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Short communication

Enhancing the performance of free-standing TiO_2 nanotube arrays based dye-sensitized solar cells via ultraprecise control of the nanotube wall thickness

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HIGHLIGHTS

- Fabricated free standing TiO_2 nanotube array with wall thickness between 10 and 30 nm.
- Investigated the dependence of DSC performance on nanotube wall thickness.
- Identified an optimal TiO_2 nanotube wall thickness at 450 ALD cycles for DSC.
- The best efficiency 4.65% obtained at 450 cycles is 1.8 times of that at 250 cycles.

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ABSTRACT

Free standing TiO_2 nanotube (TNT) array film was fabricated via template-assisted Atomic Layer Deposition (ALD) in nanoporous anodic alumina templates followed by alumina dissolution. Effect of TiO_2 nanotube wall thickness on the photovoltaic performance was studied on the dye sensitized solar cells with the TNT wall thickness precisely controlled by ALD between 250 and 550 cycles. The results show that the photovoltaic performance can be improved by optimizing the TNT wall thickness. A thick enough tube wall is crucial for forming space charge layer to allow efficient charge separation, but too thick wall thickness will reduce dye loading relatively. A photovoltaic energy conversion efficiency of 4.65% was obtained on the device with 450 ALD cycles of TNT wall thickness, which is about 1.8 times of that obtained with 250 cycles thickness.

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1. Introduction

Dye-sensitized solar cells (DSC) have attracted substantial interests in the past decades due to its low cost and high throughput manufacturing potential for large scale industrial applications [1–3]. Conventional DSCs are fabricated with mesoporous nanocrystalline TiO_2 photoelectrodes which have large internal surface areas for absorbing light-sensitive dyes. Upon illumination, electrons will be excited from the light-sensitive dyes and then injected into the wide bandgap TiO_2 nanostructured materials. However, conventional DSCs rely on a trap-limited diffusion mechanism for electron transport and collection in the mesoporous TiO_2 nanostructured materials [4]. The process is slow and inefficient which limit the photo conversion efficiency of the solar cells.

Highly ordered TiO_2 nanotube (TNT) electrodes have been demonstrated with better photovoltaic performance due to increased charge collection efficiency and enhanced light trapping ability [5,6]. Another major advantage of such one-dimensional nanostructure over planar geometries is that they can decouple the directions of light absorption and charge carrier collection [7–9]. However, the physical parameters of the TiO_2 nanotube arrays can be an obstacle because both the electric and photo property would be affected by the morphology [10–13]. It is especially important to control the physical parameters for optimizing the device performance of solar cells. Past research has successfully demonstrated the strong dependence of solar cell efficiency on nanotube length, in general with increased power efficiency on longer nanotubes [14,15]. It was estimated the electron diffusion length in TNT is of the order of 100 μm on titanium foil by considering the electron diffusion coefficient and lifetime as a function of electron quasi Fermi level [15], which indicate that electron collection in TNT arrays with varied tube lengths are all efficient and the improvement should contribute to dye loading and light absorption enhancement from the increased length. Therefore, it's reasonable to estimate that the main electrical

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obstacle may originate from the electron injection process through the interface of dye and TiO_2, which are strongly affected by the TNT wall thickness [16,17]. However, it was rarely investigated experimentally before because of the challenges in ultraprecise control of the TNT wall thickness in nanometer scale.

In order to obtain nanotubes which are particularly favorable to achieve efficient charge injections with reduced charge recombination, ultraprecise control of the wall thickness of TiO_2 nanotubes will be highly desirable. To date, highly ordered TNT arrays are generally synthesized through two approaches: 1). Self-assembled method such as Ti electrochemical anodization [18–20], hydrothermal growth [21], etc., and 2). Template-assisted approaches usually through nanoporous anodic alumina oxide (AAO) template [22,23]. Although self-assembled method provide a more direct route to the arrays, but template assisted approach could provide more precise control of the tube dimensions especially combined with Atomic Layer Deposition (ALD) [24–26]. ALD is a self-limiting nanotechnology which operates in a cyclic process and can fabricate uniform and conformal layers at atomic scale [27–29], which satisfy the requirement of ultraprecise wall thickness control for the nanotube geometric study. Past research has applied ALD on a large number of metal oxides (such as ZnO, TiO_2, ZrO_2, SnO_2 and NiO) for new electrode development [28,30].

In this paper, we use free standing TiO_2 nanotube arrays as a model material for DSC fabrication and conduct a quantitative study on the effect of the TNT wall thickness on the solar device performance. The TNT wall thickness is controlled by various ALD deposition cycles in the range of 10–30 nm. From these results, we demonstrate that the efficiency of TiO_2 nanotube array based DSC can be improved by optimizing the wall thickness. An energy conversion efficiency of 4.65% was obtained using the TNT with 450 cycles TiO_2 deposition, which is about 1.8 times of that obtained using the TNT with 250 cycles. This geometric study of TiO_2 nanotube arrays can offer a strategy toward optimizing the energy conversion efficiency with other semiconductor materials in solar cell fabrication.

2. Experimental

2.1. Free-standing TiO_2 nanotube membrane fabrication

The TiO_2 nanotube was fabricated through a template ALD technique. Commercial AAO membranes with 200 nm pores (Anodisc, Whatman) were used as the template. Briefly, the AAO template was attached on a Si substrate with the open end facing up. Diffusion mode of a Cambridge Nanotech S100 ALD system was used to deposit TiO_2 into high aspect ratio AAO nano pores. HTDMA and

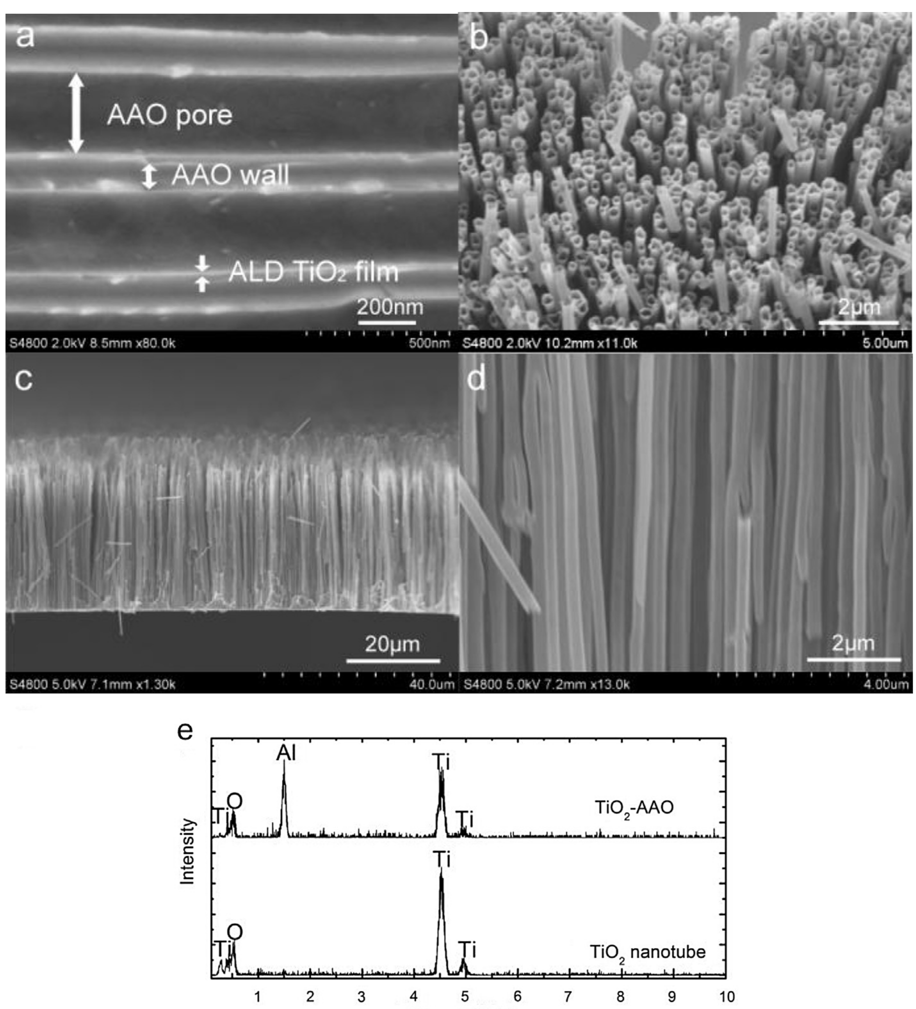

Fig. 1. (a) Cross section image of TiO_2 film deposited into AAO pores (b) top view of the obtained TiO_2 nanotube arrays after AAO etching. (c) Low magnification and (d) high magnification cross section image of TiO_2 nanotube array film. (e) EDS spectrum of TiO_2 nanotube before and after etching AAO.

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