

## Photocatalytic hydrogen evolution by Smectite-based clay composite catalyst

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Hydrogen has emerged as a promising sustainable and environmentally benign energy carrier for addressing the rapidly increasing global energy demand. Despite considerable efforts toward the development of high-performance photocatalysts for the photocatalytic hydrogen evolution reaction (HER), several fundamental limitations, including the rapid recombination of photogenerated electron-hole pairs, inefficient charge separation, and restricted charge transport, continue to hinder the efficiency and practical applicability of photocatalytic systems. Transition-metal dichalcogenides (TMDs), generally represented by the formula  $MX_2$ , where M denotes a transition metal and X represents a chalcogen, have attracted considerable attention in photocatalytic HER. In particular, amorphous  $MoS_x$ -based catalysts are regarded as promising alternatives to Pt-based noble-metal catalysts because of their relatively low cost, favorable electronic properties, and abundant catalytically active sites for proton reduction. Nevertheless, their photocatalytic performance may be limited by strong interparticle interactions that promote aggregation and consequently decrease the accessible active surface area. To overcome these limitations, support materials are commonly employed. Natural clay minerals are increasingly considered promising support materials for various photocatalytic systems owing to their large surface areas, high adsorption capacities, ion-exchange properties, and layered structures, which may facilitate charge-carrier separation and transport. In this study,  $MoS_x$  catalysts were formed on the surface of smectite, a clay mineral with a three-layered structure, through the photodeposition of a molybdenum precursor in the presence of eosin Y (EY). The catalytic activity of the resulting smectite- $MoS_x$  system was evaluated for photocatalytic hydrogen evolution under visible-light irradiation. Following the photocatalytic hydrogen evolution reaction, the formation and chemical state of  $MoS_x$  on the smectite surface were confirmed by X-ray photoelectron spectroscopy (XPS). Furthermore, the obtained results demonstrated that the presence of  $MoS_x$  markedly enhanced the HER activity compared with bare smectite.

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