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Sodium and potassium removal from brackish water by nanofiltration membrane: single and binary salt mixtures

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ABSTRACT

In this study, the removal efficiency of sodium (Na^+) and potassium (K^+) by nanofiltration (NF) process was investigated. The experiments was conducted by variation of influent concentration of Na^+ and K^+, flow rate, solution pH and also co-existing anions and cations. The results showed that with increasing influent concentration of Na^+ from 200 to 400 mg/L, the removal efficiency decreased from 68.9% to 63.1% and K^+ removal efficiency declined from 89.1% to 83.4% with increasing concentration from 20 to 60 mg/L. As the solution pH was raised from 4 to 9, the Na^+ and K^+ removal efficiency increased from 66.5% to 74.1% and 87.5% to 93.5% and correspondence with deceasing the concentration of Na^+ and K^+ in permeate water from 67.1 mg/L to 51.8 mg/L and from 2.51 mg/L to 1.32 mg/L, respectively. In addition, as the flow rate was varied of 0.4 L/min to 0.8 L/min, the Na^+ and K^+ rejection by NF descended approximately 5%. The most important of effective co-exist anions and cations on Na^+ and K^+ rejection were nitrate and magnesium, respectively. Consequently, the NF process was recognized as efficient, convenience and low cost method for Na^+ and K^+ removal from brackish water.

Keywords: Co-existing anion and cation; Membrane filtration; Nanofiltration; Salt rejection; Sodium and potassium removal

1. Introduction

Sodium is a common element in the environment and occurs widely in soils, plants, water, and foods [1]. The metallic sodium is used in the manufacture of tetraethyl lead and sodium hydride, in titanium production, as a catalyst for synthetic rubber, as a laboratory reagent, as a coolant in nuclear reactors, in electric power cables, in nonglare lighting for roads, and as a heat-transfer medium in solar-powered electric generator [2]. Sodium salts are used in water treatment, including softening, disinfection, corrosion control, pH adjustment, and coagulation and in addition in road de-icing and in the paper, glass, soap, pharmaceutical, chemical, and food industries[3–4]. High levels of sodium may aggravate existing high blood pressure and adverse cardiovascular health [5]. Factors to help reduce

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high blood pressure include a low sodium diet, increased fruit and vegetable consumption, exercise, weight control, and medication if necessary [6].

Potassium is an alkali metal and the seventh most common element on earth [7]. Although potassium is one of abundant elements, the concentration found in most natural waters rarely exceeds 20 mg/L much higher levels being found in spa waters [8]. Potassium is an essential macro mineral nutrient for human body [9]. It has some salty flavor but they also have off-flavors, like metallic or bitter taste [5]. Potassium has an important effect on heart and bone health and reduces the risk of stroke and coronary heart disease by decrease in blood pressure and attenuates the adverse effects of sodium on blood pressure [9–10]. The standard limit for sodium and potassium was determined 150 and 12 mg/L, respectively [8].

The different methods were used for Na^+ and K^+ removal and are including ion exchange, electrochemical, reverse osmosis, adsorption and freezing [11–15]. During the last decade, membrane process has been noticed considerably in removal of different contaminants from aqueous solution. Depending on pore sizes, the membrane process can be classified into four broad categories including microfiltration (MF), ultrafiltration (UF), NF and reverse osmosis (RO) membranes [16]. Todays, membrane process and cellulosic fibers used for water treatment in extensive domain [17–20]. Due to lower energy consumption and higher flux rates, the NF membrane has attracted some attention in comparison to RO [17]. The RO has ability to remove almost all cations and anions in both brackish and seawater desalination. For NF membrane, the rejection of sodium and potassium chloride is an important factor that determines the efficiency of NF membranes that are applied to desalinate brackish water [21–23]. The advantages of NF membranes process including high efficiency in removing multivalent ions, operational simplicity, lower pressure than RO membrane, no additive requirements, low energy consumption, modular construction and application to form point of use [24–25]. The charged groups and pore size diameters less than 1nm are some of NF membrane characteristics [28]. In the NF membrane process, three solute-membrane interactions are including steric exclusion (sieving), Donnan (charge) interactions, and solute-membrane affinity is governed the mass transfer of membrane [29]. The RO and NF membranes fouling causes loss of productivity so, it leads to operational cost subsequently [27,30].

In the previous study, the NF membrane used to removal of organic matter [31–32], pharmaceutical chemical [29,33–34], lactic acid from fermentation broth [35] and salts rejection including cations, anions and heavy metals [36–40] from aqueous systems such as water and industrial effluent. However, while NF rejection behavior for single components (salts, pesticides, microorganisms, etc.) is largely understood, its behavior in multi-component systems becomes complex and less predictable [41].

This research aimed to evaluate the Na^+ and K^+ removal efficiency by NF membrane under different operational condition including different solution pH, influent Na^+ and K^+ concentration and flow rate of brackish water. In addition, the effects of co-existing anions and cations (binary salt mixture) on Na^+ and K^+ rejection efficiency by NF membrane were simulated.

2. Experimental methods

2.1. Reagents and sample analysis

The salt solutions were freshly prepared immediately prior to filtration using analytical reagent-grade chemicals (Merck Co. Germany) dissolved in distilled water. The Na^+ and K^+ for permeation experiment was prepared from NaF, NaCl, $NaNO_3$, Na_2SO_4, KCl, KNO_3 and K_2SO_4 dissolved in distilled water. The Na^+ and K^+concentrations were measured by a Dr. Lange M7D Flame Photometer (Artikel-Standort. Hamburg, Germany). During experiments, solution pH, fluoride, chloride, nitrate, calcium and magnesium were measured using a glass body pH probe (CG 824 SCHOTT, Germany), SPANDS (DR5000 Spectrophotometer, Hach Co. Germany), titration method, cadmium reduction method (Hanna instruments, USA), and colorimetric methods. All test methods were adopted from standard methods for the examination of water and wastewater [42]. All experiment carried out at 20°C and Na^+ and K^+ removal efficiency was calculated by Eq. (1).

$$R(\%) = \left(1 - \frac{C_p}{C_f}\right) \times 100 \quad (1)$$

where R is Na^+ and K^+ removal efficiency (%), C_p and C_f are Na^+ and K^+ concentration in permeate and feed, respectively.

2.2. NF membrane processset up

In this study, a pilot scale commercial NF membrane was used. Table 1 shows the characteristics of NF membrane and Fig. 1 illustrates the schematic design of the NF system. The feed water is pumped onto the NF module by a peristaltic pump. In all stage, the experiments were conducted at a pressure 8 bars and 45% of recovery rate.

2.3. Experiments procedure

The experiments was carried out by changing the influent concentrations of Na^+ from 200 to 400 mg/L and K^+ initial concentrations from 20 to 60 mg/L with different influent flow rate (0.4 to 0.8 L/min). The effect of differ-

Table 1
Characteristics of NF membrane

Membrane type	Polyamide thin-film composite
Maximum operational pressure, bar	8–16
Maximum operational temperature, °C	45
Continuous operating pH range	4–11
Surface charge	Negative
Nominal cut off, Da	270
Maximum influent discharge, L/min	0.8

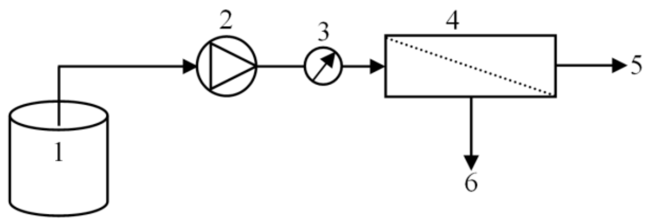

Fig. 1. Schematic of nanofilter pilote (1: feed tank, 2: pump, 3: barometer, 4: NF membrane, 5: permeate flow and 6: concentrate flow).

ent solution pH (4–9) was performed at Na^+ and K^+ initial concentrations 200 and 20 mg/L, respectively with 0.4 L/min of flow rate. In order to investigate the effect of co-existing anions and cations (binary mixture) on Na^+ and K^+ removal by NF membrane, fluoride, chloride, nitrate (as anions), sodium, potassium and magnesium (as cations) ions were chosen.

3. Results and discussion

3.1. Influent concentration of Na^+ and K^+ and flow rate

The obtained results of influent concentration of Na^+ and K^+ and flow rate are shown in Figs. 2 and 3. As shown in Fig. 2, with increasing Na^+ concentration in the influent feed water from 200 mg/L to 400 mg/L, the Na^+ removal efficiency decreased from 68.9% to 63.1%. In addition, the K^+ rejection was declined from 89.1% to 83.4% with increasing initial concentration of K^+ in feed water from 20 mg/L to 60 mg/L. The results show that with rising feed flow rate, the Na^+ and K^+ rejection decreased approximately by 5% (Figs. 2 and 3).

With increasing initial concentration of Na^+ and K^+ in feed water, the salt concentration at the membrane surface increased too. This situation led to increasing of concentration polarization for which the membrane must filter a solution with a higher Na^+ and K^+ concentration than the feed water. Therefore more salts are permeated through the membrane and the removal decreases. The results of this study are in line to other researches [43–45].

With increasing flow rate of feed water, the mass coefficient across the concentration polarization boundary layer and the degree of mixing near the membrane surface increased. It can leading the thickness reduction of the boundary layer at the interface between membrane and solution [39]. It means that with increasing flow rate of feed water from 0.4 to 0.8 L/min, the Na^+ and K^+ removal efficiency is reduced. In addition, this situation may be relayed to the influence of amount of ion discharged over membrane surface on solute transfer. This function is characteristic of situations where concentration polarization still influences the solute transfer with a non-negligible contribution of diffusion in the pores and led to dispersion and then to a lower rejection [42]. Some researchers have found that rises flow rate has decreased the retention efficiency [42], while others have presented the opposite conclusion [39,47].

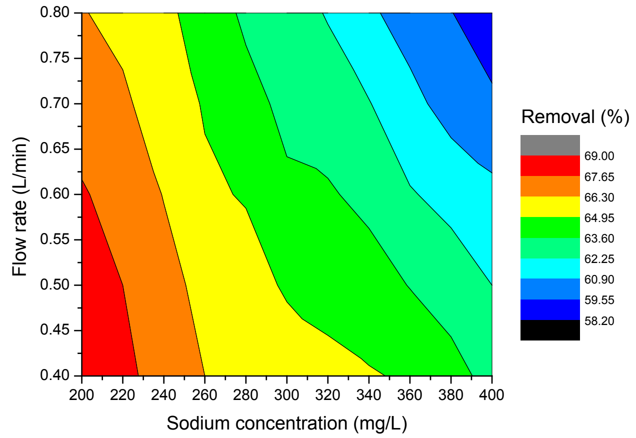

Fig. 2. Na^+ removal as function of influent concentration of Na^+ and flow rate (Influent concentration: 200–400 mg/L, flow rate: 0.4–0.8 L/min and solution pH: 7).

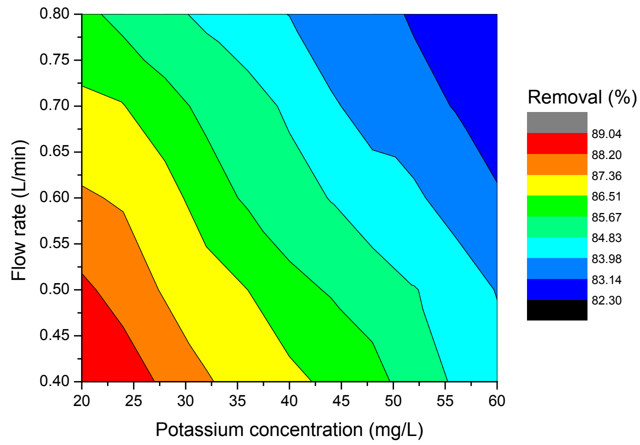

Fig. 3. K^+ removal as function of influent concentration of K^+ and flow rate (Influent concentration: 20–60 mg/L, flow rate: 0.4–0.8 L/min and solution pH: 7).

3.2. Effect of solution pH

The effect of solution pH on Na^+ and K^+ rejection by NF membrane was studied and obtained data are shown in Fig. 4. As shown in Fig. 4, when the solution pH of feed water was increased from 4 to 9, the rejection percentage of Na^+ and K^+ in the permeate water increased from 66.5% to 74.1% and 87.5% to 93.5%, respectively.

The Nernst-Planck equation is explained change in the rejection with pH and rejection difference originating from difference in chemical species. The extended Nernst-Planck equation will be written as Eq. (2) [48].

$$J_i = \left(-D_i \frac{dc_i}{dx}\right) - \left(D_i \cdot z_i \cdot c_i \cdot \frac{F}{RT} \cdot \frac{d\psi}{dx}\right) + K_i \cdot c_i \cdot J_v \qquad (2)$$

The NF membrane charge density is an important parameter to determine the rejection in charged membranes. The concentration and type of other solutes in the solution affects the membranes charge density, and finally it affects the rejection of the Na^+ and K^+ by NF membrane. With increasing solu-

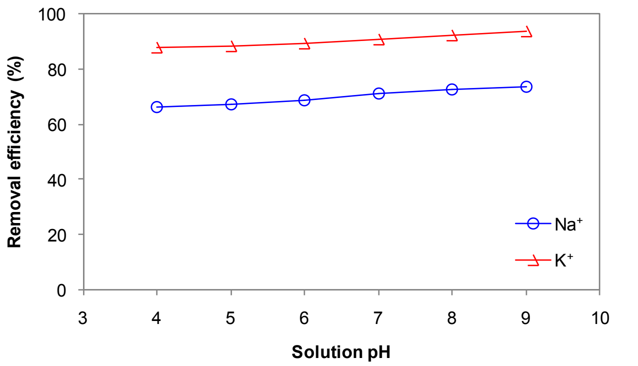

Fig. 4. Effect of solution pH on Na^+ and K^+ removal efficiency (flow rate 0.4 L/min, 20 mg/L of K^+ and 200 mg/L of Na^+).

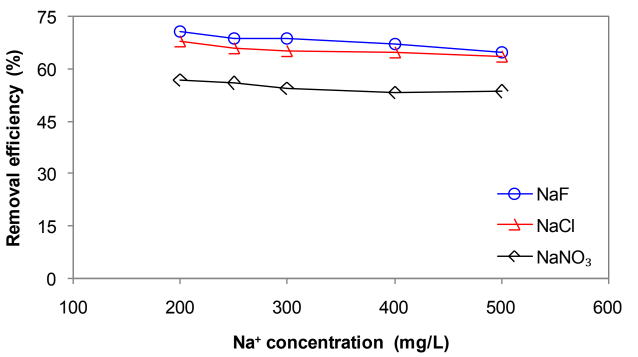

Fig. 5. Effect of co-existing anions on Na^+ removal (flow rate: 0.4 L/min and solution pH: 6).

tion pH, the removal efficiency of Na^+ and K^+ increased and presumably related to the interfering of hydronium ions with Na^+ and K^+ in neutralization of membrane surface charge. As previously mentioned, the NF membrane used in this study was negatively charged in the neutral pH. Therefore, as the solution pH was decreased, the effective charge density decreases due to isoelectric point location of the membrane at a lower pH [48,49]. The membrane zeta potential (or charge) is influences with solution pH and increasing of pH caused to increasing of the zeta potential and so the NF surface become more negative [50]. Some researchers are reported similar results in arsenic removal by NF membrane [48,50].

3.3. Effect of co-existing anions

Figs. 5 and 6 show the results of the presence of co-existing anions on Na^+ and K^+ rejection by NF membrane. The results depicted that with increasing ionic radius of co-existing anion, the Na^+ and K^+ removal efficiency decreased. The Na^+ and K^+ rejection (influent concentration 200 mg/L of Na^+ and 20 mg/L of K^+ as influent concentration) by using F^- and Cl^- as co-existing anion were 80.8% and 88.8%, respectively. These rejection percent was decreased with increasing the ionic radius of a co-existing anion. The ionic radiuses of co-existing anions are summarized in descending order as shown below:

$NO_3^- >> Cl^- > F^-$

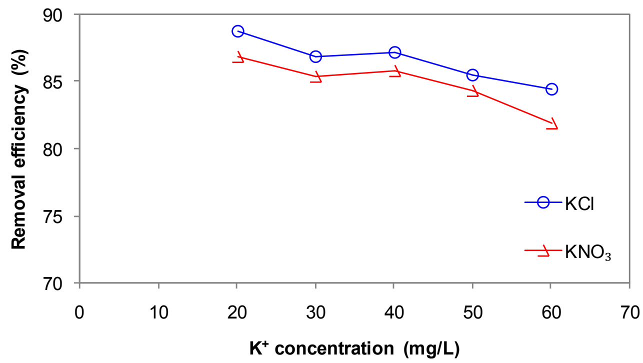

Fig. 6. Effect of co-existing anions on K^+ removal (flow rate: 0.4 L/min and solution pH: 6).

The highest Na^+ and K^+ removal efficiency was obtained with application of fluoride and chloride as co-existing anions, respectively. It may be related to lower rejection of fluoride and chloride by NF membrane that affected Na^+ and K^+ removal efficiency to maintenance of NF systems balance [37,39].

3.4. Effect of co-existing cations (binary salt mixture)

The Na^+ and K^+ removal efficiency by NF membrane from binary salt mixture are illustrated in Figs. 7 and 8. In overall, the percentage of rejection of Na^+ and K^+ was

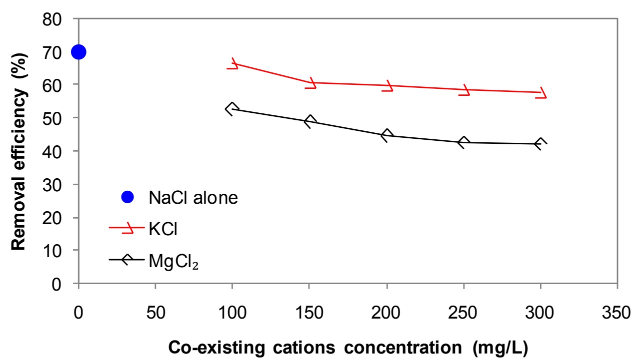

Fig. 7. Effect of co-existing cations on Na^+ removal (influent concentration of Na^+: 200 mg/L, flow rate: 0.4 L/min and solution pH: 6).

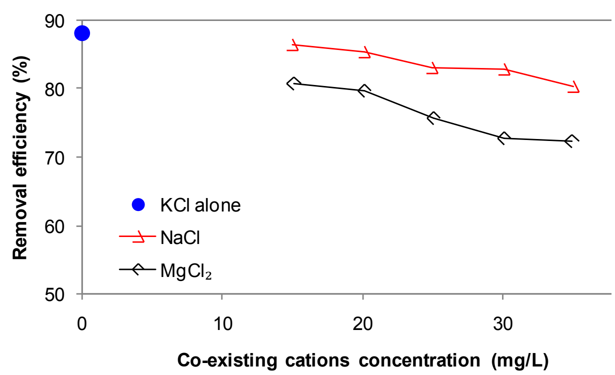

Fig. 8. Effect of co-existing cations on K^+ removal (influent concentration of K^+: 20 mg/L, flow rate: 0.4 L/min and solution pH:6).

Table 2
Building investment and operating costs of NF process

Cost type	Sub-cost	Equation	Value
Investment	Civil ($)	$55.73\ Q_f + 80.1$ n	16818.39
	Mechanical engineering ($)	$233.44\ Q_f^{0.85} + 58.75$ n	13675.93
	Electrical ($)	$3 \times 10^3 + (3.49PQf)$	3918.64
	Membrane module ($)	666 n	1326
	Total Investment cost ($)	Civil + Mechanical engineering + Electrical + Membrane module	167057.97
Operating	Maintenance ($)	2% of total investment	3341.16
	Membrane ($)	60 $/m² (6 months membrane life)	10000
	Labor ($)	400 $/month	4800
	Total operating cost ($)	Membrane cost + Labor cost + Maintenance cost	18141.16
The annualized capital cost ($/m³)		(Total cost × *CRF**) ÷ *Q*	0.44

**CRF:* $(I \times (1+i)^{n^{**}}) \div ((1+i)^{n-1})$, **n: project life

declined in the permeate water by adding any co-existing cation into feed water.

As depicted in Figs. 7 and 8, the rejection of Na^+ and K^+ by NF membrane in single mixture at influent concentration of 200 and 20 mg/L was 69.8% and 88.1%, respectively. With adding of KCl and $MgCl_2$ at 100 mg/L in the feed water (binary mixture), the Na^+ removal was diminished to 66.4% and 52.5%, respectively. In the case of K^+, the rejection percent was decreased to 86.4% and 80.8% with addition of NaCl and $MgCl_2$ at 15 mg/L, respectively. As seen in Figs. 7 and 8, in overall, with increasing any co-cation in feed water, the Na^+ and K^+ removal efficiency was declined and the highest decreasing effect on rejection of Na^+ and K^+ is related to magnesium cation. This situation presumably related to more neutralization of surface membrane charge by magnesium cation due to more positive charge. So, the effect of co-cation with positive charge can be written according to:

$$M^{2+} > M^+$$

Hydration energy of ions could also be affected on crossing ions, the more ions are hydrated so more it would be removed by NF membrane. Other researcher was adopted this results [43].

3.5. Economic evolution

The economic evolution of NF membrane process for Na^+ removal was conducted for the plant capacity of 10 m³/h according to Kumar and Pal (2013) procedure [51]. The following design criteria were considered.

- Na^+removal efficiency: 70%
- Permeate recovery rate: 45%
- Operating pressure (*P*): 8 bars
- Membrane flux: 0.12 m³/m²·h
- Membrane module area: 0.5 m²
- Project life: 20 year
- Interest rate: 15 %

Table 2 was summarized the total annualized cost of NF process.

As presented in Table 2, in this study, the total annualized cost (investment + operating cost) was calculated equal to 0.44 $/m³. Verberne and Wouters [51] showed that the total operating costs is 038 $/m³ of water produced and Kumar and Pal [54] demonstrated nearly 0.46 $/m³. This difference presumably related to operating pressure of membrane and interest rate.

4. Conclusion

Based on the carried out experiment, the following conclusions can be presented.

- With increasing Na^+ and K^+ concentration in feed water, the NF removal efficiency was decreased.
- The higher influent flow rate to NF membrane leading the lower removal efficiency of Na^+ and K^+.
- As the solution pH of feed water was increased from 4 to 9, the rejection efficiency of Na^+ and K^+ was increased.
- For co-existing anions, it was concluded that with increasing in ionic radius of anion, the $Na^+$and K^+ removal efficiency decreased.
- The highest decreasing effect on rejection of Na^+ and K^+ is related to magnesium existing as co-existing cations.

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