



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



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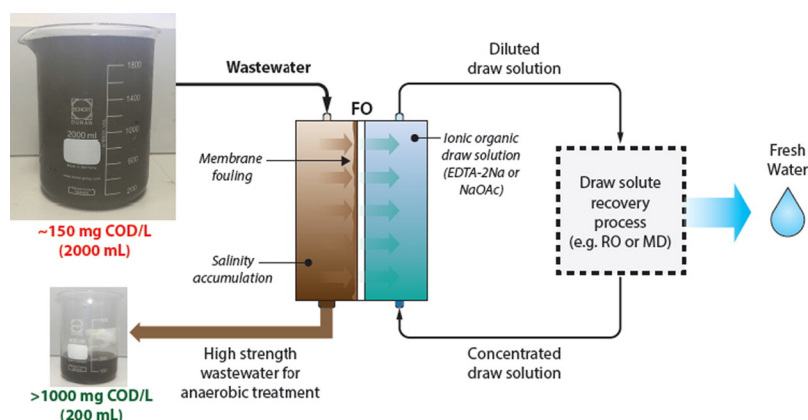
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### HIGHLIGHTS

- FO concentrates the COD of wastewater by up to approximately eight-fold.
- Low strength wastewater can be pre-concentrated by FO for anaerobic treatment.
- Ionic organic DS mitigates salinity accumulation in pre-concentrated wastewater.
- Membrane fouling is prominent, but can be fully recovered by membrane flushing.

### GRAPHICAL ABSTRACT



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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 chemical oxygen demand (COD) of raw wastewater was concentrated up to approximately eightfold at a water recovery 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 successfully 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.

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## 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 <500 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 co-generation 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  $\mu\text{m}$ . 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  $\mu\text{m}$  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

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