



Performance improvement of perovskite solar cells through enhanced hole extraction: The role of iodide concentration gradient



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ABSTRACT

Realization of fast hole extraction is an effective route for the performance enhancement of inverted perovskite solar cells (PSCs). The precise bandgap tuning in perovskite layer has been expected to accelerate hole extraction. Therefore, in this report, we demonstrate a modified one-step fabrication method for the bandgap tuning of perovskite layer by designing iodide concentration gradient. Prior to the perovskite precursor solution spin-coated, different concentration of methanaminium iodide (MAI) solution was pre-coated on PEDOT:PSS layer to form iodide concentration gradient in perovskite layer. Fast hole extraction and short lifetime due to iodide concentration gradient was confirmed by photoluminescence measurement. By optimizing the MAI concentration with 4 mg/ml, the modified PSCs exhibited a power conversion efficiency of 16.67% due to an increase in short-circuit current density (J_{sc}) from 19.66 mA/cm² to 23.52 mA/cm² and an increase in open circuit voltage from 0.97 V to 1.01 V. These values are significantly higher than PSCs using conventional one-step fabrication method without pre-coated MAI layer, which exhibit efficiency of only 14.8%. We think that this new approach is an effective way to enhance the performance of PSCs.

1. Introduction

Organic-inorganic perovskite materials have attracted increasing attention for the next-generation solar cells [1], because of their outstanding features, such as adjustable bandgap [2], high optical absorption coefficients [3], long electron–hole diffusion lengths [4] and high mobility of charge carriers [5]. The distinct advantages of perovskite solar cells (PSCs) are high performance [6] and simple fabrication process [7]. In the past few years, extensive research in perovskite material [8], device architecture [9], fabrication processes [10–12] and elemental engineering [13] has boosted the development of PSCs. The power conversion efficiency (PCE) of PSCs has shown an unprecedented increase from 3.8% [14] in 2009–22.1% in 2016 [15]. In order to improve the PCE, a variety of method has been explored to optimize the fabrication procedure, which includes one-step spin coating [16], two-step sequential deposition [17], the vacuum deposition [18] and the vapor-assisted solution process. In all these methods, the one-step spin coating method has motivated the most extensive studies for fabricating high performance PSCs, as the one-step spin coating method could form high uniform perovskite film with full coverage over the substrates.

At present, two major architectures have been developed for PSCs, i.e., the conventional configuration [19] and the inverted structures [20]. The conventional configuration in which the perovskite layer is deposited on the top of an electron transporting layer (ETL), followed by a hole transporting layer (HTL) to form a n-i-p type device, while the inverted device configuration (p-i-n), in which ETLs are used on the top of perovskite layer to reverse the direction of charge extraction. In comparison with the conventional configuration, the inverted structure devices have exhibited the advantages of simple device fabrication and small hysteresis [21–23]. However, the inverted structure suffers from shortcomings like small short-circuit current density (J_{sc}) [24], which restricts the further enhancement of PCE. The main reason for this small J_{sc} is that the hole drift acceleration is lower than that of electron inside the perovskite layer. Thus, it is worth to find ways to improve J_{sc} of the inverted structures device. In order to obtain high J_{sc} , band gap engineering [25] and interface engineering [24,26,27] have been widely employed but with little effect on large open circuit voltages (V_{oc}).

Recently, halide concentration gradient in perovskite layer has been used for designing bandgap tuning to accelerate carriers extraction. When using halide element concentration gradient, the valence band edge can be modified in greater extent than the conduction band edge

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[25], suggesting that halide concentration gradient could shuttle holes more rapidly to HTL. Previous research has taken the conversion of halide ions to modify the interface and form the halide concentration gradient to enhance the performance of PSCs, but the reported method using vaporization of HBr to form Br concentration gradient in perovskite layer was complicated and high cost. Although their obtained PCE was increased, the obtained J_{sc} was lower than that of the reference device. Their obtained J_{sc} was also still less than 23 mA/cm². Thus, it is urgent to explore a low cost and simple method not only to realize high J_{sc} but also high V_{oc} . To solve this issue, we proposed a modified one-step spin coating method, i.e., prior to the perovskite precursor solution spin-coated, an appropriate amount of methanaminium iodide (MAI) was spin-coated on PEDOT:PSS layer to form iodide concentration gradient in perovskite layer, which could accelerate the hole extraction. In addition, the pre-coated MAI could also react with the excess PbI₂ at the bottom of perovskite layer to avoid excess PbI₂ at the interface, which could reduce hysteresis [28].

In this work, we presented a simple approach to realize iodide concentration gradient in perovskite materials by pre-coating of MAI and controlling the amount of excess PbI₂ at the perovskite/HTL interfaces simultaneously, which effectively enhanced hole extraction. We systematically studied the effect of the different MAI concentration on the performance of PSCs. Consequently, with an optimal MAI concentration, the PSCs achieve a high photovoltaic performance with a PCE of 16.67%, a J_{sc} of 23.52 mA/cm², a V_{oc} of 1.01 V and a fill factor of 70.05%.

2. Experimental

2.1. Materials preparation

Methylammonium iodide (MAI, > 99.5% purity), PbI₂ (> 99.99% purity), PbCl₂ (> 99.99% purity) were supplied by Xi'an Polymer Light Technology Corp. PEDOT:PSS aqueous solution (Clevios PVP Al 4083) was purchased from Heraeus and the other chemicals solutions, including γ -butyrolactone (GBL, > 99% purity), rhodamine 101 (RhB101), dimethyl sulphoxide (DMSO, > 99.9% purity), isopropanol (IPA, 99.7% purity) were purchased from Sigma-Aldrich and used as received.

2.2. Synthesis of the MAPbI_{3-x}Cl_x perovskite precursor solution

The MAPbI_{3-x}Cl_x precursor solution was prepared by dissolving 581 mg PbI₂, 39 mg PbCl₂ and 209 mg MAI (1.24: 0.16: 1.31 M ratio) in 700 μ l GBL and 300 μ l DMSO solution. The perovskite solutions were heated at 70 °C for 12 h inside the glovebox filled with nitrogen gas.

2.3. Devices fabrication

Devices with an ITO/PEDOT:PSS/MAPbI_{3-x}Cl_x/PCBM/rhodamine101/LiF/Ag structure were prepared as follows: Etched ITO glass substrates (sheet resistance of 20 Ω \square^{-1}) was cleaned with detergent, deionized (DI) water, ethyl alcohol, acetone, and isopropanol (IPA), respectively, followed by dried in nitrogen flow and treated with UV ozone for 30 min. A thin layer of PEDOT:PSS film about 20 nm was deposited on the ITO substrate by spin-coating at 8000 revolutions per minute (rpm) for 40 s, and then annealed in ambient air at 120 °C for 15 min. After that, the substrates were moved into a glove box filled with dried nitrogen (< 1.0 (parts per million) ppm of O₂ and H₂O). A 60 μ l different concentration of MAI solution (0 mg/ml, 1 mg/ml, 4 mg/ml and 8 mg/ml, DMSO) were spin-casting on the top of PEDOT:PSS film at 4000 rpm for 40 s. A 50 μ l precursor solution of MAPbI_{3-x}Cl_x was spread onto the MAI layer. Then, the spin-coater was started at a rotation speed of 1000 rpm for 20 s and 3500 rpm for another 40 s and at 15 s before the end of spin coating program, 1 ml methylbenzene was injected onto the spinning film. Subsequently, the obtained perovskite

films were annealed at 100 °C for 15 min. Specifically, the spin-coating and heating procedures are important for realize halide concentration gradient perovskite film. After cooling, PCBM solution (23 mg/ml dissolved in CB) was spin-coated on top of the perovskite film at a rotation speed of 2500 rpm for 30 s. After that, RhB101 (0.6 mg/ml) was spin coated onto the PCBM layer at 1500 rpm for 40 s. Finally, LiF film (1.2 nm) and Ag film (100 nm) was deposited under high vacuum (< 2.5 \times 10⁻⁵ Pa) in evaporator chamber.

2.4. Devices characterizations

X-ray diffraction (XRD) patterns were acquired using a PANalytical Empyrean diffractometer equipped with Cu K α radiation (λ = 1.5406 Å). Optical absorption spectra of active layer were collected by Shimadzu UV-1800 UV-Vis spectrophotometer. The steady state photoluminescence (PL) spectra were obtained by a fluorescence spectrophotometer (Cary Eclipse, Agilent) with excitation wavelength of 530 nm. Scanning electron microscopic (SEM) images were measured by JEOL JSM-7800F field emission scanning electron microscope. The time-resolved PL measurements were carried out by a combined fluorescence lifetime and steady-state spectrometer (FLS980, Edinburgh Instruments Ltd.). Photocurrent-voltage (J-V) curves were achieved with an AM 1.5G solar simulator (Newport, 2612A) which calibrated by a silicon reference cell under a light intensity of 100 mW/cm². Electrochemical impedance spectroscopy (EIS) was investigated via an CHI660E Electrochemical Workstation (ChenHua, China) with an alternating current amplitude of 250 mV.

3. Results and discussion

We fabricated PSCs with an iodide concentration gradient and its schematic diagram of fabrication is shown in Fig. 1. For the conventional one-step spin-coating method, there is no pre-coated MAI layer. In contrast, different concentrations of MAI was pre-coated on PEDOT:PSS layer prior to the perovskite precursor solution coating for the modified one-step spin-coating method. Close to the perovskite/HTL interfaces, iodine concentration could be easily changed by the pre-coated MAI solution with different concentrations (0 mg/ml, 1 mg/ml, 4 mg/ml and 8 mg/ml). As the MAI can be dissolved in mixed solvent (DMSO/GBL 3:7) easily, the MAI film would be redissolved fleetly when the heated perovskite precursor solution dripped onto the MAI film. By this way, we could realize iodine concentration gradient near the perovskite/HTL interfaces. In the conventional one step spin-coating method, the amount of PbI₂ at the perovskite/HTL interfaces is hard to be controlled. Thus, in our modified one step spin-coating method, the MAI would help control the amount of PbI₂ at the perovskite/HTL interfaces, as the proper amount of PbI₂ can enhance the carrier generation or hole transport [29]. Fig. 1b and Fig. 1c shows the diagram of energy levels of each layer without pre-coated MAI and after pre-coated MAI, respectively. After pre-coated MAI, the bottom of the perovskite layer is full of iodine-rich region compared to the top of perovskite layer, which realize halide concentration gradient perovskite film and form the gradational energy band level. The similar mechanism for the formation of the gradational energy band level can be found in the previous report [30].

The effect of pre-coated MAI on the crystal structure was investigated by X-ray diffraction (XRD) measurements. As show in Fig. 2a, all the diffraction peaks can be assigned to the typical MAPbI_{3-x}Cl_x phase [31–33] except for the diffraction peak at 12.6° which belongs to the (001) face of PbI₂ [34]. To explore the content variation of PbI₂, the magnified PbI₂ diffraction peak was shown in Fig. 2b. It can be seen that the intensity of the diffraction peak of the (001) face of PbI₂ reduce with the increase of the concentration of MAI precursor, which supports the evidence of MAI was redissolved and took part in the reactions of perovskite crystallization. However, the intensity of the diffraction peak of PbI₂ almost has no change, when the concentration of

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