1 **Performance of a seawater-driven forward osmosis process for pre-**
2 **concentrating digested sludge centrate: organic enrichment and membrane**
3 **fouling**

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13 **Abstract:**

14 This study demonstrated the potential of seawater-driven forward osmosis for enriching organic
15 matter in digested sludge centrate. The results indicated that the cellulose triacetate membrane
16 offered better performance than the polyamide membrane in terms of organic materials
17 enrichment, fouling resistance and membrane cleaning efficiency. Membrane fouling decreased
18 the enrichment efficiency of organic matter since the deposition of suspended particulate matter
19 on the membrane surface caused fouling and loss of organic matter from the concentrated
20 sludge centrate. The results showed that increasing the draw solution concentration increased
21 flux but did not aggravate membrane fouling, however, it could reduce the efficiency of
22 physical flushing to recover the flux. Seawater showed comparable forward osmosis
23 performance to that of analytical grade NaCl as draw solutes in terms of flux and organic
24 enrichment. The results also showed that seawater as the draw solution resulted in more
25 membrane fouling and lower flux recovery compared to NaCl.

26 **Keywords:** Forward osmosis (FO); cellulose triacetate (CTA); polyamide (PA); fouling
27 behaviour; digested sludge centrate; organic matter; seawater.

Water Impact Statement:

29 Sludge centrate is a small waste stream during wastewater treatment but with a high content of
30 dissolved organic carbon and nutrients. Results from this study highlight the potential of a
31 seawater driven forward osmosis process to enrich the organic content in sludge centrate as
32 well as as key challenges for practical implementation. By enriching the organic and nutrient
33 content in sludge centrate using forward osmosis, it is possible to simultaneously reduce
34 contaminant loading and create opportunities for resource recovery.

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1. Introduction

Forward osmosis (FO) is a robust separation platform capable of treating highly complex solutions that are not suitable for conventional membrane processes [1, 2]. In FO, mass transfer through the membrane is osmotically driven. Thus, when a draw solution (DS) is readily available, the FO process can occur with very low energy input [1, 3-5]. The absence of external hydraulic pressure can also explain the low membrane fouling tendency and excellent fouling reversibility of FO. In recent years, the potential of FO to treat many complex feed solutions has been demonstrated in the literature. These complex solutions include drilling and fracking fluids from oil and gas exploration [6-9], sludge [10, 11], digested sludge centrate [2, 12, 13], and municipal wastewater [4, 14, 15].

A major obstacle to full scale deployment of FO is the lack of a suitable DS [1]. Issues associated with cost of the draw solutes, regeneration, and loss of draw solutes due to reverse diffusion can increase the operating cost, thereby hindering the feasibility of FO applications [1, 16, 17]. In this context, seawater, which is abundant and cheaply available in coastal areas, has been increasingly considered as a potential DS [1, 16]. The diluted seawater released from the process can be returned to the sea, and thus, DS regeneration is not necessary.

In a typical wastewater treatment plant, the sludge is anaerobically digested. The digested sludge is then dewatered to obtain biosolids for land application. The liquid from this dewatering process is called sludge centrate, which has a high content of suspended solids, nutrients, and organic matter [13]. Due to the difficulties associated with the treatment of this sludge centrate, in most cases, it is returned to the headworks of the treatment plant. The recirculation of untreated sludge centrate to the headworks leads to additional organic and nutrient loading, and deprive the plant from any opportunities for energy and nutrient recovery [12, 18].

The use of FO to pre-concentrate sludge centrate has been investigated in several recent studies [2, 13]. However, draw solutes such as $MgCl_2$ and NaCl are expensive and must be regenerated. On the other hand, seawater appears to be a particularly promising DS for pre-concentrating sludge centrate. Ansari et al. [12] has recently demonstrated a seawater-driven FO process for phosphorus recovery from digested sludge centrate with a specific focus on evaluating the efficiency of nutrient recovery. Results from previous studies suggest that identifying the most suitable membrane type and orientation is necessary to ensure the best performance of seawater-driven FO process for pre-concentrating sludge centrate [2, 10-12]. More importantly, understanding of the fouling process and developing strategies to control fouling need to be discussed to guarantee the long-term operation of the FO system. It is also necessary to consider all other factors affecting water flux and membrane fouling, such as membrane pre-wetting and draw solution.

This study aims to elucidate the effects of membrane materials, prewetting procedures and DS on the performance of seawater-driven forward osmosis for pre-concentrating organic matter

74 in the sludge centrate. The performance of the FO process is observed in terms of chemical
75 oxygen demand (COD) enrichment, membrane fouling and flux recovery by physical cleaning.
76 Comparison between seawater and analytical grade NaCl as the DS was made to highlight the
77 potential and challenges of using seawater for enriching COD in sludge centrate.

78 2. Materials and methods

79 2.1. Forward osmosis system and membranes

80 A lab-scale cross-flow FO system (Figure 1) was used. The FO system included a membrane
81 cell, two variable speed gear pumps (Micropump, Vancouver, Washington, USA), conductivity
82 and temperature controllers, and a digital balance to measure the flux. The FO membrane cell
83 consisted of two symmetric rectangular chambers for the feed and draw solutions, respectively.
84 The internal dimensions of each chamber were 10 cm in length, 5 cm in width and 0.2 cm in
85 height. The system was operated in the counter-current mode. The FO membranes were
86 positioned either in active layer facing feed solution (AL-FS) orientation, or active layer facing
87 draw solution (AL-DS) orientation.

88 Flat sheet FO membranes were obtained from Hydration Technology Innovations (HTI,
89 Albany, OR) and Porifera, Inc. (Hayward, California, USA). The HTI membrane had an
90 asymmetric structure and was made of cellulose triacetate (CTA) with an embedded polyester
91 mesh for mechanical support. The Porifera membrane was a thin film composite (TFC)
92 membrane consisting of a thin polyamide (PA) layer on a microporous polysulfone supporting
93 layer. Key properties of the HTI and Porifera membranes were summarised in Table 1.

94 **Table 1:** Key properties of the active layer of the FO membranes [19, 20].

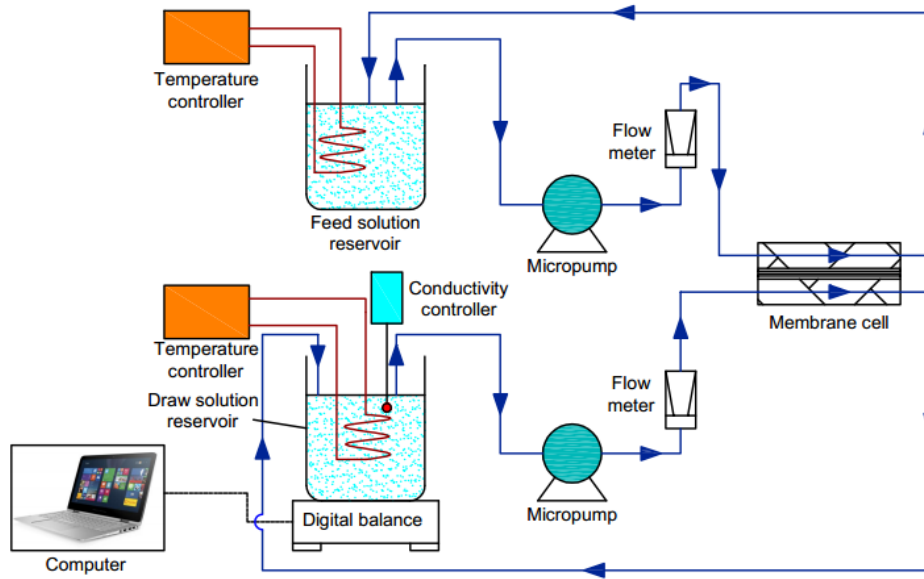| Membrane	HTI-CTA	Porifera-PA
Water permeability (A) (L/m^2.h.bar)	0.84	3.2
Salt (NaCl) permeability (B) (L/m^2.h)	0.32	0.41
Structural parameter (S) (mm)	0.57	0.46
Contact angle (^{o})	61	49.5
Surface roughness (nm)	3.8	57.4
Zeta potential at pH = 7 (mV)	-5	-16

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Figure 1: Schematic diagram of the lab-scale FO system.

98 **2.2. Feed solution and draw solution**

In this study, seawater and NaCl solutions were used as DSs. Seawater was collected from North Wollongong Beach (Wollongong, New South Wales, Australia) and was filtered using filter paper with a pore size of 1 μm prior to the experiments. NaCl solutions of 0.25, 0.5, 0.75 and 1 M were prepared using analytical grade NaCl and deionised (DI) water. Anaerobically digested sludge centrate was collected from a high-speed centrifuge at the Shellharbour Wastewater Treatment Plant (Shellharbour, New South Wales, Australia). The centrate was pre-filtered using a 0.2 mm plastic screen to remove any large objects. The compositions of seawater and digested sludge centrate were summarised in Table 2.

Table 2. Composition of seawater and digested sludge centrate (values indicated average ± standard deviation of at least three samples).

Parameters	Unit	Seawater	Digested sludge centrate
pH	-	7.3 ± 0.2	7.1 ± 0.1
Electrical conductivity	mS/cm	44.2 ± 0.3	6.8 ± 0.1
Osmotic pressure	bar	28.1 ± 0.6	6.5 ± 0.1
Total solids	g/L	31.7 ± 2.8	1.6 ± 0.1
COD	mg/L	-	420.3 ± 15.5
Ammonia	mg/L	-	520 ± 2.6
Total phosphorus (as PO_4^{3-})	mg/L	-	371.5 ± 1.4

2.3. Membrane prewetting

Prewetting was conducted by soaking in a solution containing 70% ethanol and 30% water for 45 min. Following soaking, the membrane was rinsed and preserved in DI water overnight prior to filtration experiments.

2.4. Experimental protocols

All FO experiments were conducted in four steps. In the first step, DI water was used as the feed solution (FS) for 30 min to determine the pure water flux. In the second step, DI water was substituted with sludge centrate and the FO experiment was conducted until a water recovery of 55% had been achieved. Duration of this second step varied from experiment to experiment ranging from 24 to 120 hours. At specified time intervals, 10 mL samples were collected from the feed for analysis. In the third step, the draw and feed solutions were replaced by DI water to facilitate membrane cleaning by flushing at a cross-flow velocity of 24 cm/s for 20 min. In the last step, the pure water flux was determined again using DI water under identical experimental conditions as in the first step. In all experiments, initial volumes of the feed and draw solutions were 1 and 3 L, respectively. The circulation flow rate of the feed and the draw solution was 0.8 L/min (i.e., cross flow velocity of 13 cm/s).

2.5. Calculations

Water flux (J_w) was calculated based on the change in weight of DS, and expressed as in Eq. (1):

$$J_w = \frac{m_{t_i} - m_{t_{i-1}}}{\left(t_i - t_{i-1}\right) \times \rho \times A_m} = \frac{\Delta m_i}{\Delta t_i \times \rho \times A_m} \qquad \text{(Eq. 1)}$$

Where: Δm_i: the change in weight of DS over a time interval (g)

Δt_i: a time interval (h)

ρ: the solution density (g/cm^3)

A_m: the effective membrane area (m^2)

Water recovery was determined based on the ratio of the cumulative permeate volume and the initial volume of FS, and presented as in Eq. (2):

$$\text{Water recovery (\%)} = \frac{\int_0^t A_m \times J_w \times dt}{V_{initial}} \times 100\% \qquad \text{(Eq. 2)}$$

Where: J_w: the observed water flux at time t

$V_{initial}$: the initial volume of FS

140 The draw solute flux (J_s) was calculated by Eq. (3):

141
$$J_s = \frac{C_{f,t_2} \times V_{f2} - C_{f,t_1} \times V_{f1}}{\Delta t \times A_m}$$
(Eq. 3)

142 Where: $C_{f,t2}$: The concentration of draw solute in FS at time t_2 (g/L)

143 V_{f2}: The volume of FS at time t_2 (L)

144 $C_{f,t1}$: The concentration of draw solute in FS at time t_1 (g/L)

145 V_{f1}: The volume of FS at time t_1 (L)

146 Δt: a certain period of filtration time (h)

147 **2.6. Analytical methods**

148 Key water quality parameters of digested sludge centrate and seawater were measured
149 according to standard methods. COD was determined using a Hach DRB200 COD Reactor and
150 Hach DR3900 spectrophotometer following the US-EPA Standard Method 5220. Temperature
151 and pH of solutions were measured by an Orion 4-Star Plus pH/conductivity meter (Thermo
152 Scientific, Waltham, MA).

153 The surface characteristics of the FO membranes were characterized using scanning electron
154 microscopy (SEM) (JOEL, JSM-6400LV, Japan). Prior to taking SEM images, coupon
155 membrane samples were coated with a thin layer of gold.

156 **3. Results and discussions**

157 **3.1. Pure water flux under different conditions**

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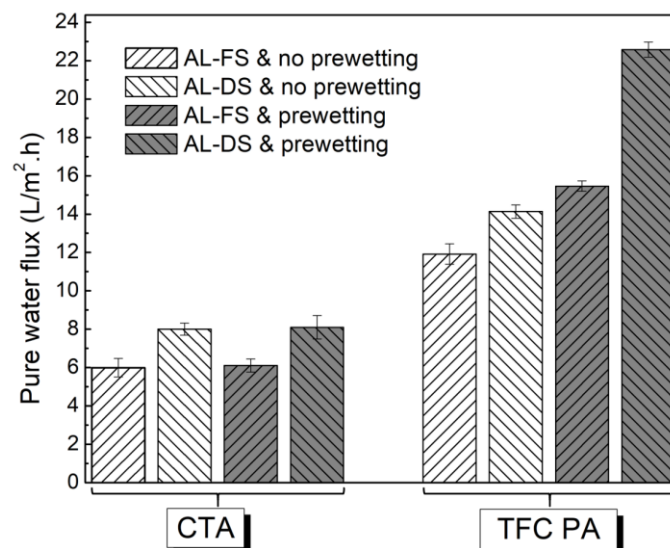
Figure 2: Pure water fluxes of CTA and PA FO membranes using seawater as DS in AL-FS and AL-DS orientations with and without prewetting.

Under the same experimental conditions, the TFC PA membrane showed a higher pure water flux than that of the CTA membrane (Figure 2). This observation could be explained by the A value (water permeability under a hydraulic pressure) and the structure parameter (S value) of these two membranes (Table 1). Indeed, the A value of the TFC PA membrane was approximately 4 times higher than that of the CTA membrane. The structural parameter value of the TFC PA membrane (460 μm) was slightly lower than that of the CTA membrane (570 μm) [19]. Previous studies have demonstrated that a smaller structural parameter results in less severe internal concentration polarization (ICP) and, thus, higher water flux[1, 19]. It is noteworthy that in the FO process, concentration polarisation could also influence the water flux. Thus, the difference in pure water flux between the TFC PA and CTA membranes was not necessarily proportional to the difference in their A and S values.

The AL-DS orientation exhibited a higher pure water flux compared to the AL-FS orientation. The difference in water flux between these two orientations was considerably less than the comparison between the TFC PA and CTA membranes discussed above. Indeed, the higher water flux under the AL-DS orientation compared to the AL-FS orientation could be solely attributed to a less severe ICP condition [1, 21, 22].

Prewetting significantly improved the pure water flux of the TFC PA membrane but had a negligible impact on the CTA membrane. As a result of prewetting, water flux of the PA membrane increased by 29.7% and 59.7% under the AL-FS and AL-DS orientation, respectively (Figure 2). There were two possible reasons for this notable increase in flux by the TFC PA membrane after prewetting, namely swelling of the PA skin layer and prewetting of the polysulfone supporting layer. The PA skin layer could swell in alcohol causing an increase in the effective pore size and thus, increased the water flux [20]. However, there was no discernible increase in the reverse salt flux due to prewetting (data not shown). Thus, swelling was not expected to be a major reason for the improvement in water flux observed here. The polysulfone supporting layer was hydrophobic and could not be completely wetted by water [23]. Compared to water, ethanol had a lower surface tension, thus, could easily penetrate into the porous structure and prewet the pores of the polysulfone supporting layer for subsequent water permeation [23, 24]. Conversely, CTA membrane was asymmetric and CTA material was readily hydrophilic. Thus, prewetting did not affect its pure water flux (Figure 2) [25].
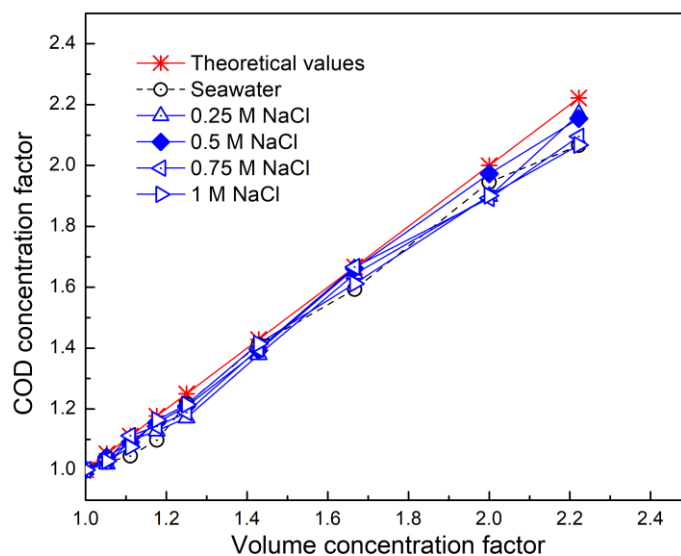
3.2. Pre-concentration of digested sludge centrate by FO
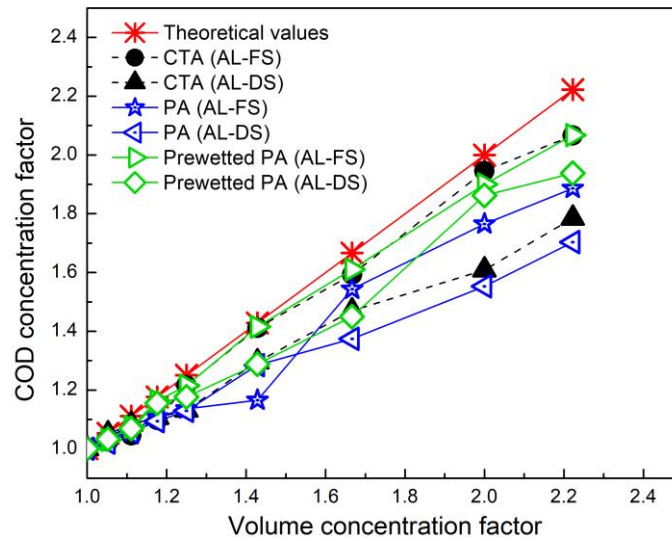
Figure 3: The performance of FO for enrichment of COD in digested sludge centrate using seawater as the DS. The theoretical COD concentration factor was calculated assuming 100% COD retention by the FO membrane.

As the sludge centrate was concentrated by FO, COD concentration increased proportionally (Figure 3). In all cases, the COD concentration factor was lower than the theoretical value assuming complete COD retention. The observed difference between the COD concentration factor and theoretical value could possibly be attributed to the deposition of particulate COD materials on the membrane surface. In fact, there was a correlation between membrane fouling and the COD enrichment results in Figure 3 as discussed further in section 3.3.

The best performance in terms of COD enrichment was from the CTA membrane with the AL-FS orientation (Figure 3). The active layer of the CTA membrane had a lower surface roughness than its own supporting layer as well as that of the TFC PA membrane [20, 26-28]. Thus, due to the hydrodynamic drag force from the cross flow, the deposition of organic substances on the CTA membrane was expected to be less compared to the TFC PA membrane. In addition, surface chemistry interaction between organic matter and the membrane surface could be a reason for the lower organic enrichment when using the PA membrane. The PA membrane has a considerable number of highly polar carboxylic functional groups on its surface and has significant affinity towards organic colloids in the FS [20]. As a result, the accumulation of organic matter on the PA membrane surface could be enhanced, thus reducing COD concentration in the bulk feed. It is noted that this combination (CTA under AL-FS orientation) also had the lowest initial water flux. However, it appears that water flux did not affect COD enrichment performance as demonstrated below.

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Figure 4: The performance of FO for enrichment of COD in digested sludge centrate using the NaCl solution as the DS and the CTA membrane in the AL-FS orientation. Note: The theoretical COD concentration factor was calculated assuming 100% COD retention by the FO membrane.

DS concentration did not affect the enrichment of COD by FO (Figure 4). No significant difference in COD enrichment was observed when the NaCl DS concentration increased from 0.25 to 1 M (Figure 4). The initial water flux was proportional to the DS concentration. Thus, results in Figure 4 also suggested that water flux did not affect COD enrichment as discussed above.

Seawater showed comparable COD enrichment performance to that of analytical grade NaCl as the draw solutes. The osmotic potential of seawater was similar to that of the 0.5 M NaCl solution [29, 30]. Seawater was readily available in coastal areas and thus it was a low-cost DS. However, in addition to NaCl, seawater contained many other salts. Some of them were sparingly soluble and might cause membrane scaling as further discussed in section 3.3.2.

3.3. Factors affecting fouling behaviour

3.3.1. Membrane properties and orientations
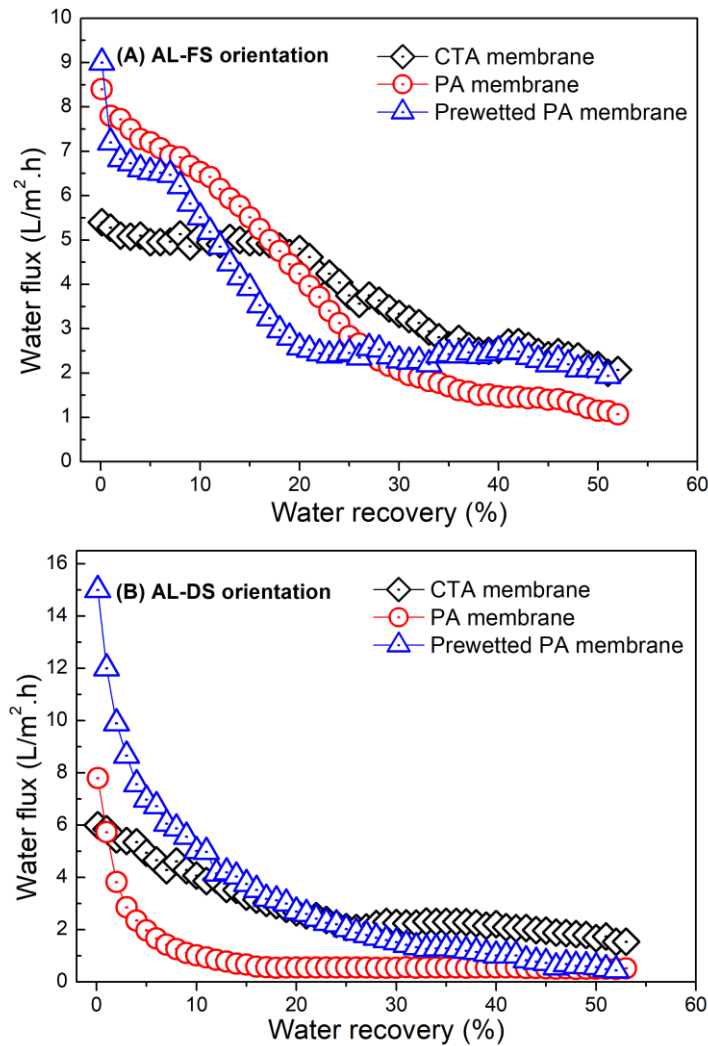
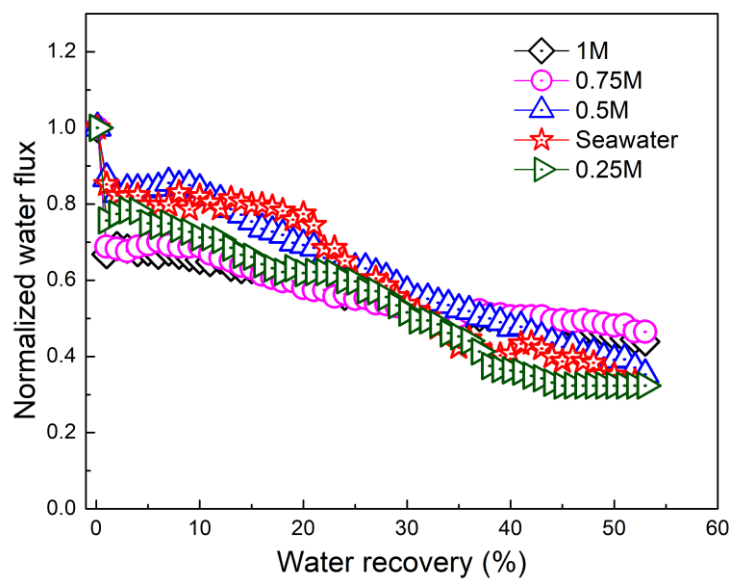**Figure 5:** Fouling behaviour of the FO membranes in (A) AL-FS orientation and (B) AL-DS orientation using seawater as DS.

In all cases, membrane fouling was significant as indicated by significant water flux decline during COD enrichment (Figure 5). The decrease in water flux could be mostly attributed to the formation of a cake layer on the membrane surface. This cake layer caused an additional hydraulic resistance and increased concentration polarisation, thus reducing water flux.

In good agreement to the data in Figure 3, the CTA membrane was less susceptible to fouling than the TFC PA membrane regardless of the membrane orientation. As described above, this result was likely due to the smooth surface of the CTA membrane. The higher roughness and prominent ridge-and-valley structure of the PA membrane surface could exacerbate the deposition of foulants, thus more severe fouling [20]. Additionally, a high density of carboxylic functional groups in the structure of the PA membrane could be potentially vulnerable to fouling

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245 [31]. In the presence of Ca^{2+} ions, carboxyl acid functional groups could lead to bridging of
246 membrane surface and Ca^{2+}-organic foulants, thus probably exacerbating organic fouling.

247 The AL-FS orientation showed less fouling than the AL-DS orientation. There were several
248 possible explanations. As mentioned above, the lower roughness of the active layer in the AL-
249 FS orientation could alleviate the accumulation of foulants on the membrane surface. FO
250 operation under the AL-DS orientation was susceptible to internal clogging since organic
251 molecules could readily penetrate the porous supporting layer. In addition, the high reverse
252 solute diffusion in the AL-DS orientation could increase the osmotic potential of the feed,
253 decreased effective driving force, and thus, reduced the water flux [21].

254 *3.3.2. Draw solution*


256 **Figure 6:** Comparison of fouling behaviour towards CTA membranes in the AL-FS
257 orientation using seawater and the NaCl solutions at different concentration as DSs.

258 As discussed in section 3.2, the CTA membrane under the AL-FS orientation demonstrated the
259 best suitability for enriching organic matter in centrate. Thus, the CTA membrane under the
260 AL-FS orientation was used for further investigation. Increasing the DS concentration led to a
261 higher initial water flux (Figure 9), but no significant impacts on membrane fouling (Figure 6).
262 The elevated initial water flux was a result of the increased driving force of water transport due
263 to an increase in the concentration gradient along the membrane. However, the extent of fouling
264 in all cases was nearly the same. This was probably because of the smoothness of the CTA
265 membrane surface that could effectively minimize the accumulation and deposition of foulants.
266 In addition, this could be explained using the theory of 'critical DS concentration'. According
267 to this concept, fouling could be less severe at below the critical DS concentration [32, 33]. It is
268 possible that the used DS concentrations in this study were lower than the critical DS
269 concentration, thus, the impacts of DS concentration on fouling were insignificant.

270 Seawater exhibited similar fouling to that from the 0.5 M NaCl solution as the DSs. This
271 observation was consistent with the data shown in Figure 4, and thus could also be explained
272 by the same reason as referred to earlier. However, it is noted that the flux profile when using
273 seawater was less stable than that of using the 0.5 M NaCl solution. Multivalent ions in
274 seawater, such as Ca^{2+}, Mg^{2+}, SO_4^{2-} and PO_4^{3-} could act as fouling promoters, and scaling
275 precursors through the reverse solute diffusion, thus increasing fouling potential.

276 *3.3.3. Fouling layer characteristics*
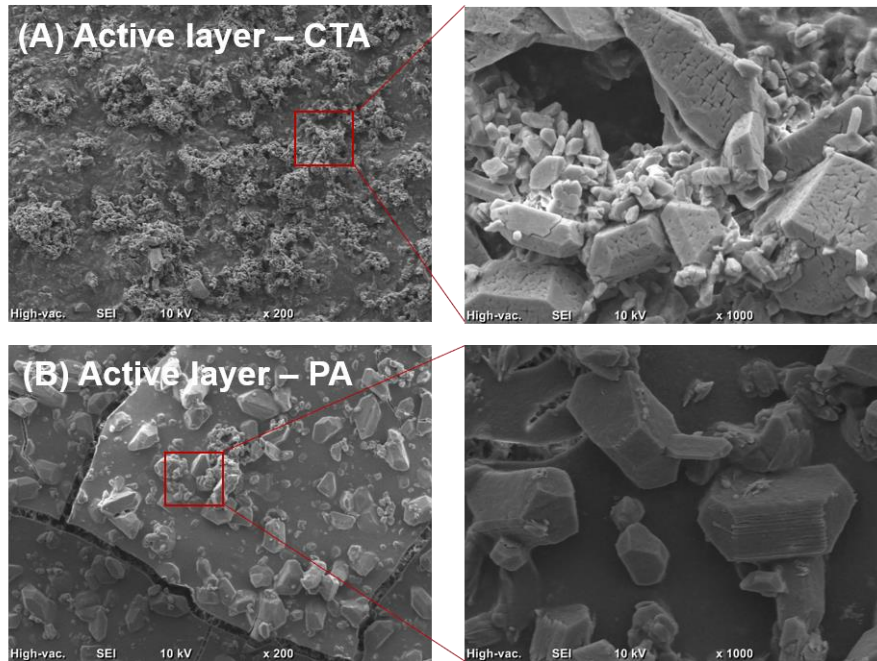
278 **Figure 7:** SEM images of the active layer of (A) the fouled CTA membrane and (B) the
279 fouled TFC PA membrane in the AL-FS orientation using seawater DS.

280 A notable contrast in the morphology of the fouling layer on the CTA and TFC PA membranes
281 could be observed (Figure 7). The fouling layer on the active layer surface of the CTA
282 membrane was loose and soft (Figure 7A) while that of the PA membrane was dense and firm
283 (Figure 7B). The observed irregular shape and size of crystals and organic cake-layer on the
284 membrane surface suggested the presence of both organic and inorganic foulants.

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3.4. Cleaning and flux reversibility
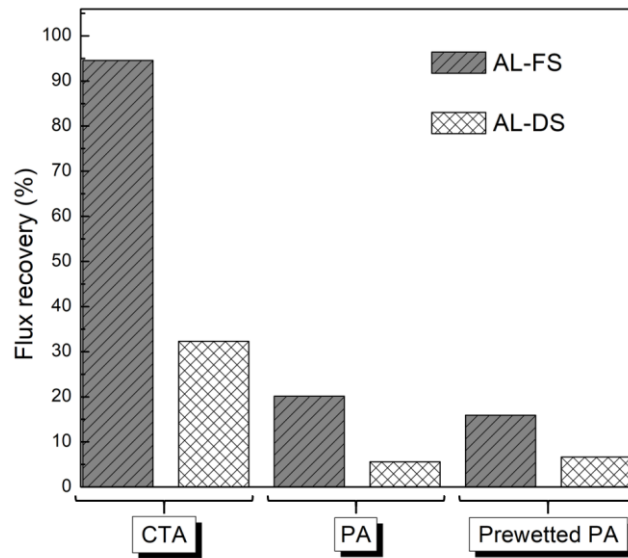
Figure 8: Flux recovery of the fouled CTA, PA and prewetted PA membranes using seawater DS after physical flushing.

As expected, flux recovery by flushing in the AL-FS orientation was higher than that in the AL-DS orientation (Figure 8). Flux recovery was proportional to the efficiency of foulant removal from the membrane surface. In the AL-FS orientation, the deposition of foulants on the membrane was a surface phenomenon and the fouling cake could be readily removed by shear force from flushing. On the other hand, in the AL-DS orientation, the feed solution was in contact with the supporting layer and due to pore clogging of the supporting layer, flux recoverability was much lower than in the AL-FS orientation (Figure 8).

The highest flux recovery (95%) was observed with the CTA membrane in the AL-FS orientation (Figure 8). Together with the high COD enrichment performance shown in Figure 3, this result suggested that the CTA membrane in the AL-FS orientation was the most suitable for sludge centrate. This result was also consistent with the loose and soft fouling layer of the CTA membrane under the AL-FS orientation previously shown in Figure 7A.

Flushing was not efficient in restoring the water flux of the PA and prewetted PA membranes. There were two possible reasons. Firstly, foulants deposited on a rough surface of the PA membrane could be sheltered from cross-flow shear force, thus decreasing the number of foulants detached from the membrane surface. Secondly, the highly polar carboxylic functional groups on the PA membrane surface were available for ionic bonding with foulants, thus improving foulant adhesion [20]. In contrast, the CTA membrane was only slightly negatively charged and did not have free carboxyl functional groups that could interact with the foulants [20].
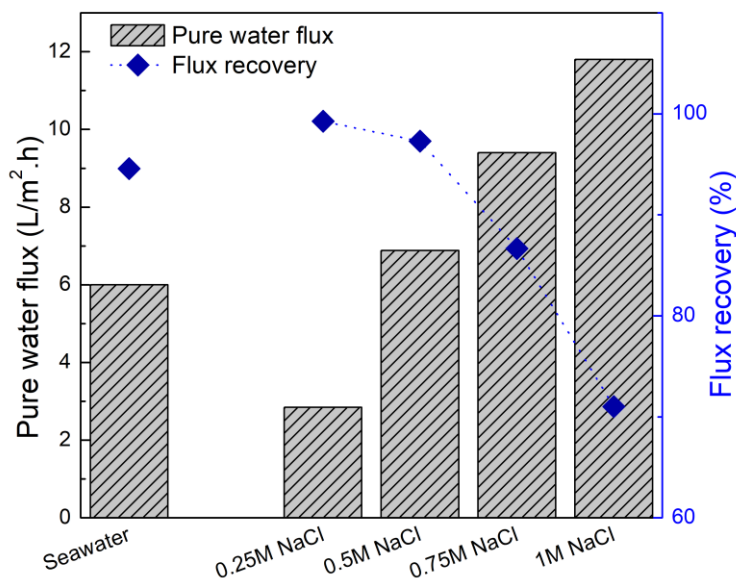
Figure 9: Comparison of pure water fluxes and flux recovery of CTA membranes in the AL-FS orientation using seawater and NaCl DS.

Increasing the DS concentration resulted in a higher pure water flux but also reduced flux recovery by flushing (Figure 9). Indeed, as the NaCl concentration increased from 0.25 to 1 M, flux recovery decreased from nearly 100% to 70%. Since the DS concentration was proportional to water flux, results in Figure 9 showed that the extent of irreversible fouling (by flushing) was inversely correlated to the initial flux. A denser and more compact fouling was formed at higher initial flux, thereby, impairing the efficiency of flushing.

Seawater had a similar osmotic potential to a 0.5 M NaCl solution. However, a slightly lower flux recovery was observed when using seawater as the DS compared to the 0.5 M NaCl solution. The complex composition of seawater could result in a less reversible fouling layer. Indeed, multivalent cations (such as Ca^{2+} and Mg^{2+}) in seawater could exacerbate fouling and render fouling layer more adhesive, thus lowering the flux recoverability by flushing.

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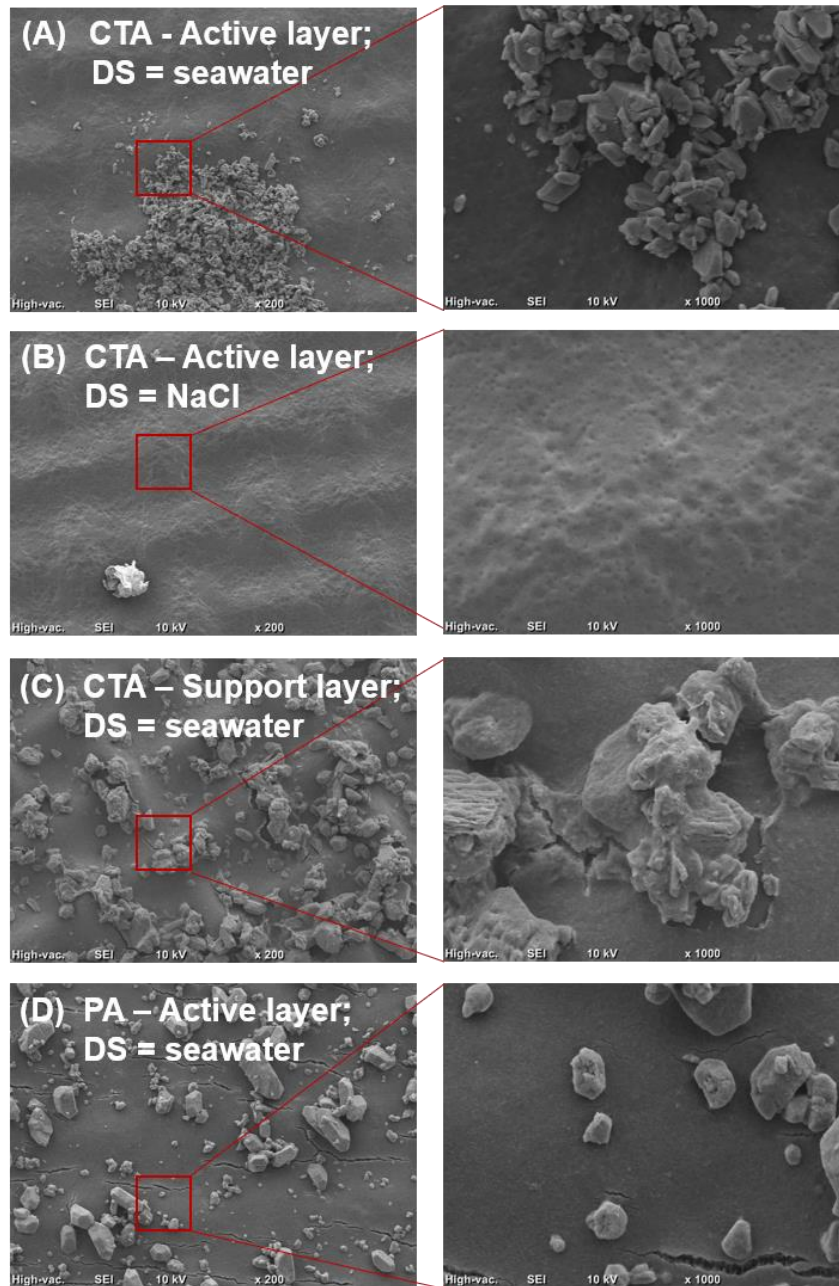
Figure 10: SEM images of (A) the active layer of the cleaned CTA membrane in the AL-FS orientation using seawater as DS, (B) the active layer of the cleaned CTA membrane in the AL-FS orientation using the 0.5 M NaCl solution as DS, (C) the supporting layer of the cleaned CTA membrane in the AL-DS orientation using seawater as DS and (D) the active layer of the cleaned PA membrane in the AL-FS orientation using seawater DS.

The complex composition of seawater and the membrane characteristics were observed to govern the cleaning efficiency of flushing (Figure 10). After flushing, the CTA membrane active surface in the AL-FS orientation (seawater as DS) still had some salt crystals and organic particles (Figure 10A). In contrast, when using the 0.5 M NaCl solution, the membrane was mostly clean after flushing (Figure 10B). This was consistent with the data in Figure 9 that indicated how the complex composition of seawater resulted in a more adhesive fouling layer.

In addition, SEM images indicated that both for CTA membrane under the AL-DS orientation (Figure 10C) and PA membrane in the AL-FS orientation (Figure 10D) flushing removed crystals significantly, but the organic layers only partly. These observations resulted from synergistic effects of membrane roughness and chemical structure of these membranes on fouling as discussed above.

4. Conclusions

Results from this study demonstrated the potential of the FO process to enrich COD in sludge centrate. Compared to the TFC PA membrane, the CTA membrane in the AL-FS orientation showed a much better COD enrichment efficiency, lower fouling, and higher flux recovery by simple flushing. There was a correlation between membrane fouling and COD enrichment efficiency. In other words, COD enrichment efficiency decreased when organic matter accumulated on the membrane surface, causing fouling. The results also showed that membrane fouling was not affected by the DS concentration (or initial water flux) possibly because of the low initial flux in this study, however, flux recovery by membrane flushing decreased as the initial water flux increased. Seawater is a potentially low-cost and effective DS for COD enrichment. However, compared to NaCl, seawater as the DS led to more severe membrane fouling and lower flux recovery by flushing. Further research is necessary to address the issue of membrane fouling when using seawater as the DS for COD enrichment in sludge centrate.

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