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Physical cleaning techniques to control fouling during the pre-concentration of high suspended-solid content solutions for resource recovery by forward osmosis

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Physical cleaning techniques to control fouling during the pre-concentration of high suspended-solid content solutions for resource recovery by forward osmosis

Abstract

The fouling propensity of digested sludge centrate, and the effectiveness of membrane flushing, air-scouring, and ultrasonication for physical cleaning were systematically evaluated. Accelerated fouling conditions were applied to simulate the long-term and intensive pre-concentration scenario that is required for phosphorus recovery from digested sludge centrate. The results suggest that membrane fouling during forward osmosis operation to pre-concentrate digested sludge centrate is mostly due to the deposition of small mineral crystals and particulate matter on the membrane surface. Both high cross-flow velocity flushing and ultrasonication were effective at preventing membrane fouling under accelerated fouling conditions. Our results also highlight the potential of intermittent membrane cleaning for achieving a higher cumulative permeate volume and lower energy consumption in comparison to continuous application to prevent membrane fouling. Among several physical cleaning regimes investigated in this study, the combination of ultrasonication and high cross-flow velocity flushing was the most effective and could maintain stable FO operation over several consecutive cleaning cycles.

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1 **Physical cleaning techniques to control fouling during the pre-concentration of high**
2 **suspended-solid content solutions for resource recovery by forward osmosis**

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4 **Desalination**

5
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26 comparison to continuous application to prevent membrane fouling. Among several physical
27 cleaning regimes investigated in this study, the combination of ultrasonication and high
28 cross-flow velocity flushing was the most effective and could maintain stable FO operation
29 over several repetitive cleaning cycles.

30 **Keywords:** forward osmosis (FO); membrane fouling; physical cleaning; ultrasonication;
31 phosphorus recovery; sludge centrate.

32

33 **1. Introduction**

34 Phosphorus is an essential fertilizer ingredient. As the supply of fossil phosphorus is
35 dwindling, the need to develop an alternative and renewable source of phosphorus has
36 emerged as a significant challenge of our time [1-4]. The expected shortage of phosphorus is
37 an imminent threat to all agricultural and industrial processes that rely on this valuable
38 element [5, 6]. Comprehensive analyses of global phosphorus flows have identified
39 wastewater discharge as a dominant pathway of non-diffuse phosphorus losses. Thus,
40 phosphorus recovery from wastewater is a promising source of this important element [7, 8].
41 In addition to the future concern of phosphorus depletion, phosphorus recovery from
42 wastewater can minimise the risk of struvite scaling on wastewater treatment equipment [9,
43 10] and prevent the discharge of nutrient that may cause eutrophication in natural waterways
44 [11-13].

45 Several approaches have been developed to recover phosphorus from wastewater. They differ
46 in regards to the source water and the method used to pre-concentrate phosphate. Source
47 waters include urine [14], raw wastewater [15-17], treated effluent [18, 19], sludge [20], and
48 digested sludge centrate (i.e. anaerobic supernatant) [21-23]. Among these source waters,
49 digested sludge centrate is an important target for phosphorus recovery because it is small in
50 volume but rich in phosphorus and readily available at any large scale wastewater treatment
51 plant [21-23]. The efficiency of phosphorus recovery, generally as struvite
52 ($\text{MgNH}_4\text{PO}_4 \cdot 6\text{H}_2\text{O}$) [10] can be enhanced by pre-concentrating phosphate prior to chemical
53 precipitation. A novel membrane filtration process with significant potential for pre-
54 concentrating phosphate for subsequent recovery is forward osmosis (FO). As a high
55 rejection membrane process, FO can effectively retain and enrich the phosphate and some of
56 the ammonia in digested sludge centrate for subsequent recovery [24-26]. Furthermore, the
57 bidirectional diffusion of protons from the feed solution into the draw solution [27] increases
58 the digested sludge centrate pH and provides a more favourable alkaline environment for
59 chemical phosphorus recovery [21, 22].

60 FO can be used to extract clean water from difficult and complex waste streams that could
61 not be processed by other conventional filtration processes. Previous studies have
62 demonstrated the low fouling propensity of FO compared with its pressure driven
63 counterparts such as reverse osmosis (RO) [28-30]. More importantly, FO membrane fouling
64 appears to be reversible [28-30]. Indeed, several lab and pilot scale tests of FO membranes

65 for the treatment of highly complex waste streams including fracking fluid [31, 32], drilling
66 mud [33], landfill leachate [34], and anaerobically digested sludge centrate [21, 22] have
67 been reported. In particular, our recent investigations [21, 22] have highlighted the challenge
68 of controlling fouling during the pre-concentration of the high suspended solid content sludge
69 centrate solution. Nevertheless, no previous studies have comprehensively evaluated the FO
70 process for a high water recovery (>80%) from digested sludge centrate that is necessary to
71 achieve viable phosphorus recovery [35]. Thus, techniques to mitigate and control fouling are
72 essential for realising the full potential of FO for high suspended solids waste streams, such
73 as digested sludge centrate [36, 37].

74 FO membrane fouling can be controlled via either a physical or chemical cleaning process
75 [38, 39]. Physical cleaning techniques such as cross-flow velocity increase or pulsated cross-
76 flow, membrane flushing, air-scouring, osmotic backwashing, and ultrasonication have been
77 studied for different applications and FO configurations [40-43]. These techniques provide
78 vigorous hydrodynamic conditions to prevent or remove the fouling cake layer from the
79 membrane surface [30, 40]. FO membrane fouling during the pre-concentration of sludge
80 centrate is expected to occur rapidly but also be readily reversible. Thus, although chemical
81 cleaning can be much more effective than physical cleaning [44, 45], it is not compatible with
82 the high cleaning frequency necessary for pre-concentrating sludge centrate for subsequent
83 phosphorus recovery. In this context, ultrasonication is a promising technique to complement
84 other physical cleaning techniques. Indeed, the potential of ultrasonication as a robust but
85 chemical free FO cleaning technique has recently been demonstrated for calcium sulfate
86 scaling [43] and supernatant from waste activated sludge thickening [42].

87 Previous investigations have demonstrated the capability of FO to effectively retain thus pre-
88 concentrate phosphate in the sludge centrate by more than five times [21, 22] to further
89 enhance the economic viability of phosphorus recovery. Preliminary results from these
90 investigations on fouling assessment also highlight the need to develop an effective
91 membrane cleaning strategy to counteract the rapid but potentially more reversible fouling
92 during the pre-concentration of sludge centrate by FO.

93 This study evaluates the propensity and characteristics of FO membrane fouling for
94 phosphorus recovery applications. Accelerated fouling conditions are applied to represent the
95 long-term and intensive concentration scenario that is required for phosphorus recovery from

96 anaerobically digested sludge centrate. We evaluated three physical membrane fouling
97 control techniques, namely, membrane flushing, air-scouring, and ultrasonication in terms of
98 fouling prevention and water flux recoverability.

99 **2. Materials and methods**

100 2.1 Materials and chemicals

101 The cellulose triacetate FO membrane was from Hydration Technologies, Inc. (Albany,
102 Oregon, USA). Analytical grade NaCl was used as the draw solute at a concentration of 3 M.
103 Wastewater was obtained after primary sedimentation from the Wollongong Water Recycling
104 Plant (New South Wales, Australia). The sludge centrate was obtained from a digested sludge
105 dewatering centrifuge from the same plant.

106 2.2 Forward osmosis system

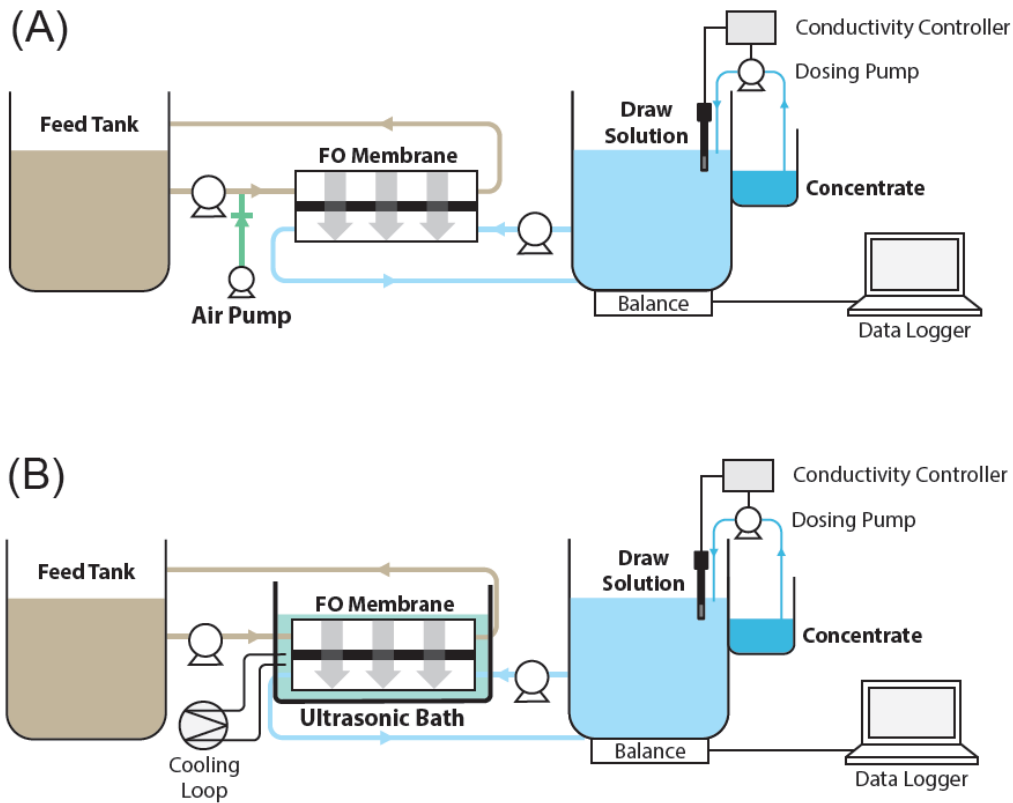
107 A lab-scale, cross-flow FO system was employed in this study. The cell was constructed of
108 two symmetric flow channels with length, width, and height dimensions of 100 mm, 50 mm,
109 and 3 mm, respectively, and an effective membrane area of 50 cm². Circulation of the feed
110 and draw solutions through the cell flow channels was achieved by two variable speed gear
111 pumps (Micropump, Vancouver, Washington, USA). The circulation flow rate was regulated
112 using two rotameters, and pump speed was adjusted to achieve the desired cross-flow
113 velocity. For all experiments, a spacer was positioned on the draw solution side of the
114 membrane cell to improve draw solution mixing. The flat-sheet membrane was sandwiched
115 between two rubber gaskets and the two perspex semi-cells. The feed solution was circulated
116 along the top semi-cell unless otherwise stated.

117 Permeate water flux was determined by recording the weight changes of the draw solution
118 tank using a digital balance (Mettler-Toledo Inc., Hightstown, New Jersey, USA) at two
119 minute intervals. Calculation of water flux was performed according to a standard procedure
120 described elsewhere [46]. All experiments were conducted using a constant 3 M NaCl draw
121 solution. The draw solution concentration (therefore osmotic pressure) was maintained
122 constant using a conductivity controlled pump, which dosed a highly concentrated stock
123 solution (5 M) of NaCl into the draw solution. Conductivity was continuously measured
124 using a conductivity probe (Cole-Parmer, Vernon Hills, Illinois, USA), and was connected to
125 a controller and a peristaltic pump to regulate the concentration of the draw solution (control

126 accuracy of ± 0.1 mS/cm). The temperature of the system was maintained at 21 °C using a
127 chiller and heater during all experiments (Neslab RTE 7, Thermo Scientific, Waltham, MA).

128 2.3 Physical cleaning

129 Three fouling control techniques were evaluated in this study. They include in-situ flushing,
130 air-scouring, and ultrasonication. In-situ flushing was achieved by increasing the circulation
131 flow rates of the feed and draw solutions. The schematics of the air-scouring and
132 ultrasonication cleaning equipment, and their assimilation with the FO system are shown in
133 Figure 1. Each fouling control technique was applied separately, either continuously for
134 fouling prevention or intermittently for membrane cleaning. The former does not interrupt the
135 FO process. The latter requires a brief suspension of the FO process for foulant removal using
136 clean water.



137

138

139 **Figure 1:** Schematic representation of an FO system with (A) air-scouring and (B)
140 ultrasonication cleaning equipment.

141 For in-situ flushing, the pump circulation flow rate was adjusted to increase the rate of cross-
142 flow velocity flushing (i.e. five times the baseline cross-flow velocity). Air-scouring was

143 achieved by connecting an air pump (Aqua One, Australia) inline to the cross-flow membrane
144 cell entry tube, via a one way valve (Figure 1A). The air supply rate was adjusted to achieve a
145 uniform mixture of water and air (approximately 3 L/min). For ultrasonic application, the
146 membrane cell was immersed inside a low frequency (i.e. 30 kHz) ultrasonic water bath
147 (ECO-CT, Ultrasonics Eco, Queensland, Australia) (Figure 1B). The gaskets and tight screws
148 of the membrane cell prevented leakage of liquid from the water bath (i.e. DI water) into the
149 membrane cell flow channels and was verified by clear water testing. The temperature of the
150 ultrasonic bath was maintained at 21 °C using a cooling loop. The cooling loop consisted of a
151 separate reservoir with a submerged stainless steel heat-exchanging coil connected to a chiller
152 (SC200-PC, Aqua Cooler, Sydney, Australia), and a peristaltic pump to circulate liquid
153 between the water bath and cooling reservoir.

154 2.4 Accelerated fouling experimental protocol

155 Accelerated fouling conditions were implemented by applying a high draw solution
156 concentration to maximise water flux and therefore increase the rate of membrane fouling.
157 The circulation flow rate for all reference experiments (i.e. without applying physical
158 cleaning) was 0.5 L/min (corresponding to a cross-flow velocity of 8.3 cm/s). An analytical
159 grade NaCl solution of 3 M was used as the draw solution and this concentration was kept
160 constant throughout the experiment using an automated control system [47]. A preliminary
161 experiment using a synthetic solution with similar background electrolytes to the sludge
162 concentrate was also conducted. The water flux was constant over the entire experiment of 12
163 hours suggesting that the increase in osmotic pressure of the feed was insignificant. Since the
164 draw solution concentration was constant and the increase in the feed osmotic pressure was
165 insignificant, any observable flux decline in this study can be solely attributed to membrane
166 fouling.

167 All experiments were performed with the membrane oriented in FO mode (i.e. active layer
168 facing the feed solution) and in a counter-current flow arrangement. The feed solution
169 volume was 1.5 L and the initial draw solution volume was 1 L.

170 2.5 Physical cleaning

171 The three fouling control techniques described in section 2.3 were applied either continuously
172 for membrane fouling prevention or intermittently for membrane cleaning. For membrane
173 fouling prevention, these techniques were continuously applied during the entire accelerated

174 fouling cycle. The water flux obtained was then compared with the reference condition (i.e.
175 circulation flow rate of 0.5 L/min, corresponding to a cross-flow velocity of 8.3 cm/s).

176 For membrane cleaning, an accelerated membrane fouling experiment was first conducted.
177 After each fouling cycle (approximately five hours) the membrane was cleaned for 30
178 minutes in-situ using one or a combination of these techniques with DI water as the carrier
179 fluid. After cleaning, flux recoverability was determined by replenishing the feed solution
180 with fresh digested sludge centrate. High cross-flow flushing was achieved by increasing the
181 circulation flow rate by fivefold (i.e. 42 cm/s), whilst the other cleaning techniques were
182 analysed at the reference flow rate for comparison. Repetitive membrane cleaning was
183 performed by operating consecutive four hour accelerated fouling cycles. At the conclusion
184 of each cleaning cycle, the feed solution was replaced with fresh sludge centrate.

185 2.6 Membrane autopsy

186 Scanning electron microscopy (SEM) coupled with energy dispersive spectroscopy (EDS)
187 (JCM-6000, JEOL, Tokyo, Japan) was used to identify the fouling layer morphology and
188 composition. The membrane samples were firstly air-dried in a desiccator and then coated
189 with an ultra-thin gold layer with a sputter coater (SPI Module, West Chester, PA).

190 2.7 Analytical methods

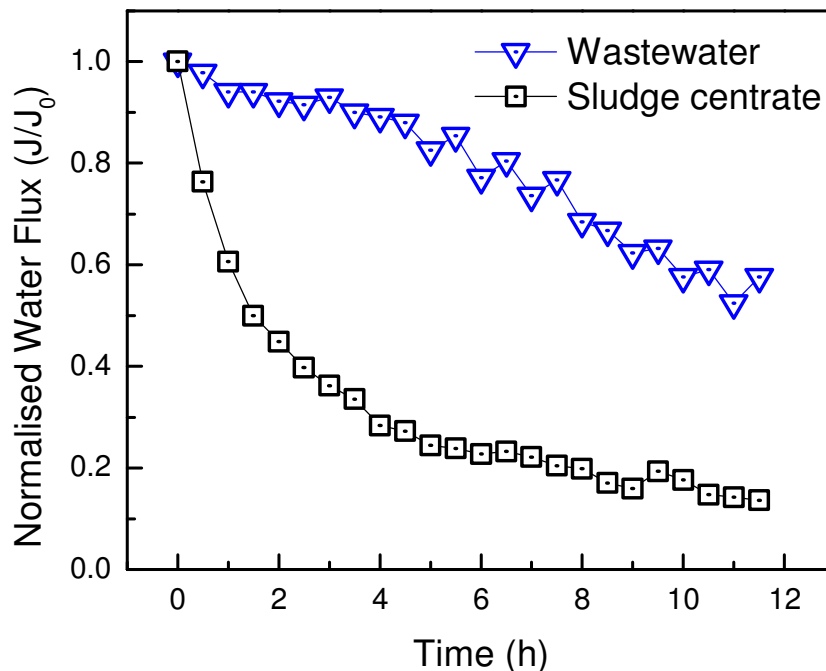
191 The water quality parameters of the wastewater and primary effluent were measured
192 following standard procedures. Total organic carbon (TOC) was analysed using a Shimadzu
193 analyser (TOC-V_{CSH}) and key ions were analysed using an inductively coupled plasma –
194 optical emission spectroscopy (ICP-OES) system (ICP-OES 710, Agilent, Australia). The
195 temperature, pH, and electrical conductivity were monitored using an Orion 4-Star
196 pH/conductivity meter (Thermo Scientific, Waltham, MA).

197 **3. Results and discussion**

198 3.1 Fouling propensity of wastewater and digested sludge centrate

199 The fouling propensity of raw wastewater and digested sludge centrate was evaluated by
200 performing FO filtration experiments under accelerated fouling conditions (Figure 2). As
201 noted in section 2.4, water flux decline can be solely attributed to membrane fouling since the
202 draw solution was maintained at 3 M NaCl and osmotic pressure increase in the feed solution
203 was negligible. For raw wastewater, the water flux gradually declined by approximately 42%

204 of its initial value after 12 hours of operation. On the other hand, digested sludge centrate
 205 showed a more severe fouling behaviour, with a sharp initial decrease and total water flux
 206 decline of 86% after 12 hours. Under these accelerated fouling conditions, water recoveries
 207 from raw wastewater and sludge centrate were approximately 50 and 21%, respectively.
 208 Compared to digested sludge centrate, the observed water flux decline when raw wastewater
 209 was pre-concentrated was less significant. Thus, sludge centrate was used in all subsequent
 210 experiments to evaluate the effectiveness of physical cleaning.



211
 212 **Figure 2:** Comparison of wastewater and digested sludge centrate fouling propensity.
 213 Fouling propensity is represented as the observed water flux decline during accelerated
 214 fouling conditions. Initial water flux of wastewater and digested sludge centrate was $20.0 \pm$
 215 $0.5 \text{ L/m}^2\text{h}$. Accelerated fouling conditions: feed solution was either wastewater or digested
 216 sludge centrate; NaCl draw solution was maintained at 3 M; cross-flow rates of both the feed
 217 and draw solutions were 0.5 L/min (corresponding to a cross-flow velocity of 8.3 cm/s).

218 The high fouling propensity of sludge centrate can be attributed to its very high solids (i.e.
 219 1.16 g/L) and mineral content (i.e. calcium and magnesium) as can be seen in Table 1. For
 220 sludge centrate, during the first two hours of FO filtration, the water flux declined rapidly,
 221 due to the significant deposition of solid particles on the membranes surface. After this point,
 222 the rate of water flux decline was much smaller. The flux profile in Figure 2 suggests that
 223 rapid cake layer formation was the prevalent cause of FO membrane fouling. The formation
 224 of a cake layer on the membrane surface can result in severe cake-enhanced concentration
 225 polarisation, thus, reducing the effective osmotic driving force. It is noteworthy that major

226 constituents in the sludge centrate including phosphate, ammonia and dissolved organics can
 227 be effectively retained by the FO process (Table 1). This attribute is essential for subsequent
 228 resource (phosphorus in this example) recovery but can also aggravate the cake-enhanced
 229 concentration polarisation phenomenon [35].

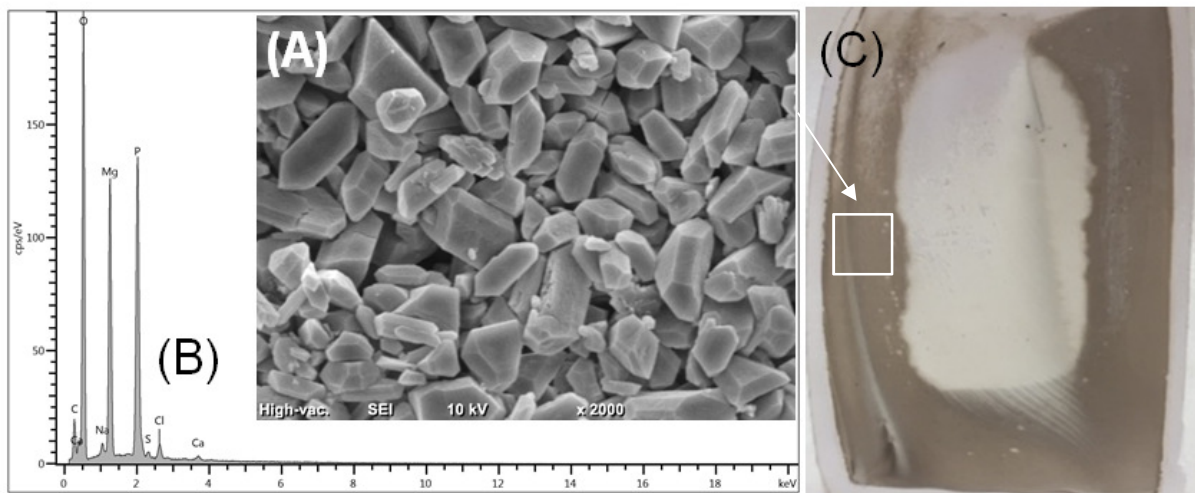
230 **Table 1:** Characteristics of raw wastewater and digested sludge centrate (average
 231 concentration \pm standard deviation from triplicate measurements). The minimum FO
 232 rejection was calculated based on experimental data from our previous study [22].
 233

Parameter	Units	Raw wastewater	Sludge centrate	Sludge centrate - Minimum FO rejection (%)
Total solids	g/L	0.64 \pm 0.03	1.16 \pm 0.03	-
Volatile solids	g/L	0.40 \pm 0.02	0.58 \pm 0.12	-
Electrical conductivity	mS/cm	1.45 \pm 0.24	5.99 \pm 0.11	-
pH	-	6.85 \pm 0.10	7.77 \pm 0.05	-
Total organic carbon	mg/L	45 \pm 10	602 \pm 16	94.3
Total nitrogen	mg/L	41 \pm 9	764 \pm 25	67.6
PO ₄ ³⁻ -P	mg/L	23 \pm 5	97 \pm 7	98.6
NH ₄ ⁺ -N	mg/L	71 \pm 12	521 \pm 22	88.3
Ca ²⁺	mg/L	-	63 \pm 5	-
Mg ²⁺	mg/L	-	14 \pm 5	-
K ⁺	mg/L	-	106 \pm 3	-

234 3.1.1 Digested sludge centrate fouling characterisation

235 Representative morphology and composition of the sludge centrate fouling layer are shown in
 236 Figure 3. The presence of irregular sized crystals suggests the dominance of inorganic
 237 membrane fouling (Figure 3A). Elementary analysis results indicated that the crystals
 238 predominantly contained carbon, oxygen, magnesium, phosphorus, and calcium (Figure 3B).
 239 Some crystals resembled an orthorhombic like shape typical of struvite, however, the
 240 presence of calcium and organic matter in solution was likely to influence the crystal size,
 241 shape, and purity. Interestingly, visual observation of the fouling layer on the membrane
 242 coupon revealed a white flaky precipitate layer at the centre and a brown area at the edge of
 243 the membrane coupon (Figure 3C). The presence of these two distinctive fouling areas is
 244 likely due to the hydraulic profile within the membrane cell. In other words, the brown
 245 sections indicate areas where suspended organic solids were more likely to accumulate.
 246 Nevertheless, detailed examination by SEM analysis revealed no discernible difference in the
 247 morphology and composition of these two areas.

248 The observed crystal morphology and the rapid flux decline shown in Figure 2, suggest that
249 bulk crystallization of minerals occurred in the digested sludge feed solution, followed by
250 particle deposition on the membrane surface [48]. However, it is noted that under the
251 accelerated fouling condition in this experiment, the water recovery was only 21%. Thus, the
252 deposition of more mineral crystals would be expected at higher water recoveries. As
253 previously mentioned, in phosphorus recovery applications, a high concentration factor is
254 necessary to improve process performance (i.e. phosphorus precipitation kinetics) and
255 economics (i.e. chemical consumption) [21, 22].

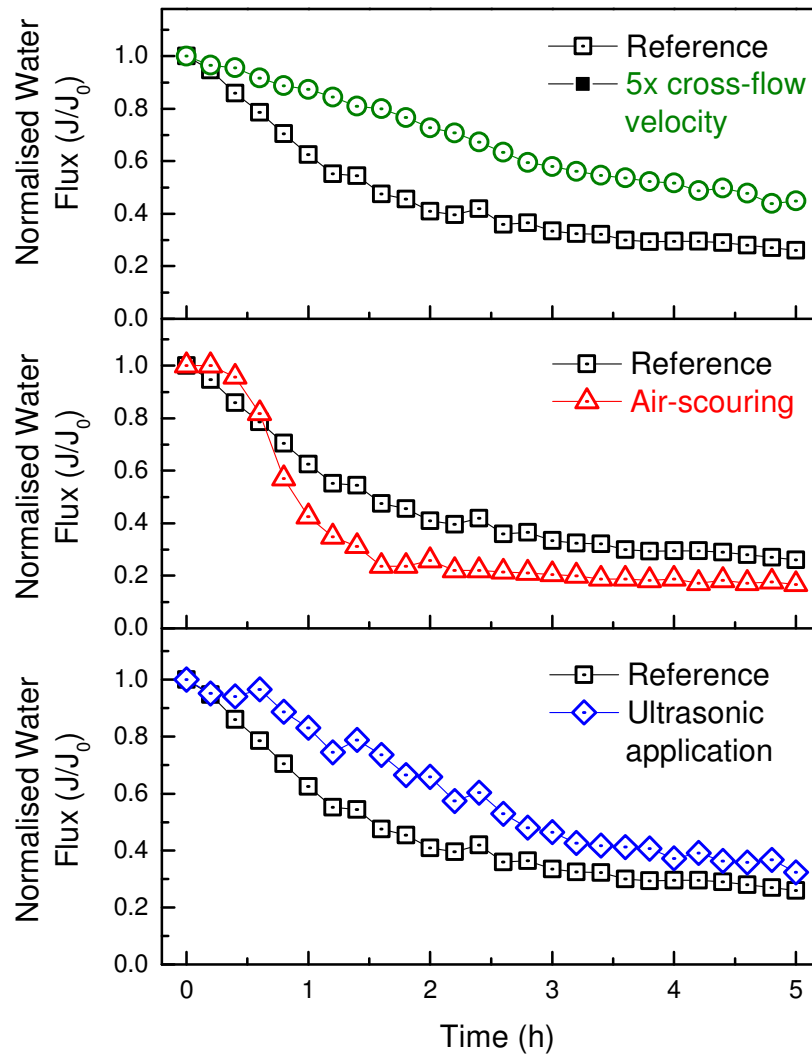


256

257 **Figure 3:** (A) SEM micrograph and (B) EDS spectra of the FO membrane surface at the
258 conclusion of the accelerated fouling experiment using digested sludge centrate as the feed
259 solution. Experimental conditions are described in Figure 2.

260 3.2 Membrane fouling prevention

261 Three fouling prevention techniques were evaluated during the pre-concentration of digested
262 sludge centrate using FO. These prevention techniques were continuously applied during the
263 accelerated fouling cycle and each presented a unique effect on water flux decline compared
264 to the reference flux decline (i.e. when no prevention technique was applied) (Figure 4).



265 **Figure 4:** Normalised water flux decline during accelerated fouling conditions with; (A) 5x
 266 cross-flow velocity (i.e. 42 cm/s), (B) Air-scouring, and (C) ultrasonic application, applied as
 267 fouling prevention techniques. Prevention techniques were continuously applied during the
 268 filtration time. Reference condition represents fouling cycle under accelerated fouling
 269 conditions. Accelerated fouling conditions: feed solution was digested sludge centrate; NaCl
 270 draw solution was maintained at 3 M; cross-flow rates of both the feed and draw solutions
 271 were 0.5 L/min (corresponding to a cross-flow velocity of 8.3 cm/s).
 272

273

274 Operating at a high cross-flow velocity (i.e. 42 cm/s or five times the reference cross flow
 275 velocity of 8.3 cm/s) and ultrasonic application effectively slowed the rate of water flux
 276 decline (Figure 4). Similarly, constant ultrasonic application reduced the severity of water
 277 flux decline compared to the reference. Increasing the cross-flow velocity is a proven
 278 technique to improve the hydrodynamic conditions close to the membranes surface as
 279 turbulence and shear force can prevent foulant accumulation [40]. On the other hand, the

280 observed benefit of applying ultrasonication was possibly due to the combined effects of
281 induced cavitation and the agitation of foulants near the membrane surface [49]. Ultrasonic
282 application also reduced the extent of concentration polarisation by rapidly mixing both the
283 feed and draw solutions close to the membrane surface, and thus improving the water flux
284 dynamics [50]. Our results are consistent with previous studies on membrane cleaning using
285 ultrasonication [42, 43, 51].

286 In contrast, air-scouring had a negative effect during the five hour fouling cycle. Water flux
287 decline during continuous air-scouring was more severe than the reference condition. Within
288 the first 30 minutes, water flux did not decline dramatically. However, after the first 30
289 minutes, water flux drastically declined as air bubbles appeared to compress the fouling layer
290 within the narrow membrane feed channel of the cross-flow module. The presence of air
291 bubbles along the membrane surface may also reduce the available surface area (where the
292 feed solution is in contact with the membrane for mass transfer), thus, limiting the rate of
293 water permeation through the membrane. This effect was verified by performing the
294 experiment with the feed active layer facing up and facing downwards in the membrane cell.
295 Negligible differences in water flux decline were observed between the two configurations
296 (data not shown). Air-scouring as a fouling prevention technique is generally a successful
297 option in membrane bioreactor applications [52]. Our results suggest that module
298 configuration is an essential parameter to consider when applying air-scouring, alongside
299 aeration intensity, optimum bubble size and membrane contact [53]. Applying air-scouring
300 for membrane fouling prevention is expected to be more viable in a submerged membrane
301 configuration.

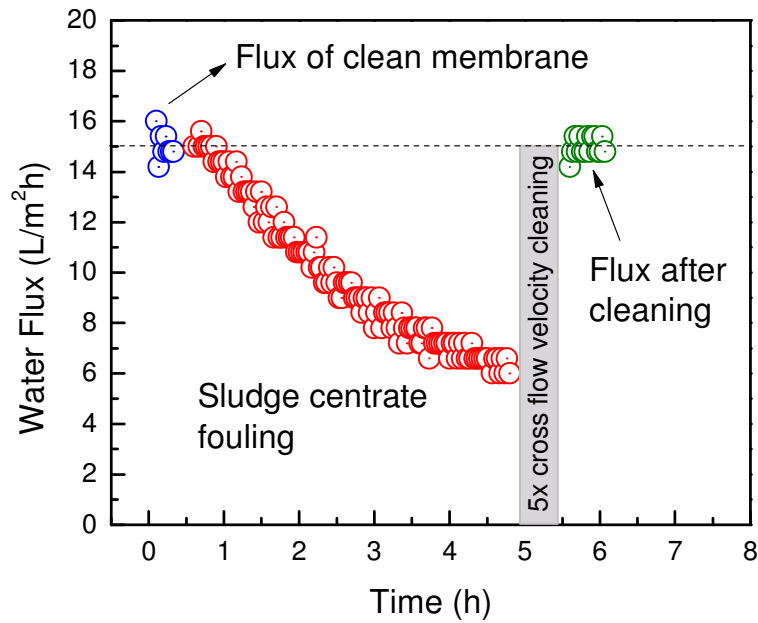
302 Increasing the cross-flow velocity during filtration cycles was the most effective strategy
303 amongst the three techniques investigated here. This achieved the highest cumulative
304 permeate volume during the five hour cycle corresponding the lowest water flux decline.
305 Variations in the cross-flow velocity rate are expected to be proportional to the water flux
306 behaviour, however, this would correspondingly influence the systems energy consumption.
307 Costs associated with circulation can be significant for FO membrane systems [54] and
308 therefore optimisation of membrane fouling prevention techniques is important for a
309 sustainable system. A similar argument can be said for ultrasonication, as continuous
310 application would not be feasible due to the extensive energy consumption required.

311 3.3 Membrane cleaning

312 3.3.1 Influence of repetitive high-cross flow velocity flushing

313 The promising results of high cross-flow velocity and ultrasonication were further
314 investigated for membrane cleaning. At the conclusion of each accelerated fouling
315 experiment, in-situ high cross-flow velocity flushing with DI water could restore the water
316 flux to the initial value (Figure 5). In comparison to the results in Figure 4A, these results
317 (Figure 5) show that applying membrane cleaning is more effective than solely implementing
318 fouling prevention over the five hour period. During the 30 minute cleaning period, foulants
319 on the membrane surface were dislodged and removed from the membrane surface.
320 Furthermore, since the feed and draw solutions were replaced with DI water, there was no
321 water permeation during membrane cleaning. This relaxation period improved the
322 effectiveness of high-cross flow velocity induced shearing on the fouling layer. Since
323 membrane cleaning can be as short as 30 mins, this approach results in a lower energy
324 requirement and only a brief suspension of the filtration process compared to continuous
325 operation at a high cross flow velocity.

326 There was evidence that high-cross flow velocity flushing could not completely remove all
327 solid particles from the membrane surface. Thus, it was not sustainable over multiple cycles
328 of repetitive cleaning during accelerated digested sludge centrate fouling (Figure 6). At the
329 conclusion of each cleaning cycle, the feed solution was replaced with fresh sludge centrate
330 and a graduate flux decline was observed after several consecutive cleaning cycles. These
331 results indicate that the effectiveness of high-cross velocity cleaning is dependent on cleaning
332 frequency.

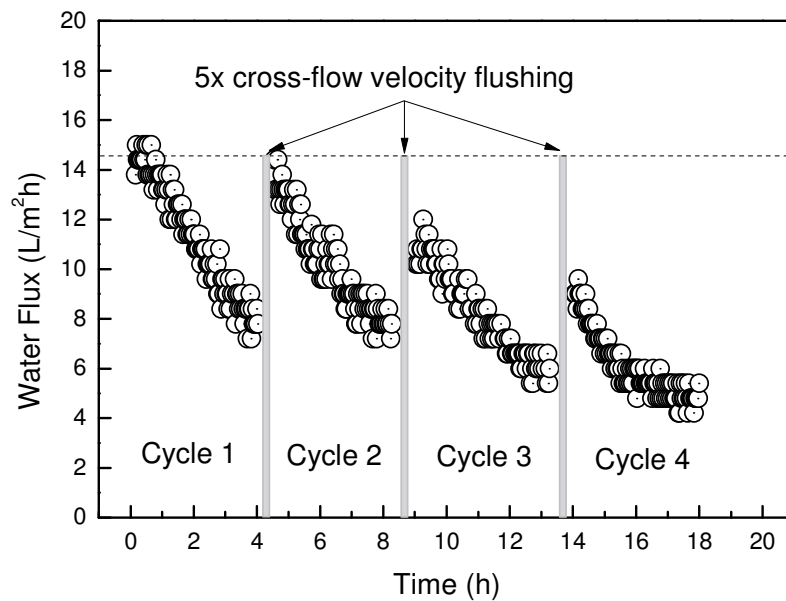


333

334 **Figure 5:** Water flux decline profile for a single digested sludge centrate fouling cycles using
 335 30 minutes in-situ high cross-flow velocity flushing (i.e. 42 cm/s) with DI water. Accelerated
 336 fouling conditions: feed solution was digested sludge centrate; NaCl draw solution was
 337 maintained at 3 M; cross-flow rates of both the feed and draw solutions were 0.5 L/min
 338 (corresponding to a cross-flow velocity of 8.3 cm/s).

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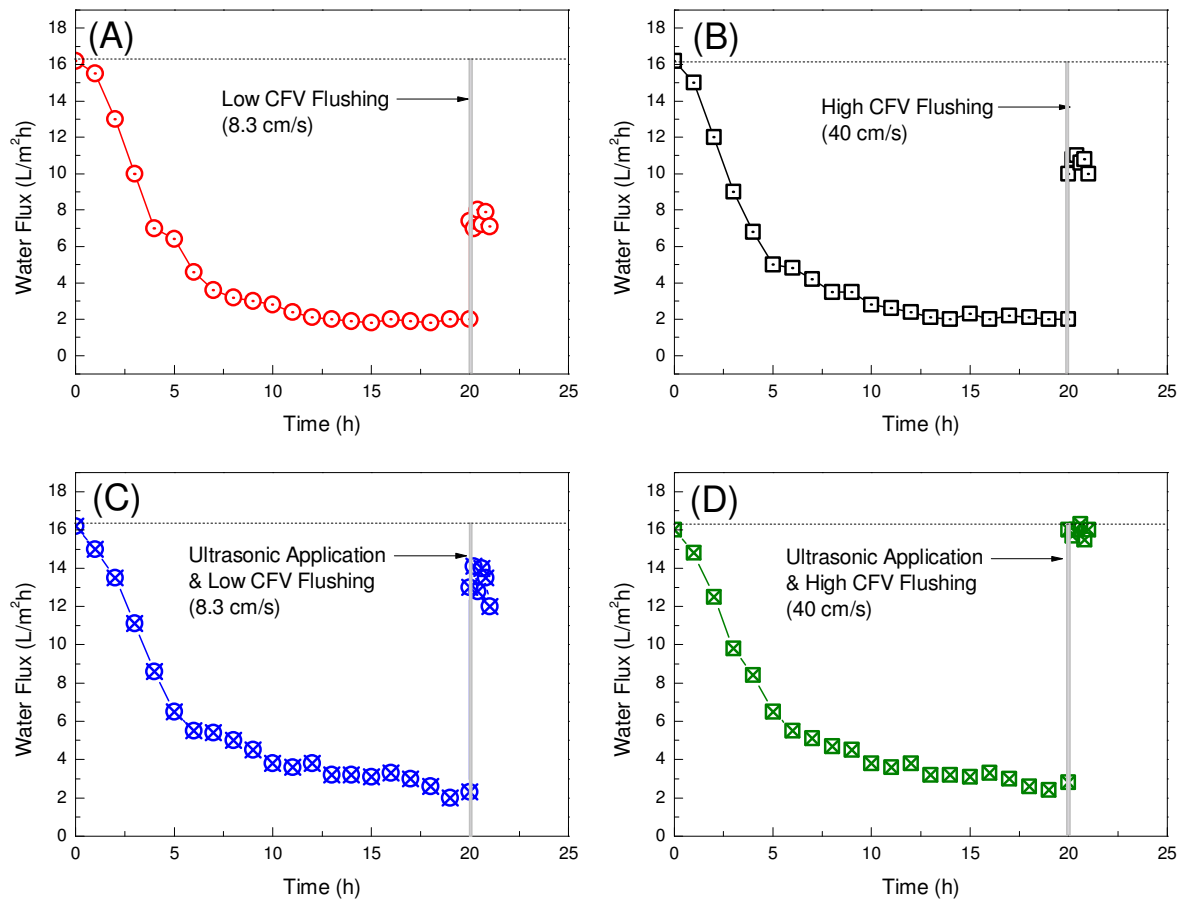


341

342 **Figure 6:** Water flux decline profile for repetitive, digested sludge centrate accelerated
 343 fouling cycles using 30 minutes in-situ high cross-flow velocity flushing (i.e. 42 cm/s) with
 344 DI water. Experimental conditions are as in Figure 5.

345 3.3.2 Complementary effects of ultrasonic cleaning and high-cross flow velocity flushing

346 Given the effectiveness of ultrasonication to prevent fouling during accelerated fouling
347 condition (section 3.2), the combination of ultrasonic cleaning and high-cross flow velocity
348 flushing was evaluated for membrane cleaning. Both the reference and five times the cross-
349 flow velocity were analysed to quantify the individual and complementary effects of these
350 two cleaning techniques. The duration of the accelerated fouling cycle was increased to
351 approximately 20 hours, to clearly distinguish the effectiveness of each cleaning strategy.
352 Figures 7A & B show how cross-flow velocity flushing at varying intensities was insufficient
353 to restore the initial water flux after a 20 hour fouling cycle. On the other hand, ultrasonic
354 application improved the water flux recovery at both rates of cross-flow velocity (Figure 7C
355 and D). The complementary effects of the two cleaning techniques were evident by the near
356 complete restoration of water flux after ultrasonic application combined with high cross-flow
357 velocity flushing (Figure 7D). The foulant materials released from the membrane surface as a
358 result of ultrasonication (i.e. high shear and turbulent conditions caused by cavitation) were
359 more readily transferred into the bulk cleaning fluid (i.e. DI water) due to the high cross-flow
360 velocity environment. Ultrasonic cleaning significantly improved simple membrane flushing
361 and has the potential to reduce the frequency of chemicals used for FO membrane cleaning.



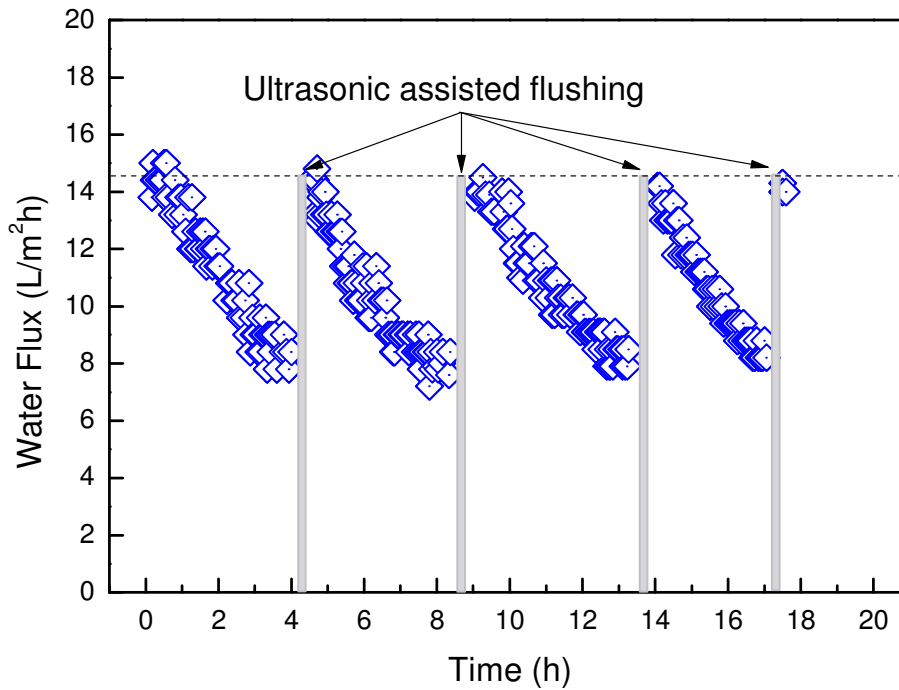
362

363 **Figure 7:** Accelerated fouling profile and water flux recovery after applying 30 minutes of
 364 (A) low cross-flow velocity (CFV), (B) high cross-flow velocity, (C) ultrasonic application
 365 with low cross-flow velocity, and (D) ultrasonic application with high cross-flow velocity.

366 Experimental conditions are as in Figure 5.

367

368 The combination of ultrasonic cleaning with high cross-flow velocity flushing was able to
 369 completely recover water flux to the initial value, over four repetitive fouling/cleaning cycles
 370 (Figure 8). These results indicate that the combination of ultrasonication and high cross-flow
 371 velocity flushing is an effective cleaning strategy. Further evaluation of ultrasonic frequency,
 372 intensity, and other operational parameters are necessary to further demonstrate process
 373 suitability and energy consumption. It is also necessary to evaluate the long term effects of
 374 ultrasonication on membrane durability after repetitive cleaning cycles.

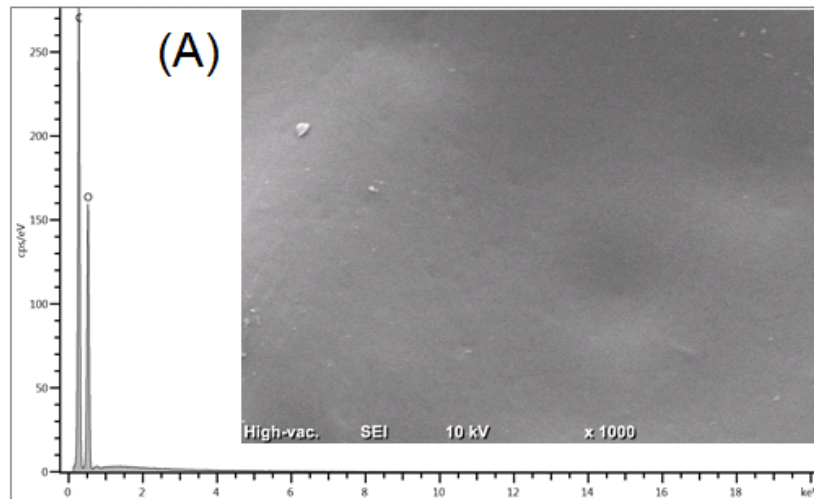


375

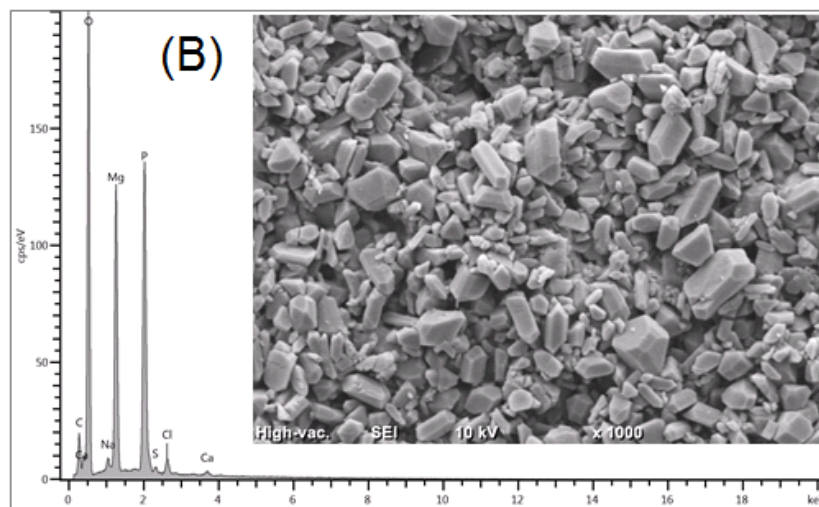
376 **Figure 8:** Water flux decline profile for repetitive, digested sludge centrate accelerated
 377 fouling cycles using 30 minutes in-situ high cross-flow velocity flushing (i.e. 42 cm/s) and
 378 ultrasonic application with DI water. Experimental conditions are as in Figure 5.
 379

380 The cleaning efficiency of ultrasonic assisted flushing is also demonstrated by comparing the
 381 pristine membrane, with the fouled and cleaned CTA membrane (Figure 9). A detailed
 382 discussion of the digested sludge centrate fouling characterisation is presented in section
 383 3.1.1. Overall, the SEM micrographs show that the application of ultrasonication with high
 384 cross-flow velocity can significantly remove all of the crystals evident in the fouling layer
 385 (Figure 9C). Furthermore, this also confirms that the dominant fouling mechanisms was bulk
 386 crystallization of minerals, followed by particle deposition on the membrane surface, as
 387 physical cleaning was capable of removing the majority of foulants [48]. In terms of the EDS
 388 spectra, the cleaned membrane indicated that traces of silicon, chlorine, and potassium
 389 remained sparsely attached to the membrane surface after the four accelerated fouling cycles
 390 (Figure 9 C). It is possible that intensified physical cleaning or chemical cleaning may be
 391 necessary to completely restore membrane performance in long term operations.

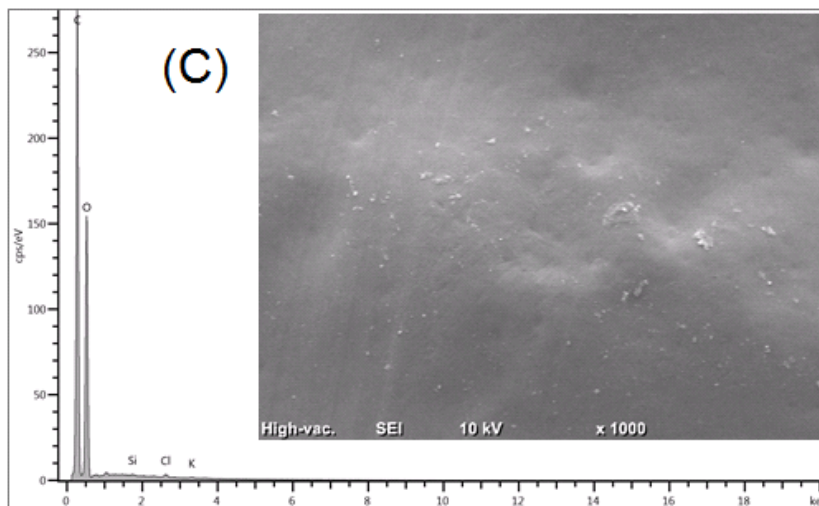
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393



394



395 **Figure 9:** SEM micrographs and EDS spectra of the (A) pristine FO membrane, (B) fouled
396 membrane, and (C) membrane after ultrasonic assisted flushing cleaning. Experimental
397 conditions are described in Figure 8.

398 **4. Conclusion**

399 Results from this study demonstrate that forward osmosis (FO) fouling associated with the
400 pre-concentration of digested sludge centrate for subsequent phosphorus recovery is
401 attributed mostly to the deposition of small mineral crystals and particulate matter on the
402 membrane surface. Thus, FO fouling during the pre-concentration of digested sludge centrate
403 can be effectively mitigated by physical cleaning. Under accelerated fouling conditions, high
404 cross-flow velocity flushing and ultrasonication could prevent membrane fouling to some
405 extent, whilst air-scouring aggravated the extent of membrane fouling. The results show that
406 periodic membrane cleaning (i.e. brief suspension of the filtration process for membrane
407 cleaning with water) was more practical than physical fouling prevention (i.e. continuously
408 applying control technique during filtration operation). The combination of ultrasonication
409 and high-cross flow velocity flushing could restore water flux to the initial value over several
410 repetitive fouling and cleaning cycles.

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