



Arsenate removal from sulphate-containing water streams by an ion-exchange membrane process



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ABSTRACT

A possible limitation of applying Donnan dialysis for arsenate removal from drinking water supplies is when sulphate is present as an accompanying anion in the water to be treated, due to strong competition between arsenate and sulphate transport through standard grade anion-exchange membranes. This work aims at evaluating the feasibility of employing an ion-exchange membrane Donnan dialysis based process with a new strategy of using sulphate as the driving counter-ion for arsenate counter-transport and removal from sulphate-containing drinking water streams. The process performance with different arsenate receiving solutions (containing chloride or sulphate as driving counter-ions and with or without arsenic precipitation) was investigated and compared. Experiments were carried out to separate arsenate from contaminated water comparing the performance of three anion-exchange membranes: one with mono-valent anion permselective properties and two standard grade membranes. As selection criteria, the transport rate of arsenate and its membrane retention were investigated. The effects of pH variation and co-ion (cation) leakage on the Donnan dialysis process efficiency were also studied. Efficient arsenic removal was achieved through integrating Donnan dialytic transport of arsenate with its simultaneous precipitation in the receiving compartment with FeSO_4 . The process proposed is environmentally friendly and with minimal maintenance requirements, which makes it especially attractive to be applied in rural areas located far from centralized drinking water supply infrastructures.

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1. Introduction

Arsenic (As) incidence has been the focus of increasing attention since a number of countries are facing the challenge of treating their drinking water supplies in order to meet the current regulation standards for As. Due to its toxicity, mutagenicity and carcinogenicity, the World Health Organization (WHO), the European Commission (EC) and the United States Environmental Protection Agency (USEPA) reduced the maximum contaminant level (MCL) from 50 $\mu\text{g/L}$ to 10 $\mu\text{g/L}$ in January 2006 [1,2]. This stiffening of regulations reflects the need of developing highly efficient technologies for selective arsenic removal from water since recent estimations indicate that over 130 million people worldwide are potentially exposed to harmful levels of arsenic in drinking water [3].

Arsenic occurrence in natural water sources is a consequence of leaching from natural deposits (rocks/minerals) in the earth crust, and, to a lesser degree, due to anthropogenic causes, such as agriculture practices and mining industry pollution. Moreover, arsenic in its inorganic form is more toxic than in its organic form [4].

Several treatment options have been applied so far for As removal from water streams, including nanofiltration [5], reverse osmosis [4], chemical precipitation and coagulation [6,7], adsorption [8–11] and ion exchange (IEX) [12–14]. Amongst them, adsorption and anion exchange processes are the most extensively studied ones, because they generally provide relatively lower costs, ease of handling and potential reuse/recycling of the adsorbents/anion-exchangers.

Different adsorbents and IEX media have been applied such as biological materials, mineral oxides, activated carbons and polymeric resins [12–15]. The US Environmental Protection Agency (US EPA) has identified IEX as the best available technology (BAT) although the arsenate removal can suffer from interferences due to the presence of accompanying anions that are normally present in drinking water sources in much higher concentrations than that of the arsenic-containing species. The following selectivity order has been reported for strong base anion (SBA) exchangers [16]: $\text{SO}_4^{2-} > \text{HAsO}_4^{2-} > \text{Cl}^- > \text{H}_2\text{AsO}_4^- > \text{HCO}_3^- > \text{OH}^-$.

On the other hand, sulphate can be present in groundwater, at levels that sometimes can exceed 1000 mg/L [<http://www.health.state.mn.us/divs/eh/wells/waterquality/sulphate.htm>], while arsenic-containing species are most commonly present within

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the $\mu\text{g/L}$ concentration range. Therefore it is extremely difficult to reduce the As concentration in such sulphate-containing waters below the MCL of $10 \mu\text{g/L}$ of As using conventional strong anion-exchange resins. Their high affinity for sulphate leads to the need of a frequent regeneration before reuse, thus producing large volumes of brines [15,17,18].

An ion-exchange membrane process, integrating Donnan dialysis with precipitation of target ionic pollutant(s), can offer the possibility of their selective removal, while minimizing the risks of secondary pollution of the treated water, which can occur in conventional IEX treatment after the resin saturation (once the target pollutant breakthrough has occurred). Although the ion-exchange membrane process has relatively slow kinetics when compared to electrodialysis, it presents several possible advantages, especially in the case of rural applications in developing countries, for being energy saving and an easy to handle process [19].

In a prior study, performed with model single arsenate solutions, it has been demonstrated that the Donnan dialysis process performance depends strongly on the type of anion-exchange membrane used especially under batch operating conditions [20]. The removal of ions by Donnan dialysis is, however, affected by several other factors, such as the nature and concentration of accompanying ions, composition and concentration of the receiving solution and pH, amongst others [21].

Therefore this work explores the feasibility of applying an ion-exchange membrane process for arsenic removal from sulphate-containing water streams. The aims of the study are as follows: (1) examine the transport behaviour of arsenate and sulphate and compare their sorption on a mono-valent anion permselective and on two standard grade anion-exchange membranes; (2) identify optimal process conditions (membrane type and composition of the receiving solution, with and without addition of a precipitant) for arsenate removal and (3) investigate the effect of co-ion (sodium) leakage on the process performance.

In this study, a set of Donnan dialysis experiments using NaCl as a receiving solution was first performed. It was investigated whether a mono-anion permselective membrane would favour the transport of monovalent arsenate in the presence of divalent sulphate. Subsequently, the performance of two membranes with no claimed mono-anion permselectivity was tested. The affinity of the membranes towards arsenate and sulphate was also studied through dedicated sorption experiments.

A new strategy of using Na_2SO_4 in the receiving solution in order to minimize the interference of the competing sulphate was then studied. Finally, an ion exchange membrane process, Donnan dialysis for membrane transport of arsenate with its simultaneous precipitation by iron using FeSO_4 in the receiving solution was evaluated.

2. Materials and methods

2.1. Reagents and solutions

All reagents ($\text{Na}_2\text{HAsO}_4 \cdot 7\text{H}_2\text{O}$, Na_2SO_4 , NaCl and FeSO_4) used were of analytical grade. The solutions were freshly prepared with deionized water (conductivity $\leq 1 \mu\text{S/cm}$). Feed solutions were spiked with sodium arsenate and sodium sulphate to the desired concentrations, whereas receiving solutions were prepared with sodium chloride, sodium sulphate or ferric sulphate depending on the aim of the experiment.

2.2. Membranes

Three commercial anion-exchange membranes (AEM) were tested: Neosepta ACS (Tokuyama Soda, Japan) which is a mono-

valent anion permselective membrane, characterized by a compact surface structure, which causes the rate of transport and efficiency of large anion transport to decrease noticeably; a low-crosslinked PC 200D membrane purchased from PCA-Polymerchemie Altmeier GmbH (Germany), which is a membrane suitable for transport of anions with a molecular mass of up to about 200 Da [<http://www.pca-gmbh.com/membrane/membrane.htm>]; and an alkali resistant Neosepta AXE 01 membrane from Tokuyama Soda, Japan, suitable for transport of anions with a molecular mass of up to about 300 Da [22]. All membranes are homogeneous and contain quaternary ammonium as fixed charged groups. Their main properties are listed in Table 1.

The membrane thickness (in their wet chloride form) was determined (average of five measurements) using a picometer after wiping the membranes with a filter paper for moisture removal.

2.2.1. Dry weight

The dry weight of membrane circle samples (with an area of 11.3 cm^2 each) was measured after drying in an oven to a constant weight at $60 \text{ }^\circ\text{C}$ for 9 h. Immediately after drying, the membrane samples, were put in a desiccator until cooling down to the room temperature before being weighed. This procedure was also applied for longer periods of drying (24 and 72 h) and the results were found to be consistent with those obtained for 9 h of drying.

2.2.2. FTIR measurements

Infrared analyses were performed on a Nicolet Nexus spectrophotometer coupled to a Continuum microscope ($15\times$ objective) with a MCT-A detector cooled by liquid nitrogen. The spectra were collected in transmission mode (4 cm^{-1} , 128 scans), using a Thermo diamond anvil compression cell in the range $4000\text{--}650 \text{ cm}^{-1}$. Periodically the system was purged with nitrogen prior to data acquisition. For all infrared spectra the CO_2 adsorption at circa $2400\text{--}2300 \text{ cm}^{-1}$ was removed.

2.3. Donnan dialysis experiments

These experiments were carried out in a dialysis cell made of plexiglass (Fig. 1), in which two compartments (136 ml each) are in contact through a circular window, where the studied membrane sample (with an area of 11.3 cm^2) was placed. Each compartment was equipped with a stirrer (with a stirring rate set to 700 rpm, to avoid/minimize concentration polarization effects). All experiments were performed in batch operation mode in an air-thermostated room at $24 \text{ }^\circ\text{C}$. At pre-defined time intervals, samples (2 ml each) were taken from the feed and receiving solution compartments for inductively coupled plasma (ICP) analyses.

In the feed (polluted water) compartment, As (in the forms of HAsO_4^{2-} and H_2AsO_4^-) and SO_4^{2-} , competing for the transport through the anion-exchange membrane are shown. In the receiver, $\text{X}^{\vee-}$ represents the driving counter-ion used (i.e. chloride or sulphate).

The data obtained were compared in terms of arsenic removal (%) from the feed water as follows:

$$\eta (\text{Removal}) (\text{As}) = \left(1 - \frac{[\text{As}]_{\text{feed}(t)}}{[\text{As}]_{\text{feed}(0)}} \right) \times 100\% \quad (1)$$

where $[\text{As}]_{\text{feed}(0)}$ and $[\text{As}]_{\text{feed}(t)}$ denote, respectively the arsenic concentrations at the time 0 and at an elapsed time, t , in the feed water compartment.

Two sets of experiments were performed. The first one with feed solutions contained $\text{Na}_2\text{HAsO}_4 \cdot 7\text{H}_2\text{O}$ and Na_2SO_4 , and a NaCl aqueous solution as the receiver. The experimental conditions are presented in Table 3, and the results obtained in Section 3.1. The

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