



## Review

## Design and architecture of metal organic frameworks for visible light enhanced hydrogen production



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## ABSTRACT

MOFs offers a potential platform for anchoring photosensitizers and catalytic moieties, encapsulating metal nanoparticles, and supporting the semiconductor to drive chemical H<sub>2</sub> production reactions by taking the advantages of large surface area, permanent pores and tunable channels as well as tailorable physical and chemical functions. Recently, considerable attention has been paid to the application of MOFs in heterogeneous photocatalysis for H<sub>2</sub> production. In this review, the latest progress of MOFs involved in the reaction of H<sub>2</sub> production from water and visible-light-enhanced H<sub>2</sub> releasing from hydrogen carriers were summarized. The successful methods to maximize photocatalytic activity of H<sub>2</sub> production from water by introducing light sensitization and anchoring catalytic sites within MOFs, tuning the band gap of MOFs by modification of functional groups were also highlighted. Also the latest advances in heterogeneous organic dye-sensitized metal nanoparticle supported on MOFs for photocatalytic H<sub>2</sub> production and visible light enhanced H<sub>2</sub> production from H<sub>2</sub> carriers were emphasize. In the end, a brief perspective to the challenges and new development directions in the development of highly active MOFs to achieve high photocatalytic activity in the H<sub>2</sub> production is also discussed, which would be of great interest in the solar energy conversion.

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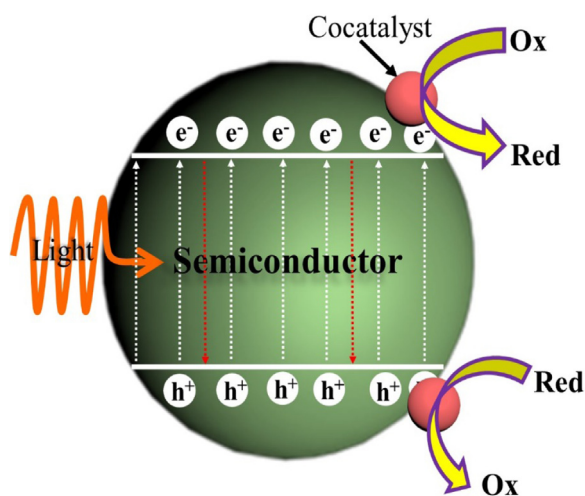


Fig. 1. Schematic illustration of photocatalytic reaction taking place on a semiconductor photocatalyst mediated by suitable redox cocatalysts.

## 1. Introduction

The worldwide problem of energy crises and environmental problem caused by the increasing use of the limited fossil fuels have received tremendous researcher's attention in the search for clean and renewable energy sources [1,2]. In fact solar energy is an inexhaustible natural resource. The magnitude of available solar energy striking on the surface of the earth ( $1.3 \times 10^5$  TW), which is much larger than the energy consumed by humanity per year ( $1.6 \times 10^1$  TW in 2010) [3], is a highly appealing source of natural energy. The conversion of solar energy into chemical energy in the form of chemical bonds is considered as a highly desirable approach to fulfill the requirement of sustainable development. Inspired by the natural photosynthesis of green plants and some other microorganisms with efficient converting solar energy into chemical energy in the form of carbohydrates or hydrogen, the field of photocatalysis has expanded rapidly over the past several years and it is recognized as the most promising technology for converting solar energy into useful chemical potential for synthetic purposes [4–6]. Especially, semiconductor-based photocatalysis has received considerable attention because of its potential applications in energy exploitation [7] and environmental purification [8], since the discovery of the Honda-Fujishima effect on water splitting for  $H_2$  production over  $TiO_2$  photoelectrodes in 1972 [9]. Increasing efforts have been devoted to explore light responsive materials for visible light driven photocatalytic reaction [10].

The development of new environment-friendly, low-cost and highly stable materials has become one of the most urgent and challenging issues related to global energy crisis and pollution problem [11]. Semiconductors are considered as one of the most promising photocatalysts for various photoredox reactions, because of suitable band gap, low cost, high chemical stability, and environmental benign [12]. Fig. 1 displays the schematic illustration of photocatalytic reaction taking place on a semiconductor photocatalyst mediated by suitable redox cocatalysts. When the energy of incident light is higher than that of bandgap, electrons are generated through band-gap excitation and transferred from the valence band to conduction band of photocatalyst particles, leaving the holes in the valence band. The photo-generated electrons may go through the following possible pathways: (1) transferred to surface active sites and subsequently involved in surface redox reaction, where the hole is involved in photocatalytic oxidation reaction; (2) captured by the defect sites in the bulk of semiconductors;

(3) recombine with photo-generated holes by releasing the excess energy to the surrounding environment.

Considering that the chemical reactions usually occur on the surface of photocatalysts, the redox reactions of adsorbed species with photoexcited electron-hole pairs and the recombination of photoexcited electron-hole pairs are two competing processes. The first pathway is favored, where the second and third pathway are not desirable. Therefore, reducing the recombination rate of photoexcited electron-hole pairs is the critical way to enhance the photocatalytic performance. On the other hand, solar light contains less than 5% of ultraviolet (UV) light, about 42–43% visible light (400–700 nm) [13], and 52–55% infrared (>700 nm). In the consideration of efficient utilization of visible light, the development of a photocatalyst with a wide range of visible light absorption is indispensable. However, most of the reported photocatalysts are only responsive to UV light [14]. To this end, numerous attempts have been made to promote the transformation of photo-excited electron-hole pairs from bulk to the surface of semiconductor and synthesis of visible light responsive semiconductor by various techniques, such as doping semiconductor with metal or nonmetal elements [15–18], morphology controlling [19,20], size turning [21], loading with plasmonic nanoparticles (such as gold, silver and copper) [22–24], forming heterojunction by coupling with narrow-band-gap semiconductors [25], or atomic structure engineering [26]. Apart from this, various types of visible light responsive semiconductors have also been explored and studied for artificial photosynthesis [27–29]. Even though recent intensive efforts and exciting achievements in the  $H_2$  production from water have been done [30], the development of visible light driven photocatalysts with narrow band gap that can efficiently utilize entire solar spectrum to driven  $H_2$  production with high catalytic activity and stability is still a challenge for the research community.

Metal-organic frameworks (MOFs) are constructed from inorganic metal ions as connecting centers and organic moieties as linkers. Such inorganic-organic polymers offer significant chemical diversity because they can be modified by functional groups and, they have been widely employed in many applications such as catalysis, gas sorption, energy storage, and membrane [31,32]. MOFs have several advantages over other inorganic materials [33]. Firstly, photoactive organic linker endows the MOFs semiconducting properties. Upon light irradiation, the organic linker acts as an antenna to capture light, electrons get excited from HOMO to LUMO state of organic linker, the photo-excited electrons further relay to nodes of MOFs through a linker-to-cluster charge-transfer mechanism (LCCT) to activate the metal nodes, subsequently involved in a heterogeneous photoredox reaction, such as  $CO_2$  reduction, water splitting, degradation of organic pollutants, or organosynthesis [34,35]. Secondly, controllable physical and chemical properties, extraordinarily high surface area and a great number of functional groups can allow the efficient adsorption and diffusion of reactant and product molecules [36]. Furthermore, it is easy to tailor the chemical properties of MOFs and at the same time to enrich structural diversity in MOFs through synthetic modification [37], the linker of MOFs can be integrally or partially replaced by other organic linker via the mix-and-match synthetic strategy [38]. Moreover, the nodes of MOFs can also be replaced by foreign metal ions [39]. It is clear that MOFs provide a great opportunity for heterogeneous photocatalysis due to the richness of metal-containing center and organic bridging linker, as well as the controllability of the synthesis. In the last five years, MOFs have been extensively exploited as photocatalysts for  $H_2$  production from water and have proved to be important alternatives to semiconductor photocatalysts and metal complexes photocatalysts. Tremendous efforts have been dedicated to design and architecture of visible-light-responsive MOFs for enhanced hydrogen production from water and the interest is continuously increasing. Thus, a review of the recent advances

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