Coupling photothermal evaporation into photocatalysis for enhanced hydrogen production from water

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Converting solar energy into hydrogen energy by photocatalysis has been considered as a promising solution to tackle energy crisis and promote sustainable development. However, the solar-to-hydrogen (STH) efficiency is not yet satisfactory due to the wide band gap of conventional photocatalysts

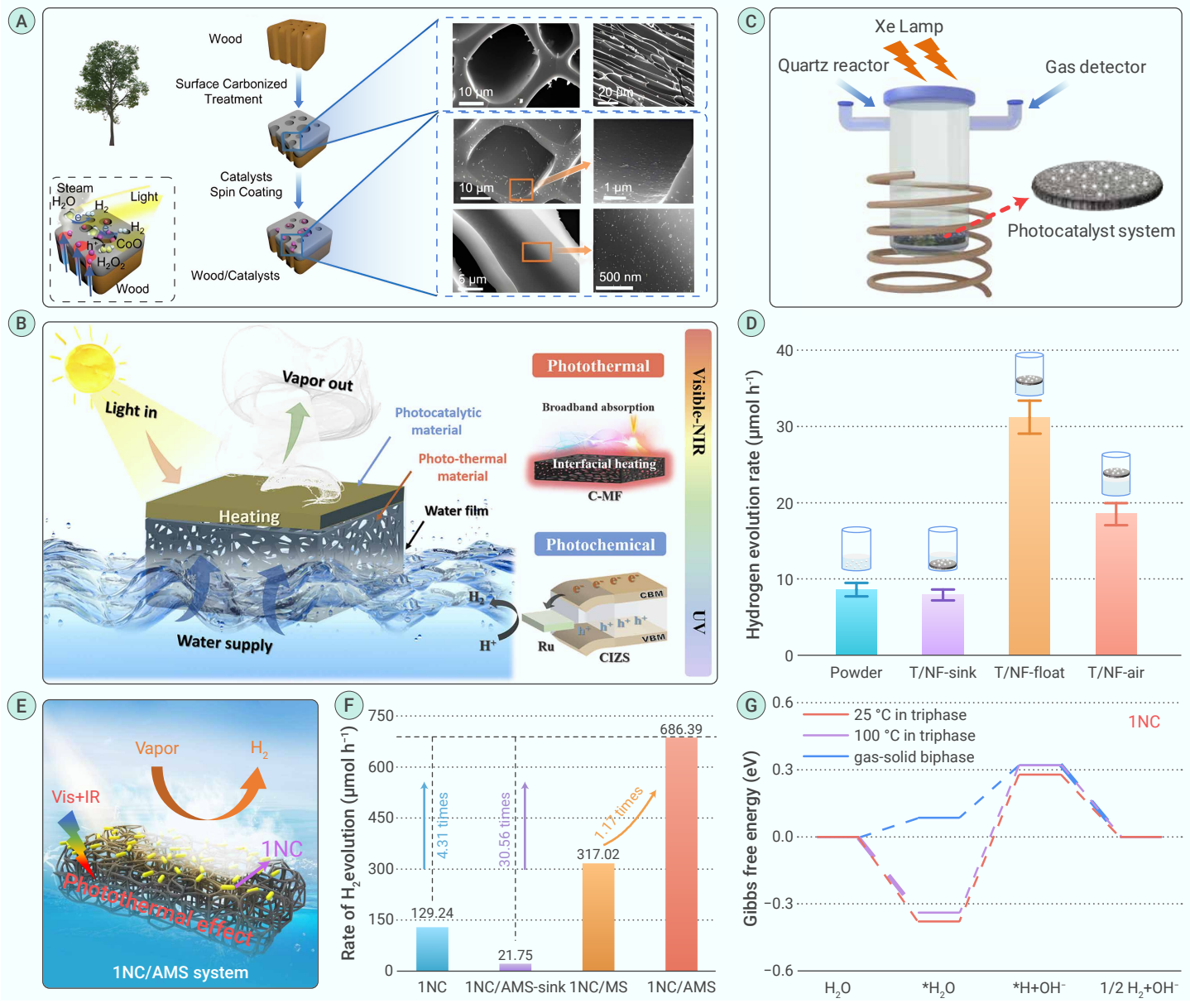

Figure 1. Innovative photothermal catalytic systems coupling photothermal evaporation into photocatalysis (A) Schematic of the fabrication process of the wood/photocatalyst structure that generates the water steam and catalyzes its splitting for hydrogen evolution.[1] Copyright (2021) Springer Nature. (B) Schematic diagram of $Cu_{0.04}In_{0.25}ZnS_y$ (CIZS@Ru) loaded on carbonized melamine foam (C-MF) for photothermal–photocatalytic coupling solar hydrogen production.[2] Copyright (2023) American Chemical Society. (C) Schematic diagram of photocatalytic hydrogen production system under alternating magnetic field conditions. (D) H_2 evolution rates of different photocatalyst systems (T/NF-sink: the catalyst sunk in water; T/NF-float: the catalyst floated at the liquid-gas interface; T/NF-air: the catalyst placed in air).[4] Copyright (2022) Wiley-Blackwell. (E) schematic diagram of a gas–solid photocatalytic reaction system (1NC/AMS: 1% Ni-CdS loaded on annealing melamine sponge). (F) H_2 evolution rates of different photocatalyst systems (1NC/AMS-sink: 1% Ni-CdS loaded on annealing melamine sponge and sunk in water; 1NC/MS: 1% Ni-CdS loaded on melamine sponge floated at the liquid-gas interface; 1NC/AMS: 1% Ni-CdS loaded on annealing melamine sponge floated at the liquid-gas interface). (G) Gibbs free energy of Ni-CdS under different photocatalyst systems.[5] Copyright (2024) Royal Society of Chemistry.

and the limited presence of UV photons in solar radiation. As a result, there has been increasing interest in the integration of photocatalysis and thermal catalysis, in a form named photothermal catalysis. This approach aims to reduce the energy consumption of thermal catalysis and improve the efficiency of photocatalysis.[1] In the traditional solid-liquid-gas triphase photothermal catalytic system for hydrogen production from water, the response of solar spectrum has been successfully expanded to achieve higher activity. However, challenges still persist, such as heat loss and sluggish mass transfer in the traditional triphase reaction, hindering its practical application. To overcome the inherent limitations, the combination of solar water evaporation and photocatalytic water splitting provides a novel approach to design innovative photothermal catalytic systems.

Recently, Guo et al. constructed an efficient photothermal catalytic system using a charred wood substrate to convert liquid water to vapor and produce hydrogen under light irradiation without requiring additional energy.[2] This biphase system enhances the kinetic transport of hydrogen, facilitating its escape from the system, while thermodynamically reduces the interfacial barrier for water molecule adsorption. In their study, this wood/CoO biphasic system exhibits an impressive photocatalytic hydrogen production rate of up to 220.74 μmol h^{-1} cm^{-2} (Figure 1A). This work highlights the effectiveness of gas-solid biphase system in promoting mass and energy transfer during the photocatalytic reaction. In addition, Chang group developed a remarkable system for photocatalytic water splitting by using $Cu_{0.04}In_{0.25}ZnS_y$@Ru (CIZS@Ru) loaded on carbonized melamine foam (C-MF) at the water-air interface, with the assistance of photothermal conversion.[3] This gas-solid system presented an impressive hydrogen evolution rate of 603 mmol h^{-1} m^{-2} under one-sun intensity illumination, which is more than twice that achieved in traditional liquid water reactions. The breakthrough aspect of this innovative catalytic system lies in the ability to synergistically combine photothermally induced water evaporation with subsequent photocatalytic water vapor splitting. This unique approach expands the utilization range of the solar spectrum and greatly improves the efficiency of photocatalytic water splitting (Figure 1B).

In a traditional photocatalytic reaction for hydrogen evolution from water, photocatalyst is dispersed in liquid water through stirring, which yet has two obvious disadvantages. Firstly, it leads insufficient utilization of light energy. Secondly, the recycling process becomes cumbersome. In response to these challenges, our group has developed an external field-assisted photothermal catalytic biphasic reaction system, which significantly improves the performance of photocatalytic water splitting through the magneto-photothermal coupling effect.[4] We designed a monolithic catalyst in the form of nickel foam supported TiO_2 (designated as T/NF) to study the impact of magneto-photothermal coupling field (Figure 1C). It is also very interesting that the photocatalytic performance could be significantly changed when the monolithic catalyst was placed at different places relative to the liquid-gas interface. Figure 1D indicates that photocatalyst possessed an excellent performance, with a hydrogen production rate of 31.2 μmol h^{-1}, when the catalyst floated (designated as T/NF-float) at the liquid-gas interface. In the presence of alternating magnetic fields, the reaction temperature of the T/NF surface can be increased due to the eddy current effect. This, in turn, leads to a more efficient charge separation. More importantly, the generated heat facilitates the coupling the gaseous hydrogen production with water evaporation, thus avoiding nucleation, growth, and desorption processes involved in the formation of kinetic hydrogen bubbles. This external field-assisted photothermal catalytic system has demonstrated a remarkable increase in the hydrogen evolution rate, reaching 104.3 μmol h^{-1}. This activity is 3.3 times that of T/NF-float system and 12.2 times that of T/NF-sink system (T/NF-sink indicates the catalyst is sunk in water).

The demand for additional energy input in the multiple-field coupled reaction systems has enabled researchers to seek for alternative approaches. Recent progress has been made by our group in the research of photothermally driven decoupling of gas evolution at the solid-liquid interface.[5] We have constructed a gas-solid biphase photocatalytic system by loading Ni-CdS onto annealing melamine sponge (1NC/AMS). The catalyst consists of CdS nanorods anchored by 1% highly dispersed nickel atoms/clusters, serving as the photocatalyst, and AMS as the vapor generator. The system, shown in Figure 1E, demonstrates that the hydrogen evolution rate of the 1NC/AMS system is up to 686.39 μmol h^{-1}, 5.31 times that of the traditional solid-liquid-gas triphase system (Figure 1F). This improvement is primarily attributed to a more favorable environment for the transfer of hydrogen at the active sites rather than the increase in surface temperature of 1NC/AMS resulting from photothermal conversion. In addition, the state of water also plays a crucial role in enhancing photocatalytic hydrogen evolution. To gain further insights into the potential mechanism behind this gas-solid photocatalytic system, we conducted DFT calculations of the Gibbs free energy of the H_2 evolution reaction (HER). The results show that Gibbs free energy barrier of the rate-limiting step is significantly reduced when HER occurs in the gas-solid biphase system compared to the solid-liquid-gas triphase system at 100 °C (Figure 1G). This indicates that the decomposition process of gaseous water in the biphase system is smoother than that of liquid water in the triphase system, leading to an accelerated photocatalytic HER. Moreover, due to the competition between various metal cations and H^+ in natural water sources, traditional solid-liquid-gas triphase photocatalytic systems exhibit unsatisfactory photocatalytic hydrogen evolution activity in natural water sources. This gas-solid biphase photocatalytic system can physically separate the metal cations from the photocatalyst by evaporation, thereby reducing the likehood of H^+ competitive reactions. Overall, this photothermally induced gas-solid biphase system, with its high solar spectral utilization, thermodynamically reduces the Gibbs free energy barrier of HER, dynamically reduces the hydrogen transport resistance at the interface, and intrinsically changes the slow mass transfer and conversion process at the triphase photocatalytic interface.

In summary, great advancements have been achieved to develop novel photothermal catalytic biphase systems to realize full-spectrum solar driven water splitting. Such photothermal catalytic systems based on an efficient and practical photothermal material substrate enable optimized energy flow, facilitating effective propagation, conversion, and utilization of the energy. Moreover, this novel catalytic system thermodynamically lowers the photocatalytic reaction barrier and kinetically reduces the hydrogen transport resistance at the interface. While the efficiency of this novel photothermal catalytic biphase systems has experienced significant growth, several challenges remain. The mechanism responsible for the enhanced mass transfer in this novel photothermal catalytic biphase system should be further investigated through theoretical calculations and in-situ characterization. We believe this photothermal-photocatalytic coupling system will achieve remarkable cost-effectiveness and hold promising potential for practical application with the concerted efforts of researchers.

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DECLARATION OF INTERESTS

The authors declare no competing interests.