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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

Ashley Ansari
University of Wollongong, ashleyz@uow.edu.au

Faisal I. Hai
University of Wollongong, faisal@uow.edu.au

Tao He
Chinese Academy Of Sciences, het@sari.ac.cn

William E. Price
University of Wollongong, wprice@uow.edu.au

Long D. Nghiem
University of Wollongong, longn@uow.edu.au

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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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Desalination

Ashley J. Ansari [a], Faisal I. Hai [a], Tao He [b], William E. Price [c], and Long D. Nghiem *,[a]

[a] Strategic Water Infrastructure Laboratory, School of Civil, Mining and Environmental Engineering, University of Wollongong, Wollongong, NSW 2522, Australia

[b] Laboratory for Membrane Materials and Separation Technology, Shanghai Advanced Research Institute, Chinese Academy of Sciences, Shanghai 201203, China

[c] Strategic Water Infrastructure Laboratory, School of Chemistry University of Wollongong, Wollongong, NSW 2522, Australia

* Corresponding author: Long D. Nghiem, Email longn@uow.edu.au; Tel: +61 2 4221 4590

Abstract:

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

1. Introduction

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

Several approaches have been developed to recover phosphorus from wastewater. They differ in regards to the source water and the method used to pre-concentrate phosphate. Source waters include urine [14], raw wastewater [15-17], treated effluent [18, 19], sludge [20], and digested sludge centrate (i.e. anaerobic supernatant) [21-23]. Among these source waters, digested sludge centrate is an important target for phosphorus recovery because it is small in volume but rich in phosphorus and readily available at any large scale wastewater treatment plant [21-23]. The efficiency of phosphorus recovery, generally as struvite ($MgNH_4PO_4 \cdot 6H_2O$) [10] can be enhanced by pre-concentrating phosphate prior to chemical precipitation. A novel membrane filtration process with significant potential for pre-concentrating phosphate for subsequent recovery is forward osmosis (FO). As a high rejection membrane process, FO can effectively retain and enrich the phosphate and some of the ammonia in digested sludge centrate for subsequent recovery [24-26]. Furthermore, the bidirectional diffusion of protons from the feed solution into the draw solution [27] increases the digested sludge centrate pH and provides a more favourable alkaline environment for chemical phosphorus recovery [21, 22].

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

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

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

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

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

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

2. Materials and methods

2.1 Materials and chemicals

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

2.2 Forward osmosis system

A lab-scale, cross-flow FO system was employed in this study. The cell was constructed of two symmetric flow channels with length, width, and height dimensions of 100 mm, 50 mm, and 3 mm, respectively, and an effective membrane area of 50 cm^2. Circulation of the feed and draw solutions through the cell flow channels was achieved by two variable speed gear pumps (Micropump, Vancouver, Washington, USA). The circulation flow rate was regulated using two rotameters, and pump speed was adjusted to achieve the desired cross-flow velocity. For all experiments, a spacer was positioned on the draw solution side of the membrane cell to improve draw solution mixing. The flat-sheet membrane was sandwiched between two rubber gaskets and the two perspex semi-cells. The feed solution was circulated along the top semi-cell unless otherwise stated.

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

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

2.3 Physical cleaning

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

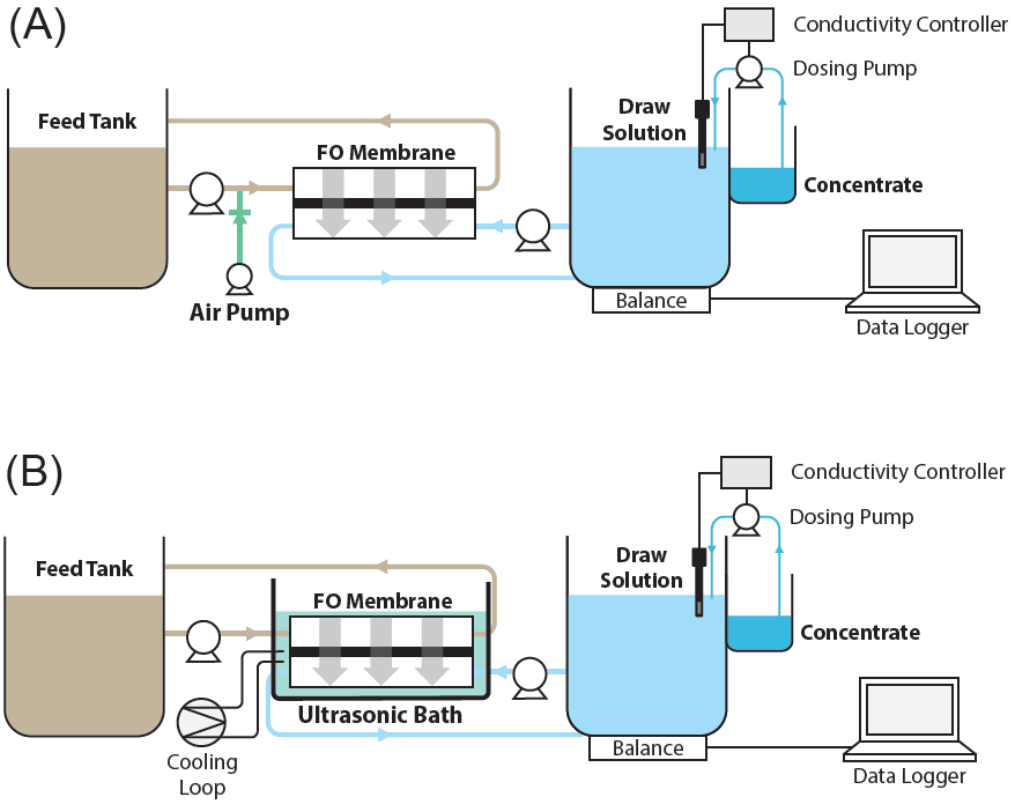

Figure 1: Schematic representation of an FO system with (A) air-scouring and (B) ultrasonication cleaning equipment.

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

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

2.4 Accelerated fouling experimental protocol

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

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

2.5 Physical cleaning

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

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

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

2.6 Membrane autopsy

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

2.7 Analytical methods

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

3. Results and discussion

3.1 Fouling propensity of wastewater and digested sludge centrate

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

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

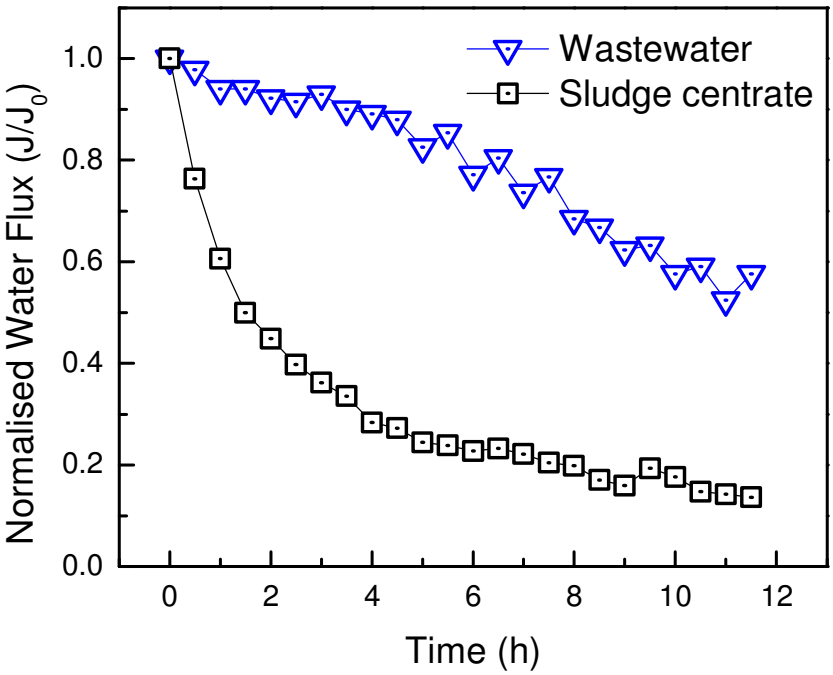

Figure 2: Comparison of wastewater and digested sludge centrate fouling propensity. Fouling propensity is represented as the observed water flux decline during accelerated fouling conditions. Initial water flux of wastewater and digested sludge centrate was 20.0 ± 0.5 L/m^{2}h. Accelerated fouling conditions: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

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

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

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

Parameter	Units	Raw wastewater	Sludge centrate	Sludge centrate - Minimum FO rejection (%)
Total solids	g/L	0.64 ± 0.03	1.16 ± 0.03	-
Volatile solids	g/L	0.40 ± 0.02	0.58 ± 0.12	-
Electrical conductivity	mS/cm	1.45 ± 0.24	5.99 ± 0.11	-
pH	-	6.85 ± 0.10	7.77 ± 0.05	-
Total organic carbon	mg/L	45 ± 10	602 ± 16	94.3
Total nitrogen	mg/L	41 ± 9	764 ± 25	67.6
PO_4^{3-}-P	mg/L	23 ± 5	97 ± 7	98.6
NH_4^+-N	mg/L	71 ± 12	521 ± 22	88.3
Ca^{2+}	mg/L	-	63 ± 5	-
Mg^{2+}	mg/L	-	14 ± 5	-
K^+	mg/L	-	106 ± 3	-

3.1.1 Digested sludge centrate fouling characterisation

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

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

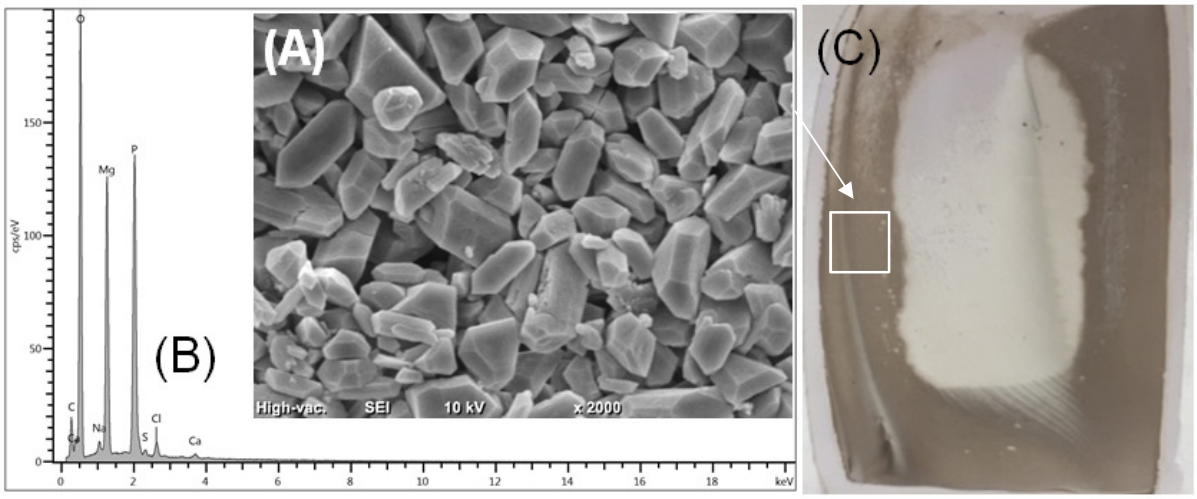

Figure 3: (A) SEM micrograph and (B) EDS spectra of the FO membrane surface at the conclusion of the accelerated fouling experiment using digested sludge centrate as the feed solution. Experimental conditions are described in Figure 2.

3.2 Membrane fouling prevention

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

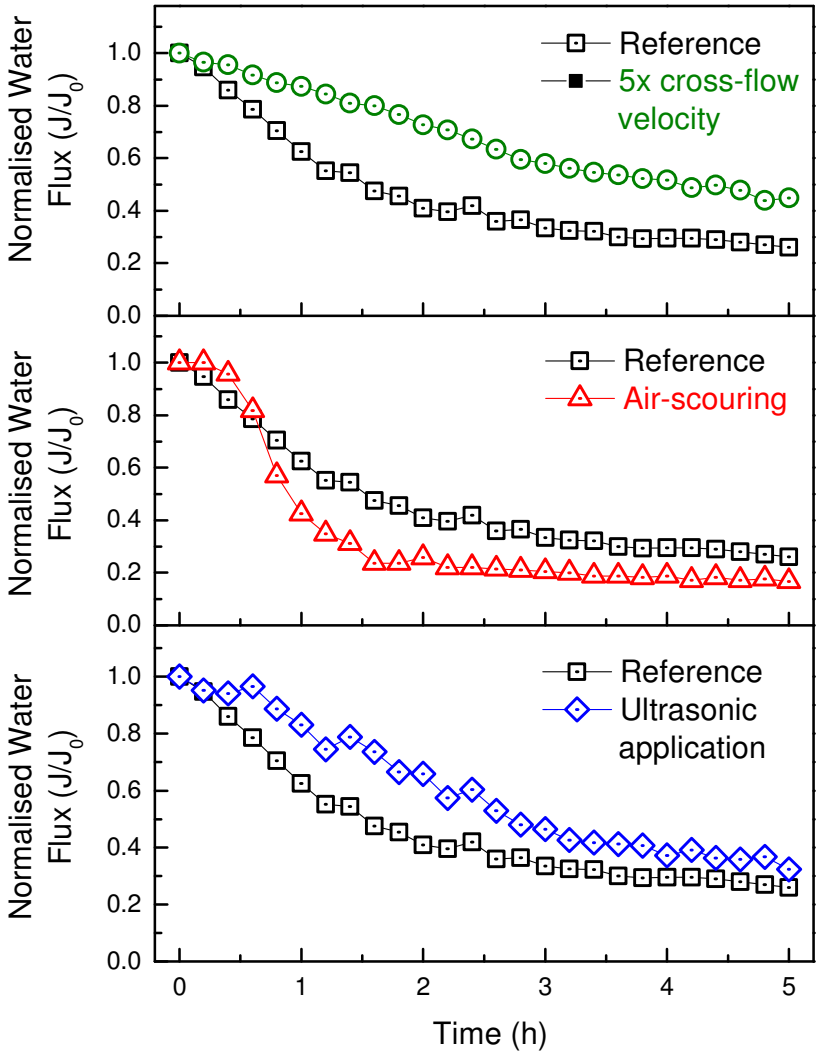

Figure 4: Normalised water flux decline during accelerated fouling conditions with; (A) 5x cross-flow velocity (i.e. 42 cm/s), (B) Air-scouring, and (C) ultrasonic application, applied as fouling prevention techniques. Prevention techniques were continuously applied during the filtration time. Reference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

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

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

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

Increasing the cross-flow velocity during filtration cycles was the most effective strategy amongst the three techniques investigated here. This achieved the highest cumulative permeate volume during the five hour cycle corresponding the lowest water flux decline. Variations in the cross-flow velocity rate are expected to be proportional to the water flux behaviour, however, this would correspondingly influence the systems energy consumption. Costs associated with circulation can be significant for FO membrane systems [54] and therefore optimisation of membrane fouling prevention techniques is important for a sustainable system. A similar argument can be said for ultrasonication, as continuous 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.

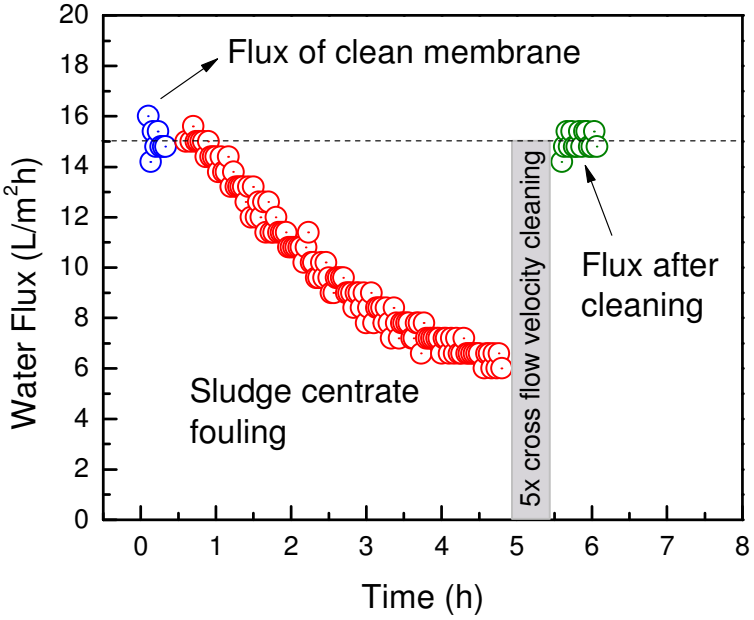

Figure 5: Water flux decline profile for a single digested sludge centrate fouling cycles using 30 minutes in-situ high cross-flow velocity flushing (i.e. 42 cm/s) with DI water. Accelerated fouling conditions: feed solution was digested sludge centrate; NaCl draw solution was maintained at 3 M; cross-flow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

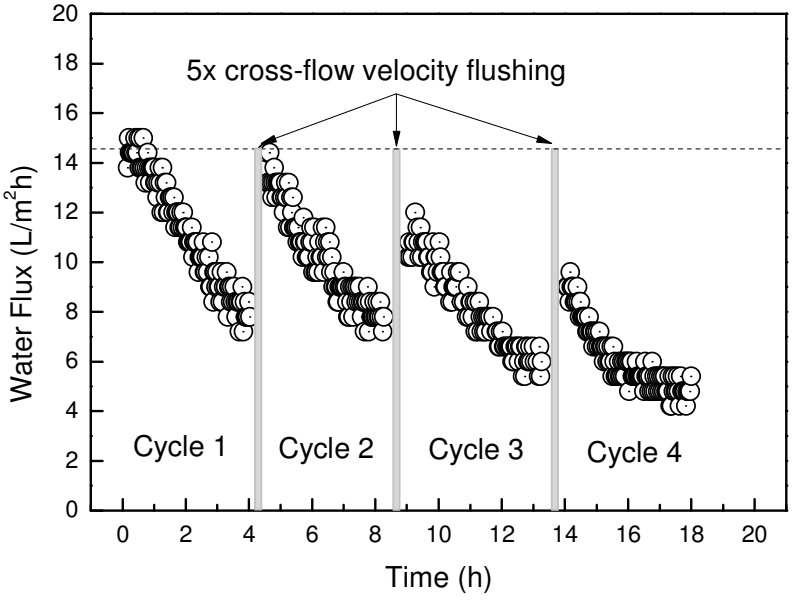

Figure 6: Water flux decline profile for repetitive, digested sludge centrate accelerated fouling cycles using 30 minutes in-situ high cross-flow velocity flushing (i.e. 42 cm/s) with DI water. Experimental conditions are as in Figure 5.

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

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

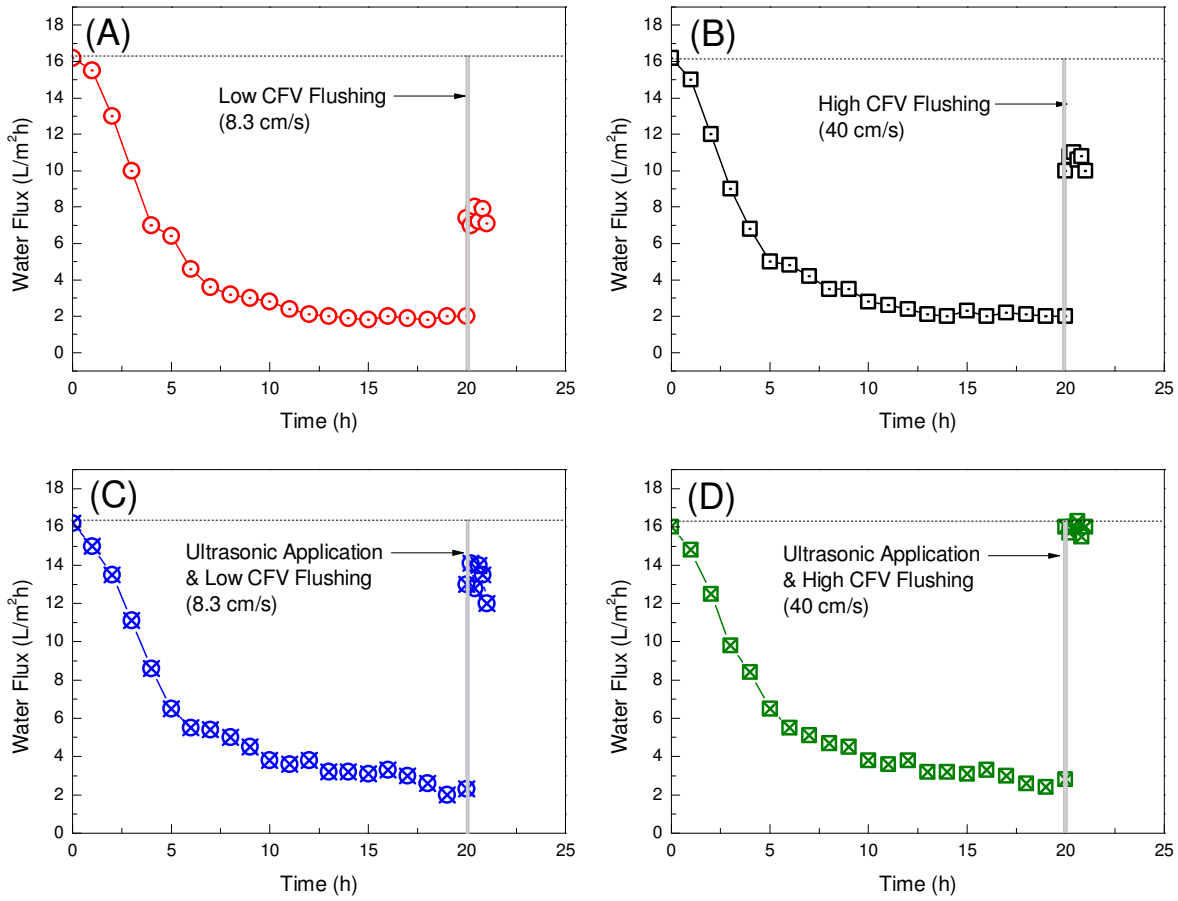

Figure 7: Accelerated fouling profile and water flux recovery after applying 30 minutes of (A) low cross-flow velocity (CFV), (B) high cross-flow velocity, (C) ultrasonic application with low cross-flow velocity, and (D) ultrasonic application with high cross-flow velocity. Experimental conditions are as in Figure 5.

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

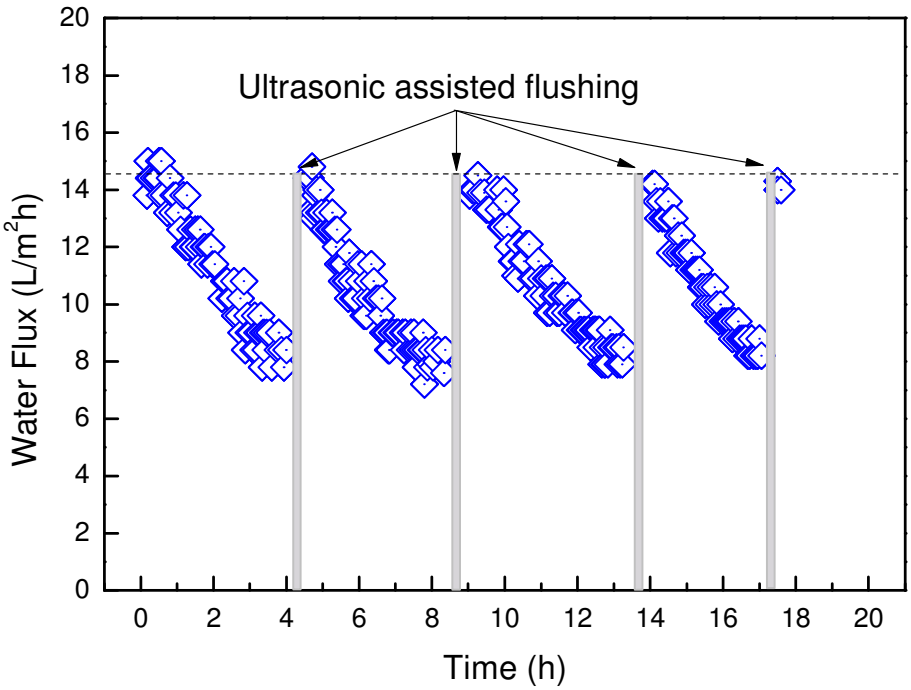

Figure 8: Water flux decline profile for repetitive, digested sludge centrate accelerated fouling cycles using 30 minutes in-situ high cross-flow velocity flushing (i.e. 42 cm/s) and ultrasonic application with DI water. Experimental conditions are as in Figure 5.

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

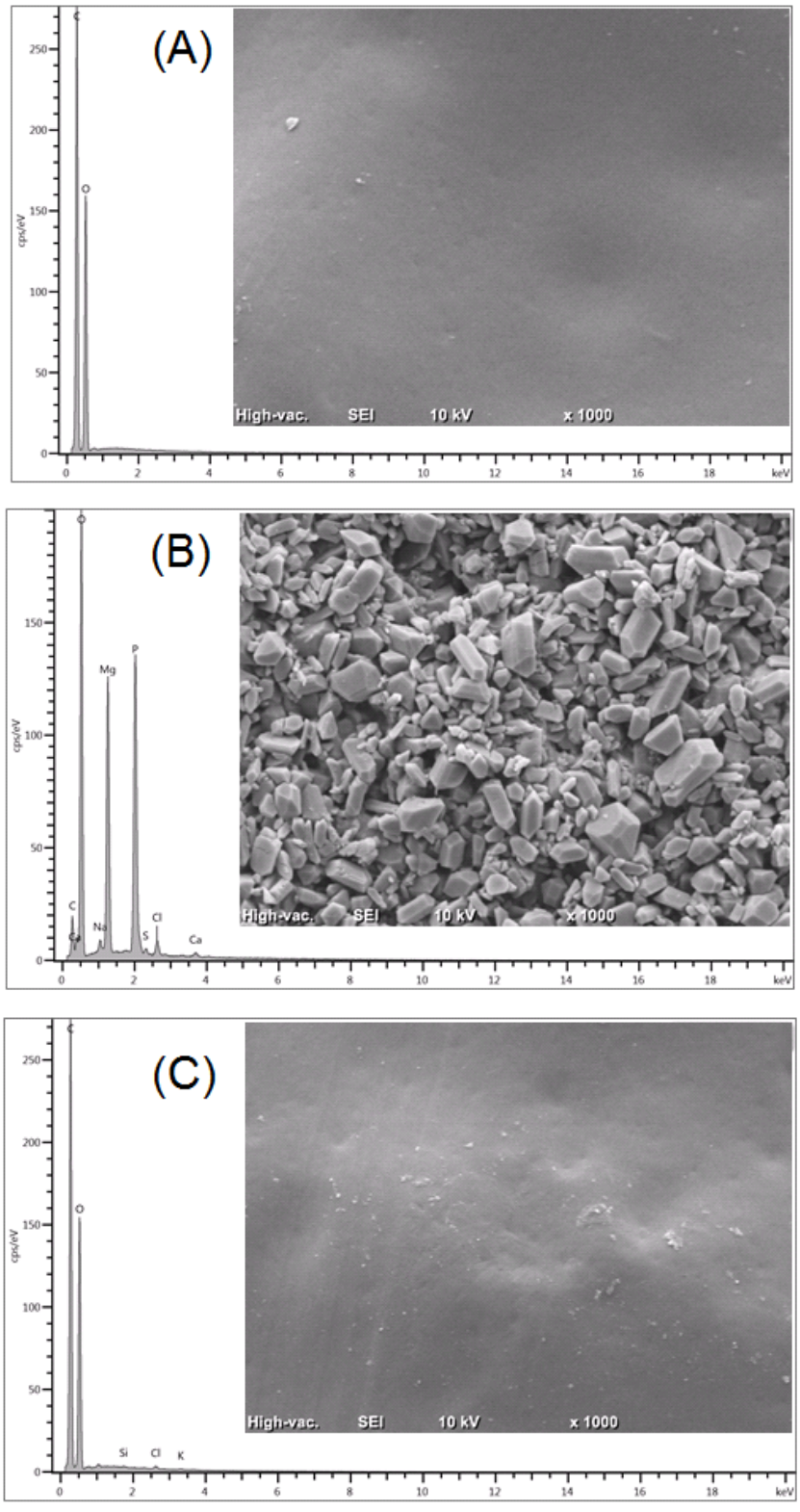

Figure 9: SEM micrographs and EDS spectra of the (A) pristine FO membrane, (B) fouled membrane, and (C) membrane after ultrasonic assisted flushing cleaning. Experimental conditions are described in Figure 8.

4. Conclusion

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

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6. References

[1] G. Calvo, A. Valero, A. Valero, Assessing maximum production peak and resource availability of non-fuel mineral resources: Analyzing the influence of extractable global resources, Resources, Conservation and Recycling, 125 (2017) 208-217.

[2] D. Cordell, J.-O. Drangert, S. White, The story of phosphorus: Global food security and food for thought, Global Environmental Change, 19 (2009) 292-305.

[3] J. Elser, E. Bennett, Phosphorus cycle: A broken biogeochemical cycle, Nature, 478 (2011) 29-31.

[4] R.W. Scholz, F.W. Wellmer, Approaching a dynamic view on the availability of mineral resources: What we may learn from the case of phosphorus?, Global Environmental Change, 23 (2013) 11-27.

[5] C.M. Lwin, M. Murakami, S. Hashimoto, The implications of allocation scenarios for global phosphorus flow from agriculture and wastewater, Resources, Conservation and Recycling, 122 (2017) 94-105.

[6] O. Krüger, C. Adam, Phosphorus in recycling fertilizers - analytical challenges, Environmental Research, 155 (2017) 353-358.

[7] K.C. van Dijk, J.P. Lesschen, O. Oenema, Phosphorus flows and balances of the European Union Member States, Science of the Total Environment, 542, Part B (2016) 1078-1093.

[8] B. Li, I. Boiarkina, B. Young, W. Yu, Substance flow analysis of phosphorus within New Zealand and comparison with other countries, Science of the Total Environment, 527–528 (2015) 483-492.

[9] N. Martí, R. Barat, A. Seco, L. Pastor, A. Bouzas, Sludge management modeling to enhance P-recovery as struvite in wastewater treatment plants, J Environ Manage, 196 (2017) 340-346.

[10] K.S. Le Corre, E. Valsami-Jones, P. Hobbs, S.A. Parsons, Phosphorus Recovery from Wastewater by Struvite Crystallization: A Review, Critical Reviews in Environmental Science and Technology
39 (2009) 433-477.

[11] L.E. de-Bashan, Y. Bashan, Recent advances in removing phosphorus from wastewater and its future use as fertilizer (1997–2003), Water Research, 38 (2004) 4222-4246.

[12] E. Desmidt, K. Ghyselbrecht, Y. Zhang, L. Pinoy, B. Van der Bruggen, W. Verstraete, K. Rabaey, B. Meesschaert, Global Phosphorus Scarcity and Full-Scale P-Recovery Techniques: A Review, Critical Reviews in Environmental Science and Technology, 45 (2014) 336-384.

[13] B.K. Mayer, L.A. Baker, T.H. Boyer, P. Drechsel, M. Gifford, M.A. Hanjra, P. Parameswaran, J. Stoltzfus, P. Westerhoff, B.E. Rittmann, Total Value of Phosphorus Recovery, Environmental Science & Technology, 50 (2016) 6606-6620.

[14] J. Zhang, Q. She, V.W.C. Chang, C.Y. Tang, R.D. Webster, Mining Nutrients (N, K, P) from Urban Source-Separated Urine by Forward Osmosis Dewatering, Environmental Science & Technology, 48 (2014) 3386-3394.

[15] G. Qiu, Y.-P. Ting, Direct phosphorus recovery from municipal wastewater via osmotic membrane bioreactor (OMBR) for wastewater treatment, Bioresource Technology, 170 (2014) 221-229.

[16] W. Luo, F.I. Hai, W.E. Price, W. Guo, H.H. Ngo, K. Yamamoto, L.D. Nghiem, Phosphorus and water recovery by a novel osmotic membrane bioreactor–reverse osmosis system, Bioresource Technology, 200 (2016) 297-304.

[17] G. Qiu, S. Zhang, D.S. Srinivasa Raghavan, S. Das, Y.-P. Ting, The potential of hybrid forward osmosis membrane bioreactor (FOMBR) processes in achieving high throughput treatment of municipal wastewater with enhanced phosphorus recovery, Water Research, 105 (2016) 370-382.

[18] R. Liu, Y. Wang, G. Wu, J. Luo, S. Wang, Development of a selective electrodialysis for nutrient recovery and desalination during secondary effluent treatment, Chemical Engineering Journal, 322 (2017) 224-233.

[19] W. Xue, T. Tobino, F. Nakajima, K. Yamamoto, Seawater-driven forward osmosis for enriching nitrogen and phosphorous in treated municipal wastewater: Effect of membrane properties and feed solution chemistry, Water Research, 69 (2015) 120-130.

[20] T. Schütte, C. Niewersch, T. Wintgens, S. Yüce, Phosphorus recovery from sewage sludge by nanofiltration in diafiltration mode, Journal of Membrane Science, 480 (2015) 74-82.

[21] M. Xie, L.D. Nghiem, W.E. Price, M. Elimelech, Toward Resource Recovery from Wastewater: Extraction of Phosphorus from Digested Sludge Using a Hybrid Forward Osmosis–Membrane Distillation Process, Environmental Science & Technology Letters, 1 (2014) 191-195.

[22] A.J. Ansari, F.I. Hai, W.E. Price, L.D. Nghiem, Phosphorus recovery from digested sludge centrate using seawater-driven forward osmosis, Sep Purif Technol, 163 (2016) 1-7.

[23] R.W. Holloway, A.E. Childress, K.E. Dennett, T.Y. Cath, Forward osmosis for concentration of anaerobic digester centrate, Water Research, 41 (2007) 4005-4014.

[24] M. Xie, H.K. Shon, S.R. Gray, M. Elimelech, Membrane-based processes for wastewater nutrient recovery: Technology, challenges, and future direction, Water Research, 89 (2016) 210-221.

[25] D.L. Shaffer, J.R. Werber, H. Jaramillo, S. Lin, M. Elimelech, Forward osmosis: Where are we now?, Desalination, 356 (2015) 271-284.

[26] K. Lutchmiah, A.R.D. Verliefde, K. Roest, L.C. Rietveld, E.R. Cornelissen, Forward osmosis for application in wastewater treatment: A review, Water Research, 58 (2014) 179-197.

[27] N.T. Hancock, T.Y. Cath, Solute Coupled Diffusion in Osmotically Driven Membrane Processes, Environmental Science & Technology, 43 (2009) 6769-6775.

[28] W.C.L. Lay, T.H. Chong, C.Y. Tang, A.G. Fane, J. Zhang, Y. Liu, Fouling propensity of forward osmosis: Investigation of the slower flux decline phenomenon, in: Water Science & Technology, 2010, pp. 927-936.

[29] S. Lee, C. Boo, M. Elimelech, S. Hong, Comparison of fouling behavior in forward osmosis (FO) and reverse osmosis (RO), Journal of Membrane Science, 365 (2010) 34-39.

[30] Q. She, R. Wang, A.G. Fane, C.Y. Tang, Membrane fouling in osmotically driven membrane processes: A review, Journal of Membrane Science, 499 (2016) 201-233.

[31] X.-M. Li, B. Zhao, Z. Wang, M. Xie, J. Song, L.D. Nghiem, T. He, C. Yang, C. Li, G. Chen, Water reclamation from shale gas drilling flow-back fluid using a novel forward osmosis–vacuum membrane distillation hybrid system, Water Science & Technology, 69 (2014) 1036-1044.

[32] G. Chen, Z. Wang, L.D. Nghiem, X.-M. Li, M. Xie, B. Zhao, M. Zhang, J. Song, T. He, Treatment of shale gas drilling flowback fluids (SGDFs) by forward osmosis: Membrane fouling and mitigation, Desalination, 366 (2015) 113-120.

[33] K.L. Hickenbottom, N.T. Hancock, N.R. Hutchings, E.W. Appleton, E.G. Beaudry, P. Xu, T.Y. Cath, Forward osmosis treatment of drilling mud and fracturing wastewater from oil and gas operations, Desalination, 312 (2013) 60-66.

[34] J. Li, A. Niu, C.-J. Lu, J.-H. Zhang, M. Junaid, P.R. Strauss, P. Xiao, X. Wang, Y.-W. Ren, D.-S. Pei, A novel forward osmosis system in landfill leachate treatment for removing polycyclic aromatic hydrocarbons and for direct fertigation, Chemosphere, 168 (2017) 112-121.

[35] A.J. Ansari, F.I. Hai, W.E. Price, J.E. Drewes, L.D. Nghiem, Forward osmosis as a platform for resource recovery from municipal wastewater - A critical assessment of the literature, Journal of Membrane Science, 529 (2017) 195-206.

[36] M. Xie, J. Lee, L.D. Nghiem, M. Elimelech, Role of pressure in organic fouling in forward osmosis and reverse osmosis, Journal of Membrane Science, 493 (2015) 748-754.

[37] Q.H. She, X. Jin, Q.H. Li, C.Y.Y. Tang, Relating reverse and forward solute diffusion to membrane fouling in osmotically driven membrane processes, Water Research, 46 (2012) 2478-2486.

[38] B. Mi, M. Elimelech, Chemical and physical aspects of organic fouling of forward osmosis membranes, Journal of Membrane Science, 320 (2008) 292-302.

[39] T. Majeed, S. Phuntsho, L. Chekli, S.-H. Lee, K. Kim, H.K. Shon, Role of various physical and chemical techniques for hollow fibre forward osmosis membrane cleaning, Desalination and Water Treatment, 57 (2016) 7742-7752.

[40] C. Boo, M. Elimelech, S. Hong, Fouling control in a forward osmosis process integrating seawater desalination and wastewater reclamation, Journal of Membrane Science, 444 (2013) 148-156.

[41] R. Valladares Linares, Z. Li, V. Yangali-Quintanilla, Q. Li, G. Amy, Cleaning protocol for a FO membrane fouled in wastewater reuse, Desalination and Water Treatment, 51 (2013) 4821-4824.

[42] N. Nguyen Cong, N. Hau Thi, C. Shiao-Shing, N. Nhat Thien, L. Chi-Wang, Application of forward osmosis (FO) under ultrasonication on sludge thickening of waste activated sludge, Water Science & Technology, 72 (2015) 1301-1307.

[43] Y.-J. Choi, S.-H. Kim, S. Jeong, T.-M. Hwang, Application of ultrasound to mitigate calcium sulfate scaling and colloidal fouling, Desalination, 336 (2014) 153-159.

[44] Z. Wang, J. Tang, C. Zhu, Y. Dong, Q. Wang, Z. Wu, Chemical cleaning protocols for thin film composite (TFC) polyamide forward osmosis membranes used for municipal wastewater treatment, Journal of Membrane Science, 475 (2015) 184-192.

[45] X. Wang, T. Hu, Z. Wang, X. Li, Y. Ren, Permeability recovery of fouled forward osmosis membranes by chemical cleaning during a long-term operation of anaerobic osmotic membrane bioreactors treating low-strength wastewater, Water Research, 123 (2017) 505-512.

[46] T.Y. Cath, M. Elimelech, J.R. McCutcheon, R.L. McGinnis, A. Achilli, D. Anastasio, A.R. Brady, A.E. Childress, I.V. Farr, N.T. Hancock, J. Lampi, L.D. Nghiem, M. Xie, N.Y. Yip, Standard Methodology for Evaluating Membrane Performance in Osmotically Driven Membrane Processes, Desalination, 312 (2013) 31-38.

[47] A.A. Alturki, J.A. McDonald, S.J. Khan, W.E. Price, L.D. Nghiem, M. Elimelech, Removal of trace organic contaminants by the forward osmosis process, Sep Purif Technol, 103 (2013) 258-266.

[48] B. Mi, M. Elimelech, Gypsum Scaling and Cleaning in Forward Osmosis: Measurements and Mechanisms, Environmental Science & Technology, 44 (2010) 2022-2028.

[49] M.O. Lamminen, H.W. Walker, L.K. Weavers, Mechanisms and factors influencing the ultrasonic cleaning of particle-fouled ceramic membranes, Journal of Membrane Science, 237 (2004) 213-223.

[50] B.S. Chanukya, N.K. Rastogi, Ultrasound assisted forward osmosis concentration of fruit juice and natural colorant, Ultrasonics Sonochemistry, 34 (2017) 426-435.

[51] H. Kim, Y. Lee, M. Elimelech, A. Adout, Y.C. Kim, Experimental Study of Ultrasonic Effects on Flux Enhancement in Forward Osmosis Process, Meeting Abstracts, MA2012-01 (2012) 90.

[52] W. Luo, F.I. Hai, W.E. Price, L.D. Nghiem, Water extraction from mixed liquor of an aerobic bioreactor by forward osmosis: Membrane fouling and biomass characteristics assessment, Sep Purif Technol, 145 (2015) 56-62.

[53] T. Wang, X. He, Z. Wu, J. Li, Direct observation of flow and bubble behavior in flat sheet modules with a distributor, RSC Advances, 7 (2017) 19050-19059.

[54] S. Zou, H. Yuan, A. Childress, Z. He, Energy Consumption by Recirculation: A Missing Parameter When Evaluating Forward Osmosis, Environmental Science & Technology, 50 (2016) 6827-6829.