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Simultaneous photoreduction of Uranium(VI) and photooxidation of Arsenic (III) in aqueous solution over g-C_3N_4/TiO_2 heterostructured catalysts under simulated sunlight irradiation

Xun-Heng Jiang[a], Qiu-Ju Xing[a], Xu-Biao Luo[a], Fei Li[a], Jian-Ping Zou[a,*], Shan-Shan Liu[a], Xiang Li[a], Xiang-Ke Wang[b,*]

[a] Key Laboratory of Jiangxi Province for Persistent Pollutants Control and Resources Recycle, Nanchang Hangkong University, Nanchang 330063, PR China
[b] School of Environment and Chemical Engineering, North China Electric Power University, Beijing 102206, PR China

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ABSTRACT

In the present work, the hererostructured catalysts of g-C_3N_4/TiO_2 were synthesized and well characterized by XRD, SEM, TEM, Raman, UV–vis diffuse reflectance spectra, PL, Mott-Schottky, and XPS. Simultaneous photoreduction of Uranium(VI) and photooxidation of Arsenic(III) was firstly achieved over the g-C_3N_4/TiO_2 catalysts. And the experimental results show that the removal rate of U(VI) decreases with the increase of As(III) concentration, whereas the photooxidation rate of As(III) to As(V) increases with the increase of As(III) concentration. Noteworthily, the photoreduction of U(VI) to U(IV) and photooxidation of As(III) to As(V) was confirmed by XPS analysis in time-scale. The experimental results of free radical capture and quantitative test indicate that holes, hydroxyl radical and superoxide radical are the major active species for photooxidation of As (III), while U(VI) was reduced to U(IV) by the photogenerated electrons. Furthermore, a possible mechanism was proposed to well explain the improved photocatalytic performance of the g-C_3N_4/TiO_2 and the competitive relationship between photoreduction of U(VI) and photooxidation of As(III). The present work develops a heterostructured catalyst for potential application to the simultaneous removal of U(VI) and As(III), and makes clear the effect of photooxidation of As(III) on photoreduction of U(VI) for the first time.

1. Introduction

With the rapid development of modern industry, the shortage of fossil fuels and global warming became very seriously environmental and social issues [1]. Nuclear power, as a clean energy, has become increasing worldwide concerns because of without greenhouse gas emission. During the past several decades, anthropocentric activities, such as uranium mining [2] and processing [3], improper nuclear waste management and nuclear safety accidents, have resulted in the release of considerable amounts of uranium into the natural environment, mostly through migration in ground or surface water systems as the highly soluble uranyl (VI) ion [4,5]. Uranium's combination of chemotoxicity and radiotoxicity can lead to irreversible kidney damage, urinary system disease [6], DNA damage [7], and the disruption of biomolecules [8]. Meanwhile, the heavy metal pollution continues to undermine the environment of the whole earth, and endanger the human healthy and survival. Arsenium is commonly found in groundwater as well as industrial wastewater. It is highly toxic and known to cause skin, lung, kidney, stomach and bladder cancers [9–11]. Furthermore, some literatures reported that arsenium contaminant will be associated during the uranium mining and utilization [12]. Thus, it is very urgent and meaningful to efficiently treat the wastewater containing uranium or arsenium [13].

Till now, several methods have been investigated to treat the uranium and arsenic wastewater. Hexavalent uranium (U(VI)) can be physically removed by solvent extraction [14], membrane filtration [15], and adsorption [16]. And U(VI) can be removed by chemical precipitation, chemical reduction [17] and biological treatment [18]. Meanwhile, the reported conventional methods for the removal of arsenium consist of adsorption [19], precipitation [20] and chemical oxidation [21]. These methods show some advantages for removing uranium and arsenium such as high adsorption capacity, rapid kinetics and high selectively [22,23]. However, due to the limited saturation of adsorption materials and the variety of surrounding environment, uranium pollution is likely to be returned to the aqueous system, leading to secondary pollution. Thus, it is necessary to develop new methods to

* Corresponding authors.
E-mail addresses: zjp_112@126.com (J.-P. Zou), xkwang@ncepu.edu.cn (X.-K. Wang).

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efficiently remove the trace amount of uranium or arsenium in the water.

Photocatalysis treatment technologies have been extensively studied to remediate contaminated water because they can well use sunlight and show good efficiency and environmental-friendly feature [24,25]. Recently, some researchers have reported that uranium and arsenium can be removed from aqueous solution over some photocatalysts under visible light or simulated sunlight irradiation. For example, Liu reported that U(VI) was reduced to U(IV) by B [26] or S [27] doped g-C_3N_4 under visible light irradiation. And TiO_2/Fe_3O_4 was used to remove U(VI) by the photocatalysis method [28]. Meanwhile, some catalysts were used to oxidize As(III) to As(V) under simulated sunlight irradiation, such as TiO_2 [29], Ferric Hydroxide [30] and Iron oxide nanoparticles [31]. However, the removal rate and efficiency of uranium and arsenium is still low and the quantum efficiency is very small because the photogenerated electrons and holes cannot be effectively used at the same time. And there are no reports on the combination of the simultaneous removal of uranium and arsenium by photocatalysis technology. In theory, the dual-purpose approaches of the coupling of the simultaneous removal of uranium and arsenium would be superior to the single-purpose ones in terms of efficiency and cost.

As well known, uranium species exist in several chemical states including U(III), U(IV), U(V), and U(VI), where the predominant chemical states are less soluble U(IV) and soluble U(VI). And the reduction potential of UO_2^{2+}/U^{4+}, UO_2^{2+}/UO_2 and U_4O_9/UO_2 is 0.267 V, 0.411 V and 0.456 V, respectively [32,33]. For arsenium, As(III) is the most prevalent form in the natural environment, which is more toxic than As(V) to environment, and As(III) is also more mobile than As(V). And the reduction potential of AsO_2/AsO_4^- and AsO_4^{3-}/AsO_2^- is 0.56 V and −0.71 V, respectively [34]. Thus the removal of soluble U (VI) was generally achieved by the reduction of U(VI) to less soluble U (IV), whereas the removal of As(III) was finished via the oxidation of As (III) to less toxic As(V) [35,36]. Furthermore, the photocatalysis technology contains photooxidation and photoreduction reactions. In order to efficiently remove U(VI) and As(III) in the water, it is desired to couple photoreduction of U(VI) with photooxidation of As(III) based on photocatalysis technology. Noteworthily, the combination of oxidation and reduction reactions would markedly improve the separation rate of photogenerated holes and electrons and enhance the quantum efficiency, leading to enhancement of removal rate of U(VI) and As(III).

To achieve the simultaneous and high-efficient removal of U(VI) and As(III) in the water, in this work, we firstly couple photoreduction of U(VI) with photooxidation of As(III) using the heterostructured catalyst of g-C_3N_4/TiO_2 under simulated sunlight irradiation. Furthermore, the photocatalytic improvement mechanisms of the heterostructured catalyst and the coupling mechanisms of simultaneous photoreduction of U(VI) and photooxidation of As(III) were systematically studied. And the results clearly indicate the enormous potential of the as-synthesized heterostructured catalyst in the treatment of radioactive and chemical toxic wastewater. The present work will pave the way for further developing high-efficient photocatalysis system for the simultaneous removal of uranium and arsenium.

2. Experiment section

2.1. Chemicals

All reagents were analytical grade and used without further purification. The detailed information of the chemicals are shown in the Supporting information (Text S1).

2.2. Syntheses

2.2.1. Synthesis of the mesoporous TiO_2

The mesoporous TiO_2 was synthesized via a modified solvothermal process based on the previously reported method [37,38]. In a typical process, 0.1 mL of diethylenetriamine was added to 80 mL of isopropyl alcohol with vigorous stirring, and 3.8 mL of titanium isopropoxide was dropwised to the above solution under vigorous stirring. The mixture was stirred for 30 min and then transferred to a 100 mL Teflon-lined stainless-steel autoclave. After treatment at 200 °C for 24 h, a white precipitate was collected by centrifugation and washed with ethanol and deionized water several times. Finally, white powder of TiO_2 was obtained after dried at 120 °C for 4 h.

2.2.2. Synthesis of g-C_3N_4

Typically, 6.5 g of urea was put into an alumina crucible with a cover and then heated to 550 °C in a muffle furnace for 2 h at a heating rate of 8 °C min^{-1} based on the previously reported method [39]. After the reaction, the alumina crucible was cooled to room temperature, and a faint yellow powder of g-C_3N_4 was obtained.

2.2.3. Synthesis of g-C_3N_4/TiO_2 composites

The synthesis process of g-C_3N_4/TiO_2 composites was similar to that of the pure g-C_3N_4 via a calcination method [40]. A certain amount of urea (3.25, 6.5, 9.75, and 13 g) was mixed with the as-prepared TiO_2 (0.25 g), then the mixture was calcined under 550 °C in a muffle furnace for 2 h at a heating rate of 8 °C min^{-1}. After the reaction, the alumina crucible was cooled to room temperature, and a series of products with the color from white to yellow were obtained. The as-prepared g-C_3N_4/TiO_2 composites with mass ratio of g-C_3N_4 and TiO_2 being 20%, 40%, 60%, and 80% are denoted as 20 CNT, 40-CNT, 60-CNT, and 80-CNT, respectively.

2.3. Analyst methods

2.3.1. Measurement of As(III) concentration

The concentration of As(III) was measured using a hydride generation atomic fluorescence spectrometry (AFS-8220) equipped with an arsenic air cathode lamp. A citric acid-sodium citrate buffer solution (pH = 5) was used as the current-carrying liquid in the analysis. The signal of AsH_3 was recorded for the determination of arsenic concentration. Prior to analysis, the samples were diluted with the buffer solution to make sure the concentration of arsenic was lower than 10 μg/L. A solution composed of 2% KOH and 20% KBH_4 was employed as the reductant to reduce As(III) to AsH_3. Total arsenic was measured by mixing the sample solution with 1 mL of concentrated hydrochloric acid and 1 mL of thiocarbamide solution (5%) for 30–60 min to reduce As(V) to As(III). During this process, only As(III) could be converted to AsH_3.

2.3.2. Measurement of As(V) concentration

Molybdenum blue method was used to analyze the concentration of As(V). Typically, 6.5 g ammonium molybdate and 0.175 g antimony potassium tartrate were dispersed in 50 mL deionized water, and then added to 100 mL (v:v = 1:1) of sulfuric acid with deionized water under vigorous stirring to form solution A. 5 g ascorbic acid was dispersed in 50 mL deionized water to form solution B. Before the detection of the As (V) concentration, 1 mL solution B and 2 mL reagent A are successively added to a 40 mL sample aliquot in a 50 mL volumetric flask. A blank solution was prepared according to the same procedure using the appropriate volume of deionized water. The concentration of the As(V) was analysed by UV–vis spectrometry (MAPADA V-1100D) at wavelength of 870 nm.

2.3.3. Measurement of U(VI) concentration

Arsenazo-III method was used to analyze U(VI) concentration. Solution A of 0.1% 2,4-dinitrophenol, solution B of 3 mol/L hydrochloric acid, solution C of chloroacetic acid-sodium acetate buffer solution, and solution D of 0.5 g/L arsenazo-III were prepared. Then the solutions A–D (according to the volume ratio of (1:3:4:4)) were mixed.

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