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Effects of Operating Conditions on the Performance of NF270 and TW30 Membranes During As (III) Removal

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Abstract The influence of operating conditions on the performance of nanofiltration (NF270) and reverse osmosis (TW30) membranes for arsenite [As (III)] removal was studied. This study aimed at following the evolution of membrane performance, especially, solute rejection and water flux with the change of operating parameters. The operating parameters as applied pressure, recovery, feed pH, feed As (III) concentration and ionic strength were considered. As (III) rejection and water flux were determined at each operating condition. Results show that increasing applied pressure contributes to improve As (III) rejection and water flux through the membrane. As (III) rejection increased from 40.2 to 71.1% and water flux from 11.65 to 38.59 $Lh^{-1}m^{-2}$ when applied pressure changed from 2 to 6 bar with TW30 membrane. In the same sense, an increase of pH at a value of 10 considerably improves As (III) removal whereas water flux was slightly impacted. On the other hand, an increase of recovery and feed As (III) concentration leads to the decrease of the system performance in term of As (III) rejection. As for ionic strength, its increase does not affect As (III) rejection but can contribute to the decrease of water flux. It was put in evidence that applied pressure and feed pH are the most important parameters which must be considered if As (III) removal wants to be improved.

Keywords: *As (III) rejection, nanofiltration, reverse osmosis, operating parameters*

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1. Introduction

Natural water pollution by arsenic is a worldwide issue and has become a challenge for engineers, scientists and for decision-makers of various countries. Indeed, arsenic can be found in high concentrations in both surface water and groundwater usually used for human consumption [1]. It is a metalloid compound which can have both inorganic and organic forms [2]. This element is present in the environment in different oxidation states [As (V), As (III), As (0) and As (-III)] and forms various organic species. However, the two states prevalent in water are trivalent arsenic [arsenite, As (III)] and pentavalent arsenic [arsenate, As (V)]. Generally, As (V) is the dominant species of arsenic in surface water, whilst As (III) is dominant in groundwater [3]. Long term exposition to inorganic arsenic may cause a wide range of health effects [4,5]. In 1993, the World Health Organization (WHO) established 10 µg L^{-1} as a provisional guideline value for arsenic in drinking water. In 2001, the United States arsenic drinking water limit of 50 µg L^{-1} was lowered to 10 µg L^{-1}, the same as in the European Union. Above the limit of 10 µg L^{-1}, arsenic in drinking water has an estimated cancer risk of 1 in 500 while 1 in 10,000 is usually the highest cancer risk allowed by the United States Environmental Protection Agency in tap water [6]. For these reasons many technologies were developed to remove arsenic from water. Among various available technologies, nanofiltration (NF) and reverse osmosis (RO) are recommended as best technologies for safe drinking water production [7]. Studies have shown that these technologies can reach a rejection ratio of total arsenic of 99% [8,9,10]. However, the rejection ratio of As (V) is higher than the one of As (III) due to the predominance of the Donnan potential over a steric effect [11,12,13] whereas trivalent compounds are more toxic than the pentavalent ones [14]. Reference [15] found that the removal efficiencies were about 99% and 55% for As (V) and As (III), respectively. Thereby, the efficiency of NF and RO processes for water treatment is mainly defined by As (III) rejection. Otherwise, it is recognized that the operating parameters play an important role in the efficiency of NF and RO processes. For example, the feed pH controls the dissociation of As (III) and modifies the surface charge density of the membrane [16,17] and then has a strong impact on rejection by electrostatic interactions. The applied pressure, recovery, feed concentration and ionic strength can also influence the efficiency of the membrane [18,19,20,21]. Certainly, various studies were carried out onto the effect of operating parameters on the performance of NF and RO membranes for As (III) rejection, but, As (III) rejection differs from one membrane to another. Indeed, it can be noticed that, reference [22], for instance, found As (III) rejection of <20% with Filmtec NF45 membrane, whereas, a rejection of >99% was obtained by [23] with Filmtec SW30HR membrane. Moreover, reference [24] found a rejection of As (III) of 50% with Filmtec NF70-4040 membrane. So, it would be interesting to study the rejection of As (III) on any other membrane. This is the aim of this study which objective is to show the effect of operating parameters on the As (III) rejection and water flux of NF270 and TW30 membranes in order to determine and to optimize the most important parameters which enhance As (III) removal.

2. Materials and Methods

2.1. Feed Solution

A 1000 mg L^{-1} As (III) stock solution was obtained by dissolving appropriate quantity of arsenic (III) oxide (As_2O_3), purity 99.95 – 100.05% provided by Sigma Aldrich, in NaOH 1 N solution and neutralized by HNO_3 1 N. The mixture was completed with pure water 15 MΩ.cm resistivity. As (III) feed solution at different concentration, ranging from 50 µg L^{-1} to 150 µg L^{-1}, was prepared by diluting stock solution with pure water. The pH of feed solution was adjusted with HNO_3 1 N or NaOH 1 N.

2.2. Membrane Unit and Procedures

Two commercial membranes were used in this study: one nanoflitration membrane (NF270) and one reverse osmosis membrane (TW30). Both membranes are thin film composite polyamide membrane provided by Dow Chemical. These membranes were mounted on two parallel spiral modules of the pilot unit provided by Deltalab/Cosimi (Figure 1).

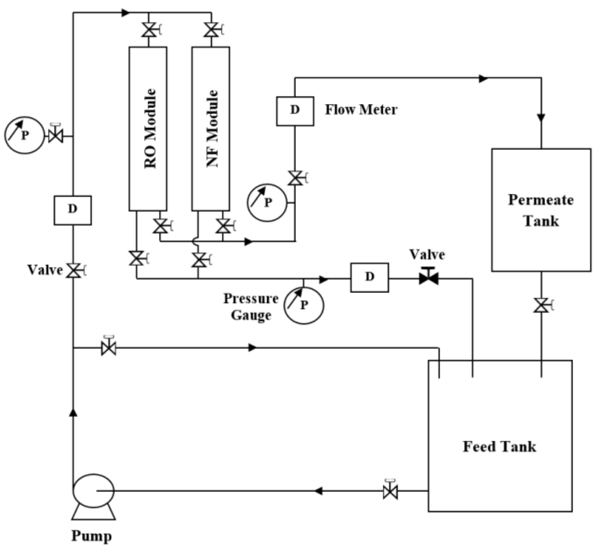

Figure 1. Schematic diagram of the pilot unit

The active membrane surface was 2.6 m². The pilot unit is made up of, apart from membrane modules, feed tank and permeate tank of capacity respectively 100 L and 20 L, high pressure pump (LOWARA 3SV19F022M) and three pressure gauges. The feed and permeate flow rates were measured at the inlet and outlet of membrane module by two flow meters (KROHNE H250/RR/M9/ESK) with an accuracy of 1.6 % of measured value. All filtration experiments were carried out in recirculation mode (i.e. both concentrate and permeate were returned back to the feed container) with an initial volume of 20 L. The applied pressure was adjusted using the concentrate outlet valve. The effect of applied pressure, recovery, feed pH, feed As (III) concentration and ionic strength on the performance of NF270 and TW30 membranes, in term of As (III) rejection and permeate flux, were studied. The Table 1 shows the different values for the variation of each studied parameter.

Table 1. Different values for the variation of each studied parameter

Membranes / Parameters	NF270	TW30
Applied pressure (bar)	1; 2; 3	2; 4; 6
Recovery (%)	30; 50; 70; 90	30; 50; 70; 90
Feed pH	4; 7; 10	4; 7; 10
Feed [As (III)] (µg L^{-1})	50; 100; 150	50; 100; 150
ionic strength effect (EC, µS cm^{-1})	150; 500; 1000	150; 500; 1000

Note: for ionic strength effect, the electric conductivity (EC) of feed water was adjusted by adding a corresponding quantity of NaCl.

The calculation formulas of some parameters is shown in the Table 2.

Table 2. Calculation of some parameters

Parameters	Relations	Definition
Permeate flux (J, L h-1 m-²)	$J = \frac{Q_p}{A}$	Q_p, permeate flow rate (L h-1) A, membrane area (m²)
Recovery (Y, %)	$Y = 100 \times \frac{Q_p}{Q_F}$	Q_p, permeate flow rate (L h-1) Q_F, feed flow rate (L h-1)
Rejection (R, %)	$R = 100 \times (1 - \frac{C_p}{C_F})$	C_p, concentration of As (III) in permeate (µg L-1) C_F concentration of As (III) in feed (µg L-1)

2.3. As (III) Analysis

2.3.1. Equipment

As (III) was analyzed by anodic stripping voltammetry (ASV) at the rotating gold electrode with 797 VA Computrace, from Metrohm, Switzerland. 797 VA Computrace is a computer controlled system for voltammetry, which consists of a measuring vessel (50 mL of capacity) where three electrodes plunge. The working electrode, rotating disk electrode, is the gold electrode tip where take place electron transfers. The auxiliary electrode, glassy carbon rod, assures current traffic and its measure. The reference electrode, Ag/AgCl/KCl reference system, constituted of an electrolyte vessel filled with 3 mol L^{-1} NaCl suprapur. This electrode controls the working electrode voltage.

2.3.2. Analysis Protocol

Before each As (III) daily determination, the gold rotating disk electrode was conditioned electrochemically. The conditioning of the gold rotating disk electrode is repeated until the baseline is constant. For As (III) measurement, the scans are in the direct current mode. 19 mL of water sample mixed with 1 mL citric acid solution 1 M and 0.1 mL Ethylendiamine tetraacetic acid disodium salt dihydrate solution 0.05 M was placed in the measuring vessel and deaerated by purging with nitrogen during 300 s. The determination of the concentration of As (III) was performed by the standard addition method. Thus, after the recording of the sample voltammogram, two additions of 1 mL of standard solution 100 µg L^{-1} were carried out. More details of this method can be obtained on Metrohm application work reference AW CH4-0503-012011.

3. Materials and Methods

3.1. Effect of Applied Pressure

Figure 2 shows the rejection of As (III) and the permeate flux as function as the applied pressure with NF270 and TW30 membranes. It can be observed an increase of As (III) rejection with pressure from 1 to 2 bar and a decrease at 3 bar for NF270 membrane, whilst for TW30 membrane, As (III) rejection increases continuously with the applied pressure. As (III) rejection with TW30 membrane increases from 40.2 to 71.1% when pressure changes from 2 to 6 bar so an increase of about 30%. This increase of As (III) rejection when applied pressure increase was also highlighted by various studies [13,19,20,25]. The increase of As (III) rejection with the increase of the pressure can be caused by the permeate dilution. Indeed, when the pressure increases, water flow rate also increases and because the solute (As (III)) passage through the membrane is slower than water crossing, a dilution of permeate can be occur which results in an increased rejection. The decrease of As (III) rejection from 7.8 to 3.4% when pressure changes from 2 to 3 bar observed with NF270 membrane can be due to the high passage of solute with pressure. In fact, NF270 membrane is loose membrane, the increase of pressure promotes the solute passage by convection contrary to TW30 where the solute passage is governed by diffusion phenomenon. According to [26], the increase of pressure can cause a decrease in retention for NF membranes. Otherwise, it can be noticed that, at the same pressure (2 bar), the As (III) rejection rate of TW30 membrane (40.2%) is higher than the one of NF270 membrane (7.8%). The great difference of the rejection efficiencies resulted from the significant difference of the respective membrane pore size. NF270 membrane is a loose NF membrane whereas TW30 is a dense membrane. This low As (III) rejection by NF270 membrane was also observed by [20]. Considering the Figure 2 (a, b), it is interesting to notice that 2 bar pressure and 6 bar pressure are the pressures which gave a best As (III) retention rate for NF270 and TW30 membranes, respectively. As for water flux, it increases for both membranes with applied pressure. Water flux changes from 10.4 to 34.7 $Lh^{-1}m^{-2}$ and from 11.7 to 38.6 $Lh^{-1}m^{-2}$ for NF270 and TW30 membranes, respectively. Reference [21] also highlighted the increase of water flux with applied pressure during arsenic filtration.

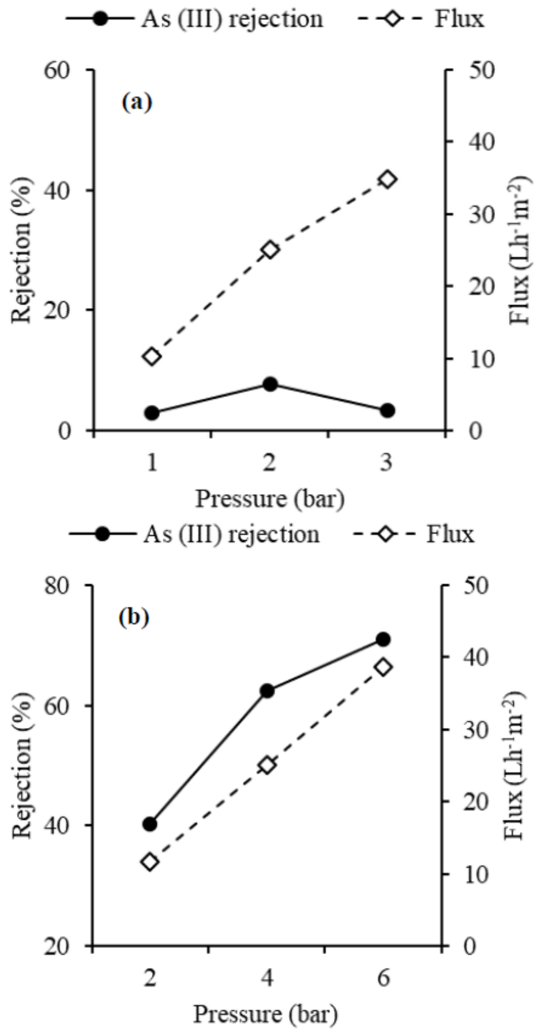

Figure 2. Effect of operating pressure on As (III) rejection and water flux (a) NF270 membrane and (b) TW30 membrane; pH: 5.2, T=25°C

3.2. Effect of Applied Pressure

Figure 3 shows a clear decrease of As (III) rejection and almost no variation for water flux when the recovery increases from 30 to 90% at constant pressure for both membranes. The rejection of As (III) changes from 9 to 1.5% and from 75.4 to 55.5% for NF270 and TW30 membranes, respectively. This decline of rejection can be caused by concentration polarization. Indeed, when the recovery increases, the retentate water flow decreases, the concentration of As (III) near retentate membrane side increases. That leads to increase of As (III) passage through the membrane. This decrease of retention due to the concentration polarization phenomenon was also highlighted by [20] during the removal of heavy metal ions by NF. Reference [27], who worked on the removal of various salts by NF45 membrane, also observed the decrease of ions rejection with increase of recovery. For water flux, it changes from 24.7 to 24 $Lh^{-1}m^{-2}$ for NF270 membrane and from 38.6 to 37.5 $Lh^{-1}m^{-2}$ for TW30 membrane, which is negligible.

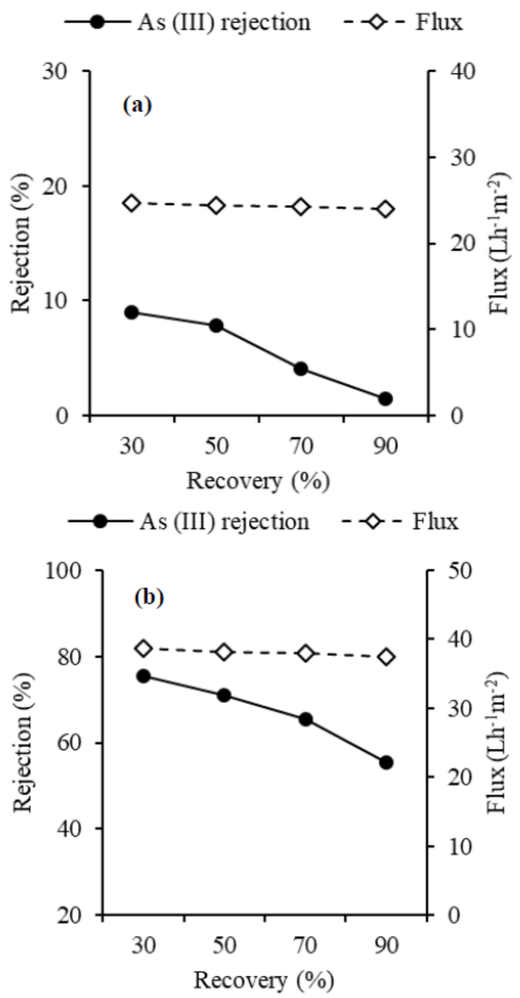

Figure 3. Effect of water recovery on As (III) rejection and water flux (a) NF270 membrane, 2 bar and (b) TW30 membrane, 6 bar; pH: 5.2, T=25°C

3.3. Effect of Feed pH

Figure 4 presents the As (III) rejection and the permeate flux as a function of pH. The variation of As (III) rejection is negligible when the pH changes from 4 to 7 for both membranes. At pH 10, As (III) rejection significantly increases: As (III) rejection varies from 5.7 to 23.8% and from 60.7 to 93.8% for NF270 and TW30, respectively. This lack of variation of As (III) rejection when the pH changes from 4 to 7 was also highlighted by [18,21]. For instance, reference [18] who worked on BQ01 NF membrane observed that, As (III) rejection rate remains constant when the feed pH changes from 4.5 to 8.5. As for [21], they observed that As (III) rejection was almost constant when the feed pH had varied from 4 to 9 for NF and RO membranes used. It could be explained by the fact that As (III) species - H_3AsO_3 - in solution remains the same when the pH change from 4 to 7. For this range of pH, species is neutral and the rejection then only depends on steric hindrance. However, at pH 10, the neutral species of As (III) - H_3AsO_3 - is converted in ionic species - $H_2AsO_3^-$ - (pka of H_3AsO_3 / $H_2AsO_3^-$= 9.22), the rejection results of the combined effect of steric hindrance and Donnan exclusion. That explains the increase of rejection observed at pH 10 due to the contribution of electrostatic rejection. For the water flux, slight variation was observed in the range of pH 4 – 10. It could result of membrane structure modification when the pH change like also highlighted by [28] when they studied the role of pH in nanofiltration of atrazine and dimethoate from aqueous solution. In fact, the pH variation can cause the expanding or shrinking of the cross linked-membrane polymer network, the lowest electroviscous effect and the highest net driving force due to lowest osmotic pressure at the membrane surface [16].

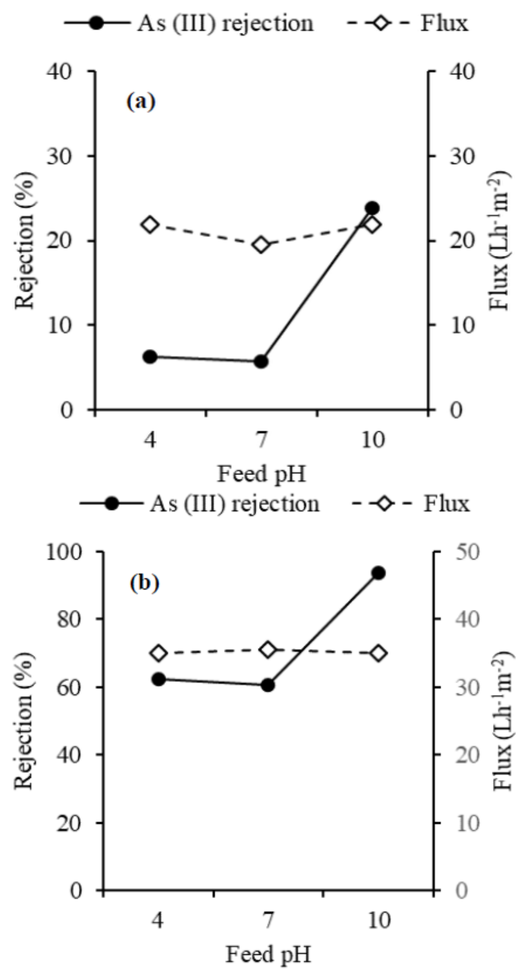

Figure 4. Effect of feed pH on As (III) rejection and water flux (a) NF270 membrane, 2 bar and (b) TW30 membrane, 6 bar; T=25°C

3.4. Effect of Feed As (III) Concentration

Figure 5 presents As (III) rejection and the permeate flux as a function of As (III) concentration. Water flux seems constant when feed As (III) concentration increases while the rejection decreases for both membranes. As (III) rejection decreases from 5.2 to 4% and from 63.3 to 57%, respectively, for NF270 and TW30 membranes when the feed As (III) concentration increases from 50 to 150 µg L^{-1}. A similar trend of As (III) rejection when the feed As (III) concentration increases was found by [11,21]. For instance, reference [11] observed with a NF membrane that the rejection of As (III) decreases from 9.8 to 2.0% when the arsenic concentration in feed increases from 20 to 90 µgL^{-1}. As for [21], they observed that, in the NF membrane process, increased feed As (III) concentrations (from 50 to 400 µg L^{-1}) caused about 10% decrease of As (III) rejection efficiency. Indeed, according to [18], when As (III) concentration in feed increases, both diffusion and convection of the uncharged As (III) species increase, resulting in an additional decrease of its rejection. The fact that water flux remains constant when the feed As (III) concentration increases from 50 to 150 µg L^{-1} shows that this increase of concentration does not affect membrane structure and does not cause concentration polarization phenomenon.

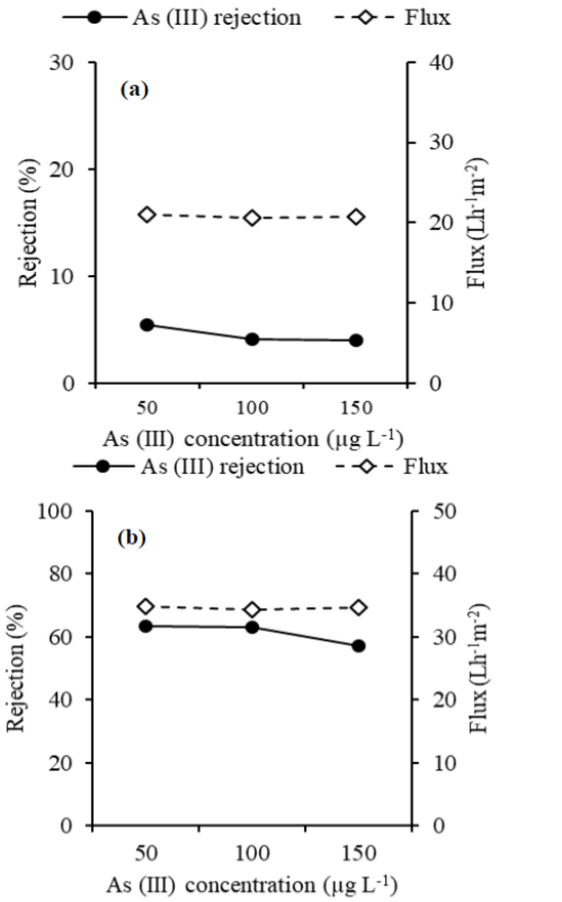

Figure 5. Effect of initial concentration on As (III) rejection and water flux (a) NF270 membrane, 2 bar and (b) TW30 membrane, 6 bar; pH: 7, 25°C

3.5. Effect of Feed As (III) Concentration

Figure 6 shows the effect of ionic strength on As (III) rejection and water flux. As (III) rejection seems unchanged whereas water flux slightly decreases when feed conductivity varies from 150 to 1000 µS cm^{-1} for both membranes. As (III) rejection is not impacted by ionic strength as it was 4.4, 4.9 and 4.7% for NF270 membrane, and, 66.3, 66 and 65.6% for TW30 membrane for 150, 500 and 1000 µS cm^{-1}, respectively. reference [11] also observed that the increase of concentration of additional salts did not affect A (III) rejection. According to [29], an increase of background salt concentration reduces the electrical double layer thickness of the membrane surface charge and contributes to enhance salt ion passage through the membrane pores due to the reduction of the electrostatic effect. But, as As (III) is uncharged at pH 7, working pH of this test, it is not influenced by the variation of membrane surface charge. Consequently, its rejection is not modified by additional salts added. For water flux, it changes from 20 to 18.2 $Lh^{-1}m^{-2}$ and from 33.4 to 31.4 $Lh^{-1}m^{-2}$ for NF270 and TW30 membranes, respectively. This slight decrease of water flux with the increase in ionic strength could result in the modification in the pore size of the membranes. Indeed, references [30,31], which worked respectively on fouling of NF membranes and on factors and mechanisms affecting rejection and flux decline with charged ultrafiltration membrane, observed that increase in ionic strength of a solution caused a decrease in the pore size of polyamide membranes. This decrease in the pore size could lead to the decrease of water flux.

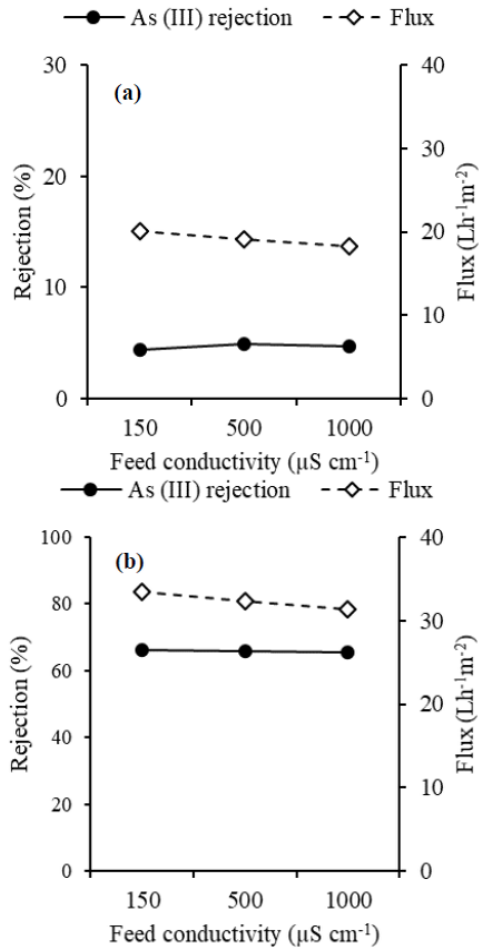

Figure 6. Effect of ionic strength on As (III) rejection and water flux (a) NF270 membrane, 2 bar and (b) TW30 membrane, 6 bar; pH: 7, T=25°C

4. Conclusions

Study of effect of operating parameters on the performance of NF270 and TW30 membranes during As (III) removal led to the following conclusions:

- For both types of membrane the influence of operating parameters is the same.
- When increasing applied pressure, As (III) rejection and water flux increased. Increase applied pressure contributes to enhance the performance of the membrane system.
- The increase of recovery leads to the decrease of the system performance for As (III) removal.
- The increase of feed pH contributes to improve As (III) rejection with few impacts on water flux.

- When feed As (III) concentration increases, As (III) rejection decreases whereas water flux remains unchanged.
- The increase of ionic strength does not have any influence on the rejection of As (III) uncharged species, but can contribute to the decrease of water flux due to pore size decrease.

According to what precede, applied pressure and feed pH seem to be the most important parameters which must be considered if As (III) rejection needs to be improved. Moreover, TW30 membrane is more efficient than NF270 membrane for As (III) removal whatever the operating conditions.

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