Membrane Fouling in Osmotically Driven Membrane Processes: A Review

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Abstract

The utilization of osmosis for engineered applications sparked off various emerging technologies relying on osmotically driven membrane processes (ODMPs). Represented by forward osmosis (FO) and pressure retarded osmosis (PRO), ODMPs show great promise to leverage the global water-energy nexus and have drawn considerable attention in recent years. However, their performance in practical applications is significantly affected by membrane fouling. Membrane fouling is a complex problem and is associated with the foulant deposition, concentration polarization and reverse solute diffusion (RSD) in ODMPs. The current paper provides a comprehensive review on membrane fouling in ODMPs with a focus on the elaboration of the factors and mechanisms governing the fouling behavior. Among those fouling factors and mechanisms, some are also applicable for pressure-driven membrane processes (e.g., reverse osmosis (RO) and nanofiltration (NF)), such as the effects of hydrodynamic conditions, feedwater composition, and membrane material and properties, and the cake-enhanced concentration polarization (CE-CP) mechanism. Others are unique for ODMPs, such as the effects of draw solution composition and membrane orientation, the internal concentration polarization (ICP) self-compensation effect, and the RSD-enhanced fouling. A general osmotic-resistance filtration model for ODMPs is presented in this paper to assist in the interpretation of the intrinsic interrelationships among those fouling factors and mechanisms. The impact and mechanisms of membrane fouling on contaminates removal are also reviewed briefly based on the limited existing literature on this topic. Finally, the available membrane fouling control strategies for ODMPs are summarized upon understanding the cause and effect of fouling. Based on the current review, future research prospects are proposed for further studying the membrane fouling in ODMPs.

Keywords: membrane fouling, osmotically driven membrane processes (ODMPs), forward osmosis (FO), pressure retarded osmosis (PRO), fouling control

Table of Contents

1. Introduction

Osmotically-driven membrane processes (ODMPs), represented by Forward Osmosis (FO) and Pressure Retarded Osmosis (PRO), are emerging membrane technologies that show great promise to address the global challenges in water and energy supply [1-5]. FO utilizes a high osmotic pressure draw solution (DS) to “draw” the water from a low osmotic pressure feed solution (FS) through a semi-permeable membrane. With a suitably selected DS for FO, the product fresh water can be sustainably separated out from the diluted DS using an appropriate separation approach [6]. PRO is a process similar to FO except that a hydraulic pressure lower than the osmotic pressure difference across the membrane is applied in the DS side. This hydraulic pressure “retards” the permeation of water from FS, converting the free energy of mixing (i.e., the salinity-gradient energy or osmotic energy) into a useful work that can be further harvested as electricity by depressurizing the permeate-enhanced DS through a hydroturbine [7].

Since 2005 when Elimelech and coworkers reported the use of FO technology for desalination [6], the publications on ODMPs (especially FO) have been expanding at an exponential rate (refer to Figure 1). The applications based on FO have been extended to various water industries (i.e., water treatment, wastewater reclamation and brackish/seawater desalination) [1, 3, 5, 8, 9]. Without the requirement of DS separation, the stand-alone FO can be used to concentrate the feed water (e.g., concentration of anaerobic digester centrate [10] and concentration of wastewater for energy and nutrients recovery [11-13]) or dilute the draw solution (e.g., fertigation [14] and osmotic dilution of seawater for desalination [15, 16]). The major advantage of the stand-alone FO process over the pressure-driven reverse osmosis (RO) and nanofiltration (NF) processes lies in the very low energy consumption (mainly for pumping the FS and DS).

With the development of DS regenerating technologies and novel draw solutes synthesis, the DS regeneration may become less energy-intensive and the cost for separating the product water from the diluted DS may be substantially reduced [2, 17, 18]. PRO is primarily used for recover the renewable salinity-gradient energy. Although proposed around 40 years ago [7], it did not achieve such rapid advancement until recent decade especially after the operation of the first prototype PRO osmotic power plant in Norway in 2008 [3, 5, 19, 20]. Recent analysis shows that PRO may be superior to its competing technology reverse electrodialysis (RED) in terms of energy efficiency and power density for extracting salinity-gradient energy [21].

Figure 1

Despite the great attractiveness of ODMPs, their performance can be significantly affected by membrane fouling – which is caused by the deposition of suspended particles or colloids, organic macromolecules, sparingly soluble inorganic compounds, microorganisms, or their mixtures on (or even inside) the membrane [3, 5]. Membrane fouling not only reduces the permeate water flux (and osmotic power output in the case of PRO), water recovery, and permeate quality, but also causes increased operating cost and shortened membrane life. Among the vast number of studies on ODMPs, a significant portion of them are related to membrane fouling (Figure 1). These fouling papers are to evaluate the impact of fouling on the process performance [10, 22-26], to explore the factors and mechanisms governing the fouling behavior [27-33], and/or to develop approaches for mitigating fouling [34-38]. Particularly, understanding the causes of fouling in ODMPs is of paramount importance for developing effective fouling-control strategies. Most of the fouling studies in ODMPs reported similar fouling mechanisms to those in pressure-driven RO/NF processes. These mechanisms are associated with the complex interplay of solution chemistry, hydrodynamic conditions and membrane surface properties [27, 39]. On the other hand, some other studies reported several fouling mechanisms that are unique in ODMPs, such as the relationship between fouling and ICP self-compensation effect [29] and the enhanced fouling due to reverse solute diffusion [40]. Due to the complex nature of fouling in ODMPs and the lack of comprehensive reviews on this important topic, a timely review on this subject is necessary and will be of help for well understanding the causes, effects, and control strategies of membrane fouling in ODMPs.

This paper focuses on the comprehensive review of membrane fouling in ODMPs based on the existing literature. The review starts with the revisit of the fundamentals of mass transport in ODMPs. Then, an osmotic-resistance filtration model differentiating all the driving forces is proposed for the general discussion of membrane fouling in ODMPs. Subsequently, the factors and mechanisms governing the fouling in ODMPs are discussed and compared with those in pressure-driven RO/NF processes. The impact of membrane fouling on contaminants removal (particularly trace contaminants) in ODMPs and the rejection mechanisms are also reviewed. Next, the approaches for mitigating membrane fouling are summarized. Finally, future research needs on fouling and its control in ODMPs are recommended.

2. Fundamentals of mass transport in ODMPs

Before reviewing membrane fouling, it is essential to understand the mass (i.e., water and solute) transport in ODMPs. In ODMPs, water in the FS of lower concentration (or higher water chemical potential) transports through the membrane into the DS of higher concentration (or lower water chemical potential). Meanwhile, draw solute in the DS diffuses into the FS in an opposite direction to the water permeation. Solute diffusion in this fashion is referred to as reverse solute diffusion (RSD) [29, 41-44], which is a unique phenomenon in the ODMPs. The rates of water permeation and RSD are affected by internal concentration polarization (ICP) within the membrane porous support layer and external concentration polarization (ECP) near the membrane surface, as the ICP and ECP can lead to the reduction of their effective driving force (i.e., the osmotic pressure difference or concentration difference across the membrane) [29, 45-49]. On the other hand, water permeation and RSD will influence the extent of ICP and ECP [29, 42, 50]. This section will revisit the fundamentals of water and solute transport with a focus on RSD, ICP, ECP, their relationships, and their influences on water flux behavior under non-fouling conditions in ODMPs. The role of RSD, ICP and ECP on membrane fouling will be further discussed in Section 3.

2.1. Water permeation and reverse solute diffusion

The membrane processes can be classified into reverse osmosis (RO), pressure retarded osmosis (PRO), forward osmosis (FO) and pressure assisted osmosis (PAO), based on the water flux direction as well as the relationship between the net applied hydraulic pressure (ΔP) and osmotic pressure gradient across the membrane ($\Delta\pi_{eff}$). These four processes are schematically illustrated in Figure 2a and their water flux can be generally described by the following equation [46]:

$$J_w = A\left(\Delta\pi_{eff} - \Delta P\right) \qquad (1)$$

where A is water permeability coefficient of the membrane; $\Delta\pi_{eff}$ is effective osmotic pressure difference across the membrane; ΔP is the net applied hydraulic pressure.

Figure 2b shows J_w as a function of ΔP for the four processes. Since this review focuses primarily on FO and PRO, the sign convention is such that a positive J_w indicates that the permeation of water from the low concentration FS to the high concentration DS (i.e., the water flux direction in FO and PRO). Accordingly, a positive ΔP indicates a higher hydraulic pressure in DS than that in FS. In Figure 2b, the relationship between J_w and ΔP for each process is summarized below:

1) For RO, $\Delta P > \Delta\pi_{eff}$ (i.e., a net hydraulic pressure applied in the high concentration solution is greater than $\Delta\pi_{eff}$). The driving force is the hydraulic pressure and water permeates from the high concentration side to the low concentration side. The magnitude of water flux $\left|J_w\right|$ increases with increasing ΔP.
2) For PRO, $0 < \Delta P < \Delta\pi_{eff}$ (i.e., a net hydraulic pressure is applied in the high concentration DS side but its value is lower than $\Delta\pi_{eff}$). The driving force is the osmotic pressure difference and water permeates from FS into DS. Water flux J_w decreases with increasing ΔP such as the pressure in DS acts to "retard" the water flux.
3) For FO, $\Delta P = 0$ (i.e., no hydraulic pressure is applied). The driving force is osmotic pressure and the water permeates from FS into DS.
4) For PAO, $\Delta P < 0$ (i.e., a net hydraulic pressure is applied in the low concentration FS side). The driving force is the osmotic pressure together with the hydraulic pressure. The magnitude of water flux J_w increases with increasing the applied pressure in FS.

It is worthwhile to note that PAO has received some attention recently [51-53], as the additionally applied pressure in FS is necessary to maintain the FS flow in a spiral-wound FO module in practical operation and can potentially increase the water recovery of an FO system.

Figure 2

The reverse solute flux (J_s) can be generally described below:

$$J_s = B\Delta C_{eff} \tag{2}$$

where B is solute permeability coefficient of the membrane; ΔC_{eff} is effective solute concentration difference across the membrane.

Besides Eq. (2), another useful quantification of revere solute diffusion (RSD) is the specific reverse solute flux (J_s / J_w) that describes the amount of draw solute diffusing into FS per unit volume of water permeating into DS [29, 42, 43, 54]. Combining Eq. (1) and (2) and relating $\Delta\pi_{eff}$ to ΔC_{eff} with van't Hoff Equation, J_s / J_w in ODMPs can be generally described by the following equation [42]:

$$\frac{J_s}{J_w} = \frac{B}{A\beta R_g T}(1 + \frac{A\Delta P}{J_w}) \tag{3}$$

where β is the van't Hoff coefficient and is a constant if the concentration and osmotic pressure of the solution follow a linear relationship, R_g is the universal gas constant, and T is absolute temperature. The positive J_s / J_w indicates the direction of solute diffusion is opposite to that of water permeation.

Figure 2c shows the theoretical J_s / J_w as a function of ΔP in different regions. Obviously, J_s / J_w is a hyperbolic curve with two asymptotic lines of $x = \Delta\pi_{eff}$ and $y = 0$ (i.e., x axis). Specifically, the relationship between theoretical J_s / J_w and ΔP in different regions is summarized below:

1) In the RO region, the negative J_s / J_w indicates that the solute diffusion has the same direction with the water flux. The absolute J_s / J_w value decreases with increasing ΔP and approaches to zero when ΔP is infinitely great.
2) In the PRO region, the theoretical J_s / J_w increases with increasing the applied hydraulic pressure [42, 55] and becomes infinitely large when the ΔP approaches to $\Delta\pi_{eff}$ due to the diminished water flux at osmotic equilibrium.
3) In the FO region where $\Delta P = 0$, J_s / J_w is a constant value for a given membrane and is only dependent on the membrane intrinsic separation properties and working temperature [29]. Theoretical J_s / J_w is independent of the operating conditions, such as DS and FS concentration, membrane orientation and hydrodynamic conditions [44, 54, 56].
4) In the PAO region, the theoretical J_s / J_w decreases with increasing the hydraulic pressure applied in FS and approaches to zero when the ΔP becomes infinitely great in FS.

In reality, the experimentally measured J_s / J_w may not exactly follow the trend in Figure 2c, because the membrane separation properties (*A* value and *B* value) may change with the operating conditions and solution chemistry. For example, it has been reported that membrane deformation at high pressure PRO and PAO operation can lead to the significant loss of membrane rejection [42, 53, 55, 57]. The high concentration DS in contact with the membrane active layer may also lead to the change of membrane separation properties [58]. RSD is a very important phenomenon in ODMPs, as it can lead to the loss of draw solute, enhancement of concentration polarization (Section 2.2 and Section 2.3), and the change of feedwater chemistry that may significantly influence the membrane fouling behavior (Section 3.1.3 and Section 3.2.5).

Power density (W) can be used to characterize the energy consumption or energy production in these processes, as expressed in Eq. (4). Figure 2d shows the relationship between W and ΔP. In the RO and PAO regions, the negative W value indicates that energy is consumed in these processes. In contrast, W value is positive in PRO region, which indicates that energy (i.e., salinity-gradient energy) can be extracted from this process. In FO region, energy is neither consumed nor produced in ideal case (where the energy for recirculating FS and DS is not considered).

$$W = J_w \Delta P \tag{4}$$

2.2. External and internal concentration polarization

Concentration polarization (CP) refers to the phenomenon that solute concentration near a membrane surface differs from that in bulk solution. The occurrence of CP not only can reduce the effective driving force across the membrane (which can in turn reduce the permeating water flux), but also may be associated with membrane fouling [59, 60]. In ODMPs, two forms of CP are applicable – external concentration polarization (ECP) and internal concentration polarization (ICP) (Figure A1 in Appendix A). ECP occurs near both sides of membrane surfaces. It can be alleviated through optimization of the hydrodynamic conditions (such as increasing cross-flow velocity). On the other hand, ICP occurs within the membrane porous support layer that acts as an unstirred layer to hinder solute diffusion. Therefore, ICP plays a dominant role in the performance of ODMPs.

ECP and ICP behave differently in different membrane orientations, as shown in Figure A1. Due to ICP and ECP, the actual solute concentration at the support-active layer interface (C_i) and on the active layer surface (C_{al}) respectively is different from that in the bulk solution. Table 1 summarizes the mathematical equations and their physical interpretations to describe the C_i and C_{al} due to dilutive CP and concentrative CP in different orientations. It should be noted that CP in ODMPs are contributed by both solution convection and reverse solute diffusion [42, 50, 55, 61].

Table 1

Based on the equations in Table1 and Eq. (1) and (3), the water flux due to ECP and ICP in ODMPs can be generally described by following equations [55].

$$J_w = K_{overall} \ln\left[\frac{\pi_{ds} + \frac{B}{A}\left(1+\frac{A\Delta P}{J_w}\right)}{\pi_{fs} + \frac{B}{A}\left(1+\frac{A\Delta P}{J_w}\right) + \frac{J_w}{A}\left(1+\frac{A\Delta P}{J_w}\right)\exp\left(\frac{J_w}{k_{ecp,al}}\right)}\right] \quad \text{(AL-FS orientation)} \quad (5)$$

and

$$J_w = K_{overall} \ln \left[\frac{\pi_{ds} + \frac{B}{A}\left(1+\frac{A\Delta P}{J_w}\right) - \frac{J_w}{A}\left(1+\frac{A\Delta P}{J_w}\right)\exp\left(\frac{J_w}{k_{ecp,al}}\right)}{\pi_{fs} + \frac{B}{A}\left(1+\frac{A\Delta P}{J_w}\right)} \right] \quad \text{(AL-DS orientation)} \quad (6)$$

where $K_{overall}$ is the overall mass transfer coefficient. $K_{overall}$ is dependent on the mass transfer coefficient within the support layer k_{icp}, that near the surface of the active layer $k_{ecp,al}$ (i.e., k_{cecp} in AL-FS orientation or k_{decp} in AL-DS orientation), and that near the surface of support layer $k_{ecp,sl}$ (i.e., k_{decp} in AL-FS orientation or k_{cecp} in AL-DS orientation). $K_{overall}$ can be determined by Eq. (7) as follows:

$$\frac{1}{K_{overall}} = \frac{1}{k_{icp}} + \frac{1}{k_{ecp,sl}} + \frac{1}{k_{ecp,al}} \quad (7)$$

2.3. Osmotic-resistance filtration model for ODMPs

While Eqs. (6) and (7) are extensively used to model water flux with emphasis on the effects of ICP and ECP, an osmotic-resistance filtration model is proposed in this paper to describe the water flux which explicitly differentiates all the driving forces and hydraulic resistances. A similar model has been shown to be valuable for the better understanding of membrane fouling in RO [62]. As schematically illustrated in Figure 3, the apparent concentration driving force for ODMPs (proportional to osmotic driving force), ΔC_{bulk}, can be divided into four components – (1) the effective concentration driving force (ΔC_{eff}), (2) the loss of driving force due to ICP (ΔC_{dicp} of DS in AL-FS orientation or ΔC_{cicp} of FS in AL-DS orientation), (3) loss of driving force due to CECP (ΔC_{cecp}), and (4) loss of driving force due to DECP (ΔC_{decp}). The mathematical equation for osmotic-resistance filtration model is expressed in Eq. (8) for AL-FS orientation and Eq. (9) for AL-DS orientation respectively, which can be obtained by rearranging Eq. (6) and Eq. (7) and relating Eq. (3) (detailed derivation is provided in Appendix A).

$$J_w = \frac{(\pi_{ds} - \pi_{fs}) - \Delta P - F_{cecp}\left(\pi_{fs} + \frac{J_s}{J_w}\beta R_g T\right) - F_{dcp}\left(\pi_{ds} + \frac{J_s}{J_w}\beta R_g T\right)}{\mu R_m}$$

(AL-FS orientation) (8)

and

$$J_w = \frac{(\pi_{ds} - \pi_{fs}) - \Delta P - F_{ccp}\left(\pi_{fs} + \frac{J_s}{J_w}\beta R_g T\right) - F_{decp}\left(\pi_{ds} + \frac{J_s}{J_w}\beta R_g T\right)}{\mu R_m}$$

(AL-DS orientation) (9)

where μ is the feedwater viscosity, and R_m is the membrane hydraulic resistance. The membrane water permeability (A) is related to μ and R_m by Eq. (10)

$$A = \frac{1}{\mu R_m} \qquad (10)$$

F_{cecp} and F_{dcp} are concentration polarization factors for CECP at the active layer side and dilutive CP (DCP, i.e., both DICP and DECP) at the support layer side in AL-FS orientation, respectively; F_{ccp} and F_{decp} are concentration polarization factors for concentrative CP (CCP, i.e., both CICP and CECP) at the support layer side and DECP at the active layer side in AL-DS orientation, respectively. The mathematical expressions for these concentration factors are described in Appendix A. It is worth noting that concentrative CP factors (both CICP and CECP) are a function of $\exp(J_w / k)$, while the dilutive CP factors (both DICP and DECP) are a function of $-\exp(-J_w / k)$.

Figure 3

The osmotic-resistance filtration model (refer to Figure 3 and Eqs. (8) and (9)) provides strong physical meanings to explain the ICP, ECP and water flux behavior in ODMPs:

(1) CECP, ICP and DECP cause significant loss of driving forces, where ICP typically plays a dominant role [46, 47, 63]. Moreover, the losses of driving forces due to CECP, ICP and DECP are specifically described in the osmotic-resistance filtration model.

(2) The loss of driving force due to concentrative CP (CCP, i.e., both CECP and CICP) in the FS side, ΔC_{ccp}, is contributed by the accumulation of solute due to FS convection (C_{fs}) and RSD (J_s / J_w) at the boundary layer interface. The relative importance of FS convection and RSD on CCP is dependent on the relative value of C_{fs} and J_s / J_w [61]. When $C_{fs} >> J_s / J_w$, FS convection dominates the CCP and the influence of RSD is negligible; when $C_{fs} << J_s / J_w$, RSD dominates the CCP and the influence of FS convection is negligible; when C_{fs} and J_s / J_w are in the same level, both feed water convection and RSD play a significant role.

(3) The loss of driving force due to dilutive CP (DCP, i.e., both DECP and DICP) in the DS side, ΔC_{dcp}, is contributed by the DS dilution due to convection and loss of draw solute due to RSD. Severe RSD leads to a great ΔC_{dcp} due to significant loss of draw solute.

(4) Dilutive ICP (DICP) in AL-FS orientation is more severe than concentrative ICP (CICP) in AL-DS orientation [29, 64], since DICP causes more loss of driving force.

(5) ECP could be neglected at the support layer side when its boundary layer thickness is much smaller than the structural parameter of support layer (i.e., $\delta_{sl} << S$) (also refer to Eq. (A5) and Eq. (A10) in Appendix A).

3. Membrane fouling in ODMPs

3.1. General aspects of membrane fouling in ODMPs

3.1.1. External fouling and internal fouling in ODMPs

In ODMPs, membrane fouling can take place at different locations [27, 30, 31, 33, 65-74]. As illustrated in Figure 4a, membrane fouling in AL-FS orientation occurs by means of deposition of foulant from FS onto the membrane active layer surface and the subsequent formation of “cake layer”, which is similar to fouling in RO. Membrane fouling in this manner is referred to as **external fouling**. In the AL-DS orientation, the occurrence of membrane fouling is more complicated. Figure 4b illustrates several possible scenarios of fouling in this orientation. If the foulant has relatively small size and can enter into the membrane porous support layer with the feed water convection, it will either be adsorbed on the pore walls of the support layer or eventually be retained by the dense active layer and deposit on the back side of the active layer surface. Subsequently, the foulant that enters into the porous support layer can further attach on the foulant that has been adsorbed on the pore walls or deposited on the back side of active layer surface, which leads to the “pore clogging”. Membrane fouling in this manner is referred to as **internal fouling** (scenario (1) in Figure 4b). Under severe fouling conditions, the foulant will continue to deposit on the outer surface of the porous support layer in addition to internal pore clogging. Membrane fouling in this manner is the combined internal and external fouling (scenario (2) in Figure 4b). If the foulant has relatively large size and cannot enter into the porous support layer, the foulant may just deposit on the outer surface of the support layer. In this case, only external fouling occurs (scenario (3) in Figure 4b). If the feed water contains a mixture of foulants with different sizes, both external fouling and internal fouling may occur (scenarios (4) and (5) in Figure 4b).

Figure 4

Compared to internal fouling, external fouling occurs on the membrane surface and is more removable by optimizing the hydrodynamic conditions of the feed water (e.g., increasing cross-flow velocity [32, 36, 73, 75], using the feed spacer [36, 65, 68], introducing the pulsed flow [36], employing the air scouring [37]). Therefore, most of the researchers recommend the AL-FS orientation for FO application to prevent undesirable internal fouling [27, 29, 76], although the ICP in AL-FS is more severe than that in AL-DS orientation [29, 64]. In addition, unlike the external fouling in RO where the foulant layer on the membrane surface is compacted by hydraulic pressure, external fouling in ODMPs is more reversible than that in RO due to the lack of hydraulic compaction of the foulant layer [32, 75]. In contrast, internal fouling occurs within the porous support layer that acts as an unstirred layer. It is more difficult to control internal fouling by simple optimization of hydrodynamic conditions, and it is also less reversible than the external fouling [35]. Internal fouling occurs mostly in PRO membranes that are usually operated in AL-DS orientation considering the better membrane mechanical stability and higher power density [42, 55]. Although osmotic backwash method is developed for the cleaning of the foulants within the support layer [33, 35], the development of more effective strategies for internal fouling control will still be an important research topic in the future.

3.1.2. Osmotic-resistance filtration model for fouling in ODMPs and fouling mechanisms

Membrane fouling in ODMPs leads to a decrease of water flux due to the deposition of foulant on the membrane surface or within the membrane support layer. Fouling leads to an increase of overall membrane resistance for water transport (or reduce the overall membrane water permeability) and may change the extent of concentration polarization as well. Based on the osmotic-resistance filtration model (Section 2.3), the water flux for the fouled membrane can be expressed by Eq. (11) and Eq. (12) for AL-FS and AL-DS orientations respectively.

$$J_{w,f}=\frac{(\pi_{ds}-\pi_{fs})-\Delta P-F_{cecp,f}\left(\pi_{fs}+\frac{J_{s,f}}{J_{w,f}}\beta R_g T\right)-F_{dcp,f}\left(\pi_{ds}+\frac{J_{s,f}}{J_{w,f}}\beta R_g T\right)}{\mu(R_m+R_f)}$$

(AL-FS orientation) (11)

and

$$J_{w,f}=\frac{(\pi_{ds}-\pi_{fs})-\Delta P-F_{ccp,f}\left(\pi_{fs}+\frac{J_{s,f}}{J_{w,f}}\beta R_g T\right)-F_{decp,f}\left(\pi_{ds}+\frac{J_{s,f}}{J_{w,f}}\beta R_g T\right)}{\mu(R_m+R_f)}$$

(AL-DS orientation) (12)

where $J_{w,f}$ is the water flux of the fouled membrane, R_f is the additional hydraulic resistance imposed by the foulant deposition (e.g., the formation of a cake layer), $F_{cecp,f}$ and $F_{dcp,f}$ are the CECP and DCP factors for the fouled membrane in AL-FS orientation respectively, $F_{ccp,f}$ and $F_{decp,f}$ are the CCP and DECP factors for the fouled membrane in AL-DS orientation respectively, and $J_{s,f}/J_{w,f}$ is the specific reverse solute flux for the fouled membrane. The overall water permeability of the fouled membrane (A_f) can be determined by

$$A_f = \frac{1}{\mu\left(R_m + R_f\right)} \tag{14}$$

Figure 5

Eqs. (11) and (12) provide significant physical meanings for the analysis of water flux behavior in ODMPs. In general, the water flux can be affected by concentration polarization (CP), reverse solute diffusion (RSD), and foulant deposition (i.e., membrane fouling). Severe CP results in two adverse effects on water flux: (1) a substantial loss of osmotic driving force and thus the decrease of water flux, and (2) the elevation of foulant concentration near the membrane surface, which tends to accelerate the foulant deposition and thus the flux decline. The CP phenomenon can be further affected by RSD. Membrane fouling due to foulant deposition may influence the water flux in different ways, which is illustrated in Figure 5 and described below:

(1) ***Introducing additional hydraulic resistance***. The formed foulant cake layer causes additional hydraulic resistance for water permeation, which decreases the overall water permeability of the fouled membrane. Consequently, the water flux is decreased even at the same effective osmotic driving force. The modeling of the cake layer resistance and its dependence on solution chemistry has been extensively discussed in pressure-driven membrane processes (refer to the review papers [62, 77]), but only few studies reported this point in ODMPs [29, 33, 78, 79].

(2) ***Inducing fouling-enhanced concentration polarization***. The cake layer may hinder the diffusion of the solute within this unstirred layer back to the bulk feed solution and thereby further enhance the solute concentration near the membrane surface. This phenomenon is known as cake-enhanced concentration polarization (CE-CP). CE-CP is due to the significant

increase of the concentrative CP factors after fouling, i.e., $F_{cecp,f} > F_{cecp}$ in AL-FS orientation and $F_{ccp,f} > F_{ccp}$ in AL-DS orientation. As a result, the CE-CP leads to the further decrease of the effective osmotic driving force and thus the water flux. The effect of CE-CP is specifically discussed in Section 3.2.4.

(3) ***Changing membrane rejection property***. The deposited foulant on the membrane may change the membrane rejection property. For instance, the foulant may seal the membrane pores/defects for solute diffusion [25, 26] and thus increase the membrane rejection and reduce the reverse solute diffusion from DS into FS. This suggests that the specific reverse solute flux in fouling condition (i.e., $J_{s,f} / J_{w,f}$) may be reduced, i.e., $J_{s,f} / J_{w,f} < J_s / J_w$, resulting in the reduced CP. Such a condition can potentially lead to the peculiar phenomenon of increased water flux upon mild fouling. This point is discussed in Section 3.2.4 and Appendix B. Where RSD is worsened upon fouling ($J_{s,f} / J_{w,f} > J_s / J_w$), it can lead to more severe CP and thus greater water flux loss.

(4) ***ICP self-compensation effect***. This effect is more pronounced in AL-FS orientation [29]. During membrane fouling, the reduced water flux is compensated by a reduced ICP level (i.e., $F_{dcp,f} < F_{dcp}$ in Eq. (11)). This effect is specifically discussed in Section 3.2.3.

3.1.3. Role of reverse solute diffusion

Reverse solute diffusion (RSD) not only can increase CP, but also may lead to the change of feedwater chemistry and composition, which may further influence the membrane fouling behavior. When the draw solutes in DS diffuse into FS, these solutes could act as fouling promoters (e.g., divalent cations) to exacerbate the organic/colloidal fouling [30, 40], or fouling precursors (e.g., Ca^{2+} and SO_4^{2-}) to accelerate the inorganic scaling [31, 80], or nutrient to favor the biofilm development. Role of RSD on fouling is illustrated in Figure 6. It will be further discussed in more details in Section 3.2.2.3 and Section 3.2.5.

Figure 6.

3.1.4. Classification of membrane fouling and foulant interactions in colloidal system

Similar to membrane fouling in RO [62, 81, 82], fouling in ODMPs can be classified into four major groups according to the characteristics of foulants:

- Colloidal fouling – deposition of colloidal particles on the membrane;
- Organic fouling – deposition and adsorption of macromolecular organic compounds on the membrane;
- Inorganic scaling – precipitation or crystallization of sparingly dissolved inorganic compounds on the membrane; and
- Biofouling – adhesion and accumulation of microorganisms, and development of biofilm on the membrane.

The specific foulants in different groups are closely associated with the characteristics of feed waters in ODMPs and can be identified using a series of tools. Table 2 summarizes the properties of major foulants and the associated identification/characterization methods. Specifically, particulates, colloids, and organic macromolecules (such as polysaccharides, humic substances and proteins) are major foulant components in raw wastewater and treated wastewater effluent [83-86]. They are also ubiquitous in natural waters (e.g., river water, seawater, and groundwater) [27, 73, 82]. Zhou et al. [85] used the gas chromatography–mass spectrometry (GC-MS) to identify the soluble microbial products (SMPs) that contained a large portion of polysaccharides, proteins and humic substances from the raw and effluent wastewater. Recently, liquid chromatography–organic carbon detection–organic nitrogen detection (LC-OCD-OND) becomes increasingly popular for the identification of these organic foulants [66, 73, 87]. The deposited organic foulants on the membrane can be identified by Fourier Transform Infrared (FTIR) spectroscopy, solid state ^{13}C-nuclear magnetic resonance (NMR) spectroscopy and high performance size exclusion chromatography (HP-SEC) [86]. Total organic carbon (TOC) measurement and UV analysis were also performed to quantify the density of organic foulant deposition on the membrane [66, 78, 82, 88, 89]. Transparent exopolymer particles (TEPs) are another important organic foulant that typically exists in natural waters. Feedwater TEP can be characterized by two methods: microscopic enumeration and colorimetric determination, both of which rely on staining with alcian blue [72, 90-92]. Silica is a major inorganic foulant and generally exists in dissolved state or as colloidal particles in seawater, brackish water and wastewater [73, 93-95]. In addition, the sparingly dissolved salts (such as calcium carbonate, calcium sulphate, and calcium phosphate) from seawater, brackish water and wastewater are large components of inorganic foulants [28, 66, 96]. These inorganic foulants deposited on the membrane are extensively characterized by the scanning electron microscopy–energy dispersive X-ray apparatus (SEM-EDX) [66, 73] and X-ray Diffraction (XRD) [80, 97-99]. Bio-foulants (microorganisms) are major components of active sludge in membrane bioreactors (MBR) [38, 66, 100-102]. They also exist largely in natural water and can cause severe biofouling in seawater or brackish water desalination [38, 73, 103, 104]. The microbial communities of the biofilm can be characterized by the analysis of DNA extracted from their live cells using the microbiological methods such as polymerase chain reaction denaturing gradient gel electrophoresis (PCR–DGGE) and fluorescence in situ hybridization (FISH) [85, 105].

Biofouling can be monitored by a series of microscopic techniques, spectroscopic techniques and other non-classified techniques. These monitoring techniques were comprehensively review previously [106].

Table 2

In practical applications of ODMPs, the membrane is often fouled by a mixture of different types of foulants from natural or industrial wastewaters and the membrane fouling involves the combination of the above four categories [66, 73]. However, the mechanisms of mixed fouling are difficult to understand due to the complexity of the mixed foulants. To investigate the membrane fouling mechanisms, many existing fouling studies select synthetic water with a single foulant type [27, 29, 30, 32, 39, 40, 107, 108]. At the meantime, there have been increased number of studies on fouling of ODMPs with more complex feed waters, particularly in the context of osmotic MBRs [66, 100, 102]. The study of single model foulant system is useful because the properties of the selected model foulant can be better controlled/determined and the foulant-foulant interaction and foulant-membrane interaction can be more easily understood. The major physicochemical properties and identification methods of the selected model foulants are summarized in Table 2. These properties (e.g., size, shape, charge and some specific functional groups) are very important factors affecting the stability of the foulants in the feed water and their tendency to foul the membrane [62]. Upon understanding the fouling mechanisms in single-foulant system, future studies may focus on the investigation of fouling mechanisms in mixed-foulant systems which are more representative of practical applications.

3.2. Colloidal fouling and macromolecular organic fouling in ODMPs

Colloidal fouling and organic fouling share many similarities [62] and are thus discussed together. Their behaviors in ODMPs are affected by similar factors and can be explained by similar fouling mechanisms. Indeed, most of these factors and mechanisms are also applicable for inorganic scaling and biofouling. Section 3.3 and section 3.4 will specifically discuss the factors uniquely available for inorganic scaling and biofouling, respectively.

3.2.1. Colloidal interactions

Foulant deposition and accumulation are controlled by foulant-foulant and foulant-membrane interactions [62, 109]. For colloidal particles or macromolecules, the foulant-foulant interaction and foulant-membrane surface interaction in the aquatic environment can be explained by the classical Derjaguin–Landau–Verwey–Overbeek (DLVO) theory (i.e., the net interaction is the sum of van der Waals (VDW) and the electrostatic interaction force (also called electrical double layer (EDL) force)) [62, 110, 111]. For microorganisms whose size are comparable to colloidal particle’s size, the DLVO theory is also useful for the explanation of their adhesion and attachment on membrane surface during biofouling [106, 112, 113]. The aggregation of foulants and/or attachment of foulants onto membrane surface require overcoming the energy barrier resulting from the superimposed VDW and EDL interactions [111, 114]. The interaction energy is affected by those physicochemical properties of the foulants, which in turn are strongly dependent on the solution chemistry (such as pH, ionic strength, and ions that can have specific interactions with foulants). Further details of DLVO theory and their applicability in membrane fouling can be found in Tang et al. [62].

In addition to the classical DLVO theory, the non-DLVO interactions typically represented by the Lewis acid-base (AB) interactions have also been employed to explain the colloidal particle aggregation, as the classical DLVO theory cannot adequately explain and/or predict some experimental observations [115, 116]. Arising from the electron-acceptor–electron-donor interaction, the AB interfacial interaction can be either repulsive for hydrophilic colloids/surfaces due to hydration force or attractive for hydrophobic ones [115]. By addition of AB, VDW and EDL interactions, van Oss developed the extended DLVO (XDLVO) theory that is now extensively used to interpret the foulant-foulant/foulant-membrane interactions during membrane fouling [112, 114, 117, 118].

3.2.2. Factors affecting the fouling in ODMPs

Colloidal and organic fouling in ODMPs is a complex problem and is affected by a series of physical and chemical factors. In general, these factors can be classified into five groups:

(1) Operating conditions, such as initial water flux, cross flow velocity, spacer, aeration, and temperature;

(2) Feedwater characteristics, such as foulant type, foulant physicochemical properties (e.g., shape, size, charge, and functional group), foulant concentration, solution pH, ionic strength, and ionic composition (e.g., divalent cation);

(3) Draw solution composition, such as draw solution concentration and draw solute type;

(4) Membrane properties, such as membrane separation and structural properties (i.e., A, B and S values), and membrane surface properties (e.g., hydrophilicity/hydrophobicity, roughness, charge density, surface functional groups); and

(5) Membrane orientation.

It is worthwhile to note that the DS composition and membrane orientation are unique for fouling in ODMPs, while the other three groups are also applicable in pressure-driven membrane processes [62, 119]. The effect of these factors on fouling in ODMPs is reviewed in detail in this section.

3.2.2.1. Effect of hydrodynamic conditions

Hydrodynamic conditions (such as water flux and cross-flow velocity) are very important factors affecting membrane fouling in both osmotically-driven and pressure-driven membrane processes. The effect of hydrodynamic conditions on membrane fouling in pressure-drive membrane processes has been extensively studied before and reviewed by previous researchers [62, 119-121]. Most of the conclusions in pressure-driven membrane processes are also applicable in ODMPs. In general, more severe membrane fouling occurs at higher water flux and lower cross-flow velocity.

Tang and co-workers investigated membrane fouling in ODMPs using a variety of organic macromolecules (alginate, humic acid, and protein), inorganic colloids and scalant (e.g., silica and gypsum) and microorganism (e.g., microalgae) as model foulants, and consistently observed that membrane fouling became more severe at higher initial water flux levels [29-31, 39, 40, 65, 68, 78, 122]. Similar observations were also reported by the researchers from other groups [27, 122, 123]. They attributed the exacerbated fouling at higher water flux to: (1) larger volume of water permeating through the membrane (and thus greater amount of foulant brought towards the membrane surface due to the water convection), (2) more severe concentration polarization, and (3) greater hydrodynamic drag force towards the membrane surface. Tang and co-workers [29, 30, 39, 40, 68] also observed that fouling did not occur when the flux was below a certain value.

This flux-dependent membrane fouling phenomenon has been extensively observed in pressure-driven membrane processes and can be explained by the classical "critical flux" theory [62, 120, 124]. Those observations further reveal that the critical flux concept is also applicable in ODMPs [29, 30, 39, 40, 65, 125]. Interestingly, Zhang et al. observed that the flux at different initial water flux levels declined to an identical pseudo-stable flux at the end of long-term gypsum scaling experiments in PRO process [31]. They further indicated that the limiting flux behavior observed by Tang et al. in pressure-driven membrane process [62, 126] may also exist in ODMPs.

Cross-flow velocity over the membrane surface is another hydrodynamic factor affecting the membrane fouling through the influence of concentration polarization and mass transfer near the membrane surface. A number of studies observed that external fouling on the membrane surface was mitigated at increased cross-flow velocities in ODMPs when the membrane active layer is facing the feed water [28, 32, 36, 73, 127]. In the cross-flow membrane filtration system, colloidal particles transport towards the membrane due to the permeate water flux (i.e., the hydrodynamic drag perpendicular to the membrane surface) and meanwhile they are moved away from the membrane surface by the cross flow (i.e., the shear rate tangential to the membrane surface) and back transport towards the bulk solution. Therefore, the accumulation of retained foulant particles near the membrane surface (i.e., concentration polarization of foulants) is mitigated by the back transport of those particles to avoid severe membrane fouling [62]. At increased cross-flow velocity, such mass transfer (i.e., back transport) can be further enhanced which leads to reduced concentration polarization and membrane fouling. In addition to the increase of cross-flow velocity, greater shear rate induced by the use of spacer, the employment of pulsed flow and the introduction of air scouring in the feed flow channel have also been demonstrated to be effective against fouling in ODMPs [36, 37, 65, 68, 71, 72]. It is interesting to note that the increase of shear rate can be effective to reduce fouling on the membrane surface (external fouling) but may not be effective to reduce fouling within the support layer (internal fouling). For example, Arkhangelsky et al. found that the increase of cross-flow velocity has negligible effect on the flux behavior caused by inorganic scaling in AL-DS orientation where the internal fouling dominates the fouling behavior [35].

Although the use of feed spacer seems to reduce CP and external fouling, Wang et al. [68] found that particles had strong tendency to preferentially deposit near the “hydrodynamic dead zones” between the spacer filaments and membrane surface. This phenomenon was further confirmed by a recent direct fouling observation study using optical coherence tomography, which correlated the preferential foulant deposition to the hydrodynamic shadow zones created due to the presence of spacer [128]. These studies may suggest the critical research needs in improved spacer design for both ODMPs and pressure-driven membrane processes.

Temperature is regarded as an important physical parameter affecting the performance of ODMPs. Various researchers observed that the water flux is improved at increased temperatures in ODMPs [42, 69, 125, 129-131]. They attributed this observation to the combined effects of (1) reduced solution viscosity that reduces the membrane resistance and thus increases the water permeability, (2) increased solute diffusivity that increases the mass transfer in the boundary layer and thus reduces CP (especially ICP), and (3) increased osmotic pressure that increases the effective driving force. Temperature can also influence the membrane fouling behavior in

ODMPs. The effect of temperature on membrane fouling is virtually through the influence of hydrodynamic conditions (e.g., initial water flux level and mass transfer of foulant) and thermodynamic conditions (e.g., solution osmotic pressure, foulant solubility/stability, and foulant-foulant/foulant-membrane interactions). Kim et al. observed that flux decline caused by organic fouling was more severe at increased draw solution temperature primarily due to increased permeation drag at increased initial flux level, while membrane was less fouled at increased feed solution temperature due to the enhanced organic back diffusion from membrane surface and increased organic solubility [125]. They further observed that fouling became more severe when the initial flux was above certain critical flux at increased both feed and draw temperature where organic convection by permeation drag dominated fouling mechanism. Similarly, Zhao and Zou observed that the inorganic scaling became more severe at higher working temperatures during brackish water desalination by FO [69]. They attributed this to the increased permeation drag force at increased initial water flux and increased saturation index at increased feedwater recovery (or concentration factor).

It should be noted that the majority of existing fouling studies in ODMPs were performed using a small membrane coupon. In large-scale applications, fouling actually occurs in membrane modules, which may significantly differ from that in membrane coupons [132, 133]. Indeed, the spacer geometry and module configuration design can significantly influence the hydrodynamic conditions along the flow channels in membrane modules [133-135]. In addition, flux distribution and concentration factor vary at different locations in membrane modules [132-134, 136]. The concentration of feed solutes and draw solutes (through RSD) in feed water can lead to elevated local concentrations at high recovery [137], which can induces higher osmotic pressure of the feed, higher ionic strength (reduced EDL interaction), greater divalent ion concentration (promotion of organic fouling due to increased Ca^{2+} and Mg^{2+} concentration), and high saturation index (increased risk of scaling). These factors can influence the fouling behavior in membrane modules (or submerged membrane reactors) possibly in a highly non-linear manner. Therefore, there is a strong need to systematically investigate the fouling in membrane modules for optimizing the module designs for ODMPs.

3.2.2.2. Effect of feedwater composition

Feedwater composition is one of the most important factors affecting the membrane fouling. The effect of feedwater composition on membrane fouling in ODMPs is similar to that in pressure-driven membrane processes (a general review of the latter can be found in [62, 86, 119]), and has been extensively studied recently [27, 39, 71, 74, 108, 123]. Generally, the extent and rate of fouling can be strongly dependent on the type, properties, and concentration of foulants in the feed water. The fouling behavior can also be strongly dependent on the feedwater chemistry (i.e., ionic strength, pH and divalent ion concentration), since the feedwater chemistry can significantly influence the physicochemical properties (e.g., surface charge [82, 89, 110, 138-140], shape and conformation [141-143]) of foulants and the foulant-foulant/foulant-membrane interactions.

A number of studies on organic fouling in ODMPs selected macromolecules (such as alginate, humic acid and proteins) as model foulant, as these macromolecules are ubiquitous in natural waters and wastewater effluents that are commonly used as feed water for ODMPs [27, 29, 30, 32, 40, 71, 72, 75, 78, 107, 122]. Consistent with the observations in pressure-driven membrane processes [62], organic fouling in ODMPs is generally more severe at increased divalent cation concentration (Ca^{2+} and Mg^{2+}) and ionic strength [27, 40, 78, 107, 123]. Despite limited studies on the pH effect on fouling in ODMPs [39, 78, 123], the typical trends observed in pressure-driven membrane processes are generally applicable: fouling by negatively charged alginate or humic acid is aggravated by decreasing the pH [82, 138, 144]; the most severe protein fouling occurs at its isoelectric point (IEP) [89, 145, 146]. The effects of ionic strength and pH on organic fouling can be well explained by the role of charge interaction in membrane fouling – reduced electrostatic repulsion (or increased electrostatic attraction) will destabilize foulant and will lead to more severe membrane fouling [82, 89, 110, 138, 139]. Increased ionic strength weakens the interaction between charged particles/molecules due to the charge screening effects (also known as EDL compression) [82, 138]. Although it is commonly known that increasing ionic strength tends to promote more severe fouling for systems in which EDL repulsion prevails [62], opposite trend (more stable foulants and reduced fouling at high ionic strength) has been reported for systems dominated by EDL attraction [147].

The effect of divalent ion (e.g., Ca^{2+} and Mg^{2+}) on organic fouling arises from their specific interaction with functional groups in foulant molecules [27, 30, 40, 71, 75, 78, 107, 122]. Ca^{2+} and Mg^{2+} are known to have strong ability to form complex with the carboxylic groups of organic macromolecules and to bridge adjacent molecules together [27, 82, 138]. The specific interaction between the divalent ions and carboxylic groups can also reduce the charge density of the foulants [110, 140]. The bridging effect together with the reduced charge density can lead to the destabilization of organic foulants and thus promote their aggregation and deposition on the membrane. The relative effect of divalent ions on fouling seems to be strongly correlated to their affinity the target organic molecules. Mi and Elimelech [27] observed that Ca^{2+} in the feed water can enhance the organic fouling by alginate, bovine serum albumin (BSA), and Aldrich humic acid (AHA) in FO processes. They further reported that the flux decline was the highest for alginate, for which they attributed to its greatest carboxylic acidities and special structural characteristics that promotes the formation of cross-linked network alginate gel through intermolecular bridging. In contrast, She et al. [30] observed that the alginate fouling is weaker than the humic acid fouling in the presence of Mg^{2+}. This may be due to the weaker affinity between Mg^{2+} and alginate than that between Mg^{2+} and humic acid [118, 148].

In addition to the foulant properties and solution chemistry, foulant concentration is another factor affecting the rate and extent of fouling in ODMPs. Similar to that in pressure-driven membrane processes [138, 145, 149], the increase of foulant concentration leads to the increase of rate and extent of initial fouling in ODMPs [31, 107, 108]. However, the fluxes for different foulant concentrations tend to approach an identical pseudo-stable flux at long-term fouling tests in both pressure-driven membrane processes [126, 145, 149, 150] and ODMPs [31]. Interestingly, by plotting the flux (J_w) against the total amount of foulants convectively transporting to membrane surface per unit area (M_f), the flux decline curves for different foulant concentrations exhibit nearly identical trend [108, 145, 149]. According to previous studies [82, 108, 145], M_f can be determined by integrating the specific foulant convective mass transport ($C_{foulant} J_w$) over time t (Eq. (~~15~~1)). In addition, She et al. [145] suggested that the rate of foulant attachment onto a membrane (dm_f / dt) not only depends on the rate of foulant convective mass transport (dM_f / dt), but also depends on the attachment coefficient α (Eq. (~~16~~15)).

$$M_f = \int_0^t C_{foulant} J_w(t) dt$$ (14~~15~~)

and

$$\frac{dm_f}{dt} = \alpha \frac{dM_f}{dt}$$ (15~~6~~)

In Eq. (14~~5~~) and (15~~6~~), dM_f / dt may be interpreted as the frequency at which the foulant molecules collide with the membrane [143]. Not all the molecules encounter the membrane can attach to its surface. Here, α represents the probability of foulant attachment resulting from a given collision event [145]. Tang et al. [62, 89, 126, 138, 145, 147, 151] further suggested that α is a complex function of water flux, cross-flow velocity, solution chemistry, foulant properties and membrane properties, but independent of foulant concentration. The increased foulant concentration can increase the frequency of foulant collision with membrane (i.e., dM_f / dt) and thus increase the rate of foulant attachment on the membrane (i.e., dm_f / dt) at the initial stage of membrane fouling where increased rate of flux decline is observed. At long-term fouling test, α gradually diminishes with the decline of water flux and a pseudo-stable condition may be reached where the hydrodynamic drag force balances the barrier force (see Section 3.2.1) and there will be no further net foulant attachment on membrane any more even though dM_f / dt is still positive.

Apart from the studies on single-foulant fouling, a handful of studies focused on the fouling by mixed-foulants in ODMPs to simulate more practical situations. In the mixed-foulant system, the inter-foulant-species interactions can further influence the fouling behavior. Liu and Mi [71] investigated the combined organic fouling and gypsum scaling in FO and observed that the coexistence of both alginate and gypsum foulants caused faster flux decline than the superimposed effect of each individual foulant. This synergistic effect was caused by the accelerated gypsum crystal nucleation and growth in the presence of alginate. Gu et al. [78] investigated the FO membrane fouling by oppositely charged alginate and lysozyme and also observed the mixed fouling was more severe than the fouling by single foulant. Moreover, they observed that the ratio of lysozyme and alginate in the cake layer composition maintained a relatively stable value regardless of the mass ratio of lysozyme and alginate in bulk feed solution. They attributed this to (1) the enhanced inter-foulant-species interactions due to the electrostatic attractions between the oppositely charged foulants, and (2) the strong correlation between the inter-foulant-species interactions and the charge properties of foulants. Arkhangelsky et al. [122] investigated the FO fouling in ternary-foulant system by alginate, BSA and silica and observed that the ternary fouling is more severe than single-foulant fouling due to the stronger binding of alginate and BSA to silica in the presence of Ca^{2+}.

3.2.2.3. Effect of draw solution composition

Draw solution (DS) is a unique feature in ODMPs and is the source of the osmotic driving force for ODMPs. The DS composition, such as the type and concentration draw solute, not only influences water flux and solute flux but also affects membrane fouling behavior in ODMPs.

Generally, the increase of DS concentration can lead to the increase of initial water flux, which can potentially exacerbate membrane fouling. The fouling studies from different groups consistently reported that more severe fouling occurred at higher DS concentrations [27, 29-31, 39, 40, 65, 68, 122]. They attributed this to the greater hydraulic drag force as a result of higher flux that promotes the foulant deposition on the membrane. In this regard, the change of DS concentration essentially leads to the change of hydrodynamic conditions. Therefore, this DS concentration-dependent fouling behavior can be well explained by the flux-dependent fouling mechanism where the hydrodynamic condition (i.e., water flux) plays a dominant role (refer to Section 3.2.2.1).

While the increase of DS concentration can increase the initial water flux level in ODMPs, it can also elevate the rate of draw solute reverse-diffusion from DS into FS [29, 43, 44]. She et al. further found that the more severe fouling at higher DS concentrations can be caused by the elevated RSD in addition to the greater initial water flux level [30, 40]. RSD can lead to the accumulation of draw solute in the FS, which can further change the feedwater chemistry and thus the membrane fouling behavior. Higher DS concentration could lead to greater rate of RSD and thus more significant change of feedwater chemistry, which may cause more severe fouling. For example, She et al. observed greater ionic strength and specific ion (i.e., Ca^{2+} or Mg^{2+}) concentration in the feed solution at higher DS concentrations for the DS with Ca^{2+}/Mg^{2+}, which caused additional organic (alginate) fouling [30, 40]. Similar phenomenon was also observed by other researchers when studying biofouling [65] and inorganic fouling [31]. Upon the occurrence of a cake layer or support layer pore clogging, a greater RSD at high DS concentration also means more severe fouling-enhanced CP (refer to Eq. (10) and Eq. (11)), a phenomenon that will be further discussed in Section 3.2.4.

In addition to the DS concentration, the DS type can influence the membrane fouling as it can eventually influence the change of feedwater chemistry [30, 31, 39, 40, 65]. The DS type can determine the type of solute reverse-diffusion into FS and rate of its diffusion [40, 43, 152], which leads to the change of feedwater chemistry in different extent (refer to Figure 6). Tang and co-workers observed more severe fouling for the DS with divalent cations (e.g., Ca^{2+} and/or Mg^{2+}) than the DS without those specific ions even at the same initial water flux level [30, 31, 39, 40, 65]. This is due to the strong interaction between the foulant in the FS and these specific ions after they reversely diffuse from DS into FS (e.g., the Ca^{2+} can complex with COO- to enhance organic fouling or react with SO_4^{2-} to form inorganic scale). Moreover, She et al. observed that more severe organic (alginate) fouling for the $Ca(NO_3)_2$ DS than the $CaCl_2$ DS although these two DSs have same specific ion (i.e., Ca^{2+}) [40]. They attributed this to the faster reverse diffusion of Ca^{2+} for the $Ca(NO_3)_2$ DS than that for the $CaCl_2$ DS, which is caused by the different diffusion rate of their counter-ions (i.e., NO_3^- diffuses faster than Cl^-). Presumably, the choice of DS can also significantly affect the ionic strength in the feed water through RSD (e.g., microscale changes due to their CP in a foulant matrix and macroscale changes due to their accumulation in the feed water at increased water recovery). Such effects can cause complex and often highly nonlinear response, potentially leading to either increased or decreased fouling (e.g., depending on the dominant EDL interaction, see Section 3.2.2.2), changed solubility of inorganic minerals (thus scaling), and even affect the biological growth (thus biofouling). In other cases, DS may contain constituents that are precursors of foulants/scalents (e.g., Ca^{2+} as a precursor of gypsum scaling [31]) or that can be used as substrate (e.g., glucose based DS [153]) or nutrients (e.g., ammonia carbonate based DS [6]) for enhanced biofouling. The RSD of such fouling promoters can lead to severe water flux loss – a problem that does not occurred in pressure-driven membrane processes and has not been adequately addressed in the current literature on ODMPs fouling. The choice of DS and management of RSD deserve greater attention in the ODMPs' research community.

The effect of DS composition (concentration and type) on membrane fouling in ODMPs is via the combined effects of hydrodynamic conditions and feedwater chemistry. While the "critical flux" theory has been extensively adopted to explain the flux-dependent fouling in pressure-driven membrane processes [62, 120, 124], it is insufficient to explain the DS concentration-dependent fouling in ODMPs without the consideration of the feed chemistry effect. She et al. further extended the critical flux concept to a concept termed as "critical draw solution concentration" to explain the fouling in OMDPs [30]. The critical draw solution concentration refers to the threshold DS concentration below which fouling is not noticeable. Significant fouling can occur above the critical DS concentration. The concept of critical DS concentration involves the water flux and reverse solute diffusion in the explanation of membrane fouling in ODMPs [30, 65]. The RSD-enhanced fouling is an important and unique fouling mechanism in ODMPs and will be reviewed in more detail in Section 3.2.5.

3.2.2.4. Effect of membrane materials, structure and properties

Membrane fouling can be strongly influenced by the membrane materials and properties. The FO/PRO membranes used in ODMPs are typically comprised of a nonporous active layer (that rejects most of the ions and solutes) and a porous support layer (that allows the passage of most ions and solutes) [154-162]. The intrinsic separation properties of the active layer and the structural properties of the support layer govern the water and solute transports, which could further influence the membrane fouling behavior. Membranes with superior separation properties and structural properties (i.e., greater water permeability and selectivity, and smaller structural parameter) could deliver greater water flux. Nevertheless, this may in turn increase the potential of membrane fouling due to the enhanced hydrodynamic drag force (refer to Section 3.2.2.1). On the other hand, membranes with inferior separation properties (i.e., smaller water permeability and selectivity) could result in more solute diffusing from DS into FS (and from FS into DS). This may also raise the risk of fouling (refer to Sections 3.2.2.2 and 3.2.2.3). In addition, the membrane structural properties may also significantly influence the internal fouling especially in the PRO applications. Nevertheless, there is little systematic research that discussed the effect of membrane separation and structural properties on the membrane fouling [29, 30, 163]. When designing or selecting membranes for FO/PRO applications in the future, more attention could be paid to the evaluation of membrane separation and structural properties on fouling not only the consideration of flux performance in non-fouling conditions.

While the membrane separation properties can indirectly influence membrane fouling through the influence of hydrodynamic conditions and solution chemistry during operation, the membrane surface properties (such as roughness, surface charge, hydrophilicity/hydrophobicity, and specific functional groups) can directly influence the foulant-membrane interaction and play a paramount role in early stage of membrane fouling. The influence of these properties on foulant-membrane interaction is mainly due to (1) non-homogeneity and membrane morphology, (2) their effect on XDLVO interactions (VDW, EDL, and acid-base interactions, refer to Section 3.2.1), and (3) specific interactions between the ions in FS (or DS) and the functional groups of the membrane [62]. Generally, membranes with smoother and more hydrophilic surfaces may be less prone to fouling. Gu et al. compared the organic fouling of cellulose triacetate (CTA) membrane and polyamide (PA) thin-film composite (TFC) membrane in ODMPs and observed that the smoother CTA membrane exhibited less fouling tendency than the PA membrane at early stage of fouling, though the PA membrane had a smaller contact angle than the CTA membrane [78]. In their study, it seems that the membrane surface roughness may have stronger influence on fouling compared to the surface hydrophilicity. Elimelech and co-workers also observed the important role of membrane surface roughness in membrane fouling in pressure-driven RO/NF processes [75, 164, 165]. The more severe fouling of PA membranes in both RO and FO is primarily attributed to the greater roughness of these membranes. The surface of PA membranes exhibited large-scale ridge-and-valley rough structure that may provide a larger surface area for particles-membrane interaction [75, 109, 165-167]. Moreover, the shear rate over the valley of the PA rough structures is presumably lower than that over the smooth surface [164, 165]. This could lead to preferential deposition of particles in the “valley” regions of the rough membranes at the initial stage of filtration, resulting in greater flux decline than the smooth membranes [109]. Despite the much smoother surface, the cellulose derived membranes have relatively a narrower range of pH tolerance, lower water permeability and salt rejection, and are more biodegradable compared to the TFC PA membranes [75, 156, 157]. As a result, TFC osmotic membranes based on PA materials are becoming increasingly popular in water and energy applications [50, 61, 155-157, 168-171]. Tiraferri et al. observed that the hydrophilicity for the PA-based membranes seems to play a significant role in reducing fouling in ODMPs [172]. The importance of hydrophilicity on improving the fouling resistance was extensively observed for pressure-driven membranes [151, 173-175]. Based on the XDLVO theory, these authors attributed the greater anti-fouling tendency of the more hydrophilic membranes to the larger repulsive acid-base interaction (or repulsive hydrophilic interaction) that prevents the attachment of colloidal particles on the membrane surface. Mi and Elimelech further observed that the rich carboxyl groups on the surface of PA membranes can increase the gypsum scaling

[28]. They found that the carboxylic groups on the PA membrane surface can specifically bind with Ca^{2+} ions and form complexes resulting in the increased Ca^{2+} concentration at the membrane surface, which promotes gypsum nucleation and the subsequent growth of both amorphous and crystalline gypsum on the membrane surface. Similarly, when studying the alginate fouling in seawater RO desalination, Jin et al. observed that the divalent ions can bridge the carboxylic groups on the PA membrane surface and that in alginate molecules, which further enhance the alginate-membrane adhesion force [118]. They suggested that the idea membrane should not only be smooth and hydrophilic but also lack of carboxylic groups (to prevent the specific interaction with calcium) to reduce the risk of alginate fouling. Surface charge can influence the electrostatic interaction between the membrane and colloids in feed solution. In general, repulsive electrostatic interaction between the charged membrane surface and the colloids in the feed solution minimizes the attachment of colloids on the membrane, thus reducing fouling. Highly negatively charged membranes often exhibit strong anti-fouling tendency [176], considering that most of the colloidal particles such as NOMs are negatively charged in the natural water ([175] and also Table 2). Nevertheless, the existing of potential charging ions (such as Ca^{2+} by binding to some negatively charged functional groups) and/or any positively charged colloids may lead to severe fouling of negatively charged membranes. For this reason, commercial antifouling RO and NF membranes are typically designed to have a neutral hydrophilic surface through surface modification [177, 178]. In addition, zwitterionic surface has been reported to be effective in fouling reduction [179]. In summary, a membrane with strong anti-fouling potential may have neutral, smooth and hydrophilic surface without carboxylic functional groups. To enhance membrane anti-fouling properties, significant amount of efforts have been spent on the modification of membrane surface to optimize the surface properties

particularly in the field of pressure-driven membranes but could be useful in ODMPs [175]. These modification approaches include surface coating [180, 181], grafting [182, 183], blending/embedding antimicrobial nanoparticles [184, 185], which is reviewed in Section 5.1.4.

It is worthwhile to point out that the membrane surface properties may only play a role in the initial fouling stage. Once the deposited foulants covers the membrane surface, the membrane surface properties are masked by the foulant properties [88]. The subsequent fouling is governed by foulant-deposited-foulant interaction instead of the foulant-membrane interaction, and thus the surface properties may have little influence on the long term fouling behavior (particularly under severe fouling conditions) [110]. For example, Liu and Mi observed that the behavior of initial gypsum scaling on the organic macromolecular conditioned FO membrane surface was strongly dependent on the chemical properties of the organic macromolecules deposited on the membrane rather than the surface properties of the virgin membrane [70]. Gu et al. [78] reported membrane-dependent FO fouling behavior under mild fouling conditions while membrane-independent fouling upon the occurrence of severe fouling (i.e., complete cake layer formation). Tang et al. observed that the flux decline during long-term fouling tended to approach a pseudo-stable value (i.e., limiting flux) that was independent of membrane surface properties during RO/NF membrane fouling by humic acid [110, 126]. Similar limiting flux behavior may also exist in ODMPs [31].

3.2.2.5. Effect of membrane orientation

Membrane orientation is another unique factor affecting the membrane fouling behavior in ODMPs. Different groups of researchers consistently observed that membrane was less prone to fouling in AL-FS orientation compared to the AL-DS orientation [29, 65, 66, 68, 76, 107]. They attributed this observation to the following reasons: (1) smoother active layer surface that can reduce the possibility of foulant deposition in AL-FS orientation, (2) vanished crossflow within the support layer that can accelerate the internal foulant deposition and/or internal concentration polarization in AL-DS orientation, (3) the retention of foulants by the dense rejection layer and their entrapment in the porous support, and (4) severe pore clogging of the support layer that enhances the ICP in AL-DS orientation. These explanations indicate that the influence of membrane orientation on fouling is indeed through the combined effects of surface properties, hydrodynamic conditions and concentration polarization. Previous researchers also observed that the external fouling in AL-FS orientation was more reversible by surface flushing [32, 72, 75, 186], while the internal fouling in AL-DS orientation exhibited less cleaning efficiency [35].

Despite the advantages of reduced fouling tendency and greater fouling reversibility in AL-FS orientation, the osmotic membrane suffers much greater ICP effect and thus delivers lower initial water flux than that in AL-DS orientation under otherwise identical conditions [29, 47, 64]. In addition, when operated in PRO process, membranes may experience reduced mechanical stability in AL-FS orientation [42]. This suggests that the advantages of AL-FS orientation in terms of fouling and cleaning are diminished when the feed solution has low-fouling potential and membrane mechanical stability is a high requirement. It should also be noted that, at increased feed solution concentration, the difference of ICP effect for both orientations becomes marginally small and the initial water fluxes in both orientations converge [42, 54, 56, 76, 187, 188]. Therefore, the selection of membrane orientation for FO/PRO applications should take into comprehensive account the influence of membrane fouling, ICP and membrane stability rather than any single effect. Some recent works have further attempted to address these issues by using membranes with a double-skin design, where a primary rejection skin is used for salt retention and a second skin for internal fouling prevention [56, 189, 190].

3.2.3. ICP self-compensation effect

It has been well recognized that the water flux in ODMPs can be significantly affected by ICP and membrane fouling. Recently, Tang and co-workers systematically investigated the coupled effects of ICP and humic acid fouling on the flux behavior of FO membrane [29]. They observed that the flux in the AL-FS orientation exhibited much more stable trend compared to that in AL-DS orientation. The superior flux stability in AL-FS orientation was also observed by other researchers [65, 66, 107]. The phenomenon is not solely due to the less fouling propensity in AL-FS orientation (Section 3.2.2.5). Tang et al further attributed it to the stronger **ICP self-compensation effect** – any attempt to reduce membrane flux was compensated by a reduction in ICP [29].

To further explain the ICP self-compensation effect, the water flux of fouled membrane was simulated and illustrated in Figure 7a [29]. During membrane fouling, flux decreases as a result of increased hydraulic resistance of the fouled membrane (assuming that the membrane rejection is not affected). Since the degree of ICP is an exponential function of water flux (refer to Section 2.2, Section 2.3 and Appendix A), a decrease in water flux also decreases the ICP, which in turn leads to increased effective driving force (i.e., concentration difference across the membrane active layer) under constant apparent driving force (also noting that $F_{dcp,f} < F_{dcp}$ in Eq. (11) and Eq. (8)). This compensates for the reduced membrane water permeability. The ICP self-compensation effect is much stronger in the AL-FS orientation, arising from the generally more severe ICP in this orientation in the first place. Therefore, only slight membrane flux decline may occur in the AL-FS orientation. While ICP self-compensation is also applicable in the AL-DS orientation, the effect is much weaker. Compounded by other fouling mechanisms (internal clogging and fouling-enhanced CP, see Section 3.2.4), the flux in AL-DS is far less stable compared to AL-FS.

Figure 7

Apart from membrane fouling, any other attempts (e.g., the dilution of DS or the concentration of FS) at reducing the water flux can trigger the ICP self-compensation effect to moderate the flux decline. Tang et al. observed that the water flux in baseline test showed more excellent stability in AL-FS orientation, although the DS was continuously diluted by the water permeating from FS [29]. As illustrated in Figure 7b, at the same flux level, the AL-FS curve is flatter since ICP is more severe in this orientation. Increasing DS concentration in this orientation is compensated by the enhanced ICP. Consequently, the net effect on flux increase is only marginal. In a similar fashion, dilution of DS in AL-FS tends to be compensated by a less severe ICP. In contrast, the ICP self-compensation effect is much weaker in the AL-DS orientation as a result of its milder ICP. Likewise, the self-compensation of ICP also takes significant effect in AL-FS orientation at the increase of FS concentration. Groups of researchers [54, 56, 76, 188] observed that the water flux decline with increasing FS concentration in AL-FS orientation was much milder than that in AL-DS orientation. The water fluxes in both orientations eventually converged toward an identical value at high FS concentrations although the water flux in AL-FS was much lower at low FS concentrations (Figure 7c). The relatively greater flux stability in AL-FS against the concentration of FS can be well explained by the ICP self-compensation effect – the attempt of increasing FS concentration to reduce the water flux is compensated by the reduced dilutive ICP.

3.2.4. Fouling-enhanced concentration polarization

While the membrane fouling can compensate the ICP and moderate the rate of flux decline through the ICP self-compensation effect, it can also enhance the concentration polarization and accelerate the rate of flux decline due to the fouling-enhanced concentration polarization (CP). Unlike the ICP self-compensation effect that is solely available in ODMPs, the fouling-enhanced CP has been observed in both pressure-driven RO/NF processes [62, 191-193] and osmotically-driven FO/PRO processes [25, 29, 32, 40, 95, 194, 195].

Membrane fouling is associated with the deposition of foulant and subsequent cake layer formation. As illustrated in Figure 8, the foulant cake layer indeed acts as a secondary filtration layer that can impose additional resistance to water transport. Moreover, this “unstirred” cake layer can hinder the back diffusion of solute within it, which leads to an enhanced solute concentration near the membrane surface – a phenomenon termed cake-enhanced concentration polarization (CE-CP) [191]. The CE-CP together with the cake layer hydraulic resistance contribute to the overall flux decline for the salt-rejecting membranes during fouling. In some cases, CE-CP effect can dominate the overall water flux decline over the effect of cake resistance. It was observed that the cake layer resistance for colloidal silica particles with relatively large size did not contribute significantly to the overall FO flux decline, whereas the flux decline for this large particles was due to the predominant CE-CP effect [32, 95]. This observation was later verified by the modeling of colloidal fouling [195] and is consistent with the observations in RO fouling studies [192, 193]. In addition, the CE-CP may reduce the membrane rejection to the solute in FS and thus reduce the permeate quality [25, 196-198].

Figure 8

Although the nature of CE-CP is similar in both pressure-driven RO/NF and osmotically-driven FO/PRO processes (i.e., the CP is enhanced by fouling, which causes additional flux decline), the factors that contribute to the enhanced CP are different. In RO/NF, the CP is contributed by the feed water convection. In comparison, in ODMPs, the concentrative CP in the feed side is not only contributed by feed water convection but also by the reverse solute diffusion (RSD) (refer to the osmotic-resistance filtration model in Section 3.1.2). Hong and co-workers observed more flux decline in FO fouling than that in RO fouling due to the significant contribution of RSD to the CE-CP in FO process [32]. On the other hand, they found that the contribution of RSD to CE-CP will not be significant if the membrane has very low solute permeability, in which case the CE-CP is mainly contributed by the feed water convection in a fashion similar to RO [195]. It is also interesting to note that while decreasing membrane rejection reduces more CE-CP in RO (as less feed solutes are retained in the cake layer), an opposite trend could be observed in ODMPs as a result of severe RSD for membranes with reduced rejection.

In the ODMPs, the membrane fouling and CP in feed solution side behaves differently in the two opposite orientations (refer to Section 2.2, Section 3.1.1, Section 3.1.2, and Section 3.2.2.5).

Thus, the membrane fouling-enhanced CP can be specifically expressed in different forms – cake-enhanced external concentration polarization (CE-ECP) in AL-FS orientation (Figure 8a and refs. [32, 95]) and pore clogging-enhanced internal concentration polarization (PCE-ICP) in AL-DS orientation (Figure 8b and refs. [29, 40]). CE-ECP is virtually through the enhancement of the solute mass transfer resistance within the external cake layer on the membrane active layer surface (i.e., the $1/k_{cecp,f} > 1/k_{cecp}$ that results in the $F_{cecp,f} > F_{cecp}$ in Eq. (11) and Eq. (8) in Section 2.3 and section 3.1.2). In contrast, for PCE-ICP, the solute mass transfer resistance within the membrane support layer is enhanced by the internal pore clogging (i.e., the $1/k_{cicp,f} > 1/k_{cicp}$ that results in the $F_{ccp,f} > F_{ccp}$ in Eq. (12) and Eq. (9) in Section 2.3 and section 3.1.2). Noting that ICP dominates the water flux in ODMPs, the PCE-ICP presumably plays a more significant role in the flux decline. It should also be noted that the much stronger ICP self-compensation effect in AL-FS orientation may moderate the rate of flux decline even though the CE-ECP is significant during membrane fouling (refer back to Figure 7c and Section 3.2.3). On the other side, PCE-ICP may cause much more severe flux decline. Nevertheless, systematic studies are still needed to explore the CE-ECP and PCE-ICP effects on ODMPs fouling.

It is interesting to note that in some cases membrane fouling may lead to the reduced concentration polarization and thus the increased water flux. Our results in Figure B1 (refer to Appendix B) show the water flux behavior of a TFC membrane in baseline test and in humic acid fouling test. Interestingly, the flux decline in baseline test was faster than that in humic acid fouling test. This phenomenon was not by accident but could be attributed to the cake-reduced concentration polarization (CR-CP). In mild fouling conditions, the deposition of humic acid on the membrane surface at relatively low initial water flux only slightly increase the hydraulic resistance to water permeation. However, it could significantly enhance the rejection (e.g., by sealing defects in the membrane rejection layer, leading to a decrease of the *B* value) [138, 199], which further reduces the RSD (i.e., $J_{s,f} / J_{w,f} < J_s / J_w$) and consequently RSD-related CP. Similarly, Xie et al. also observed that the humic acid fouling reduced the NaCl permeability but did not significantly reduce the water permeability [26]. Noting again that the CP in FO is contributed by both feed water convection and RSD (refer to Section 2.2, Section 2.3 and Section 3.1.2), the CP on the membrane active surface is thus reduced due to the reduced RSD.

3.2.5. Reverse solute diffusion-enhanced membrane fouling

Reverse solute diffusion (RSD) not only contributes to the fouling-enhanced CP (Section 3.2.4), but also leads to the enhanced membrane fouling [30, 31, 39, 40]. As illustrated in Figure 6 and Section 3.1.3, the solute (e.g., ions and/or molecules) in the DS diffusing into FS can lead to a significant change of feed solution chemistry and composition (such as ionic strength, pH, and/or specific ion concentration), which can potentially change the membrane fouling behavior (Section 3.2.2.2). The phenomenon of RSD-enhanced fouling was first reported by Tang and co-workers [39]. They observed that the reverse diffusion of Mg^{2+} from $MgCl_2$ draw solution into FS led to enhanced microalgae fouling on the FO membrane when studying the microalgae separation by FO membranes [39].

RSD-enhanced fouling is a new membrane fouling mechanism uniquely associated with ODMPs. She et al. revealed that the mechanism of RSD-enhanced fouling is related to the (1) type of draw solute diffusing into FS, (2) ability of the diffused draw solute to interact with the foulant in FS, and (3) rate of the draw solute reverse diffusion [30, 40]. The type of draw solute that can reversely diffuse into FS is associated with the draw solution composition and can influence the fouling potential (refer to Section 3.2.2.3). If the draw solute that has diffused in FS can interact and destabilize the foulant in FS, it can potentially enhance the fouling. She et al observed that the reverse diffusion of Ca^{2+}/Mg^{2+} from seawater-based DS into FS led to enhanced alginate fouling and humic acid fouling on the FO/PRO membranes [30, 40]. Moreover, the reverse diffusion of Ca^{2+} caused more severe alginate fouling than that of Mg^{2+} because the Ca^{2+} has much stronger complexation with the carboxylic group in organic foulant molecules. Although the Na^+ has negligible complex effect with macromolecular organics, it is reported that the reverse diffusion of Na^+ can led to the increase of local ionic strength near membrane surface and enhance the humic acid deposition [26]. Similar RSD-enhanced fouling phenomenon has been observed during the study of inorganic scaling and biofouling [31, 65, 80, 200, 201]. In addition to potential role of increased feed osmotic pressure, increased ionic strength, and fouling promoter, solute diffused from DS to FS may also serve as fouling/scaling precursors [31] or stubstrates/nutreints for biological growth [6, 14, 153].

While the type of draw solute reverse diffusion can potentially initiate the enhanced fouling, the rate of RSD can determine the amount of draw solute accumulation in FS and thereby the extent of eventual fouling. She et al observed that greater rate of RSD led to more accumulation of draw solute in the bulk FS and near the membrane surface, which caused more severe fouling if the specific draw solute can have strong interaction with the foulants in FS side [30, 31, 40]. The rate of draw solute reverse diffusion is dependent on a series of factors, such as DS concentration [29, 40-44, 54], type of counter ions [40, 43, 152], membrane separation properties [29, 41, 58, 156], and membrane operating conditions (e.g., the exposure to the high salt concentration and high applied pressure) [30, 42, 55, 58]. Generally, greater rate of specific draw solute reverse diffusion could be observed with higher DS concentration, greater reverse diffusion rate of its counter ion (e.g., the reverse diffusion of Ca^{2+} is faster for $Ca(NO_3)_2$ DS than that for $CaCl_2$ DS), greater membrane solute permeability, and more severe deformation at higher applied pressure in PRO. It is interesting to note that, in PRO operation, an attempt to improve the osmotic power output and mitigate fouling by increasing the applied pressure (and thus decreasing water flux) may be completely offset by the unexpectedly enhanced membrane fouling due to increased RSD, particularly for DS containing fouling promoters [30, 31]. For instance, She et al observed that both the alginate fouling and gypsum scaling on PRO membranes were significantly enhanced at increased applied pressures due to the increased reverse diffusion of fouling/scaling precursor (i.e., Ca^{2+}) as a result of membrane deformation, even though lower initial water fluxes were used at increased applied pressures [30, 31]. On the other hand, it was reported that the increase of applied pressure in feed solution in the PAO operation led to the decrease of reverse solute flux [202, 203]. This suggests that the fouling may be reduced in PAO under certain conditions compared to FO, which requires further studies for verification.

In some cases, the RSD-enhanced fouling occurs virtually through the RSD-enhanced CP. For example, when studying the inorganic gypsum scaling, Zhang et al observed that the reverse diffusion of scaling precursors from DS initially only enhanced the concentrative ICP and thus increased the gypsum saturation index within the membrane support layer in PRO process [31]. The onset of gypsum precipitation occurred when the calcium sulphate became supersaturated in the support layer. It is worthwhile to note that the RSD may mitigate fouling rather than enhance fouling in case the draw solute can act as anti-fouling agent when it reversely diffuses into FS

[204]. Chen et al. observed that the gypsum scaling was inhibited and even eliminated at increased reverse Mg^{2+} diffusion due to the competitive formations of $MgSO_4°$ and $CaSO_4·2H_2O$ [201]. Systematic studies on RSD-reduced fouling are guaranteed in the future.

3.3. Inorganic scaling in ODMPs

Inorganic scaling could be a major issue for membrane technologies applied in wastewater reclamation and brackish water desalination [11, 66, 69, 80]. The occurrence of scaling in high-pressure RO system has been systematically reviewed elsewhere [96]. Most of the findings and conclusions are applicable for explaining the scaling behavior in ODMPs. In ODMPs, inorganic scaling occurs with the deposition and growth of the sparingly soluble inorganic scale on the osmotic membranes. Many of the factors and mechanism governing the inorganic scaling in ODMPs are similar to those for colloidal and organic fouling that have been discussed in detail in Section 3.2.

Particularly, the occurrence and severity of scaling is strongly dependent on the concentration and solubility of potential scalants (or their precursors) in the feed solution. From the thermodynamic perspective, a mineral may start to crystallize when its concentration is greater than the saturation limit (i.e., upon supersaturation of the solution) [96]. In addition, the kinetics of precipitation determines the severity of scaling and can be sped up with a high concentration of nucleation sites. Unlike the colloidal and organic fouling that can occur with a low foulant concentration, the scaling only occurs if the supersaturation exceeds a critical value. Interestingly, Zhang et al. observed that although the calcium sulphate is undersaturated in the bulk feed water, severe internal gypsum scaling still occurred within the membrane support layer in PRO process

[31]. They attributed this phenomenon to the severe concentrative internal concentration polarization (CICP) of scaling precursor ions (i.e., the Ca^{2+} and SO_4^{2-}) within the support layer, which leads to the substantial increase of the ions' concentration to reach supersaturation at the interface of support layer-active layer (e.g., the saturation index increased from 0.8 in the bulk feed solution to 4.0 in the support layer due to CICP). Similar CICP effect may also be applicable to other types of fouling in ODMPs, although more studies are still needed to confirm this phenomenon.

In addition, the reverse diffusion of scaling precursor ions from DS into FS can further enhance the scaling. Zhang et al. observed that the gypsum scaling was enhanced when the $CaCl_2$ or Na_2SO_4 was used as DS [31]. When scaling precursor ions (i.e., Ca^{2+} or SO_4^{2-}) diffused into FS, the gypsum saturation index was substantially increased in the feed side especially within the support layer where CICP plays an important role. This RSD-enhanced scaling mechanism is illustrated in Figure 6 in section 3.1.3. On the other hand, the forward diffusion of scaling precursors into DS may also induce scaling providing that a high concentration of counter-ion as scaling precursor is available in the DS. For example, Zhang et al. observed the gypsum scaling formed on the membrane surface in the DS side even when small amount of SO_4^{2-} in FS diffused through the membrane into the high concentration $CaCl_2$ draw solution [31]. This mechanism of forward-diffusion-induced scaling is also illustrated in Figure 6 in Section 3.1.3.

It is worthwhile to note that most of the existing studies on inorganic scaling in ODMPs focused on the use of gypsum as model scale that is independent of solution pH and has relatively high solubility [28, 31, 35, 69, 70, 205, 206]. Only few studies reported other types of scaling [80, 186]. In reality, typical inorganic scaling can be caused by alkaline scale (e.g., calcium carbonate ($CaCO_3$) and calcium phosphate ($Ca_3(PO_4)_2$)) and silica-based scale. Different from the non-alkaline gypsum scale, crystallization of these alkaline and silica-based scales are also dependent on solution pH not only their concentration. In ODMPs, the H^+ and OH^- can diffuse through the membrane bidirectionally between DS and FS, which can potentially alter the local solution pH near the membrane surface and thus the scaling tendency of those alkaline and silica-based scales on the membrane [204]. Future study may pay more attention to the alkaline scaling and silica scaling and explore relative scaling mechanisms uniquely available in ODMPs.

3.4. Biofouling in ODMPs

Compared to organic and inorganic fouling where the foulants are non-living substances, biofouling is much more complicated. When the live microorganisms attach to the membrane surface, they start to propagate utilizing the nutrient from the surrounding environment to form the aggregated "biofilm" and are capable of colonizing almost all the surface [103, 106]. During their metabolism, the microorganisms often excrete large amount of extracellular polymeric substances (EPS) that can help to enhance the adhesion of the microorganisms on the membrane surface and protect them from the biocides and toxins in the solution [103, 106].

In ODMPs, biofouling can be identified in brackish/seawater desalination, bio-product separation, and in osmotic membrane bioreactor (OMBR) for wastewater reclamation [13, 38, 39, 65, 66, 102, 207, 208]. While biofouling has been extensively studied in pressure-driven membrane processes (refer to the review papers [103, 104, 106, 119]), only several papers reported its studies in ODMPs [38, 39, 65, 66, 207, 208]. In addition to the general fouling factors and mechanisms that are applicable for colloidal and organic fouling in ODMPs (Section 3.2), the properties of microbial communities and the nutrient concentration are critical factors for biofouling. To some extent, the biofouling potential can be represented by the microorganisms and the nutrient concentration [104, 106]. Even small amount of microorganisms in the initial feed solution can induce severe biofouling due to biofilm development if sufficient nutrient is provided for their growth. This is different from organic and inorganic fouling which can be well controlled by simply reducing the foulant concentration in the feed solution.

Flemming divided the biofilm development on membrane surface into three phase: induction, logarithmical growth, and plateau phases [209]. The first phase is mainly dependent on the concentration of microorganisms, while the second and third phases are strongly affected by the nutrient concentration in the bulk solution [106, 209]. Among all the factors for biofilm growth, the nutrient concentration is regarded as one of the most important ones [209]. To control the biofouling, it is essential to decrease the available nutrients supply. In the ODMPs-based systems, the nutrients for biofilm growth can be obtained from three major sources: (1) the contaminants in the original bulk feed water, (2) the leached material from the substratum that could be the the membrane material itself, and (3) the solutes with nutrient elements in the draw solution (that can reversely diffuse into feed water). The nutrient concentration in the feed solution has strong effect on biofilm development [106]. The adhered microorganisms on the membrane surface can also scavenge their food from the membrane if the membrane materials are nutritious for them. For example, Chen et al. observed that the CTA membrane was biologically degraded by the bacteria in an anaerobic OMBR system, as the CTA material can serve as the nutrient for their growth on the membrane surface [12]. This further leads to the loss of membrane separation performance and the increase of salt accumulation in the feed tank which are diffused from DS [12]. High concentration salt ions accumulated in feed tank can potentially influence the bioactivity of microorganisms and change the microbial communities that exist in feed bioreactor and/or attach on membrane surface [200, 210, 211]. The nutrients can also be obtained from DS due to their reverse diffusion. Bowden et al. investigated the organic ionic salts for specific use in OMBR [212]. These organic ionic salts can act as the carbon source and be biodegraded for the denitrification process, which could be beneficial to the reduction of salts accumulation in the reactor. Nevertheless, they did not investigate the adverse impact of the reverse diffusion of nutrient on the biofouling. It can be expected that the reverse diffusion of nutrients can enhance the biofouling since biofilm growth on the membrane surface can benefit from the nutrients that diffuse from DS (Figure 6). This can be a critical issue for biofouling development and should be a very important research topic for the future, because there are tremendous nutritious draw solutes available in the application of ODMPs, such as the fertilizer draw solutes [14] and the ammonia-carbon dioxide draw solutes for desalination [6].

The excreted EPS from the microorganisms' metabolism is another essential factor affecting the biofouling. EPS are important constituting materials for biofilm [86, 119], and it consists of polysaccharides, proteins, lipid, nucleic acids, etc. [86, 103, 119]. These materials are sticky and can enhance the adhesive interaction between the microorganisms and the membrane surface [106]. The influence of EPS on membrane fouling was extensively investigated in MBR systems [86, 119, 121]. Their fouling behavior in ODMPs is similar to that of macromolecular organics and can be well interpreted by the fouling mechanisms discussed in Section 3.2. For example, Zou et al. reported that the reverse diffusion of Mg^{2+} can enhance the microalgae fouling, which is virtually due to the enhanced interaction of the EPS in the feed water with the Mg^{2+} diffused from DS [39, 65].

It is worthwhile to note that different trend between biofouling and colloidal organic fouling was also observed during the study of the spacer effect. Valladares Linares et al. observed that the biofouling with a thicker (46 mil) feed spacer resulted in less flux decline than that with a thinner (28 mil) spacer in FO process [208]. This is different from the flux behavior in particulate/colloidal and organic fouling where thicker spacer with lower cross-flow velocity increases the membrane fouling and reduces the permeate flux. They further revealed that the spacer can impact the localization of biofilm on membrane and spacer filament – the membrane and spacer filament were covered a thick biofilm for the 28 mil spacer, but some membrane areas and spacer filaments were not covered by the biofilm for the 46 mil spacer.

3.5. Comparison of membrane fouling in FO, PRO and PAO processes

The general principles and fouling mechanisms detailed in Section 2 and Sections 3.1-3.4 are applicable to FO, PRO and PAO processes. However, due to the differences among FO, PRO, and PAO processes with respect to the applied hydraulic pressure, membrane orientation and reverses solute diffusion, membrane fouling in these processes could also behave differently in some cases.

Firstly, in PRO, the water flux is retarded by the applied hydraulic pressure in DS, while in PAO, the water flux is improved by the applied pressure in FS. The water fluxes for these three processes follow the trend of PAO > FO > PRO assuming other testing conditions are identical.

Therefore, when the membrane is operated in the same orientation, the membrane fouling tendency may follow the same trend as that of their water flux if the flux-dependent fouling mechanism plays a dominant role (Section 3.2.2.1). Consistently, Kim et al. [123] observed that more flux decline due to colloidal-organic fouling occurred at higher applied pressure in PAO when NaCl was used as DS. Moreover, they observed that the colloidal-organic fouling was less reversible in PAO than that in FO even though the same initial water flux was applied in both fouling experiments. They further attributed the stronger fouling tendency and less fouling reversibility in PAO to the more compacted foulant layer formed under the applied hydraulic pressure in FS side. In this regard, membrane fouling in PAO is similar to that in RO, where the applied hydraulic pressure compresses the deformable colloidal-organic foulant layer that may induce more hydraulic resistance to water permeation and is less removable through physical cleaning [75]. On the other hand, fouling is regarded to be more reversible in FO, since there is a lack of hydraulic compaction on the foulant layer.

In practical operation, PRO is typically operated in AL-DS orientation to maintain the membrane mechanical stability under the applied hydraulic pressure, while FO and PAO prefer to be operated in AL-FS orientation when exposure to a high-fouling potential feed water. Therefore, in reality, PRO usually suffers more severe fouling and flux decline than FO due to the membrane orientation effect (Section 3.1.1, Section 3.2.2.5 and ref. [27, 29]), the less ICP self-compensation effect (Section 3.2.3), and the more severe fouling-enhanced ICP (Section 3.2.4). Moreover, the internal fouling in PRO is much less removable using conventional cleaning methods compared to the external fouling in FO and PAO. This suggests that the AL-DS orientation may be a major shortcoming for practical operation of PRO under high-fouling conditions. While She et al. attempted to operate the PRO in AL-FS orientation to reduce the fouling tendency at a low applied pressure [30], more efforts are needed to further improve the membrane mechanical stability at higher applied pressures in this orientation.

Thirdly, the reverse solute diffusion (RSD) is typically more severe in PRO process than that in FO process even if both processes are operated in the same membrane orientation (refer to Section 2.1 and ref. [42, 55]). Moreover, RSD will be further accelerated due to membrane deformation in PRO [42, 55]. As a result, the RSD-enhanced fouling may be more pronounced for PRO when the DS contains fouling promoters, fouling precursors and/or nutrients. This is consistent with the observation by She. et al. that membrane fouling was more severe in PRO process than that in FO process when using the seawater-based draw solutions [30]. Interestingly, in PAO, RSD decreases with increasing the applied hydraulic pressure (Figure 2 in Section 2.1). Similar trend was experimentally observed by Oh et al. [203]. This suggests that PAO may experience less severe fouling compared to PRO and FO when the DS contains large quantities of fouling promoters/precursors (e.g., fertilizer draw solution [80]). On the other hand, excessive applied pressure in PAO could also result in membrane deformation, loss of rejection, and thus RSD-enhanced fouling under certain conditions. Up to now, systematic studies are still lacking in this regard.

Overall, membrane fouling in PRO process can be a much more serious problem than that in FO and PAO due to the severe internal fouling, fouling-enhanced CP and severe reverse solute diffusion in PRO and shall be treated as a priority task in ODMPs research.

4. Effect of membrane fouling on contaminants removal

Membrane fouling not only influences the water permeation but also affects the solute transport through the membrane. It has been extensively reported that in both ODMPs and pressure-driven RO/NF processes the contaminants removal by the fouled membrane may behave differently from that by the clean membrane [25, 26, 196, 213-218]. The mechanisms for the rejection of trace contaminants in ODMPs are very similar to that in pressure-driven membrane processes. In general, the trace contaminants removal by a clean membrane can be dominated by the individual or combined mechanisms of electrostatic repulsion (or Donnan exclusion), steric hindrance (or size exclusion), hydrophilic/hydrophobic interaction between compounds and membrane, and dipole interaction, which can significantly affect the contaminants' solubility and diffusivity in the membrane [219, 220]. During membrane fouling, the foulant layer formed on the membrane surface can modify the membrane surface properties and separation properties and may also result in the enhanced concentration polarization near the membrane surface (refer to Section 3.2.4). All of these may lead to the change of transport behavior of contaminants through the membrane. A recent review on the rejection of trace organic compounds by the FO membranes is available [219]. In the current section, we will briefly review the effect of membrane fouling on the trace contaminants (both organic and inorganic) removal from the mechanistic point of view in ODMPs.

According to the literature, the membrane fouling can either enhance or reduce the removal of contaminants due to different rejection mechanisms. Jin et al. observed that the alginate fouling can enhance the arsenite rejection as a result of the enhanced effect of size exclusion because the alginate deposited on the membrane can block the pores or membrane defects for arsenite transport [25]. Similarly, Xie et al. observed this fouling enhanced size exclusion effect led to the enhanced rejection of trace organic compounds (TOrCs) [26, 196]. They suggested that the enhanced steric hindrance was caused by the hydrated humic acid fouling layer that hindered the transport of solutes but not the permeation of water. Valladares Linares et al. also observed the increased rejection of micropollutants by the fouled membrane, but the rejection mechanisms were associated with the change of the physicochemical properties of the membrane surface due to the deposition of a more hydrophilic and negatively charged foulant layer [221]. In their study, the increased rejection of the hydrophilic ionic compounds was due to the increased electrostatic repulsion, and the increased rejection of hydrophobic neutral compounds was due to their increased adsorption onto the membrane as a result of the increased hydrophobic interaction. It is reported the rejection of hydrophobic compounds may decrease at longer-term rejection when the adsorbed TOrCs reached saturation and then break through the rejection layer [222, 223]. Cartinella et al. observed that the presence of surfactant in the simulant wastewater could result in significant increase of natural stories hormone removal [224]. They proposed two possible mechanisms to explain this phenomenon: (1) the adsorption of anionic surfactant on the membrane surface, which hinders the sorption and transport of hormones through the membrane, and (2) the sequestration of hormones due to their interaction with and uptake by the surfactant micelles in the bulk solution.

On the other hand, many researchers observed the decreased trace contaminants rejections by the fouled membrane [25, 196, 221]. The primary reason is attributed to the fouling-enhance concentration polarization that leads to the increased solute concentration near the membrane surface and thus enhanced the solute diffusion through the membrane. Xie et al. observed that high initial water flux led to the form of a compact and cohesive foulant layer, where the enhanced steric hindrance became less important but the effect of cake-enhanced concentration polarization became predominant. Thus, they observed a decrease in TOrCs rejection [196]. Jin et al. observed that the severe alginate fouling in AL-DS orientation decreased the boron rejection to nearly zero, suggesting that the pore-clogging enhanced ICP in this orientation may play a more significant role on boron rejection [25]. Valladares Linares et al. observed that the rejection of hydrophilic neutral compounds was decreased by the fouled membrane [221]. They attributed this to two reasons: (1) the increase in surface charge due to membrane fouling might promote membrane swelling and thus increase its molecular weight cut-off, and (2) enhanced micropollutants partitioning into the membrane as a result of the changed surface hydrophilicity.

Above-mentioned contaminants rejection mechanisms by fouled membranes in ODMPs have also been extensively investigated in pressure-driven RO/NF processes [214-218]. However, due to the unique properties of ODMPs, some other factors can also influence the trace contaminants rejection, such as membrane orientation [11, 25, 225, 226] and reverse solute diffusion [11, 222, 227]. In addition, in a hybrid treatment system or a pilot system where feed water contains different groups of contaminants, the overall removal efficiency of the contaminants could be influenced by the combined rejection mechanisms rather than a single one [213, 228-230]. The future work may further investigate the rejection efficiency and mechanisms in pilot and full-scale systems instead of a bench-scale system.

5. Mitigation of fouling in ODMPs

Membrane fouling is inevitable in ODMPs, But understanding of the roots and causes of membrane fouling is useful to developing more effective strategies to mitigate fouling. This section will summarize the various approaches for fouling mitigation for ODMPs.

5.1. Fouling reduction

5.1.1. Pretreatment of feed water

The purpose of feedwater pretreatment is to minimize membrane fouling by removing foulants/fouling precursors and/or adjusting the feedwater chemistry. To date, there is no publication reporting the investigation of feedwater pretreatment for ODMPs. On the other hand, the feedwater pretreatment for RO/NF membrane system has been extensively studied. In fact, many approaches for RO/NF feedwater pretreatment could be applicable to pretreat FO/PRO feed water due to the similar properties of the feed waters used in both membrane systems. The typical pretreatment approaches include: coarse/fine screening, coagulation-flocculation and sedimentation, media filtration, UF/MF membrane filtration, and/or their combinations [231-234]. Screening is a preliminary step that removes only relatively large and suspended visible solids from the influent, such as trash and debris [235]. Coagulation-flocculation and sedimentation are effective physicochemical treatment method and have been widely used to remove particulate and colloidal matters (such as silica, iron, NOM, etc.). The typical coagulants include alum and ferric salts. Systematic studies on the effect of type and dose of coagulants on the pretreatment efficiency were reported in the literature [232, 234, 236-239]. Media filtration is also used to remove particulate matters [232, 240, 241]. The nominal size range of conventional media filtration is usually considered to be 1 µm [241]. The materials reported for media filtration include activated carbon, expanded clay, anthracite coal, glass fiber, etc. [232, 240, 241]. For the well intake of seawater for RO desalination, sand filtration is extensively used as a pretreatment method to remove large-size particulate matters [232, 242]. MF/UF are becoming increasingly popular to pretreat the feed water and can remove a variety of foulants, such as biomass flocs, individual bacterial cells, colloidal particles, and macromolecular organics [234, 241, 243]. The removal efficiency is dependent on the membrane pore size or molecular weight cut off (MWCO). Typically, the nominal size for commercially available MF membranes range from 0.1-1.0 μm and UF membranes from 1-300 kDa [241]. Other feed water pretreating methods are also reported to control the colloidal fouling and inorganic scaling, including ion exchange [239] and acidification [231].

While above methods focused on the removal of foulants from feed water, some other methods aim to optimize the feedwater chemistry or limit the nutrients loading in feed water. Solution chemistry (e.g., pH, ionic strength, and specific ion composition) can significantly affect the foulant-foulant and foulant-membrane interactions, inorganic scale precipitation, and biological activity of microorganisms. The method of acidity (pH) adjustment is extensively used to control the inorganic scaling by alkaline scales (e.g., calcium carbonate and calcium phosphate) [96, 231, 244]. Hardness removal (e.g., reduce Ca^{2+} and Mg^{2+} concentration) is known to reduce the risk of organic fouling, inorganic scaling and biofouling [27, 39, 74, 119]. Typical methods for hardness removal include lime/soda softening [245], ion exchange softening [96, 246] and electrocoagulation/precipitation [247]. Nutrients are essential for the microorganism growth and biofilm development. It is reported that limitation of nutrients loading in feed water are effective strategies for biofouling control [248, 249]. Adding antiscalant in the feed water is one of the commonly methods for scaling control [96, 231]. However, some limitations of using antiscalant are reported in the literature. For example, some antiscalants in feed water showed risks of increasing biofouling potential [250] as well as enhancement of organic fouling [251].

Although feedwater pretreatment is likely to significantly reduce membrane fouling, it is important to realize that pretreatment could be an energy-, chemical-, and cost-intensive process. It may also introduce additional chemical byproducts that may further cause adverse environmental impact in the effluent of membrane concentrate. The tradeoff between outcome of membrane fouling and cost of pretreatment should be evaluated before selecting the most appropriate pretreatment methods for practical applications.

5.1.2. Proper selection of draw solution

As discussed in Section 3.2.2.3, draw solution composition is an important factor affecting the membrane fouling in ODMPs. Selection of a suitable DS for ODMPs can be a good strategy for fouling control. In principle, DS selection should consider the (1) DS type and (2) DS concentration [40]. Firstly, the draw solutes in the DS should not act as fouling promoters, fouling precursors or nutrients. In addition, they should have minimal permeability through the membrane. Thirdly, the selected DS concentration may be slightly lower than the level of “critical DS concentration”. In practice, the DS for ODMPs can be obtained from the nature (e.g., seawater) and industrial brines, or by using synthetic draw solutes. For synthetic DS, it could be easy to control its composition and concentration following the above DS selection principles. However, for the natural or industrial DS, it is usually impractical to change its composition. For DS containing fouling promoters/precursors, it may increase the fouling potential (e.g., seawater or seawater desalination brine) [30]. To select this type of DS for ODMPs, careful assessment and/or pretreatment are needed to prevent severe RSD-induced fouling.

5.1.3. Optimization of operating conditions

The typical operating parameters for ODMPs include membrane orientation, cross-flow velocity, and operating pressure in addition to flux. Membrane orientation is the first consideration before starting the operation of ODMPs. As discussed in Section 3.1.1 and Section 3.2.2.5, AL-FS orientation is preferred due to lower fouling potential. However, ICP is more severe in this orientation and the membrane mechanical stability in this orientation should be carefully evaluated for PRO process. Cross-flow velocity is another important parameter in the operation of ODMPs. In general, greater cross-flow velocity induces greater shear force on membrane surface that would lead to lower foulant deposition and concentration polarization (refer to section 3.2.2.1) and thus is preferred in the operation. In addition to the increased cross-flow velocity, the introduction of air bubbling in feed solution is also able to reduce the membrane fouling due to the air scouring on membrane surface [34, 66]. This is particularly important for the control of fouling in MBR system [119, 252]. However, there is a need to further investigate the effect of aeration intensity and air-bubble size on the fouling in ODMPs. It was also reported that the employment of spacer could alleviate fouling [36, 68]. The spacer inserted in the feed flow channel can increase the effective cross-flow velocity, turbulence and mass transfer near the membrane surface. The spacer geometry (e.g., thickness and opening ratio) can influence the FO/PRO performance [55] and potentially influence the fouling behavior. Valladares Linares et al. reported that using the thicker spacer resulted in less flux reduction during biofouling in FO, which is similar to the behavior of biofouling in RO [208]. Spacer can also significantly change the deposition pattern of particulate and organic foulants [68, 128].

Hydraulic pressure is applied in the PRO and PAO processes. The choice of operating pressure is also very important for the control of membrane fouling, since the hydraulic pressure can not only influence the water flux but also influence the reverse solute flux [42, 53, 203] both of which can potentially influence the membrane fouling. In PRO, She et al. found that that the increase of applied pressure can reduce the organic fouling for NaCl draw solution due to the reduced water flux and permeation drag force, but it can enhance fouling for $CaCl_2$ draw solution due to increased reverse diffusion of Ca^{2+} [30, 31]. These two competing effects associated with the operating pressure are significantly dependent on the draw solution composition and the membrane mechanical stability. While PAO is attracting increasing interest recently [52, 53, 203], further work is recommended to study the role of applied pressure on membrane fouling using different draw solutions.

5.1.4. Modification of membrane properties and structure

As discussed in Section 3.2.2.4, membrane fouling is significantly affected by the membrane properties. Therefore, an effective strategy to mitigate fouling and/or improve fouling reversibility is to modify the membrane properties and structures to render the membranes more fouling-resistant. To date, there have been 13 papers studying the membrane modification for application in ODMPs. As summarized in Table 3, the FO/PRO membrane modification aims to: (1) increase surface hydrophilicity [172, 181-183, 253, 254], (2) reduce surface roughness [253], (3) reduce surface charge density (i.e., make a neutral surface) [172], (4) reduce carboxyl groups in membrane surface [172, 182, 183, 253], (5) introduce specially structured polymer chains [182, 254], (6) incorporate nanoparticles [184, 185, 255], and (7) form a second NF/UF layer on the support layer side [56, 189, 190, 256]. While most of these studies focused on the modification of membrane active layer that could improve the membrane fouling resistance in FO processes, only three papers modified the support layer that may enable the membrane be operated in PRO process with reduced fouling propensity [181, 254, 255]. It is worthy to note that the surface modification can be effective to mitigate external fouling that mainly occurs in FO process, but may has little effect for the internal fouling control. More work could be done in the future to modify the membrane porous support layer to improve the membrane internal fouling resistance in the PRO process. It should also be noted that the surface modification sometimes reduce the membrane water permeability because an additional layer is formed on the membrane surface. As thus, future work may also need to evaluate the trade-off between the loss of water permeability and the improvement of anti-fouling properties of the modified membranes.

Table 3

5.2. Fouling removal and membrane cleaning

5.2.1. Physical cleaning

Physical methods have been extensively used to mitigate membrane fouling in ODMPs [33-38, 75, 257]. The physical cleaning methods mainly include membrane surface flushing and membrane backwashing [35, 75]. The membrane surface flushing relies on an enhanced shear force (e.g., cross flow) along membrane surface to remove the deposited foulants, while the membrane backwashing is via reversing the water permeation direction and using the permeation drag force to detach and remove the deposited foulants on the membrane [119]. Both methods have been demonstrated to be effective against the membrane fouling under a variety of conditions.

Mi and Elimelech systematically investigated the organic fouling reversibility of FO membranes through evaluating the membrane cleaning efficiency by surface flushing [75]. They observed that the fouling in FO was more reversible than that in RO due to the lack of hydraulic compaction of the foulant layer in FO. According to these authors, simple surface flushing could be an effective cleaning method to restore the water flux of the fouled FO membrane without chemical cleaning agent. They further observed increased FO cleaning efficiency (up to 98%) at increased cross-flow velocity and extended cleaning duration. Moreover, by introducing the air bubble, the flux of the cleaned membrane could be recovered remarkably close to 100% at a much shorter cleaning duration due to the enhanced shear force. The air scouring has also been proved to be an effective physical approach to reduce fouling in hybrid membrane systems (e.g., MBR) [119, 252]. In later studies, the surface flushing method (either by increasing cross-flow velocity or by introducing air scouring) was extensively used to clean the fouled FO membranes and study the FO membrane fouling reversibility by groups of researchers [28, 34-38, 70-72, 76, 80, 122, 123, 186, 257-259]. Despite the high cleaning efficiency of surface flushing in most studies, this cleaning method will typically consume certain amount of clean water during flushing, which would in turn reduce the actual water recovery. Moreover, the surface flush could become ineffective against internal fouling especially in PRO operation, where the sheer force may diminish within the porous support layer [35]. Instead, it was suggested that the membrane backwashing could be an effective approach to remove the foulants from the unstirred porous support layer [33, 35].

Osmotic backwashing is a typical membrane backwashing approach used for cleaning the fouled FO/PRO membranes. It is originally developed from the RO desalination industry to mitigate RO membrane fouling [260-267]. With the development of FO/PRO technology, it has been extensively used for FO/PRO membrane cleaning [10, 23, 33, 35, 37, 127, 194, 268, 269]. During the osmotic backwashing, the original feed water is replaced with a higher-salinity water and the draw solution is replaced with a lower-salinity water (or fresh water), which creates a negative water flux (i.e., the water permeation direction during backwashing is opposite to that in FO/PRO process) that is able to break up the foulant layer and move it away from the membrane. Some researchers simply swap the feed and draw streams to reverse the water permeation direction and initiate the backwashing momentarily [33, 127], whereby the clean water consumption is avoided for membrane cleaning. It has been reported that the osmotic backwashing could be effective to restore a considerable portion of water productivity against the membranes fouled by complex feed waters (e.g., the anaerobic digester centrate [10], MBR feed water [194, 268], industrial wastewater [127], and municipal wastewater [269]). Yet, some other studies reported the low efficiency of osmotic backwashing for water flux recovery [23, 35, 37]. They attributed to the accumulation of salt within foulant layer during cleaning that reduced the effective osmotic driving force. Arkhangelsky et al. further adopted the hydraulic backwashing by applying additional hydraulic pressure to clean the membrane after internal gypsum scaling within the support layer and achieved 100% water flux restoration. Indeed, the efficiency of backwashing depends on its frequency, duration and intensity [119]. Nevertheless, these parameters have not been systematically investigated to design the backwashing in ODMPs. In addition, the design of membrane backwashing for FO/PRO membrane cleaning could be more flexible than that for RO membrane cleaning and should not be limited to osmotic backwashing. The hydraulic backwashing may be a good choice when osmotic backwashing cannot work well although the former will consume additional pressure energy and clean water. Future studies on FO/PRO fouling control might consider both osmotic and hydraulic backwashing. Last but not least, the combination of surface flushing and membrane backwashing may offer better cleaning efficiency since both shear force and permeation drag force will be applied to remove the foulant layer from the membrane.

5.2.2. Chemical cleaning

Although physical cleaning exhibits great promise for mitigation of membrane fouling in ODMPs, it cannot perform well if the foulant has strong interaction with the membrane. For instance, Mi and Elimelech reported that the surface flushing was less effective for polyamide TFC membrane cleaning compared to the CTA membrane cleaning due to the stronger adhesion between alginate and TFC membrane [75]. Yoon et al. also found that the physical cleaning was not effective to recover the water flux when studying the biofouling of FO membranes [38]. Instead, they found that chemical cleaning with chlorine could effectively mitigate the biofouling where membrane should be highly chlorine resistant. Chemical cleaning depends on the chemical reactions to weaken the adhesion force between foulants and membrane. To date, only several papers have reported the chemical cleaning for membrane fouling control in ODMPs [10, 37, 38, 72, 270]. According to these papers, the chemical cleaning is able to achieve a high cleaning efficiency against the organic fouling and biofouling that could have strong interaction with the membranes. The agents used for chemical cleaning include sodium hydroxide (NaOH), sodium ethylenediaminetetraacetic acid (EDTA) and sodium hypochlorite (NaOCl) and their typical concentration ranged from 0.5% to 1%. Despite the high cleaning efficiency, chemical cleaning could not remove the foulant from membrane pores [10] and may compromise the FO membrane integrity and shorten the membrane life [37]. Moreover, the waste stream produced during chemical cleaning may cause additional environmental problems [75, 263].

5.2.3. Biological cleaning

Although physical cleaning and chemical cleaning are effective against membrane fouling in most cases, negative concerns on these cleaning methods are also reported [252]. For instance, physical cleaning could be energy-intensive and may cause membrane damage. Chemical cleaning may consume large quantities of chemicals, impact membrane integrity, influence the microbial community in MBR system, and generate waste chemical reagents. To overcome these adverse impacts, biological cleaning has been developed to remove the foulant and recover the water flux. Biological cleaning involves the use of bioactive agents (e.g., enzymatic cleaning or biological predation) for foulant removal [271]. A recent review on MBR cleaning [252] reported three biological cleaning strategies: enzymatic cleaning, energy uncoupling and quorum quenching. Briefly, enzymatic cleaning is the most widely used biological method for membrane fouling control [252]. In enzymatic cleaning, enzymatic agents can specifically interact with the biopolymeric foulant (e.g., proteins and lipids) and break up the foulant layer on the membrane surface [271, 272]. Energy uncoupling is a method using the biochemical uncouplers to inhibit adenosine triphosphate (ATP) synthesis [273]. The uncoupling of energy metabolism can affect biofilm stability, which can eventually lead to the biofilm detachment [273]. Quorum quenching is a method through quorum sensing (QS) control (e.g., the blocking of intercellular communication) to prevent biofilm formation on membrane surface [274].

6. Conclusions and recommendations

This paper presents a comprehensive review of membrane fouling in ODMPs. Firstly, an osmotic-resistance filtration model, which explicitly differentiates all the driving forces for ODMPs, is proposed to guide the overall discussion of fouling in ODMPs. Membrane fouling by colloidal particles, organic macromolecules, sparingly soluble inorganic compounds and microorganisms are reviewed respectively with a focus on the discussion of the factors and mechanisms governing their fouling behavior. The similarities and differences of membrane fouling between the FO process and PRO process are specifically outlined, and the impact and mechanisms of the membrane fouling on trace contaminants removal are briefly discussed. The membrane fouling-control strategies are also reviewed and discussed. The major conclusions are summarized below.

- As illustrated in Figure 9, membrane fouling, concentration polarization (both ICP and ECP) and reverse solute diffusion (RSD) are interrelated closely to each other in ODMPs and can be modeled using the osmotic-resistance filtration model.
- Many of the fouling factors and mechanism for ODMPs are also applicable for pressure-driven RO/NF processes, such as the effects of hydrodynamic conditions, feedwater composition, and membrane properties, and the cake-enhanced concentration polarization (CE-CP) mechanism.
- The driving force for ODMPs is osmotic pressure that arises from the use of high-concentration draw solution, which is a major difference between the ODMPs and pressure-driven membrane processes. As a result, most of the fouling factors and mechanisms

uniquely applicable for ODMPs are associated with the use of draw solution, such as the effect of draw solution composition, and the RSD-enhanced fouling and CP mechanisms.

- Membrane orientation is another important factor affecting membrane fouling in ODMPs and also a consideration for distinguishing the FO operation and PRO operation. AL-FS orientation is preferred for FO operation because it has less fouling propensity and more flux stability due to ICP self-compensation effect. On the other hand, AL-DS orientation is preferred for PRO operation due to the stronger membrane mechanical stability at high applied pressures and less severe ICP. However, this orientation is more prone to internal fouling that is less reversible.
- The impact of membrane fouling can either increase or reduce the contaminants rejection mainly due to the size exclusion mechanism or the CE-CP effect.
- To control the membrane fouling, the membrane modification may be a promising strategy to reduce the foulant-membrane interaction and thus improve the fouling reversibility that eases the membrane cleaning.

Despite of the great number of studies on the fouling in ODMPs, more research is still needed to further understand the fouling mechanisms. In particular, systematic fouling characterization for complex feed water and fouling studies at large scales are recommended. In viewing of the importance of RSD, its role in ODMPs deserves further attention (e.g., on the reverse diffusion of nutrients on the biofouling). Further studies are also required to understand the similarities and differences of ODMP fouling versus RO fouling (e.g., to better understand the role of pressure and flux), which may provide additional insights for developing tailored fouling control measures. Fouling control strategies need to be further investigated and their integration with ODMPs operations required further verification for pilot and full-scale applications.

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Appendix A. Derivation of ICP, ECP and osmotic-resistance filtration models

ECP and ICP behave differently in different membrane orientations. Figure A1 presents the schematic illustration of solute concentration profile across the membrane due to ECP and ICP in both membrane orientations. In the active-layer-facing-feed-solution (AL-FS) orientation (Figure A1a), the solute near the active layer surface is concentrated and its concentration on the active layer surface is greater than that in the bulk FS. This is referred to as concentrative ECP (CECP) [47]. However, solute in the DS side is diluted within the porous support layer and near the surface of support layer by the permeated water and its concentration in these two regions is lower than that in the bulk DS. CP in these two regions is referred to as dilutive ICP (DICP) and dilutive ECP (DECP) respectively. In contrast, the concentration polarization in the active-layer-facing-draw-solution (AL-DS) orientation (Figure A1b) is exhibited as dilutive ECP (DECP) near the active layer surface in the DS side, and concentrative ICP (CICP) within the porous support layer and concentrative ECP (CECP) near the surface of support layer in the FS side [47].

Figure A1

The actual solute concentration at the support-active layer interface (C_i) and on the active layer surface (C_{al}) due to ICP and ECP respectively can be modeled by taking into account the polarized boundary layer in three regions: (I) the external boundary layer at the active layer side with a thickness of δ_{al}, (II) the overall support layer with a structural parameter of S, and (III) the external boundary layer at the support layer side with a thickness of δ_{sl} [55, 137]. At steady state, the solute flux in both orientations due to convection and diffusion satisfies the following equation based on the one-dimensional coordinate system (x) with positive direction pointing from draw solution to the feed solution [55, 137]:

$$J_s = -J_w C(x) - D_{eff} \frac{dC(x)}{dx} \tag{A1}$$

The boundary layer conditions for Eq. (A1) are illustrated in Figure A1. C_i and C_{al} are obtained by solving Eq. (A1) at different boundary layer regions, and are expressed below in both membrane orientations.

In the AL-FS orientation:

$$C_{al} = C_{fs} \exp\left(\frac{J_w}{k_{cecp}}\right) + \frac{J_s}{J_w}\left[\exp\left(\frac{J_w}{k_{cecp}}\right) - 1\right] \text{ (concentrative ECP)} \tag{A2}$$

and

$$C_i = C_{ds} \exp\left(-\frac{J_w}{k_{dcp}}\right) - \frac{J_s}{J_w}\left[1 - \exp\left(-\frac{J_w}{k_{dcp}}\right)\right] \text{ (dilutive ICP and ECP)} \tag{A3}$$

where C_{fs} and C_{ds} are the bulk feed and draw solution concentration respectively; k_{cecp} is the mass transfer coefficient near the active layer surface due to CECP and it can be determined by Eq. (A4).

$$k_{cecp} = \frac{D}{\delta_{al}} \tag{A4}$$

k_{dcp} is the overall mass transfer coefficient that correlates the mass transfer coefficient within the support layer (i.e., k_{icp} or k_{dicp}) and that near the support layer surface (i.e., $k_{ecp,sl}$ or k_{decp}) due to dilutive CP (i.e., both DICP and DECP), and it can be determined by Eq. (A5).

$$\frac{1}{k_{dcp}} = \frac{1}{k_{dicp}} + \frac{1}{k_{decp}} = \frac{S + \delta_{sl}}{D} \tag{A5}$$

The structural parameter of membrane support layer, S, is related to the length (l_{sl}), tortuosity (τ) and porosity (ε) of the support layer and can be determined by the Eq. (A6).

$$S = \frac{\tau \times l_{sl}}{\varepsilon} \tag{A6}$$

Similarly, in the AL-DS orientation:

$$C_i = C_{fs} \exp\left(\frac{J_w}{k_{ccp}}\right) + \frac{J_s}{J_w}\left[\exp\left(\frac{J_w}{k_{ccp}}\right) - 1\right] \text{ (concentrative ICP and ECP)} \tag{A7}$$

and

$$C_{al} = C_{ds} \exp\left(-\frac{J_w}{k_{decp}}\right) - \frac{J_s}{J_w}\left[1 - \exp\left(-\frac{J_w}{k_{decp}}\right)\right] \text{ (dilutive ECP)} \tag{A8}$$

where k_{decp} is the mass transfer coefficient near the active layer surface due to DECP and it can be determined by Eq. (A9).

$$k_{cecp} = \frac{D}{\delta_{al}} \tag{A9}$$

k_{ccp} is the overall mass transfer coefficient that correlates the mass transfer coefficient within the support layer (i.e., k_{icp} or k_{cicp}) and that near the support layer surface (i.e., $k_{ecp,sl}$ or k_{cecp}) due to concentrative CP (i.e., both CICP and CECP), and it can be determined by Eq. (A10).

$$\frac{1}{k_{ccp}} = \frac{1}{k_{cicp}} + \frac{1}{k_{cecp}} = \frac{S + \delta_{sl}}{D} \tag{A10}$$

Based on Eqs. (A2) and (A3) or Eqs. (A7) and (A8), the effective concentration difference across the membrane (ΔC_{eff}) can be determined by the difference between C_i and C_{al}, whereby the conventional ICP and ECP models for ODMPs can be derived. Eq. (A5) and Eq. (A10) suggests that the influence of ECP near the support layer surface on the performance of ODMPs can be neglected if the boundary layer thickness is much smaller than the structural layer parameter of the support layer (i.e., $\delta_{sl} << S$).

In ODMPs, the effective osmotic (or concentration) driving force is lower than the osmotic pressure (or concentration) difference between the bulk DS and bulk FS due to the ICP and ECP. The osmotic-resistance filtration model explicitly differentiates all the driving forces for ODMPs. As illustrated in Figure 3a, in the AL-FS orientation, the loss of driving force due to concentrative CP (i.e., ΔC_{cecp}) and dilutive CP (i.e., ΔC_{dicp} and ΔC_{decp}) can be determined by Eq. (A11) and Eq. (A12) respectively.

$$\Delta C_{cecp} = C_{al} - C_{fs} = F_{cecp}\left(C_{fs} + \frac{J_s}{J_w} \right) \tag{A11}$$

and

$$\Delta C_{dcp} = \Delta C_{dicp} + \Delta C_{decp} = C_{ds} - C_i = F_{dcp}\left(C_{ds} + \frac{J_s}{J_w} \right) \tag{A12}$$

where F_{cecp} and F_{dcp} are concentration polarization factors for CECP and DCP respectively and are determined by Eq. (A13) and Eq. (A14).

$$F_{cecp} = \exp\left(\frac{J_w}{k_{cecp}} \right) - 1 \tag{A13}$$

and

$$F_{dcp} = 1 - \exp\left(- \frac{J_w}{k_{dcp}} \right) \tag{A14}$$

Similarly, the loss of osmotic driving force due to concentrative CP (i.e., ΔC_{cicp} and ΔC_{cecp}) and dilutive CP (i.e., ΔC_{decp}) in AL-DS orientation (refer to Figure 3b) can be determined by Eq. (A15) and Eq. (A16) respectively.

$$\Delta C_{ccp} = \Delta C_{cicp} + \Delta C_{cecp} = C_i - C_{fs} = F_{ccp}\left(C_{fs} + \frac{J_s}{J_w}\right) \tag{A15}$$

and

$$\Delta C_{decp} = C_{ds} - C_{al} = F_{decp}\left(C_{ds} + \frac{J_s}{J_w}\right) \tag{A16}$$

where F_{ccp} and F_{decp} are concentration polarization factors for CCP and DECP respectively and are determined by Eq. (A17) and Eq. (A18).

$$F_{ccp} = \exp\left(\frac{J_w}{k_{ccp}}\right) - 1 \tag{A17}$$

and

$$F_{decp} = 1 - \exp\left(-\frac{J_w}{k_{decp}}\right) \tag{A18}$$

It is interesting to note that the equations related to concentrative CP are in similar form (e.g., Eq. (A2) vs. Eq. (A7), Eq. (A11) vs. Eq. (A15), and Eq. (A13) vs. Eq. (A17)) and those related to dilutive CP are also in similar form (e.g., Eq. (A3) vs. Eq. (A8), Eq. (A12) vs. Eq. (A16), and Eq. (A14) vs. Eq. (A18)) regardless of membrane orientations.

In osmotic-resistance filtration model, ΔC_{eff} is determined by Eq. (A19).

$$\Delta C_{eff} = \Delta C_{bulk} - \Delta C_{loss,ccp} - \Delta C_{loss,dcp} = \left(C_{ds} - C_{fs}\right) - \Delta C_{cecp} - \Delta C_{icp} - \Delta C_{decp} \tag{A19}$$

Based on Eq. (A19), the water flux equation can be derived as shown in Section 2.3.

Appendix B. Fouling-reduced concentration polarization (CR-CP).

Figure B1a presents the water fluxes in baseline test and humic acid fouling test in FO process. For the baseline testing, the water flux experienced a mild decrease with the progressing of testing due to the gradual dilution of the DS and the loss of osmotic driving force. In contrast, the water flux in the fouling testing maintained almost unchanged. This is due to the fouling-reduced concentration polarization (CR-CP). In the mild fouling condition (i.e., only 10 mg/L humic acid present in the DI water and a low initial water flux (~14 L/m^{2}hr) was used in the testing), the deposition of humic acid presumably did not cause additional resistance to water permeation. But the hydrated humic acid layer on the membrane surface can block the pores for NaCl diffusion from the DS and reduce the solute flux. As shown in Figure B1b, the specific solute flux (J_s / J_w) in fouling testing (i.e., 7.6 mM) is more than three times lower than that in the baseline testing (i.e., 24.6 mM). Noting that the CP in FO is contributed by feed water convection and reverse solute diffusion (refer to the osmotic-resistance filtration model in section 3.1.2), this indicates that the CP effect for fouling testing is significantly lower than that in baseline testing since in the current case the CP is only contributed by the reverse solute diffusion when DI water is used as feed water (i.e., $C_{fs} = 0$). It is also worthwhile to note that the TFC polyamide membrane surface has ridge-and-valley structure. The local CP in the valley region is less susceptible to cross flow. Therefore, compared to the baseline testing, the reduced CP in the fouling testing results in the greater water flux.

Figure B1

Nomenclature

CP	concentration polarization
ICP	internal concentration polarization
ECP	external concentration polarization
CCP	concentrative concentration polarization
CICP	concentrative internal concentration polarization
CECP	concentrative external concentration polarization
DCP	dilutive concentration polarization
DICP	dilutive internal concentration polarization
DECP	dilutive external concentration polarization
CE-CP	cake-enhanced concentration polarization
CE-ECP	cake-enhanced ECP in AL-FS orientation
PCE-ICP	pore clogging-enhanced ICP in AL-DS orientation
FS	feed solution
DS	draw solution
RSD	reverse solute diffusion
EPS	extracellular polymeric substances
SMP	soluble microbial products

A	water permeability coefficient (L/m^2-bar)
A_f	water permeability coefficient of the fouled membrane (L/m^2-bar)
B	solute permeability coefficient (L/m^2)
C_{ds}	draw solution concentration (M)

C_{fs}	feed solution concentration (M)
C_i	concentration at the interface between active layer and support layer (M)
C_{al}	concentration on the active layer surface (M)
ΔC_{eff}	effective concentration difference across the active layer (M)
$\Delta C_{loss,icp}$ or ΔC_{icp}	loss of concentration driving force due to ICP (M)
$\Delta C_{loss,ecp}$ or ΔC_{ecp}	loss of concentration driving force due to ECP (M)
ΔC_{ccp}	loss of concentration driving force due to CCP (M)
ΔC_{cecp}	loss of concentration driving force due to CECP (M)
ΔC_{cicp}	loss of concentration driving force due to CICP (M)
ΔC_{dcp}	loss of concentration driving force due to DCP (M)
ΔC_{decp}	loss of concentration driving force due to DECP (M)
ΔC_{dicp}	loss of concentration driving force due to DICP (M)

D	diffusion coefficient (m^2/s)
J_w	water flux (L/m^2-h)
$J_{w,f}$	water flux in fouling testing (L/m^2-h)
J_s	solute flux ($mole/m^2$-h)
$J_{s,f}$	solute flux in fouling testing ($mole/m^2$-h)
J_s/ J_w	specific reverse solute flux (M)
$J_{s,f}/ J_{w,f}$	specific reverse solute flux in fouling test (M)
F_{ccp}	concentration polarization factor for CCP (-)
F_{dcp}	concentration polarization factor for DCP (-)
F_{cicp}	concentration polarization factor for CICP (-)

F_{cecp}	concentration polarization factor for CECP (-)
F_{dicp}	concentration polarization factor for DICP (-)
F_{decp}	concentration polarization factor for DECP (-)
$F_{ccp,f}$	concentration polarization factor for CCP for the fouled membrane (-)
$F_{dcp,f}$	concentration polarization factor for DCP for the fouled membrane (-)
$F_{cicp,f}$	concentration polarization factor for CICP for the fouled membrane (-)
$F_{cecp,f}$	concentration polarization factor for CECP for the fouled membrane (-)
$F_{dicp,f}$	concentration polarization factor for DICP for the fouled membrane (-)
$F_{decp,f}$	concentration polarization factor for DECP for the fouled membrane (-)
k	mass transfer coefficient (m/s)
k_{icp}	mass transfer coefficient within the support layer due to ICP (m/s)
k_{ecp}	mass transfer coefficient near the membrane surface due to ECP (m/s)
$K_{overall}$	overall mass transfer coefficient (m/s)
π_{ds}	osmotic pressure of draw solution (bar)
π_{fs}	osmotic pressure of feed solution (bar)
$\Delta\pi_{bulk}$	osmotic pressure difference between the bulk DS and bulk FS (bar)
$\Delta\pi_{eff}$	effective osmotic pressure difference across the active layer (bar)
$\Delta\pi_{loss,icp}$	loss of osmotic driving force due to ICP (bar)
$\Delta\pi_{loss,ecp}$	loss of osmotic driving force due to ECP (bar)
T	absolute temperature (K)
R_g	universal gas constant (0.083145 L bar mole^{-1} K^{-1})

R_m	hydraulic resistance of the membrane (m^{-1})
R_f	hydraulic resistance of the foulant layer on membrane (m^{-1})
β	Van't Hoff coefficient
ε	porosity (-)
l_{sl}	length of support layer (m)
τ	tortuosity of support layer (-)
δ_{sl}	ECP boundary layer thickness near the support layer surface (m)
δ_{al}	ECP boundary layer thickness near the active layer surface (m)
μ	fluid viscosity (Pa s)

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List of Tables

Table1. Mathematical equations and physical interpretations on $\boldsymbol{C_{al}}$ and $\boldsymbol{C_i}$ due to influence of ICP and ECP in ODMPs.

Table 2. Properties and identification methods of selected foulants.

Table 3. Summary of membrane modification to improve anti-fouling properties in ODMPs.

List of Figures

Figure 1. Annual number of publications on ODMPs and membrane fouling in ODMPs since 2005. Data were obtained from Scopus on7th Oct 2015. Keywords for searching are “Forward Osmosis or Pressure Retarded Osmosis” and “Forward Osmosis or Pressure Retarded Osmosis and Fouling” respectively. In searched results, the document type does not include Short Survey, Erratum, Letter, Conference Review, and Conference Paper.

Figure 2. Classification and characteristics of osmosis-related membrane separation processes. (a) Direction of water flows in RO, PRO, FO and PAO processes. In RO, water flows from high salinity draw solution (DS) to low salinity feed solution (FS) driven by the applied hydraulic pressure; in PRO and FO, water flows from low salinity FS to high salinity DS driven by the osmotic pressure; in PAO, water flows from low salinity FS to high salinity DS driven by both osmotic pressure and applied hydraulic pressure. (b) Water flux (J_w) magnitude, (b) specific solute flux (J_s/J_w) magnitude, and (c) power density (W) magnitude as a function of applied pressure in RO, PRO, FO and PAO processes.

Figure 3. Schematic illustration of osmotic-resistance filtration model in ODMPs in (a) active-layer-facing-feed-solution (AL-FS) orientation and (b) active-layer-facing-draw-solution (AL-DS) orientation. All the driving forces represented by the concentration profile across the membrane due to ICP and ECP are specifically differentiated in accordance with the osmotic-resistance filtration model. The mathematic expressions of all the concentration polarization factors (F_{cecp}, F_{dcp}, F_{ccp} and F_{decp}) are described in Appendix A. It is assumed that (1) the osmotic pressure is linearly proportional to the concentration and (2) the feed solution and draw solution contain the same solute.

Figure 4. External fouling and internal fouling in ODMPs. (a) Only external fouling occurs in AL-FS orientation, (b) both external fouling and internal fouling occur in AL-DS orientation.

Figure 5. Schematic illustration of the influence of membrane fouling on water flux behavior in ODMPs according to the osmotic-resistance filtration model.

Figure 6. Schematic illustration of role of reverse solute diffusion (RSD) on enhanced membrane fouling in ODMPs.

Figure 7. ICP self-compensation effect for the AL-FS orientation in different scenarios. (a) In the scenario of membrane fouling where water permeability is reduced. In the simulation, DS concentration is 5 M for AL-FS orientation and 1 M for AL-DS orientation respectively. (b) In the scenario of reduced DS concentration. (c) In the scenario of increased FS concentration. Other parameters for simulation: A value 2.14 × 10^{-12} m/s.Pa, B value 1.76 × 10^{-7} m/s, S value 456 µm, $\Delta P = 0$ bar, $C_{ds} = 1$ M, and $C_{fs} = 10$ mM.

Figure 8. Schematic illustration of concentration profile across the membrane due to fouling-enhanced concentration polarization. The fouling-enhanced CP leads to the $\Delta C_{eff,f} < \Delta C_{eff}$. (a) Fouling-enhanced ECP in AL-FS orientation. Note that ICP self-compensation effect also plays a significant role in this orientation, which will moderate the flux decline. (b) Fouling-enhanced ICP in AL-DS orientation. Note that fouling-enhanced ECP in this orientation may behave similar to that in AL-FS orientation and thus is not illustrated. The solid line indicates the concentration profile before fouling, while the dotted line indicates the concentration profile after fouling.

Figure 9. The intrinsic interrelationship among membrane fouling, concentration polarization (CP) and reverse solute diffusion (RSD) in ODMPs.

Figure A1. Concentration profile across the membrane due to ICP and ECP. Assuming the draw solution and feed solution contain the same solute.

Figure B1. Enhanced water flux in humic acid fouling test due to fouling-reduced concentration polarization (CR-CP). (a) water flux in baseline test and humic acid fouling test in FO process, and (b) the average J_s/J_w in baseline test and fouling test. Fouling experimental conditions: feed solution only contained 10 mg/L humic acid in DI water, draw solution 0.5 M NaCl, cross-flow velocity 23.2 cm/s, AL-FS membrane orientation, and hand-casted TFC-PA FO membrane (the properties are reported in ref. [156]). In baseline test, DI water was used as feed solution.

Tables and Figures

Table1. Mathematical equations and physical interpretations on C_{al} and C_i due to influence of ICP and ECP in ODMPs.

Membrane Orientation	Concentrative Concentration Polarization (CCP) at the FS side	Dilutive Concentration Polarization (DCP) at the DS side
AL-FS	$C_{al} = \underbrace{\underbrace{C_f \exp\left(\frac{J_w}{k_{cecp}}\right)}_{\text{Concentration of FS due to feed water convection}} + \underbrace{\frac{J_s}{J_w}\left[\exp\left(\frac{J_w}{k_{cecp}}\right)-1\right]}_{\text{Accumulation of solute due to RSD}}}_{\text{CECP near the active layer surface}}$	$C_i = \underbrace{\underbrace{C_d \exp\left(-\frac{J_w}{k_{dcp}}\right)}_{\text{Dilution of DS due to DS convection}} - \underbrace{\frac{J_s}{J_w}\left[1-\exp\left(-\frac{J_w}{k_{dcp}}\right)\right]}_{\text{Loss of draw solute due to RSD}}}_{\text{DCP at the support layer side (i.e., DECP near the support layer surface and DICP within the support layer)}}$
AL-DS	$C_i = \underbrace{\underbrace{C_f \exp\left(\frac{J_w}{k_{ccp}}\right)}_{\text{Concentration of FS due to feed water convection}} + \underbrace{\frac{J_s}{J_w}\left[\exp\left(\frac{J_w}{k_{ccp}}\right)-1\right]}_{\text{Accumulation of solute due to RSD}}}_{\text{CCP at the support layer side (i.e., CECP near the support layer surface and CICP within the support layer)}}$	$C_{al} = \underbrace{\underbrace{C_d \exp\left(-\frac{J_w}{k_{decp}}\right)}_{\text{Dilution of DS due to DS convection}} - \underbrace{\frac{J_s}{J_w}\left[1-\exp\left(-\frac{J_w}{k_{decp}}\right)\right]}_{\text{Loss of draw solute due to RSD}}}_{\text{DECP near the active layer surface}}$

Note: C_{al} is the solute concentration at the active layer surface; C_i is the solute concentration at the active-support layer interface.

Table 2. Properties and identification methods of selected foulants

Types of Fouling	Foulants	Examples	Size and shape	charge	Characterization method
Colloidal and Organic fouling	Polysaccharides [275]	Alginate	200–2000 kDa, extended random coil.	Negatively charged, ~3 meq/g (up to 6 meq/g)	Colorimetric method (phenol-sulfuric acid method at UV of 485 nm) [66, 78].
		Xanthan and gellan	100–2500 kDa, linear	Negatively charged	
		Schizophyllan	400–500 kDa, rigid rod-like	Neutral	
	Humic substance [62]	Humic acid (IHSS)	Mw of a few kDa to a few hundred kDa. Globular molecule (linear under high pH, low ionic strength, and low concentration	Negative charged (pH_{pzc}=3), typical total acidity: 5–10 meq/g	(1) TOC and UV254 [88]. (2) LC-OCD and 3D FEEM [72] (or 3D FTIR [66]).
	Proteins [139]	Bovine serum albumin (BSA)	67 kDa	pH_{IEP} = 4.7 (total acidity 1.5 meq/g including both carboxylic and amine groups)	Using protein assay kit to analyze at UV562 [78, 276].
		Bovine immunoglobulin G	155 kDa	pH_{IEP} = 6.6	
		Bovine hemoglobin	68 kDa	pH_{IEP} = 7.1	
		Bovine pancreas ribonuclease A	13.7 kDa	pH_{IEP} = 7.8	
		Lysozyme	14.4 kDa	pH_{IEP} = 11.0	
	Inorganic colloids	Silica [62]	Round	Negatively charged (pH_{pzc}~3)	SEM-EDX
		Aluminiun silicate minerals [62]	Angular	Negatively charged at pH~7	
		Ferric oxides/hydroxide [62]	Varies depending on crystalline or amorphous	Positively charged. pH_{pzc} for goethite ~9	
	Others	Transparent exopolymer particles (TEP) [72, 91, 92]	Transparent, sticky and amorphous substances. Exists in different forms (e.g. strings, disks, sheets, fibers) and sizes (up to 100 s of mm long).	-	(1) Microscopic enumeration, and (2) Colorimetric determination [90]. Both methods are based on staining with alcian blue.

Types of Fouling	Foulants	Examples	Size and shape	charge	Characterization method
		Latex	Mean diameter of 3 µm, nearly monodispersed spherical shape	-	-
Inorganic scaling	Inorganic scales	Calcium sulphate (or Gypsum) [96]	Needle-like and plate-like	Non-alkaline scale that is pH independent	SEM-EDX
		Calcium carbonate [96]	Varied from forms. Calcite: 10 µm, rhombohedral; aragonite clusters: outward oriented needles; vaterite: 0.05 to 5 µm, spherical.	Alkaline scale that is dependent on the bicarbonate alkaline and pH	
		Calcium phosphate [96]	Amorphous	Alkaline scale that is dependent on the bicarbonate alkaline and pH	
		Silicate scale [96]	crystalline or amorphous form	The formation of silica scale is dependent on the pH.	
Biofouling	Bacteria [103]	*Pseudomonas* [38]	Rod-shaped	-	polymerase chain reaction denaturing gradient gel electrophoresis (PCR–DGGE) and Fluorescence In Situ Hybridization (FISH) [86, 105]
		E. coli, [185]	Rod-shaped	-	
		Corynebacterium, [103]	Rod-shaped	-	
		B. subtilis [185]	Rod-shaped	-	
	Fungi [103]	*Penicillium*	Brush-like and flask-shaped	-	
		Trichoderma	Divergent and flask-shaped	-	
	Microalgae [39, 65]	*Chlorella sorokiniana*	Cell diameter ~5 µm,	Negatively charged at pH 7.2	

Note: “-” indicates not available.

Table 3. Summary of membrane modification to improve anti-fouling properties in ODMPs.

Base membrane	Modification method	Properties improved	Function of the membrane	Reference
TFC polyamide flat-sheet membrane	Dip coating of the functionalized superhydrophilic silica nanoparticles with positively charged ammonium groups on onto the surface of polyamide membrane with negatively charged carboxylic groups via electrostatic attraction	The modified membrane primarily has superhydrophilic surface. The negatively charged carboxyl groups are neutralized and covered by the positively charged nanoparticles.	The functionalized membrane showed improved fouling resistance and cleaning efficiency in organic fouling (BSA, alginate, and humic acid fouling) experiments	[172]
TFC polyamide flat-sheet membrane	In situ grafting Jeffamine, an amine-terminated poly(ethylene glycol) derivative, to dangling acyl chloride surface groups on the nascent polyamide active layer.	(1) the grafted Jeffamine molecules impose a steric barrier against the adsorption of the foulant molecules, (2) decreased number of surface carboxyl groups, (3) increased hydrophilicity and reduced roughness of membrane surface	The modified membrane showed significantly improved fouling resistance and fouling reversibility in the alginate fouling test.	[253]
Cellulose triacetate (CTA) flat-sheet membrane	Coating the poly amino acid 3-(3,4- ihydroxyphenyl)-L-alanine (L-DOPA), a zwitterionic polymer on the surface of membrane porous support layer	The modified membrane surfaces became more hydrophilic.	In alginate fouling experiments, the modified membrane showed improved fouling resistance and reversibility	[181]
TFC polyamide flat-sheet membrane	Grafting the polyzwitterions (PZs) (i.e., alkyne-polyMEDSAH) onto polyamide membrane surface through click chemical reaction.	The modified membrane has (1) shielded specific binding sites (e.g., carboxylic groups), (2) improved hydrophilicity, and (3) long, flexible, brush-like PZ chains that improved the steric repulsion.	Using humic acid as a model foulant, the PZ-grafted polyamide membrane showed improved antifouling performance.	[182]
TFC polyamide flat-sheet membrane	Firstly, a second interfacial reaction is carried out between ethylenediamine and acyl chloride groups on the nascent polyamide layer. Secondly, the resulting amine-rich active layer is	The modified membrane has (1) improved hydrophilicity and (2) reduced carboxylic groups.	Using alginate as organic foulant, the modified membrane showed significantly reduced fouling.	[183]

Base membrane	Modification method	Properties improved	Function of the membrane	Reference
	functionalized with FO membranes functionalized with poly (ethylene glycol) (PEG) diglycidyl ether.			
TFC polyamide hollow fibre membrane with poly(ether sulfone) (PES) support layer	The PES support layer was first coated with polydopamine (PDA) and then grafted with either poly(ethylene glycol) (PEG) or hyperbranched polyglycerol (HPG)	The PEG-grafted-PES membrane and HPG-grafted-PES membrane were more hydrophilic than pristine PES membrane. The HPG-g-PES membrane has dendritic polymeric structure, while the PEG has linear polymeric structure.	In the protein adsorption and bacteria adhesion tests, the HPG-g-PES membrane exhibited much superior resistance against bovine serum albumin (BSA) adsorption, E. coli adhesion, and S. aureus attachment. The HPG grafted membrane showed least fouling in dynamic PRO fouling test and best water flux recovery after physical cleaning.	[254]
TFC polyamide flat-sheet membrane with polysulfone (PSf) support layer	Blending the titanium dioxide (TiO_2) nanoparticles into the PSf dope solution for casting the support layer	The PSf-TiO_2 support layer of the formed thin-film nanocomposite (TFN) membrane has greater hydrophilicity and porosity but smaller structural parameter than the PSf support layer of the TFC membrane.	Using BSA as foulant, the TFN membrane showed better fouling resistance in fouling test and better water flux recovery after cleaning.	[255]
Flat sheet membrane with NF-like rejection layer through layer-by-layer (LBL) assembly and crosslinking on top surface of PAN substrate	In situ incorporation of silver nanoparticles in the rejection layer of the base membrane	The silver nanocomposite membranes exhibit excellent antibacterial properties against both Gram-positive and Gram-negative bacteria in the colony-forming units test, bacterial cell membrane compromise test, and diffusion inhibition zone test	The silver nanocomposite membranes showed great promise in biofouling control.	[185]
Cellulose triacetate (CTA) flat-sheet membrane	Silver (Ag) nanoparticles were first deposited on the membrane surface by a photo induced growth approach. Then, the	The silver nanoparticles coated on membrane exhibited strong antibacterial effect, while the TiO2 played an effective role in	In the accelerated biofouling experiment, the Ag/TiO2- coated membranes experienced less flux reduction than the pristine	[184]

Base membrane	Modification method	Properties improved	Function of the membrane	Reference
	charge-driven self-assembly of titanium dioxide (TiO2) nanoparticles took place on the layers of silver nanoparticles.	regenerating the silver nanoparticles by decomposing the organic matter that covered the silver nanoparticles	membrane. The modified membranes also achieved more water flux recovery after physical cleaning.	
Single-skinned hollow fiber membrane with RO-like polyamide TFC layer in the lumen of PAI substrate	Form a second NF-like skin at outer surface through the chemical crosslinking between PEI and PAI	The second NF-like skin acts as a barrier to reject the passage of divalent ions into the support layer	The double-skinned membrane showed lower propensity to inorganic scaling	[189]
Single-skinned flat sheet membrane with NF-like rejection layer through layer-by-layer (LBL) assembly and crosslinking on top surface of PAN substrate	Form a second skin on the bottom surface of the PAN substrate through LBL assembly and crosslinking	The bottom skin acts as a barrier to prevent the organic foulant clogging the porous support layer	The double-skinned membrane showed greater resistance to organic fouling	[56]
Single-skinned flat sheet membrane with RO-like TFC polyamide layer on the top surface of PAN substrate	Form a second skin (a self-assembled sulfonated pentablock copolymer (Nexar copolymer) layer) on the bottom of PAN substrate layer	The bottom skin acts as a barrier to reject the passage of emulsified oil particles into the porous substrate layer	The double-skinned membrane showed superior anti-fouling in emulsified oil-water separation process and greater flux restoration after physical cleaning	[190]
Single-skinned flat sheet membrane with NF-like TFC and TFN polyamide layer on the top surface of PSf substrate	A second skin was simultaneously formed through interfacial polymerization between dopamine hydrochloride (DA) and trimesoyl chloride (TMC)	The bottom skin acts as a barrier to prevent the organic foulant clogging the porous support layer	The double-skinned membrane showed greater resistance and reversibility to humic acid fouling	[256]