Challenges and potentials of forward osmosis process in the treatment of wastewater

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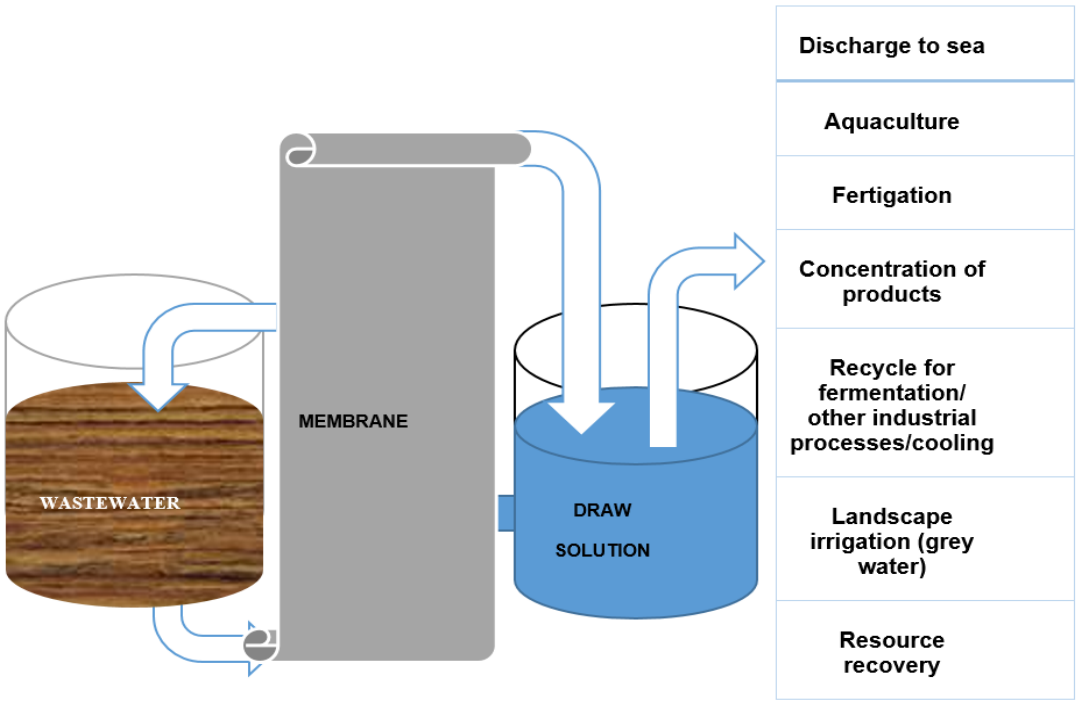
ABSTRACT

An emerging osmotically driven membrane process, forward osmosis has attracted growing attention in the field of desalination and wastewater treatment. The present study provides a critical review of the forward osmosis process for wastewater treatment focussing on most recent studies. Forward osmosis is one of the technologies that has been widely studied for the treatment of a wide range of wastewater because of its low fouling and energy consumption compared to conventional techniques for wastewater treatment. To date, forward osmosis has limited applications in the field of wastewater treatment due to several technical and economic concerns. Although membrane cost is one of the critical issues that limit the commercial application of forward osmosis, there are other obstacles such as membrane fouling, finding an ideal draw solution that can easily be recycled, concentration polarisation and reverse salt diffusion. Innovative technologies for in-situ real-time fouling monitoring can give us new insights into fouling mechanisms and fouling control strategies in forward osmosis. This study evaluated recent advancements in forward osmosis technology for wastewater treatment and the main challenges that need to be addressed in future research work.

Keywords: Wastewater, Forward Osmosis, wastewater treatment



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Nomenclature

A	**Water permeability constant, m/s.Pa**	**MNP**	**Magnetic nanoparticles**
AHA	Aldrich Humic Acid	NOM	Natural organic matter
AL	Active layer	PA	Polyamide
B	Solute permeability coefficient, m/s	PES	Polyether sulfone
CECP	Concentrative External polarisation	PRO	Pressure retarded osmosis
BSA	Bovine Serum albumin	PS	Polysulfone
CA	Cellulose acetate	R	Rejection of solute, %
CEOP	Cake enhanced Osmotic pressure	RO	Reverse Osmosis
CFV	Cross-flow velocity	RSD	Reverse salt diffusion
CP	Concentration polarisation	S	Membrane structure parameter, µm
ICP	Internal Concentration polarisation	t_s	Support layer thickness μm
CTA	Cellulose triacetate	T	Temperature, K
DICP	Dilutive internal concentration polarisation	TDS	Total dissolved Solids
D	Solute diffusion coefficient m^2/s	TFC	Thin film composite
DS	Draw solution	TOC	Total organic carbon
ECP	External concentration polarisation	UF	Ultrafiltration
EPS	Extra polymeric substances	π	Osmotic pressure, bar
FO	Forward Osmosis	σ	Reflection coefficient
Es	Specific power consumption (kWh/m^3)	*n*	Pump efficiency
FS	Feed side	τ	Tortuosity of the support layer
ICP	Internal concentration polarisation	ε	Porosity of the support layer
J_s	Salt flux, g/m^2 h	β	Van't Hoff coefficient
J_w	Water flux L/ m^2 h		
k	Mass transfer coefficient, m/s		
K	Salt resistivity, s/m		

1. Introduction

Water scarcity in the coming decades will severely affect society, ecological systems, food security, and environmental sustainability and may pose a significant threat to economic developments (Distefano and Kelly 2017). It is predicted that if the water is consumed at the current rate, by 2025, two third of the world's population may face water shortages (WWF 2018). The use of water for purposes other than sustenance (industrial processes) is therefore of great concern (Lutchmiah et al., 2014b). Amongst the various

methods to address water shortages are desalination, using waterless technologies in industrial processes, water storage in reservoirs, protecting wetlands, and several others. A possible alternative to alleviate global water scarcity is the reclamation and re-use of wastewater (Salgot and Folch 2018) using pressure-driven or membrane-based filtration techniques.

Amongst the many viable techniques for wastewater treatment, reverse osmosis (RO) is one of the most effective and widely used technology worldwide (Cath et al., 2006; Lee et al., 2010; Volpin et al., 2018). Despite its immense popularity, RO has several drawbacks such as high operating costs, CO_2 emissions, brine management, irreversible membrane fouling and requirements of extensive pre-treatment (McCutcheon and Elimelech 2006; Nguyen et al., 2018). Furthermore, RO cannot treat highly saline streams directly and is energy extensive due to using high hydraulic pressures (Chung et al., 2012). RO is also not an affordable solution due to its high capital costs (CAPEX) and operational costs (OPEX). Moreover, due to its high-energy consumption, RO can turn the water crisis into an energy crisis (Gilron 2014). Therefore, there is an urgent need to investigate cheaper, less energy-intensive and more sustainable processes for desalination and wastewater treatment.

Recently, a new osmotically driven membrane process, forward osmosis (FO) has attracted tremendous interest from researchers and scientists across the globe as one of the promising membrane processes and alternative to the RO process. The major advantage of FO over other pressure-driven membrane processes is that FO phenomena occurs spontaneously, and requires no hydraulic pressures (Mondal et al., 2017). While there has been tremendous research underway in the field of FO, approximately 30% of

FO publications are on wastewater studies since 2010 (Figure 1). Nevertheless, research in this field is still attracting a lot of attention due to the potential in wastewater treatment.

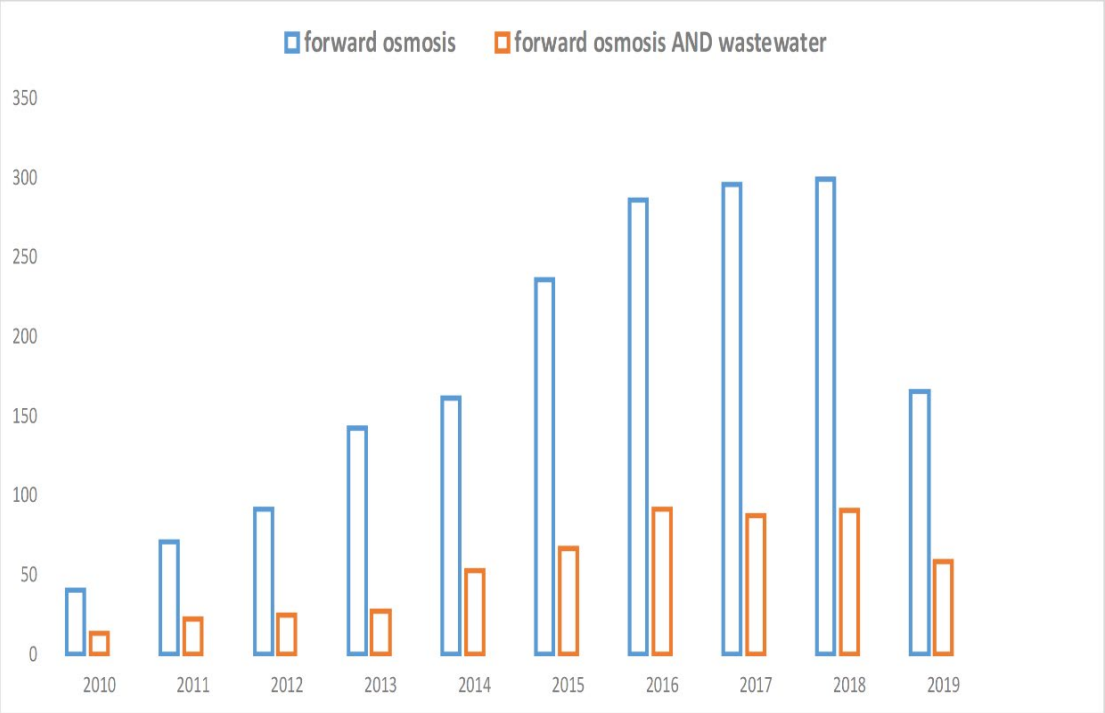


Figure 1. Number of publications on forward osmosis and forward osmosis for wastewater treatment since 2010 (search is performed on Scopus Sciencedirect using keywords forward osmosis and forward osmosis wastewater. Search is limited to research and review articles only.

Shortcomings of FO includes membrane fouling, concentration polarisation, reverse solute diffusion, lack of high selective membranes, selection of appropriate draw solution and most importantly the regeneration of the draw solution (Corzo et al., 2017; Korenak et al., 2017; Lutchmiah et al., 2014b; McCutcheon and Elimelech 2006; McGovern and Lienhard V 2014). This paper evaluates the progress in the FO process for wastewater treatment addressing draw solutions, FO membranes selection, the impact of key operating parameters on the FO process, membrane fouling and fouling mitigation strategies, in-situ real-time fouling monitoring techniques, and lastly pilot-scale studies on FO for wastewater treatment.

2. The forward osmosis process, evolution of the current flux model

FO models were designed to predict water flux in across the membrane (Wang et al., 2014). These models were evolved over time to include different physical parameters, reflecting our increased understanding of the phenomenon of osmosis flux in the FO membrane. Lee et al., (1981) was the first to introduce a flux model for pressure-retarded osmosis (PRO) in the presence of concentration polarisation (CP). PRO performance was predicted from the FO and RO measurements. Loeb et al., (1997) later described the model of Lee et al., (1981) for the FO process. This model was further revised by McCutcheon and Elimelech (2006) to incorporate the effects of concentration polarisation (Figure 2) on flux behaviour.

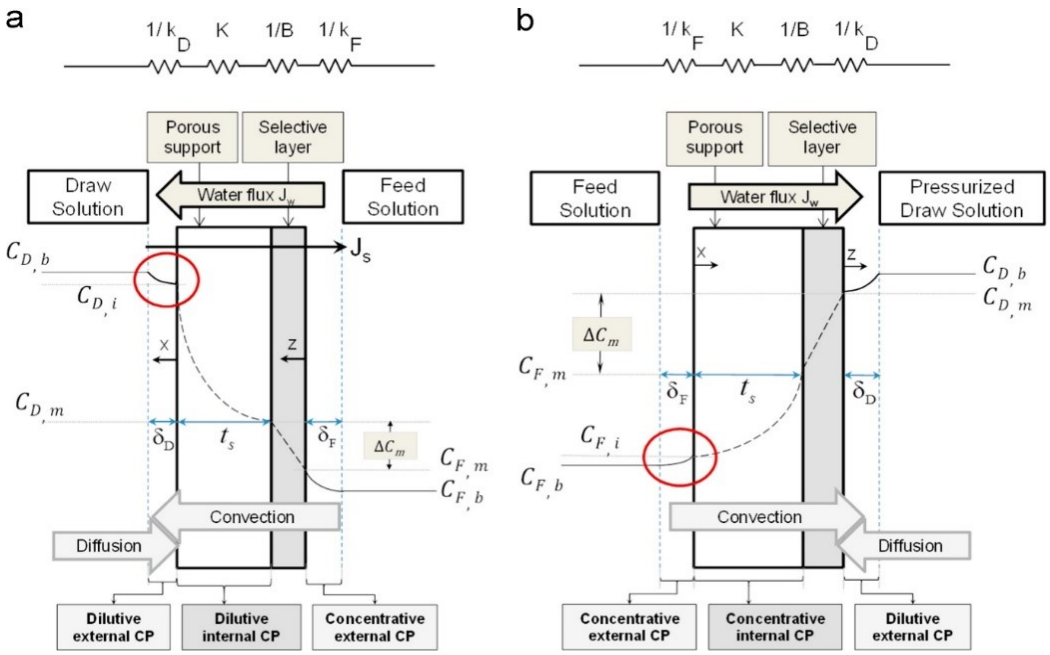


Figure 2. Solute Concentration Profiles at steady state across a TFC membrane in (a) FO mode (b) PRO mode. Reprinted from Bui et al. (2015) with permission from Elsevier

When the feed solution faces the support layer and the draw solution faces the active layer (PRO mode), the effects of concentrative internal concentration polarisation (CICP) are coupled with dilutive external concentration polarisation (DECP), with the flux being predicted as given by equation [1].

$$J_w^{PRO} = A\left[\pi_{D,b}\exp\left(-\frac{J_w}{k}\right) - \pi_{F.b}\exp(J_w K)\right] \quad [1]$$

For the FO membrane with active layer faces the feed solution and the support layer faces the draw solution (FO mode), water flux is given by equation [2].

$$J_w^{FO} = A\left[\pi_{D,b}\exp(J_w K) - \pi_{F.b}\exp\left(-\frac{J_w}{k}\right)\right] \quad [2]$$

Where k is the mass transfer coefficient, and K is the measure of how quickly a solute can diffuse into and out of the support layer and is the measurement of the severity of internal concentration polarisation (McCutcheon and Elimelech 2006). Although the above equations account for ECP and ICP in the FO process, they ignore the effects of reverse salt diffusion (RSD) in the FO process and are valid only under the assumption that FO membrane has a complete rejection to solutes.

The McCutcheon and Elimelech (2006) model does not account for the effects of reverse salt diffusion in the FO membrane. Tan and Ng (2008) criticized the model developed by McCutcheon and Elimelech (2006) and found that it over-predicts the water flux at draw solution concentrations more than 1.0 M NaCl. Yip et al., (2011) developed a mathematical model to predict water flux in the FO process including the effects of internal and external concentration polarizations with reverse salt diffusion from the draw to the feed solution:

$$J_w^{FO} = A\left\{\frac{\pi_{D,b}\exp(-J_w K) - \pi_{F,b}\exp(\frac{J_w}{k})}{1 + B/J_w\{\exp(J_w/k) - \exp(-J_w K)\}}\right\} \quad [3]$$

$$J_w^{PRO} = A\left\{\frac{\pi_{D,b}\exp(-\frac{J_w}{k}) - \pi_{F,b}\exp(J_w K)}{1 + B/J_w\left\{\exp(J_w K) - \exp(-\frac{J_w}{k})\right\}}\right\} \quad [4]$$

Although Yip et al., (2011) equations considered the effects of internal and external concentration polarisation and reverse salt diffusion that is illustrated in the denominator; they ignored mass transfer resistance at the porous support layer. Practically, the effects of ECP occurring on the porous support side of the membrane becomes substantial and cannot be ignored when the FO membrane operates at a high water flux or at low cross-flow velocities (Gruber et al., 2011; Nagy 2014). Nagy (2014) introduced a mathematical model combining the effects of concentration polarization with the effects of ECP on the porous support layer.

$$J_w^{FO} = A\left\{\frac{\pi_{D,b}\exp\left[-J_w(\frac{1}{k_D} + S/D_D\right] - \pi_{F,b}\exp(\frac{J_w}{k_F})}{1 + B/J_w\left\{\exp(J_w/k_F) - \exp\left[-J_w(\frac{1}{K_D} + \frac{S}{D_D})\right]\right\}}\right\} \quad [5]$$

$$J_w^{PRO} = A\left\{\frac{\pi_{D,b}\exp(-\frac{J_w}{k_D}) - \pi_{F,b}\exp\left[J_w(\frac{1}{k_F} + \frac{S}{D_F})\right]}{1 + B/J_w\left\{\exp\left[J_w(\frac{1}{K_F} + \frac{S}{D_F}) - \exp(-\frac{J_w}{k_D})\right]\right\}}\right\} \quad [6]$$

Where, S is the membrane structure parameter, and D_D is the diffusion coefficient of the draw solution. Technically, the impact of ECP at the porous support layer is responsible for less than 10 percent water flux decline in the forward osmosis process (Altaee et al., 2017; Bui et al., 2015). Recent laboratory-scale study on a flat sheet

membrane showed that the effects of CP depend on technical and operating parameters such as type of membrane, solutions concentrations, feed temperature, cross-flow velocity, and membrane orientation (Altaee et al., 2017). Membranes with a denser structure, such as HTI, resist internal mixing, resulting in a severe internal concentration polarization (Hawari et al., 2016). Such membranes benefit from increasing the cross-flow velocity in reducing the effect of internal concentration polarization (Zhang et al., 2010). The study also revealed that the increase in water flux resulted in more intensified ICP, but it was lower at high cross-flow velocities. Therefore, many studies recommended high cross-flow velocity to alleviate the effects of concentration polarization.

In the FO process along with the water flux, there is also reverse salt flux from the draw solution to the feed. Reverse salt flux is an intrinsic property of all osmotically driven membrane processes and has adverse effects on membrane performance in the FO process (Phuntsho et al., 2011). The reverse salt flux in the FO process can be estimated by the following equation (Johnson et al., 2018).

$$J_s = B\left[\frac{C_{Db}\exp\left(-\frac{J_w}{k}\right) - C_{Fb}\exp\left(\frac{J_w S}{D}\right)}{1 + \frac{B}{J_w}\left[\exp\left(\frac{J_w S}{D}\right) - \exp\left(-\frac{J_w}{k}\right)\right]}\right] \quad [7]$$

Where C_{Db} and C_{FB} are the bulk concentrations of the draw and feed solutions respectively. Reverse salt diffusion has adverse consequences such as decreasing the net driving force across the membrane, increasing the loss of draw solution, contaminating the feed solution (when certain draw solutions are used) and promoting membrane fouling.

3. Draw solutions in wastewater treatment applications

Analysis of the FO literature reveals that finding an ideal draw solution and efficient membrane are the main obstacles towards the commercialization of the process (McCutcheon et al., 2005). Despite the wide range of draw solutions, it is still believed that the selection of an appropriate draw solution is paramount for an efficient FO process (Cai and Hu 2016). The criteria for ideal draw solution are high osmotic pressure, low reverse salt diffusion, high diffusion coefficient to reduce ICP, low viscosity to allow easy pumping around the FO system, and ability to regenerate at a competitive cost (Cai and Hu 2016, Johnson, et al. 2018, Phuntsho, et al. 2011). Technically, there is not a single draw solution that meets every criterion of an ideal draw solution, and this continues causing confusion about a standard draw solution for a specific application such as wastewater treatment. For example, NaCl has been used widely in the FO experiments because it has high osmotic pressure, small molecular size, and high diffusion coefficient, but at the same time, it exhibits high reverse salt flux due to its smaller molecular size. On the other hand, draw solutions containing divalent ions such as $MgSO_4$ and $MgCl_2$ have lower reverse salt flux than NaCl, but the presence of divalent magnesium and calcium ions promote organic fouling by complexation and formation of intermolecular bridges among organic molecules. Diffusivity of draw solution is influenced by other factors such as temperature, viscosity and particle size (Lutchmiah et al., 2014b). According to Ge et al., (2017) viscosity of a draw solution is linked to its diffusion coefficient and inversely proportional to the water flux in the FO process although high viscosity hydrogels draw solution exhibits high water flux (Zhang et al., 2015). Hydrogel draw solution requires heat energy for regeneration and freshwater separation, which increases the cost of FO treatment. The economic availability of draw solution is also a factor to consider in selecting draw solutes. Synthesized draw solutions

in the FO studies such as magnetic nanoparticles, polyelectrolytes, zwitterions, and hydroacid complexes have excellent osmotic pressures and low reverse salt flux (Cui et al., 2014; Ge et al., 2012; Lutchmiah et al., 2014a; Na et al., 2014). Unfortunately, these draw solutions are rather expensive and have an intricate synthesis process, which complicates their commercial application.

Table 1 provides information about draw solutions and membranes used in the treatment of various wastewaters. Regeneration process is the most energy-intensive stage in the FO process but several FO applications, successfully eliminated this stage (Ansari et al., 2016; Gwak et al., 2018; Kalafatakis et al., 2017; Korenak et al., 2019; Takahashi et al., 2016; Zou and He 2016). Ansari et al., (2016) used real seawater draw solution to recover calcium phosphate from a digested sludge. CTA FO membrane (HTI) was used, and the reported water flux was 6.4 L/m^2h. In this study, the regeneration stage was not required for recovery of the draw solution and hence reduced the energy requirements of the FO process. The problem with indirect desalination is there are several ethical and environmental guidelines that strict the application of product water. In another study, a flat sheet CTA FO membrane (HTI) was used for fertilizer draw solutions preparation using a treated wastewater feed solution (Zou and He 2016). Although fertilizer draw solution from the FO process is ready for use, it may require dilution before application. Therefore, a source of desalinated water should be available, which compromises the cost of fertilizer solution. Apart from these applications, FO has recent advancements in industrial applications where regeneration of the draw solution is eliminated. One recent study suggested crude glycerol and pre-treated hydrolysed wheat straw (PHWS) as potential draw solutions for water recovery and recirculation in biorefineries using the FO process (Kalafatakis et al., 2017). The FO study applied Aquaporin A/S^{TM} membrane, and the reported water flux was 10.5 $L/m^2/h$ and 5.37

$L/m^2/h$ for crude glycerol and PHWS, respectively. Draw solution regeneration was not required as the concentrated glycerol was the draw solution while biological wastewater effluent was the feed solution. The problem, however, with these draw solutions was the presence of microbial cells, which led to biofouling of the FO membrane. Textile dyes draw solution was recently investigated for the treatment of textile wastewater with Aquaporin A/S FO membrane (Korenak et al., 2019). The regeneration step was eliminated since the draw solution was dyes from the production line. The major drawback of dyes draw solution was the high reverse salt flux across the membrane and hence contaminated the feed solution. Another study used Electroless Nickel plating solution draw solution to recover precious metal ions from PCB (printed circuit board) wastewaters using TFC Porifera membrane (Gwak et al., 2018). The FO process successfully concentrated the palladium-based wastewater-stream and without need for regeneration of the draw solution. Although water flux in the FO was relatively high (about 20 L/m^2h), membrane scaling was inevitable. Therefore, the disadvantages of using an industrial waste stream as a draw solution negate the advantages of eliminating the regeneration stage in the FO process.

There is a long list of organic and inorganic draw solutions that has been proposed by researchers (Table 1) for the treatment of a wide range of wastewaters such as sodium ligno sulphate (SLS) and di-sodium hydrogen phosphate (DHSP) (Achilli et al., 2010; Corzo et al., 2017; Lutchmiah et al., 2014a; Thiruvenkatachari et al., 2016). Special considerations are given to the compatibility and reliability of the draw solution with the FO membrane and type of wastewater to avoid major technical and operating problems. For example, researchers used potassium formate and potassium sulfate for boron removal applications, but the high pH of draw solution was not compatible with the operating condition recommended for CTA FO membrane (Corzo et al., 2017).

Researchers also used zwitterions draw solution for wastewater reclamation to reduce reverse salt flux, but it showed drawbacks such as susceptibility to biodegradation and development of biofouling (Lutchmiah et al., 2014a). On the other hand, draw solutions containing magnesium and calcium ions are easily rejected by NF membrane and more energy-efficient to regenerate, but they promote inorganic scaling (Achilli et al., 2010). Furthermore, researchers proposed magnetic nanoparticles as a draw solution because of their high rejection by FO membrane, easy regeneration using a magnetic field, and moderate osmotic pressure, but nanoparticles face problems in scale-up production, agglomeration, intricate synthesis processes or synthesis can be too expensive (Ling et al., 2010). Therefore, magnetic nanoparticles draw solution is not practical for large capacity and commercial applications.

FO process could have a niche market for the treatment of difficult wastewaters, such as mining and shale gas wastewater, where conventional treatment processes are less effective (Ge et al., 2017; Han et al., 2015; Lee and Kim 2017; Thiruvenkatachari et al., 2016). Experimental work on the FO treatment of shale gas wastewater revealed that reverse salt diffusion is a severe issue, due to the formation of irreversible scales on the active layer of the FO membrane. For example, the reverse flux of HCO_3 in the NH_3-CO_2 draw solution chemically reacted with Ca^{2+} ions in the feed to form irreversible $CaCO_3$ scales (Lee and Kim 2017).

Table 1. Draw solutions and membranes used in Wastewater treatment studies

Wastewater	Draw solute	Membrane	Water flux LMH	findings	Reference
WW with heavy metal ions	Hydro acid complex $Na_4[Co(C6H_4O_7)_2]\cdot 2H_2O$ 1M	Lab scale TFC membrane	11	Synthetic, good flux in PRO mode only.	(Cui et al., 2014)
Dye WW	Polyelectrolytes	CA hollow fiber Lab Scale	~ 15 (at 50°C)	High viscosity, Synthetic.	(Ge et al., 2012)

Digested sludge centrate	Real Seawater	CTA HTI	6.4	Phosphorous recovery from sludge. No regeneration required.	(Ansari et al., 2016)
PVC Latex	Synthetic Seawater	CTA-HTI	4.5		(Takahashi et al., 2016)
Treated WW	Fertilizers	CTA HTI	4.2	No regeneration	(Zou and He 2016)
Biorefineries WW	PHWS	Flat sheet Biomimetic membrane by Aquaporin A/S	5.37	Microbial cells in DS can lead to biofouling. No regeneration required.	(Kalafatakis et al., 2017)
Biorefineries WW	Undiluted Glycerol	Flat sheet Biomimetic membrane by Aquaporin A/S	10.5	DS can be toxic. No regeneration.	(Kalafatakis et al., 2017)
Textile WW	Green Dye mixture, Blue Dye mixture. NaCl (1M) $MgCl_2$, (1M),	Biomimetic Aquaporin A/S	~ 11.6	High RSF for dye mixtures. No regeneration is required in case of dye mixture DS.	(Korenak et al., 2019)
PCB WW	E'less Ni Plating solution	TFC Porifera	20	DS leads to inorganic scaling. No regeneration required.	(Gwak et al., 2018)
Medical Radioactive WW	MgCL2 (0.48M). NaCl (0.6M)	TFC PA membrane Porifera	20.4±1.2 to 20.8±2.1	NaCl has a higher rejection for Iodine.	(Lee et al., 2018)
WW plant effluent with antibiotics	1M NaCl without spacer 2M NaCl with spacer	CTA HTI	~13	Same flux for FO and FOwEO (electrochemical oxidation)	(Liu et al., 2015)
Construction WW	NaCl (0.6M)	CTA HTI	7.44	Feed flow rate of 2.9L/min, No spacer and pre-treatment	(Hawari et al., 2018)
Municipal WW	$K_4P_2O_7$, Sodium Polyacrylate, $MgSO_4$	TFC Flat sheet HTI FO 4040 Hollow fiber CTA	9 ~3	TFC flat sheet, $MgSO_4$, Sodium Polyacrylate, and $K_4P_2O_7$ were selected for the demo plant.	(Corzo et al., 2017)
Coal mines WW	SLS, SHMP, DHSP	CTA HTI	5.83-6.9	CTA membrane had better rejection than the RO membrane.	(Thiruvenkatachari et al., 2016)
Shale gas WW	NH_3-CO_2, NaCl	TFC Porifera	21.4	In-organic scaling in the presence of calcium ions	(Lee and Kim 2017)
Oily WW	Oxalic Acid complexes. NaCl	Lab scale TFC-PES membrane	20-23	In PRO mode oxalic acid had good flux.	(Ge et al., 2017)
Emulsified oily WW	1M NaCl	TFC Cellulose acetate butyrate (CAB) Hollow fiber Lab scale	~ 28.2	The experiment was done in the PRO mode. This membrane had excellent oil rejection.	(Han et al., 2015)
Synthetic WW	3M NaCl	TFC-ES HTI	-	Presence of cations in feed aggravates fouling in FO.	(Motsa et al., 2018)
Mercury Polluted WW	1M $MgCl_2$, 1M NaCl	TFC HTI	-	Mercury permeation into draw side.	(Wu et al., 2016)
Industrial WW	Glauber Salt 1 to 2M	Low-pressure RO membrane (Vontron)	5.3	Scaling of the membrane due to DS nature.	(Dutta and Nath 2018)
Fracking WW	KAc (4.47 M) NaGly (4.93M) KFor (4.57 M)	TFC HTI	19.51 to 24.81	Organic DS promote membrane fouling.	(Islam et al., 2019)

	NaPro(4.60 M) NaCl (4.03 M)				
Lab WW	Zwitterions	CTA HTI	4.3-4.9	Low RSF but biodegradation of the DS.	(Lutchmiah et al., 2014a)
Drilling mud and fracturing WW	NaCl 260g/L	CTA HTI	14	Presence of Humic acid and Fulvic acid in DS.	(Hickenbottom et al., 2013)
Distillery WW	$MgCl_2.6H_2O$	TFC Aquaporin A/S	2.8 to 6.3	Fouling of the membrane	(Singh et al., 2018)
Swine WW	$MgCl_2$ (0.5 M)	CTA HTI	3.12	Nutrient recovery from livestock WW.	(Wu et al., 2018)
Acidic WW	NaCl (2M)	Thin film in-organic Lab scale. (TFI)	69.0	High water flux and good rejection of heavy metals by FO.	(You et al., 2017)
Synthetic wastewater with sludge	NaCl	CTA Flat sheet Lab made	~ 15	Bioinspired surface modification improved the antifouling abilities of CTA membrane.	(Li et al., 2016)
Raw Sewage	1.5 M NaCl	CTA HTI	8	FO-MD	(Xie et al., 2013)
Secondary WW Effluent	1M NaCl	CTA HTI	4.5	FO-ED	(Zhang et al., 2013)
Synthetic WW	2M NaCl	TFC HTI	18.6	Sever fouling in the PRO mode.	(Pan et al., 2015)
Olive Mill WW	3.7 M $MgCl_2$	CTA HTI	6.01	The high viscosity of draw solution enhanced DICP.	(Gebreyohannes et al., 2015)
Oil sands produced WW	0.5 M NaCl	Lab-made TFC	18.1	The lab-scale membrane showed good performance than commercial membrane.	(Khorshidi et al., 2016)
High Nutrient sludge	0.2 M Na_3PO_4	TFC HTI	7.09	Less RSD than NaCl	(Nguyen et al., 2016)
Synthetic Dye WW	2M NaCl	TFC Lab made	12.01	Cationic dyes show more fouling tendency	(Han et al., 2016)

Until now, there is no standard draw solution for wastewater treatment yet, although the NaCl solution has been widely used in laboratory experiments (Table 1). As shown in Figure 3, 49% of the FO studies for wastewater treatment used NaCl draw solution followed by $MgCl_2$ (13%).

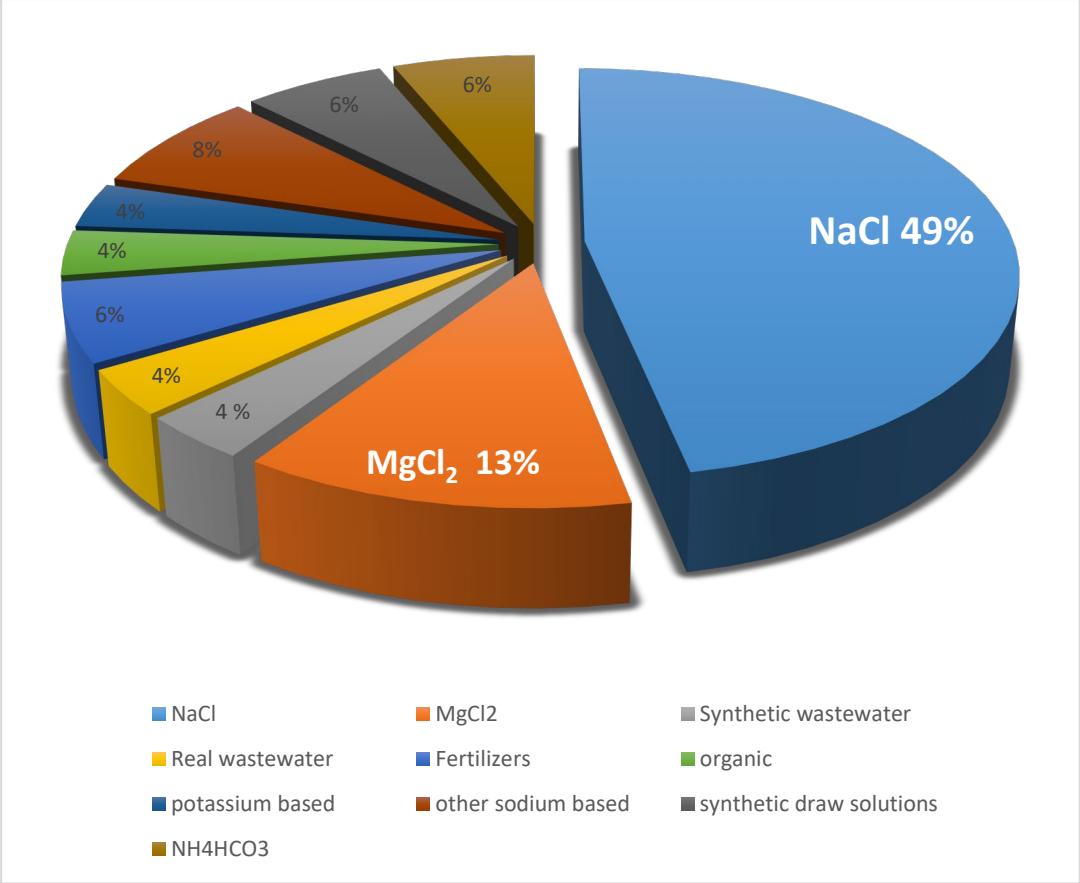


Figure 3. Percentages of different draw solutions used in wastewater treatment studies.

Future research should focus on suitable membranes for wastewater treatment, which can minimize the back diffusion of salt as well as minimize the bridging effect associated with back diffusion of divalent ions. With such a membrane available, NaCl or divalent salts would be perhaps the ideal draw solutions for wastewater treatment applications.

4. FO Membranes

FO membrane plays a critical role in wastewater treatment to minimize fouling. Figure 4 shows the percentage of different type of membranes used in wastewater treatment studies. According to our analysis, 48% of the experimental work used CTA FO membrane for the treatment of wastewater, because of its high tolerance to chlorine,

insensitive to biological degradation, and low fouling potential (Herron 2008; Lv et al., 2017; Thorsen 2004).

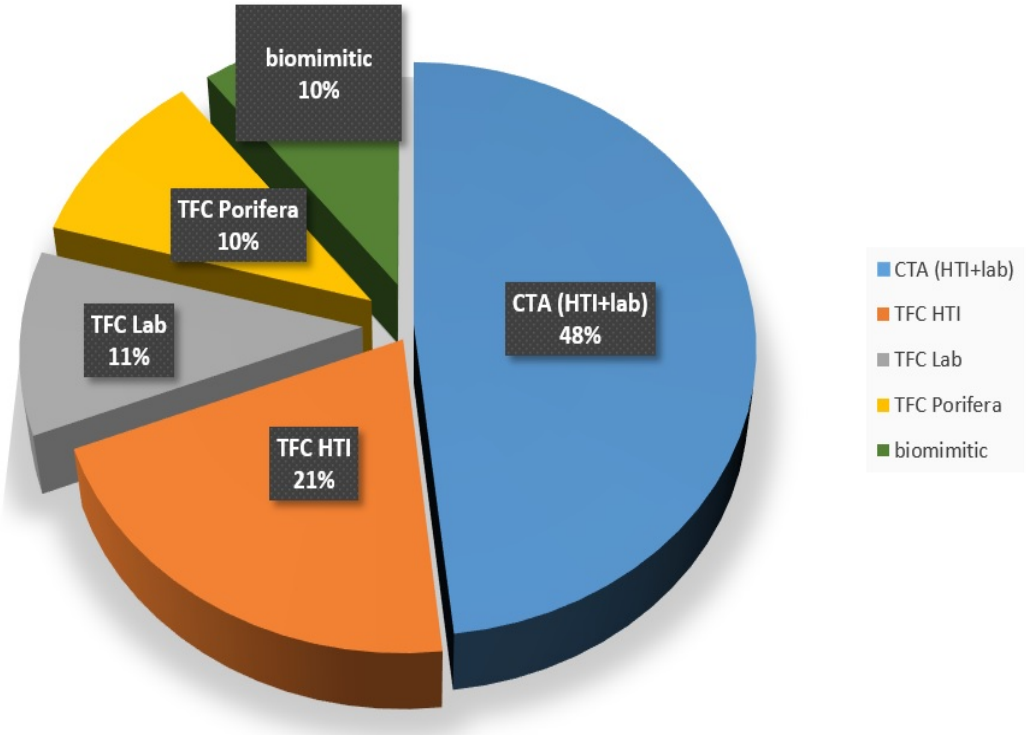


Figure 4. Percentage of different FO membranes used in various FO wastewater applications.

Despite its numerous advantages, CTA membranes have several shortcomings, such as limited pH tolerance, modest water permeability and high sodium chloride permeability (Chou et al., 2010; Ren and McCutcheon 2014; Wang et al., 2018). Commercial CTA membranes were available from Hydration Technology innovation (HTI-USA) and currently from Toyobo Company (Japan) (Nicoll 2013). Toyobo FO membrane is a modification of the RO membrane with a selective layer on the shell side and withstands a maximum hydraulic pressure < 30 bar on the shell side and < 2 bar on the bore side. On the other hand, HTI spiral wound membrane tolerates a maximum pressure of 0.69 bar (10 psi) because of the thinner structural parameter, ~400 µm (Bui et al., 2015).

Nowadays, thin-film composite (TFC) FO membranes have become the most competitive membranes for FO because of the higher water permeability than the CTA membranes (Alsvik and Hägg 2013; Ren and McCutcheon 2014). TFC membranes are commercially available from several companies including Porifera, HTI, and Oasys water Inc. with high rejection rate to nitrates, silica and organic compounds (Singh 2006). Unfortunately, TFC membranes are prone to fouling in wastewater treatment due to the high fouling environment. TFC membranes have limited tolerance to chlorine attack, the reason why many studies using complex wastewaters incorporated CTA membrane is that it can tolerate up to 1 ppm (part per million) of chlorine residues (Fam et al., 2013; Lu et al., 2017). Additionally, CTA membranes are more resistant to silica and gypsum scaling, whereas the presence of high density of carboxylic acid functional group on the surface of TFC membranes leads to its high fouling propensity (Xie et al., 2016).

In addition to fouling of membrane, concentration polarization effects have a detrimental effect on the water flux, especially at the support layer because of the limited hydrodynamic mixing, which leads to the severity in internal concentration polarization. Structure parameter (*S*) of the support layer is a function of the support layer thickness:

$$S = \frac{t_s \tau}{\varepsilon} \qquad [8]$$

Where t_s is the thickness of the support layer, τ is the tortuosity, and ε is the porosity of the support layer. Nowadays FO membranes are fabricated with structure parameter ≤ 500 μm (Bui et al., 2015). FO membranes with thinner support layer exhibit higher water flux but reducing the thickness of the support layer compromises the mechanical integrity of the FO membrane. It is also valid to argue that thinner membrane may have a shorter lifetime although there is no data available about a pilot FO system

operating for 3 years or more (assuming the lifetime of the FO membrane equals to that of the RO membrane).

Wastewater treatment, in fact, is a challenging environment that requires a membrane of high fouling resistance. Membrane module configuration plays an essential role in membrane cleaning and fouling reduction. Flat sheet and spiral wound modules require less pre-treatment than hollow fiber modules. They are also easier to clean up than hollow fiber modules, which have a high packing density (Fritzmann et al., 2007). The main drawbacks of flat sheet FO modules are the low packing density, 70-m^2 per module for Porifera compared to 650 m^2 for Toyobo hollow fiber FO membrane and 16.5 m^2 for HTI spiral wound FO module. Commercial FO modules are available now in a flat sheet (Porifera), spiral wound (HTI and Toray) and hollow fiber (Aquaporin A/S and Toyobo) configuration (Nicoll 2013).

The other challenge that faces the FO process is the cost of the membrane, which is almost ten times more expensive than RO membrane based on the HTI 16.5 m^2 element costs (Altaee et al., 2014). Unless the cost of the FO membrane is reduced, the technology cannot be considered economically competitive to the existing state-of-the-art RO technology. If the demand for the FO membrane increases, the cost will come down. It is important to mention here that the cost of Toyobo 700 m^2 FO membranes is almost comparable to that of the RO membranes (Altaee et al., 2017). However, Toyobo membranes are less common in laboratory size experiments, and they are only available in full-scale modules.

Future research should also focus on the development of chlorine resistance membranes for the treatment of wastewaters and impaired-quality feed solutions (Lu et al., 2017). Successfully, researchers were able to fabricate membrane that tolerates up

to 1000 mg/L NaOCl using layered double hydroxide nanoparticles bound on the TFC by a polydopamine-induced immobilization process. The double-layered hydroxide coating serves as a barrier against the chlorine attack and provides enhanced membrane resistance to organic fouling. Such a membrane would be a good fit for wastewater treatment, but the commercial product may take a longer time to be available. Biomimetic membranes made by Aquaporin (Denmark), on the other hand, demonstrated good performance in terms of water flux (Table 1). However, these membranes need further development as they have a shorter life span than polyamide membranes. Research should also focus on the field performance of a full-scale FO membrane rather than the FO membrane coupons in a laboratory size experiments.

5. Membrane fouling and mitigation techniques

Although membrane technologies have advantages over other mature technologies in wastewater treatment, fouling is still a major operating problem in membrane processes (Chun et al., 2017). Membrane fouling strongly affects water flux and may result in irreversible consequences that lead to membrane damage. Organic, inorganic, biofouling, and colloidal membrane fouling has been reported in the FO process treating wastewater feed solutions (Kwan et al., 2015; Lee et al., 2010; Lee and Elimelech 2006; Li et al., 2016; Mi and Elimelech 2010b; She et al., 2016; Yoon et al., 2013). Researchers have developed several strategies for fouling mitigation in the FO such as increasing feeds cross-flow velocity, chemical treatment, air scouring, altering membrane orientation, DI water bubbling, ultrasonic cleaning, and osmotic backflush. The proposed methods for mitigating membrane fouling were not always successful in restoring water flux in the FO process, especially in a real wastewater environment.

Several studies tested simulated model wastewater foulants instead of real wastewater to investigate membrane fouling in the FO process. The most common fouling problem in many membrane processes is caused by colloidal particles (Singh and Song 2007). Although colloidal fouling would be reversible in the FO process by increasing the cross-flow velocity, it becomes a more challenging problem due to the reverse salt flux from the draw solution. Experimental work revealed that particles aggregation under conditions of high salt concentration due to the reverse salt diffusion would result in a non-recoverable water flux especially at high feed solution pH. A research work by Boo et al., (2012) investigated colloidal fouling in FO using suspension of silica nanoparticles as model colloidal foulants. Silica stock solution was the feed solution, and NaCl and $LaCl_3$ were the draw solutions to investigate the influence of reverse salt flux on fouling behaviour. Colloidal fouling resulted in a recoverable water flux by physical cleaning with high cross-flow velocity, but non-recoverable water flux occurred at high salt concentration and high feed solution pH. Increasing the flow rate of feed solution to reduce colloidal fouling has negative consequences such as increasing pumping energy and pre-treatment cost of the feed solution as in the following expression (Altaee 2012).

$$Es = \frac{Q_f P_f + Q_d P_d}{n Q_p} \qquad [9]$$

Where Es is the specific power consumption, Q_f and Q_d are the flow rate of feed and draw solution, respectively, P_f and P_d are the feed and draw pressure, respectively, n is the pump efficiency, and Q_p is the permeate flow rate. According to equation [9], the high feed flow rate will result not only in increasing the pumping energy into the FO module but also the energy requirements for the pre-treatment of feed solution. This will increase the cost of the FO process for wastewater treatment. Alternatively, colloidal

fouling would be avoidable by a proper pre-treatment of the feed solution to reduce the silt density index (SDI) to less than five by the cost-effective sand filtration process (Fritzmann et al., 2007). Research should also focus on identifying a minimum cost-effective pre-treatment required for the wastewater in the FO process to avoid membrane fouling.

In forward osmosis, similar to other osmotically driven membrane processes, accumulation and growth of microorganisms on the surface of the membrane lead to biofouling (Bucs et al., 2016; Shannon et al., 2008; Vrouwenvelder et al., 2008). While other forms of fouling can be controlled with a variety of pre-treatments, biofouling is the most ubiquitous type and difficult to control due to the strong adhesion of bacteria onto the membrane surface and formation of the extracellular polymer matrix (EPS) (O'Toole et al., 2000). Biofouling can also lead to pore clogging and assist with other types of fouling such as inorganic fouling, and these channelling matters can lead to precipitation of soluble salts and eventually scaling (Abid et al., 2017; Hausman et al., 2009).

Amongst the different types of fouling in the FO process, organic fouling is perhaps the most poorly understood fouling. A wealth of literature has been published about organic fouling in the FO membrane, but most of the studies were performed using model organic foulants (Amy 2008; Lee et al., 2010; Parida and Ng 2013). Simulated organic foulants, unfortunately, are not representative of the actual organic matter in real wastewaters and hence the results from such studies would not resemble the situation of membrane fouling in real wastewater environment or pilot plant. Organic fouling is also affected by the operating mode of the FO membrane, which is found to be crucial in the wastewater treatment by the FO process. Experimental works demonstrated that FO

mode provides more stable filtration process and better cleaning efficiency than the PRO mode but water flux is lower in the FO mode than in the PRO mode due to the intensive internal concentration polarization (Tang et al., 2010; Zhao et al., 2011). Although operating the FO process in the FO mode is recommended to achieve a more stable filtration process, but that would be at the expense of lower water flux. In other words, operating the FO process in the FO mode reduces the membrane fouling, but it compromises the water flux. Practically, there is a wide range of organic substances in the real wastewater that is responsible for membrane fouling; thus laboratory results using model organic foulants are not entirely reliable (Parida and Ng 2013). Several studies have suggested that humic acid fraction of natural organic matter (NOM) is a major foulant, which controls the rate and extent of fouling, while other studies have reported that polysaccharides (hydrophilic fraction) were the main cause of severe fouling in membrane processes (Combe et al., 1999; Jones and O'Melia 2000; Shon et al., 2006; Yuan and Zydney 1999). The fouling tendency of different fractions in natural organic matter decreases as *Hydrophilic neutrals > hydrophobic acids > transliphic acids > hydrophilic charged* (Fan et al., 2001).

In reality, membrane fouling occurs due to the interaction between various fouling matters in the wastewater solution. Therefore, different types of membrane fouling take place simultaneously during the filtration process. For example, combined organic-colloidal fouling shows less reversible behaviour, particularly in the presence of Ca^{2+} ions (Kim et al., 2014). Fouling due to colloidal particles can be controlled to a certain extent by providing an efficient pre-treatment to feed solution, which guarantees its removal from the feed solution. Ultrafiltration and microfiltration membranes demonstrated high efficiency for the removal of colloidal particles from feed solutions. Nowadays, many wastewater treatment plants use membrane bioreactor (MBR)

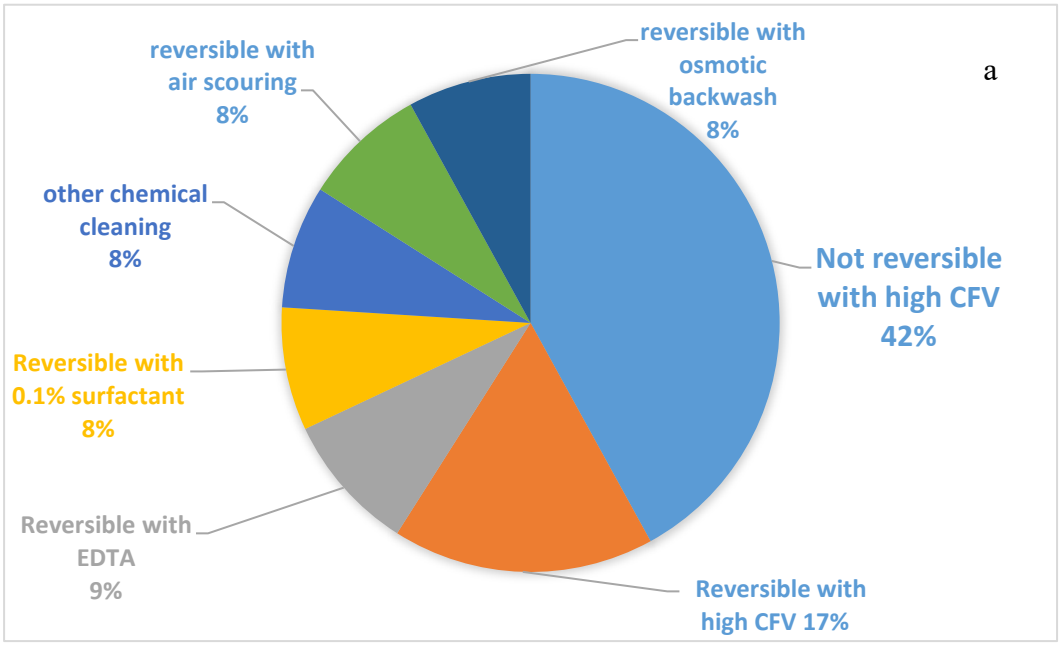
technology, which warrants the removal of colloidal particles from the treated effluent. Unfortunately, using MBR technology will increase the cost of wastewater reclamation by the FO process.

Another type of fouling in the FO process is inorganic scaling, which is mainly caused by the retention of sparingly soluble mineral salts such as calcium carbonate, calcium sulphate, barium sulphate, magnesium salts and silica (Fane 2016; Mi and Elimelech 2013). Amongst the various scaling compounds reported in the literature are silica, calcium carbonate, gypsum and calcium sulphate (Choi et al., 2014; Lee and Kim 2017; Xie and Gray 2017; Xie and Gray 2016; Zhang et al., 2016). To the best of our knowledge, no study has explored fouling due to barium sulphate scaling. A very small number of studies have discussed silica scaling and its cleaning mechanism in the FO process. Mi and Elimelech (2013) investigated silica scaling and cleaning behaviour in the FO process using cellulose acetate (CA) and polyamide (PA) FO membranes. According to the study, 100% and 60% water flux recovery was achievable in the CA and PA membrane, respectively, by increasing the cross-flow velocity from 8.5 cm/s to 21 cm/s. Silica layer on PA membrane was difficult to remove due to the strong adhesion force between the membrane surface and the silica gel. Although increasing the flow velocity of feed solution was a successful technique for fouling mitigation, it was highly dependent on the type and nature of fouling. Inorganic fouling becomes more persistent in the presence of biofouling, and physical cleaning only cannot restore the water flux. Therefore, chemical cleaning with chlorine is recommended to restore the water flux (Yoon et al., 2013). Generally, combined fouling is more resilient in a real wastewater environment, and it is more challenging to mitigate.

Several researchers suggested antiscalant-blended draw solution to mitigate inorganic fouling (Combe et al., 1999; Gwak and Hong 2017). Antiscalants are widely used in the RO desalination of seawater to minimize inorganic fouling (Fritzmann et al., 2007). Although antiscalant-blended draw solution was effective in reducing gypsum scaling, it would increase the cost of FO treatment. Several researchers in the FO process also recommended chemical cleaning for wastewater-fouled membranes (Amy 2008; Lv et al., 2017; Wang et al., 2015). Surfactant, acid and alkaline solution were used for cleaning FO membrane treating oily wastewater and municipal wastewater. Chemical cleaning was experimentally proven as the most effective approach for the restoration of the water flux (Lv et al., 2017). However, it should be carefully implemented to avoid chemicals incompatibility with the membrane material.

In another study, ultrasonic cleaning was suggested for the fouled FO membrane, but it can damage the membrane structure (Zhang et al., 2017). Furthermore, ultrasonic cleaning is expensive and hence not appropriate for a commercial wastewater treatment plant. Air scouring is another useful technique for organic fouling mitigation as well as maintaining a steady flux in both lab and pilot-scale experiments (Qin et al., 2010; Valladares Linares et al., 2013). Several researchers used spacers (feed spacers or antiscalant spacers) to mitigate colloidal and biological fouling in the FO treatment of wastewater (Amy 2008; Hawari et al., 2016; Valladares Linares et al., 2014). Spacers come in different designs and thickness, which play an essential role in minimizing the membrane fouling. Thicker spacers are more efficient for mitigating membrane fouling than thinner spacers, although thicker spacers promote organic and colloidal fouling at low cross-flow velocities (Amy 2008; Valladares Linares et al., 2014). The role of spacers in fouling mitigation until now seems controversial as spacers can hinder the cleaning efficiency in the FO process (Tow et al., 2016).

Membrane material also plays a crucial role in controlling fouling and cleaning behaviour in the FO because of the foulant-membrane interaction (Lay et al., 2012; Li et al., 2018). Although PA membranes have higher fouling potential than CTA membranes, physical cleaning is more effective for the TFC than for the CTA membranes. However, physical cleaning is not effective mitigation method for organic and biological fouling, which are the major fouling problems in the wastewater treatment. Figure 5a and 5b shows the percentage of cleaning protocols and fouling reversibility for fouling with real wastewater and model foulants respectively.



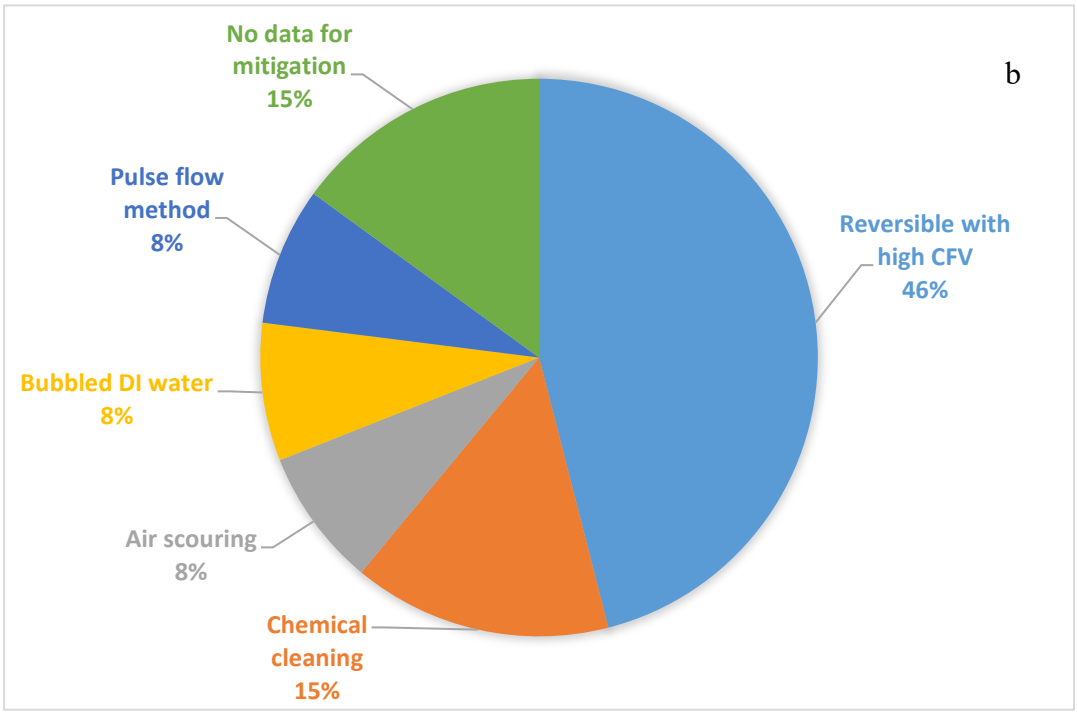


Figure 5. a) Cleaning protocols and fouling reversibility for real wastewater b) cleaning protocols and reversibility for model foulants.

Most of the fouling studies using model foulants demonstrated water flux reversibility by changing the hydrodynamic conditions such as high cross-flow velocities or osmotic backwashing. However, when treating real wastewater feeds, fouling mitigation is not possible by merely changing hydrodynamic conditions and chemical cleaning is often required. Since model fouling experiments are performed to understand the mechanisms of fouling deposition and formation, researchers should be careful when reporting the experimental results so it will not be incorrectly interpreted. Table 2 shows the type of membrane fouling in the FO treatment of wastewater and the mitigation processes.

Table 2. Fouling mitigation in forward osmosis and an overview of different fouling mitigation strategies

Fouling Type	Model Foulants/Feed Water	Draw Solution	Membrane	Initial Operating Conditions	Mitigation	Ref
Organic	Sodium alginate, bovine serum albumin (BSA) and Suwannee River humic acid.	5 M NaCl. 3M Dextrose	CA membrane by HTI	20 °C Same initial flux in all fouling tests	Reversible with high cross flow velocity (CFV)	(Lee et al., 2010)
Organic-inorganic-biofouling-colloidal	*Digested sludge centrate*	Seawater	CTA HTI	CFV of 9cm/s	Reversible with high CFV	(Ansari et al., 2016)
Inorganic scaling	Printed Circuit board wastewater	E'Less Ni Plating solution	TFC Porifera	25 degrees. CFV of 17.0cm/s	Scaling was not removed with physical cleaning, 60-96 % flux recovery with high CFV	(Gwak et al., 2018)
Organic-in-organic-biofouling-colloidal	*Coal mines wastewater*	0.25 M SHMP	CTA HTI	25-35 degrees. CFV data unknown	Reversible with high CFV	(Thiruvenkatachari et al., 2016)
Inorganic Scaling	Shale gas wastewater	Ammonium Carbon dioxide	TFC Porifera	20 degrees CFV 13.6cm/s	Reversible with chemical cleaning with EDTA.	(Lee and Kim 2017)
Inorganic scaling	*Industrial wastewater*	1M Glauber salt	Low-pressure RO membrane	-	90% reversible with high CFV	(Dutta and Nath 2018)
Organic-inorganic-	*Drilling mud and fracturing wastewater*	NaCl 260g/L	CTA HTI	CFV of 0.075 to 0.3 m/s	Reversible with modified	(Hickenbottom et al., 2013)

biofouling-colloidal					osmotic backwashing	
Biofouling	Chlorella sorokiniana with NaCl and/ or $MgCl_2$	0.25 to 2M NaCl stepped up in 30 min interval. $MgCl_2$	CTA	CFV of 22.3 cm/S, 23.0±1 °C AL-DS mode diamond spacer in draw channel	Less reversibility using high CFV	(Zou et al., 2013)
Organic-inorganic-colloidal-biofouling	Oily wastewater	2M NaCl	CTA (lab fabricated)	CFV 8.2 cm/s 25 ºC	Irreversible with high CFV 33 cm/s	(Lv et al., 2017)
Organic-inorganic-colloidal-biofouling	Oily wastewater	2M NaCl	CTA (lab fabricated)	CFV 8.2 cm/s 25 ºC	95% reversible with osmotic backwash	(Lv et al., 2017)
Organic-inorganic-colloidal-biofouling	Oily wastewater	2M NaCl	CTA (lab fabricated)	CFV 8.2 cm/s 25 ºC	90% reversible with 0.1 % HCl	(Lv et al., 2017)
Organic-inorganic-colloidal-biofouling	Oily wastewater	2M NaCl	CTA (lab fabricated)	CFV 8.2 cm/s 25 ºC	90% reversible with 0.1% EDTA	(Lv et al., 2017)
Organic-inorganic-colloidal-biofouling	Oily wastewater	2M NaCl	CTA (lab fabricated)	CFV 8.2 cm/s 25 ºC	85% reversible with 0.1% NaClO	(Lv et al., 2017)
Organic-inorganic-colloidal-biofouling	Oily wastewater	2M NaCl	CTA (lab fabricated)	CFV 8.2 cm/s 25 ºC	100 % reversible with 0.1% surfactant	(Lv et al., 2017)
Biofouling plus organic	Pseudomonas aeruginosa PA01 GFP with 10mM NaCl and 1mM $CaCl_2$ with and without Alginate	4M NaCl	CTA (HTI)	CFV of 4cm/ S and T 25.0±1 °C	Reversible with chemical cleaning with chlorine	(Yoon et al., 2013)
Organic	Sodium alginate+50mM NaCl+0.5mM $CaCl_2$	4M NaCl	CA membrane HTI. TFC (DOW chemicals)	CFV:8.5 cm/s pH:5.8 20±1ºC.	Fastest reversibility with bubbled DI water	(Mi and Elimelech 2010b)
Biofouling	*Pseudomonas aeruginosa in Synthetic Wastewater*	1.3 M NaCl 1.6 M $MgCl_2$	TFC FO (HTI)	CFV velocity of 8.5cm/s, T 25ºC	No data	(Kwan et al., 2015)
Organic	RO brine	2M NaCl	CTA HTI	CFV of 50cm/s	Reversible with high CFV	(Parida and Ng 2013)
Organic	BSA+ Aldrich Humic Acid+Sodium alginate +50mM NaCl with/ or without $CaCl_2$	1.5 or 4M NaCl	CA membrane by HTI	CFV of 8.5cm/s And 20 ± 1 °C	N/A	(Mi and Elimelech 2008)
Organic	Tannic acid + Ca ions in feed	NaCl 2M or 4M	CTA HTI	CFV of 8.5cm/s And 22 ± 1 °C	Not reversible with ultrasonic	(Zhang et al., 2017)
Inorganic	4.2mM Silica+115mM NaCl+19mM $MgCl_2$	1.5 to 4M NaCl	CA membrane HTI	CFV of 8.5cm/s And 20 ± 1 °C	Fully reversible with high CFV	(Mi and Elimelech 2013)
Inorganic	4.2mM Silica+115mM NaCl+19mM $MgCl_2$	1.5 to 4M NaCl	TFC PA membrane Dow Chemicals Company	CFV of 8.5cm/s And 20 ± 1 °C	60% reversible with high CFV	(Mi and Elimelech 2013)
Inorganic	35 mM CaCl2, 20 mM Na2SO4, and 19 mM	4M NaCl	CA membrane HTI	CFV of 8.5cm/s	Above 90% reversible	(Mi and Elimelech 2010a)

	NaCl, with Gypsum			And 20 ± 1 °C	with high CFV	
Inorganic	35 mM CaCl2, 20 mM Na2SO4, and 19 mM NaCl, with Gypsum	4M NaCl	TFC membrane Dow Chemicals	CFV of 8.5cm/s And 20 ± 1 °C	75% reversible with high CFV	(Mi and Elimelech 2010a)
In-organic Scaling	Gypsum, 24mM NaCl,25mM Na_2SO_4, 44mM $CaCl_2$	NaCl+PaspNa Or NaCl+SHMP (1M)	TFC HTI Or CTA HTI	CFV of 8.5cm/s And 25 ± 0.5 °C	Antiscalant DS can control scaling.	(Gwak and Hong 2017)
In-organic	$CaSO_4$	4M NaCl	CA Flat sheet HTI	CFV 8.0 cm/s 20±2 °C	Reversible with high CFV with DI water	(Choi et al., 2014)
Colloidal	Silica 10-20nm	4M NaCl	CA Flat sheet HTI	CFV 8.0 cm/s 20±2 °C	Partially reversible (75%) with high CFV	(Choi et al., 2014)
Colloidal	Poly disperse colloidal suspension solution	3M NaCl	CA HTI	pH 9 , CFV 8.5cm/s	Not reversible with high CFV 25.6	(Boo et al., 2012)
Colloidal	Poly disperse colloidal suspension solution	2.5 M $LaCl_3$	CA HTI	pH 9 , CFV 8.5cm/s 21.0 ± 1.0 °C.	Reversible with high CFV	(Boo et al., 2012)
Biofouling and organic, inorganic	Municipal wastewater	2M NaCl	TFC HTI	CFV 20cm/s 25.0 ± 1.0 °C	Reversible with chemical cleaning	(Wang et al., 2015)
Organic	Municipal Wastewater effluent	Seawater	CTA HTI		Reversible with Air scouring	(Valladares Linares et al., 2013)
Organic-inorganic-biofouling-colloidal	Oily wastewater	2M NaCl	Antifouling zwitterion PES TFC lab scale	CFV of 32.72 cm/s 21.0 ± 1.0 °C	Reversible with high CFV due to membrane properties	(Lee et al., 2018)
Biofouling and organic, inorganic	Municipal secondary wastewater. Synthetic municipal wastewater	3.6% NaCl for simulating natural seawater DS	CTA (HTI)	Single-phase flow with CFV of 0.04m/s. Bubbly Flow with aeration (0.4 L/min). FS and DS temperature of 35.0± 1 °C	Not reversible with bubbly flow method	(Du et al., 2017)
Organic	Soluble Algal product	NaCl $MgCl_2$ $CaCl_2$	CTA HTI and TFC Lab made	CFV of 5.5cm/s And 25 ºC	Physical cleaning Irreversible for CTA. reversible for TFC	(Li et al., 2018)
Organic	Humic acid and alginate	Red sea salt in DI water	One CTA and TFC from HTI. 2 TFC from Porifera.	CFV of 0.1m/s	Reversible with high CFV and osmotic backwashing	(Blandin et al., 2016)
Organic-inorganic	Sodium alginate, BSA and Suwannee River natural organic matter with synthetic wastewater	Seawater RO BRINE	CA HTI	CFV of 10.7cm/s 25.0±0.5 °C	None	(Boo et al., 2013)
Organic-inorganic	Alginate, BSA and Suwannee River natural organic matter with synthetic wastewater	2M NaCl 5M NaCl	CA HTI	CFV of 10.7cm/s 25.0±0.5 °C	Reversible with high CFV or or Pulse flow.	(Boo et al., 2013)

6. Fouling mitigation via chemical modification of FO membranes

The development of high performance and stable antifouling membranes that can operate in harsh operating conditions is vital for the advancement of the FO process in wastewater applications. Although the development of absolute non-fouling membranes seems extremely difficult, if not totally impossible (Rana and Matsuura 2010), manipulating the physiochemical and/or topography structure of a membrane surface can weaken the interactions between foulants and membrane surface (Jiang et al., 2016). The fouling behaviour of the FO membrane is closely related to its properties such as surface hydrophilicity, morphology, and charge. Surface modification is one of the most common, simple and flexible approaches to enhance the separation performance and antifouling properties of TFC membrane (Shen et al., 2017) as well as CTA membranes (Li et al., 2016).

Generally, an increase in surface hydrophilicity of a membrane offers better fouling resistance as most proteins and foulants are hydrophobic in nature (Rana and Matsuura 2010). For wastewater treatment applications, the majority of studies have incorporated CTA HTI membranes because of its high hydrophilicity, good mechanical strength, and high tolerance to chlorine and other oxidants (Zhao et al., 2012). The hydrophilicity of the CTA membrane can be increased by coating the porous support side of the membrane surface with zwitterionic amino acid $_{L}$- Dopa (Nguyen et al., 2013). The modified $_{L}$-Dopa coated membrane showed an excellent fouling resistance against model organic foulants (Alginic acid sodium salt, AAS + 200mg/L $CaCl_2$). CTA FO membranes modified via polydopamine (PD) coating and polyethylene glycol grafting (PD-g-PEG) have also shown excellent antifouling performance in a laboratory scale submerged

osmotic membrane bioreactors (Li et al., 2016). In the wastewater treatment by the FO process, another effort to modify CTA membrane was done by Lv et al., (2017) for the treatment of actual oily wastewater (Table 2). The CTA membrane termed as CTA-HM (home-made) used in this study was fabricated via a non-solvent phase inversion and exhibited excellent flux recovery and 3 times less reverse salt flux compared to a commercial CTA HTI membrane.

The incorporation of nanomaterials such as graphene oxide (GO) into membranes has attracted tremendous interest in recent years. CTA/graphene oxide (GO) membranes with no support layer (free standing) have exhibited excellent resistance against biofouling, reduced effects of internal concentration polarization as well as excellent mechanical stability (Wang et al., 2016a). Despite its excellent results, the cost of GO is a major impediment in scaling up and commercialization of the membrane.

There is a wealth of literature available on chemical modification of the polyamide (PA) layer of thin film composite (TFC) membrane to improve its antifouling properties for wastewater treatment applications (Table 3). Shen et al., (2017) reported a simple and effective second interfacial polymerization (SIP) of the TFC membrane with polyethyleneimine (PEI) of various molecular weights. The antifouling capabilities of the modified membranes were tested in a synthetic wastewater containing sodium alginate (SA) and divalent calcium ions. The modified TFC membranes (especially those modified with high molecular weight PEI) exhibited excellent antifouling behaviour in the synthetic wastewater. This is because of their increased hydrophilicity, less active complexion sites of carboxyl groups and enhanced steric-hindrance with PEI modification (Shen et al., 2017). Han et al., (2015) fabricated a TFC FO membrane by using cellulose acetate butyrate (CAB) as a membrane material for the hollow fiber

substrate for treatment of emulsified oily wastewater under the PRO mode. CAB was chosen due to its better chemical stability and higher free volume than conventional cellulose acetate materials (Ong et al., 2013). The outer surface of the membrane was further coated with polydopamine (PDA), and the inner surface was modified via interfacial polymerization. The modified membrane in this study showed excellent antifouling behaviour under the PRO mode, and water flux dropped to only 90% of the initial value after 12 hours of operation. The flux of the fouled membrane was restored to 97% with water rinsing and without using any chemicals. Table 3 lists some useful studies for improving the antifouling capacity of the FO membrane.

Table 3. Chemical modification of membranes to enhance antifouling properties

Chemical modification	Results	Feed / Draw solutions tested	Reference
Poly[3-(N-2-methacryloylxyethyl-N,N-dimethyl)-ammonatopropanesulfonate] (PMAPS) incorporated TFC membrane	Lower fouling propensity than a neat TFC membrane. Flux recovery of 97% compared to 70.8% of neat TFC membrane.	1000 ppm emulsified oily wastewater/ 2M NaCl	(Lee et al., 2019)
Polyamidoamine (PAMAM) dendrimer grafted TFC FO membrane	Robust antifouling capacity due to its improved surface hydrophilicity. Improved ammonia selectivity (93% rejection)	Domestic wastewater/ 5M NaCl	(Bao et al., 2019)
Thin film nanocomposite (TFN) membrane fabricated by incorporating polyoxometalate based open frameworks (POM-OFs) within the polyamide (PA) active layer of thin film composite (TFC) membrane	Enhanced hydrophilicity, good thermal and hydrostability and excellent water flux (2 times higher than neat TFC membrane). Good fit for wastewater treatment.	Bovine serum albumin (BSA) with synthetic wastewater/ 1M NaCl	(Shakeri et al., 2019)
Thin film nanocomposite (TFN) forward osmosis membrane with a polydopamine modified zeolitic imidazolate framework incorporated selective layer.	Enhanced water permeability without sacrificing the selectivity of the membrane. Excellent rejection of heavy metal ions (>96%) in the FO (active layer facing feed solution) mode.	Heavy metals wastewater/ 1M $MgCl_2$	(Qiu and He 2019)

Thin film nanocomposite (TFN) membrane containing compatible 2D copper based metal-organic framework (MOF) nanofiller in the active layer.	Increase in antifouling and biocidal abilities compared to a pristine TFC membrane. 50% higher flux than the pristine TFC.	Wastewater/ 1M NaCl	(Dai et al., 2019)
Binding of silver-based metal-organic framework (Ag-MOFs) on the surface of the active layer of TFC membrane.	Enhanced antifouling and anti-biofouling properties.	Wastewater containing Pseudomonas aeruginosa / NaCl (0.5 to 2M)	(Seyedpour et al., 2019)
Layered double hydroxides (LDHs) polyamide membrane on polysulfone (PSF) substrate blended with LDH. This membrane was termed as LDH@TFC-LDH	Superior antifouling properties. Long-term stability during chlorination tests under alkaline conditions without sacrificing the water flux.	Antifouling test: 200mg/L sodium alginate+ 50mM NaCl+0.5mM $CaCl_2$/ 2M NaCl Chlorination test: 1000ppm NaOCl/ 1M NaCl	(Lu et al., 2019)
Second interfacial polymerization (SIP) of TFC membrane with polyethyleneimine (PEI) of various molecular weights	Improved antifouling behaviour. High hydrophilicity. Better separation performance.	Synthetic wastewater with sodium alginate / 2M NaCl	(Shen et al., 2017)
Double-skinned TFC membrane with a fully porous sublayer sandwiched between a dense skin (for salt rejection) and a loose skin (for oil particle rejection). TFC layer was formed on top of the polyacrylonitrile (PAN) support followed by the formation of Nexar copolymer layer on the bottom of the support.	Outperforms a single skin membrane due to its lower fouling propensity for the treatment of emulsified oily wastewater treatment. Good resistance against internal fouling and better long-term performance.	Emulsified oily wastewater/ 0.5M NaCl	(Duong et al., 2014)

7. In-situ and real-time fouling monitoring techniques for fouling monitoring in the FO process

In-situ and real-time fouling monitoring is a hot spot for FO research, and it can give us an insight into fouling layer formation and the severity of fouling on the membrane, and thus correspondingly conduct timely cleaning. Fouling in the forward osmosis is usually evaluated through monitoring water flux decline, which generally occurs due to membrane fouling. Amongst the various technologies for monitoring real-time fouling, the simplest one is confocal laser scanning microscopy (CLSM) in combination with multiple fluorescence labelling. This technique gives insight into the structure, distribution, and function of biofilm constituents at a microscale (0.5 to 1.5 micrometre) (Yun et al., 2006). Yuan et al., (2015) used this technique to investigate the biofouling mechanism in OMBR. This study revealed that EPS plays a major role in the formation of biofouling layer and extending the operating time of OMBR leads to a reduction in the growth of microorganisms as well as easy detachment from the fouling layer. Though it is a useful technique, CLSM has several drawbacks such as photo-bleaching can kill some living organisms on the biofilm, the monochromatic laser beam can be harmful, and it can be costly as well.

The first non-invasive systematic investigation of fouling in the FO process was conducted by Wang et al., (2010) using a direct microscopic observation method. Developed by Li et al., (1998) direct observation through the membrane (DOTM) is an effective technique to study particle transport and deposition at the membrane-solution

interface. Using this technique Wang and his co-workers investigated the effects of draw solution concentration, membrane orientation, and influence of feed spacer on membrane fouling using latex particles as model foulants. In-situ observations revealed that FO mode is more resilient to fouling than the PRO mode, and increasing draw solution concentration increases particle deposition on the membrane. Furthermore, feed spacers improved initial and critical water flux in the FO process. The role of feed spacer in improving flux was also reported by another in-situ visualization of fouling study; however, the researchers concluded that feed spacers enhance water flux at the expense of hindering the cleaning efficiency (Tow et al., 2016). This study also introduced a new type of fouling due to vapour formation when the forward osmosis membrane operates in the PRO mode.

A direct observation system to monitor flux decline and fouling layer formation in a study conducted by Liu and Mi (2012) is shown in Figure 6. Direct investigation of the membrane surface revealed that foulant-foulant interaction also affects fouling. The researchers observed a rapid flux decline due to combined fouling compared to individual model foulants. The cleaning efficiency of the fouled membranes was lower in the presence of combined foulants (alginate+gypsum) than for individual foulants. This implies that the results of fouling experiments done using single model foulants (alginate, BSA, gypsum) may not translate precisely into real pilot experiments.

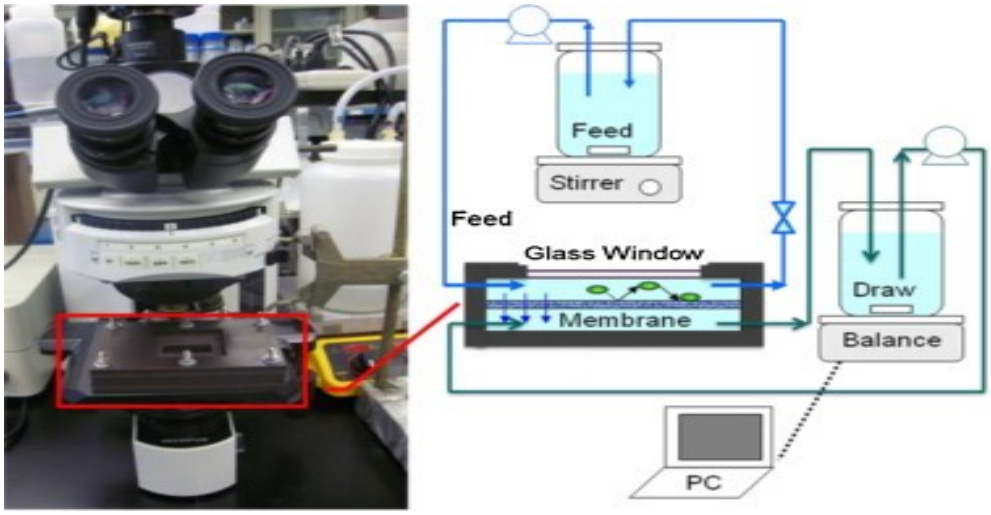


Figure 6. Direct observation system to monitor real-time fouling in FO. Adapted from Liu and Mi (2012) with permission from Elsevier.

In recent years, the application of ultrasound time- domain reflectometry (UTDR) has emerged as a popular technique for monitoring real-time membrane fouling. In UTDR, waves are sent through a medium (Figure 7) and once reflected back from the interfaces (water/membrane or fouling layer/water), the return time and magnitude of the waves are calculated through which the thickness of the fouling layer can be determined (Jiang et al., 2017). Sim et al., (2013b) used UTDR to investigate and monitor biofouling enhanced by silica dosing as an acoustic enhancer in pressure-driven membrane processes. However, since the acoustic properties at the interfaces (water/membrane or water/fouling layer) are slightly different, it is not easy to detect membrane biofouling precisely by the UTDR (Sim et al., 2013b). To the best of our knowledge, no study has reported this technology to investigate the fouling mechanism in the FO, but it is worth investigating in future works.

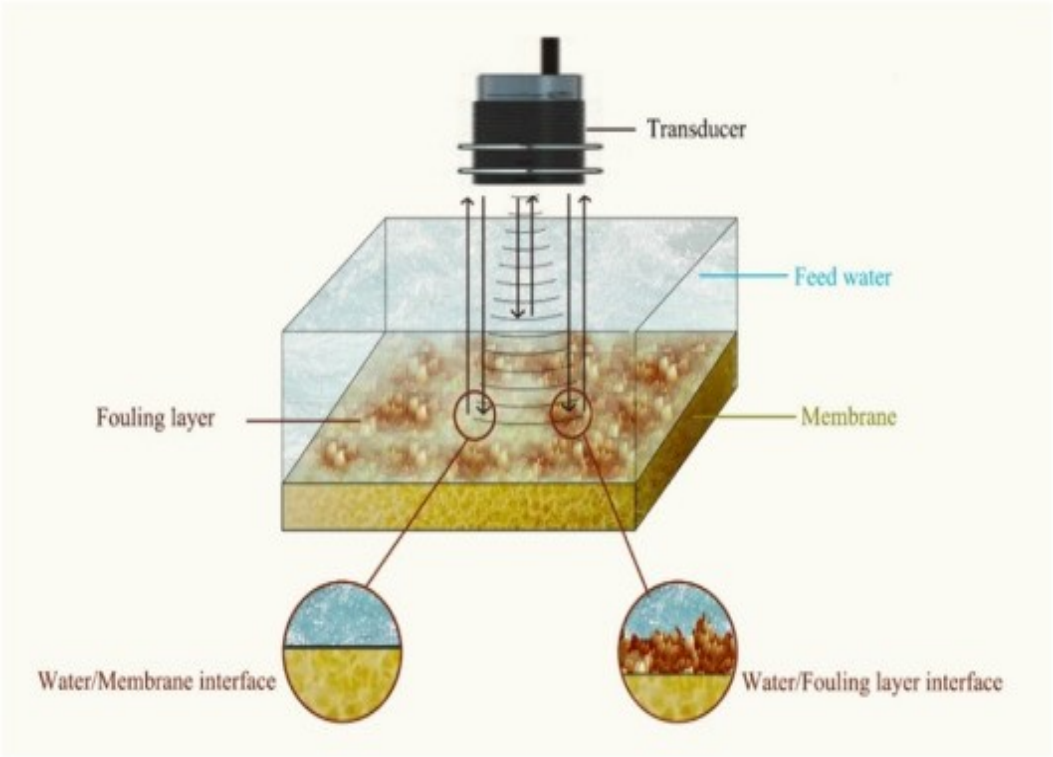


Figure 7. Schematic diagram of UTDR Adapted from Jiang et al. (2017) with permission from Elsevier.

A better alternative to UTDR is the nuclear magnetic resonance (NMR) microscopy, used to monitor biofouling in reverse osmosis for the first time by Graf von der Schulenburg et al., (2008). The study demonstrated that nuclear magnetic resonance microscopy is capable of extracting the biofilm distribution, the velocity field, impact of fouling on hydrodynamics, fouling impact on mass transport, and distribution of the molecular displacement of passive tracers. However, NMR is quite costly and has certain limitations as given in Table 4. A recent study by Valladares Linares et al., (2016) reviewed NMR along with optical coherence tomography (OCT) and oxygen imaging with planar optodes. Reported for the first time by Fujimoto and colleagues in 1991 (Katkar et al., 2018); OCT uses low coherence light to capture micrometre resolution 2D and 3D images from within the optical scattering media. The most significant advantage

of OCT in membrane fouling is that it does not need any signal enhancers or contact with the biofilm (Valladares Linares et al., 2016). Oxygen imaging with planar optodes gives useful insights to microbial activity on a membrane surface and can be helpful in concentration polarisation studies (Valladares Linares et al., 2016).

A novel non-invasive and sensitive monitoring technique for membrane fouling known as electrical impedance spectroscopy (EIS) has gained popularity in recent years (Jiang et al., 2017). The principle of this technology is that when membrane fouling starts to form, it results in change in the membrane electrical properties. The superiority of EIS over other non-invasive techniques is that EIS detect any change in the membrane surface in an almost real-time manner (Sim et al., 2013a). Both conductance and impedance show dramatic changes when a membrane is fouled by a small amount of precipitated divalent salts (Kavanagh et al., 2009). According to Cen et al., (2015) EIS can also indicate fouling type. However, EIS tests are mostly conducted on lab-scale only, and industrial applications need to be field-tested and adjusted accordingly (Jiang et al., 2017). Nevertheless, since EIS is very sensitive to any small changes that occur on the membrane, it would be an efficient technology to investigate real-time fouling behaviour in FO membranes.

Ex-situ scale observation detector (EXSOD) is another monitoring technology for membrane fouling. This technology can detect mineral scaling of the membrane at very early stages of formation before any flux decline occurs (Uchymiak et al., 2007). According to Uchymiak's study, EXSOD can also be a beneficial tool to analyse and evaluate colloidal and biofouling as well as the effectiveness of antiscalant and membrane cleaning strategies.

Table 4. Advantages and disadvantages of different real-time fouling monitoring techniques.

Technique	Advantages	Limitations	References
UTDR	• Can monitor membrane compaction, fouling, and casting processes.	• Non-constant sonic velocity can affect accuracy. • Time should be measured with nanosecond accuracy. • Limited to applications where sonic velocity is constant • Slight Change in operating conditions can lead to errors.	(Stade et al., 2015)
NMR	• A powerful technique that can provide information on topology, dynamics and 3D structure of molecules in a solution and the solid state.	• Expensive • Slow speed and shallow depth of penetration. • Diamagnetic or paramagnetic ions present in a solution can affect tools response.	(Sandeep 2019)
Optical coherence tomography	• Works without the addition of stains or signals to the biofilm. • Contact-free • Can capture micrometre resolution biofilm images • Can determine the impact of hydrodynamics on biofilm formation • Can provide information on how feed water, nutrient concentration, and salt concentration affects biofilm formation.	• High cost • Lack of commercial availability • Materials need to be opaque or translucent	(Katkar et al., 2018; Valladares Linares et al., 2016)
Oxygen imaging with planar optodes	• Can be beneficial for studies on concentration polarisation.	• Sensitive to light and background interference in some applications.	(Valladares Linares et al., 2016)
EXSOD	• Detect scale crystals before flux decline happens or at a very early stage of scale formation • Can be used to evaluate colloidal and biofouling • Can be used to evaluate the effectiveness of antiscalants and cleaning strategies.	• Inadequate spatial resolution	(Uchymiak et al., 2007)
Electrical Impedance Spectroscopy	• Sensitive monitoring method • Can detect any changes that occur on the membrane surface due to high sensitivity • Can evaluate cleaning efficiency • Can be a potential tool to indicate fouling type	• Only limited to lab scale applications	(Cen et al., 2015; Jiang et al., 2017)

8. FO pilot scale applications, energy, and economic aspects

Despite the numerous laboratory size FO experiments, there are only a few tests on a pilot scale for wastewater treatment (Table 5) using the FO process. Scaling up from a laboratory to pilot plant size provides a better perspective and information about the FO process potential and feasibility for wastewater treatment. Several pilot plant tests have been performed for the treatment of various types of wastewater. Oasys water developed the first pilot FO membrane brine concentrator (MBC) for treating high salinity brine streams from oil and gas wastewater (Coday et al., 2014). The draw solution in the FO process was a thermolytic draw solution, which consists of a mixture of ammonium bicarbonate and ammonium hydroxide dissolved in water. The thermal distillation process was applied to cut the cost of regeneration of the thermolytic draw solution (McGinnis et al., 2013). Due to the elevated concentration of feed solution, water flux was relatively low, 2-3 L/m^2 h, which indicates that a large membrane area is required. The other concern about the FO process for treatment of wastewater is the back diffusion of ammonium carbonate across the membrane, which could further contaminate the feed solution (Coday et al., 2015; Mulder and Mulder 1996). With respect to the energy requirements, a similarly configured open cycle single staged evaporative brine concentrator (no energy recovery) would need an energy input of 633 kWh/m^3 of thermal energy, which is 2.3 times higher the energy of 275 kWh/m^3 required in the FO-MBC pilot plant (McGinnis et al., 2013). Therefore, the justification for using the FO process is the elevated concentration of oil and gas wastewater, which is not suitable for treatment by the thermal or hydraulically driven membrane processes.

Osmotic membrane bioreactor (OMBR) has the potential to reduce the energy consumption of wastewater treatment and generate potable water for direct reuse (Wait

2012). Qin et al., (2010) carried out a pilot-scale FO-MBR study for the treatment of domestic sewage. The pilot study used air scouring at the feed side to mitigate the membrane fouling and results showed that the OMBR successfully reduced the energy cost of wastewater treatment and provided a stable water flux. The pilot study did not perform any economic analysis but focused only on the performance of the FO process. Cornelissen et al., (2011) extended the pilot study conducted by Qin et al. (2010) through combining the OMBR with the RO process for regeneration of the draw solution. Economic analysis of the OMBR-RO system revealed that a water flux of 15 L/m^2h using 0.5M NaCl draw solution would be required for the OMBR system to be economically competitive against a conventional MBR, which is operated at an average 16 L/m^2h water flux. The cost of the draw solution regeneration can also be reduced by choosing a proper RO membrane. For example, coupling the FO process with low-pressure reverse osmosis (LPRO) can reduce the energy consumption of the wastewater treatment. According to one study, the FO-LPRO process consumed energy equal to 1.5 kWh/m^3 when seawater is diluted with a secondary wastewater effluent compared to 4 kWh/m^3 for a high-pressure RO system (Yangali-Quintanilla et al., 2011). The FO-LPRO system for indirect desalination consumes only half of the energy used in the high-pressure RO process and can produce good quality water from impaired feeds. Furthermore, the pre-treatment of wastewater should also be included in the total energy requirement for wastewater treatment by the FO process. The capital cost of the OMBR will be higher than that of the MBR due to the additional cost of FO membranes. Currently, the cost of commercial FO membranes varies from tens of USD to few hundreds USD per square meter (Altaee et al., 2014; Altaee et al., 2017). One of the reasons for the high cost of FO membrane is the low commercial demands, and it is expected to decrease if large capacity FO plants

will be built. As such, most pilot plant studies did not provide a cost analysis of the FO process.

Table 5. Overview of pilot studies on forward osmosis for Wastewater Treatment

Feed Water	Membrane material, Module & Configuration	Configuration	Draw Solute	Water Flux L/m^{2}h	Reference
Pre-treated shale gas produced water	TFC-spiral wound (Oasys water)	FO-Thermal	5.5 M to 6.0 M NH_4HCO_3	2.6 ±0.12	(McGinnis et al., 2013)
Produced water from natural gas processing	Mechanically enhanced Flat sheet CTA module	FO-Thermal	NH_4HCO_3 (3M to 6M)	27.5	(Nelson and Ghosh 2011)
Municipal Wastewater	Aquaporin Inside ,TFC	FO-MF	Seawater Or 2M NaCl	1.1 and 9.1	(Hey et al., 2018)
Domestic sewage	CTA Flat sheet pilot-scale	FO-MBR	0.12M NaCl+$MgSO_4$	3	(Qin et al., 2010)
Wastewater effluent	CTA spiral wound	FO-RO (Closed loop)	Synthetic sea salt (30g/L)	7.8 to 7.5	(Hancock et al., 2013)
Mine impaired water	CTA spiral wound	FDFO-NF	0.95,1.8,2.84 M $(NH_4)_2SO_4$ SOA	5.9,7.5,8.8 respectively	(Phuntsho et al., 2016)
Brackish water	CTA spiral wound FO	FDFO	0.6 to1.0M $(NH_4)_2SO_4$ SOA	1 to 4	(Kim et al., 2015)

Raw Produced water from oil and gas	CTA spiral wound	FO-RO	1M NaCl Brine	3.1	(Maltos et al., 2018)
Drilling wastewater	CTA , Flat sheet spiral wound module	FO-RO Close loop	NaCl	-	(HTI 2011)
Pre-treated Municipal wastewater	CTA flat sheet HTI, Aquaporin Inside TFC	FO with physio-chemical pre-treatment	2M NaCl	13.4 and 12.0 respectively	(Hey et al., 2017)
Pre-treated Municipal wastewater	CTA FO spiral wound	FO	0.5M NaCl	No Data	(Wang et al., 2016b)

While these pilot scale system results are promising, little is known about long-term (greater than one year) fouling propensity and its effects on the efficiency of the FO process when treating complex feeds such as oil and gas wastewater (Coday et al., 2014). Future pilot studies should address the long-term performance of the FO process and the energy consumption for wastewater treatment including the regeneration process. Furthermore, standard and cost-effective technologies for fouling mitigation should be developed for wastewater reclamation by the FO process such as air bubbling, reversible flow, and chemical cleaning. Techno-economic studies on the FO treatment of wastewater should be performed, as more information about the feasibility and cost-effectiveness of the FO process is required for comparison purposes with the conventional processes. FO studies should also focus on the advanced treatments of wastewaters, such as NEWater, where high removal of nitrogen and phosphorus can be achieved by the double-barrier in the FO-RO process.

Conclusions

Forward osmosis has great potential in the field of wastewater treatment; however, challenges such as concentration polarisation, ineffective membranes, reverse salt flux, fouling and finding draw solutions that can easily be recycled are the main impediments in its commercialization. FO membranes without a support layer can be the solution for mitigating concentration polarization; however, scaling up such a membrane will take time. In selecting suitable draw solutions for wastewater treatment, NaCl seems the most compatible draw solution with wastewater. Using seawater as draw solution can eliminate the energy extensive regeneration step in forward osmosis. On the other hand, using industrial wastes as draw solution can eliminate the regeneration process but can induce irreversible fouling in the FO system. CTA HTI membranes have been widely used (in wastewater studies) due to its numerous advantages over TFC membranes despite high rejection rates of TFC membranes. CTA membranes have excellent flux recovery rates and are less prone to fouling compared to TFC membranes. CTA membranes are also more resistant to chlorine compared to TFC membranes. However, the lower flux of CTA membranes makes it hard for FO to compete economically with the current technologies. Novel TFC membranes with high flux, excellent antifouling and chlorine resistant properties can be the future of FO wastewater treatment applications.

Due to no hydraulic pressures involved, Fouling in FO is highly reversible (mostly) when model foulants are used to simulate fouling conditions. However, when real wastewater is used, fouling reversibility becomes a challenge. More research is needed to investigate fouling reversibility without chemical cleaning especially in the

presence of divalent calcium or magnesium ions in the feed or draw solution. The role of spacers in fouling mitigation is controversial and more research is needed for efficient spacer designs and location to combat fouling without hindering cleaning deficiency.

While FO cannot energetically outperform RO due to the draw solution regeneration in the FO process, pilot scale FO studies have shown promising results. For draw solution regeneration, using waste-heat or renewable energies can make the FO process competitive against current technologies. FO has a niche market in treating such complicated wastewaters where conventional treatment processes suffer from technical and operating problems.

Future studies on FO can be a long-term investigation and economic analysis of a pilot FO system and comparison with other technologies. In-situ and real-time fouling monitoring is a hot spot for FO research and can give us more insights into the fouling mechanism and correspondingly develop cleaning strategies for the FO process. A range of modern technologies are available to explore for in-situ and real-time fouling monitoring; though Electrical impedance spectroscopy has a great potential due to its high sensitivity and fewer limitations.

Acknowledgments

This work is supported by an Australian Government Research Training program scholarship.

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