## Forward osmosis for the concentration and reuse 1

#### of process saline wastewater 2

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#### 13 Abstract

14 Salt-contaminated soil generated by industrial activities has become a problem of concern over 15 the last decades. While existing treatment technologies for these soils are efficient, the 16 wastewater produced remains the major drawback. In this study, a novel application of forward 17 osmosis (FO) was tested. The FO process operated in osmotic dilution mode was investigated in 18 order to concentrate saline soil treatment wastewater and produce valuable process water. FO 19 was chosen for this application since it is a low-energy technology and process adapted for 20 complex liquid streams. Results from this study indicate that FO operated in osmotic dilution 21 mode with a 1 M CaCl<sub>2</sub> draw solution is able to recover 50% of the initial wastewater volume 22 with an initial total dissolved solids concentration of 16.3 g/L. Following treatment, the draw 23 solution can be reused as a cationic exchange solution with CaCl<sub>2</sub> concentrations between 0.41 24 M and 0.44 M. No important solute transport from the feed to the draw solution was observed, 25 except for some potassium drawn by the draw solution. This study demonstrated the potential 26 of integrating the FO technology in the soil treatment industry in order to enhance water reuse 27 in water-intensive processes.

#### 28 **Keywords**

29 Forward osmosis, wastewater, reuse, concentration, case study

#### 30 **Graphical Abstract**



## 31 32

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#### 33 1. Introduction

34 Salt-contaminated soil has become a worldwide problem of concern over the last decades and is 35 present in more than 100 countries. In 1989, according to the available data at the time, 36 approximately 932.2 million hectares of land were salt impacted[1]. Unlike hydrocarbon or 37 heavy metal contamination, salt does not have a pathological connotation. However, salt can be 38 harmful to the environment by changing the physical properties of the soil, resulting in a 39 reduction of aquifer recharge or a lixiviation of salts in groundwater, thereby reducing the 40 reserves of fresh water[2]. The origin of salt-affected soils is different depending on location. 41 This paper emphasizes punctual salinization, which is the result of accidental spills[3] or poor 42 industrial practices[4]. One of the industries responsible for a large part of punctual salinization 43 around the world is that of oil and gas. When exploiting an oil reserve, hydrocarbons are not the 44 only product extracted. Brine water, which can have a salt concentration higher than sea water, 45 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 20<sup>th</sup> century, this water was 46 discharged straight to the environment or, at best, contained in basins without retention 47 48 devices[7].

49 In 2001, the Environmental Sciences Division of the Alberta Government published the Salt 50 Contamination Assessment & Remediation Guidelines in which different remediation 51 technologies are proposed. According to this document, the most suitable treatment process for 52 site remediation is the replacement of sodium with calcium. This exchange is done by increasing 53 the calcium concentration in the soil with the addition of a calcium additive[8]. The major 54 drawbacks of this technique are the consumption of a large volume of fresh water and the 55 management of process water. The wastewater produced by the treatment process is 56 transported to a deep well according to the regulations[9]. Since many of the contaminated sites 57 are located in remote areas with scarce fresh water supplies, these local water supplies are 58 greatly depleted through the process of soil treatment and soil washing. In addition, the 59 distance from the water source and the disposal well for process water can be prohibitive to the 60 technology due to transportation costs. In the ongoing effort to reduce water consumption and 61 management costs, it is necessary to develop a dewatering system that fulfills the following 62 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].

76 When used for water treatment or for wastewater volume reduction, FO can be operated in 77 different modes. In applications in which high quality product water is not required, FO can be 78 operated in an osmotic dilution mode, where water drawn from wastewater through the 79 membrane dilutes the DS. The two end products are the diluted DS, which can be valorised, and 80 the concentrated wastewater. The osmotic dilution mode is illustrated in Fig. 1a. In applications 81 where high quality product water is required, FO can be coupled with a process such as RO to 82 systematically re-concentrate the DS and produce a high quality permeate. In the latter case, FO 83 acts as a high-level pre-treatment step before the desalination process; this pre-treatment mode 84 is illustrated in Fig. 1b.[23, 24]





86 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<sub>2</sub> solution as DS produced from the CaCl<sub>2</sub> 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.

93 Similarly to RO membranes, FO membranes act as a barrier to most salts, organic contaminants 94 and suspended particles [21]. However, because a FO system operates with low or no hydraulic 95 pressure, their configurations are different. The main distinctions between FO and RO 96 membranes is the FO membrane's lack of the thick support layer that gives physical resistance 97 to hydraulic pressure and the hydrophobicity of the membranes. Most RO membrane are made 98 of polysulphone polymers[25], a hydrophobic material, limiting the wetting of the support. 99 Specially designed FO membranes increase water flux in osmotic driven systems but are still 100 affected by CP, which is the accumulation of solute at the surface of a membrane.

101 Because of CP, the osmotic pressure difference across the active layer is much lower than the 102 bulk osmotic pressure difference resulting in much lower flux than expected. In systems where

103 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.

108 When operating in FO mode (selective layer facing the FS), solutes accumulate on the surface of 109 the selective layer; this may be called concentrative ECP and is similar to CP observed in RO. 110 When operating in PRO (pressure retarded osmosis) mode (selective layer facing the DS), the 111 draw solution in contact with the membrane is diluted by the permeating water; this is called 112 dilutive ECP. In both cases, the water flux across the membrane is reduced due to the reduction 113 of the effective osmotic driving force. ECP can be controlled hydrodynamically by increasing 114 water velocity and turbulence on the surface of the membrane [14, 17].

115 ICP is associated to osmotic-driven membrane process with asymmetric membranes consisting 116 in a dense selective layer and a porous support layer. Diffusion attracts solutes from the draw 117 solution toward the feed solution, creating an accumulation inside the membrane layers. This 118 phenomenon is particularly important in FO when operating in FO mode: the porous layer 119 becomes a thick polarized layer, substantially reducing the osmotic driving force between the 120 feed and the draw solution. Extensive research has reported higher water fluxes in PRO-mode, 121 attributed to less severe ICP. However, this orientation is more prone to membrane fouling [10, 122 26]. Unlike ECP, ICP cannot be controlled by operation factors [17].

123 In this study, we perform the first evaluation of the potential of the FO process to treat 124 wastewater generated during the treatment of salt-contaminated soils. The use of this 125 technology in the soil treatment process has the potential of providing a more energy-efficient 126 solution to conventional desalination technologies such as RO by using the difference in osmotic 127 pressure of two solutions as the primary energy source. The main objective was to evaluate the 128 performance of the FO process operated in osmotic dilution mode to concentrate saline soil 129 treatment wastewater and produce valuable process water. Results also allowed identifying the 130 most suitable commercially available membrane for this application as well as key design 131 parameters, such as water flux and salt permeation. Opportunities and challenges for large-scale 132 application of this technology are also discussed.

## 133 2. <u>Material and methods</u>

## 134 2.1. <u>Commercial FO membranes</u>

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 cm<sup>2</sup> membranes identified as HTI OsMem<sup>™</sup> TFC-ES and the cellulose triacetate (CTA) membrane kit composed of 929 cm<sup>2</sup> membranes identified as HTI OsMem<sup>™</sup> CTA-ES and 929 cm<sup>2</sup> membranes identified as HTI OsMem<sup>™</sup> CTA-NW2. Porifera kindly provided 929 cm<sup>2</sup> high flux forward osmosis membranes identified as FOMEM-0513.

#### 142 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 143 144 osmotic dilution mode with a recirculation loop on each side of the membrane. The FS tank, 145 with a capacity of approximately 4 liters, was connected to a piston pump (Hydra-cell Pump). 146 The DS tank, with a capacity of 4 or 20 liters depending on the test, was connected to a 147 peristaltic pump (Peristaltic Metering Pump Flex-pro A3Vseries). The system was equipped with 148 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 149 adequate support to the membrane. Water was circulated on each side of the membrane 150 through ten round openings having a diameter of 4.7 mm. The effective surface area of the 151 membrane is 149  $\text{cm}^2$  (10 cm wide and 15 cm long with rounded corners). A three dimension 152 153 model of the cell is illustrated at Fig. 2b.





154

Fig.2b

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

#### 156 2.3. <u>Production of saline waste</u>water

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157 In order to have wastewater representative of the lixiviation produced in the field, five lixiviation 158 columns were prepared with saline soil from two sites in Alberta, referred to as Site A (oil and 159 gas-associated facility) and Site B (well site). Two columns were filled with soil from Site A and 160 three columns were filled with soil from Site B. Each column is 1 m tall with an inner diameter of 161 10 cm and contains approximately 10 kg of soil. To facilitate the flow through the column, wood 162 chips were added to the soil. Soil treatment was performed using a 0,4 M CaCl<sub>2</sub> solution pumped 163 into the columns with a peristaltic pump at a flow rate of 0.25 mL/min during 40 days. 164 Wastewater was collected for each of the five columns in individual 1 L glass bottles and then 165 pooled in a 20 L plastic container for storage. The composition of the wastewater collected is 166 listed in Table 2.

#### 167 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.

#### 175 2.4.1. <u>Membranes characterization</u>

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.

181 
$$J_w = \frac{(V_{Draw,i} - V_{Draw,f})}{\Delta t * A}$$
(Eq.1)

182 Where  $J_w$  is the water flux across the membrane,  $V_{Draw,i}$  and  $V_{Draw,f}$  are the initial and final volume 183 of the draw solution,  $\Delta t$  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<sub>NaCl</sub>. Salt rejection for characterization
 tests was calculated using Eq. 2.

187 
$$R_{NaCl} = \left(1 - \frac{\left(\frac{mol \, NaCl \, transferred \, to \, feed}{L \, water \, removed}\right)}{1 \, M \, NaCl}\right) * 100 \tag{Eq.2}$$

188 This procedure was done in duplicate with both membrane orientation (FO and PRO mode) for 189 each of the selected membranes.

#### 190 2.4.2. Flow rate effect on water flux

191 The evaluation of the FS and DS flow rate effect on the water flux across the membrane was 192 done by using the same method used for membrane characterization, but with flow rates of

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193 100, 200 and 400 mL/min. Water flux was calculated by measuring the difference in draw 194 volume every 10 minutes. This test was only done with membranes showing the higher water 195 flux at the membrane characterization test and was performed in PRO mode since higher water 196 flux are obtain in this mode.

#### 197 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.

#### 205 2.4.4. <u>Wastewater treatment test</u>

206 The wastewater treatment test was done in duplicate. For each experiment, the initial DS and 207 wastewater (FS) volumes were 0.5 L and 1.5 L, respectively. Flows of DS and FS were adjusted to 208 100 mL/min in cross-flow (equivalent to a water velocity of 0.17 m/s). The wastewater was 209 obtained from lixiviation columns and the DS was a 1 M  $CaCl_2$  solution (laboratory grade salt). 210 For the treatment tests, CaCl<sub>2</sub> was chosen, instead of NaCl, to produce a liquid calcium 211 amendment that could be reused for soil treatment. Every 10 minutes, the change in DS volume 212 was measured to estimate the flow and calculate the water flux across the membrane using Eq. 213 1. 50 mL FS and DS samples were taken from their respective tanks at the beginning and the end 214 of each experiment for ions analysis in order to evaluate the salt permeation. Calcium, chlorides, 215 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 216 217 columns.

218 2.4.5. <u>Analytical method</u>

219 After dilution of the sample with of 4% V/V HNO<sub>3</sub> solution – in order to have a concentration in 220 the range of the calibration curve for each element – concentrations of sodium, calcium, 221 magnesium, iron and potassium were measured by inductively coupled plasma using a Perkin 222 Elmer Emission Spectrometer Optima 4300 DV. Ion chromatography analyses were done using a 223 Thermo Scientific Dionex ICS-1600 with a Dionex AS-DV sampler in order to determine the 224 concentrations of chlorides and sulfates. The mobile phase was a 4.5 mM sodium carbonate/ 1.4 225 mM sodium bicarbonate solution at a flow of 1.2 mL/min. The chromatography column was a 226 AS-22 4X250 mm Dionex IonPack.

- 227 3. <u>Results and discussion</u>
- 228 3.1. <u>Membrane selection</u>
- 229 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.

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- 240
- 241
- 242
- 243

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

Membrane	ne Company Water Flux (LMH		x (LMH)	Salt Rejection (%)		
		Spec sheet	Lab test	Spec sheet	Lab test	
	Forwa	rd osmosis membr	ane 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 Re	tarded Osmosis me	embrane orientati	ion (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	

245

246 Since the test apparatus and test conditions were different than those of the companies, there 247 was a slight difference between the laboratory test values and specification sheets values.

248 The ideal FO membrane for the application in this study would have a high water flux and salt 249 rejection, as well as low CP and fouling. With both modes of operation, all membranes showed 250 an adequate salt rejection. In terms of water flux, values were higher when operated in PRO 251 mode due to less severe ICP, as expected[10, 11, 26]. From results in Table 1, CTA membranes 252 had the lowest water flux. HTI describes these membranes as the most adapted for complex 253 water highly charged with organic matter, since they are less likely to get fouled than TFC 254 membranes. The contaminant in the wastewater targeted in this study was mostly ionic and 255 mineral; therefore, CTA membranes were not ideal for the application. HTI's TFC membrane and 256 Porifera's membrane were thus selected for the performance evaluation tests due to their high 257 water flux.

#### 258 3.1.2. Feed and draw flow effects on water flux across the membrane

259 Since energy consumption is proportional to flow rate as well as pressure, the effect of lower 260 flow rates on water flux across the membrane was measured using HTI OsMem™ TFC-ES and 261 FOMEM-0513 membranes at a specific DS solution concentration. For each membrane, one 262 experiment was conducted for each of the following FS and DS flow rates: 100, 200, 400 and 263 1000 mL/min. For each of these flow rates, surface velocities were approximately 0.33 m/s, 264 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 265 266 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 267 268 measured for each flow rates, water flux presented at Fig. 3 correspond to the average water 269 flux calculated over the first 30 minutes of each experiment. Results in Fig. 3 illustrate a flux 270 decline with lower flow rates due to ECP which is limited by the flow velocity. At a certain 271 thickness, the layer of more concentrated solution accumulated at the surface will stop 272 thickening because of the shear created by water flow. These observations are further supported by a recent study published by Hickenbottom and al. [21]. 273

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.



280 Fig. 3

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



282 Fig. 4

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

#### 285 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<sub>2</sub> solution as DS with the membrane that showed higher water flux. Both NaCl and CaCl<sub>2</sub> solution had the same initial osmotic pressure of 48.3 bar.

Fig. 4 illustrates specific flux measured in time for both HTI OsMem<sup>TM</sup> 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 ( $\pi$ ) of the draw solution (Eq. 3)[28].

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

297 Where  $\pi$  is the calculated osmotic pressure in bar, T is the temperature in Celsius and  $\sum M_j$  is 298 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<sub>2</sub>. Since Ca<sup>2+</sup> is a divalent ion, its tendency to penetrate the membrane is lower than that of Na<sup>+</sup>, therefore rejection is higher and external concentration polarization in the draw solution is reduced.

- 320 3.2. <u>Wastewater treatment using selected membrane</u>
- 321 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<sub>2</sub> solution as DS. The FO system was operated in osmotic dilution mode in order to produce a diluted CaCl<sub>2</sub> 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.



#### 334 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)

338 Water flux in Experiment 1 with saline soil treatment wastewater was initially 19.7 LMH and 339 decreased to 2 LMH. This decrease was attributed to the dilution of the DS and the 340 concentration of the FS, thus reducing the osmotic driving force. The initial osmotic pressure 341 difference was 58.7 and 58.4 bar while the final osmotic pressure difference was 8 and 11.6 bar 342 for Experiment 1 and 2 respectively. Water flux in Experiment 2 was initially 17.3 LMH and 343 decreased to 3.5 LMH. The initial water flux in Experiment 1 was slightly higher than in 344 Experiment 2, while the final water flux was slightly higher in Experiment 2 than Experiment 1. 345 These results are explained by a smaller difference in osmotic pressure at the end of the 346 treatment test in Experiment 1, thus resulting in a lower flux. However, in Fig. 5c we see that the 347 specific flux remained almost constant for the duration of the experiment. This normal variation 348 of membrane performance was also observed in a study conducted by Hickenbottom and al. on 349 drilling mud and fracturing wastewater [21].

350 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

352 the method described in section 3.1.3. where  $\pi$  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].

361 The average water fluxes obtained in Experiment 1 and Experiment 2 were lower than 362 previously observed during the preliminary tests using CaCl<sub>2</sub> as draw solute over 180 minutes, 363 39% and 54% lower, respectively. The initial osmotic pressure difference between FS and DS was 364 48.4 bar for the preliminary tests and 59.4 bar for the wastewater treatment experiments 365 (taking the osmotic pressure of the FS into account). Therefore, the initial osmotic pressure 366 difference did not cause the lower water flux. The difference can however be explained by the 367 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 368 369 decline. Indeed, TDS concentration on the FS side in wastewater treatment is initially of 16 300 370 mg/L while TDS concentration in the deionised water is approximately 4 mg/L

371 3.2.2. Solute transport during wastewater treatment:

372 Results of chemical analysis of the samples collected during the two experiments performed373 with wastewater are summarized in Table 2.

	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
lron (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

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

- : below the detection limit (BDL)

377

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.

382 The final results in the analysis of the FS and the DS after treatment indicate that sulphates and 383 magnesium did not contaminate the DS. However, salt permeation occurred for sodium and 384 potassium, causing a concentration of 157 mg/L of Na and 5.2 mg/L of K. When a mass balance 385 was conducted, it was determined that 3.6% of the sodium was transferred to the DS. Since the 386 DS could be reused as a calcium amendment, a concentration of 157 mg/L would be negligible 387 compared to the concentration of calcium. When a mass balance is conducted for the potassium 388 however, it was determined that an important percentage was transferred to the DS (6.3%). For 389 a single reuse of the DS, this would not be a concern; however in a cascade application with 390 multiple uses, potassium may accumulate and become a problem. Further studies should be 391 conducted in order to evaluate the potential accumulation of potassium in the DS in a series of 392 cascade applications as a calcium amendment.

#### 393 **4.** <u>Conclusion</u>

394 The main objective of the study was to evaluate the potential application of the FO process in 395 the treatment of wastewater generated during the treatment of salt-contaminated soils. Results 396 from the tests performed under the usual conditions of the application indicated that Porifera's 397 membrane was the most suitable membrane. However, even if the membrane design allows 398 initial water flux as high as 0.88 LMH/bar when using a 0.7 M CaCl<sub>2</sub> DS, it rapidly decreases due 399 to concentration polarization. An important challenge for large-scale application would be to 400 elaborate a design to obtain ideal specific flux. To obtain such performances, high cross flow 401 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 disposable 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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