

Simultaneous pollutant degradation and power generation in visible-light responsive photocatalytic fuel cell with an Ag-TiO₂ loaded photoanode

Beiqi Deng^a, Shaozhu Fu^{a,b}, Yanpeng Zhang^a, Yun Wang^a, Dongmei Ma^a, Shuangshi Dong^{a,b,*}

^a School of Jilin University, Changchun 130021, China

^b Key Lab of Groundwater Resources and Environment, Ministry of Education, Jilin University, Changchun 130021, China

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ABSTRACT

Photocatalytic fuel cell (PFC) has been studied for treating phenolic organic pollutants and converting chemical energy into electricity effectively. The photoanode in the anode chamber degraded pollutants by photocatalytic oxidation, while the released electrons were transported to the cathode chamber via an external circuit to participate in the reduction reaction. These reactions enable the conversion of chemical energy into electrical energy. In this study, we firstly used sol-gel method to prepare Ag-TiO₂ as the visible-light responsive catalyst. Then we compared two coating methods to obtain effective and stable photoanode with carbon foam as the support, i.e. sol-gel impregnation method and powder spray coating method. The 4-chlorophenol (4-CP), phenol and tetracycline (TC) were selected as the target pollutants. A dual chamber PFC was constructed to examine the pollutants degradation and power generation. The results showed that the photoanode prepared by powder spray coating method presented higher photoactivity and stability. The uniform and stable loading could be obtained when the loading ratio was 0.62 g/g. The degradation efficiency was maintained around 58% and the detachment of photocatalyst was 27.0% after three repeated cycles. In the PFC, the degradation efficiency of 4-CP, phenol and TC reached 32.6%, 37.9% and 96.4% in 6 h and the filling factors (*FF*) were 0.17, 0.31 and 0.44, respectively.

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1. Introduction

The phenolic compounds and antibiotics have aroused the environmental problem due to their representative poisonousness, harmfulness and refractory degradability [1–3]. The typical treatment methods for the phenolic pollutants include physical (e.g., adsorption, ultrasonic method), chemical (e.g., advanced oxidation process, chemical reduction) and biological process [4]. Physical process is limited in application due to the problem of high cost and secondary pollution [5]. The operation of biological process can be greatly subjected to the environmental factors because of the high biodegradation resistance of the pollutants [6].

Recently, the photocatalytic degradation of these organic compounds has drawn great attention, as it possesses the merit of high efficiency, low cost and no toxicity [7]. With the increasing demand of simultaneous water treatment and clean energy reclamation, the concept of a PFC was firstly proposed by Kaneko et al. using titanium dioxide as the photocatalyst in PFCs [8]. However, the

TiO₂ with wide band gap can only absorb ultraviolet energy less than 387 nm wavelength, which merely accounts for about 4% of the solar energy [9]. Accordingly, noble metals (Pt, Au, Ag and Pd) were introduced to TiO₂ to form a Schottky barrier with a large gap between the Fermi levels of TiO₂, which can effectively inhibit the recombination of electron hole pairs and improve photocatalytic efficiency [10–13]. Studies had shown that the Ag-TiO₂ particles had higher photoactivity than the pure TiO₂ and that the photocatalytic activity increased with the rise in the AgNO₃ content [14]. The photocatalytic efficiencies of the Ag-TiO₂ nanocomposite thin film samples prepared using Ag concentrations of 1%, 5% and 10% were 53%, 63% and 36%, respectively [15]. In the selection of anode substrate material, metal oxide and metal plate were usually used as the anode for their good conductive effect [16], but the system had a weak response to visible light due to the lack of photocatalytic modification [17]. Besides, some researchers used conductive glass as the substrate, but the photocatalyst loading was unstable due to the smoother surface and the smaller specific surface area [18].

In this paper, the 4-CP, phenol and TC were taken as target pollutants, and the three-dimensional carbon foam material as the

* Corresponding author.

E-mail address: dongshuangshi@gmail.com (S. Dong).

anode substrate. We used silver doped titania as the photocatalyst and compared two methods for catalyst loading to prepare the photoanode. We constructed a two-chamber PFC to investigate the effect of simultaneous 4-CP degradation and residual chemical energy to electric energy conversion. We aim to find a new way for energy recovery in the process of organic pollutant treatment.

2. Experimental

2.1. Preparation of Ag-TiO₂ powder and Ag-TiO₂ loaded carbon foam electrode

The Ag-TiO₂ photocatalyst was prepared via a modified sol-gel method based on the research of Lee et al. [14] 0.24 mmol of silver nitrate (AR) was added to a sealed bottle containing 60 mL of 4 mmol/L sodium citrate (AR). The reaction temperature was kept at 80 °C with the continuous stirring until the color of the solution changed obviously from colorless to light brown. An amount of 0.91 mL of nitric acid (AR) was slowly added to 40 mL of tetraisopropyl titanate (98%) with stirring for 5 min and then was injected into the above solution by a syringe. The mixture was stored at 50 °C for 24 h under vigorous stirring. Subsequently, the mixture was poured into a 200 mL Teflon-lined stainless steel autoclave, and maintained at 130 °C for 5 h. The resulted white composite was collected by removing the supernatant. Finally, the composites were dispersed in the absolute ethanol (99%) at different Ag-TiO₂ concentrations for direct loading (sol-gel impregnation method). Carbon foam (ERG Group, USA) with 80 pores per linear inch (ppi), and external dimensions of 2.5 × 4 × 3 mm (length × width × height), was successively soaked with 1 M nitric acid, 1 M acetone (AR) and absolute ethanol before drying at 60 °C and catalyst loading. As for the other loading method (powder spray coating method), the composites were dried at 80 °C before grinding to Ag-TiO₂ powder, which was then dispersed into absolute ethanol to obtain loading solution with concentration at 1 g/L. The solution was sprayed on the carbon foam with sprinkling. The catalyst loaded carbon foam was dried at 60 °C for 15 min after each spraying. The loading ratios were controlled at 0.31, 0.62, and 0.93 g/g (catalyst/carbon foam). The relationship between the load ratio and the photoactivity was investigated by using the carbon foams with different loading ratios to degrade the 4-CP.

2.2. Characterization

The crystal structures of TiO₂ and Ag-doped TiO₂ photocatalysts were identified by X-ray diffraction (XRD) in the 2θ range from 10°–90° with a scanning rate of 2° min⁻¹. The XRD patterns were obtained with a powder X-ray diffractometer (RINT 2500, Rigaku) using Ni-filtered Cu Kα radiation. Peak positions were compared with the standard files to identify the crystalline phases. The microstructure of the catalyst loaded carbon foam (The loading ratios was 0.62 g/g) was observed by scanning electron microscope (SEM). The sample observed with an SEM instrument (Akashi-SX-40, USA), equipped with a EDS probe for the energy dispersive spectroscopy analysis (EDS) to confirm the presence of elements by detecting X-rays.

2.3. Photocatalytic activity of photocatalyst

The photocatalytic activities of as-prepared photocatalyst powder and the photocatalyst loaded electrode were investigated by degradation of 4-CP with the photocatalyst powder dosage at 1 g/L and the concentrations of 4-CP were 10, 20, 30, 40 mg/L, respectively. A 150 W xenon lamp with 420 nm cut filter was used as the light source. The samples at given intervals were taken from the suspension and analyzed through high performance

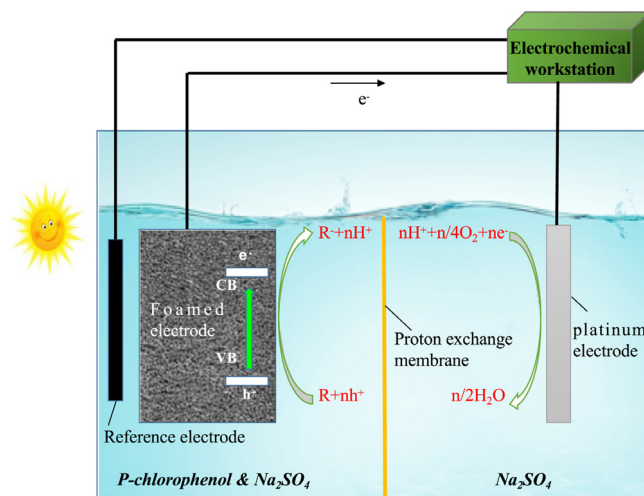


Fig. 1. The configuration of the dual chamber structured PFC with Ag-TiO₂ loaded carbon foam as the anode and the platinum electrode as the cathode. An amount of 20 mg/L of 4-CP, phenol and TC were used as anode substrates, respectively. Dissolved oxygen was used as an electron acceptor in the cathode chamber. Light energy is the motive power of the PFC system.

liquid chromatography (HPLC), the tested method was provided in Supplementary information [19]. The photocatalytic degradation efficiency could be calculated according to Eq. (1).

$$\eta = (1 - C/C_0) \quad (1)$$

where C_0 and C is the concentration of pollutant solution at initial and at the sampling time, respectively.

The pseudo first order kinetic equation (2) for linear regression analysis was employed to fit the degradation data [20,21].

$$\ln(C_0/C) = kt \quad (2)$$

where k represents the first-order reaction rate constant.

2.4. PFC experiments

Fig. 1 illustrates the configuration of the PFC. We adopted a dual chamber structured PFC with the volume of each electrode chamber at 60 mL. The two chambers were separated by a proton exchange membrane. The catalyst loaded carbon foam was used as the anode electrode, while the platinum electrode as the cathode electrode. Both the two electrode chambers contained 50 mmol/L of anhydrous sodium sulfate (AR) solution as electrolyte and the anode substrate was 20 mg/L of 4-CP, phenol or TC. Electrochemical data were recorded by a CHI-660e electrochemical workstation (Shanghai Chenhua, China). The dark condition was kept for the PFC before start-up.

3. Results and discussion

3.1. Characterization of the prepared catalyst and anode

Fig. 2a and b showed SEM images of Ag-TiO₂ loaded carbon foam anode prepared by sol-gel impregnation method and powder spray coating method, respectively. It could be seen that both methods achieve the uniform distribution of the photocatalyst powders on the surface of carbon foam structure. Fig. 2c presented the EDS spectra from the given point at Fig. 2a. The results indicated that the scanning area contained Ti, O, Ag and C. The inset table illustrated that the content of these elements for the prepared sample was determined by analyzing their weight percentage and

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