



## Research paper

# Modified surface loading process for achieving improved performance of the quantum dot-sensitized solar cells

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## ABSTRACT

Achieving high surface coverage of the colloidal quantum dots (QDs) on TiO<sub>2</sub> films has been challenging for quantum dot-sensitized solar cells (QDSCs). Herein, a general surface engineering approach was proposed to increase the loading of these QDs. It was found that S<sup>2-</sup> treatment/QD re-uptake process can significantly improve the attachment of the QDs on TiO<sub>2</sub> films. Surface concentration of the QDs was improved by ~60%, which in turn greatly enhances light absorption and decreases carrier recombination in QDSCs. Ensuing QDSCs with optimized QD loading exhibit a power conversion efficiency of 3.66%, 83% higher than those fabricated with standard procedures.

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## 1. Introduction

Quantum dot-sensitized solar cells (QDSCs) have emerged as a kind of promising alternatives to existing photovoltaic devices due to their low cost and simple production procedure [1–3]. These QD-based photovoltaics possess several exclusive advantages because of the unique and versatile characteristics of QDs, including spectral tunability with tunable band gap by changing their size, high absorption coefficient to produce more excited carriers, as well as the potential of multiple exciton generation and hot-electron injection to exceed the Schottky–Queisser limit [4–8].

Operative mechanism of QDSCs resembles that of a typical dye-sensitized solar cell (DSSC), except for the dye sensitizer being replaced by QDs. QDs absorb photons from sunlight and inject electrons into the TiO<sub>2</sub> conduction band [9]. It is known that the achievable photocurrent of these sensitized solar cells is intrinsically limited by the total amount of the photons that the sensitizers can absorb and also the open-circuit voltage is closely related to the surface properties of the TiO<sub>2</sub> [10]. It is thus highly desirable to

achieve a high surface coverage of the QDs on TiO<sub>2</sub> films for fabricating high-efficiency QDSCs. Adsorption of the QDs on mesoporous TiO<sub>2</sub> films can be carried out via the following two approaches: (a) *in situ* method. QDs are directly grown on TiO<sub>2</sub> nanoparticles by direct chemical reaction of ionic species using the methods of chemical bath deposition (CBD) [11,12] or successive ionic layer adsorption and reaction (SILAR) [13]. (b) Post-synthesis assembly approach (also referred to as the *ex situ* method). Colloidal QDs were pre-synthesized and then attach to TiO<sub>2</sub> films in a way of ligand coordination [14–16]. The first method involves a nucleation and growth mechanism leading to a high surface coverage of the QDs on TiO<sub>2</sub> films but rendering rather difficult the control of the size distribution of the QDs, and ultimately an internal recombination of the solar cells [10]. Use of the pre-synthesized colloidal QDs in the alternative approach of the *ex situ* method can avoid these disadvantages. It enables a better control on the morphological characteristics of the QDs and subsequently a potentially enhanced electronic properties. Deposition of the colloidal QDs on TiO<sub>2</sub> mesoporous films is typically carried out by directly soaking the TiO<sub>2</sub> films into QD solution and allow it to stay for an extended period of time [17,18]. However, because of the weak coordination between colloidal QDs and TiO<sub>2</sub> nanoparticles, surface concentration of these pre-synthesized QDs still remains at a relatively low level and the maximal control over the surface distribution has not yet been fully realized due to the insufficient understanding of the QD/TiO<sub>2</sub> inter-

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face [1,10,19,20]. Previous efforts that have been made to improve this situation mainly focused on surface functionalization of the TiO<sub>2</sub> nanoparticles or the QD itself with bifunctional molecular groups like mercaptopropionic acid (MPA), trioctylphosphine (TOP), and thioglycolic acid (TGA), etc. These approaches were indeed helpful in increasing surface loading, however, they suffer from charge transport problems caused by either the insulating organic layers or the non-ignorable distance between nanoparticles [21–23].

Herein, we propose a robust and efficient method to improve the QD loading on TiO<sub>2</sub> surface. S<sup>2-</sup> treatment of the QD-attached TiO<sub>2</sub> films was previously reported to trigger an inorganic ligand exchange on QD surface, leading to an improved photovoltaic performance of the solar cells due to the enhanced charge transport between QDs and TiO<sub>2</sub> nanoparticles [24]. In this paper, from considerations of the nature of the interactions between QDs and TiO<sub>2</sub> nanoparticles, as well as the interactions between colloidal QDs and their capping ligands, we show that this facile post-treatment methodology can further optimize the QD/TiO<sub>2</sub> interface and lead to a dramatic increase in effective QD uptake on TiO<sub>2</sub> films, which contributes to an enhanced light absorption and suppressed charge recombination, and thus an improved photovoltaic performance of the solar cells.

## 2. Results and discussion

Initial adsorption of the CdSe QDs on TiO<sub>2</sub> films was carried out by directly immersing the TiO<sub>2</sub> films into prepared QD solution (more details can be found in Supporting Information). Adsorption of the QDs on TiO<sub>2</sub> films was carefully monitored by their UV–vis absorption spectra. Fig. 1a shows the temporal evolution of the intensity of the absorbance at the first absorption onset (excitonic absorbance) of the CdSe QDs in TiO<sub>2</sub> films. It indicates that the adsorption of the QDs on TiO<sub>2</sub> surface saturates to a plateau after 60–70 h of exposure, after which no significant increase was observed. S<sup>2-</sup> treatment was then performed by immersing the saturated film into Na<sub>2</sub>S solution (0.08 M in 1:1 vol/vol water and methanol). After that, the treated TiO<sub>2</sub> film was allowed to re-immersion in QD solution for another 48 h. Surprisingly, as shown in Fig. 1b, significant increase in absorption intensity was observed, indicating a continue increase in QD uptake upon re-immersion into the QD solution. To further confirm this increase of QD amount on TiO<sub>2</sub> surface, cross-section scanning electron microscopy (SEM) micrograph with energy dispersive spectroscopy (EDS) elemental mapping were recorded (Fig. S1). It was found that the initial saturated TiO<sub>2</sub> film showed a Ti/Cd/Se atomic ratio of 88.50:5.75:5.75 (Cd/Ti, 6.5%), while after re-immersion it showed a Ti/Cd/Se atomic

ratio of 82.74:8.62:8.64 (Cd/Ti, 10.4%), indicating a 60% enhancement in QD loading. However, importantly, we note that since the formation of a CdS monolayer is quite possible during our S<sup>2-</sup> treatment/QD re-uptake process, the actual increase in CdSe QDs may be lower than this value, i.e. part of the increase in atomic ratio of Cd/Ti could come from CdS monolayers.

Influence of the S<sup>2-</sup> treatment on adsorption behavior of the QDs was further investigated. As shown in Fig. 2, after about 72-h initial QD adsorption, QD uptake on TiO<sub>2</sub> surface saturates to a plateau. A slight decrease in absorbance intensity was observed when immersing the QD/TiO<sub>2</sub> films in Na<sub>2</sub>S methanol/water solution for 10 h, indicating a certain degree desorption of the QDs from TiO<sub>2</sub> films. In fact, exposure of the QD/TiO<sub>2</sub> films to the pure solvent (water and methanol) also results in a decrease in QD amount as revealed by Fig. 2. This behavior is quite similar to those observed in Bazzan's et al. study on dye's adsorption behavior and they ascribe the desorption to the effect of hydrolysis of those weak linkages that formed between the adsorbent and the TiO<sub>2</sub> nanoparticles [25]. Next, for both Na<sub>2</sub>S solution and pure solvent, further increase in QD uptake on TiO<sub>2</sub> films was observed when re-immersion into the QD solution except that the former gives a more significant rise. The enhancement in QD uptake when re-immersion into the pure water/methanol solvent can be attributed to the rinse of those weakly bound QDs and then a subsequent reorganization of QDs on TiO<sub>2</sub> surface [25]. While the more

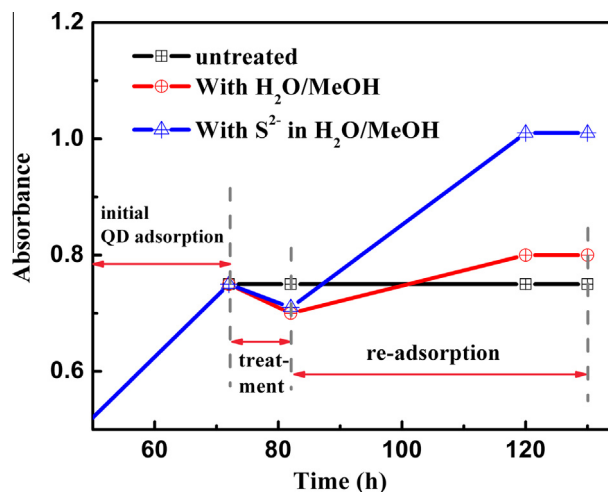


Fig. 2. Temporal evolution of the excitonic absorbance of the CdSe QDs in TiO<sub>2</sub> films.

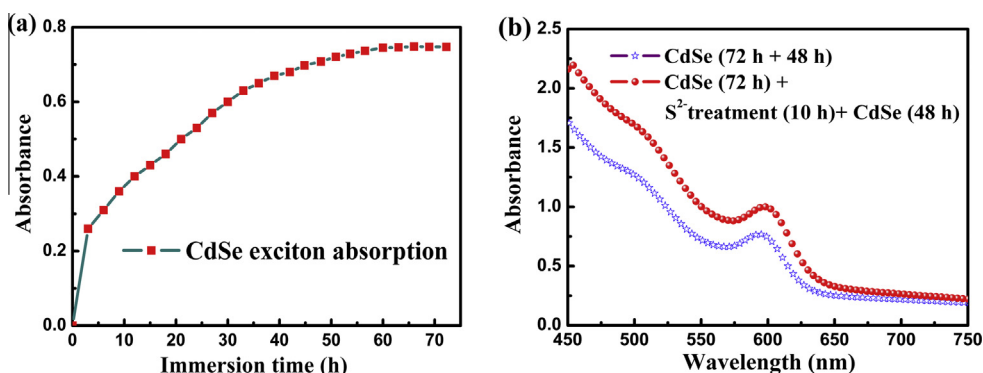


Fig. 1. (a) Absorbance at peak maximum vs. immersion time in CdSe QD solution. (b) UV–vis absorption spectra of the QD-sensitized TiO<sub>2</sub> films prepared by immersing the TiO<sub>2</sub> films in QD solution for total 120 h (blue line); S<sup>2-</sup> treatment after 72 h initial QD adsorption, followed by re-immersion in QD solution for 48 h (red line). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

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