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Solar Energy Materials & Solar Cells

journal homepage: www.elsevier.com/locate/solmat

Interface studies of the planar heterojunction perovskite solar cells



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ARTICLE INFO

Article history:

Received 30 May 2016

Received in revised form

19 July 2016

Accepted 27 July 2016

Available online 10 August 2016

Keywords:

Perovskite solar cells

Electron-collection interlayer

Rhodamine 101

Ultraviolet photoelectron spectroscopy

Kelvin probe force microscopy

ABSTRACT

Planar perovskite solar cells (PSCs) with poly(3,4-ethylenedioxythiophene): poly(styrenesulfonate) (PEDOT:PSS) and [6,6]-phenyl-C₆₁-butyric acid methyl ester (PCBM) have attracted much interest because they can be fabricated by low temperature process and exhibit high power conversion efficiency (PCE). The PCE is strongly dependent on the interface properties between the PCBM layer and cathode. In this work, solution-processable material, rhodamine 101, was used as the interfacial layer between the PCBM layer and Ag cathode. These interfacial materials significantly increase the fill factor so as to the PCE of the PSCs. The optimal power conversion efficiencies are 13.2% (CH₃NH₃PbI₃) and 14.8% (CH₃NH₃PbI_{3-x}Cl_x) for the devices with rhodamine 101 as the electron collection interlayer, saliently higher than that (8.5% and 12.0%) of control devices without an electron-collection interlayer. The effect of the interfacial material is carefully examined by time-resolved photoluminescence spectroscopy (TR-PL), ultraviolet photoelectron spectroscopy (UPS), and kelvin probe force microscopy (KPFM) techniques. Decreased work-function and energy barrier, enhanced electron extraction and prolonged free carrier lifetime all contributed to the photovoltaic performance improvement.

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1. Introduction

Perovskite solar cells (PSCs) with alkylammonium lead halides like CH₃NH₃PbI₃ for light absorption have attracted considerable attention because of their low fabrication cost and high power conversion efficiency (PCE) [1–14]. PCEs over 20% have been reported recently [14]. The organic perovskite materials have high light absorption coefficient and long charge diffusion length. Two types of device structures, one with a scaffold layer and the other with a planar structure, have been reported in literature. The morphology of the perovskite layer is the key factor for the photovoltaic performance. It is quite sensitive to the experimental conditions. Both two-step sequential deposition and one-step deposition were developed for the preparation of the perovskite layer [15–22]. Compared to PSCs with a scaffold layer, the planar PSCs have a simpler cell architecture that can avoid the infiltration problems of the sensitizer and hole transport materials into the scaffold layer, thus reduce the charge recombination and improve the performance reproducibility [21–28].

Planar PSCs with organic materials for hole and electron collections can be fabricated by low temperature process, so flexible

substrates and roll-to-roll printing technology can be used. In most planar PSCs, poly(3,4-ethylenedioxythiophene): poly(styrenesulfonate) (PEDOT:PSS) acts as the hole transport material and [6,6]-phenyl-C₆₁-butyric acid methyl ester (PCBM) acts as the electron transport material, leading to high photovoltaic performance. The photovoltaic performance of planar PSCs strongly depends on the perovskite layer and the interface properties between PCBM and the cathode. For perovskite layer, optimal perovskite morphology can maximize the light absorption as well as charge generation and transport. However, perovskite thin film with poor quality can cause electrical shorting, poor charge dissociation, poor charge transport and high charge recombination [23–25]. The perovskite morphology could be controlled by the nucleation and growth of the perovskite crystals, and could be influenced by the experimental conditions, including the concentration, composition, purity and solvent of the precursor solution, deposition temperature, the surface properties of the layer underneath, and the post-coating annealing conditions [25–30].

On the other hand, the interface properties could be affected by energy mismatch between PCBM and electrode, as well as the potential reaction between decomposed products of perovskite (like I₂) and electrode etc. Although aluminum (Al) with a low work function of 4.1 eV was generally used as the cathode of PSCs, it can be easily oxidized and corroded by the perovskite layer during electrical test in ambient conditions. As a result, the

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photovoltaic performance degrades rapidly. When a stable metal like silver (Ag) is used to replace Al as the cathode, the performance stability and reproducibility of the devices could be greatly improved. However, there is an energy offset between the PCBM layer and Ag, because the work function of Ag is 4.6 eV, which mismatches with the lowest unoccupied molecular orbital (LUMO) of PCBM (−4.0 eV). Introducing an interlayer can relieve this problem and even give rise to Ohmic contact. Moreover, potential reaction between Ag and decomposed products of perovskite could be avoided, which enhance the device stability. Previously, lithium fluoride (LiF), bathocuproine (BCP), and fullerene (C₆₀) have been investigated as the interfacial material for electron collection [31–34]. However, these materials are processed by thermal deposition, which increases the fabrication cost, makes the fabrication process complicated, and is not compatible with roll-to-roll printing technique. Thus, solution-processable interfacial materials have the potential application in PSCs. Recently, several solution processed interlayers have also been used in the PSCs, such as fullerene surfactant (Bis-C60) [8], Polyelectrolyte (PEIE) [35], amino-functionalized polymer (PN4N) [36,37], perylene–diimide derivative (PDINO) [38], Crown-ether functionalized fullerene (PCBC) [39], 2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline (BCP) [40] etc. It was found that the interface modification layer can lower the electrode work-function by introducing interfacial dipole, which increases the electrical potential across the device. Hence, the charge carrier generation and extraction efficiency enhanced to produce high device performance. However, the effect of interface modification on electronic transport and charge carrier dynamics has not been fully understood. In order to further enhance device performance, it is desirable to understand the mechanism of interface engineering.

In this work, the solution-processable materials, rhodamine 101, were chosen to modify the interface between PCBM and Ag. They can greatly enhance fill factor (FF) of the PSCs. FF is only 0.53 for the control cells without an interfacial layer, and it increases to 0.75 for the devices with rhodamine 101 as the interfacial layer. Correspondingly, the PCE is improved from 8.5% to 13.2% (with rhodamine 101). Time-resolved photoluminescence spectroscopy (TR-PL), ultraviolet photoelectron spectroscopy (UPS), and kelvin probe force microscopy (KPFM) were utilized to probe the interface between PCBM and Ag electrode. It was found that an ohmic contact formed between the PCBM layer and Ag due to the surface dipole caused by rhodamine 101 layer, which improved the electron extraction, lowered the series resistance, and thus enhanced the device performance.

2. Experimental section

2.1. Materials

Patterned ITO glass substrates (10 Ω/□) were supplied by NSG group. PEDOT:PSS (Clevios P VP Al 4083) was purchased from Heraeus Holding GmbH. Methylammonium iodide (MAI) was obtained from Dyesol Ltd. and 1-Material Inc. [6,6]-phenyl-C₆₁-butyric acid methyl ester (PCBM) was bought from Nano-C Inc. Other materials, including lead (II) iodide (PbI₂, 99% purity), lead (II) chloride (PbCl₂, 99.999% purity), dimethyl sulfoxide (DMSO, anhydrous, ≥ 99.9% purity), γ-butyrolactone (GBL, ≥ 99% purity), rhodamine 101, LiF (≥ 99% purity), chlorobenzene (anhydrous, 99.8% purity) and isopropanol (IPA, anhydrous, 99.5% purity), were supplied by Sigma-Aldrich. All materials are used without further purification.

2.2. Fabrication and characterization of perovskite solar cells

ITO glass substrates were cleaned sequentially in detergent, deionized water, acetone, and isopropanol by sonication for 20 min. After drying under N₂ stream, substrates were further treated with UV-ozone for 15 min. A PEDOT:PSS layer with a thickness of ~40 nm was prepared by spin coating Clevios P 4083 on ITO substrates at 5000 rpm for 1 min and subsequently annealed at 140 °C for 15 min in air. The substrates with PEDOT:PSS were then transferred into a glove box filled with highly pure N₂. The perovskite layer was formed by spin coating a solution consisting 1.4 M PbI₂ (or 1.26 M PbI₂+0.14 M PbCl₂) and 1.35 M MAI in cosolvent of DMSO:GBL (3:7 vol ratio) at 1000 rpm for 20 s, and then at 5000 rpm for 60 s. After 45 s of the start of the spin coating, 200 μl toluene was dripped. Then, they were annealed at 100 °C for 15 min or 20 min. The thickness of the perovskite thin films was around 260 nm. The PCBM layer with a thickness of about 55 nm was deposited by spin coating a chlorobenzene solution of 20 mg/mL PCBM at 2000 rpm for 40 s. The interfacial layer was prepared on PCBM by spin coating IPA solution of 0.05 wt% rhodamine 101. The devices were completed by thermally deposition a layer of 100 nm-thick Ag in a vacuum of < 1 × 10^{−6} Torr. Each device had an area of 0.11 cm². The perovskite solar cells were encapsulated with cover glass slides in the glove box. The photovoltaic stability of PSCs was investigated by storing the encapsulated devices in ambient conditions for 24h. The photovoltaic performance of the PSCs was tested in air with a computer-programmed Keithley 2400 source/meter and a Newport's Oriel class A solar simulator, which simulated the AM1.5 sunlight with energy density of 100 mW/cm² and was certified to the JIS C 8912 standard. IPCEs of PSCs were measured with a 300 W Xenon Lamp (Oriel 6258) and a Cornerstone 260 Oriel 74125 monochromator.

2.3. Material characterization

X-ray diffraction (XRD) patterns were acquired using a Bruker D8 Advance XRD Instrument. UV–vis Absorption spectra were recorded with a Shimadzu UV-1800 spectrophotometer. Scanning electron microscopy (SEM) images were obtained with a Zeiss Supra-40 SEM. Atomic force microscopy (AFM) images were acquired using a Veeco NanoScope IV Multi-Mode AFM operated in tapping mode. Film thickness was determined by a surface profilometer (KLA Tencor, Alpha-Step IQ). Kelvin probe force microscopy (KPFM, noncontact) (Electric-Lever, Olympus, Japan) was performed with Pt-coated tip and applied a 3 V DC voltage under a lift height of 50 nm.

3. Results and discussion

The crystallinity of the CH₃NH₃PbI₃ films could affect the efficiency of charge generation, charge transport, and exciton diffusion length [41]. The CH₃NH₃PbI₃ thin films prepared with toluene dripping was investigated by X-ray diffraction (XRD) and UV–vis spectroscopy. As shown in Fig. 1a, the CH₃NH₃PbI₃ thin film has strong diffraction patterns at 14.2°, 28.5°, and 31.9°. These XRD patterns are assigned to (110), (220), and (310) of CH₃NH₃PbI₃, respectively. The diffraction pattern at 12.7° originates from PbI₂ because of the excess PbI₂ over CH₃NH₃I (PbI₂:CH₃NH₃I=1.4:1.35). The beneficial effect of excess PbI₂ in the perovskite thin film has been reported in several papers, and this is contributed for the high performance of the devices. All the XRD patterns of CH₃NH₃I disappear for the perovskite film, indicating that all CH₃NH₃I converts into CH₃NH₃PbI₃. The CH₃NH₃PbI₃ films were also characterized by UV–vis spectroscopy (Fig. 1b). The thin film has strong

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