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In-situ hydrothermal synthesized γ-Al_2O_3/O-g-C_3N_4 heterojunctions with enhanced visible-light photocatalytic activity in water splitting for hydrogen

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ABSTRACT

In this work, γ-Al_2O_3 and hydrogen peroxide treated g-C_3N_4 (O-g-C_3N_4) were combined through a novel in-situ hydrothermal method to form heterojunction structured photocatalysts. These photocatalysts were characterized by X-ray diffraction (XRD), scanning electron microscopy (SEM), Fourier transform infrared spectroscopy (FT-IR), X-ray photoelectron spectroscopy (XPS), UV–vis diffuse reflectance spectroscopy and photoluminescence spectroscopy (PL). FT-IR results indicate that oxygen functional groups can be grafted on the surface of O-g-C_3N_4 by hydrogen peroxide treatment. The visible light photocatalytic hydrogen evolution rate was investigated in 10 vol% TEOA aqueous solution. The optimal Al_2O_3 mass content is set to be 20 wt% and the corresponding hydrogen evolution rate is 1288 μmol/h/g which is approximately 6, 3 folds that of pristine g-C_3N_4 and O-g-C_3N_4 respectively and 1.6 folds that of mechanical mixed composite with the same Al_2O_3 mass content. The photocurrent density–time curves were carried out under visible light illumination for four on–off cycles. The electrochemical impedance spectroscopy (EIS) measurements verified the enhanced separation efficiency of electron–hole pairs. This work raised a new method to form the heterojunction structured photocatalysts and achieved a remarkable improvement of the photocatalytic activity in water splitting for hydrogen under visible light irradiation.

1. Introduction

With the deterioration of the global environment, hydrogen is widely considered as a promising fuel to maintain the development of society. Since Fujishima and Honda first discovered the water splitting effect in 1972 [1], a huge amount of work has been implemented to find applicable semiconductor materials for photocatalytic water splitting [2–5]. In 2008, Wang et al. [6] reported the polymeric organic semiconductor g-C_3N_4 as a favorable photocatalyst in water splitting for hydrogen, thus lots of attention has been drawn because of its high photocatalytic ability with relative narrow band gap of 2.7 eV, low preparation cost and good thermal stability [7–12]. However, the photocatalytic activity of pristineg-$C_3N_4$is limited on account of its low charge mobility, high excitation dissociation energy, inappropriate position of valance band, poor visible photon absorption ability and low specific surface area etc. [13]. With the purpose of ameliorating the photocatalytic activity of pristine g-C_3N_4, numerous strategies have been developed, such as modified synthesis methods [14], morphology modification [4,15–17], metal [11,18,19] or non-metal ions [20,21] doping, construction of g-C_3N_4 based composites [3] and dye sensitization [22]. It has been testified that their value for the improvement of photocatalytic efficiency of pure g-C_3N_4.

Al_2O_3 is widely used as catalyst's support due to its excellent chemical stability, high specific surface area and good thermal stability [23,24]. In the field of photocatalysis, Al_2O_3 has not been widely used as active site because of its broad band gap. Recently, Li et al. [25,26] reported that Al_2O_3/g-C_3N_4 composites can remarkably improve the photocatalytic activity in degradation of RhB of pure g-C_3N_4, the multiple defect sites of amorphous Al_2O_3 can act as the electron acceptors thus reduce the recombination efficiency between electrons and holes, and then enhance the photocatalytic activity.

In present work, we successfully combined Al_2O_3 and surface modified g-C_3N_4 (O-g-C_3N_4) through a novel in-situ hydrothermal method when introducing oxygen functional groups on the surface of g-C_3N_4. It has been demonstrated that this method

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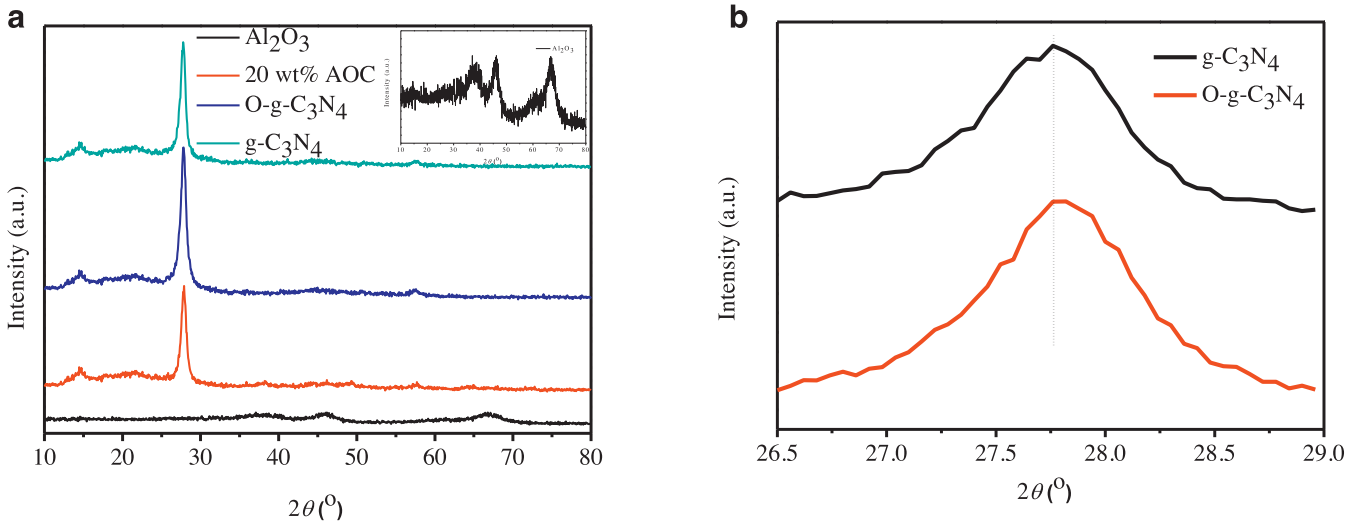

Fig. 1. XRD patterns of Al_2O_3, g-C_3N_4, O-g-C_3N_4 and 20 wt% AOC.

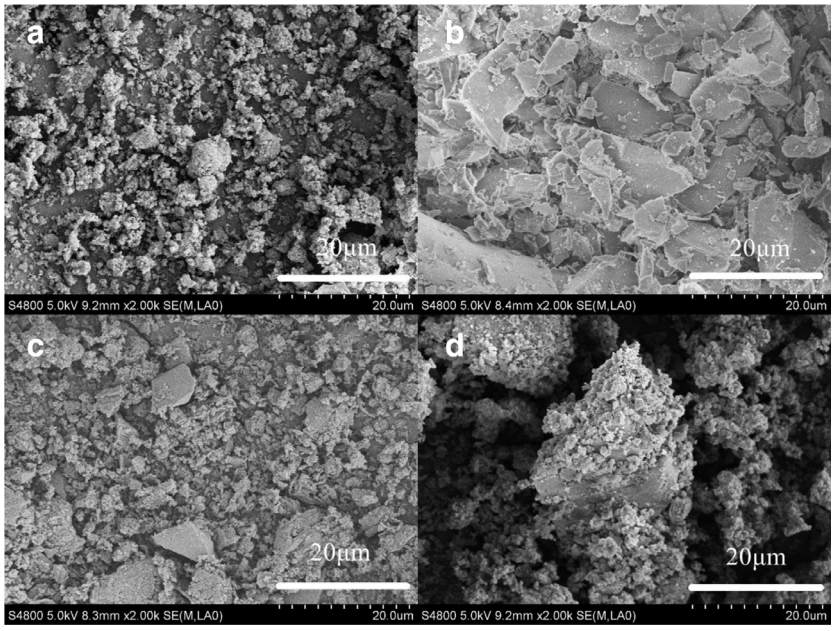

Fig. 2. SEM images of (a) O-g-C_3N_4, (b) Al_2O_3, (c) 20 wt% AC and (d) 20 wt% AOC.

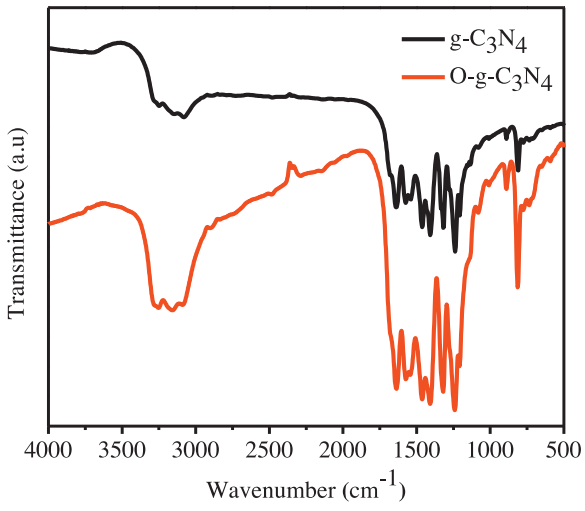

Fig. 3. FT-IR spectroscopy of g-C_3N_4 and O-g-C_3N_4.

successfully formed the heterojunction structured photocatalytic composites and these composites can be used in the field of visible light photocatalytic water splitting for hydrogen. Compared with the pristine g-C_3N_4, O-g-C_3N_4 and mechanical mixed composites, the composites showed remarkable enhanced photocatalytic activity in water splitting for hydrogen under visible light illumination. The influence of Al_2O_3 mass content on the hydrogen evolution efficiency is also studied and then discussed in detail.

2. Experimental

2.1. Materials

Melamine, triethanolamine, urea, aluminum nitrate nonahydrate and 30 wt% hydrogen peroxide were purchased from Chengdu Kelong Reagent Factory. All reagents were used as received without further treatment.