Materials Science in Semiconductor Processing 87 (2018) 92–99

Contents lists available at ScienceDirect

Materials Science in Semiconductor Processing

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

Improving perovskite/carbon interfacial contact in carbon based perovskite solar cells by changing two-step spin coating sequence

Ali Mashreghi*, Kiamehr Maleki

Department of Materials Science and Engineering, Shiraz University of Technology, Shiraz 71555-313, Iran

ARTICLE INFO

Keywords:
Perovskite solar cell
Carbon electrode
Interfacial contact

ABSTRACT

Hole-transport-material free and carbon based perovskite solar cells (PSCs) are promising low-cost and highly-efficient solar cells. It has been reported in literature that incorporation of carbon black (CB) nanoparticles in carbon layer improved perovskite/carbon interfacial contact and, as a result, increased power conversion efficiency (η). In the present work, an alternative route was proposed and it was evaluated. In conventional fabrication route, carbon paste is coated on two-step deposited perovskite layer, but in proposed alternative route, carbon paste is first deposited on PbI_2 layer and then CH_3NH_3I solution is infiltrated through it to convert PbI_2 to perovskite. PSCs fabricated by conventional and alternative routes, using a carbon paste which didn't contain CB nanoparticles, showed average η of 4.3% ± 1.0% and 7.7% ± 1.6%, respectively. It was demonstrated that improvement of perovskite/carbon interfacial contact, due to protrusion of perovskite crystallites between graphite flakes, was the main reason of this efficiency enhancement. PSCs fabricated by conventional route and by using carbon paste, which contained CB nanoparticles, showed η of 5.9% ± 1.3%.

1. Introduction

First application of organo-lead halide perovskite ($CH_3NH_3PbX_3$, X = Cl, Br, I) in third-generation solar cell was reported by Miyasaka et al. in 2009 [1]. They used $CH_3NH_3PbI_3$ and $CH_3NH_3PbBr_3$ as quantum dot sensitizers in liquid-state dye sensitized solar cell (DSSC) with power conversion efficiencies (η) of 3.81% and 3.13%, respectively. Afterwards, Park et al. [2] used organo-lead halide perovskite in solid-state DSSC and obtained η of 6.5%. Since then, solid-state DSSC based on perovskite light absorber was termed "perovskite solar cell (PSC)" and its efficiency was drastically improved up to more than 20% [3–5]. The main advantages of organo-lead halide perovskite light absorber are direct band gap, high light absorption coefficient, ambipolar charge transport and its solution based processing, which make it suitable for low-cost and high-efficiency third generation photovoltaic [6,7]. However, two main limitations exist, which must be overcome before commercialization of PSCs. One limitation is expensive metals used as counter electrode, e.g. gold and silver, which must be deposited by complicated and expensive vacuum deposition techniques. The second limitation is high cost and low stability of hole-transport-materials (HTMs) used for high efficiency PSCs.

To overcome the above mentioned limitations, Han's group proposed a carbon counter electrode for HTM-free PSCs, with η of 6.64%, in 2013 [8]. In later works [7,9–13], the efficiency was further increased. This type of PSC has fully printable porous layers of TiO_2/ZrO_2/Carbon, which is infiltrated by $CH_3NH_3PbI_3$ precursor solution to form perovskite within pores. However, a challenge of this PSC type is incomplete infiltration of perovskite precursor solution through thick mesoporous layers.

Another fabrication route is direct deposition of low temperature carbon paste on pre-deposited perovskite layer [14–18]. In reference [18], carbon paste was prepared by mixing graphite powder with composite solution of butadiene-styrene rubber and ethyl cellulose in ethyl acetate. The prepared paste was printed on perovskite layer to form carbon counter electrode. It was suggested that unlike evaporated metal counter electrode, which has quite good contact with perovskite layer, the graphite flakes have poor contact with it. So, this decreased the PSC efficiency due to the increase of hole transfer resistance across perovskite/counter electrode interface. To improve perovskite/counter electrode interfacial contact, carbon black (CB) nanoparticles were added to carbon paste. It was shown that CB nanoparticles filled the pores between graphite flakes and perovskite layer and, as a result, power conversion efficiency was increased.

It seems that another way of improving the contact between perovskite and graphite flakes is protruding perovskite crystallites between graphite flakes. In this work, to realize this idea, perovskite layer was

* Corresponding author.
E-mail address: mashreghi@sutech.ac.ir (A. Mashreghi).

https://doi.org/10.1016/j.mssp.2018.07.014
Received 12 February 2018; Received in revised form 25 May 2018; Accepted 7 July 2018
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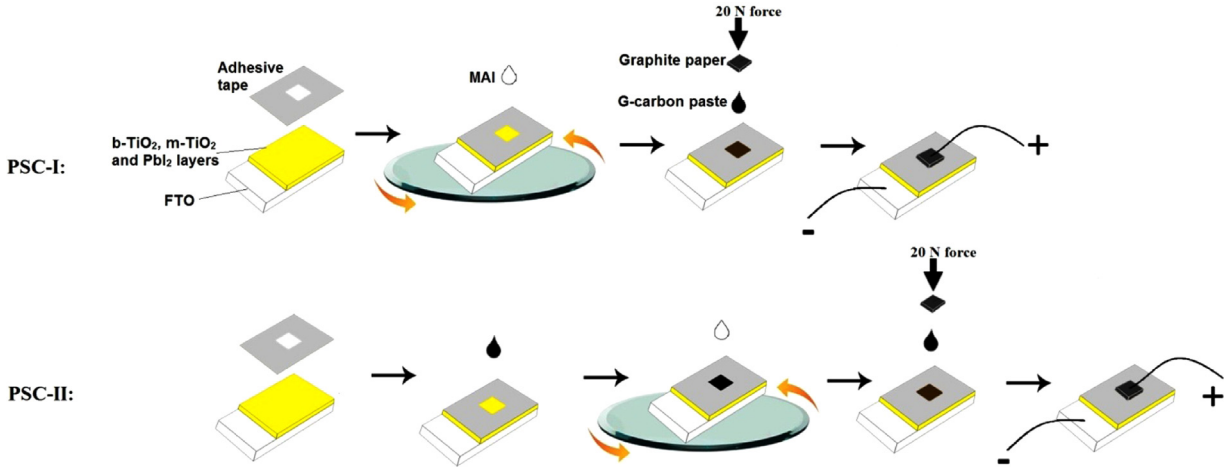

Fig. 1. Schematic illustration of fabrication steps of PSC-I and PSC-II.

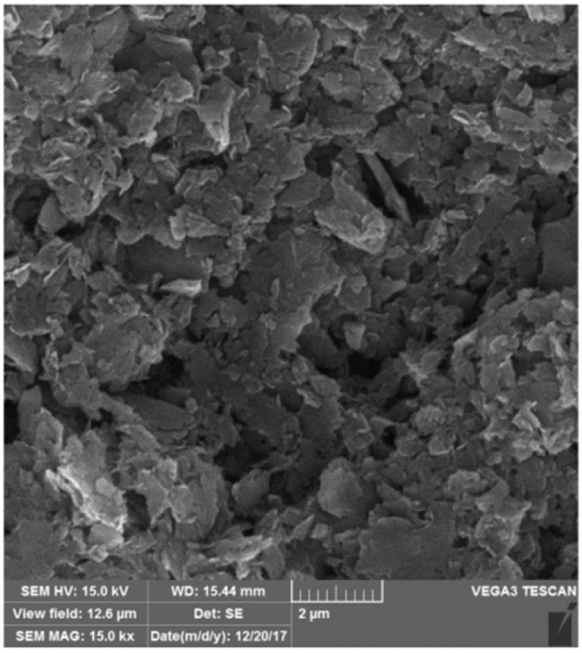

Fig. 2. Top-view SEM image of carbon layer.

formed by a modification of two-step spin coating method. In conventional two-step deposition route, similar to that of reference [18], perovskite layer is formed and carbon layer is deposited on it. In modified and our proposed route, the carbon layer is deposited on PbI_2 layer and then infiltrated by CH_3NH_3I solution to convert PbI_2 to $CH_3NH_3PbI_3$. We show that this route can improve perovskite/counter electrode interfacial contact, comparable to or even better than that obtained by incorporation of CB nanoparticles in carbon layer.

2. Experimental method

2.1. Materials

Fluorine doped tin oxide (FTO) coated glass substrates with 15 Ω/□ sheet resistance were purchased from SharifSolar. Titanium dioxide nanoparticle slurry was prepared by mixing titanium dioxide nanoparticle paste (SharifSolar) with ethanol (Merck) in 1:3.5 weight ratio. Lead iodide and titanium tetra-isopropoxide (TTIP) were purchased from Sigma-Aldrich. N,N-dimethylformamide (DMF), iso-propyl alcohol (IPA) and ethyl acetate were purchased from Dae-Jung. Methylammonium iodide (MAI) was purchase from SharifSolar. Graphite powder (150–200 mesh) and CB nanoparticles (50 nm) were purchased from SinChem and US-nano, respectively. Butadiene-styrene rubber and ethyl cellulose were obtained from Takhte-Jamshid Petrochemical Co. and SharifSolar, respectively.

Ethyl acetate, DMF and IPA were dried before use, but other chemicals were used as received, without further purification.

2.2. Preparation of carbon paste

Carbon paste was prepared according to reference [18]. 0.025 g butadiene-styrene rubber (2.5 wt%) and 0.025 g ethyl cellulose (2.5 wt %) were dissolved in 20 ml ethyl acetate. Then, 0.95 g graphite powder was added to the obtained solution and the mixture was milled for 18 h. Glass balls were used as grinding medium. Hereafter, this paste will be named "G-carbon paste". Moreover, for comparison, another paste was prepared, similar to G-carbon paste, but by adding 0.2 g CB nanoparticles (20 wt%, together with 55 wt% graphite powder). This paste will be named "CB-carbon paste".

2.3. Solar cell fabrication

FTO substrates were ultrasonically cleaned in aqueous detergent, distilled water, acetone and ethanol baths, each for 5 min. TiO_2 blocking layer (b-TiO_2) was deposited on FTO by spin coating a mild acidic solution of TTIP at 2000 rpm for 30 s. The formed layer was dried at 120 °C for 20 min and it was annealed at 500 °C for 1 h. Mesoporous TiO_2 (m-TiO_2) layer was deposited by spin coating TiO_2 slurry at 5000 rpm for 30 s. After drying at 120 °C for 10 min, it was sintered at 500 °C for 30 min. Perovskite layer was deposited by two-step spin coating method [19], but through two different routes. In one route, 1 M PbI_2 solution was spin coated on m-TiO_2 layer at 2000 rpm for 40 s and it was dried at 80 °C for 10 min. To define the effective area of solar cell, the PbI_2 layer was masked with an adhesive tape which had a square window of 0.14 cm^2. Then, 6 mg/ml MAI solution in IPA was spin coated on PbI_2 layer at 4000 rpm for 20 s, after loading time of 3 min. The masked yellow PbI_2 layer was remained unchanged and color of unmasked PbI_2 region was changed from Yellow to dark brown. After spinning, perovskite layer was annealed at 100 °C for 5 min. Then, G-carbon paste was drop casted on perovskite layer through the

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