



Noble-metal-free heterostructure for efficient hydrogen evolution in visible region: Molybdenum nitride/ultrathin graphitic carbon nitride

Shuaiqi Gong^a, ZhengJun Jiang^a, Penghui Shi^{a,b,*}, Jinchen Fan^{a,b}, Qunjie Xu^{a,b,*}, Yulin Min^{a,b,*}

^a Shanghai Key Laboratory of Materials Protection and Advanced Materials in Electric Power, Shanghai University of Electric Power, Shanghai 200090, PR China

^b Shanghai Institute of Pollution Control and Ecological Security, Shanghai 200092, PR China

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ABSTRACT

Molybdenum nitride (MoN) has been attracting increasing attention in recent years for the application of electrocatalytic hydrogen evolution reaction. However, the application of MoN for photocatalytic H₂ evolution has seldom been reported. Here, we report that the sample with the heterostructure between Mo₂N and 2D ultrathin g-C₃N₄ nanosheet (CN) is fabricated via boil bathing method for the use of the visible light photocatalytic H₂ evolution. Without using any noble metal cocatalyst, the H₂ evolution rate of Mo₂N/CN under visible light irradiation reaches 0.89 μmol g⁻¹ h⁻¹, which is ≈148 times higher than that of pure CN. The photoelectron-chemical experiments suggest that the carriers in Mo₂N/CN can separate and transfer easier than those in CN due to the effect of Mo₂N as the modification of CN. The combination of the Mo₂N and CN represents not only a simple but also an economical and powerful method for the highly effective photocatalytic H₂ generation in the visible region.

1. Introduction

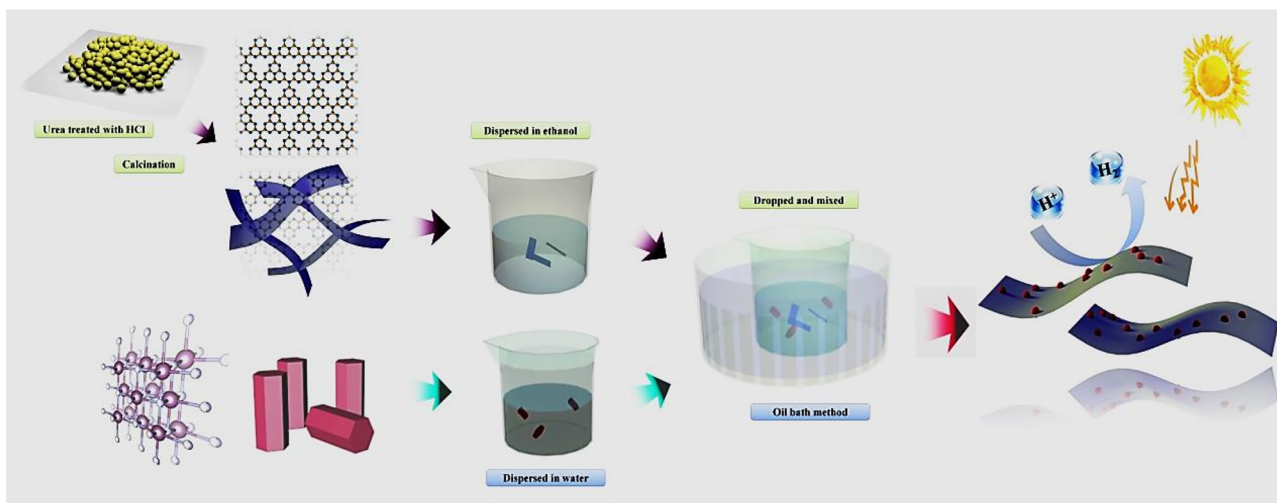
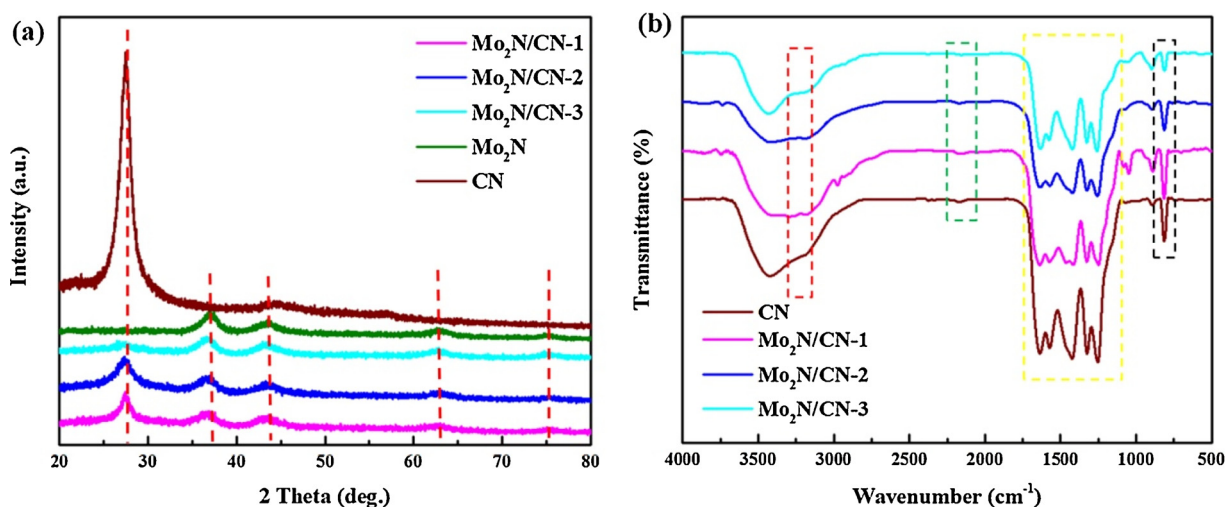
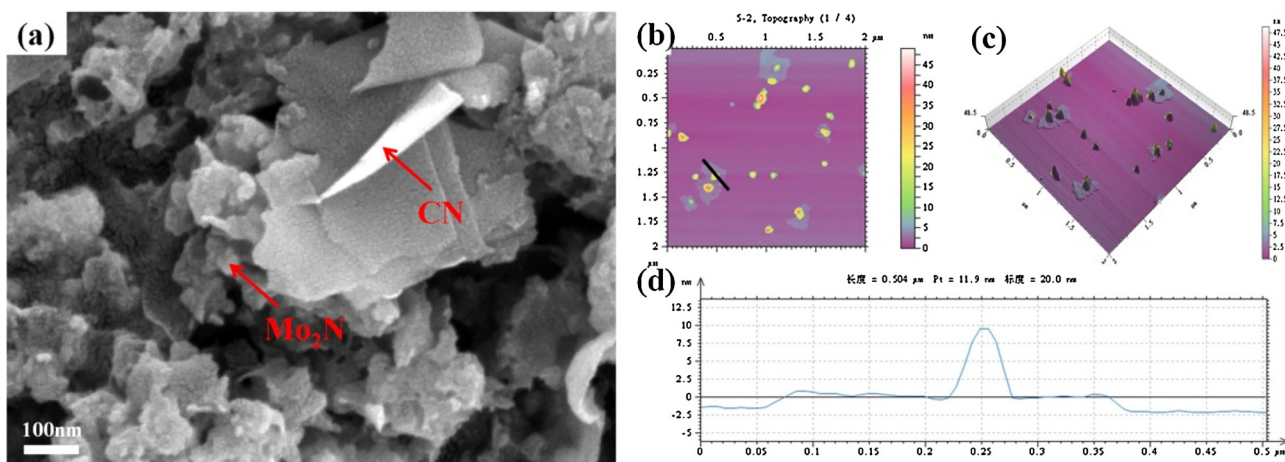
Concentrating on the energy crisis and the increasing environmental issues which are caused by the excessive consumption of fossil fuels, renewable and clean energy sources are intensively demanded in modern society [1]. Hydrogen (H₂) is undeniably pursued as one of the most promising candidates for green energy carries to solve the energy problems because of its zero carbon footprints and high mass-specific energy density [2,3]. Among all kinds of hydrogen evolution processes, splitting water using solar energy is believed to be the most sustainable option. In the past decades, researchers always aim to find high performance and low-cost photocatalysts toward water splitting. At present, the energy conversion efficiency of solar-to-hydrogen is still too low although H₂ can be obtained directly using semiconductor photocatalysts under light irradiation. The main barriers are the lack of suitable photocatalysts which can meet the following requirements: (1) absorbing broadband light; (2) efficient separation and fast transfer of photogenerated electron-hole pairs [4]. To address these requirements, many great efforts have been made by using diversified semiconductor photocatalysts or doping noble metals such as Pt as cocatalysts to promote charge separation. However, the terrestrial scarcity and high cost of noble metals limit the viability of H₂ production and commercial applications. So, these obstacles have been motivated to design and develop non-noble metal cocatalysts with photocatalysts to

promote the efficient charge separation from the semiconductor to the surface on the cocatalyst, from which H₂ is obtained by reducing H₂O [5,6].

Currently, a wealth of materials responding to the visible-light spectrum which occupies around 46% of the solar light are designed as photocatalysts to achieve effective solar hydrogen production from water, such as sulfide [7], nitride [8], vanadates [9], phosphides [10], and metal oxides [11]. However, their commercial applications are hindered due to the high cost, risks for the environment or inadequate scalability [12]. Polymeric graphitic carbon nitride (g-C₃N₄) has recently shown promise for driving photocatalytic H₂ production owing to its chemical stability and amazing electronic structure [13–16]. The bulk g-C₃N₄ is usually synthesized by thermal polymerization of melamine, urea, or dicyanamide. It possesses a large bandgap at about 2.7 eV, and its highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) situated at +1.6 V and -1.1 V versus normal hydrogen electrode, respectively. Although it is cheap and non-toxic, the pristine g-C₃N₄ is still limited by the poor visible light absorption (λ < 460 nm), fast recombination of photoinduced electron-hole pairs, low surface area, and limited active sites [15,17]. In order to challenge these limitations, various strategies have been made to improve the photocatalytic properties of g-C₃N₄, such as synthesizing specific nanostructures of g-C₃N₄ [18,19], doping with metal (e.g. Pt) or non-metal, (e.g. C [20], S [21]), building heterostructure between g-

* Corresponding authors at: College of Environmental and Chemical Engineering, Shanghai University of Electric Power, 200090, PR China.

E-mail addresses: shipenghui@shiep.edu.cn (P. Shi), xuqunjie@shiep.edu.cn (Q. Xu), ahaqmylin@126.com (Y. Min).

Scheme 1. The process of synthesizing Mo₂N/CN.Fig. 1. XRD patterns of CN, Mo₂N, and Mo₂N/CN-x (a), and FTIR patterns of CN and Mo₂N/CN-x (b).Fig. 2. SEM image of Mo₂N/CN (a). AFM (b, c, d) images of Mo₂N/CN and the corresponding height profile of the black line part in (b).

C₃N₄ and other semiconductors. For instance, Xiong's group [22] builds metal-to-ligand charge transfer between Pt²⁺ and g-C₃N₄, achieving broad light-absorption at the visible-near-infrared area and high H₂ production rate.

However, high cost and scarcity of Pt have limited practical

applications. It has been proved that loading cocatalysts can not only speed up electron transport but also act as electron sinks to provide further charge separation across the interface [23]. Many non-noble metal cocatalysts have received much attention such as two-dimensional (2D) molybdenum disulfide (MoS₂) owing to its high mobility for

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