Factors governing the pre-concentration of wastewater using forward osmosis for subsequent resource recovery

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Abstract

This study demonstrated a technique using forward osmosis (FO) to pre-concentrate the organic matter in raw wastewater, thereby transforming low strength wastewater into an anaerobically digestible solution. The chem- ical oxygen demand (COD) of raw wastewater was concentrated up to approximately eightfold at a water recov- ery of 90%. Thus, even low strength wastewater could be pre-concentrated by FO to the range suitable for biogas production via anaerobic treatment. Excessive salinity accumulation in pre-concentrated wastewater was suc- cessfully mitigated by adopting ionic organic draw solutes, namely, sodium acetate, and EDTA-2Na. These two draw solutes are also expected to benefit the digestibility of the pre-concentrated wastewater compared to the commonly used draw solute sodium chloride. Significant membrane fouling was observed when operating at 90% water recovery using raw wastewater. Nevertheless, membrane fouling was reversible and was effectively controlled by optimising the hydrodynamic conditions of the cross-flow FO system.

Keywords: Forward osmosis (FO); Wastewater; Pre-concentration; Ionic organic draw solution; Anaerobic digestion; Membrane fouling

1. Introduction

The shift from aerobic to anaerobic biological treatment processes is a necessary step to achieve energy efficient wastewater treatment and to facilitate resource recovery practices (Frijns et al., 2013; Verstraete et al., 2009; Wei et al., 2014). Anaerobic treatment has two major advantages over aerobic treatment, namely energy recovery via methane production and reduced energy input, since aeration is not required (Appels et al., 2008). Furthermore, anaerobic effluent represents a practical platform for nutrient recovery (Ansari et al., 2016; Xie et al., 2014b).

In general, municipal wastewater is not suitable for direct anaerobic treatment. Indeed, given the low organic matter content of municipal wastewater (indicated by a chemical oxygen demand (COD) of usually b500 mg/L), the thermal energy and physical footprint required for anaerobic treatment can be excessive. Importantly, anaerobic treatment requires a feed solution in excess of 1000 mg COD/L to ensure system stability and process efficiency (Khanal, 2009). An innovative approach to overcome the challenges associated with the anaerobic treatment of municipal wastewater involves the initial pre-concentration of organic matter prior to feeding the digester.

The net energy recovery of anaerobic systems is theoretically proportional to the COD of the feed solution. Thus, pre-concentrating the organic matter in wastewater can significantly benefit the economics of anaerobic treatment processes. An ideal pre-concentration process would essentially separate water and non-aqueous components, to produce high quality water for reuse and a concentrate stream suitable for anaerobic treatment. Previously suggested methods include dynamic sand filtration, dissolved air flotation, and bio-flocculation (Frijns et al., 2013; Verstraete et al., 2009). However, these systems have limited organics retention capability and effluent from these processes still requires membrane filtration to produce water suitable for reuse. High rejection membrane processes such as nanofiltration (NF) and reverse osmosis (RO) can pre-concentrate the organic content of wastewater. Yet, they are not suitable for direct wastewater treatment and require extensive pre-treatment to control membrane fouling. Thus, the application of advanced separation technologies which can handle complex wastewater and achieve low energy treatment will be pivotal to developing sustainable wastewater treatment practices.

Forward osmosis (FO) is a membrane process with significant advantages when applied to wastewater treatment for fresh water production and resource recovery (Lutchmiah et al., 2014b; Xie et al., 2016). Unlike pressure driven membrane processes, the driving force of water permeation for FO is the osmotic pressure gradient between the feed solution (wastewater) and the draw solution (e.g. NaCl) (Cath et al., 2006). FO can directly pre-concentrate wastewater without significant external energy input (Alturki et al., 2013; Cath et al., 2006; Lutchmiah et al., 2014a,b). Furthermore, the nature of the driving force means that the process has a low fouling propensity and fouling can be highly reversible (Mi and Elimelech, 2010; Mi and Elimelech, 2013; She et al., 2016). Therefore, treatment of complex matrices such as wastewater by FO is feasible and key constituents including organic matter and nutrients can be retained in the concentrate. Fresh water can also be recovered from the draw solution by applying an additional desalination process such as NF (Nguyen et al., 2015), RO (Holloway et al., 2014; Luo et al., 2016), or membrane distillation (MD) (Nguyen et al., 2016; Xie et al., 2013). In particular, as a thermally driven desalination processes, MD presents a unique opportunity, as the required thermal energy could be supplied by solar thermal energy or from biogas cogeneration produced from the subsequent anaerobic digestion of pre-concentrated wastewater (Duong et al., 2016).

FO is recognised as a promising approach to pre-concentrate wastewater prior to anaerobic treatment (Ansari et al., 2015; Lutchmiah et al., 2014a; Wei et al., 2014; Zhang et al., 2014), however this approach is yet to be fully explored. Recent studies have focused almost exclusively on the integration of FO and anaerobic treatment to form an anaerobic osmotic membrane bioreactor (An-OMBRs) (Chen et al., 2014; Gu et al., 2015; Yin Tang and Ng, 2014) or to filter anaerobic effluent (Ding et al., 2014; Ding et al., 2016; Onoda et al., 2015). To date, very few studies have investigated the use of FO for direct treatment of municipal wastewater (Wang et al., 2016; Xie et al., 2013; Zhang et al., 2014). The FO wastewater pre-concentration concept allows for the simultaneous extraction of clean water for beneficial reuse whilst pre-concentrating wastewater to a higher strength suitable for anaerobic treatment. In this approach, a higher degree of control and accessibility exists for the FO component as it is not confined within a bioreactor, as is the case for An-OMBRs. In their recent work, Zhang et al. (2014) demonstrated the FO wastewater pre-concentration process, however due to the limitations of their experimental set-up, could only demonstrate a COD concentration factor of approximately three. Wang et al. (2016) presented the treatment performance of a spiral wound FO module to concentrate wastewater. Nevertheless, issues of salinity accumulation and anaerobic treatment integration were not addressed by Wang et al. (2016).

Although there is growing interest in the application of FO for wastewater pre-concentration and subsequent energy/resource recovery, the assessment of key performance factors has not been systematically investigated to date. Several challenges must be addressed for the implementation of the proposed FO wastewater pre-concentration process. Firstly, salinity accumulation is a major problem for high retention membrane systems such as FO, and particularly when combined with a sensitive biological process (Lay et al., 2010; Luo et al., 2014; Nawaz et al., 2013). Secondly, membrane fouling remains a prominent challenge for the sustained wastewater filtration of such complex wastewater solutions (Lutchmiah et al., 2011; Valladares Linares et al., 2013; Xie et al., 2013; Zhang et al., 2014).

This study aims to elucidate the key factors governing FO membrane performance during wastewater pre-concentration. The effectiveness of FO at pre-concentrating wastewater was examined by evaluating the ability of the FO membrane to retain COD at different water recoveries. Next, we evaluated the use of ionic organic draw solutes to mitigate salinity build-up. The effect of the selected draw solution on the produced clean water flux, COD, and pH of the concentrated wastewater was also investigated. Lastly, the extent of membrane fouling was assessed and hydrodynamic conditions were optimised. Both batch and continuous flow experiments were conducted to observe FO membrane fouling behaviour with real wastewater under intense pre-concentration conditions. Overall, this study proposes the importance of draw solution selection and optimised hydrodynamic conditions for the application of FO for wastewater pre-concentration.

2. Materials and methods

2.1. Materials and chemicals

Cellulose triacetate (CTA) membrane with a non-woven support was used in this study and was acquired from Hydration Technologies Innovation (Albany, Oregon, USA). The overall thickness of this non-woven CTA membrane is 144 μm. The average pore size is expected to be similar to that of a CTA membrane with embedded support which has been reported to be 0.37 nm by Xie et al. (2014a). Experiments were conducted with analytical grade draw solutes, at a constant osmotic pressure of 60 bar. The concentration of each draw solution at this pressure was calculated using OLI Stream Analyzer (OLI Systems, Inc., Morris Plains, New Jersey, USA). Sodium chloride, sodium acetate, and EDTA-2Na were used as draw solutions and the concentrations corresponding to 60 bar osmotic pressure were 1.27, 1.49, and 0.61 M, respectively.

Primary effluent (i.e. wastewater after primary sedimentation) was obtained from Wollongong Wastewater Treatment Plant (Wollongong, Australia). All batch experiments were conducted using both low and moderate strength wastewater, to represent the variability of municipal

wastewater influent quality. Moderate strength wastewater was obtained during a dry weather period. Low strength wastewater was obtained immediately after a wet weather period.

2.2. Forward osmosis system

A lab-scale, cross-flow FO membrane system was used. The membrane cell had two symmetric flow channels both with length, width, and height of 250, 50, and 2 mm, respectively, resulting in an effective membrane area of 125 cm^2. The feed and draw solutions were continuously circulated through each flow channel by two variable speed gear pumps (Micropump, Vancouver, Washington, USA). The flow rate was regulated by two rotameters and was adjusted to achieve the desired cross-flow velocity. The majority of experiments were operated with 1 L/min (corresponding to a cross-flow velocity of 16.7 cm/s). A spacer was placed on the draw solution side of the membrane to improve mixing.

The draw solution reservoir was positioned on a digital balance (Mettler-Toledo Inc., Hightstown, New Jersey, USA) and weight changes were recorded to calculate permeate water flux. A reservoir containing a highly concentrated stock solution (5 M) was also placed on the digital balance and was automatically dosed into the draw solution to maintain a constant osmotic pressure during experiments. The conductivity of the draw solution was monitored using a conductivity probe (Cole-Parmer, Vernon Hills, Illinois, USA), and was connected to a controller and a peristaltic pump to automatically regulate the draw solution concentration (control accuracy of ±0.1 mS/cm).

2.3. Experimental protocol

All experiments were conducted in FO mode (i.e. active layer facing the feed solution). Analytical grade solutes were dissolved in DI water at concentrations corresponding to an osmotic pressure of 60 bar. Water flux was measured according to the standard procedure previously described by Cath et al. (2013). Water recovery was used to represent the FO water extraction rate and was calculated based on the ratio of the cumulative permeate volume and the initial feed solution volume.

For batch experiments, the FO system was used to process primary effluent until a water recovery of 90% had been achieved. The initial volume of primary effluent (i.e. feed solution) was 2 L, and the solution was continuously filtered until 90% of the feed solution had permeated through the membrane (i.e. permeate volume of 1.8 L). Water flux was continuously monitored. The conductivity, pH, and temperature of each solution were also regularly measured. A 10 mL sample was withdrawn from the feed solution at specific time intervals for COD analysis as a measure of the strength of the wastewater or concentrated solution. All batch experiments were conducted in duplicate.

A continuous flow experiment was also conducted whereby 5 L of primary effluent was firstly processed to achieve 90% water recovery, leaving 0.5 L of pre-concentrated solution. At this point, the membrane was flushed with DI water to remove the fouling layer. The system was then continuously operated using a feeding and concentrate withdrawal regime (maintaining 90% water recovery). Two Masterflex peristaltic pumps (Cole-Parmer, Vernon Hills, Illinois, USA) were used to supply fresh primary effluent into the feed solution reservoir and to withdraw concentrate. The experiment was terminated approximately 90 h after membrane flushing, when the water flux had reduced to half of the initial water flux. Sodium chloride was used as the draw solution for all continuous flow experiments.

Detailed reverse solute flux experiments were conducted to elucidate solute transport behaviours of the ionic organic draw solutes. The feed solution consisted of 3 L of DI water and the respective draw solution had a constant osmotic pressure of 60 bar. The conductivity, pH, and temperature of solutions were measured hourly. The reverse draw solute flux of each draw solution was measured by monitoring the changes of conductivity in the feed solution over time. A 20 mL sample was also withdrawn from the DI water feed solution reservoir for subsequent analysis of sodium and total organic carbon (TOC) to determine the reverse solute flux of sodium, and acetate and EDTA, respectively.

2.4. Analytical methods

Key water quality parameters of the primary effluent were measured according to standard methods. COD was measured using a Hach DRB200 COD Reactor and Hach DR3900 spectrophotometer (program number 435 COD HR) following the US-EPA Standard Method 5220. Adequate dilutions and adjustments were made to minimise chloride interference during sample measurements. A Shimadzu analyzer (TOC-V_{CSH}) was used to determine TOC concentration. An inductively coupled plasma-optical emission spectroscopy (ICP-OES) system (ICP-OES 710, Agilent, Australia) was used to determine the sodium ion concentration in the samples. Temperature, pH, and electrical conductivity were measured using an Orion 4-Star pH/conductivity meter (Thermo Scientific, Waltham, MA).

3. Results and discussion

3.1. FO pre-concentration of organic matter in wastewater

Low strength wastewater can be pre-concentrated by FO up to the range suitable for anaerobic digestion (i.e. approximately 1000 mg COD/L). In this study, both low strength (137 ± 8 mg COD/L), and moderate strength wastewater (356 ± 13 mg COD/L) were pre-concentrated until 90% water recovery was achieved (Fig. 1A). The FO process predominantly extracted clean water, therefore enriching the concentration of organic matter in the feed solution. Results show that the FO process consistently pre-concentrated COD up to approximately eightfold, independent of the initial wastewater COD. The low and moderate strength wastewater COD concentrations were increased up to 982 ± 61 and 2893 ± 70 mg/L, respectively. These results demonstrate the suitability of FO for pre-concentrating wastewater, and its robustness for treating wastewater with variable influent quality. Furthermore, pre-concentrating wastewater with FO produces a reduced solution volume (i.e. ten times reduction at 90% water recovery) that is rich in organics and is arguably more amenable to anaerobic digestion compared to directly digesting raw wastewater.

The concentration of COD in wastewater increased proportionally with the FO system water recovery (Fig. 1B). The FO membrane effectively retained a large proportion of organic matter in the feed solution, shown by the comparability of the experimental COD concentration with the calculated mass balance (i.e. assuming 100% COD retention in the feed solution). The experimental results were only slightly lower than values obtained from mass balance calculation and this observation can possibly be explained by the accumulation of solid organics within the membrane cell. In other words, a portion of the bulk pre-concentrated wastewater COD gradually formed a cake layer on the membrane surface. Therefore, the measured feed solution COD concentration was lower than expected, particularly at high water recoveries where solids content was high. To a lesser degree, the observed COD pre-concentration behaviour may also relate to the incomplete rejection of COD by the FO membrane (i.e. 99% rejection) (Valladares Linares et al., 2013). Theoretically, the COD concentration factor could be further maximised by increasing water recovery, or when higher strength wastewater is used as the feed solution (i.e. N500 mg COD/L), yet this would further exacerbate the issues of salinity accumulation (Section 3.2.1) and membrane fouling (Section 3.3). The eightfold concentration of COD achieved in this study is substantially higher than previous studies (i.e. threefold COD concentration) (Zhang et al., 2014) and is attributed to the longer process filtration time and potentially the lower initial COD of the wastewater.

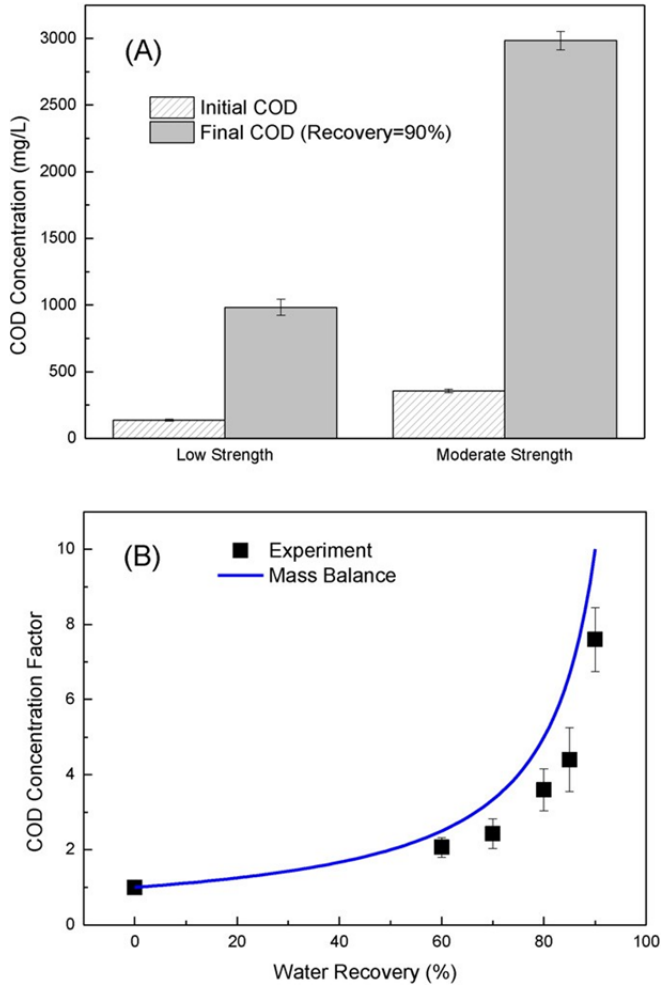

Fig. 1. (A) Initial and final (i.e. at water recovery of 90%) COD concentrations for low and moderate strength wastewater. Error bars represent the standard deviation of triplicate COD sample measurements. (B) Variation of experimental and calculated wastewater COD concentration factor during FO pre-concentration. Error bars represent the standard deviation of triplicate COD sample measurements from duplicate experiments. The initial wastewater COD for low and moderate strength wastewater were 137 ± 8 mg/L, and 356 ± 13 mg/L, respectively. Mass balance assumes 100% COD retention in feed solution. Experimental conditions: primary effluent feed solution (2 L); π = 60 bar, NaCl 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).

The enhanced organic content of FO concentrated wastewater can enable this solution to be fed into an anaerobic digester, and is arguably more effective when compared to direct anaerobic digestion of dilute wastewater. The net energy recovery from an anaerobic digester is theoretically proportional to the feed COD concentration, and therefore the FO system water recovery (Wei et al., 2014). Thus, the increased COD concentration of FO pre-concentrated wastewater would increase energy recovery per unit volume of digestate. Furthermore, since 90% of the initial water content has been extracted by the FO process for further treatment, the volume of feed that requires heating to optimum mesophilic conditions (i.e. 35 °C) during anaerobic treatment is lowered tenfold (when compared with raw wastewater). In addition, when the FO process is combined with other desalination processes, high quality water can be reclaimed for reuse (Chekli et al., 2016). Overall, FO presents a direct and robust approach to wastewater treatment, by focusing on pre-concentrating organic matter to facilitate subsequent anaerobic digestion for energy recovery.

3.2. Ionic organic draw solutes for wastewater pre-concentration

3.2.1. Salinity accumulation

Salinity accumulation is a major hindrance for high retention membrane systems such as FO, particularly when coupled with a biological process (Luo et al., 2014). Intensive pre-concentration of wastewater by FO leads to the accumulation of salinity in the feed solution via two mechanisms. Firstly, the natural salinity of wastewater is retained by the FO membrane, and therefore the salt concentration increases proportionally to the system water recovery. Secondly, salt leaks from the draw solution into the feed solution (i.e. reverse draw solute flux) and can also significantly contribute to salinity accumulation (Cath et al., 2006). Salinity accumulation in FO systems can have detrimental effects on water flux, as the osmotic pressure of the feed solution is increased, thereby reducing the effective osmotic driving force. More importantly for this application, high salt content within the pre-concentrated wastewater can have adverse effects on subsequent anaerobic treatment processes (Ansari et al., 2015).

A promising approach to mitigate salinity build-up in FO pre-concentrated wastewater involves the use of ionic organic draw solutes. When sodium chloride was used as the draw solution, the conductivity of wastewater significantly increased as water recovery increased (Fig. 2A). On the other hand, ionic organic draw solutes such as sodium acetate and EDTA-2Na presented a significantly lower conductivity compared to sodium chloride, demonstrating effective mitigation of salinity accumulation. A similar result was expected by Bowden et al. (2012) when using ionic organic draw solutes in an aerobic osmotic

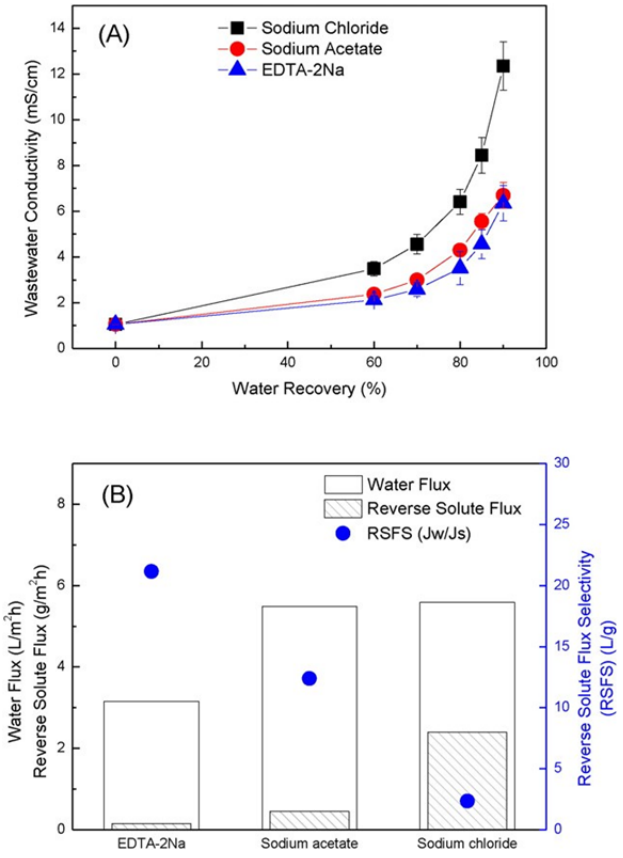

Fig. 2. (A) Variation of wastewater conductivity during wastewater pre-concentration for sodium chloride, sodium acetate, and EDTA-2Na. Experimental conditions: primary effluent feed solution (2 L); π = 60 bar draw solution; cross-flow rates of both feed and draw solutions were 1 L/min (corresponding to a cross-flow velocity of 16.7 cm/s). The initial wastewater conductivity was 1.05 ± 0.02 mS/cm. (B) Water flux, reverse solute flux, and RSFS of sodium chloride, sodium acetate, and EDTA-2Na. Experimental conditions: as above, with DI water feed solution (4 L). Error bars represent the standard deviation of measurements from duplicate experiments.

membrane bioreactor. Because each experiment pre-concentrated wastewater to 90% water recovery, the main contributor to the variance in salinity was the reverse draw solute flux. As shown in Fig. 2B, the extent of salt accumulation was inversely related to the magnitude of reverse solute flux selectivity (RSFS) for each draw solution. Both sodium acetate and EDTA-2Na exhibited a larger RSFS compared to sodium chloride, indicating that a smaller amount of solute diffused through the membrane for a constant permeate volume. Thus, adopting ionic organic draw solutions could achieve a pre-concentrated solution with a lower salinity, without compromising the achievable organic content in pre-concentrated wastewater.

The lower reverse solute flux behaviour of sodium acetate and EDTA-2Na can be explained by the mobility of the draw solute molecule. Both draw solutes have a lower diffusivity compared to sodium chloride, as acetate and EDTA ions are significantly larger than chloride (Ansari et al., 2015). Thus, solute diffusion from the draw solution to the feed solution is restricted. This has implications for the attainable water flux for each draw solution (Section 3.2.3). Binary ion analysis for sodium acetate showed a similar performance to sodium chloride, whereby both the cation and anion diffused into the feed solution at a similar rate (Fig. 3A). In contrast, binary ion analysis for EDTA-2Na revealed the potential decoupling of sodium and EDTA diffusion rates (Fig. 3B). In other words, sodium tended to diffuse through the FO membrane at a faster rate than EDTA. This is likely due to the large size and high negative charge of EDTA, minimising EDTA diffusion through the membrane (Hau et al., 2014). Nonetheless, despite the identified decoupling of the EDTA-2Na draw solute, compared to sodium chloride and sodium acetate, the reverse salt flux with respect to only sodium was still insignificant. The combination of EDTA with solutes other than sodium has also shown potential to minimise reverse solute flux and would greatly benefit the FO pre-concentration process (Nguyen et al., 2015).

3.2.2. COD content of pre-concentrated wastewater

In addition to mitigating salinity build-up, ionic organic draw solutes enhance COD when pre-concentrating low strength wastewater. At 90% water recovery, both sodium acetate and EDTA-2Na displayed higher COD concentrations compared to sodium chloride (Fig. 4A). This may be due to the reverse solute flux of the ionic organic draw solutes, enhancing the COD concentration of the low strength wastewater. Although reverse solute flux is generally viewed as a hindrance for the FO process, in the case of ionic organic draw solutes, the mechanism could be beneficial for subsequent anaerobic treatment. For example, unlike sodium chloride which inhibits methane production during anaerobic treatment, the presence of sodium acetate or EDTA-2Na in pre-concentrated wastewater can benefit methane production (Ansari et al., 2015). By adopting ionic organic draw solutes when treating low strength wastewater, opportunities exist to operate at a favourably lower water recovery, whilst attaining the desired COD range and allowable salinity level. On the other hand, for moderate strength wastewater, the contribution of reverse solute flux to COD concentration was negligible (Fig. 4B). The higher initial COD of the wastewater may have masked the contribution by reverse solute flux, and was possibly the reason why all three draw solutes displayed similar COD concentration performance.

3.2.3. Effect of draw solute on water flux decline

During the batch wastewater pre-concentration experiments, the choice of draw solute did not significantly affect water flux decline even at high water recovery values (Fig. 5). This suggests that both membrane fouling and salinity accumulation did not significantly contribute to water flux decline under these conditions (i.e. small processing volume and 90% water recovery cycle). As discussed in Section 3.3, continuous operation did result in more severe membrane fouling. For these batch experiments, the osmotic pressure of the pre-concentrated wastewater was significantly lower than the draw solution throughout the experiment. Flux decline was likely caused by the sparse accumulation of foulants on the membrane surface, as the implemented hydrodynamic conditions (i.e. increased cross-flow velocity) prevented excessive build-up of foulant materials.

Although the draw solution did not affect water flux decline, the initial water flux was significantly governed by the draw solution. Sodium chloride and sodium acetate gave similar initial water fluxes (5.5 and 5.4 L/m^2 h, respectively) at the same osmotic pressure (i.e. 60 bar), whilst the initial water flux of EDTA-2Na was significantly lower (3.3 L/m^2 h). EDTA-2Na exhibited the lowest water flux, owing to the negative effects of internal concentration polarisation (McCutcheon and Elimelech, 2006). This has limitations regarding the scale-up of FO systems using EDTA based draw solutions, since a large membrane area would be required. Nonetheless, since FO is an osmotically driven process, other operational costs would not be significantly impacted.

3.2.4. Effect of draw solute on pre-concentrated wastewater pH

For all three draw solutions, the wastewater pH gradually increased during the pre-concentration process (Fig. 6). This is a result of the net diffusion of hydrogen ions from the feed to the draw solution. Hydrogen ion diffusion occurs in order to maintain solution electroneutrality, as a result of reverse solute flux (Hancock and Cath, 2009; Xie et al., 2014b). When EDTA-2Na was used, the wastewater pH tended to increase at a fractionally slower rate compared with the other two draw solutions, and may be due to the significantly lower reverse solute flux rate of EDTA-2Na. Additionally, despite the lower reverse solute flux of sodium acetate compared to sodium chloride, the basic nature of highly concentrated sodium acetate solution may have contributed to the observed high wastewater pH. Results indicate that independent of the selected draw solution, FO pre-concentrated wastewater will have a high pH (approximately pH 8) and thus may need adjustment prior to feeding into an anaerobic reactor.

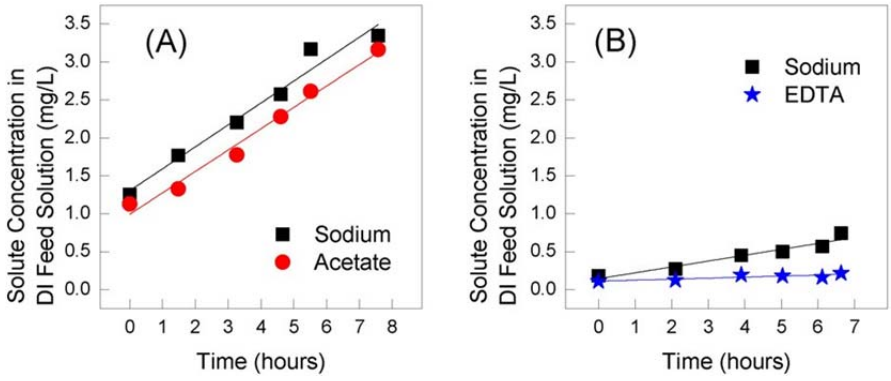

Fig. 3. Ionic organic draw solution binary ion diffusion analysis with linear regressions. (A) Sodium acetate and (B) EDTA-2Na. Experimental conditions: as in Fig. 2B.

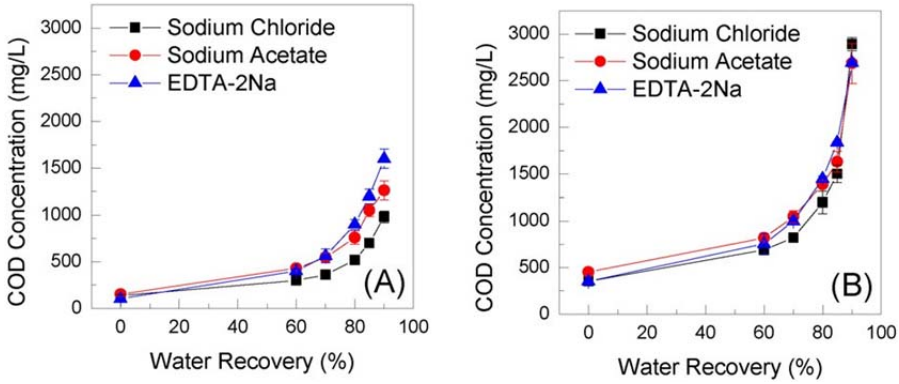

Fig. 4. Variation of COD concentration during wastewater pre-concentration for (A) low strength and (B) moderate strength wastewater. Experimental conditions: primary effluent feed solution (2 L); π = 60 bar draw solution; cross-flow rates of both feed and draw solutions were 1 L/min (corresponding to a cross-flow velocity of 16.7 cm/s). Error bars represent the standard deviation of triplicate COD measurements.

3.3. Membrane fouling

Sustained wastewater pre-concentration inevitably leads to membrane fouling. As shown in Fig. 7, when a cross-flow velocity of 9 cm/s was applied, severe membrane fouling was evident by a rapid water flux decline. Importantly, a water recovery of only 70% could be achieved as the water flux had reduced to below 1 L/m^2 h.

We increased the cross flow velocity as a hydraulic fouling control method during the continuous flow experiment. The difference in water flux decline patterns between the two cross-flow velocities (i.e. 9 cm/s and 17 cm/s) was significant (Fig. 7). When the cross-flow velocity was approximately doubled, water flux decline was considerably lower, and the target water recovery of 90% could be achieved in one cycle. Increasing the cross-flow velocity provides additional shear force, which hinders the accumulation of foulants on the membrane surface (Boo et al., 2013). For the higher cross flow velocity, the water flux decline was minimal up to a water recovery of 70%. From this point onwards, water flux declined more rapidly, most likely due to the increased solids content of the pre-concentrated wastewater at high water recoveries. Despite the flux declining by approximately half at a water recovery of 90%, the increased cross-flow velocity was shown to effectively reduce the rate of water flux decline for the complex pre-concentrated wastewater solution. High cross-flow velocity flushing regimes can be further optimised to lower the energy consumption of this membrane fouling control strategy. However, this aspect is beyond the scope of our current study.

3.4. Fouling reversibility and water flux sustainability

Increasing the applied cross-flow velocity resulted in less flux decline during wastewater pre-concentration. However, after one cycle, water flux still declined to approximately 50% of the initial value. After membrane flushing, the initial water flux was completely restored (Fig. 8), demonstrating the reversibility of FO membrane fouling. Furthermore, this water flux recoverability highlights the negligible contribution of feed water salinity increase to water flux decline. The FO process inherently inhibits fouling due to the nature of the osmotic driving force. The absence of hydraulic pressure promotes a loose and highly reversible fouling layer. In addition, FO generally operates at a low water flux and therefore a lower fouling rate (Shaffer et al., 2015). For these reasons, simple membrane flushing is a highly effective cleaning strategy.

Longer-term water flux behaviour was observed by continuously operating the FO system with the pre-concentrated wastewater solution (i.e. fixed 90% water recovery) after one pre-concentration cycle. In other words, after 70 h of operation, fresh primary effluent was fed into the FO feed solution and concentrate was withdrawn to maintain a constant 90% system water recovery. From 70 h onwards, the water

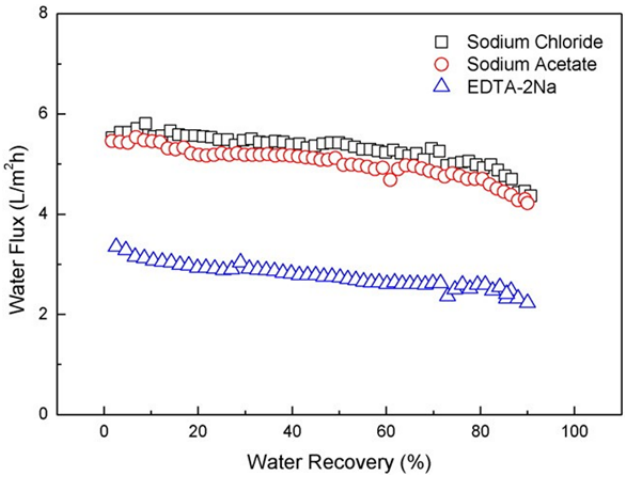

Fig. 5. Water flux decline during batch wastewater pre-concentration. Experimental conditions: primary effluent feed solution (2 L); π = 60 bar draw solution; cross-flow rates of both feed and draw solutions were 1 L/min (corresponding to a cross-flow velocity of 16.7 cm/s).

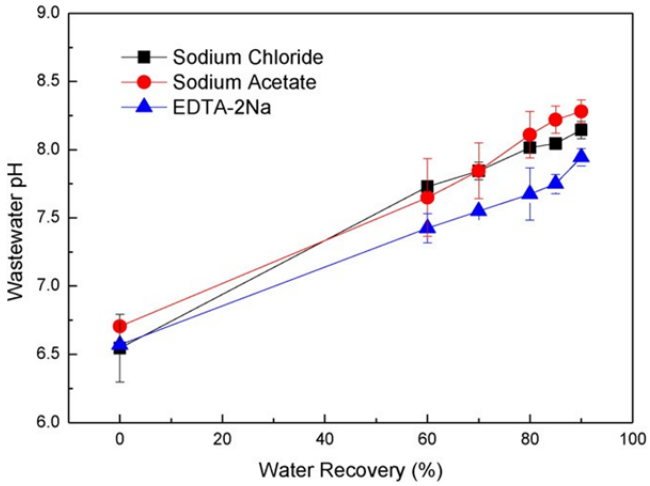

Fig. 6. Variation of pre-concentrated wastewater pH during batch wastewater pre-concentration experiments. Experimental conditions: primary effluent feed solution (2 L); π = 60 bar draw solution; cross-flow rates of both feed and draw solutions were 1 L/min (corresponding to a cross-flow velocity of 16.7 cm/s). Error bars represent the standard deviation of measurements from duplicate experiments.

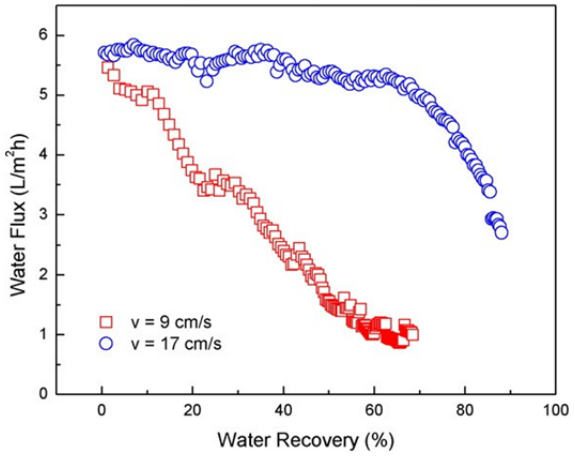

Fig. 7. Effect of applied cross flow velocity on water flux during the continuous flow experiment. Experimental conditions: primary effluent feed solution (5 L); π = 60 bar, NaCl draw solution; cross-flow rates of both feed and draw solutions were adjusted to achieve desired cross-flow velocity.

flux gradually declined due to the continuous exposure to the pre-concentrated wastewater. Interestingly, the rate of water flux decline gradually decreased and appeared to reach a steady state at approximately 150 h. This may indicate that the fouling cake layer had reached a maximum thickness, due to the cross flow conditions. Nonetheless, membrane fouling remains a prominent hurdle for FO systems and further efforts are required to investigate the effectiveness of other fouling mitigation methods during wastewater pre-concentration.

4. Conclusion

Pre-concentration of wastewater using FO presents a feasible approach to maximise the content of organic matter and possibly improve the digestibility of wastewater. In this study, the FO system achieved a COD concentration factor of approximately eight for low and moderate strength wastewater, at a water recovery of 90%. Specifically, FO allows for the pre-concentration of wastewater to the COD range (i.e. N1000 mg/L) suitable for biogas production via anaerobic treatment, even with low strength primary effluent obtained during wet weather. Furthermore, the importance of draw solution selection is emphasised, as ionic organic draw solutes benefited the pre-concentration process in two ways. Both sodium acetate and EDTA-2Na solutes effectively mitigated excessive salinity build-up in the pre-concentrated wastewater due to their lower reverse solute fluxes. Additionally, the ionic organic draw solutes enhanced the COD of low strength pre-concentrated wastewater, and are expected to benefit the digestibility of the solutions in terms of biogas production compared to sodium chloride. Significant membrane fouling was observed when operating at 90% water recovery using raw wastewater during the continuous flow experiment. However, this was reversible and could be controlled by optimising the hydrodynamic conditions during the FO process. Further developments of this FO wastewater pre-concentration process are recommended, including sustainable membrane fouling mitigation strategies and techno-economic evaluation at a pilot scale level.

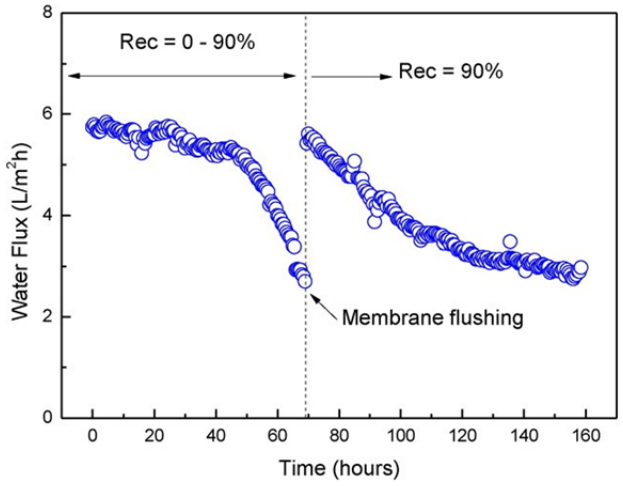

Fig. 8. Variation of water flux during the continuous flow experiment for one pre-concentration cycle and at a fixed 90% water recovery (i.e. Rec = 90%). Experimental conditions: primary effluent feed solution (5 L); π = 60 bar, NaCl draw solution; cross-flow rates of both feed and draw solutions were 1 L/min (corresponding to a cross-flow velocity of 16.7 cm/s).

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