

Supporting Information for

Ionic liquid-assisted perovskite crystal film growth for high performance planar heterojunction perovskite solar cells

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Experimental

Materials

An aqueous dispersion of PEDOT: PSS (Baytron[®] PVP Al 4083) was obtained from Heraeus Co. 1-ethylpyridinium chloride (1-EC) was purchased from ACROS. Fullerene derivative (PCBM) with purity over 99% was obtained from Solenne B.V., Netherlands. N, N-dimethylformamide (DMF, purity 99.5%) obtained from J&K Scientific. $\text{PbAc}_2 \cdot 3\text{H}_2\text{O}$ (99.995%, metals basis, Alfa Aesar). PbAc_2 was dehydrated in vacuum drying oven at 75°C for 1 h prior to use. $\text{CH}_3\text{NH}_3\text{I}$ was synthesized using the same method as published in literature. ITO-coated glass with a sheet resistance of 10 Ω / square is used for device fabrication. The routine cleaning procedure includes ultra-sonication in a solution of detergent, deionized water in sequence.

Preparation of the perovskite precursor

$\text{CH}_3\text{NH}_3\text{PbI}_3$ precursor solution with a concentration of 0.5 M was prepared by mixing lead salts (PbAc_2) and $\text{CH}_3\text{NH}_3\text{I}$ in DMF. The mole ratio of PbAc_2 : $\text{CH}_3\text{NH}_3\text{I}$ mixture was 1:3 according to the literature. Different content in quality of 1-EC additive (0 wt%, 0.25 wt%, 0.5 wt%, 0.75 wt%, 1 wt%, 1.25 wt%) was added to this precursor solution.

Device fabrication and performance measurements

The preparation of the solar cell is described as follows. The washed ITO coated glasses were dried under an infrared lamp for 15min. The substrate was then treated with UV-ozone for 15 min. The PEDOT: PSS layer of about 40 nm was fabricated by spin coating at 3000 rpm for 30 s on ITO-coated glass substrates, followed by baking at 120°C for 30 minutes in air. The precursor

solutions were spin-coated on top of the PEDOT: PSS layer at 2000 rpm for 40 s. The perovskite-coated films were then annealed on a hot-plate at 90°C for 10 min. The measured thickness of the perovskite film was about 180 nm. The humidity is 19%. For the electron transport materials, 24 mg/mL PCBM dissolved in o-dichlorobenzene solution was then sequentially casted onto the CH₃NH₃PbI₃ film by spin coating at 1000 rpm for 60 s in ambient condition and then dried in air on a 100 °C hot-plate for 10 minutes. Finally, 150 nm Al are thermally evaporated under a vacuum with a pressure of 1×10⁻⁴ Pa to form planar device structure. The characterization procedures were performed in an ambient atmosphere at room temperature. The active area of each device was about 0.075 cm² defined by a shadow mask. The J-V characteristics of the un-encapsulated devices were measured under intensity of 100 mW/cm² illuminations by using a 150W solar simulator with an AM1.5 G filter. The light intensity was calibrated with an Oriel mono-Si reference cell (CROWNTECH PVM 272 certificated by NREL). The surface roughness and morphology of the CH₃NH₃PbI₃ layers were characterized by Bruker INNOVA[®] SPM in tapping mode. The thickness of above layer was measured by a surface profiler (KLA Tencor[®] Alpha Step D-100). More detailed information on device fabrication and device characterization can be found in our previous paper.

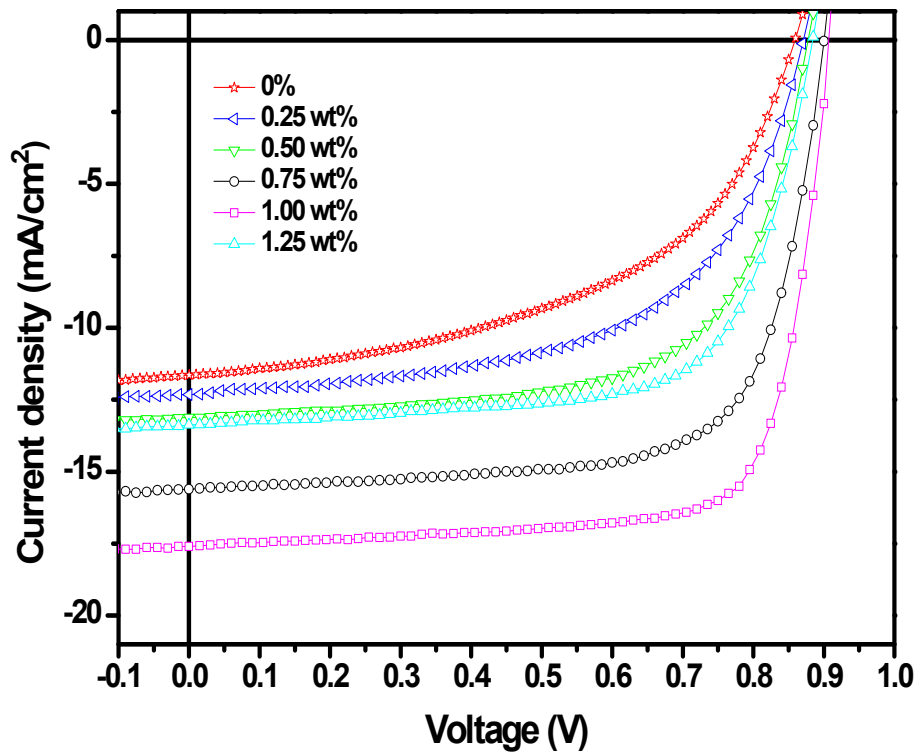


Fig.S1 The J-V characteristics for perovskite solar cells with different concentration of 1-EC additive.

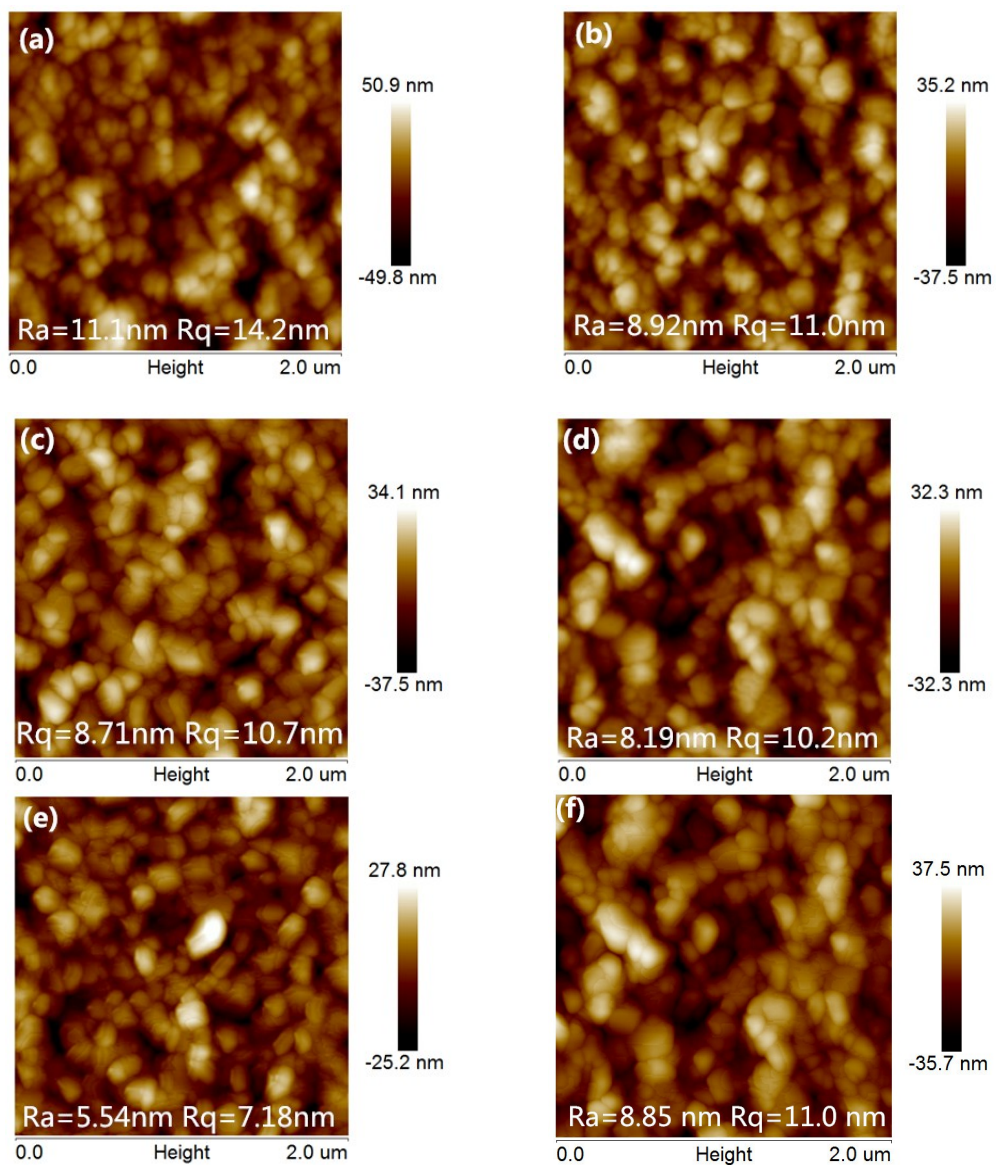


Fig. S2 The AFM topography of the surface morphology of pristine $\text{CH}_3\text{NH}_3\text{PbI}_3$ (a) and with different content of 1-EC as additive. (b: 0.25wt%; c: 0.5wt%; d: 0.75wt%; e: 1wt%; f: 1.25wt%).

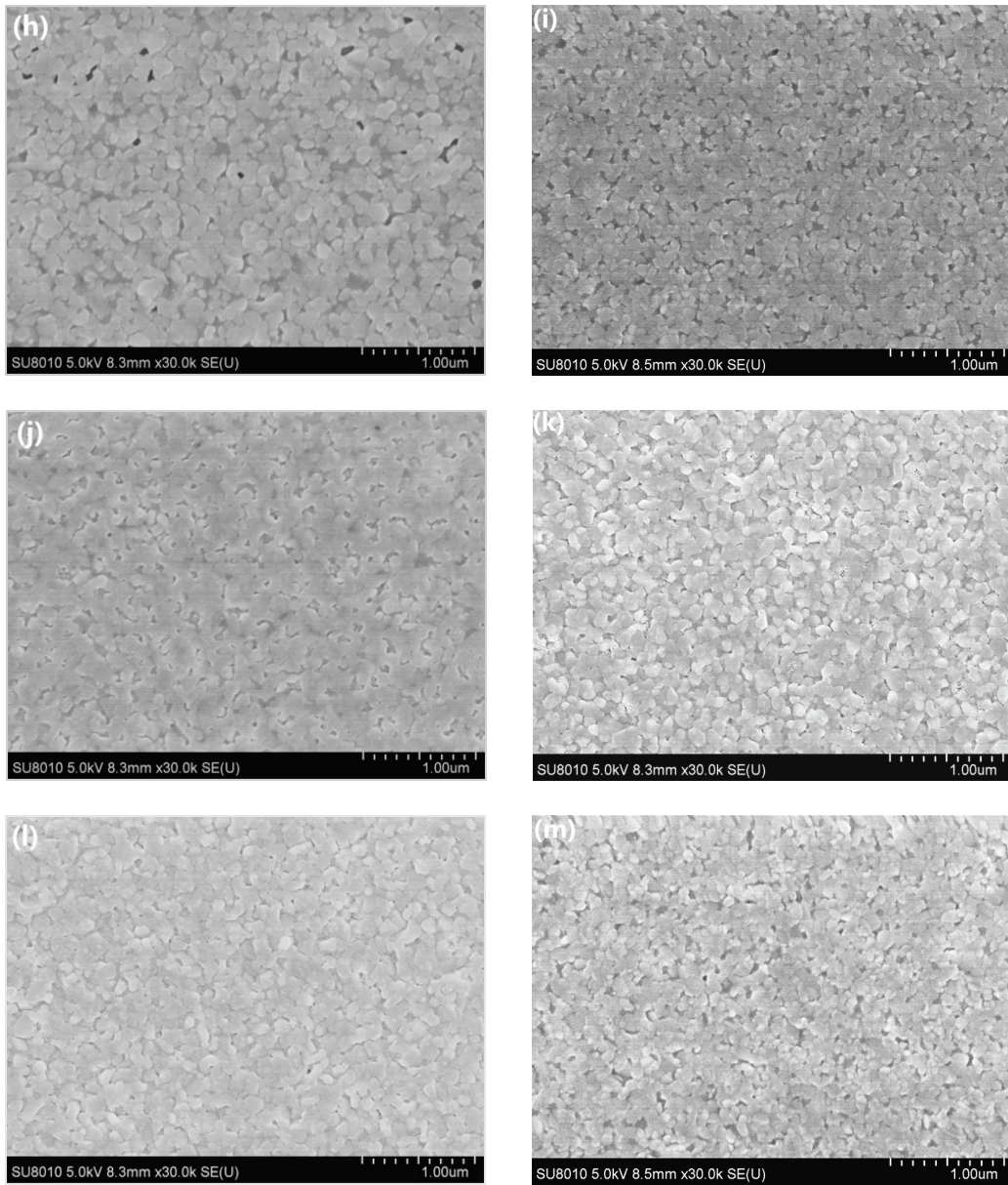


Fig. S3 The SEM images of perovskite film with different content of 1-EC (h-pristine; i-0.25wt%; j-0.5wt%; k-0.75wt%; l-1wt%; m-1.25wt%).

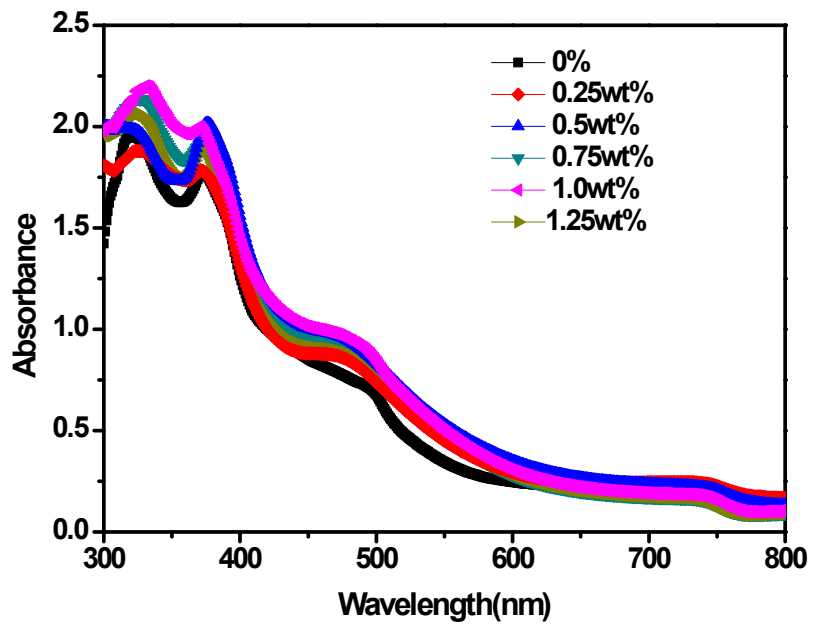


Fig. S4 The UV-Vis absorption for pristine CH₃NH₃PbI₃ film and with different content of 1-EC as additive.

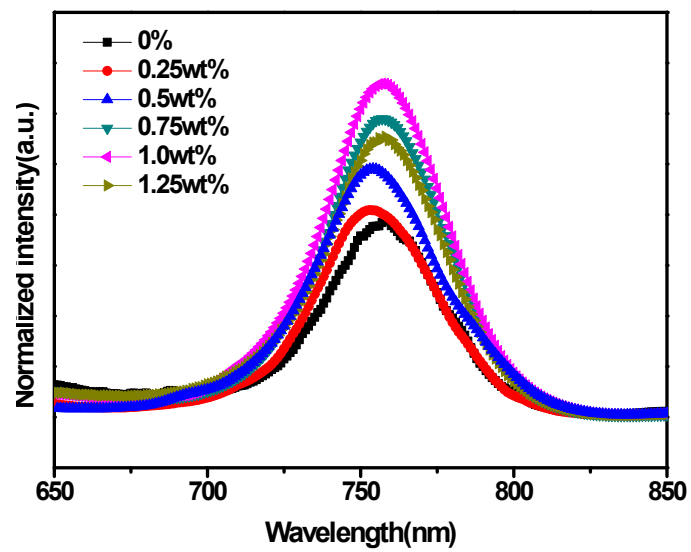


Fig. S5 The steady-state PL spectra (b) for pristine $\text{CH}_3\text{NH}_3\text{PbI}_3$ film and with different content of 1-EC as additive.

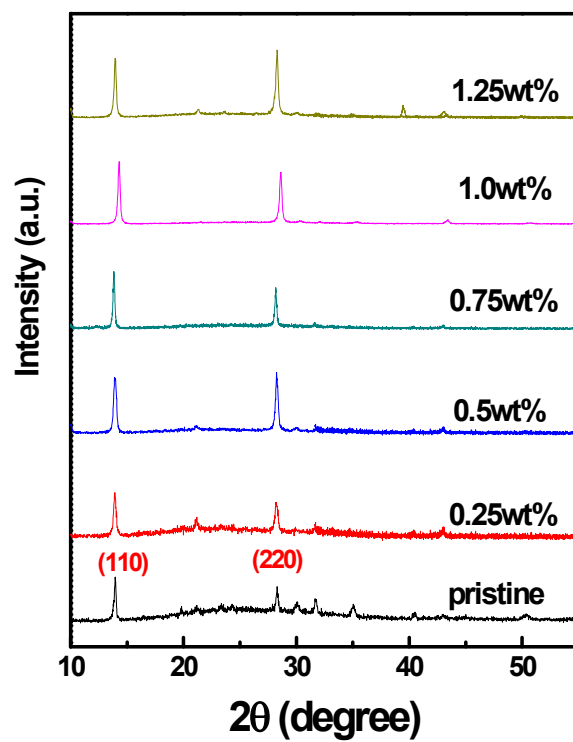


Fig. S6 The XRD patterns of $\text{CH}_3\text{NH}_3\text{PbI}_3$ films formed on PEDOT: PSS with different concentration of 1-EC additive.

Table S1 Peak parameters of the average crystallite sizes in pristine CH₃NH₃PbI₃ film and the films formed with 1.0wt% 1-EC as additive estimated from the Scherrer formula.

Parameters	Perovskite films	
	With 1.0 wt% 1-EC additive	Without additive
FWHM (β)	0.20	0.28
Bragg angle (θ) primary peak (degree)	7.13	7.01
X-ray wavelength (λ)	0.154	0.154
Constant (K)	0.9	0.9
D _{XRD} (nm)	40.03	28.59