



Improving the photovoltaic parameters in Quantum dot sensitized solar cells through employment of chemically deposited compact titania blocking layer

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HIGHLIGHTS

- Deposited titania compact layer by a facile room temperature chemical bath method.
- Employed this to mitigate back electron transfer at TCO/Electrolyte interface.
- Compact layer incorporation has improved the solar cell performance by 130%.

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ABSTRACT

Incorporation of compact blocking layer at the Transparent Conducting Oxide (TCO)/Electrolyte interface is an effective method to improve the device performance in QDSSC through mitigation of electron re-combinations at this interface. This paper reports the most facile and cost effective method of depositing a rutile titania Compact Layer (CL) over Fluorine doped Tin Oxide (FTO) substrate and its application in titania based CdS QD sensitized solar cells. The deposited compact layers are characterized to study their structural, optical, morphological and electrochemical properties using X-Ray Diffractometry, UV–Visible spectroscopy, Scanning electron microscopy, Cyclic Voltammetry and Contact Angle measurements. Sandwich solar cells are fabricated using these CL based electrodes and characterized using Electrochemical Impedance Spectroscopy, Open Circuit Voltage Decay and J-V characteristics. The CL incorporated CdS QDSSC showed more than 100% increase in the photoconversion efficiency (**1.68%**) as compared to its bare FTO counterpart (**0.73%**) proving the efficacy of employed strategy.

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1. Introduction

Quantum dot sensitized solar cell (QDSSC) is apparently one of the most promising architectures of third generation photovoltaics in view of its low cost and inorganic materials base. The efficiency of QDSSC is just over 9% [1] as against their potential of reaching 44% [2]. One of the greatest challenges for the research community around the world is to bridge the gap between the existing and projected photovoltaic efficiencies of these cells. Efforts are on to study the various factors influencing the performance of QDSSC. So

far it is being construed that the non-ideal nature of various solid liquid interfaces/junctions present in these excitonic solar cells give rise to many electron loss mechanisms and so stand in the way of attaining reasonable efficiencies in them. The principle of operation of QDSSC is analogous to Dye sensitized solar cells (DSSC) except for the difference that in the former Quantum dots are used for sensitization and in the latter it is the dye [3].

Back electron transfer (BET) or back reaction is one of the important loss mechanisms in DSSC/QDSSC. It is an accepted fact that in DSSC/QDSSC, BET takes place via two path ways [4,5]. One is the loss of electron through its transfer from conduction band of titania to oxidant species in the electrolyte and the other is its loss from the transparent conducting oxide to the electrolyte species. As the electron transfer process to electrolyte species is slow compared to the average transit time for the collection of electrons,

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BET due to the former may be neglected under short circuit conditions [4,5] even though the contribution of the latter is still finite and non-negligible. However, under open circuit conditions, to maintain a photo stationary state, rate of photo injected electrons is balanced by the rate of loss of electrons to the redox species through back reaction where in both path ways influence the magnitude of recombination. Thus in all, there are two arguments in vogue with respect to the locus of recombination in this type of solar cells [4,5]; one suggests that, the recombination takes place throughout the nanocrystalline photoanode, that is at the $\text{TiO}_2/\text{QD}/\text{electrolyte}$ interface while the other claims TCO/Electrolyte interface to be the main locus, where majority of the electrons are lost in recombination [6]. These studies also discuss the dependence of recombination path ways as a function of photo voltage or incident light intensity. They suggest that BET from titania to redox species is dominant at high photovoltages and at low photo voltages the BET contribution at TCO/electrolyte interface becomes considerably important [6]. Hence, for accomplishing better photoconversion efficiencies, it is imperative to arrest the recombination of electron what so ever, irrespective of its locus and its dependence on the magnitude of photovoltage or intensity of incident light.

Previously, many researchers have reported on strategies to minimize the recombinations at $\text{TiO}_2/\text{QD}/\text{Electrolyte}$ interface by means of surface passivations [6,7]. However, improvement of the TCO/Electrolyte interface has not received the attention that it really deserves. This may probably be due to the under estimation of its contribution in the recombination. With reference to the above, present article focuses on a facile chemical means to improve this interface and elaborates on how such an improvement can contribute to the enhancement in the photovoltaic performance of a nano structured solar cell.

Introduction of an electron blocking layer between the transparent conducting oxide and the porous metal oxide film is a widely accepted approach that may boost the performance of a photoelectrochemical solar cell. Such an intermediate compact layer is believed to insulate TCO from the redox species, and hence reduces the probable electron recombination at this interface. There are certain reports in the literature with regard to DSSC that, introduction of such a compact layer besides reducing the BET at TCO/electrolyte interface, is also known to improve various photovoltaic parameters and hence the overall photo conversion efficiency of the solar cell [8–12]. There are propositions in the literature of DSSC stating that compact layer is not really necessary at the TCO/Electrolyte interface since the redox Fermi level of Polyiodide, the most generally employed electrolyte in DSSC, matches with that of FTO thus reducing the probability of BET [13,14]. However, many researchers have presented considerable improvement in the photovoltaic efficiency of the cells after using a compact blocking layer [8–12]. In respect of QDSSC [15–18], the idea of compact layer seems to have worked well to reduce the recombination, since Polysulphide is used as the electrolyte, whose redox level is positive with respect to that of FTO. Though, there are many reports in the literature in synthesis and incorporation of such blocking layers, majority are for DSSC and only a countable research communications are available for QDSSC. Also, the syntheses reported either involved complex procedures like Magnetron sputtering and spray pyrolysis [19,20]. Some of the syntheses reported though facile, involved high temperature treatments and energy intensive techniques which are principally, in contravention with the cost-effective architectures like DSSC/QDSSC [20]. Besides different techniques, different materials are used for the design of compact layers. Blocking layers made up of Al_2O_3 , dense titania, Nb doped titania etc are reported in the literature and their effect in the performance enhancement is discussed in DSSC/QDSSC [19,20]. Compact Rutile titania layer is one of the effective ways to impede

back reaction of electrons and is used as an under layer for porous titania photo electrode in various DSSC to show improved solar cell performance [21]. The methods adopted to fabricate rutile titania compact layer so far have been either complex or cost intensive when used with DSSC. As stated earlier, the report here in presents the most facile and inexpensive technique to prepare an efficient compact blocking layer and its successful application as a strategy to improve the overall performance of QDSSC by considerable arresting electron recombinations at the FTO/electrolyte interface.

2. Experimental

Detailed procedure of deposition of nanocrystalline rutile titania films has been discussed in one of our previous reports where in authors have proposed application of these chemically deposited films as compact blocking layers against recombination of electrons in QDSSC [22]. However, the deposition method is discussed briefly here. Few ml of Titanium trichloride is added to four times its volume of double distilled water and stirred uniformly for a few minutes in a beaker. A few ml of NaOH is added to the solution under stirring to adjust its pH to be from 1.5 to 2.0. Clean, Fluorine doped tin oxide (FTO) coated glass substrates are now immersed into the baths maintained at three different pH. The substrates are named CL1(pH1.5), CL2(pH1.75) and CL3(pH 2.0) in the increasing order of pH of the baths to indicate the Compact Layer (CL) deposition over them. Throughout the course of the chemical reaction, the temperature of the deposition baths is maintained at 300 K. The substrates are taken out after 70 h with films deposited on them, washed in a gush of tap water to remove aggregations and then dried at room temperature.

Basic structural and morphological studies of the deposited CLs are carried out using X-Ray Diffractometry (XRD, PHILLIPS PW 1840), and Field Emission Scanning Electron Microscopy (FE-SEM, ZEISS). Ultra Violet-Visible spectro photometry (UV-VIS, JASCO V-670) is used to record transmittance spectra. The thickness of the deposited CL films is measured using transmittance spectra employing swanpoel's method [23]. Cyclic voltammetric studies are conducted for the compact depositions over FTO and bare FTO samples in polysulphide electrolyte with Platinum (pt) as the counter electrode and Ag/AgCl as the reference electrode using Potentiostat/Galvanostat (Ivium Soft:Vertex) at a scan speed of 100 mV/s and in the range -1 V to $+1$ V. Contact angle measurements are conducted for the bare and compact layer coated FTOs by drop casting polysulphide electrolyte using an optical microscope.

Photoanodes are prepared by doctor blading slurry of Degussa P25 titania, on few bare and CL films. Care is taken to keep the porous titania layer thickness to be nearly same in all the films and it is ~ 8 μm . The photo anodes so prepared are loaded with CdS QDs using Successive Ionic Layer Adsorption and Reaction (SILAR) method as reported in the literature [24]. Initially, Cadmium nitrate and Sodium sulphide are used as precursors of cadmium and sulphide ions by preparing their 0.5 M solutions in ethanol and methanol respectively. The titania photoelectrodes are now dipped in these solutions to deposit CdS QDs on its surface with intermittent rinsing in pure ethanol and methanol solutions respectively after the film dipping in cadmium and sulphur precursors. This constitutes one SILAR cycle. Six such SILAR cycles are conducted on each photoelectrode film to sensitize them with CdS QDs. Sandwich cells were fabricated using polysulphide as the electrolyte and pt as the counter electrode to conduct Electrochemical Impedance Spectroscopy (EIS) in dark for the frequency range between 10^6 -0.1 Hz, at a bias of -0.4 V and signal amplitude of 10 mV using Potentiostat/Galvanostat (Ivium Soft: Vertex). Same cell configuration and experimental equipment is used to study Open Circuit Voltage Decay (OCVD) employing a white light LED source at

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