



Citation for published version:

Liu, S, Song, W, Meng, M, Xie, M, She, Q, Zhao, P & Wang, X 2022, 'Engineering pressure retarded osmosis membrane bioreactor (PRO-MBR) for simultaneous water and energy recovery from municipal wastewater', *Science of the Total Environment*, vol. 826, 154048. <https://doi.org/10.1016/j.scitotenv.2022.154048>

DOI:

[10.1016/j.scitotenv.2022.154048](https://doi.org/10.1016/j.scitotenv.2022.154048)

Publication date:

2022

Document Version

Peer reviewed version

[Link to publication](#)

Publisher Rights

CC BY-NC-ND

University of Bath

Alternative formats

If you require this document in an alternative format, please contact:
openaccess@bath.ac.uk

General rights

Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

Take down policy

If you believe that this document breaches copyright please contact us providing details, and we will remove access to the work immediately and investigate your claim.

1 **Engineering pressure retarded osmosis membrane bioreactor (PRO-**
2 **MBR) for simultaneous water and energy recovery from municipal**
3 **wastewater**

4
5 Shuyue Liu^a, Weilong Song^{a,*}, Manli Meng^a, Ming Xie^b, Qianhong She^c, Pin Zhao^a,
6 Xinhua Wang^a

7 ^a Jiangsu Key Laboratory of Anaerobic Biotechnology, School of Environmental and
8 Civil Engineering, Jiangnan University, Wuxi 214122, PR China

9 ^b Department of Chemical Engineering, University of Bath, Bath, BA2 7AY, UK

10 ^c School of Civil and Environmental Engineering, Nanyang Technological University,
11 Singapore 639798, Singapore

12

13 *Corresponding author:

14 [Tel:+86-13081495594](tel:+86-13081495594), Email: swl@jiangnan.edu.cn (W. Song)

15

16

17

18

19

20

21

22

23 **Abstract**

24 Osmotic membrane bioreactors (OMBR) have gained increasing interest in wastewater
25 treatment and reclamation due to their high product water quality and fouling resistance.
26 However, high energy consumption (mostly by draw solution recovery) restricted the
27 wider application of OMBR. Herein, we propose a novel pressure retarded osmosis
28 membrane bioreactor (PRO-MBR) for improving the economic feasibility. In
29 comparison with conventional FO-MBR, PRO-MBR exhibited similar excellent
30 contaminants removal performance and comparable water flux. More importantly, a
31 considerable amount of energy can be recovered by PRO-MBR (4.1 kWh/100 m²·d),
32 as a result of which, 10.02% of the specific energy consumption (SEC) for water
33 recovery was reduced as compared with FO-MBR (from 1.42 kWh/m³ to 1.28 kWh/m³).
34 Membrane orientation largely determined the performance of PRO-MBR, higher power
35 density was achieved in AL-DS orientation (peak value of 3.4 W/m²) than that in AL-
36 FS orientation (peak value of 1.4 W/m²). However, PRO-MBR suffered more severe
37 and complex membrane fouling when operated in AL-DS orientation, because the
38 porous support layer was facing sludge mixed liquor. Further investigation revealed
39 fouling was mostly reversible for PRO-MBR, it exhibited similar flux recoverability
40 (92.4%) to that in FO-MBR (95.1%) after osmotic backwash. Nevertheless, flux decline
41 due to membrane fouling is still a restricting factor to power generation of PRO-MBR,
42 its power density was decreased by 38.2% in the first 60 min due to the formation of
43 fouling. Overall, in perspective of technoeconomic feasibility, the PRO-MBR
44 demonstrates better potential than FO-MBR in wastewater treatment and reclamation

45 and deserves more research attention in the future.

46 **Keywords:** pressure retarded osmosis; forward osmosis; membrane bioreactor; energy
47 recovery; wastewater treatment

48 **1. Introduction**

49 An osmotic membrane bioreactor (OMBR) that integrates an activated sludge
50 process with a forward osmosis (FO) membrane was firstly proposed by Cornelissen et
51 al. at 2008 ([Cornelissen et al., 2008](#)). In the past decade, OMBR technology has aroused
52 increasing interest in the field of wastewater treatment and reclamation due to the
53 advantages of better product water quality and lower fouling tendency as compared
54 with traditional membrane bioreactors (MBRs) (Nguyen et al., 2015; Wang et al., 2017;
55 Xu et al., 2020). However, there are still bottlenecks in OMBR that hinder its wider
56 application in wastewater treatment and reclamation, e.g., low water flux, salt
57 accumulation, membrane fouling and draw solute recovery (Lee and Hsieh, 2019; Wang
58 et al., 2016a). Draw solute recovery is an essential component in OMBR, by which the
59 draw solute is recycled and the high-quality product water is obtained. Currently, the
60 common approaches for draw solute recovery, including reverse osmosis (RO),
61 nanofiltration (NF) and membrane distillation (MD), consume a large amount of energy
62 to drive the separation process (Eriksson et al., 2005; Luo et al., 2017; Vinardell et al.,
63 2020), directly resulting in a substantial increase in energy consumption and operational
64 cost of OMBR. This is regarded as one of the biggest obstacles on the development and
65 application of OMBR for wastewater treatment and reclamation.

66 In the operation of FO filtration, there is a natural concentration gradient between

67 the two sides of membrane, i.e., a high concentration draw solution (DS) and a low
68 concentration feed solution (FS). Osmotic energy is generated upon the water passes
69 through semipermeable membrane and mixes with the draw solution in the FO process
70 (R. Pattle, 1954). Recent years, osmotic energy has attracted increasing interest because
71 it is a new clean energy that can be sustainably generated with no constraints of the
72 meteorological and geographical conditions (Einarsson and Wu, 2021; Shi et al., 2021).
73 Pressure retarded osmosis (PRO) is one of the most promising technologies for
74 harnessing osmotic energy (Helfer et al., 2014; Son et al., 2016; Thorsen and Holt,
75 2009). During PRO operation, the DS is pressurized and fed into membrane module by
76 a high-pressure pump, and the water from FS permeates into the DS side through the
77 membrane against the hydraulic pressure, then the volume-expanded DS is
78 depressurized via a hydro-turbine to convert osmotic energy to electric power.
79 Compared with conventional FO, the PRO process not only demonstrates similar solute
80 rejection performance but also recovers osmotic energy (Patel et al., 2014; Sakai et al.,
81 2016; Wan and Chung, 2015). The obtained energy can be further utilized to
82 compensate for the energy need of water recovery process.

83 Inspired by osmotic energy recovery in the PRO process, replacing the FO process
84 in OMBR with a PRO process with aim to simultaneously recover osmotic energy and
85 clean water seems to be a potential way to improve the energy efficiency of OMBR.
86 Based on this, present study proposed a novel PRO-MBR of integrating the bioreactor
87 with the PRO process. Existing studies on PRO process mostly employed clean water,
88 river water or low-strength wastewater as FS to evaluate the power generation

89 performance (Kim et al., 2015; O'Toole et al., 2016; Wan and Chung, 2015). The power
90 density of PRO varied significantly with different FS since the concentration and
91 composition of FS closely relate to the water flux and membrane fouling in PRO, which
92 directly or indirectly determines the energy recovery efficiency (Bar-Zeev et al., 2015;
93 She et al., 2017a, 2013; Yip and Elimelech, 2011). To the best of our knowledge, there
94 has been no study focusing on the power generation performance of PRO with sludge
95 mixed liquor as FS. Only one previous paper of ours reported the fouling characteristics
96 in PRO coupled with activated sludge process (Meng et al., 2020). Thus, the power
97 generation performance of PRO-MBR and how much the energy consumption can be
98 reduced as compared with conventional FO-MBR, as well as how the membrane
99 fouling influences the power generation performance in PRO-MBR deserve to be
100 further studied.

101 To this end, a lab-scale PRO-MBR system was established and a comparative
102 study with conventional FO-MBR was then conducted under AL-DS (active layer
103 facing FS) and AL-FS (active layer facing DS) mode. The contaminants removal
104 performance, water flux, power generation performance, membrane fouling behavior
105 and fouling reversibility were comprehensively investigated for both PRO-MBR and
106 FO-MBR with the aim to assess the potential of the PRO-MBR for wastewater
107 treatment and energy recovery.

108 **2. Materials and methods**

109 2.1 Experimental setup

110 A laboratory-scale PRO-MBR comprised of a bioreactor and an FO membrane

111 module was established in this study (Fig. S1). The bioreactor with an effective volume
112 of 1.7 L was full of activated sludge (collected from municipal WWTP), and an aeration
113 diffuser was placed at the bottom. The membrane module was constituted by two
114 identical flow channels (85 mm × 50 mm × 1.5 mm) for FS and DS streams, respectively,
115 with membrane coupon mounted between the two channels. A commercial FO
116 membrane made of cellulose triacetate (CTA) (supplied by Hydration Technologies
117 Innovations, Albany, OR) with an effective membrane area of 25.5 cm² was used in this
118 study. Both the active layer and the support layer of the FO membrane were filled with
119 a tricot-type spacer (She et al., 2017b). The mixed liquor in the bioreactor was
120 circulated by a peristaltic pump (BT100-2J, Longer Precision Pump, China) through
121 the FS flow channel with a cross-flow velocity of 10.3 cm/s, meanwhile a NaCl solution
122 with a concentration of 2 M (osmotic pressure of 9.9 MPa) was pressurized and
123 circulated by a high-pressure pump (DP-130, Xinxishan, China) through the DS flow
124 channel, with a cross-flow velocity of approximately 177 cm/s. The DS tank was placed
125 on a digital balance (PL6001E, Mettler Toledo, China), and the DS weight change was
126 continuously recorded by a computer. To make a fair comparison, a FO-MBR with the
127 entire system the same except without applied hydraulic pressure on DS stream was
128 operated in parallel. The DS solution was circulated by another identical peristaltic
129 pump through the DS flow channel with a cross-flow velocity of 10.3 cm/s.

130 2.2 Operation conditions

131 During the whole experiment, the PRO-MBR and FO-MBR were operated at
132 temperature of 25 ± 1 °C. The hydraulic retention time (HRT) varied in the range of 32

133 to 74 h along with the flux variation in the operation of FO, and no sludge was
134 discharged during the experiment. Synthetic domestic wastewater was used as the feed
135 water with chemical oxygen demand (COD), total organic carbon (TOC), total
136 phosphorus (TP), total nitrogen (TN) and $\text{NH}_4^+\text{-N}$ concentrations of 373.3 ± 17.2 mg/L,
137 81.96 ± 1.68 mg/L, 2.08 ± 0.13 mg/L, 38.24 ± 1.68 mg/L and 24.88 ± 1.50 mg/L,
138 respectively. The composition of synthetic wastewater was set according to that
139 reported in literature (Wang et al., 2014). The sludge collected from a secondary
140 sedimentation tank at the Taihu Xincheng Wastewater Treatment Plant (Wuxi, China)
141 was employed as the seed sludge. It was cultivated in the same bioreactor with synthetic
142 wastewater for approximately 15 days before starting the operation. The initial sludge
143 concentration in the PRO-MBR and FO-MBR were both 3.0 g/L for mixed liquor
144 suspended solids (MLSS) and 2.1 g/L for mixed liquor volatile suspended solids
145 (MLVSS). The aeration rate was approximately 100 L/h, and the corresponding DO
146 concentration in the bioreactors were maintained in the range of 4-5 mg/L.

147 Membrane orientation is a critical factor that largely determines the water flux and
148 membrane fouling behavior in FO and PRO processes (Kim et al., 2016). Therefore,
149 both AL-FS orientation and AL-DS orientation were applied in the operation of PRO-
150 MBR and FO-MBR. As for PRO-MBR, the additional hydraulic pressure applied on
151 the DS side was set as 6 bar (0.6 MPa), which ensured that the FO membrane was
152 maintained mechanically stable in both orientations. The pristine FO membrane was
153 first preconditioned for 4 h in the membrane module in advance to obtain its stable
154 initial water flux (She et al., 2017a).

155 In addition, at the end of each experiment, the fouled membrane was *in situ*
156 physically cleaned for 1 h using 0.08 M NaCl as the FS and deionized water as the DS
157 (i.e., osmotic backwash), according to the method reported in previous literature (Yuan
158 et al., 2015). The DI water flux was retested for the membranes after cleaning and
159 compared with that of pristine membrane, based on which the fouling reversibility was
160 then assessed.

161 2.2 Analytical methods

162 The contaminants concentrations in the permeate, mixed liquor supernatant and
163 feed water were periodically measured for both PRO-MBR and FO-MBR. The
164 concentrations of $\text{NH}_4^+\text{-N}$, $\text{PO}_4^{3-}\text{-P}$, TN, TO, MLSS and MLVSS were determined
165 according to the standard method (APHA, 1998), and the TOC concentration was
166 analyzed by a TOC analyzer (TOC-Vcsh, Shimadzu, Japan).

167 The water flux (J_w) was calculated via the variation of DS weight (according to
168 Eq. (1)), which was continuously recorded by a digital balance connected to a computer.

$$169 \quad J_w = \frac{\Delta V}{A \times \Delta t} \quad (1)$$

170 where ΔV (L) is the collected permeate volume over a pre-determined duration Δt (h),
171 A is the active membrane area (m^2). To eliminate the impacts of the initial water flux of
172 different FO membranes, the normalized flux was used to characterize the water flux
173 performance during the operation of PRO-MBR and FO-MBR. The water flux was
174 normalized by Eq. (2).

$$175 \quad J' = \frac{J_w}{J_0} \quad (2)$$

176 where J' is the normalized flux, J_0 is the initial water flux of the FO membrane ($\text{L}/(\text{m}^2$

177 h)). In addition, the water fluxes after fouling and after physical cleaning were measured
178 to evaluate the flux recoverability in PRO-MBR and FO-MBR. The flux recovery rate
179 was calculated by Eq. (3).

$$180 \quad R = \frac{J_2 - J_1}{J_0 - J_1} \quad (3)$$

181 where R is the flux recovery rate (%), J_1 is the water flux of the fouled FO membrane
182 before physical cleaning ($L/(m^2 h)$), and J_2 is the water flux of the fouled FO membrane
183 after physical cleaning ($L/(m^2 h)$).

184 Power density is widely used to assess the power generation performance of PRO.
185 It is defined as the osmotic energy output per unit membrane area (Han et al., 2016b)
186 and it can be calculated by Eq. (4).

$$187 \quad W = \frac{J_w \times \Delta P}{36} \quad (4)$$

188 where W is the power density (W/m^2), J_w is the water flux of the FO membrane ($L/(m^2$
189 h)), and ΔP is the effective hydraulic pressure difference across the membrane (bar).

190 Specific energy consumption (SEC) was usually used to evaluate the energy
191 efficiency of water recovery process (Seo et al., 2019). SEC is defined as the energy
192 consumed for generating one unit volume of product water and it can be calculated for
193 PRO and FO by Eq. (5).

$$194 \quad SEC = SEC_{pumping} + SEC_{DS\ regeneration} - SEG \quad (5)$$

195 where $SEC_{pumping}$ is the energy consumption of pumping FS/DS, $SEC_{DS\ regeneration}$ is the
196 energy consumption of DS generation process, specific energy generation (SEG) is the
197 energy generated by PRO while unit volume of product water is generated. The
198 $SEC_{pumping}$ of pump was calculated by Eq. (6). and the W_{pump} of high-pressure pump

199 (for pressurizing DS) with energy recovery device was calculated by Eq. (7) (Kim et
200 al., 2013).

$$201 \quad SEC_{pumping} = \frac{W_{pump} \times 24h}{V_{product\ water}} \quad (6)$$

$$202 \quad W_{pump} = \Delta P \times Q_{DS} \times (1 - \eta_{ERD}) \quad (7)$$

203 where W_{pump} is the pump power, Q_{DS} is the DS flow rate, η_{ERD} is the efficiency of energy
204 recovery device (95% in present study), $V_{product\ water}$ is the product water volume per day
205 (m^3). RO is the normally employed way for DS regeneration in FO, thus $SEC_{DS\ regeneration}$
206 is calculated based on the RO as DS regeneration process in present study. The software
207 ROSA 9.1 (Dow Filmtec) was used to simulate and calculate the SEC of RO for DS
208 generation (to be 1.38 kWh/ m^3); Meanwhile, the SEC of RO for DS regeneration
209 (under similar operation conditions) reported in literature was in the range of 1.37-1.5
210 kWh/ m^3 (Chia et al., 2021; Kim et al., 2015; Seo et al., 2019; Zaviska et al., 2015).

211 Therefore, present study takes 1.4 kWh/ m^3 for the following calculation in reference of
212 both the simulated value and the reported value. The energy generated by PRO process
213 can be further utilized to reduce the SEC. The SEG can be calculated as per Eq. (8).

$$214 \quad SEG = \frac{W \times \eta \times 10^{-3} \times 24h \times A}{V_{product\ water}} \quad (8)$$

215 where η is the energy conversion efficiency (95% in present study).

216 At the end of experiments, the fouled FO membranes were carefully collected for
217 fouling characteristic analyses. A field emission scanning electron microscope (FESEM)
218 (S-4800, Hitachi, Japan) and an energy-dispersive X-ray (EDX) analyzer (Falcon,
219 EDAX Inc., USA) were used to characterize the morphology and chemical composition
220 of the fouled FO membranes. In addition, a confocal laser scanning microscope (CLSM,

221 LSM 710, ZESIS, Germany) was applied to characterize the distributions of organic
222 foulants and biofoulants on the fouled FO membrane surfaces and within porous
223 support layer. The target foulants, including α -D-glucopyranose and β -D-
224 glucopyranose polysaccharides, proteins and microorganisms, were stained by
225 concanavalin A (ConA), calcofluor white (CW), fluorescein isothiocyanate (FITC) and
226 SYTO 63, respectively, before characterization. Details of the specific methods of the
227 SEM, EDX and CLSM analyses can be found in our previous publications ([Wang et al.,](#)
228 [2016b; Yuan et al., 2015](#)).

229 **3. Results and discussion**

230 3.1 Contaminants removal performance

231 Firstly, contaminants removal performance of PRO-MBR and FO-MBR were
232 investigated and compared. The two identical MBRs were operated in parallel for more
233 than 30 days to achieve stable biological treatment performance before the start-up of
234 PRO-MBR and FO-MBR. Table 1 summarizes the concentrations of TOC, NH_4^+ -N, TN
235 and TP in the influent, supernatant and permeate, as well as their corresponding removal
236 rates in PRO-MBR and FO-MBR.

237 Excellent removal performances of organic matters and nutrients were achieved in
238 both PRO-MBR and FO-MBR regardless of the membrane orientation. The TOC
239 removal rate and NH_4^+ -N removal rate were $> 96\%$ and $> 98\%$, respectively, for both
240 PRO-MBR and FO-MBR; moreover, no TOC and NH_4^+ -N accumulation was observed
241 in the supernatant, thus this result should be mainly attributed to the biodegradation of
242 microorganisms in the bioreactor. In addition, effective removal of TN ($> 96\%$) and TP

243 (approximately 100%) were also achieved in both PRO-MBR and FO-MBR.
244 Considering the dominating aerobic condition in the MBRs, such high removal
245 performance of TN and TP should be mainly attributed to the high rejection ability of
246 FO membrane to nitrite, nitrate and phosphate. As a result, high-quality product water,
247 with TOC < 3 mg/L, $\text{NH}_4^+\text{-N}$ < 1 mg/L, TN < 1 mg/L and TP not detected, were
248 achieved in both PRO-MBR and FO-MBR. Overall, the contaminants removal
249 performance of the PRO-MBR was comparable with that of FO-MBR, and consistent
250 with previous reports on the osmotic MBRs for treating municipal wastewater (Qiu et
251 al., 2016; Vinardell et al., 2021). PRO-MBR (same as FO-MBR) combines the
252 biodegradation and bioconversion effects of bioreactor with the high retention effect of
253 FO membrane, by which high-efficiency pollutants removal was achieved and high-
254 quality water recovery can be guaranteed.

255 It is noteworthy that in a typical PRO process with wastewater as FS, the
256 contaminants cannot be removed but be retained and accumulated in the FS side. Hence,
257 management of the concentrate should be carefully considered. However, there was no
258 TOC and $\text{NH}_4^+\text{-N}$ accumulation phenomenon in the FS during the operation of PRO-
259 MBR, as suggested by the contaminant concentrations in the supernatant (shown in
260 Table 1), due to the biodegradation and bioconversion effects of microorganisms in the
261 bioreactor. With regard to TN, it can be readily removed by applying A/O-MBR or
262 employing biofilm system. Therefore, the treatment of PRO concentrate, which could
263 inevitably increase the cost and induce secondary pollutants, can be avoided and the
264 sustainability and technoeconomic of PRO process will be improved. In addition,

265 previous study reported that OMBR exhibited lower fouling propensity compared with
 266 direct FO process for municipal wastewater in long-term operation, because much of
 267 the potential organic foulants in wastewater was degraded by bacteria in MBR (Sun et
 268 al., 2016). Thus, a combination of MBR with PRO should be advantageous to fouling
 269 control in PRO. In summary, such a novel PRO-MBR system is potentially able to
 270 achieve simultaneous energy and water recovery in a sustainable way.

271 **Table 1**

272 TOC, NH₄⁺-N, TN and TP concentrations in the influent, sludge supernatant and FO permeate and
 273 their removal rates (average ± standard deviation from triple measurements) in FO-MBR and PRO-
 274 MBR.

Contaminants	Concentrations and removal rates	PRO-MBR	PRO-MBR	FO-MBR	FO-MBR
		AL-DS	AL-FS	AL-DS	AL-FS
TOC	Influent (mg/L)	78.49 ± 4.73	77.49 ± 3.56	78.88 ± 1.57	77.56 ± 2.83
	Sludge supernatant (mg/L)	4.99 ± 2.41	3.50 ± 2.75	3.86 ± 1.15	4.36 ± 0.95
	FO permeate (mg/L)	2.77 ± 1.51	2.07 ± 1.45	2.86 ± 0.76	2.74 ± 0.10
	Removal rate (%)	96.47 ± 1.10	97.33 ± 1.12	96.37 ± 0.51	96.46 ± 0.10
NH ₄ ⁺ -N	Influent (mg/L)	25.06 ± 1.64	25.43 ± 0.89	24.86 ± 0.75	25.34 ± 0.75
	Sludge supernatant (mg/L)	0.28 ± 0.21	0.46 ± 0.28	0.34 ± 0.12	0.51 ± 0.19
	FO permeate (mg/L)	0.37 ± 0.07	0.32 ± 0.03	0.26 ± 0.05	0.39 ± 0.04
	Removal rate (%)	98.52 ± 1.18	98.74 ± 0.26	98.95 ± 0.69	98.46 ± 0.62
TN	Influent (mg/L)	28.73 ± 2.34	28.36 ± 1.85	29.64 ± 1.95	28.49 ± 2.18
	Sludge supernatant (mg/L)	30.58 ± 2.17	30.47 ± 1.24	31.24 ± 1.55	29.98 ± 1.87
	FO permeate (mg/L)	0.78 ± 0.06	0.89 ± 0.05	0.85 ± 0.08	0.61 ± 0.09
	Removal rate (%)	97.29 ± 1.28	96.86 ± 0.39	97.13 ± 0.90	97.86 ± 0.68
TP	Influent (mg/L)	1.95 ± 0.05	2.08 ± 0.04	2.06 ± 0.01	2.12 ± 0.03
	Sludge supernatant (mg/L)	0.28 ± 0.20	0.34 ± 0.18	0.12 ± 0.09	0.36 ± 0.13
	FO permeate (mg/L)	ND	ND	ND	ND
	Removal rate (%)	100	100	100	100

275

276 3.2 Water flux performance

277 The water flux profiles of PRO-MBR and FO-MBR with different membrane
 278 orientations are shown in Figure 1a and Figure 1b, respectively. Generally, water flux
 279 in the FO-MBR was slightly higher (stable flux of 12.1 and 10.8 LMH for AL-DS and
 280 AL-FS, respectively) than in PRO-MBR (stable flux of 11.3 and 8.5 LMH for AL-DS
 281 and AL-FS, respectively) in both two membrane orientations. In PRO system, the draw
 282 solution is pressurized in order to convert the osmotic power to mechanical energy (Shi

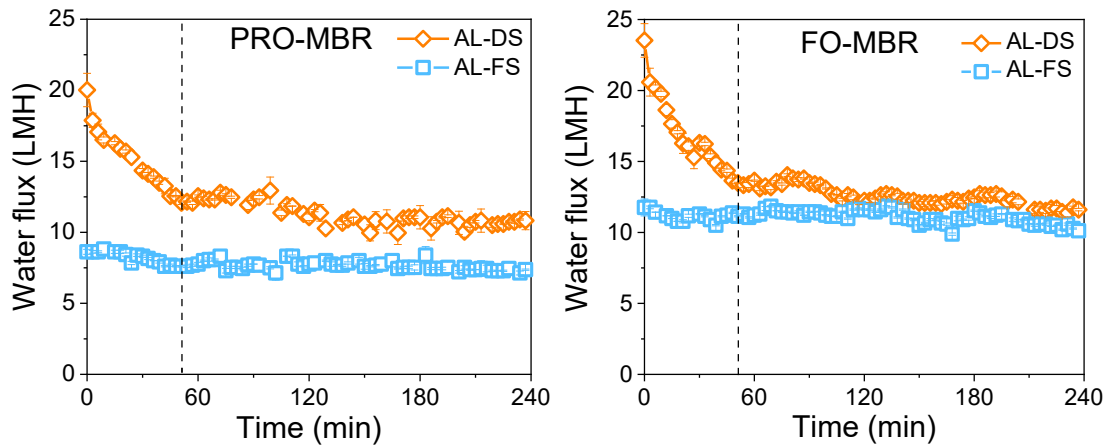
283 [et al., 2021](#)). This additional hydraulic pressure reduces the permeation driving force
284 (osmotic pressure difference) across the membrane thus inducing water flux decline. In
285 present study, a hydraulic pressure of 0.6 MPa was applied on DS side which was much
286 lower than the osmotic pressure difference across membrane (approximately 9.0 MPa)
287 with 2 M NaCl as DS and domestic wastewater as FS. Therefore, PRO-MBR can
288 achieve comparable water flux with that in conventional FO-MBR.

289 Membrane orientation is a critical operational parameter for FO and PRO
290 processes, which substantially influences the water flux performance, the fouling
291 propensity and the membrane stability. The water flux performances of PRO-MBR in
292 AL-DS and AL-FS orientation were compared, consequently. In general, the water flux
293 of membrane operated in AL-DS orientation was consistently higher than that of under
294 AL-FS orientation, i.e., both a higher initial flux (20.0 LMH versus 8.6 LMH) and a
295 higher stable flux (10.6 LMH versus 7.5 LMH) were achieved under the AL-DS
296 orientation. Similar result was also obtained in FO-MBR. The better water flux
297 performance under AL-DS orientation, which was expectable in FO, can be attributed
298 to the less severe internal concentration polarization (ICP) effect under AL-DS
299 orientation than that under AL-FS orientation ([McCutcheon and Elimelech, 2006](#); [Tang](#)
300 [et al., 2010](#)). As for PRO operated in AL-FS orientation, with the porous and thick
301 support layer facing DS, the mixing of high concentration DS and permeate from FS
302 was retarded in support layer, thus resulting in the dilution of DS at the interface of
303 active layer and support layer, and consequently the reduction of osmotic pressure
304 difference across the membrane (permeation driving force). While in AL-DS

305 orientation, with support layer facing FS, the concentrative ICP effect in support layer
306 is relatively lower because the low concentration of FS, thus the influence on osmotic
307 pressure difference is much lower than that in AL-FS orientation. The higher the water
308 flux is, the higher the power density can be achieved in PRO process, therefore the AL-
309 DS orientation is normally adopted for PRO.

310 In contrast, the FO-MBR is normally operated under AL-FS orientation to avoid
311 serious membrane fouling in the support layer of the FO membrane (Honda et al., 2015).
312 Indeed, as shown in Figure 1, the water flux decline in AL-DS orientation was more
313 significant than that in AL-FS orientation for both FO-MBR and PRO-MBR, though
314 the initial flux in AL-DS orientation was much higher. The water flux profiles of
315 membrane operated in AL-DS orientation exhibited typical 2-stage decline curve for
316 both FO-MBR and PRO-MBR, i.e., the water flux in AL-DS orientation dropped
317 rapidly in the first 50 minutes and then stabilized, however, the water flux maintained
318 a relatively stable level in AL-FS orientation during the whole operation period. In AL-
319 DS orientation, where the porous support layer facing mixed liquor, the pollutants can
320 be easily carried into and adsorbed within support layer, moreover the activated sludge
321 also can be directly deposit on support layer surface, which collectively caused rapid
322 flux decline at the beginning of operation; and once a stable cake layer was formed on
323 support layer surface, the penetration of pollutants into support layer might be slowed
324 down due to barrier effect of cake layer, therefore the flux variation proceeded to a
325 gradual decline phase. This result implied that membrane fouling behavior was highly
326 dependent on the membrane orientations in PRO-MBR. Considering the power

327 generation efficiency, PRO is normally operated in AL-DS orientation, however the
328 fouling propensity need to be seriously considered for PRO-MBR in which sludge
329 mixed liquor is used as FS (facing the support layer of membrane).



330

331 **Figure 1** Water flux profiles in PRO-MBR and FO-MBR with different membrane orientations
332 (i.e., AL-DS and AL-FS)

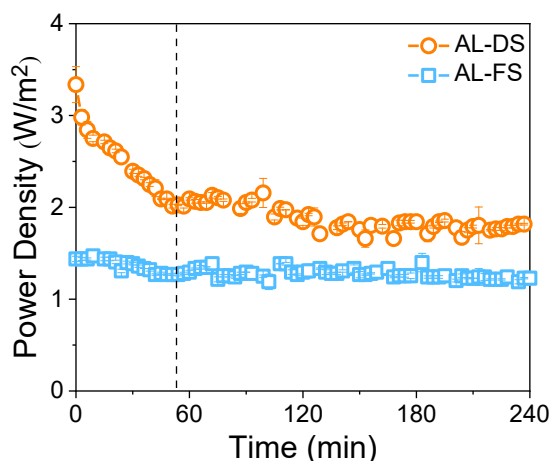
333 3.3 Power generation performance

334 Previous studies demonstrated that substantially different power generation
335 performances were obtained in PRO process in different membrane orientations (AL-
336 FS and AL-DS). Though PRO was normally recommended to operate in AL-DS
337 orientation considering the higher power density and better membrane stability, there is
338 still controversy on which orientation is better when wastewater (with high fouling
339 potential) is used as FS. In AL-DS orientation, membrane is more prone to fouling with
340 porous support layer facing wastewater, as a consequence, the advantage of high power
341 density and techno-economic will be compromised.

342 This study, for the first time, investigated the power generation performances of
343 PRO-MBR (with sludge mixed liquor as FS) operated in AL-DS and AL-FS orientation.
344 Figure 2 presents the power density curves of PRO-MBR in AL-DS and AL-FS

345 orientation. The power density profiles of PRO-MBR (for both two orientations) were
346 observed to follow the similar variation trend of membrane fluxes (as shown in Fig. 1).
347 Based on the fact that the power density is directly proportional to the water flux
348 (according to Eq. (4)), PRO-MBR operated in AL-DS orientation (with a better flux
349 performance than in AL-FS orientation) undoubtedly achieved a higher power density,
350 i.e., the power density ranged from 3.4-1.8 W/m² in the AL-DS orientation while it was
351 only around 1.4 W/m² in the AL-FS orientation. Likewise, this can be simply explained
352 by the fact that the dilutive ICP in AL-FS mode was more severe than the concentrative
353 ICP in AL-DS mode, thus leading to lower flux and poorer power density.

354 It was reported that with the same membrane orientation of AL-DS, similar DS
355 concentration (2 M NaCl) and applied pressure (6.0-6.5 bar) on the DS side, the peak
356 power density of the PRO process was normally around 4.0 W/m² (Kim et al., 2016;
357 She et al., 2013, 2012b). On the other hand, it was reported that the power density of a
358 PRO process was largely compromised due to membrane fouling when real wastewater
359 was used as the feed (Wan and Chung, 2015). Thus, considering the high concentration
360 and complexity of sludge mixed liquor as FS, it is reasonable that the maximum power
361 density achieved in PRO-MBR (3.4 W/m² in AL-DS orientation) was lower than that
362 in ideal condition.



363

364

Figure 2 Power density profiles of PRO-MBR operated in different membrane orientations (i.e., AL-DS and AL-FS).

365

366 3.4 Techno-economic analysis

367

To evaluate how much energy consumption can be reduced by replacing FO with

368

PRO in a OMBR system, the specific energy consumption (SEC) of FO and PRO under

369

AL-DS orientation were analyzed and compared (as shown in Table 2). Energy

370

consumption of a conventional FO system was basically comprised of two aspects: the

371

pumping for FS and DS and the RO process for DS regeneration. As shown in Table 2,

372

the SEC of conventional FO system was 1.427 kWh/m³, in which RO for DS

373

regeneration was the dominant energy consuming component (1.4 kWh/m³). This

374

accounted for 98.2% of the total energy consumption. In contrast, besides the equal

375

energy consumption of RO for DS regeneration, additional hydraulic pressure was

376

applied on DS side in the PRO process, thus the energy consumption was relatively

377

higher (1.451 kWh/m³) than that of conventional FO process.

378

However, osmotic energy was harvested during the PRO process, then the osmotic

379

energy can be further converted to electricity energy as energy supplement by a turbo

380

device. In present study, 0.168 kWh energy was generated along with per m³ water

381 production by PRO process. Considering this additional energy supplement, the net
382 specific energy consumption of the PRO process eventually came to 1.297 kWh/m³, a
383 reduction of 10.09% was achieved via replacing FO with PRO in a OMBR system with
384 otherwise conditions identical. Overall, with the ability of recovering osmotic energy
385 while wastewater treatment, the PRO-MBR showed better economicalness than
386 conventional FO-MBR in the fields of wastewater treatment and reclamation.

387 It is noteworthy that membrane fouling is a critical factor affecting the power
388 generation performance in PRO-MBR. As shown in Figure 2, PRO exhibited the
389 maximum power density (as high as 3.4 W/m²) at the very beginning of operation,
390 however it declined rapidly as the operation proceeded and stabilized at 1.8 W/m².
391 Correspondingly, the energy generation performance decreased from 0.317 kWh/m³ to
392 0.168 kWh/m³ (a reduction of 47.05%). This can be attributed to the formation of
393 fouling layer on support layer of FO membrane during the initial filtration (as discussed
394 in Section 3.2). If such membrane fouling can be mitigated (e.g., applying bio-carriers,
395 quorum quenching strategy, fabricating FO membrane with low S value, etc.), the
396 power density and technoeconomic competitiveness of PRO-MBR could be largely
397 improved. Furthermore, in present study, a relatively low applied hydraulic pressure (6
398 bar) was employed in PRO with the aim to prevent membrane deformation under long-
399 term operation. The applied hydraulic pressure is lower than the theoretical optimum
400 (around 45 bar for present study) for power generation. Therefore, fabricating FO
401 membrane with high mechanical strength (able to withstand high hydraulic pressure)
402 could be another approach to improve the power generation performance of PRO-MBR.

403 In summary, the results of present study preliminarily demonstrated the good techno-
 404 economic potential of the PRO-MBR, while there is still a big room for improvement.

405 **Table 2**

406 The specific energy consumption of FO and PRO

	FS/DS feeding pump ^a (kWh/m ³)	High-pressure pump on DS ^b (kWh/m ³)	RO for DS regeneration ^c (kWh/m ³)	Specific Energy generation ^d (kWh/m ³)	Specific energy consumption (kWh/m ³)
FO	0.027	-	1.4	-	1.427
PRO	0.011	0.040	1.4	0.168	1.283

407 ^a The feeding pump energy consumption was calculated as: $W_{\text{pump}} \times 24 \text{ h} / V_{\text{water production}}$.

408 ^b The energy consumption of high-pressure pump with energy recovery device was calculated as: $\Delta P \times Q_{\text{DS}} \times (1 - \eta_{\text{ERD}}) \times 24 \text{ h} / V_{\text{water production}}$.

410 ^c Energy consumption of RO for DS regeneration was calculated to be 1.38 kWh/m³ by ROSA 9.1 (Dow Filmtec)
 411 was; moreover, the SEC of RO for DS regeneration reported in literature was in the range of 1.37-1.5 kWh/m³ (Chia
 412 et al., 2021; Kim et al., 2015; Seo et al., 2019; Zavisla et al., 2015); present study takes 1.4 kWh/m³ for the following
 413 calculation in reference of both the simulated value and the reported values.

414 ^d The specific energy generation of PRO was calculated as: $W_{\text{PRO}} \times \eta_{\text{PRO}} \times 10^{-3} \times 24 \text{ h} \times A / V_{\text{water production}}$.

415 W_{pump} and $V_{\text{water production}}$ refer to pump power and water production per day; ΔP , Q_{DS} , η_{ERD} refer to applied pressure
 416 on DS, DS flow rate and energy recovery efficiency, respectively; W_{PRO} , η_{PRO} and A refer to power density of PRO,
 417 energy conversion efficiency of PRO and the effective membrane area, respectively.

418 3.5 Membrane fouling characteristics

419 It was showed in previous section that the power density in PRO-MBR was
 420 highly influenced by membrane fouling. Understanding the fouling characteristics in
 421 PRO-MBR is quite essential for developing effective fouling control strategy, and
 422 thereby further improving the power density and sustainability of PRO-MBR.

423 Because of the fact that hydraulic conditions in PRO-MBR was different with
 424 that in FO-MBR, the fouling characteristics in PRO-MBR would be distinct from that
 425 in FO-MBR as well. In addition, unlike the AL-FS membrane orientation that is
 426 normally adopted in FO, PRO process is usually operated in AL-DS mode (porous
 427 support layer facing FS) to achieve higher power density. Therefore, in the case of
 428 sludge mixed liquor as FS, the fouling process could be even more complex in PRO-
 429 MBR. With the aim to clarify the fouling characteristics in PRO-MBR, the fouled
 430 membranes of FO-MBR and PRO-MBR (in both AL-DS and AL-FS orientations) were

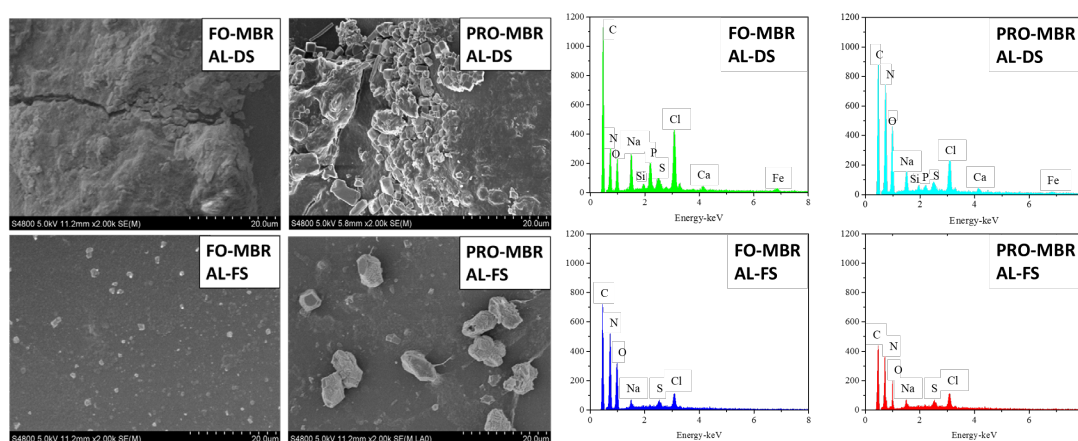
431 collected at the end of experiments and characterized by SEM, EDS and CLSM.

432 Figure 3 presents the SEM images of the side of membranes facing FS (sludge
433 mixed liquor). It is obvious that a sludge cake layer had formed on membranes in AL-
434 DS orientation (support layer facing FS) for both FO-MBR and PRO-MBR, while the
435 fouling on membranes in AL-FS orientation (active layer facing FS) was negligible.
436 Compared to the dense and smooth active layer, the support layer with porous and thick
437 structure was very prone to fouling. It was reported that in the PRO process treating
438 municipal wastewater, most of the fouling occurred in the pores of the support layer
439 (Han et al., 2016a; She et al., 2017b). However, the observed significant sludge cake
440 layer on support layer of membranes in present study indicated that with activated
441 sludge mixed liquor as FS (in AL-DS orientation), the fouling was not only distributed
442 within the pores of support layer but also deposited on the surface of support layer.

443 The element composition of the fouling layers on membranes were further
444 analyzed by EDS. As shown in Figure 3, C, N, O, Na, Cl, P and S were the major
445 elements on membranes fouled in AL-DS orientation for both PRO-MBR and FO-MBR.
446 The presence of Na and Cl on fouled membrane surfaces was the result of reverse salt
447 transport from DS side (Luján-facundo et al., 2017). Additionally, since the pristine
448 CTA-FO membrane only contains C and O, the abundant N element and considerable
449 P and S content suggested that organic fouling or biofouling was formed on membrane
450 surfaces, which was consistent with the finding of sludge cake layer via SEM images.
451 Furthermore, Ca element was also observed on membrane fouled in AL-DS orientation,
452 though with a low intensity, which suggested inorganic ions was involved in the

453 membrane fouling (via complexation or scaling effects). In contrast, the Ca and P
 454 element were undetected on the membranes fouled in AL-FS orientation for both PRO-
 455 MBR and FO-MBR, moreover, the peak intensities of other elements were generally
 456 lower than those on membranes fouled in AL-DS orientation. This result further
 457 confirmed that membrane fouling in AL-DS orientation was more severe and complex.

458 Considering the complexity of membrane fouling in the AL-DS orientation
 459 (porous support layer facing mixed liquor), the cross-section of the membranes fouled
 460 in the AL-DS orientation was further investigated by SEM-EDS. As shown in Figure
 461 2S, fouling took place as expected within the porous support layer of membranes in
 462 both FO-MBR and PRO-MBR. It is noteworthy that unlike the fouling layer on the
 463 surface of support layer, intensive accumulation of Ca and P within support layer of
 464 membranes was observed from the EDS mapping images, implying that inorganic
 465 scaling as a result of the precipitation of Ca and P ions probably took place within
 466 support layer (She et al., 2017a).



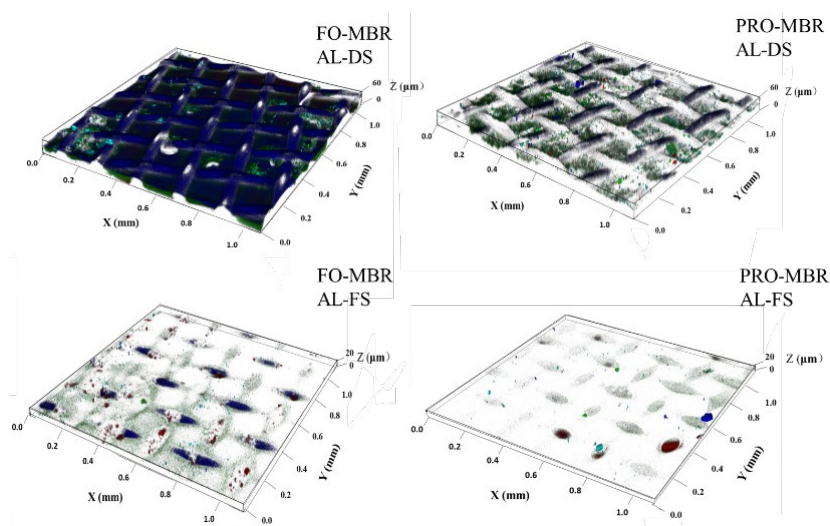
467
 468 **Figure 3** SEM images (left) and EDS spectra (right) of the fouled FO membranes in the PRO-
 469

469 MBR and FO-MBR.

470 Biofouling is normally regarded as the dominant fouling type in membrane

471 bioreactor. To achieve a deeper understanding of the biofouling characteristics in PRO-
472 MBR, the distributions and the contents of bio-foulants (e.g., polysaccharides, proteins
473 and microorganisms) on fouled membranes were further analyzed by CLSM coupled
474 with multiple fluorescence labeling (Li et al., 2019; Wang et al., 2016b; Yuan et al.,
475 2015). As shown in Figure 4, the surface of membranes fouled in AL-DS orientation
476 (for both PRO-MBR and FO-MBR) were covered with thick biofouling layers (both
477 around 60 μm thick); Since the support layer of FO membrane was approximately 30
478 μm thick, it can be inferred that the foulants were indeed located not only within the
479 pores but also on the surface of the support layer. By contrast, the biofouling layers on
480 membranes fouled in the AL-FS orientation were much thinner (approximately 20 μm
481 thick), and the foulants were all deposited on the surface of the active layer. This finding
482 was in consistence with above observations of the fouled FO membranes via SEM and
483 EDX, that membrane fouling was more severe and complex in the AL-DS orientation.

484 A quantitative analysis was further conducted on the fouling layers. The
485 biovolume of various bio-foulants in fouled membrane was calculated by *PHLIP*
486 software (Yuan et al., 2015) and the results are summarized in Table 3. The total
487 biovolume of polysaccharides, proteins and microorganisms on membranes fouled in
488 AL-DS orientation were 30.98 $\mu\text{m}^3/\mu\text{m}^2$ and 16.92 $\mu\text{m}^3/\mu\text{m}^2$ for FO-MBR and PRO-
489 MBR, respectively, which were much higher than those in membrane fouled in AL-FS
490 orientation, i.e., 3.29 $\mu\text{m}^3/\mu\text{m}^2$ in PRO-MBR and 4.84 $\mu\text{m}^3/\mu\text{m}^2$ in FO-MBR. This result
491 further demonstrated that biofouling on membranes fouled in AL-DS orientation was
492 much more significant.



493

494 **Figure 4** CLSM images of the fouled FO membranes in different membrane orientations in the
 495 PRO-MBR and FO-MBR (the cyan, blue, green and red colors represent α -D-glucopyranose, β -D-
 496 glucopyranose, proteins, and microbial cells, respectively).

497

498 **Table 3**

499 Biovolume of the foulants on the fouled FO membranes in PRO-MBR and FO-MBR (calculated by
 500 PHLIP).^a

		α -D- glucopyranose ($\mu\text{m}^3/\mu\text{m}^2$)	β -D- glucopyranose ($\mu\text{m}^3/\mu\text{m}^2$)	Proteins ($\mu\text{m}^3/\mu\text{m}^2$)	Total cells ($\mu\text{m}^3/\mu\text{m}^2$)	Sum ($\mu\text{m}^3/\mu\text{m}^2$)
PRO-	AL-DS	0.21 ± 0.07	7.13 ± 0.71	6.51 ± 0.33	3.07 ± 0.66	16.92 ± 1.77
MBR	AL-FS	0.88 ± 0.14	0.63 ± 0.06	1.06 ± 0.21	0.72 ± 0.08	3.29 ± 0.49
FO-	AL-DS	2.01 ± 0.64	11.13 ± 1.03	9.99 ± 0.42	7.85 ± 0.78	30.98 ± 1.87
MBR	AL-FS	2.00 ± 0.09	1.07 ± 0.04	1.37 ± 0.08	0.40 ± 0.03	4.84 ± 0.24

501

^a Values are given as the mean values \pm standard deviation (number of measurements: n = 3).

502

Above results collectively indicated that membrane orientation largely
 503 determined the fouling behavior in PRO-MBR and FO-MBR. The PRO-MBR, in which
 504 membrane was normally operated in AL-DS orientation, suffered more severe and
 505 complex membrane fouling, as compared with FO-MBR (membrane normally operated
 506 in AL-FS orientation). From another point of view, AL-FS orientation could be a more
 507 promising option in the scenario of PRO-MBR if the shortcomings of severe ICP and
 508 membrane stability (leading to poor power density and membrane damage) can be well

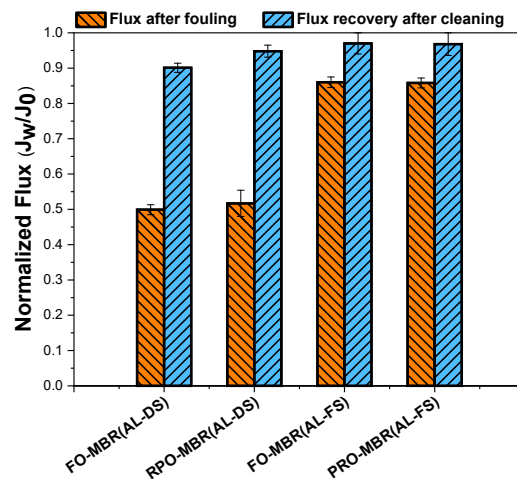
509 addressed.

510 Additionally, it is interesting to observe that the biofouling in PRO-MBR was
511 obviously less than those in FO-MBR when they were both operated in AL-DS
512 orientation (as shown in Figure 4). The biovolume of polysaccharides, proteins and
513 microorganisms on membranes fouled in PRO-MBR (in AL-DS orientation) were 7.33
514 ± 1.77 , 6.51 ± 0.33 and $3.07 \pm 0.66 \mu\text{m}^3/\mu\text{m}^2$, respectively, which were all lower than
515 those in FO-MBR (polysaccharides of $13.14 \pm 1.69 \mu\text{m}^3/\mu\text{m}^2$, proteins of 9.99 ± 0.42
516 $\mu\text{m}^3/\mu\text{m}^2$ and microorganisms of $7.85 \pm 0.78 \mu\text{m}^3/\mu\text{m}^2$). In total, the biofoulants on
517 membrane fouled in PRO-MBR was 45% (in volume) less than those in FO-MBR. Such
518 reduction of biofouling in support layer of membrane in PRO-MBR could be attributed
519 to the result of reverse salt transport. Due to the applied additional hydraulic pressure
520 on DS side, the reverse salt transport was enhanced, thus more salts passed through the
521 active layer, and accumulated in support layer because of the ICP effect. The high
522 salinity stress induced strong inhibitory effect on bioactivity, hence the biofouling was
523 largely restrained. Previous studies generally believed that reverse solute diffusion will
524 enhance the organic fouling in PRO process because the divalent ions (e.g. Ca^{2+}) from
525 DS can promote aggregation of alginate and induce severe pore clogging and cake layer
526 formation (She et al., 2013, 2012a). However, local salinity stress in support layer
527 induced by RSD and its inhibitory effect on the biofouling were not considered in
528 previous studies. Our study provided a new understanding to the effect of RSD on
529 membrane fouling in PRO process.

530 3.6 Fouling reversibility

531 Fouling reversibility is an important factor that determines the sustainability and
532 technoeconomic of MBR system (Song et al., 2018, 2017). At the end of experiment,
533 the fouled membranes in PRO-MBR and FO-MBR were physically cleaned and the
534 fouling reversibility was then evaluated.

535 Figure 5 shows the normalized fluxes of the fouled membranes in PRO-MBR and
536 FO-MBR before and after physical cleaning. Generally, the flux loss of membranes
537 fouled in AL-FS orientation was significantly larger than that in AL-DS orientation for
538 both PRO-MBR and FO-MBR. The normalized flux of membrane after fouling was
539 only 0.51 for PRO-MBR in AL-DS orientation, which was much lower than those for
540 FO-MBR and PRO-MBR in AL-FS orientation (0.85 and 0.86, respectively). This
541 result was in agreement with the result of previous sections that membrane fouling was
542 more severe in AL-DS orientation. In AL-DS orientation, the porous and thick support
543 layer of FO membrane faced the sludge mixed liquor, complex foulants in sludge mixed
544 liquor was easily deposited within the pores, and the aeration scouring effect at
545 membrane surface was unable to completely remove the foulants in support layer, thus
546 leading to inevitable flux decline.



547

548 **Figure 5** Normalized fluxes of the fouled FO membranes in the PRO-MBR and FO-MBR before
549 and after physical cleaning.

550 After osmotic backwash of 3 h, the membrane flux was almost completely
551 recovered (both above 95%) for membranes operated in AL-FS orientation for both
552 PRO-MBR and FO-MBR, which indicted that fouling formed in AL-FS orientation (on
553 active layer surface) was mostly reversible. As for membranes oriented in AL-DS
554 orientation, a comparable flux recovery rate of 92.4% was also achieved by physical
555 cleaning for PRO-MBR, suggesting that most of the fouling in support layer was
556 reversible too. Previous study reported that the membrane fouling in FO-MBR (in AL-
557 FS orientation) normally presented high reversibility, the flux recovery rate of 98% was
558 easily achieved by just osmotic backwash (Yuan et al., 2015). This should be mainly
559 attributed to the very low hydraulic pressure applied in FO process. Unlike that in RO
560 and NF processes (driven by high hydraulic pressure), the FO process was driven by
561 osmotic pressure (exclusively on water molecules) difference across the semipermeable
562 membrane, thus the force driving foulants to membrane is much weaker.

563 Nevertheless, the severe flux loss during operation of PRO-MBR in AL-DS
564 orientation, though mostly reversible, signifies the requirement of high cleaning
565 frequency and operational cost. Additionally, power density is directly proportional to
566 the membrane flux in PRO process, thus the decline of flux also means decrease of
567 power generation performance. Hence, flux decline due to membrane fouling is a
568 critical restricting factor to the performance of RPO-MBR. In view of this, operating
569 PRO-MBR in AL-FS orientation seems a potential way to alleviate membrane fouling,
570 however, as mentioned previously, the severe ICP and membrane stability need to be

571 addressed before.

572 3.7 Implications

573 Comparative analysis (as summarized in Table 4) showed that the PRO-MBR
574 exhibited similar excellent contaminants removal performances to that of FO-MBR for
575 municipal wastewater treatment. Additionally, operating flux comparable with that in
576 FO-MBR was also obtained in PRO-MBR under identical operation conditions. More
577 importantly, with the application of PRO process, a considerable amount of energy can
578 be extracted from the osmosis process (not available in FO-MBR), and be further
579 utilized to reduce system energy consumption. Energy consumption is an important
580 factor that determines the feasibility of osmotic MBR in practical application. In this
581 sense, the PRO-MBR system exhibited better application potential than conventional
582 FO-MBR in the field of wastewater treatment and reclamation.

583 Membrane fouling was an important hindrance to the performance of PRO-MBR.
584 About 40% of the power density was compromised by membrane fouling in PRO-MBR.
585 The power generation performance of PRO-MBR could be further improved if effective
586 fouling control strategies can be developed, e.g., applying bio-carriers, quorum
587 quenching bacteria or antifouling FO membrane material. Furthermore, given the more
588 complex fouling mechanisms, especially biofouling, in PRO-MBR, future research
589 attention should also focus on clarifying its fouling characteristic.

590 The choice of membrane orientation is of paramount importance for PRO-MBR.
591 Present study found that the energy generation efficiency achieved in AL-DS
592 orientation (4.1 kWh/100 m²·d) was 28.1% higher than that in AL-FS orientation (3.2

593 kWh/100 m²·d) with otherwise conditions identical. The relatively lower energy
 594 generation efficiency in AL-FS orientation should be attributed to the more severe ICP
 595 in support layer for FO membrane operated in AL-FS orientation, which induced lower
 596 operating flux, and lower power density as well. Furthermore, the membrane stability
 597 was also a big concern for PRO process in AL-FS orientation. However, the inherent
 598 advantage of less prone to fouling makes the AL-FS orientation still a potential option
 599 for PRO-MBR, in which the severe fouling problem is a critical factor limiting its power
 600 density. Therefore, future study on ICP mitigation strategy in AL-FS orientation and
 601 high-strength FO membrane should be very necessary.

602 **Table 4**

603 Performance comparison between the FO-MBR and the PRO-MBR.^a

	FO-MBR		PRO-MBR		
	AL-DS	AL-FS	AL-DS	AL-FS	
Operating flux (LMH)	13.54 ± 2.31	11.09 ± 0.45	12.19 ± 2.08	7.79 ± 0.42	
Removal rate (%)	TOC	96.51 ± 0.51	96.66 ± 0.10	96.47 ± 1.10	97.33 ± 2.12
	TP	100	100	100	100
	NH ₄ ⁺ -N	97.49 ± 0.69	98.68 ± 0.62	98.52 ± 1.18	98.74 ± 0.26
	TN	96.51 ± 0.90	95.96 ± 0.68	97.29 ± 1.28	96.86 ± 0.39
Flux recovery rate (%)	90.10 ± 1.31	97.04 ± 3.45	94.83 ± 1.71	96.82 ± 3.22	
Specific energy consumption ^b (kWh/m ³)		1.427	1.283	1.288	
Energy generation efficiency ^c (kWh/100 m ² ·d)	-	-	4.1	3.2	

604 ^a Values are given as the mean values ± standard deviation (number of measurements: n = 3).

605 ^b Energy generated by PRO was also considered.

606 ^c Energy generation efficiency was defined as the energy generated by unit membrane area per day.

607 4. Conclusion

608 A novel PRO-MBR was proposed and compared with conventional FO-MBR in this
 609 study. PRO-MBR exhibited comparable contaminants removal and water flux
 610 performances as compared with FO-MBR. Additionally, a considerable amount of
 611 energy (4.1 kWh/100 m²·d) was generated in PRO-MBR, by which the SEC for water
 612 recovery was reduced by 10.02% as compared with FO-MBR. The performance of

613 PRO-MBR was largely determined by membrane orientation, peak power density of
614 3.4 W/m^2 was achieved in AL-DS orientation, while that in AL-FS orientation was only
615 1.4 W/m^2 (because of the severe ICP). However, PRO-MBR suffered more severe and
616 complex membrane fouling when operated in AL-DS orientation. Flux decline induced
617 by membrane fouling restricted the power generation performance of PRO-MBR,
618 especially in AL-DS orientation, the power density was decreased by 38.2% due to the
619 formation of fouling. Future study on PRO-MBR should focus on the control of severe
620 membrane fouling in AL-DS orientation; Moreover, AL-FS orientation could also
621 become a potential option if severe ICP issue was mitigated.

622 **CRedit authorship contribution statement**

623 **Shuyue Liu:** Conceptualization, Methodology, Investigation, Data curation, Writing -
624 original draft. **Weilong Song:** Conceptualization, Supervision, Methodology, Data
625 curation, Review & editing, Project administration. **Manli Meng:** Methodology,
626 Investigation, Data curation. **Ming Xie:** Review & editing. **Qianhong She:** Review &
627 editing. **Pin Zhao:** Project administration, Review & editing. **Xinhua Wang:**
628 Conceptualization, Supervision, Funding acquisition.

629 **Acknowledgments**

630 This work was supported by the National Natural Science Foundation of China [grant
631 number 51978312]; the Six Major Talent Peaks of Jiangsu Province [grant number
632 2018-JNHB-014]; and the Program to Cultivate Middle-aged and Young Science
633 Leaders of Colleges and Universities of Jiangsu Province.

634 **Supporting information**

635 Detailed information on additional figures and foulants extracting method can be found
636 in the Supporting Information

637 **References**

- 638 APHA, 1998. Standard Methods for the Examination of Water and Wastewater-
639 twentieth ed. Am. Public Heal. Assoc. Washington, D.C.
640 <https://doi.org/10.1016/j.cej.2017.02.125>
- 641 Bar-Zeev, E., Perreault, F., Straub, A.P., Elimelech, M., 2015. Impaired Performance
642 of Pressure-Retarded Osmosis due to Irreversible Biofouling. *Environ. Sci.*
643 *Technol.* 49, 13050–13058. <https://doi.org/10.1021/acs.est.5b03523>
- 644 Chia, W.Y., Chia, S.R., Khoo, K.S., Chew, K.W., Show, P.L., 2021. Sustainable
645 membrane technology for resource recovery from wastewater: Forward osmosis
646 and pressure retarded osmosis. *J. Water Process Eng.* 39, 101758.
647 <https://doi.org/10.1016/j.jwpe.2020.101758>
- 648 Cornelissen, E.R., Harmsen, D., de Korte, K.F., Ruiken, C.J., Qin, J.J., Oo, H.,
649 Wessels, L.P., 2008. Membrane fouling and process performance of forward
650 osmosis membranes on activated sludge. *J. Memb. Sci.* 319, 158–168.
651 <https://doi.org/10.1016/j.memsci.2008.03.048>
- 652 Einarsson, S.J., Wu, B., 2021. Thermal associated pressure-retarded osmosis
653 processes for energy production: A review. *Sci. Total Environ.* 757, 143731.
654 <https://doi.org/10.1016/j.scitotenv.2020.143731>
- 655 Eriksson, P., Kyburz, M., Pergande, W., 2005. NF membrane characteristics and
656 evaluation for sea water processing applications. *Desalination* 184, 281–294.

657 <https://doi.org/10.1016/j.desal.2005.04.030>

658 Han, G., Zhou, J., Wan, C., Yang, T., Chung, T., 2016a. Investigations of inorganic
659 and organic fouling behaviors , antifouling and cleaning strategies for pressure
660 retarded osmosis (PRO) membrane using seawater desalination brine and
661 wastewater. *Water Res.* 103, 264–275.
662 <https://doi.org/10.1016/j.watres.2016.07.040>

663 Han, G., Zhou, J., Wan, C., Yang, T., Chung, T.S., 2016b. Investigations of inorganic
664 and organic fouling behaviors, antifouling and cleaning strategies for pressure
665 retarded osmosis (PRO) membrane using seawater desalination brine and
666 wastewater. *Water Res.* 103, 264–275.
667 <https://doi.org/10.1016/j.watres.2016.07.040>

668 Helfer, F., Lemckert, C., Anissimov, Y.G., 2014. Osmotic power with Pressure
669 Retarded Osmosis: Theory, performance and trends - A review. *J. Memb. Sci.*
670 453, 337–358. <https://doi.org/10.1016/j.memsci.2013.10.053>

671 Honda, R., Rukapan, W., Komura, H., Teraoka, Y., Noguchi, M., Hoek, E.M.V.,
672 2015. Effects of membrane orientation on fouling characteristics of forward
673 osmosis membrane in concentration of microalgae culture. *Bioresour. Technol.*
674 197, 429–433. <https://doi.org/10.1016/j.biortech.2015.08.096>

675 Kim, D.I., Kim, J., Hong, S., 2016. Changing membrane orientation in pressure
676 retarded osmosis for sustainable power generation with low fouling. *Desalination*
677 389, 197–206. <https://doi.org/10.1016/j.desal.2016.01.008>

678 Kim, D.I., Kim, J., Shon, H.K., Hong, S., 2015. Pressure retarded osmosis (PRO) for

679 integrating seawater desalination and wastewater reclamation: Energy
680 consumption and fouling. *J. Memb. Sci.* 483, 34–41.
681 <https://doi.org/10.1016/j.memsci.2015.02.025>

682 Kim, J., Park, M., Snyder, S.A., Kim, J.H., 2013. Reverse osmosis (RO) and pressure
683 retarded osmosis (PRO) hybrid processes: Model-based scenario study.
684 *Desalination* 322, 121–130. <https://doi.org/10.1016/j.desal.2013.05.010>

685 Lee, D.J., Hsieh, M.H., 2019. Forward osmosis membrane processes for wastewater
686 bioremediation: Research needs. *Bioresour. Technol.* 290, 121795.
687 <https://doi.org/10.1016/j.biortech.2019.121795>

688 Li, L., Wang, X., Xie, M., Wang, Z., Li, X., Ren, Y., 2019. In situ extracting organic-
689 bound calcium: A novel approach to mitigating organic fouling in forward
690 osmosis treating wastewater via gradient diffusion thin-films. *Water Res.* 156,
691 102–109. <https://doi.org/10.1016/j.watres.2019.03.018>

692 Luján-facundo, M.J., Soler-cabezas, J.L., Mendoza-roca, J.A., Vincent-vela, M.C.,
693 Bes-piá, A., 2017. A study of the osmotic membrane bioreactor process using a
694 sodium chloride solution and an industrial effluent as draw solutions. *Chem.*
695 *Eng. J.* 322, 603–610. <https://doi.org/10.1016/j.cej.2017.04.062>

696 Luo, W., Phan, H. V., Li, G., Hai, F.I., Price, W.E., Elimelech, M., Nghiem, L.D.,
697 2017. An Osmotic Membrane Bioreactor-Membrane Distillation System for
698 Simultaneous Wastewater Reuse and Seawater Desalination: Performance and
699 Implications. *Environ. Sci. Technol.* 51, 14311–14320.
700 <https://doi.org/10.1021/acs.est.7b02567>

701 McCutcheon, J.R., Elimelech, M., 2006. Influence of concentrative and dilutive
702 internal concentration polarization on flux behavior in forward osmosis. *J.*
703 *Memb. Sci.* 284, 237–247. <https://doi.org/10.1016/j.memsci.2006.07.049>

704 Meng, M., Liu, S., Wang, X., 2020. Pressure retarded osmosis coupled with activated
705 sludge process for wastewater treatment: Performance and fouling behaviors.
706 *Bioresour. Technol.* 307, 123224. <https://doi.org/10.1016/j.biortech.2020.123224>

707 Nguyen, N.C., Chen, S.S., Nguyen, H.T., Ngo, H.H., Guo, W., Hao, C.W., Lin, P.H.,
708 2015. Applicability of a novel osmotic membrane bioreactor using a specific
709 draw solution in wastewater treatment. *Sci. Total Environ.* 518–519, 586–594.
710 <https://doi.org/10.1016/j.scitotenv.2015.03.011>

711 O’Toole, G., Jones, L., Coutinho, C., Hayes, C., Napoles, M., Achilli, A., 2016.
712 River-to-sea pressure retarded osmosis: Resource utilization in a full-scale
713 facility. *Desalination* 389, 39–51. <https://doi.org/10.1016/j.desal.2016.01.012>

714 Patel, R., Chi, W.S., Ahn, S.H., Park, C.H., Lee, H.K., Kim, J.H., 2014. Synthesis of
715 poly(vinyl chloride)-g-poly(3-sulfopropyl methacrylate) graft copolymers and
716 their use in pressure retarded osmosis (PRO) membranes. *Chem. Eng. J.* 247, 1–
717 8. <https://doi.org/10.1016/j.cej.2014.02.106>

718 Qiu, G., Zhang, S., Srinivasa Raghavan, D.S., Das, S., Ting, Y.P., 2016. Towards
719 high through-put biological treatment of municipal wastewater and enhanced
720 phosphorus recovery using a hybrid microfiltration-forward osmosis membrane
721 bioreactor with hydraulic retention time in sub-hour level. *Bioresour. Technol.*
722 219, 298–310. <https://doi.org/10.1016/j.biortech.2016.07.126>

723 R. Pattle, 1954. Production of Electric Power by mixing Fresh and Salt Water in the
724 Hydro-electric Pile. *Nature* 174, 660.

725 Sakai, H., Ueyama, T., Irie, M., Matsuyama, K., Tanioka, A., Saito, K., Kumano, A.,
726 2016. Energy recovery by PRO in sea water desalination plant. *Desalination* 389,
727 52–57. <https://doi.org/10.1016/j.desal.2016.01.025>

728 Seo, J., Kim, Y.M., Chae, S.H., Lim, S.J., Park, H., Kim, J.H., 2019. An optimization
729 strategy for a forward osmosis-reverse osmosis hybrid process for wastewater
730 reuse and seawater desalination: A modeling study. *Desalination* 463, 40–49.
731 <https://doi.org/10.1016/j.desal.2019.03.012>

732 She, Q., Jin, X., Li, Q., Tang, C.Y., 2012a. Relating reverse and forward solute
733 diffusion to membrane fouling in osmotically driven membrane processes. *Water*
734 *Res.* 46, 2478–2486. <https://doi.org/10.1016/j.watres.2012.02.024>

735 She, Q., Jin, X., Tang, C.Y., 2012b. Osmotic power production from salinity gradient
736 resource by pressure retarded osmosis: Effects of operating conditions and
737 reverse solute diffusion. *J. Memb. Sci.* 401–402, 262–273.
738 <https://doi.org/10.1016/j.memsci.2012.02.014>

739 She, Q., Wong, Y.K.W., Zhao, S., Tang, C.Y., 2013. Organic fouling in pressure
740 retarded osmosis: Experiments, mechanisms and implications. *J. Memb. Sci.*
741 428, 181–189. <https://doi.org/10.1016/j.memsci.2012.10.045>

742 She, Q., Zhang, L., Wang, R., Krantz, W.B., Fane, A.G., 2017a. Pressure-retarded
743 osmosis with wastewater concentrate feed: Fouling process considerations. *J.*
744 *Memb. Sci.* 542, 233–244. <https://doi.org/10.1016/j.memsci.2017.08.022>

745 She, Q., Zhang, L., Wang, R., Krantz, W.B., Fane, A.G., 2017b. Pressure-retarded
746 osmosis with wastewater concentrate feed : Fouling process considerations. *J.*
747 *Memb. Sci.* 542, 233–244. <https://doi.org/10.1016/j.memsci.2017.08.022>

748 Shi, Y., Zhang, M., Zhang, H., Yang, F., Tang, C.Y., Dong, Y., 2021. Recent
749 development of pressure retarded osmosis membranes for water and energy
750 sustainability: A critical review. *Water Res.* 189, 116666.
751 <https://doi.org/10.1016/j.watres.2020.116666>

752 Son, M., Park, H., Liu, L., Choi, Hyeongyu, Kim, J.H., Choi, Heechul, 2016. Thin-
753 film nanocomposite membrane with CNT positioning in support layer for energy
754 harvesting from saline water. *Chem. Eng. J.* 284, 68–77.
755 <https://doi.org/10.1016/j.cej.2015.08.134>

756 Song, W., Li, Z., Ding, Y., Liu, F., You, H., Qi, P., Wang, F., Li, Y., Jin, C., 2018.
757 Performance of a novel hybrid membrane bioreactor for treating saline
758 wastewater from mariculture: Assessment of pollutants removal and membrane
759 filtration performance. *Chem. Eng. J.* 331, 695–703.
760 <https://doi.org/10.1016/j.cej.2017.09.032>

761 Song, W., You, H., Li, Z., Liu, F., Qi, P., Wang, F., Li, Y., 2017. Membrane fouling
762 mitigation in a moving bed membrane bioreactor combined with anoxic biofilter
763 for treatment of saline wastewater from mariculture. *Bioresour. Technol.* 243.
764 <https://doi.org/10.1016/j.biortech.2017.07.092>

765 Sun, Y., Tian, J., Zhao, Z., Shi, W., Liu, D., Cui, F., 2016. Membrane fouling of
766 forward osmosis (FO) membrane for municipal wastewater treatment: A

767 comparison between direct FO and OMBR. *Water Res.* 104, 330–339.
768 <https://doi.org/10.1016/j.watres.2016.08.039>

769 Tang, C.Y., She, Q., Lay, W.C.L., Wang, R., Fane, A.G., 2010. Coupled effects of
770 internal concentration polarization and fouling on flux behavior of forward
771 osmosis membranes during humic acid filtration. *J. Memb. Sci.* 354, 123–133.
772 <https://doi.org/10.1016/j.memsci.2010.02.059>

773 Thorsen, T., Holt, T., 2009. The potential for power production from salinity
774 gradients by pressure retarded osmosis. *J. Memb. Sci.* 335, 103–110.
775 <https://doi.org/10.1016/j.memsci.2009.03.003>

776 Vinardell, S., Astals, S., Jaramillo, M., Mata-Alvarez, J., Dosta, J., 2021. Anaerobic
777 membrane bioreactor performance at different wastewater pre-concentration
778 factors: An experimental and economic study. *Sci. Total Environ.* 750, 141625.
779 <https://doi.org/10.1016/j.scitotenv.2020.141625>

780 Vinardell, S., Astals, S., Mata-Alvarez, J., Dosta, J., 2020. Techno-economic analysis
781 of combining forward osmosis-reverse osmosis and anaerobic membrane
782 bioreactor technologies for municipal wastewater treatment and water
783 production. *Bioresour. Technol.* 297, 122395.
784 <https://doi.org/10.1016/j.biortech.2019.122395>

785 Wan, C.F., Chung, T.S., 2015. Osmotic power generation by pressure retarded
786 osmosis using seawater brine as the draw solution and wastewater retentate as
787 the feed. *J. Memb. Sci.* 479, 148–158.
788 <https://doi.org/10.1016/j.memsci.2014.12.036>

789 Wang, X., Chang, V.W.C., Tang, C.Y., 2016a. Osmotic membrane bioreactor
790 (OMBR) technology for wastewater treatment and reclamation: Advances,
791 challenges, and prospects for the future. *J. Memb. Sci.* 504, 113–132.
792 <https://doi.org/10.1016/j.memsci.2016.01.010>

793 Wang, X., Chen, Y., Yuan, B., Li, X., Ren, Y., 2014. Impacts of sludge retention time
794 on sludge characteristics and membrane fouling in a submerged osmotic
795 membrane bioreactor. *Bioresour. Technol.* 161, 340–347.
796 <https://doi.org/10.1016/j.biortech.2014.03.058>

797 Wang, X., Wang, C., Tang, C.Y., Hu, T., Li, X., Ren, Y., 2017. Development of a
798 novel anaerobic membrane bioreactor simultaneously integrating microfiltration
799 and forward osmosis membranes for low-strength wastewater treatment. *J.*
800 *Memb. Sci.* 527, 1–7. <https://doi.org/10.1016/j.memsci.2016.12.062>

801 Wang, X., Zhao, Y., Yuan, B., Wang, Z., Li, X., Ren, Y., 2016b. Comparison of
802 biofouling mechanisms between cellulose triacetate (CTA) and thin-film
803 composite (TFC) polyamide forward osmosis membranes in osmotic membrane
804 bioreactors. *Bioresour. Technol.* 202, 50–58.
805 <https://doi.org/10.1016/j.biortech.2015.11.087>

806 Xu, X., Zhang, H., Gao, T., Wang, Y., Teng, J., Lu, M., 2020. Customized thin and
807 loose cake layer to mitigate membrane fouling in an electro-assisted anaerobic
808 forward osmosis membrane bioreactor (AnOMEBR). *Sci. Total Environ.* 729,
809 138663. <https://doi.org/10.1016/j.scitotenv.2020.138663>

810 Yip, N.Y., Elimelech, M., 2011. Performance limiting effects in power generation

811 from salinity gradients by pressure retarded osmosis. *Environ. Sci. Technol.* 45,
812 10273–10282. <https://doi.org/10.1021/es203197e>

813 Yuan, B., Wang, X., Tang, C., Li, X., Yu, G., 2015. In situ observation of the growth
814 of biofouling layer in osmotic membrane bioreactors by multiple fluorescence
815 labeling and confocal laser scanning microscopy. *Water Res.* 75, 188–200.
816 <https://doi.org/10.1016/j.watres.2015.02.048>

817 Zaviska, F., Chun, Y., Heran, M., Zou, L., 2015. Using FO as pre-treatment of RO for
818 high scaling potential brackish water: Energy and performance optimisation. *J.*
819 *Memb. Sci.* 492, 430–438. <https://doi.org/10.1016/j.memsci.2015.06.004>

820

821 **Table Captions**

822 **Table 1** TOC, NH₄⁺-N, TN and TP concentrations in the influent, sludge supernatant
823 and FO permeate and their removal rates (average ± standard deviation from triple
824 measurements) in FO-MBR and PRO-MBR.

825 **Table 2** The specific energy consumption of FO and PRO.

826 **Table 3** Biovolume of the foulants on the fouled FO membranes in PRO-MBR and FO-
827 MBR (calculated by PHLIP).^a

828 **Table 4** Performance comparison between the FO-MBR and the PRO-MBR.^a

829

830 **Figure Captions**

831 **Figure 1** Water flux profiles in PRO-MBR and FO-MBR with different membrane
832 orientations (i.e., AL-DS and AL-FS).

833 **Figure 2** Power density profiles of PRO-MBR operated in different membrane

834 orientations (i.e., AL-DS and AL-FS).

835 **Figure 3** SEM images (left) and EDS spectra (right) of the fouled FO membranes in
836 the PRO-MBR and FO-MBR.

837 **Figure 4** CLSM images of the fouled FO membranes in different membrane
838 orientations in the PRO-MBR and FO-MBR (the cyan, blue, green and red colors
839 represent α -D-glucopyranose, β -D-glucopyranose, proteins, and microbial cells,
840 respectively).

841 **Figure 5** Normalized fluxes of the fouled FO membranes in the PRO-MBR and FO-
842 MBR before and after physical cleaning.

843