Morphological and structural study on low cost SnO2 counter electrode and its applications in quantum dot sensitized solar cells with polysulfide electrolyte

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panelG.K.R. Senadeera a b, W.I. Sandamali a b, J.M.K.W. Kumari b, T. Jaseetharan b c, Janith Weerasinghe d e, Prashant Sonar d e, V.P.S. Perera a, J.C.N. Rajendra a, N. Karthikeyan a, M.A.K.L. Dissanayake b

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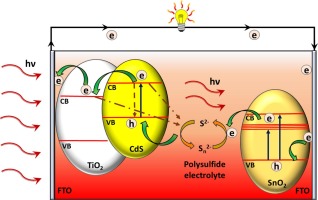
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Abstract

Fabrication of efficient CdS [quantum dot](https://www.sciencedirect.com/topics/materials-science/quantum-dot) sensitized solar cell with a novel stable counter electrode based on a thin film of SnO2 is revealed. The film was characterized by using Scanning Electron Microscopy (SEM), High-Resolution Tunneling Microscopy, X-ray diffraction (XRD) and UV–Visible spectroscopic techniques. Photovoltaic performances and Electrochemical Impedance Spectroscopic techniques (EIS) were performed on FTO/TiO2/CdS/polysulfide/SnO2/FTO device under the light illumination of 100 mW cm−2 and comparison was done with the conventional Pt counter electrode. Impressive 43 % efficiency enhancement in these solar cells was achieved compared with the Pt based devices. Porous thick nanostructure of SnO2 with crystal defects such as oxygen vacancies and Sn vacancies arising from lattice structures as confirmed by SEM, Raman and, XRD spectroscopy could be some of the reasons for this enhancement. Excellent photo enhanced electrocatalytic activity against the [polysulfide](https://www.sciencedirect.com/topics/materials-science/polysulfide) electrolyte is confirmed by EIS and Cyclic Voltammetry studies.

Graphical abstract



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Introduction

Among the third generation of photovoltaics, quantum dot sensitized solar cells (QDSSCs) have been attracting much attention owing to their unique properties existing in the quantum dots, such as tunable band gaps, multiple exciton generation, and large extinction coefficient [1], [2]. According to recent literature, significant performance enhancements have been achieved in these solar cells with an increased interest in reaching the maximum theoretical efficiency of ∼ 66 %, which is much greater than conventional solar cells [2]. Even though numerous research investigations have been carried out on these QDSSCs, still their power conversion efficiencies and stability are much lower than commercially available conventional Si solar cells.

The main components of QDSSCs are quantum dot sensitized photoanode, counter electrode (CE), and the electrolyte sandwiched in-between them. Light capturing quantum dot decorated, wide bandgap mesoporous semiconductors like TiO2, SnO2, ZnO, etc. are the most popular photoanodes employed in these devices [3]. Here the quantum dots are responsible for generating electron-hole pairs from the harvested light, while wide bandgap semiconductors provide electron transport pathways in aid of the current generation and reducing electron-hole recombination. Polysulfide redox mediator (Sn2−/S2−) is preferred as the electrolyte for the quantum dot regeneration process and hole transportation from the counter electrode. In this context, reduction and oxidation take place at the counter electrode and therefore CE plays a key role in the conversion of Sx2− ions to S2− ions and vice versa, within the electrolyte which eventually affects the overall performance of the device. Generally, CEs of these QDSSCs consist of a thin film of platinum (Pt) deposited on a conducting substrate as the catalytic materials due to its high electro-catalytic activity [2], [3]. Even though it has been widely used as a CE for QDSSCs, several factors have been proved to be disadvantageous for its continuous use. It was reported that Pt shows a reduction in catalytic activity when contacted with polysulfide electrolyte, due to surface adsorbed sulfur and polysulfide which reduce catalyst active surface area and increase charge transfer resistance at the electrolyte/CE interface [4], [5]. On the other hand, fabrication processes of these rarely available and expensive noble metal, Pt counter electrodes involved expensive sputtering and electrochemical depositing techniques which would limit the large-scale commercial applications of these devices [3], [5]. In order to replace these Pt-based counter electrodes with cheaper materials with good electronic conductivity and efficient electro-catalytic effects for polysulfide reduction, various potential alternative materials such as those prepared with carbon [6], Au [7], structured carbon material [8], CoS [9] and CuS [10], Cu2S [11], were used as the catalyst materials of the counter. Moreover, conducting polymer materials, such as polythiophene (h = 0.09 %), polypyrrole (h = 0.41 %) and poly(3,4-ethylene dioxythiophene) (h = 1.35 %) have been also suggested [1]. In this context, recently Sajjad et al. have compared the device performances of CdS based QDSSCs fabricated with carbon counter electrodes having either polysulfide electrolyte or iodide electrolyte in these solar cells and achieved higher efficiency in carbon-based CE devices with an overall efficiency of 0.38 compared with 0.10 for iodide based cells [12]. Therefore, finding a low-cost suitable counter electrode for these sulphide-based electrolytes in QDSSCs is one of the major challenges in this area of research. With the aim of addressing these problems, in this study we report comparative photovoltaic studies by using various experimental techniques, on low cost easily fabricable SnO2 counter electrode based CdS QDSSCs with its counterpart comprising Pt-based devices.

Tin (IV) oxide (SnO2) could become a promising candidate due to its high chemical stability, high electron mobility, low cost, and environmentally friendly nature. SnO2 is a wide bandgap semiconductor widely utilized in batteries, sensors, supercapacitors, and the glass coating industry [13], [14], [15], [16]. However, SnO2 CEs are less explored due to their low electrocatalytic activity reported and need improvements with further modifications. Contradictorily, the notable photocatalytic activity of SnO2, revealed in literature, opens pathways for exploring it as a photoactivated CE material [17], [18], [19]. In recent studies, SnO2 has been widely used as the CE composite material for dye-sensitized solar cells. It was reported that SnO2 nanoparticles act as a binder in the composite and increase the effective surface area due to the porous nature of the CE [20]. Moreover, it was also reported that Pt-SnO2 nanocomposite CE records improved the photo-conversion efficiency in dye-sensitized solar cells when compared with the conventional Pt CE [13]. Studies related to Pt-decorated SnO2 nanotubes and hierarchical SnO2@SnS2 CEs showed the utilization of the fast electron transport property of SnO2 in developing CEs [14], [15]. A nonstoichiometric SnO2-δ nanocrystal-based CE reports a remarkable performance of dye-sensitized solar cells [16] and preparation of SnO2 CE under an N2 atmosphere imparts electronic structure changes leading to different catalytic activities [21]. However, to our knowledge, no studies have been reported on the feasibility of using thin films of SnO2 counter electrodes in QDSSCs so far. Therefore, in order to explore the possibilities of employing SnO2 thin films as the counter electrodes in QDSSCs, in this study, CEs were fabricated by depositing SnO2 films on fluorine-doped tin oxide (FTO) glass substrates using a simple spray pyrolysis technique and thermal annealing. Performances of CdS sensitized TiO2 photoanodes were compared by fabricating solar cells with both, Pt CEs prepared by sputtering a thin film of Pt on FTO glass (commercial) and SnO2 CEs prepared by spray coating a film SnO2 on FTO glass substrates.

Section snippets

Materials used

Fluorine doped tin oxide (FTO) coated glass (7 Ω cm−2, Solaronix), tin (IV) oxide (15 % in H2O colloidal dispersion, Alfa Aesar), titanium dioxide P90 powder (Evonik), titanium dioxide powder P25 (Degussa), Triton X-100 (Sigma Aldrich), glacial acetic acid (99 %, Fisher), sulfur (99 %, Daejng), sodium sulfide hydrate (>60 %, Sigma Aldrich), cadmium (II) chloride (99.99 %, Sigma Aldrich), potassium chloride (99 %, Aldrich) polyethylene glycol (99.8 %, Sigma Aldrich), ethanol (95 %, Sigma

Results and discussion

Basic operation of the photovoltaic device fabricated in this study is explained schematically as shown in Fig. 1. In the photoanode, CdS quantum dots (bandgap ∼ 2.4 eV, VB ∼ - 6.2 eV) generate electron-hole pairs upon light irradiation and TiO2, a wide bandgap semiconductor (bandgap ∼ 3.2 eV), provides electron pathways for the generated electrons to move to the external circuit. At the photoanode/ electrolyte interface S2- ions in polysulfide electrolyte (redox potential ∼ - 5.0 eV) oxidize

Conclusion

Simple and low-cost alternative counter electrode for Pt in sulfide electrolytes-based quantum dot sensitized solar cells was fabricated successfully and its performances were tested, with CdS QDSSCs, as a comparison of Pt counter electrode. The spray-coated nanoporous thick structure of SnO2 provides reactive sites for the efficient electrocatalytic reduction of Sn2– to S2– ions at the CE promoting fast hole recovery for CdS quantum dot regeneration and increased current density as reflected

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Data availability

The raw/processed data required to reproduce these findings cannot be shared at this time due to technical limitations but could be provided on request