Electrochimica Acta xxx (2015) xxx–xxx

Contents lists available at ScienceDirect

Electrochimica Acta

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

Structural alternation of tandem dye-sensitized solar cells based on mesh-type of counter electrode

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ARTICLE INFO

Article history:
Received 6 December 2014
Received in revised form 17 April 2015
Accepted 17 April 2015
Available online xxx

Keywords:
Tandem dye-sensitized solar cell
Mesh counter electrode
Structural alternation
Long-term stability

ABSTRACT

Tandem dye-sensitized solar cells (DSCs) are very effective to improve light absorption characteristics and overall performance. The structure of conventional tandem DSC is an assembly of two independent DSCs. Therefore, additional TiO_2 layer, Pt film, and transparent conductive oxide (TCO) electrodes weaken incident light to the bottom cell and complicate the fabrication as compared with standard DSCs. Here, this work proposed the structural alternation of tandem DSC as a solution. Mesh type of counter electrode was inserted between top and bottom cells instead of TCO electrodes. Two photo electrodes shared electrolyte and counter electrode in this structure. High aperture ratio of mesh increased light penetration into bottom cell and led to the performance improvement. Structural alternation also simplified the fabrication. It could be fabricated like standard DSCs. After dye arrangement and TiO_2 layer of bottom cell were controlled, the photovoltaic performance of proposed tandem DSC was enhanced and it was higher than conventional tandem DSC. Finally, the long-term stability of proposed tandem DSC was secured by the control of sealing walls.

1. Introduction

Dye-sensitized solar cells (DSCs) have attracted lots of attention in the photovoltaic research field since first development in the early 1990s [1,2]. Its simple fabrication process, low manufacturing cost and short energy payback time strengthened the cost-competitiveness. Its unique characteristics such as transparency, various colors, and flexibility made DSCs specialized in the portable, wearable, and building integrated photovoltaics. Although highly efficient perovskite solar cells were recently appeared [3–5], more stable DSC is still an attractive and competitive photovoltaic device. To enhance DSC performance, highly transparent electrodes with low resistivity, conductive semiconductor oxide materials, electrolyte for better charge transportation, and sensitizers with strong absorption were researched so far [6–14]. Within a range of related researches, the improvement on the absorption of sensitizer is very important for large current and high efficiency because it is directly connected to the photo-generation. Many dyes were developed so far but only a few dyes such as N719, N749, and N3 were widely used [15–17]. There is still the demand of the strong absorption in the wide wavelength range although dye absorption was much enhanced so far. Tandem DSCs were developed to supplement insufficient absorption of single dye [18–22]. Multiple dyes of different absorption characteristics were stacked and overall absorption was enhanced and widened. The structure of conventional tandem DSC is shown in Fig. 1(a). It is an assembly of two independent DSCs. Tandem DSCs definitely harvest more photons and have higher performance than standard DSCs. However, TiO_2/dye layer, Pt film, and two TCO layers are additionally necessary. Especially, expensive Pt and TCOs are one of crucial causes of the increase in manufacturing cost. In addition, its fabrication becomes complicated. As a solution, this work proposed the structural alternation of tandem DSC shown in Fig. 1(b). In this structure, two photo electrodes share mesh type of counter electrode and redox electrolyte. Therefore, its fabrication is similar to a standard DSC. TCO-less counter electrode leads to manufacturing cost saving and the increase in incident light intensity to the bottom cell. N719 and N749 dyes were combined in this tandem DSC. In order to verify the effect of structural alternation, photovoltaic properties, cyclic voltammetry (CV), incident photon to current conversion efficiency (IPCE), and long-term stability were examined.

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http://dx.doi.org/10.1016/j.electacta.2015.04.105

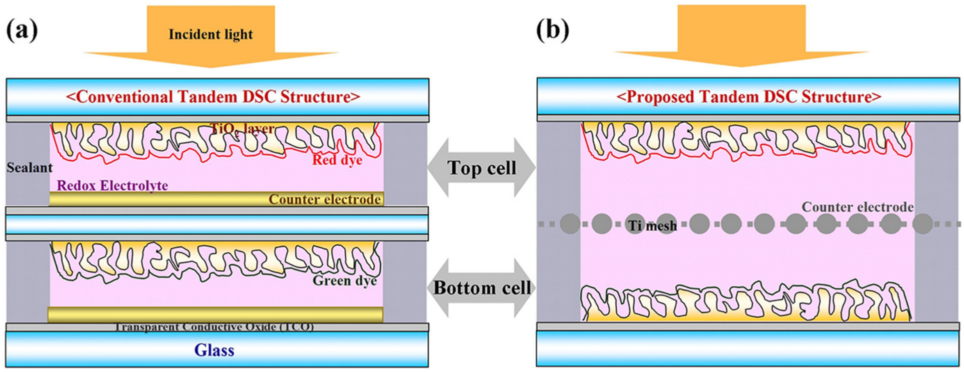

Fig. 1. Structural diagrams of (a) conventional and (b) proposed tandem DSCs.

2. Experimental

Proposed tandem DSCs were fabricated as follows. Fluorine-doped tin oxide (FTO) substrates (15 Ω/sq., Pilkington) were used for the photo electrodes. 16 × 13 mm^2 sized FTO substrates were sequentially cleaned by sonicating in acetone, ethyl alcohol, and distilled water. They were dried using a stream of nitrogen. TiO_2 paste was prepared by general procedure [23]. TiO_2 paste was uniformly coated on FTO substrate by doctor-blade method. TiO_2 layer changed to nano-porous structure after heat treatment at 450 °C for 30 min. The thickness of TiO_2 layer was about 7 μm. Nano-porous TiO_2/FTO electrodes were soaked into N719 ($RuC_{58}H_{86}N_8O_8S_2$) or N749 ($C_{69}H_{117}O_6N_9S_3Ru$) dye solution. Dye adsorption time was 16 h because our previous work verified that dye adsorption was saturated after 12 h [24,25]. Ti mesh (Unique Wire Weaving Co., Inc.) was used for the counter electrode. Mesh was uniformly weaved by Ti strings with a diameter of 76.2 μm. Its aperture ratio was 88.4% and the transmittance was enough secured. 16 × 13 mm^2 sized Ti mesh was cleaned by sonicating in ethyl alcohol. Dried Ti mesh was dipped into a 100 mM H_2PtCl_6 in isopropanol and dried at 120 °C. After that, it was sintered at 400 °C for 30 min. N719 and N749 dye-sensitized photo electrodes and Pt deposited mesh counter electrode were sealed using thermoplastic sealants (SX 1170-25, Solaronix). Sealants were put on two photo electrodes and mesh counter electrode was inserted between photo electrodes. Assembly was sealed at 100 °C by pressing. In order to find stable sealing condition, the thickness of sealants was varied from 25 to 75 μm. Redox electrolyte which consisted of 0.5 M LiI, 0.05 M I_2 and 0.5 M 4-tertbutylpyridine in acetonitrile was injected through a pre-drilled hole into the bottom electrode. The fabrication was completed after holes were sealed. In order to compare photovoltaic properties, conventional tandem DSC was also prepared as a reference. As shown in Fig. 1(a), one-sided FTO substrates were prepared for top and bottom electrodes and double-sided FTO substrate was prepared for intermediate electrodes. Nano-porous TiO_2 layer was deposited on top and intermediate FTO substrates. 10 mM H_2PtCl_6 in isopropanol was spin-coated on bottom FTO and another side of intermediate FTO substrates with a rotating speed of 3000 rpm for 60 s and sintered at 400 °C for 30 min. Three electrodes were sequentially sealed. Dye adsorption and electrolyte injection were identically conducted as described above.

All completed tandem DSCs were kept under the dark and open-circuit conditions for 24 h to allow electrolyte to penetrate into the nano-pores. Their photovoltaic properties were measured under dark and irradiated conditions. 1 sun (air mass 1.5, 100 mW/cm^2) light was irradiated and I-V (current-voltage) characteristic curves were measured by a source meter (Model 2400, Keithley Instrument, Inc.). IPCE (SM-250-P1, Bunkoukeiki) was measured in the wavelength range from 250 to 1100 nm. During irradiance and characterization, a black mask was fitted by the active area of 0.20 cm^2. CV was investigated using the electrochemical analysis instrument (SP-150, Biologic SAS). It was performed at a scan rate of 100 mV/s in the potential range from −2 to 2 V. For stability measurement, conventional and proposed tandem DSCs were continuously exposed to irradiation. Their photovoltaic properties were measured up to 1000 h.

3. Results and Discussion

In conventional tandem DSCs, short and long wavelength absorptive dyes were arranged on top and bottom cells, respectively [18–22]. However, optical path to bottom cell was changed by structural alternation of proposed tandem DSCs. Accordingly, some photons were lost by absorption, scattering, and reflection of electrolyte although its rate was not large. In addition, the performance of N719 DSC is generally higher than that of N749 DSC despite its relatively narrow absorption range [26,27]. Therefore, it was necessary to compare photovoltaic characteristics according to dye arrangement. Fig. 2 shows (a) photovoltaic properties and (b) IPCE of N749 top/N719 bottom and N719 top/N749 bottom tandem DSCs. Two tandem DSCs showed different open-circuit voltage (V_{OC}). Tandem DSC of N719 top/N749 bottom had slightly higher V_{OC} than that of N749 top/N719 bottom. N719 DSC generally has higher V_{OC} than N749 DSC. Our standard N719 and N749 DSCs had V_{OC} of 0.75 V and 0.71 V, respectively. Other researches also reported same tendency [28–30]. Consequently, V_{OC} of N719 top/N749 bottom tandem DSC had relatively high V_{OC} because its N719 top cell was fully photo-generated by 1 sun illumination. On the other hand, short-circuit current density (J_{SC}) of N749 top/N719 bottom tandem DSC was higher than that of N719 top/N749 bottom tandem DSC although standard N719 DSC had larger photocurrent than N749 DSC. It meant that N719 bottom cell had more photo-generation than N749 bottom cell. These results were verified by IPCE results in Fig. 2(b). N749 top/N719 bottom tandem DSC had high photo-generation in whole wavelength range. Especially, it had stronger absorption in long wavelength region. In other words, photons in the wavelength range over 700 nm were not enough for the photo-generation of N749 bottom cells while the photo-generation was conducted by incident photons in short wavelength. This obviously came from the change of optical path and the disturbance of electrolyte. As a result, N749 top/N719 bottom tandem DSC had larger J_{SC} and higher performance than N719 top/N749 bottom tandem DSC in spite of relatively low V_{OC}. N749 top/N719 bottom tandem DSC had 0.69 V of V_{OC}, 15.85 mA/cm^2 of J_{SC}, 0.62 of fill factor (FF), and 6.78% of efficiency while N719 top/N749 bottom tandem DSC had 0.73 V

Please cite this article in press as: H. Seo, et al., Structural alternation of tandem dye-sensitized solar cells based on mesh-type of counter electrode, Electrochim. Acta (2015), http://dx.doi.org/10.1016/j.electacta.2015.04.105