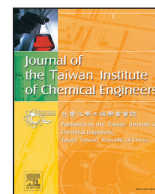




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Influence of photoanode architecture on light scattering mechanism and device performance of dye-sensitized solar cells using TiO₂ hollow cubes and nanoparticles

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ABSTRACT

Herein, we report the impact of light scattering mechanism on photovoltaic and photoelectrochemical performance of dye-sensitized solar cell (DSC) devices composed of TiO₂ nanoparticles and hollow cubes. DSCs are designed by two different light scattering modes (i.e., mode I in form of single layer electrode containing nanoparticles and hollow cubes and mode II in the form of double layer electrode comprising active and scattering layers made of nanoparticles and mixtures of nanoparticles and hollow cubes, respectively). The synthesized anatase-TiO₂ hollow cubes (200–400 nm) and nanoparticles (15–30 nm) are employed to enhance the optical length and light harvesting of photoanodes, respectively. Although the charge transfer resistances at Pt/electrolyte (R_{Pt}) and TiO₂/dye/electrolyte (R_{ct}) interfaces are decreased by mode I, it is not an appropriate approach for efficiency improvement of the devices due to hindering electrolyte diffusion and decreasing fill factor. In contrast, the photovoltaic performances of DSCs designed by mode II are successfully enhanced due to remarkable decrease in R_{Pt} and R_{ct} . Not only mode II improves light scattering but also it makes a balance between electron transfer and dye sensitization, leading to an increase in cell efficiency up to 9.31% compared to 7.0% for the reference device composed of pure nanoparticles.

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1. Introduction

Dye-sensitized solar cells (DSCs) are regarded as one of the most promising alternatives to the conventional Si-based solar cells owing to their relatively high efficiency, low production cost and environmentally friendly fabrication process. DSC devices can be prepared in various forms such as flexible, polymeric, bioderived photoanodes and polymer electrolytes [1,2]. Furthermore, the other advantage of DSCs such as capability of effectively performance in dark conditions (such as in the dawn and dusk or in cloudy weather) makes them an excellent choice for indoor applications [3]. During the past decades, many efforts have been focused on improvement of DSCs efficiency by the development of their components and materials (e.g., photoanode, electrolyte, dye and counter electrode). In DSCs, the photoanode as the key component has a notable impact on device efficiency. Light scattering and harvesting, dye adsorption, electron injection and collection are the most important parameters of the photoanode influence on photovoltaic characteristics of the device. Among oxide semiconduc-

tor materials are generally used as the photoanode of DSCs, TiO₂ is a favorable compound due to its unique structural and photoelectric properties as well as nontoxicity, biocompatibility and low cost. TiO₂ nanostructures have excellent properties for photocatalysis, sensing, self-cleaning coating, and photovoltaic applications [4–8]. Moreover, among TiO₂ crystal structures the anatase phase is expected to show higher performance for DSC applications due to its active surface chemistry, high electron mobility, and large band gap energy [9]. TiO₂ shows two different characteristics of hydrophilic and hydrophobic for DSC application [10]. In order to reach high conversion efficiency, high specific surface area, fast electron transport, and outstanding light-scattering properties play crucial roles in enhancement of DSCs performance. However, it is generally difficult to find all these factors together at the same time. As an alternative to overcome this problem, multilayer structures containing TiO₂ nanoparticles as the active layer and TiO₂ nanostructures with light scattering capability such as hierarchical flowers [11], dandelion-like particles [12], rod like nanocrystals [13], microspheres [14], nanotubes [15] and hollow spheres [16] as the top layer have been employed. It has been reported DSCs made of three-dimensional (3D) TiO₂ structures show higher power energy conversion efficiency than those composed of 1D TiO₂ morphologies [17,18]. TiO₂ hollow cubes, as 3D structures, have been

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Table 1Preparation conditions of solvothermally grown TiO₂ hollow cubes (HCs).

Processing parameters	Temperature (°C)	Time (h)	Heating rate (°C/min)	Sample
Temperature	200	12	5	HC-200-12-5
	220	12	5	HC-220-12-5
Time	200	24	5	HC-200-24-5
	200	48	5	HC-200-48-5
	200	72	5	HC-200-72-5
Heating rate	200	12	1	HC-200-12-1
	200	12	2.5	HC-200-12-2.5

introduced as anode material for DSC and lithium ion battery applications [19]. For example, Shi et al. [20] prepared double layer DSCs with efficiency of 6.4% made of P25 nanoparticles as the active layer and TiO₂ hollow boxes as the scattering layer. Niu et al. [21] fabricated multilayer DSCs with various arrangements of layers made of TiO₂ hollow cubes and nanoparticles. They obtained the highest cell efficiency of 6.4% for the DSC prepared by four layers of P25 and two layers of hollow cubes. Triple layer DSC composed of P25 nanoparticles as the first layer, TiO₂ nanotube arrays as the middle layer and TiO₂ hollow boxes as the upper layer reported by Hu et al. [15] showed efficiency of 6.0%. So far, no comprehensive study has been carried out on the impact of light scattering mechanism (e.g., mode I in form of single layer electrode comprising nanoparticles and scattering particles and mode II in the form of double layer electrode containing active and scattering layers). For the single layer cells mixtures of TiO₂ nanoparticles and hollow cubes are prepared, whereas for the double-layer films an active layer composed of pure nanoparticles and scattering layer with mixtures of TiO₂ nanoparticles and hollow cubes is fabricated.

Herein, we proposed a concept of DSCs made of composite electrodes in two different light scattering arrangement modes (i.e., in forms of single- and double-layer films) using TiO₂ nanoparticles and hollow cubes for improvement of their light scattering, dye sensitization and electron transfer. In addition, the impact of phase composition of the composite electrodes (e.g., nanoparticle: hollow cube weight ratio) on photovoltaic performance of the devices was studied.

For this purpose, we synthesized TiO₂ nanoparticles and hollow cubes by solvothermal method. The effect of hydrothermal processing parameters such as process time, temperature and heating rate on morphology of hollow cubic structures was investigated.

2. Experimental

2.1. Synthesis of TiO₂ nanoparticles

We synthesized TiO₂ nanoparticles (NPs) by a combination of solvothermal and modified sol-gel methods analogous to procedure reported previously [22]. TiO₂ sol was first prepared using titanium tetraisopropoxide (TTIP) with a purity of 97% (Aldrich, UK) as a titanium source and 1-octanol with a purity of 99% (Merck, Germany) as solvent and deionized water. First, TTIP was mixed with 1-octanol and stirred for 30 min at room temperature to form transparent precursor solution. During stirring, deionized water was added dropwise to the solution with [H₂O]: Ti ratio of 3.3:1 in order to form TiO₂ sol. The prepared sol was transferred into 38 mL Teflon-lined stainless steel autoclave and heated at 180 °C for 4 h for hydrothermal process. Finally, the collected nanoparticles were washed with dilute ethanol several times and dried at room temperature.

2.2. Preparation of TiO₂ hollow cubes

We prepared TiO₂ hollow cubes (HCs) by template- and surfactant-free solvothermal reaction and subsequent calcination, analogous to the procedure previously reported [20]. However, we used TTIP as titanium precursor as an alternative to titanium tert-butoxide. In the first step, 1.6 mL hydrofluoric acid (HF) with purity of 38% (Aldrich, UK) was added into 15 mL acetic acids with purity of 99.8% (Merck, Germany) and stirred for 15 min. 5 mL TTIP was slowly added next to the solution and stirred for 30 min. The prepared solution was transferred into 38 mL Teflon-lined stainless steel autoclave, heated at 200 °C for 12 h and subsequently cooled naturally to room temperature. The collected precipitates (i.e., the white cubic TiOF₂) were washed with absolute ethanol three times, dried at 80 °C for 4 h and calcined at 500 °C for 2 h at a heating rate of 5 °C/min to achieve TiO₂ hollow cubes. The impact of solvothermal processing parameters such as process time, temperature and heating rate on morphology of hollow cubic structures was also studied (See Table 1).

2.3. Device construction

We fabricated two categories of DSC devices, with an average thickness of 22 μm, in order to study the impact of light scattering mechanism (i.e., in the forms of single- and double-layer electrodes made of mixtures of nanoparticles and hollow cubes) on the performance of the devices. The first category was consisted of five single-layer DSCs made of mixtures of TiO₂ NPs and HCs. The second category consisted of four double-layer electrodes containing the active layer made of pure NPs and the scattering layer composed of mixtures of NPs and HCs with various weight ratios (See Table 2). We also studied the influence of nanoparticles: hollow cubes weight percentage on photovoltaic characteristics of devices for each category. The photoanode electrodes were prepared using a homemade new formulated paste reported elsewhere [23]. Before deposition, FTO-coated glass substrates (1.5 cm × 2 cm and 7 Ω/sq) were washed with mild detergent, distilled water and subsequently with ethanol and acetone in an ultrasonic bath for 10 min. A compact thin layer of TiO₂ was deposited using TiCl₄ solution (0.04 M) followed by heat treatment at 450 °C for 30 min. Subsequently, different titania pastes were prepared using TiO₂ NPs and HCs with various weight percentages, deposited by spin coating and sintered at 400 °C for 2 h. A post-treatment with TiCl₄ solution was applied to the newly sintered TiO₂ electrodes and then annealed again at 450 °C for 30 min. For dye loading, the resulting photoanode electrodes were soaked in 0.3 mM ruthenium (II) dye (Ruthenium 535-bis TBA dye, or N719, Solaronix) solution for 18 h in a dark room. The dye-loaded photoanodes were rinsed with ethanol, dried in air and immediately used for photovoltaic measurements. The counter electrode was made by applying a drop of H₂PtCl₆ solution onto FTO and annealing at 450 °C for 30 min. The counter electrode and dye-adsorbed photoanode were attached

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