



Performance of nanofiltration and reverse osmosis membranes for arsenic removal from drinking water

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ABSTRACT

The removal of arsenic was investigated by four types of thin-film polyamide nanofiltration (NF) (NF270, NF90) and reverse osmosis (RO) (XLE, BW30) membranes in a flat-sheet module. The influence of membrane types, pressure, pH, and pre-oxidation step on the removal of arsenic (As(III)) was investigated. Initial As(III) concentration was 100 µg/l for all of the experiments. Flux was determined over a pressure range of 3.5–10 bar for both NF and RO membranes. Experiments were conducted at pH 3.5, 5, 7.5, and 10 to evaluate the effect of pH on As(III) removal. The impact of pre-oxidation and oxidant concentrations on As(III) removal were also evaluated. It was found that the percentage of As(III) removal of RO membranes were in the range of 97–99 for all transmembrane pressure applied. In the range of operating conditions, As(III) and As(V) rejection were found almost equally good by RO membranes. RO permeate met the WHO and Turkish standard for arsenic. Pre-oxidation step improved the rejection performance of NF. Nevertheless, NF permeate did not meet the standards in the range of operating conditions.

Keywords: Arsenite; Arsenic removal; Oxidation; Nanofiltration; Reverse osmosis

1. Introduction

Arsenic in ground and surface waters has been reported above the drinking water standards of different countries around the world. The literature shows that two oxidation states dominant in these water sources are arsenate, As(V), and arsenite, As(III), respectively. The ingestion of water-containing arsenic causes serious effects on human health, such as lung,

bladder, liver, renal, and skin cancers [1]. Considering the health effect of arsenic, the World Health Organization (WHO) established an allowable limit of 10 µg/l for arsenic in drinking water [2]. In Turkey, Republic of Turkish Ministry of Health set an allowable limit of 10 µg/l. In some regions of Turkey, arsenic concentration was found higher than the drinking water standards ranging from 10 to 7,754 µg/l [3,4]. Altas et al. determined arsenic concentration in 62 stations utilized as drinking water

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resources by the local community in Aksaray, Turkey. In this study, arsenic concentrations were found to be ranging between 10 and 50 $\mu\text{g}/\text{l}$ in 22 stations and $>50 \mu\text{g}/\text{l}$ in five stations [4].

A number of treatment technologies, such as ion exchange, adsorption [5–8], coagulation/flocculation [3,9], lime softening and membrane processes [10–15] have been investigated to remove arsenate and arsenite from drinking waters. Membrane processes, such as nanofiltration and reverse osmosis, can be considered promising technologies for arsenic removal in terms of high efficiency, easy operation, high effluent water quality, modularity, and flexibility. Up to now, various research studies have been performed to remove arsenic from drinking water by NF and RO membrane modules [12–20]. These studies were summarized in Table 1. It is clearly seen from Table 1 that operational conditions play an important role in determining the success of the processes. The change in the applied pressure, pH, form of arsenic, concentration of arsenic, and the existence of organic matter may have significant effects on rejection. As an interesting example, Oh et al. used RO membrane to remove arsenic from contaminated water applying low pressure through bicycle pump. High removal rates were achieved in this study [21].

Based on the developments in membrane technology, new thin-film polyamide negatively charged NF and RO membranes have been produced. These negatively charged membranes can ensure the highest degree of purification by means of size exclusion and electrostatic charge (Donnan exclusion) effects. All kinds of dissolved ions including arsenic can be effectively removed from water by NF and RO membranes which largely assume negative surface potential.

In this paper, the performances of negatively charged four flat-sheet NF and RO membranes were investigated with tap water spiked with arsenite. Commercial polyamide (NF90) and polypiperazine (NF270) NF and polyamide (XLE, BW30) RO membranes were chosen to evaluate the performances of membranes in terms of permeate flux and arsenic rejection as a function of transmembrane pressure and pH. It is known that membranes have higher removal efficiencies of As(V) than As(III) [15,16,18,22]. For this reason, a pre-oxidation step that oxidizes As(III) to As(V) were also incorporated in the study to improve the membrane performance. Hypochlorite was used as an oxidizing agent in the pre-oxidation step. The effect of pre-oxidation was also evaluated relevant to hypochlorite concentration.

2. Materials and methods

2.1. Standards and reagents

The stock solution, 1,000 mg/L As(III) was prepared dissolving appropriate amount of analytical grade NaAsO_2 (3.75 g/l as As) (Merck, Germany; 99% purity) in deionized (DI) water (MilliQ A10, Millipore). Appropriate amount of As(III) added to tap water to obtain As(III) concentration of 100 $\mu\text{g}/\text{l}$. The pH of the solution was adjusted to 3.5, 5, 7.5, and 10 using hydrochloric acid (HCl) and sodium hydroxide (NaOH). Pre-oxidation tests were conducted with sodium hypochlorite (6–14% active chlorine) (Merck, Germany) solution. Table 2 shows the characteristics of the tap water.

2.2. Experiments

Four type commercial flat-sheet thin-film NF (NF270, NF90) and RO membranes (XLE, BW30) were supplied by Dow-Filmtec. Table 3 summarizes the characteristics of the membranes used in this study [23–28]. The nominal MgSO_4 rejections of NF270 and NF90 have been reported to be 97 and $>97\%$, respectively, according to the manufacturer's specification. NF270 is designed to remove high percentage of TOC, divalent ions, and color, while NF90 is a tighter membrane designed to remove high percentages of salts, nitrate, iron, and organic compounds. Low-energy XLE membrane is designed for lowest pressure brackish water RO originally. The XLE membrane provides removal of both monovalent and divalent ions [23,29].

A laboratory-scale membrane module with a flat-sheet membrane cell (Sterlitech) was used in this study. The effective membrane area and operating volume were 14.6 cm^2 and 300 mL, respectively. System is capable to handle pressures up to 69 bar and maximum bear temperature at 55 bar is 121 °C. The membrane module was operated in dead-end mode. The experimental setup of the membrane module consisted of a nitrogen gas cylinder to maintain constant-pressure filtration, an external 2.5-L reservoir between the pressure source and the filtration cell, and a dead-end filtration cell with a capacity of 300 mL. Membrane module was placed on a magnetic stirrer for complete agitation of the solution during operation (Fig. 1).

Filtered water was weighted, and the data were collected using a personal computer to calculate flux. Weight was recorded at 30 s intervals by computer to observe of flux variation during filtration. Arsenic removal was studied under several conditions mainly to determine the removal of As(III) with direct filtration through NF (NF270, NF90) and RO membranes

Table 1
Arsenic removal studies by membrane

| Membrane type | Arsenic form | R_{NaCl} (%) or MWCO | Removal (%) | Refs. |
|--|-----------------------|------------------------|--|-------|
| Nitto NTR7450 | As(III):50 µg/l | 50% (0.75 MPa) | As(III):21.5, As(V):82.5 (P:1 MPa, pH 7.5) | [12] |
| Toray UTC70 | As(V):50 µg/l | 99.52% (0.75 MPa) | As(III):80.5, As(V):99.2 (P:1 MPa, pH 7.5) | [13] |
| NF90 | As(III)-(V):100 µg/l | 220–310 | As(III) ≈ 50, As(V) ≈ 90 (P:10 bar, pH 7) | [14] |
| NF70 | pre-oxidation (ozone) | 350 | As(III) ≈ 35, As(V) > 90 (P:10 bar) | [15] |
| NF40 | As(V):100–1,000 µg/l | 1,000 | As(III):15, As(V) > 90 | [14] |
| Dow-Filmtec NF90 | As(V):100–1,000 µg/l | 200–400 | As(V) > 94 (P:6 bar, pH 8) | [15] |
| Microdyn-Nadir N30F | As(III):20–90 µg/l | 400 | As(V):78 (P:6 bar, pH 8) | [16] |
| Toray NF | As(V):20–90 µg/l | N/A | As(III) < 10 | [17] |
| As(V):20–90 µg/l | As(V):90–100 | | | |
| As(III):62–152 µg/l pre-oxidation (KMnO ₄) | N/A | | As(III):63, As(V):98 (P:12 kgf/cm ² , pH 7) | [18] |
| As(III):75 µg/l | | | As(III):57, As(V):96 (P:12 kgf/cm ² , pH 7) | [19] |
| As(V):105 µg/l Preoxidation (KMnO ₄) | | | As(III):60, As(V):96 (P:12 kgf/cm ² , pH 7) | [20] |
| As(III):18.5 µg/l | | | As(V):>98 (P:16 kgf/cm ² , pH 7.2) | [21] |
| As(V):25.5 µg/l | | | | [22] |
| Filmtec NF70 | | N/A | As(III) ≈ 55, As(V) > 98 | [23] |
| Desal HL4040 | | 300 | As(III) ≈ 20, As(V) > 98 | [24] |
| Hydranautics 4040UHA | | 300 | As(III) ≈ 30, As(V) > 98 | [25] |
| Fluid Systems TFC4921 | | N/A | As(III) ≈ 60, As(V) > 95 | [26] |
| Fluid Systems TFC 4820 | | N/A | As(III) ≈ 75, As(V) > 98 | [27] |
| Desal AG4040 | | N/A | As(III) ≈ 70, As(V) > 98 | [28] |
| Hydranautics 4040 LSA | | N/A | As(III):87, As(V) > 98 | [29] |
| Filmtec NF270 | Total As:185–208 µg/l | – | Total As > 91 (P:698 kPa, pH 8.35) | [30] |
| Filmtec NFc | | | Total As > 85 (P:698 kPa, pH 8.35) | [31] |
| Hydranautics CPA2 | | | Total As > 99 (P:698 kPa, pH 8.35) | [32] |
| Desal HL4040 | As(III):50–400 µg/l | 300 | As(III) > 78 (P:0.55 MPa, pH 9.5) | [33] |
| Desal AK | | | | [34] |

Table 2
Tap water characteristics

| Parameters | Values | Parameters | Values |
|--------------------------------------|--------|-----------------------|--------|
| SO ₄ ²⁻ (mg/l) | 120 | Alkalinity (mg/l) | 100 |
| PO ₄ ³⁻ (mg/l) | 0.16 | F ⁻ (mg/l) | 0.14 |
| NO ₃ ⁻ (mg/l) | 1.5 | pH | 7.5 |
| Cl ⁻ (mg/l) | 97 | Temperature (°C) | 21.7 |

(XLE, BW30). Initial As(III) concentration was 100 µg/l for all the experiments performed. All the experiments were carried out at a constant temperature of 20 ± 1 °C. Flux was determined over a pressure range of 3.5–10 bar for both NF and RO membranes. Experiments were conducted at pH 3.5, 5, 7.5, and 10 to evaluate the effect of pH on As(III) removal. Tests were conducted with sodium hypochlorite to determine the effect of pre-oxidation. The impact of oxidant concentrations on As(III) removal was also evaluated. Samples were subjected to duplicate arsenic analysis after filtration and pre-oxidation/filtration.

2.3. Analytics

Total arsenic was determined using Perkin-Elmer Analyst 400 graphite furnace atomic absorption spectrometer equipped with a graphite tube atomizer and programmable auto sampler. Argon gas of high purity

was used to purge the volatilized matrix materials and protect the heated graphite tube from air oxidation. Arsenic electrodeless discharge lamps were used at a wavelength of 193.7 nm with a slit width of 2.7 nm. Operating currents of electrodeless discharge lamps were 400 mA. Magnesium nitrate (Mg(NO₃)₂) and palladium (inorganic ventures) were selected as the matrix modifier for this study. As(V) was analyzed using WTW PhotoLab 6600 UV–vis Spectrophotometer according to Funing and Daren method [30].

Arsenic removal (%) was calculated from the arsenic concentration difference of feed water (C₀) and permeate (C) using the following equation:

$$\text{Arsenic removal (\%)} = [(C_0 - C)/C_0] \times 100 \quad (1)$$

3. Results and discussion

3.1. Effect of transmembrane pressure on flux and removal

The effect of transmembrane pressure on permeate flux was determined at 100 µg/l of initial As(III) concentration for all of the used NF and RO membranes. As expected, increase in transmembrane pressure caused an increase in steady-state flux for all of the used membranes (Fig. 2). When the transmembrane pressure increased from 3.5 to 10, flux increased from 18 to 66 and 15 to 36 L/m² h for NF270 and NF90, respectively. Flux increased from 11 to 24 L/m² h and 3 to 7 L/m² h with the increase in transmembrane

Table 3
Characteristics of the NF and RO membranes

| Membrane | NF270 | NF90 | XLE | BW30 |
|--|-----------------------|--------------------|--------------------|------------------|
| Manufactures | Dow/Filmtec | Dow/Filmtec | Dow/Filmtec | Dow/Filmtec |
| Material | Polypiperazine | Polyamide | Polyamide | Polyamide |
| Membrane type | Flat sheet | Flat sheet | Flat sheet | Flat sheet |
| Surface charge | Negative | Negative | Negative | Negative |
| Maximum operating temp. | 45 °C | 45 °C | 45 °C | 45 °C |
| pH range | 2–11 | 2–11 | 2–11 | 2–11 |
| MWCO | 200 | 150 | 100 | 100 |
| Contact angle | 10.00 | 50.29 | 69.29 | 51.16 |
| Zeta potential | –5 to –16 mV | –15 to –30 mV | –10 to –25 mV | –6 mV |
| NaCl rejection ^a | 50% | 85–95% | 98.6% | 99.4% |
| MgSO ₄ rejection ^a | 97% | >97% | 99.2% | 99.7% |
| Pure water permeability (Lm ² h ⁻¹ bar ⁻¹) | 5.1–11.5 ^b | 4–6.4 ^c | 2–5.7 ^d | 2–3 ^e |

^aPermeate flow and salt rejection based on the following test conditions: 2,000 mg/l NaCl and MgSO₄, 70 psi (0.48 MPa), 77 °F (25 °C), and 15% recovery [23].

^bPure water permeability values obtained from Ref. [24–26].

^cPure water permeability values obtained from Refs. [24,25,27].

^dPure water permeability values obtained from Refs. [24,25].

^ePure water permeability values obtained from Refs. [24,28].

Note: Characteristics given without superscript obtained from manufacturer.

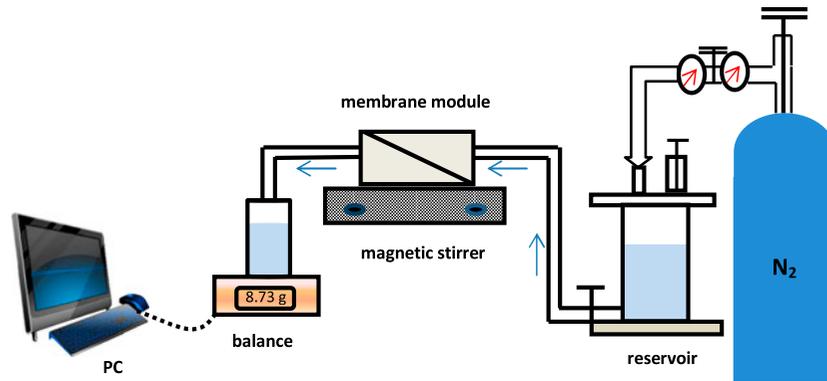


Fig. 1. Experimental setup of membrane module.

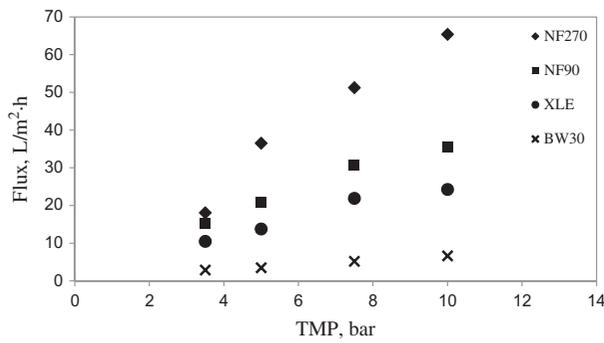


Fig. 2. Effect of transmembrane pressure on permeate flux (pH 7.5; operating temperature: 20°C).

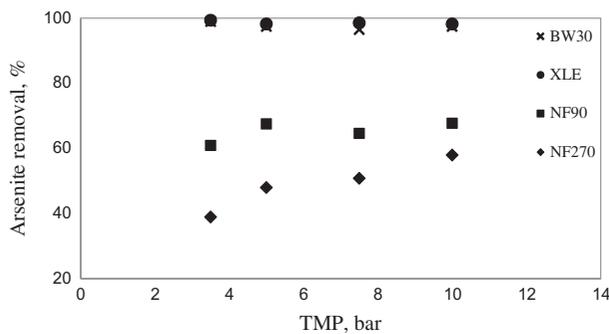


Fig. 3. Effect of transmembrane pressure on the removal of As(III).

pressure from 3.5 to 10 bar for XLE and BW30 membranes, respectively. NF270 gave the highest flux, depending on their polypiperazine structure. Additionally, NF90 is a tighter membrane than NF270. This is another reason for the higher flux obtained by NF270.

The effect of transmembrane pressure on the removal of arsenic was determined for all of the used

NF and RO membranes under the following conditions: Initial As(III) concentration = 100 µg/l, pH 7.5, 20°C. Transmembrane pressure was varied from 3.5 to 10 bar. Fig. 3 shows that As(III) rejection of RO membranes (XLE and BW30) were significantly higher than that of NF membranes. The percentage of As(III) removal of these RO membranes varied from 97 to 99% for all transmembrane pressure applied. Both RO permeate (XLE and BW30) met the WHO and Turkish standard for arsenic. Similarly, Teychene et al. achieved >99% removal of As(III) by BW30 at pH 7.6 [31].

Most of the arsenic is in the form of uncharged H_3AsO_3 at typical drinking water pH level (pH 6–9) and therefore less efficiently rejected. However, because selected RO membranes have a comparable pore size to that of molecular H_3AsO_3 , obtained higher rejection ratio of RO membrane at pH 7.5 in this study, could be explained by strong steric effect of RO. As(III) removal rate of NF membranes was not as sufficient as that of RO membranes. Nevertheless, arsenite rejection for NF90 and NF270 membranes increased slightly with increasing transmembrane

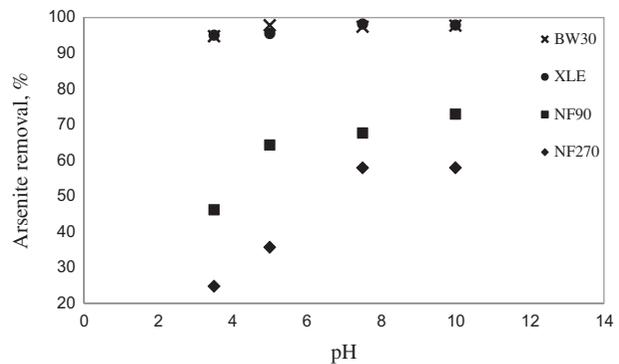


Fig. 4. Effect of pH on arsenite removal.

pressure. Removal percentage of As(III) with NF90 (>62%) was higher than that of NF270 (>40%) over the pressure range investigated based on lower molecular weight cutoff for NF90. But As concentration in the permeate was still significantly higher than that of WHO and Turkish standard for arsenic. Moore found similar result in his study using groundwater to remove 38–44 $\mu\text{g}/\text{l}$ of arsenic with XLE, NF90, and NF270 membrane [29]. But arsenic guideline value could not be achieved with NF or XLE membrane alone in the mentioned study in spite of low initial arsenic concentration.

3.2. Effect of pH on the removal of arsenite

The effect of pH on arsenite removal at 100 $\mu\text{g}/\text{l}$ of initial As(III) concentration for NF (NF270, NF90) and RO (XLE, BW30) membranes is shown in Fig. 4. Applied pressure and temperature were fixed at 10 bar and 20°C, respectively. As shown in Fig. 4, pH 7.5 gives the highest removal rate for all of the RO and NF membranes except NF90. At this pH, XLE and BW30 RO membranes gave an arsenic removal percentage of 98.23 and 97.47%, respectively.

RO membrane was abundant of surface negative charges. In fact, all the used membranes in this study are negatively charged and they become more negative as the pH value increases. Thus, it is expected that charge exclusion must play an important role beside steric hindrance. But charge exclusion had only a minor effect on the As(III) rejection for NF90 which is made of polyamide and did not have a significant effect on the As(III) rejection for NF270. The percentages of arsenic removal at pH 7.5 were 67.72 and 57.96 for NF90 and NF270, respectively. It is partially surprising because the molecular weight of As(III) was 125.94 g/mol (0.24 nm) which was higher than that of NF membranes [20].

Because As(III) exists as a neutral molecule at pH 5–8, removal of As(III) by membrane is expected to be unaffected by change in pH at this pH region. Arsenite is dissociable with the first $\text{p}K_a$ at 9.2. Above this pH, it is found that As(III) removal increases with the increase in pH [20,32]. H_2AsO_3^- is the dominant species in solution at $\text{pH} > 9.2$. The increase in As(III) removal by NF270 is considerably lower than NF90 with increase in pH beyond pH 7.5. This phenomenon can be explained by lower monovalent ion retention rate of NF270 than NF90. Kosutic et al. also revealed only a minor increase in the removal rate of NF270 with the increase in pH. Findings of the present research are similar to those of the research conducted by Kosutic et al. [19].

3.3. Effect of pre-oxidation by hypochlorite

Pre-oxidation step were incorporated in the study in order to improve membrane performance. Initial As (III) concentration, applied pressure, pH, and temperature were fixed at 100 $\mu\text{g}/\text{l}$, 10 bar, 7.5, and 20°C, respectively. Hypochlorite was used as an oxidizing agent in the pre-oxidation step. Stoichiometrically, approximately 100 $\mu\text{g}/\text{l}$ hypochlorite is required for the complete oxidation of 100 $\mu\text{g}/\text{l}$ As(III). It was found in our unpublished study that, when NaClO was added in stoichiometric amount, the oxidation performance of 100 $\mu\text{g}/\text{l}$ As(III) was found as 19%. At least 10 times higher hypochlorite concentrations than the stoichiometric amount were required for complete oxidation. This must be due to the presence of some reducing agents in tap water and/or use of only a portion of chlorite in the oxidation process. Similar results have been also reported by Sorlini and Gialdini [2]. They found that when the ground water was spiked with As(III) to obtain initial As(III) concentration of 50 $\mu\text{g}/\text{l}$ and when NaClO was added 70 times higher than the stoichiometric requirement, 95% oxidation was achieved within 5 min. In the present study, it is found that all of the As(III) was oxidized to As(V) at the dose of 1 mg/l or higher hypochlorite. Based on this finding, oxidant concentrations were varied from 1 to 10 mg/l. The effect of pre-oxidation relevant to hypochlorite concentration was shown in Fig. 5.

According to the results, it is reasonable to conclude that the As(III) and As(V) removal performance of RO membranes were almost equally in the range of operating conditions. Oxidation of As(III) to As(V) had negligible effect on the performance of RO membranes, but had a significant effect on the performance of NF membranes. The percentage of As(III) removal of XLE and BW30 membrane were found as high as 98.23 and 97.47%, respectively. Total arsenic rejection

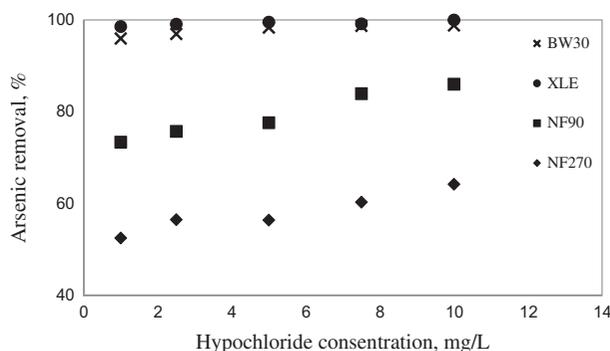


Fig. 5. The effect of pre-oxidation step on arsenic removal.

reached 100% with the hypochlorite dose of 10 mg/l. Hering and Elimelech (1996) also found that the removal performances of RO membranes are about equal for As(III) and As(V) [33]. With the same dose of hypochlorite, the percentage of As(V) rejection reached 86 and 64% for NF90 and NF270, respectively. The performance of NF90 was found better than NF270. Despite this improvement, arsenic concentration of the permeate was still higher than allowable limit of WHO and Turkish standard for NF90 and NF270.

It must be pointed out that the molecular weight of As(V) is 140.94 g/mol which is higher than As(III) and selected NF membranes. But, it must be emphasized also that in the range of pH 4–6, dominant forms of arsenate is monovalent H_2AsO_4^- ; while above pH 7, divalent HAsO_4^{2-} is the dominant form. Consequently, significant increase in arsenic rejection by NF90 membrane can be explained by its high salt rejection rate (both monovalent (NaCl) and divalent (MgSO_4)). On the other hand, although similar rejection rates for divalent salts (MgSO_4) were obtained by polypiperazine NF270 membrane, arsenic rejection rate of NF270 membrane did not increase significantly. A minor increase obtained in removal rate of As(V) compared to As(III) by NF270 can be explained by better but inadequate removal performance of NF270 for divalent ions than uncharged H_3AsO_3 ion due to its relatively high molecular weight cutoff in comparison with the NF90. In terms of As(V), charge exclusion seems to play a role for NF90 membrane beside steric effect.

4. Conclusions

The present study considered the effect of membrane types, pressure, pH, and pre-oxidation step on the removal of arsenic from water. The removal of arsenic was investigated by four types of negatively charged thin-film composite NF (NF270, NF90) and RO (XLE, BW30) membranes. The results of the study may be summarized as follows.

As(III) rejection of RO membranes was significantly higher than that of NF membranes. The percentage of As(III) removal of RO membranes varied from 97 to 99% for all transmembrane pressure applied. Applied pressure of 4 bar may be sufficient to achieve high As(III) removal rate. In any case, RO permeate met the WHO and Turkish standard for arsenic. As the selected RO membranes have a comparable pore size to that of H_3AsO_3 , obtained higher As(III) rejection ratio of RO at pH 7.5 in this study, could be explained by strong steric effect of RO membranes. As(III)

removal rate of NF membranes was not as good as RO membranes. Rejection of As(III) for NF90 and NF270 membranes increased slightly with increasing transmembrane pressure. But As(III) concentration in the permeate was still significantly higher than that of WHO and Turkish standard for arsenic. In contrast to expectations, beyond pH 9, charge exclusion had only minor effect on the As(III) rejection for polyamide NF90 membrane. There were no significant effects on the As(III) rejection for polypiperazine NF270 membrane. This phenomenon can be explained by lower monovalent ion retention rate of NF270 than NF90.

In the range of operating conditions, As(III) and As(V) rejection were found almost equally good by RO membranes. The percentage of As(III) removal of XLE and BW 30 membranes were as high as 98.23 and 97.47%, respectively. Arsenic rejection reached 100% with the hypochlorite dose of 10 mg/l. Oxidation of As(III) to As(V) had negligible effect on the performance of RO membranes, but had a significant improvement, especially on the performance of NF90 membrane. This improvement depends on divalent HAsO_4^{2-} formation followed by oxidation. It must be pointed out that arsenic removal was relatively low for NF270 despite reported high divalent salt rejection rate according to the manufacturer's specifications. The percentage of arsenite removal was found as 67.72 and 57.96 for NF90 and NF270, respectively; while with the dose of 10 mg/l of hypochlorite, the percentage of arsenic rejection reached 86 and 64% for NF90 and NF270, respectively. Despite this improvement, the arsenic concentration of the permeate is still higher than the allowable limit of WHO and Turkish standard for NF90 and NF270.

It is concluded that charge exclusion cannot be sufficient to remove arsenic from water by negatively charged thin-film composite NF membrane. Different membrane materials and different oxidation techniques that will not harm the membrane should be tried further.

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