



Effect of the adsorbed concentration of dye on charge recombination in dye-sensitized solar cells

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ABSTRACT

Charge recombination is the most important factor limiting the power conversion efficiency of dye-sensitized solar cells (DSCs). Lots of factors affecting the charge recombination in DSCs have been investigated except the adsorbed concentration of dye on the surface of TiO₂ film. By electrochemical impedance spectroscopy (EIS) analysis, it is found that with the adsorbed concentration of dye decreasing, both charge transport resistance at the TiO₂/electrolyte interface and electron life time within the TiO₂ photoanode increase, which indicates that the charge recombination in DSC decreases. Owing to this effect, the DSC keeps the fill factor as high as 0.75–0.78 even though the TiO₂ film thickness exceeds 20 μm.

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1. Introduction

Dye-sensitized solar cells (DSCs) are one of the most promising technologies for sunlight to electricity conversion. DSCs have obtained 12.3% of the photoelectric conversion efficiency (η) recently [1]. However, the η has still been far below the 30% of its theoretical maximum value [2]. The charge recombination, i.e., the electron loss in DSCs, reduces V_{oc} , I_{sc} and ff , and thus is one of the principal factors limiting power conversion efficiency [3].

The charge recombination in DSCs could take place between the injection electrons and triiodide in electrolyte at the semiconductor/electrolyte interface or oxidized dye at the semiconductor/dye interface. As the former is calculated to take priority kinetically to the latter [4], the oxidized dye molecules have been neglected in the charge recombination of DSCs. However, some evidences implied that dye molecules could have great influence on the charge recombination in DSC.

The studies on the dependence of electron transport property on the incident light intensity in DSC indicated that the recombination of the injection electrons with oxidized dye molecules (D^+) becomes important at high light intensity under the open-circuit condition where stationary electron densities in excess of 10^{17} cm^{-3}

(corresponding to more than one electron in a nanoparticle) [5,6]. In addition, the dye molecules are not independent from the electrolyte related electron recombination. Grätzel et al. [7] reported that the local I_3^- concentration had important influence on the electron lifetime. Under illumination, I_3^- is formed “in situ” by dye regeneration at the TiO₂/electrolyte interface, which is also proved by Raman spectra of dye-sensitized solar cells in situ investigated by Shi et al. [8]. The high local I_3^- concentration produced in the porous network under light is expected to accelerate the recapture of conduction band electrons by the local I_3^- and shortens their lifetime within the TiO₂ film. Peter and Wijayantha [9] reported that the recombination of electrons with tri-iodide was faster under high light intensity than under low light intensity and the electron lifetime depended inversely on the square root of the light intensity, i.e. $\tau_n \propto I_0^{-0.5}$. High local I_3^- concentration may result not only from high light intensity but also from high adsorbed concentration of dye on the surface of TiO₂ film. It is reasonable to wonder if the adsorbed concentration of dye has similar effects on the charge recombination in DSCs.

In this paper, the dye-adsorption capacity of anatase nanocrystalline film was first investigated to control the adsorbed concentration of dye in TiO₂ photoanode. The effect of the adsorbed concentration of dye on the charge recombination in DSCs was investigated by means of electrochemical impedance spectroscopy (EIS). And the photoelectric conversion performance characteristics of the DSC with different adsorbed concentrations of dye were measured.

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2. Material and methods

2.1. Materials

All chemicals used are analytical grade. Tetrabutyl titanate, anhydrous ethanol, Nitric acid, acetylacetone (ACA), polyethylene glycol 2000, ultra pure water and Triton X-100 were used as received. *cis*-di(thiocyanato)-*N,N'*-bis(2,2'-bipyridyl-4-carboxylate-4'-tetrabutyl ammonium carboxylate)ruthenium(II) (N719 dye), liquid electrolyte solution DHS-E23 (mixed by LiI, I₂, tert-butylpyridine (TBP), acetonitrile (ACN)) and Conducting glass plates (FTO, sheet resistance is 25 Ω/□) (used as a substrate for deposition of TiO₂ films) were purchased from Dalian Heptachroma Solar Tech. Co. Ltd.

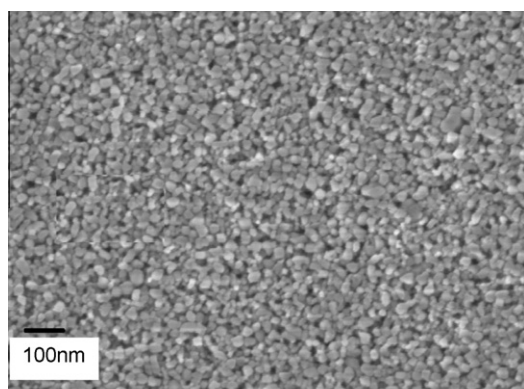
2.2. TiO₂ photoanode preparation and cell assembly

The TiO₂ photoanode was prepared by the hydrothermal method according to the literature [10]. A TiO₂ nano-particles were first synthesized in an autoclave at 200 °C. The porous TiO₂ films were prepared from the TiO₂ nanoparticle paste on FTO glass by annealing at 450 °C for 30 min. After annealing process, the TiO₂ films were cooled to about 80 °C and then immersed in an ethanol solution of N719 over night for adsorbing dye to obtain the TiO₂ photoanodes. The TiO₂ photoanodes were clamped with the counter electrode (a platinised FTO) into a sandwich-type cell. A drop of electrolyte solution was injected into the space between the clamped electrodes to obtain a DSC for testing of the photoelectric conversion performance characteristics. The effective size of the DSC is 0.25 cm².

2.3. Measurements and characterizations

The photoelectric conversion performance characteristics of DSCs were measured with IV Test Station 2000 (Crowntech, USA) which consisted of Sourcemeater 2400 (Keithley, USA), Solar Simulator 92250A-1000 (Newport, USA) and Lamp power supply 69907 (Newport, USA). The light intensity corresponding to AM 1.5 (100 mW cm⁻²) was calibrated using a standard silicon solar cell.

The electrochemical impedance spectroscopy (EIS) measurements were performed with a computer-controlled potentiostat (CHI604D, USA). The frequency range is 0.01 Hz–100 kHz. The magnitude of the alternative signal is 10 mV. All impedance measurements were carried out under a bias illumination of 100 mW cm⁻² (calibrated using a standard silicon solar cell) from a 500 W xenon light source and under open-circuit conditions. The obtained spectra were fitted with ZSimpWin software (v3.1, Princeton Applied Research) in terms of appropriate equivalent circuits.



The morphology and microstructure of TiO₂ films were characterized by field-emission scanning electron microscopy (FE-SEM) (Leo-Supra35, Germany). The crystal structures of the samples were characterized by X-ray diffractometer (XRD) (Philips PW 1830) using Ni-filtered Cu Kα radiation (λ = 1.54056 Å).

The TiO₂ film thickness was measured by a Stylus Profiler (Dektak8, Veeco, USA). The dye-loading of TiO₂ film was determined by desorbing the dye from the TiO₂ surface into NaOH aqueous solution (pH = 13) and measuring its adsorption spectrum by UV–Vis spectrophotometer (SP-2102UV, China). And then the concentration of desorbed dye was analyzed with the pre-measured concentration-absorbance curve of N719 dye to obtain the amount of dye adsorbed on the surface of TiO₂ film.

3. Results and discussion

3.1. Microstructure and dye-adsorption capacity of TiO₂ films

Fig. 1 shows the FE-SEM image of the microstructure of the TiO₂ film prepared. The TiO₂ film is composed of interconnected nano-particles of ~20 nm in diameter. The XRD pattern of the TiO₂ film shows distinguishable peaks at 2θ of 25.5°, 38.0°, 48.0°, 54.0°, 55.1°, 62.7°, 69.1° and 75.0°, corresponding to the characteristic peaks of anatase [11].

After immersing the resultant TiO₂ nanocrystalline film into the N719 dye ethanol solution with different concentrations or for different soaking time, the dye-loading of TiO₂ film varies within a certain range. The adsorption of dye molecules on the surface of TiO₂ film is known as a chemical adsorption process [12], which means that the dye-adsorption reaches a saturation state when the dye-loading does not increase any further. Fig. 2 shows the dye-loading of TiO₂ film obtained in different adsorption conditions.

By soaking the TiO₂ film in the dye solution for 24 h, the dye-loading increased with the concentration of dye solution increasing and became constant when the concentration of dye solution exceeded 0.3 mM, as shown in Fig. 2a. Alternatively, by soaking TiO₂ film in the dye solution with a concentration of 0.5 mM, the dye-loading increased with the soaking time increasing and remained unchanged when the soaking time exceeds 24 h, as shown in Fig. 2b.

The adsorbed concentration of dye (C₀) is defined by the adsorbed amount of dye (D₀) in unit specific surface area of TiO₂ film (S) by:

$$C_0(\text{mol m}^{-2} \text{kg}^{-1}) = \frac{D_0(\text{mol})}{S(\text{m}^2 \text{kg}^{-1})} \quad (1)$$

For TiO₂ films composed by the nano-particles with a certain size and shape, thus the same specific surface area and pore structure, the available surface area for dye molecules adsorption

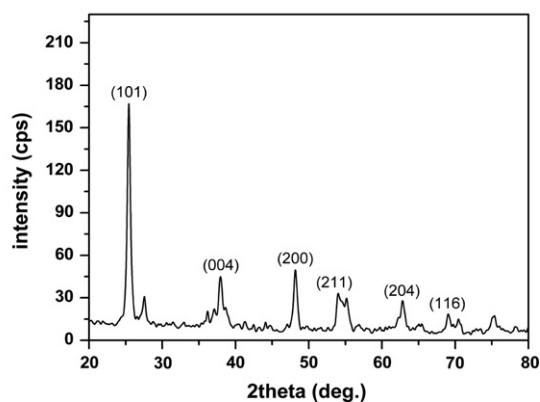


Fig. 1. FE-SEM picture of TiO₂ film (left) and XRD pattern of the TiO₂ film (right).

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