



Effect of physico-chemical parameters on inorganic arsenic removal from aqueous solution using a forward osmosis membrane



Priyanka Mondal^a, Nick Hermans^a, Anh Thi Kim Tran^{a,b}, Yang Zhang^c, Yanyan Fang^d, Xiaolin Wang^d, Bart Van der Bruggen^{a,*}

^aDepartment of Chemical Engineering, Process Engineering for Sustainable Systems (ProcESS), KU Leuven, W. de Croijlaan 46, B-3001 Leuven, Belgium

^bFaculty of Chemical and Food Technology, HCM University of Technical Education, Vietnam

^cSeparation and Conversion Technology, Flemish Institute for Technological Research (VITO), Boeretang 200, B-2400 Mol, Belgium

^dBeijing Key Laboratory of Membrane Materials and Engineering, Department of Chemical Engineering, Tsinghua University, Beijing, China

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ABSTRACT

The rejection of arsenic by a forward osmosis (FO) membrane, and the effects of relevant physico-chemical factors on the separation have been systematically investigated. MgSO₄ and glucose solutions were used as two potential draw solutions. More than 98% rejection was observed when the initial As(V) concentration was 500 μg L⁻¹, yielding an As concentration in the permeate below the maximum contamination level (MCL). It was demonstrated that the rejection of As was higher when the membrane active layer faces the feed solution (AL-FS) compared to the rejection when the membrane active layer faces the draw solution (AL-DS). However, for As(III), it was observed that the rejection was low at lower pH (3–12.6% within pH 3–7) and oxidation of As(III) at neutral pH increased the rejection to 95.7%. Thus, oxidation before FO is suggested as an essential pretreatment for total As removal in the neutral pH range. Therefore, FO is thought to have potential as a barrier for As removal in an integrated treatment system. However, recovery of the draw solutions by using reverse osmosis (RO), nanofiltration (NF) or membrane distillation for producing As-free water is one of the critical challenges that have to be overcome before implementation in field conditions.

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Introduction

Over the past decade, arsenic (As) contamination in groundwater has resulted in mass-poisoning of millions of people across the globe [1]. Several natural sources, e.g. arsenical minerals and sediments, as well as anthropogenic sources, e.g. mining, burning of fossil fuels, wood preservatives and application of arsenical fungicides, pesticides and herbicides are responsible for As poisoning of groundwater [2]. The oxyanions of As(V) [H₃AsO₄, H₂AsO₄¹⁻, HAsO₄²⁻] and As(III) [H₃AsO₃ and H₂AsO₃¹⁻] are the major species found in groundwater, while organo-As species are commonly observed in industrially polluted water [1]. The predominance of the As species depends on the pH and the redox conditions of the groundwater [3]. Since the consumption of As contaminated water is considered as the principle route of As poisoning [4], the maximum contamination level (MCL) of As in drinking water has been lowered to 10 μg L⁻¹ (previously 50 μg L⁻¹) by several national and international organizations including the WHO [5].

Currently there are various removal technologies that can be applied for the removal of As from aqueous solutions. These include oxidation, coagulation–flocculation, adsorption, ion exchange, and membrane technology [6]. Recently innovative carbon based adsorbents have been synthesized (zerovalent iron doped multiwall carbon nanotube and powdered chicken feather) that can directly remove As(III) considerably [7,8] but always certain conditions need to be maintained prior to the application of these adsorbents. Moreover, it was shown that these technologies alone are not (always) sufficient to remove As to a concentration below the MCL, because of several drawbacks and limitations [6]. For example, toxic chemicals and carcinogens are produced as by-products during oxidation and a large dosage is required for coagulation–flocculation. Interferences from competitive anions and regeneration of adsorbents for multiple uses are disadvantages for adsorption. Solid clogging is an important drawback for ion exchange together with competitive ion interferences. Moreover, environmental recontamination of As from the toxic waste of the above processes needs to be considered [6]. Although membrane technologies (RO and NF) have some advantages over the above technologies such as the high As removal efficiency and lower production of toxic solid waste, the removal efficiency is very low when contaminated water contains a high concentration of

* Corresponding author.

E-mail address: bart.vanderbruggen@cit.kuleuven.be (B. Van der Bruggen).

As. Therefore, developing new technologies to provide As-free water to the society has become a great challenge for the scientific community.

In the last decade, forward osmosis (FO) has gained considerable interest due to its low energy demand [9], high rejection efficiency, low membrane fouling, lower pressure requirements and thus, simple equipment set up compared to pressure driven membrane filtration [10,11]. In this process, due to osmosis, relatively pure water passes across the semi-permeable membrane from a region of lower osmotic pressure to a higher osmotic pressure and as a result, separation of solutes takes place [10]. FO membranes have not only been used extensively for desalination of water [11–14] but have also been successful laboratory-scale treatment of waste water such as domestic wastewater [15], landfill leachate [16], activated sludge [17], hydrophobic trace organic contaminants removal [18] and boron removal [19]. However, selection of a proper draw solution and its recovery to produce fresh water are critical issues that need to be overcome. The important criteria for a proper draw solution are to (i) have a high osmotic pressure, (ii) have minimal internal concentration polarization (ICP) and (iii) be easily separated from product water. ICP plays an important role in FO as it decreases the reverse salt flux and water flux by decreasing the draw solute concentration at the support layer-active layer interface. The specific reverse solute flux (ratio of reverse solute flux to forward water flux) is an important parameter to evaluate the performance of FO membranes. The decrease in specific reverse solute flux leads to higher membrane selectivity that simultaneously increases FO membrane performance [11]. However, this parameter is independent on the concentration of the draw solution and the membrane support layer structure but is determined by the selectivity of the active layer of the membrane [20]. A draw solution with higher diffusion coefficient and lower molecular size can minimize ICP but simultaneously increases the reverse draw solute flux, which is considered an important disadvantage of the FO process [21]. The performance of an FO membrane decreases with increasing draw solute flux. Similarly, the separation of the draw solution from the product water also needs to be considered. Therefore, a draw solution should be selected depending on the specific application and the characteristics of the membranes. Several innovative draw solutions have been used to respond to these requirements, such as magnetic nanoparticles [22], hydrophilic magnetic nanoparticles [23], polymeric hydrogels [24] and organic compounds [25,26]. These draw solutions can be recovered by applying a magnetic field [22], a canister separator [25], by deswelling [24], heating [27] or by using ultrafiltration for solutes with large particle size [23].

In a recent study, Jin et al. [28] investigated the effect of organic fouling and membrane orientation on the removal of As(III) and boron in laboratory-scale crossflow FO membrane filtration experiments. In optimized conditions, about 60% rejection of As(III) from aqueous solution was obtained. Recently, Butler et al. [29] reported 88.3% removal of As(V) at concentration of $10 \mu\text{g L}^{-1}$ from feed water. However, the performance of FO membranes in removing the various As species from aqueous solution in the presence of different environmental conditions, has not yet been reported. The various factors and related mechanisms that control the removal of As by FO membranes need to be elucidated for a better understanding of the separation mechanisms.

The aim of the present study is to provide new insights in the use of FO membranes as a barrier for As species in aqueous solution. The separation of As(V) was investigated via a commercially available FO membrane using either MgSO_4 or glucose as the draw solution. The effect of several physico-chemical parameters, e.g. membrane orientation, the initial As concentration, and the draw solution concentration on the rejection of As was extensively studied. The removal of As and the effect of pH on the removal of dominant As species [As(V) and As(III)] were also examined, to determine the efficiency

of FO for total As removal from water. The effect of above parameters on As separation and water flux was elucidated with the help of solute diffusion coefficients, membrane pore radius, hydrated radius, stoke radius along with previously established mathematical expressions. Such an elucidation of these parameters on As(V) removal by using MgSO_4 and glucose as draw solution has not yet been reported to the best of our knowledge. Lastly, oxidation experiment was also performed to improve total As removal which simultaneously ascertained the feasibility of hybrid/integrated treatment technology.

Materials and methods

Chemicals

The draw solutions were prepared by dissolving either MgSO_4 (Chem Lab NV, Belgium) or glucose (Fischer Chemical, Fair Lawn, New Jersey) in de-ionized water (Millipore Milli-Q 18 M Ω). $\text{Na}_2\text{AsO}_4 \cdot 7\text{H}_2\text{O}$ (RPL, Belgium) and As_2O_3 (Riedel-de Haen, Germany) were used as a source of As(V) and As(III), respectively. HNO_3 1 M (Merck, Germany) and NaOH 1 M (Merck, Germany) solutions were used to adjust the pH of the solution and was determined by using a pH electrode (Orion pH meter model 420A). KMnO_4 (Merck, Germany) was used for the oxidation of As(III). All the chemicals were of analytical grade and used without further treatment.

The molar concentration of glucose and MgSO_4 was 0.5 M and 0.25 M respectively except the experiments related to effect of draw solution concentration. The required molar concentration of the draw solutions with respect to osmotic pressure difference can be approximated using Morse equation:

$$\pi = iMRT \quad (1)$$

where π is the generated osmotic pressure (bar), i is dimensionless Van't Hoff factor, M is the molar concentration of the draw solute (mol L^{-1}), R is the ideal gas constant ($8.314 \times 10^{-2} \text{ L bar K}^{-1} \text{ mol}^{-1}$) and T is the absolute temperature (K).

For simplicity the ideal Van't Hoff factors is assumed to be valid in the tested concentration range. For MgSO_4 the ideal Van't Hoff factor is two (assuming full ionization), while for glucose (does not ionize in water) it is one. Therefore, the molar concentration for glucose (0.5 M) was chosen to be two times higher than MgSO_4 (0.25 M) to generate the same osmotic pressure.

Forward osmosis membrane and membrane characterization

The forward osmosis membrane used in this investigation was provided by Hydration Technology Innovation (HTI, Scottsdale, AZ). The membrane is asymmetric and consists of a cellulose triacetate active layer with an embedded polyester mesh as the mechanical support. The active layer is dense and moderately hydrophobic. The detailed characteristics of such FO membrane have been discussed extensively in other studies [30,31].

The contact angle measurement of the membrane surface was performed by using a contact angle goniometer (DSA 10 Mk2, Krüss, Germany). The equilibrium contact angle was measured using a standard sessile drop method. The average value of the contact angles on both sides of the membrane (active and passive layer) was taken based on ten measurements. The membrane was submerged into water and later dried in desiccators prior to measuring the contact angle. The measured contact angles for the active layer and support layer were $67.15 \pm 0.9^\circ$ and $78.52 \pm 2.1^\circ$, respectively.

Forward osmosis laboratory system

A laboratory scale plate and frame module was used as the FO system. The membrane cell used during the experiment had a dimension of $8 \text{ cm} \times 8 \text{ cm} \times 0.05 \text{ cm}$ (length \times width \times depth). The

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