



Efficient removal of radioactive iodide ions from water by three-dimensional Ag₂O–Ag/TiO₂ composites under visible light irradiation



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HIGHLIGHTS

- 3D Ag₂O–Ag/TiO₂ composites have been synthesized through a facile method.
- 3D Ag₂O–Ag/TiO₂ composites exhibit large photocatalytic adsorption capacity, high selectivity, and excellent trace removal performance of I[−] under visible light.
- 3D Ag₂O–Ag/TiO₂ composites could be easily separated and regenerated.
- The adsorption capacity of Ag₂O for I[−] is enlarged 4.4 times by the photooxidation of Ag/TiO₂.
- The cooperative effects mechanism between Ag₂O and Ag/TiO₂ is proposed and verified.

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ABSTRACT

Three-dimensional Ag₂O and Ag co-loaded TiO₂ (3D Ag₂O–Ag/TiO₂) composites have been synthesized through a facile method, characterized using SEM, EDX, TEM, XRD, XPS, UV–vis DRS, BET techniques, and applied to remove radioactive iodide ions (I[−]). The photocatalytic adsorption capacity (207.6 mg/g) of the 3D Ag₂O–Ag/TiO₂ spheres under visible light is four times higher than that in the dark, which is barely affected by other ions, even in simulated salt lake water where the concentration of Cl[−] is up to 590 times that of I[−]. The capability of the composites to remove even trace amounts of I[−] from different types of water, e.g., deionized or salt lake water, is demonstrated. The composites also feature good reusability, as they were separated after photocatalytic adsorption and still performed well after a simple regeneration. Furthermore, a mechanism explaining the highly efficient removal of radioactive I[−] has been proposed according to characterization analyses of the composites after adsorption and subsequently been verified by adsorption and desorption experiments. The proposed cooperative effects mechanism considers the interplay of three different phenomena, namely, the adsorption performance of Ag₂O for I[−], the photocatalytic ability of Ag/TiO₂ for oxidation of I[−], and the readsorption performance of AgI for I₂.

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1. Introduction

With the frequent occurrence of nuclear accidents, a large number of radioactive elements, especially ¹³¹I, are discharged into the environment, which threatens our health [1,2]. Meanwhile, radioactive iodide is incessantly used in biological experiments,

medical treatments, and medical diagnoses [3,4], especially in the radioactive iodine treatments for thyroid cancer, a significant amount of radioactive waste is generated from the patients. Therefore, efficient removal is of great importance. The most common method for the disposal of iodine waste is an anion exchange resin or charcoal column [5,6]. However, these operations are inconvenient and time consuming, so the need for new materials to remove radioactive waste is highlighted. Recently, some materials, such as, saponite [7], activated carbon [8], zeolite [9], heavy metal (Hg²⁺) compounds [10], and silver oxide composites [11,12], have been reported to remove radioactive I[−]. Remarkably, Ag₂O composites

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have shown better I^- removal ability, but most of the removal processes are only through monotonous chemical adsorption.

Photocatalytic technology, developed in recent years, provides an ideal method to treat wastewater [13,14]. Titanium dioxide (TiO_2) is one of the most widely used photocatalytic materials owing to its long-term physical and chemical stability, low cost and high catalytic efficiency. However, its large-scale industrial application is greatly limited because of the lack of visible-light utilization, the high charge recombination rate and the difficulty of separation [15]. To handle these problems, substantial TiO_2 -based composite photocatalysts have been developed [16–18]. In particular, the deposition of silver on multi-dimensional TiO_2 (Ag/TiO_2) has been of considerable interest because Ag nanoparticles can either extend the TiO_2 excitation under visible irradiation due to the localized surface plasmon resonance (SPR) effect or serve as electron trap to depress electron/hole (e^-/h^+) recombination by forming Schottky barrier at the interface [19–23]. Multi-dimensional TiO_2 can make the Ag/TiO_2 photocatalyst easier to separate and should have large specific surface area to achieve high photocatalytic activity. It has been shown experimentally that I^- can be easily oxidized to I_2 by Ag/TiO_2 photocatalyst, but I_2 is more apt to combine with I^- to form polyiodide ion (I_3^-) than to adsorb onto the Ag/TiO_2 surface [24–26]. Hence, the photocatalyst of Ag/TiO_2 can be used to remove radioactive I^- from water, only when its ability to capture I_2 is improved.

In this paper, we aim to develop a novel photoactive adsorbent that can efficiently remove radioactive I^- from water through the cooperative effects between adsorption and photocatalysis under visible light. We utilized large specific surface area 3D TiO_2 microspheres [27] from our previous work as a support for Ag_2O and Ag nanoparticles that were co-loaded onto the surface of the 3D TiO_2 microspheres. The morphology, microstructure, chemical compositions and optical properties of the 3D Ag_2O-Ag/TiO_2 composites were determined by a series of characterization methods. Their adsorption kinetics and isotherms for I^- were studied, and the removal efficiencies of I^- , including trace level I^- , from deionized water, seawater, and salt lake water by the 3D Ag_2O-Ag/TiO_2 composites were evaluated, respectively. Additionally, the effects of visible light and pH on I^- removal were examined, and the regeneration of the composites was also taken into account. Furthermore, we proposed a photocatalytic adsorption mechanism based on EDX, Raman, XRD, and XPS analyses of the Ag_2O-Ag/TiO_2 composites after the adsorption of I^- , then the rationality of the mechanism was examined by experiments. To the best of our knowledge, this is the first time that the photocatalytic adsorption of 3D Ag_2O-Ag/TiO_2 composites for radioactive I^- from water has been reported.

2. Materials and methods

2.1. Materials

$TiCl_4$ was obtained from Tianjin Suzhuang Chemical Reagent Ltd. (Tianjin, China), and 38% HCl was received from Tianjin Kernel Chemical Reagent Co. (Tianjin, China). $AgNO_3$ was purchased from Tianjin Yingdai Precious Chemistry Reagent Factory (Tianjin, China). KI , purchased from Tianjin Jiangtian Chemical Reagents Co., Ltd., China, was used as the source of I^- . All chemicals were used as received, without further purification.

2.2. Preparation of 3D Ag_2O-Ag/TiO_2

The 3D TiO_2 spheres were synthesized using a self-assembly process; detailed synthetic information can be found in our previous paper [27]. In brief, 500 mL of HCl (3 mol/L) was added to a flask, and 200 mL of $TiCl_4$ was subsequently added at $0^\circ C$ under vigorous stirring for 1 h resulting in the stock solution. Next, 400 mL

of the stock solution, which was diluted 10 times with deionized water, was added to a 5000-mL beaker and stirred at $60^\circ C$ for 3 h. The precipitate was washed with water three times and dried at $70^\circ C$ for 6 h. Finally, the TiO_2 spheres were obtained. The Ag_2O-Ag/TiO_2 composites were prepared as follows [19,28]: 10.0 g of TiO_2 spheres were added to 5000 mL of deionized water, and the solution was sonicated for 20 min. Next, 1.0 g of $AgNO_3$ was added to the dispersed solution and then stirred under ultraviolet irradiation for 2 h. The final product was washed with deionized water several times and dried under vacuum at $40^\circ C$ for 12 h. The processes of fabricating 3D Ag_2O-Ag/TiO_2 composites are illustrated in Scheme 1.

2.3. Characterization of 3D Ag_2O-Ag/TiO_2

X-ray diffraction (XRD) analysis was performed using a Bruker AXS D8 Focus X-ray diffractometer (Germany) equipped with $Cu K\alpha$ radiation. The patterns were recorded from 10° to 80° at 45 kV with an intensity of 30 mA. Raman spectra were obtained using a laser Raman spectrometer (DXR Microscope, American) with an excitation wavelength of 532 nm at room temperature. The X-ray photoelectron spectroscopy (XPS) of the samples was observed using a PerkinElmer PHI 1600 ESCA X-ray photoelectron spectrometer with a monochromatic $Mg K\alpha$ radiation, and the binding energies were normalized to the $C1s$ peak at 284.6 eV. Scanning electron microscopy images (SEM) and energy-dispersive X-ray analysis (EDX) were recorded on a Hitachi S-4800 instrument. Transmission electron microscopy (TEM) was performed with a JEM-2100F (JEOL, Japan) at an accelerating voltage of 100 kV. UV-vis diffuse reflectance spectra (DRS) of the samples were obtained using a SHIMADZU UV-2550 spectrophotometer using $BaSO_4$ as reference. The Brunauer-Emmett-Teller (BET) surface area and Barrett-Joiner-Halenda (BJH) pore-size distribution measurements were tested on a NOVA-2000 volumetric gas sorption instrument (Quantachrome, USA). The concentration of trace I^- was measured by inductively coupled plasma mass spectrometry (ICP-MS) on Perkin Elmer spectrophotometer (NexION 300D).

2.4. Adsorption studies

2.4.1. I^- adsorption kinetics and isotherms

Solutions with different concentrations of I^- were prepared using KI . For the adsorption kinetic study, 0.1 g of the 3D Ag_2O-Ag/TiO_2 material was added to 100 mL of the solutions with an initial concentration of 220 mg/L. After a specified time, the concentration of I^- in the solution was measured by iodine blue spectrophotometry (MT/T 892-2000). For the adsorption isothermal study, 0.1 g of the adsorbent was added to 100-mL samples with different concentrations and stirred for 5 h at room temperature. Experiments were always exposed to ambient visible light produced by indoor fluorescent lamps. The adsorption capacity (q_{I^-}) for I^- was obtained from the following equation:

$$q_{I^-} = \frac{(C_0 - C_R)V}{W} \quad (1)$$

where V (L) is the volume of solution, C_0 and C_R (mg/L) are the concentrations of the initial and remaining I^- , respectively, and W (g) is the weight of the 3D Ag_2O-Ag/TiO_2 adsorbent.

2.4.2. Effect of visible light on I^- removal

3D TiO_2 (0.1 g) and 3D Ag_2O-Ag/TiO_2 (0.1 g) materials were added to 100 mL of the deionized water with 220 mg/L of I^- , respectively, and then the solution was stirred at $25^\circ C$ for 5 h in the dark. In comparison, 0.1 g of 3D Ag_2O-Ag/TiO_2 composites was used in

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