

Efficient and non-hysteresis $\text{CH}_3\text{NH}_3\text{PbI}_3/\text{PCBM}$ planar heterojunction solar cells



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ABSTRACT

Highly efficient and non-hysteresis organic/perovskite planar heterojunction solar cells were fabricated by low-temperature, solution-processed method with a structure of ITO/PEDOT:PSS/ $\text{CH}_3\text{NH}_3\text{PbI}_3$ /PCBM/Al. The high-quality perovskite thin film was obtained using a solvent-induced-fast-crystallization deposition involving spin-coating the $\text{CH}_3\text{NH}_3\text{PbI}_3$ solution followed by top-dropping chlorobenzene with an accurate control to induce the crystallization, which results in highly crystalline, pinhole-free, and smooth perovskite thin film. Furthermore, it was found that the molar ratio of CH_3NH_3 to PbI_2 greatly influence the properties of $\text{CH}_3\text{NH}_3\text{PbI}_3$ film and the device performance. The equimolar or excess PbI_2 was facile to form a flat $\text{CH}_3\text{NH}_3\text{PbI}_3$ film and produced relatively uniform perovskite crystals. Perovskite solar cells (PSCs) with high-quality $\text{CH}_3\text{NH}_3\text{PbI}_3$ thin film showed good performance and excellent repeatability. The power conversion efficiency (PCE) up to 13.49% was achieved, which is one of the highest PCEs obtained for low-temperature, solution-processed planar perovskite solar cells based on the structure ITO/PEDOT:PSS/ $\text{CH}_3\text{NH}_3\text{PbI}_3$ /PC₆₁BM/Al. More importantly, PSCs fabricated using this method didn't show obvious hysteresis under different scan direction and speed.

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1. Introduction

Low-temperature and solution-processed solar cells have attracted more and more attention in recent years since they are compatible with flexible substrate for large-scale, high-throughput, and low-cost roll-to-roll (R2R) printing manufacturing [1–3]. Recently, organolead halide perovskites have arisen as excellent earth abundant photovoltaic materials and show many advantages including small bandgap, strong absorption, long carrier diffusion length, high carrier mobility, and so on [4–7]. It was reported that the certified power conversion efficiency (PCE) of perovskite solar cells (PSCs) is over 20% [8]. There are several device architectures and deposition techniques used for fabricating highly efficient PSCs [7,9–17]. Typically, perovskite materials are deposited on mesoporous metal oxide scaffold such as TiO_2

[7,14], Al_2O_3 [12], ZrO_2 [9]. Then a hole transport layer (HTL) is deposited subsequently, which is normally one of organic semiconductor materials, for example, spiro-MeOTAD [5,8,11,12,18,19], poly-triarylamine (PTAA) [14], P3HT [20–22]. However, the mesoporous scaffold layer has to be sintered at high temperature (usually above 450 °C) [8,14,19]. The high-temperature sintering is a potential obstacle for the further development and commercialization of PSCs, especially in flexible large-scale R2R production process [5,8]. It is particularly critical to explore the possibilities of preparing high-performance PSCs with a simple structure under low temperature.

Templating from organic photovoltaics (OPVs), planar heterojunction (PHJ) structure fabricated at low temperature was introduced into PSCs [1,10,23–25]. Planar architecture potentially provides enhanced flexibility for device optimization, multijunction construction, and the investigation of device physics, but it requires tremendous effort to fabricate high-quality perovskite thin film [10,16,26]. The solution-processed fabrication is highly recommended to prepare PHJ PSCs due to its low cost and high output as compared with vacuum evaporation. However, it is very difficult to control the formation of perovskite thin film with good

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coverage and without pinholes, which are important to the device performance [27]. The low-temperature, solution-processed PHJ PSCs was first reported by Jeng et. al. in 2013, in which the *PCE* is as low as 3.9% [10]. They pointed out that it is a great challenge to form a thick and uniform perovskite thin film. Then, a relatively higher *PCE* of 9.22% was achieved based on the structure of ITO/PEDOT:PSS/Perovskite/PCBM/C₆₀/BCP/Al by optimizing the precursor ratio and utilizing multilayer modification [28]. However, the *PCEs* are still relatively lower than commonly high-temperature processed PSCs.

The anomalous hysteresis is another issue for both high-temperature mesoporous and low-temperature processed PHJ PSCs [8,14,29–31]. Snaith et. al. pointed out that the defects states including interface states or interstitial defects (iodide or methylammonium) and ferroelectric properties are the possible reasons for producing the hysteresis [29,31]. Huang et. al. utilized multilayer modification in PSCs with the structure of ITO/PEDOT:PSS/CH₃NH₃PbI₃/PCBM/C₆₀/BCP/Al and significantly eliminated the anomalous hysteresis [28].

Herein, we used a low-temperature, solution-processed method combining with a solvent treatment technique, *i.e.*, solvent-induced-fast-crystallization deposition (SIFCD), to form a very continuous and flat CH₃NH₃PbI₃ layer on ITO/PEDOT:PSS substrate, and further fabricate perovskite/fullerene PHJ solar cells. The morphological, absorbance and crystallographic properties of perovskite thin film as well as PSCs performance were studied in detail. It was found the morphology of perovskite films and the size of perovskite crystals are sensitive to the precursor composition, and a 1:1 precursor stoichiometry could lead to the best performance. Using a simple structural PHJ PSCs based on ITO/PEDOT:PSS/CH₃NH₃PbI₃/PCBM/Al, a *PCE* as high as 13.49% was achieved, which is one of the highest *PCEs* based on this device structure [1,10]. More interestingly, the performance of these simple structural PSCs showed good repeatability and no obvious hysteresis, which have great potential application in commercial R2R production.

2. Experimental section

2.1. Materials

Organolead halide perovskite precursor solution was made with mixing methylammonium iodide (CH₃NH₃I, 99%, Jingge, Wuhan) and lead iodide (PbI₂, 99%, Zhengpin, Shanghai) in *N,N*-dimethylformamide (DMF, J&KSeal) with various molar ratios (CH₃NH₃I/PbI₂) at 1:0.6, 1:0.8, 1:1, 1:1.2. The precursor solution was vigorously stirred over night at 60 °C, and was filtered with a 0.22 μm PVDF filter before the deposition. Fullerene derivative [6,6]-phenyl C₆₁-butyric acid methyl ester (PCBM) was purchased from American Dye Source, and was dissolved in anhydrous chlorobenzene (CB, J&KSeal). All materials were used directly without any purification.

2.2. Device fabrication

Fig. 1 shows the structure of PSCs. The patterned indium tin oxide (ITO) coated glass was used as the substrate, which was ultrasonically cleaned in acetone, detergents, distilled water and isopropyl alcohol for 15 min respectively, then dried by hot air and treated by UV-ozone for 15 min. The PEDOT:PSS layer (Baytron, PVP Al 4083) with a thickness of about 50 nm was spin-coated at 3000 rpm onto the patterned ITO substrate and dried on hot plate at 150 °C for 15 min. The deposition of CH₃NH₃PbI₃ layer was performed in a glove box (both H₂O and O₂ <1.0 ppm) and the SIFCD method was used to produce

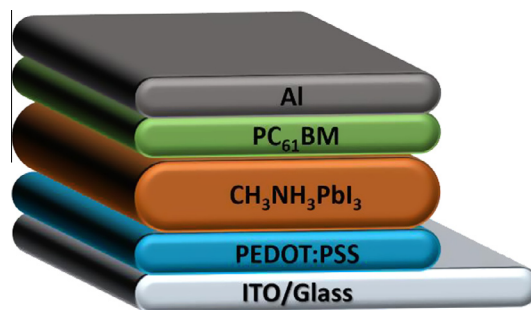


Fig. 1. The schematic of PSC structure.

high-quality film, in which a poor solvent was introduced to induce the formation of perovskite thin film [14,26]. The scheme of perovskite films prepared by the SIFCD method is shown in Fig. S1a. Firstly, CH₃NH₃PbI₃ solution (50 μL) was first dropped onto a PEDOT:PSS coated ITO substrate, of which the size is 1.5 cm × 1.5 cm. The substrate was then spun at 4000 rpm. When the precursor roll out, anhydrous CB solvent (50 μL), as a poor solvent, was quickly dropped onto the center of the substrate (The detailed speed-time profile for spin coating process with CB drop-casting was shown in Fig. S1b). The color of just deposited CH₃NH₃PbI₃ layer was changed from transparent to light brown. The thickness of perovskite film was controlled using the concentration of precursor solution. The total precursor concentration was varied from 450 mg/ml to 650 mg/ml with an interval of 50 mg/ml, correspondingly to form 210 nm to 370 nm perovskite films with a similar interval of about 40 nm. The samples were subsequently treated at 100 °C for 10 min. After cooling to room temperature, PCBM solution was spin-coated onto the CH₃NH₃PbI₃ layer with different thicknesses. Finally, a bar-like defined Al electrode (100 nm) by mask was deposited on top of PCBM film by thermal evaporation under the vacuum of ~4 × 10⁻⁶ mbar, resulting in an active area of 0.09 cm².

2.3. Characterization

Crystallographic properties of CH₃NH₃PbI₃ films were characterized by X-ray diffraction (XRD, Rigaku D, Max 2500, Japan). Absorption properties of CH₃NH₃PbI₃ films were characterized by UV-vis spectrophotometer (UV-vis, TV-1800PC, Pgeneral). The morphology was characterized by atomic force microscopy (Agilent Technologies 5500 AFM/SPM System, USA) with tapping-mode. Current density–voltage (*J*–*V*) characteristics of PSCs were measured by digital Source Meter (Keithley, model 2420). During the measurements, the scan speed was located at 0.3 V/s and the scan direction is from +1.2 V to –1.2 V if without special description. A solar simulator (91160s, Newport, AM 1.5G) was used for *PCE* test. Light intensity was 100 mW/cm² calibrated by a standard silicon solar cell. The thickness of the perovskite and PCBM films were measured with both surface profiles (Dektak 150, Veeco, USA) and AFM.

3. Results and discussion

The precursor ratio is closely related to the quality of CH₃NH₃PbI₃ layer and the performance of PSCs. To achieve a high-quality CH₃NH₃PbI₃ film, the precursor ratio of CH₃NH₃I to PbI₂ was varied from 1:0.6 to 1:1.2 and the total precursor concentration was fixed at 450 mg/ml. Fig. 2 shows the XRD patterns of CH₃NH₃PbI₃ films with different molar ratios. The strong diffraction peaks at 2θ = 13.98°, 28.22°, and 31.67° appear for all samples, corresponding to the (1 1 0), (2 2 0) and (3 1 0) planes of CH₃NH₃PbI₃

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