Electrochimica Acta 109 (2013) 162–167

Contents lists available at ScienceDirect

Electrochimica Acta

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

Magnetic field enhanced photoelectrochemical response of a nanostructured titanium dioxide anode

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ARTICLE INFO

Article history:
Received 30 March 2013
Received in revised form 3 May 2013
Accepted 9 July 2013
Available online 22 July 2013

Keywords:
Photoelectrochemical fuel cell
Response to magnet field
Photon excitation
TiO_2 nanotube

ABSTRACT

The concept of permanent-magnet photoelectrochemical fuel cell is proposed. The fuel cell contains a TiO_2 nanotube anode and a platinum cathode. Water was used as the fuel. The anode has a self-organized high aspect ratio TiO_2 nanotube layer made through electrochemical oxidation of Ti. Photocatalysis and permanent magnetic field excitation experiments were performed. The photoelectrochemical responses of the anode to visible (Vis) and ultraviolet (UV) light were studied to examine the magnetic field effect. The open circuit voltages were 0.0564, 0.1625, 0.1898 and 0.376 V, under the excitation of Vis, UV, magnet and magnet + UV, respectively. Current density was measured under different linear potential scanning conditions. It reached a value of 5.23 mA/m^2 just under the excitation of the permanent magnet at a bias voltage of 2 V. Combining the magnetic action and the photovoltaic effect, the titanium dioxide photosensitive anode showed a maximum photocurrent density of 296.05 mA/m^2 at 2 V bias. It is concluded that the absolute value of open circuit voltage (OCV) of the fuel cell due to the exposure to the magnet field is much higher than that due to either UV or Vis light illumination. However, UV light excitation results in higher current density in the measurement.

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1. Introduction

Fossil fuels are consumed very rapidly, creating an urgent need for research on clean and renewable energy conversion methods. Photoelectrochemical fuel cells (PEFCs) have been widely studied as promising clean energy conversion devices. They are considered to be used for water splitting [1], decomposing pollutants [2,3] and generating electrical energy [4]. The renewable energy source of PEFCs is solar light. With the development of nanotechnology, nanostructured anodes [5] are adopted in photo fuel cell construction. TiO_2 is a promising material proposed for such an application as early as in 1972 [6]. The first self-organized anodic oxide using Ti was reported in 1999 [7]. The effect of processing conditions on the geometry of TiO_2 nanotubes was reported in 2005 [8]. The wide applications of TiO_2 nanotube include, but are not limited to photocatalysis [9–11], photovoltaics [12–14], electrochromics [15], and cell-implant interaction enhancement [16].

TiO_2 nanotube is an n-type semiconducting material which is active under photonic, magnetic and/or electric excitations [17–21]. Magnetic field is an important energy source which has been utilized over a long period of time [22,23]. Magnetic field effect on oxygen redox reaction in a polymer electrolyte fuel cell was studied earlier [24]. When a permanent magnet was set behind a cathode, the performance of the polymer electrolyte fuel cell was enhanced. This is because of the magnetic attractive force toward O_2 gas. Wang et al. [25] studied the magnetic particle assisted methanol oxidation and oxygen reduction processes. They developed a method of switching fuel cells that can enable on-demand power generation in order to meet the specific needs of power consuming units. The Hall Effect [26–28] describes the magnetic field induced electric response in a solid state semiconducting material. The nanostructured titanium dioxide anode/electrolyte interface in the fuel cell is a liquid junction which is similar to a junction in those solid semiconducting materials. The magnetic field induced electrification phenomenon should be similar to the Hall Effect as found. To test this hypothesis, we performed experimental studies as described in this paper.

The rest of this paper is organized as follows. First, we shall present how a new fuel cell called magnet-photo fuel cell (MPFC) is developed through the addition of a permanent magnet field to the TiO_2 anode of the PEFC. Then, the magnet field induced changes

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http://dx.doi.org/10.1016/j.electacta.2013.07.081

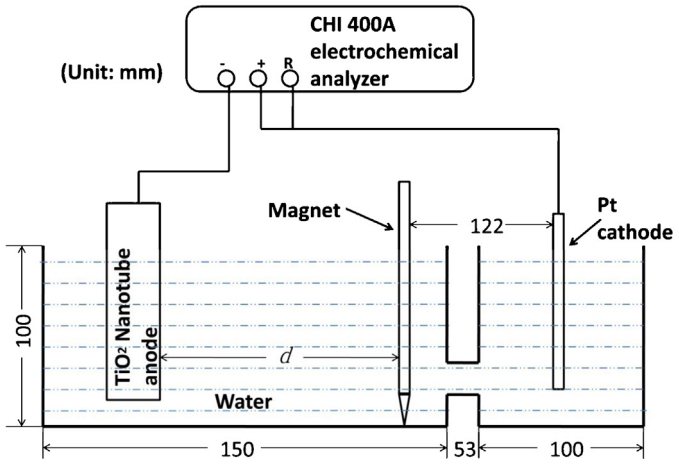

Fig. 1. The configuration of the magnet-photo fuel cell.

in the voltage and current density of the fuel cell will be demonstrated. Finally, the conclusion is drawn that the additional electric field generated by the magnetic field can enhance the open circuit voltage of the fuel cell.

2. Materials and experimental

2.1. Chemicals

All chemicals were used as received from Alfa Aesar without further purification. Ti foil (99.9% purity, 0.127 mm), H_3PO_4 (85% wt in water), NaF (98% purity) were used as the start materials to prepared the fuel cell anode and to make the electrolyte for titanium dioxide nanotube growth. The materials for making the fuel cell compartment are stainless steel (304Y) and glass (the configuration of the fuel cell is shown in Fig. 1).

2.2. TiO_2 nanotube preparation

The electrochemical oxidation method was used to synthesize the self-organized TiO_2 nanotubes. The size of the titanium foil was 3.2 mm × 40 mm. Before being used for the electrochemical oxidation or anodization, the titanium foil was cleaned step-by-step using acetone, ethanol and distilled water. Then it was dried in air stream. The TiO_2 nanotubes on the Ti were fabricated in a solution containing 1.0 M H_3PO_4 and 0.2 M NaF. The Ti foil was used as the anode and a platinum wire was used as the cathode. The voltage and time for anodizing Ti were 20 V and 18 h. The nanotubes were cleaned using distilled water and the samples were dried in air. The anodization process was conducted at room temperature (25 °C). The TiO_2 nanotube was annealed in air at 450 °C for 4 h in order to convert the amorphous structure of TiO_2 into an anatase crystalline structure.

2.3. Magnet-photo electrochemical catalytic response measurement

A magnet-photo electrochemical fuel cell was made using the TiO_2 nanotube sheet as the anode, Pt wire as the cathode, and water as the fuel. The ultraviolet (UV) lamp used was the model UVL-21 (365 nm UV, 4 W, 0.16 A) lamp with the illumination intensity of 40 mW/cm^2. The visible light (Vis) source was a 250 W incandescent light bulb. To generate the required magnet excitation, we used a small, regular cylindrical magnet with a dimension of 11.80 mm (d) × 11.80 mm (h). The operation method of the magnet excitation was as follows. We simply inserted the magnet into the solution. The field is applied to the fuel cell. Taking the magnet out from the fuel cell, we removed the field. A CHI 400 A electrochemical workstation was used to measure the open circuit voltages and current densities of the fuel cell and to supply the bias potentials. The term "bias potential" stands for the applied voltage crossing the fuel cell with respect to the reference Pt.

We used the two-electrode system for electrochemical testing, as shown in Fig. 1. As compared with the three-electrode system using the Ag/AgCl reference electrode, the two electrode method was more stable since there was no chemical reaction during the test. Typically, the Ag/AgCl or Hg/$HgCl_2$ reference electrode is used in electrochemical studies. The shift in standard potentials of these reference electrodes may occur under the influence of operating temperature, composition of solutions, and the polarization levels. As compared with the three electrode system, the counter electrode and the reference electrode are the same of Pt in the two-electrode system. Being a noble metal, Pt has better stability than the Ag/AgCl or Hg/$HgCl_2$ reference electrode. The reference electrode lead and the counter electrode lead were connected together in the two electrode system. Therefore, the potential drift could be reduced significantly. The main part of the container was made of stainless steel with glass windows, which allows light to pass through. The details of the whole system are shown in Fig. 1. The distance between the anode and magnet was demonstrated by *d*. This distance can be adjusted in order to examine the effect of *d* on the performance of the magnet-photo fuel cell.

3. Results and discussion

3.1. Surface morphology of the TiO_2 nanotube photoanode

Fig. 3 shows a high magnification transmission electron microscopic (TEM) image of the nanostructured anode material, from which we can see that the TiO_2 nanotubes have a diameter of about 120 nm. The wall thickness is about 40 nm. The bundle of the TiO_2 nanotubes (NTs) as shown by this image reveals that the length of the nanotubes is over 1 μm. The increase in the concentration of NaF could result in deeper penetration of more F^- into the Ti substrate. Thus, the length of the NTs can be increased significantly.

If the electrochemical oxidation parameters such as the applied DC voltage and processing time were changed, the size of the TiO_2 NTs can also be changed using different electrolytes consisting of either HF or NH_4F. Energy dispersive X-ray spectrum was obtained and shown in Fig. 4. The result shows that Ti and O are the major elements at the anode. The peak of O is found at 0.525 kV and Ti peaks are at 0.401, 0.452, 4.551 and 4.932 kV. Quantitative analysis gives the atomic ratio of Ti:O = 1:2, which means that the nanotubes have the stoichiometric composition of titanium dioxide. The weight percentage of each element is also calculated and reveals that the weight ratio of Ti:O equals to 3:2. The signal from fluorine ion is found because the ion penetrates into the wall of the nanotubes. The remaining ion is the reason for the peak located at 0.677 kV. It should be noted that the Cu peaks are from the TEM specimen holder. In the TEM experiment, a copper mesh was used to hold the nanotube sample.

The open circuit voltage and current density under linear voltage scanning were tested under permanent magnetic field and light excitation. The effect of the distance between the TiO_2 NTs anode and magnet were also studied. The open circuit voltage test in the case of excited by the magnet showed very promising results. The value is even higher than that with UV light excitation. The open circuit voltage reflects the driving force for the entire fuel cell system. It is noted that the open circuit voltage is related to the dynamics response of the electron–hole charge separation under external magnetic field or photon excitation. The current density is an indication of the reaction kinetics of the electrode process.

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