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1	Engineering pressure retarded osmosis membrane bioreactor (PRO-
2	MBR) for simultaneous water and energy recovery from municipal
3	wastewater
4	
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Osmotic membrane bioreactors (OMBR) have gained increasing interest in wastewater 24 25 treatment and reclamation due to their high product water quality and fouling resistance. However, high energy consumption (mostly by draw solution recovery) restricted the 26 wider application of OMBR. Herein, we propose a novel pressure retarded osmosis 27 membrane bioreactor (PRO-MBR) for improving the economic feasibility. In 28 comparison with conventional FO-MBR, PRO-MBR exhibited similar excellent 29 contaminants removal performance and comparable water flux. More importantly, a 30 considerable amount of energy can be recovered by PRO-MBR (4.1 kWh/100 m<sup>2</sup>·d), 31 as a result of which, 10.02% of the specific energy consumption (SEC) for water 32 recovery was reduced as compared with FO-MBR (from 1.42 kWh/m<sup>3</sup> to 1.28 kWh/m<sup>3</sup>). 33 34 Membrane orientation largely determined the performance of PRO-MBR, higher power density was achieved in AL-DS orientation (peak value of 3.4 W/m<sup>2</sup>) than that in AL-35 FS orientation (peak value of 1.4 W/m<sup>2</sup>). However, PRO-MBR suffered more severe 36 and complex membrane fouling when operated in AL-DS orientation, because the 37 porous support layer was facing sludge mixed liquor. Further investigation revealed 38 fouling was mostly reversible for PRO-MBR, it exhibited similar flux recoverability 39 (92.4%) to that in FO-MBR (95.1%) after osmotic backwash. Nevertheless, flux decline 40 due to membrane fouling is still a restricting factor to power generation of PRO-MBR, 41 its power density was decreased by 38.2% in the first 60 min due to the formation of 42 fouling. Overall, in perspective of technoeconomic feasibility, the PRO-MBR 43 demonstrates better potential than FO-MBR in wastewater treatment and reclamation 44

45 and deserves more research attention in the future.

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

48 **1. Introduction** 

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

66

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

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

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

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

To this end, a lab-scale PRO-MBR system was established and a comparative study with conventional FO-MBR was then conducted under AL-DS (active layer facing FS) and AL-FS (active layer facing DS) mode. The contaminants removal performance, water flux, power generation performance, membrane fouling behavior and fouling reversibility were comprehensively investigated for both PRO-MBR and FO-MBR with the aim to assess the potential of the PRO-MBR for wastewater 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 $\times$ 50 mm $\times$ 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 cm <sup>2</sup> 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 expect 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

During the whole experiment, the PRO-MBR and FO-MBR were operated at temperature of  $25 \pm 1$  °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 NH <sub>4</sub> <sup>+</sup> -N concentrations of $373.3 \pm 17.2$ mg/L,
137	$81.96 \pm 1.68$ mg/L, $2.08 \pm 0.13$ mg/L, $38.24 \pm 1.68$ mg/L and $24.88 \pm 1.50$ 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.

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

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

161 2.2 Analytical methods

The contaminants concentrations in the permeate, mixed liquor supernatant and feed water were periodically measured for both PRO-MBR and FO-MBR. The concentrations of  $NH_4^+$ -N,  $PO_4^{3-}$ -P, TN, TO, MLSS and MLVSS were determined according to the standard method (APHA, 1998), and the TOC concentration was analyzed by a TOC analyzer (TOC-Vcsh, Shimadzu, Japan).

167 The water flux (J<sub>w</sub>) 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  $J_w = \frac{\Delta V}{A \times \Delta t} \tag{1}$ 

where  $\Delta V$  (L) is the collected permeate volume over a pre-determined duration  $\Delta t$  (h), A is the active membrane area (m<sup>2</sup>). To eliminate the impacts of the initial water flux of different FO membranes, the normalized flux was used to characterize the water flux performance during the operation of PRO-MBR and FO-MBR. The water flux was normalized by Eq. (2).

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

where J' is the normalized flux,  $J_0$  is the initial water flux of the FO membrane (L/ (m<sup>2</sup>

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

$$R = \frac{J_2 - J_1}{J_0 - J_1} \tag{3}$$

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

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

187 
$$W = \frac{J_w \times \Delta P}{36} \tag{4}$$

where W is the power density (W/m<sup>2</sup>),  $J_w$  is the water flux of the FO membrane (L/ (m<sup>2</sup>)

189 h)), and  $\Delta P$  is the effective hydraulic pressure difference across the membrane (bar).

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

193 PRO and FO by Eq. (5).

$$SEC = SEC_{pumping} + SEC_{DS \ regeneration} - SEG \tag{5}$$

where  $SEC_{pumping}$  is the energy consumption of pumping FS/DS,  $SEC_{DS regeneration}$  is the energy consumption of DS generation process, specific energy generation (SEG) is the energy generated by PRO while unit volume of product water is generated. The SEC<sub>pumping</sub> of pump was calculated by Eq. (6). and the W<sub>pump</sub> of high-pressure pump (for pressurizing DS) with energy recovery device was calculated by Eq. (7) (Kim etal., 2013).

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

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

where  $W_{pump}$  is the pump power,  $Q_{DS}$  is the DS flow rate,  $\eta_{ERD}$  is the efficiency of energy 203 recovery device (95% in present study), V<sub>product water</sub> is the product water volume per day 204 (m<sup>3</sup>). RO is the normally employed way for DS regeneration in FO, thus SEC<sub>DS regeneration</sub> 205 is calculated based on the RO as DS regeneration process in present study. The software 206 ROSA 9.1 (Dow Filmtec) was used to simulate and calculate the SEC of RO for DS 207 generation (to be 1.38 kWh/m<sup>3</sup>); Meanwhile, the SEC of RO for DS regeneration 208 (under similar operation conditions) reported in literature was in the range of 1.37-1.5 209 kWh/m<sup>3</sup> (Chia et al., 2021; Kim et al., 2015; Seo et al., 2019; Zaviska et al., 2015). 210 Therefore, present study takes 1.4 kWh/m<sup>3</sup> for the following calculation in reference of 211 both the simulated value and the reported value. The energy generated by PRO process 212 can be further utilized to reduce the SEC. The SEG can be calculate d as per Eq. (8). 213

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

where  $\eta$  is the energy conversion efficiency (95% in present study).

At the end of experiments, the fouled FO membranes were carefully collected for fouling characteristic analyses. A field emission scanning electron microscope (FESEM) (S-4800, Hitachi, Japan) and an energy-dispersive X-ray (EDX) analyzer (Falcon, EDAX Inc., USA) were used to characterize the morphology and chemical composition of the fouled FO membranes. In addition, a confocal laser scanning microscope (CLSM, LSM 710, ZESIS, Germany) was applied to characterize the distributions of organic foulants and biofoulants on the fouled FO membrane surfaces and within porous support layer. The target foulants, including  $\alpha$ -D-glucopyranose and  $\beta$ -Dglucopyranose polysaccharides, proteins and microorganisms, were stained by concanavalin A (ConA), calcofluor white (CW), fluorescein isothiocyanate (FITC) and SYTO 63, respectively, before characterization. Details of the specific methods of the 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

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

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

(approximately 100%) were also achieved in both PRO-MBR and FO-MBR. 243 Considering the dominating aerobic condition in the MBRs, such high removal 244 performance of TN and TP should be mainly attributed to the high rejection ability of 245 FO membrane to nitrite, nitrate and phosphate. As a result, high-quality product water, 246 with TOC < 3 mg/L,  $NH_4^+$ -N < 1 mg/L, TN < 1 mg/L and TP not detected, were 247 achieved in both PRO-MBR and FO-MBR. Overall, the contaminants removal 248 performance of the PRO-MBR was comparable with that of FO-MBR, and consistent 249 with previous reports on the osmotic MBRs for treating municipal wastewater (Qiu et 250 251 al., 2016; Vinardell et al., 2021). PRO-MBR (same as FO-MBR) combines the biodegradation and bioconversion effects of bioreactor with the high retention effect of 252 FO membrane, by which high-efficiency pollutants removal was achieved and high-253 254 quality water recovery can be guaranteed.

It is noteworthy that in a typical PRO process with wastewater as FS, the 255 contaminants cannot be removed but be retained and accumulated in the FS side. Hence, 256 257 management of the concentrate should be carefully considered. However, there was no TOC and NH<sub>4</sub><sup>+</sup>-N accumulation phenomenon in the FS during the operation of PRO-258 MBR, as suggested by the contaminant concentrations in the supernatant (shown in 259 Table 1), due to the biodegradation and bioconversion effects of microorganisms in the 260 bioreactor. With regard to TN, it can be readily removed by applying A/O-MBR or 261 employing biofilm system. Therefore, the treatment of PRO concentrate, which could 262 inevitably increase the cost and induce secondary pollutants, can be avoided and the 263 sustainability and technoeconomic of PRO process will be improved. In addition, 264

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.

#### Table 1 271

TOC, NH4+-N, TN and TP concentrations in the influent, sludge supernatant and FO permeate and 272

their removal rates (average ± standard deviation from triple measurements) in FO-MBR and PRO-273

274 MBR.

IDR.					
Contaminants	Concentrations and removal rates	PRO-MBR	PRO-MBR	FO-MBR	FO-MBR
Contaminants	Concentrations and removal rates	AL-DS	AL-FS	AL-DS	AL-FS
	Influent (mg/L)	$78.49 \pm 4.73$	$77.49\pm3.56$	$78.88 \pm 1.57$	$77.56 \pm 2.83$
TOC	Sludge supernatant (mg/L)	$4.99 \pm 2.41$	$3.50\pm2.75$	$3.86 \pm 1.15$	$4.36\pm0.95$
TOC	FO permeate (mg/L)	$2.77 \pm 1.51$	$2.07 \pm 1.45$	$2.86\pm0.76$	$2.74\pm0.10$
	Removal rate (%)	$96.47 \pm 1.10$	$97.33 \pm 1.12$	$96.37\pm0.51$	$96.46\pm0.10$
	Influent (mg/L)	$25.06 \pm 1.64$	$25.43\pm0.89$	$24.86 \pm 0.75$	$25.34\pm0.75$
NH4 <sup>+</sup> -N	Sludge supernatant (mg/L)	$0.28\pm0.21$	$0.46\pm0.28$	$0.34\pm0.12$	$0.51\pm0.19$
INП4 -IN	FO permeate (mg/L)	$0.37\pm0.07$	$0.32\pm0.03$	$0.26{\pm}~0.05$	$0.39{\pm}~0.04$
	Removal rate (%)	$98.52 \pm 1.18$	$98.74\pm0.26$	$98.95{\pm}0.69$	$98.46{\pm}0.62$
	Influent (mg/L)	$28.73\pm2.34$	$28.36 \pm 1.85$	$29.64 \pm 1.95$	$28.49\pm2.18$
TN	Sludge supernatant (mg/L)	$30.58\pm2.17$	$30.47 \pm 1.24$	$31.24\pm1.55$	$29.98 \pm 1.87$
110	FO permeate (mg/L)	$0.78\pm0.06$	$0.89\pm0.05$	$0.85\pm0.08$	$0.61\pm0.09$
	Removal rate (%)	$97.29 \pm 1.28$	$96.86\pm0.39$	$97.13\pm0.90$	$97.86\pm0.68$
	Influent (mg/L)	$1.95\pm0.05$	$2.08\pm0.04$	$2.06\pm0.01$	$2.12\pm0.03$
TD	Sludge supernatant (mg/L)	$0.28\pm0.20$	$0.34\pm0.18$	$0.12 \pm 0.09$	$0.36\pm0.13$
TP	FO permeate (mg/L)	ND	ND	ND	ND
	Removal rate (%)	100	100	100	100

275

#### 3.2 Water flux performance 276

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

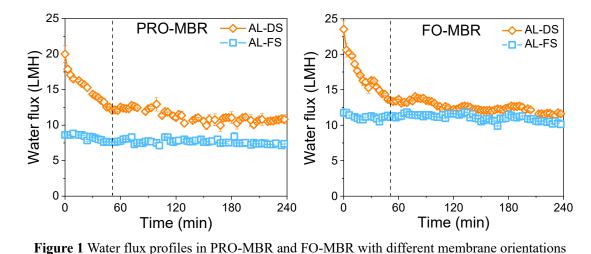
et al., 2021). This additional hydraulic pressure reduces the permeation driving force
(osmotic pressure difference) across the membrane thus inducing water flux decline. In
present study, a hydraulic pressure of 0.6 MPa was applied on DS side which was much
lower than the osmotic pressure deference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

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

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

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

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



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

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332

333

3.3 Power generation performance

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

This study, for the first time, investigated the power generation performances of PRO-MBR (with sludge mixed liquor as FS) operated in AL-DS and AL-FS orientation. 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 $3.4-1.8 \text{ W/m}^2$ in the AL-DS orientation while it was
351	only around 1.4 $W/m^2$ 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
354 355	It was reported that with the same membrane orientation of AL-DS, similar DS concentration (2 M NaCl) and applied pressure (6.0-6.5 bar) on the DS side, the peak
355	concentration (2 M NaCl) and applied pressure (6.0-6.5 bar) on the DS side, the peak
355 356	concentration (2 M NaCl) and applied pressure (6.0-6.5 bar) on the DS side, the peak power density of the PRO process was normally around 4.0 $W/m^2$ (Kim et al., 2016;
355 356 357	concentration (2 M NaCl) and applied pressure (6.0-6.5 bar) on the DS side, the peak power density of the PRO process was normally around 4.0 W/m <sup>2</sup> (Kim et al., 2016; She et al., 2013, 2012b). On the other hand, it was reported that the power density of a
355 356 357 358	concentration (2 M NaCl) and applied pressure (6.0-6.5 bar) on the DS side, the peak power density of the PRO process was normally around 4.0 W/m <sup>2</sup> (Kim et al., 2016; She et al., 2013, 2012b). On the other hand, it was reported that the power density of a PRO process was largely compromised due to membrane fouling when real wastewater
355 356 357 358 359	concentration (2 M NaCl) and applied pressure (6.0-6.5 bar) on the DS side, the peak power density of the PRO process was normally around 4.0 W/m <sup>2</sup> (Kim et al., 2016; She et al., 2013, 2012b). On the other hand, it was reported that the power density of a PRO process was largely compromised due to membrane fouling when real wastewater was used as the feed (Wan and Chung, 2015). Thus, considering the high concentration

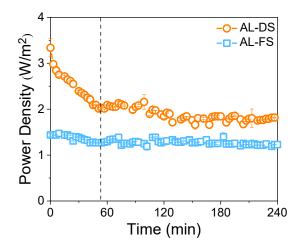


Figure 2 Power density profiles of PRO-MBR operated in different membrane orientations (i.e.,
 AL-DS and AL-FS).

366 3.4 Techno-economic analysis

363

To evaluate how much energy consumption can be reduced by replacing FO with 367 PRO in a OMBR system, the specific energy consumption (SEC) of FO and PRO under 368 AL-DS orientation were analyzed and compared (as shown in Table 2). Energy 369 370 consumption of a conventional FO system was basically comprised of two aspects: the pumping for FS and DS and the RO process for DS regeneration. As shown in Table 2, 371 the SEC of conventional FO system was 1.427 kWh/m<sup>3</sup>, in which RO for DS 372 373 regeneration was the dominant energy consuming component (1.4 kWh/m<sup>3</sup>). This accounted for 98.2% of the total energy consumption. In contrast, besides the equal 374 energy consumption of RO for DS regeneration, additional hydraulic pressure was 375 applied on DS side in the PRO process, thus the energy consumption was relatively 376 higher  $(1.451 \text{ kWh/m}^3)$  than that of conventional FO process. 377

However, osmotic energy was harvested during the PRO process, then the osmotic energy can be further converted to electricity energy as energy supplement by a turbo device. In present study, 0.168 kWh energy was generated along with per m<sup>3</sup> water production by PRO process. Considering this additional energy supplement, the net specific energy consumption of the PRO process eventually came to 1.297 kWh/m<sup>3</sup>, a reduction of 10.09% was achieved via replacing FO with PRO in a OMBR system with otherwise conditions identical. Overall, with the ability of recovering osmotic energy while wastewater treatment, the PRO-MBR showed better economicalness than conventional FO-MBR in the fields of wastewater treatment and reclamation.

It is noteworthy that membrane fouling is a critical factor affecting the power 387 generation performance in PRO-MBR. As shown in Figure 2, PRO exhibited the 388 maximum power density (as high as  $3.4 \text{ W/m}^2$ ) at the very beginning of operation, 389 however it declined rapidly as the operation proceeded and stabilized at 1.8 W/m<sup>2</sup>. 390 Correspondingly, the energy generation performance decreased from 0.317 kWh/m<sup>3</sup> to 391  $0.168 \text{ kWh/m}^3$  (a reduction of 47.05%). This can be attributed to the formation of 392 fouling layer on support layer of FO membrane during the initial filtration (as discussed 393 in Section 3.2). If such membrane fouling can be mitigated (e.g., applying bio-carriers, 394 quorum quenching strategy, fabricating FO membrane with low S value, etc.), the 395 power density and technoeconomic competitiveness of PRO-MBR could be largely 396 improved. Furthermore, in present study, a relatively low applied hydraulic pressure (6 397 bar) was employed in PRO with the aim to prevent membrane deformation under long-398 term operation. The applied hydraulic pressure is lower than the theoretical optimum 399 (around 45 bar for present study) for power generation. Therefore, fabricating FO 400 membrane with high mechanical strength (able to withstand high hydraulic pressure) 401 could be another approach to improve the power generation performance of PRO-MBR. 402

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

1	6,	1			
	FS/DS	High-pressure	RO for DS	Specific Energy	Specific energy
	feeding pump	pump on DS <sup>b</sup>	regeneration <sup>c</sup>	generation d	consumption
	$a (kWh/m^3)$	$(kWh/m^3)$	(kWh/m <sup>3</sup> )	$(kWh/m^3)$	$(kWh/m^3)$
FO	0.027	-	1.4	-	1.427
PRO	0.011	0.040	1.4	0.168	1.283

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

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

<sup>c</sup> Energy consumption of RO for DS regeneration was calculated to be 1.38 kWh/m<sup>3</sup> by ROSA 9.1 (Dow Filmtec)
was; moreover, the SEC of RO for DS regeneration reported in literature was in the range of 1.37-1.5 kWh/m<sup>3</sup> (Chia et al., 2021; Kim et al., 2015; Seo et al., 2019; Zaviska et al., 2015); present study takes 1.4 kWh/m<sup>3</sup> for the following calculation in reference of both the simulated value and the reported values.

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

 $415 \qquad W_{pump} \ and \ V_{water \ production} \ refer \ to \ pump \ power \ and \ water \ production \ per \ day; \ \Delta P, \ Q_{DS}, \ \eta_{ERD} \ refer \ to \ applied \ pressure$ 

416 on DS, DS flow rate and energy recovery efficiency, respectively; W<sub>PRO</sub>, η<sub>PRO</sub> and A refer to power density of PRO,

energy conversion efficiency of PRO and the effective membrane area, respectively.

418 3.5 Membrane fouling characteristics

It was showed in previous section that the power density in PRO-MBR was 419 highly influenced by membrane fouling. Understanding the fouling characteristics in 420 421 PRO-MBR is quite essential for developing effective fouling control strategy, and thereby further improving the power density and sustainability of PRO-MBR. 422 Because of the fact that hydraulic conditions in PRO-MBR was different with 423 424 that in FO-MBR, the fouling characteristics in PRO-MBR would be distinct from that in FO-MBR as well. In addition, unlike the AL-FS membrane orientation that is 425 normally adopted in FO, PRO process is usually operated in AL-DS mode (porous 426 support layer facing FS) to achieve higher power density. Therefore, in the case of 427

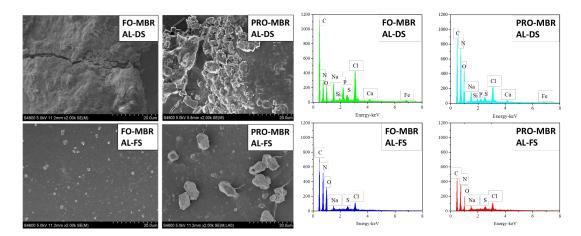
- 428 sludge mixed liquor as FS, the fouling process could be even more complex in PRO-
- 429 MBR. With the aim to clarify the fouling characteristics in PRO-MBR, the fouled
- 430 membranes of FO-MBR and PRO-MBR (in both AL-DS and AL-FS orientations) were

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

Figure 3 presents the SEM images of the side of membranes facing FS (sludge 432 mixed liquor). It is obvious that a sludge cake layer had formed on membranes in AL-433 DS orientation (support layer facing FS) for both FO-MBR and PRO-MBR, while the 434 fouling on membranes in AL-FS orientation (active layer facing FS) was negligible. 435 Compared to the dense and smooth active layer, the support layer with porous and thick 436 structure was very prone to fouling. It was reported that in the PRO process treating 437 municipal wastewater, most of the fouling occurred in the pores of the support layer 438 439 (Han et al., 2016a; She et al., 2017b). However, the observed significant sludge cake layer on support layer of membranes in present study indicated that with activated 440 sludge mixed liquor as FS (in AL-DS orientation), the fouling was not only distributed 441 442 within the pores of support layer but also deposited on the surface of support layer. The element composition of the fouling layers on membranes were further 443 analyzed by EDS. As shown in Figure 3, C, N, O, Na, Cl, P and S were the major 444 elements on membranes fouled in AL-DS orientation for both PRO-MBR and FO-MBR. 445 The presence of Na and Cl on fouled membrane surfaces was the result of reverse salt 446 transport from DS side (Luján-facundo et al., 2017). Additionally, since the pristine 447 CTA-FO membrane only contains C and O, the abundant N element and considerable 448 P and S content suggested that organic fouling or biofouling was formed on membrane 449 surfaces, which was consistent with the finding of sludge cake layer via SEM images. 450 Furthermore, Ca element was also observed on membrane fouled in AL-DS orientation, 451

452 though with a low intensity, which suggested inorganic ions was involved in the

membrane fouling (via complexation or scaling effects). In contrast, the Ca and P 453 element were undetected on the membranes fouled in AL-FS orientation for both PRO-454 455 MBR and FO-MBR, moreover, the peak intensities of other elements were generally lower than those on membranes fouled in AL-DS orientation. This result further 456 confirmed that membrane fouling in AL-DS orientation was more severe and complex. 457 Considering the complexity of membrane fouling in the AL-DS orientation 458 (porous support layer facing mixed liquor), the cross-section of the membranes fouled 459 in the AL-DS orientation was further investigated by SEM-EDS. As shown in Figure 460 461 2S, fouling took place as expected within the porous support layer of membranes in both FO-MBR and PRO-MBR. It is noteworthy that unlike the fouling layer on the 462 surface of support layer, intensive accumulation of Ca and P within support layer of 463 464 membranes was observed from the EDS mapping images, implying that inorganic scaling as a result of the precipitation of Ca and P ions probably took place within 465 support layer (She et al., 2017a). 466





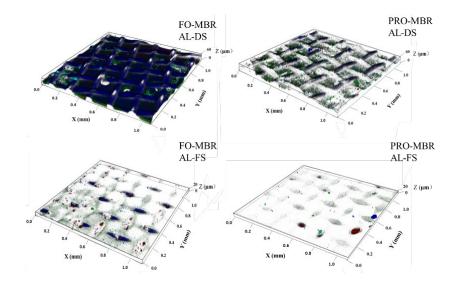
468 Figure 3 SEM images (left) and EDS spectra (right) of the fouled FO membranes in the PRO-

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 $\mu$ m thick); Since the support layer of FO membrane was approximately 30
478	$\mu$ m 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 $\mu m$
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 $\mu m^3/\mu m^2$ and 16.92 $\mu m^3/\mu m^2$ 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 $\mu m^3/\mu m^2$ in PRO-MBR and 4.84 $\mu m^3/\mu m^2$ in FO-MBR. This result
491	further demonstrated that biofouling on membranes fouled in AL-DS orientation was
492	much more significant.



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  $\alpha$ -D-glucopyranose,  $\beta$ -D-

glucopyranose, proteins, and microbial cells, respectively).

496

497

#### 498 Table 3

Biovolume of the foulants on the fouled FO membranes in PRO-MBR and FO-MBR (calculated byPHLIP).<sup>a</sup>

		α-D- glucopyranose (μm <sup>3</sup> /μm <sup>2</sup> )	β-D- glucopyranose (μm <sup>3</sup> /μm <sup>2</sup> )	Proteins (µm <sup>3</sup> /µm <sup>2</sup> )	Total cells (μm <sup>3</sup> /μm <sup>2</sup> )	Sum (µm³/µm²)
PRO-	AL-DS	$0.21\pm0.07$	$7.13\pm0.71$	$6.51\pm0.33$	$3.07\pm0.66$	$16.92\pm1.77$
MBR	AL-FS	$0.88\pm0.14$	$0.63\pm0.06$	$1.06\pm0.21$	$0.72\pm0.08$	$3.29\pm 0.49$
FO-	AL-DS	$2.01\pm0.64$	$11.13\pm1.03$	$9.99\pm0.42$	$7.85\pm0.78$	$30.98 \pm 1.87$
MBR	AL-FS	$2.00\pm0.09$	$1.07\pm0.04$	$1.37\pm0.08$	$0.40\pm0.03$	$4.84\pm0.24$

<sup>a</sup> Values are given as the mean values  $\pm$  standard deviation (number of measurements: n = 3).

Above results collectively indicated that membrane orientation largely determined the fouling behavior in PRO-MBR and FO-MBR. The PRO-MBR, in which membrane was normally operated in AL-DS orientation, suffered more severe and complex membrane fouling, as compared with FO-MBR (membrane normally operated in AL-FS orientation). From another point of view, AL-FS orientation could be a more promising option in the scenario of PRO-MBR if the shortcomings of severe ICP and membrane stability (leading to poor power density and membrane damage) can be well 509 addressed.

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

530 3.6 Fouling reversibility

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

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

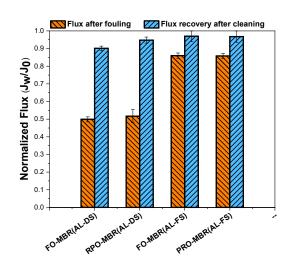


Figure 5 Normalized fluxes of the fouled FO membranes in the PRO-MBR and FO-MBR before and after physical cleaning.

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

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

571 addressed before.

# 572 3.7 Implications

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

The choice of membrane orientation is of paramount importance for PRO-MBR. Present study found that the energy generation efficiency achieved in AL-DS orientation (4.1 kWh/100 m<sup>2</sup>·d) was 28.1% higher than that in AL-FS orientation (3.2

593	kWh/100 $m^2 \cdot d$ ) 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.<sup>a</sup>

		FO-MBR		PRO-MBR	
		AL-DS	AL-FS	AL-DS	AL-FS
Operating flux (LMH)		$13.54 \pm 2.31$	$11.09\pm0.45$	$12.19\pm2.08$	$7.79\pm0.42$
Removal rate (%)	TOC TP NH4 <sup>+</sup> -N TN	$\begin{array}{c} 96.51 \pm 0.51 \\ 100 \\ 97.49 \pm 0.69 \\ 96.51 \pm 0.90 \end{array}$	$\begin{array}{c} 96.66 \pm 0.10 \\ 100 \\ 98.68 \pm 0.62 \\ 95.96 \pm 0.68 \end{array}$	$96.47 \pm 1.10 \\ 100 \\ 98.52 \pm 1.18 \\ 97.29 \pm 1.28$	$97.33 \pm 2.12$ 100 $98.74 \pm 0.26$ $96.86 \pm 0.39$
Flux recovery rate (%)		$90.10\pm1.31$	$97.04\pm3.45$	$94.83 \pm 1.71$	$96.82\pm3.22$
Specific energy cor (kWh/m <sup>3</sup> )	nsumption <sup>b</sup>	1.	427	1.283	1.288
Energy generation efficiency <sup>c</sup> $(kWh/100 m^2 d)$		-	-	4.1	3.2

 $\frac{(kWh/100 \text{ m}^2 \cdot \text{d})}{\text{a Values are given as the mean values } \pm \text{ standard deviation (number of measurements: n = 3).}$ 604

605 606 <sup>b</sup> Energy generated by PRO was also considered.
 <sup>c</sup> 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 m <sup>2</sup> ·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

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

# 622 CRediT authorship contribution statement

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

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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						
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- 821 Table Captions
- **Table 1** TOC, NH<sub>4</sub><sup>+</sup>-N, TN and TP concentrations in the influent, sludge supernatant
- and FO permeate and their removal rates (average  $\pm$  standard deviation from triple
- measurements) in FO-MBR and PRO-MBR.
- **Table 2** The specific energy consumption of FO and PRO.
- **Table 3** Biovolume of the foulants on the fouled FO membranes in PRO-MBR and FO-
- 827 MBR (calculated by PHLIP).<sup>a</sup>
- **Table 4** Performance comparison between the FO-MBR and the PRO-MBR.<sup>a</sup>

- 830 **Figure Captions**
- Figure 1 Water flux profiles in PRO-MBR and FO-MBR with different membrane
- 832 orientations (i.e., AL-DS and AL-FS).
- **Figure 2** Power density profiles of PRO-MBR operated in different membrane

- 834 orientations (i.e., AL-DS and AL-FS).
- **Figure 3** SEM images (left) and EDS spectra (right) of the fouled FO membranes in
- the PRO-MBR and FO-MBR.
- 837 Figure 4 CLSM images of the fouled FO membranes in different membrane
- orientations in the PRO-MBR and FO-MBR (the cyan, blue, green and red colors
- 839 represent  $\alpha$ -D-glucopyranose,  $\beta$ -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.