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A low-cost, high-efficiency and durable homogeneous system for molecular hydrogen

Qifeng Liu,^a Bingrui Liu,^a Qian Zhang,^a Jianping Gao^a and Jingjun Ma^{a,b*}

ABSTRACT High-efficiency hydrogen evolution is obtained from a novel fluorescein homogeneous system, comprising a noble metal free fluorescein as the photosensitizer, Pt nanoparticle as the cocatalyst and TEOA as the holes scavenger in an aqueous solution under visible light irradiation. The highest H₂ production rate is 491.40 μ mol/h ($\lambda > 420$ nm) and quantum efficiencies of our system are among the highest reported for the molecular hydrogen systems in the visible light range. Unlike many reported molecular hydrogen systems are either unstable under illumination and expensive, or have weak photocatalytic performances, our low-cost system exhibits excellent activity and endurance. We believe that this low-cost, high-efficiency and durable homogeneous system for molecular hydrogen may result in new breakthroughs for the hydrogen energy and solar energy utilization.

Keywords: Solar energy materials; organic; hydrogen evolution; homogeneous system; high-efficiency and durable.

1. Introduction

The conversion of solar energy into conveniently usable energy in the form of hydrogen gas by homogeneous system is one of the most promising approaches toward production of renewable and clean energy.[1] These systems universally utilize a photosensitizer (PS) for solar light harvesting, a sacrificial agent to serve as holes scavenger and a cocatalyst for active catalytic sites.[2] The great challenges that exist in respect of molecular hydrogen system are applying earth-abundant materials and improving of the efficiency and durability. Historically, noble metal-based photosensitizers, such as Ru^I -,[3] Ir^{III} -,[4] Pt^{II} -[5] and Re^{I} -[6] based photosensitizers have been the most common PS choices. However, the character of noble metallic composition, inefficiency and non-resistant may limits practical applications in homogeneous system for molecular hydrogen. A more promising homogeneous system for molecular hydrogen is reported that do not contain a noble metal of photosensitizer (PS).[7] The system utilizes the

brominated xanthene-based dye Eosin Y as the photosensitizer. The system, however, is subject

to photodecomposition of PS and inefficiency for H₂ production, furthermore it is inactivate only after 30 h, as is another system using the halogen-containing xanthene-based dyes including Rose Bengal, Eosin B, Erythrosin B and Phloxine B as the PS.[8] The C-Br, C-Cl and C-I bonds of these xanthene-based dyes are also viewed as the source of their photodecomposition through reductive quenching and C-X cleavage from the reduced PS.[9] Compared with halogen-substituted xanthenebased dyes, the unsubstituted fluorescein is far more stable under light irradiation.

Here, we report a low-cost, high-efficiency and durable homogeneous system for molecular hydrogen, which uses a noble metal free fluorescein as the photosensitizer, Pt nanoparticle as the cocatalyst and triethanolamine (TEOA) as the holes scavenger. The highest H_2 production rate and TON

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