Journal of Power Sources 371 (2017) 96–105

Contents lists available at ScienceDirect

Journal of Power Sources

journal homepage: www.elsevier.com/locate/jpowsour

A membrane electrode assembled photoelectrochemical cell with a solar-responsive cadmium sulfide-zinc sulfide-titanium dioxide/mesoporous silica photoanode

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HIGHLIGHTS

- A MEA based PEC is developed for electricity generation by degrading organics.
- Adding SBA-15 into the photoanode increases specific surface area and pore volume.
- Enhanced transport at the photoanode with 15% SBA-15 results in the best performance.
- Parametric study is performed to optimize the performance with the MEA design.

ARTICLE INFO

Keywords:
Photoelectrochemical cell
Membrane electrode assembly
Mesoporous silica
Air-breathing cathode
Cell performance

ABSTRACT

In this work, a membrane electrode assembled photoelectrochemical cell (PEC) is developed for the electricity generation by degrading the organic compounds. The photocatalyst is prepared by the incorporation of mesoporous silica SBA-15 into TiO_2 and the photosensitization of CdS-ZnS to enhance the photoanode performance, while the cathode employs the air-breathing mode to enhance the oxygen transport. The experimental results show that the developed PEC exhibits good photoresponse to the illumination and the appropriate SBA-15 mass ratio in the photoanode enables the enhancement of the performance. It is also shown that the developed PEC yields better performance in the alkaline environment than that in the neutral environment. Increasing the KOH concentration can improve the cell performance. There exist optimal liquid flow rate and organics concentration leading to the best performance. Besides, it is found that increasing the light intensity can generate more electron-hole pairs and thus enhance the cell performance. These results are helpful for optimizing the design.

1. Introduction

Each year, a significant quantity of wastewater that contains abundant chemical energy is discharged into water body, causing severe water pollution and threatening the human health [1]. To resolve this challenging issue, several wastewater treatment technologies have been developed, such as the cavitation [2], photocatalytic oxidation [3], photocatalytic ozonation [4], Fenton's chemistry ozonation [5] and so on [6]. However, these technologies mainly focus on the efficiency and velocity of degrading organic compounds, leading to a large amount of energy to be wasted [7,8]. Hence, recovering the potential chemical energy stored in wastewater and turning it into a useful energy form are essential to simultaneously alleviate the environmental and energy problems. Photoelectrochemical cell (PEC) is one of such kind technologies to meet this demand [9]. Upon illumination, the degradation of a variety of organic pollutants into nontoxic materials and electricity generation can be realized simultaneously [10]. Because of this feature, the PEC has become an increasing-attention research field [11].

Previous studies on the PEC mainly focused on the development of highly-efficient photoanode catalysts [12–14]. For instance, Liu et al. [12] developed the photocatalytic fuel cell (PFC) with a TiO_2-nanotube-

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http://dx.doi.org/10.1016/j.jpowsour.2017.10.049
Received 13 April 2017; Received in revised form 5 October 2017; Accepted 16 October 2017
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array-based photoanode to use refractory organic compounds as substrates for electricity generation, which was also a typical PEC system. Antoniadou et al. [13] developed the visible-light responsive photocatalyst by adding CdS-ZnS for water splitting in the presence of sacrificial agents. Although significant progress on the photoanode catalyst has been made, the performance of existing photoanode is still limited by low specific surface area [15]. Hence, mesoporous materials with large specific surface area and pore volume have been incorporated into the photocatalysts to increase the specific surface area, the dispersion of nanoparticles, and the photocatalytic activity [16,17]. In addition to the photoanode catalysts, the performance of the PEC is also greatly influenced by the reactor design. Traditional PECs usually employ single-compartment and double-compartment designs [11,18]. For single-compartment PEC, the mixture of organics and electrolyte is supplied to the electrodes [19,20], so that the organics reaching to the cathode can form the mixed potential and thus lower the cathode performance [21]. For double-compartment PEC, the photoanode and cathode are separated by an ion exchange membrane [22,23], in which large distance between the electrodes causes the problems of large ion transfer resistance and gross structure. Moreover, the oxygen required by the cathodic reactions usually comes from the oxygen dissolved electrolyte [24]. In this case, not only the oxygen transport is limited due to low solubility of oxygen but also the ancillary devices for the oxygen supply are required. To enhance the oxygen transport and eliminate ancillary devices associated with the oxygen supply, Li et al. [25] developed a micro PFC with an air-breathing cathode to replace the oxygen dissolved cathode.

Membrane electrode assembly (MEA), in which the anode, membrane and cathode are sandwiched together by hot-press, has been widely employed in conventional fuel cells [26,27]. The utilization of the MEA can make the fuel cell system more compact and flexible. Using the MEA design, Seger and Kamat used the TiO_2 photoanode to replace Pt and developed a MEA based photoelectrochemical cell to generate the photocurrent and hydrogen under UV irradiation [28]. However, this photoanode could only respond to the UV light and had low specific surface area, limiting the improvement in the photoelectrochemical reactions. Hence, we proposed a membrane electrode assembled photoelectrochemical cell in this work to simultaneously degrade the organics and generate electricity upon the visible light irradiation. The photoanode was formed via incorporating mesoporous material (silica, SBA-15) into the TiO_2 followed by the quantum-dot-sensitization of CdS-ZnS, termed as CdS-ZnS-TiO_2/SBA-15. This photoanode could not only respond to the visible light but also have large specific surface area and pore volume, thereby improving the photocatalytic activity and mass transport efficiency. The air-breathing cathode was employed to enhance the oxygen transport and simplify the PFC system. The performance of the developed PEC with the MEA design was accessed by using ethanol as a model organic pollutant under various conditions.

2. Experimental

2.1. Construction of photoanode

The composite photoanode of CdS-ZnS-TiO_2/SBA-15 was prepared by the following three steps: the preparation of the TiO_2/SBA-15 colloid, the construction of the TiO_2/SBA-15 film on the carbon paper (Toray 090, Japan) and the quantum-dot sensitization by CdS-ZnS. In the first step, the TiO_2/SBA-15 colloid was prepared by the sol-gel method [29]. Here, a given amount of TiO_2 nanoparticles (Aeroxide P25, Acros, Belgium) and a given amount of SBA-15 particles (Nathmay, China) were put into an agate mortar and grinded for 5 min to make them uniformly mixed. The mixed powders were then added into the mixture of 120 mL deionized water and 0.4 mL acetylacetone (Sigma-Aldrich, USA) with magnetic stirring. After that, 0.2 mL Triton X-100 (Sigma-Aldrich, USA) was added to facilitate the spreading of the colloid. Finally, 2.4 g polyethylene glycol (Aladdin, China) was added into the solution and magnetically stirred for 12 h. In the second step, the carbon paper was cut into the size of 2.0 cm × 2.2 cm and ultrasonically cleaned in deionized water for 10 min. After drying, an adhesive tape was used to cover one edge of the carbon paper, forming an exposed area of 2.0 cm × 2.0 cm for coating TiO_2/SBA-15. Then the TiO_2/SBA-15 colloid was sprayed on the exposed carbon paper by a spray gun until the total photocatalyst loading of TiO_2/SBA-15 was about 3 mg/cm^2. The loading was determined by the mass difference between the carbon papers without/with the coated photocatalysts over the active area of the electrode. During the spray process, the mass of the carbon paper with the coated photocatalysts was frequently measured until the target loading was achieved. After that, the adhesive tape was removed and the photocatalysts coated carbon paper underwent the calcination at 550 °C for 2 h. In this work, we prepared four photoanodes with various SBA-15 mass ratios defined as the ratio of the SBA-15 mass to the TiO_2 mass, The SBA-15 ratio ranged from 15%, 30%, 45% to 60%, which were represented by CdS-ZnS-TiO_2/15% SBA-15, CdS-ZnS-TiO_2/30% SBA-15, CdS-ZnS-TiO_2/45% SBA-15 and CdS-ZnS-TiO_2/60% SBA-15, respectively. The third step aimed to form the CdS-ZnS composite on TiO_2/SBA-15 film by successive ionic layer adsorption and reaction (SILAR) [30]. Two aqueous solutions were prepared, one containing $Cd(NO_3)_2{\cdot}4H_2O$ (Aladdin, China) and $Zn(NO_3)_2{\cdot}6H_2O$ (Aladdin, China) with a molar ratio of 3 and the other one containing $Na_2S{\cdot}9H_2O$ (Aladdin, China). The concentration of metal ions as well as the concentration of sulfur ions were 0.1 M. The TiO_2/SBA-15 coated carbon paper was put into the $Cd(NO_3)_2$ and $Zn(NO_3)_2$ mixed solution for 4 min and then washed in deionized water. After that, it was put into the $Na_2S{\cdot}9H_2O$ solution for 4 min and washed in deionized water again. This sequence corresponded to one SILAR cycle. 4 SILAR cycles were repeated and then dried with N_2 stream. Finally, the CdS-ZnS-TiO_2/SBA-15 coated carbon paper was put into an oven at 100 °C for 10 min. It was found that the prepared photoanode showed yellow color because of the quantum-dot-sensitization by CdS-ZnS, indicating that the visible-light responsive photoanode has been successfully prepared.

2.2. Construction of cathode

The cathode was made of the carbon black coated carbon paper (HCP120, Hesen, China) with deposited Pt black (JM, Hesen, China). The cathode was prepared by the following three steps. First, 0.1 g Pt black electrocatalyst was mixed with 2.5 g distilled water. Then 2.5 g isopropanol (Aladdin, China) and 0.5 g Nafion® perfluorinated resin (D520, 5% wt. DuPont Co. USA) were added. The prepared mixture was then applied on the carbon black coated carbon paper (2.0 cm × 2.0 cm) by a spray gun. Subsequently, the electrode was heated at 80 °C for 30 min. The Pt loading was about 1 mg/cm^2.

2.3. Assembly of membrane and electrodes

Before the MEA fabrication, 5% Nafion® perfluorinated resin solution was firstly sprayed onto the surface of the prepared cathode. To do this, 0.4 mg Nafion® solution was sprayed onto the side of the cathode with catalysts. After that, the ionomer coated cathode with the catalyst coated side toward the membrane was hot-pressed with the Nafion® membrane (211, Hesen, China) at 0.5 MPa and 135 °C for 3 min. Subsequently, the photoanode was directly attached to the other side of the Nafion® membrane without hot-pressed, forming a membrane electrode assembly with the active area of 2.0 cm × 2.0 cm = 4 cm^2.

2.4. PEC fixture

As shown in Fig. 1a, the fabricated MEA was sandwiched between two electrical current collectors. A serpentine flow field was drilled in the both current collectors, which served as the passages of light,

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