



Assessment of different nanofiltration and reverse osmosis membranes for simultaneous removal of arsenic and boron from spent geothermal water

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ABSTRACT

One of the factors that determine agricultural crops' yield is the quality of water used during irrigation. In this study, we assessed the usability of spent geothermal water for agricultural irrigation after membrane treatment. Preliminary membrane tests were conducted on a laboratory-scale set up followed by mini-pilot scale tests in a geothermal heating center. In part I, three commercially available membranes (XLE BWRO, NF90, and Osmonics CK- NF) were tested using a cross-flow flat-sheet membrane testing unit (Sepa CF II, GE-Osmonics) under constant applied pressure of 20 bar. In part II, different spiral wound membranes (TR-NE90-NF, TR-BE-BW, and BW30) other than the ones used in laboratory tests were employed for the mini-pilot scale studies in a continuous mode. Water recovery and applied pressure were maintained constant at 60% and 12 bar, respectively. Performances of the membranes were assessed in terms of the permeate flux, boron and arsenic removals. In laboratory tests, the permeate fluxes were measured as 94.3, 87.9, and 64.3 L m⁻² h⁻¹ for XLE BWRO, CK-NF and NF90 membranes, respectively. The arsenic removals were found as 99.0%, 87.5% and 83.6% while the boron removals were 56.8%, 54.2%, and 26.1% for XLE BWRO, NF90 and CK-NF membranes, respectively. In field tests, permeate fluxes were 49.9, 26.8 and 24.0 L m⁻² h⁻¹ for TR-NE90-NF, BW30-RO and TR-BE-BW membranes, respectively. Boron removals were calculated as 49.9%, 44.1% and 40.7% for TR-BE-BW, TR-NE90-NF and BW30-RO membranes, respectively. Removal efficiencies of arsenic in mini-pilot scale membrane tests were all over 90%. Quality of the permeate water produced was suitable for irrigation in terms of the electrical conductivity (EC) and the total dissolved solids (TDS) for all tested membranes with respect to guidelines set by the Turkish Ministry of Environment and Urbanisation (TMEU). However, XLE BWRO, CK-NF and NF90 membranes failed to meet the required limits for irrigation in terms of boron and arsenic concentrations in the product water. The permeate streams of TR-BE-BW, TR-NE90-NF and BW30-RO membranes complied with the irrigation water standards in terms of EC, TDS and arsenic concentration while boron concentration remained above the allowable limit.

1. Introduction

Energy and water demand are the major challenges facing most nations in the world nowadays. The main reasons can be summarized as increases in population, industrialization and standards of living (Baba

et al., 2019). With increase in the global population, the water consumption per capita has also increased over the last century, resulting in a six-fold rise in groundwater withdrawals. Primary (for domestic use) and secondary (for agriculture and other commercial purposes) water demands are expected to put significant pressures on presently available

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water sources (Gude, 2016; Dana et al., 2019). Globally, the agri-food chain accounts for 80–90% of total global freshwater use and 70% alone is for the irrigation water (Bundschuh et al., 2017). Numerous studies have shown that volume of irrigation water is the first to be reduced instead of drinking or process water needs when the water supply has continued to be inadequate (Smith et al., 2015). A reduction in the irrigation water supplies leads to a reduction in the agricultural activities and thus affects livelihood, sustainability and the food security of any society.

In order to promote a sustainable development and to protect environment, there is an increasing pressure across the globe to increase use of the renewable energy sources (Zheng et al., 2015). In the 21st century, the renewable energy development primarily centered on biomass, solar, wind while lesser importance was attributed to hydro and geothermal energies due to the fact that prior was severely affected by climate and the latter was localized by nature.

Turkey is located within the Mediterranean Earthquake Belt, the complex deformation of which results from the continental collision between the African and Eurasian plates. As a result, Turkey has a considerably high level of geothermal energy potential with more than 60,000 MW due to its geological and tectonic settings (Ozbey-Unal et al., 2018; Baba and Sözbilir, 2012). Geothermal resources may be assessed for several uses, including energy production, horticulture, balneotherapy, recreation, heat pumps, cooling, aquacultural pond heating, industrial process heat, greenhouse, and space heating etc (Tomaszewska et al., 2017). Geothermal energy is considered to be safe, efficient and reliable as well as environmentally friendly if emissions caused by it are properly controlled and managed (Alimonti and Soldo, 2016). Furthermore, the geothermal fluids withdrawn for any purpose should be either re-injected back into the reservoirs or treated before use in order to ensure sustainable utilization. If these options are not exercised, the geothermal energy cannot be considered as a clean energy as stated by Melikoglu (Melikoglu, 2017).

Baseline water stress levels are becoming more severe as the water demand by sectors – public, agriculture, industry – exceed available freshwater resources (World Resources Institute, 2020). As a result, there is a paradigm shift regarding the water resources in recent years, assessing the use of the treated wastewater streams for various purposes. Since the agriculture sector needs high amounts of water, we would like to evaluate the untapped source of the spent geothermal brine for irrigation. Successful use of geothermal water for agricultural irrigation was performed in the Kebili region of Tunisia, where 95% of low enthalpy geothermal water with a salinity of 4 g L^{-1} has been directly used for greenhouse heating and oasis irrigation (Ben Mohamed, 2003). However, a detailed evaluation of its composition reveals that the spent geothermal brine has to be treated before use in irrigation.

The geothermal fluid is withdrawn from the aquifer with high flow rates during utilization and it contains B, As, NH_3 , SiO_2 in their neutral state and some noble gases along with Ca^{2+} , Mg^{2+} , Li^+ , HCO_3^- , SO_4^{2-} , CO_3^{2-} , F^- , I^- , K^+ , Na^+ and Cl^- at high concentrations. There are also some heavy metals such as Rb, Cr, Hg, Ag, Mn, Fe, Cu, Pb, Ni, Cs in geothermal brines (Ozbey-Unal et al., 2018; Awerbuch et al., 1976). Due to its high salt content, release of the geothermal fluids to the environment would evidently results in salinization and sodification of agricultural fields, the expense of which could well outweigh the benefits of the extracted energy. Tolerance to the salt content varies amongst the irrigated crops. While broccoli, tomato and beetroot can tolerate an EC of $2.5\text{--}2.8 \text{ mS cm}^{-1}$, okra and peas can only resist an EC of $1.0\text{--}1.7 \text{ mS cm}^{-1}$ (Shahbaz et al., 2012). Among the components listed, the most problematic ones are generally boron and arsenic, since their concentrations are usually found above the allowable limits.

High boron concentrations in irrigation water induce the boron toxicity with signs of the yellowish spots on leaves and fruits (Kabay et al., 2010; Yoshizuka et al., 2010). Boron also has an ability to form complexes with Cd, Ni, Pb, and Cu ions, which have a higher toxicity than those heavy metals alone when it presents in groundwater (Yavuz

et al., 2013). For that reason, the World Health Organization (WHO) sets a limit of 2.4 mg L^{-1} as the allowable concentration for boron in drinking water while the limit in irrigation water is 1 mg L^{-1} .

Arsenic is also a well-known toxic and cancer-causing metalloid, which is found in a wide variety of geothermal water sources. Contamination of aquatic bodies by geothermal water containing arsenic may hinder their use for drinking or irrigation purposes (Bundschuh and Maity, 2015). According to the guideline of World Health Organization (WHO) (2011) water for human consumption should not contain arsenic at a concentration greater than $10 \mu\text{g L}^{-1}$.

After extracting energy from the geothermal fluid, the spent geothermal water may reasonably be considered a potential option for agricultural irrigation and industrial applications. Though studies indicate that converting the processed geothermal water into a drinking water supply is theoretically possible, it requires knowledge of environmental and health repercussions (Gallup, 2007). As mentioned by Quist-Jensen et al. (2015), it is much easier to produce water for agricultural irrigation than for drinking purposes because it is difficult to meet all the required quality criteria for drinking water compared to irrigation water. Nevertheless, the different plants require different water quality standards, for that reason it is possible to tailor the reclaimed water when it is intended to be used for the irrigation purposes. The treatment cost of these saline fluids has to be included in any project that aims to utilize this resource (Ben Mohamed, 2003). Various separation processes have been proposed in addition to reverse osmosis (RO) to achieve desalination, including ion exchange, electrocoagulation, adsorption or continuous electrodeionization (CEDI) (Ruiz-García et al., 2019).

The aim of this study is the valorization of the spent geothermal water from a geothermal heating center for agricultural irrigation purposes. Pressure driven membrane processes such as nanofiltration (NF) and RO were employed to remove impurities from the spent geothermal water prior to agricultural irrigation. For that purpose, firstly laboratory membrane tests were conducted using XLE-BWRO, NF90 and CK-NF membranes by means of a cross-flow flat sheet membrane test system. Then, field tests were conducted using spiral-wound NE2540-90 (TR-NE90-NF), RE2540-BE (TR-BE-BW), and BW30-2540 (BW30-RO) membranes on a mini-pilot scale membrane treatment system installed on-site at the geothermal heating center. Qualities of the product water streams were evaluated by monitoring the boron and arsenic concentrations.

2. Materials and methods

2.1. Laboratory tests

A SEPA CF II (GE-Osmonics) cross-flow flat-sheet membrane test system was used to investigate performances of the flat sheet membranes (XLE BWRO, NF90, and CK-NF) with respect to their arsenic and boron rejections. The membrane test system accommodates flat sheet membranes with an active area of 140 cm^2 . The membrane properties were given in Table 1.

Geothermal water samples used in this study were taken from the geothermal wells of Izmir Geothermal Energy Co., Izmir, Turkey. Initially, hot geothermal water (around 100°C) was transferred to the laboratory with a 30 L of polyethylene container and was allowed to cool down. Properties of the geothermal water used in this study were listed in Table 2.

After the geothermal water was cooled down, it was transferred to the feed tank for the membrane test system. Each membrane was conditioned by soaking in deionized water for 24 h prior to testing. The cross-flow membrane test system used in this study was illustrated in Fig. 1. A constant pressure of 20 bar was applied throughout the tests. Each membrane test lasted for 4 h and this study was conducted in a close loop where permeate and concentrate streams were fed back to the feed tank. Samples were taken from the fresh feed before initiating each

Table 1
Membrane specifications given by the manufacturers (Lenntech 2020; Toray Advanced Materials Korea Inc. 2020).

Membrane	Producer	pH Range	Active area (m ²)	Maximum pressure (bar)	Maximum Temperature (°C)
XLE-BWRO	Dow Film Tech	2–11	0.014	41.0	45
NF90	Dow Film Tech	2–11	0.014	41.0	45
CK-NF	GE Osmonics	3–8	0.014	31.0	30
TR-NE90-NF	Toray Advanced Materials Korea Inc.	2–11	2.5	41.4	45
BW30-RO	Dow Film Tech	2–11	2.6	41.0	45
TR-BE-BW	Toray Advanced Materials Korea Inc.	2–11	2.5	41.4	45

Table 2
Geothermal water characteristics.

Parameter	Stored reinjection water	Fresh geothermal water
pH	8.40	8.52
Conductivity (µS/cm)	1836	1807
TDS (mg/L)	1224	1230
Total Alkalinity (mg CaCO ₃ /L)	480	509
HCO ₃ (mg CaCO ₃ /L)	460	475
HCO ₃ (mg/L)	561	580
CO ₃ (mg/L)	12	21
Cl (mg/L)	222	199
SO ₄ (mg/L)	163	164
F (mg/L)	5.4	7.0
Na (mg/L)	360	411
K (mg/L)	26	32
Mg (mg/L)	14	7.7
Ca (mg/L)	68	25
Li (mg/L)	1.2	1.4
Sr (mg/L)	0.65	0.65
B (mg/L)	5.7	12
Si (mg/L)	43.1	55
SiO ₂ (mg/L)	92	118
As (mg/L)	0.08	0.17
Fe (mg/L)	<0.1	<0.1
Ba (mg/L)	0.08	0.13

test. Samples were also collected from permeate and concentrate streams at 30, 60, 120, 180 and 240th min of each membrane test.

2.2. Pilot tests

A mini-pilot membrane test system installed at a geothermal heating center was used for field tests. The geothermal water sample used in these studies was the spent geothermal brine collected from the reinjection water reservoir (Fig. 2). Initially, spent geothermal water taken from the reinjection line with a temperature of around 60 °C was pumped to the reservoir. The stored reinjection water (with temperature about 30 °C) was pumped to our mini-pilot scale membrane test system having nanofiltration (NF) and reverse osmosis (RO) membranes as shown in Figs. 3 and 4. The feed water was first filtered through the sand filter and the cartridge filters for pretreatment and then stored in another tank prior to membrane tests. In order to eliminate precipitation of the insoluble salts on the membranes' surface, PRI-3000 A (Ropur) type antiscalant was employed at the concentration of 5 mg L⁻¹ of geothermal water using a dosage pump. Interestingly, there was some

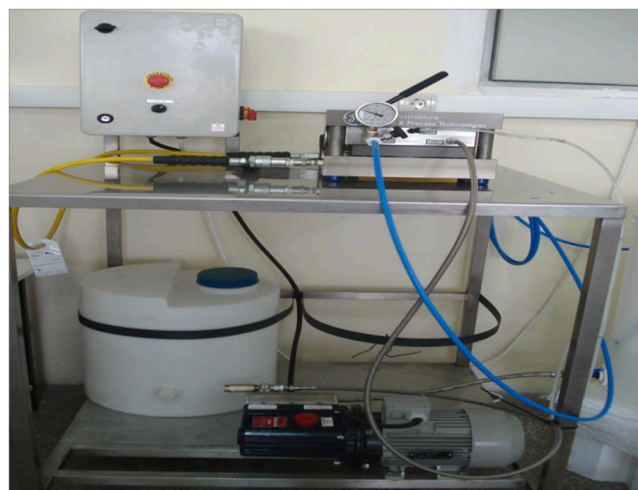


Fig. 1. Cross-flow membrane test system used in laboratory membranes tests.



Fig. 2. Spent geothermal water reservoir.

difference in the concentrations of some ions between the fresh geothermal water and the spent geothermal water stored for reinjection. For instance, there were significant changes in the concentrations of arsenic, boron, SiO₂, HCO₃⁻, Si, Ba, Ca²⁺, Mg²⁺, etc. The main reason for having a lower arsenic concentration in the spent geothermal water stored for re-injection was probably due to the adsorption of arsenic on the sediments of the reservoir (Morales-Simfors et al., 2020). Furthermore, as temperature of the reservoir decreases, rate of the arsenic sorption on sediments increases as explained in the literature (Bonte et al., 2013). Also, it is well known that silica precipitates at a lower temperature. There was also a significant decrease in concentration of boron in the spent geothermal water stored for re-injection. According to literature, boron makes complexes with heavy metals which tend to settle at the bottom of the reservoir (Ozbey-Unal et al., 2018; Cengeloglu et al., 2008).

For the preliminary field tests, the spiral wound NF (TR-NE90-NF) and RO membranes (TR-BE-BW and BW30-RO) were employed. Properties of the membranes used in this study were summarized in Table 1. Performances of each single membrane were investigated at constant pressure (12 bar). Water recovery was adjusted by decreasing the flow rate of concentrate stream (manually) and it was maintained constant at 60% throughout the tests for each membrane. Concentrate stream from the mini-pilot system was discharged to the reinjection line of the



Fig. 3. Mini-pilot scale membrane test system front view (left), side view (right).

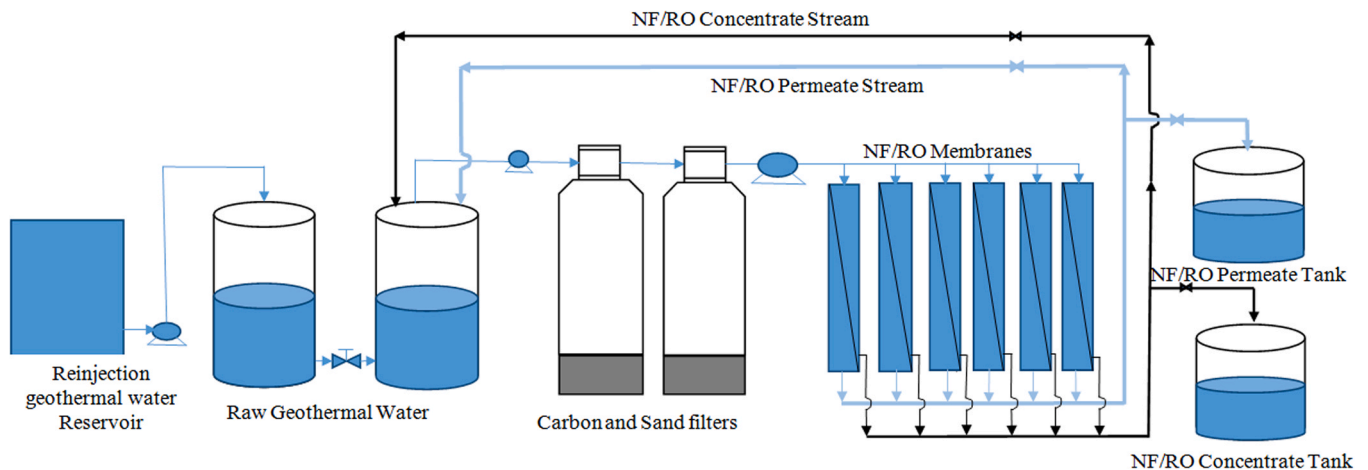


Fig. 4. Flow diagram of mini-pilot scale membrane test system installed at geothermal heating center.

heating center. Each membrane test was conducted in a continuous mode and lasted for 4 h.

Conductivity, pH, total dissolved substances (TDS), and salinity measurements were done using the Hach-Lange HQ14D model multi-meter during each test. Boron concentrations in feed, permeate and concentrate samples were determined by the curcumin method using Jasco SSE-343 V-530 UV/Vis spectrophotometer (in both laboratory and pilot studies). Shimadzu AA-7000 model atomic absorption spectrophotometer (AAS) was used to measure the concentration of total arsenic in feed, permeate and concentrate samples obtained in the laboratory membrane tests.

For the field membrane tests, concentrations of total-As, Na, K, Mg, Ca, SiO₂, Ba, Fe, Si, Sr and Li were measured using the inductively coupled plasma SM 3120 B (ICP) method. The total alkalinity (mg CaCO₃/L), HCO₃⁻ (mg/L), and CO₃²⁻ (mg/L) were determined using the standard method 2320B. Concentrations of SO₄²⁻ and F⁻ ions were determined using the standard methods with chemical kits while Cl⁻ ion concentration was determined using the standard iodometric method (4500-Cl B).

3. Results and discussion

3.1. Preliminary membrane tests in the laboratory

Laboratory membrane tests were conducted first to investigate the

performances of each membrane using a cross-flow flat sheet membrane test system, as shown in Fig. 1. Fresh geothermal water used in the laboratory tests was collected from a geothermal well in the field and its properties were given in Table 2. The applied pressure was maintained constant at 20 bar in all membrane tests and each membrane was assessed according to their permeate fluxes. As illustrated in Fig. 5, the

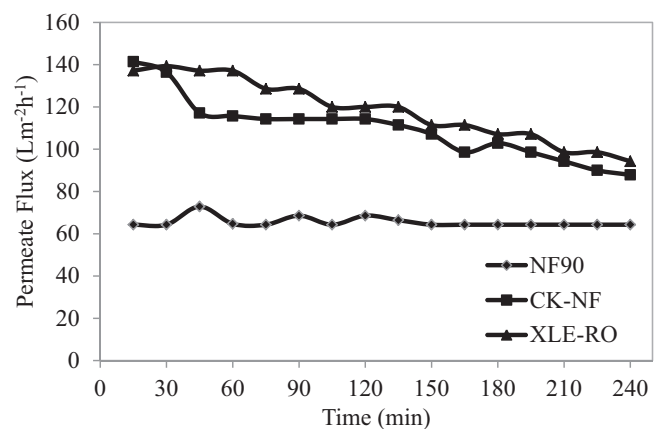


Fig. 5. Permeate fluxes vs. time plots for XLE-RO, NF90 and CK-NF membranes.

lowest flux was obtained using the NF90 membrane with a permeate flux of $64 \text{ L m}^{-2} \text{ h}^{-1}$. The permeate flux of the NF90 membrane increased up to $72 \text{ L m}^{-2} \text{ h}^{-1}$ in the first 45 min of operation, then it dropped to $64 \text{ L m}^{-2} \text{ h}^{-1}$ again at 60th min. The highest permeate fluxes of 141 and $137 \text{ L m}^{-2} \text{ h}^{-1}$ were obtained using the CK-NF and XLE-RO membranes, respectively, at the beginning of the membrane tests. There was a rapid decline in the permeate flux of CK-NF membrane from $141 \text{ L m}^{-2} \text{ h}^{-1}$ to $117 \text{ L m}^{-2} \text{ h}^{-1}$ in 45th min. Interestingly, the XLE-RO membrane showed a different trend in terms of permeate flux decline. Its permeate flux was stable in the first 60 min of operation ($137 \text{ L m}^{-2} \text{ h}^{-1}$) then the permeate flux began to decline from $137 \text{ L m}^{-2} \text{ h}^{-1}$ in 60th min to $128 \text{ L m}^{-2} \text{ h}^{-1}$ in 75th min of operation. A sudden change of the permeate flux in the first 60 min was expected because while the surface of membranes was free of contaminants initially, there was a rapid accumulation of contaminants on the surface of membranes as the operation started, which caused a rapid decline in the permeate flux. This phenomenon was also known as membrane conditioning. After 60 min, the membrane has stabilized and any flux decline was considered to be due to the membrane fouling. However, towards the end of the test (240th min), the permeate flux of the XLE-BWRO membrane was better than that of the CK-NF membrane with a permeate flux of $94 \text{ L m}^{-2} \text{ h}^{-1}$ while CK-NF membrane gave a permeate flux of $89 \text{ L m}^{-2} \text{ h}^{-1}$ at 240th min. It was observed that the permeate fluxes of the XLE-RO and CK-NF membranes declined towards end of the membrane test while the NF90 membrane showed a stable permeate flux throughout the study period. That means the NF90 membrane has more fouling propensity compared to the other membranes (XLE-RO and CK-NF) used in this study.

The XLE-BWRO membrane showed a better performance with a maximum 99% of arsenic removal followed by the NF90 and then CK-NF membranes exhibiting maximum arsenic removals of 88% and 83%, respectively (Fig. 6). The higher rejection of arsenic by the XLE-RO membrane than by the NF90 and CK-NF membranes can be attributed to the pore sizes of respective membranes. The XLE-RO membrane is a dense, nonporous membrane, where transport takes place through the polymer void-free spaces under a driving force (pressure difference) as reported by Kosutic et al. (2008). The NF90 and CK-NF membranes were reported to have an average porosity of 0.36 nm (Liu et al., 2018) and 0.57 nm (Mohammad et al., 2010), respectively. Therefore, the mechanism of contaminants' removal from the geothermal water is generally by the molecular sieving. That probably resulted in higher arsenic removal with the XLE-RO membrane than with the NF90 and CK-NF membranes. The average concentration of arsenic in permeates of the NF90 and CK-NF membranes were 26 and $18 \mu\text{g L}^{-1}$, respectively. Those values were higher than the recommended value in drinking water set by WHO (2011). These findings are in accordance with the study by Gonzalez et al. (2019), where the NF membranes failed to decrease arsenic concentration below the maximum allowable limit set by WHO.

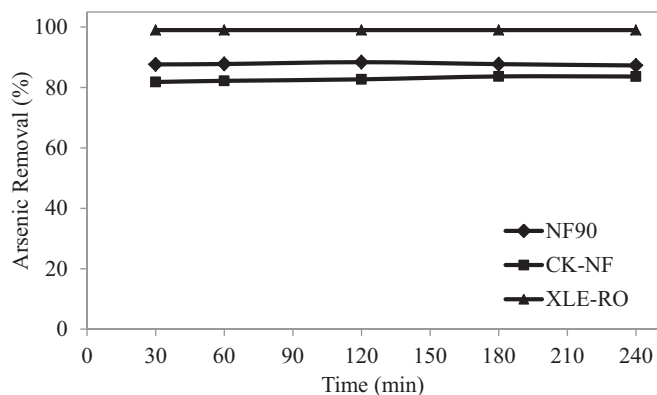


Fig. 6. Arsenic removals vs. time plots of XLE-RO, NF90 and CK-NF membranes.

Arsenic concentrations obtained in their study were reported as 35 and $21 \mu\text{g L}^{-1}$ for the NF270 and Alfa Laval NF membranes, respectively (at pH 8).

In our studies, the arsenic concentrations in the permeate samples of the XLE-BWRO membrane were below the detection limit ($2 \mu\text{g L}^{-1}$) of AAS. Hence, permeate of the XLE-BWRO membrane has complied with the maximum permissible value set by WHO for arsenic in drinking water. The RO membrane used in this study showed a better performance in terms of the arsenic removal than the findings reported by Tomaszewska and Bodzek (Tomaszewska and Bodzek, 2013), where only 84% of arsenic removal was obtained using the BWRO membrane at 11 bar of applied pressure.

Boron removal performances of the XLE-RO, CK-NF, and NF90 membranes were depicted in Fig. 7. The lowest boron removal was obtained with the CK-NF membrane with a maximum boron removal of 26%. The maximum boron removal by the XLE-RO membrane (60%) was higher than that by the NF90 (54%) and CK-NF membranes. The first one hour of operation in pressure driven membrane processes is regarded as membrane conditioning. When membranes come in contact with water, polymers on the surface of the membranes become swollen as it is saturated with water molecules. As a result of this interaction, voids in the matrix of the polymer on the active layer of the membranes decrease while separation via size-exclusion increases (Ben-David et al., 2006). Although there seemed to be some difference at the beginning of the test, there was no significant difference towards the end (240th min) in terms of boron removal by the XLE-BWRO (56%) and NF90 (54%) membranes. Tomaszewska and Bodzek (Tomaszewska and Bodzek, 2013) also reported a maximum boron removal from geothermal water as 48% using the BW30HR-440i RO membrane at a pressure of 11 bar. Boron concentration in geothermal water was reduced from 8.98 to 4.51 mg L^{-1} at the natural pH of the geothermal water.

In another study by Koseoglu et al. (2010), a 60% of boron removal was achieved at a pressure of 15.5 bar using the NF90 membrane at pH 8 with a reduction in boron concentration from 8.5 to 9.0 mg L^{-1} to 4.0– 4.5 mg L^{-1} by NF90 membrane.

In our laboratory-scale membrane tests, the minimum boron concentrations in the permeate samples after 4 h of operation were 4.7, 5.0 and 7.5 mg L^{-1} for the NF90, XLE-BWRO, and CK-NF membranes, respectively. Unfortunately, these results showed that the product water obtained using these membranes could not be utilized in agricultural irrigation due to the high boron concentration in the product water.

Boron removal increased towards the end of the run (240th min), especially in the case of the CK-NF and NF90 membranes due to narrowing pores of membranes by scalants. However, this has affected the productivity of membranes as well, causing a significant decline in the membrane flux especially for the CK-NF membrane (Fig. 5).

Boron in water is generally found in the form of weak boric acid and it dissociates following the reactions given below (Eqs. 1–3) at their respective pH values (Redondo et al., 2003; Kołtuniewicz and Drioli,

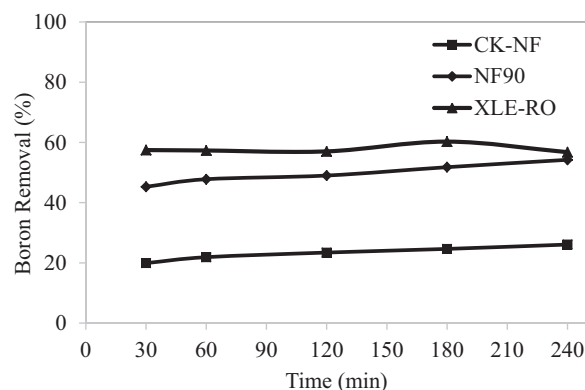


Fig. 7. Boron removal vs. time plots for XLE-RO, NF90 and CK-NF membranes.

2008):



Hence, at pH below the dissociation pH of boric acid, molecular form as boric acid is the most predominant form of boron found in water, and hydration of this molecule is weak due to the absence of ionic charge (Tomaszewska and Bodzek, 2013). As the pH of solution increases, percentage of boron in its ionic form also increases. Since pH of geothermal brine used in this study was in the range of 8.40–8.56, boron was expected to be dominant in its nonionic form (boric acid). Therefore, the mechanism of boron separation in this study was considered to be due to mostly by size exclusion. Boric acid is very small in its molecular form (~0.155 nm) compared with the polyamide aggregated pores (~0.350–0.450 nm) and network pores in the active layer of membranes (~0.100–0.300 nm) (Shultz et al., 2018). Furthermore, boric acid and water have similar transportation behavior during membrane separation processes due to similarities in hydrogen bondings (Roy et al., 2011). That was the reason for inadequate boron removal by membranes at geothermal water pH. Therefore, the removal of boron with the RO membrane requires elevated pH values. On the other hand, high pH is problematic in a single pass RO operation due to the high alkalinity and hardness resulting in the excessive consumption of caustic, which could cause scaling on the membrane surface (Sanguanpak et al., 2015). Therefore, the most practical way of reducing the boron concentration below limits comprises two or more passes where the first pass should be operated at the natural pH and second pass at elevated pH (Tomaszewska et al., 2018). Tomaszewska and Bodzek (2013) and Yavuz et al. (2013) have investigated effect of pH on boron removal from the geothermal water and their findings clearly showed higher boron removals at elevated pH.

3.2. Pilot tests in the geothermal heating center

Performances of different commercial spiral wound membranes (TR-NE90-NF, TR-BE-BW, and BW30-RO) were investigated using the spent geothermal water stored in a well before reinjection. The membrane tests conducted in this part were in a continuous mode and the applied pressure was 12 bar. The running time was 4 h, while 60% of water recovery was maintained constant throughout the mini-pilot scale tests. All tests were carried out at pH of geothermal water. Performances of each membrane were assessed with respect to their permeate fluxes along with their arsenic and boron removals (Fig. 8).

The TR-NE90-NF membrane gave the highest permeate flux with an average value of $49.9 \text{ L m}^{-2} \text{ h}^{-1}$ (Fig. 9). The average fluxes of the

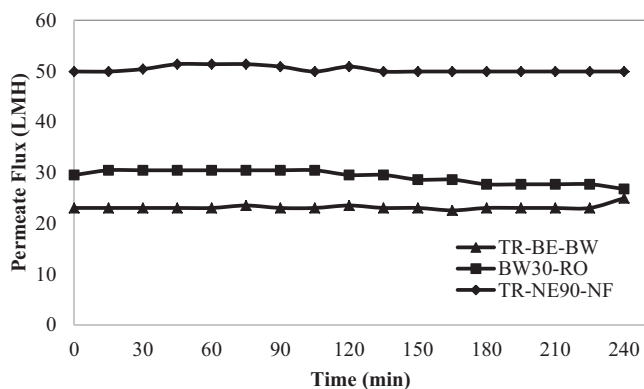


Fig. 8. Permeate flux vs. time plots for TR-NE90-NF, TR-BE-BW and BW30-RO membranes.

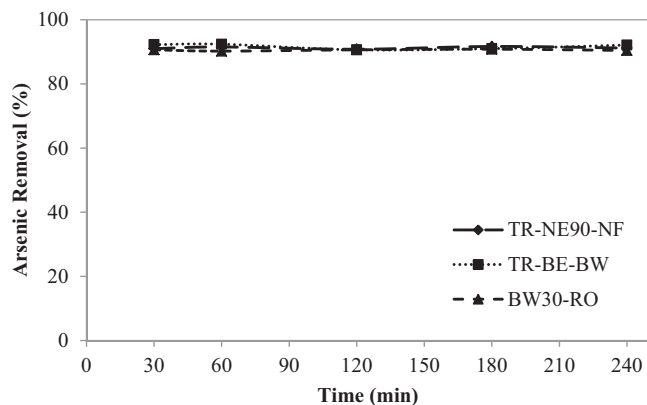


Fig. 9. Arsenic removal performances of TR-NE90-NF, TR-BE-BW and BW30-RO membranes.

BW30-RO and TR-BE-BW membranes were 26.8 and $24.0 \text{ L m}^{-2} \text{ h}^{-1}$, respectively. It was also observed that there was no significant flux decline throughout operation. Since the TR-NE90-NF membrane was NF membrane, it was expected this membrane to show superiority in terms of permeate flux when compared to RO membranes, which had no pores (dense membranes). In addition to that, the RO membranes had perfect hydrogen bonding and π - π stacking in their polyamide polymer, making it tight and hence less water and ions permeability are achieved with the RO membranes than with the NF membranes (Heo et al., 2013).

The results found in this study agreed well with the findings by Gündoğdu et al. (2019) and Sert et al. (2017) where the NF membranes showed better performance than the RO membranes in terms of the permeate flux when operated at the same operational conditions.

Several studies have demonstrated a successful removal of arsenic from different water sources by the NF and RO membranes (Chang et al., 2014; Abejón et al., 2015; Schmidt et al., 2016; Víctor-Ortega and Ratnaweer, 2017; Van der Bruggen and Vandecasteele, 2003; Yu et al., 2013). The arsenic removal by the RO membranes were reported as 78–90% by Chang et al. (2014) who tested different RO membranes under the various operational conditions. Abejón et al. (2015) studied on minimization of cost and the energy consumption with 90–97% of total arsenic removal in their research. Schmidt et al. (2016) investigated the total arsenic removal from groundwater for sustainable drinking water production using a pilot system, achieving 70–99% of total arsenic removal. Víctor-Ortega and Ratnaweer (2017) also found a 77–99% of total arsenic removal in a two-step RO membrane configuration for the production of drinking water. The NF membranes showed a remarkable performance, as reported by Chang et al. (2014) for arsenic removal as 65–80%. Van der Bruggen and Vandecasteele (2003) reported 47–98% of total arsenic removal using the NF membranes applied to remove pollutants from the surface water and groundwater samples. Fang and Deng (2014) also reported 43–96% of arsenic removal from the synthetic wastewater.

In our study, arsenic and boron removal performances of the tested membranes were depicted in Figs. 9 and 10, respectively. All membranes showed similar results with an average maximum arsenic removal of 91%. Concentrations of arsenic in the permeate streams were less than $10 \mu\text{g L}^{-1}$, which was also the maximum permissible level of arsenic in drinking water as recommended by WHO. Hence, the product water produced by all tested membranes could be considered as safe in terms of arsenic concentration.

According to the results obtained, the TR-BE-BW membrane gave the highest score with 49.9% of average boron removal from the spent geothermal water during 4 h of operation. The average boron removals obtained were 44.1% and 40.7% by the TR-NE90-NF and BW30-RO membranes, respectively. Boron concentrations in the permeate streams of three membranes at the end of operation were 3.3, 3.4 and

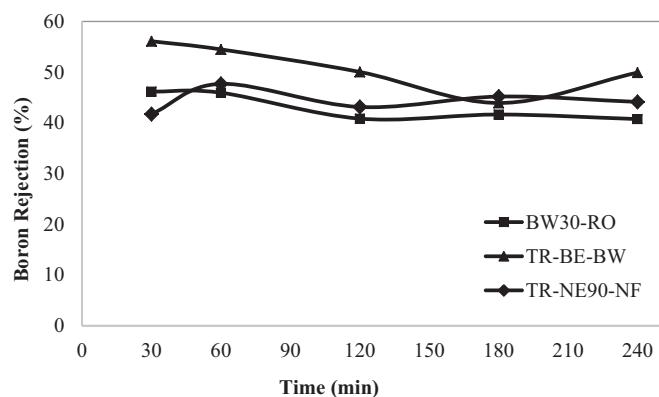


Fig. 10. Boron removal performances of TR-NE90-NF, TR-BE-BW and BW30-RO membranes.

3.9 mg L⁻¹ for BW30-RO, TR-NE90-NF and TR-BE-BW membranes, respectively. Interestingly, the TR-NE90-NF membrane exhibited a better performance to some extent in terms of boron removal than the BW30-RO membrane, even though the initial boron concentration in the study by using the TR-NE90-NF membrane was 6.1 mg L⁻¹ while it was 5.6 mg L⁻¹ for the BW30-RO membrane.

Many researchers have discussed the possibility of increasing boron removal from different water sources by modifying the surface charges of membranes or by adjusting the affinity of boron towards membranes (Roy et al., 2011; Li et al., 2018, 2020; Liu et al.; Wang et al., 2018). Koseoglu et al. (2010) investigated the performances of different commercially available membranes for reclamation of the geothermal water. According to their findings, the Toray UTC-70C membrane has shown better performance in terms of boron removal from geothermal water (90%) than the BW30-RO membrane (77%) at geothermal water pH and 20 bar of operational pressure. New generation Toray membranes have high boron rejections, probably due to membrane modification, and even their NF membrane is competing with the BW30-RO membrane, as demonstrated in that study.

Indeed, we have also obtained that TR-BE-BW membrane showed better boron removal than the BW30-RO membrane, although the boron concentration in the product water was the highest amongst the used membranes. The initial boron concentration in the feed stream during the study with TR-BE-BW membrane was 7.8 mg L⁻¹ which was slightly higher than the case for the tests using other two membranes. As boron concentration increases, the degree of dissociation of boric acid to its ionic form decreases at constant pH, pressure, and temperature (Dickson, 1990). As reported in the literature (Tomaszewska et al., 2017), percentage of boron in its ionic form decreased from 95.5% to 88.4% (pH was 10) when initial boron concentration in the geothermal water increased.

Although boron is an essential micronutrient for plants, a high concentration of boron in the irrigation water was reported to have a toxic effect on some plants (Roessner et al., 2006). Table 3 shows the sensitivity of some crops towards boron. Many countries have also continued to apply their own criteria in terms of the boron concentration in both drinking and irrigation water (Wang et al., 2014).

According to WHO (2011), the maximum level of boron concentration accepted in drinking water is 2.4 mg L⁻¹. Hence, water produced by these membranes (BW30-RO, TR-NE90-NF, and TR-BE-BW) were all above the allowable limits set by WHO. Therefore, if the product water is to be considered for agricultural irrigation, it had to be blended with freshwater having very low boron in it (e.g., tap water). For instance, blending permeate of the TR-BE-BW membrane which has 3.9 mg L⁻¹ of boron with fresh water at 50% ratio would decrease the concentration of boron in water below 2.0 mg L⁻¹, meeting the standards set by WHO.

Furthermore, reduction of boron in the permeate stream was also possible by increasing the operating pressure as done by Yavuz et al.

Table 3

The tolerances of different crops in response to boron in irrigation water (Yilmaz et al., 2008).

Sensitive (1 mg L ⁻¹)	Semi-sensitive (2 mg L ⁻¹)	Resistant (4 mg L ⁻¹)
Walnut	Sunflower	Asparagus
Plum	Potato	Date Palm
Pear	Cotton	Palm
Fig	Tomato	Sugar beet
Apple	Pea	Onion
Grape	Olive	Cabbage
Peach	Barley	Lettuce
Cherry	Corn	Carrot
Apricot	Wheat	Broad bean
Grape fruit	Cruet	Turnip
Lemon	Gruel	Bean
Orange		Clover

(2013). However, further increase of pressure would decrease concentration of both monovalent and divalent ions which are needed for plant growth. In addition, this would increase not only the concentration polarization on the membrane surface but also the process cost. Another option for further reduction of boron concentration in the product water was to adjust the pH of geothermal water. As mentioned in the literature (Tomaszewska and Bodzek, 2013; Yavuz et al., 2013), higher boron removal was obtained at high pH.

Some other physicochemical analyses conducted for the permeate streams in both laboratory-scale and mini-pilot scale tests were summarized in Table 4. Quality of all permeates obtained from the membranes employed has met the limits set by Turkish Ministry of Environment and Urbanisation (TMEU) in terms of EC and TDS (The technical procedure communication for wastewater treatment plants, 20.03.2010).

4. Conclusions

The simultaneous removal of arsenic and boron from fresh geothermal water and spent geothermal water in the geothermal heating center was conducted by laboratory-scale and mini-pilot scale tests, respectively. From the results obtained in the laboratory, it was observed that the XLE BWRO membrane gave a better performance than the CK-NF and NF90 membranes in terms of simultaneous removal of arsenic and boron from geothermal water. During the mini-pilot tests, it was seen that all the membranes tested were successful in the removal of arsenic from spent geothermal water to reduce its concentration below the recommended level (10 µg L⁻¹) set by WHO. However, these membranes failed to reduce the boron concentration to the permissible level at pH of the geothermal water. Further studies will be conducted to improve efficiency of membranes in the integrated configurations for reclamation and reuse of spent geothermal water in agricultural irrigation.

CRedit authorship contribution statement

Yakubu A. Jarma: Conducting research, performing the experiments, collecting data, writing the paper. **Aslı Karaoğlu:** Performing the experiments, collecting data. **Özge Tekin:** Performing the experiments, collecting data. **Alper Baba:** Management responsibility for the research activity. **H. Eser Ökten:** Reviewing and editing the paper. **Barbara Tomaszewska:** Critical review, commentary. **Kamil Bostancı:** Water analyses. **Müşerref Arda:** Provision of reagents, materials, instrumentation, or other analysis tools. **Nalan Kabay:** Oversight and leadership responsibility for the research activity planning and execution.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence

Table 4

Other quality results of the product waters of the membranes tested in laboratory and pilot tests.

Parameter	CK-NF	XLE BWRO	NF90	TR-NE90-NF	TR-BE-BW	BW30-RO	TMEU limits (Ministry of Environment and Urbanisation, Republic of Turkey, 2010)
EC ($\mu\text{S}/\text{cm}$)	140.00	36.48	55.00	42.00	23.60	50.00	<700
TDS (mg/L)	64.64	17.02	25.69	19.84	11.02	25.00	<500
Salinity (%)	0.06	0.02	0.02	0.02	0.01	0.02	–
pH	6.52	6.40	6.63	8.15	7.76	7.25	–

the work reported in this paper.

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