Chemical Engineering Journal 273 (2015) 277–285

Contents lists available at ScienceDirect

Chemical Engineering Journal

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

Photocatalytic hydrogen evolution with simultaneous photocatalytic reforming of biomass by Er^{3+}:$YAlO_3$/Pt–TiO_2 membranes under visible light driving

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HIGHLIGHTS

- Er^{3+}:$YAlO_3$ was mixed with TiO_2 to solve the problem of low sensitivity of TiO_2 to visible-light.
- Er^{3+}:$YAlO_3$/TiO_2 catalyst onto glass substrates as membranes could be reused more times.
- Hydrogen evolution and glucose degradation can be carried out simultaneously in the same system.

ARTICLE INFO

Article history:
Received 5 January 2015
Received in revised form 13 March 2015
Accepted 18 March 2015
Available online 24 March 2015

Keywords:
Upconversion luminescence agent
Er^{3+}:$YAlO_3$/Pt–TiO_2 membrane
Visible light photocatalyst
Hydrogen evolution
Reforming of biomass

ABSTRACT

A photocatalyst capable of responding to visible light, Er^{3+}:$YAlO_3$/Pt–TiO_2 membrane, was synthesized via sol–gel dip-coating method by using glass substrates as a supporter. X-ray diffraction (XRD), scanning electron microscope (SEM), transmission electron microscope (TEM) and energy dispersive X-ray spectroscopy (EDX) were performed to reveal the crystallinity, surface morphology and chemical composition of Er^{3+}:$YAlO_3$/Pt–TiO_2 membranes. The photocatalysis capability of Er^{3+}:$YAlO_3$/Pt–TiO_2 membrane was measured through the photocatalytic hydrogen evolution with simultaneous photocatalytic reforming of biomass under simulated visible light irradiation. In addition, mass ratio, heat-treated temperature and initial solution pH as principal influence factors on the photocatalysis capability of Er^{3+}:$YAlO_3$/Pt–TiO_2 membranes were studied. The reused times was also investigated. It was found that the hydrogen generation and glucose degradation could be carried out simultaneously in the same system and the Er^{3+}:$YAlO_3$/Pt–TiO_2 membrane exhibited a much better visible light photocatalytic activity than pure Pt–TiO_2 membrane. Furthermore, the membranes are recyclable. Therefore, these novel composite membrane photocatalysts will have wide applications for photocatalytic hydrogen evolution with simultaneous wastewater treatment on practical application.

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1. Introduction

The fossil fuel shortage and the pollution caused by increasingly energy demands has becomes issues of global concern. However, the pollutants containing high levels of organics are also regarded as an available energy source. In fact, the degradation of organic pollutants alone is also a waste of energy. Therefore, the elimination of pollutions with simultaneously producing renewable energy has been attracting worldwide interest. The solar energy is one of the most promising energy due to its free, easily accessible and inexhaustible characteristics. However, it needs to be captured and converted. Sequentially, most researchers attempt to convert solar energy to a more useful energy form [1]. Hydrogen, an attractive alternative clean energy [2], is expected to become a major energy source in the future. Nowadays, hydrogen is mainly produced by water electrolysis and natural gas (including naphtha, heavy oil, refinery gas and coke oven gas) steam reforming [3]. Considering the policies of energy saving and emission reduction, hydrogen should be produced from renewable energy. Therefore, Photocatalytic hydrogen production from water by solar light has been considered as one of the most promising method to solve energy shortage and environmental pollution problems [4]. Many researches have been done in this field [5–7]. Nevertheless, the efficiency of photocatalytic hydrogen generation is very low because of the lack of an electron donor. In order to solve this problem, many researches have involved sacrificial agents as electron donors, such as methanol, formic acid Na_2S and EDTA [8], etc.

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http://dx.doi.org/10.1016/j.cej.2015.03.090
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Unfortunately, these improvements are based on the consumption of nonrenewable sacrificial reagents. Therefore, the real photocatalytic hydrogen evolution from water is now quite far away from practical application. As a means of dealing with the problems, the biomass glucose can be chosen as the sacrificial agent. Glucose is one of the most important building blocks in carbohydrate family and also good source for hydrogen production because it has a high hydrogen/carbon ratio and can be easily obtained from renewable biomass such as cellulose, starch and related industrial waste [9]. The original research was carried out by Kawai and Sakata in 1980. Their research suggested that biomass, such as cellulose, starch, wheat straw, and others waste materials, after a series of photocatalytic reactions can produce hydrogen [10]. These works demonstrate that hydrogen production from biomass is achievable by photocatalytic process. The method of photocatalytic hydrogen evolution from biomass glucose decomposition not only achieves the purpose of environmental remediation, but also provides the practical and low cost technologies in the hydrogen based energy system.

One of the most challenging tasks in the photocatalytic hydrogen evolution is to develop a photocatalyst that can work efficiently under solar light irradiation. Various kinds of semiconductors to photocatalytic reaction have gained increasing attention of researchers around the world [11]. Among various semiconductors, titanium dioxide (TiO_2) has been considered as the most promising material and extensively application due to its inexpensiveness, high physical and chemical stability, non-toxicity, and excellent photocatalytic activity [12], but it is still far from being a perfect and ideal photocatalyst. The main disadvantage of TiO_2 is that it is active only under UV light due to its wide band gap (E_{bg} = 3.2 eV) [13]. However, the UV light is less than 5.0% of solar spectrum. Some actions have been taken to improve the utilization ratio of sunlight when TiO_2 is used as the photocatalyst. Many researchers carried out metal ions (Au or Pt) [14] or non-metallic elements (C or S) doped TiO_2 [15], and narrow gap semiconductor coupling [16] to extend the range of absorption wavelength. The absorption spectrum of TiO_2 has been widened to a certain extent by means of above methods. Nevertheless the photocatalytic performance of the catalyst becomes lower along with its band gap narrow down. In contrast with the traditional ideas, we have been engaged in the research of TiO_2 combined with upconversion luminescence agents to perform the simulated visible light photocatalytic degradation of pollutants and achieved some satisfactory results in the past few years [17,18]. The high degradation effect can be attributed to some special properties of the upconversion luminescence agents, which can generate one high energy photon by absorbing two or more incident low energy photons. As well known some upconversion luminescence agents, such as $NaYF_4$:Yb/Er, $CaSc_2O_4$:Yb/Er, Er^{3+}:$Y_3Al_5O_{12}$ and Er^{3+}:$YAlO_3$ have been studied detailedly [19,20]. Among them, the Er^{3+}:$YAlO_3$ has been extensively used in many fields on account of its chemical stability and high upconversion luminescence efficiency. So, in this article the Er^{3+}:$YAlO_3$ was used as upconversion luminescence agents into TiO_2 to obtain a novel Er^{3+}:$YAlO_3$/Pt–TiO_2 photocatalyst to carry out the visible light photocatalytic hydrogen evolution with simultaneous reforming of biomass. Nevertheless, suspended Er^{3+}:$YAlO_3$/Pt–TiO_2 powder generally present some shortcomings. That is, the Er^{3+}:$YAlO_3$/Pt–TiO_2 particles are difficult to separation from aqueous solution, also the filtration process have high requirements for instruments and equipment in the experiment and practical application. It is harmful to regeneration and recycle of catalysts [21]. Moreover, the catalyst particles may become a cause of turbidity in the drainage as a secondary pollutant. In order to solve these problems and realize this technology for large scale water treatment, many studies have implemented to support TiO_2 on different substrate materials as membranes or thin films in photocatalytic degradation of organic dye in recent years [22]. Thus, we designed the photocatalyst as the form of membrane supported on the glass substrate by sol–gel method. The Er^{3+}:$YAlO_3$/Pt–TiO_2 composite membranes can be easily separated from water and are advantageous to regeneration and recycle, which has a great potential in photocatalytic hydrogen evolution and organic wastewater treatment using solar energy.

In this study, the Er^{3+}:$YAlO_3$/TiO_2 membrane was prepared through sol–gel and calcination method. Lastly, the Pt as the cocatalyst was deposited on the surface of composite membrane and the Er^{3+}:$YAlO_3$/Pt–TiO_2 membrane was obtained and used as a photocatalyst for the photocatalytic hydrogen evolution with simultaneous photocatalytic reforming of biomass under visible light irradiation. Meanwhile, some main influence factors on the visible light photocatalytic activity of Er^{3+}:$YAlO_3$/Pt–TiO_2 membranes were studied. Furthermore, the mechanism of upconversion luminescence process of Er^{3+}:$YAlO_3$ and the visible light photocatalytic hydrogen evolution principle of Er^{3+}:$YAlO_3$/Pt–TiO_2 membrane under visible light irradiation were proposed.

2. Experimental

2.1. Materials and apparatus

Er_2O_3 (99.99% purity), Y_2O_3 (99.99% purity) and $Al(NO_3)_3·9H_2O$ (analytically pure), citric acid (analytically pure) and HNO_3 (analytically pure) (Veking Company, China) were used to synthesize the Er^{3+}:$YAlO_3$ as upconversion luminescence agent. $Ti(OBu)_4$ (TBT, analytically pure, Sinopharm Chemical Reagent Co., Ltd., China) as titanium dioxide (TiO_2) precursor was used to prepare the TiO_2 membrane. Hexachloroplatinic acid hexahydrate ($H_2PtCl_6·6H_2O$, 97% purity, Sinopharm Chemical Regent Co., Ltd, China) was used as co-catalyst precursor for preparing the Er^{3+}:$YAlO_3$/Pt–TiO_2 photocatalysts. Glucose (analytically pure) (Veking Company, China) was used as sacrificial agent. All the reagents were of analytical purity grade, and were used without further purification. Experiments were conducted in a photocatalytic reactor. The volume of the reactor was 500 mL. The outside of the reactor was a water jacketed to keep a stable reaction temperature (25 °C). The sheet glass (75 mm (length) × 25 mm (width) × 2.2 mm (thickness)) was used as the supports of Er^{3+}:$YAlO_3$/Pt–TiO_2 membrane. 300 W xenon lamp (LX-300, Deruifeng hardware electrical appliance businesses, China) was used as simulated visible light irradiation source. Water in this experiment was purified by a Milli-Qwater system (Millipore Company, USA) and used throughout.

2.2. Preparation of visible light photocatalyst (Er^{3+}:$YAlO_3$/Pt–TiO_2 membranes)

2.2.1. Synthesis of Er^{3+}:$YAlO_3$ as upconversion luminescence agent

The upconversion luminescence agent of Er^{3+}:$YAlO_3$ was synthesized by the sol–gel and calcination method. Er_2O_3 (0.0128 g) and Y_2O_3 (2.2715 g) powders were dissolved in HNO_3 (30 mL) with magnetic stirring and heated (about 60 °C) until transparent yttrium nitrate and erbium nitrate solutions were obtained. Then, appropriate amount of aluminum nitrate solution was added to the above transparent solution and the uniform solution was obtained. Then citric acid as chelating agent and co-solvent was added into the above mixture solution (mol ratio of citric acid: metal ion is 3:1). The resulting solution was stirred and heated at 50–60 °C until the transparent sol was successfully prepared. Then, the sol was heated at 80 °C for 24 h and the xerogel was finally obtained, which was ground into fine homogeneous powders. The powders were heated at 500 °C for 50 min, and then were continuously heated up to 1100 °C and kept for 2.0 h and allowed

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