

1 **Exploring the Differences between Forward Osmosis and Reverse Osmosis Fouling**

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38 **Abstract**

39 A comparison of alginate fouling in forward osmosis (FO) with that in reverse osmosis (RO)  
40 was made. A key experimental finding, corroborated by membrane autopsies, was that FO is  
41 essentially more prone to fouling than RO, which is opposite to a common claim in the  
42 literature where deductions on fouling are often based solely on the water flux profiles. Our  
43 theoretical analysis shows that, due to a decrease in the intensity of internal concentration  
44 polarization (ICP), and thus an increase in the effective osmotic driving force during FO fouling  
45 tests, the similarity of experimental water flux profiles for FO and RO is in accordance with  
46 there being greater fouling in FO than RO. The specific foulant resistance for FO was also  
47 found to be greater than that for RO. Possible explanations are discussed and these include the  
48 influence of reverse solute diffusion from draw solution. Whilst this explanation regarding  
49 specific foulant resistance is dependent on the draw solution properties, the finding of greater  
50 overall foulant accumulation in FO is considered to be a general finding. Additionally, the  
51 present study did not find evidence that hydraulic pressure in RO plays a critical role in foulant  
52 layer compaction. Overall this study demonstrated that although FO has higher fouling  
53 propensity, it offers superior water flux stability against fouling. For certain practical  
54 applications this resilience may be important.

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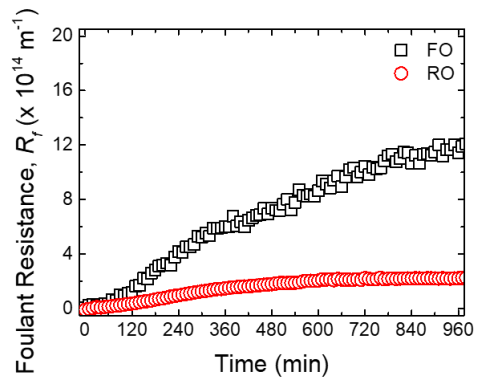
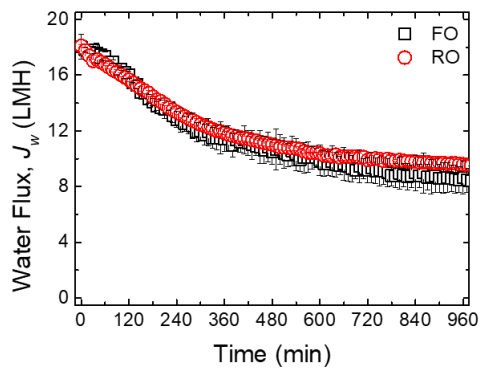
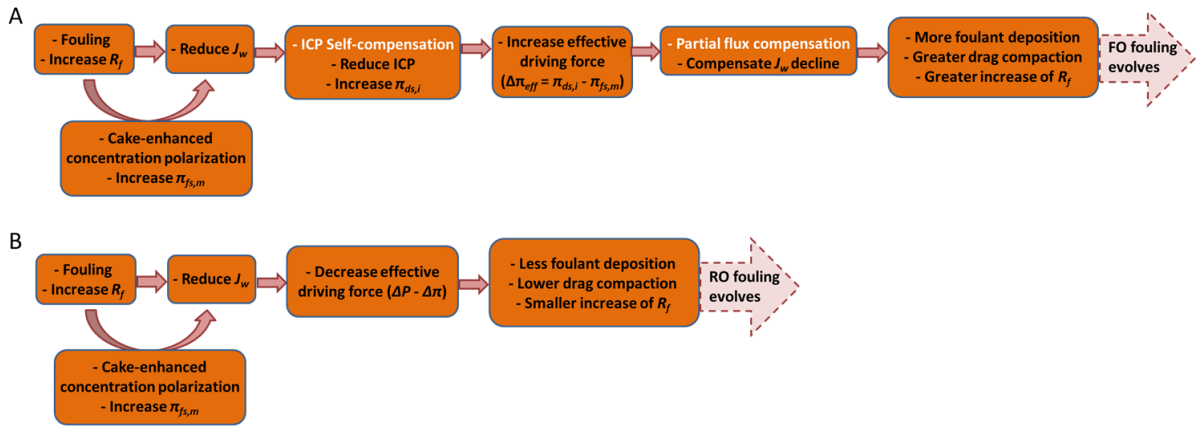
57 **Keywords:** forward osmosis; reverse osmosis; fouling; internal concentration polarization  
58 (ICP); cake-enhanced concentration polarization (CECP)

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61 **Graphic Abstract**

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## 67 **1. Introduction**

68 Forward osmosis (FO) has received considerable interest in the recent decade for various  
69 applications such as desalination [1-4], wastewater treatment [3-6], trace contaminant removal  
70 [7-9], and resource recovery [10, 11]. In an FO process a draw solution (DS) with a higher  
71 osmotic pressure on one side of a selective membrane draws the water from a feed solution  
72 (FS) with a lower osmotic pressure on the other side of the membrane [1]. Unlike pressure-  
73 driven reverse osmosis (RO) that is a relatively energy-intensive process, osmotically driven  
74 FO process only requires minimum electrical energy for pumping the DS and FS solutions. In  
75 those special cases where the application does not require the regeneration of the DS (e.g., the  
76 osmotic dilution of the fertilizer-based DS [12] and/or the concentration of the FS for nutrient  
77 recovery [10]), FO has an outstanding advantage in terms of lower energy consumption. Also  
78 it has been suggested that a hybrid FO system that incorporates a DS regeneration process may  
79 also outperform conventional RO when treating challenging feedwaters (e.g., the feedwater  
80 with high salinity or specific challenging contaminants) [4, 13, 14]. Whilst energy consumption  
81 is a major factor in the evaluation between FO and RO [13, 15, 16], membrane fouling is  
82 another important consideration when comparing the performance of FO and RO in practical  
83 applications [3-5, 13, 14, 17].

84

### 85 **1.1. Critical review of prior studies on the comparison of fouling in FO and RO**

86 Owing to the different driving forces for FO and RO (i.e., osmotic pressure vs. hydraulic  
87 pressure), fouling behaviour between FO and RO has been presumed to be different. The  
88 comparison of membrane fouling between FO and RO processes has been studied extensively  
89 and it has been broadly claimed that osmotically driven FO has lower fouling tendency and  
90 greater fouling reversibility than pressure-driven RO [13, 18-28]. These studies attributed their  
91 claim to the lack of hydraulic compaction of the foulant layer in the FO process, which resulted

92 in the formation of different foulant layer structure in FO compared to that in RO. They stated  
93 that in FO the foulant layer is looser and less compacted and thereby the fouled membrane can  
94 be easily cleaned by a brief water rinsing, whereas in RO the foulant layer is densely compacted  
95 and tightly held on the membrane under the action of hydraulic pressure, thereby resulting in a  
96 reduced cleaning efficiency [19, 24].

97

98 On the other hand, a number of other studies have reported opposing observations [29-32]. Lay  
99 et al. did not observe differences in the flux decline between FO and RO fouling and they  
100 attributed this to the low initial water flux that was said to be below the critical flux [29], which  
101 today might be better termed threshold flux [33]. For alginate and silica fouling, Jang et al.  
102 observed in a laboratory study that fouling propensity was the highest for FO compared with  
103 RO and membrane distillation [30]. Tow et al. developed a method of *in situ* membrane fouling  
104 quantification and found greater foulant accumulation with FO than with RO, which suggests  
105 that fouling in FO might be more severe than RO despite the observed lower flux decline in  
106 FO [31]. In addition, their study did not find any evidence that the thinner cake layer (less  
107 foulant accumulation) in RO could be attributed to the hydraulic pressure compaction [31]. In  
108 an earlier study on alginate and silica fouling in RO under constant flux operation, Fane and  
109 Chong observed no clear difference in the trans-membrane pressure (TMP) profiles for a flux  
110 of 40 l/m<sup>2</sup>hr (well above the critical flux for both foulants) with varying feed pressures from  
111 22 to 30 bar, suggesting that foulant layer compaction is physically related to water flux not  
112 hydraulic pressure *per se* [32].

113

## 114 **1.2. Analysis of possible reasons for the different findings on FO and RO fouling**

115 The inconsistent findings on FO and RO fouling between different groups of researchers  
116 probably lie in the discrepancies with respect to experimental methods used, as well as in the

117 different analytical approaches. Firstly, in most of the prior studies experimental conditions for  
118 FO and RO were not comparably controlled. For example, (i) in many cases the apparent  
119 driving force for RO (i.e., hydraulic pressure,  $\Delta P$ ) was maintained constant during the entire  
120 RO experimental test, whereas that for FO (i.e., osmotic pressure difference between the bulk  
121 DS and the bulk FS,  $\Delta\pi$ ) was gradually decreasing during the FO experimental test as the DS  
122 was gradually diluted and the FS was gradually concentrated [19-21, 23, 24, 30, 31, 34]; (ii)  
123 different types of membranes were used for FO and RO tests, for which different membrane  
124 properties may influence the fouling behaviour [26, 31]. Secondly, in many prior studies the  
125 reported water flux for RO was directly observed from experiments, whereas for FO it was not  
126 the experimentally observed flux but a corrected one by using experimental fouling flux and  
127 baseline flux under non-fouling conditions [19-24]. Typically, the approach to correct the  
128 observed FO flux was to eliminate the effects of DS dilution and the FS concentration during  
129 the test. However, the approach of flux correction did not take into account the effects of  
130 concentration polarization (CP) especially internal concentration polarization (ICP) that is  
131 strongly dependent on the solution concentration and will significantly influence the observed  
132 flux via the change in effective driving force [35-37]. Thirdly, the majority of prior studies  
133 compared the fouling propensity between FO and RO based on the extent of flux decline [19-  
134 24, 26, 30, 34]. However, in both FO and RO, especially FO, temporal changes in flux do not  
135 properly reflect the evolution of foulant accumulation on the membrane, because the flux  
136 decline is related not only to the hydraulic resistance of the foulant layer accumulated on the  
137 membrane but also to the CP that will result in the decrease of effective driving force [20, 31,  
138 36-39]. It is also noted that the foulant layer formed on the membrane might influence the  
139 degree of external CP through the process of “cake-enhanced concentration polarization” [38].  
140 Although Tow et al. developed a method to quantify membrane fouling by employing two  
141 parameters – cake structural parameter (that is related to cake-enhanced concentration

142 polarization) and pore hydraulic diameter (that is related to hydraulic resistance of foulant  
143 layer), it appears that their study only focused on the analysis of the former under conditions  
144 where cake hydraulic resistance is negligible [31].

145

### 146 **1.3. Definition of fouling and objectives of the current study**

147 The controversy over FO and RO fouling in prior studies has provided an impetus for us to  
148 perform an insightful comparison of fouling between FO and RO processes. It is noted that the  
149 majority of prior studies comparing fouling between FO and RO were based solely on the water  
150 flux profiles [18-26]. It was generally assumed that a water decline was an appropriate metric  
151 for fouling behaviour in both cases [18-26]. However, this overlooks a key difference between  
152 fouling in FO compared with that in RO. This is because water flux decline is dependent not  
153 only on fouling but also on driving force (i.e., osmotic pressure for FO and hydraulic pressure  
154 for RO), as shown below:

$$155 \quad J = \frac{F}{\mu R} \quad (1)$$

156 where  $J$  is water flux,  $F$  is driving force,  $\mu$  is viscosity of the solution, and  $R$  is the overall  
157 hydraulic resistance of membrane and foulant layer.

158 Now in this study we specifically define that *fouling* is the accumulation of foulant on the  
159 membrane, and is quantified by the foulant layer resistance ( $R_f$ ), which is consistent with  
160 previous studies quantifying fouling of desalination membranes [32, 40-42]. As CP is flux  
161 dependent, fouling will change the effective driving forces in FO and RO because of changes  
162 in CP. Due to ICP changing with water flux, the changes in effective driving force are  
163 particularly significant for FO [36, 37]. Therefore, for FO processes, an examination of the  
164 decline in flux in isolation does not properly reflect the extent of fouling (i.e., the evolution of  
165 foulant accumulation on the membrane).

166

167 Consequently the current study aims to compare the fouling between FO and RO focusing on  
168 the comparison of hydraulic resistances of the foulant layers with due allowance for the CP  
169 effects. A specific objective is to have a mechanistic understanding of the differences of fouling  
170 and its influences on water flux between FO and RO. To enable a fair comparison our  
171 experiments were designed to use the same membranes, have essentially the same initial water  
172 flux, and have well controlled conditions including constant overall driving force, throughout  
173 the complete experiments as detailed in section 2.2.

174

## 175 **2. Materials and Methods**

### 176 **2.1. Chemicals and membranes**

177 Unless otherwise stated, all the chemicals used in this study were of analytical grade. Ultrapure  
178 deionised (DI) water which was supplied by a Milli-Q Ultrapure water system (Millipore  
179 Singapore Pte Ltd) with a resistivity of 18.2 M $\Omega$  cm was used to prepare all the solutions.  
180 Sodium salt of alginic acid (alginate, Sigma-Aldrich St. Louis, MO) was used as model foulant  
181 to study membrane fouling. It gives gel-layer fouling rather than cake-layer fouling but the  
182 term cake-enhanced concentration polarization (CECP) has been retained in this paper. The  
183 feed solution in both FO and RO fouling experiments was composed of 45 mM NaCl, 5 mM  
184 CaCl<sub>2</sub> and 200 mg/L alginate. The draw solution for FO experiments was composed of 1.5 M  
185 NaCl. The initial volume of the feed solution and draw solution was 5 L.

186

187 A cellulose triacetate (CTA) membrane provided by Hydration Technology Innovations (HTI,  
188 Albany, OR) was used in both FO and RO experimental tests. The CTA membrane comprised  
189 a dense selective layer and a porous support layer embedded within a polyester woven mesh  
190 fabric. This membrane has been widely used as a model membrane to compare fouling in FO  
191 and RO [19-21, 31]. The reason for the use of the same membrane in both FO and RO tests is



192 to eliminate the influence of membrane materials on fouling and thus generate a fair  
193 comparison between fouling in FO and RO.

194

## 195 **2.2. FO and RO membrane fouling experiments**

196 The same experimental setup was used for FO and RO experimental tests with only slight  
197 modification between the two different test modes (Fig. S1 in Supporting Information S1). This  
198 setup has also been used in our previous osmotic membrane fouling experiments and benefits,  
199 *inter alia*, from being able to maintain a constant draw concentration [43]. The setup had a PLC  
200 control system that allowed automatic control of experimental operation and data acquisition.  
201 For FO tests, both FS and DS were recirculated with Hydra-Cell positive displacement  
202 diaphragm pumps (Fig. S1A). The FO membrane test cell (CF042, Sterlitech Corporation) was  
203 comprised of two symmetric Delrin half-cells (top cell and bottom cell) with identical  
204 dimension of flow channel (85 mm length  $\times$  39 mm width  $\times$  2.3 mm height). A net spacer was  
205 placed in the DS flow channel to enhance the mixing and mass transfer of DS [44]. The DS  
206 cross-flow velocity was 11.1 cm/s. The draw solution conductivity (and thus concentration)  
207 was maintained constant by dosing with a more concentrated NaCl solution. The feed solution  
208 conductivity was monitored with time to estimate the reverse solute flux following the same  
209 methods described previously [44]. For RO tests, only FS was recirculated, while the permeate  
210 water was collected directly in a permeate tank (Fig. S1B). There was dosing of the feed with  
211 DI water to ensure concentration was kept constant. The RO membrane test cell had the same  
212 FS flow channel as that for FO. The permeate channel was filled with sintered porous metal  
213 plate (with  $\sim$ 20  $\mu$ m mean pore size) that could fully support the membrane against deformation  
214 in the RO test. The feed and permeate conductivity were monitored with time to estimate the  
215 rejection.

216

217 For both FO and RO tests, the FS tank was placed on a digital balance and the FS mass (and  
218 thus FS volume and foulant concentration) was maintained constant via continuous dosing with  
219 DI water (see Fig S1B). Small amounts of salt leakage did occur from the DS side but the  
220 increase in bulk FS concentration was marginal due to large volume (5 L) of FS used in the  
221 experiments and its influence on the bulk DS and FS osmotic pressure difference is negligible  
222 according to conductivity monitoring. The mass change of the DI water with time was recorded  
223 and used to determine the water flux. No feed spacer was placed in the FS channel to accelerate  
224 fouling. The membrane active layer was facing the FS. The cross-flow velocity of FS was 7.4  
225 cm/s. The apparent driving forces for both RO and FO (i.e., the applied hydraulic pressure for  
226 RO and the osmotic pressure difference between the bulk DS and the bulk FS for FO) were  
227 maintained constant.

228

229 After each fouling test, the fouled membrane was either cleaned via surface flushing to  
230 investigate the fouling reversibility or autopsied to determine the foulant deposition. During  
231 surface flushing, the FS was replaced with DI water and the cross-flow velocity of FS was  
232 increased to 29.6 cm/s. For the FO fouled membrane the DS was also replaced with DI and not  
233 recirculated; for the RO fouled membrane the applied hydraulic pressure was reduced to zero.  
234 In both cases the surface flushing was performed for 30 minutes. The foulant mass load (i.e.,  
235 amount of foulant deposited on unit area of membrane surface) was determined by foulant  
236 extraction followed by measurement of the total organic carbon (TOC) using a similar protocol  
237 reported elsewhere [43]. The protocol is briefly summarized in Supporting Information S2.

238

### 239 **2.3. Determination of foulant resistance for fouled membranes in FO and RO**

240 Before the determination of the foulant resistance on the fouled membranes ( $R_f$ ), the clean  
241 membrane resistance ( $R_m$ ) was first measured via a RO test using a foulant-free feed solution

242 with the same background electrolyte used for the fouling test. The  $R_m$  for the clean membrane  
 243 was estimated using the following osmotic-resistance filtration (ORF) model for RO that was  
 244 reported elsewhere [45] and can be simplified from the universal ORF model for osmotically  
 245 driven membrane processes (ODMPs) ([37] and Appendix A).

$$246 \quad J_w = \frac{\Delta P - \eta_{rej} \pi_{fs} \exp\left(\frac{J_w}{k_{ecp}}\right)}{\mu R_m} \quad (2)$$

247 where  $\Delta P$  is the effective applied hydraulic pressure,  $\eta_{rej}$  is the solute rejection that was  
 248 determined based on conductivity measurement of permeate and feed water,  $\pi_{fs}$  is the osmotic  
 249 pressure of the feed solution (that can be correlated by the van't Hoff equation  $\pi = C\beta R_g T$   
 250 where  $C$  is concentration,  $\beta$  is van't Hoff coefficient,  $R_g$  is the universal gas constant and  $T$  is  
 251 temperature),  $J_w$  is the water flux,  $k_{ecp}$  is the mass transfer coefficient near the membrane  
 252 surface, and  $\mu$  is the viscosity of the feed solution. The membrane resistance  $R_m$  is related to  
 253 the water permeability coefficient ( $A$ ) by  $A = 1/\mu R_m$ . Note that external concentration  
 254 polarization (ECP) has been incorporated in Eq. (2) and that for the feed channel  $k_{ecp}$  can be  
 255 estimated following the approach reported elsewhere [46].

256  
 257 The structural parameter ( $S$ ) of the FO membrane was determined by inputting  $\pi_{ds}$  and  $\pi_{fs}$ ,  
 258 and the foulant-free experimentally obtained parameters (i.e.,  $J_w$ ,  $J_s/J_w$ ,  $R_m$ ) into the following  
 259 equation that is rearranged from the ORF model [37].

$$260 \quad S = \frac{D}{J_w} \ln \left[ \frac{\pi_{ds} + \frac{J_s}{J_w} \beta R_g T}{\left(\pi_{fs} + \frac{J_s}{J_w} \beta R_g T\right) \exp\left(\frac{J_w}{k_{ecp}}\right) + \mu R_m J_w} \right] \quad (3)$$

261 The value of  $k_{ecp}$  was the same value as that estimated for RO because the membrane cell for  
 262 the FO tests and RO tests had the same feed-side flow channel hydrodynamics.

263

264 The foulant resistance ( $R_f$ ) on the RO fouled membrane was determined by inputting  $\pi_{fs}$ ,  $\Delta P$ ,  
 265 and the experimentally obtained  $J_{w,f}$ ,  $\eta_{rej,f}$  into Eq. (4) which, through the term  $k_{ecp,f}$ ,  
 266 includes an adjustment for cake-enhanced concentration polarization (CECP).

$$267 \quad J_{w,f} = \frac{\Delta P - \eta_{rej,f} \pi_{fs} \exp\left(\frac{J_{w,f}}{k_{ecp,f}}\right)}{\mu(R_m + R_f)} \quad (4)$$

268 where  $J_{w,f}$  is the fouling water flux,  $\eta_{rej,f}$  is the membrane rejection during the RO fouling  
 269 test, and  $k_{ecp,f}$  is the overall mass transfer coefficient across the foulant layer and external  
 270 concentration polarization boundary layer. As shown by Eq. (5)  $k_{ecp,f}$  is dependent on both  
 271 the external concentration polarization (ECP) and CECP at the feed side. Thus  $k_{ecp,f}$  consists  
 272 of two terms, one is related to the mass transfer within the foulant layer on the membrane  
 273 ( $k_{ecp,f^*}$ ) and the other to the ECP boundary layer above the foulant layer ( $k_{ecp,0}$ ).

$$274 \quad \frac{1}{k_{ecp,f}} = \frac{1}{k_{ecp,0}} + \frac{1}{k_{ecp,f^*}} = \frac{\delta}{D} + \frac{S_f}{D} = \frac{\bar{S}_f}{D} \quad (5)$$

275 where  $\delta$  is the boundary layer thickness adjacent to the foulant layer and it can be estimated  
 276 from  $k_{ecp}$  for an empty channel [46];  $S_f$  is the structural parameter of the foulant layer that has  
 277 an analogous definition to the membrane structural parameter [31];  $\bar{S}_f$  is the sum of  $\delta$  and  $S_f$   
 278 and is defined as the overall effective thickness of the CP boundary layer that incorporates both  
 279 CECP within the foulant cake layer and the external CP adjacent to the foulant layer. For the  
 280 calculation of  $R_f$ , a range of  $\bar{S}_f$  from 125  $\mu\text{m}$  to 719  $\mu\text{m}$  (where 125  $\mu\text{m}$  is the ECP boundary  
 281 layer thickness) was selected based on the nature of alginate fouling [47]. Clearly the CECP  
 282 effect is negligible when  $\bar{S}_f = 125 \mu\text{m}$ . The selected range of  $\bar{S}_f$  was rationalized via the  
 283 sensitivity analysis as detailed in Appendix B. The numerator of Eq. (4) represents the effective  
 284 driving force for RO during fouling and is used to calculate the RO effective driving force.

285

286 The  $R_f$  on the fouled FO membrane was calculated using  $\pi_{ds}$ ,  $\pi_{fs}$ , and the experimentally  
 287 obtained parameters ( $J_{w,f}$ ,  $J_{s,f}/J_{w,f}$ ,  $R_m$  and  $S$ ) based on the ORF model given below [37].

$$288 \quad J_{w,f} = \frac{(\pi_{ds} - \pi_{fs}) - F_{ecp,f} \left( \pi_{fs} + \frac{J_{s,f}}{J_{w,f}} \beta R_g T \right) - F_{dcp} \left( \pi_{ds} + \frac{J_{s,f}}{J_{w,f}} \beta R_g T \right)}{\mu(R_m + R_f)} \quad (6)$$

289 where the external concentration polarization (ECP) factor,  $F_{ecp,f}$ , at the feed side and dilutive  
 290 concentration polarization (DCP) factor,  $F_{dcp}$ , at the draw side are expressed by Eq. (7) and  
 291 Eq. (8), respectively.

$$292 \quad F_{ecp,f} = \exp \left( \frac{J_{w,f}}{k_{ecp,f}} \right) - 1 \quad (7)$$

$$293 \quad F_{dcp} = 1 - \exp \left( -\frac{J_{w,f}}{k_{dcp}} \right) = 1 - \exp \left( -\frac{J_{w,f}}{D/S} \right) \quad (8)$$

294 Eq. (6) incorporates the effect of reverse solute diffusion (i.e.,  $J_s/J_w$ ), internal concentration  
 295 polarization (included in  $F_{dcp}$ ), and cake-enhanced concentration polarization (included in  
 296  $F_{ecp,f}$ ). The term  $k_{ecp,f}$  in Eq. (7) was determined by Eq. (5) following similar approaches to  
 297 those for RO. Although a precise value of  $\bar{S}_f$  was not determined in this study, the selected  
 298 range of  $\bar{S}_f$  readily indicates the trend of the calculated  $R_f$  for FO and RO (also refer to  
 299 Appendix B). The numerator of Eq. (6) represents the effective driving force for FO during the  
 300 fouling test and is used to calculate the FO effective driving force. The effect of different  
 301 scenarios of  $k_{ecp,f}$  on the calculated  $R_f$  and effective driving forces for FO and RO fouled  
 302 membranes will be evaluated and compared. ORF models show that the mass transfer  
 303 limitation for RO (Eq. (4)) only lies on the feed side but for FO (Eq. (6)) it lies on both the feed  
 304 and draw (permeate) sides which concurs with an earlier analysis [48]. As shown later ICP (or  
 305  $k_{dcp}$ ) at the draw side plays a significant role in determining the difference between FO and  
 306 RO fouling behaviours. It is important to note that the calculation of  $R_f$  for both FO and RO  
 307 fouled membranes (Eq. (4) and Eq. (6)) is based on the experimentally measured parameters,

308 which is essentially similar to the method for calculating clean membrane resistance  $R_m$  (or  
309 clean membrane water permeability  $A$ ) widely used in the research community [36, 41, 49, 50].

310

### 311 **3. Results and Discussion**

#### 312 **3.1. Comparison of water flux performance between FO and RO**

313 For both FO and RO tests, the initial water flux was controlled at the same level of ~18 LMH  
314 and the respective overall driving forces were maintained constant. Prior to fouling tests,  
315 baseline tests without adding foulant in the FS were performed. The results show that baseline  
316 fluxes for both FO and RO were almost constant during the entire test (Fig. S2 in Supporting  
317 Information S3). Therefore, the flux decline during the fouling test is solely due to the addition  
318 of foulant in the FS.

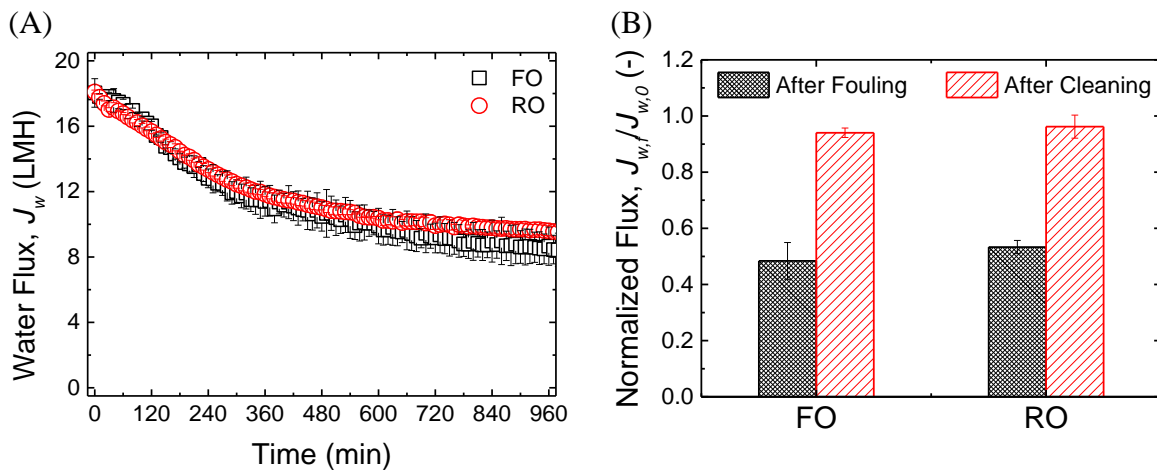
319

320 Fig. 1 shows the water flux behavior during FO and RO fouling tests and flux recovery after  
321 membrane cleaning by water flushing. As shown in Fig. 1a, the water flux decline due to  
322 membrane fouling in both FO and RO followed nearly the same trend. Similar observations  
323 have been reported previously [19, 21, 24]. After the physical cleaning, water fluxes for both  
324 FO and RO recovered significantly (Fig. 1b) with water flux recovery values of ~94% for FO  
325 and ~96% for RO. Given the error bars the difference is not statistically significant. This  
326 observation is different from that reported in previous studies in which flux recovery in FO  
327 was generally much greater than that in RO [19-22, 24, 51]. Our results show that FO and RO  
328 can have similar water flux decline trends during fouling and similar water flux recovery after  
329 physical cleaning.

330

331 However, as introduced in Section 1, the water flux profiles alone do not reflect the extent of  
 332 membrane fouling. The subsequent sections will provide an in-depth analysis of membrane  
 333 fouling in both FO and RO via the comparison of  $R_f$  in both processes.

334



335 Fig.1. Comparison of FO and RO performance. (A) Water flux behavior during membrane  
 336 fouling test, (B) water flux recovery after membrane cleaning. In the FO test DS was 1.5 M  
 337 NaCl; in the RO test applied hydraulic pressure was 17.6 bar. Other fouling experimental  
 338 conditions: FS contained 200 mg/L alginate, 45 mM NaCl and 5 mM  $\text{CaCl}_2$ ; DS contained 1.5  
 339 M NaCl; no spacer was placed in FS flow channel, and a diamond net-type spacer was placed  
 340 in DS flow channel; membrane active layer facing feed solution (AL-FS); cross-flow velocity  
 341 in FS flow channel was 7.4 cm/s and that in DS flow channel was 11.1 cm/s. During membrane  
 342 cleaning (surface flushing), the FS was replaced with DI water and cross-flow velocity  
 343 increased to 29.6 cm/s for 30 minutes.

344

### 345 3.2. Comparison of fouling between FO and RO based on foulant resistance

346 Fig. 2 shows the foulant resistance  $R_f$  during FO and RO fouling calculated from the osmotic-  
 347 resistance filtration models using the experimentally measured  $R_m$  of  $3.26 \times 10^{14} \text{ m}^{-1}$ ,  $S$  of 425  
 348  $\mu\text{m}$ ,  $J_{w,f}$  from Fig. 1, specific reverse solute flux ( $J_{s,f}/J_{w,f}$ ) for FO from Fig. S3 in Supporting  
 349 Information S3, and rejection ( $\eta_{rej,f}$ ) for RO from Fig. S4 in Supporting Information S3. The

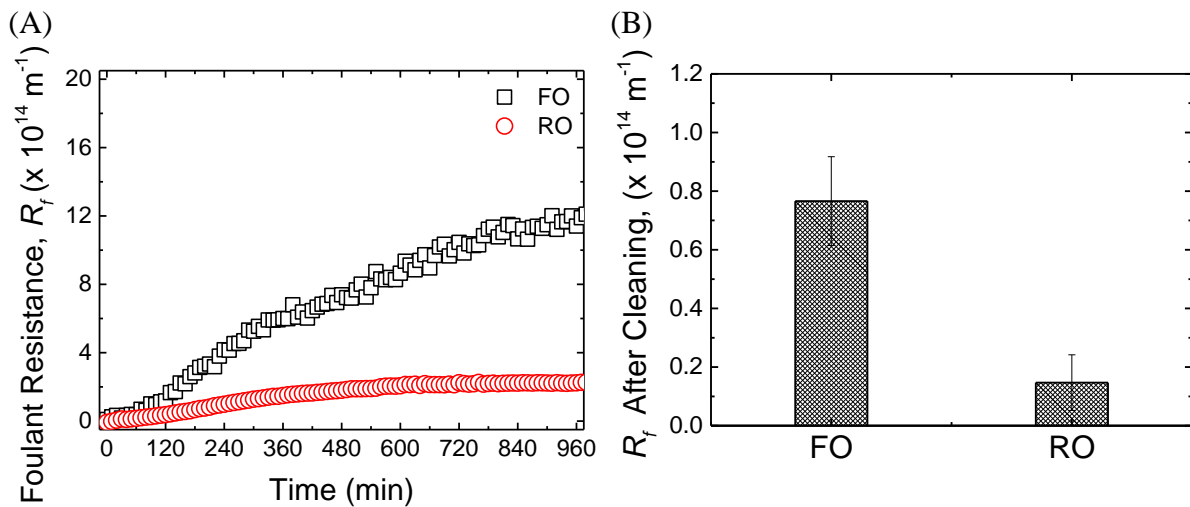
350 calculation of foulant resistance  $R_f$  incorporated the effect of cake-enhanced concentration  
351 polarization as detailed in Appendix B. It was found that  $R_f$  for FO increased to a far greater  
352 extent than that for RO with the progress of fouling (Fig. 2a). At the end of the 16-hour fouling  
353 test,  $R_f$  for FO ( $\sim 12.11 \times 10^{14} \text{ m}^{-1}$ ) was over 5 times that for RO ( $\sim 2.27 \times 10^{14} \text{ m}^{-1}$ ). This  
354 comparison of foulant resistances reveals that for our experimental conditions FO is more prone  
355 to foulant accumulation than RO. More foulant accumulation in FO accords with the finding  
356 of Tow et al. who reported that  $S_f$  for FO would be increasingly greater than RO [31]. A  
357 sensitivity analysis of the effect of cake-enhanced concentration polarization on the calculated  
358  $R_f$  was performed for different scenarios with  $\bar{S}_f$  varying from 125 to 719  $\mu\text{m}$ . It was found  
359 that the overall trend for all of the scenarios is similar to that in Fig. 2a (refer to Fig. B1 in  
360 Appendix B).

361

362 Fig. 2B shows that after physical cleaning the residual foulant resistance  $R_f$  for FO ( $\sim 0.77 \times 10^{14}$   
363  $\text{m}^{-1}$ ) was also much greater than that for RO ( $\sim 0.15 \times 10^{14} \text{ m}^{-1}$ ), although water flux recovery  
364 for both processes was almost the same (Fig. 1B). This shows that basing conclusions solely  
365 upon a comparison of water fluxes, as is common e.g. [19-22, 24, 51], can be misleading. The  
366 trends of water flux (Fig. 1) and foulant resistance (Fig. 2) are reconciled in Section 3.4.

367





369

370 Fig. 2 – (A) Foulant resistance  $R_f$  during membrane fouling, and (B) Foulant resistance  $R_f$  after  
 371 membrane cleaning.  $R_f$  was calculated based on the osmotic-resistance filtration models (Eq.  
 372 (4) for RO and Eq. (6) for FO) using the experimentally measured water flux in Fig. 2, specific  
 373 reverse solute flux ( $J_s/J_w$ ) from Fig. S3 in Supporting Information S3 for FO, rejection for RO  
 374 from Fig. S4 in Supporting Information S3, and basic membrane parameters ( $R_m$  and  $S$ ), and  
 375 incorporating the cake-enhanced concentration polarization.

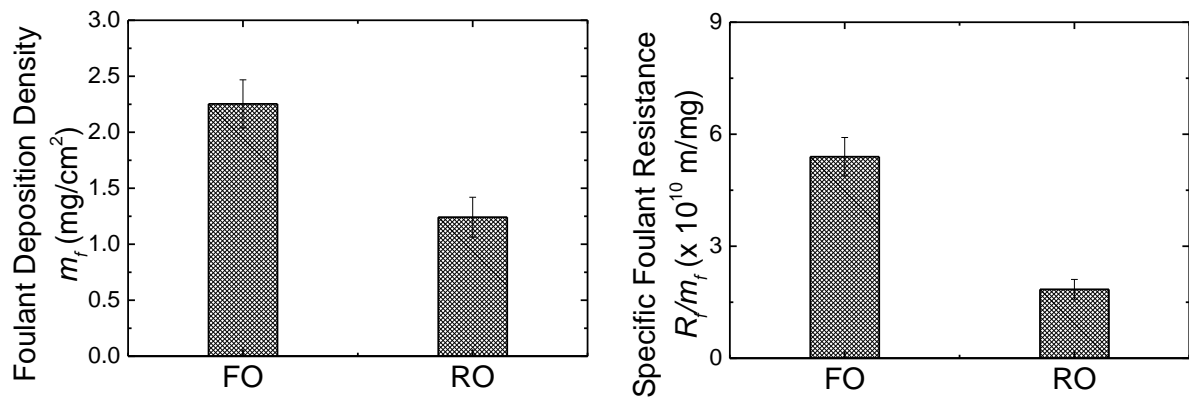
376

377 To further examine the extent of fouling, the fouled membranes were autopsied to ascertain the  
 378 foulant mass deposition density ( $m_f$ ). As shown in Fig. 3a, at the end of the fouling test the  
 379 amount of alginate depositing on the unit area of membrane surface for FO ( $\sim 2.25 \text{ mg/cm}^2$ )  
 380 was nearly 2 times of that for RO ( $\sim 1.24 \text{ mg/cm}^2$ ). Interestingly, the specific foulant resistance  
 381 ( $R_f/m_f$ ) as shown in Fig 3b indicates that the unit amount of alginate depositing on the  
 382 membrane for FO caused greater hydraulic resistance than that for RO;  $R_f/m_f$  for FO  
 383 ( $\sim 5.40 \times 10^{10} \text{ m/mg}$ ) is approximately 3 times of that for RO ( $\sim 1.85 \times 10^{10} \text{ m/mg}$ ).

384

(A)

(B)



385 Fig. 3 – (A) Foulant deposition density,  $m_f$ , (B) Specific foulant resistance  $R_f/m_f$ . To calculate  
 386 the  $R_f/m_f$ ,  $R_f$  was the value at the end of fouling test collected from Fig. 2.

387

388 That greater values of both  $m_f$  and  $R_f/m_f$  were observed for FO is contradictory to some previous  
 389 studies in which it is generally claimed that the foulant layer is less compacted in FO than RO  
 390 owing to the lack of hydraulic pressure in FO [19, 21-24]. Thus experiments specifically  
 391 designed to investigate the effect of hydraulic pressure on the compaction of the foulant layer  
 392 were undertaken, which is discussed later in Section 3.3.

393

394 With regard to the finding of greater values of both  $m_f$  and  $R_f/m_f$  for FO (in comparison to RO)  
 395 it is noted that this does accord with the findings of Song and Elimelech [52] who modelled  
 396 particle transport rates toward a nonporous membrane. They found a significant increase in  
 397 particle deposition upon an increase in salt concentration. Now in FO there is a significantly  
 398 higher salt concentration adjacent to the membrane due to reverse salt diffusion and so greater  
 399 particle deposition can be anticipated. Using experimental evidence provided by the work of  
 400 Sim et al. [53] it was shown that an increase in ionic strength of the feed solution leads to  
 401 increases in cake thickness and decreases in cake porosity which accords exactly with the  
 402 experimental findings reported above for FO. The mechanisms for fouling differences in FO  
 403 and RO are discussed in more detail in section 3.4.

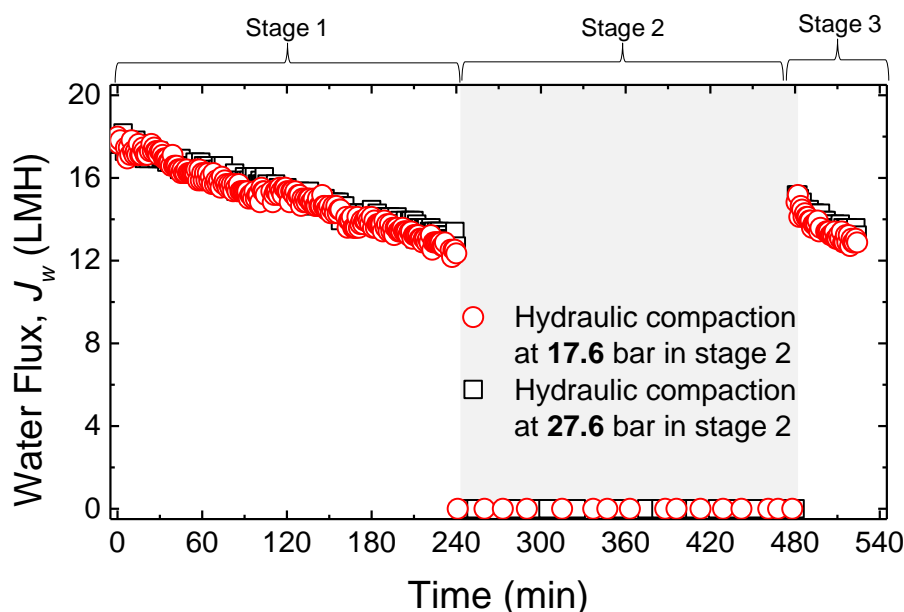
404

### 405 3.3. Effect of hydraulic pressure on the compaction of foulant layer

406 As shown in Fig. S5 in Supporting Information S4, the experiment was divided into three stages:  
407 (1) foulant layer development stage, (2) solely hydraulic pressure compaction stage, and (3)  
408 performance re-evaluation stage. The test results are shown in Fig. 4. The first stage is the  
409 initial 4-hour constant-pressure (~17.6 bar) RO fouling test, at the end of which a foulant layer  
410 had formed on the membrane; the water flux had declined over 30%. In the second stage, the  
411 permeate valve was closed to ensure the permeate water flux was zero, thus eliminating the  
412 flux-induced hydrodynamic drag compaction and only leaving the hydraulic pressure  
413 (maintained at ~17.6 bar or elevated to ~27.6 bar) to “compact” the foulant layer for another 4  
414 hours. In the third stage, the permeate valve was opened again and the permeate water flux was  
415 re-measured under the same pressure used in the first stage (~17.6 bar).

416

417 If the hydraulic pressure plays a more critical role in the “compaction” of the foulant layer than  
418 the water permeation drag force as previously claimed [24], then one would have expected to  
419 find upon reopening of the permeate valve that the hydraulic resistance of the foulant layer ( $R_f$ )  
420 had increased and the water flux had decreased. However, the opposite was found. As shown  
421 in Fig. 4 the water flux was elevated significantly after the fouled membranes had been solely  
422 “compacted” by the hydraulic pressure in the second stage. Importantly it was found that the  
423 level of the water flux elevation was independent of the pressure used to solely “compact” the  
424 foulant layer. The elevated water flux is likely to be due to the removal of some of the foulant  
425 layer by the cross-flow shear force in the absence of flux-induced drag. Our results suggest  
426 that it is the hydrodynamic drag force due to flux rather than the hydraulic pressure *per se* that  
427 plays a critical role in the compaction of the alginate fouling layer.



429

430 Fig. 4 – Effect of hydraulic pressure on the compaction of foulant layer. Water flux at different  
 431 stages is shown. Stage 1 is normal RO operation at 17.6 bar; in stage 2 foulant layer is  
 432 compacted only by hydraulic pressure at either 17.6 bar or 27.6 bar in which the permeate valve  
 433 is closed and permeate water flux is zero; in stage 3 the permeate valve is reopened and the  
 434 water flux is re-evaluated at 17.6 bar after the sole hydraulic compaction in stage 2. Other  
 435 experimental conditions: FS contained 200 mg/L alginate, 45 mM NaCl and 5 mM CaCl<sub>2</sub>; no  
 436 spacer is placed in FS flow channel; cross-flow velocity in FS flow channel is 7.4 cm/s.

437

438 The above findings are in agreement with other previous studies [32, 54, 55]. When studying  
 439 RO and NF membrane fouling by humic acid [54], Tang and Leckie observed a limiting flux  
 440 that is independent of applied pressures (initial water fluxes) and membrane properties,  
 441 suggesting that the foulant layer compaction might not be dominated by pressure but by flux;  
 442 otherwise a limiting flux would not be observed. During the investigation of RO membrane  
 443 fouling under constant-flux operation (see Fig. S6 in Supporting Information S4) and [32],  
 444 Fane et al. found no clear difference in TMP profiles with varying feed pressure for either silica  
 445 or alginate fouling as long as the water flux was maintained constant. They concluded that cake

446 filtration is related to the differential pressure across the fouling layer that is physically related  
447 to flux (Eq. (4)) rather than to the absolute pressure itself [56]. In a recent study Tow and  
448 Lienhard found that alginate gel compaction by high feed hydraulic pressure does not occur  
449 and suggested that other explanations should be sought for FO's fouling resistance relative to  
450 RO [55]. In the following sections we will explore further the mechanisms governing the  
451 different fouling behaviours between FO and RO observed in this study.

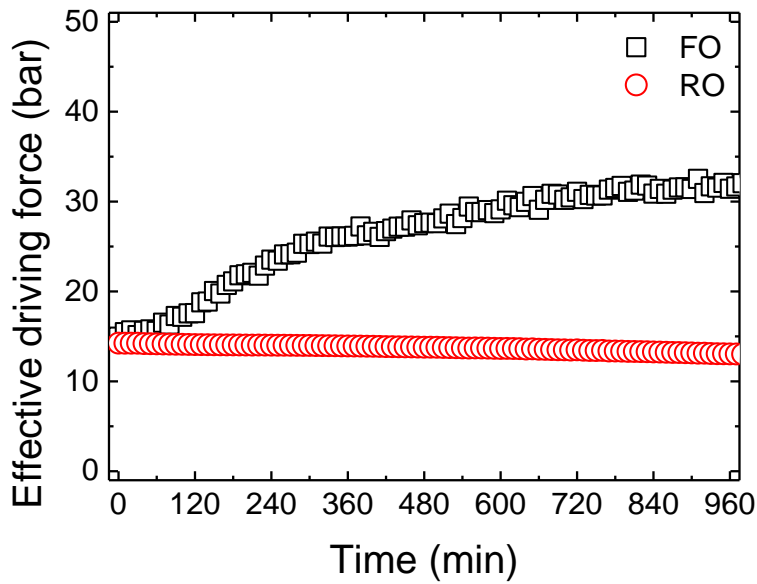
452

### 453 **3.4. Mechanisms for the different fouling behaviours between FO and RO**

#### 454 **3.4.1. ICP self-compensation effect**

455 The difference in foulant accumulation between FO and RO can be attributed primarily to the  
456 different responses of their driving forces to the water flux. For FO the effective osmotic  
457 driving force is significantly influenced by the ICP that is exponentially proportional to the  
458 water flux [35, 36]. A small variation of water flux can result in a significant variation of ICP  
459 and thus effective driving force [36]. Fig. 5 shows the effective driving forces for FO and RO  
460 during the fouling tests. Despite the same effective driving force at the beginning of fouling  
461 tests, the effective driving force for FO increased significantly with the progress of fouling test,  
462 while that for RO slightly decreased. At the end of fouling test the effective driving force for  
463 FO became nearly three times of that for RO. In an earlier study of modelling the effective  
464 driving force for FO and RO under the same extent of fouling, Lay et al. also found that the  
465 effective driving force for FO was greater than that for RO [29].

466



467

468 Fig. 5 – Comparison of effective driving force in FO and RO during the fouling test. Effective  
 469 driving force is the numerator of osmotic-resistance filtration models (Eq. (4) for RO and Eq.  
 470 (6) for FO) and is calculated using the experimentally obtained water flux in Fig. 2, specific  
 471 reverse solute flux ( $J_s/J_w$ ) from Fig. S3 in Supporting Information S3 for FO, rejection for RO  
 472 from Fig. S4 in Supporting Information S3, and basic membrane parameters ( $R_m$  and  $S$ ), and  
 473 incorporating the cake-enhanced concentration polarization.

474

475 The progressively increased effective driving force in FO is due to the ICP self-compensation  
 476 effect [36, 37]. That is, the decreased water flux due to membrane fouling results in a decrease  
 477 in ICP, which in turn leads to an increase in the effective osmotic driving force. The different  
 478 evolution of fouling in FO and RO is elaborated through simulation as discussed in detail in  
 479 Section 3.5 and as shown in Fig. 8 later. Here a pictorial explanation is given. Although there  
 480 are not discrete steps, one can view the evolution of the flux decline as consisting of a number  
 481 of components as depicted in Fig. 6A. The increase in the effective driving force in FO leads  
 482 to partial flux compensation which in turn leads to greater foulant accumulation. More  
 483 accumulation leads to a further decrease in water flux and with the decreased water flux (and

484 the consequent ICP self-compensation) the process continues until there is a balance between  
485 foulant being convected to the surface and foulant being removed by crossflow.

486

487 In contrast, the effective driving force for RO (i.e., the difference between the hydraulic  
488 pressure and the osmotic pressure) responds much less significantly to the change of water flux,  
489 noting that in RO only external CP changes with flux but hydraulic pressure is maintained  
490 constant. It could even decrease with the progression of fouling due to increased cake-enhanced  
491 concentration polarization (Fig. 6B). Thus, the compensation for partial flux decline is much  
492 weaker or does not exist for RO. Consequently, the increase of foulant resistance for RO is  
493 much smaller than that for FO (Fig. 2A) and the foulant deposit in RO is smaller than FO (Fig.  
494 3A).

495

496 The evolution of foulant accumulation ( $R_f$ ) can also be explained mathematically by  
497 differentiating the water flux equation  $J = F/\mu R$  with respect to time ( $t$ ), which is easy to show  
498 that

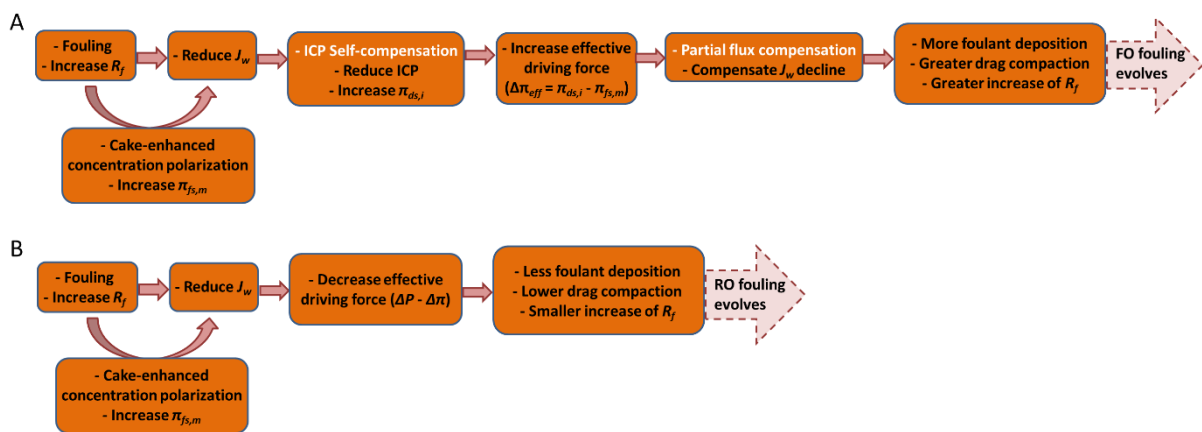
$$499 \quad \frac{dR/dt}{R} = \frac{(-dJ/dt)}{J} + \frac{dF/dt}{F} \quad (9)$$

500 where  $J$  is water flux,  $F$  is driving force, and  $R$  is resistance. Thus the relative increase in  
501 resistance at any point during the evolution of the resistance is the sum of the relative flux  
502 decline and the relative increase in driving force. In the case of RO the third term is negligible  
503 or very small but in the case of FO it is not. Therefore for the similar water flux decline profile,  
504 the increase in resistance for FO is increasingly greater than RO.

505

506 It is well known that for a compressible filter cake that the porosity at the bottom, i.e., nearest  
507 the support (it is membrane in our case), is lowest. This is because the bottom layers of the  
508 cake have to support the drag forces imparted on the top layer of the cake. If there is more drag

509 (due to larger deposit and  $R_f$  as in FO at compensated partial flux), the bottom of the cake is  
 510 more compressed. Our measured specific cake resistance is the cake average value, but this  
 511 may be dominated by the effect of the bottom layer. This offers a partial explanation for the  
 512 greater specific cake resistance for FO than RO (Fig. 3B) or more exactly an explanation for  
 513 augmentation of the higher specific cake resistance. This might also explain that, under the  
 514 same surface flushing conditions, the residual foulant resistance for FO fouled membrane was  
 515 greater than RO fouled membrane (Fig. 2B), since the bottom cake layer might dominate the  
 516 overall specific cake resistance. A second reason for higher specific resistance in FO is that  
 517 the diffusiophoretic gradient is higher within the cake layer [57] due to reverse solute diffusion  
 518 and as noted in this previous study this could lead to cake compaction by diffusiophoresis (DP);  
 519 this will be discussed further in Section 3.4.3.



520  
 521 Fig. 6 – Evolution of membrane fouling in (A) FO and (B) RO. The relationship linking fouling,  
 522 water flux, ICP self-compensation, cake-enhanced concentration polarization (CECP), and  
 523 effective driving forces in FO and RO is schematically illustrated.

524

### 525 3.4.2. Cake-enhanced concentration polarization (CECP)

526 The results in Fig. 5 on the analysis of effective driving force also suggest that cake-enhanced  
 527 concentration polarization (CECP) might play a less important role in FO in the AL-FS  
 528 orientation compared to that in RO for the alginate fouling in this study. For RO, CECP could



529 result in the decrease of effective driving force that further aggravates the decrease of water  
530 flux. However, for FO, CECP would not change the trend where the effective driving force  
531 tends to increase with the progress of fouling. The reasoning again relates to the ICP self-  
532 compensation effect – the decreased ICP at the draw side due to the decrease of water flux by  
533 fouling was much more significant than the cake-enhanced CP at the feed side in this study.  
534 This was further demonstrated through sensitivity analysis for a wide range of scenarios - see  
535 Fig. B1b in Appendix B. This shows that the increase of effective driving force for FO could  
536 be moderately slowed down at an increased CECP, but the overall trend (effective driving force  
537 for FO significantly > RO) remains unchanged as long as the fouling continues to lead to an  
538 increase of foulant resistance ( $R_f$ ). This finding supports an earlier study on the modelling of  
539 the effect of feed concentration on FO water flux, where She et al. suggested that CECP might  
540 not be important for FO in the AL-FS membrane orientation due to the strong ICP self-  
541 compensation effect [37].

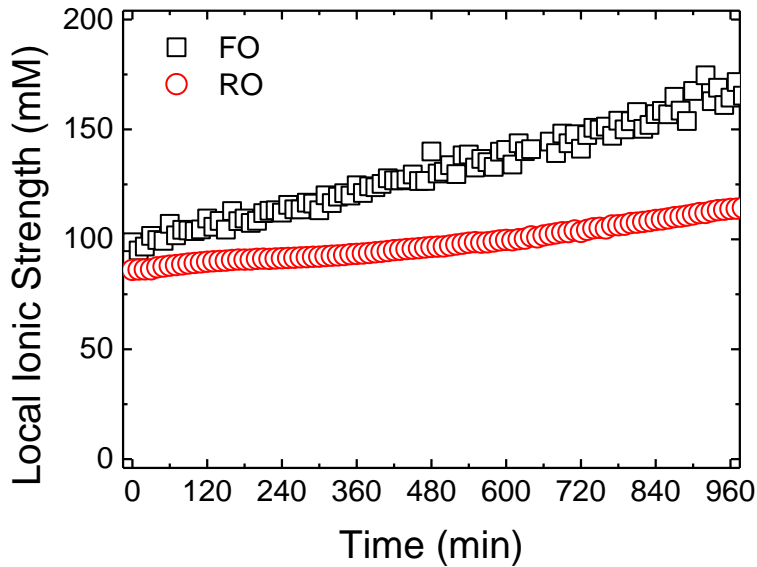
542

### 543 **3.4.3. Reverse solute diffusion (RSD)**

544 The reverse diffusion of draw solute into the FS can influence the fouling behaviour (either  
545 increasing or decreasing fouling) due to the change of local feed solution chemistry near the  
546 FO membrane surface, which has been identified to be a unique fouling mechanism for  
547 osmotically driven membrane processes [37, 43, 44]. In the current study the reverse diffusion  
548 of NaCl from DS into FS would elevate the ionic strength of FS. As shown in Fig. 7, it was  
549 estimated, based on the approach reported previously [58, 59], that the local ionic strength near  
550 the active layer surface was elevated from ~98 mM at the beginning to ~167 mM at the end of  
551 the fouling test due to both CECP and reverse solute diffusion (RSD). In comparison, during  
552 the RO fouling test the bulk FS ionic strength is constant (~60 mM) and the local ionic strength  
553 near the active layer surface was elevated from ~86 mM to ~117 mM due to CECP. It has been

554 reported that with an increased ionic strength, the alginate fouling rate reduces when the feed  
555 solution has a high  $\text{Ca}^{2+}$  concentration ( $> 1 \text{ mM}$ ) [60, 61] due to the reduced binding affinity  
556 between  $\text{Ca}^{2+}$  and carboxyl units of the organic compounds [60-63]. Owing to the relatively  
557 high  $\text{Ca}^{2+}$  concentration (5 mM) in the feed solution in the present study, it was expected that  
558 the increased ionic strength at the feed side in FO due to reverse diffusion of NaCl could lead  
559 to a decreased specific cake resistance. However experimental results in Fig. 2 show that the  
560 specific cake resistance as well as the overall foulant resistance was greater for FO compared  
561 with RO. This suggests there would be other reasons. In addition to the ICP self-compensation  
562 as discussed in Section 3.4.1, another potential contributing effect is diffusiophoresis (DP) [57].  
563

564 In FO, due to RSD, there would be a steeper concentration gradient of salinity across the foulant  
565 layer, which aligns with estimates in Fig 7. This would invoke a stronger diffusiophoresis (DP)  
566 effect in FO than RO, particularly if the feed solution is of low salinity. This stronger effect in  
567 FO may not only lead to a great specific resistance but could also augment the degree of  
568 deposition. Whilst the greater foulant load in FO compared to RO is definitely due in part to  
569 the decrease in the intensity of ICP with time, and hence the increase in effective driving force,  
570 it may be augmented by DP. Previous work has shown that the critical flux for a feed consisting  
571 principally of humic acid had a lower value with an RO membrane compared to the value for  
572 a UF membrane. (Taheri paper JMS 2015). Now this was partially attributed to DP because  
573 for the RO membrane (unlike the UF one) salt gradients would be established. The plateau  
574 fluxes in Fig. 1 (which can be taken as a measure of the critical fluxes) are lower for FO than  
575 RO by around 10-15% and this accords with DP having a potential role in determining the net  
576 flux of foulants towards the membrane surface.  
577



578

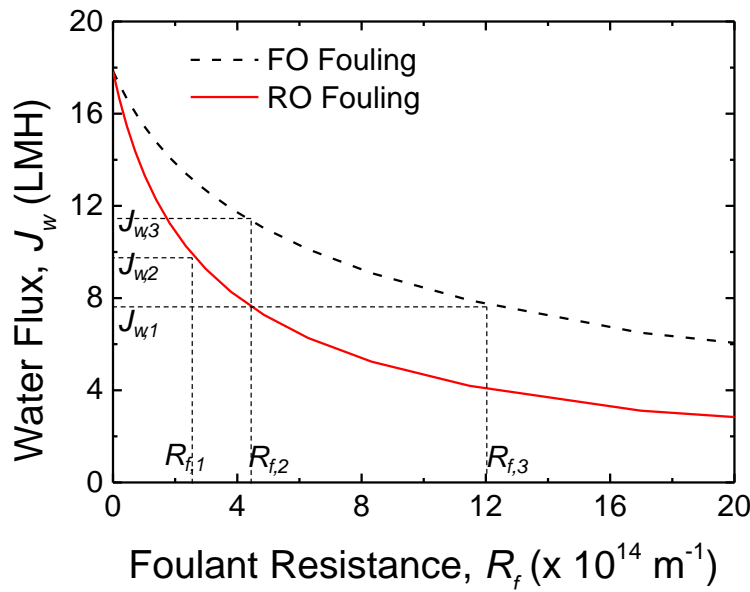
579 Fig. 7 – Estimated local ionic strength near the membrane active layer surface for FO and RO  
 580 during the fouling test. The calculation of local ionic strength followed the method reported  
 581 previously [58, 59] incorporating cake-enhanced concentration polarization.

582

### 583 3.5. Implications

584 The above experimental results show that although FO is more prone to fouling in terms of  
 585 more foulant accumulation and greater foulant hydraulic resistance than RO, the water flux in  
 586 FO might be more stable against fouling, which could enable FO to be a more resilient process  
 587 in some applications. This is further elaborated in this section by the simulation of FO and RO  
 588 water fluxes as a function of the extent of fouling (i.e., foulant resistance) in Fig. 8. The slope  
 589 of Fig. 8 was mathematically derived in Appendix C to further help the analysis of the fouling  
 590 behaviour. For the same extent of fouling (i.e., at the same  $R_f$  when  $R_f > 0$ ) it is apparent that  
 591 the water flux for FO is intrinsically higher than that for RO, demonstrating the superiority of  
 592 FO to RO in terms of water flux performance. However, the same extent of fouling will not be  
 593 a stable condition in a practical operation. This is because a higher flux in FO under such  
 594 conditions would bring more foulants towards the membrane and lead to a greater  
 595 hydrodynamic drag force, which would result in more foulant accumulation in FO and in turn

596 more flux decline, as exhibited in our experimental observations (see Sections 3.1 and 3.2). Fig.  
 597 8 also shows that at the same level of water flux during fouling tests the foulant resistance for  
 598 FO has to be greater than that for RO. This concurs with our experimental observations in  
 599 Section 3.2. As illustrated in Fig. 8, in some cases FO can exhibit higher flux even at more  
 600 severe fouling (e.g., water flux of FO at foulant resistance of  $R_{f,2}$  is greater than that of RO at  
 601  $R_{f,1}$ ).



602  
 603 Fig. 8. Simulation of water flux of FO and RO as a function of foulant resistance. The  
 604 simulation is based on the osmotic-resistance filtration models (Eq. (4) and Eq. (6)) assuming  
 605 that membrane fouling only leads to the increase of  $R_f$  while other membrane parameters (solute  
 606 permeability coefficient  $B$  value and structural parameter  $S$  value) are unchanged. Cake-  
 607 enhanced concentration polarization (CECP) is considered in the simulation by assuming  $\bar{S}_f$   
 608 increases from 125  $\mu\text{m}$  to 500  $\mu\text{m}$ . For the clean membrane the  $R_m$  is  $3.26 \times 10^{14} \text{ m}^{-1}$ , the  $B$   
 609 value is  $4.47 \times 10^{-7} \text{ m/s}$ , and the  $S$  value is 425  $\mu\text{m}$ .

610  
 611 During membrane cleaning, the water flux for FO can be recovered to a higher level than RO  
 612 even though the fouled FO membrane is not cleaned to the same extent as the fouled RO  
 613 membrane. This point is illustrated in Fig. 8. Considering that the foulant resistance for FO is

614 reduced from  $R_{f,3}$  to  $R_{f,2}$  after membrane cleaning (Fig. 8), the water flux for FO will still be  
615 greater than that for RO when the foulant resistance is reduced from  $R_{f,2}$  to  $R_{f,1}$  in Fig. 8. Again  
616 this indicates that the change of water flux in FO in response to a given change of foulant  
617 resistance (i.e. fouling) is much less than that in RO. This also explains why fouling  
618 reversibility, based on measured water fluxes, appears to be more effective for FO than RO  
619 [19-25, 27]. This is not due to the foulant layer in FO being less compacted (indeed on the  
620 contrary we found the specific resistance to be higher for FO) but because the change in ICP  
621 (and thus the change in effective driving force) in FO leads to a higher flux in the presence of  
622 residual fouling.

623

624 The above modelling does not incorporate the influence of diffusiophoretic deposition (DP).  
625 As elegantly illustrated in Fig.9 of their paper, Guha et al [57] showed that for filtration-based  
626 particle deposition leading to convective cake formation one can often expect filtration-based  
627 ion concentration polarization which leads to diffusiophoretic movement augmenting particle  
628 deposition and this in turn creates both a greater level of cake formation and compaction, and  
629 further enhanced ion concentration polarization. Thus there is a positive feed-back loop further  
630 enhancing fouling.

631

632 In summary, the advantage of greater water flux stability of FO over RO is due to the ICP self-  
633 compensation effect for FO that can result in a partial water flux compensation and leverage  
634 the water flux decline by increasing the effective driving force. Our results suggest that,  
635 contrary to earlier reports, FO does not benefit from less foulant compression due to its low  
636 hydraulic pressure operation. While ICP is generally regarded as a detrimental effect for FO,  
637 the current study reveals that ICP can also have an upside in that it helps to maintain water flux  
638 stability. An interesting corollary to this is that the quest for FO membranes with smaller and

639 smaller  $S$  values to reduce ICP needs to consider whether there is an optimal  $S$  value that  
640 balances the magnitude of flux decline and the resilience that the ICP compensation imparts  
641 upon the system.

642

#### 643 **4. Conclusions**

644 In this study the differences in membrane fouling between FO and RO were explored under  
645 comparably controlled experimental conditions in which the apparent driving forces for FO  
646 and RO were maintained constant. Sodium alginate was the foulant. It was found that:

- 647 1. Water flux decline during both FO and RO fouling tests followed broadly the same trend  
648 and water flux recovery after membrane cleaning for both FO and RO reached a similar  
649 level. However, the driving forces of FO and RO respond differently to the progression of  
650 fouling and as a result the foulant resistance for FO was increasingly greater than that for  
651 RO.
- 652 2. Membrane autopsy after the fouling tests showed that more foulant had been deposited on  
653 the FO fouled membrane than the RO fouled membrane. Also, the specific foulant  
654 resistance was greater with FO than RO.
- 655 3. The dominant reason for the higher fouling propensity in FO is due to the change of ICP  
656 and effective driving force in response to the evolution of fouling; it is true for all systems  
657 with manifest ICP.
- 658 4. Calculations suggest that CECP does not play an important role in flux decline in FO due  
659 to the dominance of the ICP self-compensation effect.
- 660 5. The reverse diffusion of draw solute into feed solution could also influence fouling in FO  
661 in two ways. Firstly directly as a result in the change of feed solution chemistry, an effect  
662 that is strongly dependent on the draw solution properties. Secondly RSD will also  
663 influence the salinity gradient across the FO foulant layer. This gradient will be greater in

664 FO than the corresponding one for RO and probably led to diffusiophoresis (DP). The role  
665 of DP in FO is worthy of further investigation.

666 6. No evidence was found that hydraulic pressure in RO plays a critical role in the compaction  
667 of alginate fouling layers. Furthermore the generally observed high flux reversibility of  
668 FO after membrane cleaning is probably due to the change of ICP (and thus effective  
669 driving force) in response to fouling rather than the lack of compaction due to hydraulic  
670 pressure.

671 7. Overall and notwithstanding its higher fouling propensity, FO was found to exhibit higher  
672 flux stability against membrane fouling. Excluding those applications where the reverse  
673 salt flux generates additional fouling FO is potentially a more resilient process than RO.

674

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683

#### 684 **Appendix A. Derivation of osmotic-resistance filtration model for RO**

685 The osmotic-resistance filtration model of Eq. (A1) is originally derived for osmotically driven  
686 membrane processes and differentiates all the driving forces incorporating concentration  
687 polarization and reverse solute diffusion [37].

$$688 \quad J_w = \frac{(\pi_{ds} - \pi_{fs}) - F_{ecp} \left( \pi_{fs} + \frac{J_s}{J_w} \beta R_g T \right) - F_{dcp} \left( \pi_{ds} + \frac{J_s}{J_w} \beta R_g T \right)}{\mu R_m} \quad (A1)$$

689 where  $F_{ecp}$  and  $F_{dcp}$  are the concentrative external concentration polarization (ECP) factor at  
 690 the feed side and dilutive concentration polarization (DCP) factor at the draw side respectively.  
 691 They are expressed by Eq. (A2) and Eq. (A3).

$$692 \quad F_{ecp} = \exp\left(\frac{J_w}{k_{ecp}}\right) - 1 \quad (A2)$$

$$693 \quad F_{dcp} = 1 - \exp\left(-\frac{J_w}{k_{dcp}}\right) = 1 - \exp\left(-\frac{J_w}{D/S}\right) \quad (A3)$$

694 Eq. (A1) is also applicable for RO and can be expressed as Eq. (A4) considering the direction  
 695 of water flux and solute flux as well as the redefinition of signs to represent the parameters for  
 696 RO.

$$697 \quad J_w = \frac{\Delta P - (\pi_{fs} - \pi_p) - F_{ecp}\left(\pi_{fs} - \frac{J_s}{J_w}\beta R_g T\right)}{\mu R_m} \quad (A4)$$

698 At equilibrium DCP does not exist in RO and the specific solute flux  $\left(\frac{J_s}{J_w}\right)$  can be correlated to  
 699 the permeate concentration ( $c_p$ ) by Eq. (A5).

$$700 \quad c_p = \frac{J_s}{J_w} \quad (A5)$$

701 Inserting Eq. (A2) and Eq. (A5) into Eq. (A4) yields the expected equation:

$$702 \quad J_w = \frac{\Delta P - (\pi_{fs} - \pi_p) \exp\left(\frac{J_w}{k_{ecp}}\right)}{\mu R_m} \quad (A6)$$

703 By assuming that the concentration and osmotic pressure follow the van't Hoff equation, the  
 704 solute rejection in RO can be expressed below.

$$705 \quad \eta_{rej,f} = 1 - \frac{c_p}{c_f} = 1 - \frac{\pi_p}{\pi_f} \quad (A7)$$

706 Inserting Eq. (A7) into Eq. (A6) yields

$$707 \quad J_w = \frac{\Delta P - \eta_{rej} \pi_{fs} \exp\left(\frac{J_w}{k_{ecp}}\right)}{\mu R_m} \quad (A8)$$

708



709 **Appendix B. Sensitivity analysis of the effect of cake-enhanced concentration polarization**  
710 **(CECP) on the calculated values of foulant resistance  $R_f$**

711 This section shows the results of a sensitive analysis in which the influence of assumed levels  
712 of cake-enhanced concentration polarization (CECP) on the calculated  $R_f$  for FO and RO were  
713 explored.  $R_f$  was calculated for the following four scenarios using the experimentally  
714 measured data (i.e.,  $J_w, J_s/J_w, R_f, S, \pi_{ds}$  and  $\pi_{fs}$ ).

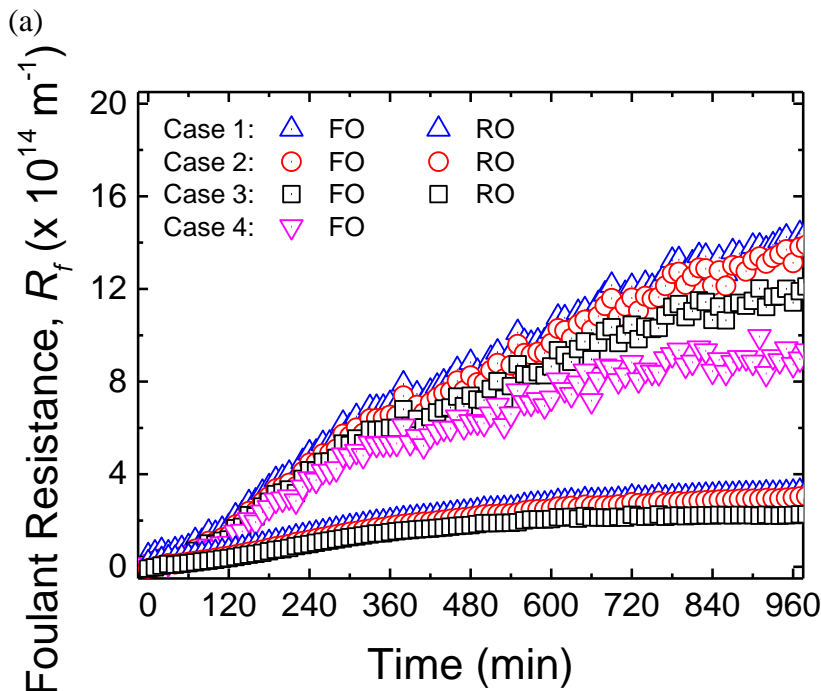
- 715 • Case (1): ECP at the feed side is neglected (i.e., assuming  $\bar{S}_f = 0$  in Eq. (5)).
- 716 • Case (2): ECP at the feed side is considered but CECP is neglected (i.e., assuming  $S_f = 0$   
717 and  $\bar{S}_f = \delta$  in Eq. (5); using  $\bar{S}_f$  of 125  $\mu\text{m}$  that is estimated for empty flow channel  
718 following the method reported elsewhere [46]).
- 719 • Case (3): CECP is considered and  $S_f$  is the same for both FO and RO (i.e., assuming that  
720  $\bar{S}_f$  for both FO and RO increases at the same rate with the progress of fouling test from 125  
721  $\mu\text{m}$  at the beginning of fouling test to 422  $\mu\text{m}$  at the end of fouling test).
- 722 • Case (4): CECP is considered and  $S_f$  for FO becomes increasingly greater than that for RO  
723 based on the analysis of Tow et al. [31] (i.e.,  $\bar{S}_f$  for FO increases faster with the progress  
724 of fouling than that for RO; specifically it was assumed that  $\bar{S}_f$  for FO increases from 125  
725  $\mu\text{m}$  to 719  $\mu\text{m}$  whilst that for RO increases from 125  $\mu\text{m}$  to 422  $\mu\text{m}$  during the fouling  
726 tests).

727 As shown in Fig. B1a, for all the scenarios  $R_f$  for both FO and RO increased with the progress  
728 of fouling test. Moreover, although the increase of concentration polarization from Case (1) to  
729 (4) at a fixed time could decrease the calculated  $R_f$  for both FO and RO, for all scenarios  $R_f$   
730 for FO becomes increasingly greater than that for RO. Note that for Case (4) the selected range  
731 of  $\bar{S}_f$  for FO from 125  $\mu\text{m}$  to 719  $\mu\text{m}$  over the testing period approaches an extreme condition  
732 in which a further faster increase in  $\bar{S}_f$  (i.e. a more severe rate of foulant accumulation) would

733 result in a decrease in the calculated  $R_f$  (see Fig. B1c for Case (5)) which is unrealistic. In  
 734 addition, the assumption of a faster increase of  $\bar{S}_f$  for FO in Case (4) has already suggested that  
 735 foulant accumulation for FO is more severe than that for RO [31]. As there is much greater  $R_f$   
 736 for FO with the progress of fouling, these results further corroborate our observation that FO  
 737 has a greater fouling propensity than RO even under the extreme conditions considered here.  
 738 Although a more valid method needs to be developed to determine the accurate  $\bar{S}_f$  value, the  
 739 sensitive analysis with  $\bar{S}_f$  varying within the boundary conditions can clearly demonstrate that  
 740  $R_f$  for FO is always greater than RO under the experimental conditions in the current study.

741

742



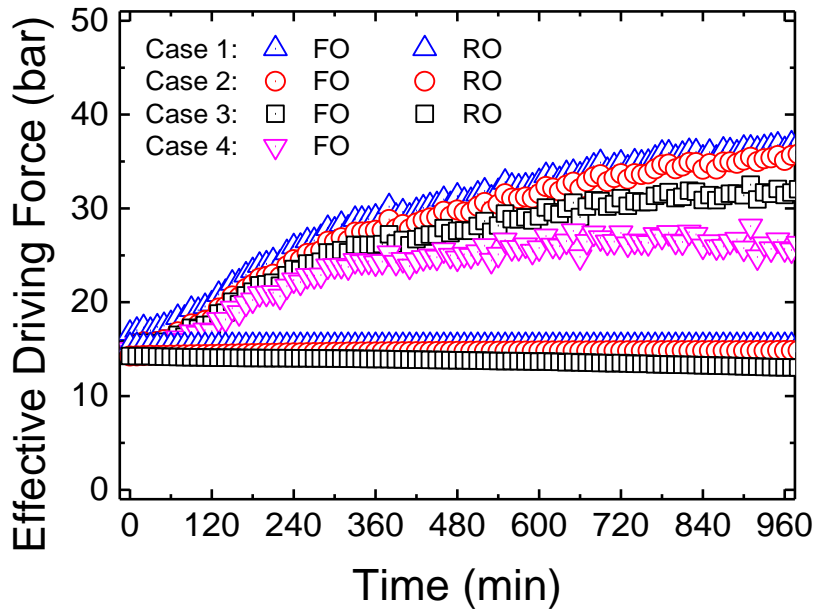
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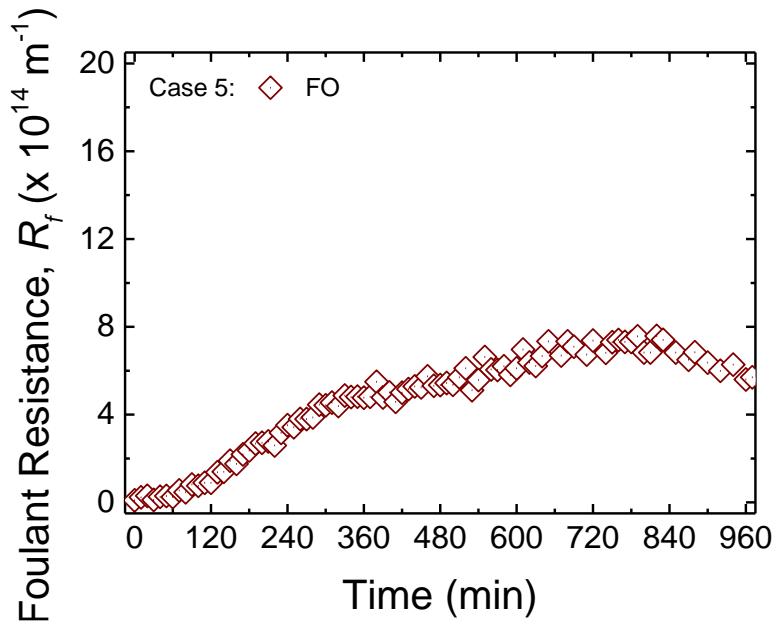
746

(b)



747  
748  
749

(c)



750  
751

752 Fig. B1 – Calculated foulant resistance ( $R_f$ ) (a) and effective driving force (b) for FO and RO  
753 based on the osmotic-resistance filtration model in different scenarios: (1) cake-enhanced  
754 concentration polarization (CECP) is neglected (i.e., assuming that the ECP boundary layer  
755 thickness ( $\delta$ ) is zero), (2) CECP is considered but cake-enhanced concentration polarization is  
756 neglected (i.e., using  $\delta$  of 125  $\mu\text{m}$  that is estimated for empty flow channel following the  
757 method reported elsewhere [46]), (3) CECP is considered and the foulant layer structural  
758 parameter ( $S_f$ ) is the same for both FO and RO (i.e.,  $(\delta + S_f)$  for both FO and RO increases at  
759 the same rate with the progress of fouling test from 125  $\mu\text{m}$  at the beginning to 422  $\mu\text{m}$  at the  
760 end of fouling test), and (4) CECP is considered and the foulant layer structural parameter ( $S_f$ )  
761 for FO becomes increasingly more greater than that for RO (i.e.,  $(\delta + S_f)$  for FO increases  
762 faster with the progress of fouling than that for RO;  $(\delta + S_f)$  for FO increases from 125  $\mu\text{m}$  to  
763 719  $\mu\text{m}$ , while that for RO increases from 125  $\mu\text{m}$  to 422  $\mu\text{m}$ ). For the calculation, the

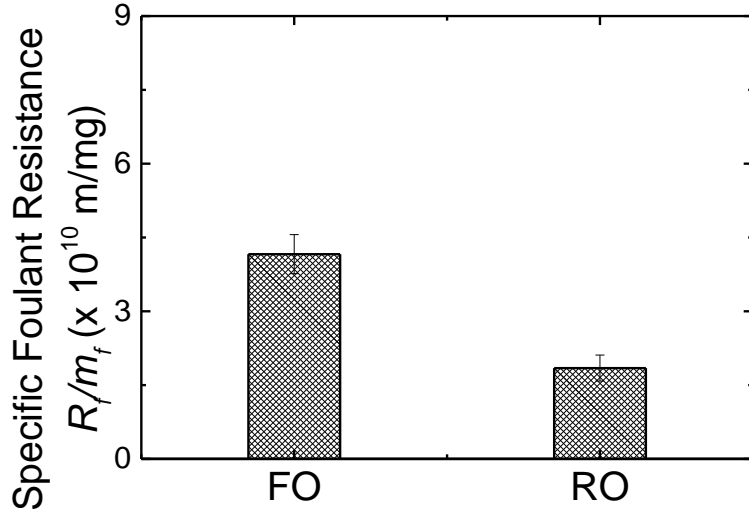
764 experimentally obtained clean membrane resistance  $R_m$  is  $3.26 \times 10^{14} \text{ m}^{-1}$  and structural  
765 parameter  $S$  is  $425 \text{ }\mu\text{m}$ . (c) foulant resistance ( $R_f$ ) for case (5) when the  $\bar{S}_f$  has a faster increase  
766 than that in case (4).  
767

768 Fig. B1b shows the effective driving force for FO and RO. For all the Cases the effective  
769 driving force for FO increased with the progress of fouling test and became increasingly greater  
770 than that for RO. In contrast to FO, the effective driving force for RO behaved differently for  
771 different scenarios: it maintained constant for Case (1), increased gradually with fouling test  
772 for Case (2), and decreased gradually with fouling test for Case (3). Although the increase of  
773 concentration polarization from Case (1) to Case (4) led to the decrease of effective driving  
774 force for both FO and RO at a fixed time of fouling test, the effective driving force for FO was  
775 always increasing with the fouling test and becoming increasingly greater than that for RO.  
776 This suggests that (1) the different response of the effective driving force to fouling test  
777 between FO and RO is the major reason for their different fouling behaviours, and (2) CECP  
778 for FO plays a much less important role in flux decline than it does for RO.

779

780 Fig. B2 shows the calculated specific foulant resistance ( $R_f/m_f$ ) using the experimentally  
781 measured foulant deposition density ( $m_f$ ) and calculated  $R_f$  in Case (4) that is under the  
782 extreme conditions. Interestingly, the specific foulant resistance ( $R_f/m_f$ ) for FO is still  
783 consistently greater than RO even under the case of extreme conditions.

784



785

786 Fig. B2 – Specific foulant resistant ( $R_f/m_f$ ) for FO and RO.  $R_f/m_f$  was calculated using the  $R_f$   
 787 from Fig. B1a in Scenario (4) and the experimentally measured  $m_f$  in Fig. 3A.

788

### 789 Appendix C. Mathematic derivation of flux decline with respect to foulant accumulation

790 For simplicity we write  $R = R_m + R_f$  in the development of (C2) and (C3). We ignore feed

791 side and draw side external concentration polarization for FO (i.e., equation (C2)) and feed side

792 external concentration polarization for RO (i.e., equation (C3)).

793 For FO: 
$$J = \frac{\pi_{ds} \cdot \exp\left(-\frac{J}{k_{dcp}}\right) - \pi_{fs}}{\mu R} \quad (C1)$$

794 Hence for FO the rate of flux decline with respect to fouling is:

795 
$$\frac{dJ}{dR_f} = \frac{-\left(\pi_{ds} \cdot \exp\left(-\frac{J}{k_{dcp}}\right) - \pi_{fs}\right)}{\mu R^2} / \left[1 + \pi_{ds} \cdot \exp\left(-\frac{J}{k_{dcp}}\right) / k_{dcp} R\right] \quad (C2)$$

796 However for RO the corresponding equation to (C2) is:

797 
$$\frac{dJ}{dR_f} = \frac{-\Delta P}{\mu R^2} \quad (C3)$$

798 At the beginning of both experiments  $R \approx R_m$  and the only difference between (C2) and (C3),

799 is the denominator in square brackets in (C2). These expressions are related to the gradient of

800 the curves in Fig. 8. The clear implication is that for a given deposition of foulant (equating to

801 a given  $\Delta R$ ) the change in flux will be smaller in FO than in RO. Now foulant accumulation

802 can be expected to continue at a decreasing rate until there is a balance between deposition, by  
803 convective flow to and through the membrane, and removal by shear [64]. Now given that flux  
804 is declining more slowly with respect to a given amount of foulant accumulation in FO than  
805 RO there will naturally be a greater amount of foulant deposition in FO before the limiting flux  
806 is reached.

807

#### 808 **Appendix D. Supplementary material**

809 The supplementary data can be found online via the link of <http://>

810

811

812

813 **Abbreviations**

814

815	AL-FS	active layer facing feed side
816	CECP	cake-enhanced concentration polarisation
817	CEOP	cake-enhanced osmotic pressure
818	CP	concentration polarisation
819	CTA	cellulose triacetate
820	DCP	dilutive concentration polarisation
821	DI	de-ionised
822	DS	draw solution
823	ECP	external concentration polarisation
824	FO	forward osmosis
825	FS	feed solution
826	ICP	internal concentration polarisation
827	NF	nanofiltration
828	ODMPs	osmotically driven membrane processes
829	ORF	osmotic-resistance filtration
830	RO	reverse osmosis
831	RSD	reverse solute diffusion

832

833

834

835 **Nomenclature**

836

837	$A$	water permeability coefficient ( $\text{m}^3/\text{m}^2\text{-Pa}$ )
838	$B$	solute permeability coefficient ( $\text{m}^3/\text{m}^2$ )
839	$C$	concentration ( $\text{moles}/\text{m}^3$ )
840	$D$	diffusion coefficient ( $\text{m}^2/\text{s}$ )
841	$F_{cecp}$	concentration polarization factor for CECP (dimensionless)
842	$F_{dcp}$	concentration polarization factor for DCP (dimensionless)
843	$F_{ecp}$	concentration polarization factor for ECP (dimensionless)
844	$J_s$	solute flux ( $\text{m}^3/\text{m}^2 \text{ s}$ )
845	$J_w$	water flux ( $\text{m}^3/\text{m}^2 \text{ s}$ )
846	$J_{w,f}$	fouling water flux ( $\text{m}^3/\text{m}^2 \text{ s}$ )
847	$k$	mass transfer coefficient ( $\text{m}/\text{s}$ )
848	$k_{cecp}$	mass transfer coefficient near the membrane surface ( $\text{m}/\text{s}$ )
849	$k_{ecp,f}$	overall mass transfer coefficient across the foulant layer and external concentration polarization boundary layer ( $\text{m}/\text{s}$ )
850		
851	$k_{ecp,f}^*$	mass transfer coefficient within the foulant layer on the membrane ( $\text{m}/\text{s}$ )
852	$k_{ecp,o}$	mass transfer coefficient to the ECP boundary layer above the foulant layer ( $\text{m}/\text{s}$ )
853	$M$	molar ( $\text{moles}/\text{m}^3$ )
854	$m_f$	mass deposition density ( $\text{g}/\text{m}^2$ )
855	$R_f$	foulant resistance ( $\text{m}^{-1}$ )
856	$R_f/m_f$	specific foulant resistance ( $\text{m}/\text{g}$ )
857	$R_g$	universal gas constant ( $8.3145 \text{ m}^3 \text{ Pa mol}^{-1} \text{ K}^{-1}$ )
858	$R_m$	hydraulic resistance of the membrane ( $\text{m}^{-1}$ )
859	$S$	structural parameter ( $\text{m}^{-1}$ )

860	$S_f$	structural parameter of the foulant layer ( $m^{-1}$ )
861	$\bar{S}_f$	overall effective thickness of the CP boundary layer (m)
862	$T$	temperature (K)
863	$TMP$	transmembrane pressure (Pa)
864	$TOC$	total organic carbon ( $g/m^3$ )
865	$\beta$	van't Hoff coefficient
866	$\delta$	boundary layer thickness (m)
867	$\mu$	fluid viscosity (Pa s)
868	$\eta_{rej}$	solute rejection
869	$\pi_{ds}$	osmotic pressure of the draw solution (Pa)
870	$\pi_{fs}$	osmotic pressure of the feed solution (Pa)
871	$\pi_{fs,m}$	osmotic pressure of the feed solution at the membrane surface (Pa)
872	$\pi_i$	osmotic pressure of the draw at the interface between the active layer of the
873		membrane and the support
874	$\Delta\pi$	osmotic pressure difference across the membrane (Pa)
875	$\Delta P$	effective applied hydraulic pressure (Pa)
876		
877		



878 **References**

- 879 [1] T.Y. Cath, A.E. Childress, M. Elimelech, Forward osmosis: Principles, applications, and  
880 recent developments, *J. Membr. Sci.*, 281 (2006) 70-87.
- 881 [2] S. Zhao, L. Zou, C.Y. Tang, D. Mulcahy, Recent developments in forward osmosis:  
882 Opportunities and challenges, *J. Membr. Sci.*, 396 (2012) 1-21.
- 883 [3] R. Valladares Linares, Z. Li, S. Sarp, S. Bucs, G. Amy, J.S. Vrouwenvelder, Forward  
884 osmosis niches in seawater desalination and wastewater reuse, *Water Res.*, 66 (2014) 122-139.
- 885 [4] G. Blandin, A. Verliefde, J. Comas, I. Rodriguez-Roda, P. Le-Clech, Efficiently Combining  
886 Water Reuse and Desalination through Forward Osmosis—Reverse Osmosis (FO-RO) Hybrids:  
887 A Critical Review, *Membranes*, 6 (2016) 37.
- 888 [5] K. Lutchmiah, A.R.D. Verliefde, K. Roest, L.C. Rietveld, E.R. Cornelissen, Forward  
889 osmosis for application in wastewater treatment: A review, *Water Res.*, 58 (2014) 179-197.
- 890 [6] X. Wang, V.W.C. Chang, C.Y. Tang, Osmotic membrane bioreactor (OMBR) technology  
891 for wastewater treatment and reclamation: Advances, challenges, and prospects for the future,  
892 *J. Membr. Sci.*, 504 (2016) 113-132.
- 893 [7] B.D. Coday, B.G.M. Yaffe, P. Xu, T.Y. Cath, Rejection of Trace Organic Compounds by  
894 Forward Osmosis Membranes: A Literature Review, *Environmental Science & Technology*,  
895 48 (2014) 3612-3624.
- 896 [8] X. Wang, J. Zhang, V.W.C. Chang, Q. She, C.Y. Tang, Removal of cytostatic drugs from  
897 wastewater by an anaerobic osmotic membrane bioreactor, *Chem. Eng. J.*, 339 (2018) 153-161.
- 898 [9] X. Jin, Q. She, X. Ang, C.Y. Tang, Removal of boron and arsenic by forward osmosis  
899 membrane: Influence of membrane orientation and organic fouling, *J. Membr. Sci.*, 389 (2012)  
900 182-187.
- 901 [10] J. Zhang, Q. She, V.W.C. Chang, C.Y. Tang, R.D. Webster, Mining Nutrients (N, K, P)  
902 from Urban Source-Separated Urine by Forward Osmosis Dewatering, *Environ. Sci. Technol.*,  
903 48 (2014) 3386-3394.
- 904 [11] A.J. Ansari, F.I. Hai, W.E. Price, J.E. Drewes, L.D. Nghiem, Forward osmosis as a  
905 platform for resource recovery from municipal wastewater - A critical assessment of the  
906 literature, *J. Membr. Sci.*, 529 (2017) 195-206.
- 907 [12] S. Phuntsho, H.K. Shon, S. Hong, S. Lee, S. Vigneswaran, A novel low energy fertilizer  
908 driven forward osmosis desalination for direct fertigation: Evaluating the performance of  
909 fertilizer draw solutions, *J. Membr. Sci.*, 375 (2011) 172-181.
- 910 [13] D.L. Shaffer, J.R. Werber, H. Jaramillo, S. Lin, M. Elimelech, Forward osmosis: Where  
911 are we now?, *Desalination*, 356 (2015) 271-284.
- 912 [14] L. Chekli, S. Phuntsho, J.E. Kim, J. Kim, J.Y. Choi, J.-S. Choi, S. Kim, J.H. Kim, S. Hong,  
913 J. Sohn, H.K. Shon, A comprehensive review of hybrid forward osmosis systems: Performance,  
914 applications and future prospects, *J. Membr. Sci.*, 497 (2016) 430-449.
- 915 [15] R.K. McGovern, J.H. Lienhard V, On the potential of forward osmosis to energetically  
916 outperform reverse osmosis desalination, *J. Membr. Sci.*, 469 (2014) 245-250.
- 917 [16] N.M. Mazlan, D. Peshev, A.G. Livingston, Energy consumption for desalination — A  
918 comparison of forward osmosis with reverse osmosis, and the potential for perfect membranes,  
919 *Desalination*, 377 (2016) 138-151.
- 920 [17] T.-S. Chung, L. Luo, C.F. Wan, Y. Cui, G. Amy, What is next for forward osmosis (FO)  
921 and pressure retarded osmosis (PRO), *Sep. Purif. Technol.*, 156 (2015) 856-860.
- 922 [18] A. Achilli, T.Y. Cath, E.A. Marchand, A.E. Childress, The forward osmosis membrane  
923 bioreactor: A low fouling alternative to MBR processes, *Desalination*, 238 (2009) 10-21.
- 924 [19] B. Mi, M. Elimelech, Organic fouling of forward osmosis membranes: Fouling  
925 reversibility and cleaning without chemical reagents, *J. Membr. Sci.*, 348 (2010) 337-345.

- 926 [20] S. Lee, C. Boo, M. Elimelech, S. Hong, Comparison of fouling behavior in forward  
927 osmosis (FO) and reverse osmosis (RO), *J. Membr. Sci.*, 365 (2010) 34-39.
- 928 [21] B. Mi, M. Elimelech, Silica scaling and scaling reversibility in forward osmosis,  
929 *Desalination*, 312 (2013) 75-81.
- 930 [22] Y. Kim, M. Elimelech, H.K. Shon, S. Hong, Combined organic and colloidal fouling in  
931 forward osmosis: Fouling reversibility and the role of applied pressure, *J. Membr. Sci.*, 460  
932 (2014) 206-212.
- 933 [23] S.E. Kwan, E. Bar-Zeev, M. Elimelech, Biofouling in forward osmosis and reverse  
934 osmosis: Measurements and mechanisms, *J. Membr. Sci.*, 493 (2015) 703-708.
- 935 [24] M. Xie, J. Lee, L.D. Nghiem, M. Elimelech, Role of pressure in organic fouling in forward  
936 osmosis and reverse osmosis, *J. Membr. Sci.*, 493 (2015) 748-754.
- 937 [25] F. Lotfi, L. Chekli, S. Phuntsho, S. Hong, J.Y. Choi, H.K. Shon, Understanding the  
938 possible underlying mechanisms for low fouling tendency of the forward osmosis and pressure  
939 assisted osmosis processes, *Desalination*, (2017).
- 940 [26] Y. Yu, S. Lee, S.K. Maeng, Forward osmosis membrane fouling and cleaning for  
941 wastewater reuse, *Journal of Water Reuse and Desalination*, 7 (2017) 111-120.
- 942 [27] J. Lee, S. Kook, C. Lee, I.S. Kim, Effect of intermittent pressure-assisted forward osmosis  
943 (I-PAFO) on organic fouling, *Desalination*, 419 (2017) 60-69.
- 944 [28] G. Blandin, A.R.D. Verliefde, P. Le-Clech, Pressure enhanced fouling and adapted anti-  
945 fouling strategy in pressure assisted osmosis (PAO), *J. Membr. Sci.*, 493 (2015) 557-567.
- 946 [29] W.C.L. Lay, T.H. Chong, C.Y. Tang, A.G. Fane, J. Zhang, Y. Liu, Fouling propensity of  
947 forward osmosis: Investigation of the slower flux decline phenomenon, *Water Sci. Technol.*,  
948 61 (2010) 927-936.
- 949 [30] Y. Jang, H. Cho, Y. Shin, Y. Choi, S. Lee, J. Koo, Comparison of fouling propensity and  
950 physical cleaning effect in forward osmosis, reverse osmosis, and membrane distillation,  
951 *Desalination and Water Treatment*, 57 (2016) 24532-24541.
- 952 [31] E.W. Tow, J.H. Lienhard V, Quantifying osmotic membrane fouling to enable  
953 comparisons across diverse processes, *J. Membr. Sci.*, 511 (2016) 92-107.
- 954 [32] A.G. Fane, T.H. Chong, J. Zhang, W.C.L. Lay, The Effect of Flux and Pressure on Fouling  
955 in Reverse Osmosis Desalination, *Proceedings IDA World Congress*, (2009) Paper DB09-128.
- 956 [33] R.W. Field, G.K. Pearce, Critical, sustainable and threshold fluxes for membrane filtration  
957 with water industry applications, *Adv. Colloid Interface Sci.*, 164 (2011) 38-44.
- 958 [34] Z. Xie, N. Nagaraja, L. Skillman, D. Li, G. Ho, Comparison of polysaccharide fouling in  
959 forward osmosis and reverse osmosis separations, *Desalination*, 402 (2017) 174-184.
- 960 [35] J.R. McCutcheon, M. Elimelech, Influence of concentrative and dilutive internal  
961 concentration polarization on flux behavior in forward osmosis, *J. Membr. Sci.*, 284 (2006)  
962 237-247.
- 963 [36] C.Y. Tang, Q. She, W.C.L. Lay, R. Wang, A.G. Fane, Coupled effects of internal  
964 concentration polarization and fouling on flux behavior of forward osmosis membranes during  
965 humic acid filtration, *J. Membr. Sci.*, 354 (2010) 123-133.
- 966 [37] Q. She, R. Wang, A.G. Fane, C.Y. Tang, Membrane fouling in osmotically driven  
967 membrane processes: A review, *J. Membr. Sci.*, 499 (2016) 201-233.
- 968 [38] E.M.V. Hoek, M. Elimelech, Cake-Enhanced Concentration Polarization: A New Fouling  
969 Mechanism for Salt-Rejecting Membranes, *Environ. Sci. Technol.*, 37 (2003) 5581-5588.
- 970 [39] T.H. Chong, F.S. Wong, A.G. Fane, Implications of critical flux and cake enhanced  
971 osmotic pressure (CEOP) on colloidal fouling in reverse osmosis: Experimental observations,  
972 *J. Membr. Sci.*, 314 (2008) 101-111.
- 973 [40] C.Y. Tang, Y.N. Kwon, J.O. Leckie, Characterization of humic acid fouled reverse  
974 osmosis and nanofiltration membranes by transmission electron microscopy and streaming  
975 potential measurements, *Environ. Sci. Technol.*, 41 (2007) 942-949.

976 [41] A. Tiraferri, N.Y. Yip, A.P. Straub, S. Romero-Vargas Castrillon, M. Elimelech, A method  
977 for the simultaneous determination of transport and structural parameters of forward osmosis  
978 membranes, *J. Membr. Sci.*, 444 (2013) 523-538.

979 [42] A.J. Karabelas, D.C. Sioutopoulos, Toward improvement of methods for predicting  
980 fouling of desalination membranes — The effect of permeate flux on specific fouling resistance,  
981 *Desalination*, 343 (2014) 97-105.

982 [43] Q. She, Y.K.W. Wong, S. Zhao, C.Y. Tang, Organic fouling in pressure retarded osmosis:  
983 Experiments, mechanisms and implications, *J. Membr. Sci.*, 428 (2013) 181-189.

984 [44] Q. She, X. Jin, Q. Li, C.Y. Tang, Relating reverse and forward solute diffusion to  
985 membrane fouling in osmotically driven membrane processes, *Water Res.*, 46 (2012) 2478-  
986 2486.

987 [45] Q. She, D. Hou, J. Liu, K.H. Tan, C.Y. Tang, Effect of feed spacer induced membrane  
988 deformation on the performance of pressure retarded osmosis (PRO): Implications for PRO  
989 process operation, *J. Membr. Sci.*, 445 (2013) 170-182.

990 [46] E.M.V. Hoek, A.S. Kim, M. Elimelech, Influence of crossflow membrane filter geometry  
991 and shear rate on colloidal fouling in reverse osmosis and nanofiltration separations,  
992 *Environmental Engineering Science*, 19 (2002) 357-372.

993 [47] C.Y. Tang, T.H. Chong, A.G. Fane, Colloidal interactions and fouling of NF and RO  
994 membranes: A review, *Adv. Colloid Interface Sci.*, 164 (2011) 126-143.

995 [48] R.W. Field, J.J. Wu, Mass transfer limitations in forward osmosis: Are some potential  
996 applications overhyped?, *Desalination*, 318 (2013) 118-124.

997 [49] T.Y. Cath, M. Elimelech, J.R. McCutcheon, R.L. McGinnis, A. Achilli, D. Anastasio, A.R.  
998 Brady, A.E. Childress, I.V. Farr, N.T. Hancock, J. Lampi, L.D. Nghiem, M. Xie, N.Y. Yip,  
999 Standard Methodology for Evaluating Membrane Performance in Osmotically Driven  
1000 Membrane Processes, *Desalination*, (2012).

1001 [50] B. Kim, G. Gwak, S. Hong, Review on methodology for determining forward osmosis  
1002 (FO) membrane characteristics: Water permeability (A), solute permeability (B), and structural  
1003 parameter (S), *Desalination*, 422 (2017) 5-16.

1004 [51] B. Mi, M. Elimelech, Gypsum scaling and cleaning in forward osmosis: Measurements  
1005 and mechanisms, *Environ. Sci. Technol.*, 44 (2010) 2022-2028.

1006 [52] L. Song, M. Elimelech, Particle Deposition onto a Permeable Surface in Laminar Flow, *J.*  
1007 *Colloid Interface Sci.*, 173 (1995) 165-180.

1008 [53] L.N. Sim, Y. Ye, V. Chen, A.G. Fane, Investigations of the coupled effect of cake-  
1009 enhanced osmotic pressure and colloidal fouling in RO using crossflow sampler-modified  
1010 fouling index ultrafiltration, *Desalination*, 273 (2011) 184-196.

1011 [54] C.Y. Tang, J.O. Leckie, Membrane independent limiting flux for RO and NF membranes  
1012 fouled by humic acid, *Environ. Sci. Technol.*, 41 (2007) 4767-4773.

1013 [55] E.W. Tow, J.H. Lienhard V, Unpacking compaction: Effect of hydraulic pressure on  
1014 alginate fouling, *J. Membr. Sci.*, 544 (2017) 221-233.

1015 [56] A. Rushton, A.S. Ward, R.G. Holdich, *Solid-Liquid Filtration and Separation Technology*,  
1016 Wiley-VCH, 1996.

1017 [57] R. Guha, X. Shang, A.L. Zydney, D. Velegol, M. Kumar, Diffusiophoresis contributes  
1018 significantly to colloidal fouling in low salinity reverse osmosis systems, *J. Membr. Sci.*, 479  
1019 (2015) 67-76.

1020 [58] M. Zhang, D. Hou, Q. She, C.Y. Tang, Gypsum scaling in pressure retarded osmosis:  
1021 Experiments, mechanisms and implications, *Water Res.*, 48 (2014) 387-395.

1022 [59] M. Zhang, Q. She, X. Yan, C.Y. Tang, Effect of reverse solute diffusion on scaling in  
1023 forward osmosis: A new control strategy by tailoring draw solution chemistry, *Desalination*,  
1024 401 (2017) 230-237.

- 1025 [60] P. van den Brink, A. Zwijnenburg, G. Smith, H. Temmink, M. van Loosdrecht, Effect of  
1026 free calcium concentration and ionic strength on alginate fouling in cross-flow membrane  
1027 filtration, *J. Membr. Sci.*, 345 (2009) 207-216.
- 1028 [61] S. Meng, Y. Liu, Transparent exopolymer particles (TEP)-associated membrane fouling  
1029 at different Na<sup>+</sup> concentrations, *Water Res.*, 111 (2017) 52-58.
- 1030 [62] J. BAUMY, J., G. BRULE, Effect of pH and ionic strength on the binding of bivalent  
1031 cations to  $\beta$ -casein, *Lait*, 68 (1988) 409-417.
- 1032 [63] K.J. Thomas, C.V. Rice, Revised model of calcium and magnesium binding to the  
1033 bacterial cell wall, *BioMetals*, 27 (2014) 1361-1370.
- 1034 [64] R.W. Field, J.J. Wu, Modelling of permeability loss in membrane filtration: Re-  
1035 examination of fundamental fouling equations and their link to critical flux, *Desalination*, 283  
1036 (2011) 68-74.  
1037