Journal of Molecular Catalysis A: Chemical 423 (2016) 356–364

Contents lists available at ScienceDirect

Journal of Molecular Catalysis A: Chemical

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

Fabrication of graphene-modified nano-sized red phosphorus for enhanced photocatalytic performance

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ARTICLE INFO

Article history:
Received 15 March 2016
Received in revised form 18 July 2016
Accepted 20 July 2016
Available online 21 July 2016

Keywords:
Nano red phosphorus
Graphene
Hydrogen evolution
Visible light photocataysis

ABSTRACT

In this present paper, nano-sized red phosphorus particles (NRP) were prepared through a mechanical milling and ultrasonic flotation method. The as-prepared red phosphorus nano particles were loaded on the surface of graphene. The residual oxidative bonds on the surfaces of the red phosphorus and graphene were carried out to form effective interface contact. The present results indicated that comparing with large size commercial red phosphorus particles, NRP possesses higher specific surface area and photocatalytic activity. The NRP/graphene composite shows a great adsorption capacity for rhodamine (RhB). Meanwhile, its visible-light-driven decolorization performance for RhB was 5 times higher than that of pure NRP, which can decolorize of RhB completely in only 3 min. So, NRP/graphene composite photocatalyst possess great potential in the area of environment organic pollution purge. Electrochemical impedance spectra (EIS) and Mott-Schottky spectra results of these samples indicated that a downward bend of the conduction band of NRP was formed when it contacted with graphene, resulting in a rapid electron injection into the graphene. Therefore, the probability of recombination of photogenerated carriers was inhibited effectively, thus enhanced the overall photocatalytic performance of the composite material.

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1. Introduction

Photocatalysis has been considered as one of the key techniques for solving energy resource and environmental problems [1,2]. Since Fujishima reported the photocatalytic hydrogen production of TiO_2 in 1972 [3], photocatalytic techniques have continued to develop. Some novel visible light photocatalysts such as $BiVO_4$ [4,5], InGaN [6,7], $BaTaO_2N$ [8] and $CuIn_xGa_{1-x}Se_2$ [9] have been developed and achieved excellent photocatalytic performances. Compared with these complicated multi-component photocatalysts, some photocatalysts with simpler compositions, including elementary sulfur [10] and red phosphorus [11], have attracted attention in recent years. Among these materials, red phosphorus is a type of stable semiconductor, with a band width of 1.7 eV that its light absorption range can almost cover the visible light range [11]. In addition, red phosphorus possess appropriate conduction band electrical potential and valence band electrical potential to decompose water into hydrogen and oxygen, which make it a potentially excellent semiconductor material for photocatalysis [11].

Wang et al. [11] made the first report of the visible light photocatalytic water reduction performance of red phosphorus, and calculated the electrical potentials of the conduction and valence bands of it using density function theory (DFT). The researchers then prepared P_4/YPO_4 composite photocatalyst through hydrothermal reaction [12]. It was found that, after hydrothermal treatment by Y^{3+} ions, the specific surface area of red phosphorus was increased greatly. Simultaneously, the separation efficiency of the photoinduced carriers were enhanced by the heterojunction effect between the red phosphorus and YPO_4. Yuan et al. [13] prepared a g-C_3N_4 layer coated red phosphor/g-C_3N_4 heterojunction photocatalyst; they found that the photocatalytic hydrogen evolution performance on water and photocatalytic CO_2 reduction performance, were significantly enhanced when the mass fraction of g-C_3N_4 reached 30%. Xia et al. [14] studied the photocatalytic antibacterial performance of red phosphorus excited by visible light and sunlight, and found that the oxygen species ($\bullet OH$, $\bullet O^{2-}$, H_2O_2) formed using red phosphorus under light irradiation could oxidize the cell membrane of E.coli, which eventually achieved the goal of antibacterial action.

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http://dx.doi.org/10.1016/j.molcata.2016.07.039
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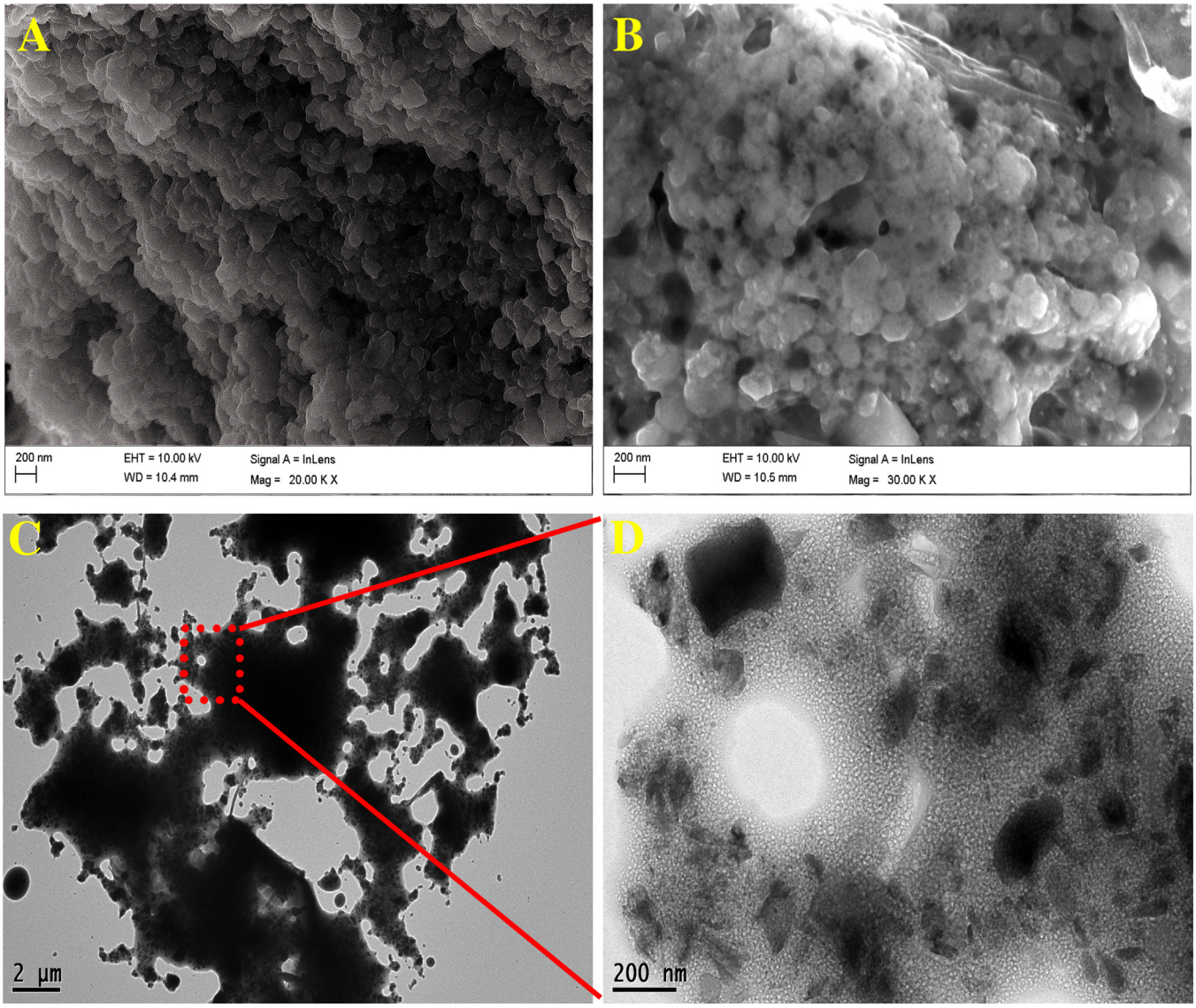

Fig. 1. SEM images of (A) NRP; (B) NRP/G 2; TEM of NRP/G2 (C) low resolution and (D) higher resolution.

However, the photocatalytic performance of red phosphorus is possibly limited by the following two key factors. Firstly, the red phosphorus previously applied in photocatalysis presented a large particle size, which decreases the number of active sites on the surface of red phosphorus, thereby limiting the photocatalytic performance. Secondly, the crystallinity of red phosphorus semiconductors is relatively low, increasing the probability of recombination between photo-induced electrons and holes. Therefore, developing a feasible method to prepare nano-sized red phosphorus photocatalysts, as well as increasing the electron mobility of red phosphorus photocatalysts, are both importance.

In this work, red phosphorus was modified to address the above-mentioned two problems. Since red phosphorus is a relatively soft material, nano-sized red phosphorus would be prepared through a combination of mechanical milling and ultrasonic floatation methods. In addition, Graphene is a two-dimensional material with very high electron mobility [15–19]. Currently, amount of studies indicated that [20–25], when graphene was compounded with other semiconductor materials, graphene could significantly increase the electron mobility of photocatalysts through the interfacial photo-induced electrons transfer process; In addition, graphene could promote the separation efficiency of light-induced electrons and holes in semiconductor photocatalysts. Therefore, in the present paper, nano-sized red phosphorus was prepared through a simple method, and then the as-prepared red phosphorus nanoparticles were loaded on the surface of graphene, which was expected to achieve the objects of increasing active sites for photocatalytic reaction, meanwhile promoting the mobility and separation efficiency of photo-induced electrons also.

2. Experimental

2.1. Preparation of nanoparticles red phosphorus (NRP)

In this study, all the reagents were purchased from Aladdin Industrial Corporation of analytical grade, and used directly without any purification. Red phosphorus was prepared by mechanical boll milling and flotation method. In brief, firstly 10 g commercial red phosphorus was annealed at 400 °C (with a rate of temperature up/down 5 °C/Min) for 4 h in the N_2 atmosphere to enhance the crystallinity. After that, 4 g annealed red phosphorus was grinded by a ball mill at a revolving speed 350 r/min for 12 h with water. Finally, 0.5 g grinded red phosphorus was dispersed in 100 mL anhydrous alcohol for 30 min by ultrasonic dispersion method.

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