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Design and fabrication of a high performance inorganic tandem solar cell with 11.5% conversion efficiency



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ABSTRACT

Tandem solar cell is a design that combines two types of solar cells to benefit their advantages. We show a new concept for achieving highly efficient dye sensitized and quantum dot tandem solar cells. The new tandem cell further enhances the performance of the device, leading to a power conversion efficiency more than 11% under 1.5 Air Mass. To the best of our knowledge, this is the first time that the efficiency over 11 percent is achieved based on tandem solar cell. X-ray diffraction, Transmission Electron Microscopy, Scanning Electron Microscopy, Current-Voltage measurments, Intensity modulated photocurrent spectroscopy, intensity modulated photovoltage spectroscopy, Energy Dispersive X-ray spectroscopy, Brunauer-Emmett-Teller, Barrett-Joyner-Halenda and absorption spectroscopy were used to characterize the fabricated solar cells.

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1. Introduction

Typical dye-sensitized solar cells (DSSCs) have been regarded as a promising branch of study compared to conventional siliconbased photovoltaic devices because of their low cost and high environmental friendliness [1]. Up to now, weak light absorption is one of the most serious problems that has limited the efficiency of DSSCs. Titanium dioxide (TiO₂) which has limited absorption in the ultraviolet region [2] is one of the most prominent materials that are used as photoanode in DSSCs [3]. In addition, all dyes which were used in DSSCs have weak absorption for instance black dye and N719 dye [4]. Black dye absorbs visible light strongly at 340 and 640 and N719 absorbs strongly at 535 nm, but they drastically have reduced extinction coefficients in other wavelengths [5]. Till now, there have been many reports for improving efficiency of DSSC and quantum-dot (QD) DSSCs (QDSSCs). Various strategies including modifying photoanode by using 3D hierarchical architectures, mesoporous structures, nanorod, nanotube, and nanosheet [6-13], doping TiO₂ with different ions [14-16], using

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http://dx.doi.org/10.1016/j.electacta.2017.09.013 0013-4686/© 2017 Elsevier Ltd. All rights reserved. plasmonic materials (eg. gold and silver) [17,18], and making tandem solar cell. The reported tandem solar cells have different structures: 1) "n-n tandem structured DSSC" with two different dye-sensitized n-type semiconductive electrodes [19–23]; 2) "n-p tandem structured DSSC" with one dye sensitized n-type semiconductive electrode connected with a dye-sensitized p-type semiconductive electrode [24–27]; 3) dye sensitized solar cell in combination with other types of solar cells [28,29].

Here, we show a new photoanode for achieving high efficiency consisting a tandem solar cell with core-shell structure including CdSe/CdS@TiO₂ nanolayers. The outer TiO₂ shell is sensitized with black dye as in a typical DSSC. The presented novel tandem system is based on the so-called co-sensitization method in which photoanode is sensitized with both QDs and dye, simultaneously [30,31]. This helps improving the efficiency by widening the solar absorption range, increasing surface photovoltage response, making new pathways for electron transport, and decreasing electron and hole recombination [31]. However, a main problem in this system is CdS corrosion by the iodine in electrolyte or dye degradation by polysulfide electrolyte. For solving this problem, a TiO₂ or Al₂O₃ thin film layer has been introduced between CdS and the electrolyte [32–35]. In this work, the thin TiO₂ layer has been considered as both separator and charge separator layer. With this



strategy, the TiO₂ substrate which in previous reports was used underneath the quantum layer is removed. The TiO₂ layer which is located in the middle, collects the electrons from its surrounding dye and CdSe/CdS layers and transfers them to the front FTO electrode. By having lower surface trap density and decreased trapping/detraaping events, the thin layer of TiO₂ complies the easy and fast transport of the electrons and results in high efficiency of the solar cells. Moreover, using QDs and dyes as cosensitizers is very effective in increasing the photocurrent response of the final device since first, two types of antenna are absorbing the light energy for electron-hole separation and second, CdSe/CdS core play a crucial role as a barrier to prevent charge recombination in TiO₂. To the best of our knowledge, this is the first time that this high efficiency is achieved for a tandem solar cell.

2. Materials and Methods

2.1. Chemicals and characterization

Titanium (IV) butoxide (TBOT 97%), Trioctylphosphine oxide (TOPO 99%), 1-octanethiol (>98.5%), 1-octadecene (ODE, 90%), trioctylphosphine (TOP, 97%), oleylamine (OAm, 70%), sulphur powder (99.999%) and poly(methyl methacrylate) (PMMA, 350k MW) were obtained from Aldrich. Selenium powder (99.999%), cadmium nitrate tetrahydrate (Cd(NO₃)₂·4H₂O, 99.99%), myristic acid (MA, 99%), oleic acid (OLA, 90%) Cadmium oxide (CdO, 99.998%), and octadecylphosphonic acid (ODPA, 97%) were purchased from Alfa Aesar. Methoxy-polyethylene-glycol thiol PEG-SH (5000 MW) was obtained from Rapp Polymer GmbH, Tuebingen, Germany. The pore size distribution and surface area were investigated by Brunauer-Emmett-Teller (BET) and Barrett-Joyner-Halenda (BJH) analyses with a Micrometrics ASAP 2000 apparatus after degassing the samples at 200 °C for 10 h. GC-2550TG (Teif Gostar Faraz Company, Iran) was used for all chemical analyses. The specific surface areas were estimated by using the BET method, and the pore size distributions were determined with the BJH method by using the nitrogen desorption branches of the isotherms. For characterization of the products XRD patterns were recorded by a Rigaku D-max C III, X-ray diffractometer using Nifiltered Cu Ka radiation. The energy dispersive spectrometry (EDS) analysis was studied by XL30, Philips microscope. Transmission electron microscope [36] images were obtained on a Philips EM208S transmission electron microscope with an accelerating voltage of 100 kV. Solar cells were characterized by I-V (solar simulator - Luzchem Research, Inc, IVIUMSTAT - Ivium Technologies), Intensity modulated photovoltage spectroscopy (IMVS) and Intensity modulated photocurrent spectroscopy (IMPS) (IVIUM MODULIGHTMODULE - Ivium Technologies) spectra under A.M 1.5.

2.2. CdSe core synthesis

CdSe cores were synthesized according to a literature method [37]. Briefly, 560 mg ODPA, 120 mg CdO and 6 g TOPO were mixed in a 100 mL flask. The mixture was heated to 150 °C and degassed under vacuum for 1 hour. The reaction mixture was heated to 320 °C to form a colorless clear solution under nitrogen flow. After adding 2.0 mL TOP to the solution, the temperature was increased up to 380 °C, at which point Se/TOP (120 mg Se in 1 mL TOP) solution was injected into the flask. The reaction was ended by removing the heat when the CdSe core nanocrystals reached the desired size. The resulting CdSe particles were precipitated by adding acetone and were dispersed in hexane.

2.3. CdS shell growth

CdS shell growth followed a previous reported modified method [38]. Briefly, a hexane solution containing 200 nmol of CdSe QDs was dispersed in a mixture of 6 mL of OAm and 6 ml of ODE. The reaction solution was degassed under vacuum at room temperature for 1 hour and 120 °C for 20 min to completely remove the hexane, water and oxygen inside the reaction solution. After that under nitrogen flow and magnetic stirring, the reaction solution was brought up to 310°C with a heating rate of 18 °C min⁻¹. In another beaker, 0.1 mmol cadmium nitrate tetrahydrate was added to 5 mL of OAm to form Cd-oleate complexes. During the heating, cadmium oleate (Cd-oleate, diluted in 12 mL ODE) and octanethiol began to be injected dropwise into the growth solution at a rate of $3 \text{ mL} \text{ h}^{-1}$ using a syringe pump when the temperature reached 240 °C. After finishing precursor injection, 1 mL oleic acid was guickly injected and the solution was further heated at 310 °C for 60 min. A solution of 0.05 mmol sulfur powder dissolved in small amount of ODE was finally injected to the reaction flask, and then the reaction mixture was allowed to cool to 250 °C for formation of CdS shell. The resulting CdSe/CdS core/shell QDs were precipitated by adding acetone, and then redispersed in hexane.

2.4. Synthesis of Uniform Porous TiO₂ Shell Nanostructures

The uniform porous TiO_2 shell core-shell structures were prepared via a previous reported method [39]. The CdSe@CdS nanoparticles were loaded in absolute ethanol (100 mL), following by mixing with concentrated ammonia solution (0.30 mL, 28 wt%) under ultrasound for 15 min. After that 0.75 mL of TBOT was added dropwise during 5 min, and the reaction was allowed to proceed for 24 h at 45 °C under continuous magnetic stirring. The resultant products were separated and collected, and washed with deionized water and ethanol 3 times. Then, the obtained powders were dried at 100 °C for 20 h.

2.5. Solar cell fabrication

To prepare the DSSC working electrodes, the FTO glass used as current collector (Solar 4 mm thickness, 10 Ω , Nippon Sheet Glass, Japan) was first cleaned in a detergent solution using an ultrasonic bath for 15 min, and then rinsed with water and ethanol. After treatment in a plasma system for 18 min, electrophoretic deposition (EPD) was utilized to deposit CdSe/CdS@TiO2 core shells and P25 NPs-based films used in tandem cell and DSSCs, respectively. During EPD, positively charged nanoparticles are deposited on the negatively charged cleaned FTO glass (cathode) while a pure steel mesh was used as anode. The linear distance between the two electrodes was about 2 cm. Power was supplied by a Megatek Programmable DC Power Supply (MP-3005D). The applied voltage was 10–15 V. The deposition cycle was 4 and 8 times with each time of 15–20 s, and the temperature of the electrolyte solution was 25 °C. Electrolyte solution and the amount of additives are important for creation a surface with high quality. Based on our pervious works we used optimal concentrations of additives in the EPD bath [22,24,40–43] containing P25–iodine–acetone–acetyl acetone–water: P25 1 g, I_2 120 140 mg L⁻¹, acetone 48–56 mL L⁻¹, and water 20 mLL⁻¹. The particle charging in this system is achieved via adsorption of protons, which are formed by the ketoenol reaction:

 $CH_3COCH_3 \leftrightarrow CH_3C(OH)CH_2$

 $CH_3C(OH)CH_2 + I_2 \rightarrow CH_3COCH_2I + H^+ + I^-$

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