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An efficient photoelectrochemical cell functioning in the presence of organic wastes

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ABSTRACT

This work presents a design and studies the function of a photo-fuel cell, which can photocatalytically consume organic substances to produce electricity. The design resembles hydrogen fuel cells but, in the present case, hydrogen ions are produced photocatalytically in the presence of nanocrystalline titania. Results are presented for ethanol and glycerol but many other substances can be used as well. The cell can function in natural solar light by exploiting the UV portion of the solar spectrum. The produced current was enhanced when titania was combined with cadmium sulphide, which absorbs visible light. © 2009 Elsevier B.V. All rights reserved.

1. Introduction

Semiconductor-mediated photocatalytic [1-5] and photoelectrochemical [6-9] processes have been widely investigated in recent years as potentially efficient, economic and environmentally friendly methods for a wide range of industrial applications, including mineralization of organic pollutants [1–3], disinfection of water [4] and air [5] as well as production of fuels [7,8,10,11] and electricity [9]. In spite of the remarkable progress that has been made in fundamental research, in the development of solar photocatalysts [12-14] and in photoreactor design [15,16], the practical achievements in the field of solar energy conversion and storage have been modest. This is mainly due to the low quantum yield of photocatalytic reactions and the relatively low overall efficiency of photocatalyst/photoreactor configurations. In our recent studies, we have shown that the efficiency of photocatalytic hydrogen [10,17–20] or electricity [21,22] production from irradiated aqueous Pt/TiO₂ suspensions [10,17–19] or immobilized Pt/TiO₂ films [20-22] may be remarkably enhanced in the presence of organic compounds in solution. Here we present a novel design for an efficient photoelectrochemical cell (PEC), which is able to produce electricity from aqueous solutions of waste organic compounds when irradiated with simulated or natural solar light. The cell comprises a TiO₂-coated photoanode

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and a cathode, which consists of a combination of a proton transfer membrane and a Pt/carbon cloth (Pt/C) electrode. Our findings suggest that the present PEC design may provide the basis for the development of efficient devices for the production of electricity from renewable resources with simultaneous degradation of waste organic compounds in solution.

2. Experimental

2.1. Materials

Unless otherwise indicated, reagents were obtained from Aldrich and were used as received. The nanocrystalline titania was commercial Degussa P25. Millipore water was used in all experiments. SnO₂:F transparent conductive electrodes (FTO, resistance 8 Ω/\Box) were purchased from Hartford, USA. Nafion membrane N117 was purchased from Ion Power, Inc. and carbon cloth, 20% wet proofing, from BASF Fuel Cell, Inc., USA. Carbon black, Vulcan XC72R, was a gift from Cabot Corporation. Pt/carbon black electrocatalyst (30% on Vulcan XC72) was also purchased from BASF Fuel Cell, Inc., USA.

2.2. Preparation of the anode

Nanocrystalline titania (n-TiO₂) films were deposited on FTO transparent conductive electrodes by the following procedure. Degussa P25 was dispersed in a mixture of water, ethanol and

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polyethyleneglycol (PEG) 2000 as in previous publications [21-23]. Briefly, 0.3 g of Degussa P25 was dispersed by vigorous stirring in 3 ml of a solution made of about 10 ml water, 10 ml of ethanol and 1 g of PEG 2000. The resulting material was a white paste, which can be easily cast on any solid substrate. The viscosity of the paste can be controlled by evaporating part of ethanol in a rotary evaporator. Films were thus made by doctorblading the paste by hand on the FTO electrodes. After casting, the electrodes were calcined at 550 °C. The procedure was repeated once more. The final quantity of titania on the electrode was about 20 mg, while the dimension of the film was $2.5 \times 3.0 = 7.5$ cm². Due to the high calcination temperature employed, the film adhered well on the SnO₂:F electrode and withstood the hard alkaline conditions during cell operation. External electric connection was made by using an auto-adhesive copper ribbon. Cadmium sulfide was deposited on n-TiO₂ films by adsorption of the reagents from aqueous solutions. For this, we first prepared an aqueous solution of 0.1 mol/L $Cd(NO_3)_2$ and an aqueous solution of 0.1 mol/L Na₂S. The as-prepared n-TiO₂/FTO electrode was dipped for 5 min in $Cd(NO_3)_2$ solution, which resulted in adsorption of Cd²⁺ ions on the titania nanocrystalline film. Then the electrode was taken out of the solution, thoroughly washed with water and subsequently dipped in the Na₂S solution for 5 min. During the contact with the sulfur-ion-containing solution, CdS was formed on the titania film. The procedure was repeated ten times, by taking care to wash with water all the excess ions possibly adsorbed. The deposition of CdS was monitored by absorption spectrophotometry. Ten dipping procedures were judged sufficient to saturate the titania film with CdS.

2.3. Preparation of the cathode

An amount of 0.246 g of carbon black was mixed with 8 mL of distilled water by vigorous mixing in a mixer (about 2400 rpm) until it became a viscous paste. This paste was further mixed with 0.088 mL polytetrafluorethylene (Aldrich, Teflon 60% wt. dispersion in water) and then applied on a carbon cloth cut in the necessary dimensions. This has been achieved by first spreading the paste with a spatula, preheating at 80 °C and finally heating in an oven at 340 °C. Subsequently, the catalytic layer was prepared as follows: 1 g of Pt/carbon black electrocatalyst (30% on Vulcan XC72) was mixed with 8 g of nation perfluorinated resin (5 wt% solution in lower aliphatic alcohols and water, Aldrich) and 15 g of a solution made of 7.5 g H_2O and 7.5 g isopropanol. The mixture was ultrasonically homogenized and then applied on the previously prepared carbon cloth bearing carbon black. Then the electrode was heated at 80 °C for 30 min and the procedure was repeated as many times as necessary to load about 0.5 mg of

Pt/cm². The thus prepared Pt/carbon cloth (Pt/C) electrode is ready for use.

2.4. Preparation of the cathode–nafion membrane assembly

The next step was to paste the nafion membrane on the Pt/C electrode. A piece of nafion membrane was cut in the dimensions necessary to apply on the Pt/C electrode, leaving uncovered only a small part for electric connection. The two pieces were placed one on the top of the other, were sandwiched between two teflon sheets and placed between two metallic plates, which could be electrically heated with temperature control and pressed with a hydraulic press. Then the plates were heated at 120 °C and as soon as this temperature was reached, a small pressure was applied. Heat and pressure helps nafion membrane to permanently stick on the Pt/C electrode. Teflon sheets are used only to protect the assembly from direct contact with the hot metal plates.

2.5. Construction of the cell

The main body of the cell was made of prexiglass appropriately machined to receive the anode electrode on one side and the nafion–Pt/C assembly on the other. The size of the active window was 2.5×3 cm. The distance between the anode and the nafion–Pt/C assembly was 5 mm. A small hole on the top allowed introduction of the electrolyte and could be easily sealed with an adhesive tape. Electric connections could be directly made with crocodile clips. Fig. 1 shows a picture of the cell in operation under natural sunlight.

2.6. Characterization of the cell

Current–voltage characteristics of the electrodes were obtained by using a Keithley 2601 source meter. Illumination of the sample was made by using various light sources applying in each case: a 450 W Xenon lamp, a home-made black–light source employing 4 W tubes, a solar simulator (Atlas Suntest CPS⁺) and natural solar light. The intensity of the incident radiation in each case was measured with an Oriel (Newport) 70260 Radiant Power Meter.

3. Results and discussion

A very important aspect of processes involving semiconductormediated photocatalytic degradation of organic wastes in aqueous media concerns energy yield and energy conversion routes.



Fig. 1. PEC cell (left) powering a small motor with rotating disk (right). The system runs under natural (solar) light. The dimensions of the plexiglass body of the cell is $5.5 \times 6 \times 5$ cm. The distance between anode and cathode was about 5 mm.

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