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1 **A novel forward osmosis reactor assisted with microfiltration for deep thickening**
2 **waste activated sludge: performance and implication**

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27 **Abstract**

28 Waste activated sludge (WAS) treatment has gained growing interests for its
29 increasingly capacity and high process cost. Sludge thickening is generally the first process
30 of the WAS treatment. However, traditional sludge thickening approach was restrained by
31 large footprint, low thickening efficiency, and tendency of releasing phosphorus. Here, we
32 reported a novel microfiltration (MF) membrane assisting forward osmosis (FO) process
33 (MF-FO) for sludge thickening. The MF-FO reactor achieved a sludge thickening of the
34 mixed liquor suspended solids (MLSS) concentration from approximately 7 to 50 g/L after
35 10-day operation. More importantly, the effluent quality after FO filtration was superior
36 with total organic carbon (TOC), ammonia nitrogen ($\text{NH}_4^+\text{-N}$), nitrate nitrogen ($\text{NO}_3^-\text{-N}$)
37 and total phosphorus (TP) of 1.94 ± 0.46 , 0.02 ± 0.07 , 4.55 ± 1.59 and 0.24 ± 0.26 mg/L,
38 respectively. Additionally, the integration of MF membrane successfully controlled the
39 salinity of the MF-FO reactor in a low range of 1.6-3.1 mS/cm, which mitigated the flux
40 decline of FO membrane and thus prolonged the operating time. In this case, the flux
41 decline of FO membrane in the MF-FO reactor was mainly due to the membrane fouling.
42 Furthermore, the fouling layer on the FO membrane surface was a gel layer mainly
43 composed of biofoulants and organic foulants when the MLSS concentration was less than
44 30 g/L, while it turned to a cake layer when the MLSS concentration exceeded 30 g/L.
45 Results reported here demonstrated that the MF-FO reactor is a promising WAS thickening
46 technology for its excellent thickening performance and high effluent quality of FO
47 membrane.

48 **Keywords:** forward osmosis; microfiltration; waste activated sludge; sludge thickening;

49 membrane fouling

50 **1. Introduction**

51 With increase of municipal wastewater capacity and improvement of the wastewater
52 treatment process, waste activated sludge (WAS), a by-product of wastewater treatment, is
53 growing substantially in daily operation (Zhu et al., 2012). It is estimated that the cost of
54 WAS treatment is equivalent to wastewater treatment (Pei et al., 2010; Yuan et al., 2011;
55 Nguyen et al., 2013, 2015; Collard et al., 2017). Accordingly, the treatment and disposal of
56 WAS has aroused growing interests. Sludge thickening is the widely adopted approach to
57 decrease the water content of WAS for achieving the reduction of sludge volume. Currently,
58 the general sludge thickening methods are applied for WAS including gravity thickening,
59 dissolved air flotation (DAF) thickening and centrifugal thickening (Wang et al., 2008a).
60 Taking the gravity thickening process for example, it has some drawbacks such as a large
61 footprint, a low thickening efficiency, a tendency of releasing phosphorus during long
62 sludge retention time (SRT), and emission of unpleasant odors (Wang et al., 2008a; Zhu et
63 al., 2012; Lee et al., 2017). In addition, it should be pointed out that the sludge supernatant
64 with high concentrations of organic matters, nitrogen and phosphorus from the traditional
65 thickening technologies has to be further treated via returning to WWTPs or discharging
66 after post-treatment, which not only enhances the cost of thickening process but also
67 complicates the thickening process.

68 Conventional sludge thickening technologies are low in efficacy and high in energy
69 consumption, thus dedicated studies have devoted to developing new sludge thickening
70 processes. Among these novel processes, applying microfiltration (MF) membrane for

71 sludge thickening is an interesting attempt (Wang et al., 2008a, 2008b, 2009; Wu et al.,
72 2009; Kim et al., 2013), which utilizes the effective separation of water and solid via MF
73 membrane to achieve thickening of WAS. It can effectively reduce the water-content of
74 WAS to about 97%, which is same as the traditional thickening technologies (Wang et al.,
75 2008a, 2008b, 2009). In addition, it possesses a less footprint, a higher solid recovery and a
76 better supernatant water quality compared with the traditional thickening technologies
77 (Kim et al., 2010, 2013). Therefore, the MF process has been considered as a potentially
78 alternative method for WAS thickening.

79 Although the MF process has many advantages over the traditional thickening
80 technologies, it also has some bottlenecks retarding its wide application including serious
81 membrane fouling and membrane permeate needing further treatment. Compared to
82 conventional membrane separation technology, forward osmosis (FO) remains a unique,
83 attractive and emerging technology after it was first proposed decade ago. FO utilizes a
84 draw solution (DS) with a high osmotic pressure to “draw” the water from a feed solution
85 (FS) with a low osmotic pressure through a semi-permeable membrane (She et al., 2016;
86 Wang et al., 2016a). Based on the fact that FO membrane has high rejection and superior
87 water flux stability against fouling (Gu et al., 2013; Li et al., 2019), a novel sludge
88 thickening technology using FO membrane instead of MF membrane has been proposed
89 (Hau et al., 2014; Nguyen et al., 2015, 2016; Lee et al., 2017; Ng et al., 2019; Sun et al.,
90 2019; Zhao et al., 2019). In comparison with the MF process, the FO process had a better
91 effluent quality and a similar thickening efficiency. However, the operation of FO process
92 in most studies does not last for long time (less than 24 hours), and there is lack of

93 evaluating membrane fouling and effluent quality in these reports. Besides, the WAS is
94 only concentrated to about 35 g/L owing to the decline of FO membrane flux. The flux
95 decline of FO membrane is mainly attributed to salt accumulation induced by high MLSS
96 and reverse salt transport (RST) (Zhu et al., 2012). Salt accumulation is a common
97 phenomenon in FO membrane reactor. For instance, many literatures have reported the
98 cause and damage of salt accumulation in osmotic membrane bioreactor (OMBR) for
99 wastewater treatment (Qiu et al., 2013; Wang et al., 2014a, 2016a, 2017a; Yang et al.,
100 2018).

101 Recently, some researchers proposed a novel method to control salt accumulation in
102 the OMBR via combining MF or UF membrane for discharging soluble salt (Wang et al.,
103 2014b; Holloway et al., 2015). Inspiring by the successfully using MF membrane for
104 alleviating salt accumulation in the OMBR, we intend to integrate MF membrane with FO
105 membrane (called MF-FO process). In the MF-FO process, salinity build-up can be
106 effectively controlled by the MF membrane, and thus enlarging the operation time of FO
107 membrane. In this case, a deep thickening of WAS might be achieved in the MF-FO
108 process, which will not only enhance the economic performance of MF-FO process but
109 also be beneficial to follow-up treatment via reducing sludge volume. Although the
110 integration of MF membrane and FO membrane has been widely reported in the OMBR
111 process for wastewater treatment, this is the first attempt on applying the hybrid MF plus
112 FO process for thickening sludge. The prior reported experimental techniques and data in
113 the literatures cannot be justified and directly used in the context of sludge management.
114 The objectives of this study are to concentrate WAS to about 50 g/L via FO membrane with

115 a help of MF membrane for controlling salt accumulation and to further investigate the
116 effluent quality and fouling behavior of FO membrane at a high MLSS condition.

117 **2. Materials and methods**

118 **2.1. Experimental set-up and operating conditions**

119 A laboratory-scale MF-FO reactor with an effective volume of 3.8 L was used in this
120 study (see Fig. 1). The WAS with the MLSS concentration of 4-5 g/L collected from the
121 Wuxi Xincheng WWTPs was directly pumped into the reactor. Both an FO and an MF
122 membrane module (with an effective area of 0.024 m² and 0.032 m², respectively) were
123 immersed in the reactor. The FO membrane made of cellulose triacetate (CTA) (Hydration
124 Technologies Inc., United States) had an orientation of active layer facing the WAS
125 (AL-FS). A NaCl solution with a concentration of 1 M used as the draw solution was
126 recirculated from draw solution tank to FO membrane with a flow rate of 0.4 L/min. In
127 addition, a conductivity controller (OKD-650, Shenzhen OK Instrument Technology Co.,
128 Ltd., China) equipped with a NaCl solution of 5 M was applied for keeping the draw
129 solution concentration constant at 1 M. The MF membrane made of polyvinylidene
130 fluoride (PVDF) (Zizheng Environment Inc., China) with a nominal pore size of 0.20 μm
131 was operated under the mode of stable flux, and its water flux was controlled by a
132 peristaltic pump. In order to alleviate both MF and FO membrane fouling, aeration was
133 introduced with an aeration rate of 200 L/min. In each cycle, the WAS was continuously
134 pumped into the reactor, and the MF and FO membrane modules were continuously
135 operated until the MLSS concentration of thickening sludge reached approximately 50 g/L.
136 After that, the influent WAS and the operation of membrane modules were simultaneously

137 stopped and then the thickening sludge was discharged from the reactor. After simple
138 physical cleaning of the FO membrane and chemical cleaning of MF membrane with 0.1%
139 NaClO, respectively, the reactor started a new cycle. During the whole experiment, the
140 reactor was operated at the temperature of 25 ± 1 °C.

141 Fig. 1

142 **2.2. Analytical methods**

143 Measurements of ammonia nitrogen ($\text{NH}_4^+\text{-N}$), nitrate nitrogen ($\text{NO}_3^-\text{-N}$) and total
144 phosphorus (TP) were conducted by Amver Salicylate Method (HACH 2606945),
145 Persulfate Digestion Method (HACH 2714100/2672245) and Molybdovanadate Method
146 (HACH 2767245), respectively. Total organic carbon (TOC) concentration was determined
147 by a TOC analyzer (TOC- V_{CPH} , Shimadzu, Japan). Mixed liquor volatile suspended solids
148 (MLVSS) and mixed liquor suspended solid (MLSS) were measured according to the
149 Standard methods (Chinese NEPA, 2002). Procedures for soluble microbial products (SMP)
150 and bound extracellular polymer substances (BEPS) extractions have been described in
151 previous studies (Chen et al., 2011; Wang et al., 2012), and the sum of polysaccharides
152 (applied the phenol sulfuric acid method (Zhang et al., 2020)) and proteins (determined by
153 a modified Lowry method (Winters et al., 2005)) was used to represent the concentrations
154 of SMP and BEPS. A field emission scanning electron microscope (FESEM) (SU8010,
155 Hitachi, Japan) was applied for capturing the surface images of the pristine, fouled and
156 cleaned FO membranes. A confocal laser scanning microscope (CLSM, LSM 710, Carl
157 Zeiss, Germany) was used for analyzing the spatial distributions of biofoulants including
158 microorganisms, proteins and polysaccharides on the fouled and cleaned FO membrane

159 samples, and the specific staining method and the fluorescent probes can be found in
160 previous reports (Yuan et al., 2015; Wang et al., 2016b; Liu et al., 2017b).

161 **3. Results and discussion**

162 **3.1. Sludge thickening and reduction**

163 Variations of MLSS and MLVSS concentrations of the thickening sludge during the
164 operation of MF-FO reactor are summarized in Fig. 2. It was observed that MLSS and
165 MLVSS concentrations rapidly increased in both cycles, i.e., the MLSS and MLVSS
166 concentrations reached to more than 50.0 and 20.4 g/L from 6.4 and 2.4 g/L, respectively,
167 after operating 10 days. According to previous literature on sludge thickening via single
168 MF membrane (Wang et al., 2008a, 2008b) and single FO membrane (Zhu et al., 2012), the
169 final MLSS concentration was in the range of 30-40 g/L. The difference in the performance
170 of sludge thickening between the MF-FO reactor and the MF or FO reactor can be
171 attributed to different operating conditions such as membrane area, initial water flux and
172 reactor volume. In brief, the MF-FO reactor successfully achieved a deeper thickening of
173 WAS.

174  Fig. 2

175 In addition, sludge reduction was also occurred during the operation of the MF-FO
176 reactor. Specifically, MLSS and MLVSS sludge reduction efficiency in both cycles was in
177 the range of 5.8%-6.3% and 11.6%-15.8%, respectively. It is noteworthy that sludge
178 digestion generally accompanied the variations of extracellular polymer substances (EPS)
179 in sludge (Wang et al., 2009). To verify this hypothesis, the variations of EPS concentration
180 in the thickening sludge during the operation of the MF-FO reactor are presented in Fig. 3.

181 It was found that the sludge EPS concentration decreased during the process of WAS
182 thickening in both cycles, indicating that the EPS was utilized by the microorganisms due
183 to lack of nutrients (Wang et al., 2009; Zhu et al., 2017).

184 Fig. 3

185 3.2. Effluent water quality

186 Variations of $\text{NH}_4^+\text{-N}$, $\text{NO}_3^-\text{-N}$, TP and TOC concentrations of the influent sludge
187 supernatant, thickening sludge supernatant, FO and MF permeates in the MF-FO reactor
188 are illustrated in Table 1. TOC, $\text{NH}_4^+\text{-N}$, $\text{NO}_3^-\text{-N}$ and TP concentrations of the influent
189 sludge supernatant in both cycles were 10.95 ± 5.72 , 1.22 ± 0.78 , 7.77 ± 2.29 and $2.26 \pm$
190 1.77 mg/L, respectively. As for TOC and TP concentrations, their accumulation was
191 observed in the thickening sludge supernatant. TOC and TP concentrations in the FO
192 membrane permeate were below 3.0 and 1.0 mg/L, respectively, due to the high rejection of
193 FO membrane for organic compounds and phosphate. However, owing to the rejection of
194 MF membrane much worse than that of FO membrane, the TOC and TP concentrations
195 (14.68 ± 13.48 and 0.90 ± 0.73 mg/L, respectively) was higher in the MF membrane
196 permeate. It is interesting to note that there was no $\text{NH}_4^+\text{-N}$ accumulation in the thickening
197 sludge supernatant. It could be attributed to the DO concentration in the range of 1-2 mg/L
198 owing to the aeration for alleviation membrane fouling, thus resulting in the conversation
199 of $\text{NH}_4^+\text{-N}$ to $\text{NO}_3^-\text{-N}$ in the MF-FO reactor, which was evident by the increase of $\text{NO}_3^-\text{-N}$
200 concentration in the thickening sludge supernatant from the initial value of 10.63 ± 0.14 to
201 20.44 ± 0.95 mg/L. As a result, there was no significant difference in the $\text{NH}_4^+\text{-N}$
202 concentration between the MF and FO membrane permeates (below 0.3 and 0.7 mg/L,

203 respectively). However, owing to the different rejection for NO_3^- -N, the NO_3^- -N
204 concentration was approximately 16 mg/L in the MF membrane permeate while it was
205 below 8 mg/L in the FO membrane permeate. In general, the permeate water quality in the
206 MF-FO was excellent and was mainly driven by higher rejection of FO membrane for
207 various contaminants.

208 Table 1

209 3.3. Water flux profile of FO and MF membranes

210 Variations of FO and MF membrane flux during the two cycles are shown in Fig. 4.
211 According to previous reports on alleviating salt accumulation of FO process via MF
212 membrane with water flux in the range of 2-6 LMH (Liu et al., 2017; Wang et al., 2017a;
213 Zhu et al., 2018), an MF flux value of 2.3 LMH was selected for mitigating the salinity
214 build-up in the MF-FO reactor. During the whole operation of the MF-FO reactor, the MF
215 flux was stable in the range of 2.16-2.36 LMH although the TMP of MF membrane
216 increased to approximately 10 kPa in each cycle. According to previous reports on the MF
217 membrane for sludge thickening (Wang et al., 2008a, 2008b; Kim et al., 2010, 2013), the
218 flux of MF membrane was significantly declined from an initial flux of approximately 15
219 LMH due to severe fouling at a high MLSS condition. Thus, the stable water flux of MF
220 membrane in the MF-FO process was owing to a mild membrane fouling operating at a low
221 flux. With the help of the MF membrane, the conductivity of the thickening sludge in the
222 MF-FO reactor was maintained in the range of 1.6-3.1 mS/cm, which is a low salinity
223 environment with no inhibition on microorganisms (Lay et al., 2010; Wang et al., 2014b).

224 The FO membrane was cleaned by the physical method (Wang et al., 2014a, 2017b)

225 after Cycle 1 and then was continued to apply in Cycle 2. As for the water flux of FO
226 membrane, a significant decrease could be observed in both cycles (as shown in Fig. 4),
227 and the corresponding flux decline was from 6.9 to 4.0 LMH and from 5.4 to 2.3 LMH in
228 the two cycles, respectively. According to previous literature (Wang et al., 2014a, 2014b,
229 2016a; Zhu et al., 2018), the flux decline of FO membrane is mainly owing to salinity
230 accumulation within the bioreactor and membrane fouling. As aforementioned, there was
231 no salt accumulation in both cycles of the MF-FO reactor. Thus, the flux decline of FO
232 membrane in the MF-FO reactor was attributed to the membrane fouling. Nevertheless, the
233 water flux of the FO membrane can be restored up to 86% of the pristine FO membrane
234 after simply physical cleaning (shown in Fig. S1). It suggested that the recovery of FO
235 membrane permeability was high in the MF-FO reactor even though the fouling tendency
236 was severe at a high MLSS condition. In addition, the flux decline rate of FO membrane
237 became quicker in Cycle 2 compared to Cycle 1, suggesting that FO membrane fouling was
238 more severe in Cycle 2. It might be attributed to the fact that the irreversible foulants
239 formed on the FO membrane surface in Cycle 1 can not be fully removed by simple
240 physical cleaning, and subsequently the remaining foulants resulted in more foulants
241 accumulating on the FO membrane surface in Cycle 2.

242 Fig. 4

243 **3.4. Evaluation of FO membrane fouling**

244 As mentioned above, a mild fouling of MF membrane was observed in the MF-FO
245 reactor due to operating at a low flux condition, while membrane fouling was the major
246 reason for the flux decline of FO membrane. Additionally, the fouling of FO membrane at a

247 high MLSS concentration especially reaching the MLSS value of approximately 50 g/L
248 was barely reported. Thus, fouling behaviors of FO membrane in the MF-FO reactor were
249 further investigated as follows.

250 During the operation of the MF-FO reactor, it is interesting to note that the MLSS
251 concentration of 30 g/L was an important turning point. When the MLSS concentration was
252 less than 30 g/L, the FO membrane surface was covered with a gel layer. However, a thick
253 cake layer started to develop on the FO membrane surface when the MLSS concentration
254 exceeded 30 g/L. In this case, the FO membrane fouling behaviors in the MF-FO reactor
255 were comprehensively analyzed at the MLSS concentration of 30 and 50 g/L, respectively,
256 in order to better understanding the FO membrane fouling at a high MLSS concentration.
257 Surface morphology of the fouled and physically cleaned FO membranes in the MF-FO
258 reactor is illustrated in Fig. 5. Compared with the pristine FO membrane (Fig. 5(a)), some
259 dispersed pollutants could be observed on the FO membrane surface at the MLSS
260 concentration of 30 g/L from both the visual observation (Fig. 5(b-1)) and the SEM image
261 (Fig. 5(b-2)). As for the fouled FO membrane at the MLSS concentration of 50 g/L, a thick
262 layer of mud cake appeared on its surface (Fig. 5(c-1)), and no grid-like structure of
263 CTA-FO membrane could be found at all from the SEM image (Fig. 5(c-2)). These results
264 indicated that more foulants were deposited on the FO membrane surface at the MLSS
265 concentration of 50 g/L and the fouling was significantly different between the MLSS
266 concentration of 30 and 50 g/L. Furthermore, these fouled FO membrane surfaces were
267 observed after physical cleaning. It could be seen that almost all foulants have been
268 removed from the surface of FO membrane at both MLSS concentrations according to the

269 optical images (Fig. 5(d-1) and (e-1)). However, some foulants still remain on the surface
270 of the FO membrane especially at the MLSS concentration of 50 g/L (Fig. 5(d-2) and (e-2)).
271 It indicated that there was some irreversible foulants, which could not be removed by the
272 merely physical cleaning, and the irreversible fouling would be aggravated as the sludge
273 concentration increased.

274 Fig. 5

275 In order to further understand the composition of the foulants on the FO membrane
276 surface, the reversible and irreversible foulants were collected from the fouled membrane
277 surface (see Section S1), and their quantities were analyzed in terms of TS and VS
278 concentrations. From Table 2, it demonstrated that the amount of reversible foulants was
279 much more than that of irreversible foulants regardless of the MLSS concentration,
280 suggesting that the reversible fouling was the dominant fouling type of FO membrane in
281 the MF-FO reactor (Nguyen et al., 2019). And the quantity of the foulants was approaching
282 1600 g/m^2 at the sludge concentration of 50 g/L, which was ten times more than that at 30
283 g/L. It further demonstrated that the fouling of FO membrane was more severe at the
284 MLSS concentration of 50 g/L. Moreover, all the ratios of VS/TS were more than 0.6,
285 indicating that the organic foulants and biofoulants were dominant foulants of the FO
286 membrane in the MF-FO reactor regardless of the sludge concentration.

287 Table 2

288 Considering the significant contribution of organic fouling and biofouling to the
289 membrane fouling in the MF-FO reactor, the typical organic foulants and biofoulants
290 including proteins, polysaccharides and microorganisms on the FO membrane at different

291 sludge concentrations were further investigated by the CLSM coupled with multiple
292 fluorescence labeling. Unfortunately, the distribution of organic foulants and biofoulants on
293 the surface of the FO membrane at sludge concentration of 50 g/L was unable to observe
294 by the CLSM because the cake layer was more than 3 mm (see Fig. S2), which was too
295 thick for the dye to penetrate and the laser to break through. However, the fouled FO
296 membrane at MLSS concentration of 30 g/L could be observed by the CLSM owing to its
297 thinner fouling layer. Its thickness of the fouling layer was approximately 30 μm (see Fig.
298 6), which was even much thinner than other fouling layers formed on FO membranes in the
299 OMBRs (Yuan et al., 2015; Wang et al., 2016b; Zhu et al., 2018). It was interesting to note
300 that the proteins and polysaccharides were the major foulants while the microorganisms
301 could only be observed in small area, suggesting that EPS secreted by the microorganisms,
302 rather than the microorganisms themselves, significantly affected the fouling behaviors of
303 the FO membrane even at such a high sludge concentration. Compare to other reports on
304 the OMBRs at a relatively low MLSS concentration (Yuan et al., 2015; Wang et al., 2016b;
305 Zhu et al., 2018), the distributions of β -D-glucopyranose polysaccharides and proteins were
306 more dispersed on the FO membrane surface in the MF-FO reactor, which could be
307 attributed to the lower concentration of EPS (Yuan et al., 2015; Wang et al., 2016b).

308 Fig. 6

309 **3.5. Implications**

310 The MF-FO process simultaneously integrates the FO and MF membranes in a single
311 reactor for deep thickening the WAS. Compared with the MF process only containing the
312 MF membrane, the MF-FO process achieved a better thickening result, i.e., the thickening

313 sludge concentration was enhanced from approximately 30 g/L to 50 g/L, and the effluent
314 quality of FO membrane permeate was much better in the MF-FO process. Additionally,
315 the MF membrane has a mild fouling in the MF-FO process owing to applying a lower flux.
316 Compared with the FO process only having the FO membrane, the water flux of FO
317 membrane dropped more slowly because the salinity in the MF-FO reactor was controlled
318 at a low range with the help of the MF membrane. The better flux performance of FO
319 membrane resulted in a longer operation time of the MF-FO process and thus a higher
320 thickening sludge concentration. Based on the above facts, the MF-FO process exhibited
321 huge potential in sludge thickening.

322 In a wastewater treatment plant, the MF-FO process can be directly used as a sludge
323 treatment unit instead of the traditional sludge thickening process. However, the MF-FO
324 process still has some limitations that need to be overcome before it becomes an industrial
325 process. For instance, the MF membrane permeate needs to be further treatment for
326 meeting with the discharge or reuse standard, and the high energy consumption of aeration
327 for mitigating membrane fouling should be reduced. In addition, a certain level of sludge
328 digestion was observed in the MF-FO process due to the microaerobic environment
329 induced by the aeration. It implied that a simultaneous thickening and digestion of WAS
330 might be achieved in the MF-FO process via adjusting the DO concentration or the
331 retention time of WAS.

332 **4. Conclusion**

333 After operating 10 days of the MF-FO reactor, the MLSS concentration reached about
334 50 g/L from the initial concentration of about 7 g/L. Owing to the high rejection of FO

335 membrane, an excellent water quality of FO membrane permeate was obtained, i.e., the
336 TOC, $\text{NH}_4^+\text{-N}$, $\text{NO}_3^-\text{-N}$ and TP concentrations were 1.94 ± 0.46 , 0.02 ± 0.07 , 4.55 ± 1.59
337 and 0.24 ± 0.26 mg/L, respectively. However, the water quality of MF membrane permeate
338 was worse than the FO permeate and needed to be further treatment for meeting with the
339 discharge standard. The integration of MF membrane successfully maintained the salinity
340 of the MF-FO reactor in a low range of 1.6-3.1 mS/cm, which mitigated the flux decline of
341 FO membrane and thus prolonged system operating time. The flux decline of FO
342 membrane was mainly due to the membrane fouling, particularly the reversible fouling
343 within the MF-FO reactor. In addition, the MLSS concentration had a significant influence
344 on the fouling mechanisms, whereby the fouling layer was a gel layer mainly composed of
345 biofoulants and organic foulants when the MLSS concentration was less than 30 g/L while
346 it turned to a cake layer when the MLSS concentration exceeded 30 g/L.

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351 of Water Treatment.

352 **Appendix A. Supplementary information**

353 Detailed information on additional figures and foulants extracting methods can be
354 found in the Supporting Information.

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490 **Table Captions**

491 Table 1 Concentrations of $\text{NH}_4^+\text{-N}$, $\text{NO}_3^-\text{-N}$, TP and TOC in the influent sludge supernatant,
492 thickening sludge supernatant, FO and MF permeates in the MF-FO reactor^a.

493 Table 2 Analyses of the foulants on the surface of the FO membrane in the MF-FO reactor.

494 **Figure Captions**

495 Fig. 1. Schematic diagram of the MF-FO reactor.

496 Fig. 2. Changes of MLSS and MLVSS concentrations of the thickening sludge during the
497 operation of MF-FO reactor.

498 Fig. 3. Variations of SMP and BEPS concentrations in the thickening sludge during the
499 operation of MF-FO reactor.

500 Fig. 4. Flux variations of both FO and MF membranes in the MF-FO reactor.

501 Fig. 5. Optical (1) and SEM (2) images of FO membrane surfaces in the MF-FO reactor: (a)
502 of the pristine membrane; (b) and (c) of the fouled membranes at MLSS concentration of
503 30 and 50 g/L, respectively; (d) and (e) of physical cleaned membranes at MLSS
504 concentration of 30 and 50 g/L, respectively. Scale bars in all SEM images correspond to
505 500 μm .

506 Fig. 6. CLSM images of the fouled FO membrane in the MF-FO reactor: (a) total cells; (b)
507 proteins; (c) $\alpha\text{-D-glucopyranose polysaccharides}$; (d) $\beta\text{-D-glucopyranose polysaccharides}$;
508 (e) all foulants.