CHAPTER 55

Fouling Study on Forward Osmosis Process

Authors

5.1 Background

5.1.1 What is the membrane fouling?

Since the world has been suffered from water scarcity, membrane filtration process has been emerging as the most promising technology in the water treatment system and has developed significantly over the past few decades. To date, a number of filtration systems have been applied to advanced water treatment (i.e., seawater desalination and waste-water reuse) and even traditional water treatment processes. Although the membrane filtration systems are expected as the most feasible alternative for water treatment, several problems still remaine for efficient operation of membrane filtration. Among its problems, fouling which is the deposition of undesirable materials on the membrane surface and/or pores and which reduces the permeate flux and operation efficiency, is one of the major challenges that should be overcome in membrane technology. Membrane fouling can not only cause severe flux decline but also affect the quality of the water produced. Severe fouling often requires intense chemical cleaning or membrane replacement in order to maintain membrane performance which leads to increase of operating costs in a treatment plant.

The fouling mechanism of membrane can be divided into two steps. The first step is foulant adsorption on the clean membrane surface, which is defined as membrane-foulant interaction (i.e., initial rapid fouling). The fouling rate of the first step is decisive in overall efficiency. The next step of the fouling mechanism is its accumulation on the membrane surface, which is defined as foulant–foulant interaction. This step takes up the greater part of the fouling mechanism (i.e., long-term gradual fouling). Between the two steps of membrane fouling, foulant adsorption on the membrane surface, named as the membrane-foulant interaction, is mainly affected by the membrane surface property and usually determines the initial fouling rate. However the foulant–foulant interaction is not clearly distinguishable with respect to the membrane surface property as the clean membrane surface has already been covered by a number of foulants due to the initial membrane-foulant interaction.

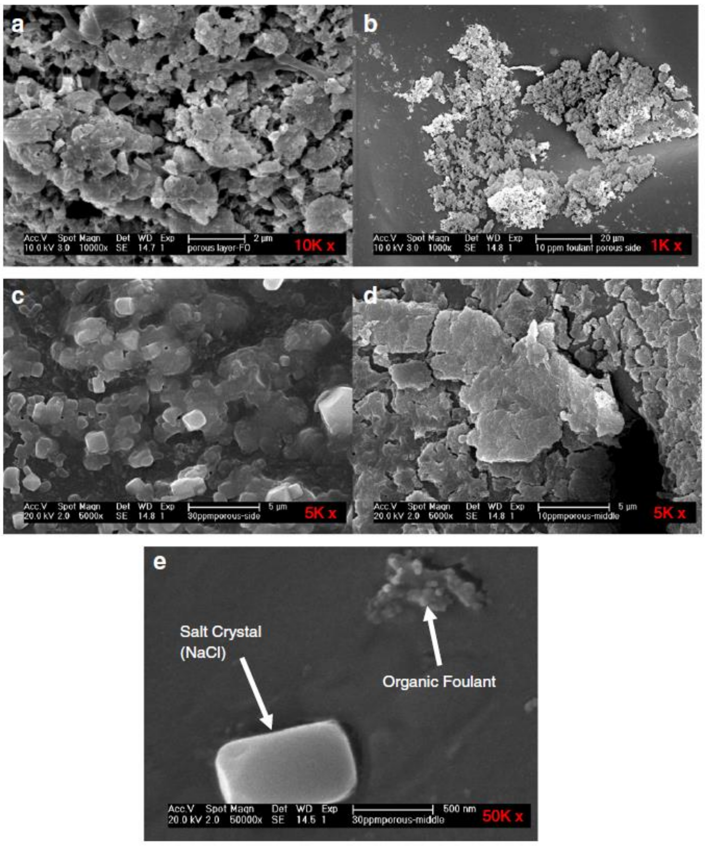
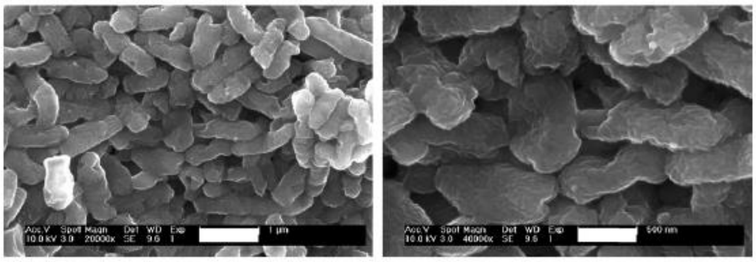
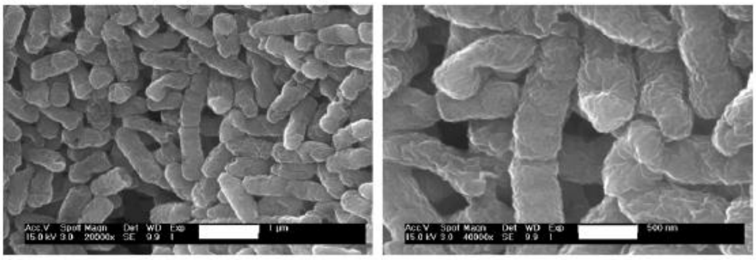


Figure 5.1. SEM images of FO membrane fouling (Parida and Ng, 2013)

(a) PA01 dead cell layer (in a synthetic wastewater medium)



(b) PA01 dead cell layer (in 0.01 mM $LaCl_3$)



(c) PA01 biofilm layer (in a synthetic wastewater medium)

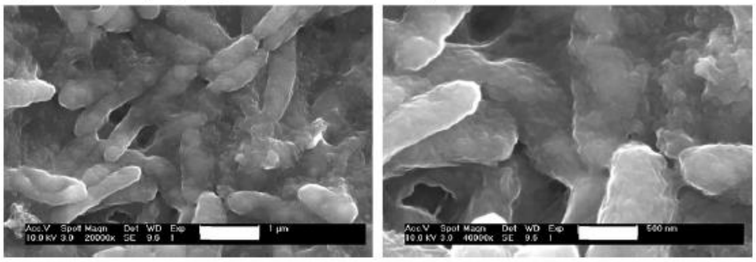


Figure 5.2. SEM images of biofilm layer on RO membrane (Herzberg and Elimelech 2007)

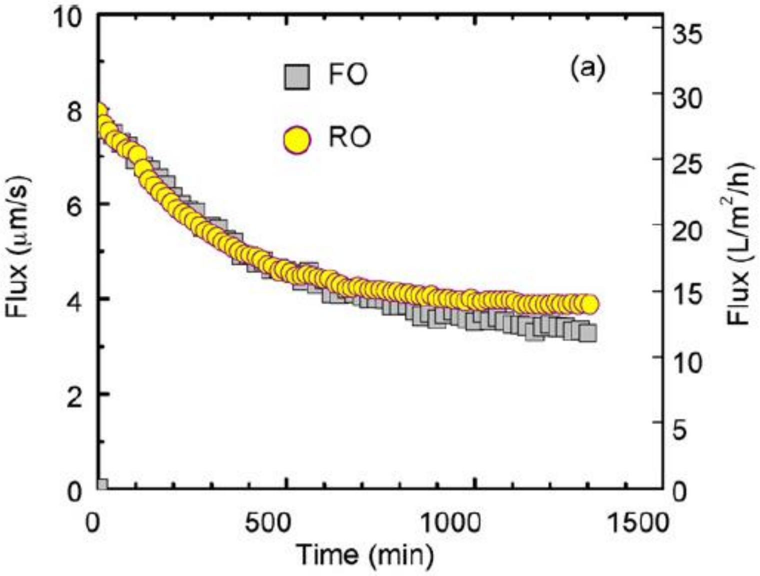


Figure 5.3. Flux decline during alginate fouling on FO and RO membranes (Mi and Elimelech 2010)

There are various types of foulants which cause membrane fouling: colloidal (clays, flocs), biological (bacteria, fungi), organic (oils, polyelectrolytes, humics) and scaling (mineral precipitates) (Baker 2004). The fouling type is determined by each of those foulants as colloidal fouling, biofouling, organic fouling and inorganic fouling.

Moreover, based on the attachment strength of foulants to the membrane surface, fouling can be divided into reversible and irreversible fouling. Reversible fouling can be removed by physical cleaning such as a strong shear force of backwashing or increased cross flow velocity. Formation of a strong matrix of fouling layer with solutes during a continuous operation of membrane filtration results in that reversible fouling being transformed into an irreversible fouling layer. Irreversible fouling, a strong attachment of particles, cannot be removed by physical cleaning thus it requires chemical cleaning (Choi et al. 2005).

5.1.2 Fouling properties of reverse osmosis (RO) process

Reverse osmosis (RO) process has been considered as one of the most promising technologies in hydraulic pressure-driven membrane process. However, RO also has suffered from fouling, especially in desalination. It was found that major factors affecting RO membrane fouling are membrane surface characteristics, feed water solution chemistry and hydrodynamic conditions.

1) Surface characteristics

Surface characteristics of membrane are of paramount importance in RO fouling. Membrane materials are known to affect foulant–membrane interactions, thereby influencing the initial stage of membrane fouling and the efficiency of membrane cleaning. Below are general characteristics of membrane and its effects on fouling (Table 1). From these studies, it can be concluded that RO membrane should have the smoother, more negatively charged, and less hydrophobic properties for reducing fouling potential of RO membranes.

Table 5.1. Membrane characteristics and its foul ing contributions

Paremeter	Remark
• Functional groups	RO fouling is greatly affected by the characteristics of the functional groups of the membrane surface and foulants. Membrane with higher adhesive interaction force with foulants shows greater fouling

	potential due to high attractive force and vice versa (Yang et al. 2010).
• Roughness	Roughness plays an important role on colloidal fouling (Vrijenhoek et al. 2001, Elimelech et al. 1997). The membranes with rougher surface are liable to colloidal fouling as the valleys created by the rough surface produce wells of low interaction energy in which colloidal particles preferentially deposit (Hoek et al. 2003).
• Surface charge	RO membranes with the negatively charged surface exhibit low fouling tendency. Most foulants are negatively charged and thus the electrostatic repulsion between negatively charged foulants and the negatively charged membrane surface prevents foulant adhesion (Shim et al. 2002, Childress and Deshmukh 1998, Brant et al. 2006a).
• Hydrophobicity	Hydrophobic membranes suffer more severely from membrane fouling than hydrophilic membranes due to the strong hydrophobic interaction, which could allow thick fouling layers on the membrane surface (Jucker and Clark 1994, Brant and Childress 2004, Boussu et al. 2007).
• Chemical heterogeneity	Heterogeneously charged RO membrane exhibits high fouling tendency because membrane surface chemical heterogeneities may affect the foulant transport and adsorption on the membrane surface (Sakuma et al. 1989, Lee et al. 2011, Lee et al. 2010a, Brant et al. 2006b).

2) Solution chemistry

With regard to the solution chemistry, pH, and divalent cations have been reported to be key factors affecting natural organic matter (NOM) fouling (Lee and Elimelech 2006; Hong and Elimelech 1997). Organic matters usually have functional groups such as carboxyl (−COOH) and phenolic groups (−OH) which are directly affected by the feed water pH and divalent cation concentration. It has been known that organic fouling is usually accelerated with decreasing pH and increasing divalent cation (i.e., calcium ions) concentration (Seidel and Elimelech 2002; Lee et al. 2005; Lee et al. 2006; Jones and O’Melia 2000). In these conditions (i.e., low pH and high divalent cation concentration), charge property of organic matters diminishes through the neutralization of functional groups as well as organic-calcium complexation. These results in the accelerated accumulation of organic matters on the membrane surface enhance the foulant–membrane interaction as well as foulant–foulant interactions (Lee and Elimelech 2006; Hong and Elimelech 1997; Seidel and Elimelech 2002; Lee et al. 2005). Especially, under typical surface water ionic strength condition (i.e., 10 mM), organic fouling was significantly affected by the solution chemistry (i.e., pH and calcium concentration), showing more rapid flux decline with decreasing pH and increasing calcium concentration. However, these visible impacts of feed water pH and calcium concentration on organic fouling almost disappeared at the condition of high ionic strength such as the TDS of seawater. These results obviously showed that the influence of solution chemistry such as feed water pH and calcium concentration would have much less impacts on organic fouling of RO membranes during seawater desalination (Yu et al. 2010).

3) Hydrodynamic condition

Hydrodynamic conditions such as initial permeate flux and cross flow velocity also affect RO membrane fouling. This indicates that an increased applied pressure (and water flux) and reduced cross flow velocity can cause faster flux decline (Wu et al. 1999). In case of initial flux, higher initial flux tends to increase severe membrane fouling as a result of increased permeate drag force in addition to the enhanced concentration polarization. After the long-term period operation, the fluxes for all membranes became slowly changed, and the fluxes are considered pseudo stable. Unlike initial flux, there is a limiting value for the pseudo stable flux under a given condition, and increasing initial flux beyond the limiting value does not lead to increase of the pseudo stable flux. Therefore, membrane having fluxes larger than limiting flux tends to reach the limiting value. On the other hand, membrane having fluxes below limiting flux seems to be liable to membrane fouling (Wang and Tang 2011; Tang and Leckie 2007). In case of cross flow velocity, cross flow velocity affects the shear induced inertial force and lateral migration, so it may affect membrane fouling tendency. However, the shear induced inertial force and lateral migration depend strongly on the particle size and are of paramount importance in relatively large particles (size ~ or > 100 nm) (Bacchin et al. 1995; Bacchin et al. 2006). Thus shear induced inertial force and lateral migration are less important for the small particle sizes (<< 100 nm) (Wang and Tang 2011; Bacchin et al. 2006).

5.1.3 Fouling properties of forward osmosis process

As mentioned above, there are several factors affecting membrane fouling in RO such as membrane characteristics, feed water solution chemistry, and hydrodynamic condition. During FO, fouling mechanism steps are identical, foulant adsorption on the clean membrane surface which is defined as membrane-foulant interaction and its accumulation on the fouled membrane surface which is defined as foulant–foulant interaction. Similarly, because of its identical fouling behavior steps between RO and FO, those factors which affect membrane fouling during RO process also affect FO membrane fouling.

It was revealed that surface characteristics such as functional groups, roughness, surface charge, hydrophobicity and heterogeneity of the FO membrane surface are important factors that affect membrane fouling rate and extent (Hoek et al. 2003; Mi and Elimelech 2010; Gu et al. 2013). However, since RO membrane is a polyamide thin-film composite membrane and FO membrane is a cellulose based (CTA) membrane, fouling trend could be different between RO and FO due to different membrane surface characteristics. It will be discussed in next section specifically.

Fouling studies on FO were investigated in terms of the effect of solution chemistries on FO fouling (Zhu and Elimelech 1995; Singh and Song 2007, 2005; Faibish et al. 1998). They found that feed solution (FS) chemistry such as pH and divalent cations is important factors for membrane fouling, especially natural organic matter (NOM) fouling. Furthermore, hydrodynamic condition such as initial water flux and cross-flow velocity is a crucial factor during FO fouling formation (Tang et al. 2010; Lay et al. 2010) because it affects membrane fouling extent and rate. However, caused by distinction of fundamental process mechanisms between FO and RO, there are some differences of fouling mechanisms. The distinction of membrane process mechanisms differences leads to different fouling rate and extent between FO and RO. Therefore, it is significant to study the fundamental process mechanisms of both processes.

First, each process has different driving force. The driving force of FO is osmotic pressure difference between FS and DS, while RO is hydraulic pressure driven process. It is very important because one of the main forces that build up fouling layer on membrane surface is driving force for water permeation. Since osmotic pressure is chemical pressure, it is possible that its impact on fouling formation may be different from hydraulic pressure that is physical pressure.

The second difference of process is solute flux direction. During RO operation, salt is dissolved in the FS and passes through the membrane as identical direction of water flux.

During FO process, on the contrary, salt passes through the membrane from DS to FS due to its higher ion concentration of the DS then the FS (Phillip et al. 2010). It is called reverse salt diffusion (or reverse solute flux, RSF).

As a result, these two major differences (driving force and salt flux direction) could be related to the difference of flux decline rate and extent and lead to different fouling mechanisms between RO and FO. For example, it is confirmed that accelerated cake-enhanced osmotic pressure (A-CEOP) in FO is mostly due to reverse salt diffusion from the DS which is accumulated between membrane surface and fouling layer then leads to much thicker layer which results in more flux decline compared to RO. Consequently, it reduces the net driving force for water permeation by increase of fouling layer resistance in FO. Moreover, it is found that the structure of the organic fouling layer is also determined by the applied hydraulic pressure (Lee et al. 2010b). FO fouling is revealed that fouling is reversible in FO while irreversible in RO. Since FO does not require hydraulic pressure, deformable foulants make loose and sparse fouling layer that can easily be removed by physical cleaning.

2. FO fouling

5.2.1 RSF and A-CEOP

Cake-enhanced osmotic pressure (CEOP) phenomenon is that the cake layer (one of the fouling types) prevents the back diffusion of salt feed bulk solution thereby resulting in increased osmotic pressure adjacent the membrane surface, thus water flux decreases. Lee et al. (2010) reported a relationship between RSF and fouling formation, now it is well known as A-CEOP phenomenon that is caused by solute from DS and it contributes to much thicker cake layers compared to RO except BSA fouling. The relationship between RSF and fouling is proved by comparing water flux decline when NaCl and dextrose as draw solute. Dextrose was effectively retained by the membrane because of its much larger hydrated radius than that of NaCl, leading to negligible reverse solute flux of dextrose from the draw to the feed side therefore A-CEOP phenomena were only observed for NaCl solution (Fig. 5.4).

The colloidal fouling is also affected by RSF. In addition to RSF, colloidal particle size contributes to fouling severity (Lee et al. 2010; Boo et al. 2012). In both papers, when small silica particle, approximately 20 nm, was used as foulant, water flux decline was mild compared to large particle, 139 nm and 300 nm respectively. This is because the small colloidal particles have large back diffusion compared to the convective permeate flow, thereby resulting in a thin cake layer which has short pathway of back diffusion, thus CEOP is insignificant.

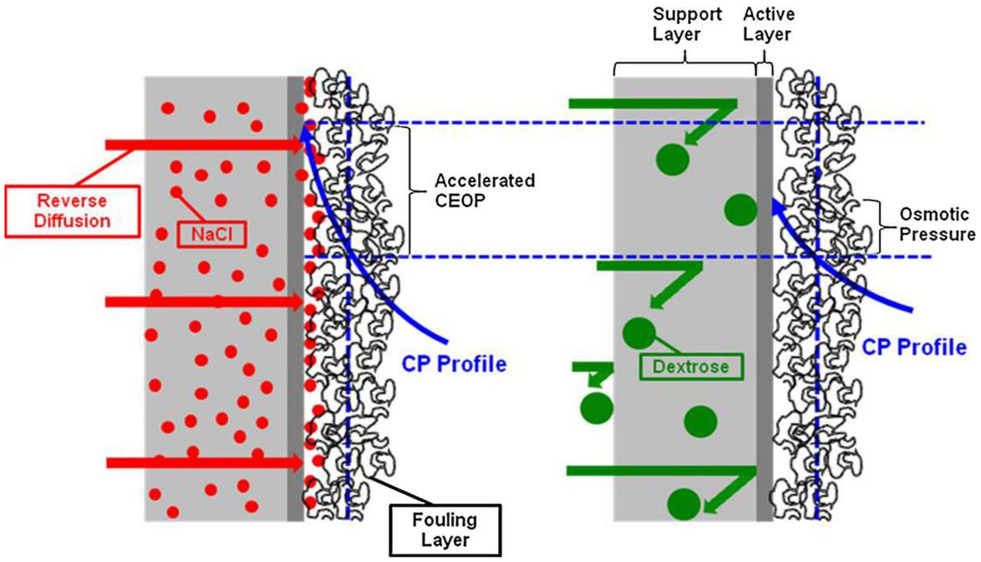


Figure 5.4. A conceptual illustration of the effect of draw solute reverse diffusion on CEOP (Lee et al. 2010)

5.2.2 Membrane orientation

Since FO uses no hydraulic pressure, it is possible that operating FO process on two membrane orientations that the membrane active layer is placed against the FS (FO mode), and the membrane active layer is against the DS (PRO mode). Mi and Elimelech (2008) observed that membrane orientation has different effects on the organic foulants such as alginate, BSA, and AHA. It is interesting to note that fouling propensity depends on foulant type and membrane orientation. For alginate, fouling is not affected by membrane orientation, with a similar flux decline obtained in FO and PRO modes. In contrast, for BSA and AHA, the flux decline is more severe in the PRO mode than in the FO mode (Fig. 5.5). This observation is most notable with AHA. The different membrane orientation effects suggest that the different organic foulants have different fouling mechanisms.

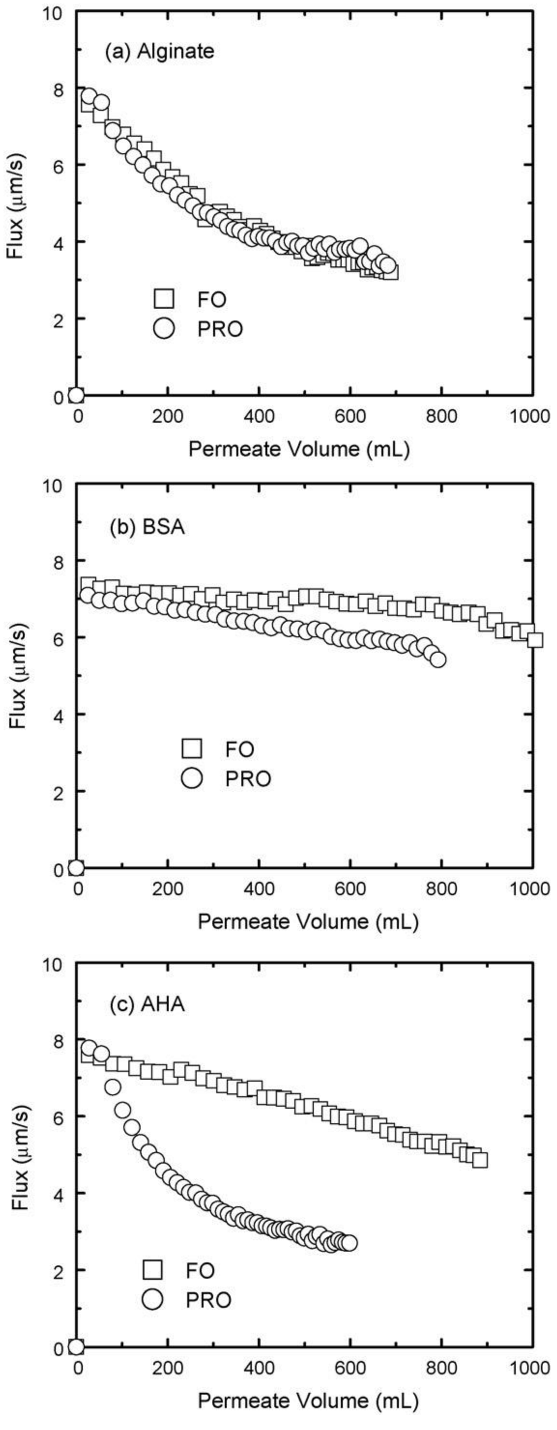


Figure 5.5. Effect of membrane orientation on FO membrane fouling by alginate, BSA, and AHA (Mi and Elimelech 2008)

Since the FO membrane has an asymmetric structure, an active layer on top of a porous support layer, membrane fouling occurs on different surfaces in FO and PRO modes. For instance, in FO mode, foulant deposition (or accumulation) occurs on top of the active layer. On the other hand, foulant deposition takes place within the porous structure of the membrane in PRO mode. The biggest difference between two modes is that cross-flow velocity vanishes within the porous support layer, thus the influence of hydrodynamic shear forces is absent at the initial stage of fouling in PRO mode. As the two membrane orientations provide different hydrodynamic conditions during membrane fouling, the membrane orientation also should be considered as a key factor of membrane fouling in FO. Figure 5.5. shows that water flux variation for alginate is identical on both modes, which means that hydrodynamic interactions do not play crucial role in alginate fouling. Chemical interactions, like calcium binding, play a dominant role because highly built gel layer is relatively unaffected by hydrodynamic condition change. However, in case of BSA and AHA, Figure 5.5. shows that hydrodynamic condition change influences fouling formation significantly by comparing water flux variation on both FO and PRO modes. In addition, the remarkable flux decline with AHA on the PRO mode is attributed to cake layer formation due to lack of shear force as well as hindered back diffusion of AHA aggregates in the porous structure (Mi and Elimelech 2008). These results are in good agreement with previous studies (C.Y. Tang et al. 2010; V. Parida and H.Y. Ng 2013b)

5.2.3 Solution chemistry (pH and Calcium binding)

1) pH

Solution chemistry has a prominent effect on the electrokinetic properties of colloidal particles and on the rate of fouling in membrane separation (Zhu and Elimelech 1995; singh and Song 2007, 2005; Masliyah 1998; Faibish et al. 1998). Boo et al. (2012) investigated the relationship between solution pH on colloidal fouling propensity and both monodisperse and polydisperse colloidal suspensions. Regardless of pH, there were no water flux differences when monodisperse colloidal particles were used as foulants. On the other hand, at high pH (pH 9), water flux for the polydisperse suspension was suddenly dropped while mild water flux decline was observed at pH 4 for identical polydisperse suspension (Fig.5.6 and Fig.5.7). This is because hydrodynamic radius of the large particles, 139 nm, significantly increased for high pH (pH 9) and high ionic strength (1 M of NaCl) solutions. Increased hydrodynamic radius of particles are unstable that tend to aggregate each other, consequently result in fouling formation.

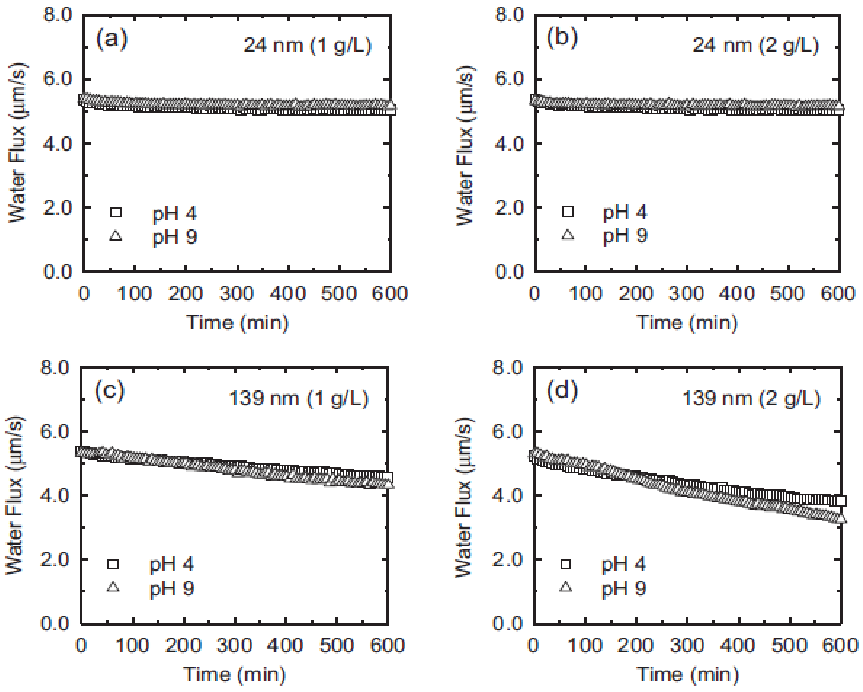


Figure 5.6. Flux-decline curves obtained during the FO colloidal fouling runs with silica particles at pH 4 and 9 (Boo et al. 2012)

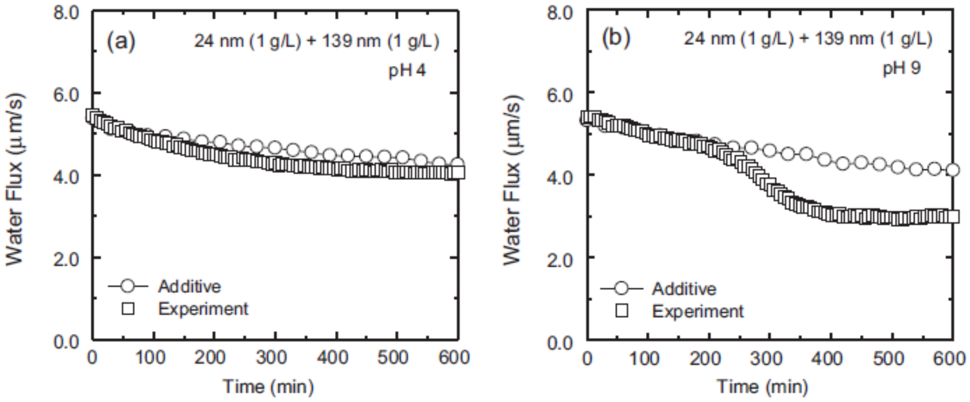


Figure 5.7. Flux decline curves for FO fouling experiments with the polydisperse silica suspension (1 g/L of 24 nm particles plus 1 g/L of the 139 nm particles) (Boo et al. 2012)

These phenomena were proven by comparing NaCl and $LaCl_3$ as a draw solute. Water flux was dropped when NaCl was used as a draw solute while no water flux changes when $LaCl_3$ was used. The first reason is apparently RSF. In addition to RSF, it was observed that pH of FS decreased due to reverse diffusion of protons generated by the hydrolysis of $LaCl_3$. Since low pH makes colloidal particles stable, such pH decreasing effect by $LaCl_3$ solution also helps low fouling propensity (Fig. 5.8).

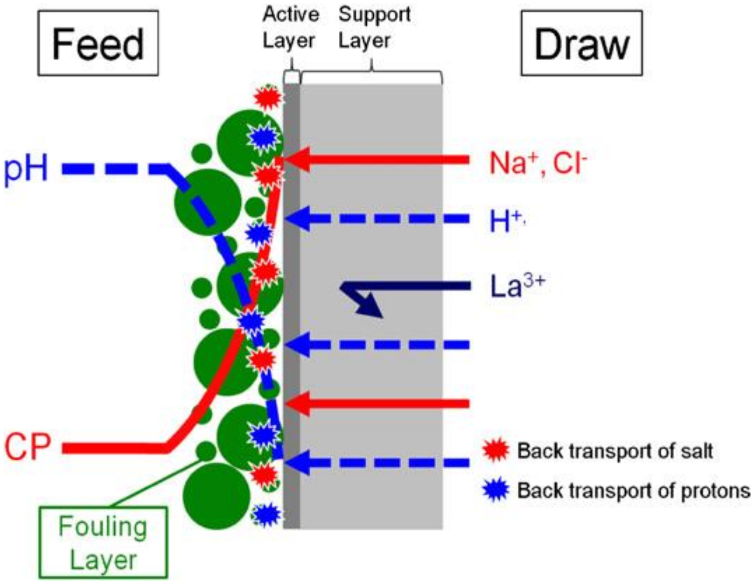


Figure 5.8. A conceptual illustration for the effect of reverse diffusion of DS on the concentration polarization (CP) and pH profiles within the fouling layer (Boo et al. 2012)

2) Calcium binding

It is well known that calcium binds with carboxylic acid groups in NOM and causes accelerated fouling (Li et al. 2007). In FO process, fouling severity is different from membrane orientation. Parida and Ng (2013a) reported Ca^{2+} effect in FO process. When the FO active layer was subject to a FS containing calcium ion and organic foulants, the flux remained nearly constant throughout 20 h of the experimental run. However, in the PRO mode, the water flux sharply dropped within the first 1–2 h operation, followed by a gradual drop. According to the results, the previous tests without Ca^{2+} addition showed that the water flux in PRO mode after 20 h dropped by 36% (from 33.0 to 24.0 LMH) for 30 ppm of TOC in the FS, and by 45% (from 33.0 to 18.0 LMH) in the case with 50 ppm of TOC. With the inclusion of Ca^{2+}, as high as 85% reduction in water flux (from 33.0 to 4.8 LMH) was observed in the PRO mode. It shows that not only Ca^{2+} binding but also membrane orientation is a key parameter in FO process because loose and rough structure of porous layer exacerbates fouling on PRO mode. On contrary, no severe fouling was observed in FO mode. Hence, although calcium–foulant interactions could cause fouling, the smooth structure of the dense layer of the FO membrane impeded fouling. Furthermore, high cross flow velocity (50 cm/s) was enough to wash off foulants from the membrane surface, thus reducing the effects of external concentration polarization (ECP) on the densely fouled membrane surface.

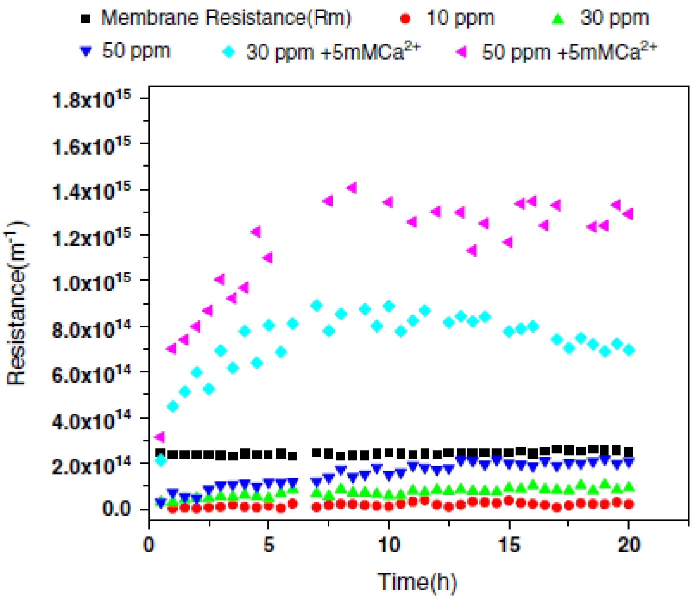


Figure 5.9. Resistance (m^{-1}) versus experimental time in the absence and presence of Ca^{2+} (5 mM) in both FO and PRO modes (Parida and Ng 2013a)

5.2.4 Initial flux

As FO is an osmotically driven membrane process, the DS concentration plays a critical role in FO performance (Tang et al. 2010; Mi and Elimelech 2008; Lay et al. 2010). The effects of initial flux on organic fouling are shown in Figure 5.10. on PRO mode.

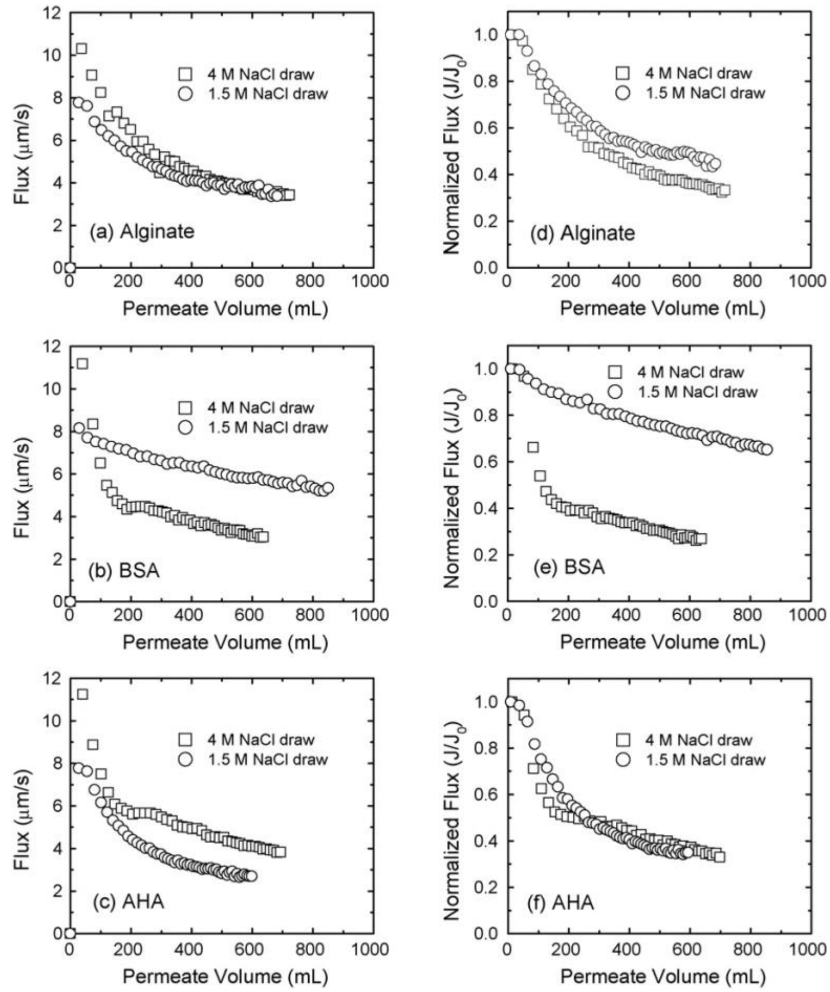
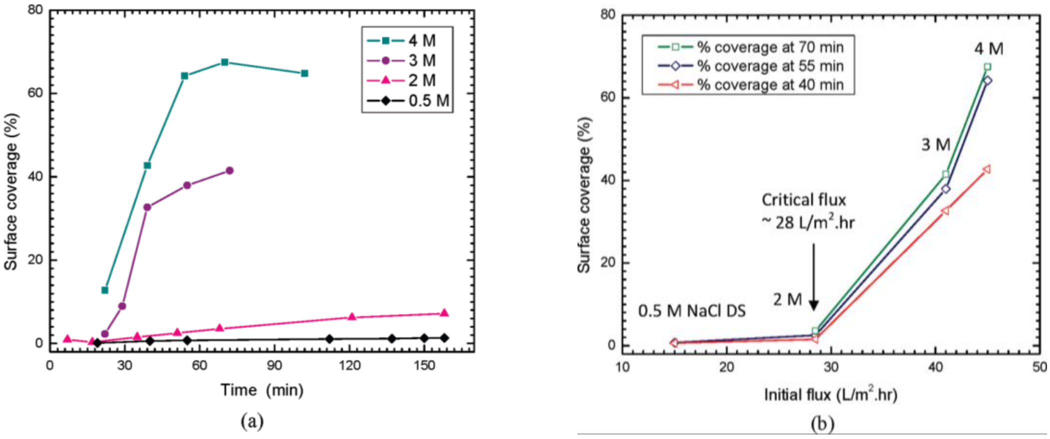


Figure 5.10. Effect of initial flux on membrane fouling in the PRO mode (Mi and Elimelech 2008)

In the case of BSA, the higher initial flux caused higher flux decline. Since the influence of initial flux on membrane fouling is principally attributed to the permeation drag resulting from convective flow toward the membrane, the results indicate that the stronger permeation drag exerted on BSA molecules results in the formation of a cake layer. The large difference in the fouling behavior with BSA for low and high permeation drags reflects a transition in the fouling layer from a loose fouling layer structure to a much more compact cake layer. For alginate and AHA, the effect of initial flux is relatively small because the fouling layers under both conditions are already in the form of a cake layer. Once a cake layer forms, fouling becomes much less sensitive.

Wang et al. (2010) studied the effect of DS concentration for FO particle fouling on PRO mode (Fig.5.11). Apparently, the DS concentration critically affects to FO fouling. While particle deposition was negligible at the low DS concentration at 0.5 M, small patches were observable for a 2 M DS. At higher DS concentrations, the surface coverage was extensive (about 40% for 3 M DS and about 70% for 4 M DS at 70 min). Corresponding to the dramatic particle deposition at higher DS concentrations, the flux decline was also more severe for the 3 and 4 M concentration of DS. The greater fouling propensity at higher DS concentrations may be explained by the higher flux levels as a result of increased FO driving force. For pressure-driven membrane processes, it is well-known that elevated flux levels inevitably lead to severe permeability loss (Tang et al. 2007; Bacchin et al. 2006). In contrast, fouling is minimal when flux is below some threshold values, which is known as the critical flux behavior (Tang et al. 2007; Bacchin et al. 2006). The study demonstrated that the critical flux concept is also applicable to the osmotically driven FO process.



(a) (b)

Figure 5.11. Effect of DS concentration and FO flux level on FO fouling (Wang et al. 2010)

The strong dependence of the critical flux value on DS chemistry is a unique feature for FO fouling. In this regard, a concept of DS type-dependent critical concentration may also be of practical interest. When there was no Mg^{2+} in the original feed water, the critical concentration of $MgCl_2$ was below 0.5 M while that for NaCl was between 2 M and 4 M for algae fouling in the present case. Zou et al. (2011) suggests a strong interplay between membrane flux and DS chemistry on FO fouling in addition to the well documented interplay between flux and solution FS chemistry.

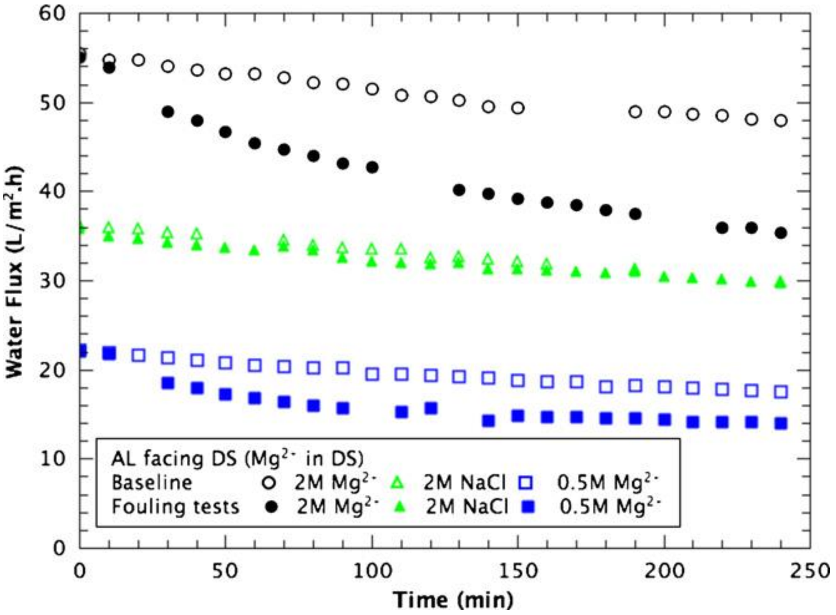


Figure 5.12. Effect of DS type (NaCl vs. $MgCl_2$) on FO fouling (Zou et al. 2011)

5.2.5 Membrane material type (PA vs CA)

Mi and Elimelech (2010) compared the CA and PA (or TFC: thin film composite) FO membranes in the RO mode to elucidate the effects of membrane materials on alginate fouling and cleaning behavior. Compared to CA membrane, PA membrane has a lower average adhesion force which is measured by atomic force microscopy (AFM) but those 18% of its adhesion forces are distributed in the range of from 1.2 to 2.0 mN/m while the maximum adhesion force of the CA membrane is 1.2 mN/m. Consequently, these small but sticky sites on the PA membrane may cause more severe fouling at the initial stage.

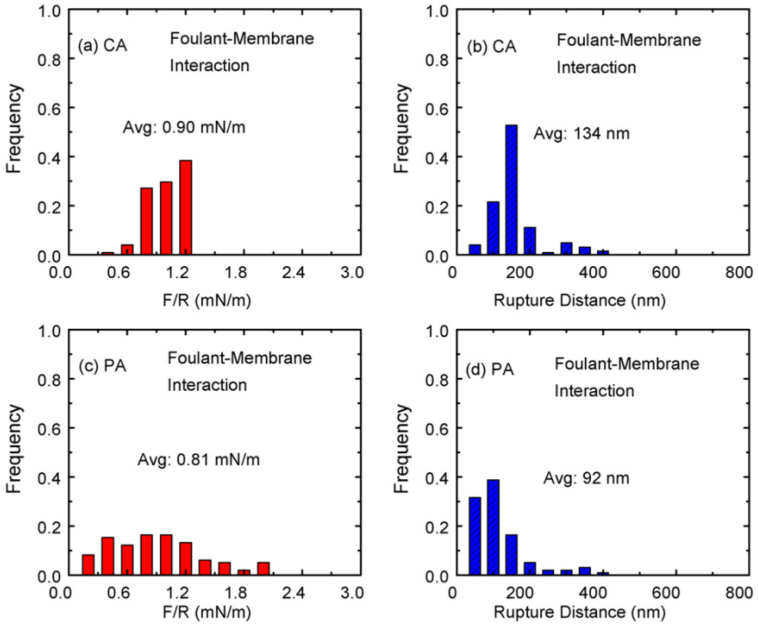


Figure 5.13. Adhesion force measurements of foulant-membrane interactions for the cellulose acetate (CA) and polyamide (PA) membranes (Mi and Elimelech 2010)

Gu et al (2013) also compared CTA FO membrane and TFC FO membrane in terms of flux performance and foulant mass deposition during FO fouling.

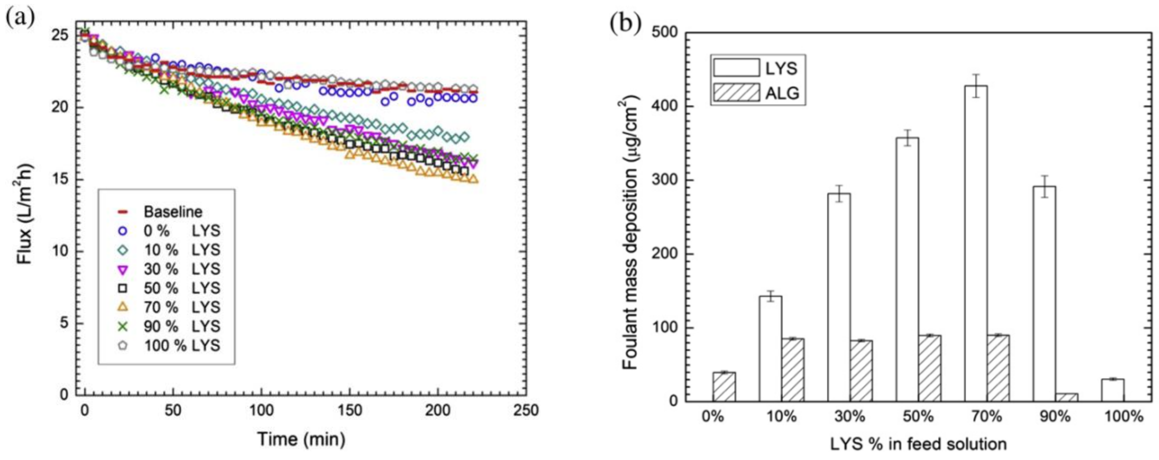


Figure 5.14. Effect of foulant composition in FS on FO fouling of CTA membrane (Gu et al. 2013). For FS containing either only sodium alginate (ALG) (0% Lysozyme (LYS)) or only LYS (100% LYS), it showed clearly that the TFC membrane was more likely to have severe fouling compared to the CTA membrane. However, when two foulants were mixed, LYS and ALG (50% LYS in feed), both the CTA and the TFC membranes experienced severe flux decline and the final flux values towards the end of fouling tests were similar (~15 LMH for CTA and 13 LMH for TFC). Moreover, the deposited foulant mass and composition for the

two membranes were nearly identical. In both cases, the unfavorable interaction between the foulant species in the FS (i.e., the electrostatic attraction between LYS and ALG) caused foulants deposition on the membrane surface, thus severe flux decline. Results from the current study suggest that: (1) Under mild FO fouling conditions, TFC FO membranes can have greater fouling tendency compared to CTA FO membranes due to their greater surface roughness (the surface roughness plays a more dominant role over surface hydrophilicity in membrane fouling, noting that the TFC membrane was more hydrophilic than the CTA membrane); (2) Under severe FO fouling conditions, membrane surface properties play a less important role; (3) Although FO is believed to have superior fouling resistance in the AL-FS orientation, fouling can occur even at moderate flux levels, especially for TFC membranes or for unstable FS.

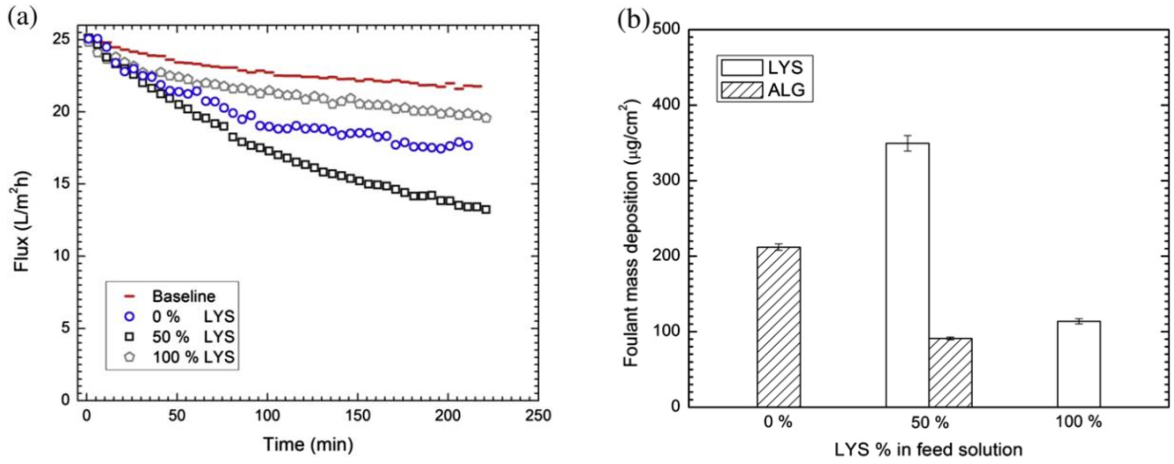


Figure 5.15. Effect of foulant composition in FS on FO fouling of TFC membrane (Gu et al. 2013)

5.3. Fouling mitigation methods

In this part, various strategies that mitigate fouling formation in FO process are discussed

5.3.1 Spacer

Inserting spacer on the feed side is the easiest and effective way to reduce fouling on membrane surface because it generates shear force that promotes turbulence near the membrane surface on the feed side. In Figure 5.16, the feed channel spacer enhances mass transfer in the boundary layer near the membrane surface, thereby resulting in less accumulation of foulants on the membrane surface (Boo et al., 2013). The flux decline is notably diminished during fouling runs with spacers. It clearly demonstrates the less accumulation of organic foulants on the membrane surface. Similarly, the amount of algal biomass deposition was reduced by the inclusion of feed spacer in the PRO mode (Zou et al. 2013).

Meanwhile, it was reported that feed spacer induces accumulation of particles near and under the spacer filament (Wang et al. 2010). Such particle accumulation was likely due to the low shear force region caused by spacer, physical blockage, due to the relatively large particle sized latex used in the study. However, approximately 10% flux decline was observed without feed spacer for 2 h fouling run, whereas no considerable flux loss with regard to the baseline was observed during fouling run with the feed spacer in the FO cell. The paper may hypothesize that the membrane area covered with the spacer has relatively marginal contribution to the overall permeate water so that local deposition of particles underneath the spacer has little effect on the flux stability.

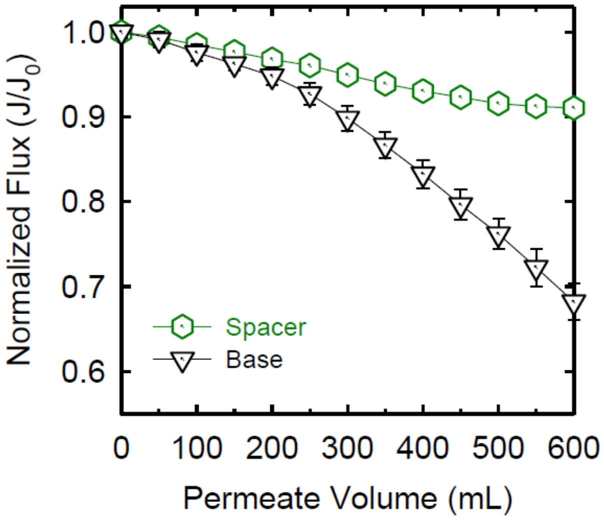


Figure 5.16. Impact of hydrodynamic control strategies on FO fouling mitigation using feed channel spacer (Boo et al. 2013)

5.3.2 Cross flow velocity

The main advantage of using FO is that it has lower irreversible fouling than pressure-driven membrane processes due to the lack of applied hydraulic pressure. Lee et al. (2010) compared the fouling behaviors in regard to effect of cross flow velocity during FO and RO (Figure 5.17). Although flux decline is much severe in FO than RO, but the reduced flux is readily recovered when the cross flow velocity increases during the FO fouling run and that the following flux decline rate lessens after increasing the cross-flow velocity. It should be noted that this simple physical cleaning by increasing cross flow velocity is not effective in RO fouling. Another result shows that fouling propensity decreases as cross flow velocity increases. Table 5.3 shows the effect of fouling mitigation by increasing cross flow velocity.

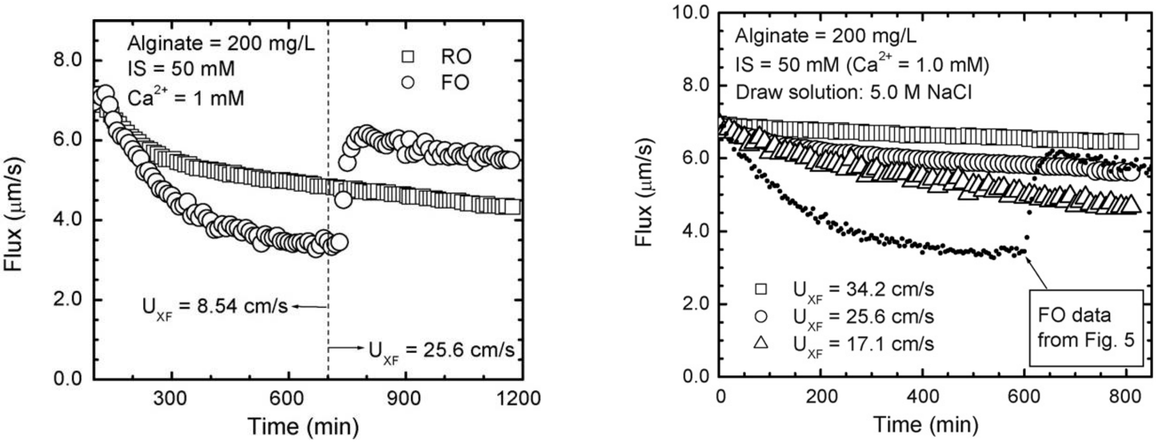


Figure 5.17. Impact of cross flow velocity on fouling behavior (Lee et al. 2010)

Table .5.3. Effect of fouling mitigation by increasing cross flow velocity

Ref	Foulants	pH	Cross flow velocity	Duration	Efficiency
Li et al. (2012)	Seawater pre-filtered by a 10 mm capsule filter	8.2	4.2 cm/s to 16.7 cm/s	Fouling run	Flux decline reduced
Boo et al.	1 g/L of 139 nm	4	8.5 cm/s to	Not	Fully

(2012)	silica		25.6 cm/s	mentioned	reversible
		9			80%
Boo et al. (2013)	75 mg/L of alginate, SRNOM, and BSA	6.5-7.5 of feed, 7.5-8.5 of draw	10.7 cm/s to 32.1 cm/s	Fouling run	Flux decline reduced
Mi and Elimelech (2010)	200mg/L alginate + 0.5 mM $CaCl_2$	5.8	8.5 cm/s to 21 cm/s	15 min	Fully reversible
Liu and Mi (2012)	Gypsum scaling	7.5	8.5 cm/s to 21 cm/s with air bubbles	20 min	Fully reversible
	Combined gypsum-alginate				75%

5.3.3 Air scouring

Air scouring has been proved to be an effective way to reduce membrane fouling in membrane bioreactors. Mi and Elimelech (2010) investigated the fouling reversibility of FO, introducing air bubbles into the DI water stream for air scouring. As shown in Figure 5.18, using air bubble combined with increasing cross flow shorten cleaning time. The enhanced cleaning efficiency by air bubbles is primarily due to the shear force increased overall on the membrane and to more turbulence generated by the introduction of bubbles (Ndinisa et al. 2006). Yangali-Quintanilla et al. (2011) obtained the same result introducing air scouring that the FO membranes were hydraulically cleaned for 30 min, operating with real seawater and secondary wastewater effluent (SWWE) in FO. For the 28% flux decline, the reversible fouling was 98.8%. On the other hand, Liu and Mi (2012) found that flux recovery by air scouring for combined gypsum and alginate fouling was low. The synergistic fouling effect is mainly a result of the intensified gypsum scaling with alginate molecules. Alginate molecules act like nuclei in gypsum crystal growth, thus significantly increasing the size of gypsum crystal and accelerating crystallization kinetics.

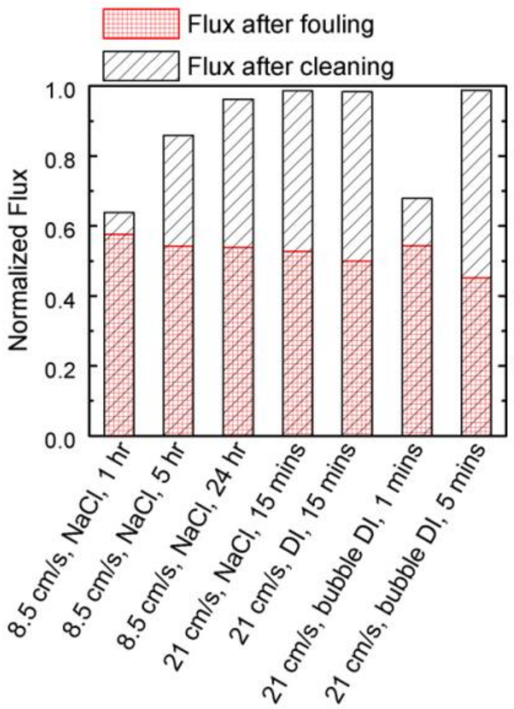


Figure 5.18. Cleaning efficiency under various crossflow, bubble and DS conditions (Mi and Elimelech 2010)

5.3.4 Cross flow type

Boo et al. (2013) investigated novel method for mitigating fouling. Pulsed flow has potential to remove foulants from the membrane surface. In Figure 5.19. the notable effect of pulsed flow on organic fouling mitigation is clearly demonstrated. The mechanisms by which pulsed flow controls membrane fouling include: (1) creating a highly turbulent flow regime, which leads to lower foulant concentration polarization, (2) increasing the overall shear force near to the membrane surface, which hinders foulant accumulation on the membrane surface, and (3) inducing membrane movement, which dislodges foulants from the membrane surface.

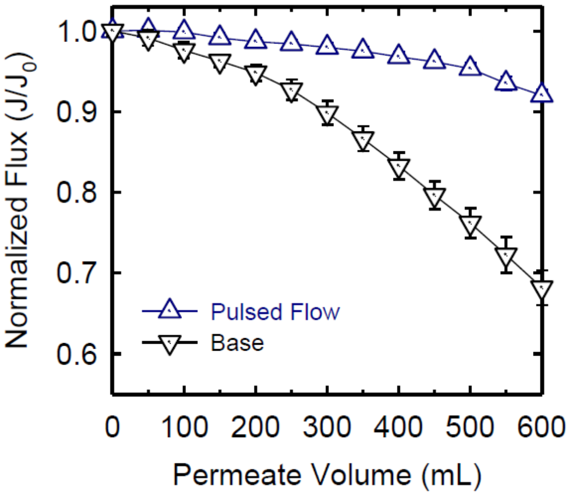


Figure 5.19. Impact of hydrodynamic control strategies on FO fouling mitigation: pulsed flow (Boo et al. 2013)

5.3.5 Osmotic backwashing

Osmotic backwashing was developed to remove foulants on membrane surface. As shown in Figure 5.20., when DS is replaced by deionized (DI) water, the osmotic pressure gradients is in reverse and the permeate (i.e. backwash water) flows from draw (DI water) to feed sides. Therefore, the deposited foulants are possibly separated by this opposite flow and then washed away by the cross flow.

Kim et al. (2012) applied osmotic backwashing to clean FO membranes after being fouled by organic and inorganic foulants (Figure 5.21). Various backwashing conditions under two different membrane orientations were also studied for better cleaning efficiency. For equivalent permeation drag force in both membrane orientation, initial water flux was adjusted to be the same.

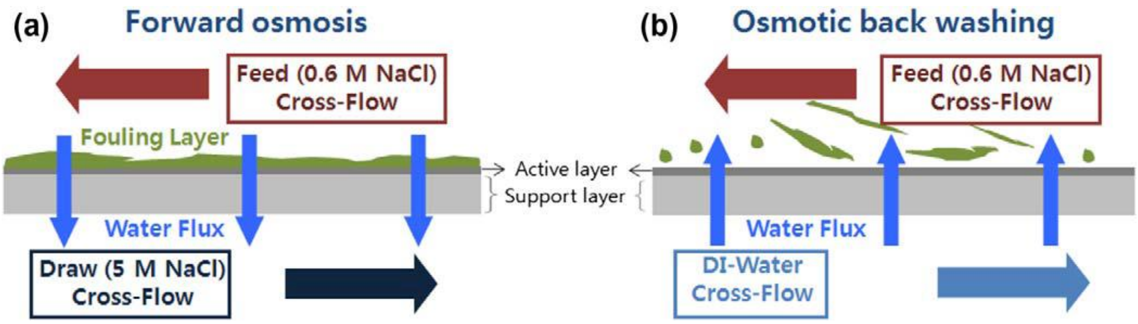


Figure 5.20. A conceptual illustration of the osmotic back washing (Kim et al. 2012)

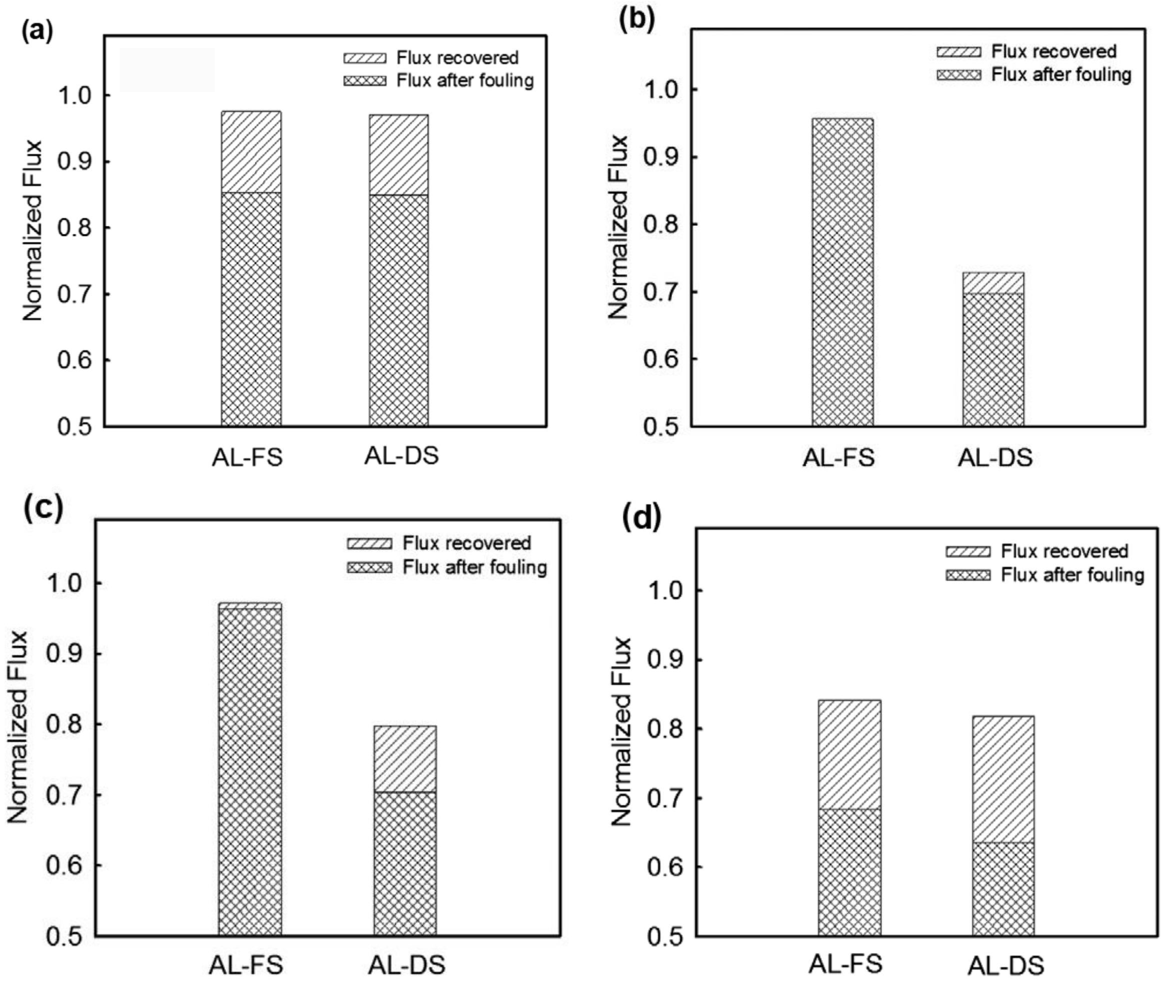


Figure 5.21. Fouling and osmotic backwashing experiments with 200 mg/L of alginate (a), 200mg/L of humic acid (b), 2 g/L of 20 nm silica particles (c) and 2 g/L of 100 nm silica particles (d) (Kim et al. 2012)

Although water flux decline showed difference depending on foulants type and membrane orientation, overall efficiency of osmotic backwashing is remarkably higher. The difference may be caused by various fouling parameters that are discussed above. For example, humic acid (HA) is effectively removed by shear force on FO mode. On the other hand, HA captures in porous structure of support layer in PRO mode and then is not removed effectively through the osmotic backwashing. It can be interpreted in a way that hydrophobicity of HA strengthens chemical interaction with FO membrane surface and possibly increased their attachments to the inside pores of PRO mode.

In the case of experiments with 20 nm particles, the flux decline on PRO mode is more severe than that on FO mode. The shear force generated by cross flow is not able to effectively flush small colloidal particles entrapped inside pores of support layer. However, a noticeable flux recovery on PRO mode was found after osmotic backwashing, although flux was not completely recovered. In contrast, fouling behaviors of 100 nm SiO_2 particles are almost the same during FO mode and PRO mode runs. These findings can be explained by CEOP at the presence of particle cake layer formed by large silica colloidal particles. Furthermore, large colloids are less removed from the membrane surface because of small diffusion coefficient of large particles. However, achieving a flux recovery is over 85% at the end of osmotic backwashing. The results are summarized in Table.5.4.

Table 5.4. Fouling characteristics in terms of foulant types, orientation and back washing

Foulants	Characteristic	Orientation	Flux decline	Back washing efficiency	Cause
Alginate	Hydrophilic	FO mode	Mild	High	Less chemically interacted
		PRO mode	Mild	High	Less chemically interacted
Humic acid	Hydrophobic	FO mode	Mild	Low	Chemically interacted
		PRO mode	Severe	Low	Chemically interacted Attachment to inside pore
Silica	20 nm	FO mode	Mild	Low	Shear force
		PRO mode	Severe	Medium	Entrapped inside pore
	100 nm	FO mode	Severe	Medium	CEOP Small diffusion coefficient
		PRO mode	Severe	Medium	CEOP

					Small diffusion coefficient

The osmotic backwashing is also applied to an osmotic membrane bioreactor (OsMBR) system that utilizes a submerged FO membrane in the bioreactor. FO offers advantage of lower fouling propensity than pressure driven systems, and therefore, requires less frequent backwashing compared to the MF or UF process.

Figure 5.22 shows the relationship between water flux and osmotic backwashing. The flux decreases by nearly 20% from the initial flux of the virgin membrane after 14 d. Osmotic backwashing was able to recover approximately 50% of this flux loss, leaving an overall 10% lower flux due to irreversible fouling. Similar trend was observed after 21 and 28 d. After an initial phase of irreversible fouling (occurring in the first 14 days), later phases of fouling were more reversible and the water flux remains a constant value over time. These results were in agreement with previous studies (Achilli et al. 2008; Lay et al. 2012).

On the other hand, Valladares Linares et al. (2013) produced conflicting results that osmotic backwash did not help recover flux. This research showed the cleaning efficiency of several methods to remove NOM-fouling on the active layer of FO membrane submerged in a municipal secondary wastewater effluent (SWWE). This research suggests hypothesis regarding ineffective osmotic backwashing: (1) as replacing the feed and draw with 4% NaCl and DI respectively, the interaction of the salt in the cleaning solution with the foulants might have enhanced the pore blockage and thus the water flux kept decreasing and (2) hypothesis is that salt accumulation in the fouling layer reduces the osmotic pressure gradient, and then, eventually reduces the flux through the membrane.

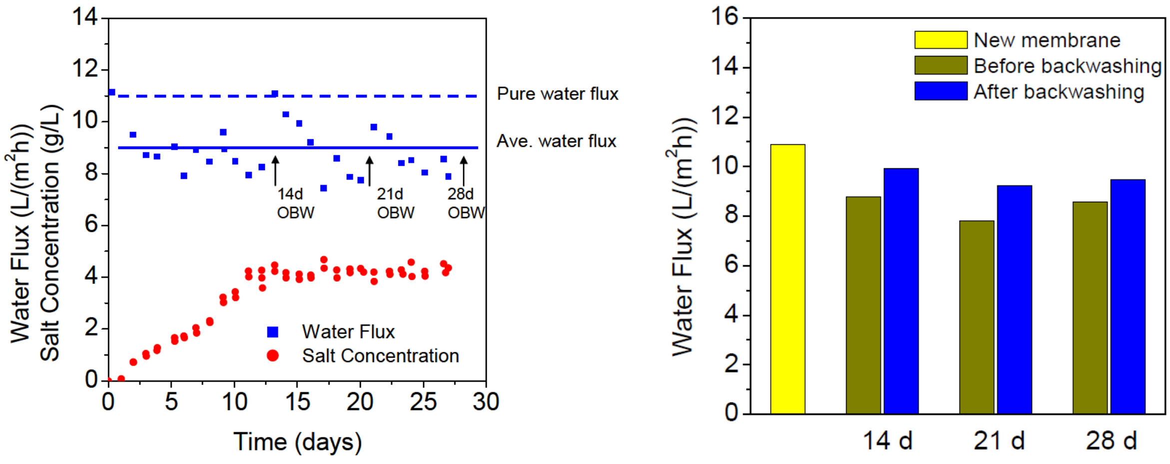


Figure 5.22. Comparison between water flux for the new membrane, and before/after osmotic backwashing at 14, 21, and 28 d (Holloway et al. 2007)

5.4 Concluding remarks

5.5 Abbreviations/Nomenclature

CP	concentration polarization
FO	forward osmosis

5. 6 References

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