



# Construction of dual-channel for optimizing Z-scheme photocatalytic system



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## ABSTRACT

A ternary all-solid-state visible-light Z-scheme photocatalytic system  $\text{BiVO}_4/\text{Au}@\text{CdS}$  is prepared by means of a two-step route, which is selectively built on the {010} facet of  $\text{BiVO}_4$  crystals in this system. Vectorial migration of photogenerated charge carriers through two order transmission channels in Z-scheme photocatalytic system further prolongs the mean free path of electrons, realizing effective separation of photogenerated charge carriers and possesses high redox ability. As expected, such  $\text{BiVO}_4/\text{Au}@\text{CdS}$  Z-scheme photocatalytic system exhibits highly efficient photocatalytic performance. Our current work is expected to offer new insight into construction of dual separation channels for rational design and compound of photocatalyst with high performance.

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## 1. Introduction

In the past decades, semiconductor photocatalysis as a promising and eco-friendly technology has attracted extensive attention due to the energy shortages and environmental crisis problem [1–6]. Owing to the limitation of narrow light responsive range and fast recombination rate of photogenerated charge carriers [7–11], photocatalytic technology doesn't achieve breakthrough in term of its practical application. Therefore, it is extremely urgent to develop visible-light-responsive photocatalysts with superior carrier separation efficiency.

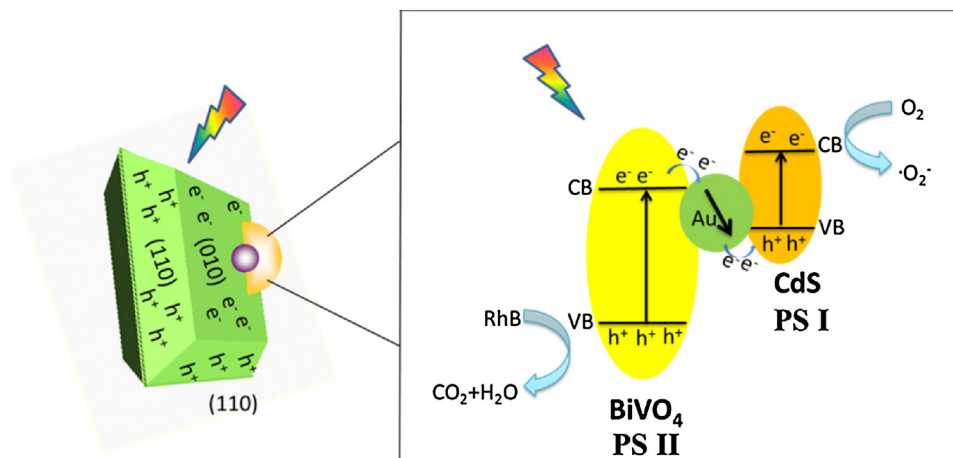
As known, the so-called Z-scheme photocatalytic process resembles natural photosynthesis which is the most important light-dependent reaction in nature. Both of photosynthesis and Z-scheme photocatalytic process include electrons (or energy) exchange among various functional components and energy transfer at the interface. Although natural photosynthesis undergoes a series of photo-induced species transmission among nanoscale architectures generally containing light-harvesting complexes, protein-metal clusters and a group of redox biocatalysts, it still can achieve high conversion efficiency. A crucial factor is the efficient electronic energy-transfer from the photo-excited chromophores to the catalytic clusters for chemical reactions [12], which is based on the accurate assemble of various functional components assuring energy transfer in an orderly way. Therefore, an efficient

photochemical system could be achieved by selecting appropriate components. Artificial Z-scheme photocatalytic system contains the photosystem I and II (hereafter referred to as PS I and PS II), as shown in Scheme 1 [13]. In Z-scheme photocatalytic process, the photogenerated electrons transfer from the conduction band (CB) of PS II to the valence band (VB) of PS I [14–17]. On one hand, the separation efficiency of photogenerated carriers can be improved through carriers transfer at the interface. On the other hand, separated electrons or holes possess higher redox ability, which is more beneficial to involve in related reactions. Therefore Z-scheme photocatalytic system has great potential to effectively utilize solar energy. In the case of artificial Z-scheme photocatalytic system, however, the complicated interfacial environment may lead to serious energy lose (low transfer rate of carriers [18,19] and opposite charge carriers accumulate at the surface [17,20]), resulting the efficiency of artificial Z-scheme photocatalytic system far away from the theoretical conversion efficiency. Thus, developing an efficient strategy to relieve the above-mentioned issues becomes a key step for optimizing the energy utilization efficiency of Z-scheme photocatalytic system.

Fortunately, some previous studies have proved that introducing electron mediators (including Au [21–23], Ag [16,24,25], reduced graphene oxide [26] and so on) at the interface between PS I and PS II could reduce the transmission impedance of electrons and improve electron transfer rate across the interface, in which electron mediators act as a transfer channel for carriers. Regrettably, because free electron and hole are photo-excited simultaneously at the surface of PS I or PS II, and the isotropic migration of photo-induced electron-hole pairs from the bulk to

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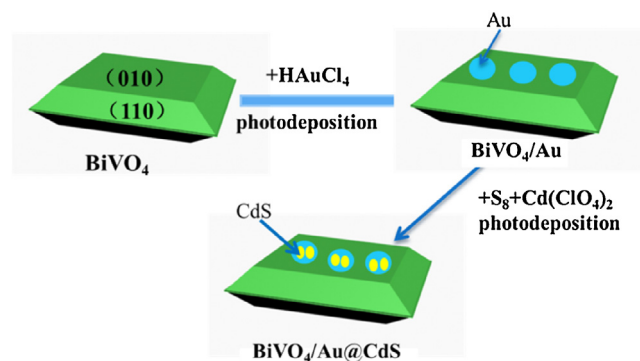
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**Scheme 1.** The proposed mechanism of photodegradation of RhB by  $\text{BiVO}_4/\text{Au}@ \text{Cds}$  under visible light irradiation.

surface of each semiconductor, opposite charge carriers can accumulate inevitably in the surface and its density will gradually increase, inducing serious surface self-recombination [20]. It's not difficult to imagine that surface self-recombination would be suppressed if extra pre-screening procedure is integrated in PS II to assure photogenerated electrons are selectively collected on the PS II. Alternatively, as we know, crystal facet engineering of various semiconductors ( $\text{TiO}_2$  [10,27],  $\text{BiVO}_4$  [28–30],  $\text{ZnO}$  [31],  $\text{WO}_3$  [32],  $\text{BiOI}$  [33]) could provide a new opportunity to drive free electrons/holes to the specific facets in PS II, due to the differences in the energy levels in the conduction band (CB) and valence band (VB) between two specific facets, achieving spatial charge separation. Namely, the photogenerated electrons transfer from a facet with more negative CB potential to another facet, while holes transfer in the opposite direction to the facet with more negative CB potential in PS II. In this case, facet engineering can act as a separation channel. Therefore, a predictive model, in which PS I is rationally arranged on the electron-gathered facets in PS II with a function of facet engineering collector and metal nanostructure embedding in the interface between PS II and PS I, is expected to be an integration of separation channel and transfer channel to improve energy transfer rate and decrease serious energy lose in the interface. Besides, this predictive system should be integration between the complementarities of interfacial composite engineering and crystal facet engineering. On one hand, interfacial metal nanostructure may transfer electron from PS II to PS I to maintain the constant potential difference between two facets. On the other hand, electrons gathering caused by facet-driving could provide more transferable electrons owing to suppression of bulk and surface self-recombination in PS II.

In order to realize the above project, the first thing is to choose a photocatalyst PS II with special facets. As a promising visible-light driving semiconductor photocatalyst, monoclinic bismuth vanadate ( $m\text{-BiVO}_4$ ) has attracted considerable attention due to its narrow band gap, good chemical and thermal stability, non-toxicity and good photocatalytic performance under visible light irradiation [30,34,35]. Recent studies confirmed that the conduction band edge and valence band edge of the {110} facet of  $m\text{-BiVO}_4$  are 0.42 and 0.37 eV respectively, which is higher than those of its {010} facet. The difference of individual energy band levels between {110} and {010} facets leads to the migration of holes and electrons to different facet, respectively [28,29]. Therefore,  $\text{BiVO}_4$  with special facets is chosen as the PS II in Z-scheme photocatalytic system. Au can be chosen as a transmission channel to transfer electrons. Besides, Au nanoparticle also can be an anchor for depositing CdS onto Au nanoparticle due to the strong affinity of Au atoms to sulphur [36].



**Fig. 1.** Schematic of the preparation route of  $\text{BiVO}_4/\text{Au}@ \text{Cds}$  Z-scheme photocatalytic system through a two-step process.

CdS with suitable band gap energy can be as the PS I in Z-scheme photocatalytic system.

Here, we report the all-solid-state visible-light Z-scheme photocatalytic system  $\text{BiVO}_4/\text{Au}@ \text{Cds}$ , which is selectively built on the {010} facet of  $\text{BiVO}_4$  crystals in this system. The schematic of the preparation route of the Z-scheme photocatalytic system is given in Fig. 1. Au nanoparticles only distribute on the {010} facets of  $\text{BiVO}_4$  crystals by facile photo-deposition method. The growth of CdS is initialized by illuminating the suspension containing the prepared Au/ $\text{BiVO}_4$ , S and  $\text{Cd}(\text{ClO}_4)_2$ . Under irradiation, S molecules are reduced to  $\text{S}^{2-}$  ions, which then selectively adsorbed on the surface of Au. The  $\text{S}^{2-}$  ions react with  $\text{Cd}^{2+}$  and form CdS, so that the CdS nanoparticles are naturally formed onto the surfaces of the Au nanoparticles [3,18,22]. The photocatalytic activity is investigated by photo-degradation organic dye. Such the Z-scheme photocatalytic system by means of meticulous design possesses the highest photocatalytic performance. Our current work is expected to offer new insight into the construction of multiple separation channels for rational design and synthesis of photocatalyst with high activity.

## 2. Experimental

### 2.1. Preparation

#### 2.1.1. Materials

Bismuth nitrate pentahydrate ( $\text{Bi}(\text{NO}_3)_3 \cdot 5\text{H}_2\text{O}$ , >99.0%), ammonium vanadate ( $\text{NH}_4\text{VO}_3$ , >99.9%), ammonia water ( $\text{NH}_3 \cdot \text{H}_2\text{O}$ , 25–28 wt%), nitric acid ( $\text{HNO}_3$ , 65–68 wt%), gold chloride ( $\text{HAuCl}_4 \cdot 4\text{H}_2\text{O}$ ), absolute ethanol and cadmium sulfide (CdS)

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