



A microfluidic all-vanadium photoelectrochemical cell for solar energy storage

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ABSTRACT

In this work, a microfluidic all-vanadium photoelectrochemical cell (μ VPEC) was designed for the solar energy storage. The miniaturization design could enhance the photon and mass transport, reduce the internal cell resistance, and improve the uniformity of the light distribution. Because of these advantages, the developed μ VPEC was able to yield good performance. Experimental results indicated that the developed μ VPEC showed good photoresponse and operation stability. Besides, its performance was also evaluated under various operating conditions, including the membrane thickness, the light intensity and the vanadium ion concentrations. It was shown that although the vanadium ion permeation was small with thicker membrane, the increased proton transfer resistance decreased the photocurrent density. The increase of the light intensity could produce more photo-generated electron-hole pairs, which could improve the photoelectrochemical reaction rate and the conversion rate. It was also found that the photocurrent density was increased with the vanadium ion concentration as a result of the enhanced mass transfer. The results obtained in this work reveal that the developed all-vanadium photoelectrochemical cell shows the promising potential for the solar energy storage.

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1. Introduction

Owing to the grand challenge of energy crisis, the replacement of fossil fuels by renewable energy becomes inevitable to ensure the sustainable development. To date, a considerable number of researchers have devoted their efforts to the development of renewable energy, including the solar energy, wind energy and tidal energy, etc [1–4]. Solar energy, as one of main renewable energy resources, has attracted wide attention of the researchers because it is clean, extensive and inexhaustible [5]. Presently, main commercial solar energy utilizations rely on the photothermal and photoelectric conversions by converting the solar energy into heat and electricity, respectively [6,7]. However, both solar utilization technologies face the problem of the intermittence. Hence, the energy storage is usually required in these applications.

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Typically, the common methods for the energy storage during the solar energy utilization processes include the solar-thermal energy storage, electrochemical energy storage and photochemical energy storage [8–12]. Among them, vanadium redox flow battery (VRB), proposed by Maria Skyllas-Kazacos and co-workers in 1985, has been regarded as one of the most competitive candidates for large-scale energy storage [13–15]. The VRB can offer lots of advantages, such as long life cycle, large storage capacity, high efficiency and safety [16,17]. Moreover, the electrolyte metal ions are all vanadium, which can effectively avoid the cross contamination in the electrolyte solution and be easily recycled [18,19]. In recent, Liu and his co-authors combined the photoelectrochemical cell with the vanadium redox flow battery to construct a photoelectrochemical vanadium redox battery for solar energy storage [20–23]. This method can directly convert the solar energy into the chemical energy via the photoelectrochemical reactions. However, there are still some drawbacks in their designs. For example, they used the batch reactor design instead of continuous flow mode. Large-scale reactors not only result in large internal resistance between the electrodes but also increase the photon and mass

transport resistance, which inhibits the reaction rate and energy conversion efficiency. Besides, the photoanode in their system was prepared by directly depositing the TiO₂ on the FTO glass using a doctor blade. The resultant poor connection between the TiO₂ film and FTO glass caused relatively low electron transfer, which may be unfavorable for preventing the recombination of the photo-excited electron-hole pairs.

In recent, the incorporation of microfluidics into the reactor design has been widely used in the photoreactors [24–27], because microfluidics has large surface-to-volume ratio to enhance the mass and photon transfer. Moreover, because of the microreactor design, the light distribution can be more uniform. Besides, previous works have also demonstrated that the addition of a compact TiO₂ layer between the porous TiO₂ layer and the conductive substrate could not only ensure the strong adhesion of the TiO₂ film to the conductive substrate but also provide high conductivity to prevent the recombination of the photo-excited electron-hole pairs, while the top porous TiO₂ layer could still provide sufficient path for the mass and photon transport and large specific surface area for the photoelectrochemical reactions [11,28–30]. All these merits are beneficial for the improvement in the photoelectrochemical reaction rate and conversion efficiency. In line with this idea, a microfluidic all-vanadium photoelectrochemical cell (μ VPEC) was proposed for the solar energy storage in the present work. The morphology and chemical composition of the photoanode were characterized using the FESEM and XRD. The performance of the developed μ VPEC was also evaluated under various design and operating parameters, including the thickness of the proton exchange membrane, the light intensity as well as the vanadium ion concentration.

2. Experimental

2.1. Preparation of the electrolyte

Prior to the experiment, the photoanode and cathode electrolytes were firstly prepared. The photoanode electrolyte was prepared by dissolving a certain amount of vanadium (IV) sulfate oxide hydrate (VOSO₄·xH₂O) (99.9%, Sigma-Aldrich, USA) in 2.0 M H₂SO₄. In this work, three photoanode electrolytes with the VO²⁺ concentration ranging from 10.0 mM, 50.0 mM–100.0 mM were prepared. For the cathode electrolyte, a certain amount of V₂O₃ powder (95%, Alfa Aesar, USA) was firstly dissolved in 200 mL 5.0 M H₂SO₄ aqueous solution via the evaporation reflux at a temperature of 120 °C in a magnetic stirring oil bath. After all the powders was dissolved, the prepared solution was cooled down to room temperature. Distilled water was then added into the prepared solution to maintain the H₂SO₄ concentration at 2.0 M, which was the same as the photoanode electrolyte. Similarly, three corresponding cathode electrolytes with the V³⁺ concentrations of 10.0 mM, 50.0 mM and 100.0 mM were prepared.

2.2. Fabrication of the photoanode

In this work, the photoanode was formed by depositing the TiO₂ film on the FTO glass (resistance 10 Ω per square, Meijingyuan Technology Co., China). The TiO₂ film was consisted of two layers. One was the dense layer at the bottom, the other was the top porous layer. The detailed preparation procedure is presented as follows. First of all, the FTO conducting glass was cleaned by sonication in acetone, isopropanol and ethanol mixture solution with the volume ratio of 1:1:1. The cleaned FTO conducting glass was then washed with distilled water followed by the drying in an oven for 8 h at 80 °C. After cleaning, the bottom dense layer coated onto the FTO conducting glass was formed by

the spin-coating method using the densely packed TiO₂ layer sol (Kunshan Sunlaite New Energy Technology Co., China). Here, 0.01 mL densely packed TiO₂ layer sol was spun onto the FTO glass by the spin coater with a speed of 1000 rpm for 18 s and 3000 rpm for 30s subsequently. After that, the FTO glass coated with the TiO₂ sol was placed into a tube furnace and calcined at 500 °C for 0.5 h with the temperature rise rate of 10 °C/min to form the bottom dense layer. The top porous layer was prepared by the wet spraying method. In this step, 12 g TiO₂ nanoparticles (Aeroxide P25, Acros, Belgium) were firstly added into a mixture solution consisting of 120 mL DI water and 0.4 mL acetylacetone (Sigma-Aldrich, USA) with magnetic stirring. Then 0.2 mL of a Triton X-100 (Sigma-Aldrich, USA) was added to facilitate the spreading of the colloid. Afterwards, 2.4 g polyethylene glycol (Aladdin, China) was added. The solution was then mixed for 12 h with magnetic stirring. The prepared colloid was sprayed onto the dense layer by a spray gun and finally calcined at 550 °C for 2 h with the temperature rise rate of 10 °C/min to form the top porous TiO₂ layer. The coated area was about 1.0 \times 1.0 cm² and the final loading of TiO₂ was about 3.0 mg/cm².

2.3. Assembly and experimental setup

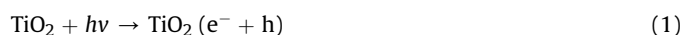
As schematically shown in Fig. 1a, The μ VPEC was consisted of a photoanode, two reaction microchambers separated by a Nafion membrane, a cathode and a cover plate, which were assembled together by bolts. The photoanode and cathode had one inlet and one outlet, respectively. The triangular shape inlets and outlets were designed to ensure that the supplied liquid could be uniformly distributed on the entire electrodes. The depth of the reaction microchambers was 500 μ m, which can greatly reduce the internal resistance of the cell and promote the mass transport. The commercial cathode of the Pt coated carbon paper (DMFC Cathode, Alfa Aesar, Great Britain) with the active area of 1.0 \times 1.0 cm² was chosen as the cathode. The loading of Pt was about 1.0 mg/cm².

The schematic diagram of the experimental system is shown in Fig. 1b. As seen, it was consisted of a syringe pump (LSP04-1A, Longer, China), a 300-W Xe lamp (CEL-HXF300, Beijing Zhongjiaojinyuan technology company, China), a data collecting instrument (34972A, Angilet, America), two liquid containers and a μ VPEC. During the operation, the photoanode and cathode electrolyte solutions were introduced into the cell by the syringe pump. The Xe lamp provided the illumination to the μ VPEC. The light intensity was controlled by adjusting the distance between the lamp and μ VPEC, and measured by the visible radiometer (FZ-Z, Photoelectric Instrument Factory of Beijing Normal University, China). The photocurrent was recorded by the data collecting instrument.

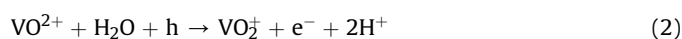
3. Results and discussion

3.1. Working principle of μ VPEC

During the working process, the photoanode and cathode electrolytes were continuously supplied into the reaction microchambers, respectively. Upon illumination, the photo-excited electron-hole pairs were generated in the TiO₂ photoanode as follows,



Because the photo-generated holes have strong oxidation ability, the vanadium ions VO²⁺ could be oxidized to VO₂⁺,



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