



In situ growth of zinc oxide nanoribbons within the interstices of a zinc stannate nanoplates network on compacted woven metal wires and their enhanced solar energy application

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ABSTRACT

A novel hybrid film is designed and prepared by in situ growth of ZnO nanoribbons in the interstices of Zn₂SnO₄ nanoplates network on the compacted woven metal wires through a simple process. The ZnO nanoribbons present in the ZnO/Zn₂SnO₄ composite film can enhance light harvesting, accelerate electron transport and induce a negative shift in the flat-band potential. Benefiting from its advantageous structure and composition, the ZnO/Zn₂SnO₄ film can be applied in many fields. For the flexible dye-sensitized solar cells (FDSSCs) employing optimized ZnO/Zn₂SnO₄ as a photoanode, the conversion efficiency reaches 2.41% corresponding to ~36.2% improvement relative to the Zn₂SnO₄ nanoplates-based FDSSCs. Moreover, ZnO/Zn₂SnO₄-based FDSSC shows relatively good mechanical stability and long-term stability, retaining 95.1% and 93.3% of its initial efficiency after ten consecutive bending tests and after 15 days under sunlight, respectively; Additionally, the immobilized ZnO/Zn₂SnO₄ on the metal wires exhibits 96.8% photocatalytic degradation efficiency against an organic dye under UV light, and the photocatalytic performance can be restored almost completely by a simple chemical treatment. More importantly, the in situ growth technique demonstrated in this work can be adopted to fabricate other composite oxides on flexible substrates with high curvature surfaces for additional practical applications in flexible devices.

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1. Introduction

The synthesis of semiconductors with controllable composition and morphology on flexible wire-shaped substrates has gained enormous attention because of the growing demand for lightweight, wearable integrated devices in practical applications [1,2]. For instance, conductive wire-based dye-sensitized solar cells show several promising advantages over the traditional rigid fluorine-doped tin oxide (FTO) glass-based solar cells, such as foldability,

stitchability, convenient transport and the capability to supply power for portable electronic devices [3]. In another example, immobilized photocatalysts prepared on a flexible woven wire mesh can be easily separated from the reaction medium and reused again after washing [4]. The use of conventional separation techniques, such as centrifugation or filtering, can then be omitted, avoiding the reduction of the photocatalyst mass in repeated photocatalytic operations. Unfortunately, the performance characteristics of flexible wire-shaped substrate-based DSSCs and photocatalysts are still inferior to those of the traditional forms [5].

In previous studies, the simple binary oxides TiO₂ and ZnO have been the most widely applied materials as photoanodes of DSSCs and photocatalyst [6–8]. Furthermore, some ternary oxides have been introduced in this field over the past few years [9,10]. Zinc stannate (Zn₂SnO₄), which has been reported to possess a larger electron mobility (10–15 cm² V⁻¹ s⁻¹) than TiO₂ (10⁻² cm² V⁻¹ s⁻¹) and excellent acid-base resistance, is especially notable in this

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regard [11,12]. The former feature results in the faster transport of photogenerated electrons, and the latter feature contributes to the improvement of the long-term stability of the DSSCs. Moreover, Zn_2SnO_4 has a longer electron lifetime and slower electron–hole recombination rate than TiO_2 in DSSCs [13]. Numerous experiments have shown that Zn_2SnO_4 is a good candidate material for highly efficient solar cells and photocatalysts [14,15]. Furthermore, current research has demonstrated that Zn_2SnO_4 could be coupled with a semiconductor that possesses different but matched energy level structure to form a heterojunction, achieving efficient charge separation and extended range of light absorption. To date, $\text{TiO}_2/\text{Zn}_2\text{SnO}_4$ [16], $\alpha\text{-Fe}_2\text{O}_3/\text{Zn}_2\text{SnO}_4$ [17], $\text{WO}_3/\text{Zn}_2\text{SnO}_4$ [17], $\text{ZnO}/\text{Zn}_2\text{SnO}_4$ [18], and $\text{SnO}_2/\text{Zn}_2\text{SnO}_4$ [19] composite heterostructures have been synthesized and showed superior photoelectric and photocatalytic performances compared to pure Zn_2SnO_4 . Among these semiconductor/ Zn_2SnO_4 compounds, ZnO and Zn_2SnO_4 are both well-known photoelectrodes and photocatalysts; when ZnO is coupled with Zn_2SnO_4 , the conduction band (CB) of ZnO is higher than that of Zn_2SnO_4 so that Zn_2SnO_4 could play the role of a sink for the photogenerated electrons [20]. This charge transfer process will promote the separation of the photogenerated electron–hole pairs by suppressing their recombination and will ultimately enhance the photoelectric and photocatalytic performances. Generally, these Zn_2SnO_4 composites were prepared by fabricating double-layered composite films with an under-layer and a light-scattering over-layer. The under-layer was composed of the low-dimensional nanomaterials owing to a high surface area of zero-dimensional nanoparticles or lower electron transfer resistance of a one-dimensional array [21,22]. Solid nanostructures with large sizes often serve as the over-layer because they are suitable for use as light-scattering layer to improve the light harvesting efficiency [23]. However, these bilayer electrodes were usually prepared by vertically stacking another metal oxide over the base metal oxide on the flat plate substrates. Such a configuration would increase the thickness of the film and therefore lead to the strong recombination and reduced light absorption of the under layer. Additionally, a higher internal/interfacial resistance would appear, arising from the imperfect interconnectivity between two layers. Furthermore, it was difficult to grow this complicated structure on some flexible wire-shaped substrates with high-curvature surface so that few studies have been conducted to date on semiconductor/ Zn_2SnO_4 for FDSSCs and immobilized photocatalysts [24].

Herein, we report an innovative structural design for introducing ZnO nanoribbons within the interstices of a Zn_2SnO_4 nanoplates network grown on compacted woven metal wires (CWMWs) and used as photoanodes of FDSSCs and of immobilized photocatalysts for the degradation of organic dye. We aim to introduce an appropriate amount of ZnO nanoribbons in the Zn_2SnO_4 nanoplates network by a simple in situ growth method in order to strengthen the interfacial contact of the two components. In this way, ZnO nanoribbons with a tunable length that can be adjusted from hundreds of nanometres to micrometres would serve as light-scattering centres and increase light scattering. At the same time, one-dimensional ZnO nanoribbons present in the composite film will promote electron transfer. More importantly, cascaded energy band matching could form among the CBs of ZnO , Zn_2SnO_4 and the substrates so that efficient electron transport can be expected. Various characterization methods were adopted to confirm our hypothesis and the feasibility of the preparation of the composite material on the compacted woven metal wires by the two-step process. The overall conversion efficiency and photocatalytic properties of the $\text{ZnO}/\text{Zn}_2\text{SnO}_4$ were significantly superior

to those of the Zn_2SnO_4 nanoplates.

2. Experimental

2.1. Electrode preparation

Fabrication of Zn_2SnO_4 nanoplate arrays on CWMWs: the CWMWs (4 cm × 2 cm) were dipped into acetone and cleaned with an ultrasonic washer for half an hour. Zn_2SnO_4 nanoplate arrays on CWMWs were fabricated by a precursor template technique. For the preparation of the dense Zn_2SnO_4 seed layer, 0.05 mol of ZnCl_2 and 0.025 mol of $\text{SnCl}_4 \cdot 5\text{H}_2\text{O}$ were added in a 100 mL ethyl alcohol with ultrasonic vibrating. The mixture was kept at 40 °C for 10 h in a water bath. The cleaned meshes were impregnated in the above-mentioned sol for several minutes, taken out, and dried by infrared irradiation; this process was performed twice, and the meshes were finally sintered at 500 °C for 10 min in air. Next, the treated CWMW was placed into a 30 mL aqueous solution of $\text{SnCl}_4 \cdot 5\text{H}_2\text{O}$ (0.6 mmol), $\text{Zn}(\text{CH}_3\text{COO})_2 \cdot 2\text{H}_2\text{O}$ (1.2 mmol) and NaOH (7.25 mmol) in a 50 mL Teflon-lined autoclave. The reaction vessel was heated in an electric oven at 200 °C for 10 h, and then the mesh was taken away from the cool vessel and rinsed with deionized water. The dried samples were calcined at 500 °C for 30 min in air (labelled “P0”). For comparison purposes, the electrode prepared without the seed layer under the same experimental conditions was labelled “P”.

Fabrication of $\text{ZnO}/\text{Zn}_2\text{SnO}_4$ hybrid film on CWMWs: ZnO seed solutions were prepared by dissolving 0.4 mmol of $\text{Zn}(\text{CH}_3\text{COO})_2 \cdot 2\text{H}_2\text{O}$ and NaOH with three concentrations: (1) 1 mM (2) 2 mM and (3) 3 mM in 100 mL of alcohol under rapid stirring. Zn_2SnO_4 nanoplate arrays on CWMWs were dipped in the ZnO seed solution for 1 h, taken out, dried and then calcined at 350 °C for 10 min in the air to prepare the ZnO seed layer. The modified Zn_2SnO_4 nanoplate on the CWMWs was obliquely immersed into 30 mL of water containing 4.5 mmol of $\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ and 1.5 mmol of hexamethylenetetramine (HMT) in a 50 mL Teflon-lined autoclave. The sealed autoclave was kept at 120 °C for 3 h. The hybrid film was heated at 500 °C for 30 min in air. The corresponding three samples prepared in the seed solution with different concentrations were labelled “P1” “P2” and “P3”, respectively.

2.2. Materials characterization

Scanning electron microscopy (SEM, FEI NOVA NanoSEM230, USA) was used for the examination of the particle morphology and size. X-ray diffraction (XRD, 200 mA, 40 kV) with $\text{Cu-K}\alpha$ radiation and transmission electron microscopy (TEM, JEOL 3010, Japan) were also applied for further investigations of the phase and structure of the samples. The adsorbed dye amount on the surface of the film was quantified by soaking the flexible photoanodes into a 0.1 M NaOH aqueous solution at 25 °C for 2 h. The concentration of dye in solution was examined using a UV–vis spectrophotometer (UV-2550, Shimadzu). The photocurrent–voltage characterization (J - V) was performed using an Oriel 92251A-1000 sunlight simulator (AM 1.5 globe, 100 mW cm^{-2}) with an active area of 0.4 cm^2 . The UV–vis diffuse reflectance spectrum was recorded using a UV–vis spectrophotometer (UV-2550, Shimadzu). The incident-photon-to-current conversion efficiency (IPCE) spectra were obtained using a PEC-S20 instrument (Pecell). Intensity-modulated photocurrent/photovoltage spectra (IMPS/IMVS) were conducted using an electrochemical workstation (Zahner, Zennium).

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