

Chemically exfoliated MoS2 on TiO2 mesocrystal with efficient photocatalytic hydrogen evolution

Over the past decades, the rapid depletion of fossil fuels and serious environmental pollution were started to strongly affect the modern society. With concerning the deficiency of human energy and alleviating the environmental pollution, solar energy as the sustainable source is becoming the clean, cheap, renewable energy to fulfill rising global demand in our life. Since 1970s, solar driven hydrogen evolution (HER) has been studied extensively with a growing amount of attention. Thus it is strongly desired to design the low cost photocatalysts with efficient HER.

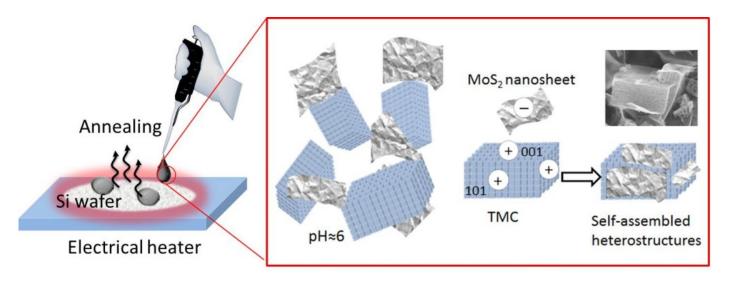


Fig. 1. Schematic illustration of synthesis of MoS₂/TMC.

We reported a facile synthetic approach involving the assembly of MoS_2 nanosheets as co-catalysts onto external surfaces of TiO_2 mesocrystals (TMCs) after mild impregnation and annealing (Fig. 1). The prepared TMCs are impregnated with the chemically exfoliated MoS_2 nanosheets in solution with continuous sonication. In the precursor solution, negatively charged MoS_2 can be adsorbed on positively charged TiO_2 due to the electrostatic interaction. Finally, the MoS_2 /TMC heterostructures were obtained after the suspension dropped on Si wafer was annealed at 160 °C for 10 min.

The photocatalytic HER was evaluated under UV light irradiation, where lactic acid was used as both a hole scavenger and an abundant source of H^+ ions. A negligible amount of H_2 gas was detected on MoS_2 or TMC alone. Interestingly, the MoS_2 /TMC exhibited higher activity (0.55 mmol h^{-1} g^{-1}) than in a nanoparticle-based system using P25 (Fig. 2a). The charge transfer dynamics was further revealed by time-resolved diffuse reflectance (TDR) spectroscopy for the first time (Fig. 2b). In the period of 0–10 ps, it was obvious that the transient absorption observed for MoS_2 modified

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TMC decayed rapidly, when compared to pure TMC. After 10 ps, the transient absorption diminished gradually. It is noteworthy that MoS₂/P25 exhibits similar decay times to those of pure P25.

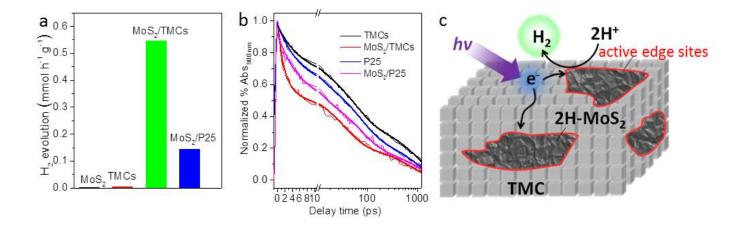


Fig. 2. (a) Comparison of the hydrogen generation rates of different samples. (b) Normalized transient absorption traces observed at 900 nm. (c) Reaction mechanism of MoS₂/TMCs in HER.

In the case of MoS₂/TMC, the TMC with good electrical conductivity promotes the efficient interfacial electron transfer from the excited TiO₂ to the active sites on MoS₂ nanosheets, resulting in the retardation of the charge recombination and efficient electron harvesting by MoS₂. This is likely to be the main reason why MoS₂/TMC exhibits a higher photocatalytic activity. The 3D architectures of MoS₂/TMC with promising superiority are expected to become a potential competitor to earth-abundant catalysts for the HER. It is believed that the superstructure-based MoS₂ can act as a potential noble-metal-free photocatalyst with superior hydrogen generation efficiency, compared to conventional nanoparticle-based systems.

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