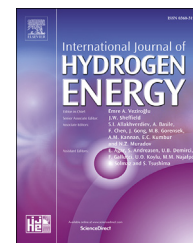


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Sol-gel synthesis of $\text{Er}^{3+}:\text{Y}_3\text{Al}_5\text{O}_{12}/\text{TiO}_2-\text{Ta}_2\text{O}_5/\text{MoO}_2$ composite membrane for visible-light driving photocatalytic hydrogen generation

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ABSTRACT

By using TiO_2 and Ta_2O_5 colloids, a stable and efficient visible-light driven photocatalyst, $\text{Er}^{3+}:\text{Y}_3\text{Al}_5\text{O}_{12}/\text{TiO}_2-\text{Ta}_2\text{O}_5/\text{MoO}_2$ composite membrane, was successfully prepared via sol-gel dip coating method at room temperature. The XRD, FTIR, SEM, TEM and EDX results confirm that approximately spherical $\text{Er}^{3+}:\text{Y}_3\text{Al}_5\text{O}_{12}$ nanoparticles were embedded in $\text{TiO}_2-\text{Ta}_2\text{O}_5$ matrix. UV-vis absorption and PL spectra of $\text{Er}^{3+}:\text{Y}_3\text{Al}_5\text{O}_{12}$ were also determined to confirm the visible absorption and ultraviolet emission. The photocatalytic hydrogen generation was carried out by using methanol as sacrificial reagent in aqueous solution under visible-light irradiation. Furthermore, some main influence factors such as heat-treated temperature, heat-treated time and molar ratio of TiO_2 and Ta_2O_5 on visible-light photocatalytic hydrogen generation activity of $\text{Er}^{3+}:\text{Y}_3\text{Al}_5\text{O}_{12}/\text{TiO}_2-\text{Ta}_2\text{O}_5/\text{MoO}_2$ composite membrane were studied in detail. The experimental results showed that the photocatalytic hydrogen generation activity of $\text{Er}^{3+}:\text{Y}_3\text{Al}_5\text{O}_{12}/\text{TiO}_2-\text{Ta}_2\text{O}_5/\text{MoO}_2$ composite membrane heat-treated at 550 °C for 3.0 h was highest when the molar ratio of TiO_2 and Ta_2O_5 was adopted as 1.00:0.50. And that a high level photocatalytic activity can be still maintained after four cycles. In addition, a possible mechanism for the visible-light photocatalytic hydrogen generation of the $\text{Er}^{3+}:\text{Y}_3\text{Al}_5\text{O}_{12}/\text{TiO}_2-\text{Ta}_2\text{O}_5/\text{MoO}_2$ membrane was proposed based on PL spectra.

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Introduction

Hydrogen energy is recognized as clean and green fuel. Recently, as a representative of the zero emission energy, so hydrogen energy stands out from many energy sources [1–3].

Simultaneously, hydrogen energy is also an ideal alternative energy, which is expected to replace fossil fuels and relieve energy crisis. Therefore, many scientists have been struggling for decades to explore how to obtain sustainable and clean hydrogen [4–6]. The conversion and store of energy in the

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form of photocatalytic water splitting to hydrogen generation holds great promise for facilitating sustainable development. Since Fujishima and Honda found that TiO_2 electrodes can be used as catalyst for hydrogen generation in 1972, numerous studies related to the photocatalytic performance of metal oxide semiconductor have received much attention [7–11]. Up to now, TiO_2 semiconductor catalyst has been still widely used in every field due to its good advantages of non-toxicity, rich source, environment-friendly and easy to obtain economic benefits [12–14]. High photocatalytic hydrogen generation capability of catalyst is closely associated with the utilization rate of light and the recombination rate of photo-generated electrons (e^-) and holes (h^+). In order to further enhance the photocatalytic hydrogen generation activity of TiO_2 , we mainly discussed the two aspects in detail.

On the one hand, under light irradiation with appropriate wavelength, the excited electrons (e^-) from valence band (VB) into conduction band (CB) of TiO_2 , which have the strong reducibility, and can sufficiently restore the hydrogen ions (H^+) to generate hydrogen (H_2) [15,16]. However, photo-generated electrons (e^-) and holes (h^+) of TiO_2 have high recombination rates, which decrease the photocatalyst hydrogen generation activity of TiO_2 . As we all know, oxidation reaction and reduction reaction occur simultaneously during the photocatalytic reaction process. We can appropriately enhance oxidation capacity of TiO_2 system to induce the oxidation-reduction reaction smoothly. Therefore, it is necessary to select another photocatalyst that has the lower VB potential and slightly higher CB potential compared with that of TiO_2 , then to combine with TiO_2 . It not only can completely separate photo-generated electron-hole pairs, but also facilitate oxidation-reduction reaction. Among the numerous photocatalyst, Ta_2O_5 , a wide band gap ($E_g = 3.9$ eV) semiconductor, has relatively high efficiency of photocatalytic hydrogen generation and it meets the above criteria [17–20]. TiO_2 combined with Ta_2O_5 can form an assumed electron-hole recombination center that consists of VB of TiO_2 and CB of Ta_2O_5 , which consumed the h^+ in the VB of TiO_2 and electrons (e^-) in CB of Ta_2O_5 . And then, photo-generated holes (h^+) in the VB of Ta_2O_5 oxidize sacrificial agent and photo-generated e^- in the CB of TiO_2 reduce hydrogen ions (H^+) to hydrogen (H_2) [21–24].

On the other hand, TiO_2 ($E_g = 3.2$ eV) and Ta_2O_5 ($E_g = 3.9$ eV) can be stimulated only under ultraviolet-light ($\lambda < 380$ nm) irradiation to carry out the photocatalytic reaction due to their wide band gap. Nevertheless, ultraviolet-light makes up only 5.0% of the total incoming solar radiation [25,26]. In order to enhance utilization ratio of visible-light and obtain adequate ultraviolet-light, a lot of researches have been performed. Among the numerous researches, the study of up-conversion luminescence has been considered to be one of the most promising methods for enhancing utilization efficiency of visible-light because it can convert low-energy visible-light into high-energy ultraviolet-light [27–29]. As a representative of up-conversion luminescence agent, $\text{Er}^{3+}:\text{Y}_3\text{Al}_5\text{O}_{12}$, has a broad development prospect, which can commendably meet the requirements of TiO_2 – Ta_2O_5 composite for ultraviolet-light [30,31]. In this way, photocatalytic activity of TiO_2 is significantly enhanced.

In addition, in the catalyst system, we use membrane as a convenience method for recycle and separate material for

effectively solving the problem of catalyst particles in suspension [32]. In order to fabricate the membrane of photocatalyst with nanostructures, several preparation methods such as thermal evaporation, electron-beam evaporation, sputtering, pulsed laser deposition and several other techniques have been used [33–36]. In the present study, $\text{Er}^{3+}:\text{Y}_3\text{Al}_5\text{O}_{12}/\text{TiO}_2$ – Ta_2O_5 composite membrane was prepared by a sol-gel dip coating method fixed on the glass substrate, which avoids the loss of catalyst and achieves the purpose of repeatedly uses for composite catalyst. In recent years, the research showed that the co-catalyst play an auxiliary role in terms of photocatalyst hydrogen generation because it can rapid transfer electrons (e^-) and act as active sites for hydrogen generation [37–39]. To further improve the photocatalytic activity in this study, we obtained a new $\text{Er}^{3+}:\text{Y}_3\text{Al}_5\text{O}_{12}/\text{TiO}_2$ – $\text{Ta}_2\text{O}_5/\text{MoO}_2$ composite membranes photocatalyst used the low-cost MoO_2 as co-catalyst.

Here, an up-conversion luminescence agent $\text{Er}^{3+}:\text{Y}_3\text{Al}_5\text{O}_{12}$ and a co-catalyst MoO_2 were obtained by sol-gel and hydrothermal method, respectively. And then, a novel visible-light photocatalyst, $\text{Er}^{3+}:\text{Y}_3\text{Al}_5\text{O}_{12}/\text{TiO}_2$ – $\text{Ta}_2\text{O}_5/\text{MoO}_2$ composite membrane, was successfully prepared by sol-gel dip coating method. The research investigated the visible-light photocatalytic performance of $\text{Er}^{3+}:\text{Y}_3\text{Al}_5\text{O}_{12}/\text{TiO}_2$ – $\text{Ta}_2\text{O}_5/\text{MoO}_2$ by using methanol aqueous solution as sacrificial agent. In addition, the effects of heat-treated temperature, heat-treated time and the molar ratio of TiO_2 and Ta_2O_5 on photocatalytic activity are explored. The mechanism of up-conversion luminescence process of $\text{Er}^{3+}:\text{Y}_3\text{Al}_5\text{O}_{12}$ and excitation principle of $\text{Er}^{3+}:\text{Y}_3\text{Al}_5\text{O}_{12}/\text{TiO}_2$ – $\text{Ta}_2\text{O}_5/\text{MoO}_2$ membrane as photocatalyst for visible-light photocatalytic hydrogen generation was proposed. This study on $\text{Er}^{3+}:\text{Y}_3\text{Al}_5\text{O}_{12}/\text{TiO}_2$ – Ta_2O_5 composite membrane may offer a more promising method for the large-scale solar-light photocatalytic hydrogen generation.

Experimental

Materials

Er_2O_3 (99.999%), Y_2O_3 (99.999%) and Al (NO_3) $_3$ ·9 H_2O (99%, analytically pure), citric acid ($\text{C}_6\text{H}_8\text{O}_7$) (analytically pure) and HNO_3 (65%, analytically pure, Veking Company, China) were used to synthesize the up-conversion luminescence agent ($\text{Er}^{3+}:\text{Y}_3\text{Al}_5\text{O}_{12}$). Tetrabutyl titanate (TBT) ($\text{Ti}(\text{OBu})_4$, $\text{C}_{16}\text{H}_{36}\text{O}_4\text{Ti}$, analytically pure, Sinopharm Chemical Reagent Co., Ltd., China) and Tantalum ethylate ($\text{Ta}(\text{OEt})_5$, 99.9%, Sinopharm Chemical Reagent Co., Ltd., China) were used to prepare the TiO_2 – Ta_2O_5 composite membrane. MoO_3 (99.5%, Shanghai Huayi Group Huayuan Chemical Industry Co., Ltd, China), Mo (99.5%, Aladdin Industrial Corporation, China), NH_4Cl (99.5%, Sinopharm Chemical Reagent Co., Ltd, China) were used as co-catalysts precursor. All chemicals were used without further purification and all experiments were carried out using deionized water.

Synthesis of $\text{Er}^{3+}:\text{Y}_3\text{Al}_5\text{O}_{12}$

The up-conversion luminescence agent, $\text{Er}^{3+}:\text{Y}_3\text{Al}_5\text{O}_{12}$, was prepared by sol-gel and calcination method [27–31]. At first,

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