reduce carrier recombination rate, and at the same time improve the carrier mobility and charge carrier collection efficiency [28–30]. All of this will deliver decent power conversion efficiency (PCE).

In this work, we demonstrate a simple approach for fast preparation method at annealing temperature of 220 °C, and the film will be shaped in 15 s. The precursor composition was changed from MAPbI<sub>3</sub> to (FAPbI<sub>3</sub>)<sub>0.85</sub>(MAPbBr<sub>3</sub>)<sub>0.15</sub> for one-step deposition. The effects of high-temperature treatment on the crystallinity, morphology of perovskite film and associated photovoltaic performance were systematically investigated. Results showed that higher annealing temperature would accelerate transition rate from FAI, PbI<sub>2</sub>, MABr, and PbBr<sub>2</sub> to (FAPbI<sub>3</sub>)<sub>0.85</sub>(MAPbBr<sub>3</sub>)<sub>0.15</sub> and produce perovskite film with better crystallinity, enhanced crystal orientation and larger crystal grain, which was further confirmed by an XRD analysis. The TAS results showed that the carrier lifetime increased from 200.6 to 440.1 ns by high temperature treatment. The largest crystal grains around 800 nm had been produced at 220 °C, which delivered a highest PCE of 17.1%. In addition the as-prepared perovskite solar cell have higher stability. The device without any encapsulation did not deteriorate with the PCE maintaining over 90% for 3 months after its preparation while the device prepared by conventional method only held 3 weeks.

## 2. Experimental

### 2.1. Materials

Lead (II) Iodide (99.99%, trace metals basis) [for Perovskite precursor] was purchased from TCI, CH<sub>3</sub>NH<sub>3</sub>I from Xi'an Polymer Light Technology Corp., TiO<sub>2</sub> (particle size: about 30 nm, crystalline phase: anatase) TiO<sub>2</sub> from dysol. DMF, chlorobenzene, lithium bis (trifluoromethylsulphonyl) imide (Li-TFSI) and 4-tert-butylpyridine (tBP) from Aldrich, and spiroMeOTAD from Borun New Material Technology Co., Ltd. All chemicals were directly used without further purification. Patterned FTO-coated glass substrates with a sheet resistance of 15 Ω sq<sup>-1</sup> were cleaned sequentially by ultrasonication in mild detergent, deionized water, acetone and ethanolamine. The substrates were exposed to UV-ozone for 20 min prior to the spin coating step.

### 2.2. Solar cell device fabrication

A thick dense blocking layer of TiO<sub>2</sub> (bl-TiO<sub>2</sub>) was deposited onto F-doped SnO<sub>2</sub> (FTO) substrate by spin coating to prevent direct contact between the FTO and the hole-conducting layer. A TiO<sub>2</sub> blocking layer was deposited on the cleaned FTO by spray pyrolysis, using dry air as

carrier gas, at 460 °C from a precursor solution of 0.6 ml of titanium diisopropoxide and 0.4 ml of bis (acetylacetonate) in 7 ml of anhydrous isopropanol. A mesoporous TiO<sub>2</sub> layer (dysol, particle size: about 30 nm, crystalline phase: anatase, diluted to w/w=1/6 in ethanol) about 160–180 nm thickness film was deposited by spin coating at 4000 rpm for 20 s onto the bl-TiO<sub>2</sub>/FTO substrate. After spin coating, the substrate was immediately dried on a hotplate at 80 °C, and the substrates were then calcined at 500 °C to remove organic components. The perovskite film was deposited by spin coating onto the TiO<sub>2</sub> substrate. The prepared FAI powders and PbI<sub>2</sub> (TCI) for 1.2 M (FAPbI<sub>3</sub>)<sub>0.85</sub>(MAPbBr<sub>3</sub>)<sub>0.15</sub> solution were stirred in a mixture of DMSO and DMF (v/v=8:2) at 60 °C for completely dissolved. The resulting solution was coated onto the mp-TiO<sub>2</sub>/bl-TiO<sub>2</sub>/FTO substrate by a consecutive two-step spin-coating process at 1100 and 5000 r.p.m for 20 s and 30 s respectively. During the second spin-coating step at last 10 s, the substrate was treated with weak corrosive mixed anti-solvent (chlorobenzene) to form the perovskite crystal, and the substrate was then heated at 220 °C for 15 s on a hotplate under low speed flow clean dry air. Then quickly transferred to the aluminum plate for radiating to prevent the film from continuous overheating. When the temperature decreased to 80–100 °C, annealing at 105 °C for another 5 min. (The conventional method was that the film coated with perovskite precursor was directly heated at 100 °C for 1 h). FAI was synthesized from hydroiodic acid reacting with formamidine acetate according to reference. The HTM layer was deposited by spin-coating at 4000 rpm for 20 s, using a solution of spiroMeOTAD, 4-tert-butylpyridine, and lithium bis (trifluoromethylsulphonyl) imide and the Co (III)-complex. Finally, Gold (80 nm) counter electrode was deposited by thermal evaporation on top of the device to form the back contact.

### 2.3. Characterization

The PCE and *J*-*V* curves were measured by using a solar simulator (Newport, Oriel Class A, 91195A) with a source meter (Keithley 2420) at 100 mA/cm<sup>2</sup> illumination AM 1.5 G and a calibrated Si-reference cell certified by NREL. The *J*-*V* curves for all devices were measured by masking the active area with a black mask 0.09 cm<sup>2</sup>. UV-vis absorption spectra were recorded on a UV-vis spectrometer (Hitachi U-3300). Incident photon to current efficiency (IPCE) were confirmed as a function of wavelength from 300 to 800 nm (PV Measurements, Inc.), with dual Xenon/quartz halogen light source, measured in DC mode with no bias light used. The setup was calibrated with a certified silicon solar cell (Fraunhofer ISE) prior to measurements. Film morphology was investigated by using a high-resolution scanning electron microscope equipped with a Schottky Field Emission gun. PL spectra were recorded by exciting the perovskite films deposited onto mesoporous TiO<sub>2</sub> at 490 nm with a standard 450 W Xenon CW lamp. The signal was recorded by a spectrofluorometer (photon technology international) and analyzed by the software Fluorescence. X-ray diffraction spectra of the perovskite films were recorded on an X'Pert MPD PRO (PANalytical). The data were collected at room temperature in the 2θ range 10–50°. The automatic divergence slit (10 mm) and beam mask (10 mm) were adjusted to the dimension of the films. A baseline correction was applied to all X-Ray thin film diffractograms to compensate for the broad feature arising from the FTO glass and anatase substrate. Transient absorption spectra (TAS) was record on LKS (Applied photophysics). The cells were measured before evaporating gold counter with a pump light wavelength of 500 nm and a probe light wavelength of 760 nm. Repetition rate of 5 Hz, the energy of laser device is 150 μJ/cm<sup>2</sup>.

## 3. Result and discussion

Fig. 1 depicted the preparation process, the precursor solution was spin-coated on the TiO<sub>2</sub> substrate to form perovskite layer followed by

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