



Metal oxide sandwiched dye-sensitized solar cells with enhanced power conversion efficiency fabricated by a facile and cost effective method



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ARTICLE INFO

Keywords:

Metal oxide semiconductors
Dye-sensitized solar cells
TiO₂/ZnO photoanode
Pt-free CuO/RGO counter electrode
Electroplating

ABSTRACT

Developing efficient and cost effective photoanode and counter electro materials have been a persistent objective by a wide community for efficient, cost effective and stable dye-sensitized-solar cells (DSSCs). We have developed a unique and inexpensive way of co-electroplating-annealing method to synthesize metal oxides and employed it to prepare ZnO material on top of doctor bladed TiO₂ as photoanode and CuO on top of reduced graphene oxide (RGO) as counter electrode. By sandwiching these two electrodes with I⁻/I₃⁻ redox couple in-between we have shown the improvement of photovoltaic properties with ZnO barrier layer using Pt or cost effective CuO/RGO counter electrodes (CEs). This paper provides a comprehensive guide to prepare those electrodes cost effectively and fabricate metal oxide sandwiched DSSCs providing up to 6.91% power conversion efficiency with 26.5% enhancement compared to the conventional DSSCs with TiO₂ photoanode and Pt CE under AM1.5 simulated solar light. This work also addresses the reduction of fabrication cost of the cells to make them more economically viable as it is

1. Introduction

Among various forms of solar cells dye-sensitized solar cells (DSSCs) are of great interest due to good price to performance ratio, short payback time, abundance of constituent materials, less temperature dependence and integration possibility with other applications [1]. Although DSSCs offer significant economic and environmental advantages compared to the conventional photovoltaic devices there are still some challenges preventing the large scale commercialization [1,2]. Although the theoretical conversion efficiency of DSSCs is 20.2% [2,3], which is comparable to Si solar cells maximum achievable conversion efficiency has been 13% using panchromatic sensitizers so far [4,5].

A typical DSSC is composed of three major components, which are namely photoanode sensitized by dye molecules, electrolyte as redox couple and counter electrode as cathode, respectively. In a typical cell mesoporous TiO₂, electrolyte with I⁻/I₃⁻ redox couple and Pt are used as photoanode, mediator and counter electrode material, respectively [6,7] and the assembly of all those components was demonstrated for the first time by Gratzel in 1991 [7] and later on people in academia and industry started to pay great attention to this technology [8]. Since then significant efforts have been made on growing new materials to optimize photoanode, sensitizer, electrolyte and photocathode and

designing new device layer structures to enhance and achieve close to theoretical power conversion efficiency (PCE). Researcher around the world focus mostly on two parts. One is to minimize the recombination process and improving the charge transport ability by modifying photoanode [9] and another is to increase the stability and catalytic activity of counter electrode using novel materials [10]. Along with improving these two parts we should also take economic and environmental considerations into account and therefore PV cells should be composed of abundant and nontoxic materials and should be prepared by an easy and cost effective method.

Optimum photoanode layer, which is used as electron transporter in conventional DSSCs, exhibits the following properties. High surface area: increases the dye loading capacity and therefore the light harvesting efficiency. As the morphology of the structures varies from 3D (bulk) to 0D (quantum dot) through 1D (nanowire) and 2D (thin film, nanosheet, etc.) the specific surface area increases and therefore the number of dye adsorbed on the wide band gap material increases as well [9,11]. As a result of that more carriers are generated and current density of the cells are boosted. High electron mobility: increases the photogenerated charge transport towards the electrode. Although TiO₂ is a widely used material in DSSCs it has low electron mobility (0.1–4 cm² V s⁻¹) [11] limiting the photogenerated charge transport. In order to compensate this some of the other materials such as ZnO,

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SnO_2 , Nb_2O_5 can be employed [12–16]. For example, ZnO having much higher electron mobility ($200\text{--}300\text{ cm}^2\text{ V s}^{-1}$ for bulks and $1000\text{ cm}^2\text{ V s}^{-1}$ for single crystal nanowires) [17,18] can be used as a barrier layer in a synergetic manner with TiO_2 to increase the transport rate of the electrons. Being inert to the redox electrolyte: reduces the electron back recombination rate, which hinders the drop of open circuit voltage (V_{oc}) of the cell. Latter two properties determine the collection efficiency of the photogenerated carriers as given by $k_{trans}/(k_{trans} + k_{brec})$, where k_{trans} and k_{brec} represent electron transport rate and back recombination rate, respectively. Since the first property determines the number of photogenerated carriers all the properties of optimum photoanode play a crucial role to increase the open circuit voltage, short circuit current (I_{sc}) and PCE of the solar cells.

On the other hand, counter electrode (CE), another part of the sandwiched cell structure, works as an electron collector that transports electrons flowing through the load to the electrolyte and catalyzes the redox couples in the electrolyte to regenerate sensitizers. Optimum CE should exhibit high electro catalytic activity, high electrical conductivity, resistance to corrosion as well as high specific surface area and should be composed of abundant materials to bring the cost of the cells down using low cost preparation methods. Pt is a widely used CE material meeting the first two requirements but failing in the latter ones, as it is rare and expensive material and susceptible to corrosion by iodide electrolyte. Various materials such as single-walled carbon nanotubes, multi-walled carbon nanotubes (MWCNTs), mixture of those with graphene and some polymeric materials, NiO, FeS_2 , reduced graphene oxide (RGO)/MWCNTs/NiO, RGO/ Cu_2S and so on were employed as an alternative to the conventional Pt CE so far [19–29]. Some of those showed comparable performance while some of the others showed superior performance to the conventional Pt electrode. Scientists usually mix the materials with carbon allotropes to increase the electrical conductivity of the electrode as mentioned to be one of the most have property of the CEs [20,30–33].

In this paper, we have developed a unique co-electroplating-annealing growth method for metal oxides and demonstrated the growth of ZnO and CuO structures using this facile and cost effective method. ZnO on top of doctor bladed TiO_2 was utilized as hybrid photoanode and CuO on top of RGO prepared by modified Hummer's method was used as Pt substitute in counter electrode. As mentioned earlier people mostly focus on improving either photoanode or counter electrode to boost the power conversion efficiency of the solar cells and reduce the cost without sacrificing the efficiency, but we simultaneously work on the improvement of both sides to obtain enhanced power conversion efficiency of the cells using a more economic and environmental friendly method.

2. Experimental

2.1. Fabrication of photoanode and counter electrode

Before all the steps taken place transparent conductive oxide (TCO) glass substrates were thoroughly cleaned using the typical cleaning procedure [34]. All the processes to prepare both electrodes are illustrated in Fig. 1. In order to prepare photoanode first TiO_2 paste was prepared by dissolving P25 nanoparticles in sulfuric acid and then the paste was applied onto TCO substrate via doctor blade method. After that the substrate was annealed on the hotplate at $310\text{ }^\circ\text{C}$ for 30 min and became ready for the next step where we employed the electroplating method.

Electroplating solution was prepared by dissolving 60 mg of Zn (NO_3)₂ into 40 ml deionized water under continues stirring for 10 min. After that Cu wire and ITO substrate were immersed into the solution and those used as anode and cathode, respectively as seen in Fig. 1(b). By applying a small potential (3 V causes 50 mA/cm^2 current density) between those electrodes Zn ions are obtained in the solution and with the help of potential difference positively charged Zn ions move

towards negative potential.

Since we connect the negative pole of the power supply to the substrate 30 s later grayish Zn metal is deposited on the TiO_2 /TCO surface as illustrated in Fig. 1. It is important to note that the electroplating time determines the thickness of the metal layer and therefore the thickness of the metal oxide layer. Right after the electroplating procedure the substrate was transferred onto the hotplate at $310\text{ }^\circ\text{C}$ and leaved there for 30 min to react with oxygen to form ZnO materials on top of TiO_2 layer.

For the counter electrode side we first dip coated the substrate into RGO prepared by the modified Hummer's method as explained in [35] to cover the TCO substrate with graphene layer. After that continuous cuboid CuO thin film was deposited using exactly the same method used for ZnO. However, in order to deposit copper metal we used $\text{CuCl}_2\cdot 2\text{H}_2\text{O}$ as a Cu precursor. After covering substrate with metal species we put the substrate on $310\text{ }^\circ\text{C}$ hotplate to anneal the metal layer in ambient atmosphere. After some time (1 min found to be the ideal) we can see the color changes from orange-brown to black on the films, which is the indication of chemical reaction between Cu and oxygen forming CuO. According to our experiments, devices fabricated with longer deposition time for both electrodes show worse performance and the reason can be explained as follows; when the thickness get larger the chance for the free carrier movement through the long distance becomes low. Consequently, recombination process takes place and thus decreasing the power conversion efficiency. In addition, when the film layer gets thicker film cracking occurs due to the stresses and this causes to have some shunting phenomena. It is important to note that the indicated times are under the aforementioned conditions, but we can also control the thickness in a good precision by using a programmable current supply rather than the voltage supply.

2.2. Fabrication of the DSSCs

In this paper, we fabricated three different DSSCs to figure out the effects of each new layer. The fabricated cells have the following structural layers; Cell#1: TCO/ TiO_2 |I-/I3 redox couple|Pt/TCO, Cell#2: TCO/ TiO_2 /ZnO |I-/I3 redox couple|Pt/TCO, and Cell#3: TCO/ TiO_2 /ZnO |I-/I3 redox couple|CuO/RGO/TCO. In the rest of the paper we are going to use Cell#1, Cell#2 and Cell#3 as a notation to indicate the aforementioned cell structures. After the preparation of photoanode as explained above it was immersed into N719 ethanolic dye solution at room temperature for 12 h, rinsed with ethanol and N_2 blow-dried. In order to construct DSSCs the working electrode (photoanode) and counter electrode were sandwiched together with a $30\text{ }\mu\text{m}$ Surlyn and then hot pressed at $110\text{ }^\circ\text{C}$ for 30 s to construct a monolithic structure. The iodine/potassium iodide electrolyte was injected from the hole on the back side via vacuum backfilling to complete the circuit and then it was sealed with Surlyn and thin cover glass by heating.

2.3. Characterization of structural, optical and photovoltaic properties

Morphology of the structures on different samples were characterized by using FEI QUANTA FRG 450 scanning electron microscope (SEM) at 15 kV accelerating voltage and chemical composition of the materials were identified using energy dispersive spectroscopy (EDS) detector embedded in the SEM microscope. Crystalline/structural properties and phase identifications were investigated by x-ray diffraction (XRD – Panalytical Empyrean) studies using $\text{CuK}\alpha$ radiation ($\lambda = 1.5406\text{ \AA}$). The optical properties were investigated using dual beam UV–VIS spectrometer (Shimadzu, UV1800) in the wavelength range of $300\text{--}1100\text{ nm}$ and the solid samples in the way of the light coming from the monochromator was used. Photovoltaic performance of the DSSCs were evaluated using Keithley 2400 source meter under AM1.5G simulated solar light and the cell size was adjusted to 0.16 cm^2 using a black dyed metal mask. The incident photon to current conversion efficiency (IPCE) was measured using a homemade IPCE apparatus and a

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