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Enhanced efficiency via blocking layers at photocathode interfaces in cobaltmediated tandem dye-sensitized solar cells



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ABSTRACT

The efficiency of tandem pn-dye-sensitized solar cells (pn-DSCs) has been limited by the poor performance of the available p-DSCs. A facile approach of introducing a compact NiO blocking layer into an active NiO photocathode network of p-DSCs and pn-DSCs is presented to enrich the power conversion efficiency (PCE) using a $Co^{2+/3+}$ redox mediator. Two photoelectrodes sensitized with different dyes (SQ for n-DSC and P1 for p-DSC) have a complementary nature in absorbing solar irradiation at different wavelengths. The constructed pn-DSC demonstrates an enhanced photovoltage compared to n-DSC, resulting in an overall PCE of 1.486%. This PCE is further improved to 1.913% by the addition of an optimized NiO blocking layer to the p-DSC; this was prepared *via* spin-coating with a nickel acetate precursor solution. All photovoltaic parameters were significantly increased with the introduction of a blocking layer compared to a bare cell.

1. Introduction

In the coming years, dye-sensitized solar cells (DSCs) are expected to be one of the most promising photovoltaic devices as low-cost alternatives to traditional silicon-based solar cells (Hagfeldt et al., 2010; Ooyama and Harima, 2012). Since the pioneering work reported by O'Regan and Gratzel (1991), a growing volume of work has been dedicated to developing both n-type dye-sensitized solar cells (n-DSCs) and p-type dye-sensitized solar cells (p-DSC). However, their power conversion efficiency (PCE) continues to lag behind the efficiency of silicon photovoltaic devices (Mathew et al., 2014). One disadvantage of conventional DSCs is their inability to capture the full spectrum of solar irradiation owing to the limited spectral range of absorbance of the photosensitizers (Ning et al., 2010; Yum et al., 2011). A promising strategy is to construct a new photovoltaic architecture, tandem dyesensitized solar cell (pn-DSC), to utilize the energy produced from the different regions of the solar spectrum. The tandem device consists of a dye-sensitized n-type semiconductor (e.g., TiO₂), a dye-sensitized p-type semiconductor (e.g., NiO), and a redox mediator between them. The combination of a photoactive anode and photoactive cathode is capable of surpassing the Shockley-Queisser limit of 33% for a single-junction device to attain 42% for a multi-junction device (He et al., 2000; Nattestad et al., 2010; Sobus and Ziolek, 2014; Vos, 1980). Another advantage of a series connected tandem solar cell is the enhancement of the photovoltage compared to each component (n-DSC and p-DSC), leading to an increment in the PCE. The maximum photovoltage for an n-DSC (Voc_n) is determined by the difference in the energy level of the conduction band of TiO₂ and the redox potential of the electrolyte, whereas the maximum photovoltage obtained for a p-DSC (Voc_p) is determined by the energy difference between the redox potential of the electrolyte and the valence band of NiO. Theoretically, the photovoltage of a tandem device (Voc_{pn}) is the sum of the photovoltage obtained from each component (Voc_{pn} = Voc_n + Voc_p) (Hagfeldt et al., 2010; He et al., 2000; Mishra et al., 2009; Nattestad et al., 2010; Ooyama and Harima, 2012; Ornso et al., 2015). Furthermore, the platinized counter electrode in traditional DSCs is replaced by a photocathode; hence, the overall fabricating cost of the solar cells can be considerably reduced. These advantages make tandem dye-sensitized solar cells one of the most encouraging candidates for commercial application.

Despite many attempts to develop a tandem structure, the PCE of a pn-DSC continues to be considerably less than that of an n-DSC, owing primarily to the poor performance of the p-DSCs (Odobel et al., 2012; Wood et al., 2016). Therefore, an improvement in the p-type PCE is essential to enhance the efficiency of pn-DSCs. In recent years, p-DSC reports have focused on designing novel sensitizers and redox systems (Favereau et al., 2013; Gennari et al., 2014; Maufroy et al., 2015; Perera et al., 2015; Powar et al., 2013; Qin et al., 2010; Wood et al., 2014; Zhang et al., 2015), manufacturing different types of p-type semiconductors (Langmar et al., 2015; Shi et al., 2014; Sumikura et al., 2008a; Xu et al., 2014), and improving the quality of the NiO photocathode (Awais et al., 2014; Gibson et al., 2013; Lepleux et al., 2009;

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Powar et al., 2012; Sumikura et al., 2008b; Zhang et al., 2011). Among these reports, the development of an electrolyte system can significantly increase the performance of the p-DSCs, primarily attributable to an increment in the photovoltage. Recently, Udo Bach's et al. reported the highest efficiency to date for p-DSCs based on novel electrolyte systems with a remarkable Voc of 645 mV and 724 mV, providing an overall PEC of 1.3% and 2.5% (Perera et al., 2015; Powar et al., 2013). On the other hand, many groups have doped metal atom into NiO active layer to improve the photovoltaic performance of p-DSCs. Introducing metal atoms is an advantage for acquiring the desired properties, such as electrical conductivity, tuning the band edge energies, increased charge carrier transport properties, increased charge density of states and Fermi energy (Natu et al., 2012; D'Amario et al., 2014; Wang et al., 2014; Wei et al., 2016; Huang et al., 2014; Zannotti et al., 2015).

Another effective approach to improve the performance of p-type DSCs is to apply the compact NiO layer as a blocking layer between the fluorine-doped tin oxide (FTO) substrate and nickel oxide working electrode. This approach has been commonly used in n-DSCs to retard charge recombination occurring at the electrolyte/FTO interface (Cameron et al., 2005; Hart et al., 2006; Li et al., 2013; Yu et al., 2009). Compact NiO films can be fabricated by several methods including RF-magnetron sputtering, spin-coating, and atomic layer deposition (Ho et al., 2016; Perera et al., 2015; Zhang et al., 2011, 2012; Zhu et al., 2014). Among these, compact NiO layer fabrication using the spin-coating technique is an inexpensive and simple method compared to the other methods.

The design of this work was to build on our previous success with photocathode having optimized NiO blocking layer (Ho et al., 2016). The introduction of an NiO compact layer can effectively improve the electronic contact between the FTO and mesoporous NiO film and suppress the hole-electron recombination processes that occurs at the FTO/electrolyte, resulting in an increased overall photovoltaic performance (Ho et al., 2016; Perera et al., 2015). Fig. 1 presents the structure and operational principle of a tandem pn-DSC with approximate energy levels. The cell contains a photoanode (n-type) and a photocathode (p-type) with a compact NiO blocking layer in a sandwiched configuration with a Co^{2+/3+} complex as an electrolyte between them. The compact NiO blocking layer was prepared by spin-coating with a precursor solution of nickel acetate tetrahydrate followed by thermal decomposition to form a compact NiO layer onto the FTO. Finally, the mesoporous NiO film was coated onto the NiO blocking layer using a screen-printing

method. Prepared photocathode films were then applied in the p-type DSCs and tandem pn-DSCs. To the best of our knowledge, this is the first report of the application of a compact NiO layer in a tandem pn-DSC. The effect of this blocking layer on the properties of the device was scrutinized in detail, including photocurrent-photovoltage (I-V) characteristics, incident photon-to-current conversion efficiency (IPCE), and electrochemical impedance spectroscopy (EIS).

2. Experimental section

2.1. Chemical reagents

All chemicals were used as received without further purification. Nickel oxide nanopowder (NiO, 20 nm in size) was purchased from Inframat. Nickel acetate tetrahydrate (Ni(CH₃COO)₂·4H₂O), cobalt(II) chloride hexahydrate (CoCl₂·6H₂O), α -terpineol, ethyl cellulose, ethanol, acetonitrile, 2-methoxyethanol, hydrogen peroxide, 2,2'-bipyridine, lithium perchlorate (LiClO₄), and 4-*tert*-butylpyridine (*t*BP) were purchased from Sigma-Aldrich. Chloroplatinic acid hexahydrate (H₂PtCl₆) and TiO₂ paste (18-NRT) were purchased from Dyesol. Fluorine-doped tin oxide (FTO)-coated glass substrate was obtained from Pilkington (8 Ω /cm²). The organic chromophore P1 and SQ were synthesized following reported methods (Lee et al., 2014; Qin et al., 2008). The cobalt complexes [Co(bpy)₃](PF₆)₂ and [Co(bpy)₃](PF₆)₃ (bpy = 2,2'-bipyridine) were synthesized according to our previously published procedure (Lee et al., 2015).

2.2. Device fabrication

The conducting glass substrates (FTO) were cleaned sequentially with deionized water, ethanol, and acetone using an ultrasonic bath for 20 min. The compact NiO blocking layer was deposited by spin-coating 0.5 M nickel acetate tetrahydrate in 2-methoxyethanol onto the cleaned FTO glass at 3000 rpm for 30 s. This process was repeated three times to obtain films with a desired thickness. These samples were sintered at 450 °C for 30 min for thermal decomposition of the nickel acetate tetrahydrate to form a compact NiO blocking layer (Afzal et al., 1991; Hong et al., 2006; Hussein et al., 1994).

The NiO paste was prepared according to a previously reported procedure (Nattestad et al., 2010). In summary, a slurry of 6 g of NiO nanopowder in ethanol was mixed with 20 mL of 10 wt% ethanolic ethyl cellulose solution and 40 mL α -terpineol and evaporated using a



Fig. 1. Schematic diagram of tandem dye-sensitized solar cells with the molecular structures adjacent.

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