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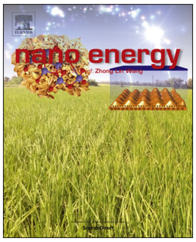

RAPID COMMUNICATION

Improving the photovoltaic performance and flexibility of fiber-shaped dye-sensitized solar cells with atomic layer deposition

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Abstract
A conformal and thin TiO_2 film fabricated with atomic layer deposition (ALD) improves the performance of fiber-shaped dye-sensitized solar cells (FDSSCs). Electrical contact at Ti/TiO_2 is improved through insertion of a uniform and pinhole-free TiO_2 film. The film thickness varies from 5 nm to 40 nm, and the high power conversion efficiency of 7.41% is achieved from a typical device with a 15 nm TiO_2 film and a 10 μm TiO_2 nanoparticles layer. The compact and high-quality TiO_2 film improves Ti/TiO_2 interfacial property. Surface modification hastens electron transport, which thus improves the charge collection efficiency. Both photocurrent density and bending performance are significantly enhanced compared with those of the corresponding FDSSCs without ALD treatment.

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Introduction

Dye-sensitized solar cells (DSSCs) are potential photoelectric conversion devices because of their low cost and simple device fabrication with the best conversion efficiency of more than 12% [1-4]. Researchers have recently reported a novel type of low-dimensional fiber-shaped DSSCs (FDSSCs)

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[5-10] that are fabricated on conducting fibers and have similar advantages as conventional planar DSSCs. The unique advantages of fiber-shaped devices, such as being transparent conductive oxide free and their 3D light harvesting capability, also allow their application to rapidly developing fields of wearable and/or portable electronic devices [11,12]. Moreover, FDSSCs can exhibit excellent bending and easy shape-changing abilities while in use. Improving flexibility and stability is highly important in the application of FDSSCs.

The power conversion efficiency (PCE) of FDSSCs has been gradually improved in recent years [13-15]. The most facile photoanode fabrication method is repeated dip-coating of Ti wires in TiO_2 nanoparticle (NP) suspensions until the TiO_2 NPs layer on the Ti wire surface exceeds 20 μm in general [16]. Introducing a compact layer into solar cells can effectively improve device performance. A kind of semiconductor oxide with improved conductivity, such as SnO_2, is used as a compact layer to construct a SnO_2/TiO_2 junction on the Ti wire substrate; thus, the conversion efficiency of the entire device increases to 5.8% because of the recombination shielding effect [17]. The PCE of FDSSCs reaches up to 7.2% with a 3 μm TiO_2 blocking layer synthesized by sintering of titanium isopropoxide on Ti wires and another 10 μm TiO_2 NPs layer [18]. Inserting a blocking layer does not only reduce the usage of TiO_2 NPs but also improves the PCE of FDSSCs, retains the high efficiency during bending, and results in improved flexibility. Nevertheless, electron transport kinetics caused by an effective compact layer on the substrate surface during FDSSCs fabrication has been insufficiently reported.

This study proposes a new method of forming a pinhole-free and dense TiO_2 blocking layer on a Ti wire surface because of the novel property of the precise growth of a TiO_2 compact layer with atomic layer deposition (ALD). ALD has attracted increasing attention because of its self-limited growth process to generate a conformal and ultrathin film [19-21]. Generally, an ALD process cycle comprises sequential alternating pulsing precursors into a vacuum chamber with the use of a carrier gas, as well as an intermediate purging step during two precursor pulses aimed to remove any excess precursors and reaction by-products. Precursors are loaded with carrier gas like nitrogen to avoid forming thick oxide layers. Film deposition is performed by repetition of the cycle until the desired film thickness is achieved. An excellent advantage of ALD against other deposition methods is its uniform deposition on various 3D topographies. For example, although a TiO_2 film can be formed through the sputter-coating method, deposition can be generally conducted on a planar surface. In addition, a sputtered film comprises 10–15 nm TiO_2 NPs that hardly covers the surface completely [22]. Another general way of forming a TiO_2 blocking layer is to sink the substrate into a $TiCl_4$ aqueous solution. However, $TiCl_4$ treatment produces a TiO_2 film that comprises about ten nanometers rod-shaped particles after annealing at 500 °C for 30 min [23,24]. Hence, the ALD technique is considered a facile way to precisely achieve a conformal, pinhole-free, and ultrathin film deposition on round Ti wires.

The photoanode in this study is uniformly coated by a compact and ultrathin TiO_2 film with the use of ALD before it absorbs TiO_2 NPs, as shown in Figure 1. ALD allows the film thickness to directly influence the performance of the assembled FDSSCs based on the aforementioned photoanode. The device improves its efficiency, stability, and flexibility with a thinner TiO_2 NPs layer. The characteristics and effects of a conformal TiO_2 film on charge transfer and collection are investigated comprehensively. The results show that the photocurrent significantly increases as the film thickness reaches up to 15 nm, and PCE improves by 26% to 7.41%. By contrast, the required TiO_2 NPs layer decreases by half.

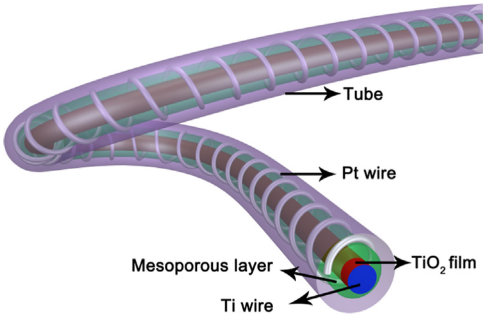

Figure 1 Schematic of the FDSSC structure with the conformal TiO_2 film deposited with ALD.

Materials and methods

Preparation of the photoanode

A Ti wire (diameter: 250 μm, Alfa Aesar) was ultrasonicated in acetone and ethanol respectively for 15 min before usage. After the Ti wire was washed with deionized water several times, the titanium dioxide film on the Ti wire was deposited via ALD inside a Picosun Sunale R-200 reactor at $T = 300$ °C with N_2 as both the carrier and purge gas. The TiO_2 film thicknesses were controlled by regulation of the deposit time. Deposition was performed with successive pulses of titanium tetrachloride (TDA, Aldrich, Germany, 25 °C) and deionized water (18.2 MΩ, 25 °C) with nitrogen as the carrier gas (150 sccm). After the 100 ms TDA or 100 ms H_2O pulse, the precursors were confined inside an ALD reactor for 2 s to ensure their complete exposure inside the Ti wire surface nanostructures. All excess precursors and reaction by-products were purged out for 6 s. The thickness of TiO_2 film was measured by operation of similar cycles on a silicon wafer via spectroscopic ellipsometry, and the growth rate was found to be ~0.066 nm/cycle.

For comparison, photoanodes were fabricated on both as-treated Ti wires and common Ti wires without ALD treatment by dip-coating these wires in TiO_2 suspensions prepared according to a previous report [25]. Different TiO_2 NPs layer thicknesses can be easily obtained by controlling the dip-coating times.

The electrode was sintered at 500 °C for 30 min and subsequently sensitized with 0.5 mM N719 (purchased from Dalian Heptachroma, China) in ethanol for 24 h at room temperature.

Assembly of fiber-shaped solar cells

The devices were fabricated according to previous reports [10,26]. Briefly, a Pt wire (diameter: 50 μm) served as the

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