

# The Factors Affecting the Performance of the FO-RO Hybrid System

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**Abstract** - Forward osmosis (FO) and reverse osmosis (RO) hybrid process provides an opportunity for safe and beneficial reuse of wastewater also it provides a pre-treatment step for RO desalination. We investigated the influence of different parameters (draw solution concentration, cross-flow velocity rates, feed water pH and types of membranes and their properties and the applied pressure) on the FO process during the osmotic dilution of seawater as part of the simultaneous desalination and wastewater reuse by FO and RO hybrid system. Synthetic wastewater (SWW), containing combined organic and inorganic foulants, was used as the FS. The interactions between foulants and membrane were examined by using the CTA membrane and the TFC membrane. Membrane materials play an important role in the membrane-foulant interactions consequently on controlling membrane fouling and cleaning behavior in the FO process. Moreover, the elevated DS concentration boosts fouling of the FO membrane and diminishes cleaning efficiency. It was found that the cross-flow velocity was the dominant factor governing FO membrane fouling as the increase in the cross-flow velocity result in less fouling.

**Keywords:** Forward Osmosis, Hybrid System, Desalination, Wastewater Treatment, Membrane Fouling.

## 1. Introduction

The sustainability of water and energy resources are being threatened due to rapid population growth and therefore, developing low-energy separation technologies is crucial to meet the increasing water demand through unconventional sources [1-4]. Membrane technologies are currently the most widely applied techniques to produce clean water and reverse osmosis (RO) is the most employed membrane process for desalination (up to 70% of the installed desalination plants) [5]. Although the RO desalination plants consume significantly less energy than it was decades ago due to the efficient energy recovery devices and improved membrane materials, desalination still remains an energy-intensive process [6, 7]. Besides, the energy required for RO desalination has almost reached its thermodynamic limit and the remaining opportunities to further reduce its energy consumption will require additional processes which ultimately increase the total cost of the final water [8]. Moreover, RO still suffers from severe membrane fouling; affecting its long-term performance and the cleaning of membranes not only has considerable environmental issue but also pose a significant plant downtime. Therefore, any novel low-cost desalination technology that could circumvent those issues will have significant impact in sustaining the water and energy sources.

Recently, forward osmosis (FO) has received increased interest as an emerging low-cost desalination technology. The term low-cost has been often attributed to this process since it relies on a natural driving force (i.e. the osmotic pressure difference across the membrane) that draws the water from saline feed water (e.g. seawater) to a highly saline draw solution (DS). Apart from its apparent low-energy requirements, FO process also showed much lower fouling potential compared to other conventional pressure-driven membrane processes such as RO [9, 10]. Fouling has been found to be physically reversible in most cases, reducing the need for chemical cleaning [11, 12]. However, one of the main barriers that impede the commercialization of this process is the separation of the produced water from the draw solution [13-15]. In fact, the success of FO for clean water production is greatly dependent on how efficient (i.e. performance and cost) the DS separation and recovery process is [16].

In the last decade, several hybrid FO systems (i.e. FO coupled with another process) have been developed for various applications, including mainly seawater and brackish water desalination, wastewater treatment and both (i.e. simultaneously) [17]. For the latest, the hybrid FO-RO system has attracted increased attention since FO can be used as an advanced desalination pretreatment process to dilute the seawater and therefore moderate the energy requirement during

RO desalination [18-21]. Besides, the low salinity of most wastewaters makes them suitable candidates for this osmotic dilution [22]. The main advantage of this hybrid process is that the FO process operates in the osmotic dilution mode (i.e. both the concentrated feed and diluted draw solutions are the target) which eliminates the energy associated with the DS recovery process [23]. This hybrid process can be further extended if, after the RO process, the second FO process is used to further concentrate the wastewater which can be then used for agriculture applications (e.g. nutrient recovery) and, at the same time, dilute the RO brine for sustainable discharge.

In the present study, we investigated the long-term operation of the FO process during the osmotic dilution of the seawater using wastewater. This study focused on evaluating and understanding the fouling behaviour in the FO process for this specific application. The effect of different operating parameters (e.g. cross flow rate, feed water pH, applied pressure) on the fouling tendency has also been investigated.

## 2. Materials and Methods

### 2.1. Feed and Draw Solutions

The feed solution (FS) consisted of a synthetic wastewater (SWW), and its composition is shown in Table 1. This composition simulates secondary treated effluent usually found in the biologically treated sewage effluent (BTSE) [40]. All samples were filtered through 0.45  $\mu\text{m}$  membrane to measure dissolved organic carbon (DOC) by using the Dohrmann Phoenix 8000 UV-persulfate TOC analyser equipped with an autosampler.

Synthetic seawater (SSW) was used as the draw solution (DS) by mixing 0.6 M sodium chloride (NaCl) solution in DI water to obtain a final concentration of 0.6 M (i.e. conductivity: 55.1 mS/cm, TDS: 35 g/L, pH: 6.8).

### 2.2. Forward Osmosis Membrane

A commercial flat-sheet asymmetric cellulose triacetate (CTA) FO membrane (Hydration Technology Innovations or HTI, Albany, USA) and commercial thin film composite polyamide (TFC) FO membrane (Toray Industry Inc., Korea) were employed in this study. The CTA membrane was composed of a cellulose triacetate layer with an embedded woven support mesh [41-43]. The TFC membrane was made of a thin selective polyamide active layer on top of a porous polysulfone support layer [44].

Table 1. Composition of the synthetic wastewater used in this study.

Compounds	Concentration (mg L <sup>-1</sup> )	Main molecular weight (Daltons)
<b>Organic Compounds</b>		
Beef extract	1.8	298, 145, 65
Peptone	2.7	34265, 128, 80
Humic acid	4.2	1543, 298
Tannic acid	4.2	6343
Sodium lignin sulfonate	2.4	12120
Sodium lauryl sulphate	0.94	34265
Arabic gum powder	4.7	925, 256
Arabic acid (polysaccharide)	5.0	38935
<b>Inorganic Compounds</b>		
(NH <sub>4</sub> ) <sub>2</sub> SO <sub>4</sub>	7.1	-
K <sub>2</sub> HPO <sub>4</sub>	7.0	-
NH <sub>4</sub> HCO <sub>3</sub>	19.8	-
MgSO <sub>4</sub> •7H <sub>2</sub> O	0.71	-

### 2.3. FO Performance Tests for Seawater Desalination and Wastewater Reuse

The performance and fouling tests of the FO process were conducted using a lab-scale FO membrane unit consisting of an acrylic FO cell with internal dimensions of 7.7 cm length, 2.6 cm width, and 0.3 cm depth (effective membrane area of 2.0.10<sup>-3</sup> m<sup>2</sup>). The schematic layout of the FO-RO hybrid process is similar to the unit used in our previous study [29]. Both the FS and DS were supplied at cross-flow velocities of 8.5 cm/s (i.e. 400 mL/min or Reynolds number (Re): 455),

unless otherwise stated, under counter-current flow and in FO mode (i.e. active layer facing the FS). The temperature of the FS and DS was maintained at 25 °C using an automated heater/chiller control system connected to a water bath. All FO experiments were conducted in the batch mode of operation. The DS and FS were recycled back to their respective tanks after passing through the FO membrane cell thereby making process a batch operation. The initial volumes of both DS and FS were fixed at 2.0 L each. Before each experiment, the FO membrane was stabilised for 30 minutes using DI water on both sides of the membrane. Once stabilised, the FO water flux was measured continuously (i.e. with a 3-minute time interval) by placing the DS tank on a digital mass scale which was connected to a computer that automatically records the change in mass over time due to permeate flux.

Pressure-assisted osmosis (PAO) experiments were also conducted using the same FO membrane cell but the channel on the DS side of the FO cell was filled with spacers in the DS channel in order to prevent membrane deformation during pressure based operation. Spacers were of two different types one had a diamond shape that allows water to pass freely in the draw channel. The width and length of the spacers were designed to fit the channel dimensions in the test cell. The second type of spacers had smaller pore size and was obtained from commercially available spiral wound FO membrane module. Four sheets of this spacer were used to fill the remaining space in the draw channel. PAO process was operated at an applied pressure of 4, 8 and 12 bar.

### **3. Results and Discussion**

#### **3.1. Influence of Draw Solution Concentration and Membrane Properties on the FO Performance**

The type of membrane or the membrane properties is also known to strongly affect the rate of membrane fouling in the FO process [11, 26, 41, 45]. The surface properties of the active layer (such as roughness, charge, functional groups) and the structural characteristics of the support layer will dictate not only the water and solute transport across the membrane but also influence its interactions thus affecting the fouling behaviour on both side of the membrane [31]. The two most commonly employed FO membranes: CTA and TFC FO membranes were used for this study.

In fact, the surface of the PA TFC membranes has large-scale ridge-and-valley structures resulting in a much higher surface area favourable for foulants-membrane interaction [41, 46, 47] compared to much smoother surface for the CTA membrane [41, 48]. Interestingly, comparing the water flux patterns of peaks and lows for the two membranes, this regular pattern was observed much earlier for the TFC membrane compared to the CTA FO membrane. This is most likely due to the higher permeate flux obtained with the TFC membrane, resulting in higher hydrodynamic drag force and thus faster accumulation of foulants on the membrane surface compared to the CTA membrane. In fact, CTA membranes have generally lower water permeability compared to the PA-based TFC membranes [49].

To assess the influence of membrane properties and DS concentration on the FO process in the FO-RO hybrid system, the performance of the CTA membrane was compared with thin film composite polyamide (TFC) membrane. The water flux of six different NaCl concentrations (0.2, 0.4, 0.6, 1, 2 and 3 M) as the DS and DI water as the FS on both CTA and TFC membranes are presented in Fig.1. The presented results shows quite different water flux behavior for the CTA and the TFC membranes [46]. When synthetic seawater (i.e. 0.6 M NaCl) was employed as the DS, it resulted in a water flux of 9 LMH and 15 LMH for the CTA and TFC membranes, respectively. Besides, for all NaCl concentrations tested, the water flux was higher when the TFC membrane was employed. This is related to different support layer structure for the CTA and the TFC membrane [47].

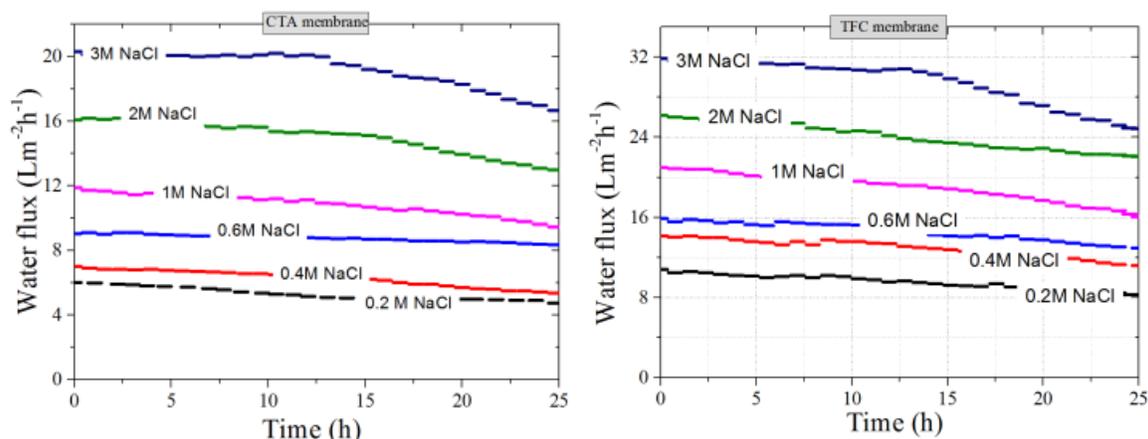


Fig. 1: Variation of permeate flux with time under different DS concentrations with (a) CTA membrane and (b) TFC membrane.

### 3.2. Influence of Feed Solution pH on the FO Performance

The feedwater composition and the feed water chemistry (pH, ionic strength and the presence and concentration of divalent ions) have been demonstrated to strongly affect the fouling behaviour in FO as it influences the properties (such as membrane surface charge) of the foulants as well as the foulant-foulant and foulant-membrane interactions [36-42]. In this study however, we investigated the effect of feed water pH on the development of flux pattern that we have described earlier [31]. Fig. 2 shows that the permeate flux obtained at higher pH (i.e. pH 8) was lowest while at pH 3 the water flux was not only higher but also the flux pattern observed was more pronounced. A visual observation of the feed water stored in the bottle shows that after 3 days, the foulant in the feed water at pH 3 formed large aggregates that settled down at the bottom of the bottle. This is most likely due to charge neutralisation that reduces the electrostatic repulsion as the foulant charge reaches closer to the point of zero charge resulting in the foulant aggregation [43]. These large aggregates is likely to deposit on the surface of the membrane probably forming a loose and more porous cake layer without significantly affecting the permeate flux or which may be easily be removed by under the influence of the velocity shear force on the membrane surface.

To further understand how the organic matter behaves under different pH conditions, feed water samples under different pH conditions were analysed using LC-OCD. The results show that, the feed water at pH 3 is characterised by the presence of higher percentage of larger molecular weight organic compounds compared to other pH conditions. Organic compounds with larger molecular weight have lower fouling potential compared to the lower molecular weight or size compounds since smaller size organic foulant can more easily penetrate through the membrane pores thereby causing pore blocking and more severe membrane fouling and flux decline [24]. This is likely the reason why at pH 8, a more severe fouling was observed. At pH 8, the organics in the feed water is mainly composed of hydrophilic compounds (87%) with higher percentage of lower molecular weight compared to the results obtained at pH 6.8. Therefore, at this pH, it is more likely that these smaller organic foulants penetrate the membrane pores and cause more severe fouling.

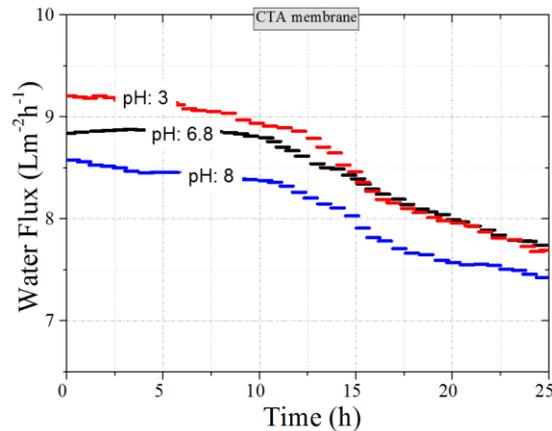


Fig. 2: Variation of the permeate flux with time during the FO process at different FS pHs.

### 3.3. Influence of Cross-Flow Rate and Pressure in the Performance of FO Process

Cross-flow velocity is a well-known hydrodynamic factor which affects the rate of membrane fouling as it directly influences the concentration polarization and mass transfer near the membrane surface [12, 25-27]. Three different cross-flow velocities were tested in this study: 100, 400 and 700 mL/min corresponding to cross-flow velocities of 2.1, 8.5 and 14.7 cm/s respectively. The results in Fig. 3 a indicate that the permeate flux improves slightly when the FO process is operated at higher cross-flow velocities which corroborates well with previous studies [12, 25-27]. In a cross-flow membrane filtration system, particles present in the feed water are subjected to two main forces: the hydrodynamic drag perpendicular to the membrane surface which forces the foulant particles to move towards the membrane and the shear rate tangential to the membrane surface which causes the particles to move back towards the bulk solution. Therefore, at high cross-flow velocities, the accumulation of foulant particles is reduced due to the higher tangential shear force [28]. At a lower cross-flow velocity (say at 100 mL/min in this study), the fouling cake layer formed is expected to be formed faster and thicker as the tangential shear force is lower. This is due to reduced mass transfer rate at the membrane surface which therefore increases the external concentration polarisation (CP) or ECP on the feed side facing the membrane active layer. At higher crossflow velocities, the water flux is slightly higher this further supports the earlier statement that related to the build-up of a loose foulant cake layer and its subsequent removal due to crossflow velocity shear.

Pressure-assisted osmosis (PAO) has been recently developed as a novel method for combining FO and RO principles whereby an additional hydraulic driving force is applied on the feed side of the FO process to simultaneously enhance the permeate flux and further dilute the DS beyond the point of osmotic equilibrium [29-40]. In this study, the PAO process was tested at three different operating pressures (4, 8 and 12 bar) and the results on the flux behaviour are presented in Fig 3 b.

As expected, the permeate flux increased significantly with the application of the hydraulic pressure with a specific flux gain of 1.31, 1.50 and 1.25 LMH/bar at 4, 8 and 12 bar, respectively as reported in the previous PAO studies [29, 36, 39-41]. At the same time however, more severe fouling was also observed at higher applied pressure which can be related to the more severe ECP effect as PAO process is operated at higher flux which increased the hydrodynamic drag force at increased flux and hence faster deposition and build-up of fouling cake layer. Besides, it has also been recently demonstrated that in PAO, in addition to the cake-enhanced osmotic pressure (i.e. CP effect), fouling layer compaction (i.e. comparable to the fouling behaviour in RO process) is also expected to occur at higher applied pressure [12]. This explains for the significant permeate flux drop observed at 12 bar applied pressure in Fig 3 b. By combining the hydraulic driving force and the osmotic driving force it increases the water flux and the fouling rate.

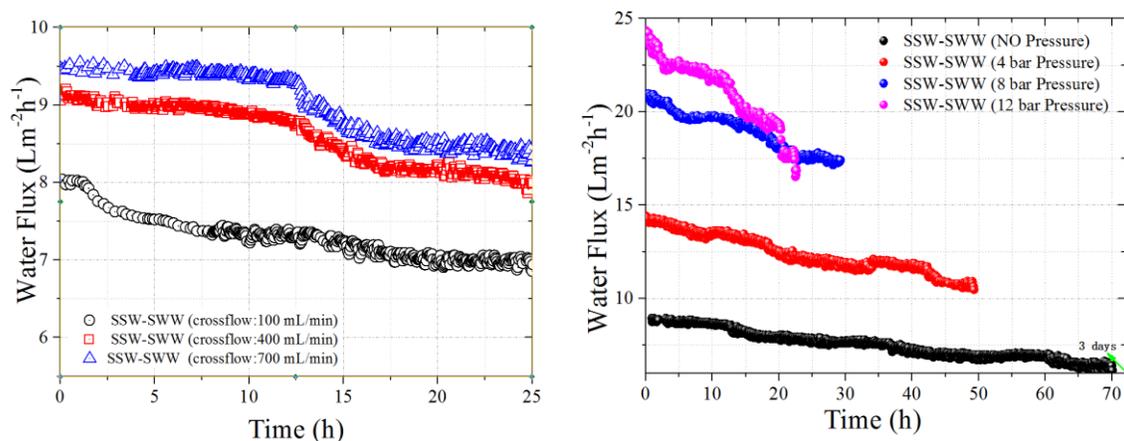


Fig. 3: (a) Variation of the permeate flux with time during the FO process at different cross-flow rates (b) at different applied pressures.

#### 4. Conclusions

This study investigated influences of different parameters in the FO and PAO processes during osmotic dilution of seawater using wastewater, a concept applicable for simultaneous wastewater treatment and seawater desalination by RO process. Results from the long-term FO and PAO operations revealed different flux behavior for CTA and TFC membranes that suggests the membrane materials play an important role during this experiment. The study also investigated the influence of various process operating parameters such as cross-flow velocity rates, feed water pH and types of membranes and their properties and the applied pressure on this FO performance. The FS pH could affect the interference between different foulants. The cross flow velocities also affect water flux behavior and membrane fouling, despite, the negative effect was more significant with low crossflow velocity. Applying high hydraulic pressure on the feed for improving the water flux caused increases the membrane fouling rate and severity. Operating the FO under the PAO mode significantly improves the water permeate flux but at the same time, increases the membrane fouling rate and severity. A simple physical or hydraulic cleaning was able to restore the initial flux by up to 90% for FO mode however, under the PAO mode, chemical cleaning was necessary to obtain similar level of flux recovery.

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