



Performance evaluation of titanium dioxide based dye-sensitized solar cells under the influence of anodization steps, nanotube length and ionic liquid-free redox electrolyte solvents



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ABSTRACT

In this work, highly ordered titanium dioxide (TiO₂) nanotube (NT) arrays were synthesized on titanium foil using electrochemical anodization method. The morphological aspects of the nanotubes based on different anodization duration and number of anodization steps (maximum two) have been investigated. The nanotube arrays subsequently used as photoanode in a dye-sensitized solar cell (DSSC) assembly. The studies on the effects of different solvents for triiodide/iodide redox electrolyte and NT length towards the performance of DSSC were conducted. It is known that electrolyte solvent can significantly affect the photovoltaic conversion efficiency. It is noteworthy that longer NT length tends to yield higher efficiency due to better dye adsorption.

However, when the NTs exceeded certain length the efficiency decreases instead. Meanwhile, a comparison of DSSC performance based on number of anodization steps on titanium was performed. Highly ordered NT arrays could be obtained using two-steps anodization, which proved to have positive effects on the DSSC performance. The highest photovoltaic conversion efficiency in this work is 2.04%, achieved by two-step anodization. The corresponding average nanotubes length was ~18 μm, with acetonitrile (ACN) as the redox electrolyte solvent.

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1. Introduction

Solar energy is one of the most intensively subjects studied by many scientists. High efficiency p-n junction solar cells have long been achieved, however, they requires expensive materials and complicated fabrication process. In contrast, dye-sensitized solar cells (DSSCs) would be a more economical alternative, since they are easier and cheaper to produce in large scale. In addition, light-weighted and flexible versions could be made. It is noteworthy that they have relatively high solar energy conversion efficiencies [1–3].

DSSC was first introduced by O'Regan and Gratzel in 1991, where TiO₂ had been demonstrated as the photoanode [2]. Despite the availability of many wide bandgap metal oxides such as ZnO, Nb₂O₅, and SnO₂, TiO₂ remained a popular choice for DSSC applications [4,5]. A typical DSSC design consists of a redox electrolyte sandwiched between the space of the

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photoanode and cathode separated by a thin layer of chemically inert film. In general, the photoanode is made of nanocrystalline TiO₂ anchored by a monolayer of ruthenium complex dye e.g. N3 or N719, which plays a role as light harvesters. As for the cathode, it is typically comprised of transparent conductive oxides (TCO) such as indium tin oxide (ITO) coated with thin layer of platinum.

Screen-printed anatase phase of TiO₂ nanoparticles is commonly used as the anode of DSSC [6]. The nanoparticles are randomly packed in this architecture, allowing better dye absorption due to larger surface area [6]. However, the three dimensional networks formed from such packing possessed high density of electron trapping centers and grain boundaries, which lead to random electron diffusion, also promote recombination of injected electrons from the dye towards the acceptor band of TiO₂, ultimately hindering the performance of DSSC [7]. To improve the charge collection efficiency, one dimensional (1D) TiO₂ nanostructures such as nanowire and nanotube are utilized instead [8–14]. Comparing the aforementioned with nanoparticles, they are expected to exhibit better photovoltaic and photo-to-electric performance due to greater electron transportation, charge collection and light scattering effect [8–11].

Light illumination towards DSSC can be done from either the front or backside. For front side illumination, light is shone from the photoanode towards the cathode. This requires the photoanode to be sufficiently transparent for light to propagate through. There are two ways to achieve front side illumination. The first being complete anodization of sputtered titanium thin film (usually less than 500 nm), which left with only TiO₂ nanotubes, on a conductive glass substrate [9]. Alternatively, TiO₂ nanotube membrane that detached from its parent metal could be transferred to the conductive glass substrate, achieving front side illumination as well [10]. Backside illumination, on the other hand, operates oppositely compared to that of front side. This feature is adapted in DSSC designs when the photoanode is opaque, e.g. TiO₂ nanotube arrays on Ti foil grown via anodization [12–14]. Under this condition, light is transmitted through the platinized ITO, which located on top of the nanotube array. Losses in terms of light intensity are to be expected due to numerous factors such as light absorption by the redox electrolyte and substantial light reflection at the counter electrode. Although such losses would hinder the DSSC's performance, it still can be compensated by the characteristics of the photoanode. This includes longer nanotubes that can only be achieved through direct anodization on Ti foil, which promotes stronger light scattering effects, improving photovoltaic performances [15,16]. In addition, this approach is simple yet low-cost, without relying on expensive sputtering and membrane transfer process [12].

Redox electrolyte is one of the key components that affects the performance of DSSC. Extensive research activities have been focusing on the effects of gel electrolytes, ionic liquids, and various additives towards the efficiency of DSSC [17–24]. However, studies on solvents for the redox electrolyte are less emphasized, moreover, the characteristics of additives and ionic liquid-free redox electrolyte.

In the present work, additives or ionic liquid-free back-illuminated TiO₂ NT-based DSSC successfully fabricated. The one-step and two-step anodized NT length is controlled by varying the anodization duration from 2 to 24 h. The synthesized NT arrays were served as the photoanode of DSSC. The photovoltaic performance of the nanotube-based DSSCs as a function of solvent of redox electrolyte, NT length and anodizing step was studied and compared.

2. Experimental details

2.1. Nanotube preparation

Titanium foil (Strem Chemical, 99.7% purity and 0.127 mm thickness) was cut and sonicated in acetone and distilled water for 10 min each to remove surface contaminants. For the electrochemical setup, the titanium anode is distanced 2 cm away from a platinum wire cathode. Both of them were partially immersed in an electrolyte containing ethylene glycol (EG) mixed with 0.3 wt% NH₄F and 5% vol. distilled water. This setup would be used in both one-step and two step anodization process. All the processes would take place at room temperature condition with anodization duration as the variable in order to obtain nanotubes of different length.

The one-step anodization process was carried out by applying a constant voltage of 60 V between the electrodes. Meanwhile, for two-step anodization, the Ti foil was first anodized at 60 V for 1 h. Next, the anodized sample was removed from the electrochemical cell followed by sonication in distilled water until the TiO₂ membrane detached from its parent metal. Additional sonications in ethanol and distilled water (10 min each) were done to ensure the removal of remained TiO₂ nanotubes. Finally, the treated foil was re-anodized in the same electrolyte with similar voltage for the second time.

In post anodization, regardless of one or two step process, the samples would be sonicated in distilled water for 5 min. After drying them in N₂ stream, the samples were crystallized through annealing under air ambient for 3 h at 500 °C. For comparison purposes some of the samples would be left without going through the annealing process.

The morphological aspects of the anodized samples were viewed using field emission scanning electron microscope (FESEM). The obtained micrographs were subsequently processed using Image J software to determine the diameter and length of the nanotubes. On the other hand, X-ray diffractometer was used to characterize the structural properties of both as-anodized and annealed samples.

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