Electrochimica Acta 284 (2018) 253-259

Contents lists available at ScienceDirect

Electrochimica Acta

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

Efficient planar heterojunction perovskite solar cells employing a solution-processed Zn-doped NiO_X hole transport layer



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ARTICLE INFO

Article history: Received 16 April 2018 Received in revised form 17 July 2018 Accepted 23 July 2018

Keywords: Metal doping NiO_X Zn Perovskite solar cells Planar heterojunction

ABSTRACT

Optimal interface of planar heterojunction perovskite solar cells determines efficient charge transport and collection for achieving high power conversion efficiency. Nickel oxide (NiO_X) is one of the most well-known hole transport materials for perovskite solar cells because of its wide band gap, *p*-type transport characteristic, good chemical stability and high optical transparency. However, pristine NiO_X possesses unsatisfactory electrical properties such as high surface trap density or low electrical conductivity which could deteriorate the device performance. We thus adopted Zn as a dopant to employ the Zn-doped NiO_X as a hole transport layer in the planar heterojunction PSCs as considering that not only Zn has a similar atomic size with Ni but also Zn forms highly crystalline oxide materials. As a result, 5% Zndoped devices achieved a power conversion efficiency up to 13.72% with improvements of a V_{OC} by 4.0%, a J_{SC} by 16.7% and a fill factor by 8.9% as compared to the un-doped NiO_X counterpart.

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1. Introduction

In the past decade, perovskite semiconductors have rapidly emerged as a promising photovoltaic material with high power conversion efficiency (PCE) of the solar cell devices [1-5]. State-ofthe-art certified PCEs of the perovskite solar cells (PSCs) have achieved > 22% by integrated efforts in the field of materials engineering and device optimization. The high performance of PSCs in a few years demonstrates the potential of the PSCs among the solar cell technologies [6-8]. The breakthrough of PSCs is attributed to the following attractive properties: an appropriate band-gap range between 1.2 and 2.3 eV, an excellent light absorption coefficient (>100 cm⁻¹ at 550 nm), a long charge-carrier lifetime and excellent charge transport properties [9,10]. The low materials cost and solution processing of the PSCs have a bright future as a viable alternative in the field of solar energy conversion technologies. The planar heterojunction PSCs with p-i-n structure received considerable attention in recent years due to their simple device architecture, straightforward solution processing and tunable device performance using various interfacial materials [11-16]. More importantly, they have shown not only less hysteresis phenomena

* Corresponding author. E-mail address: wodndwjd@khu.ac.kr (J.W. Jung). than the counterpart with n-i-p structure, but also high environmental stability of the devices [17]. In addition, facile printing technique at low temperature of afforded the p-i-n devices to be suitable for the flexible devices [18–21].

In order to improve the planar heterojunction PSCs, interfacial optimization is very critical because charge transport and collection is mainly promoted by the interfaces of the devices. In particular, hole transport layer (HTL) for PSCs determines not only short circuit current density (I_{SC}) from charge carrier transport/collection but also open circuit voltage (V_{OC}) from the minimized energy losses. Several types of *p*-type semiconductors such as poly(3,4ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS). V₂O_x, WO₃, Cu₂O, CuSCN, CuI, have been used in planar heterojunction PSCs to improve the device performance [22-26]. Nickel oxide (NiO_X) is another hole transport layer which is widely used in electronic devices. Large band gap, p-type charge transport characteristic and deep valence band of NiO_X deliver appropriate optoelectronic properties for efficient PSCs. These advantages of NiO_X for planar heterojunction PSCs leaded the comparable PCEs to the counterpart with *n-i-p* structure. However, pristine NiO_X suffers from unsatisfactory electrical properties or surface trap sites, which could deteriorate the device performance and stability. Surface treatment or passivation has been used to improve these problems of NiO_X for using in PSCs. Metal doping is another viable way to improve the electrical properties of NiO_X. Jen and co-workers have



reported that Cu-doping of NiO_X significantly improved the electrical properties, leading to enhanced PCE of the planar heterojunction PSCs [27]. Most recently, Chen et al. demonstrated that Csdoped NiO_X exhibits better electron conductivity and higher work function, resulting in 19.4% efficiency [28]. Other recent works also demonstrated that doping of NiO_X is effective method to improve the NiO_X-based PSCs [29,30].

With regard to optimal doping of NiO_X, we herein report Zndoped NiO_X for solution-processed HTL in planar heterojunction PSCs. The effect of Zn-doping of NiO_X was studied by investigating the morphology, optoelectronic properties, carrier dynamics and energy level variations of Zn-doped NiO_X thin film. Finally, we analysed the influence of Zn-doping of NiO_X on the photovoltaic performance of planar heterojunction. As a result, the 5% Zn-doped devices exhibited overall PCE up to 13.72% that is a 24%-improved value for the device using pristine NiO_X as HTL (improvements for a V_{OC} by 4.0%, a J_{SC} by 16.7%, a fill factor (FF) by 8.9%). Moreover, Zndoped NiO_X allowed improved hysteresis for J-V measurement and stabilized PCE, demonstrating that Zn is an effective dopant for NiO_X of planar heterojunction PSCs.

2. Experimental section

2.1. Materials

The precursor solution of Zn-doped NiO_X, 122.22 mg of Ni(O-COCH₃)₂·4H₂O (0.49 mmol) and Zn(OCOCH₃)₂·4H₂O of various ratios (0, 0.5, 1.0, 2.5, 5.0 mol%) were dissolved in 2-methoxyethanol (6.3 mL) containing a small amount of monoethanolamine (30 µl). After the solution was stirred at room temperature for 2 h, a homogeneous precursor solution was formed and then it was filtered by PTFE-D filters with a pore size of 0.2 µm. The perovskite precursor solution was prepared as reported elsewhere [31]. Pbl₂ (461 mg), CH₃NH₃I (159 mg) and DMSO and (78 mg) (1:1:1 M ratio) was mixed in 600 mg of DMF solution at room temperature with vigorous stirring for 1 h, followed by filtering using PTFE-D filter with a pore size of 2 µm.

2.2. Fabrication of thin-film perovskite solar cells

The ITO-coated glass substrates $(20 \Omega/sq)$ were cleaned sequentially with water, acetone, and isopropanol under sonication for 30 min, and then the substrates were treated with UV-O₃. After the Zn-doped NiO_x layer was formed by spin-coating onto the ITOcoated glass (3000 RPM for 60 s), the substrate was annealed in ambient atmosphere for 1 h at 340 °C. After the substrates were cooled to RT, the perovskite layer was formed by spin-coating the precursor solution at 4000 rpm for 25 s. After 7 s, 400 µL of chlorobenzene was poured on top of the substrates during spin-coating. After thermal annealing at $100 \degree C$ for $10 \min$, the PC₆₁BM $(15 \text{ mg mL}^{-1} \text{ in chlorobenzene})$ and bis-C60 surfactant (2 mg mL⁻¹) in isopropyl alcohol) were then sequentially deposited by spincoating at 1500 rpm for 60 s and 3000 rpm for 60 s, respectively [32]. Finally, 100 nm of Ag was evaporated under high vacuum $(<5 \times 10^{-6}$ Torr). The device area was defined by the shadow mask as 10 mm².

2.3. Characterization

Elemental investigation was carried out using X-ray Photoelectron Spectroscopy (K Alpha, Thermo Science). Absorption and transmittance measurements were carried out using spectrometer (Cary 100, Agilent). Thin film morphology was observed using an atomic force microscope (CoreAFM, Nanosurf) in tapping mode and scanning electron microscopy (S-4800, Hitach). Work function of NiOx thin films were measured by Kelvin Probe (SKP5050, KP Technology) by using Au film as a reference. The J–V curves were obtained from parameter analyzer (4200-SCS, Keithley) under AM 1.5 G (100 mW cm⁻²) illumination, which was calibrated using a

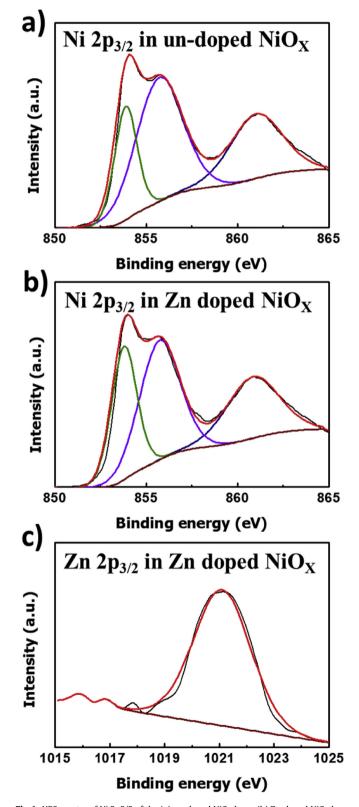


Fig. 1. XPS spectra of Ni 2p3/2 of the (a) un-doped NiO_X layer, (b) Zn-doped NiO_X layer and (c) Zn 2p3/2 of the Zn-doped NiO_X.

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