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Regular Article

Efficient As(III) removal by magnetic CuO-Fe_3O_4 nanoparticles through photo-oxidation and adsorption under light irradiation

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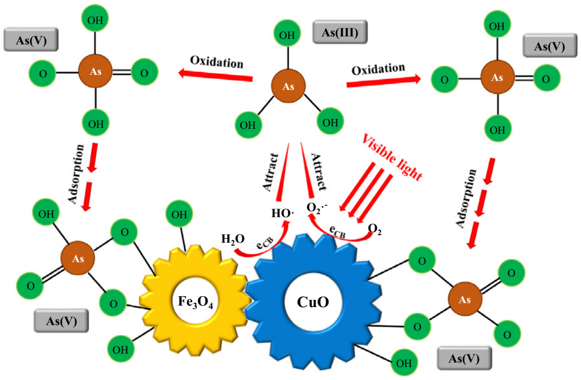
GRAPHICAL ABSTRACT



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ABSTRACT

A novel bifunctional CuO-Fe_3O_4 magnetic material was synthesized and characterized, to remove As(III) through photo-oxidation and adsorption. Photo-oxidation of As(III), adsorption characteristics of CuO-Fe_3O_4 nanoparticles, the effect of solution pH, ionic strength and coexisting ions on As(III) removal were investigated. Under light irradiation, As(III) could be completely oxidized to less toxic As(V) by CuO-Fe_3O_4 nanoparticles within 60 min in the photo-oxidation reaction. Simultaneously, As(V) could be adsorbed onto the surface of nanoparticles with high efficiency. The adsorption of As(V) was well fitted by the pseudo-second-order model and the Freundlich isotherm model, respectively, and the maximum adsorption capacities of the nanoparticles was 118.11 mg/g at pH 7.0. As(III) could be effectively removed by CuO-Fe_3O_4 nanoparticles at initial pH range from 4 to 8. Among all the common coexisting ions investigated, except for chloride and sulfate, carbonate, silicate and phosphate decreased the As(III) removal by competing with arsenic species for adsorption sites. The synthesized magnetic CuO-Fe_3O_4 combined the photocatalytic property of CuO and the high adsorption capacity of both CuO and Fe_3O_4, which make it have significant potential applications in the arsenic wastewater treatment.

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1. Introduction

Arsenic, a ubiquitous, toxic and carcinogenic element in the environment, is of worldwide serious concern [1]. The drinking water containing arsenic can cause a variety of diseases, such as dermatosis, the cancers of nasal passages and viscus [2]. To mini-

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mize these health risks, the World Health Organization (WHO) has stipulated the limited maximum concentration of arsenic was 10 $\mu g \cdot L^{-1}$ in drinking water [3]. In natural water, arsenic exists in two predominant inorganic species, arsenite (As(III)) and arsenate (As(V)) [4]. As(III) is usually the dominant arsenic in groundwater [5,21,22], which is much more toxic, soluble, and mobile than As (V) [6], and therefore deserves more attention.

A variety of technologies, including electric coagulation [7], chemical precipitation [8], ion exchange [9], membrane filtration [10], ultrafiltration [11], reverse osmosis [12] and adsorption [13] have been applied to remove arsenic. Adsorption is one of the most promising approaches to remove arsenic from water. Ferric (hydr) oxides are highly preferred for arsenic removal among various adsorbents, due to high affinity to arsenic, low cost, ease of synthesis, and environmental friendliness [14,15]. However, most adsorbents are highly effective for the removal of As(V) relative to As(III) [16]. To achieve higher arsenic removal, an oxidation pretreatment is employed to convert As(III) to As(V) prior to adsorption [17].

Recently, composite adsorbents containing ferric oxides have gained considerable attention, because the composites inherit the advantages of parent oxides and have obviously joint effect [18,19]. Zhao et al. [20] and Chao el al [21] synthesized an integrated Fe-Mn binary oxide to enhance the As(III) removal through the oxidation property of MnO_2 and then adsorption. But the oxidation capacity of Fe-Mn oxides eventually disappeared with the consumption of MnO_2 by the oxidation of As(III) [22]. Since the UV/TiO_2 system could provide a robust and low-cost approach without any damage of adsorbent, many efforts [23,24] have been made on the photo-oxidation of As(III) with TiO_2 under UV light. Yu et al. [25] reported the synthesis Fe-Ti binary nanoparticles to remove arsenic. Although As(III) is completely oxidized to As(V) by photo-oxidation, the adsorption capacity of TiO_2 is very low, resulting in the inefficient removal of As(V) [26]. Therefore, there is urgent demand for economical, effective, magnetic and reliable composites to remove As(III) through photo-oxidation and adsorption.

Copper oxide is a p-type semiconductor material with a narrow band gap of 1.4 eV [27], which is non-toxic and its constituents are available in abundance. Owing to its special properties of a large surface to volume ratio, increased activity, special electronic properties and unique optical properties [28], CuO is attractive for various applications, especially for catalysis and adsorption. Due to the photoconductive and photochemical properties of CuO, many studies have focused on its applications in photocatalysis [29]. Movever, CuO adsorbent is effective in removing arsenic from water [30]. Martinson and Reddy [31] reported that cupric oxide was an effective sorbent for both As(V) and As(III) removal over a wide pH range and performed well in the presence of competing anions. Therefore, it can be anticipated that a Fe-Cu binary oxide may possess excellent capability of the As(III) removal and can be easily separated from water with a simple magnetic process after photo-oxidation and adsorption.

On the basis of the above consideration, this study mainly focused on the As(III) removal by bifunctional CuO-Fe_3O_4 nanoparticles through photo-oxidation and adsorption. Meanwhile, the adsorption characteristics of composites were evaluated, and the effects of photo-oxidation time, initial pH, ion strength and coexisting anions on the As(III) removal were also examined. The photo-oxidation and adsorption mechanisms for the As(III) removal was also taken into consideration.

2. Materials and methods

2.1. Materials

All chemicals used were analytical grade and were purchased from Sinopharm Chemical Reagent Co. Ltd (Shanghai, China). $NaAsO_2$ [As(III)] as Arsenic source was obtained from Beijing Chemical Reagent Factory (Beijing, China). All stock and fresh solutions were prepared with ultrapure water. As(III) stock solutions (1 g/L) was prepared by dissolving $NaAsO_2$ in ultrapure water. As (III) working solutions were freshly prepared by diluting arsenic stock solutions with ultrapure water.

2.2. Preparation of CuO-Fe_3O_4 nanoparticles

The bifunctional Fe-Cu composites were synthesized by a one-step coprecipitation method, according to the following procedure: Ferric chloride ($FeCl_3 \cdot 6H_2O$, 4.61 g) and ferrous chloride ($FeCl_2 \cdot 4H_2O$, 1.99 g) were dissolved in 400 mL ultrapure water at a molar ratio of 1.7:1. The solution was stirred magnetically for 30 min, and then copper chloride ($CuCl_2 \cdot 2H_2O$, 0.125 M) was slowly added into the solution. After being stirred for several minutes, 2 g methenamine was added to the above suspension. Under vigorous magnetic stirring, 0.5 M sodium hydroxide (NaOH) solution was added dropwise to raise the mixture solution pH to around 10.0. After addition, the suspension formed was continuously stirred for 2 h at 90 °C water bath, aged at room temperature for 8 h, and then washed several times with ultrapure water till alkali free. The molar ratio of CuO to Fe_3O_4 in the synthesized composite was 1:1. The obtained material appeared fine powders, and dried in a vacuum oven at 80 °C for 12 h. Finally, the dry material was stored in a desiccator for further use.

2.3. Adsorbent characterization

The crystal phase of the synthesized materials was detected on an X-ray Diffractometer (XRD-6100, Shimadzu, Japan) operated at 40 kV and 30 mA with Cu/Kα radiation with 2θ ranging from 10° to 80°. The morphologies of the synthesized materials were examined by the scanning-electron microscope coupled to energy dispersive spectrometer (SEM- EDS, Hitachi S-3500N, Japan) and the transmission electron microscopy (TEM, JEOL-2010, Japan). The Brunauer-Emmett-Teller (BET) specific surface area and pore volume were calculated from N_2 adsorption/desorption isotherms determined using an automated nitrogen adsorption analyzer (ASAP 2020, Micromeritics, America). A vibrating sample magnetometer (VSM, JDM-13, China) was applied to determine the magnetic properties. X-ray photoelectron spectroscopy (XPS, ESCALAB250Xi, Thermo, UK) was conducted to analyse the species of arsenic.

2.4. Batch experiments

The adsorption and photocatalytic activities of the samples were tested by the As(III) removal at ambient temperature. A Xe lamp (NBET Technology Co., Ltd, China) as light source is used to simulate the sunlight. The As(III) solutions were oxidized and adsorbed under visible light ($\lambda > 420$ nm) provided by the 300 W Xe lamp equipped with circulating cooling water and two cutoff filters (UVCUT 400 and VisREF 780). The distance between the light source and the samples was 6 cm. In the experiments, 10 mg of adsorbent was dispersed into 50 mL of As(III) solution at the concentration of 10 mg/L, the ion strength was adjusted to 0.01 mol/L $NaNO_3$, and then the solution containing adsorbent was transferred into a photocatalytic reactor (OCRS-IV TYPE, China) at an agitation speed of 150 rpm. For the photo-oxidation and adsorption experiments, the suspension was stirred magnetically 60 min with light, and then continuously stirred 660 min in dark to reach the adsorption equilibrium. Adsorption isotherms were obtained by varying the initial arsenic concentrations from 1 to 50 mg/L at ppm level and 0.05 to 0.8 mg/L at ppb level. To determine the contact time required to reach equilibrium, time profiles

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