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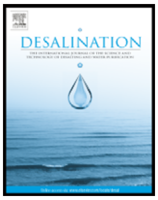

Performance of nanofiltration membrane in a vibrating module (VSEP-NF) for arsenic removal

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ABSTRACT

This study investigated the performance of a vibratory shear enhanced process (VSEP) using a nanofiltration (NF) membrane system for the removal of arsenic from drinking water as a function of retentate arsenate concentration, feed water composition, transmembrane pressure (TMP), vibration amplitude and pH. Two commercially available NF membranes having different pore sizes were used to compare their relative efficacy for arsenic removal. The relationship between arsenic rejection and the effects of operating conditions was systematically elucidated. Results reported here indicate that the removal of As(V) by the UTC-70 membrane was influenced slightly with the variation in operating conditions, whereas the removal of As(V) by the NTR-7450 membrane was strongly dependent on retentate arsenate, feed water composition, TMP, vibration amplitude and pH. The removal of As(III) by both NF membranes increased significantly with increasing TMPs, vibration amplitudes i.e. shear rates, and pH. In general, the removal of As(V) was higher than the removal of As(III) for all the conditions examined, signifying that the oxidation state appears to play a significant role in the arsenic separation behavior. In addition, the reported results suggested that the effects of operating conditions on the rejection of arsenic are also pore size dependent. Operation of the system at different vibration amplitudes suggested the significance of enhanced shear rates on the stable performance of the NF membranes for arsenic rejection.

1. Introduction

The occurrence of arsenic levels far above the drinking water standard has been reported in different places of the world. Arsenic in natural water primarily occurs in the forms of As(V) and As(III), known as Arsenate and Arsenite respectively. As(V) predominates in oxic surface water and As(III) predominates in anoxic ground water [1,2]. Arsenic is well known for its high toxicity and carcinogenic properties. Recent epidemiological studies have revealed the greater risk of cancer from the ingestion of arsenic than previously suspected [3]. As a result, the World Health Organization (WHO) has lowered the maximum permissible level for arsenic in drinking water from 50 µg/L to 10 µg/L. Therefore, research into methods of arsenic removal has been increased for the development of appropriate removal technologies. Of the available arsenic removal technologies, the membrane filtration process has emerged as a promising new route for high quality water purification due to its uniqueness over the conventional treatment processes [4]. The presence of arsenic in the form of $HAsO_4^{2-}$ in natural surface water has attracted the application of NF membranes, which are well suited for the removal of divalent ions. Recent developments in membrane technology have produced NF membranes with superior selectivity and increased water flux at much lower operating pressures. Arsenic removal by NF membranes is dependent on a number of factors i.e., pore size distribution, membrane morphology, surface charge, membrane material, module configuration and operating conditions, including solution concentration, operating pressure and solution pH. However, NF membranes are more prone to concentration polarization and irreversible fouling especially during the filtration of water containing organic substances. The presence of a fouling layer can drastically alter the characteristics of the membrane surface including surface charge and hydrophobicity [5–7]. Consequently, in addition to a decline in permeate flux, fouling can lead to considerable variation in the membrane separation efficiency [1,8,9]. Several researchers [4,10] have suggested that a pretreatment step that prevents colloidal and natural organic matter (NOM) fouling would improve the performance of membrane filtration for arsenic rejection. Nonetheless, to date, there have been no investigations that have tested this hypothesis. In recent years, VSEP technology has been used successfully to treat extremely challenging feed solutions e.g. waste waters as well as landfill leachates [11,12]. In all cases higher vibration amplitude resulted in higher permeate flux and higher solute rejection. The application of

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VSEP to treat potable water sources or other high quality water has been investigated much less extensively. In this work, a VSEP-NF membrane system is employed to remove arsenic from drinking water. Unlike the conventional cross flow membrane filtration process, this system is capable of producing intense shear force (1.35×10^5 per second) at the membrane surface by the torsional oscillation of the membrane. Thus it thins the concentration polarization layer formed at the membrane surface and improves the solute rejection through increasing solute back transport velocity [13–15]. Understanding the impacts of operating variables on rejection of arsenic species by NF membranes is of paramount importance from design as well as operational points of view. The consequence that results from the variation of the retentate As(V) concentration, feed water composition, TMP, vibration amplitude and pH on the removal of arsenic from river water by a VSEP-NF system is not well understood yet. In the open literature, there is currently little information with respect to the influence of operating conditions on the removal of arsenic by VSEP-NF systems. An understanding of the VSEP-NF process for arsenic removal can reveal the bottlenecks and the range of applicability of the system.

This paper aimed to:

(I) investigate the influence of retentate As(V) concentration and extended operation on the removal of arsenic from drinking water by a VSEP-NF system.
(II) compare the effect of feed water compositions on the As(V) removal and the As(V) concentration factor.
(III) determine the effectiveness of a VSEP-NF system for arsenic removal under various operating conditions such as TMP, vibration amplitudes and pH.

Experiments were conducted with two commercially available NF membranes. Arsenic removal was related to the membrane and arsenic species characteristics under different operating conditions. On the basis of the results, the effects of membrane operating conditions on arsenic removal were delineated and relevant mechanisms were elucidated.

2. Materials and methods

2.1. NF membrane

Two annular type NF (denoted as NTR-7450 and UTC-70) membranes were employed in this investigation. Of the membranes, NTR-7450 was supplied by Nitto Denko Co. Ltd., Japan and UTC-70 was supplied by Toray Industry Ltd., Japan. According to the manufacturer's recommendation, NTR-7450 is a sulfonated polyethersulfone and UTC-70 is a polypiperazine-amide membrane with an area of 0.045 m^2. The nominal NaCl rejections of NTR-7450 and UTC-70 have been reported to be 50% and 99.52% (1500 mg/L NaCl solution at 0.75 MPa) respectively. NTR-7450 is a loose NF membrane designed to provide removal of divalent ions, and total organic carbon (TOC). UTC-70 is a tight NF membrane designed to provide removal of both divalent and some monovalent ions. These two NF membranes have been chosen as they represent an array of filtration capabilities and energy requirement.

2.2. Chemicals and reagents

All stock solutions were prepared using reagent grade chemicals dissolved in de-ionized water. The arsenite, As(III), standard was prepared from solid standard arsenic trioxide, As_2O_3 (Wako Pure Chemical Industries, Ltd., Japan) dissolved in de-ionized water, pH 14.0 with sodium hydroxide. The Arsenate, As(V), was prepared from sodium salt heptahydrate, $Na_2HAsO_4.7H_2O$ (Wako Pure Chemical Industries, Ltd., Japan) dissolved in de-ionized water having a resistance of approx. 18 MΩ cm and filtered through a 0.22 μm membrane filter prior to use.

2.3. VSEP-NF membrane system

A small pilot scale VSEP-NF system as shown in Fig. 1 was used to treat arsenic contaminated water. Feed water containing arsenic was held in a 300 L feed tank, which was pumped to the VSEP unit by the feed pump. The temperature of the re-circulating feed water was also maintained at an approximately constant level (20 ± 2) °C by means of a thermostatically controlled cooling coil connected to a refrigeration unit. The VSEP apparatus is composed of four main components, i.e., a driving system that generates vibration, a membrane module (Filter pack), a torsion spring which transfers vibration to the membrane module and a system for controlling vibration. It can vibrate the membrane with a frequency of 60.75 Hz and maximum amplitude of 25 mm. The generated vibration is transmitted to the filter pack through the torsion spring by the mechanism of torsional oscillation waves. The strong shear forces on the membrane surface resulting from this vibration increases the back transport of the solute, hence improving its rejection rate [13–15]. The membrane filtration system was controlled by a programmable logic controller (PLC). The process conditions were monitored using on-line flow, pressure and temperature sensors. The membrane operating conditions were logged by the PLC every 5 s.

2.4. Experimental protocol

This study comprised of four main experimental parts: Part I consists of four NF test runs to evaluate the effect of retentate As(V) concentration and filtration time on the removal of arsenate. Part II included the four experimental runs to study the influence of feed water compositions on the Concentration Factor (CF) of As(V). Part III included the evaluation of the influence of source water compositions on the removal of As(V). Part IV included the evaluation of the effects of operating conditions e.g. TMP, vibration amplitudes and pH on the removal of As(III) and As(V).

(I) To evaluate the effect of retentate arsenate concentration and filtration time on the removal of As(V) by the two NF membranes, four experimental runs were conducted in the As(V) spiked Chitose river water. Two test runs for each membrane were conducted under vibration and non-vibration mode respectively. The feed water was spiked with 50 μg/L As (V). The TMP and the vibration amplitude were maintained at 310 kPa and 13 mm respectively.
(II) To investigate the influence of source water compositions on the concentration of arsenate by NTR-7450 and UTC-70 membranes, four experimental runs were conducted in the As(V) spiked Chitose river water and tap water at Kami Ebetsue water treatment plant in Japan. Run 1 and Run 2 were conducted with the NTR-7450 membrane in the As(V) spiked tap and river water respectively. Run 3 and Run 4 were conducted with the UTC-70 membrane in the As(V) spiked tap and river water respectively. The TMP and vibration amplitude were maintained at 310 kPa and 13 mm respectively. A 300 L feed tank once filled with the required amount of water was spiked to the desired arsenic level of 50 μg/L As(V). During the course of experimentation (Runs 1–4), only retentate was recycled to the feed tank. Each of the NF test runs continued for about 200 h. Chemical cleaning was done prior to each change in the following experimental run.
(III) To evaluate the effect of source water compositions on the removal of As(V), four experimental runs with both NF membranes were conducted in the Chitose river water and tap water respectively. The retentate and permeate were recycled to the feed tank that contained 50 μg/L As(V). The TMP and vibration amplitude were maintained at 560 kPa and 13 mm respectively.
(IV) In order to examine the influence of operating conditions such as TMP, vibration amplitude and pH on the removal of arsenic