

Regular Article

Enhanced visible light activated hydrogen evolution activity over cadmium sulfide nanorods by the synergetic effect of a thin carbon layer and noble metal-free nickel phosphide cocatalyst

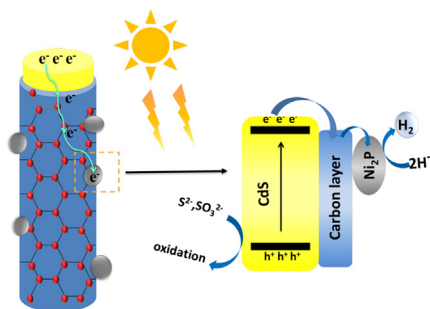


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GRAPHICAL ABSTRACT

Cheap and nontoxic carbon and nickel phosphide were used to modification the CdS nanorods. The synthesized CdS@C-Ni₂P shows excellent hydrogen evolution activity and stability.



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ABSTRACT

Photocatalytic water splitting is considered to be a promising strategy for addressing the global energy crisis through the expanded use of solar energy. Herein, cadmium sulfide (CdS) nanorods modified with a thin conductive carbon layer and a nickel phosphide co-catalyst, referred to as cadmium sulfide coated with a carbon layer and nickel phosphide (CdS@C/Ni₂P, where @ indicates a core-shell structure), were synthesized and applied as a novel composite photocatalyst for water splitting. The optimized CdS@C/Ni₂P composite showed a high photocatalytic hydrogen generation rate of 32030 μmol h⁻¹ g⁻¹, which was approximately 19 times as high as that of pure CdS. We believed that the thin carbon layer acted as an electron acceptor to promote charge transfer and protect the CdS nanorods from photocorrosion. In addition, the surface loading of the nickel phosphide (Ni₂P) cocatalyst was able to further draw photogenerated electrons from the cadmium sulfide coated with a carbon layer (CdS@C) heterojunction and provide active sites for hydrogen evolution. Thus, greatly enhanced hydrogen generation was achieved through a combination of carbon coating and surface cocatalyst loading. This development provides a new way to design composite photocatalysts with multiple junctions for efficient water splitting performance.

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1. Introduction

The depletion of fossil resources and the deepening crisis of environmental pollution from their consumption have motivated considerable effort to develop sustainable technologies [1–6]. Converting solar energy into hydrogen fuel is considered to be a promising way to generate clean and renewable energy [7–14]. In particular, solar-driven photocatalytic water splitting has received considerable attention owing to the high product purity generated by this simple process [15–17]. To date, many different semiconductor based photocatalysts have been developed and studied for solar-driven hydrogen generation [18–20]. The design of inexpensive photocatalysts with good activity and stability is considered to be essential to the conversion process.

Visible light driven cadmium sulfide is regarded as a promising material for photocatalytic hydrogen evolution among various photocatalysts because of its superior characteristics, such as a suitable band gap (~ 2.4 eV) and appropriate potential conduction band [21–23]. Unfortunately, bare CdS is not yet ready for practical applications owing to the relatively low efficiency of electron-hole separation and a tendency to undergo photocorrosion under irradiation [24,25]. A thin layer coated on the host photocatalyst can often facilitate the separation of light-induced electron-hole pairs. For example, Tang et al. showed that the intimate coaxial interfacial contact between a MoS₂ shell and CdS core facilitated the transfer of photoexcited electrons to MoS₂ [26]. As a promising material widely applied in the energy conversion field, carbon can also serve as a coating layer owing to its unique features, such as high conductivity, low-toxicity, and good chemical stability [27–29]. Recently, it was reported that carbon coated cuprous oxide nanorods showed enhanced photocatalytic performance, attributed to the carbon layer facilitating charge transfer [30]. Considering the remarkable properties of carbonaceous materials and the limitations of CdS photocatalytic systems, coating CdS with a thin carbon layer might be a suitable strategy for constructing efficient photocatalytic systems.

Active sites play an important role in the process of photocatalytic hydrogen production. Currently, noble-metal-based co-catalysts have been widely applied in photocatalytic systems to provide active sites for catalytic hydrogen evolution [31–33]. However, noble metals are rare and expensive. Therefore, there is a need to find noble-metal free co-catalysts with high efficiency. Of non-precious earth-abundant co-catalysts, transition metal phosphides have attracted considerable attention and have been confirmed to be suitable co-catalysts which can provide active sites to promote the hydrogen evolution of semiconductors [34,35]. For example, Xu et al. reported that Ni₂P can act as a highly efficient co-catalyst when loaded on the surface of carbon nitride [36].

In order to combine all the advantages mentioned above and improve the photocatalytic performance of CdS, we designed and synthesized a highly efficient ternary photocatalyst system in which a carbon layer and Ni₂P nanoparticles were used to modify CdS nanorods. The obtained hydrogen production yield reached 32030 $\mu\text{mol h}^{-1} \text{g}^{-1}$, which was 19 times as high as that of pristine CdS. The carbon layer coated on the CdS nanorods featured high conductivity and good chemical stability, providing electron transport pathways and maintaining the stability of host photocatalyst. Furthermore, surface modification with Ni₂P nanoparticles further improved the charge carrier separation and provided an abundance of active sites. Thus, excellent photocatalytic performance and stability were realized from the well-designed CdS@C/Ni₂P semiconductor composites.

2. Experimental

2.1. Materials

All chemicals were purchased and used without further purification. Cadmium chloride (CdCl₂·2.5H₂O, Sinopharm Chemical Reagent Co., Ltd, AR, $\geq 99.0\%$), thiourea (NH₂CSNH₂, Sinopharm Chemical Reagent Co., Ltd, AR, $\geq 99.0\%$), ethylenediamine (C₂H₈N₂, Sinopharm Chemical Reagent Co., Ltd, AR, $\geq 99.0\%$), ascorbic acid (C₆H₈O₆, Sinopharm Chemical Reagent Co., Ltd, AR, $\geq 99.7\%$), nickel nitrate hexahydrate [Ni(NO₃)₃·6H₂O, Sinopharm Chemical Reagent Co., Ltd, AR, $\geq 98.0\%$], sodium hydroxide (NaOH, Sinopharm Chemical Reagent Co., Ltd, AR, $\geq 96.0\%$), sodium hypophosphite (NaH₂PO₂, Aladdin Industrial Corporation, AR, $\geq 99.0\%$), sodium sulfide (Na₂S, Aladdin Industrial Corporation, AR, $\geq 99.99\%$), sodium sulfite (Na₂SO₃, Sinopharm Chemical Reagent Co., Ltd, AR, $\geq 97.0\%$), sodium sulfate (Na₂SO₄, Sinopharm Chemical Reagent Co., Ltd, AR, $\geq 97.0\%$) were used as provided.

2.2. Preparation of CdS nanorods

CdS nanorods were prepared by a reported method [37]. 9.26 g of CdCl₂·2.5H₂O and 9.26 g of thiourea in 120 mL ethylenediamine were added to a 200 mL Teflon-lined autoclave, which was maintained at 160 °C for 36 h. The yellow precipitates were collected and washed with absolute ethanol and deionized water several times. The obtained products were dried at 60 °C for 8 h in a vacuum oven.

2.3. Synthesis of CdS@C composites

CdS@C heterostructure was prepared through a hydrothermal method. A 0.1-g portion of CdS nanorods were added into a mixture of 30 mL ultrapure water and 30 mL ethanol. After stirring for 1 h, 0.01 g of ascorbic acid was added into the solution. After stirring for another 1 h, the above solution was transferred to a 100 mL Teflon-lined stainless-steel autoclave and treated at 200 °C for 2 h. After cooling to room temperature, the sample was collected, washed several times and dried at 60 °C for 8 h.

2.4. Preparation of Ni₂P nanoparticles

Ni₂P nanoparticles were synthesized according to a reported method [38]. First, an excess amount of NaOH solution (0.5 M) was added dropwise into 100 mL of an aqueous solution containing Ni(NO₃)₂·6H₂O (200 mg) and sodium citrate (50 mg) under vigorous stirring. After stirring for 30 min, the precipitates were collected by centrifugation and dried at 80 °C overnight in vacuum oven. Then, the Ni(OH)₂ and NaH₂PO₂ were mixed and ground in a mortar with a mass ratio of 1:5. The mixture was placed in a tube furnace and annealed at 300 °C for 1 h under an argon atmosphere (the ramp rate is 2 °C/min). The so-formed black solid was ground into powder, washed with deionized water and ethanol, and then dried in a vacuum oven overnight.

2.5. Preparation of CdS@C/Ni₂P composites

A certain amount of the prepared Ni₂P composites was added into 30 mL of ethanol and it was exposed to ultrasonic for 30 min to obtain a suspension. Then, 0.2 g of the CdS@C composites was added into the Ni₂P dispersion under stirring. After stirring for 1 h, the resulting suspension was transferred to a rotary evaporator to remove most of the ethanol before the products were dried in vacuum oven. We used the same method to obtain four samples

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