



# Enhance the performance of dye-sensitized solar cells by balancing the light harvesting and electron collecting efficiencies of scattering layer based photoanodes



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## ABSTRACT

Thin titanium dioxide (TiO<sub>2</sub>) semiconductor layer with different scattering layers are investigated in dye-sensitized solar cells (DSSCs). Usually, the scattering layer is placed after the photoanode films in order to harvest more incident light. The scattering layer based on rutile phase TiO<sub>2</sub> is prepared, and placed in the different position of the photoanode films (on the surface of the FTO glass, between the two layers of transparent TiO<sub>2</sub> film and after the transparent TiO<sub>2</sub> film). We use STT, TST and TTS as marks (T and S represent the transparent TiO<sub>2</sub> layer and the scattering layer respectively). The result of this study indicates that STT which has the lowest incident light harvesting efficiency demonstrates the highest electron collection efficiency, while TTS which has the highest incident light harvesting efficiency sacrifices the electron collection efficiency greatly. It is discovered that TST, of which the incident light harvesting efficiency basically remains unchanged compared to TTS, reveals higher electron collection efficiency and achieves the maximum photovoltaic conversion efficiency (7.0%). By applying UV-Visible and diffuse reflectance spectroscopy, electrochemical impedance spectroscopy (EIS), the effects of the incident light harvesting and electron collection efficiencies on different cells are analyzed. It makes the best use of this scattering layer and has a reference for the application of other scattering layer types.

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## 1. Introduction

Dye-sensitized solar cells (DSSCs) provide a promising alternative to conventional photovoltaic devices in renewable-energy research on account of their distinctive features, such as lower manufacturing costs and relatively higher conversion efficiencies [1,2]. Typical DSSCs consist of nanocrystalline TiO<sub>2</sub> film [3], sensitizing dyes [4], electrolytes containing redox couple [5], and platinum-coated counter electrodes [6]. When the dye molecules absorb incident photons, excited states dye molecules inject electrons into conduction band (CB) of TiO<sub>2</sub>, from which the electrons are transported into the back contact and flow in the external circuit [7]. Therefore, for DSSCs, there are many factors that can influence the photovoltaic conversion efficiency, among which the foremost

factors are the incident light harvesting and electron collection efficiencies of the cells [8].

Many researches had been done to enhance the light harvesting efficiency, such as exploiting new dye that can absorb a wider range of spectrum of the sunlight; using titanium tetrachloride (TiCl<sub>4</sub>) to dispose the photoanode films in order to increase specific surface area [9,10]; adding a scattering layer after the photoanode films with the purpose of limiting the light within the films [11,12]. Applications of the scattering layer have been reported many times, for instance, adding some metallic oxide (ZnO, TiO<sub>2</sub>) whose particle size is hundreds of nanometers to the film [13,14]; introducing hierarchical TiO<sub>2</sub> ball consist of tiny nanometer crystal and so on [15]. When light collides with the large TiO<sub>2</sub> particle having sub-micrometer size, the light scatters strongly, which increase the path length of the incident light in the nanocrystalline TiO<sub>2</sub> films [16,17]. Researchers pay much attention to this factor; however, another key factor is usually neglected, which is electron collection efficiency. For traditional double photoanode structure, the layer superposed on to FTO glass is made up of large number of anatase phase TiO<sub>2</sub> particles which play a weak scattering effect. Then, a

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scattering layer is added after it in order to increase the scattering effect. However, it is noticed that there will be a large part of photoinduced electrons located far away from the FTO glass while more electrons are produced. Most of the electrons will get recombination with electrolyte owing to the long transmission path [18]. Therefore, it is important to balance the incident light harvesting and the electron collection efficiencies of photoanode films in order to optimize cell performance [19].

More complex structure of the scattering layer geometries was reported to enhance the photovoltaic conversion efficiency in DSSCs [20,21]. In this work, we report the preparation of four photoelectrodes, the reference electrode TT is based on pure anatase phase  $\text{TiO}_2$  film which the thickness is  $10\ \mu\text{m}$  (it consists of two transparent layers and each layer thickness controls in  $5\ \mu\text{m}$ ). The other three electrodes are mentioned above: STT, TST, and TTS (the thickness of the scattering layer also controls in  $5\ \mu\text{m}$ ). Then the incident light harvesting and the electron collection efficiencies of each film are analyzed. Furthermore it will come to the conclusion that TST, the incident light harvesting efficiency is almost consistent with TTS, reveals higher electron collection efficiency and also achieves the maximum photovoltaic conversion efficiency. Our work is based on traditional rutile phase  $\text{TiO}_2$  scattering layer of our laboratory, but it is proposed that when designing photoanode structure, two factors should be taken into account: the incident light harvesting efficiency and the electron collection efficiency, which are of great referential significance for the introduction of other scattering layer types into the photoanode.

## 2. Experimental

### 2.1. Materials

Titanium isopropoxide (TTIP,  $\text{Ti}[\text{OCH}(\text{CH}_3)_2]_4$ ) was used as Ti precursor. Triton X-100 was selected as emulsifier of  $\text{TiO}_2$  slurry. Polyethylene glycol (PEG, molecular weight of 20000) was obtained from Sinopharm Chemical Reagent Corporation (China). Fluorine-doped  $\text{SnO}_2$  conductive glass (FTO, sheet resistance  $10\text{--}15\ \Omega\ \text{sq}^{-1}$ ) was purchased from Asahi glass, Japan. The dye (N719) was purchased from Solaronix, Switzerland. Iodine ( $\text{I}_2$ , 99.8%) was obtained from Beijing Yili chemicals, China. Lithium iodine (LiI, 99%), guanidine thiocyanate (GNCS) and 4-tert-butylpyridine (TBP) were obtained from Acros. Propylene carbonate (PC) was obtained from Sinopharm Chemical Reagent Corporation (China). All the reagents used were analytical grade.

### 2.2. The preparation of scattering layer paste and pure $\text{TiO}_2$ paste

10 mL of TTIP mixed with 2.1 g acetic acid was dropped slowly to 50 mL of deionized water under vigorous stirring for about 1 h. Then 0.68 mL of nitric acid was added to the mixture solution and stirred for 3 h at  $80\ ^\circ\text{C}$  so as to obtain transparent sol. The formed transparent sol was filtered to wipe off insoluble impurity. After that, the 63 mL of sol adjusted by deionized water was heated at  $220\ ^\circ\text{C}$  in a 100 mL autoclave for 12 h [22]. When it cooled down at room temperature, mixed the solution with some ethanol, centrifuged the liquid under 7000 revolutions per minute, cleaned several times using ethanol, and then dried the precipitate at  $80\ ^\circ\text{C}$  to get the hydrothermal powders. 4 g hydrothermal powders was ball-milled with 10 mL of ethanol and 0.8 mL of diacetone for 24 h, then 12.5 mL of ethanol and 4.5 mL of deionized water were added to above mixture for another 12 h, after then 1 g rutile powder was added to agate jar for about 30 min, finally, we got the scattering layer paste. The preparation of pure  $\text{TiO}_2$  paste was similar to that of the scattering layer paste before heating at  $220\ ^\circ\text{C}$  in a 100 mL autoclave for 12 h. When it cooled down at room temperature, and then followed

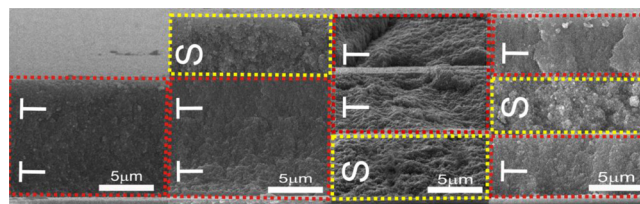


Fig. 1. Typical SEM images of cross profile of each photoelectrode.

by addition of nitric acid (0.4 mL) and dispersion by sonication. The solution was then concentrated to 25 mL by rotary evaporator. Finally, 0.56 g PEG and 0.5 mL of Triton X-100 were added to form the pure  $\text{TiO}_2$  slurry.

### 2.3. Preparation of photoelectrodes and DSSCs

Four kinds of  $\text{TiO}_2$  photoelectrode were prepared by doctor-blading, respectively, and then they were sintered at  $500\ ^\circ\text{C}$  for 30 min. The mesoporous photoelectrodes were preheated at  $120\ ^\circ\text{C}$  for 30 min; after cooling down, they were immersed in a 0.5 mM ethanolic N719 solution for 12 h at  $60\ ^\circ\text{C}$ . After taking them out, the excessive dye molecules on the electrodes were washed away by ethanol for several times, and then the electrodes were dried with a hair dryer. Then the sandwich-type DSSCs were assembled with an electrolyte layer containing 0.03 M  $\text{I}_2$ , 0.05 M LiI, 0.1 M PMII (1-methyl-3-propyl imidazolium iodide), 0.5 M TBP, 0.1 M GNCS in mixed solvent of acetonitrile and propylene carbonate ( $V/V = 1:1$ ) between the photoelectrodes and Pt counter electrode prepared by sputtering method [23].

### 2.4. Characterization

Field emission scanning electron microscopy, FESEM (JEOL, 6700, Japan) was applied to study the morphology and the thickness of the anode films. To measure the amount of dye adsorbed on the  $\text{TiO}_2$  surface, NaOH solution (0.1 M) was used for desorption and UV-vis spectrometer (Varian Carry 5000) was used to measure the absorption spectra. Diffuse reflectance and transmittance spectra were also measured on Carry 5000. The photocurrent-voltage characteristics of the cells were recorded by applying an external potential bias to the device under simulated illumination (Newport, 91192) with a power density of  $100\ \text{mW}\ \text{cm}^{-2}$  and the active area of the cell was controlled by a  $0.25\ \text{cm}^2$  mask. Electrochemical impedance spectroscopy measurements were performed on a CHI 600 C (Shang Hai, China) under the open circuit voltage ( $V_{oc}$ ) bias at illumination with the frequency ranging from 100 kHz to 0.1 Hz, and the magnitude of modulation signal was 0.01 V. IPCE spectra were recorded between 400 nm and 800 nm wavelength range under the irradiation of a 300 W xenon lamp (Newport, USA).

## 3. Results and discussion

### 3.1. Material characterization

#### 3.1.1. Morphologies and dye desorption experiment

The SEM images of cross profile in each photoelectrode were shown in Fig. 1. The location of the scattering layer can be distinguished clearly due to the large size of rutile phase  $\text{TiO}_2$  particles which show a comparatively coarse appearance. A good connection can be seen at the interface of scattering layer/FTO/transparent layer, which will not obviously hinder electron transmission. The thickness of the film TT is about  $10\ \mu\text{m}$ , while the thickness of other three films basically remains the same, controlled in  $15\ \mu\text{m}$ . The introduction of the scattering layer directly leads to the increased

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