



Research article

Arsenic removal from water using iron-coated seaweeds



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ABSTRACT

Arsenic is a semi-metal element that can enter in water bodies and drinking water supplies from natural deposits and from mining, industrial and agricultural practices. The aim of the present work was to propose an alternative process for removing As from water, based on adsorption on a brown seaweed (*Sargassum muticum*), after a simple and inexpensive treatment: coating with iron-oxy (hydroxides). Adsorption equilibrium and kinetics were studied and modeled in terms of As oxidation state (III and V), pH and initial adsorbate concentration. Maximum adsorption capacities of 4.2 mg/g and 7.3 mg/g were obtained at pH 7 and 20 °C for arsenite and arsenate, respectively. When arsenite was used as adsorbate, experimental evidences pointed to the occurrence of redox reactions involving As(III) oxidation to As(V) and Fe(III) reduction to Fe(II), with As(V) uptake by the adsorbent. The proposed adsorption mechanism was then based on the assumption that arsenate was the adsorbed arsenic species. The most relevant drawback found in the present work was the considerable leaching of iron to the solution. Arsenite removal from a mining-influenced water by adsorption plus precipitation was studied and compared to a traditional process of coagulation/flocculation. Both kinds of treatment provided practically 100% of arsenite removal from the contaminated water, leading at best in 12.9 µg/L As after the adsorption and precipitation assays and 14.2 µg/L after the coagulation/flocculation process.

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1. Introduction

Arsenic is a toxic and carcinogenic metalloid, naturally present in the environment but also introduced by human activities, such as mining, combustion of fossil fuels and agriculture (Ungureanu et al., 2015a). It is linked to a number of cancers, but also to many other adverse effects in health. Arsenic contamination of groundwater and the possibility of long term effects are a documented problem in many countries of the world, such as Bangladesh, India, Thailand, Taiwan, Chile, China and Argentina (Mandal and Suzuki, 2002; Sharma and Sohn, 2009), and in specific regions of many other countries (Buschmann et al., 2007; Mandal and Suzuki, 2002; Ungureanu et al., 2015a). Literature has been documenting As levels in a range from less than 0.5–5000 µg L⁻¹ (Shankar et al., 2014).

Groundwater is used for drinking water supply and arsenic removal is then of unquestionable importance. Many wastewaters, such as mining effluents, for which stricter requirements have been imposed, also require efficient techniques for removing arsenic, as well as other contaminants such as sulfate, selenium, and antimony (Liang, 2014). Arsenic has four oxidation states, As(V), As(III), As(0) and As(-III), but the predominant forms in the environment are As(V) and As(III), which are also the two common forms found in mine waters (Liang, 2014).

There are several methods for remediation of As-contaminated waters, as recently reviewed by Ungureanu et al. (2015a). Presently available techniques for As removal from water involve oxidation (photochemical, photocatalytic, biological and in situ), adsorption (with activated alumina, iron-based sorbents, zero-valent iron, indigenous filters, metal organic frameworks and miscellaneous sorbents), ion exchange, membrane technologies (micro, ultra and nano filtration and reverse osmosis) and coagulation-flocculation. Most of the available removal techniques are more efficient for arsenate than arsenite and treatment technologies are believed to be more effective by using a two-step approach: initial oxidation from arsenite to arsenate followed by

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the removal of arsenate (Nicomel et al., 2016). Coagulation/flocculation, conventionally employed in drinking-water treatment processes, is not a specific method for As removal, but its ability to reduce arsenic levels has been demonstrated in many studies (Pallier et al., 2010). Adsorption is a relatively simple method, efficient on the removal of different contaminants from aqueous solution. However, the adsorption of arsenic on many organic adsorbents is not easily achieved, due to the anionic character of the species (depending on the pH and redox conditions) and due to its weak tendency to complex with many functional groups. Natural and synthetic iron oxides predominate in literature as potential adsorbents for As (Giles et al., 2011). However, the fine particles that typically constitute iron and iron oxides are usually difficult to handle and limit the future scale-up and use in continuous-systems (fixed-bed column, for instance) (Ogata et al., 2011). These drawbacks have motivated the impregnation or coating of different supporting materials with iron, in order to produce effective adsorbents for As: activated carbons (Arcibar-Orozco et al., 2014; Chang et al., 2010), fiber glass and cellulose (Kumar et al., 2008), chitosan (Gupta et al., 2009), alginate beads (Sigdel et al., 2016) and clays (Mishra and Mahato, 2016). Iron coating creates specific adsorption sites in the incorporated Fe particles and changes the chemical activity of the adsorbent, shifting the pH at the point of zero charge (Arcibar-Orozco et al., 2014), which eventually facilitates electrostatic interactions between As and the adsorbent surface.

In the last decade, great attention has been given to adsorbents based on natural, waste and readily-available materials, trying to develop low-cost strategies for water treatment. Marine seaweeds, in natural forms, are highly effective on the removal of cationic metal species from solution, but previous studies (Filote et al., 2016) have showed their ineffectiveness towards arsenic. In the present study, the brown marine algae *Sargassum muticum* was coated with iron-oxy(hydroxides) and studied as adsorbent for As(III) and As(V). According to the authors' knowledge, this kind of iron-treatment using seaweeds as support material has not been tried before. Adsorption equilibrium and kinetics were studied, conversion between arsenite and arsenate species assessed and the stability of the adsorbent evaluated. The efficiency of As removal from a contaminated groundwater sample by adsorption and by coagulation/flocculation was compared.

2. Materials and methods

2.1. Arsenic solutions

Arsenic was studied under the trivalent (arsenite) and the pentavalent (arsenate) oxidation states. As(III) solutions were prepared by dilution of a As_2O_3 standard solution 1003 ± 3 mg-As/L (obtained commercially, from SCP Science). As(V) solutions were prepared by dilution of a standard 994 ± 10 mg-As/L, prepared with $\text{HAsNa}_2\text{O}_4 \cdot 7\text{H}_2\text{O}$ (Sigma-Aldrich; analytical grade).

2.2. Preparation of iron-coated algae

Sargassum muticum (SM) was collected at Viana do Castelo beaches, on Portugal's northern coast. After preliminary washing with tap water and drying (60°C), the algae was ground to particle sizes ranging between 5 and 12 mm and then washed with distilled water and dried again at 60°C in the oven. The iron-coated algae (denoted as Fe-SM) was prepared based on the procedure reported by Kumar et al. (2008). SM (dosage 10 g/L) was put in contact with FeCl_3 0.1 mol L^{-1} solution, under mechanical stirring (200 rpm). The iron was precipitated by adjusting the pH to 5, using NaOH

10 mol L^{-1} solution. The stirrer was switched off and the suspension was let to settle for a day. The mixture was filtered, using a strainer (pore size 2 mm), in order to recover the iron-coated algae. The liquid and the precipitated iron that remained in suspension were discarded. The adsorbent was washed with distilled water and dried at 60°C .

2.3. Adsorbent characterization

Raw and iron-coated algae were analyzed in order to determine the total iron content. Samples (0.5 g) of SM and Fe-SM were acid-digested with 12.0 mL HCl 37%, 4.0 mL HNO_3 65% (analytical-grade acids from Sigma-Aldrich and Scharlau, respectively), and 5.0 mL distilled water, in glass tubes at 150°C for 2 h. Total iron dissolved in the digested liquid was analyzed by flame atomic absorbance spectrometry (FAAS), in the GBC 932 plus spectrometer, and the iron content in the solid samples calculated by mass balance.

Infrared spectra for raw (SM) and iron-treated algae, before (Fe-SM) and after As adsorption (Fe-SM-As(III) and Fe-SM-As(V)) were obtained using a Shimadzu FTIR, model IRAffinity, equipped with a solid analysis module PIKE Easidiff, in a wavenumber range of $400\text{--}4000\text{ cm}^{-1}$, 50 scans and with a resolution of 8.0 cm^{-1} . Spectra were obtained in duplicate, using algae samples ground to fine powders.

The point of zero charge (pH_{PZC}) is the pH value at which the adsorbent has a null electrical charge density on the surface. The pH_{PZC} of Fe-SM was measured by the mass titration method (Noh and Schwarz, 1989). A 0.01 mol L^{-1} NaCl solution was put in contact with different dosages of iron-coated seaweed ($1\text{--}20\text{ g L}^{-1}$), in duplicate. Blank experiments were also carried out in order to discount the possible effect of CO_2 from air. The suspensions were placed in an orbital shaker (180 rpm), inside a thermostatic chamber (20°C) for 24 h. The final pH was measured and plotted against the adsorbent dosage. The limiting pH which is found for high adsorbent dosages was identified as the pH_{PZC} .

2.4. Analytical methods

2.4.1. Arsenic

Total arsenic concentration in the liquid phase (dissolved As(III)+As(V)) was determined in the working range $7\text{--}50\text{ mg L}^{-1}$, by flame atomic absorption spectrometry (FAAS), using a GBC 932 plus spectrometer, at 197.3 nm, current 5 mA and nitrous oxide-acetylene flame. Lower concentrations were measured by graphite furnace atomic absorption spectrometry (GFAAS), using a GBC GF 3000, SenAA Dual equipment, at 197.3 nm and using a 8 mA lamp current. Whenever necessary, dilutions were made with ultrapure water, in order to obtain As concentrations falling in the linear working range ($10\text{--}50\text{ }\mu\text{g/L}$).

As(III) concentrations were determined by Square Wave – Cathodic Stripping Voltammetry (SW-CSV) at a hanging mercury drop electrode (HMDE), in HCl and copper(II) solutions. A potentiostat/galvanostat AUTOLAB model PGSTAT 10, equipped with a stirring module and a multi-mode electrode stand model 663 VA (Metrohm) was used. A hanging mercury drop electrode, an Ag/AgCl/KCl 3 mol L^{-1} and a glassy carbon rod were used as working, reference and auxiliary electrodes, respectively. GPES software from Eco Chemie was used to control the voltammetric procedure and to acquire data. Analytical and instrumental parameters are presented in Table 1. The electrolyte solution was composed by HCl 2.0 mol L^{-1} and Cu(II) 25 mg L^{-1} . Apart from these concentrations and deposition time (which were optimized specifically to get a maximum peak current), the other conditions applied were based on the work conducted by Ferreira (2009). Standard addition

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