

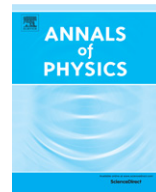


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Recent progress in photoelectrochemical water splitting for solar hydrogen production

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H I G H L I G H T S

- Some promising photoelectrode materials with narrow band gaps are discussed.
- Several strategies have been introduced to improve photoelectrodes' conversion efficiencies.
- Loading dual layer catalysts has been an effective method to enhance photoelectrodes' stability.

A R T I C L E I N F O

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A water splitting photoelectrochemical (PEC) cell can convert solar energy to hydrogen fuels directly. The challenges for practical application are to fabricate photoelectrodes with high efficiency, good durability and low cost. In this review, we focus on recent progress of some promising photoelectrode materials, including BiVO_4 , $\alpha\text{-Fe}_2\text{O}_3$, Ta_3N_5 photoanodes and $\text{Cu}_2\text{ZnSnS}_4$ photocathodes. Several new strategies to enhance the performance of a PEC cell, such as surface exfoliation, suppressing back reaction and loading dual-layer catalysts, are discussed.

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1. Introduction

Solar energy is regarded as one of the most promising renewable energies to support the sustainable development of human society. To utilize sunlight, three main problems should be solved:

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capture, conversion and storage. In photovoltaic cells, solar energy is converted into electric energy, which is difficult to be stored in a large scale at present. Moreover, the yield of electricity is unsteady owing to daily and seasonal variability of sunlight. Hydrogen is an ideal clean energy carrier for future. A photoelectrochemical (PEC) cell integrates capture and conversion in one step inexpensively. Since 1972 Fujishima and Honda reported photoelectrochemical water splitting on a TiO_2 photoanode, PEC cells have been investigated intensively [1]. There have been some excellent reviews on the fundamental principles of photoelectrochemical system [2,3]. Briefly, for semiconductor photoelectrodes, electrons are excited from valence band to conduction band under illumination and electron–hole pairs are generated. The electron–hole pairs are separated and react with water to produce hydrogen and oxygen. If the valence band maximum is lower than the water oxidation potential and conduction band minimum is higher than proton reduction potential, photogenerated electrons and holes are able to realize water splitting without a bias.

The challenges of large-scale application of PEC cells are to fabricate photoelectrodes with high efficiency, good durability and low cost. The band gap of TiO_2 is too wide so that it only absorbs a small portion of UV light in sunlight, which leads to a very low theoretical conversion efficiency. A p–n tandem cell, is considered as the most promising cell with high efficiency and low cost. In this cell, a n type semiconductor and a p type semiconductor are used as a photoanode for water oxidation and a photocathode for proton reduction, respectively. Up to date, some high-efficiency photoanodes with narrower band gaps, such as WO_3 (2.6 eV), Fe_2O_3 (2.1 eV) and BiVO_4 (2.4 eV), have been explored [4,5]. WO_3 and Fe_2O_3 photoanodes have been studied intensively for about forty years. Their conversion efficiencies have been enhanced remarkably. A BiVO_4 photocatalyst was firstly reported by Kudo's group in 1998. Up to date, its performance has been close to its maximum theory value. However, the conversion efficiencies of these metal oxide photoanodes are still low for practical application. In the recent 10 years, some nitrides and oxynitrides, such as TaON (2.5 eV), Ta_3N_5 (2.1 eV) and LaTiO_2N (2.1 eV), have been firstly reported as photocatalysts by Domen's group, which will be promising candidate photoanode materials for solar water splitting in future [6–8]. Moreover, some new photocathode materials have also been studied. In this review, we mainly discuss the recent progress (2013–2014) of some visible-light-response photoelectrodes, including $\alpha\text{-Fe}_2\text{O}_3$, BiVO_4 , Ta_3N_5 photoanodes, and $\text{Cu}_2\text{ZnSnS}_4$ (CZTS) photocathodes.

2. BiVO_4

Monoclinic sheelite BiVO_4 has attracted great interest since in 1998 Kudo and colleagues reported its high photocatalytic ability for water oxidation under visible light illumination [9]. The band gap of BiVO_4 is about 2.4 eV, with valence band maximum at *ca.* +2.4 eV and conduction band minimum a little lower than H^+ reduction potential, which suggests a bias is necessary for solar water splitting. There are good reviews of crystal and electronic structure of BiVO_4 [3,10]. The conduction band of BiVO_4 consists mainly of V 3d orbitals. V ions locate in non-interconnecting VO_4 tetrahedrons. As a result, the electrons have to hop between these tetrahedrons to transfer [11]. The carrier mobility of undoped BiVO_4 is $\sim 4.4 \times 10^{-2} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, which is 1–2 order lower than typical semiconductor photoanode materials, such as WO_3 [12]. Besides, severe recombination of photogenerated electron–hole pairs exists in the bulk and on the surface, which makes photoelectrochemical performance of a BiVO_4 photoanode very poor. Various attempts have been carried out to improve the performance, such as doping, loading co-catalysts and heterojunctions.

2.1. Doping

Doping is a widely used strategy to improve carrier transport properties of BiVO_4 . Luo and Zou et al. have reported that Mo^{6+} and W^{6+} are effective dopants [13]. Incident photon-to-electron conversion efficiency (IPCE) of Mo-doped BiVO_4 reaches 42% at around 400 nm, much higher than pure BiVO_4 . Moreover, it has been revealed that the Mo-rich segregation on the surface acted as recombination centers in Mo-doped BiVO_4 [14]. After removing the segregation with a facile electrochemical pretreatment, the photocurrent was further improved remarkably. Bard

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