2018

Assessing the integration of forward osmosis and anaerobic digestion for simultaneous wastewater treatment and resource recovery

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

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

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

Hao H. Ngo
University of Technology Sydney, h.ngo@uts.edu.au

Wenshan Guo
University of Technology Sydney, wguo@uts.edu.au

See next page for additional authors

Assessing the integration of forward osmosis and anaerobic digestion for simultaneous wastewater treatment and resource recovery

Abstract

This study assessed the performance and key challenges associated with the integration of forward osmosis (FO) and anaerobic digestion for wastewater treatment and resource recovery. Using a thin film composite polyamide FO membrane, maximising the pre-concentration factor (i.e. system water recovery) resulted in the enrichment of organics and salinity in wastewater. Biomethane potential evaluation indicated that methane production increased correspondingly with the FO pre-concentration factor due to the organic retention in the feed solution. At 90% water recovery, about 10% more methane was produced when using NaOAc compared with NaCl because of the contribution of biodegradable reverse NaOAc flux. No negative impact on anaerobic digestion was observed when wastewater was pre-concentrated ten-fold (90% water recovery) for both draw solutes. Interestingly, the unit cost of methane production using NaOAc was slightly lower than NaCl due to the lower reverse solute flux of NaOAc, although NaCl is a much cheaper chemical.

Disciplines

Engineering | Science and Technology Studies

Publication Details

Ansari, A. J., Hai, F. I., Price, W. E., Ngo, H. H., Guo, W. & Nghiem, L. D. (2018). Assessing the integration of forward osmosis and anaerobic digestion for simultaneous wastewater treatment and resource recovery. Bioresource Technology, 260 221-226.

Authors

Ashley Ansari, Faisal I. Hai, William E. Price, Hao H. Ngo, Wenshan Guo, and Long D. Nghiem

RESEARCH HIGHLIGHTS

- No negative effect on CH_4 production at 10 folds wastewater pre-concentration
- At 90% water recovery, CH_4 production using NaOAc was 10% more than NaCl as DS
- The unit cost of methane production was highly sensitive to the reverse salt flux
- The unit cost of methane production using NaOAc was slightly lower than NaCl
- Membrane fouling was limited to surface deposition and was readily removed by flushing

Assessing the integration of forward osmosis and anaerobic digestion for simultaneous wastewater treatment and resource recovery

Revised Manuscript Submitted to

Bioresource Technology

CESE2017

March 2018

Ashley J. Ansari [a], Faisal I. Hai [a], William E. Price [b], Huu H. Ngo [c], Wenshan Guo [c], and Long D. Nghiem [c,*]

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

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

[c] Centre for Technology in Water and Wastewater, School of Civil and Environmental Engineering, University of Technology Sydney, Ultimo NSW 2007, Australia

---

* Corresponding author:

Long D. Nghiem; Email: duclong.nghiem@uts.edu.au; Ph +61 2 9514 2625

Abstract

This study assessed the performance and key challenges associated with the integration of forward osmosis (FO) and anaerobic digestion for wastewater treatment and energy recovery. Using a thin film composite polyamide FO membrane, maximising the pre-concentration factor (i.e. system water recovery) resulted in the enrichment of organics and salinity in wastewater. Biomethane potential evaluation indicated that methane production increased correspondingly with the FO pre-concentration factor due to the organic retention in the feed solution. At 90% water recovery, about 10% more methane was produced when using NaOAc compared with NaCl because of the contribution of degradable reverse NaOAC flux. No negative impact on anaerobic digestion was observed when wastewater was pre-concentrated ten-fold (90% water recovery) for both draw solutes. Interestingly, the unit cost of methane production using NaOAc was slightly lower than NaCl due to the lower reverse solute flux and higher methane production.

Keywords: Forward osmosis (FO); reverse solute flux; biomethane potential (BMP) analysis; draw solution selection; sewer mining.

1. Introduction

In a circular economy, wastewater is considered as a source of water, energy, and nutrients, rather than a waste. As such, there is a growing demand for low impact wastewater treatment systems that provide water reuse and are able to recover nutrients and energy (Desmidt et al., 2014; Puyol et al., 2016). This demand has driven the development of innovative technologies to tap into the resource potential of wastewater. Membrane-based technologies have been essential for advanced water purification in reuse applications (Shannon et al., 2008; Xie et al., 2016). Similarly, anaerobic digestion has evolved as a key technological pathway for the realisation of energy and nutrient recovery from wastewater (Frijns et al., 2013; Verstraete et al., 2009).

Anaerobic digestion is a promising platform for low energy wastewater treatment and resource recovery. Indeed, the conventional activated sludge process requires significant electrical energy consumption for aeration. Anaerobic digestion has been widely used for the treatment of sludge originating from wastewater treatment plants, however, there are several technical challenges associated with applying anaerobic digestion for direct wastewater treatment. One such difficulty is the dilute nature of wastewater that significantly increases the digester heating requirement per unit of biogas production and thus influences the economic viability of the process. In addition, methane loss due to dissolution in the effluent is significant at a low production rate. For low-strength wastewater, processes that pre-concentrate chemical oxygen demand (COD) and nutrients (e.g. phosphorus) represent one avenue to improving the economics of biogas recovery from anaerobic treatment units (Jin et al., 2017; Wan et al., 2016).

High retention membranes such as forward osmosis (FO) can be strategically integrated with anaerobic digestion to achieve simultaneous wastewater treatment and resource recovery (Ansari et al., 2017; Wang et al., 2016). The major advantages of FO compared to other membrane processes include, low hydraulic pressure operation, low fouling propensity, easy cleaning, and a high rejection of a broad range of contaminants. FO can also be coupled with a draw solution regeneration process such as membrane distillation (MD) and reverse osmosis to directly extract clean water from raw wastewater, while simultaneously concentrating wastewater organics for subsequent anaerobic digestion (Luo et al., 2017; Nguyen et al., 2016; Shahzad et al., 2017). Anaerobically digesting FO pre-concentrated

wastewater can produce biogas, which can be utilised by a combined heat and power engine to produce electricity and thermal energy. Surplus electricity can be supplied to the grid and the produced thermal energy can be used for MD and the anaerobic process. This latter process also converts biologically bound phosphorus into a soluble form, thus allowing phosphorus recovery as struvite ($MgNH_4PO_4 \cdot 6H_2O$) or hydroxyapatite ($Ca_{10}(PO_4)_6(OH)_2$).

Interest in combining FO with anaerobic treatment has significantly increased in recent years due to the potential advantages of low-energy wastewater stabilisation and resource recovery. Recent studies have investigated FO-anaerobic integration in terms of draw solution selection (Kim et al., 2016), process configurations (Qiu et al., 2016; Wang et al., 2017b; Zhang et al., 2017), membrane cleaning (Wang et al., 2017a), trace organic contaminant removal (Kim et al., 2017), microbial composition (Wu et al., 2017), and energy dynamics (Onoda et al., 2017). However, there is a lack of studies which thoroughly assess the key FO operating parameters that essentially govern anaerobic digestion performance.

Water recovery and the selected draw solution can influence the composition of pre-concentrated wastewater in terms of organics retention and salinity accumulation. The incompatibility between high salinity and anaerobic microorganisms represents the most prominent challenge associated with integrating FO with anaerobic treatment. Salinity accumulation is inherently associated with the FO process. However, appropriate draw solution selection can potentially reduce the amount of solute diffusing into the feed solution. On the other hand, water recovery determines the accumulation of existing dissolved solutes in wastewater. Determining the influence of these FO operating parameters on anaerobic treatment is imperative to evaluate the feasibility and optimise biogas production from FO pre-concentrated wastewater.

This study aims to evaluate the process performance and investigate the key challenges associated with integrating FO with anaerobic treatment. Specifically, this study optimises the FO concentration factor (i.e. system water recovery) to balance the organic content and salt concentration in pre-concentrated wastewater and their combined effects on methane production. Representative inorganic and ionic organic draw solutes, namely sodium chloride (NaCl) and sodium acetate (NaOAc) were compared in terms of FO membrane performance and the digestibility of pre-concentrated wastewater. Optimised parameters and cleaning

techniques are applied to mitigate salinity accumulation (i.e. alternative draw solute) and membrane fouling (i.e. physical flushing).

2. Materials and methods

2.1 Forward osmosis system

The lab-scale FO system used in this study consisted of a cross-flow membrane cell with an effective membrane area of 50 cm^2. The membrane cell comprised of two symmetric flow channels for the feed and draw solutions to contact the membrane. Each flow channel had length, width, and height dimensions of 100 mm, 50 mm, and 3 mm, respectively. The flat-sheet membrane was positioned between two rubber gaskets and two semi-cells made of perspex. The feed and draw solutions were circulated through the membrane cell channels via two variable-speed gear pumps (Micropump, Vancouver, Washington, USA). The pump speed was adjusted to maintain the system cross-flow velocity, and the circulation flow rate was regulated using two rotameters. A diamond shaped spacer with a thickness of 1 mm was placed within the draw solution flow channel to improve mixing.

The flux dynamics of the system were determined according to the standard procedure described by Cath et al. (2013). The weight change of the draw solution tank was measured using a digital balance (Mettler-Toledo Inc., Hightstown, New Jersey, USA) to determine the permeate water flux. The osmotic pressure of each draw solution was kept constant during each FO experiment by controlling the solution conductivity. The draw solution conductivity was continuously measured using a conductivity probe (Cole-Parmer, Vernon Hills, Illinois, USA). A peristaltic pump connected to a controller dosed highly concentrated stock solution (5 M) into the draw solution as the measured conductivity fell below the specified range at a control accuracy of (±0.1 mS/cm). This re-concentration system was also placed on a digital balance to ensure accurate flux measurements due to weight changes.

2.2 Biochemical methane potential experimental set-up

The biochemical methane potential (BMP) experimental set-up consisted of 16 fermentation bottles (Wiltronics Research, Ballarat, Victoria, Australia). Each BMP bottle was filled with 500 mL of inoculum and 250 mL of the simulated FO pre-concentrate. The fermentation bottles were submerged in a water bath at a constant temperature of 35.0 ± 0.1 °C (Ratek Instruments, Boronia, Victoria, Australia). Each bottle was sealed with a rubber bung

attached to a water filled S-shaped air lock, and flexible plastic tubing was used to transfer biogas to the collection gallery. The gas collection gallery included 16 inverted 1000 mL plastic measuring cylinders, filled with a 1 M NaOH solution. The NaOH solution sequestered the CO_2 and H_2S in the biogas, whilst the CH_4 gas displaced the NaOH inside the cylinder. Daily measurements of CH_4 gas production were recorded.

2.3 Materials and chemicals

Wastewater (after primary sedimentation) and digested sludge were obtained from the Wollongong Wastewater Treatment Plant (WWTP) in New South Wales, Australia. The wastewater was used as a feed solution for FO pre-concentration experiments, whilst the digested sludge was used as the inoculum for the BMP experiments. Basic characteristics of the solutions are summarised in Table 1.

[Table 1]

Draw solutions were prepared using analytical grade NaCl or NaOAc. The draw solution concentration was determined by OLI Stream Analyzer (OLI Systems, Inc., Morris Plains, New Jersey, USA) calculations to achieve an equivalent osmotic pressure of 30 bar (similar to that of seawater).

To accurately assess the effect of FO water recovery and draw solution on methane production, BMP experiments were conducted using a synthetic wastewater solution. The actual concentrate originating from the FO system was not used in the BMP experiments, as the liquid volume produced by the lab-scale FO system was too small. Instead, a synthetic solution was made to simulate the pre-concentration of wastewater components, as well as the contribution of reverse draw solute flux. The concentrated stock solution was prepared to contain 4 g/L glucose, 1 g/L peptone, 0.35 g/L urea, 0.175 g/L KH_2PO_4, 0.175 g/L $MgSO_4$, 0.1 g/L $FeSO_4$, and 2.25 g/L NaOAc. This stock solution was then diluted to accurately simulate the COD of the initial primary effluent as well as the experimentally measured COD amount in FO pre-concentrated wastewater at 50, 80 and 90% water recovery. A pre-determined amount of analytical grade NaCl or NaOAc was then added to the synthetic feed to simulate salinity increase corresponding to each water recovery values as calculated from the FO experimental results. Pure nitrogen gas was used to flush the BMP bottles and a 1 M

sodium hydroxide (NaOH) solution was used to absorb the carbon dioxide (CO_2) and hydrogen sulphide (H_2S) from the biogas.

A thin film composite (TFC) FO membrane was used in this study and was supplied by Porifera (Porifera Inc., Hayward, CA). This had a polyamide active layer with a porous polysulfone layer for support. The membrane was positioned in FO mode (i.e. active layer facing the feed solution) for all experiments.

2.4 Experimental protocol

For the FO experiments, wastewater from the Wollongong WWTP was used as the feed solution. Analytical grade NaCl or NaOAc was dissolved in DI water to obtain the final concentration of 0.65 or 0.72 M, respectively, corresponding to the osmotic pressure of seawater (approximately 30 bar). The system water recovery was calculated based on the ratio of the cumulative permeate volume and the initial feed solution volume. The FO system was operated continuously until 90% of water had been recovered from the feed solution. The initial volume of wastewater feed solution was 2 L, corresponding to a total concentrate volume of 0.2 L. The water flux was continuously monitored, whilst the wastewater conductivity, pH, and temperature were frequently measured. At specific time intervals, samples of 10 mL volume were withdrawn from the feed solution for COD analysis to represent the organic content in solution. The circulation flow rates were maintained at 1 L/min giving a cross-flow velocity of 16.7 cm/s.

At the conclusion of the experiment, the membrane was flushed at a higher cross flow velocity for 30 minutes. This was achieved by replacing the feed and draw solutes with DI water and doubling the cross-flow velocity (i.e. 33.4 cm/s). After flushing, fresh wastewater was used as the feed solution to verify the water flux recoverability at the initial conditions.

After experimentally determining the pre-concentrated wastewater characteristics (i.e. COD and salinity), a synthetic wastewater solution and each draw solute was used to simulate the wastewater at 50, 80, and 90% water recovery. The COD results from the FO experiments were used to represent the COD increase in wastewater. The synthetic wastewater solution described in Section 2.1 was prepared to obtain the COD value at each corresponding water recovery, and also provided the expected salinity related to only FO rejection of feed water. Alternatively, the contribution of reverse solute flux was provided by adding a specified

amount of either NaCl or NaOAc to the synthetic wastewater solution. This reverse solute flux contribution ($Salt_{RSF}$) was estimated using salinity measurement assuming: $Salt_{RSF} = Salt_{Total} - Salt_{WW}$, where $Salt_{Total}$ is the measured salt concentration at each water recovery value and $Salt_{WW}$ is the calculated salt concentration from the wastewater due to FO rejection. This concentration ($Salt_{WW}$) was calculated using a mass balance, assuming complete rejection of any salts in wastewater as equivalent NaCl. The salinity of the feed solution was determined using electrical conductivity measurements and calibration curves were then used to determine salt concentration.

The simulated FO pre-concentrate was mixed with digested sludge in each BMP bottle. An inoculum volume of 500 mL and a substrate volume of 250 mL was selected, corresponding to an inoculum/substrate ratio of 2:1. A reference condition was used to represent the methane production of the inoculum, and real wastewater (i.e. FO feed solution with 0% water recovery) was also used as a separate condition for comparison to the synthetic wastewater. Prior to the BMP experiment, the bottles were purged with nitrogen gas, sealed, and submerged in the water bath. The flexible plastic tubing was connected to the biogas collection gallery. All BMP experiments were conducted in duplicate and biogas measurements were recorded daily. The contents of each bottle was characterised before and after the BMP experiment in terms of pH, conductivity, and COD.

2.5 Analytical methods

Standard methods were used during the analysis of basic water quality parameters. The temperature, pH, and electrical conductivity were monitored using an Orion 4-Star pH/conductivity meter (Thermo Scientific, Waltham, MA). COD samples were analysed using a Hach DBR200 COD Reactor and Hatch DR/2000 spectrophotometer (program number 435 COD HR) following the US-EPA Standard Method 5220 D. Total solids (TS) and volatile solids (VS) of the primary effluent were determined within three days after sample collection. All samples were stored at 4 ℃ in the dark.

2.6 Draw solute cost

Replenishment costs were calculated based on the pure water performance of the FO system at the draw solute concentration corresponding to 30 bar osmotic pressure. The replenishment cost only considered the loss of salt due to reverse draw solute flux. Losses from the draw

solute recovery process (i.e. RO or MD) were assumed to be insignificant. The initial cost of draw solution was also neglected as it can be reused in the process. Current average wholesale price of NaCl and NaOAc was used. The cost of draw solute replenishment per ML of permeate produced by the FO system was determined and a system water recovery of 90% was evaluated. Next, experimentally determined values of methane production and the draw solute replenishment costs were used to calculate the unit cost of methane production for each draw solute.

3. Results and discussion

3.1 Pre-concentration performance using thin film composite membrane

Pre-concentrating wastewater with the TFC FO membrane resulted in a substantial increase in COD (i.e. approximately eight-fold) at a water recovery of 90% (Figure 1). Organic matter enrichment for NaOAc was higher than NaCl, due to the contribution of organic reverse draw solute flux. For both draw solutions, the maximum COD was slightly lower than the theoretical COD amount. As no fouling mitigation strategy was implemented for this experiment, it is possible that surface deposition of organics was an important fouling mechanism, and thus, resulting in a lower bulk COD concentration than theoretically possible (i.e. ten-fold). In practice, the fouling layer can be re-suspended into the feed solution during membrane cleaning, and thus contribute to the feed COD amount.

[Figure 1]

Similar to the enrichment of COD in pre-concentrated wastewater, the level of salinity also increased as the FO experiment progressed (Figure 2). Wastewater conductivity increased for two reasons. The natural salinity of the wastewater (approximately 1 mS/cm) accumulated within the feed solution, due to rejection by the membrane and the concentrating effect. The reverse diffusion of the draw solute into the feed solution also contributed to salinity accumulation. The relative contribution of these two mechanisms is shown in Figure 2 and compared to the theoretically calculated conductivity increase due to the concentration of wastewater (i.e. ignoring reverse draw solutes flux). NaOAc exhibited a similar increase in conductivity compared with the theoretical wastewater salt accumulation, owing to the small amount of reverse solute flux (2.2 g/m^2h) compared with NaCl (12.4 g/m^2h). Deviation from the theoretical salt accumulation behaviour was likely due to the impact of flux dynamics and

membrane fouling on salt rejection at high water recoveries. In contrast, the reverse solute flux of NaCl contributed to salinity accumulation by approximately 50% higher than NaOAc. The results highlight the potential negative impacts associated with using highly diffusive inorganic draw solutions, such as NaCl.

[Figure 2]

3.2 Effect of forward osmosis concentration factor on methane production

Variations in wastewater characteristics at FO water recoveries of 50, 80, and 90% were simulated in batch anaerobic BMP experiments (Table 2). For both draw solutions, the conditions were simulated based on the experimentally determined values for salt concentration (i.e. conductivity) and organic content (i.e. COD) during the FO wastewater pre-concentration experiments. Wastewater COD was simulated using synthetic wastewater and the remaining conductivity requirement was supplied with the relevant amount of each draw solute (i.e. NaCl or NaOAc). Higher FO system water recovery resulted in an increase in both conductivity and COD (Table 2). It has been reported that conductivity and COD could have adverse and opposing effects on methane production by anaerobic treatment (Appels et al., 2008). COD loading up to 1,000 mg/L can significantly benefit the anaerobic process in terms of methane conversion. Conversely, high conductivity solutions can seriously affect methanogenic health and inhibit methane production.

[Table 2]

The cumulative methane production over a period of 30 days demonstrated the varying effect of FO water recovery and draw solute selection on the digestibility of pre-concentrated wastewater (Figure 3). Firstly, the methane production of real wastewater (i.e. unconcentrated) was only slightly higher than the reference condition (i.e. inoculum only) and can be attributed to marginal difference in total COD for these two conditions, as well as due to variations in the inoculum characteristics (Table 2). This result demonstrates the difficulties associated with digesting low-strength wastewaters for the purpose of biogas recovery. In all cases, pre-concentrating wastewater using FO improved the total wastewater COD, thus tended to increase methane production over the evalutation period. For both NaCl (Figure 3A) and NaOAc (Figure 3B), the cumulative methane production increased as the system water recovery was maximised. Overall, a minor effect of the reverse draw solute flux

on methane production was observed. This was likely due to the presence of sufficient biodegradable matter in the pre-concentrated wastewater, or because of the applied inoculum/ substrate ratio of 2:1, which may have masked the total salinity.

[Figure 3]

Methane production increased linearly with increasing pre-concentration factor and indicates an improvement in digester performance owing to the FO process. At the pre-concentration factor of ten (i.e. 90% water recovery), methane production was improved by approximatley five and seven times for NaCl and NaOAc, respectively (Figure 4). Comparing the two draw solutions, NaOAc could produce a larger amount of methane compared with NaCl (i.e. approximately 10%), due to lower reverse solute flux and degradable nature of NaOAc. Therefore, in terms of concentrated wastewater digestibility, no apparent negative effect on anaerobic treatment was observed when wastewater was pre-concentrated by ten times (equivalent to 90% water recovery) and with an inoculum/ substrate ratio of 2:1. Although FO reverse solute flux of inorganic draw solutions has been reported to negatively affect anaerobic treatment (Li et al., 2017), these results show that careful selection of FO operating parameters and digester loadings could potentially improve the process performance. In effect, pilot-scale assessment is necessary to evaluate the feasibility of operating at a high FO system water recovery and to determine the optimum anaerobic digester loading rate.

When comparing this process to the direct digestion of raw wastewater, a number of additional advantages of using FO to pre-concentrate wastewater exist. These include a substantially reduced digester volumetric loading (i.e. 10% of intial wastewater volume) and therefore, a smaller amount of anaerobic effluent. Furthermore, FO pre-concentration can provide a foulant-free draw solution for a subsequent desalination process to recover fresh water.

[Figure 4]

3.3 Unit cost of methane production

The costs associated with replenishing the draw solute as a result of reverse solute flux are shown in Table 3. Table 3 also includes the unit cost of methane production for both NaCl and NaOAc in terms of FO draw solute replenishment. Although the wholesale price of NaCl

is significantly lower than that of NaOAc, the high reverse solute flux of NaCl resulted in a slightly higher replenishment cost. The unit costs of methane production using NaOAc and NaCl were \$0.53 and \$0.64 per m^3 of methane, respectively. At 90% water recovery, there was about 10% increase in the volume of methane produced using NaOAc in comparison to NaCl (section 3.2). However, this contribution is insignificant compared to the difference in reverse solute flux between NaOAc and NaCl (Table 3). Results in Table 3 indicate that the unit cost of methane production is highly sensitive to the reverse solute flux. Further improvement in FO membrane fabrication is expected and can lower the cost of methane production from wastewater. It is noteworthy that Table 3 can be only used to compare the unit cost of methane production between NaOAc and NaCl. The calculation in Table 3 did not take into account the potential revenue from clean water production and further research is necessary for an overall economic analysis of methane production from pre-concentrated wastewater by anaerobic digestion.

[Table 3]

3.4 Water flux decline and flux recoverability

At the same osmotic pressure, water flux decline was evaluated for both NaCl and NaOAc (Figure 5). Although the initial water flux of NaOAc (16.6 L/m^2h) was slightly lower than that of NaCl (17.4 L/m^2h), both draw solutes exhibited a similar flux decline in the initial stages of the experiment. Subsequently, NaOAc fouling was more severe and indicated the possible interaction between the draw solute and membrane fouling layer (Luo et al., 2016; She et al., 2012). The total experimental duration to achieve 90% water recovery for NaCl and NaOAc was 65 and 72 hours, respectively. Despite the observed membrane fouling, 30 minutes of in-situ membrane flushing could completely recover water flux, indicating that no significant irreversible fouling occurred and that fouling was limited to surface deposition (Figure 5). The results in this study show that the rate of membrane fouling using the TFC membrane was higher compared with the CTA membrane used in a previous study (Ansari et al., 2016). This can mostly be attributed to the significantly larger initial water flux of the TFC membrane.

[Figure 5]

4. Conclusion

Pre-concentrating wastewater using the TFC FO membrane effectively concentrated COD by approximately eight-folds. Although the resultant pre-concentrated wastewater solution was highly saline, no apparent effect on methane production was observed for both draw solutes at the maximum water recovery value (i.e. 90%) during biomethane potential assessment. Overall, the pre-concentrated wastewater containing NaOAc resulted in a higher methane production to that of NaCl. Additionally, the unit cost of methane production using NaOAc was slightly lower than NaCl. FO membrane fouling was limited to surface deposition, thus, allowed for effective cleaning via membrane flushing at a high cross flow velocity.

Acknowledgements

This research was supported by the Australian Government Research Training Program Scholarship (Ashley Ansari) and the Australian Research Council's Discovery Project funding scheme (project DP140103864).

5. References

Ansari, A.J., Hai, F.I., Guo, W., Ngo, H.H., Price, W.E., Nghiem, L.D. 2016. Factors governing the pre-concentration of wastewater using forward osmosis for subsequent resource recovery. *Science of the Total Environment*, **566–567**, 559-566.

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

Appels, L., Baeyens, J., Degrève, J., Dewil, R. 2008. Principles and potential of the anaerobic digestion of waste-activated sludge. *Progress in Energy and Combustion Science*, **34**(6), 755-781.

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

Desmidt, E., Ghyselbrecht, K., Zhang, Y., Pinoy, L., Van der Bruggen, B., Verstraete, W., Rabaey, K., Meesschaert, B. 2014. Global Phosphorus Scarcity and Full-Scale P-Recovery Techniques: A Review. *Critical Reviews in Environmental Science and Technology*, **45**(4), 336-384.

Frijns, J., Hofman, J., Nederlof, M. 2013. The potential of (waste)water as energy carrier. *Energy Conversion and Management*, **65**, 357-363.

Jin, Z., Meng, F., Gong, H., Wang, C., Wang, K. 2017. Improved low-carbon-consuming fouling control in long-term membrane-based sewage pre-concentration: The role of enhanced coagulation process and air backflushing in sustainable sewage treatment. *Journal of Membrane Science*, **529**, 252-262.

Kim, Y., Chekli, L., Shim, W.-G., Phuntsho, S., Li, S., Ghaffour, N., Leiknes, T., Shon, H.K. 2016. Selection of suitable fertilizer draw solute for a novel fertilizer-drawn forward osmosis–anaerobic membrane bioreactor hybrid system. *Bioresource Technology*, **210**, 26-34.

Kim, Y., Li, S., Chekli, L., Woo, Y.C., Wei, C.-H., Phuntsho, S., Ghaffour, N., Leiknes, T., Shon, H.K. 2017. Assessing the removal of organic micro-pollutants from anaerobic membrane bioreactor effluent by fertilizer-drawn forward osmosis. *Journal of Membrane Science*, **533**, 84-95.

Li, S., Kim, Y., Phuntsho, S., Chekli, L., Shon, H.K., Leiknes, T., Ghaffour, N. 2017. Methane production in an anaerobic osmotic membrane bioreactor using forward osmosis: Effect of reverse salt flux. *Bioresource Technology*, **239**, 285-293.

Luo, W., Hai, F.I., Price, W.E., Elimelech, M., Nghiem, L.D. 2016. Evaluating ionic organic draw solutes in osmotic membrane bioreactors for water reuse. *Journal of Membrane Science*, **514**, 636-645.

Luo, W., Phan, H.V., Li, G., Hai, F.I., Price, W.E., Elimelech, M., Nghiem, L.D. 2017. An Osmotic Membrane Bioreactor–Membrane Distillation System for Simultaneous Wastewater Reuse and Seawater Desalination: Performance and Implications. *Environmental Science & Technology*, **51**(24), 14311-14320.

Nguyen, N.C., Nguyen, H.T., Chen, S.-S., Ngo, H.H., Guo, W., Chan, W.H., Ray, S.S., Li, C.-W., Hsu, H.-T. 2016. A novel osmosis membrane bioreactor-membrane distillation hybrid system for wastewater treatment and reuse. *Bioresource Technology*, **209**, 8-15.

Onoda, S., Masumori, Y., Miyoshi, T., Yasukawa, M., Takahashi, T., Matsuyama, H. 2017. Evaluation of energy production from municipal wastewater using forward osmosis process and anaerobic membrane bioreactor. *Desalination and Water Treatment*, **65**, 1-10.

Puyol, D., Batstone, D.J., Hülsen, T., Astals, S., Peces, M., Krömer, J.O. 2016. Resource Recovery from Wastewater by Biological Technologies: Opportunities, Challenges, and Prospects. *Frontiers in Microbiology*, **7**, 2106.

Qiu, G., Zhang, S., Srinivasa Raghavan, D.S., Das, S., Ting, Y.-P. 2016. Towards high through-put biological treatment of municipal wastewater and enhanced phosphorus recovery using a hybrid microfiltration-forward osmosis membrane bioreactor with hydraulic retention time in sub-hour level. *Bioresource Technology*, **219**, 298-310.

Shahzad, M.A., Khan, S.J., Siddique, M.S. 2017. Draw solution recovery using direct contact membrane distillation (DCMD) from osmotic membrane bioreactor (Os-MBR). *Journal of Water Process Engineering*.

403 Shannon, M.A., Bohn, P.W., Elimelech, M., Georgiadis, J.G., Marinas, B.J., Mayes, A.M.
404 2008. Science and technology for water purification in the coming decades. *Nature*,
405 **452**(7185), 301-310.

406 She, Q.H., Jin, X., Li, Q.H., Tang, C.Y.Y. 2012. Relating reverse and forward solute
407 diffusion to membrane fouling in osmotically driven membrane processes. *Water*
408 *Research*, **46**(7), 2478-2486.

409 Verstraete, W., de Caveye, P.V., Diamantis, V. 2009. Maximum use of resources present in
410 domestic "used water". *Bioresource Technology*, **100**(23), 5537-5545.

411 Wan, J., Gu, J., Zhao, Q., Liu, Y. 2016. COD capture: a feasible option towards energy self-
412 sufficient domestic wastewater treatment. *Scientific Reports*, **6**, 25054.

413 Wang, X., Chang, V.W.C., Tang, C.Y. 2016. Osmotic membrane bioreactor (OMBR)
414 technology for wastewater treatment and reclamation: Advances, challenges, and
415 prospects for the future. *Journal of Membrane Science*, **504**, 113-132.

416 Wang, X., Hu, T., Wang, Z., Li, X., Ren, Y. 2017a. Permeability recovery of fouled forward
417 osmosis membranes by chemical cleaning during a long-term operation of anaerobic
418 osmotic membrane bioreactors treating low-strength wastewater. *Water Research*,
419 **123**, 505-512.

420 Wang, X., Wang, C., Tang, C.Y., Hu, T., Li, X., Ren, Y. 2017b. Development of a novel
421 anaerobic membrane bioreactor simultaneously integrating microfiltration and
422 forward osmosis membranes for low-strength wastewater treatment. *Journal of*
423 *Membrane Science*, **527**, 1-7.

424 Wu, Y., Wang, X., Tay, M.Q.X., Oh, S., Yang, L., Tang, C., Cao, B. 2017. Metagenomic
425 insights into the influence of salinity and cytostatic drugs on the composition and
426 functional genes of microbial community in forward osmosis anaerobic membrane
427 bioreactors. *Chemical Engineering Journal*, **326**, 462-469.

428 Xie, M., Shon, H.K., Gray, S.R., Elimelech, M. 2016. Membrane-based processes for
429 wastewater nutrient recovery: Technology, challenges, and future direction. *Water*
430 *Research*, **89**, 210-221.

431 Zhang, H., Jiang, W., Cui, H. 2017. Performance of anaerobic forward osmosis membrane
432 bioreactor coupled with microbial electrolysis cell (AnOMEBR) for energy recovery
433 and membrane fouling alleviation. *Chemical Engineering Journal*, **321**, 375-383.

434

411 **List of Tables**

412 **Table 1.** Characterisation of real wastewater and digested sludge inoculum (average
413 concentration ± standard deviation from at least three samples).

Parameters	Units	Wastewater	Digested sludge
Chemical oxygen demand (COD)	mg/L	288 ± 10	4,000 ± 60
Electrical conductivity (EC)	µS/cm	977 ± 4	5,230 ± 8
Total solids (TS)	%	0.07 ± 0.02	1.7 ± 0.5
Volatile solids (VS)	%	0.03 ±0.01	1.1 ± 0.3

414

415 **Table 2.** Variation in pre-concentrated wastewater conductivity and COD simulated in BMP
416 experiments for NaCl and NaOAc. The calculated total COD in each BMP bottle (750 mL) is
417 also shown. Two BMP experiments were performed and each condition was conducted in
418 duplicate.

Condition	FO water recovery (%)	Pre-concentrated wastewater conductivity (µS/cm)	Pre-concentrated wastewater COD (mg/L)	Total COD in each BMP bottle (mg)
Reference	-	-	-	4,000
Real wastewater	0	977	288	4,072
Synthetic wastewater + NaCl	50	2,449	540	4,135
	80	7,846	1,079	4,270
	90	16,750	2,280	4,570
Synthetic wastewater + NaOAc	50	1,889	540	4,675
	80	6,122	1,079	6,306
	90	8,900	2,280	7,588

419

420

Table 3. Draw solute replenishment cost and unit cost of methane production using NaCl and NaOAc. Draw solute replenishment costs were based on the average wholesale salt cost and the pure water flux performance (J_w and J_s) for each draw solution at 30 bar osmotic pressure. Draw solute cost per methane produced was determined at 90% FO water recovery.

Parameter	**Units**	**NaCl**	**NaOAc**
Water flux (J_w)	L/m^2h	18.1	16.9
Reverse solute flux (J_s)	g/m^2h	12.4	2.2
Specific reverse solute flux (J_s/J_w)	$g/L_{permeate}$	0.69	0.13
Salt cost	$/kg	0.05	0.3
Replenishment cost	$/ML_{permeate}$	34.25	39.23
Specific methane production at 90% FO water recovery	L CH_4/ L substrate	0.48	0.66
Unit cost of methane production	$/m^3$ CH_4 produced	0.64	0.53

List of Figure Captions

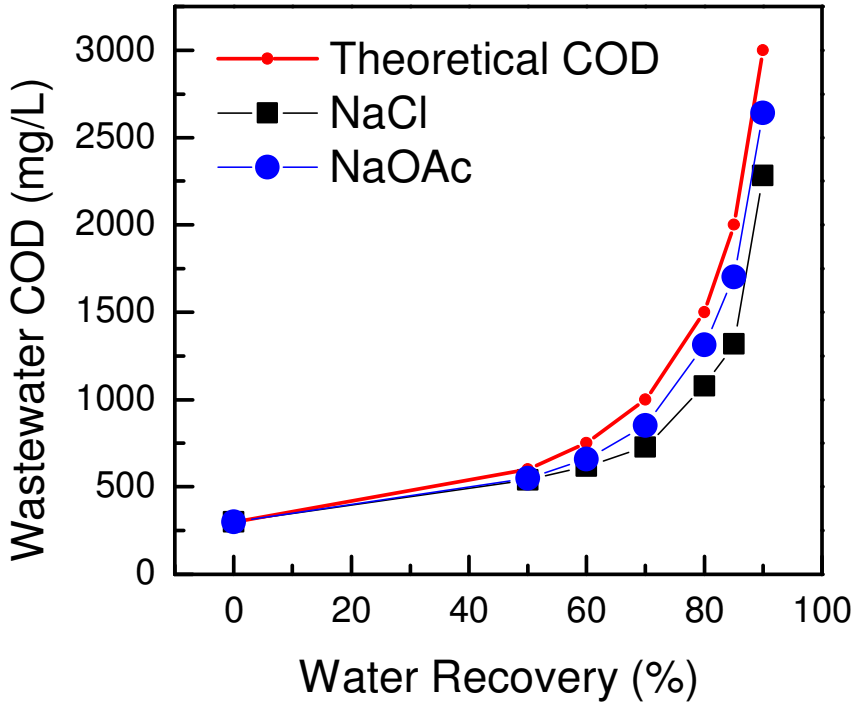
Figure 1: Pre-concentration of wastewater COD using NaCl and NaOAc draw solutions with the TFC FO membrane. Theoretical COD increase is shown assuming 100% COD retention. Experimental conditions: primary effluent feed solution (2 L); π = 30 bar draw solution; cross-flow rates of both feed and draw solutions were 1 L/min (corresponding to a cross-flow velocity of 16.7 cm/s).

Figure 2: Variation in wastewater conductivity for NaCl and NaOAc draw solutions. Theoretical salt accumulation ($Salt_{Acc}$) from natural wastewater salinity only (i.e. excluding reverse draw solute flux) is shown assuming 100% salt retention Experimental conditions as in Figure 6.1.

Figure 3: Average cumulative methane production over the 30 day evaluation period at various wastewater (WW) pre-concentration stages using (A) NaCl and (B) NaOAc FO draw solutions. Error bars represent n=4 measurements, including two BMP experiments with each condition performed in duplicate.

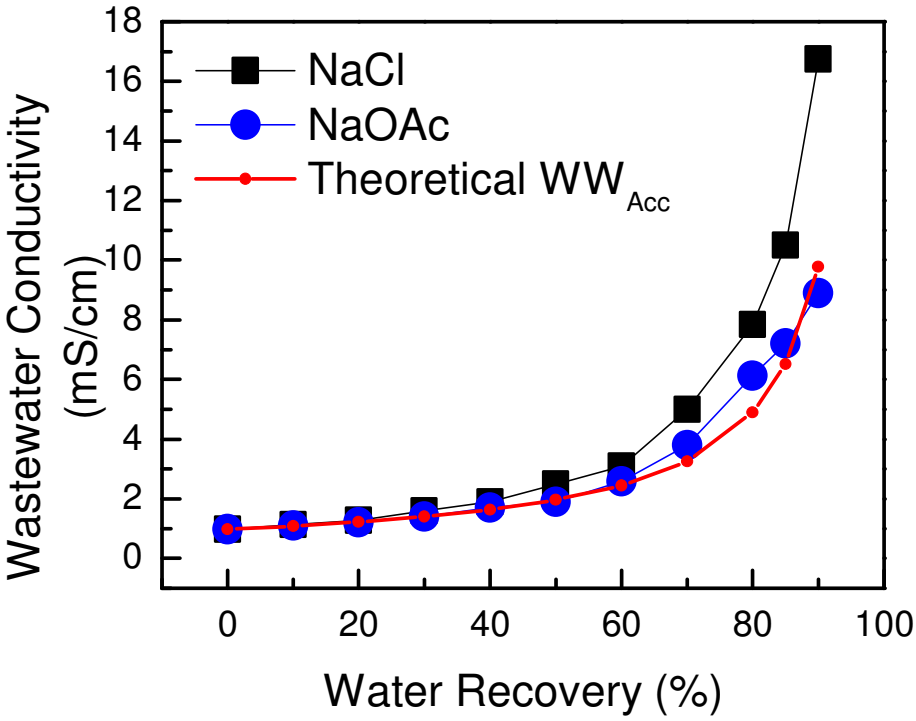
Figure 4: Specific methane production over the experimental period, indicating no negative effect of pre-concentrated wastewater up to 90% water recovery. Experimental conditions as in Figure 6.3. Error bars represent n=4 measurements, including two BMP experiments with each condition performed in duplicate.

Figure 5: Water flux decline and recoverability during FO pre-concentration with TFC membrane. After achieving 90% water recovery, membrane flushing was performed for 30 min using DI water at double the experimental cross-flow velocity (i.e. 33.4 cm/s)). Experimental durations corresponding to 90% recovery were 65 and 72 hours for NaCl and NaOAc, respectively. Initial water flux was 17.4 L/m^{2}h for NaCl and 16.6 L/m^{2}h for NaOAc. Experimental conditions as in Figure 1.



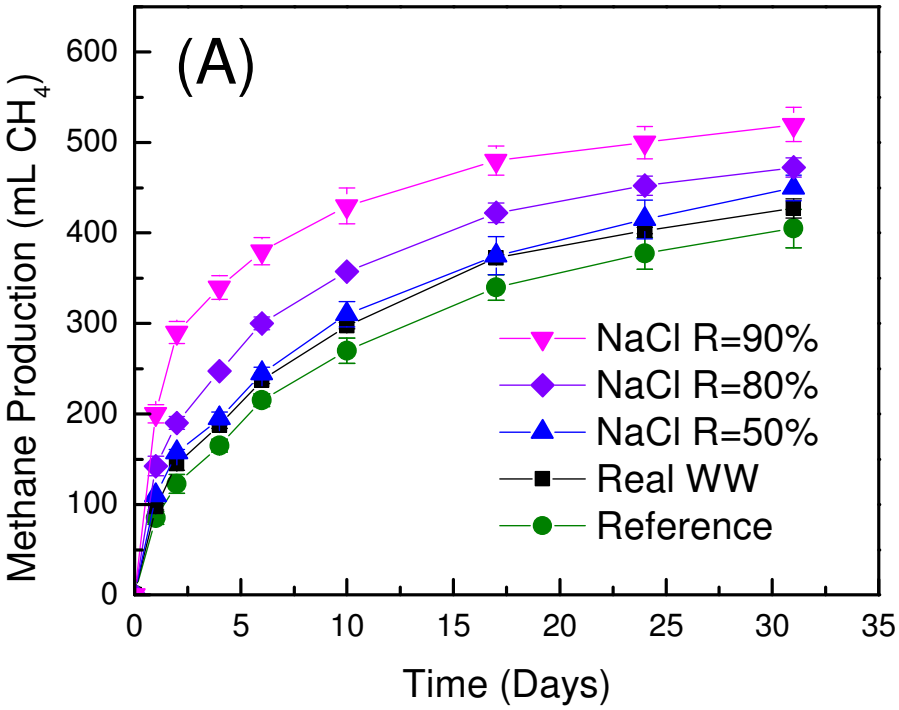
451

452 **Figure 1**

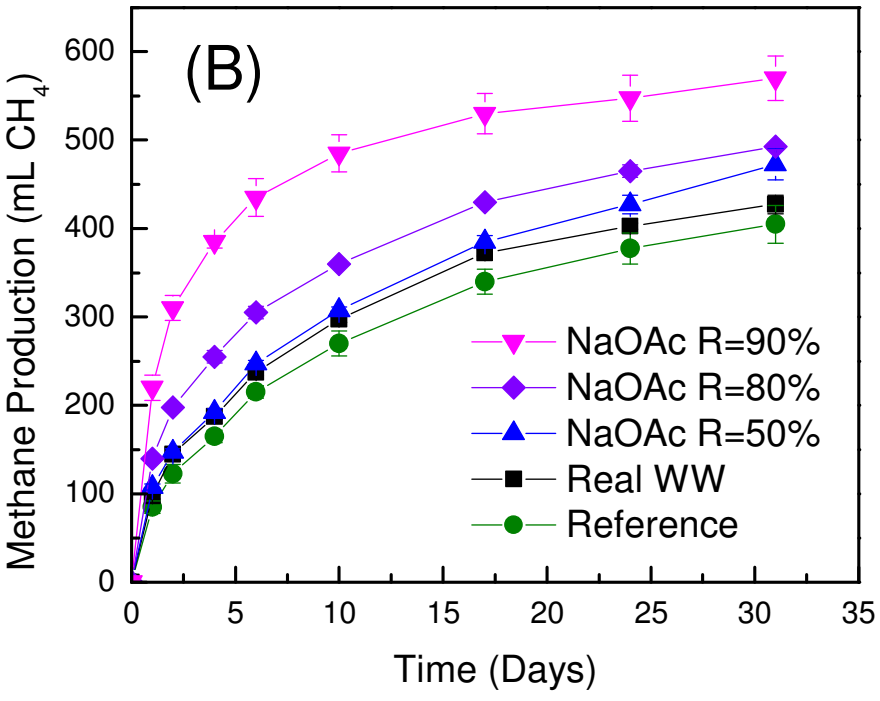


453

454 **Figure 2**



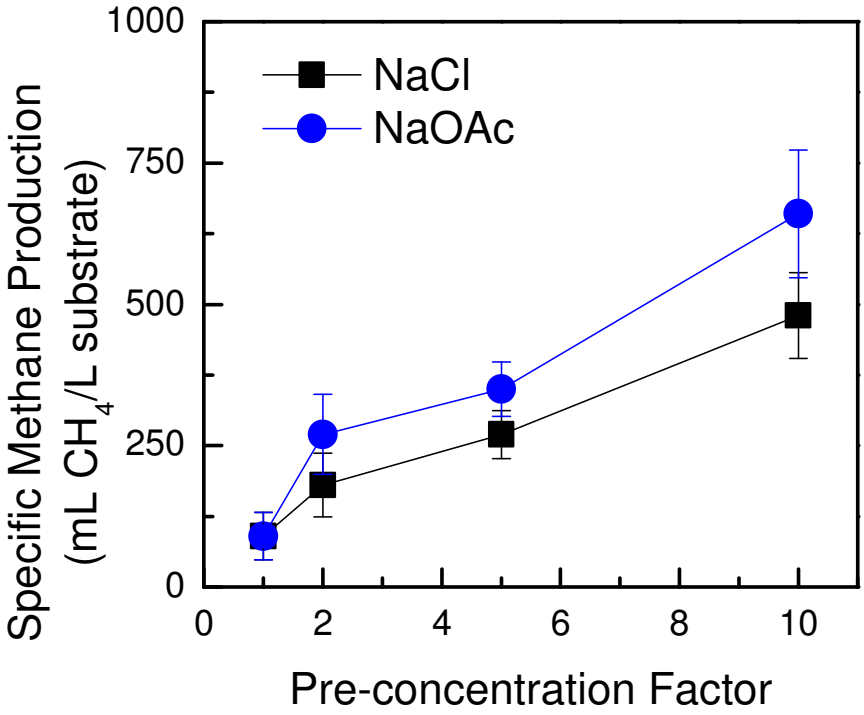
455



456

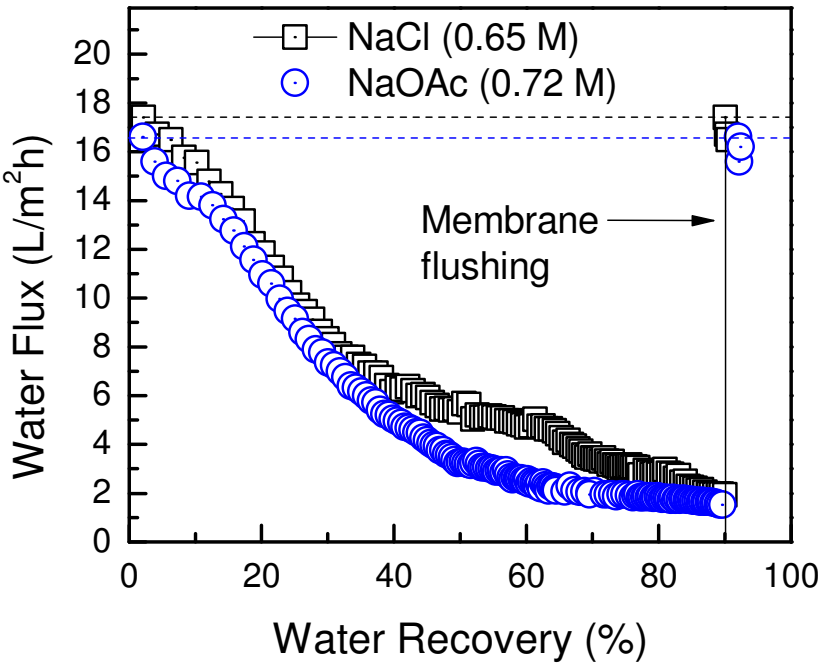
457 **Figure 3**

458



459

460 **Figure 4**



461

462

463 **Figure 5**