



# Dye-sensitized solar cells with titania concave mirror



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## ABSTRACT

A layer of titania semi-spherical shell is assembled onto the titania nano-crystalline film to serve as photo-anode in dye-sensitized solar cells. The shell performs as a concave mirror. Unlike random scattering centers, the unique structure reflects full band of the incident light back to the titania nano-crystalline film in a convergent way. As a result, conversion efficiency is drastically enhanced by the semi-spherical shell coating with a maxima ratio of over forty-seven percent on cells with an effective area of 100 mm<sup>2</sup>.

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## 1. Introduction

Effort to extend the light path in dye-sensitized solar cells (DSCs) is a synergy to improve the conversion efficiency since the first report of DSC in 1991 [1], no matter what kind of electron generation, separation and transportation is designed. DSCs typically consist of a TiO<sub>2</sub> thin film on transparent conducting oxide substrate as the photo-anode, dye-molecules to convert incident photons to electrons, liquid electrolyte to connect the inner circuit and help the regeneration of dye molecules. There are many reports on the above aspects to improve cell performance, for example, extending the spectral response of dye molecules so as to generate more electrons on low energy band photons; [2,3] growing arrays of nanostructure [4,5] to provide a fast electron transport pathway so as to enhance electron collecting efficiency; novel electrolyte to overcome the long-term instability and corrosive nature of triiodide/iodide electrolyte [6]. Although these progresses make independent improvement on the DSCs' efficiency, they are sometimes mutually restrained. For example, a thicker film is highly expected for the ultimate absorption of the incident light. But nanoparticle thin films come to their critical thickness of around ten micrometers where the recombination of electrons is competitive to the forward process of electron transportation [7]. Although the nanowire/tube arrays provide a direct pathway for the fast transportation of electrons on the electrode, they are with

low surface area. As a result, fewer dye molecules are loaded on the surface of these structures [8]. Apart from these efforts on electron generation, separation and transportation, the extension of light path is a general route for the performance enhancement.

The light path in a DSC can be extended by the light-scattering effect [9] with top-coatings of large-sized particles on thin solid film as a back reflector [10] or by the incorporation of large-sized particles in the thin solid film as random scattering centers [11]. These particles can be substituted with voids [12,13], photonic crystals [14–17], crystals with featured facets [18], and even polymer or silver mirror attached to the back of the cathode [19,20]. Conversion efficiency enhancements from around 8% [19] to over 40% [10] are reported. Nevertheless, there are several problems remain unsolved.

One of the key problems is that the reflected lights propagate randomly after the incident light is scattered by the reflecting or scattering centers. A considerable part of the scattered light is not returned back into the cell. They leak out of the cells instead due to the random orientation of the scattered light [9,10]. As a result, energy loss is a due process. Furthermore, as a back reflector exterior to the cell, the counter electrode must be transparent. Even though, the reflected light must travel through the counter electrode and the electrolyte, which means the reflected light will be partially absorbed and energy loss is destined, before it can be captured by dye molecules [19,20]. The problem seems to be solved by the incorporation of large particles, voids or photonic crystals inside the cell, no matter they are within the thin solid film or on the top of it. Besides the low dye loadings due to less surface area of large particles and pore structure of voids and photonic crystals [12,13]. These structures face another problems. The

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thickness of these nanocrystalline constructed structures is typically in the tenth of micrometer. The recombination of photo-generated electrons with electrolyte will compete with the electron transfer process. It is highly expected to increase the film thickness for more dye loadings. But the recombination possibility increases with increasing film thickness. Furthermore, the full illumination band of the incident light cannot be trapped by photonic crystals with a determined narrow band. And their physical properties are strongly dependent on the ordered structure. All the difficulties imply that much work needs to be done on tuning of the optical bands and large scale applications [16]. So, how to design a full band reflector with fewer light leakages and less electron recombination is a very important issue in extending the light pathway in DSCs.

In this work, ordered two-dimensional (2D) TiO<sub>2</sub> shell as a concave back reflector is prepared by the replica of polystyrene template. The back reflector is coated on top of the nc-TiO<sub>2</sub> film. The effective area of DSCs is 10 mm × 10 mm. The structure is characterized by XRD, TEM, and SEM. DSC parameters such as  $I_{sc}$ ,  $V_{oc}$ , FF,  $\eta$ , and IPCE are tested. An efficiency enhancement of 47.5% is received from photo-anodes with titania shells of 900 nm in diameter.

## 2. Experimental

### 2.1. Chemicals

All the chemicals used in this experiment were analytical grade without further treatments except that styrene (St) was alternately washed with 5% NaOH and deionized water in order to remove polymerization inhibitor.

### 2.2. Synthesis of titanium dioxide nanoparticles and monodisperse PS

A typical process to nanocrystalline TiO<sub>2</sub> (nc-TiO<sub>2</sub>) was as follows. 20 mL tetrabutyl titanate was injected into 36 mL deionized water under vigorous stirring for 1 h to form a white precipitate. The white precipitate was washed several times with deionized water. 3.9 mL tetramethylammonium hydroxide was then added to the precipitate under stirring at 120 °C for 3 h. After that, the color of the turbid solution changed to translucent blue-white. The solution was treated hydrothermally in the reactor at 210 °C for 4.5 h. The product was obtained by centrifugation, washing and collected for further characterization.

To prepare mono-disperse PS beads, 95 mL of water and 8 mL of St were added to a three-necked flask. A small amount of KPS initiator was introduced to the flask. The reaction was conducted at 70 °C under nitrogen atmosphere for 24 h. The PS colloid suspension was collected and washed with deionized water and EtOH several times, then dried for further application.

### 2.3. Preparation of photo-anode with TiO<sub>2</sub> shell

nc-TiO<sub>2</sub> films were prepared using doctor blade technique [9]. Then ordered monolayer of PS spheres was assembled onto nc-TiO<sub>2</sub> films according to previous literature [21]. The PS coated nc-TiO<sub>2</sub> films were fixed at 100 °C for 1 h before their application as template to TiO<sub>2</sub> shell.

Tetra-*n*-butyl titanate (TNBT), diethanolamin (DEA), ethanol (EtOH), and deionized water with a mass ratio of 0.5:0.24:7.44:0.06 were mixed to form a sol. DEA was used to control the hydrolysis and condensation rates of TNBT. The fixed PS coated nc-TiO<sub>2</sub> films were immersed into the sol, then left to stand for 1 min and taken out from the sol. The films conducted gelation process at room temperature for 24 h. After that, the films were calcined at 450 °C for 2 h to transform the gels to TiO<sub>2</sub> and remove

PS spheres. These films coated with TiO<sub>2</sub> shells were used as photo-anodes in DSCs. Films of mere nc-TiO<sub>2</sub> were also used as photo-anodes in DSCs to illustrate the concave mirror effects of TiO<sub>2</sub> shells.

### 2.4. Assembly of DSCs

The photo-anodes were immersed in a 300 μM solution of Ru-based N719 dye in ethanol overnight. They were rinsed with ethanol to remove non-chemisorbed dye then dried. The photo-anodes were assembled with a platinum coated counter electrode using a thin thermoplastic spacer (Surlyn, 1702, 25 μm). One drop of electrolyte (0.5 M LiI, 0.05 M I<sub>2</sub>, 0.3 M 1,2-dimethyl-3-propylimidazolium iodide in 3-methoxypropionitrile) was introduced into the space via capillary action.

### 2.5. Characterization

The phase and crystallization of the products were characterized by powder X-ray diffraction (XRD) using a Rigaku (Japan) D/Max2550VB+/PC X-ray diffractometer equipped with Cu K $\alpha$  radiation ( $\lambda = 1.5418 \text{ \AA}$ ) at a scanning rate of 8°/min for  $2\theta$  ranging from 20° to 80°. The operation voltage and current were 40 kV and 40 mA, respectively. The size and morphology were observed using Philips-FEI Quanta 200 scanning electron microscope (SEM) with an operation voltage of 20 kV. The current–voltage ( $I$ – $V$ ) characteristics of the cells were measured using a Keithley 2602 source-meter under simulated solar illumination (AM1.5-spectrum, 100 mW cm<sup>-2</sup>) at room temperature. The incident photon to current efficiency (IPCE) tests were performed on Zolix Solar-CellScan100 with a calibrated reference cell. The UV–vis transmission spectra were measured with an AvaSpec-2048-2 Spectrometer.

The loading amount of the dye in photo-anode was characterized by UV–vis absorption technique. The dye was desorbed from photo-anode into NaOH solution in water/ethanol (1.0 M, 50:50, v/v). A UV–vis spectrophotometer (Hitachi U-3900/3900H) was employed to measure the dye concentration of the desorbed-dye solution.

## 3. Results and discussion

A typical XRD pattern of the collected nc-TiO<sub>2</sub> powder is illustrated in Fig. 1a. All the diffraction peaks are readily indexed to pure TiO<sub>2</sub> with anatase structure (JCPDS No. 21-1272). The intense peaks suggest that the products are well crystallized. No other impurity peak is observed. A representative TEM image of the as-prepared sample confirms that the sizes of the TiO<sub>2</sub> crystals are in nano-meter scale (Fig. 1b). They are fairly well dispersed crystals of 9–20 nm in size (Fig. 1c). The small sizes provide large surface areas, while the good dispersibility favors dye loading [9]. nc-TiO<sub>2</sub> was applied on the FTO glass using the doctor-blade technique followed by heat treatment. The top surface of the nc-TiO<sub>2</sub> films can be controlled with surface undulation heights less than several hundred nanometers and adjacent step heights less than 50 nm (Fig. 1d), which favors the assembly of periodical PS spheres and afterward treatment to TiO<sub>2</sub> shells. If the surface roughness of nc-TiO<sub>2</sub> film is further increased, the periodicity of PS texture will be lost during the assembly process. As a result, the afterward treatment to TiO<sub>2</sub> shell will not lead to concave mirrors. Instead, a TiO<sub>2</sub> film coating is formed.

During the fabrication of periodic titania shell, it is pivotal to prepare high-quality PS templates with uniform morphology. The mono-dispersed PS beads were synthesized by a surfactant-free polymerization method [22]. The size distribution of the PS spheres was controlled through the concentration of the St. The

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