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CdS quantum dots sensitized single- and multi-layer porous ZnO nanosheets for quantum dots-sensitized solar cells

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ABSTRACT

The photoelectrodes of quantum dots-sensitized solar cells (QDSCs) combining porous ZnO nanosheets and CdS quantum dots (QDs) were constructed and examined. The single- and multi-layer porous ZnO nanosheets were in situ grown on ITO conducting glass by electrodeposition at the deposition potentials of −0.5 and −0.8 V (vs. SCE), respectively, followed by a heat treatment. CdS QDs attached on porous ZnO nanosheets were synthesized by successive ionic layer adsorption and reaction method. The conversion efficiency achieved by the QDSC based on CdS QDs-sensitized multi-layer porous ZnO nanosheets (M–ZnO/CdS electrode) was 1.16% which was much higher than that based on CdS QDs-sensitized single-layer porous ZnO nanosheets (S–ZnO/CdS electrode). This was due to the more intense absorption of visible light by the M–ZnO/CdS electrode than that by the S–ZnO/CdS electrode.

1. Introduction

Dye-sensitized solar cells (DSSCs) are of great interest as an alternative to conventional silicon photovoltaics due to its cost-effective manufacture and low environmental impact [1–3]. The photoelectrode of DSSCs usually includes a mesoporous oxide film (such as TiO_2, ZnO and SnO_2) consisting of interconnected nanoparticles and a monolayer of dye molecules (act as sensitizers) coated on the surface of a mesoporous oxide film [1–3]. In addition to organometallic or organic dyes, semiconductor nanocrystals (also known as quantum dots (QDs)) have recently been considered to be promising sensitizers, so-called quantum dots-sensitized solar cells (QDSCs). In general, QDs have some important advantages over dyes, such as the adjustable band gap, high extinction coefficients and large intrinsic dipole [4–6]. Furthermore, the quantum confinement effect of QDs makes it possible to generate multiple electron–hole pairs per photon through impact ionization effect, which could be exploited to improve the efficiency of QDSCs in the future [7].

Very recently, CdS and CdSe QDs have been used to modify one-dimensional (1D) nanostructure arrays (nanowire or nanotube arrays) to obtain QDSCs due to that 1D nanostructure arrays can improve electron transport by avoiding particle-to-particle hopping in mesoporous networks [8–10]. However, insufficient internal surface area of 1D nanostructure arrays limits the conversion efficiency at a relatively low level owing to the insufficient QDs loading and light harvesting [8–10]. In addition to 1D nanostructure arrays, the two-dimensional (2D) nanostructures (nanosheets or nanobelts) in situ grown on conducting substrate are also expected to improve the electron transport. Besides, 2D nanostructures can offer a large specific surface area for the deposition of sensitizers (dyes or QDs) [11,12]. The photoelectrode of DSSCs based on ZnO nanosheets has achieved a really high performance [11,13] and it is suggested that the higher performance photoelectrode of DSSCs or QDSCs might be obtained if nanosheets could be made into porous structures, because porous nanosheets can facilitate the diffusion of electrolyte and offer a larger specific area [14].

In this communication, the porous ZnO nanosheets with single- and multi-layer structures were in situ grown on conducting substrate (ITO glass) by electrodeposition at two different deposition potentials followed by heat treatment. These photoelectrodes were then combined with CdS QDs by depositing the QDs using the successive ionic layer adsorption and reaction (SILAR) method.

2. Experimental details

2.1. Preparation

ZnO nanosheets are in situ grown on ITO conductive glass by electrodeposition in the electrolyte containing 0.1 M $Zn(NO_3)_2$ and 0.1 M KCl [15]. Electrodeposition experiment was carried out in a two-electrode cell with a piece of pure zinc as counter electrode. Electrodeposition temperature was controlled at 60 °C by immersing the cell in a water bath and electrodeposition was carried out under potentiostatic condition. The duration of electrodeposition is 30 min. The single and multi-layer nanosheets were electrodeposited at the deposition potentials of −0.5 V and −0.8 V (vs. SCE), respectively. The as-deposited ZnO nanosheets were sintered at 450 °C for 30 min in air atmosphere to obtain porous structure.

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The single- and multi-layer porous ZnO nanosheets were sensitized with CdS QDs by SILAR method. The SILAR method involved the successive immersion of single- and multi-layer porous ZnO nanosheets in the aqueous solutions of 0.05 M $Cd(NO_3)_2$, water, 0.05 M Na_2S and water to deposit CdS QDs [16].

2.2. Characterizations

The X-ray diffraction (XRD) pattern was recorded on a diffractometer with monochromatized Cu Ka radiation. The field-emission gun scanning electron microscope (FESEM, Apollo 300) with an energy dispersive X-ray (EDX) spectroscopy system was used to evaluate the morphology and elemental composition of the samples. A transmission electron microscope (TEM, JEM-2100F) was used to qualitatively confirm the presence of the CdS QDs on the ZnO nanosheets. Ultraviolet–Visible (UV–Vis) absorption spectra of photoelectrodes were recorded on a GBC spectrometer (Cintra 10e).

To study the photoelectrochemical performance of electrodes, a two-electrode photoelectrochemical cell was constructed with CdS QDs-sensitized porous ZnO nanosheets as the working electrode and a platinum foil as the counter electrode, respectively. The electrolyte was the aqueous solution of 2 M Na_2S and 3 M S [17]. A xenon lamp (500 W) with the illumination intensity of ~100 mW cm^{-2} and a wavelength range of 380–700 nm was used as a light source. A CHI 600A electrochemical analyzer was employed to record the current and voltage obtained under illumination with an active area of 0.25 cm^2 and dark condition.

3. Results and discussion

Fig. 1(A) shows the ZnO nanosheets electrodeposited at the potential of −0.5 V (vs. SCE) followed by heat treatment. It can be readily indicated that the nanosheets are hexagonal in shape and the porous and thickness of ZnO nanosheets is from 100 to 300 nm. The cross-section view (inset in Fig. 1(A)) shows that the ZnO nanosheets deposited at −0.5 V (vs. SCE) is a single-layer structure and all nanosheets stand almost vertically on the substrate with the height from 2 to 3 μm. Fig. 1(B) shows the top view of the ZnO nanosheets electrodeposited at the potential of −0.8 V (vs. SCE) followed by heat treatment. It can be easily seen that the nanosheets are also hexagonal in shape and is highly porous with the thickness below 100 nm. The nanosheets seem to stand with different angles to the substrate and some nanosheets are almost parallel to the substrate. The cross-section view (inset in Fig. 1(B)) shows that the ZnO nanosheets deposited at −0.8 V (vs. SCE) is a multi-layer structure and the thickness of the multi-layer ZnO nanosheets is calculated to be about 12–15 μm. It is expected that this multi-layer nanosheet structure would be favorable for light absorption because the light that passes through the gaps between adjacent nanosheets at the lower layer might be absorbed by the upper layer. Fig. 1(C) is a high-magnification SEM image of a single porous ZnO nanosheet, revealing that many nanopores are dispersed uniformly in the nanosheets with the dimension about tens of nanometers. The XRD pattern (Fig. 1(D)) of the multi-layer porous ZnO nanosheets demonstrates that the nanosheets are only composed of hexagonal ZnO.

Fig. 2(A) shows a SEM image of CdS QDs-sensitized multi-layer porous ZnO nanosheets (M–ZnO/CdS electrode). It can be readily indicated that the deposition of CdS QDs does not obviously change the morphology of porous nanosheets and the hexagonal porous nanosheets can still be observed. The EDX spectroscopy in Fig. 2(B) clearly shows the peaks of S and Cd elements accompanying the peaks of Zn and O elements, confirming the presence of Cd and S on the ZnO surface. Fig. 2(C) presents a TEM image of a single CdS QDs-sensitized ZnO nanosheet, indicating that the nanosheet is still hexagonal in shape and highly porous after the deposition of CdS QDs. A HRTEM image taken from the white square area in Fig. 2(C) is shown in

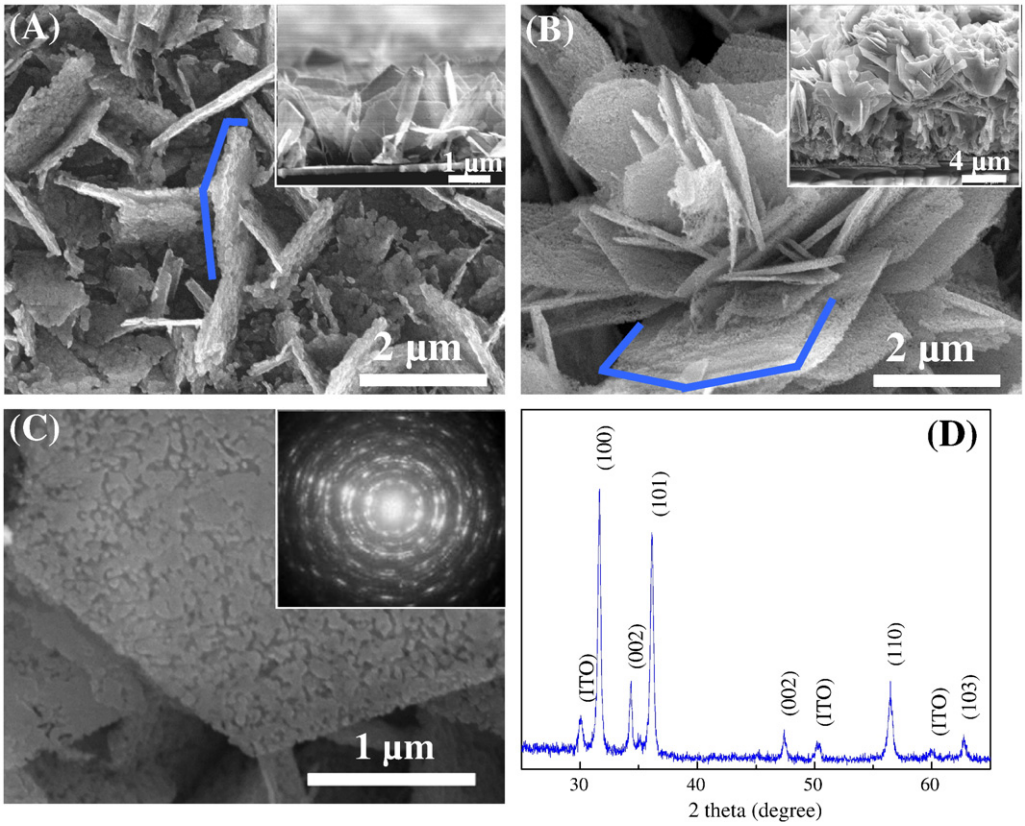

Fig. 1. SEM images of (A) single- and (B) multi-layer porous ZnO nanosheets, (C) high-magnification SEM image of a single porous nanosheet, and (D) XRD pattern of multi-layer porous ZnO nanosheets. The insets in (A) and (B) are the corresponding cross-section views. The inset in (C) is the electron diffraction pattern of a single porous nanosheet.