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Adsorption combined with superconducting high gradient magnetic separation technique used for removal of arsenic and antimony

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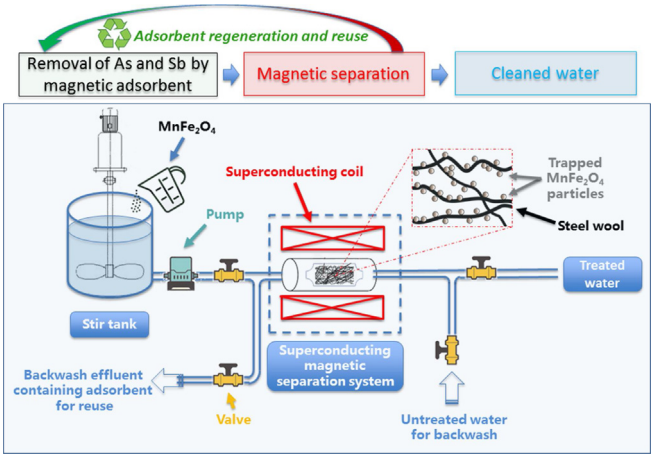
HIGHLIGHTS

- $MnFe_2O_4$ shows adsorption and catalytic oxidation efficacy towards As and Sb.
- Effective As and Sb removal by $MnFe_2O_4$ adsorption combined with HGMS system.
- The maximum separation capacities of HGMS system are well predicted using Thomas model.
- HGMS separation capability is affected by flow rate and steel wool filling ratio.

GRAPHICAL ABSTRACT



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ABSTRACT

Manganese iron oxide ($MnFe_2O_4$), an excellent arsenic(As)/antimony(Sb) removal adsorbent, is greatly restricted for the solid-liquid separation. Through the application of superconducting high gradient magnetic separation (HGMS) technique, we herein constructed a facility for the in situ solid-liquid separation of micro-sized $MnFe_2O_4$ adsorbent in As/Sb removal process. To the relative low initial concentration 50.0 $\mu g\,L^{-1}$, $MnFe_2O_4$ material sorbent can still decrease As or Sb below US EPA's drinking water standard limit. The separation of $MnFe_2O_4$ was mainly relied on the flow rate and the amount of steel wools in the HGMS system. At a flow rate 1 $L\,min^{-1}$ and 5% steel wools filling rate, the removal efficacies of As and Sb in natural water with the system were achieved to be 94.6% and 76.8%, respectively. At the meantime, nearly 100% micro-sized $MnFe_2O_4$ solid in the continuous field was readily to be separated via HGMS system.

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In a combination with the experiment results and finite element simulation, the separation was seemed to be independent on the magnetic field intensity, and the maximum separation capacities in various conditions were well predicted using the Thomas model (R^2 = 0.87–0.99).

1. Introduction

Elemental arsenic (As) and antimony (Sb) belong to Group VA of the periodic table, and these two elements can exist together in ore deposits. For example, 7% arsenic oxide and 30% antimony oxide have been detected from the mineral senarmontite [1,2]. Nearly all arsenic and antimony compounds are considerably toxic, especially the inorganic ones [3]. The contamination of drinking water by arsenic and antimony, largely due to human activities [4], is a major health problem in some Asian countries such as China [5], India [6], and Vietnam [7]. To reduce health risks, drinking-water quality guidelines set by World Health Organization (WHO, 4th edition) were as low as 10 $\mu g\,L^{-1}$ for As and 20 $\mu g\,L^{-1}$ for Sb [8]. And even more strict standards for Sb, in the range of 5–15 $\mu g\,Sb\,L^{-1}$, were adopted by United States Environmental Protection Agency (US EPA), China, Japan, and European Union countries. Furthermore, the combined pollution of As and Sb can occur in the aquatic environment due to their common origin and similar chemical characteristics, and the co-exposure to high levels of antimony and arsenic might not only raise the health risks but also increase the requirement for more reliable As and Sb-containing water treatment technologies [9]. In the past decade, various techniques such as coagulation [10], membrane filtration [11], biological treatment [12], and adsorption [13–15] have been proposed for the aqueous As and Sb removal; of these, adsorption is the most popular method due to its simplicity of operation, low cost, and high efficiency. The nano/micro-sized particles, i.e. Fe-Mn binary oxide [16], activated carbon [17], and granular ferric oxide [15], exhibited excellent ability and stability in pollutants removal, and were investigated as adsorbents for the remediation of As and Sb-contaminated aqueous systems. However, the applications of conventional adsorption technology are greatly limited due to the difficulty in separation of the used nano/micro-adsorbent from aqueous solutions [18]. Fortunately, the magnetic properties of some adsorbent materials have allowed the feasible process of solid-liquid separation to be successfully employed by simple magnetic field [19,20]. But a current unresolved question remains, how can a powerful magnetic field be incorporated to water treatment processes?

Since 1960s, the High Gradient Magnetic Separation (HGMS) system was introduced as a consequence of the breakthroughs in magnetic technology and using HGMS system in the fields of environment has received significant attention in the last few years [20–22]. In principle, HGMS systems commonly employed a porous matrix of magnetizable material (steel wool or stacked iron balls) to create high gradient magnetic interfaces for interaction, when placed inside a working field volume thus increasing separation efficiency [23]. The high-strength magnetic working field (more than 1 T) can be formed using superconducting coil, which shown great energy saving potential because of it has no resistance while being cooled to below the characteristic critical temperature (T_c). The HGMS system with high magnetic separation capability, low energy consumption, low environmental impact, and much compact structure is expected as a promising separation unit which could combine with adsorption technology to achieve the removal of contaminants by using magnetic adsorbents. On the other hand, iron oxides, e.g. γ-Fe_2O_3 [24], Fe-Mn bimetallic oxide [25], and MFe_2O_4 (M = Mn [26], Co [27], Cu [28], and so on), exhibited good adsorption capacity and excellent magnetic characteristic, and were widely-used as inexpensive adsorbents for the removal of As and Sb in aqueous solution. Magnetite (Fe_3O_4) is the most common magnetic material; however, its adsorption efficiency towards As and Sb is relatively low [29]. The incorporation of Mn into Fe_3O_4 could not only provide more effective sites available for pollutants adsorption but also retained the magnetic properties of the material. Parsons et al. successfully prepared $MnFe_2O_4$ adsorbent by the microwave assisted hydrothermal method, and this adsorbent showed 22.3 and 1.35 times higher adsorption capacity towards As(III) and As(V) as compared to the pure Fe_3O_4 [30]. Rooygar et al. investigated the feasibility of using $MnFe_2O_4$ to remove Sb(III), and the maximum adsorption capability was determined to be as high as 10.66 $mg\,g^{-1}$ [31]. Hou et al. reported the high arsenic removal by manganese (Mn)-based adsorbent, and they attributed it to Mn^{2+} release caused by As(III) oxidation and the formation of more Mn vacancy defects, which activates the adsorption site greatly [32].

To our best knowledge, no research results on the removal of pollutant by magnetic adsorbent combined with the pilot-scale superconducting HGMS system have been reported in the literature. Therefore, the main objectives of this research were to: (1) used the synthetic water to evaluate the arsenic and antimony adsorption behaviors onto magnetic $MnFe_2O_4$ particles; (2) developed a continuous pilot-scale adsorption combined with superconducting magnetic separation system and evaluated its feasibility to separate the magnetic $MnFe_2O_4$ particle for the simultaneous removal of antimony and arsenic; and finally (3) determined the optimal operating conditions of this system and identified its performance for natural contaminated water sample.

2. Experimental

2.1. Materials

$MnFe_2O_4$ magnetic particles (99.90%) used in this study were purchased from Beijing DK nano technology Co., Ltd (Beijing, China). Analytical grade reagents potassium antimony(III) L(+)-tartrate hemihydrate ($C_4H_4KO_7Sb{\cdot}0.5H_2O$), potassium antimonite(V) ($K_2H_2Sb_2O_7{\cdot}4H_2O$), sodium metaarsenite ($NaAs^{III}O_2$), and sodium arsenate dodecahydrate ($Na_3As^{V}O_4{\cdot}12H_2O$) were purchased from Sinopharm Chemical Reagent Co., Ltd. (Beijing, China). The stock solutions of As(III), As(V), Sb(III), and Sb(V) were prepared, respectively by dissolving appropriate salt in deionized water. The synthetic As/Sb-containing water samples (Table 1) were freshly prepared by diluting the stock solutions to make the desired concentrations with dechlorinated tap water. The natural river water sample containing As and Sb was collected from the Zijiang river located in Loudi City, Hunan Province of China, and was used directly without any pretreatment. In addition, the standard solutions containing 1000 $mg\,L^{-1}$ of As and Sb were purchased from the National Research Center for Certified Reference Materials of China for hydride generation-atomic fluorescence spectroscopy (HG-AFS).