



Elucidating the opto-electrical properties of solid and hollow titania scattering layers for improvement of dye-sensitized solar cells



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ABSTRACT

The light scattering method has been adapted in dye-sensitized solar cells (DSCs) for optical absorption enhancement. In DSCs, particle-size of TiO₂ should be inline with the scattering wavelength range. Scattering particles can be used either by forming a bilayer structure with TiO₂ nanocrystalline film or into the bulk of TiO₂ nanocrystalline film. For improving the DSCs performances these scattering layers aim to refract/reflect the incident light by extending the traveling distance of UV–Visible/near-IR light within the dye-sensitized TiO₂ nanocrystalline film. In this work, the scattering layers with two different particle-sizes (~200 nm-solid and ~400 nm-hollow) were deposited as an additional layer on the top of dye-sensitized TiO₂ nanocrystalline film and the morphological properties were studied. By using various opto-electrical characterization techniques, the influence of these scattering layers for two different classes of DSCs prepared from N3 (UV–Vis) and SQ2 (near-IR) dyes were investigated.

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1. Introduction

The power conversion efficiencies of dye-sensitized solar cells (DSC) are progressively improving. [1,2] The complete DSC is composed of several layers i.e. dye-sensitized TiO₂ nanocrystalline film on transparent conducting oxide (TCO) substrate, liquid iodide electrolyte and counter electrode (TCO + Pt/carbon) [3,4]. Several investigations have been implemented to improve the DSC efficiencies, primarily on photo-anode (TiO₂/dye). [5–7] In DSCs the photo-anode (dye-sensitized TiO₂ layer + TiO₂ scattering (Sc) layer) absorbs the incoming photons and the excited electrons from the dye are injected into TiO₂ layer. These injected electrons flow through the outer electrical circuit up to the counter electrode; electrons are transferred back to the dye by the reduction of liquid electrolyte i.e. iodine to triiodide. [8], [9–11] In the photo-anode, the dye-sensitized mesoporous (m)-TiO₂ layer is usually prepared from a 20 nm sized anatase phase semiconductor which ensures a larger surface area for loading larger quantities of dye. [12] This m-TiO₂ layer is transparent and allows the transmittance of

incident light of a longer wavelength range (IR to UV–Vis). The anatase phase 200 nm TiO₂ particle layers are often used to scatter the incident light, in order to enhance light harvesting. [13] This larger 200 nm TiO₂ particle-size layers will have low surface area for dye-sensitization (negligible dye loading) and enhance the light scattering, with minor input of incident light. [14]

In DSCs, the Sc layers can be incorporated by various methods such as a mixture of nanoparticles, double layered nanoparticles or different methods of producing single-layer structures which can also serves as light scattering. [15] Out of all these methods, double layered nanoparticles/submicron structures have proved to be promising by obtaining higher yields in DSCs and have been extensively studied [16–20]. Nevertheless, only fewer studies are concerned on the usage of nanoparticles/submicron structures for near-IR absorbing DSCs compared to Ru-complex DSCs. [6,21–28] Moreover, various forms of solid (S) and hollow (H) TiO₂ based nanoparticle layers are also used for photocatalytic applications apart from solar cells. [29–33] The importance of different particle-size TiO₂ Sc layers above m-TiO₂ layers was studied as a part of this work. These Sc TiO₂ layers were prepared from two different particle-sizes ~200 nm-S and ~400 nm-H structures. The scattering wavelength of these two different particle-size particles matches the absorption wavelength of the dyes. Two different classes of DSCs were prepared from N3 dye (absorbs in UV–Vis region) and SQ2 dye (absorbs in near-IR). By using scanning electron microscopy

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(SEM) and X-ray diffraction (XRD) measurements the morphological and structural properties of these larger particle-size TiO_2 layers were probed. The influence of these larger particle-size TiO_2 **Sc** layers in N3/UV-Vis and SQ2/near-IR DSCs were expounded by using various opto-electrical characterization techniques such as light current voltage (I-V), external quantum efficiency (EQE) and electrochemical impedance spectroscopy (EIS) measurements.

2. Experimental details

Glass/FTO (fluorine doped tin oxide) substrates of $15 \Omega/\text{sq}$. were supplied from Dyesol Inc. The TiO_2 pastes of (T/SP) for transparent **m-TiO₂** layers, (R/SP) for solid reflective add-on **Sc** layers, 5-carboxy-2-[[3-[(2,3-dihydro-1,1-dimethyl-3-ethyl-1 H-benzo[e]indol-2-ylidene)methyl]-2-hydroxy-4-oxo-2-cyclobuten-1-ylidene)methyl]-3,3-dimethyl-1-octyl-3 H-indolium (SQ2) and cis-diisothiocyanato-bis(2,2'-bipyridyl-4,4'-dicarboxylic acid) ruthenium(II) (N3) dyes were purchased from Solaronix. T/SP is a light-yellow viscous paste with 100% anatase phase nano-particles size from 15 to 20 nm. R/SP is a white viscous paste with the nano-particle size greater than 100 nm. Preceding the DSCs assembly, all the FTO substrates were thoroughly cleaned and treated with 40 mM TiCl_4 solution at 60°C for 30 min and calcinated at 500°C for 30 min. The **m-TiO₂** layers were deposited onto TiCl_4 treated glass/FTO substrates using doctor-blade and calcinated at 500°C for 30 min. The solid-scattering (**S-Sc**) TiO_2 layers were deposited using the commercially available Ti-Nanoxide R/SP paste using doctor-blade technique. For developing hollow-scattering (**H-Sc**) TiO_2 layers, initially polystyrene spheres (PS) paste was prepared using the method of mesoporous inverse opal photonic crystals synthesis [34]. Later, this paste was deposited on **m-TiO₂** layers by using doctor-blade technique. The 500 nm PS were supplied from Bangs Laboratories, Inc. Finally, all the **Sc** layers were calcinated at 500°C for 30 min in presence of dry oxygen. The complete thickness of the TiO_2 layers (**m-TiO₂** + **S-Sc/H-Sc**) was $\sim 12 \mu\text{m}$ to $\sim 13 \mu\text{m}$. These layers were treated with 40 mM TiCl_4 solution at 60°C for 30 min and sintered at 500°C for 30 min. These FTO/ TiO_2 layered electrodes were dipped in 0.4 mM of N3 or 0.1 mM (with 10 mM cheno) of SQ2 [35] ethanoic solutions for 4 h [36]. Onto the cleaned FTO substrates, Pt-counter electrode layers were deposited by brushing from Pt-solution (Solaronix) and calcinated at 450°C for 30 min. The dye-sensitized TiO_2 and Pt-layered electrodes were sealed together using $25 \mu\text{m}$ thermoplastic films (Solaronix) and filled with homemade liquid iodide electrolyte [37,38].

Photovoltaic I-V performances of these DSCs were recorded by a class A solar simulator purchased from Newport Spectra Physics together with a Keithley 2400 Source meter under simulated 0.77 sun illumination (filter AM 1.5) at $\sim 25^\circ\text{C}$ with an active area of 0.2064 cm^2 . By using UV-Vis absorption measurements, the absorption of dye adsorbed **m-TiO₂** layers with and without **Sc** layers was almost constant. XRD patterns of the **Sc** layers were recorded by grazing incidence diffractometer (Bruker D8) dedicated to polycrystalline thin films (Sol-X detector, room temperature, Cu K alpha) in locked mode. The SEM morphology was characterized by FEG-ESEM XL30 (FEI) with an accelerating voltage of 5 kV under high vacuum. The EQE measurements were performed with the monochromator model GM 252 and the lamp model LH 151 N (supplied from Kratos), the power meter with Si photo detector purchased from Thorlabs. The EIS measurements were performed by using Potentiostat/galvanostat (BioLogic SP200/EIS; Princeton Applied Research - EG&G Instruments 263A) at frequency range of 100 mHz to 100 kHz.

3. Results and discussion

The schematic representation of DSCs with **S-Sc** and **H-Sc** TiO_2 layers are depicted in Fig. 1. The reflection and refraction of light in different directions is mainly caused due to the orientation of nanoparticles,

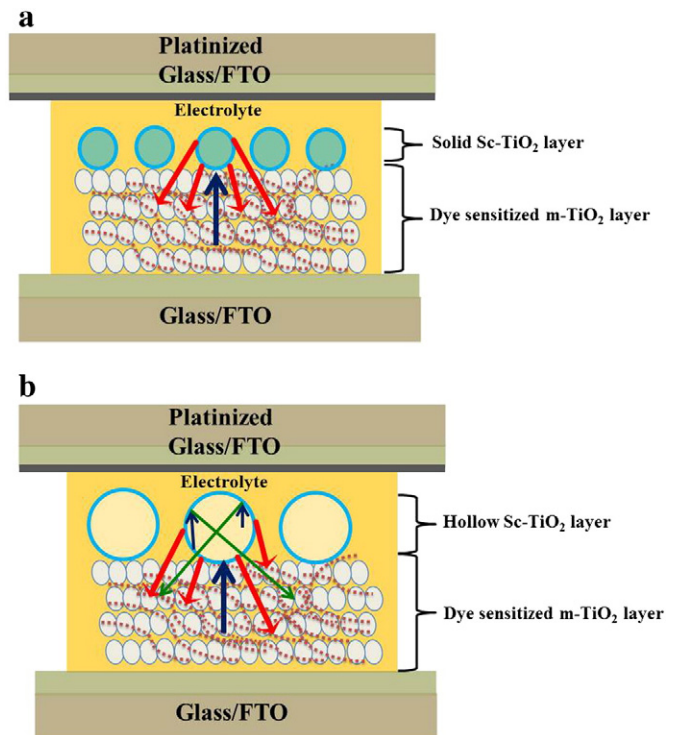


Fig. 1. Schematic representation of DSCs with **S-Sc** (a) and **H-Sc** (b) TiO_2 layers.

roughness and its boundary conditions. In **S-Sc** TiO_2 layer, reflection of light is taking place. In **H-Sc** TiO_2 layer the light undergoes both reflection and refraction. This is due to the hollowness of the nanoparticles that are responsible for slowing down of light at a certain wavelength range while traveling through layer and causing a slight change in the direction of light. The size of the nanoparticles can influence the direction of light. The refraction is also dependent on the refractive index of the TiO_2 layers. The light scattering at the **H-Sc** TiO_2 layer boundary between the shell and inner hollow pore might be more dynamic than **S-Sc** TiO_2 layer. [39,40] Thus, these multiple-refractions inside the **H-Sc** TiO_2 layer can increase the amount of light scattering which in turn could enhance the absorption window. The direction of the scattered light can have an adverse effect on the N3 and SQ2 DSC devices performances.

The SEM images of **S-Sc** and **H-Sc** TiO_2 layers are shown in Fig. 2 (a, b). From these images the well-dispersed and uniform TiO_2 **Sc** layers can clearly be seen. In the preparation of **H-Sc** layers, PS of 500 nm was used and after the final calcination they resulted in the formation of **m**-layers. In the **H-Sc** TiO_2 layers, the TiO_2 particle-size shrinks by 15–20% [41,42]. From these images, it can be clearly observed that the **H-TiO₂** spherical particle-size was $\sim 400 \text{ nm}$ and larger than **S-TiO₂** spherical particle-size. From the XRD recordings as shown in Fig. 3 it can be concluded that mostly crystalline anatase phase of TiO_2 was present in these **Sc** layers. The XRD peak at 25° was sharper for **S-Sc** TiO_2 layer than **H-Sc** TiO_2 layer. This might be due to the presence of more amount of anatase phase of TiO_2 in **S-Sc** layer. These **Sc** layers are required for efficient DSCs. [15]

With this two particle-size **Sc** layers above **m-TiO₂** layers, a study was made to understand the influence on UV-Vis region absorbing N3 DSCs and near-IR absorbing SQ2 DSCs. The experimental light I-V parameters of the DSCs with **S-Sc** and **H-Sc** TiO_2 layers are shown in Fig. 4 (a, b) and presented in Table 1. It can be noted in N3-dye DSCs that the current density (J_{sc}) and open-circuit voltage (V_{oc}) were decreased by usage of a **H-Sc** TiO_2 layer, when compared to a **S-Sc** TiO_2 layer. However, in the case of SQ2 DSCs, significant enhancement in J_{sc} can be observed with the **H-Sc** TiO_2 layer while V_{oc} remained unaffected. From the I-V measurements the higher photo-current in

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