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The enhanced photocatalytic performance of Z-scheme two-dimensional/two-dimensional heterojunctions from graphitic carbon nitride nanosheets and titania nanosheets

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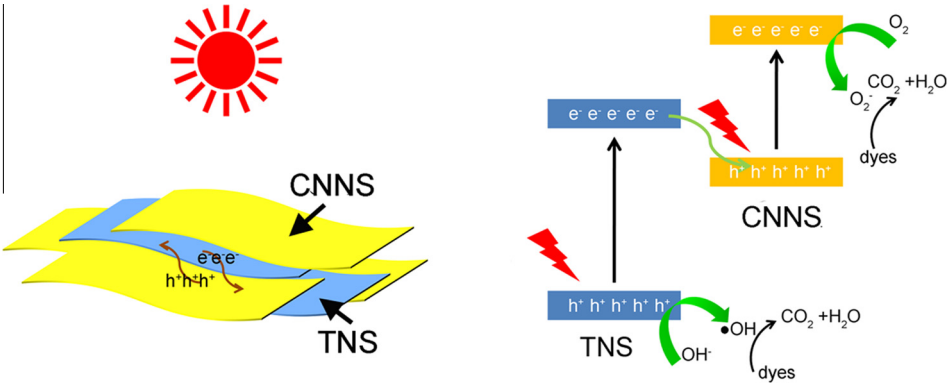
GRAPHICAL ABSTRACT



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ABSTRACT

A direct solid state Z-scheme photocatalytic system was fabricated by assembling two-dimensional (2D) g-C_3N_4 nanosheets (CNNS) and titania nanosheets (TNS), which were obtained from the delamination of their corresponding layered precursors. By introducing TNS, the interlayer restacking of CNNS was effectively prohibited, forming uniform CNNS/TNS composites. The tightly contacted CNNS/TNS interface promoted the charge transfer and therefore improved the separation ratio of photogenerated electron-hole pairs. The photocatalytic performance of CNNS/TNS in various mass ratios was investigated for dye degradation, and the degradation rate of optimal sample 0.7CNNS/0.3TNS was 2.34 and 48.5 times higher than those of proton flocculated pure CNNS and TNS, respectively. Superoxide radicals and hydroxyl radicals were determined as the main active species by the quenching experiment. Moreover, the enhanced generation of superoxide radicals and hydroxyl radicals was confirmed by the absorption spectra of nitroblue tetrazolium and the photoluminescence spectra of 2-hydroxy terephthalic acid, respectively. Finally, we proposed a possible Z-scheme mechanism based on the theoretical calculation and the experimental results.

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1. Introduction

Encouraged by the pioneer work on graphene at 2004 [1], tremendous research interest recently has been focused on two-dimensional (2D) nanostructures, which exhibit promising applications in optics, electronics, magnetism, catalysis and so on because of their novel physicochemical properties [2–5]. Especially, the unique characteristics of 2D nanostructures, high aspect-ratio, large surface area, ultrathin thickness, have portrayed a magnificent prospect for photocatalysis [6–8]. All the constituted atoms of 2D materials are on the surface or near the surface, which provides a short distance for the photogenerated carriers to diffuse to the surface, therefore prohibiting the recombination of electron and hole. Furthermore, the plentiful surface groups are favorable for anchoring co-catalysts and three dimensional (3D) porous pillared structures can be fabricated using 2D materials as building blocks [9–11].

Graphitic carbon nitride, g-C_3N_4 (CN), has attracted numerous attentions as a novel visible light photocatalyst due to its metal-free nature, high thermal and chemical stability, narrow band gap (−2.7 eV) and facile preparation using low-cost precursors [12,13]. In spite of the small specific surface area and low quantum yield of bulky CN, a great variety of approaches have been employed to improve the photocatalytic activity, including decreasing the particle size [14–17], doping with metal or non-metal element [18–20], fabricating heterojunctions using other semiconductors with matched band structure [21–25].

As the graphite analogue, the exfoliation of layered CN into 2D nanosheets is of significant importance from the theoretical or practical perspective [17,26]. G. Liu et al. firstly achieved extremely thin CN nanosheets (CNNS) by a solid state exfoliation via thermal etching of pristine CN in air [15]. Furthermore, the ultrasound-assisted liquid exfoliation of CN was subsequently realized in water and other solvents [16,27]. The well dispersed colloidal nanosheets provide the opportunity to fabricate various nanostructures using it as building block. Nevertheless, the exfoliation yield and the monolayer ratio in both thermal oxidation exfoliation and ultrasonic exfoliation are unsatisfactory. By an intercalation reaction of H_2SO_4 into the interlayer of CN, Y. Zhu et al. obtained delaminated CNNS with a >60% monolayer ratio [28]. What is more, an almost ~100% exfoliation yield was gained and the surface charge was able to be controlled by adjusting the pH value [29]. Very recently, we developed an alkali-treating method to exfoliate CN [30]. Besides the high exfoliation efficiency and yield, the generated CNNS retained much more visible light absorption than the nanosheets from acidic exfoliation.

It is well known that a Z-scheme photocatalytic system is in favor of the separation of photoinduced electrons and holes on different semiconductors, and meanwhile maintains the redox ability of photoexcited carriers. The traditional Z-scheme systems involve two individual semiconductors and the electron mediator that is usually noble metals or the redox pairs of Fe^{3+}/Fe^{2+} and IO_3^-/I^- [31,32]. However, the difficulty of these conventional Z-scheme systems in practical application inspirits the development of direct solid-state Z-scheme systems [33–37]. In view of the relative high conduction band (CB) position of CN (−1.2 V vs. NHE), the photogenerated electrons on CB have a strong reduction ability. If fabricating conventional CN based heterojunctions (Fig. 1a), the reduction ability of electrons would be weakened after the charge transfer, despite the charge separation was improved. Thereby it is desirable to establish CN based Z-scheme photocatalytic system (Fig. 1b) that does not only suppress the recombination of photogenerated electron-hole pairs, but also preserves the strong redox ability of charges in the meantime.

Herein we reported an efficient 2D-2D coupled Z-scheme photocatalyst by electrostatic assembling of delaminated CNNS and titania nanosheets (TNS). The intimate interface and the enhanced contact area in 2D-2D combination facilitated the charge transfer. Following the Z-scheme mechanism, the photogenerated electrons were accumulated on CNNS and photoexcited holes were collected on TNS, therefore promoting the separation of carriers as well as retaining the powerful reduction ability of electron and the strong oxidation property of holes. The morphology, structure and optical property of the obtained 2D-2D composited CNNS/TNS were systematically studied, and its photocatalytic activity was investigated by photodecomposing organic pollutants. In addition, the possible photocatalytic mechanism was proposed based on various experimental results.

2. Experimental section

2.1. Synthesis

2.1.1. Delamination of CN

The pristine CN was synthesized by thermal polycondensation of melamine, which was described in published literatures [38], and the resultant yellow product was collected and milled into a powder for further use. The procedure of CN exfoliation was according to the alkali-treating method in our previous report [30]. Typically, 1 g CN powders were dispersed into NaOH aqueous solution (50 mL, 0.2 mol L^{-1}), which was further heated at 373 K for 12 h. After pouring out the clear supernatant, the sediment was re-dispersed into 50 mL deionized water. The mixture was centrifuged at 10,000 rpm for 5 min to remove precipitation, and then we obtained colloidal CN nanosheets, denoted as CNNS. Additionally, the centrifuged partial precipitation could be dispersed into water and formed CNNS colloid suspension by the next water washing. Finally, almost all sediment could be dispersed.

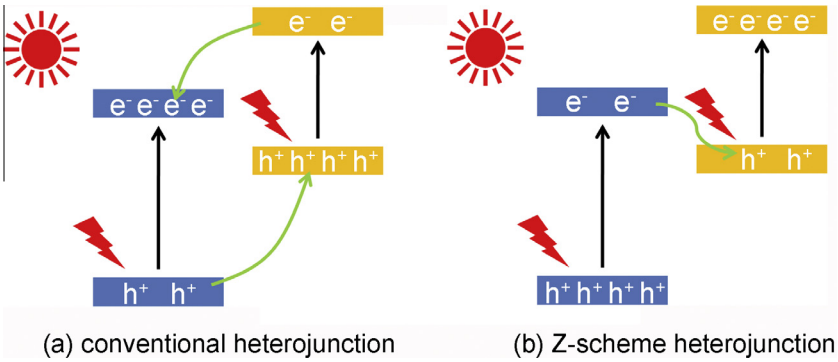

Fig. 1. Schematic illustration of the charge transfer in (a) the conventional heterojunction photocatalyst and (b) the Z-scheme photocatalyst.

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