



ZnO nanowalls grown at low-temperature for electron collection in high-efficiency perovskite solar cells



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ABSTRACT

This paper reports on the fabrication of ZnO nanowalls for use as an electron collecting layer (ECL) in $\text{CH}_3\text{NH}_3\text{PbI}_3$ perovskite solar cells (PSCs). Two-dimension ZnO nanowalls were grown using a low-temperature chemical bath method with a thin Al film as a seed layer. We compared electron collecting layers based on nanowalls and sol-gel derived ZnO thin film in PSCs and sought to identify the mechanism underlying the collection of electrons. The proposed ZnO nanowalls achieved a fill factor significantly higher than that of ZnO thin films, which translated into a remarkable improvement in power conversion efficiency, reaching 13.6% under AM 1.5G illumination.

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1. Introduction

Perovskite based solar cells (PSCs) have been attracting considerable attention [1,2], since Miyasaka et al. first incorporated perovskite semiconductors in photovoltaic devices in 2009 [3]. By 2012, several groups had made remarkable progress in boosting the power conversion efficiency (PCE) of perovskite solar cells from 4% to over 10% [4,10]. The highest PCE recorded from these devices exceeded 20% [11]. Typically, the device architecture of PSCs includes a mesoporous TiO_2 film as an electron collection layer (ECL); however, these devices tend to have low electron mobility ($0.1\text{--}4 \text{ cm}^2/\text{V s}$) [12], and must be fabricated using complex methods at high temperatures. ZnO exhibits crystallization at low temperature process and enables electron mobility of up to several hundred ($110\text{--}138 \text{ cm}^2/\text{V s}$) [13–15]. ZnO thin films can be prepared using a simple solution process that does not require expensive equipment or extreme growth conditions. ZnO thin films have recently been introduced into planar perovskite solar cells to function as an efficient ECL in PSCs [16–22].

Many ZnO nanostructures with one-dimensional (1D) and two-dimensional (2D) morphologies, such as nanorods, nanobelts, nanowalls and nanoflowers, have been synthesized [23–26]. ZnO nanowalls (NWs) provide a 2D network and surface area larger

than that of ZnO nanorods [27]. NWs have considerable application potential in energy-storage devices [28], field emission devices [29], and biological sensors [30,31]. The application of ZnO nanostructures in PSCs has attracted considerable attention as a substitute for mesoporous TiO_2 nanostructures in conventional PSCs [32–34]. However, PSCs based on ZnO nanostructures present a low fill factor (FF). Furthermore, no previous study has reported on the use of ZnO NWs as an electron collection layer in PSCs.

In this study, we developed a $\text{CH}_3\text{NH}_3\text{PbI}_3$ PSCs using ZnO NWs as an electron collection layer to improve the FF and thereby enhance the PCE of the resulting PSCs. ZnO NWs were deposited on an indium tin oxide (ITO)/glass substrate using an inexpensive aqueous solution route at low temperature. Through the study of charge carrier dynamics in $\text{CH}_3\text{NH}_3\text{PbI}_3$ layers deposited on ZnO NWs, we observed rapid electron behavior highly conducive to the efficient collection of electrons. The performance of the proposed device and underlying electronic mechanism are detailed in the following.

2. Experimental

ZnO nanostructures were grown on an ITO/glass substrate using a chemical bath method [35] using an evaporating layer of Al (c.a. 20 nm) with a precursor solution of zinc acetate dehydrate (0.02 M) and hexamethylenetetramine (0.02 M). Following the growth of NWs at 80 °C for 6 h, the samples were removed from the solution and cleaned with DI water and dried at 60 °C. A sol-

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gel ZnO precursor solution (0.5 M) was prepared by combining monoethanolamine (MEA) and zinc acetate $[Zn(CH_3COO)_2 \cdot 2H_2O]$ at a molar ratio of 1:1 in 2-ethoxyethanol followed under stirring over a period of 24 h. ZnO thin films were spin-coated (5000 rpm) onto an ITO/glass substrate for 30 s and then annealed at 150 °C for 20 min. The $CH_3NH_3PbI_3$ precursor solution (1.25 M) was prepared by combining PbI_2 (Aldrich) and CH_3NH_3I (Ruilong Inc., Taiwan) in a mixed solvent of DMSO and GBL (with volume ratio of 1:1) under stirring for 3 h at 60 °C. A layer of perovskite $CH_3NH_3PbI_3$ was spin-coated (5000 rpm) onto the samples over a period of 30 sec and subsequently treated with toluene drop-casting [36] over ITO substrates coated with ZnO NWs and a ZnO thin film before undergoing annealing at 100 °C for 5 min. Spiro-OMeTAD was then deposited by spin-coating (4000 rpm) for 30 s for use as a hole transport layer. The spiroOMeTAD (2,2',7,7'-tetraakis[N,N-di(4-methoxyphenyl)amino]-9,9'-spirobifluorene) precursor solution was prepared by dissolving 80 mg spiroOMeTAD, 28.5 μL 4-tert-butylpyridine, and 17.5 μL lithium bis(trifluoromethyl-sulphonyl)imide solution (520 mg in 1.0 mL acetonitrile) in 1 mL chlorobenzene. Finally, the sample was transferred to a vacuum chamber for the deposition of an Ag electrode (100 nm). XRD data were collected using a Rigaku RINT 2100 diffractometer and the surface morphology was studied using a field-emission scanning electron microscope (FE-SEM, Hitachi S4100 and SU8000). The J-V characteristics of the cells were obtained using a Keithley 2400 source measuring unit under a simulated AM1.5G light (Wacom solar simulator) at 100 $mW\ cm^{-2}$. All the J-V curves, including the forward and reverse scan, were measured using a voltage step of 0.01 V and a delay time of 200 ms (i.e. a scanning speed of 0.05 V/s). The incident photo-to-current

conversion efficiency (IPCE) was measured using the QE-R3011 measurement system in air (Enlitech Inc., Taiwan).

3. Results and discussion

3.1. Characterization results

Fig. 1(a) presents photographic images of bare ITO, ZnO thin film produced using the sol-gel method, and transparent NW films deposited on patterned ITO substrates. **Fig. 1(b)** presents an SEM image of the ZnO NWs with the inset showing an SEM image in cross-section. As shown in the SEM images, the large number of porous (sponge-like) ZnO nanostructures resulted in a very large surface area. The thickness of the film and sidewalls was measured at c.a. 320 and 30–80 nm, respectively.

Fig. 1(c) presents a transmission electron microscopy (TEM) image of the proposed ZnO NWs, the sidewall of which is composed of uniformly sized ZnO nanocrystals with grain size of less than 10 nm. Selected area electron diffraction (SAED) patterns (**Fig. 1(d)**) of the ZnO NWs reveals seven diffraction rings, which perfectly match the index positions associated with bulk ZnO ceramics, which indicate the crystallinity of the ZnO NWs with a hexagonal wurtzite structure.

Fig. 2 presents the dense, uniform surface morphology and large grain size of $CH_3NH_3PbI_3$ deposited over the ZnO thin film and NWs. The smaller number of pinholes was reduced (in the cross-sectional view) in the $CH_3NH_3PbI_3$ deposited on the ZnO NWs which can provided a scaffold, similar to a mesoscopic TiO_2 configuration, for $CH_3NH_3PbI_3$ growth and carrier-collection [37].

After charge transfer processes, we carried out electrochemical impedance spectroscopy on $ITO/CH_3NH_3PbI_3/P3HT/Ag$ and $ITO/ZnO/CH_3NH_3PbI_3/P3HT/Ag$ devices (Figure 4) as a powerful characterization technique for studying the surface charge transfer properties of photovoltaic cells [38], and has recently been used to investigate the charge transport and recombination dynamics in the perovskite solar cells [39–43]. **Figure 4(a)** shows the current density plots of devices both with and without ZnO thin film. ZnO nanowall

ITO ZnO thin film ZnO nanowall

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