

57 (2016) 22980–22993 October



Forward osmosis for treatment of oil sands produced water: systematic study of influential parameters

Amrit Bhinder^a, Brian A. Fleck^a, David Pernitsky^b, Mohtada Sadrzadeh^{a,*}

^aDepartment of Mechanical Engineering, University of Alberta, 6-074 NINT Building, Edmonton AB T6G 2G8, Canada, Tel. +1 780 492 9099; Fax: +1 780 492 2200; email: sadrzade@ualberta.ca (M. Sadrzadeh) ^bSuncor Energy Inc., P.O. Box 2844, 150-6th Ave. SW, Calgary, AB T2P 3E3, Canada

Received 27 May 2015; Accepted 7 October 2015

ABSTRACT

Steam-assisted gravity drainage (SAGD) is a thermally enhanced heavy oil recovery method which is widely practiced for the bitumen extraction from the oil sands in Alberta, Canada. This study is the first application of forward osmosis (FO) for the treatment of SAGD produced water with the intent to reuse the treated water. The effects of temperature, flow rate, and pH of the feed water (produced water) and concentration and flow rate of the draw solution (salt solution) on the water flux as well as undesired diffusion of organic matter toward the draw solution were studied. Since no interaction between parameters is predicted, a fully saturated L_{16} Taguchi design was used to investigate these five parameters each at four levels. It was found that increasing the feed water temperature and the draw solution concentration enhanced the water flux. The change in feed pH did not have any significant effect on water flux. Increasing the flow rate of both the BFW and the draw solution reduced the concentration polarization layer on both sides, thus increased the efficiency of the separation process. Analysis of variance showed that the feed water temperature and the draw solution concentration were the most influential parameters. This study provides valuable insights regarding the feasibility of the FO process for the treatment of oil sands produced water.

Keywords: Forward osmosis; SAGD produced water; Taguchi method; ANOVA

1. Introduction

Steam-assisted gravity drainage (SAGD) is a thermally enhanced heavy oil recovery method which is widely practiced for bitumen extraction from oil sands in Alberta, Canada. In this process, steam is injected through a horizontal well into the bitumen-containing formation to decrease the viscosity of the bitumen and affect its extraction. An emulsion of steam condensate and heated bitumen flows down along the periphery of the steam chamber to the production well which is located below the injection well. This emulsion is then pumped to the surface where the bitumen and water are separated and the water is treated for reuse as boiler feed water [1].

Presented at EuroMed 2015: Desalination for Clean Water and Energy Palermo, Italy, 10–14 May 2015. Organized by the European Desalination Society.

1944-3994/1944-3986 © 2015 Balaban Desalination Publications. All rights reserved.

^{*}Corresponding author.

Fig. 1 shows a conventional SAGD water treatment plant. After initial bitumen water separation, the produced water is de-oiled by gravity skim tanks and induced static flotation (ISF) where the majority of the oil is removed from the water. Finally, the free oil content in the produced water is reduced to below 20 mg/L by passing it through walnut shell filters. The de-oiled water is then treated in a warm lime softener (WLS) for the removal of silica, followed by the removal of suspended solid in after filters. Finally, a weak-acid cation exchanger (WAC) is used to remove the residual multivalent cations like Ca²⁺ and Mg²⁺. After the treatment process, this water is then used as feed water in once through steam generators (OTSGs).

The conventional treatment process is not designed to remove TDS and dissolved organic matter. Typical SAGD water composition after conventional treatment is summarized in Table 1. For this harsh quality of water, having a high level of TDS and silica, to be used as boiler feed water, OTSGs are used. OTSGs are robust and are able to handle BFW containing higher level of TDS (<8,000 ppm) and Ca/Mg/Si than conventional drum boilers [2]. To compensate for the poor water quality, the quality of steam in the OTSGs is typically limited between 75 and 80% to ensure that a sufficient volume of water is available for cooling the inner surfaces of the tubing in the radiant section and to prevent any impurities in the BFW from being deposited on the tube surface. The steam is separated from the liquid at the exit of the steam generator and

Table 1 Specification of a typical BFW

Parameter	Specification
Conductivity (mS)	2.5–3.5
TDS (mg/L)	<3,500
pH	9.8–10.5
DOM (mg/L)	500-600
Silica (mg/L as SiO_2)	<75

is sent into the reservoir. The separated liquid is characterized by high concentration of dissolved solids and organics and is known as boiler blow down water (BBD). A portion of BBD is recycled back to the WLS, while the remainder is disposed off.

The inability of the conventional water treatment method to meet BFW specifications has resulted in the fouling and failure of boiler tubes in the field [3,4]. Fouling of the boiler tubes is a major problem in the SAGD operation as it results in periodic shutdowns of the operation for cleaning and maintenance, which results in a significant loss in production of bitumen [5]. Several research efforts are being made to better understand the mechanism of fouling, but still it is unclear whether the high levels of organic carbon deposited on the tubes, in addition to silica, are due to the deposition and coking of free and emulsified oil, or if they are due to temperature-related precipitation of organic matter.



Fig. 1. Flow diagram of a typical SAGD water treatment process (figure adapted from [6]).

In order to prevent fouling of the steam generators and to recycle as much water as possible, oil sands companies are seeking novel water treatment technologies. Membrane-based separation processes have been extensively used in treating produced water due to their clear-cut advantages over conventional processes, i.e. lower operating cost and energy consumption. Membrane-based treatment processes have been found to be efficient in treating water with high oil content, low mean particle size, and high flow rates. Numerous studies have been published on the application of microfiltration (MF) and ultrafiltration (UF) [7-12] for the treatment of oily produced water. For the separation of a broader range of contaminants, like silica, dissolved organic matter and salt, tighter membrane processes like nanofiltration (NF) and reverse osmosis (RO), have been used [13-17]. Several studies have also been published on the use of NF and RO for treating oil sands process affected water (OSPW) associated with surface mining for bitumen extraction, and recently, a study was conducted on the use of NF membrane processes for the SAGD produced water treatment [6], but to date no published studies have been found on the use of FO for the treatment of SAGD-produced water. In the present study, FO was investigated for the first time on model BFW obtained from a SAGD operation to reduce the concentration of TDS and dissolved organic matter.

Forward osmosis (FO) has gained ground in the field of water treatment and desalination in the past decade [18]. Several studies have been published in the last decade showing the wide scope of application of FO in different areas of water treatment, which include desalination of seawater and brackish water [19,20], concentration of landfill leachates [21], treatment of municipal [22–24], industrial wastewater [25,26], and processing of food and beverages [27,28].

Cath et al. [26] used seawater as a draw solution, due to its low cost and high availability in coastal regions, to produce drinking water using water from a domestic wastewater treatment plant and impaired surface water as feed. The main motive of this study was to investigate the performance of FO in conjugation with RO. Investigation of membrane fouling tendency, rejection of inorganic compounds, nutrients and also preliminary economic evaluation of the hybrid system were done. Great improvement was observed in terms of rejection, low membrane fouling and low cost with a dual-barrier FO-RO process. Hutchings et al. [29] tested the applicability of FO for reclaiming drilling wastewater for reuse as the base fluid for hydraulic fracturing. The system operated with a 26% w/w NaCl draw solution for treating drilling wastewater with a 5,000 ppm TDS а

concentration. The FO system was able to recover 70% of the water from a typical wastewater pit and a significant reduction in the concentration of heavy metals and salts was found in the recovered water. Another study was made by Hickenbottom et al. [30] on FO operation for the treatment of drilling mud and fracturing wastewater. Bench scale tests were performed on waste streams using a commercial CTA membrane and a 26% w/w NaCl draw solution. It was reported that at least 80% of the O&G drilling wastewater volume was recovered. A high rejection of inorganic and organic compounds was achieved during the pilot testing. Minimal irreversible fouling was observed and the effectiveness of osmotic backwashing to remove the fouling layer was also demonstrated.

The above-mentioned studies investigated the general feasibility of FO, but did not determine optimal performance parameters. The performance of the FO system depends upon various parameters like temperature, flow rate, draw solution concentration and the pH of the feed, which will affect the water flux as well as the rejection rate of the membrane. In this study, the effects of all major factors like temperature, flow rate and pH of the feed water and the concentration and flow rate of the draw solution on the FO of model BFW obtained from a SAGD operation were investigated. The undesired diffusion of organic matter toward the draw solution was also reported. Since no interaction between parameters was predicted, a fully saturated L₁₆ Taguchi design was used to investigate these five parameters each at four levels. Using a Taguchi experimental design, the results of the experiments were analyzed to identify (1) the optimal process conditions in terms of water flux, (2) the role of individual parameters, and (3) the response for the conditions which were not experimented. Analysis of variance (ANOVA) was used for the statistical analysis of the results and also to determine the contribution of each factor toward response variation.

2. Materials and methods

2.1. Preparation of feed

A model SAGD BFW (Table 2) was prepared by diluting BBD water obtained from a SAGD treatment plant located in the Athabasca oil sands region of Alberta, Canada. Hot samples were collected, shipped in sealed containers and were kept in an inert atmosphere with a nitrogen blanket. 950 g of BBD water was diluted five times to obtain the properties of BFW. About 1 M solutions of NaOH and HCl (Sigma Aldrich) were added to the model BFW to get the required pH of the solution.

Table 2 Properties of BFW modeled from BBD water

Parameters	BBD water	Modeled BFW
Conductivity (mS)	16.2	3.5
TDS (mg/L)	9,800	2,000
pН	10.5	10.0
TOC (mg/L)	2,700	550
Silica (mg/L as SiO_2)	125	35
Ca^{2+} (mg/L)	2.80	0.5
Mg^{2+} (mg/L)	0.64	0.15
Iron (mg/L)	0.70	0.25

2.2. Membrane

A semipermeable polyamide thin-film composite (TFC) membrane with embedded polyester screen support was acquired from Hydration Technology Innovation (HTI Albany, OR). As compared to the commonly available asymmetric cellulose triacetate (CTA) membranes, the TFC membranes are relatively new class of membranes for FO process and have superior performance than the CTA membranes.

Based on manufacturing data, the maximum operating temperature and workable pH range of the TFC membrane was 71 °C and 2–11, respectively. In the present work, the threshold operating conditions were deliberately reached to test the membrane filtration performance under the harsh conditions which would be expected during full-scale industrial application of the system.

2.3. Cross flow FO setup

A bench scale FO test cell system was obtained from Sterlitech Corporation to evaluate the performance of the FO membrane under varying operating conditions. The schematic view of the FO setup is shown in Fig. 2. The system consists of two 9 L tanks one for the draw solution and the other for the feed solution, connected to the FO membrane element cell. The effective filtration area of the cell is 140 cm^2 . Variable speed gear pumps were used to circulate the solution through the system. The flow rate, conductivity, and temperature were measured using digital sensors which came integrated with the system and the reading could be conveniently taken from the control panel screen on the system. A 16 kg weighing balance (Mettler Toledo, model: MS16001L, Switzerland) was used to measure the change in the weight of the draw tank. The temperature of the feed side was increased by circulating bath heater (Polyscience,

model: MX-CA11B, USA), while the temperature of the draw solution was controlled by a recirculating chiller (Polyscience, model: 6560M11A120C, USA).

2.4. Total organic carbon analysis

Draw solution samples collected at the end of the experiment were analyzed for total organic carbon (TOC), which is the indicator of the amount of dissolved organic carbon present in the SAGD produced water. TOC was measured by using a combustion type TOC analyzer (Shimadzu, model TOC-V; detection range 3–25,000 mg/L). The TOC analyzer is sensitive to high salt concentration in the solution, so the samples were diluted 10 times before they were sent for analysis.

2.5. Experimental methodology

To study the effect of all the influential parameters on the water flux one at a time, 4^5 numbers of experiments would have to be carried out. Instead, the Taguchi method was used for the experimental design to study the effect of all factors with minimum number of experiments. Five factors, each with four levels, were chosen for the experiment as shown in Table 3. The experimental matrix was designed by selecting a L₁₆ orthogonal array. The layout of the L₁₆ array used for the experimental design is presented in Table 4.

The Taguchi method utilizes orthogonal arrays to group the parameters affecting the result and the levels at which these parameters should be varied, thus significantly reducing the number of experimental configurations. The advantage of using orthogonal arrays is that it is possible to separate out the effect of each factor at a different level. For example, the mean response for temperature at 25, 40, 50 and 55 °C can be calculated by taking the average of the response for the experiments 1–4, 2–8, 8–12, and 8–16, respectively. The mean response for other factors at each level can be calculated in a similar way [31].

The results obtained from the set of experiments as described in the orthogonal array are analyzed in the Taguchi method using a statistical index of performance called signal-to-noise (S/N) ratio. The S/N ratio is the function of both the mean and the standard deviation of the result. Therefore, the S/N ratio is the logarithmic (to the base 10) ratio of the mean (result) to the deviation (noise) of the result from the desired value. Depending upon the quality characteristic to be optimized, there are three standard S/N ratios (i) bigger-is-best, (ii) smaller-is-best, and (iii) nominal-is-best [32]. In our study, the quality



Fig. 2. Schematic of bench scale FO setup.

Table 3 Controllable factors and their levels

	Level	s		
Factor	1	2	3	4
Feed temperature (°C)	25	40	50	55
Feed pH	8.5	9.5	10.5	11.5
Feed flow rate (LPM)	0.5	1.0	2.0	3.0
Draw conc. (M)	0.5	1.0	2.0	3.0
Draw flow rate (LPM)	0.5	1.0	2.0	3.0

characteristic to be optimized was the water flux, so the bigger-is-best response was chosen. For the biggeris-best response, the following relation is used to find the S/N ratio:

$$S/N = -10\log\left(\frac{1}{n}\sum_{i=1}^{n}\frac{1}{J_{w_i}^2}\right)$$
(1)

where *n* is the number of all observations used for the calculation of the S/N. Whatever may be the quality characteristic, the transformations of the S/N ratio are such that it is always interpreted as the bigger-thebetter [32]. After analyzing the signal-to-noise ratio, ANOVA is performed for estimating error variance and for determining the relative importance of various parameters.

2.6. Experimental procedure

Sixteen experiments were conducted as shown in Table 4 to evaluate the performance of the FO membrane. The initial draw (NaCl) solution and feed solution (model BFW) volumes were 2 and 4.75 L, respectively. The TFC membrane was operated in FO mode (i.e. the active layer was facing the feed solution). The experiments were performed under the given set of operating conditions for duration of 10 h and the flux of the draw solution and the conductivity change of the feed and draw solution were monitored throughout the experiment. The temperature of the draw solution was maintained at 28 ± 3 °C throughout all experiments. The average flux over a period of 10 h was used for analysis in our study.

3. Results and discussion

From our past experience and from related works in the literature, five important influential parameters that affect the water flux in FO were selected: temperature, draw concentration, flow rate of feed and draw solution and pH. Fig. 3(a) and (b) shows variation of flux, draw conductivity, and feed conductivity with time corresponding to experiment number 2 and 9 in Table 4. As can be seen, the flux remains constant throughout the experiment for run 2, while it decreases with time for run 9. For run 9, this decline in flux was partially due to exceeding the critical flux

Run no.	Feed temperature (°C)	Feed pH	Feed flow rate (LPM)	Draw conc. (M)	Draw flow rate (LPM)
1	25	8.5	0.5	0.5	0.5
2	25	9.5	1.0	1.0	1.0
3	25	10.5	2.0	2.0	2.0
4	25	11.5	3.0	3.0	3.0
5	40	8.5	1.0	2.0	3.0
6	40	9.5	0.5	3.0	2.0
7	40	10.5	3.0	0.5	1.0
8	40	11.5	2.0	1.0	0.5
9	50	8.5	2.0	3.0	1.0
10	50	9.5	3.0	2.0	0.5
11	50	10.5	0.5	1.0	3.0
12	50	11.5	1.0	0.5	2.0
13	55	8.5	3.0	1.0	2.0
14	55	9.5	2.0	0.5	3.0
15	55	10.5	1.0	3.0	0.5
16	55	11.5	0.5	2.0	1.0

Table 4 Experimental design using L₁₆ orthogonal array



Fig. 3. Water flux, draw conductivity, and feed conductivity as a function of time for experiment no 2 and 9 corresponding to Table 4.

of the FO membrane. At a flux higher than a critical value, the permeation drag pushes the foulants toward the membrane surface and consequently reduces the flux. Another reason behind such a sharp flux decline is simply reduction of the driving force due to the rapid concentration of the feed as well as dilution of the draw solution. For the case where the flux is low, there is not much change in the driving force, and hence the flux decline is less.

Table 5 shows the experimental results for average water flux and the corresponding S/N ratio calculated using Eq. (2). Each experiment was replicated twice to confirm the reproducibility of the results. To get a better understanding of the effect of each parameter, the Taguchi method uses the plots of marginal mean and

the S/N ratio, as shown in Figs. 4 and 5, respectively. These graphs are based on data given in Table 6. It should be noted that these graphs just show the trend effect of each factor, and it would be incorrect to use these graphs to predict values that were not experimented.

It can be seen from Figs. 4 and 5 that draw concentration, feed and draw flow rate and temperature have positive effect on the water flux. According to Fig. 5(b), increasing the pH from 8.5 to 10.5 slightly increases the flux but, thereafter it has no influence. From the Taguchi design, it can be concluded that the flux is highly dependent on the change in draw solution concentration and feed water temperature (Fig. 5(a) and (d)). These results are consistent with

Run no.	Feed temperature (°C)	Feed pH	Feed flow rate (LPM)	Draw conc. (M)	Draw flow rate (LPM)	Flux (LMH) Run 1	Flux (LMH) Run 2	Avg flux (LMH)	S/N (dB)
1	25	8.5	0.5	0.5	0.5	4.3	4.4	4.4 ± 0.0	12.8
2	25	9.5	1.0	1.0	1.0	6.8	5.9	6.7 ± 0.6	16.5
3	25	10.5	2.0	2.0	2.0	11.3	11.8	11.5 ± 0.4	21.2
4	25	11.5	3.0	3.0	3.0	20.1	18.5	19.3 ± 1.1	25.7
5	40	8.5	1.0	2.0	3.0	16.0	18.1	17.0 ± 1.5	24.7
6	40	9.5	0.5	3.0	2.0	21.0	19.8	20.4 ± 0.8	26.2
7	40	10.5	3.0	0.5	1.0	13.3	12.9	13.1 ± 0.3	22.3
8	40	11.5	2.0	1.0	0.5	13.4	12.3	12.9 ± 0.8	22.2
9	50	8.5	2.0	3.0	1.0	22.3	23.8	23.0 ± 1.1	27.3
10	50	9.5	3.0	2.0	0.5	19.5	20.8	20.1 ± 0.9	26.1
11	50	10.5	0.5	1.0	3.0	17.8	17.7	17.6 ± 0.1	24.9
12	50	11.5	1.0	0.5	2.0	13.3	14.6	13.9 ± 0.9	22.3
13	55	8.5	3.0	1.0	2.0	16.5	19.2	17.8 ± 2	25.0
14	55	9.5	2.0	0.5	3.0	16.6	15.5	16.0 ± 0.8	24.1
15	55	10.5	1.0	3.0	0.5	25.5	24.7	25.1 ± 0.6	28.0
16	55	11.5	0.5	2.0	1.0	19.5	20.0	19.8 ± 0.4	25.9

Table 5 Average response and S/N ratio for each run

the previous literature [33,34]. Increasing feed and draw flow rate slightly improved water flux (Fig. 5(c) and (e)). This is attributed to the decrease in the concentration polarization effect on both sides of the membrane.

3.1. Effect of feed temperature

The produced water generated by the SAGD operation is typically at a high temperature [6]. Here, we tried to test the membrane at temperatures other than room temperature to investigate the performance of FO process at condition as much similar to practical application. So the temperature of the feed solution was varied between 25 and 55 °C. Higher temperatures than 55 °C could not be achieved with the current setup. According to Fig. 4(a), an increase in temperature has a positive effect on the permeate flux. Change in the temperature of a solution influences its thermodynamic properties like viscosity and osmotic pressure as well as the diffusivity of dissolved solids. The osmotic pressure of a solution is given by:

$$\pi = icRT \tag{2}$$

where i is the Van't Hoff factor, c is the molar concentration, R is the universal gas constant and T is the absolute temperature. Based on Eq. (2), increasing the temperature of the feed solution will increase its osmotic pressure, which is not desirable because it decreases the net driving force in the FO process.

However, it was shown by Phuntsho et al. [33] that the increase in osmotic pressure with temperature is marginal and becomes insignificant when compared to the high osmotic pressure of the draw solution. Two main causes that contribute to flux enhancement by increasing the temperature are decrease in water viscosity and swelling of the membrane. With an increase in temperature there is a decrease in the viscosity of water from $0.893 \times 10^{-6} \text{ m}^2/\text{s}$ at 25°C to the $0.51 \times 10^{-6} \text{ m}^2/\text{s}$ at 55°C [35]. This increases the pure water permeability (A) of the membrane which is an inverse function of viscosity given by $A = 1/\mu R_{\rm m}$, where μ is the viscosity of the solution and $R_{\rm m}$ is the intrinsic hydraulic resistance of the membrane [36]. More importantly, more relaxation of the polymeric membrane matrix at elevated operating temperatures can intensify the membrane swelling with water, thereby allowing more water to pass through it.

3.2. Effect of draw solution concentration

From Fig. 4(c), it can be observed that the draw solution concentration has a strong influence on water flux. The standard flux equation in the FO process is given by:

$$J_{\rm w} = A(\pi_{\rm D} - \pi_{\rm F}) \tag{3}$$

where *A* is the pure water permeability, π_D and π_F are osmotic pressure of the draw and feed solutions respectively. As NaCl is well dissociated in the



Fig. 4. Effect of (a) temperature, (b) pH, (c) feed flow rate, (d) draw concentration, and (e) draw flow rate on water flux.

solution, it can be seen from Eq. (2) that increasing the concentration of the draw solution proportionally increases the osmotic pressure. So, higher permeate flux is observed with an increase in draw solution concentration due to the increased driving force (Eq. (3)). But in asymmetric membranes at higher concentration of draw solutions, a non-linear dependence of flux with driving force is observed, and the flux can no longer be predicted by Eq. (3). This is due to the phenomena of concentration polarization [34]. But

the Taguchi method was not able to capture this non-linearity in the flux behavior, possibly due to the simultaneous influence of other factors on the flux.

3.3. Effect of flow rates

Fig. 4(c) and (e) show the effect of the feed flow rate and the draw flow rate on the water flux. Both the feed flow rate and the draw flow rate showed similar trends on flux behavior. As can be seen, the flux is



Fig. 5. Effect of (a) temperature, (b) pH, (c) feed flow rate, (d) draw concentration, and (e) draw flow rate on S/N ratio.

almost unaffected by the flow rate when it is increased from 0.5 to 2 LPM, but there is a small jump in the flux when the flow rate is increased to 3 LPM. Flux enhancement at higher flow rates is well studied in the literature for pressure driven processes [37,38], and a similar explanation can be applied in case of FO. For the feed side, the increase in feed flow rate will reduce the thickness of the concentration boundary layer near the membrane thereby enhancing the mass transfer coefficient. Therefore, the severity of the external concentration polarization (ECP) is decreased at higher flow rates. Another factor which contributes to the enhancement of flux in real feed system is the reduced fouling tendency at higher flow rates [38,39].

The reason for moderate flux enhancement due to an increase in draw flow rate is not well established, since internal concentration polarization is the dominant phenomena on the draw solution which is not affected by the draw flow rate, as it occurs inside the porous support layer. A confirmation test (as will be

Factor	Level	Response (Flux)	Response (S/N)
Temperature (°C)	25	10.47	19.06
L	40	15.87	23.82
	50	18.47	25.13
	55	19.68	25.73
pН	8.5	15.57	22.39
•	9.5	15.81	23.21
	10.5	16.84	24.12
	11.5	16.27	24.03
Feed flow (LPM)	0.5	15.55	22.46
	1.0	15.50	22.86
	2.0	15.87	23.68
	3.0	17.57	24.57
Draw concentration (M)	0.5	11.65	20.38
	1.0	13.75	22.14
	2.0	17.11	24.45
	3.0	21.97	26.78
Draw flow (LPM)	0.5	15.61	22.25
· · · /	1.0	15.65	23.00
	2.0	15.72	23.67
	3.0	17.51	24.83

Table 6

Effect of influential parameters

shown later) showed a 28% increase in flux with a change in the draw flow rate from 1 to 3 LPM when other parameters remained constant. A plausible explanation can be made based on the model developed by Suh and Lee [40]. Previous researchers neglected the phenomena of ECP on the draw side but this phenomenon can be significant at low cross-flow velocities and high flux.

So, when the draw flow rate is increased, the ECP phenomenon is reduced, causing less difference in solution concentration in the bulk region and near the membrane. This consequently, leads to slightly higher salt concentration inside the porous support of the membrane.

But the improvement in flux cannot be explained just by the improvement in the mass transfer coefficient on draw side, another reasonable argument can be made on the basis of the dilution of the draw solution along the membrane by the inflow of permeate as given by Xu et al. [39]. The phenomena of dilution of the draw solution can be severe at low cross flow velocity and high permeate flux. Excessive dilution of the draw solution may lead to a decrease in concentration potential across the membrane and thus reduce water flux through the membrane. However at higher cross flow velocity the diluted draw solution is replenished quickly, leading to an enhancement in permeate flux. It should be noted that, Xu et al. performed their study on spiral wound membranes, which have a higher membrane surface area than the flat sheet membrane used in out experiment, thus achieving a much higher dilution factor.

3.4. Effect of pH

The pH of the BFW obtained from the SAGD plant varies between 9.8 and 10.5, so it was very important to test the performance of the membrane at these conditions. Therefore, the pH of the feed solution was varied from 8.5 to 11.5. There was a minimal change in flux when the pH was changed from 11.5 to 8.5 (Fig. 4(b)). But this was contrary to what has been observed in pressure-driven membrane processes. Sadrzadeh et al. [6] reported a 20% decrease in flux when the pH was changed from 10.5 to 8.5 in a NF separation process. It was observed that silica nanoparticles and organic matter precipitated on the membrane surface at lower pH due to decreased inter-particle repulsion as well as particle-membrane repulsion, thus accelerating the formation of a cake layer. This cake layer formation choked the membrane and hence decreased the permeate flux through it. However, according to the literature, the fouling layer formed in a FO process is less compact compared to that formed in a pressure driven membrane process [41]. Thus, there was no noticeable change in flux when the pH is decreased from 10.5 to 8.5. Hence, it can be concluded that FO was less susceptible to fouling by the BFW as compared to the pressure driven membrane process.

3.5. Rejection of organics

SAGD BFW has a wide variety of organic contaminants with different chemical properties and molecular weight [42]. With the wide range of molecular weights and chemical properties the removal of organic compounds from the BFW is a big challenge. So, the efficiency of FO TFC membrane was tested for the filtration of organic compounds. The samples of draw solution were collected after each experiment and analyzed for DOM. TOC results showed that the rejection of the organics was in the range of 85–96%. The TOC rejection did not show any particular trend indicating that the membrane had a stable performance even at operating condition close to its threshold limits.

4. ANOVA

Results obtained from Taguchi can be coupled with ANOVA to determine the relative significance of each parameter on the response and also to determine whether the variation in response is due to a change of parameter level or due to experimental noise. ANOVA uses sum of squares (SS), degree of freedom (dof), and mean square to find the associated *F*-value, which is then compared to the *F*-value obtained from the statistical table to check for the significance of a factor. A brief overview of the terms which we come across when using ANOVA is given below [32,43]:

The SS of a factor is given by:

$$SS_A = \sum_{i=1}^{K_A} \left(\frac{A_i^2}{n_{A_i}}\right) - \frac{T^2}{N}$$
(4)

where K_A is the number of levels of a factor A ($K_A = 4$ for all factors in this study), n_{A_i} is the number of all observations at level i of factor A ($n_{A_i} = 8$), A_i is the sum of all observations of level i of factor A, and T is the total sum of all observation. The SS of the error is computed using the following equation:

$$SS_e = SS_T - (SS_A + SS_B + \cdots)$$
(5)

where SS_T is the total SS and is given by:

$$SS_T = \sum_{i=1}^{N} J_{w_i}^2 - \frac{T^2}{N}$$
(6)

where *N* is the total no of observations. The SS and SS_T are the basic calculations needed for ANOVA. From these quantities, variance can be calculated by dividing the SS by the dof (*v*).

$$V_A = \frac{\mathrm{SS}_A}{v_A} \tag{7}$$

where v_A is the degrees of freedom of a factor A and is given by $v_A = K_A - 1$. From this, the *F*-value can be calculated as follows:

$$F_A = \frac{V_A}{V_e} \tag{8}$$

where V_e is the variance for the error term which can be obtained by calculating the error SS_e and dividing it by error degrees of freedom (v_e), i.e. $V_e = SS_e/v_e$, $v_e = v_T - (v_A + v_B + \cdots)$ and v_T which is total degrees of freedom equal to (N - 1). Now the calculated *F*-value is compared to the *F*-value obtained from the statistical tables at various risks (α) using v_A and v_e . If the calculated value of *F* is greater than the extracted one, then it can be concluded that the effect of the parameter is significant. Table 7 provides the statistical results based on the experimental data. The term *P* in the table is the percentage influence of each factor on the response.

Comparing the obtained value of *F* with the extracted value of *F* (*F* = 3.24) from the table at $\alpha = 0.05$, it can be concluded that the variance of all factors except pH is significant compared with the variance of error, and hence, all these factors have significant effects on the response. The *p*-value of the draw concentration and the temperature is very high

Table 7 Statistical results based on experimental data

	Flux–mean					
Factor	SS	dof	Variance	F	p (%)	
Draw concentration	486.5	3	162.1	203.5	51.1	
Temperature	401.4	3	133.8	167.9	42.1	
Feed flow rate	22.8	3	7.6	9.6	2.4	
Draw flow rate	20.6	3	6.8	8.6	2.16	
pН	7.5	3	2.5	3.1	0.7	
error	12.7	16	0.8	-	-	

Comparison of flux results obtained from confirmation test run and by Taguchi prediction												
Temperature (°C)			Draw		Flux (LMH)							
	pН	pН	pН	pН	pН	pН	pН	Feed flow rate pH (LPM)	concentration (M)	Draw flow rate (LPM)	Taguchi prediction	Experimental value
50	9.5	1	1	1	14.7	15.9	7.5					
25	9.5	1	1	3	8.6	8.6	0.1					
25	9.5	3	1	1	8.8	11.1	20.7					
25	11.5	1	1	1	7.2	7.1	1.4					

1

2

9.0

16.9

10.0

17.6

10

4.0

2

2



Fig. 6. (a) Flux at different draw concentration and temperature, (b) flux at different feed flow rates and temperatures, (c) flux at different draw flow rate and temperatures, and (d) flux at different pH and different draw concentration.

as compared to feed flow rate and draw flow rate which brings us to the conclusion that draw solution concentration and temperature are the most influential factors in the FO process.

Table 8

25

40

9.5

10.5 2

1

For optimizing the performance of the system, a balance should be established between water flux and rejection of dissolved organic matter. But for these experiments, the TOC results did not follow any trend and the rejection rate varied from 85 to 96%. Hence, water flux was chosen as the sole criteria for the optimization of performance. As a result, high temperature, high draw concentration, high feed and draw flow rates and the raw feed pH of 10.5 are recommended for maximizing the response. Finally, after finding the significant factors by using the Taguchi method, the response for all combination of levels could be predicted with considerable accuracy. These predictions should be confirmed by running confirmation experiments. The results of the confirmation runs and the predicted values by Taguchi methods are presented in Table 8. As can be observed, the predicted results and experimental data match relatively well. Hence, it is shown that acceptable results can be obtained from the reduced number of experiments. Some of the results predicted by Taguchi analysis are presented in Fig. 6.

5. Conclusion

The FO separation process was applied for the first time on SAGD BFW. The effect of all important factors (temperature, pH, draw concentration, feed and draw flow rate) on water flux was studied at the same time using a Taguchi experimental design. The TOC rejection was between 85 and 96%. The rejection of TOC did not follow any particular trend indicating that membrane performance was not affected by high operating temperature and pH. Water flux was chosen as the sole criteria for optimization of system performance. High temperature, high draw concentration, high feed and draw flow rates and a pH of 10.5 are recommended for optimal performance of the process. ANOVA results revealed that draw solution concentration and temperature were the most influential parameters.

Less membrane fouling was observed and minimal treatment of the feed was required in the FO operation as compared to RO and NF, which will ensure longer operations with the same membrane and reduced maintenance costs. Moderately high quality of water was achieved by the FO process at moderately high water flux, which demonstrates the high efficiency of this process for SAGD produced water treatment. It is with mentioning that, the efficiency of a FO operation is highly dependent on the type of draw solution and the regeneration process used for its recovery. The application of pressure-driven process to recover the draw solution will not bring upon a reduction in the energy consumption. Recent studies showed that using standalone pressure-driven process consumes less energy than the hybrid FO system [44]. Hence, it is inaccurate to say FO uses lower energy than RO because energy is required in separating the draw solutes from the solvents. However, FO can be operated by using waste heat (pinch analysis) instead of using a high-grade electrical energy that can reduce the dependence on fossil fuels. [45]. For this, one of the very promising draw solutions is the solution of ammonium bicarbonate in water that can be recovered by heating the draw solution up to 60 C which decomposes ammonium bicarbonate into ammonia and carbon dioxide gas. The draw solution can be regenerated by recombining these two gases in water at room temperature. Thus, waste heat can be used for a FO process to regenerate the draw solution lowering the overall energy consumption.

Altogether, in the near future, FO can be considered as an alternative to the conventional SAGD treatment processes especially when the efforts to develop an energy efficient regeneration process for the draw solution succeed.

References

- H. Peng, K. Volchek, M. MacKinnon, W.P. Wong, C.E. Brown, Application on to nanofiltration to water management options for oil sands operation, Desalination 170 (2004) 137–150.
- [2] P. Gosselin, S.E. Hrudey, M.A. Naeth, A. Plourde, R. Therrien, G. Van Der Kraak, Z. Xu, Environmental and health impacts of Canada's oil sands industry, The Royal Society of Canada, Ottawa, Canada 2010.
- [3] D.W. Jennings, A. Shaikh, Heat-exchanger deposition in an inverted steam-assisted gravity drainage operation. Part 1. Inorganic and organic analyses of deposit samples, Energy Fuels 21 (2007) 176–184.
- [4] S. Wang, E. Axcell, R. Bosch, V. Little, Effects of chemical application on antifouling in steam-assisted gravity drainage operations, Energy Fuels 19 (2005) 1425–1429.
- [5] T. Pugsley, D. Pernitsky, J. Grundler, E.E. Johnsen, Fouling of heat transfer surfaces in a steam assisted gravity drainage (SAGD) *in situ* facility for the recovery of oil sands bitumen., Proc. International Conference on Heat Exchanger Fouling and Cleaning—2013, Budapest, Hungary (2013), pp. 116–123.
- Budapest, Hungary (2013), pp. 116–123.
 [6] M. Sadrzadeh, J. Hajinasiri, S. Bhattacharjee, D. Pernitsky, Nanofiltration of oil sands boiler feed water: Effect of pH on water flux and organic and dissolved solid rejection, Sep. Purif. Technol. 141 (2015) 339–353.
- [7] B.A. Farnand, T.A. Krug, Oil removal from oilfield produced water by cross flow ultrafiltration, J. Can. Pet. Technol. 28 (2000) 18–24.
- [8] A. Zaidi, K. Simms, S. Kak, The use of microultrafiltration for the removal of oil and suspended solids from oilfield brines, Wat. Sci. Tech. 25 (1992) 163–176.
- [9] I.W. Cumming, R.G. Holdich, I.D. Smith, The rejection of oil using an asymmetric metal microfilter to separate an oil in water dispersion, Water Res. 33 (1999) 3587–3594.
- [10] E. Gorouhi, M. Sadrzadeh, T. Mohammadi, Microfiltration of oily wastewater using PP hydrophobic membrane, Desalination 200 (2006) 319–321.
- [11] R.S. Faibish, Y. Cohen, Fouling and rejection behavior of ceramic and polymer-modified ceramic membranes for ultrafiltration of oil-in-water emulsions and microemulsions, Colloids Surf., A: Physicochem. Eng. Aspects 191 (2001) 27–40.
- [12] T. Bilstad, E. Espedal, Membrane separation of produced water, Water Res. 34 (1996) 239–246.

- [13] Z. Wang, Y. Zhao, J. Wang, S. Wang, Studies on nanofiltration membrane fouling in the treatment of water solutions containing humic acids, Desalination 178 (2005) 171–178.
- [14] K.L. Jones, C.R. O'Melia, Protein and humic acid adsorption onto hydrophilic membrane surfaces: Effects of pH and ionic strength, J. Membr. Sci. 165 (2000) 31–46.
- [15] C.A. Dyke, C.R. Bartels, Removal of organics from offshore produced waters using nanofiltration membrane technology, Environ. Prog. 9 (1990) 183–186.
- [16] F.T. Tao, S. Curtice, R.D. Hobbs, J.L. Sides, J.D. Wieser, C.A. Dyke, D. Tuohey, P.F. Pilger, Reverse osmosis process successfully converts oil field brine into freshwater, Oil Gas J. 38 (1993) 88–91.
- [17] M. Çakmakce, N. Kayaalp, I. Koyuncu, M. Cakmakce, Desalination of produced water from oil production fields by membrane processes, Desalination 222 (2008) 176–186.
- [18] T. Cath, A. Childress, M. Elimelech, Forward osmosis: Principles, applications, and recent developments, J. Membr. Sci. 281 (2006) 70–87.
- [19] P. Venketeswari, O.S. Leong, N.H. Yong, Seawater desalination using forward osmosis process, J. Water Reuse Desalin. 4 (2014) 34.
- [20] Q. Zhao, N. Chen, D. Zhao, X. Lu, Thermoresponsive magnetic nanoparticles for seawater desalination, ACS Appl. Mater. Interfaces 5 (2013) 11453–11461.
- [21] R. York, R. Thiel, E. Beaudry, Full-scale experience of direct osmosis concentration applied to leachate management, in: 6th International Waste Management Landfill Symposium, Cagliari, Italy, 1999, pp. 359–366.
- [22] E. Cornelissen, D. Harmsen, K. Dekorte, C. Ruiken, J. Qin, H. Oo, L.P. Wessels, Membrane fouling and process performance of forward osmosis membranes on activated sludge, J. Membr. Sci. 319 (2008) 158–168.
- [23] N.C. Nguyen, S.-S. Chen, H.-Y. Yang, N.T. Hau, Application of forward osmosis on dewatering of high nutrient sludge, Bioresour. Technol. 132 (2013) 224–229.
- [24] X. Zhang, Z. Ning, D.K. Wang, J.C. Diniz da Costa, Processing municipal wastewaters by forward osmosis using CTA membrane, J. Membr. Sci. 468 (2014) 269–275.
- [25] S. Zhang, P. Wang, X. Fu, T.-S. Chung, Sustainable water recovery from oily wastewater via forward osmosis-membrane distillation (FO-MD), Water Res. 52 (2014) 112–121.
- [26] T.Y. Cath, N.T. Hancock, C.D. Lundin, C. Hoppe-Jones, J.E. Drewes, A multi-barrier osmotic dilution process for simultaneous desalination and purification of impaired water, J. Membr. Sci. 362 (2010) 417–426.
- [27] K.B. Petrotos, H.N. Lazarides, Osmotic concentration of liquid foods, J. Food Eng. 49 (2001) 201–206.
- [28] M.I. Dova, K.B. Petrotos, H.N. Lazarides, On the direct osmotic concentration of liquid foods. Part I: Impact of process parameters on process performance, J. Food Eng. 78 (2007) 422–430.
- [29] N.R. Hutchings, E.W. Appleton, R.A. Mcginnis, Making high quality frac water out of oilfield waste, in: SPE Annual Technical Conference and Exhibition, Society of Petroleum Engineers, Florence, Italy, 2010, pp. 1–10.
- [30] K.L. Hickenbottom, N.T. Hancock, N.R. Hutchings, E.W. Appleton, E.G. Beaudry, P. Xu, T.Y. Cath,

Forward osmosis treatment of drilling mud and fracturing wastewater from oil and gas operations, Desalination 312 (2013) 60–66.

- [31] W.H. Yang, Y.S. Tarng, Design optimization of cutting parameters for turning operations based on the Taguchi method, J. Mater. Process. Technol. 84 (1998) 122–129.
- [32] M. Sadrzadeh, A. Razmi, T. Mohammadi, Separation of different ions from wastewater at various operating conditions using electrodialysis, Sep. Purif. Technol. 54 (2007) 147–156.
- [33] S. Phuntsho, S. Vigneswaran, J. Kandasamy, S. Hong, S. Lee, H.K. Shon, Influence of temperature and temperature difference in the performance of forward osmosis desalination process, J. Membr. Sci. 415–416 (2012) 734–744.
- [34] J.R. McCutcheon, M. Elimelech, Influence of concentrative and dilutive internal concentration polarization on flux behavior in forward osmosis, J. Membr. Sci. 284 (2006) 237–247.
- [35] J. Kestin, M. Sokolov, W.A. Wakeham, Viscosity of liquid water in the range -8°C to 150°C, J. Phys. Chem. Ref. Data 7 (1978) 941.
- [36] X. Jin, A. Jawor, S. Kim, E.M.V. Hoek, Effects of feed water temperature on separation performance and organic fouling of brackish water RO membranes, Desalination 239 (2009) 346–359.
- [37] P. Bacchin, D. Si-Hassen, V. Starov, M. Clifton, P. Aimar, A unifying model for concentration polarization, gel-layer formation and particle deposition in cross-flow membrane filtration of colloidal suspensions, Chem. Eng. Sci. 57 (2002) 77–91.
- [38] T.H. Chong, F.S. Wong, A.G. Fane, Implications of critical flux and cake enhanced osmotic pressure (CEOP) on colloidal fouling in reverse osmosis: Experimental observations, J. Membr. Sci. 314 (2008) 101–111.
- [39] Y. Xu, X. Peng, C.Y. Tang, Q.S. Fu, S. Nie, Effect of draw solution concentration and operating conditions on forward osmosis and pressure retarded osmosis performance in a spiral wound module, J. Membr. Sci. 348 (2010) 298–309.
- [40] C. Suh, S. Lee, Modeling reverse draw solute flux in forward osmosis with external concentration polarization in both sides of the draw and feed solution, J. Membr. Sci. 427 (2013) 365–374.
- [41] Y. Kim, M. Elimelech, H.K. Shon, S. Hong, Combined organic and colloidal fouling in forward osmosis: Fouling reversibility and the role of applied pressure, J. Membr. Sci. 460 (2014) 206–212.
- [42] S. Guha Thakurta, A. Maiti, D.J. Pernitsky, S. Bhattacharjee, Dissolved organic matter in steam assisted gravity drainage boiler blow-down water, Energy Fuels 27 (2013) 3883–3890.
- [43] T. Mohammadi, A. Moheb, M. Sadrzadeh, A. Razmi, Separation of copper ions by electrodialysis using Taguchi experimental design, Desalination 169 (2004) 21–31.
- [44] D.L. Shaffer, J.R. Werber, H. Jaramillo, S. Lin, M. Elimelech, Forward osmosis: Where are we now? Desalination 356 (2015) 271–284.
- [45] R.L. McGinnis, M. Elimelech, Global challenges in energy and water supply: The promise of engineered osmosis, Environ. Sci. Technol. 42 (2008) 8625–8629.