Forward osmosis for the concentration and reuse of process saline wastewater

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Abstract

Salt-contaminated soil generated by industrial activities has become a problem of concern over the last decades. While existing treatment technologies for these soils are efficient, the wastewater produced remains the major drawback. In this study, a novel application of forward osmosis (FO) was tested. The FO process operated in osmotic dilution mode was investigated in order to concentrate saline soil treatment wastewater and produce valuable process water. FO was chosen for this application since it is a low-energy technology and process adapted for complex liquid streams. Results from this study indicate that FO operated in osmotic dilution mode with a 1 M CaCl_2 draw solution is able to recover 50% of the initial wastewater volume with an initial total dissolved solids concentration of 16.3 g/L. Following treatment, the draw solution can be reused as a cationic exchange solution with CaCl_2 concentrations between 0.41 M and 0.44 M. No important solute transport from the feed to the draw solution was observed, except for some potassium drawn by the draw solution. This study demonstrated the potential of integrating the FO technology in the soil treatment industry in order to enhance water reuse in water-intensive processes.

Keywords

Forward osmosis, wastewater, reuse, concentration, case study

Graphical Abstract

1. Introduction

Salt-contaminated soil has become a worldwide problem of concern over the last decades and is present in more than 100 countries. In 1989, according to the available data at the time, approximately 932.2 million hectares of land were salt impacted[1]. Unlike hydrocarbon or heavy metal contamination, salt does not have a pathological connotation. However, salt can be harmful to the environment by changing the physical properties of the soil, resulting in a reduction of aquifer recharge or a lixiviation of salts in groundwater, thereby reducing the reserves of fresh water[2]. The origin of salt-affected soils is different depending on location. This paper emphasizes punctual salinization, which is the result of accidental spills[3] or poor industrial practices[4]. One of the industries responsible for a large part of punctual salinization around the world is that of oil and gas. When exploiting an oil reserve, hydrocarbons are not the only product extracted. Brine water, which can have a salt concentration higher than sea water, is also present[5]. About 8 to 10 barrels (1270 to 1590 L) of brine water are extracted for each barrel of crude oil produced.[6] During the first half of the 20th century, this water was discharged straight to the environment or, at best, contained in basins without retention devices[7].

In 2001, the Environmental Sciences Division of the Alberta Government published the *Salt Contamination Assessment & Remediation Guidelines* in which different remediation technologies are proposed. According to this document, the most suitable treatment process for site remediation is the replacement of sodium with calcium. This exchange is done by increasing the calcium concentration in the soil with the addition of a calcium additive[8]. The major drawbacks of this technique are the consumption of a large volume of fresh water and the management of process water. The wastewater produced by the treatment process is transported to a deep well according to the regulations[9]. Since many of the contaminated sites are located in remote areas with scarce fresh water supplies, these local water supplies are greatly depleted through the process of soil treatment and soil washing. In addition, the distance from the water source and the disposal well for process water can be prohibitive to the technology due to transportation costs. In the ongoing effort to reduce water consumption and management costs, it is necessary to develop a dewatering system that fulfills the following requirements: low-energy, low-cost and field-scale size.

Forward osmosis (FO), a water treatment technology that has recently gained research interest, meets these requirements. Forward osmosis is a technical term describing the natural phenomenon of osmosis which is the transport of water molecules across a semi-permeable membrane[10]. In a FO separation system, pure water from the solution needing to be concentrated (called feed solution or FS) is transported through the membrane using a highly concentrated solution (called draw solution or DS).

Therefore, unlike other membrane separation processes, FO does not depend on high hydraulic pressure to withdraw water from the feed solution. Process performance is also less likely to deteriorate in cases where foulant compaction is related to applied hydraulic pressures[13, 14]. Because of this low-fouling characteristic, FO has many potential applications where other membrane processes have failed. The main categories of applications are: wastewater

treatment [15, 16], water purification [17], seawater desalination [18], power generation [19] and specific industrial applications [20-22].

When used for water treatment or for wastewater volume reduction, FO can be operated in different modes. In applications in which high quality product water is not required, FO can be operated in an osmotic dilution mode, where water drawn from wastewater through the membrane dilutes the DS. The two end products are the diluted DS, which can be valorised, and the concentrated wastewater. The osmotic dilution mode is illustrated in Fig. 1a. In applications where high quality product water is required, FO can be coupled with a process such as RO to systematically re-concentrate the DS and produce a high quality permeate. In the latter case, FO acts as a high-level pre-treatment step before the desalination process; this pre-treatment mode is illustrated in Fig. 1b.[23, 24]

Fig. 1a : Osmotic dillution mode

Fig. 1b : Pre-treatment mode

Fig. 1 FO process operation modes: Fig. 1a FO process operated in osmotic dilution mode, Fig. 1b FO process operated in pre-treatment mode

The operation mode of greater interest for the treatment of saline soil treatment wastewater is the osmotic dilution mode (Fig 1a) using a highly concentrated CaCl_2 solution as DS produced from the CaCl_2 which is already used in the soil treatment process as described in this study. The resulting diluted DS becomes a liquid calcium amendment that can be reused for soil treatment.

Similarly to RO membranes, FO membranes act as a barrier to most salts, organic contaminants and suspended particles [21]. However, because a FO system operates with low or no hydraulic pressure, their configurations are different. The main distinctions between FO and RO membranes is the FO membrane's lack of the thick support layer that gives physical resistance to hydraulic pressure and the hydrophobicity of the membranes. Most RO membrane are made of polysulphone polymers[25], a hydrophobic material, limiting the wetting of the support. Specially designed FO membranes increase water flux in osmotic driven systems but are still affected by CP, which is the accumulation of solute at the surface of a membrane.

Because of CP, the osmotic pressure difference across the active layer is much lower than the bulk osmotic pressure difference resulting in much lower flux than expected. In systems where no hydraulic pressure is applied, both external CP (ECP) and internal CP (ICP) exist [10].

ECP is observed in pressure-driven membrane processes when convective permeate flow causes a build-up of solute on the membrane's active layer surface. This form of CP is not limited to pressure-driven processes, and also occurs during osmotic-driven membrane processes, with both membrane orientations.

When operating in FO mode (selective layer facing the FS), solutes accumulate on the surface of the selective layer; this may be called concentrative ECP and is similar to CP observed in RO. When operating in PRO (pressure retarded osmosis) mode (selective layer facing the DS), the draw solution in contact with the membrane is diluted by the permeating water; this is called dilutive ECP. In both cases, the water flux across the membrane is reduced due to the reduction of the effective osmotic driving force. ECP can be controlled hydrodynamically by increasing water velocity and turbulence on the surface of the membrane [14, 17].

ICP is associated to osmotic-driven membrane process with asymmetric membranes consisting in a dense selective layer and a porous support layer. Diffusion attracts solutes from the draw solution toward the feed solution, creating an accumulation inside the membrane layers. This phenomenon is particularly important in FO when operating in FO mode: the porous layer becomes a thick polarized layer, substantially reducing the osmotic driving force between the feed and the draw solution. Extensive research has reported higher water fluxes in PRO-mode, attributed to less severe ICP. However, this orientation is more prone to membrane fouling [10, 26]. Unlike ECP, ICP cannot be controlled by operation factors [17].

In this study, we perform the first evaluation of the potential of the FO process to treat wastewater generated during the treatment of salt-contaminated soils. The use of this technology in the soil treatment process has the potential of providing a more energy-efficient solution to conventional desalination technologies such as RO by using the difference in osmotic pressure of two solutions as the primary energy source. The main objective was to evaluate the performance of the FO process operated in osmotic dilution mode to concentrate saline soil treatment wastewater and produce valuable process water. Results also allowed identifying the most suitable commercially available membrane for this application as well as key design parameters, such as water flux and salt permeation. Opportunities and challenges for large-scale application of this technology are also discussed.

2. Material and methods

2.1. Commercial FO membranes

The semi-permeable membranes used were acquired from Hydration Technology Innovation (HTI) and Porifera. Two Basic FO Membrane Kits were bought from HTI Water: the thin film composite (TFC) membrane kit composed of 929 cm² membranes identified as HTI OsMem™ TFC-ES and the cellulose triacetate (CTA) membrane kit composed of 929 cm² membranes identified as HTI OsMem™ CTA-ES and 929 cm² membranes identified as HTI OsMem™ CTA-NW2. Porifera kindly provided 929 cm² high flux forward osmosis membranes identified as FOMEM-0513.

2.2. Bench-scale forward osmosis system

The test apparatus used for all the tests is presented in Fig. 2a. The system was operated in osmotic dilution mode with a recirculation loop on each side of the membrane. The FS tank, with a capacity of approximately 4 liters, was connected to a piston pump (Hydra-cell Pump). The DS tank, with a capacity of 4 or 20 liters depending on the test, was connected to a peristaltic pump (Peristaltic Metering Pump Flex-pro A3Vseries). The system was equipped with a custom-made FO test cell with symmetric flow channels on both sides of the membrane. The flow channels on each side of the membrane were filled with a mesh spacer to provide adequate support to the membrane. Water was circulated on each side of the membrane through ten round openings having a diameter of 4.7 mm. The effective surface area of the membrane is 149 cm² (10 cm wide and 15 cm long with rounded corners). A three dimension model of the cell is illustrated at Fig. 2b.

Fig.2a

Fig.2b

Fig. 2a) Bench-scale FO system and 2b) Custom made FO test cell

2.3. Production of saline wastewater

In order to have wastewater representative of the lixiviation produced in the field, five lixiviation columns were prepared with saline soil from two sites in Alberta, referred to as Site A (oil and gas-associated facility) and Site B (well site). Two columns were filled with soil from Site A and three columns were filled with soil from Site B. Each column is 1 m tall with an inner diameter of 10 cm and contains approximately 10 kg of soil. To facilitate the flow through the column, wood chips were added to the soil. Soil treatment was performed using a 0,4 M CaCl₂ solution pumped into the columns with a peristaltic pump at a flow rate of 0.25 mL/min during 40 days. Wastewater was collected for each of the five columns in individual 1 L glass bottles and then pooled in a 20 L plastic container for storage. The composition of the wastewater collected is listed in Table 2.

2.4. Experimental procedures

Prior to each test, membranes were placed in a container filled with distilled water for 30 minutes then rinsed with distilled water on both sides. To fit membranes in the cross-flow cell, membranes were cut while placed on an aluminum foil sheet in order to ensure that they were not damaged during the process.

All tests were performed with solutions at 24°C. Electrical conductivity was measured for the FS and the DS before and after each test using a Thermo Scientific Orion 013005MD conductimeter.

2.4.1. Membranes characterization

For membrane characterization, a volume of 1.5 L of distilled water was used as FS and a volume of 0.5 L of a 1 M NaCl solution was used as DS. The feed flow rates on each side were set at 1 L/min in cross-flow for 30 minutes. No hydraulic pressure was applied on the membrane. At the end of the test, the volumes of the DS and the FS were measured in order to calculate the water flux across the membrane. Water flux across the membrane was calculated using Eq. 1.

$$J_w = \frac{(V_{Draw,i} - V_{Draw,f})}{\Delta t * A} \quad (\text{Eq.1})$$

Where J_w is the water flux across the membrane, $V_{Draw,i}$ and $V_{Draw,f}$ are the initial and final volume of the draw solution, Δt is the duration of the test and A is the membrane coupon area.

FS conductivity was measured at the beginning and at the end of the test in order to measure the NaCl concentration and calculate the salt rejection, R_{NaCl} . Salt rejection for characterization tests was calculated using Eq. 2.

$$R_{NaCl} = \left(1 - \frac{\left(\frac{\text{mol NaCl transferred to feed}}{\text{L water removed}} \right)}{1 \text{ M NaCl}} \right) * 100 \quad (\text{Eq.2})$$

This procedure was done in duplicate with both membrane orientation (FO and PRO mode) for each of the selected membranes.

2.4.2. Flow rate effect on water flux

The evaluation of the FS and DS flow rate effect on the water flux across the membrane was done by using the same method used for membrane characterization, but with flow rates of

100, 200 and 400 mL/min. Water flux was calculated by measuring the difference in draw volume every 10 minutes. This test was only done with membranes showing the higher water flux at the membrane characterization test and was performed in PRO mode since higher water flux are obtain in this mode.

2.4.3. Effect of CP on long term water flux

Effect of combined ICP and ECP on long term water flux was evaluated on membranes showing the higher water flux at the membrane characterization test. This evaluation was performed using a volume of 2.5 L of distilled water as FS and a volume of 15 L of a 1 M NaCl solution as DS. This large volume of draw water was required in order to maintain the difference in osmotic pressure stable as water was attracted from the feed. The flow rates on each side were set at 100 mL/min in cross-flow for 180 minutes. Every 10 minutes, the change in DS volume was measured to estimate the flow and calculate the water flux across the membrane using Eq. 1.

2.4.4. Wastewater treatment test

The wastewater treatment test was done in duplicate. For each experiment, the initial DS and wastewater (FS) volumes were 0.5 L and 1.5 L, respectively. Flows of DS and FS were adjusted to 100 mL/min in cross-flow (equivalent to a water velocity of 0.17 m/s). The wastewater was obtained from lixiviation columns and the DS was a 1 M CaCl_2 solution (laboratory grade salt). For the treatment tests, CaCl_2 was chosen, instead of NaCl, to produce a liquid calcium amendment that could be reused for soil treatment. Every 10 minutes, the change in DS volume was measured to estimate the flow and calculate the water flux across the membrane using Eq. 1. 50 mL FS and DS samples were taken from their respective tanks at the beginning and the end of each experiment for ions analysis in order to evaluate the salt permeation. Calcium, chlorides, iron, magnesium, potassium, sodium and sulfates were selected for analysis as they represent more than 85% of the total dissolved solids based on the previous analyses made on lixiviation columns.

2.4.5. Analytical method

After dilution of the sample with of 4% V/V HNO_3 solution – in order to have a concentration in the range of the calibration curve for each element – concentrations of sodium, calcium, magnesium, iron and potassium were measured by inductively coupled plasma using a Perkin Elmer Emission Spectrometer Optima 4300 DV. Ion chromatography analyses were done using a Thermo Scientific Dionex ICS-1600 with a Dionex AS-DV sampler in order to determine the concentrations of chlorides and sulfates. The mobile phase was a 4.5 mM sodium carbonate/ 1.4 mM sodium bicarbonate solution at a flow of 1.2 mL/min. The chromatography column was a AS-22 4X250 mm Dionex IonPack.

3. Results and discussion

3.1. Membrane selection

3.1.1. Validation of spec sheet information

In a paper describing the potential of FO, Coday and *al.* [27] listed the current FO manufacturers and their commercial status; all the commercial and pre-commercial companies listed were contacted. Only HTI water and Porifera were able to provide membrane samples for lab scale

tests. Membranes could have been selected according to the specification sheet. However, the test conditions were different for each supplier. Therefore, in order to compare the performances of these membranes in identical experimental conditions, they were characterized following the same method described in section 2.4.1. Water flux and salt rejection obtained from this characterization in FO mode and PRO mode are presented in Table 1.

Table 1 Membrane characterisation results for different membranes operated in FO and PRO mode

Membrane	Company	Water Flux (LMH)		Salt Rejection (%)	
		Spec sheet	Lab test	Spec sheet	Lab test
Forward osmosis membrane orientation (FO)					
TFC-ES	HTI	18	17 ± 1	99,4	99,4
CTA-ES	HTI	9,0	9,3 ± 0,7	99,0	99,2
CTA-NW2	HTI	6,6	5,3 ± 0,7	98,0	99,0
FOMEM-0513	Porifera	33	29 ± 1	99,6	99,4
Pressure Retarded Osmosis membrane orientation (PRO)					
TFC-ES	HTI	36	39 ± 1	99,4	99,7
CTA-ES	HTI	12	14 ± 1	99,0	99,1
CTA-NW2	HTI	12	13 ± 1	98,0	99,2
FOMEM-0513	Porifera	58	49 ± 1	99,6	99,3

Since the test apparatus and test conditions were different than those of the companies, there was a slight difference between the laboratory test values and specification sheets values.

The ideal FO membrane for the application in this study would have a high water flux and salt rejection, as well as low CP and fouling. With both modes of operation, all membranes showed an adequate salt rejection. In terms of water flux, values were higher when operated in PRO mode due to less severe ICP, as expected[10, 11, 26]. From results in Table 1, CTA membranes had the lowest water flux. HTI describes these membranes as the most adapted for complex water highly charged with organic matter, since they are less likely to get fouled than TFC membranes. The contaminant in the wastewater targeted in this study was mostly ionic and mineral; therefore, CTA membranes were not ideal for the application. HTI's TFC membrane and Porifera's membrane were thus selected for the performance evaluation tests due to their high water flux.

3.1.2. Feed and draw flow effects on water flux across the membrane

Since energy consumption is proportional to flow rate as well as pressure, the effect of lower flow rates on water flux across the membrane was measured using HTI OsMem™ TFC-ES and FOMEM-0513 membranes at a specific DS solution concentration. For each membrane, one experiment was conducted for each of the following FS and DS flow rates: 100, 200, 400 and 1000 mL/min. For each of these flow rates, surface velocities were approximately 0.33 m/s, 0.133m/s, 0.067 m/s and 0.033 m/s respectively. The FO system was operated in PRO mode with deionised water as FS and 500 mL of 1M NaCl solution as DS in order to be consistent with the conditions encountered in practice. In this experiment, flux vary with time as the concentration of the DS solution changes due to dilution. In order to compare the water flux measured for each flow rates, water flux presented at Fig. 3 correspond to the average water flux calculated over the first 30 minutes of each experiment. Results in Fig. 3 illustrate a flux decline with lower flow rates due to ECP which is limited by the flow velocity. At a certain thickness, the layer of more concentrated solution accumulated at the surface will stop thickening because of the shear created by water flow. These observations are further supported by a recent study published by Hickenbottom and *al.* [21].

From 100 mL/min to 400 mL/min, there was no significant difference in flux between Porifera's and HTI's membrane. Because surface velocities are low at these flow rates, ECP is more significant. This could explain why the flux is the same between 100 mL/L and 400 mL/ for Porifera's and HTI's membrane while an important difference was observed in the flow rates.

However, in order to reduce energy consumption assuming similar head loss, a FS and DS flow rate of 100 mL/min was selected for subsequent testing of the performance of the FO system.

Fig. 3

Fig. 3 Average water flux calculated over a period of 30 minutes at different FS and DS flow rates

Fig. 4

Fig. 4 Specific flux across the membrane for the selected membrane operated in PRO mode evaluated over a period of 180 minutes

3.1.3. Long term influence of CP on membrane performances

In order to evaluate the combination of both internal and external CP effects on the membrane over the long term, tests were conducted with 2.5 L of deionised water as FS and 15L of 1 M NaCl solution as DS. Then, in order to evaluate the effect of draw solutes, a supplementary test was conducted with 2.5 L of deionised water as FS and 15L of 0.7 M CaCl_2 solution as DS with the membrane that showed higher water flux. Both NaCl and CaCl_2 solution had the same initial osmotic pressure of 48.3 bar.

Fig. 4 illustrates specific flux measured in time for both HTI OsMem™ TFC-ES and FOMEM-0513 membranes. Each series of data was obtained from a single experiment performed with a new membrane sample. The specific flux was obtained by dividing the measured water flux by the calculated osmotic pressure (π) of the draw solution (Eq. 3)[28].

$$\pi = 8,27 * 10^{-2} * (273 + T) * \sum M_j \quad (\text{Eq. 3})$$

Where π is the calculated osmotic pressure in bar, T is the temperature in Celsius and $\sum M_j$ is the sum of the molality concentration of all constituents in a solution.

In PRO mode, solutes from the DS get concentrated in the selective layer while water permeates through the membrane. As the FS is drawn, solute concentration inside the selective layer gets higher resulting in a decrease on water flux over time, thus explaining the decreased of water flux observed in Fig. 4.

Over a period of 180 minutes with NaCl DS, specific flux for Porifera's membrane fell from 0.71 LMH/bar to 0.54 LMH/bar, while for HTI's membrane, it fell from 0.65 LMH/bar to 0.46 LMH/bar. For the test with Porifera's membrane, initial and final FS TDS were 4 mg/L and 341 mg/L respectively and initial and final DS TDS were 58 500 mg/L and 53 800 mg/L respectively. For the test with HTI's membrane, initial and final FS TDS were 2 mg/L and 291 mg/L respectively and initial and final DS TDS were 58 500 mg/L and 54 600 mg/L respectively. The loss in specific flux observed with both membranes is due to increased ECP as feed TDS

increased over time. This loss was similar since TDS in the FS increased similarly in both experiments. Therefore, the membrane selected to perform wastewater treatment tests was made based on higher water flux. Porifera's membrane's average water flux was 22% higher than HTI's membrane, making it a better choice for the treatment of saline soil treatment wastewater.

Water flux across Porifera's membrane was higher when using CaCl_2 0,7M as DS even if the calculated osmotic pressure using Eq. 3 was equal for the NaCl 1M DS. Over a period of 180 minutes, the water flux fell from 0.88 LMH/bar to 0.64 LMH/bar with CaCl_2 . Since Ca^{2+} is a divalent ion, its tendency to penetrate the membrane is lower than that of Na^+ , therefore rejection is higher and external concentration polarization in the draw solution is reduced.

3.2. Wastewater treatment using selected membrane

3.2.1. Process performances

Two replicate wastewater treatment experiments were conducted with initial volumes of 1.5 L of wastewater obtained from lixiviation columns as FS and 0.5 L of 1 M CaCl_2 solution as DS. The FO system was operated in osmotic dilution mode in order to produce a diluted CaCl_2 solution that can be valorized as a liquid calcium amendment. The target performance was based on obtaining a calcium concentration acceptable for cationic exchange soil treatment, namely between 0.4 and 0.5 M, and recovering the highest percentage of FS.

The FS volume and the water flux across the FOMEM-0513 membrane as a function of time are illustrated at Fig. 5a and 5b respectively. The DS was able to draw approximately 50% of the initial volume of wastewater after 375 minutes of operation, resulting in a calcium concentration of 0.41 M and 0.44 M for Experiment 1 and Experiment 2, respectively. Average total dissolved solids in the FS between both experiments increased from 15.7 g/L to 21.5 g/L over the duration of the experiment.

Fig. 5a

Fig. 5b

Fig. 5c

Fig. 5 a) Water flux as a function of time and b) FS volume as a function of time and c) Specific flux as a function of time all obtained during osmotic dilution conducted with wastewater (two replicates: Experiment 1 and Experiment 2)

Water flux in Experiment 1 with saline soil treatment wastewater was initially 19.7 LMH and decreased to 2 LMH. This decrease was attributed to the dilution of the DS and the concentration of the FS, thus reducing the osmotic driving force. The initial osmotic pressure difference was 58.7 and 58.4 bar while the final osmotic pressure difference was 8 and 11.6 bar for Experiment 1 and 2 respectively. Water flux in Experiment 2 was initially 17.3 LMH and decreased to 3.5 LMH. The initial water flux in Experiment 1 was slightly higher than in Experiment 2, while the final water flux was slightly higher in Experiment 2 than Experiment 1. These results are explained by a smaller difference in osmotic pressure at the end of the treatment test in Experiment 1, thus resulting in a lower flux. However, in Fig. 5c we see that the specific flux remained almost constant for the duration of the experiment. This normal variation of membrane performance was also observed in a study conducted by Hickenbottom and al. on drilling mud and fracturing wastewater [21].

To evaluate the influence of both internal and external CP on system performances, specific flux as a function of time was evaluated and is illustrated at Fig. 5c. Specific flux was obtained using the method described in section 3.1.3. where π is the difference in osmotic pressure between

the FS and the DS. In order to calculate the osmotic pressure of the DS and the FS, it was assumed that salt passage was negligible and therefore that the total amount of salts on each side on the membrane remained the same.

Experiment 1 showed an average value of water flux of 0.39 LMH/bar, whereas Experiment 2 showed an average value of 0.30 LMH/bar. Unlike what is shown in Fig. 4, water flux showed no evidence of variation during the 375 minutes of operation. The difference in these results is caused by the presence of salt on both sides of the membrane, resulting in a greater polarization of the membrane, since salts in the FS accumulates in the support layer[10].

The average water fluxes obtained in Experiment 1 and Experiment 2 were lower than previously observed during the preliminary tests using CaCl_2 as draw solute over 180 minutes, 39% and 54% lower, respectively. The initial osmotic pressure difference between FS and DS was 48.4 bar for the preliminary tests and 59.4 bar for the wastewater treatment experiments (taking the osmotic pressure of the FS into account). Therefore, the initial osmotic pressure difference did not cause the lower water flux. The difference can however be explained by the ICP occurring on the FS side in wastewater treatment experiments, which is less important when testing the membrane with deionised water. Fouling of the membrane contribute to this flux decline. Indeed, TDS concentration on the FS side in wastewater treatment is initially of 16 300 mg/L while TDS concentration in the deionised water is approximately 4 mg/L

3.2.2. Solute transport during wastewater treatment:

Results of chemical analysis of the samples collected during the two experiments performed with wastewater are summarized in Table 2.

Table 2 Concentration of selected ions in the FS and the DS before and after treatment of the wastewater by FO

	Draw solution				Feed solution			
	Initial		Final		Initial		Final	
	mg/L	mg total (for 0,5 L)	mg/L	mg total (for 1,23L)	mg/L	mg total (for 1,5 L)	mg/L	mg total (for 0,77 L)
Sodium (Na)	-	-	157	194	3538	5307	5610	4314
Calcium (Ca)	39950	19975	16030	19733	1441	2162	2450	1884
Magnesium (Mg)	-	-	-	-	235	353	390,5	300
Iron (Fe)	-	-	-	-	-	-	-	-
Potassium (K)	-	-	5,2	6	64	95	95	73
Chlorides (Cl)	65078	32539	25959,5	31956	8128	12192	13107	10079
Sulphates (SO4)	-	-	-	-	445	668	669,5	515
Osmotic pressure (bar)	69,9		27,7		10,5		16,9	

- : below the detection limit (BDL)

During treatment, approximately 50% of the FS volume was transferred to the DS which should have caused a two-fold increase in ion concentration. However, precipitation on the surface of the membrane led to slightly lower concentration with a final average concentration of 1.7x the initial concentration.

The final results in the analysis of the FS and the DS after treatment indicate that sulphates and magnesium did not contaminate the DS. However, salt permeation occurred for sodium and potassium, causing a concentration of 157 mg/L of Na and 5.2 mg/L of K. When a mass balance was conducted, it was determined that 3.6% of the sodium was transferred to the DS. Since the DS could be reused as a calcium amendment, a concentration of 157 mg/L would be negligible compared to the concentration of calcium. When a mass balance is conducted for the potassium however, it was determined that an important percentage was transferred to the DS (6.3%). For a single reuse of the DS, this would not be a concern; however in a cascade application with multiple uses, potassium may accumulate and become a problem. Further studies should be conducted in order to evaluate the potential accumulation of potassium in the DS in a series of cascade applications as a calcium amendment.

4. Conclusion

The main objective of the study was to evaluate the potential application of the FO process in the treatment of wastewater generated during the treatment of salt-contaminated soils. Results from the tests performed under the usual conditions of the application indicated that Porifera's membrane was the most suitable membrane. However, even if the membrane design allows initial water flux as high as 0.88 LMH/bar when using a 0.7 M CaCl₂ DS, it rapidly decreases due to concentration polarization. An important challenge for large-scale application would be to elaborate a design to obtain ideal specific flux. To obtain such performances, high cross flow velocities are preferred. Current spiral wound element have high headloss, limiting cross-flow

402 velocities and therefore membrane performances. In terms of treatment efficiency for this
403 particular application related to the treatment of saline soil treatment wastewater, the results
404 are promising. With a recovery of almost 50 % of the initial volume with very little energy
405 consumption compared to RO and low salt permeability, the results obtained in this study
406 demonstrated the potential of the FO technology in the production of valuable process water
407 and reuse of water in industrial processes. The concentrate, which represents only 50% of the
408 initial volume, can be disposed of by deep-well injection without regulation violation, which
409 reduces the cost of disposal of the waste stream. For large-scale application, the low energy
410 consumption would result in an important reduction of operation costs. However, important
411 work is still required in order to develop a compact unit with high surface area and high water
412 flux for field scale application and to optimize the technology.

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References:

1. Rengasamy, P., *World salinization with emphasis on Australia*. Journal of Experimental Botany, 2006. **57**(5): p. 1017-1023.
2. Reading, L.P., et al., *Hydraulic Conductivity Increases in a Sodic Clay Soil in Response to Gypsum Applications: Impacts of Bulk Density and Cation Exchange*. Soil Science, 2012. **177**(3): p. 165-171 10.1097/SS.0b013e3182408f4f.
3. Massoud, M.S., M. Al-Sarawi, and S.A. Wahba, *Variations in the Chemical Properties of Soils Contaminated with Oil Lakes in the Greater Burgan Oil Fields, Kuwait*. Water, Air, and Soil Pollution, 2000. **118**(3-4): p. 281-297.
4. Rooney, D.J., K.W. Brown, and J.C. Thomas, *The Effectiveness of Capillary Barriers to Hydraulically Isolate Salt Contaminated Soils*. Water, Air, and Soil Pollution, 1998. **104**(3-4): p. 403-411.
5. Abdou, M., et al., *Finding value in formation water*. Oilfield Review, 2011. **23**(1): p. 24-35.
6. Adams, D.J., T. Pickett, and J. Murchinson, *Variation in selenium reduction by naturally-occurring microorganism*, in *Annual Meeting Abstracts*, A.A.o.P. Geologists, Editor. 1998.
7. Weathers, M.L., et al., *Reclamation of saltwater-contaminated soil in Big Lake field*. 1994. Medium: X; Size: pp. 737-743.
8. Alberta, G.o., *Salt Contamination Assessment & Remediation Guidelines*, E.S. Division, Editor. 2001.
9. Regulator, A.E., *Directive 051: Injection and Disposal Wells - Wells Classifications, Completions, Logging, and Testing Requirements*. 1994.
10. Lutchmiah, K., et al., *Forward osmosis for application in wastewater treatment: A review*. Water Research, 2014. **58**(0): p. 179-197.
11. Phuntsho, S., et al., *Assessing the major factors affecting the performances of forward osmosis and its implications on the desalination process*. Chemical Engineering Journal, 2013. **231**: p. 484-496.
12. Ge, Q., M. Ling, and T.-S. Chung, *Draw solutions for forward osmosis processes: Developments, challenges, and prospects for the future*. Journal of Membrane Science, 2013. **442**(0): p. 225-237.
13. Lay, W.C.L., et al., *Factors affecting flux performance of forward osmosis systems*. Journal of Membrane Science, 2012. **394-395**(0): p. 151-168.
14. Cath, T.Y., A.E. Childress, and M. Elimelech, *Forward osmosis: Principles, applications, and recent developments*. Journal of Membrane Science, 2006. **281**(1-2): p. 70-87.
15. Holloway, R.W., et al., *Forward osmosis for concentration of anaerobic digester centrate*. Water Research, 2007. **41**(17): p. 4005-4014.
16. Cornelissen, E.R., et al., *Membrane fouling and process performance of forward osmosis membranes on activated sludge*. Journal of Membrane Science, 2008. **319**(1-2): p. 158-168.
17. Wang, R., et al., *Characterization of novel forward osmosis hollow fiber membranes*. Journal of Membrane Science, 2010. **355**(1-2): p. 158-167.
18. Xu, Y., et al., *Effect of draw solution concentration and operating conditions on forward osmosis and pressure retarded osmosis performance in a spiral wound module*. Journal of Membrane Science, 2010. **348**(1-2): p. 298-309.
19. McCutcheon, J.R., R.L. McGinnis, and M. Elimelech, *Desalination by ammonia-carbon dioxide forward osmosis: Influence of draw and feed solution concentrations on process performance*. Journal of Membrane Science, 2006. **278**(1-2): p. 114-123.

- 468 20. Buckwalter, P., et al., *Dewatering microalgae by forward osmosis*. Desalination, 2013.
469 **312**(0): p. 19-22.
- 470 21. Hickenbottom, K.L., et al., *Forward osmosis treatment of drilling mud and fracturing*
471 *wastewater from oil and gas operations*. Desalination, 2013. **312**(0): p. 60-66.
- 472 22. Yang, Q., K.Y. Wang, and T.-S. Chung, *A novel dual-layer forward osmosis membrane for*
473 *protein enrichment and concentration*. Separation and Purification Technology, 2009.
474 **69**(3): p. 269-274.
- 475 23. Cath, T.Y., et al., *A multi-barrier osmotic dilution process for simultaneous desalination*
476 *and purification of impaired water*. Journal of Membrane Science, 2010. **362**(1–2): p.
477 417-426.
- 478 24. Hancock, N.T., N.D. Black, and T.Y. Cath, *A comparative life cycle assessment of hybrid*
479 *osmotic dilution desalination and established seawater desalination and wastewater*
480 *reclamation processes*. Water Research, 2012. **46**(4): p. 1145-1154.
- 481 25. Lee, K.P., T.C. Arnot, and D. Mattia, *A review of reverse osmosis membrane materials for*
482 *desalination—Development to date and future potential*. Journal of Membrane Science,
483 2011. **370**(1–2): p. 1-22.
- 484 26. Gray, G.T., J.R. McCutcheon, and M. Elimelech, *Internal concentration polarization in*
485 *forward osmosis: role of membrane orientation*. Desalination, 2006. **197**(1–3): p. 1-8.
- 486 27. Coday, B.D., et al., *The sweet spot of forward osmosis: Treatment of produced water,*
487 *drilling wastewater, and other complex and difficult liquid streams*. Desalination, 2014.
488 **333**(1): p. 23-35.
- 489 28. B.V., L. *Osmotic pressure calculator*. 2015 [cited 2015 2015-01-21]; Available from:
490 <http://www.lenntech.com/calculators/osmotic/osmotic-pressure.htm>.

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