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Highly efficient visible-light driven photocatalytic hydrogen production from a novel Z-scheme Er^{3+}:$YAlO_3$/Ta_2O_5-V^{5+}||Fe^{3+}-TiO_2/Au coated composite

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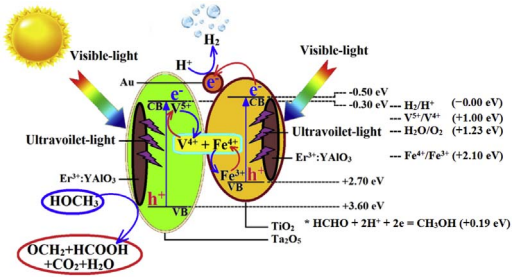
HIGHLIGHTS

- A novel Z-scheme system of Ta_2O_5-V^{5+}||Fe^{3+}-TiO_2 was designed for the first time.
- Er:YAP/Ta_2O_5-V^{5+}||Fe^{3+}-TiO_2/Au shows high visible-light photocatalytic activity.
- Er:YAP can offer ultraviolet-light to drive Ta_2O_5-TiO_2 photocatalytic H_2 evolution.
- V^{5+} and Fe^{3+} doping can promote the photo-generated electron and hole separation.
- Au nanoparticles as active sites can enhance visible-light photocatalytic activity.

GRAPHICAL ABSTRACT



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ABSTRACT

In this work, the preparation of a novel Z-scheme photocatalyst, Er^{3+}:$YAlO_3$/Ta_2O_5-V^{5+}||Fe^{3+}-TiO_2/Au coated composite, for visible-light photocatalytic hydrogen production is reported for the first time. In this photocatalyst, Au nanoparticles as conduction band co-catalyst provide more active sites to enrich electrons. Ta_2O_5-V^{5+}||Fe^{3+}-TiO_2 as composite redox cycle system thoroughly separates the photo-generated electrons and holes. In addition, Er^{3+}:$YAlO_3$ as up-conversion luminescence agent (from visible-light to ultraviolet-light) provides enough ultraviolet-light for satisfying the energy demand of wide band-gap semiconductors (TiO_2 and Ta_2O_5). The photocatalytic hydrogen production can be achieved from methanol as sacrificial agent (electron donor) under visible-light irradiation. The main influence factors such as initial solution pH and molar ratio of TiO_2 and Ta_2O_5 on visible-light photocatalytic hydrogen production activity of Er^{3+}:$YAlO_3$/Ta_2O_5-V^{5+}||Fe^{3+}-TiO_2/Au coated composite are discussed in detail. The results show that the Er^{3+}:$YAlO_3$/Ta_2O_5-V^{5+}||Fe^{3+}-TiO_2/Au coated composite with 1.0:0.5 M ratio of TiO_2 and Ta_2O_5 in methanol aqueous solution at pH = 6.50 displays the highest photocatalytic hydrogen production activity. Furthermore, a high level of photocatalytic activity can be still maintained within three cycles under the same conditions. It implies that the prepared Z-scheme Er^{3+}:$YAlO_3$/Ta_2O_5-V^{5+}||Fe^{3+}-TiO_2/Au coated composite may be a promising photocatalyst utilizing solar energy for hydrogen production.

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1. Introduction

Energy is the foundation of human production activities as well as social and economic development. Now, the severe energy shortage and excessive reliance on fossil fuels have caused a series of environment issues, which have affected the normal life of human beings [1,2]. Many scientists are pushing ahead with research on the renewable and environment-friendly energy so as to effectively solve these global issues. The solar energy is really inexhaustible source, which will likely play a crucial role in the development of new energy sources in some extent [3,4]. Hydrogen, a special energy carrier, is clean and, in practice, can be generated by photocatalytic water splitting. This means that the hydrogen can be obtained through the conversion of solar energy. The metal oxide semiconductors are considered to be the most promising photocatalysts in the field of photocatalytic hydrogen production from water splitting [5–7]. Since Fujishima and Honda firstly found semiconductor titanium dioxide (TiO_2) could be used as catalyst for hydrogen production in 1972, up to now, it has been attracted considerable attentions in some areas such as photosensitive solar cells, nonlinear optical applications and waste water recovery [8–10]. TiO_2 has many outstanding advantages, such as non-toxicity, rich source and corrosion resistance, but it also has some drawbacks including high recombination rates of photo-generated electron-hole pairs and large band-gap. Particularly, the recombination rate of photo-generated electron-hole pairs is faster than the rate of chemical interaction between TiO_2 and sacrificial agent, reducing the photocatalytic hydrogen production efficiency of TiO_2 [11,12]. Hence, searching superior method to improve transfer efficiency photo-induced charge carrier in photocatalyst is necessary. Combination of two different semiconductors and metal ion doping has been considered to be valid methods for separating photo-generated electrons (e^-) and hole (h^+), and thereby enhancing photocatalytic hydrogen production activity [13–15]. In addition, metal ion doping also can effectively broaden the range of the optical response. It is assumed that a more effective Z-scheme photocatalytic system would be formed if the two methods of different semiconductor combination and metal ion doping are used in conjunction.

Among the numerous photocatalysts, tantalum pentoxide (Ta_2O_5), as a remarkable semiconductor photocatalyst, has been considered to be almost comparable to the landmark photocatalyst of TiO_2 owing to its excellent optical properties characteristic [16–19]. In this context, we would like to report the composite Z-scheme photocatalyst that comprises of wide band-gap semiconductor TiO_2 with more negative conduction band (CB) and the other wide band-gap semiconductor Ta_2O_5 with more positive valence band (VB). However, the disadvantage of conventional Z-scheme is that the electrons (e^-) flow rate is slow and lacks a conductive channel. In order to promote the effective transfer of photo-generated electrons (e^-), it has been reported that the existence of Au nanoparticles (Au NPs) and graphene served as conductive passageway to effectively enhance the recombination efficiency of electrons (e^-) and hole (h^+) [20,21]. However, the introduction of conductive channels often increases the operating distance of electrons (e^-) and hole (h^+). To address this issue, for the first time, we doped different metal ions into the bases of TiO_2 and Ta_2O_5, respectively, to form a composite center of circular redox reaction. In general, the doping of transition metal ions (Mn, Co, Ni, Fe and V) can significantly widen light response range of wide band-gap photocatalysts and enhance the visible-light photocatalytic activity [22–26]. In addition, the bimetallic co-doping (Ni-Cu, Zn-Fe and Cr-Nb) into catalyst has also been proved to be an effective strategy for charge separation [27–31]. Specifically, the bimetallic co-doping couple of Fe^{3+} and V^{5+} is of great interest due to their diversity of valence states [32–38]. The generated Fe^{4+} in Fe^{3+} doped TiO_2 and the generated V^{4+} in V^{5+} doped Ta_2O_5 have strong oxidation power and strong reduction power, respectively. In this way, Fe^{4+} and V^{4+} easily undergo rapid redox reaction in controlled recombination center, which can be constructed by Fe^{3+}-TiO_2 and Ta_2O_5-V^{5+}. Because of the existence of composite redox cycle system (Ta_2O_5-V^{5+}||Fe^{3+}-TiO_2), the photogenerated electrons (e^-) on the conduction band (CB) of Ta_2O_5 can recombine with holes (h^+) on the valence band (VB) of TiO_2, so that the holes (h^+) on the valence band (VB) of TiO_2 and electrons (e^-) on the conduction band (CB) of Ta_2O_5 are completely separated, obviously improving the photocatalytic activity.

However, as wide band-gap semiconductors, TiO_2 (E_{bg} = 3.2 eV) and Ta_2O_5 (E_{bg} = 3.9 eV) can be excited only under appropriate wavelength of ultraviolet-light irradiation ($\lambda \leq 387$ nm and $\lambda \leq 342$ nm, respectively) and that the ultraviolet-light only accounts for a small portion of solar light, which limits the application of TiO_2 and Ta_2O_5 [39–42]. In order to enhance the utilization ratio of solar light, some researchers have made tremendous efforts to expand the light responding region and obtained remarkable achievements. For example, the use of up-conversion of luminescent agents such as Er^{3+}:$Y_3Al_5O_{12}$, Y_2SiO_5:Pr^{3+}, Li and Tm^{3+},Yb^{3+}:$NaYF_4$ have displayed the important role for expanding the light responding region because they can convert low energy photons to higher energy photons [43,44]. That is, these up-conversion of luminescent agents can transform the visible-light into ultraviolet-light [45–48]. Therefore, as an outstanding representative up-conversion luminescence agents, the Er^{3+}:$YAlO_3$ is used to combine with Ta_2O_5-V^{5+}||Fe^{3+}-TiO_2 composite photocatalys to provide the rich ultraviolet-light for satisfying the genuine requirement [49,50].

To some extent, the problems on recombination of photo-generated electrons (e^-) and holes (h^+) and low utilization rate of solar light can be solved by using above mentioned methods. However, there is still a challenge that is to accelerate the electron transfer rate for high photocatalytic hydrogen production efficiency. The studies showed that the loading of some noble metals (Au, Pt and Ag) as co-catalyst could transfer the electrons on the conduction band (CB) of photocatalysts and then make them be captured by hydrogen ions (H^+) forming hydrogen [51–53]. Therefore, in this work the Au nanoparticles as co-catalyst were deposited on the surface of TiO_2 to enrich photo-generated electrons (e^-), thus further increasing the photocatalytic hydrogen evolution activity [54–57].

Herein, a superior up-conversion luminescence agent, Er^{3+}:$YAlO_3$, was synthesized by sol-gel methods. And then, a novel visible-light driven nanocomposite photocatalyst, Er^{3+}:$YAlO_3$/Ta_2O_5-V^{5+}||Fe^{3+}-TiO_2/Au coated composite, was successfully prepared via sol-gel-hydrothermal method. It was placed in methanol aqueous solution to carry out an efficiency test of the hydrogen production under visible-light irradiation. The photocatalytic experiments showed that the present Er^{3+}:$YAlO_3$/Ta_2O_5-V^{5+}||Fe^{3+}-TiO_2/Au coated composite possessed of excellent photocatalytic activity for producing hydrogen under visible-light irradiation, which was much higher than that of Er^{3+}:$YAlO_3$/Fe^{3+}-TiO_2. In addition, the reusability of the Er^{3+}:$YAlO_3$/Ta_2O_5-V^{5+}||Fe^{3+}-TiO_2/Au was evaluated for the long-term application. This photocatalyst is expected to be applied in photocatalytic hydrogen generation by utilizing solar energy. The possible mechanisms of up-conversion luminescence of Er^{3+}:$YAlO_3$ and the related excitation principle of Er^{3+}:$YAlO_3$/Ta_2O_5-V^{5+}||Fe^{3+}-TiO_2/Au photocatalyst were also discussed in detail.

2. Experimental

2.1. Materials and reagents

Er_2O_3 (99.999%, Veking Company, China), Y_2O_3 (99.999%, Veking Company, China) and $Al(NO_3)_3{\cdot}9H_2O$ (99%, analytically pure, Veking Company, China), citric acid ($C_6H_8O_7$) (analytically pure, Veking Company, China) and HNO_3 (65%, analytically pure, Veking Company, China) were used to synthesize the up-conversion luminescence agent (Er^{3+}:$YAlO_3$). Tetrabutyl titanate (TBT) ($Ti(OBu)_4$, $C_{16}H_{36}O_4Ti$, analytically pure, Sinopharm Chemical Reagent Co, Ltd, China) and Tantalum ethylate ($Ta(OEt)_5$, $C_{10}H_{25}O_5Ta$, 99.9%, Sinopharm Chemical