



Efficient adsorption of Selenium(IV) from water by hematite modified magnetic nanoparticles



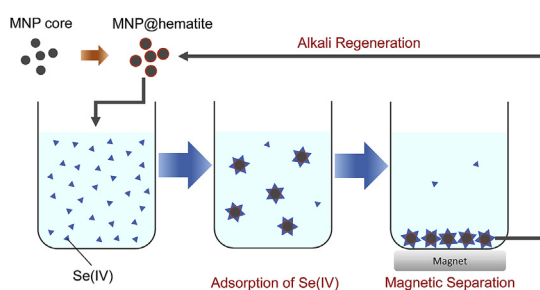
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HIGHLIGHTS

- 0.1 g/L of MNP@hematite could reduce Se(IV) from 100 to < 10 µg/L within 10 min.
- MNP@hematite could adsorb Se(IV) efficiently at pH from 4 to 9.
- Common coexisting anions and NOM did not obviously affect Se(IV) removal.
- MNP@hematite could remove Se(IV) and Sb(III) at trace level simultaneously.
- MNP@hematite could be regenerated in NaOH solution and reused for Se(IV) removal.

GRAPHICAL ABSTRACT



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ABSTRACT

Selenium (Se) could enter the environment through different anthropogenic sources, posing potential health risk. The removal of trace Se(IV) from water by hematite coated magnetic nanoparticle (MNP@hematite) under different solution conditions was investigated through batch experiments. The adsorption capacity of Se(IV) by MNP@hematite was 25.0 mg/g. 100 µg/L of Se(IV) could be rapidly decreased to below 10 µg/L by 0.1 g/L of MNP@hematite in 10 min. MNP@hematite could effectively remove Se(IV) in a wide pH range from 4 to 9. Se(IV) could form inner-sphere complex with MNP@hematite. Thus, the presence of chloride, nitrate, and sulfate (concentration of each anions <10 mM) did not affect the removal of Se(IV); whereas, carbonate (10 mM), silicate (1 mM), and phosphate (0.1 mM) yet reduced the adsorption efficiency by competing adsorption sites. Humic acid and alginate (up to 6 mg/L) did not have obviously influence on the removal of Se(IV). MNP@hematite particles were able to simultaneously remove Se(IV) and Sb(III) at trace level very efficiently in 10 min. The magnetic adsorbent can be easily recycled and regenerated in 10 mM NaOH for 15 min. In five consecutive cycles, the adsorption and desorption efficiencies were over 97% and 82%, respectively. MNP@hematite could be employed as effective adsorbent for Se(IV) removal from water.

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1. Introduction

Selenium is widely used in a variety of products such as photosensitive materials, insecticides, and semiconductors, thus it can be released into environment from sources such as the

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discharge of industrial wastewater and agricultural runoff (Bleiman and Mishael, 2010; Findik et al., 2014). Although it is one of the most important essential elements, consumption of excess selenium yet can cause loss of hair and nails, dysfunction of digestive system and even neurological damage (PerezCorona et al., 1997). The maximum contaminant level (MCL) of selenium in drinking water is regulated to be 10 $\mu\text{g/L}$ in EU, Japan, and China. Selenite (SeO_3^{2-} , Se(IV)) and selenate (SeO_4^{2-} , Se(VI)) are the predominant forms present in aqueous environment. Since Se(IV) is more toxic than Se(VI), great efforts have been taken to develop Se(IV) treatment techniques in past years.

Different techniques including co-precipitation, solvent extraction/ion-exchange, membrane separation, multiple evaporation techniques, and adsorption have been used to remove Se(IV) from water (Mentasti et al., 1989; Manning and Burau, 1995; Morrison et al., 2002; Simmons and Wallschlager, 2005; Richards et al., 2011; Awual et al., 2014, 2015a, 2015b). Among these techniques, adsorption has been drawn most attentions due to its cost effectiveness and easy operation. Various types of adsorbents such as zirconium sulfate (Takada et al., 2004), Fe/Fe₃C (Loyo et al., 2008), iron-manganese oxide (Gonzalez et al., 2010), zero-valent Iron (Ling et al., 2015), Al(III)/SiO₂ (Chan et al., 2009), pyrite (Liu et al., 2015), zirconium-based metal-organic frameworks (Howarth et al., 2015), hematite ($\alpha\text{-Fe}_2\text{O}_3$) (Jang et al., 2015; Lounsbury et al., 2016), Fe/Si coprecipitates (Chan et al., 2016), MFe₂O₄ (Sun et al., 2015), poly(1,8-diaminonaphthalene) (Findik et al., 2014), and organic ligand based composite adsorbent (Awual et al., 2014, 2015a, 2015b) have been used to remove Se(IV) from water. Se(IV) could be efficiently adsorbed by many of these adsorbents through the formation of inner-sphere complexation (Alswat et al., 2016). However, when the adsorbents are dispersed into water as fine particles, it would be very difficult to separate them from water after use. The difficulty in separation thus definitely would limit their practical application.

Due to the ease recycle by simply using external magnetic force, magnetic nanoparticles (MNPs) has recently drawn great attention in water treatment process (Tang and Lo, 2013). However, comparing with non-magnetic adsorbents, the studies focusing on the employment of MNPs to remove Se(IV) in water are limited. Only very few MNPs adsorbents such as magnetic nanoparticle-graphene oxide (You et al., 2014) and iron oxide nanoparticle/multi-walled carbon nanotube (Lee and Kim, 2016) have been fabricated and utilized to adsorb Se(IV) from water. Due to smaller adsorption capacity, bare MNPs such as magnetite without modification would not be able to efficiently remove Se(IV). Previous studies have shown that hematite ($\alpha\text{-Fe}_2\text{O}_3$) could be employed as an effective adsorbent of Se(IV) (Jang et al., 2015; Lounsbury et al., 2016). Therefore, it is expected that MNPs modified with hematite (MNP@hematite) not only could be employed to effectively capture Se(IV) from water but also would be conveniently separated from solution after treatment. Moreover, to our best knowledge, MNPs modified with hematite has never been utilized to remove trace Se(IV) from water.

The objective of this study thus is to investigate the adsorption of Se(IV) by MNP@hematite under different solution conditions. This study is mainly focusing on trace Se(IV) (~100 $\mu\text{g/L}$) removal at neutral pH (7.0). The influence of pH, interfering anions, and natural organic matters (NOM) on the removal of Se(IV) were investigated. Simultaneous removal of Se(IV) with Sb(III) was investigated. In addition, the reusability of the adsorbent was also tested.

2. Materials and methods

2.1. Materials

NaOH, HCl, and HNO₃ (guaranteed reagents) were purchased from Beijing Chemical Work. Humic acid sodium salt (Acros Organics, Geel, Belgium) and alginic acid sodium salt (Aldrich, St. Louis, MO, USA) were employed as representative NOM. All the rest of chemicals with analytical grade used in this study were from Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China) or Xilong Chemical Co., Ltd. (Shantou, Guangdong, China). The stock solutions of Se(IV) were prepared by dissolving sodium selenite pentahydrate with ultrapure water (>18.2 M Ω cm, Milli-Q, Millipore, Billerica, MA).

2.2. Adsorbent preparation and characterization

The Fe₃O₄ MNP core and MNP@hematite were fabricated following the procedure described in our previous study (Shan et al., 2014). Briefly, MNP core was synthesized via the conventional co-precipitation method under basic conditions, while hematite coating was prepared via a heterogeneous nucleation technique (Alswat et al., 2016). The detailed protocol of synthesis was provided in Supplementary Information (Text S1). The chemical composition of MNP@hematite was identified by powder X-ray diffraction (XRD, DMAX-2400, Rigaku, Tokyo, Japan). The hysteresis loop was measured to explore the magnetic properties of prepared materials by using the alternating gradient magnetometer (AGM, 2900-04C, Princeton Measurements Corporation, Princeton, NJ, USA). Transmission electron microscope (TEM, Tecnai G2 T20, FEI, Hillsboro, OR, USA) was employed to determine the surface morphology and size of MNP@hematite, while the zeta potentials of MNP@hematite at pH from 3 to 11 were measured by Zetasizer Nano ZS90 (Malvern Instruments Ltd., Worcestershire, UK). The BET surface areas were obtained from N₂ adsorption-desorption isotherms using an Accelerated Surface Area & Porosimetry System (ASAP 2010, Micromeritics Instrument Ltd., Norcross, GA, USA).

2.3. Adsorption experiments

The adsorption kinetic experiments were performed to determine the time required to reach equilibrium. 30 mg of MNP@hematite or MNP core was dispersed into flask containing 300 mL of 100 $\mu\text{g/L}$ Se(IV) solution (initial pH 7.0), which was then shaken at 160 rpm under 25 °C. 2 mL of sample was collected and filtered through a 0.22 μm polyethersulfone membrane at desired time intervals. The collected samples were then analyzed for the residual Se(IV) concentration.

In typical batch adsorption experiments, 5 mg of MNP@hematite or MNP core was dispersed into a 100-mL Erlenmeyer flask containing 50 mL of ~100 $\mu\text{g/L}$ Se(IV) solution under ultrasonic wave for 2 min. The flasks were then shaken at 180 rpm in a thermostatic orbit incubator under 25 °C until the equilibrium was reached. The initial Se(IV) solution pH was maintained to be 7.0 \pm 0.1 (unless stated otherwise). The influence of solution pH on the adsorption of Se(IV) was evaluated by adjusting the initial solution pH from 3 to 11 with either HCl or NaOH (0.1 M or 1 M). After the adsorption process, the final pH and the released concentration of Fe was measured. Adsorption isotherm at 25 °C was acquired by varying the initial Se(IV) concentrations from 1 to 120 mg/L. By introducing the corresponding sodium salts (with concentration from 0 to 10 mM) into Se(IV) solution, the effects of coexisting anions (chloride, sulfate, nitrate, carbonate, silicate, and phosphate) on the adsorption of Se(IV) was investigated. To determine the influence of NOM, humic acid and alginic acid sodium salts were

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