



Large efficiency improvement in nanoporous dye-sensitized solar cells via vacuum assistant dye adsorption

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ABSTRACT

The positive effects of the vacuum assistant dye adsorption (VADA) process on the performances of the dye-sensitized solar cells (DSSCs) were investigated experimentally. Using commonly used materials and processes as a demonstration, nanoporous films and the resulted solar cells with different dye adsorption processes were fabricated and characterized for comparison. The amounts of dyes adsorbed by the TiO₂ nanoporous films were found to be enhanced greatly compared with dyes adsorbed in ambient condition. In accordance, a relative increase of about 29% for the short-circuit currents (I_{sc}) and 32% for the efficiencies (η) were observed for the resulted DSSCs. Photocurrent-voltage measurement indicated that the lifetime of the photogenerated charge carriers changed little. Hence, such improvements were originated from the application of VADA process, which realized full contact between the dye solution and TiO₂ nanoparticles by driving away the air bubbles concealed in the nanoporous film. The VADA process may be also applied in other kinds of solar cells, such as solid state DSSCs and organic or flexible solar cells to enhance the dye adsorption and further increase their efficiencies.

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1. Introduction

Dye-sensitized solar cells (DSSCs) based on the photosensitization of nanocrystalline TiO₂ electrodes are regarded as a promising cost-effective alternative to silicon-based solar cells since the pioneering work of O'Regan and Grätzel [1,2]. A typical DSSC is composed of nanocrystal semiconductor film, sensitizing dye, electrolyte, and counter electrode. Among them, the dyes play central role of harvesting the incident sunlight and generating electron–hole pairs. The resulted electron is injected into the conduction band of the TiO₂ and finally diffuses across the interconnecting network of TiO₂ nanoparticles to the electrode and is collected to supply the external loads.

Such a procedure indicates that, the output voltage can be improved hardly because of the firm restriction by the thermodynamic potentials of TiO₂ and electrolyte. However, the current density can be enlarged easier as long as more electrons are generated and transferred to the electrode [3]. Hence, enhancing the dye adsorption on the surfaces of semiconductor nanocrystals is an effective route to obtain high efficiency in DSSCs. Indeed, relating to the pioneering work of O'Regan and Grätzel [1,2], nanoporous structures with large surface areas were adopted to

achieve a high dye adsorption density, which proved orders of increase in current density and cell efficiency. Since then, a lot of work has focused on the enlargement of surface areas to enhance the amount of absorbed dyes by reduction of nanoparticle sizes or utilization of novel structures [4–10]. In contrast, the dye adsorption techniques changed little with the assumption that the dye solution can fill the nanopores in the film completely during the dye adsorption process. As a typical process for dye adsorption, porous nano-semiconductor film was directly immersed into dye solution for a certain time [11–17]. However, we found that there were enormous tiny air bubbles concealed in the porous film, which prevented the dye solution from contacting the nanoparticles. As a result, parts of the particle surfaces left dye-nonsorbed. Moreover, the closer to the transparent conducting oxide (TCO) glass, the smaller amount of dye adsorption because of the difficulty in solution penetration. Obviously, the dyes closer to the TCO glass contribute much to the final current density because the generated electrons can move to the electrode with less probability of being recombined with the redox electrolyte or scattered by defects and grain boundaries. Hence, it would be beneficial if most of the concealed air pores were eliminated.

Here a vacuum assistant dye adsorption (VADA) process was introduced to the procedure of cell fabrication. We will show that a distinctly increased amount of dye adsorption can be obtained because of the relatively complete dye solution infiltration. We also

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show that the performances of the resulted DSSC can be improved remarkably in accordance.

2. Experimental

2.1. Preparation of nanoporous TiO₂ films

A simple procedure was used to prepare TiO₂ films. FTO conducting glass was used as substrates after being cleaned ultrasonically by acetone and alcohol, respectively. A mixture of TiO₂ film powder (Degussa P25, 2 g), deionized water (3.5 ml), glacial acetic acid (0.2 ml) and Triton X-100 (1 drop) was ground for 30 min to form a thick paste. The paste was coated on the surface of FTO glass plates by automatic film applicator coater (ZEHNTNER ZAA 2300) with controllable thickness. After dried in temperature humidity chamber, the thin film was annealed at 550 °C for 30 min to form a nanoporous film with interconnected networks.

2.2. Dye adsorption in ambient or vacuum conditions

Different dye adsorption processes were used for comparison as indicated in Fig. 1. For dye adsorption in ambient condition (defined as A process), the TiO₂ nanoporous film was immersed in dye solution (N719 24.3 mg and ethanol 100 mL were mixed to form solution) for 24 h. For dye adsorption in vacuum condition (defined as B process), a special designed device was used to provide vacuum environment. TiO₂ nanoporous film was placed at the bottom of the device and pumped. When the inside pressure was close to about 6×10^{-1} Pa, the valve was turned off and the pressure was kept for 2 min. Then the dye solution was dropped in to cover the TiO₂ film. Finally, the air was let in and kept for 24 h.

2.3. Preparation of counter Pt-electrodes

To prepare the counter electrode, a hole (0.8 mm in diameter) was drilled in the FTO glass. Then the FTO glass was cleaned ultrasonically by acetone and alcohol. After being dried in the air, Pt catalyst about 3 nm in thickness was sputtered on the FTO glass by precision etching coating system (Model 682, Gatan).

2.4. DSSCs assembly

The dye adsorbed TiO₂ electrode and Pt-counter electrode were assembled into a sandwich type cell and sealed with a hot-melt film (ionomer Surlyn 1702, Dupont) of 60 μm in thickness as described elsewhere [18]. Then the electrolyte (0.1 M LiI, 0.05 M I₂, 0.5 M 4-tertbutylpyridine in acetonitrile) was filled in and the hole was sealed using the hot-melt ionomer film made of Surlyn 1702 and a cover glass.

2.5. Measurements and characterizations

UV–visible absorption spectra were measured with a UV–Vis–Nir spectrophotometer (UV-3600, SHIMADZU) after the dyes were desorbed from TiO₂ film by NaOH solution (0.01 M). The applied scan rates were 240 nm/min and spectral resolution was 1 nm over the spectral region of interest. The baseline for the calculation of the absorption spectrum was the UV–Vis absorption spectrum obtained from the deionized water. The thickness of TiO₂ films were measured by surface profiler (DEKTAK 150, Veeco).

Photocurrent–voltage characteristics of solar cells were measured using a voltage–current source monitor (Orzel Sol 3A Newport) under illumination by a Newport solar simulator (AM 1.5, 100 mW cm⁻²) with a scan rate of 10 mV s⁻¹.

Time-resolved photovoltage was measured to study the photovoltage decay of DSSC and lifetime of the photogenerated charge carriers. Laser (FDSS 532-150, Crylas GmbH) and oscillograph (WAVE PRO 7Zi, LeCroy) were used in the measurement system. The resistance and the DC power we used in the electrical circuit were 40 MΩ and 3 V, respectively.

3. Results and discussion

As a demonstration, the materials and processes were selected as common and simple as possible to minimize the disturbance of other factors. Commercial TiO₂ nanoparticles (Degussa P25) were chosen as the basic material for fabrication of DSSC, which were usually used as contrast or comparison. Commonly used dye (N719) and electrolyte (LiI, I₂) were also selected. Furthermore, all the techniques for improving the efficiency were abandoned, such as introduction of novel structures and post-treatment with TiCl₄ [19].

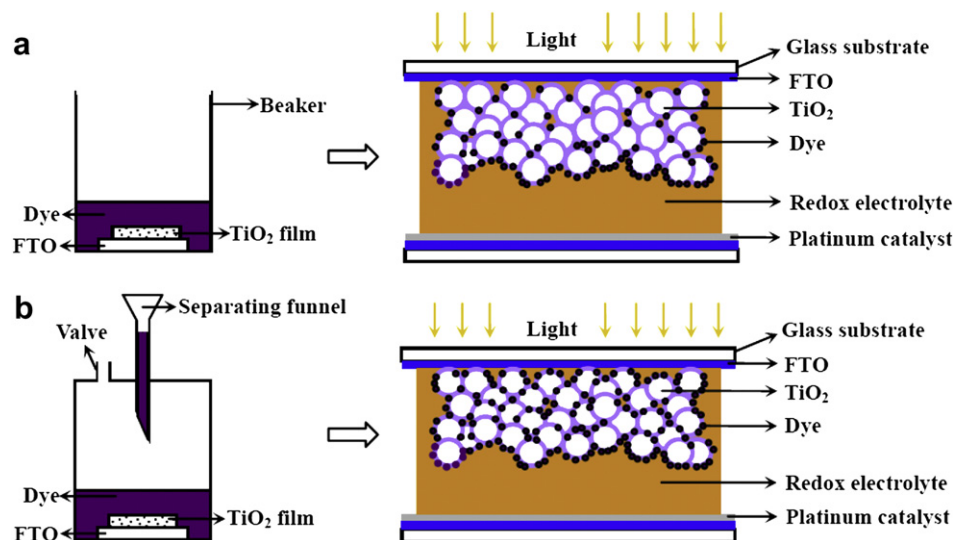


Fig. 1. Schematic of dye adsorption processes: a) Dye adsorption without vacuum; b) Dye adsorption by vacuum assistance.

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