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Three-Dimensional Carbonaceous Aerogels Embedded with Rh-$SrTiO_3$ for Enhanced Hydrogen Evolution Triggered by Efficient Charge Transfer and Light Absorption

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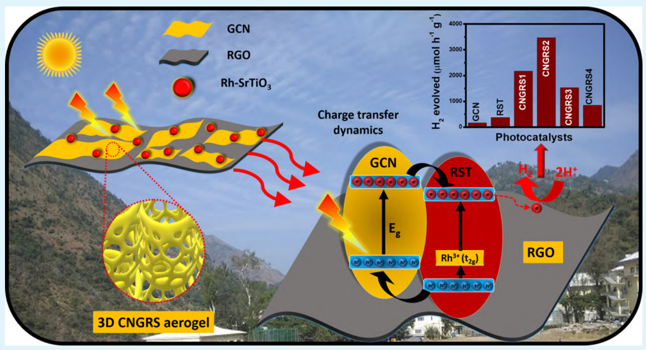

ABSTRACT: Photocatalytic and photoelectrocatalytic hydrogen generation from water splitting by utilizing the visible spectrum of sunlight has been recognized as one of the promising energy conversion applications. Herein, we report the three-dimensional (3D) superstructure of g-C_3N_4 and reduced graphene oxide embedded with Rh-doped $SrTiO_3$ nanoparticles as ternary aerogels for efficient hydrogen production. The optimized aerogel exhibits high competency for visible light harvesting due to the unique 3D morphology and shows excellent hydrogen evolution performance with quantum efficiencies of 51.1 and 26.9% at 450 and 600 nm monochromatic wavelengths, respectively. The 3D arrangement of integrated components helps in enhanced light absorption due to multiple reflections of incident light within the system and provides a high surface area with abundant reaction sites. Moreover, the ternary heterojunction facilitates efficient charge transfer owing to the suitable band positions of each component as evidenced by fluorescence lifetime, photocurrent, and impedance spectroscopic measurements, resulting in enhanced photocatalytic performance. In addition, the photoelectrocatalytic hydrogen evolution activity reveals the multifunctional nature of the synthesized catalysts. Thus, the hybrid design of the photocatalytic system realizes efficient hydrogen production in suspension and demonstrates the potential of aerogel-based materials as next-generation photocatalysts.

KEYWORDS: *g-C_3N_4, reduced graphene oxide, aerogel, photocatalysis, hydrogen evolution*

1. INTRODUCTION

The use of sunlight for photocatalytic hydrogen (H_2) production is considered as a sustainable, cleaner, and one of the alternative technologies to meet the present and future energy requirements. In addition, it can also be utilized as feedstock for different catalytic processes, such as for carbon dioxide reduction and nitrogen fixation to synthesize value-added liquid fuels, like methanol and ammonia, which are easy to store and handle.[1−3] In this regard, H_2 production through heterogeneous photocatalysis has gained considerable attention in comparison to the other fossil-fuel-based H_2 production methods like steam reforming, which liberates a huge amount of greenhouse gas, CO_2, as a byproduct.[4,5] Nevertheless, for efficient harvesting of sunlight, it is highly desirable to develop narrow-band-gap photocatalytic materials, which can produce H_2 from water under visible light as it constitutes a major portion of the solar spectrum.[6,7] In this regard, several photocatalysts, including binary, ternary, and multicomponent systems, have been synthesized to alleviate the basic problems (low absorption and fast charge recombination) of this process and to enhance the solar-to-H_2 conversion efficiency.[8−10] However, the assimilation of the merits of each component in such systems for achieving high light absorption and decreased recombination is a complex task, which can be carried out by opting smart synthesis routes for their meticulous design and development.

In this regard, 3D porous aerogel photocatalysts have attracted considerable attention from researchers.[11,12] Although several other photocatalysts with different morphological features including zero-dimensional (0D), one-dimensional (1D), and two-dimensional (2D) structures have been reported in the literature, 3D morphology has several advantages over them. The 3D structures not only provide a high surface area with enhanced accessibility but also aid in the efficient adsorption of reactants and hence in the subsequent

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