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ORIGINAL PAPER

Homogeneous Photocatalytic Hydrogen Evolution System with Assembly of CdSe Quantum Dots and Graphene Oxide

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Abstract
Hydrogen is a promising energy carrier to replace traditional fossil fuel and could be obtained by the artificial photosynthesis. Herein, a homogenous system combined CdSe quantum dots (QDs) and graphene oxide (GO) (GO/CdSe QDs) were prepared by simple ultrasonication and stirring method for highly efficient photocatalytic hydrogen evolution. Notably, the optimized photocatalytic H_2 evolution rate reached as high as 33.88 mmol g^{-1} h^{-1} for GO/CdSe QDs, which is about 7.9 times than that of pure CdSe QDs. The X-ray diffraction (XRD), Fourier-transform infrared spectroscopy (FT-IR) and transmission electron microscopy (TEM) confirmed that the GO has been successfully coupled with CdSe QDs and has ignorable influence on the original crystal structure and morphology of CdSe QDs. Importantly, we found that the hydrophilicity of GO is necessary to ensure its intact interaction with CdSe QDs for the continuous H_2 production. The significant activity enhancement mainly attributed to the acceleration of charge migration and dynamics of proton reduction after introducing GO, which has been proved by electrochemical measurements, photoluminescence spectroscopy (PL) and dynamic decay of samples.

Keywords CdSe quantum dots · Graphene oxide · Hydrogen · Homogeneous photocatalysis

1 Introduction

Developing clean and renewable energy resources to replace finite and non-renewable fossil fuels is of great importance to address great challenges of worldwide energy and environment issues in the contemporary era. [1, 2] Hydrogen with high-energy–density and carbon neutrality has been reckoned as a critical energy storage vector to completely exploit the benefit of sustainable and renewable energy. [3–5] Production of hydrogen via solar-powered water splitting, the so-called artificial photosynthesis, is indisputably one of the most important approaches due to use of the environmentally friendly solar power source. [6–8]

Till now, numerous photocatalytic systems have been developed, which are mainly categorized into heterogeneous and homogeneous catalytic systems. [9–11] In case of a heterogeneous catalytic system, the reactant and catalytic centers exist in different phases, and the commonly involved photocatalysts are bulk semiconductors such as transition metal oxides/sulfide (TiO_2, ZnO, CdS, and so on). [12–15] While homogeneous system converges the active centers and reactants in the same phase, offering easy accessibility of active centers to the reactant, which results in high photocatalytic activity. [16–18] Present study of homogeneous photocatalytic systems mainly focused on molecular catalysts comprising of noble metal centers and specific organic ligand. In 1970s, Lehn and Sauvage firstly observed that light irradiation can drive H_2 production in a homogeneous system with $[Ru(bpy)_3]^{2+}$ (bpy = 2,2′-bipyridine). [19–21] After that, Bernhard group reported the Rh-based complexes

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