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Research Article

Efficiency of Low-Pressure Reverses Osmosis (RO) in Desalination and TOC Removal from Caspian Seawater and Tajan River

Laleh R. Kalankesh[a], Mohammad Ali Zazouli[b*], Ahmad Mansouri[c*]

[a] *Department of Environmental Health Engineering, Faculty of Health, Health Sciences Research Center, Student Research Committee, Mazandaran University of Medical Sciences, Sari, Iran.*
[b] *Department of Environmental Health Engineering, Faculty of Health, Health Sciences Research Center, Mazandaran University of Medical Sciences, Sari, Iran.*
[c] *Department of Environmental Health Engineering, Faculty of Health, Mazandaran University of Medical Sciences, Sari, Iran.*

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ABSTRACT

Water scarcity is a critical issue in Caspian Sea regions of Iran. Thus, people may use polluted water or saline brackish groundwater, estuarine water or seawater. This paper deals with the application of Low-Pressure reverse osmosis (RO) for removing salt and Total Organic Carbon (TOC) in synthetic and Caspian Sea waters. The study aims to achieve optimization at different pressures (30, 50, 70, and 90 PSI) with synthetic seawater at initial salt concentrations (5, 25, and 35 g/L TDS) at various retention time intervals (15, 30, 60, 90, and 120 minutes). The results showed that the low-pressure RO system was able to reject 95 %, 57 %, and 46 % of 5, 25, and 35 g/L of TDS from synthetic seawater. In addition, rejection efficiency was achieved at 86 % and 78 % for Caspian seawater and Tajan River, respectively. In addition, optimal conditions (pressure: 70 PSI, time: 120 min) for salt rejection included 16-23 %, 93-94, 52-56 %, 88-90, and 22 % for 35g/L TDS, Tajan River, 5g/L TDS, 25g/L TDS, and Caspian seawater, respectively. Moreover, TOC rejection was achieved at >95 % and >97 % of Tajan River and Caspian seawater, respectively, at an overall 120-minute interval. In the case of growing environmental pollution that is discharged into Caspian sea including industrial and agricultural effluents from rivers, this study proposed the suggested pilot as a simple design that will significantly reduce salt, TOC, and TDS.

1. INTRODUCTION

Access to sufficient water is one of the challenging issues worldwide. Water scarcity refers to not only water availability but also water quality. Three major sectors of water demand include municipal, industrial, and agricultural sectors. Municipal and industrial demands form the largest part of these demands. Coastal areas with an enormous saline water source have a good potential to compensate the water shortage in these areas. In addition, the desalination of saline water in arid and semi-arid regions has received enough attention. Over the years, various desalination technologies have been developing for finding a suitable alternative method to remove salinity (i.e., reverse osmosis, RO; electrodialysis, ED; nanofiltration, NF). The selection of a suitable desalination method depends on the condition and geographical position of each country and, therefore, varies. Although, in the last decades, distillation has been a preferable technical method for the desalination of seawater, membrane desalination techniques have become a popular desalination method these days [1, 2]. However compared to other membrane technology, reverse osmosis has received double thumbs up due to high energy efficiency and lower operating costs. Fouling and biofouling are considered to be critical issues in the RO membrane system, which has been used for desalinating seawater [3]. Meanwhile, the desalination of seawater with a high organic concentration is posing a major challenge [4]. Therefore, addressing the possibility of simultaneous removal of salinity and Total Organic Carbon (TOC) in seawater by reverse osmosis is of great interest. A functional Ro system, including pre-treatment, can remove a large part of the organic substances. It has been reported that RO system can recover 30 % and 40 % of seawater [5]. In addition, important design parameter and running conditions such as initial salt concentration, pressure, pH, temperature, and flow rate affect the desalination efficiency in the RO system. Lectures reported that by reducing the feedwater pH from 7.2 to 7.0, the water recovery improved from 22.5 % to 34.2 % [6]. In addition, by applying the system through two stages of SWRO desalination process, 60 % of the overall water recovery has been attained [7]. Further, a designed multi-stage RO system with micro-filtration as a pre-treatment technique, water recovery successfully increased from 30 % up to 50 %. Caspian Sea with about 13,500 - 16,000 ppm salinity could be an attractive site to test RO efficiency in removing TOC and salinity simultaneously [8]. In addition, Tajan River is one of the most important water sources that connects to the Caspian Sea with high contamination. Concerning the lack of potable water source, the Iranian government has focused on the desalination of the Caspian Sea and rivers, entering the Caspian Seawater. Therefore, this paper investigates salinity and TOC removal efficiency at different initial salt concentrations and pressures by the application of the RO system. Water sources in the northern part of Iran are exposed to highly organic pollutants; therefore, investigating these cases through OR system is considered to be necessary. In addition, salinity and TOC removal efficiency of actual Caspian seawater and Tajan River is determined in optimal conditions.

*Corresponding Author's Email: mzazouli@mazums.ac.ir (M.A. Zazouli) & meeladmansoori@gmail.com (A. Mansouri)

2. MATERIALS AND METHOD

2.1. Water sampling site

Water samples were collected from the Caspian Sea water and Tajan River in November 2018. The study area in the Farah Abad Region in the north of Iran (Mazandaran Province) is located at an approximately N37 ◦ 15′ latitude and E50 ◦ 23′ latitude. The Caspian Sea with a salinity rate of about 13,500 - 16,000 ppm could be an attractive site to test the possibility of producing potable water by RO system with low costs [9]. The Caspian Sea, 1,200 km long and 320 km wide, has a surface area of approximately 371,000 km^2 and is the largest, completely enclosed body of water on Earth, and Tajan River is one of the main rivers that flows into the Caspian Sea (Fig. 1).

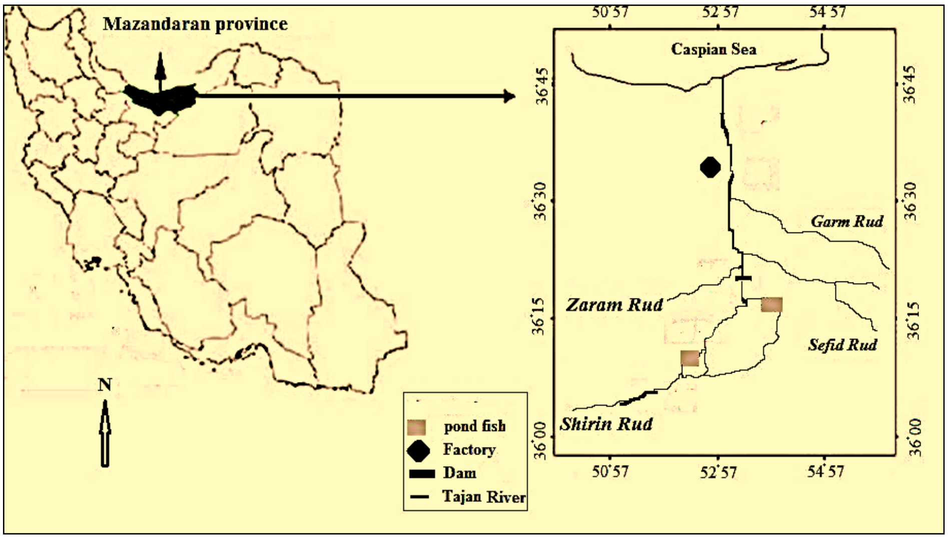

Figure 1. Diagram of the studied area and direction of Tajan river flowing from south to north [10].

2.2. Lab-scale RO membrane

A schematic diagram of the lab-scale RO membrane system is shown in Figure 1. An experimental program was implemented by using a Film Tec spiral-wound RO element, FT30 SW30-2521 (2.4" diameter, 21" length). The Ro membrane system including an accumulator (damper) for steady flow circulation and a backpressure (bypass) valve was performed with cross-flow filtration. The feedwater pumped through the membrane system with a high-pressure pump (DIAPHRAGM PUMP HF8367, serial number 1234567, made in Taiwan) is separated from the concentrate and, then, permeated. The experiment was carried out in the full circulation mode, where the retentate and permeate returned to the feed tank in order to maintain a constant concentration. Microfiltration cartridge was used for pretreating Caspian seawater and Tajan River before flowing into the RO membrane (Figs. 2a and 2b).

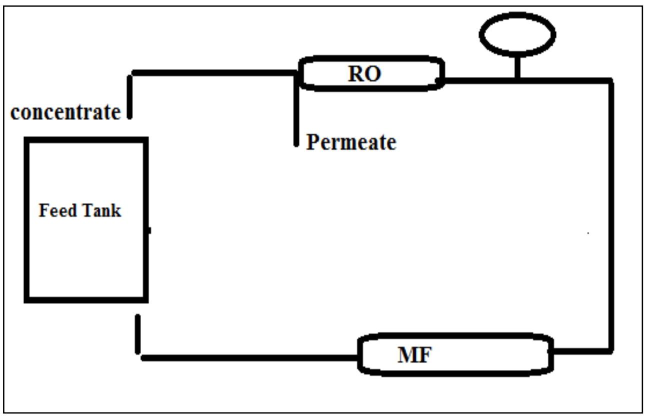

Figure 2a. Schematic diagram for Reverse Osmosis system used to evaluate TOC and salt removal efficiency from Caspian seawater and Tajan River.

Figure 2b. Actual photo of the RO system pilot for the desalination and TOC removal test.

2.3. Quality control (QC)

Methods recommended for performing the chemical analysis of serious pollutants in water include detailed QC procedures and requirements that should be followed closely throughout the evaluations. General procedures should include the analysis of a procedural blank, a matrix duplicate, a matrix spike for every 10-20 samples processed, and surrogate spike compounds (for organic analyses only). Analytical precision can be measured by analyzing 1 sample in triplicate or duplicate for every 10-20 samples analyzed. If duplicates are analyzed, the relative percent difference should be reported; however, if triplicates are analyzed, the percent relative standard deviation should be reported. Analytical bias can be measured by analyzing SRM, a matrix containing a known amount of a pure reagent. Recoveries of surrogate spikes and matrix spikes should be used to measure precision and bias; the results of these analyses should be well documented.

2.4. Membrane filtration experiments

Te experiment was carried out in the full circulation mode, where the retentate and permeate returned to the feed tank in order to maintain constant concentration. The experiments were carried out with the following protocol; for the first 30 min, the membrane was rinsed with DI water, followed by measuring the permeability of pure water as a reference. Then, an experiment with a particular feed solution was carried out until a steady state was reached. The permeate flux and samples were taken at the start, at the end, and at several predetermined time intervals (every 15 min). The permeate fluxes denoted by J and J0 (as LMH) were calculated by the ratio between the permeate flux (QP) and the membrane surface area as in the following Eq. (1) [11].

$$J = \frac{Q_P}{A} \tag{1}$$

Retention solute of TOC and conductivity was calculated according to Eq. (2) [11].

$$R\,\% = \frac{C_f \ - C_p}{C_f} * 100 = 1 - \frac{C_p}{C_f} * 100 \tag{2}$$

where R% is the retention percent, and C_f and C_p are solute concentrations in feed and permeate, respectively. After each experiment, the membrane was rinsed with DI water.

This process was implemented at pressure rates of 30 to 70 PSI, a feed flow rate of 1/h, and various initial salt concentrations of 5, 25, and 35 g/L.

2.5. Synthetic seawater

Synthetic seawater was prepared following the ASTM D1141-98 [12]. Deionized water (DI) was used for the preparation of all stock solutions and Synthetic seawater. Synthetic seawater was prepared with 371.73 g of NaCl (99 % NaCl, Samchun) and 62.35 g of sodium sulfate (98.5 % Na_2SO_4, Samchun), which was added to 300 ml of Stock 1 and 150 mL of Stock 2 (Table 1). Optimum values were determined separately for each experimental series.

Table 1. Chemical ingredients for synthetic seawater.

	Chemicals	Molecular weight	Weight (g)
Stock solution 1	$MgCl_2\ 6H_2O$	95.21	1133.82
	$CaCl_2$ (anhydrous)	10.98	120.71
	$SrCl_2 \cdot 6H_2O$	158.53	4.31
Stock solution 2	KCl	74.55	69.46
	$NaHCO_3$	84.01	20.10
	KBr	119.01	10.06
	H_3BO_3	61.83	2.71
	NaF	41.99	0.30

2.6. Analytical methods

The TOC of the feeds permeates and TOC-VCSH TOC Analyzer (Japan) analyzed retentate samples in accordance with the Standard Method 5310C. Conductivity (μS cm^{-1}) and pH of the feeds, permeates, and retentate were measured according to Standard Methods [13]. Electrical conductivity (EC) was measured by AQUA LYTIC-SensoDirect CON200, Germany, and pH of the solution was measured by an EUTECH-PH5500 electrode.

3. RESULTS AND DISCUSSION

3.1. Characterizations of Caspian seawater and Tajan river water

The characterization of Caspian seawater and Tajan River samples is presented in Table 2. A detailed characterization of the samples shows that Caspian seawater and Tajan River have a high pH (~9.3), resulting from the contact with $CaCO_3$ (calcite) and $MgCO_3$ (dolomite). In addition, the high conductivity of sea waters results from high salinity and hardness of water. In addition, TOC of the sample was ~5-6 mg/L. Concerning the obtained results of analyzing the samples, the possibility of using an RO system for the desalination of seawater because of human activities, cesspools, irrigation return flows, overexploitation (leading to aquifer salinization), municipal wastewaters, solid waste disposal, or wastewater treatment plants is debatable.

Table 2. Physicochemical characteristics of Caspian seawater and Tajan River.

Property	Caspian seawater	Tajan River
Calcium*	5533 ± 446	1156 ± 56
TDS (Total Dissolved Solid)*	10400 ± 165	3320 ± 250
Magnesium*	442 ± 50	246 ± 32
SO_4*	1500 ± 100	752 ± 33
Cl*	4516 + 76	2892 ± 345
Sodium*	5100 ± 70	1261 ± 261
Electrical conductivity**	18.36 ± 2.7	15.6 ± 2.3
pH	8.69 ± 0.9	8.00 ± 0.5
TOC*	5.5	5.7 ± 0.6
* mg/L **μs/cm^2		

3.2. Selection of optimal operating condition for RO

3.2.1. Effect of operation pressure

Operation pressure has the highest effect on permeation flux among the other operating parameters in membrane efficiency [14]. The permeation flux of RO membrane increases with the increasing pressure. This behavior can be attributed to the fact that when pressure increases, the feedwater is allocated and forced through the membrane to emerge as purified product water [15]. Water transport is pressed on the membrane pressure, and one of the important parameters affects the water activity of the membrane. On the other hand, as the pressure increases, the water activity will also increase exponentially and, in turn, will increase the degree of swelling; therefore, an increase in pressure results in the increasing flux. Desalination efficiency of the RO system was evaluated at different pressures (30, 50, 70, 90 PSI) and at a 5 g/L initial TDS concentration at a 120-minute time interval [16]. The results show that the desalination and TDS rejection by RO were high in percentage. Based on experimental data, as pressure increases from 30 to 90 PSI, TDS rejection increases from 89 % to 92.23 % (Fig. 3) and, also, salt rejection increases from 82.5 to 94.9 % (Fig. 4). In addition, time is another important factor in RO membrane performance. The result shows that time has an inverse effect on the membrane efficiency, and this can be related to the fouling of membrane pore size over time.

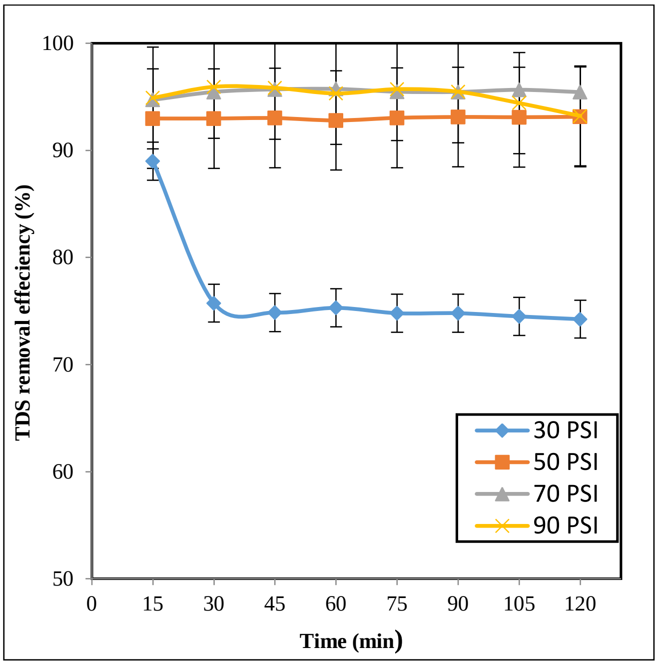

Figure 3. Effect of operation pressure on TDS rejection in TDS, 5 g/L pressures (30, 50, 70, and 90 PSI) during 120 min hydraulic retention time.

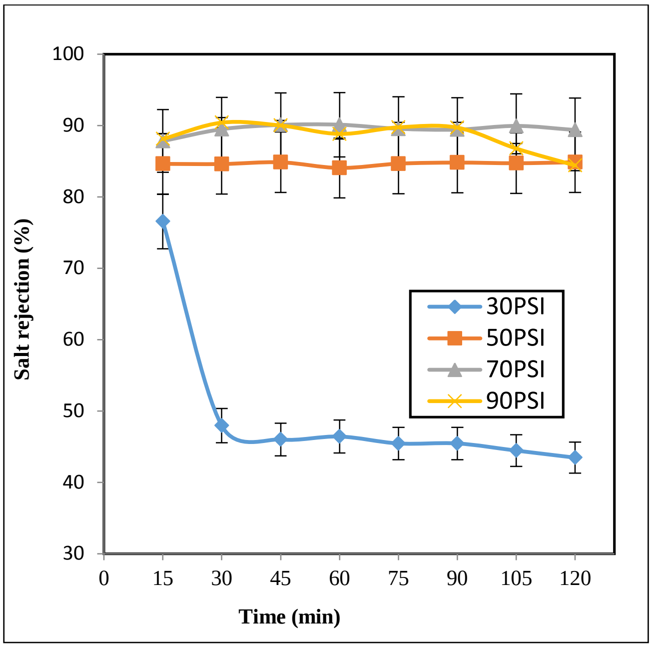

Figure 4. Effect of different levels of pressure on salt rejection in 5 g/L NaCl, pressures (30, 50, 70, and 90 PSI) during 120 min hydraulic retention time.

3.2.2. Effect the initial salt concentration of feed water rejection

In order to determine the rejection efficiency of membrane, experiments were carried out at three various salt concentrations (5, 25, 35 g/L NaCl) at 70 PSI. The data show that conductivity rejection for Caspian sea was nearly 50-60 %, while rejection for Tajan River water and 5 g/L NaCl was fairly high and nearly constant irrespective of the operating conditions (88-95 %). In addition, it was observable that the conductivity rejection increased as the feed salt concentration (conductivity) increased. Meanwhile, an interesting behavior was seen at 5 g/L NaCl (Fig. 4). Salt rejection of RO is a critical issue in membrane separation. Meanwhile, salt activities and water chemistry have significant effect on the salt rejection properties of the membrane. The detailed mechanisms for salt rejection are not very clear yet. However, the solution-diffusion model and the Donnan exclusion model are two popular mechanisms that play important roles in the salt rejection in RO membrane [17]. In the Solution-Diffusion Mechanism, solvent and solute dissolve in the homogeneous nonporous surface layer of the membrane and are transported by a diffusion mechanism in an uncoupled manner [18, 19]. It has been demonstrated that the nonporous surface layer membrane leads to high salt rejection and different mass transfer rates of the solvent and solute due to differences in solubility, and diffusivity is the main mechanism that leads to salt rejection. Therefore, no membrane is ideal in the sense that it absolutely rejects salts. The different transport rates lead to clear salt rejection [16, 20]. In the Donnan exclusion mechanism, an interaction between the solute and membrane occurs, which is known as charge effects. The surface of RO membrane with polymeric materials that contact the aqueous medium is charged. In addition, the effect of ions distribution at the membrane-solution interface attracts the charged membrane; this electrostatic repulsion is known as a Donnan exclusion.

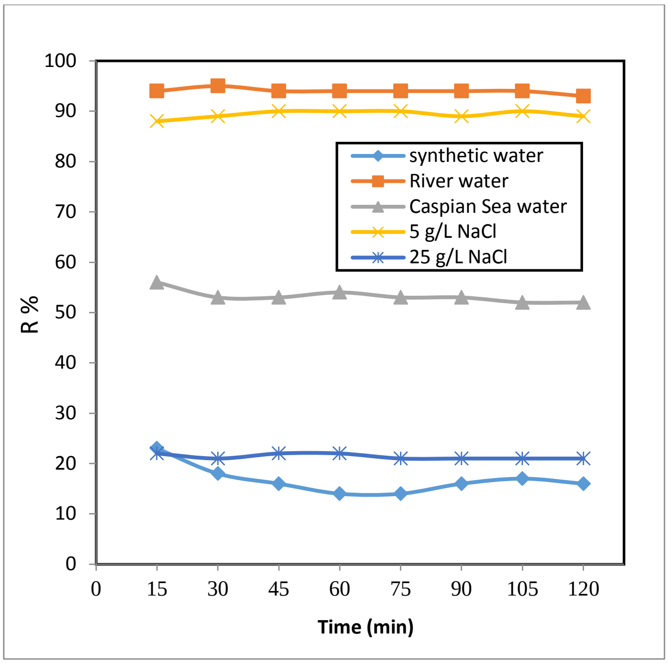

Figure 5. Effect of the initial salt concentration on the RO system efficiency at 70 PSI and at a 120 min retention time interval in synthetic seawater, Tajan River, and Caspian Seawater sources.

3.3. Total Organic Carbon rejection

The efficiency of RO membrane in TOC rejection of synthetic seawater, Tajan River, and Caspian Seawater was evaluated in the 70 PSI at a 120 min time interval (Fig. 5). The average TOC concentration in the RO system was found to be very effective in removing the most critical organic materials. The examination of all synthetic and real seawater and Tajan River shows that the RO system is able to reduce >97 % of TOC (Fig. 6). The investigation of TOC rejection showed that time was not an important factor in the removal efficiency, and TOC removal efficiency varied between 95-97 % and ~ 98 % for Tajan River and Caspian seawater, respectively, during

120 min. However, RO system shows that salt rejection is more variable than organic rejection. However, the degree of DOC removal was strongly affected by feed solution chemistry. Over the last 20 years, removing TOC by membrane filtration has become a challenging issue [21]. Shammiri and Dawas found that when the feedwater pH reduces from 7.2 to 7.0, even if no scale inhibitor has been added, the water recovery of SWRO plant has improved from 22.5 % to 34.2 %, without any damage to the membrane surface due to scaling [6]. Kurihara et al. designed a Brine Conversion System (BCS) that consists of two stages for SWRO desalination process, where 60 % water recovery has been attained, overall [22, 23]. Kim et al. designed a multi-stage RO system for the desalination of seawater at a 5 m^3/h pilot plant using micro filtration as a pre-treatment technique; the results showed that the water recovery successfully increased from 30 % up to 50 % [7]. Suess et al. reported that 14 % of TOC was adsorbed through the Ro system application [24], and Jin and Zimmerman suggested that the abiotic interaction between NOM and the carbonate aquifer matrix was a substantial controlling factor in NOM removal [25].

There are several mechanisms including charge repulsion, size exclusion, and hydrophobic interaction that affect the TOC removal by RO membrane system [26]. Natural Organic Maters are the most important parts of TOC. Previous studies have reported that membrane surface with a positive charge tends to adsorb negatively functional groups of NOM. It seems that the surface charge of the membrane plays an important role in the removal mechanism of TOC. Laine et al. reported that hydrophilic membranes showed better performance than hydrophobic ones in permeability [27]. It should be noted that the skin layer has the main role in both water flux and selectivity of the membranes. On the one hand, reducing the thickness of the membrane skin layer results in higher water flux, which can also have important implications for the retention of contaminants.

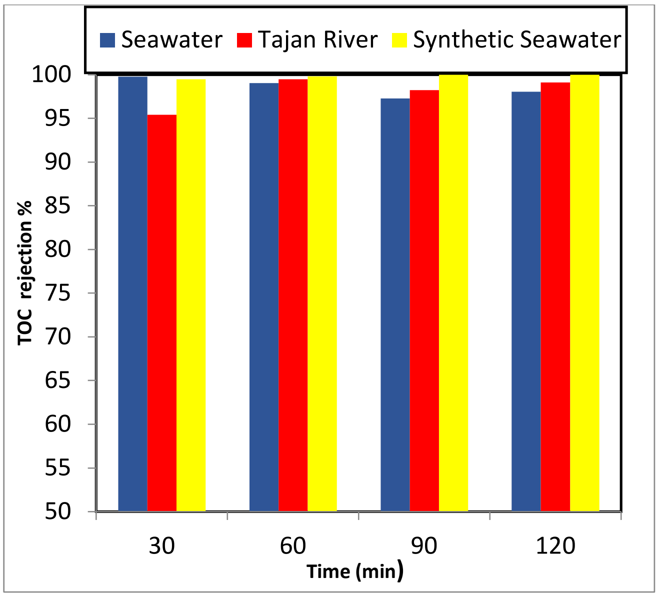

Figure 6. Comparison of the TOC rejection from Caspian seawater and Tajan River by the RO system at an applied 70 PSI pressure, 120 min contact retention time, and initial TOC concentration (35 g/L).

4. CONCLUSIONS

In this study, the feasibility and efficiency of removing salt and TOC simultaneously by applying the RO membrane system from the Caspian Sea and Tajan River were investigated and proven to be high. The suggested pilot was a simple design with materials that would significantly reduce salt and TOC (>95 %) from Caspian seawater as one of the important rivers whose effluent is flowing into the Caspian Sea (Tajan River). In addition, it is possible to achieve higher removal efficiency results in the optimum condition experiment (>98 %). Moreover, concerning the growing environmental pollution discharged into Caspian Sea such as industrial and agricultural effluents from rivers, finding a suitable alternative is necessary. These facts encourage the use of the proposed RO system on a commercial scale as tertiary treatment, and further additional research on the process is required to allow for a more detailed economic evaluation.

5. ACKNOWLEDGEMENT

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