

Arsenic removal from Water by Nanofiltration Membrane

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1. Introduction

Elevated arsenic levels can cause skin cancer and internal malignancies after continued exposure for 10–30 years. Berg et al. (2001) firstly issued a publication on arsenic contamination of the Red River alluvial tract in Hanoi, capital city of Vietnam. After treatment tap water generated from all treatment plants contained more than $10 \mu\text{g l}^{-1}$ of arsenic. In 1993, the World Health Organization (WHO) recommended a maximum arsenic concentration of 10 ppb as a provisional guideline. In 2002, Vietnamese government adopted this guideline as Vietnamese arsenic drinking water standard (TCVN, 2002). Among technologies for arsenic removal, Nanofiltration (NF) has been identified as a potential alternative for reverse osmosis (RO). The objective of this study is to investigate arsenic removal by a commercial NF membrane (NE90) with respect to performance and mechanism. The knowledge gained from this study will aid in the enhancement of arsenic removal in water treatment facility.

2. Materials and Methods

2.1. Solution chemistries

All salt solutions utilized in this research were prepared fresh using NaCl (Merck), CaCl₂ (Fluka), NaHCO₃ (Sigma) and Na₂SO₄ (Sigma). NaH₂AsO₄·7H₂O (Sigma) and As₂O₃ (Aldrich) were used to prepare 1 g l^{-1} arsenate (As(V)) and 1 g l^{-1} and arsenite (As(III)) stock solutions respectively.

2.2. Membrane and membrane test unit

A thin film composite (TFC) polyamide NF membrane (NE90) in the form of (6cm x 12cm) flat sheet was used. The membrane characteristics are presented elsewhere (Nguyen M. C., 2006). A bench-scale plate-and-frame single membrane filtration test unit was constructed for the NF studies. The membrane cell operated in cross-flow mode with feed flow rate of $400 \text{ cm}^3 \text{ min}^{-1}$ at 25°C; both permeate and retentate were recycled; pressure was varied from 96.5 to 386.0 kPa (20–80 psi).

2.3. Sample analysis

The separation of the individual NaCl, Na₂SO₄ and CaCl₂ salt solutions was evaluated from conductance measurements using conductivity meter (Thermo Orion 3 Star, USA). Arsenic concentration was analyzed by GF-AAS in comparison with ICP-MS for both permeate and feed samples.

Keywords : Arsenic; Arsenic removal; Nanofiltration membrane; Water

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3. Results and discussion

3.1. Individual salt rejection

CaCl₂, NaCl and Na₂SO₄ are usually utilized as model individual salts for investigation of the rejection performance of NF membrane. The characteristics of these salts as well as other solutes of consideration are listed elsewhere (Nguyen M. C., 2006). Observed order of rejection (Na₂SO₄ > NaCl > CaCl₂) is in agreement with the Donnan exclusion, electrostatic repulsion between membrane surface's charge and co-ions (Nguyen M. C., 2006). The highest rejection of Na₂SO₄ is attributed to the highest Donnan exclusion of negative bivalent ion (SO₄²⁻) while the lowest rejection of CaCl₂ is ascribed to the lowest Donnan exclusion of positive bivalent ion (Ca²⁺).

3.2. Arsenic removal performance and mechanism

With relatively high molecular weight, both As(III) and As(V) would be rejected from NF membrane by steric exclusion. Aside from this mechanism, as negatively charged ions, As(V) species would experience Donnan repulsion from negative NF membrane such as NE90.

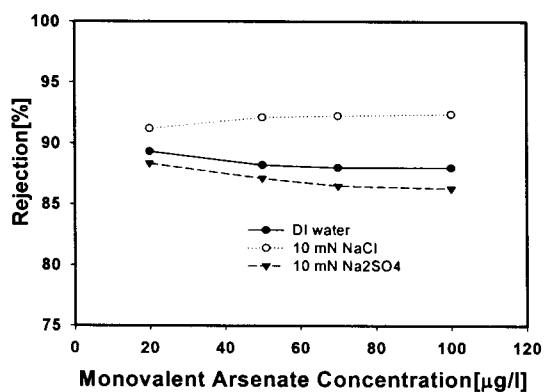


Fig. 1. Monovalent arsenate removal from DI water, 10 mN NaCl and 10 mN Na₂SO₄ solutions at pH 5.8, T 25°C, P 386 kPa, and flow rate of 0.4 L min⁻¹.

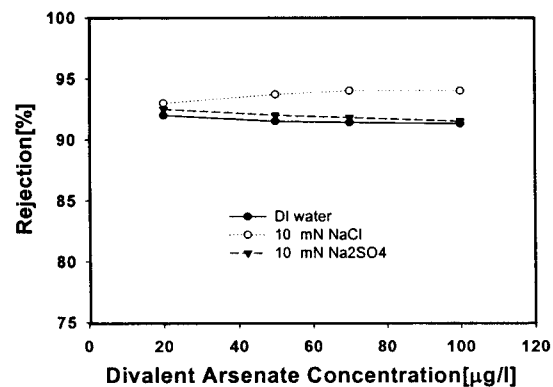


Fig. 2. Divalent arsenate removal from DI water, 10 mN NaCl and 10 mN Na₂SO₄ solutions at pH 8.0, T 25°C, P 386 kPa, and flow rate of 0.4 L min⁻¹.

3.2.1. Monovalent arsenate removal from DI water, NaCl solution, Na₂SO₄ solution

Figure 1 shows the removal of H₂AsO₄⁻ from DI water, 10 mN NaCl and 10 mN Na₂SO₄ solutions, respectively. The empirical rejection results ($R_{SO_4^{2-}} \approx 97\%$; $R_{H_2AsO_4^-} \approx 88\%$, Nguyen M. C., 2006) prove the predominance of the Donnan potential over steric effect i.e. SO₄²⁻, the divalent anion is rejected at higher rate compared to H₂AsO₄⁻, the monovalent anion regardless of their diffusivity order. However, the Donnan theory individually cannot account for effects of both NaCl and Na₂SO₄ on H₂AsO₄⁻ removal. Inside such confined and negatively charge pore matrix of the NE90 membrane, effective mobility order of co-ions Cl⁻, H₂AsO₄⁻ and SO₄²⁻ is evaluated as follow: Cl⁻ < H₂AsO₄⁻ < SO₄²⁻ (Nguyen M. C., 2006). Consequently the presence of more mobile co-ion, Cl⁻, enhances retention of

H_2AsO_4^- while the less mobile co-ion, SO_4^{2-} , worsens H_2AsO_4^- separation (Preferential passage phenomenon, postulated by Hodgson, 1970; Vrijenhoek and Waypa, 2000; Childress and Elimelech, 2000).

3.2.2. Bivalent arsenate removal from DI water, NaCl solution, Na_2SO_4 solution

At pH 8.0 As(V) predominantly exists as HAsO_4^{2-} (Brookins, 1988). The individual rejection of HAsO_4^{2-} is lower than that of SO_4^{2-} ($R_{\text{SO}_4^{2-}} \approx 97\%$; $R_{\text{HAsO}_4^{2-}} \approx 91\%$, Nguyen M. C., 2006). This phenomenon was also observed by Vrijenhoek and Waypa (2000). The presence of Cl^- , more mobile ion, not only improves the arsenic removal but also reverses the effect of initial HAsO_4^{2-} concentration on the arsenic separation i.e., arsenic separation increases with increasing initial HAsO_4^{2-} concentration (see Figure 2). Vrijenhoek and Waypa (2000) also reported the same phenomenon as investigating effect of Cl^- on As(V) removal in the presence of NaHCO_3 .

3.2.3. Membrane capacity for arsenic removal from synthetic water

The rejection of As(V) from synthetic water (1 mN NaHCO_3 + 10 mN NaCl) increased from 89% to 96% with increasing As(V) concentration from $20 \mu\text{g l}^{-1}$ to $100 \mu\text{g l}^{-1}$. At pH 7.0, As(V) exists in both forms H_2AsO_4^- and HAsO_4^{2-} which are, in consistence with the solute diffusivity order, less mobile than Cl^- and HCO_3^- (Nguyen M. C., 2006). Therefore the presence of Cl^- and HCO_3^- enhances As(V) removal. The As(III) removal decreased from 43% to 40% as the initial As(III) concentration increases from $20 \mu\text{g l}^{-1}$ to $100 \mu\text{g l}^{-1}$. At pH 7.0, As(III) prevalently occurs as neutral species H_3AsO_3 whose transport through the NE 90 membrane could be described well by Spiegler and Kedem model (Spiegler and Kedem, 1983). As the feed H_3AsO_3 concentration increases both diffusion and convection of the H_3AsO_3 species increase, in consequence the arsenic rejection decreases.

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