Materials Research Bulletin 108 (2018) 176–186

Contents lists available at ScienceDirect

Materials Research Bulletin

journal homepage: www.elsevier.com/locate/matresbu

Target preparation of multicomponent composites Au@CdS/g-C_3N_4 as efficient visible light photocatalysts with the assistance of biomolecules

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ARTICLE INFO

Keywords:
Photodegradation
core-shell structure
Z-scheme structure
g-C_3N_4
L-cysteine

ABSTRACT

In this work, we aimed to develop a novel method for the target preparation of multicomponent composites Au@CdS/g-C_3N_4 towards high visible light photocatalytic activity with the assistance of L-cysteine biomolecules. Through the interaction of L-cysteine biomolecules with Au nanoparticles and g-C_3N_4 bulks, CdS layer were in-situ precipitated on the surface of Au nanoparticles to form core-shell structure Au@CdS and Z-scheme structure Au@CdS-g-C_3N_4 with good interface bonding in composites. By taking advantage of this feature, the highest reaction rate constant of 8%Au@CdS/g-C_3N_4 for methyl orange dye and phenol degradation was 3.08- and 1.48-folds higher than pure g-C_3N_4 bulks, which was ascribed to the strong interaction between different substances for efficient charges transferring and separation as evidenced by the studies of X-ray photoelectron spectroscopy, steady-state and transient photoluminescence spectroscopy. This work highlights the utilization of biomolecules for the target preparation of composites and its application on environmental remediation.

1. Introduction

Photocatalysis technology efficiently utilizing solar energy has been considered as one of the most compelling strategies to solve the energy crisis and environmental pollution simultaneously around the globe. By rationally designing the structures and compositions of photocatalysts, the catalytic performance can be greatly enhanced [1]. Among various methods, construction of heterojunctions by coupling two or more kinds of semiconductors with suitable CB and VB position is a commonly used pathway, which can effectively expand the optical absorption range of materials, improve the charge separation efficiency and accelerate the surface catalytic reaction process [2,3]. Recently, the heterojunctions formed by sulfides and g-C_3N_4 have attracted extensive attention of researchers owing to their energetically matching and narrow band gap which can harvest the solar light with high efficiency. Up to now, different heterojunctions such as CdS/g-C_3N_4 [4–6], MoS_2/g-C_3N_4 [7–13], SnS_2/g-C_3N_4 [14,15], NiS/g-C_3N_4 [16], $CdIn_2S_4$/g-C_3N_4 [17] and $MgInS_2$/g-C_3N_4 [18] and so on, have been extensively investigated. Compared with the single ingredient photocatalyst, the construction of heterojunctions can effectively accelerate the separation of e^-/h^+ pairs and further retard their recombination, resulting in the enhanced photocatalytic activity [19,20]. In addition, g-C_3N_4 is a kind of semiconductor with conjugated structure. The surface hybridization between g-C_3N_4 and sulfides makes it more conducive to the formation of bond binding, which can increase the photocatalytic stability of sulfides [21].

In different types of heterojunctions, the Z-scheme structures consisting of solid electron mediators and photoreaction systems are especially attractive because of its special photoinduced charge behavior in contrast to the conventional heterojunctions, which can effectively overcome the shortcomings of conventional heterojunctions and obtain the higher photoinduced charge separation efficiency and stronger redox ability [22,23]. Unlike the oxide photocatalysts TiO_2 (3.2 eV) [24–26] and WO_3 (2.7 eV) [27,28], CdS is one of the most widely used sulfides for the design of Z-scheme structures because of its narrow band (2.4 eV), which has strong visible light response ability similar as Ag_3PO_4 [29–31] and AgBr [32,33]. It can enable the Z-scheme structures capture more photons. For instance, Li et al. fabricated the Z-scheme structure g-C_3N_4/Au/CdS by chemical deposition method using thioacetamide as sulfur source and Au nanoparticles as electron mediator. The photocatalytic experiments for dyes degradation, including RhB, MO and MB, verified that both holes and electrons generated by g-C_3N_4/Au/CdS had strong redox ability [34]. Wang et al. synthesized CdS/Au/g-C_3N_4 heterojunctions by photodeposition

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https://doi.org/10.1016/j.materresbull.2018.09.009
Received 21 May 2018; Received in revised form 4 September 2018; Accepted 6 September 2018
Available online 07 September 2018
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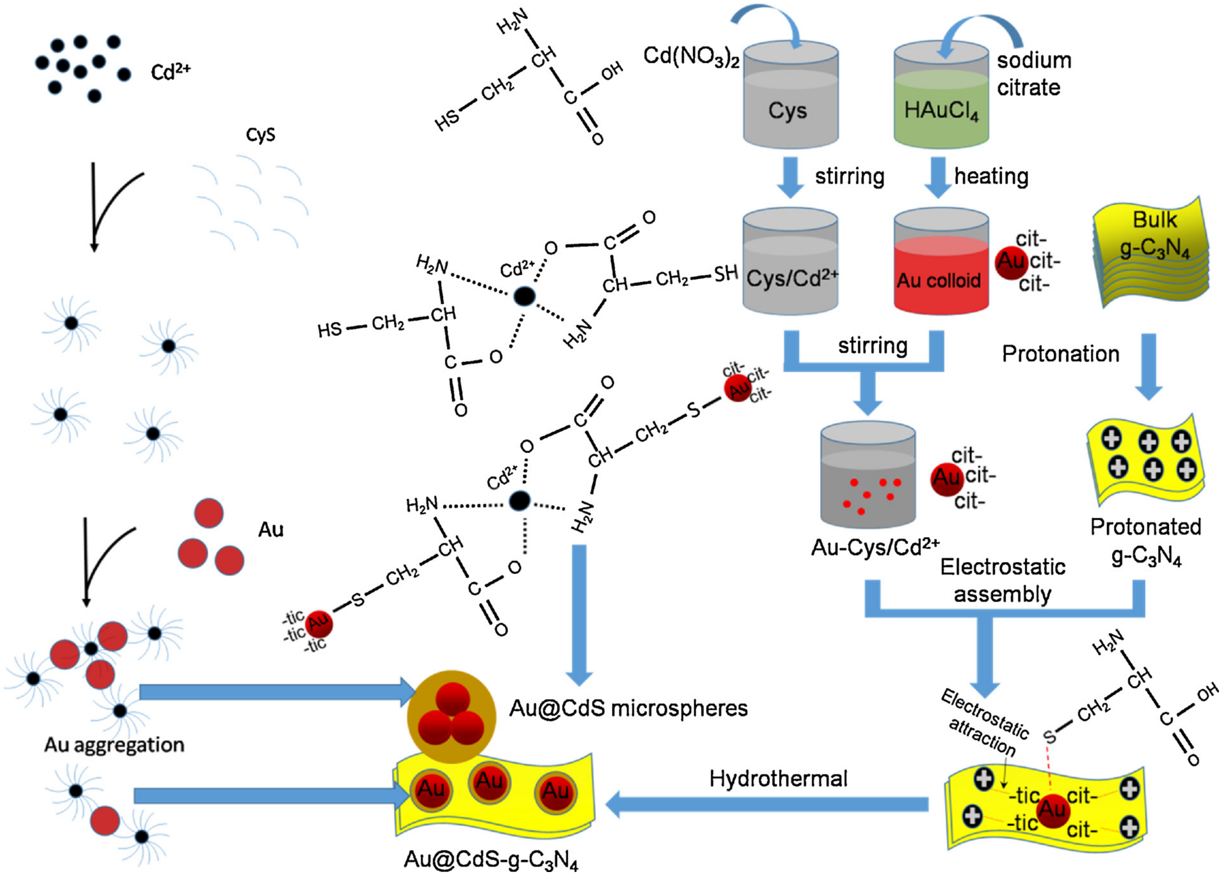

Scheme 1. Schematic illustration for the preparation of Au@CdS/g-C_3N_4 composites.

method with S_8 as sulfur source. The hydrogen production activity of Z-scheme structure is 125.8 times of pure g-C_3N_4 [35]. Very recently, Z-scheme structure CdS/rGO/g-C_3N_4 was prepared by hydrothermal method using graphene as electron mediator and DIMS as sulfur source, which demonstrates the long-term stability and photocatalytic activity for atrazine degradation [36]. In addition, a number of ternary sulfides, such as CdZnS [37], $Cd_xZn_{1-x}S$ [38], $ZnIn_2S_4$ [39], and $CuInS_2$ [40] are also reported to construct the Z-scheme structures with g-C_3N_4. However, a good bonding interface is of paramount significance to the construction of all-solid-state Z-scheme structures and it is also the key to ensure effective charge separation in the heterojunctions. Due to the homogeneous nucleation process of sulfides in commonly used methods, the morphology and distribution of heterojunctions cannot be well controlled. Moreover, the interface between sulfides and noble metal nanoparticles (solid mediators) majorly relies on their affinity [22]. Thus, the surface resistance is large and it is difficult to construct Z-scheme structure. Our group has synthesized the ternary composite CdS/Au/g-C_3N_4 by photodeposition method. Both II-type heterojunction CdS-g-C_3N_4 and Z-scheme heterojunction CdS-Au-g-C_3N_4 were observed in the composites, indicating the weak interaction role between CdS and Au nanoparticles, which in turn affects the final photocatalytic performance of materials [41].

L-cysteine is a kind of biomolecule containing different functional groups, including hydrosulphonyl (-SH), amine (-NH_2) and carboxyl (−COOH). Its special structure enables it to combine with the most metal cations by ligand bonds and release the sulfur ions for the synthesis of various metal sulfide structures [42–45]. More interestingly, the metal ions usually form the chelation bonds with the basic function groups amine (-NH_2) and acidic function groups carboxyl (−COOH) of L-cysteine [46,47]. The hydrosulphonyl groups of L-cysteine can be used as an anchor linked with Au nanoparticles [47–49], which makes it a green and cost-effective candidate for the fabrication of Z-scheme structure where Au nanoparticles serve as the electron mediator. Herein, we report an L-cysteine biomolecules assisted hydrothermal route to fabricate the multicomponent composites Au@CdS/g-C_3N_4. The Au nanoparticles were firstly synthesized by a traditional citrate reduction method. These citrate protected Au nanoparticles can couple with L-cysteine biomolecules through the linkage with hydrosulphonyl groups [47–49]. With the remaining amine and carboxyl groups bonded with metal ions, L-cysteine functions not only the sulfur source but also the directing agent to improve the interfacial bonding of CdS layer and Au nanoparticles. Moreover, the negative charged Au nanoparticles may closely integrate with protonated g-C_3N_4 bulks through the electrostatic adsorption [50]. Therefore, Au nanoparticles as the electron transfer mediator may tightly attach with both semiconductors, promoting the target preparation of multicomponent composites Au@CdS/g-C_3N_4 with Z-scheme structure. More importantly, the structures and properties of composites can be easily controlled through controlling the mass ratios of the precursors. The as-fabricated multicomponent composites Au@CdS/g-C_3N_4 demonstrated the significantly enhanced photocatalytic activity for the degradation of phenol and methyl orange (MO) under visible light irradiation as compared to pure g-C_3N_4 bulks. This work highlights the utilization of biomolecules as the directing agent to construct the multivariate heterojunctions based on sulfides and g-C_3N_4.

2. Experimental method

2.1. Photocatalyst preparation

2.1.1. Preparation of g-C_3N_4 bulks

The g-C_3N_4 bulks were prepared through a thermo-polymerization method reported in our previous literatures [41]. In typical, urea and dicyandiamide with a mass ratio of 7:3 was first ground by a

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